

# **Integrated thermal-fluid and statistical modeling for fault prediction in fixed-bed dry bottom coal gasifiers**

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## ABSTRACT

In this study, an integrated thermal-fluid and statistical modeling approach is developed for fixed-bed dry bottom coal gasifier fault prediction. This research is aimed to address the problem of current gasifier fault prediction models relying on data driven statistical techniques that do not incorporate thermal-fluid modeling. Both gasifier thermal-fluid and statistical modeling approaches are established. However, in the reviewed literature, there is no evidence of integrated thermal-fluid and statistical modeling approaches for gasifier fault prediction.

A steady-state thermal-fluid model of the fixed-bed dry bottom coal gasifier is developed in this study. This model predicts mass flow rates for the five gases that mainly comprise syngas. As such, five fundamental thermodynamic equations are selected and applied in the model. These are three mole balances, an energy balance, and a chemical reaction equilibrium condition.

Software for the Engineering Equation Solver (EES) is utilized to develop the thermal-fluid model. It is implemented so that abnormal gasifier operating conditions, where negative flow rates are predicted, can be isolated from typical data. When compared with a reference gasifier observation, the average accuracy of this model's predictions is comparable with that of a fixed-bed gasifier model available in literature.

Statistical modeling is undertaken using the R Statistical Software (R). To predict where the gasifier's operation transitions from normal to abnormal conditions, statistical classification models are applied to the thermal-fluid model's outputs. A classification model is selected that predicts a bounded region wherein the gasifier's operations are normal.

The existence of gasifier faults is discussed with respect to groups of predicted abnormal observations. Observations are presented on a plot of the selected classification model, and groups are identified, with a statistical clustering technique, according to the gasifier's inlet flow rates. The thermal-fluid model's validity is established through an analysis of the changes evident between these groups. When compared with conventional data driven fault prediction for fixed-bed dry bottom coal gasifiers, the selected classification model possesses a higher accuracy.

**Keywords:** Thermal-Fluid, Statistical, Modeling, Fault, Abnormal, Fixed-Bed, Coal, Gasifier, Syngas

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# NOMENCLATURE

## Abbreviations

| Abbreviation                    | Unit                                 |
|---------------------------------|--------------------------------------|
| °C                              | degrees Celsius                      |
| K                               | Kelvins                              |
| kg/m <sup>3</sup>               | kilograms per cubic meter            |
| kg/kmol                         | kilograms per kilomole               |
| kg/rev                          | kilograms per revolution             |
| kg/s                            | kilograms per second                 |
| kg/t                            | kilograms per tonne                  |
| kJ/kg                           | kilojoules per kilogram              |
| kJ/kg-K                         | kilojoules per kilogram Kelvin       |
| kJ/kmol                         | kilojoules per kilomole              |
| kJ/kmol-K                       | kilojoules per kilomole Kelvin       |
| kmol/s                          | kilomoles per second                 |
| kPa                             | kilopascals                          |
| kW                              | kilowatts                            |
| m <sub>n</sub> <sup>3</sup> /kg | normalized cubic meters per kilogram |
| m <sub>n</sub> <sup>3</sup> /s  | normalized cubic meters per second   |
| rev/h                           | revolutions per hour                 |
| rev/s                           | revolutions per second               |
| s/h                             | seconds per hour                     |

$10^3\text{m}_n^3/\text{h}$  thousand Normalized cubic meters per hour

t/h tonnes per hour

## Acronyms

| Acronym | Full title                   |
|---------|------------------------------|
| AD      | air dried                    |
| AR      | as received                  |
| AF      | ash free                     |
| CT      | classification tree          |
| EES     | Engineering Equation Solver  |
| FC      | fixed carbon                 |
| LHS     | left hand side               |
| MMA     | mineral matter ash           |
| PCA     | principal component analysis |
| R       | R Statistical Software       |
| RHS     | right hand side              |
| SVM     | support vector machine       |
| WAF     | water and ash free           |
| WF      | water free                   |
| VM      | volatile matter              |

## Symbols

| Symbol       | Heading/Sub-heading | Variable                     |
|--------------|---------------------|------------------------------|
| $Ac$         |                     | accuracy                     |
| $cl$         |                     | actual class                 |
| $f_{com}$    |                     | combustion value             |
| $\in$        |                     | element of                   |
| $FAR$        |                     | false-alarm rate             |
| $\forall$    |                     | for all                      |
| $Gn$         |                     | Gini index                   |
| $Ash_{rm}$   |                     | grate ash removal rate       |
| $Gr t_{frq}$ |                     | grate rotational frequency   |
| $\cap$       |                     | intersection of              |
| $min$        |                     | minimum of                   |
| $\ln$        |                     | natural logarithm            |
| $NDR$        |                     | non-detection rate           |
| $num$        |                     | number of                    |
| $\%$         |                     | percent                      |
| $cl'$        |                     | predicted class              |
| $ $          |                     | such that                    |
| $\Sigma$     |                     | summation of                 |
| $\omega$     |                     | total number of observations |
| $R_{unv}$    |                     | universal gas constant       |

## classification tree

$sp_b$  split  $b$

$Tg_a$  terminal region  $a$

proportion of observations in terminal region  $a$

$pr_{a,cl}$  class  $c/$

regions for split  $b$

$Rg_{2b}$  above

$Rg_{2b-1}$  below

## constant pressure specific heat

$cp_{ash}$  ash

$cp_{coal}$  coal

$cp_{liqH2O}$  liquid water

$cp_{FC\&VM}$  water and ash free coal

## density at Normal conditions

$\rho_{CO2}$  carbon dioxide

$\rho_{CO}$  carbon monoxide

$\rho_{H2}$  hydrogen

$\rho_{CH4}$  methane

$\rho_{O2}$  oxygen

## elemental symbol

$C$  carbon

$H$  hydrogen

|     |          |
|-----|----------|
| $N$ | nitrogen |
|-----|----------|

|     |        |
|-----|--------|
| $O$ | oxygen |
|-----|--------|

|     |        |
|-----|--------|
| $S$ | sulfur |
|-----|--------|

**enthalpy**

change

|                  |     |
|------------------|-----|
| $\Delta h_{ash}$ | ash |
|------------------|-----|

difference at inlet

|                   |      |
|-------------------|------|
| $\Delta h_{coal}$ | coal |
|-------------------|------|

|                     |        |
|---------------------|--------|
| $\Delta h_{O_2,in}$ | oxygen |
|---------------------|--------|

|                      |       |
|----------------------|-------|
| $\Delta h_{H_2O,in}$ | steam |
|----------------------|-------|

difference at outlet

|                       |                |
|-----------------------|----------------|
| $\Delta h_{CO_2,out}$ | carbon dioxide |
|-----------------------|----------------|

|                     |                 |
|---------------------|-----------------|
| $\Delta h_{CO,out}$ | carbon monoxide |
|---------------------|-----------------|

|                      |          |
|----------------------|----------|
| $\Delta h_{H_2,out}$ | hydrogen |
|----------------------|----------|

|                       |         |
|-----------------------|---------|
| $\Delta h_{CH_4,out}$ | methane |
|-----------------------|---------|

|                       |       |
|-----------------------|-------|
| $\Delta h_{H_2O,out}$ | steam |
|-----------------------|-------|

|                  |     |
|------------------|-----|
| $\Delta h_{tar}$ | tar |
|------------------|-----|

formation

|                |                |
|----------------|----------------|
| $h_{CO_2,frm}$ | carbon dioxide |
|----------------|----------------|

|              |                 |
|--------------|-----------------|
| $h_{CO,frm}$ | carbon monoxide |
|--------------|-----------------|

|                |      |
|----------------|------|
| $h_{coal,frm}$ | coal |
|----------------|------|

|               |          |
|---------------|----------|
| $h_{H_2,frm}$ | hydrogen |
|---------------|----------|

|                    |           |                         |
|--------------------|-----------|-------------------------|
| $h_{liqH_2O,frm}$  |           | liquid water            |
| $h_{CH_4,frm}$     |           | methane                 |
| $h_{O_2,frm}$      |           | oxygen                  |
| $h_{H_2O,frm}$     |           | steam                   |
| $h_{SO_2,frm}$     |           | sulfur dioxide          |
| $h_{FC\&VM,frm}$   |           | water and ash free coal |
|                    | ideal gas |                         |
| $h_{CO_2,out,idl}$ |           | carbon dioxide          |
| $h_{CO,out,idl}$   |           | carbon monoxide         |
| $h_{H_2,out,idl}$  |           | hydrogen                |
| $h_{CH_4,out,idl}$ |           | methane                 |
| $h_{H_2O,out,idl}$ |           | steam                   |
|                    | inlet     |                         |
| $h_{coal}$         |           | coal                    |
| $h_{O_2,in}$       |           | oxygen                  |
| $h_{H_2O,in}$      |           | steam                   |
| $h_{wj,in}$        |           | water jacket            |
|                    | outlet    |                         |
| $h_{CO_2,out}$     |           | carbon dioxide          |
| $h_{CO,out}$       |           | carbon monoxide         |
| $h_{H_2,out}$      |           | hydrogen                |
| $h_{CH_4,out}$     |           | methane                 |

|               |       |
|---------------|-------|
| $h_{H2O,out}$ | steam |
|---------------|-------|

|              |              |
|--------------|--------------|
| $h_{wj,out}$ | water jacket |
|--------------|--------------|

enthalpy-flow

inlet

|                  |      |
|------------------|------|
| $\dot{H}_{coal}$ | coal |
|------------------|------|

|                   |        |
|-------------------|--------|
| $\dot{H}_{O2,in}$ | oxygen |
|-------------------|--------|

|                    |       |
|--------------------|-------|
| $\dot{H}_{H2O,in}$ | steam |
|--------------------|-------|

outlet

|                     |                |
|---------------------|----------------|
| $\dot{H}_{CO2,out}$ | carbon dioxide |
|---------------------|----------------|

|                    |                 |
|--------------------|-----------------|
| $\dot{H}_{CO,out}$ | carbon monoxide |
|--------------------|-----------------|

|                    |          |
|--------------------|----------|
| $\dot{H}_{H2,out}$ | hydrogen |
|--------------------|----------|

|                     |         |
|---------------------|---------|
| $\dot{H}_{CH4,out}$ | methane |
|---------------------|---------|

|                     |       |
|---------------------|-------|
| $\dot{H}_{H2O,out}$ | steam |
|---------------------|-------|

tar

|                       |            |
|-----------------------|------------|
| $\Delta\dot{H}_{tar}$ | difference |
|-----------------------|------------|

|                     |           |
|---------------------|-----------|
| $\dot{H}_{tar,frm}$ | formation |
|---------------------|-----------|

|                 |       |
|-----------------|-------|
| $\dot{H}_{tar}$ | total |
|-----------------|-------|

entropy

|                   |                |
|-------------------|----------------|
| $S_{CO2,out,idl}$ | carbon dioxide |
|-------------------|----------------|

|                  |                 |
|------------------|-----------------|
| $S_{CO,out,idl}$ | carbon monoxide |
|------------------|-----------------|

|                  |          |
|------------------|----------|
| $S_{H2,out,idl}$ | hydrogen |
|------------------|----------|

|                   |         |
|-------------------|---------|
| $S_{CH4,out,idl}$ | methane |
|-------------------|---------|

|                    |       |
|--------------------|-------|
| $S_{H_2O,out,idl}$ | steam |
|--------------------|-------|

### equilibrium constant

|            |             |
|------------|-------------|
| $K_{meth}$ | methanation |
|------------|-------------|

|           |                 |
|-----------|-----------------|
| $K_{wgs}$ | water gas shift |
|-----------|-----------------|

### error

|       |                     |
|-------|---------------------|
| $CER$ | classification rate |
|-------|---------------------|

|       |               |
|-------|---------------|
| $SSE$ | sum of square |
|-------|---------------|

### relative

|             |                |
|-------------|----------------|
| $RE_{CO_2}$ | carbon dioxide |
|-------------|----------------|

|           |                 |
|-----------|-----------------|
| $RE_{CO}$ | carbon monoxide |
|-----------|-----------------|

|            |          |
|------------|----------|
| $RE_{H_2}$ | hydrogen |
|------------|----------|

|             |         |
|-------------|---------|
| $RE_{CH_4}$ | methane |
|-------------|---------|

### Gibbs function

|                |                |
|----------------|----------------|
| $g_{CO_2,out}$ | carbon dioxide |
|----------------|----------------|

|              |                 |
|--------------|-----------------|
| $g_{CO,out}$ | carbon monoxide |
|--------------|-----------------|

|               |          |
|---------------|----------|
| $g_{H_2,out}$ | hydrogen |
|---------------|----------|

|                |         |
|----------------|---------|
| $g_{CH_4,out}$ | methane |
|----------------|---------|

|                |       |
|----------------|-------|
| $g_{H_2O,out}$ | steam |
|----------------|-------|

### heat absorption rate

|                       |     |
|-----------------------|-----|
| $\Delta\dot{Q}_{ash}$ | ash |
|-----------------------|-----|

|                      |              |
|----------------------|--------------|
| $\Delta\dot{Q}_{wj}$ | water jacket |
|----------------------|--------------|

## heat loss

$Ht_{loss}$  fraction

$\dot{Q}_{loss}$  rate

## heat of combustion

$h_{coal,com}$  coal

## heating value

$HHV_{WAF}$  higher

$LHV_{WAF}$  lower

## mass flow rate

$\dot{m}_{wj}$  water jacket

developed during devolatilization

$\dot{m}_{CO_2,dvl}$  carbon dioxide

$\dot{m}_{CO,dvl}$  carbon monoxide

$\dot{m}_{H_2,dvl}$  hydrogen

$\dot{m}_{CH_4,dvl}$  methane

developed during drying

$\dot{m}_{H_2O,dry}$  steam

developed during gasification

$\dot{m}_{CO_2,gas}$  carbon dioxide

$\dot{m}_{CO,gas}$  carbon monoxide

$\dot{m}_{H_2,gas}$  hydrogen

$\dot{m}_{CH_4,gas}$  methane

$\dot{m}_{H_2O,gas}$  steam

inlet

$\dot{m}_{coal}$  coal

$\dot{m}_{C,in}$  fixed carbon

$\dot{m}_{O_2,in}$  oxygen

$\dot{m}_{H_2O,in}$  steam

outlet

$\dot{m}_{ash}$  ash

$\dot{m}_{CO_2,out}$  carbon dioxide

$\underline{\dot{m}}_{CO_2,out}$  carbon dioxide vector

$\dot{m}_{CO,out}$  carbon monoxide

$\underline{\dot{m}}_{CO,out}$  carbon monoxide vector

$\dot{m}_{C,out}$  fixed carbon

$\dot{m}_{H_2,out}$  hydrogen

$\underline{\dot{m}}_{H_2,out}$  hydrogen vector

$\dot{m}_{CH_4,out}$  methane

$\underline{\dot{m}}_{CH_4,out}$  methane vector

$\dot{m}_{H_2O,out}$  steam

$\underline{\dot{m}}_{H_2O,out}$  steam vector

tar

$\dot{m}_{C,tar}$  carbon

$\dot{m}_{H,tar}$  hydrogen

$\dot{m}_{S,tar}$  sulfur

$\dot{m}_{tar}$  total

**mass fraction**

air dried coal

$Tar_{AD,coal}$  tar

as received ash

$FC_{AR,ash}$  fixed carbon

as received coal

$FC_{AR,coal}$  fixed carbon

$Mst_{AR,coal}$  moisture

$VM_{AR,coal}$  volatile matter

ash free coal

$FC_{AF,coal}$  fixed carbon

$Mst_{AF,coal}$  moisture

$VM_{AF,coal}$  volatile matter

tar

$C_{WAF,tar}$  carbon

$H_{WAF,tar}$  hydrogen

$S_{WAF,tar}$  sulfur

water and ash free coal

$C_{WAF,coal}$  carbon

$FC_{WAF,coal}$  fixed carbon

|                 |                   |                 |
|-----------------|-------------------|-----------------|
| $H_{WAF,coal}$  |                   | hydrogen        |
| $N_{WAF,coal}$  |                   | nitrogen        |
| $O_{WAF,coal}$  |                   | oxygen          |
| $S_{WAF,coal}$  |                   | sulfur          |
| $VM_{WAF,coal}$ |                   | volatile matter |
|                 | water free coal   |                 |
| $FC_{WF,coal}$  |                   | fixed carbon    |
| $VM_{WF,coal}$  |                   | volatile matter |
|                 | <b>molar mass</b> |                 |
| $M_C$           |                   | carbon          |
| $M_{CO_2}$      |                   | carbon dioxide  |
| $M_{CO}$        |                   | carbon monoxide |
| $M_{CH_4}$      |                   | methane         |
| $M_{O_2}$       |                   | oxygen          |
| $M_{H_2O}$      |                   | steam           |
| $M_S$           |                   | sulfur          |
| $M_{SO_2}$      |                   | sulfur dioxide  |
|                 | hydrogen          |                 |
| $M_{H_2}$       |                   | diatomic        |
| $M_H$           |                   | elemental       |

## mole flow rate

developed during devolatilization

$\dot{n}_{CO_2,dvl}$  carbon dioxide

$\dot{n}_{CO,dvl}$  carbon monoxide

$\dot{n}_{H_2,dvl}$  hydrogen

$\dot{n}_{CH_4,dvl}$  methane

developed during drying

$\dot{n}_{H_2O,dry}$  steam

developed during gasification

$\dot{n}_{CO_2,gas}$  carbon dioxide

$\dot{n}_{CO,gas}$  carbon monoxide

$\dot{n}_{H_2,gas}$  hydrogen

$\dot{n}_{CH_4,gas}$  methane

$\dot{n}_{H_2O,gas}$  steam

inlet

$\dot{n}_{C,in}$  fixed carbon

$\dot{n}_{O_2,in}$  oxygen

$\dot{n}_{H_2O,in}$  steam

$\dot{n}_{H_2O\&O_2}$  sum of steam and oxygen

outlet

$\dot{n}_{C,out}$  fixed carbon

$\dot{n}_{sgs}$  sum of syngas

|                   |     |          |
|-------------------|-----|----------|
|                   | tar |          |
| $\dot{n}_{C,tar}$ |     | carbon   |
| $\dot{n}_{H,tar}$ |     | hydrogen |
| $\dot{n}_{S,tar}$ |     | sulfur   |

### mole fraction

|                |        |                 |
|----------------|--------|-----------------|
|                | inlet  |                 |
| $y_{O_2,in}$   |        | oxygen          |
| $y_{H_2O,in}$  |        | steam           |
|                | outlet |                 |
| $y_{CO_2,out}$ |        | carbon dioxide  |
| $y_{CO,out}$   |        | carbon monoxide |
| $y_{H_2,out}$  |        | hydrogen        |
| $y_{CH_4,out}$ |        | methane         |
| $y_{H_2O,out}$ |        | steam           |

### molecular formula

|           |  |                  |
|-----------|--|------------------|
| $Al_2O_3$ |  | aluminum oxide   |
| $NH_3$    |  | ammonia          |
| $CaO$     |  | calcium oxide    |
| $CO_2$    |  | carbon dioxide   |
| $CO$      |  | carbon monoxide  |
| $COS$     |  | carbonyl sulfide |
| $Fe_2O_3$ |  | ferric oxide     |

|         |                  |
|---------|------------------|
| $H_2$   | hydrogen         |
| $HCN$   | hydrogen cyanide |
| $H_2S$  | hydrogen sulfide |
| $CH_4$  | methane          |
| $N_2$   | nitrogen         |
| $O_2$   | oxygen           |
| $SiO_2$ | silica           |
| $SO_2$  | sulfur dioxide   |
| $H_2O$  | water            |

#### Normal volume developed per unit mass of coal

|                  |                 |
|------------------|-----------------|
| $CO_{2WAF,coal}$ | carbon dioxide  |
| $CO_{WAF,coal}$  | carbon monoxide |
| $H_{2WAF,coal}$  | hydrogen        |
| $CH_{4WAF,coal}$ | methane         |

#### Normal volume flow rate

developed during devolatilization

|                      |                 |
|----------------------|-----------------|
| $\dot{V}_{CO_2,dvl}$ | carbon dioxide  |
| $\dot{V}_{CO,dvl}$   | carbon monoxide |
| $\dot{V}_{H_2,dvl}$  | hydrogen        |
| $\dot{V}_{CH_4,dvl}$ | methane         |
|                      | inlet           |
| $\dot{V}_{O_2,in}$   | oxygen          |

|                      |              |                 |
|----------------------|--------------|-----------------|
|                      | total outlet |                 |
| $\dot{V}_{CO_2,out}$ |              | carbon dioxide  |
| $\dot{V}_{CO,out}$   |              | carbon monoxide |
| $\dot{V}_{H_2,out}$  |              | hydrogen        |
| $\dot{V}_{CH_4,out}$ |              | methane         |
| $\dot{V}_{sgs}$      |              | syngas          |

### pressure

|                 |                              |                  |
|-----------------|------------------------------|------------------|
| $P_{frm}$       | formation enthalpy reference |                  |
|                 | inlet                        |                  |
| $P_{O_2,in}$    |                              | oxygen           |
| $P_{H_2O,in}$   |                              | steam            |
| $P_{H_2O\&O_2}$ |                              | steam and oxygen |

### outlet

|                |  |                 |
|----------------|--|-----------------|
| $P_{CO_2,out}$ |  | carbon dioxide  |
| $P_{CO,out}$   |  | carbon monoxide |
| $P_{H_2,out}$  |  | hydrogen        |
| $P_{CH_4,out}$ |  | methane         |
| $P_{H_2O,out}$ |  | steam           |
| $P_{sgs}$      |  | syngas          |

### principal component

|                    |  |          |
|--------------------|--|----------|
| $\underline{pc_x}$ |  | vector x |
| $pc_x$             |  | x        |

component x loading

|              |                 |
|--------------|-----------------|
| $\phi_{x,4}$ | carbon dioxide  |
| $\phi_{x,3}$ | carbon monoxide |
| $\phi_{x,2}$ | hydrogen        |
| $\phi_{x,5}$ | methane         |
| $\phi_{x,1}$ | steam           |

component x score

|                 |                      |
|-----------------|----------------------|
| $pc_x^*$        | new observation      |
| $pc_{x,\varpi}$ | observation $\varpi$ |
| $pc_{x,w}$      | observation $w$      |

support vector machine

|                   |                         |
|-------------------|-------------------------|
| $\beta_0$         | beta 0                  |
| $\delta_0$        | delta 0                 |
| $\gamma$          | gamma                   |
| $K$               | kernel function         |
| $Mi$              | margin width            |
| $Cs$              | total margin violations |
|                   | observation $\varpi$    |
| $\alpha_{\varpi}$ | alpha                   |
|                   | observation $w$         |
| $f_w$             | class factor            |
| $\varepsilon_w$   | margin violation        |

outlet mass flow rate  $j$

$\dot{m}_{j,out}^*$  new observation

$\dot{m}_{j,out,\varpi}$  observation  $\varpi$

$\dot{m}_{j,out,w}$  observation  $w$

### temperature

$T_{ash,avg}$  average ash

$T_{frm}$  formation enthalpy reference

inlet

$T_{ash,in}$  ash

$T_{coal}$  coal

$T_{H2O\&O2}$  steam and oxygen

outlet

$T_{ash,out}$  ash

$T_{sgs}$  syngas

$T_{tar}$  tar

### volume percentages

predicted

$V_{CO2}$  carbon dioxide

$V_{CO}$  carbon monoxide

$V_{H2}$  hydrogen

$V_{CH4}$  methane

reference

|              |                 |
|--------------|-----------------|
| $Ref_{CO_2}$ | carbon dioxide  |
| $Ref_{CO}$   | carbon monoxide |
| $Ref_{H_2}$  | hydrogen        |
| $Ref_{CH_4}$ | methane         |

# CHAPTER 1: INTRODUCTION

## 1.1 Background

The coal gasifier is a technology that underpins many chemical processes important in modern society. It is used to convert coal, containing carbon, to synthesis gas, also known as syngas, which is mainly comprised of carbon monoxide ( $CO$ ) and hydrogen ( $H_2$ ) (Speight, 2020:41,70). Syngas can be converted into various commodities including synthetic natural gas, ammonia, diesel, and petrol (Dancuart *et al.*, 2004:483; Speight, 2020:140,409). An advantage reported for coal gasification is a lower environmental impact than other uses of this abundant fossil fuel (Aasberg-Petersen *et al.*, 2004:354; Meehan, 2017:15; Speight, 2020:43,51,125). There are various types of coal gasifiers currently in use, where the operations of one of these will be investigated in this study (Higman & van der Burgt, 2008:91).

Fixed-bed dry bottom coal gasifiers will be the main focus of the current research. With these gasifiers, coal is loaded from above, which is gasified with steam ( $H_2O$ ) and oxygen ( $O_2$ ) that are supplied from below (Higman & van der Burgt, 2008:95; Speight, 2020:68,70). To ensure that the ash formed can be removed as a solid, a limit is imposed on the maximum allowable temperature at the base (Aasberg-Petersen *et al.*, 2004:371). This gasifier's most distinctive features are the ammonia, tars, phenols, and various hydrocarbons contained in the exiting syngas (Aasberg-Petersen *et al.*, 2004:361; Higman & van der Burgt, 2008:97).

Over time, process deviations that negatively influence gasification operations can arise. These deviations will be referred to as faults throughout this study. Potential faults are indicated by changes in gasifier data, such as in the temperature and pressure measurements, that are of greater magnitude than the fluctuations normally present (Guo *et al.*, 2021:6). One fixed-bed dry bottom coal gasifier fault, known as channel burning, involves localized ash formation that leads to excessive variability in the syngas' composition and exit temperature (Aasberg-Petersen *et al.*, 2004:369; Keyser *et al.*, 2006:1444).

The modeling of gasifier operations, for process prediction and performance adjustment, is an active area of study (Higman & van der Burgt, 2008:16). Some approaches incorporate thermal-fluid principles and empirical relationships, while others are data driven statistical techniques. Examples of the former include the work by Luckos & Bunt (2011), He *et al.* (2013), and Ismail *et al.* (2020), and examples of the latter include the work by Coetzer *et al.* (2014) and Guo *et al.* (2021).

Although the thermal-fluid approaches are established, it is reported that the prediction accuracy of these, in terms of syngas yield and composition, needs to be improved (Mularski *et al.*, 2020:24-25). The statistical approaches can yield accurate predictions, but consideration must be given to the applicability of the specific techniques to the commercial process. Obtaining accurate predictions, for new observations, requires that these approaches model the actual process, instead of random patterns present due to variance in the original measurements (James *et al.*, 2014:8,22,32).

Both the thermal-fluid and statistical approaches are applied in practice. While the former provides a starting point for gasifier operations, the latter is used, after measurements have been taken, to model the process and improve performance (Higman & van der Burgt, 2008:27). This leads to the significance of performing research into the integrated use of these gasification modeling approaches. Furthermore, in the literature that has been reviewed, no evidence was found of a combined thermal-fluid and statistical approach for gasifier fault prediction.

## **1.2 Problem Statement**

Although the faults that can occur in fixed-bed dry bottom coal gasifiers are known, the relationships between these and the thermal-fluid process parameters are not defined sufficiently, in literature, for fault prediction. The consequence of this is that faults cannot be isolated from gasifier measurements, as required to train statistical prediction models. Current statistical fault prediction models for gasifiers rely on data driven techniques that are not integrated with a fundamental understanding of the gasification process.

## **1.3 Research Aim and Objectives**

The research aim is to develop and apply an integrated thermal-fluid and statistical approach for the identification and prediction of fault conditions in a fixed-bed dry bottom coal gasifier. The objectives that underpin the research aim include:

- Developing a thermal-fluid model for normal gasifier operations, which in this study refer to conditions where faults are not present.
- Defining the conditions under which abnormal operations, where faults may be present, are evident in the outputs of the thermal-fluid model.
- Training several statistical learning models to classify typical gasifier data as normal or abnormal.
- Selecting the statistical model that is most applicable to the prediction of abnormal operating conditions.

The following objectives are outside the scope of this research:

- Performing experiments where a gasifier is operated under either normal or fault conditions.
- Developing fault prevention procedures.

#### **1.4 Research Methodology**

The research methodology begins with a study into the operation of fixed-bed dry bottom coal gasifiers, where some faults that can occur will be identified. During this study, the relationship between these faults and the process' thermal-fluid environment, the temperature and pressure in the gasifier, will be investigated. A study will also be completed into the application of statistical learning models. The models investigated will include those that have already been applied to coal gasification or fault prediction.

A thermal-fluid model that outputs key performance indicators, for normal gasifier operations, will be developed. The conditions under which these indicators designate abnormal operations will be defined. In order to predict fault conditions, the thermal-fluid model will be applied to typical data. Accuracy will be determined by comparing the model's outputs, for a given set of gasifier operating conditions, with reference values from literature.

For statistical modeling of abnormal operations, the data will be randomly divided into a training and one or more validation sets. Statistical models will be generated using the training set, and the accuracies of these will be quantified using the validation sets (James *et al.*, 2014:176). Several statistical models, which will be identified later in this study, will be trained. The key performance indicators, output by the thermal-fluid model, will be utilized in these statistical models.

The interpretability of these statistical models, with respect to the gasification process, will be investigated. This is to verify that the models are realistic and do not generate predictions that are contrary to established gasification operations, fault mechanisms, or thermal-fluid knowledge. After rigorous evaluation of the developed models, one will be selected according to several criteria that are still to be defined. The selected model would, in practice, be used as part of some fault detection and prevention procedures. For this reason, feature analysis will be performed to identify the variables that are most likely responsible for abnormal operating conditions. Exact reasons for individual abnormal observations will not be determined. However, this analysis will provide an engineering perspective, in general, of the gasifier's cause and effect relationships for abnormal operations.

The research methodology is visually summarized in the flow chart provided in Figure 1.1.

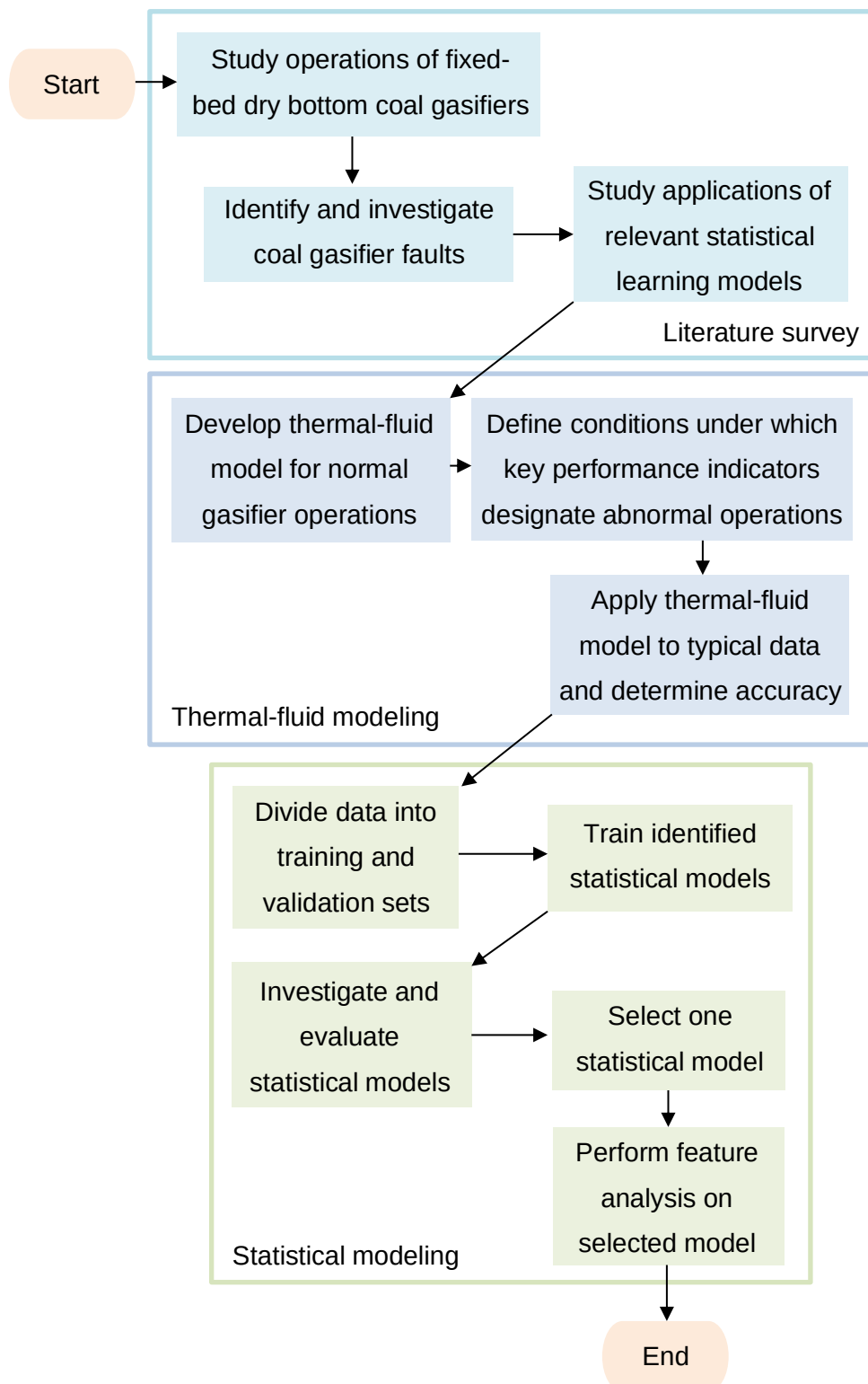


**Figure 1.1: Research Methodology Flow Chart**

## **1.5 Chapter Layout**

This dissertation is structured as follows. Chapter 2 provides literature on the operations, faults, and thermal-fluid modeling of fixed-bed dry bottom coal gasifiers. The most significant chemical reactions that occur in gasifiers are discussed, along with the statistical modeling that is pertinent to this study. Coal analyses are also discussed, and the applicable results thereof are provided.

Chapter 3 discusses the aim and underlying theory of the thermal-fluid model that is developed in this study. As such, the fundamental equations required for this model are provided. Chapter 4 describes the implementation of the thermal-fluid model, and reference gasifier data from literature are provided. The execution of this model in the Engineering Equation Solver (EES) software is also discussed, and the model's calculations are verified.

Chapter 5 presents the statistical models that are applied in this study. The theory underlying these are discussed, and the developed statistical models are evaluated. Results from the thermal-fluid model are also analyzed to determine if changes predicted by the model, for variations in the inlet flow rates, are valid. Chapter 6 provides conclusions from this study and recommendations for further work.

## **CHAPTER 2: LITERATURE SURVEY**

### **2.1 Introduction**

This chapter provides and discusses literature relevant to this study. This begins with an introduction to the operation of fixed-bed dry bottom coal gasifiers in Section 2.2.1. The chemical reactions that occur during gasification are discussed in Section 2.2.2, after which Section 2.2.3 introduces the thermal-fluid principles applicable for modeling this process. Measures used for gasification efficiency are presented in Section 2.2.4, which are followed by a discussion of fixed-bed dry bottom coal gasifier faults in Section 2.2.5. Section 2.2.6 provides notes regarding how this process can be adjusted and controlled, after which syngas contaminants are listed in Section 2.2.7.

Various statistical learning techniques that have been applied to gasification modeling and fault prediction are discussed in Sections 2.3.1 and 2.3.2, respectively. Section 2.4 introduces some of the most frequently applied coal analyses. Section 2.5 concludes the literature survey.

### **2.2 Fixed-Bed Dry Bottom Coal Gasifiers**

#### **2.2.1 Process Operation**

In fixed-bed dry bottom coal gasifiers, coal is loaded at the top, travels down the length, and forms a bed on the grate at the bottom (Higman & van der Burgt, 2008:91; Speight, 2020:65,68). The coal is supplied at specific intervals from a lock-hopper that is pressurized with syngas, where a mechanical stirrer is used to distribute the coal uniformly (Higman & van der Burgt, 2008:95,209). Oxygen and steam are supplied from the bottom of the gasifier and pass through the grate that distributes the flows (Aasberg-Petersen *et al.*, 2004:369; Higman & van der Burgt, 2008:95; Speight, 2020:65,70). These move in a counter-current direction to the coal, where the steam to coal ratio is adjusted to control the composition of the syngas (Higman & van der Burgt, 2008:11; Speight, 2020:46).

The start-up operation of this gasifier is as follows. Coal is loaded into the gasifier and heated with steam to a specified temperature, after which it is ignited by adding air. The air is gradually replaced with oxygen and steam, once the bed is burning uniformly (Supp, 1990:23).

Fixed-bed gasifiers must be fed lump coal composed of coarse particles, usually between 5 and 100 mm in diameter (Aasberg-Petersen *et al.*, 2004:369,372; Higman & van der Burgt, 2008:91; Keyser *et al.*, 2006:1439; van Dyk *et al.*, 2006:249-250). The size of the particles is

adjusted and controlled by crushing the coal and sieving out the lumps that will be gasified (Higman & van der Burgt, 2008:206). In this process, the upper and lower size limits are controlled directly by the equipment used to crush and sieve the coal. The particle size distribution within these limits varies, as this is affected by mining and sizing processes and coal properties (Coetzer & Keyser, 2003:267).

To remove the ash formed in the gasifier, the grate is rotated and the ash is sent to a lock-hopper (Aasberg-Petersen *et al.*, 2004:369,373; Higman & van der Burgt, 2008:93,95). The ash is removed as a solid, requiring that the temperature at the bottom be kept below the ash's fusion point (Aasberg-Petersen *et al.*, 2004:371; Higman & van der Burgt, 2008:22).

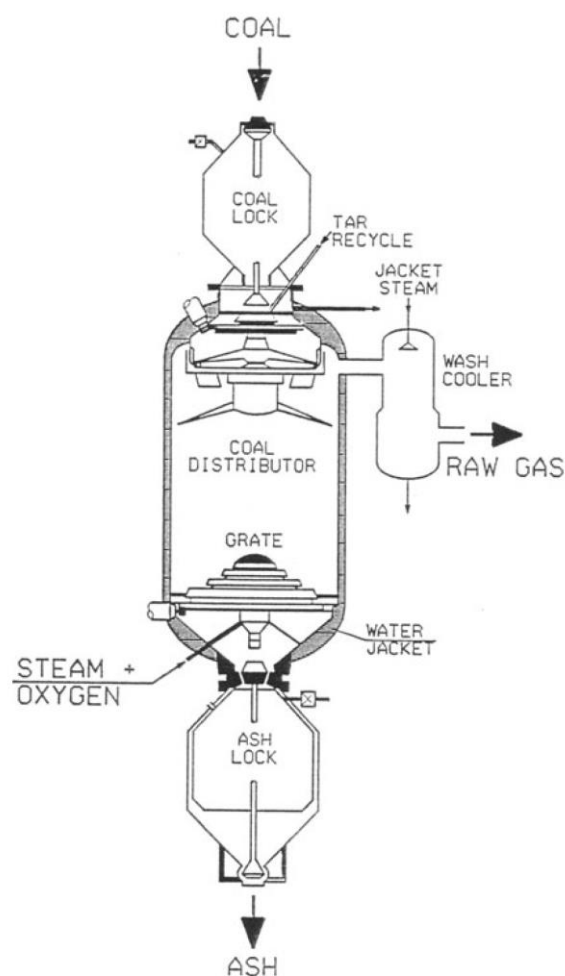


**Figure 2.1: Fixed-Bed Dry Bottom Coal Gasifier Diagram – Supp, 1990:18**

Some of the steam utilized in the gasifier is produced with heat generated in the reactor. This is accomplished through a water jacket that forms part of and cools the gasifier's walls. The water fed into the jacket needs to be at a similar pressure to that inside the gasifier. This limits

the saturated steam quality, the fraction of steam in the total amount of water by mass, that can be developed (Borgnakke & Sonntag, 2017:44; Higman & van der Burgt, 2008:95,213).

The raw syngas exiting a gasifier contains various contaminants that must be removed before it can be utilized. As the processes employed to remove the contaminants operate at temperatures below that of the exiting raw syngas, the gas must first be cooled (Higman & van der Burgt, 2008:217). This process begins with a wash cooler that adds a fraction of gas liquor to the raw syngas. Gas liquor is systematically removed from the syngas as it flows through a series of heat exchangers and is cooled down further. These use the heat transferred from the syngas to heat water and generate low pressure steam for later use (Aasberg-Petersen *et al.*, 2004:367; Supp, 1990:18,20). Figure 2.1 is a schematic diagram of the fixed-bed dry bottom coal gasifier, and Figure 2.2 is a piping and instrument diagram for the gasifier and its steam generation and syngas cooling processes (Supp, 1990:18,20).

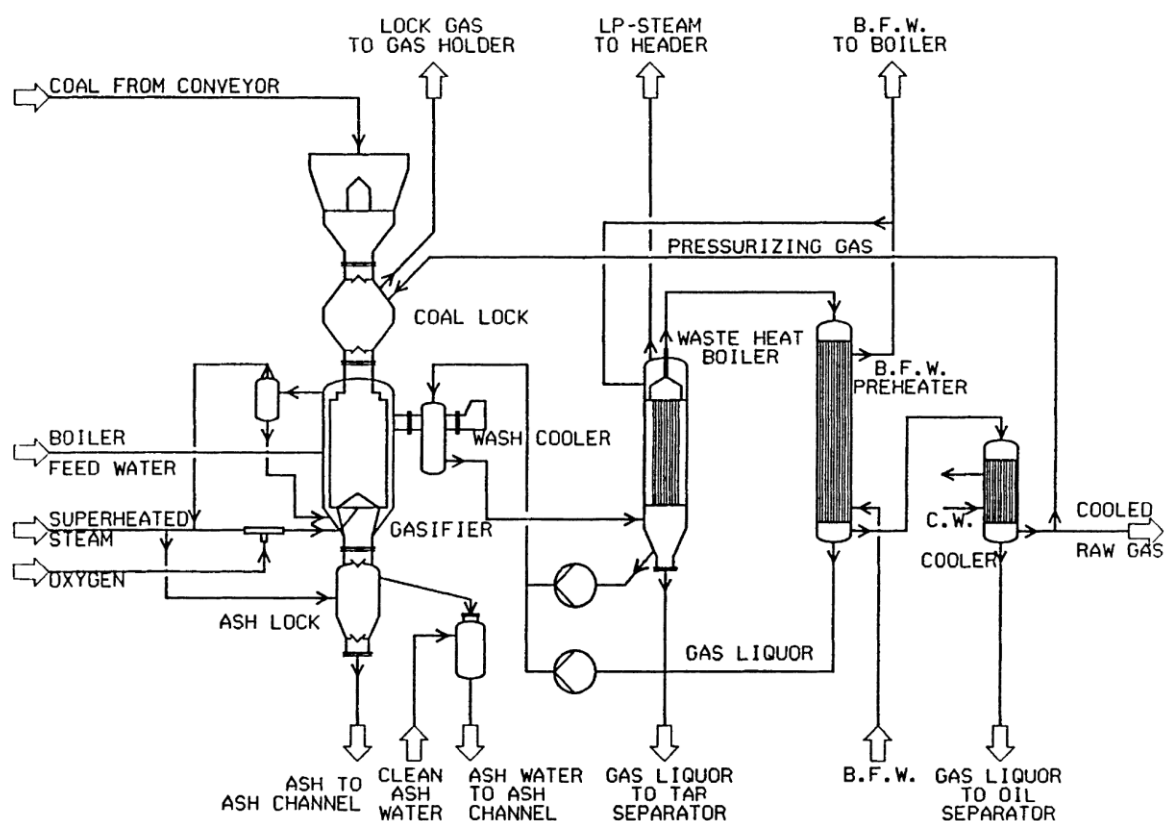


**Figure 2.2: Gasifier Piping and Instrument Diagram – Supp, 1990:20**

### 2.2.2 Chemical Reactions

Along the length of the fixed-bed dry bottom coal gasifier, four regions are present where different chemical reactions occur. These are, starting at the top, drying, devolatilization, gasification, and combustion. These regions overlap to a degree and cannot be completely separated from each other (Higman & van der Burgt, 2008:95-96). Through investigation into coal's chemical properties and composition, along the length of a gasifier, the existence of these regions has been verified (Bunt, 2006:157). Some authors, including Gräbner (2015:249), refer to a pyrolysis instead of a devolatilization region, while other authors, including Aasberg-Petersen *et al.* (2004:360) and Speight (2020:99-100,104), use these terms interchangeably.

Near the top, the exiting raw syngas heats the entering coal (Aasberg-Petersen *et al.*, 2004:372). This results in the drying and thermal decomposition of coal into char and various hydrocarbons. These hydrocarbons mainly include tars and phenols and are referred to as either volatiles or pyrolysis products, depending on which term is used for this gasifier region. In this gasifier, these are not broken down in further pyrolysis reactions and are contained in the exiting syngas (Aasberg-Petersen *et al.*, 2004:361; Gräbner, 2015:109,112; Higman & van der Burgt, 2008:34,97). The term devolatilization can thus be used to define the pyrolysis of coal that occurs without the additional pyrolysis of the resulting hydrocarbons. This term will be used throughout the study.

At the end of the devolatilization region, the coal is almost completely converted to char (Bunt, 2006:154). Below this, the char is gasified with steam and carbon dioxide ( $CO_2$ ) to form hydrogen, carbon monoxide, and methane ( $CH_4$ ) (Bunt, 2006:154; Higman & van der Burgt, 2008:95; Speight, 2020:137). Near the bottom, the remaining char combusts in oxygen to form ash and carbon dioxide (Higman & van der Burgt, 2008:95).

Equations 2.1 through 2.9 are the most significant reactions that occur in fixed-bed coal gasifiers (Aasberg-Petersen *et al.*, 2004:362; Bunt, 2006:12; Gräbner, 2015:108-109; Higman & van der Burgt, 2008:12-13; Speight, 2020:71,133,137).





where Equations 2.1 through 2.4 involve oxidation. Equations 2.5 and 2.6 are both reversible and endothermic, with the former the Boudouard and the latter the water gas reaction. Equations 2.7, 2.8, and 2.9 are, respectively, the reversible exothermic reactions for water gas shift, hydrogasification, and methanation.

### 2.2.3 Thermodynamic Reaction Models

The aims of modeling the coal gasification process include predicting the syngas' throughput and composition. Calculating the relative quantities of oxygen and steam required per unit of coal is also an aim, as well as producing process reference sets for control purposes. These models use calculations that incorporate thermodynamics, energy and mass balances, and the composition of the inlet streams (Higman & van der Burgt, 2008:16).

Gasifier designs begin from an application of specialized thermodynamics knowledge. Thermodynamics in general are used to calculate the future state of a system. When reaction rates in a gasifier are relatively high, thermodynamic equilibrium can be accurately assumed between carbon and the main gaseous components. In fixed-bed dry bottom coal gasifiers, although this is a valid assumption in the lower regions, it is not valid further up where devolatilization occurs (Higman & van der Burgt, 2008:11-12).

Gasification reactions are continually directed towards thermodynamic equilibrium. This allows thermodynamic data to be used to predict the composition of syngas and estimate the amounts of oxygen and steam required (Speight, 2020:71). However, thermodynamics alone cannot be used to predict the overall carbon conversion in gasifiers, as this calculation requires the inclusion of reaction kinetics and diffusion modeling (Aasberg-Petersen *et al.*, 2004:362).

Assuming thermodynamic equilibrium in reversible reactions allows the relative quantities of reactants and products to be calculated, with the additional assumption that these are ideal

gases. This calculation involves equating a reaction's equilibrium constant, at a given temperature, with a function of the relative quantities of reactants and products (Borgnakke & Sonntag, 2017:555,570-571; Higman & van der Burgt, 2008:13-15). The water gas shift reaction's equilibrium, at the gasifier outlet conditions, controls the quantities of carbon monoxide, carbon dioxide, water, and hydrogen in the syngas (Speight, 2020:45).

Calculating the quantities of reactants and products under non-equilibrium conditions requires the inclusion of reaction rates in the gasification model (Higman & van der Burgt, 2008:14). These are specified for the reactions that control the syngas' composition. In the fixed-bed coal gasifier, these are the water gas, Boudouard, and hydrogasification reactions (Higman & van der Burgt, 2008:36,95). The equations for these three reactions can be manipulated algebraically to produce the equations for the water gas shift and methanation reactions, which implies that these do not need to be included in the model (Higman & van der Burgt, 2008:19).

A balance is used to ensure that the energies entering and exiting the gasifier are equal. The former is the thermal and chemical energy in the inlet streams. The latter is the sum of the thermal and chemical energy in the outlet streams and the heat transferred from the gasifier (Higman & van der Burgt, 2008:19).

The mass balances that are most important for gasifier models are those for carbon, hydrogen, and oxygen (Higman & van der Burgt, 2008:19). Dalton's model for gas mixtures, which assumes that a mixture is comprised of ideal gases that do not interact, is also required in gasification models (Borgnakke & Sonntag, 2017:418-419; Higman & van der Burgt, 2008:19).

Reaction kinetics are used to define the path taken and time required for a system to reach its future state (Higman & van der Burgt, 2008:11). The available information on coal gasification kinetics is limited due to its complexity. This complexity stems from its dependence on coal's varying chemical properties and the mass transfer mechanisms of diffusion present during the gasification of solid coal particles (Higman & van der Burgt, 2008:33; Speight, 2020:72). The results from studies into this have also had only a limited impact on gasifier designs (Higman & van der Burgt, 2008:33).

The calculations completed with thermal-fluid gasifier models are used to determine the process' initial operating conditions. Once it has been operated and measurements have been recorded, the collected data are analyzed and used to direct the adjustments aimed at improving the process (Higman & van der Burgt, 2008:27). Furthermore, studies performed with models always need to be validated with data collected from gasifiers. This must include data regarding coal properties throughout the gasifier's bed (Conradie, 2016:65).

#### 2.2.4 Efficiency Measures

The efficiency measures applied to coal gasification are related to measures of feedstock conversion, syngas yield, energy, or exergy (He *et al.*, 2013:752). Two commonly used measures are cold gas efficiency and carbon conversion (Higman & van der Burgt, 2008:28). Cold gas efficiency provides the percentage of chemical energy from the coal that is available in the syngas (Aasberg-Petersen *et al.*, 2004:370; Higman & van der Burgt, 2008:28). Carbon conversion defines the percentage of carbon from the coal that is present in the syngas produced (Higman & van der Burgt, 2008:28).

Even though these are commonly used, both cold gas efficiency and carbon conversion provide limited information on the gasification process. These can also be misleading when the aim of gasification is syngas production. As such, it is more important to consider the quantities of carbon monoxide and hydrogen produced, with the aim of improving the  $H_2$  to CO ratio (Higman & van der Burgt, 2008:29-30).

#### 2.2.5 Process Faults

Coal gasifier faults have already been defined within the context of this study. Faults can occur due to the use of coal possessing properties that are not suitable for gasification. One example of this is the inconsistent gasifier performance that can result from variations in the coal's ash content (Aasberg-Petersen *et al.*, 2004:373). Another fault, which involves bridging of the feedstock, is known to cause uneven gas flow (Speight, 2020:83). As already noted, potential faults are identified by fluctuations not normally present in the process measurements of temperature and pressure. These fluctuations may be contained in either or both measurements.

The localized ash formation along a specific path through a gasifier's bed is a fault referred to as channel burning. This occurs if the flow velocity through that specific path is relatively larger than that through the other available paths (Keyser *et al.*, 2006:1444). It has been reported that channel burning is accompanied by both a raw syngas  $CO_2$  content in excess of 40% and increased carbon in the removed ash (Gräbner, 2015:260).

Channel burning and, larger than normal, pressure fluctuations can occur due to the use of coal with a particle size distribution near the maximum allowed in the gasifier (Bunt, 2006:129). This type of coal feed will be defined, in this study, as being composed of a coarse particle size distribution, also referred to as a heterogenous coal feed.

An excessive pressure drop will be observed in a gasifier if the bed does not have sufficient permeability and the flow of syngas is blocked. This occurs in fixed-bed gasifiers when the coal used has excessive fines and cakes easily (Aasberg-Petersen *et al.*, 2004:369; Higman & van der Burgt, 2008:91). Coal fines are particles usually between 0.15 and 1 mm in diameter, and caking defines the tendency of heated coal particles to fuse together (Galvin & Iveson, 2013:264; Honaker, 2013:302; van Dyk *et al.*, 2006:251). The excessive pressure drop can also be accompanied by channel burning, where both result in, a larger than normal, variability in the syngas' composition and exit temperature (Aasberg-Petersen *et al.*, 2004:369). Coal feeds containing excessive fines will also be defined, in this study, as being heterogenous.

Channel burning and the formation of hot spots have been observed in fixed-bed gasifiers due to a localized temperature increase above the grate. These resulted in inconsistent bed temperatures and can be caused by a non-uniform distribution of oxygen as it enters the gasifier (Bunt, 2006:163; Speight, 2020:83).

#### **2.2.6 Process Control**

In this subsection, additional information is provided regarding process control and fault prevention for fixed-bed dry bottom coal gasifiers. Relationships between process variables and gasifier efficiency are discussed, and variables that can be applied to estimate the temperature in a gasifier are introduced. The coal particle size distribution and its relationship to the pressure drop across a gasifier is also discussed.

This gasifier's exergy-based efficiency can be maximized by adjusting both the steam and oxygen to coal ratios. This was determined in a study performed using a thermal-fluid model of this coal gasifier, which was implemented in the Aspen Plus simulation software. It was observed that when either of the ratios was varied, the exergy efficiencies passed through a maximum. For a given ratio's varied magnitudes, each of the efficiencies reached a maximum at approximately the same ratio. When the oxygen to coal ratio was increased, the existence of a maximum was attributed to the fact that this initially resulted in more char combustion. The efficiencies decreased when this was outweighed by the conversion of carbon monoxide into additional carbon dioxide. This was due to the presence of excess oxygen. When the steam to coal ratio was increased, the maximum observed was linked to the initial promotion of the water gas and water gas shift reactions. Beyond a specific point, the efficiencies decreased with the reduced gasifier performance due to lower operating temperatures. These lower temperatures resulted from the excess steam (He *et al.*, 2013:754-756).

As it directly influences the syngas' composition, the temperature in the gasifier must be controlled. However, it is difficult to accurately measure the temperatures throughout a gasifier. This is mainly due to the environment in which these measurements need to be taken, which limits the type of measuring equipment that can be used. Accurately interpreting these measurements is also difficult due to a gasification phenomenon, which involves what is referred to as the chemical wet bulb temperature (Higman & van der Burgt, 2008:226-227).

There are some process variables that can be used to estimate the gasifier's temperature at specific points. One is the methane content of the syngas, which can be used to calculate an approximate outlet temperature for the gasifier. Another is the steam produced in the water jacket. This provides a measure of the heat flux through the gasifier's wall, which can in turn be used to estimate the temperature inside the gasifier (Higman & van der Burgt, 2008:27).

Steam controls the temperature in the gasifier by cooling the bottom region where it enters (Bunt, 2006:150; Higman & van der Burgt, 2008:17). For this reason, to prevent ash in the combustion region from melting due to a localized temperature increase above its fusion point, additional steam can be inlet to the gasifier (Bunt, 2006:147,154). Although steam is used to lower the temperature in a gasifier, this must be provided at a temperature that is high enough to prevent condensation. For this reason, the steam's temperature must be above the saturation point of water, at the pressure inside the gasifier (Higman & van der Burgt, 2008:18).

A laboratory scale fixed-bed gasifier was used to study how the particle size distribution changes during gasification (Conradie, 2016:4). The results showed that the width of the distribution increases as the coal is gasified, even when the particle size is strictly controlled (Conradie, 2016:59). The particle size distribution in turn affects the pressure drop across a gasifier. Using a computational fluid dynamics simulation, it was shown that wider particle size distributions, with the same average particle size, result in larger pressure drops (Keyser *et al.*, 2006:1442). The pressure drop in a fixed-bed dry bottom coal gasifier is also directly related to the production rate of syngas. This means that an increase in the mass flow rate of syngas produced will result in a larger pressure drop (Luckos & Bunt, 2011:921).

The use of a heterogenous coal feed has already been noted as causing channel burning and larger than normal, and excessively varying, pressure drops. Thus, a narrow particle size distribution is necessary for gasification, especially at higher syngas production rates. However, this does not preclude the possibility of channel burning, as the width of the distribution can increase during gasification.

### **2.2.7 Syngas Contaminants**

As already noted, the syngas that exits the gasifier contains contaminants that must be removed. These are not the focus of this study but are worth noting. These include tars, phenols, and pitches, which are referred to as volatiles in the context of this study to ensure consistency with the terminology already in use (Aasberg-Petersen *et al.*, 2004:361; Higman & van der Burgt, 2008:97). Also included are sulfur and chlorine compounds, solids, and ammonia (Higman & van der Burgt, 2008:217,220).

## **2.3 Statistical Modeling**

### **2.3.1 Gasification Process**

As already noted, statistical approaches are used in practice to model coal gasification. These utilize data, and the patterns present therein, without focusing on the thermal-fluid nature of the process. One of the most important applications of statistical modeling to coal gasification is the selection of a measurement reference set. This set defines the nearest to optimal operation of the fixed-bed gasifier and is required for process monitoring (Coetzer *et al.*, 2014:52; Higman & van der Burgt, 2008:16).

One procedure, based on principal component analysis, that can be followed to select this reference set was demonstrated for fixed-bed dry bottom coal gasifiers by Coetzer *et al.* (2014:61). Principal component analysis is a dimension reduction technique that determines the directions along which a set of measurements vary the most. These directions, referred to as principal components, are used in models generated from the data. This is applied when there are a large number of correlated variables and using a smaller set of latent variables is desirable. The assumption here is that this smaller set of principal components defines a large portion of the variability that is present in the original data (James *et al.*, 2014:374-375).

Prior to the utilization of thermal-fluid model's outputs, in the statistical models discussed in Section 1.4, principal component analysis can be applied. The reason for this is that dimension reduction can make patterns present in data appear more clearly. Here, the aim would be to clearly discriminate between normal and abnormal observations.

### **2.3.2 Fault Detection**

Statistical fault detection involves using binary classification to determine when a process has deviated from normal operations. It is necessary here to differentiate between this and fault

classification. Fault classification is more complicated and involves also predicting the type of fault observed (Huang *et al.*, 2020:9115,9121).

Principal component analysis, in addition to its application in the selection of reference data sets, is established as a fundamental multivariate statistical processing monitoring method. Fault detection has been accomplished through the integration of principal component analysis and techniques for the selection of relevant process variables. In the study that was performed, the selection of variables with causal relationships to faults was indicated as significantly contributing to the accuracy of fault detection during subsequent process monitoring (Jiang *et al.*, 2016:385).

The use of qualitative trend analysis, in combination with sliding window trend extraction, to detect potential faults in a coal gasifier has been demonstrated (Guo *et al.*, 2021:1). In the research that was performed, this data driven statistical method was used to determine the magnitude and direction of changes in the data, which are referred to as trends. Within specific time intervals, potential faults are detected whenever changes occur that are larger in magnitude than those typically observed (Guo *et al.*, 2021:6-7). This form of trend-based fault detection will be considered for application later in this study.

During a study into a new fault classification model, the relative accuracies of different statistical models were compared for the Tennessee Eastman process (Huang *et al.*, 2020:9115,9122). It was first observed that the proposed slow feature analysis and relevance vector machine model was more accurate than the other models investigated. In addition, it was observed that a support vector machine, an unmodified decision tree, and a modified decision tree with slow feature analysis all had approximately the same accuracy. These models consistently had accuracies between 55% and 70%. Furthermore, the accuracies of these were 13% to 23% below that of the model proposed in the study and 17% to 27% above that of the least accurate model. The least accurate model was an unmodified relevance vector machine (Huang *et al.*, 2020:9122).

Additional studies that utilized statistical techniques for process monitoring include Rossouw *et al.* (2020), Shang *et al.* (2015), and Guo *et al.* (2016). Rossouw *et al.* (2020:1-2,12) utilized principal component analysis, along with principal component biplots, to monitor and visualize the performance of coal gasifiers. Shang *et al.* (2015:3667,3680) and Guo *et al.* (2016:116,125) employed related statistical techniques, slow feature analysis and probabilistic slow feature analysis, respectively, in order to differentiate between normal and abnormal operating conditions.

As mentioned in Section 1.4, the conditions, under which the thermal-fluid model's outputs indicate abnormal operations, are still to be defined. Thus, the decision to select between fault detection and classification, for application in this study, cannot yet be made. However, it is clearly indicated that the selection of gasification process variables, according to a knowledge of the relevant fault conditions, is paramount for use in statistical modeling. The significance of principal component analysis, for use in this study, is also clearly indicated.

In addition, from the research presented in this section, support vector machines and decision trees will be considered for application later in this study. This is the case since, in the study by Huang *et al.* (2020:9122), the accuracies of support vector machines and decision trees, when used for fault classification, were demonstrated to be above 50%. Furthermore, in that study, the accuracies of these statistical models were not much lower than that of the best performing model, a relevance vector machine with slow feature analysis (Huang *et al.*, 2020:9122). As such, slow feature analysis, although a valid statistical technique for fault prediction, will not be utilized in this study. This is since the sole application of data driven techniques is not the focus of this study, and the integration of slow feature analysis with other statistical models is beyond the scope of this study.

## **2.4 Coal Analyses**

This section introduces some of the standard analyses that are applied to determine coal properties. The results of these analyses can be used to make predictions regarding the impact of specific coal properties on the gasification process (Gräbner, 2015:25). When analysis results are reported, different experimental bases can be specified. For this reason, the specific basis used must be taken into account when interpreting and applying an analysis' results (Gräbner, 2015:36).

The bases on which coal analysis results can be reported will be introduced first. Of all the available bases, as received (AR) is the closest to the coal's condition at a gasifier's inlet. This defines that coal is analyzed in the state that it is received in, where it contained volatile matter (VM), fixed carbon (FC), mineral matter ash (MMA), and close to its original moisture content (Gräbner, 2015:36).

Another basis is air dried (AD). This refers to the condition in which the moisture on the surface of the coal is removed before the analysis begins. AD should not be confused with water free (WF), which is the basis where all moisture is removed from coal prior to the analysis. The final basis that is worth noting here is water and ash free (WAF). This is where all moisture and MMA are removed before the coal is analyzed (Gräbner, 2015:36-37).

The first analysis introduced is used to provide a general perspective on coal quality. This is referred to as proximate analysis, and it defines that all the matter coal is comprised of can be divided into four basic groups. These groups are moisture, VM, FC, and MMA (Gräbner, 2015:44). The four groups are important to consider, since these are directly related to the four gasifier regions introduced in Section 2.2.2. In the two upper regions, moisture and VM are removed from the coal as this is converted to char. Thus, char is mainly comprised of FC and MMA. In the two lower regions, as already noted in Section 2.2.2, char is gasified and combusted to produce syngas and ash. This defines that FC is the quantity of carbon available in coal for gasification.

Another standard procedure that is often applied is ultimate analysis, which is used to determine the elemental composition of coal. The elements usually measured in this analysis are carbon (C), hydrogen (H), oxygen (O), sulfur (S), and nitrogen (N). Results from ultimate analysis are reported on a WAF basis (Gräbner, 2015:49,51-53).

To aid in predicting the volatiles released from coal, a test procedure known as the Fischer assay may be employed. This involves heating a coal sample and measuring the quantity of gas, tar, and water released (Gräbner, 2015:48-49). The temperature to which that sample is heated has been shown to affect the composition of volatiles released (Pretorius, 2016:85). As such, this temperature should correlate with process conditions in the gasifier, as the aim is to reproduce these experimentally.

If more data on the volatiles released is required, additional analyses can be completed on these. What is of significance to this study is the composition of gas released, which can be determined using chromatography. Here, gas chromatography is used to measure the quantity of  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  released during devolatilization (Pretorius, 2016:47). The elemental composition of the tar, which can be determined using ultimate analysis, is also significant (Pretorius, 2016:45,47).

#### **2.4.1 Proximate and Ultimate Analyses Data**

This subsection provides proximate and ultimate analyses data, as mass fractions in Tables 2.1 and 2.2, respectively, that will be used in this study. Mass fractions are dimensionless. The coal and ash data are from a study by Bunt (2006:i,iii), and the tar data are from a study by Pretorius (2016:65-66), where tar was produced at a temperature of 920°C. Based on data from proximate and ultimate analyses, it is highly likely that the coal source used by Bunt (2006,i,iii) was among those used by Pretorius (2016:30). For this study, data for coal, from this hypothesized singular source, will be used whenever possible.

**Table 2.1: Coal and Ash Proximate Analyses Data – Bunt 2006:i**

| <b>Matter</b>   | <b>Coal – AR</b> | <b>Coal – WF</b> | <b>Ash – AR</b> |
|-----------------|------------------|------------------|-----------------|
| <b>FC</b>       | 0.471            | 0.489            | 0.081           |
| <b>VM</b>       | 0.218            | 0.227            | 0.014           |
| <b>MMA</b>      | 0.275            | 0.285            | 0.906           |
| <b>Moisture</b> | 0.036            | 0                | 0               |

**Table 2.2: Coal and Tar Ultimate Analyses Data – Bunt 2006:iii; Pretorius 2016:66**

| <b>Element</b> | <b>Coal – WAF</b> | <b>Tar – WAF</b> |
|----------------|-------------------|------------------|
| <b>C</b>       | 0.795             | 0.773            |
| <b>H</b>       | 0.042             | 0.049            |
| <b>O</b>       | 0.127             | 0.144            |
| <b>S</b>       | 0.016             | 0.014            |
| <b>N</b>       | 0.021             | 0.020            |

#### **2.4.2 Fischer Assay Data**

This subsection provides Fischer assay data, as mass fractions in Table 2.3, that will be used in this study. The data, given at two different final temperatures, are from a study by Pretorius (2016:54). Additional data from this study, regarding the approximate composition of the gases in Table 2.3, are presented in Table 2.4 (Pretorius, 2016:63,118-119).

The decision needs to be made as to whether the data at 720°C or 920°C will be applied in this study. For this reason, the temperature in the devolatilization region of a fixed-bed dry bottom coal gasifier needs to be investigated. It has been shown that the average temperature in this region is between 461.6°C and 711.4°C. Furthermore, the surface temperature there is between 787.0°C and 1,003.7°C (Bunt, 2006:94). Given that 720°C is between the upper

limit of the average and the lower limit of the surface temperatures, the data corresponding to this temperature will be used throughout the study.

**Table 2.3: Fischer Assay Data – Pretorius 2016:54**

| <b>Volatile</b> | <b>720°C – AD</b> | <b>920°C – AD</b> |
|-----------------|-------------------|-------------------|
| <b>Char</b>     | 0.756             | 0.735             |
| <b>Tar</b>      | 0.031             | 0.031             |
| <b>Water</b>    | 0.110             | 0.112             |
| <b>Gas</b>      | 0.102             | 0.121             |

**Table 2.4: Gases Released during Devolatilization – Pretorius 2016:118-119**

| <b>Gas</b>                                       | <b>720°C – WAF</b> | <b>920°C – WAF</b> |
|--------------------------------------------------|--------------------|--------------------|
| <b><math>H_2</math> (<math>m_n^3/kg</math>)</b>  | 0.09               | 0.15               |
| <b><math>CO</math> (<math>m_n^3/kg</math>)</b>   | 0.04               | 0.06               |
| <b><math>CO_2</math> (<math>m_n^3/kg</math>)</b> | 0.05               | 0.05               |
| <b><math>CH_4</math> (<math>m_n^3/kg</math>)</b> | 0.07               | 0.07               |

## 2.5 Conclusions

The operation of fixed-bed dry bottom coal gasifiers has been introduced and discussed. This included information regarding how the gasifier is started, what size coal particles are supplied, and how ash is removed. An introduction was also provided to the process employed to remove contaminants from syngas. Gasifier regions, in which specific chemical reactions predominantly occur, were described. The most significant chemical reactions that occur in a gasifier were listed, after which thermodynamic modeling of the gasification process was discussed. Requirements for applying thermodynamic equilibrium to a chemical reaction were provided, and introductory details regarding the application of energy and mass balances were discussed.

Efficiency measures that are frequently applied to the coal gasification process were provided and discussed. The established process faults that can occur in fixed-bed dry bottom coal gasifiers were also introduced and described. In addition, information regarding process control for gasifiers was also provided. This included a discussion on how the oxygen to coal and steam to coal ratios, respectively, impact process efficiency. The effect that coal particle size distributions have on the process' performance was also noted, and additional details regarding syngas contaminants were provided.

Statistical modeling techniques for gasification and fault detection were introduced. This included a discussion on the application of principal component analysis to reduce the dimensions of a data set. Qualitative trend analysis, support vector machines, and decision trees were also preliminarily selected for consideration in this study. An introduction to coal analyses, as applicable to this study, was lastly provided. This included information regarding proximate and ultimate analyses, coal bases, and volatile matter analyses.

## **CHAPTER 3: THERMAL-FLUID MODEL THEORY**

### **3.1 Introduction**

In the previous chapter, literature pertaining to fixed-bed dry bottom coal gasifiers was provided and discussed. This included material regarding the operation, modeling, and faults of these gasifiers. Statistical fault prediction was also discussed, and results were presented for coal analyses that are pertinent to this study.

This chapter discusses the development of the thermal-fluid model that will be used to process fixed-bed dry bottom coal gasifier data. In Section 3.2, this model's aims are described, after which the high-level assumptions that must be made prior to the model's development are discussed. The software in which the model will be implemented is also noted in Section 3.2.

Section 3.3 introduces the equations that will be implemented in the model for three fundamental thermodynamic principles that are applicable to coal gasification. These are mole and energy balances, and chemical equilibrium for reversible reactions. The implementations of these three principles are discussed in detail in Sections 3.4, 3.5, and 3.6, respectively.

Section 3.4 defines how three separate balances are applied for the moles of carbon, hydrogen, and oxygen entering and exiting the gasifier. Section 3.5 defines how an energy balance is applied to the gasifier. Here, attention is provided to the problem of calculating the energy, both chemical and thermal, conveyed by the various streams entering and exiting the gasifier.

Section 3.6 discusses the implementation of a thermodynamic equilibrium condition for one of the reversible reactions that occur during gasification. Section 3.7 provides a summary of the thermal-fluid model's input and output variables, and Section 3.8 concludes the chapter.

### **3.2 Modeling Aim**

The thermal-fluid model, when solved, must output values that are applicable to the isolation of abnormal observations in gasifier data. Gasifier performance indicators include the mass flow rate and composition of syngas, where, as noted in Section 2.2.5, changes in these accompany faults. As such, the model must calculate the mass flow rate and composition of syngas, given available process measurements. The thermal-fluid model's inputs will include, amongst other variables, measurements of gasifier temperature and pressure, which are noted in Section 2.2.5 as being affected by process faults. Therefore, the use of flow rates calculated by the thermal-fluid model, from these process measurements, for the prediction of

gasifier faults is justifiable. This also allows the integrated thermal-fluid and statistical approach for fault prediction, which is the focus of this study, to be further developed. The gasifier process measurements available for use in the thermal-fluid model, along with symbols for these, are indicated in Table 3.1 (Bunt, 2006:51,54). Reference values for these measurements are provided in Section 4.2. It should be noted that the composition of syngas is measured after the contaminants, discussed in Sections 2.2.1 and 2.2.7, are removed in downstream gas processing units (Bunt, 2006:54, He *et al.*, 2013:752). Therefore, instead of utilizing the measured composition of syngas, which is only available after processing, the calculated composition at exit conditions will be employed for fault prediction in this study.

**Table 3.1: Available Gasifier Process Measurements – Bunt, 2006:51,54**

| Process Measurement                | Symbol              |
|------------------------------------|---------------------|
| Coal mass flow rate                | $\dot{m}_{coal}$    |
| Steam mass flow rate               | $\dot{m}_{H_2O,in}$ |
| Oxygen Normal volume flow rate     | $\dot{V}_{O_2,in}$  |
| Water jacket mass flow rate        | $\dot{m}_{wj}$      |
| Steam and oxygen inlet temperature | $T_{H_2O\&O_2}$     |
| Syngas outlet temperature          | $T_{sgs}$           |
| Syngas outlet pressure             | $P_{sgs}$           |

In order to implement mass and energy balances in the model, it must be assumed that gasification can be modeled as a steady-state process. Assuming a steady-state process simplifies these balances, as it implies that there is no storage of mass or energy in the gasifier (Borgnakke & Sonntag, 2017:138). This assumption is valid for gasifier data, given that measurements are recorded and averaged over a period of time (Borgnakke & Sonntag, 2017:138; Bunt, 2006:50,54). It is further assumed that syngas comprises  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ , and  $H_2O$  (Gräbner, 2015:130). This assumption requires that the model predicts mass flow rates for these five gases. As such, the model must be based on five equations in order to predict five flow rates per solution. These equations will be developed from the aforementioned three fundamental thermodynamic principles. Since the Engineering

Equation Solver (EES) includes a library of thermodynamic property functions, the model will be developed using this software (Nellis & Klein, 2009:1089).

### **3.3 Fundamental Thermodynamic Equations**

Mass balances, energy balances, and thermodynamic equilibrium were introduced in Section 2.2.3. It is therefore necessary to consider how five equations can be developed from these three fundamental thermodynamic principles. An application of the first law of thermodynamics results in a single equation defining that the total energy entering a steady-state system, or gasifier, must equal that exiting it (Borgnakke & Sonntag, 2017:67,70). Consequently, the model's first equation is formulated by implementing this law in the form of an energy balance. The remaining four equation formulations will follow from available mass balances and equilibrium conditions.

Here, it must be noted that mass and mole balances are different applications of the same principle, namely the law of mass conservation. This is due to the fact that providing the quantity of particles, measured in moles, is equivalent to reporting the mass of those particles (Borgnakke & Sonntag, 2017:8,132-133). Thus, the correct formulation of either is applicable in the model. The mass or mole balances ensure that the quantity of each element's atoms entering and exiting a steady-state system, or gasifier, is equal.

In this study, mole balances will be applied. These will be implemented for elemental carbon, hydrogen, and oxygen. In Section 2.2.3, these are noted as being the most important mass balances for gasifier models. Since these balances supply three additional equations, only one thermodynamic equilibrium condition, for a reversible chemical reaction, needs to be specified.

There are a total of five chemical reactions listed in Section 2.2.2 that are reversible. These are the water gas, Boudouard, hydrogasification, water gas shift, and methanation reactions. As discussed in Section 2.2.3, syngas composition is controlled by the water gas, Boudouard, and hydrogasification reactions, which can be manipulated to produce the water gas shift and methanation reactions. For this reason, selection of a reversible reaction for equilibrium modeling will be made from either the former three or the latter two reactions. To ensure that the model's assumptions remain realistic, this selection will be made between the water gas shift and methanation reactions. This is since, as stated in Section 2.2.3, reaction equilibrium calculations rely on an assumption that all the reactants and products are ideal gases. Thus, it is more realistic to apply this calculation to a reaction, such as these, where the reactants and products are gases.

Reaction equilibrium will be calculated at the syngas' outlet conditions. This is required since the syngas, comprising unknown flow rates of  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ , and  $H_2O$ , exits the gasifier at these outlet conditions. Of these five gases,  $H_2$ ,  $CO$ , and  $H_2O$  are present in both the water gas shift and methanation reactions. However,  $CO$  and  $CH_4$  are only present in the water gas shift and methanation reactions, respectively. Thus, only four of these gases will be used in the equilibrium calculation, regardless of which reaction is selected. This selection will need to be made once more information is available regarding the differences between these equilibrium calculations. This selection, made preliminarily in Section 3.6, is evaluated in Section 4.6.2. As indicated in Section 4.6.2, the simultaneous inclusion of both equilibrium conditions in the thermal-fluid model is unsuitable, and there is only one that can be implemented with relative accuracy.

### 3.4 Mole Balances

In this section, the model's mole balances are provided. Regarding these balances, the following should be noted. Firstly, terms for FC at the gasifier's inlet and outlet are provided. This is since, as stated in Section 2.4, FC defines the percentage of carbon in coal or ash that can be gasified. Secondly, none of the coal's VM, MMA, or moisture is accounted for in the balances. This results from two assumptions, where the first is that all the MMA, which enters a gasifier, exits without undergoing chemical reactions. The second assumption is that the VM and moisture, which enter a gasifier, exit with syngas. The flow rate of steam in the water jacket is also not accounted for in the mole balances. This is since the flow is separate and the same quantity of water that enters the jacket exits it.

Subscripts are applied in the balances to specify compounds and streams. The term compound is used to refer both to molecules, which include  $H_2$  and  $CO$ , and the groups of matter that coal is comprised of according to proximate analysis. In order to refer to specific gasifier inlet or outlet flows, the term stream is applied. The streams considered in the thermal-fluid model are as follows. The coal,  $H_2O$ , and  $O_2$  supplied to the gasifier are inlet streams, while the outlet streams include ash and respective total flow rates for the gases comprising syngas. In order to account for the fact that coal undergoes drying, devolatilization, gasification, and combustion in different regions along the gasifier's length, the syngas outlet stream is divided into three subsets. These stream subsets are referred to as gasification, devolatilization, and drying. The gasification stream includes the flow rates developed during combustion and gasification reactions, which are governed by the mole balances. The devolatilization stream is used to account for the gases released during the thermal decomposition of coal into char, according to the empirical data provided in Section 2.4.2. Lastly, the drying stream is employed to denote the quantity of moisture removed from coal

during drying, according to the empirical data provided in Section 2.4.1. As such, the flow rates of  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  released from coal during drying and devolatilization, are not included in the mole balances. However, as indicated in Section 3.5.6, these are summed with the respective flow rates developed during gasification for calculation of terms applied in the energy balance.

Mole balances are implemented for the elements of carbon, hydrogen, and oxygen in Equations 3.1, 3.2, and 3.3, respectively.

$$\dot{n}_{C,in} = \dot{n}_{C,out} + \dot{n}_{CO,gas} + \dot{n}_{CO_2,gas} + \dot{n}_{CH_4,gas} \quad [3.1]$$

$$2\dot{n}_{H_2O,in} = 2\dot{n}_{H_2O,gas} + 2\dot{n}_{H_2,gas} + 4\dot{n}_{CH_4,gas} \quad [3.2]$$

$$\dot{n}_{H_2O,in} + 2\dot{n}_{O_2,in} = \dot{n}_{H_2O,gas} + \dot{n}_{CO,gas} + 2\dot{n}_{CO_2,gas} \quad [3.3]$$

here the mole flow rates of compounds, entering or exiting the gasifier in various streams, are represented by  $\dot{n}$ . Subscripts utilized to denote the respective flow rate's compound and stream. Throughout this study, unless otherwise stated, mole flow rates are in kmol/s. As provided in an element's balance, the product of an integer and  $\dot{n}$  defines the mole flow rate of the respective element that is available in the specified compound present in the designated stream. The mole flow rates of FC inlet to and outlet from the gasifier, respectively, are represented by  $\dot{n}_{C,in}$  and  $\dot{n}_{C,out}$ . Flow rates for the  $H_2O$  and  $O_2$  inlet to the gasifier are  $\dot{n}_{H_2O,in}$  and  $\dot{n}_{O_2,in}$ , respectively. The predicted mole flow rates of  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  developed during combustion and gasification reactions are  $\dot{n}_{H_2O,gas}$ ,  $\dot{n}_{H_2,gas}$ ,  $\dot{n}_{CO,gas}$ ,  $\dot{n}_{CO_2,gas}$ , and  $\dot{n}_{CH_4,gas}$ , respectively.

### 3.4.1 Coal and Ash

This subsection discusses how the inlet and outlet FC mole flow rates are calculated, where these are present in coal and ash, respectively. This calculation is simpler for the former, since the flow rate of coal entering the gasifier is measured. However, an assumption must be made for the latter regarding the rate at which ash is removed by the gasifier's grate.

Gasifier inlet streams are specified in terms of mass and Normal volume flow rates (Bunt, 2006:54; Gräbner, 2015:259). Molar masses are required when converting between mass and mole flow rates (Borgnakke & Sonntag, 2017:417). Table 3.2 provides molar masses for five elements that, as mentioned in Section 2.4, are usually measured in an ultimate analysis (Walker *et al.*, 2014:A12-A14). Molar masses for  $H_2O$ ,  $O_2$ ,  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ , and sulfur dioxide ( $SO_2$ ), as available in EES, are provided in Table 3.3. Throughout this study, unless

stated otherwise, molar masses are provided in kg/kmol. The conversion between Normal volume and mass flow rates is discussed in Section 3.4.2.

**Table 3.2: Molar Masses – Walker *et al.*, 2014:A12-A14**

| Element  | Molar mass (kg/kmol) |
|----------|----------------------|
| <b>C</b> | 12.01115             |
| <b>H</b> | 1.00797              |
| <b>O</b> | 15.9994              |
| <b>S</b> | 32.064               |
| <b>N</b> | 14.0067              |

**Table 3.3: Molar Masses Extracted from EES**

| Element               | Molar mass (kg/kmol) |
|-----------------------|----------------------|
| <b>H<sub>2</sub>O</b> | 18.015               |
| <b>O<sub>2</sub></b>  | 31.999               |
| <b>H<sub>2</sub></b>  | 2.0159               |
| <b>CO</b>             | 28.001               |
| <b>CO<sub>2</sub></b> | 44.010               |
| <b>CH<sub>4</sub></b> | 16.043               |
| <b>SO<sub>2</sub></b> | 64.064               |

Equation 3.4 is used to calculate the mass flow rate of FC entering the gasifier, and Equation 3.5 is used to convert this to a mole flow rate.

$$\dot{m}_{C,in} = FC_{AR,coal} * \dot{m}_{coal} \quad [3.4]$$

$$\dot{n}_{C,in} = \dot{m}_{C,in} * M_C^{-1} \quad [3.5]$$

here  $\dot{m}_{C,in}$  is the mass flow rate of FC inlet to the gasifier. The mass fraction of FC in the coal on an AR basis is  $FC_{AR,coal}$ ,  $\dot{m}_{coal}$  is the mass flow rate of coal entering the gasifier, and  $M_C$  is the molar mass of carbon. Throughout this study, unless stated otherwise, mass flow rates are in kg/s.

The rotational frequency of the grate, measured in rev/h, determines the removal rate of ash from a gasifier. This is the case since the grate is designed to remove a specific volume of ash every revolution (Gräbner, 2015:253). It has been reported that at a rotational frequency of 3 rev/h, ash is removed from gasifiers at a mass flow rate of 9 t/h (Bunt, 2006:49). Based on this, it will be assumed that ash is removed at a rate of 3,000 kg/rev, on average.

Equations 3.6 and 3.7 are used to calculate the mass flow rates of ash and FC exiting the gasifier, respectively. In order to convert the mass flow rate of FC in ash to a mole flow rate, Equation 3.8 is applied.

$$\dot{m}_{ash} = Ash_{rm} * Grt_{frq} \quad [3.6]$$

$$\dot{m}_{C,out} = FC_{AR,ash} * \dot{m}_{ash} \quad [3.7]$$

$$\dot{n}_{C,out} = \dot{m}_{C,out} * M_C^{-1} \quad [3.8]$$

here  $\dot{m}_{ash}$  and  $Ash_{rm}$  are, respectively, the mass flow and grate removal rates of ash, where the latter is in kg/rev. The rotational frequency of the grate, in rev/s, is  $Grt_{frq}$ , and the mass flow rate of FC in ash is  $\dot{m}_{C,out}$ . On an AR basis, the mass fraction of FC in ash is  $FC_{AR,ash}$ .

### 3.4.2 Steam and Oxygen

This subsection describes how the mole flow rates of  $H_2O$  and  $O_2$  entering a gasifier are calculated. However, before these calculations are explained, the unit conversion of Normal volume to mass flow rates must be discussed. The reason for this is that, although the inlet flow rate of  $H_2O$  is reported on a mass basis, in t/h,  $O_2$  is reported on a Normal volume flow basis, in  $10^3 m_n^3/h$  (Bunt, 2006:50). Volume flow rates are also reported for gases exiting a gasifier (Gräbner, 2015:259). The densities of respective gases are required for this unit conversion, where these will be calculated at a condition defined as Normal (Gerhart *et al.*,

2017:163; Gräbner, 2015:259). The temperature and pressure of 15°C and 101.325 kPa, respectively, will be used for the condition defined as Normal throughout the study.

The mole flow rate of inlet  $H_2O$  is calculated with Equation 3.9, while the related Equation A.1, for calculating the inlet mole flow rate of  $O_2$ , is provided in Appendix A.2.

$$\dot{n}_{H_2O,in} = \dot{m}_{H_2O,in} * M_{H_2O}^{-1} \quad [3.9]$$

where the mass flow rate and molar mass of  $H_2O$  are represented by  $\dot{m}_{H_2O,in}$  and  $M_{H_2O}$ , respectively.

Equation 3.10 is applied to convert the Normal volume flow rate of  $O_2$  to a mass flow rate.

$$\dot{m}_{O_2,in} = \dot{V}_{O_2,in} * \rho_{O_2} \quad [3.10]$$

here  $\dot{m}_{O_2,in}$  and  $\dot{V}_{O_2,in}$  are, respectively, the mass and Normal volume flow rates of  $O_2$ . The Normal density of oxygen is  $\rho_{O_2}$ . Throughout the study, unless otherwise stated, volume flow rates are in  $m^3/s$ , and densities are in  $kg/m^3$ .

### 3.4.3 Raw Syngas

This subsection introduces a unit conversion that is necessary for syngas' flow rates, since mole and mass flow rates are required in the various equations of Section 3.5.6. This unit conversion is provided for  $H_2O$  in Equation 3.11. The related equations for converting between mole and mass flow rates for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  in the gasification stream are provided as Equations A.2 through A.5 in Appendix A.2.

$$\dot{m}_{H_2O,gas} = \dot{n}_{H_2O,gas} * M_{H_2O} \quad [3.11]$$

here the mass flow rate of  $H_2O$  in the gasification stream is  $\dot{m}_{H_2O,gas}$ .

## 3.5 Energy Balance

In this section, the model's energy balance, and equations for the variables present therein, are provided. The following should be noted regarding these variables. Firstly, the term enthalpy-flow is applied to define the relative thermal and chemical energy, per unit of time, conveyed by a given flow. In Section 2.2.3, the necessity of including thermal and chemical energy, from the streams entering and exiting a gasifier, in the energy balance was introduced. As such, enthalpy-flow variables for compounds in the gasifier's inlet and outlet streams are provided in the energy balance. Chemical energy is defined at a reference condition, and

thermal energy is calculated between this reference condition and the stream's respective temperature and pressure (Borgnakke & Sonntag, 2017:93,507-508). Secondly, variables for the heat absorption rates of gasifier flows that do not undergo chemical reactions are provided in the energy balance. Lastly, a variable is also included to account for heat loss to the environment.

The energy balance is implemented in Equations 3.12.

$$\begin{aligned} \dot{H}_{coal} + \dot{H}_{H_2O,in} + \dot{H}_{O_2,in} + \Delta\dot{Q}_{ash} + \Delta\dot{Q}_{wj} \\ = \dot{Q}_{loss} + \dot{H}_{H_2O,out} + \dot{H}_{H_2,out} + \dot{H}_{CO,out} + \dot{H}_{CO_2,out} + \dot{H}_{CH_4,out} + \dot{H}_{tar} \end{aligned} \quad [3.12]$$

here  $\dot{H}_{coal}$  is the enthalpy-flow provided to the gasifier by the inlet coal. Enthalpy-flow provided by the  $H_2O$  and  $O_2$  entering the gasifier are, respectively,  $\dot{H}_{H_2O,in}$  and  $\dot{H}_{O_2,in}$ . The heat absorption rate of ash and water jacket steam are  $\Delta\dot{Q}_{ash}$  and  $\Delta\dot{Q}_{wj}$ , respectively, and  $\dot{Q}_{loss}$  is the rate of heat loss to the environment. Enthalpy-flow provided by the  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  exiting the gasifier are  $\dot{H}_{H_2O,out}$ ,  $\dot{H}_{H_2,out}$ ,  $\dot{H}_{CO,out}$ ,  $\dot{H}_{CO_2,out}$ , and  $\dot{H}_{CH_4,out}$ , respectively. These account for gases formed during drying, devolatilization, and gasification. The enthalpy-flow provided in tar is  $\dot{H}_{tar}$ . Throughout this study, unless otherwise stated, enthalpy-flow, heat absorption rates, and the rate of heat loss are in kW.

### 3.5.1 Coal

This subsection describes how the enthalpy-flow of coal is calculated. Based on the proximate analysis results in Table 2.1, it is assumed for this calculation that ash is entirely comprised of MMA. This assumption allows energy available in the coal's FC, VM, and moisture to be calculated separately from the energy contained in ash. As such, the enthalpy-flow of coal accounts for chemical and thermal energy present in the FC, VM, and moisture entering a gasifier. Thus, for this enthalpy-flow calculation, the mass flow rate of ash is subtracted from that of coal. In addition, the calculations for this assumption require a hypothetical coal basis that will be referred to as ash free (AF).

Equation 3.13 is applied to calculate the enthalpy-flow of coal.

$$\dot{H}_{coal} = (\dot{m}_{coal} - \dot{m}_{ash}) * h_{coal} \quad [3.13]$$

here  $h_{coal}$  is the enthalpy of coal, and throughout this study, unless stated otherwise, enthalpies are in kJ/kg.

Enthalpy is a thermodynamic property applied during energy transfer calculations and is specifiable in a form that accounts for the relative thermal and chemical energy in a compound (Borgnakke & Sonntag, 2017:93,507-508). Coal enthalpy is calculated with Equation 3.14.

$$h_{coal} = h_{coal,frm} + \Delta h_{coal} \quad [3.14]$$

where  $h_{coal,frm}$  is the enthalpy of formation for coal. This defines coal's chemical energy and is calculated at reference conditions, which are 25°C and 100 kPa, throughout this study. The change in enthalpy for coal, between reference and gasifier inlet conditions, is  $\Delta h_{coal}$  and defines coal's thermal energy.

The enthalpy of formation for coal is calculated using Equations 3.15 through 3.18 (Gräbner, 2015:56,90).

$$h_{coal,frm} = (1 - Mst_{AF,coal}) * h_{FC\&VM,frm} + Mst_{AF,coal} * h_{liqH2O,frm} \quad [3.15]$$

$$h_{FC\&VM,frm} = h_{coal,com} * (1 - f_{com}) \quad [3.16]$$

$$h_{coal,com} = -32,763.3 * C_{WAF,coal} - 141,789.2 * H_{WAF,coal} - 9,276.8 * S_{WAF,coal} \quad [3.17]$$

$$f_{com} = 0.01379 * \ln(O_{WAF,coal}) - 0.01393 * O_{WAF,coal}^{0.75} + 1.02111 \quad [3.18]$$

where  $h_{FC\&VM,frm}$  and  $h_{liqH2O,frm}$  are the formation enthalpies of coal, on a WAF basis, and liquid water, respectively. The mass fraction of moisture in coal, on an AF basis, is  $Mst_{AF,coal}$ , for which a calculation is provided later in this section. Heat of combustion, in kJ/kg, for coal is  $h_{coal,com}$ , and  $f_{com}$  is a dimensionless combustion value based on the fraction of oxygen in coal. The mass fractions of carbon, hydrogen, sulfur, and oxygen present in coal, on a WAF basis, are  $C_{WAF,coal}$ ,  $H_{WAF,coal}$ ,  $S_{WAF,coal}$ , and  $O_{WAF,coal}$ , respectively.

Equation 3.19 is applied to calculate the change in enthalpy for coal.

$$\Delta h_{coal} = cp_{coal} * (T_{coal} - T_{frm}) \quad [3.19]$$

here  $cp_{coal}$  is the constant pressure specific heat of coal. The temperature of coal at the gasifier inlet is  $T_{coal}$ , and  $T_{frm}$  is the reference temperature that formation enthalpy is calculated at. Furthermore, unless stated otherwise, constant pressure specific heats are in kJ/kg-K and temperatures are in °C.

Equation 3.20 is used to calculate the constant pressure specific heat of coal (Gräbner, 2015:90).

$$cp_{coal} = (1 - Mst_{AF,coal}) * cp_{FC\&VM} + Mst_{AF,coal} * cp_{liqH2O} \quad [3.20]$$

here  $cp_{FC\&VM}$  and  $cp_{liqH2O}$  are the constant pressure specific heats of coal, on a WAF basis, and liquid water, respectively. A value of 4.187 kJ/kg-K will be used for the specific heat of liquid water in this model (Gräbner, 2015:91).

If  $T_{coal}$  is less than or equal to 400°C, Equation 3.21 is used to calculate the constant pressure specific heat of coal on a WAF basis. Otherwise, Equation 3.22 is applied (Gräbner, 2015:90).

$$cp_{FC\&VM} = 0.83 * (1 + 0.08 * VM_{WAF,coal}) * \left[ 1 + 0.2 * \frac{T_{coal}}{100} - 0.0033 \left( \frac{T_{coal}}{100} \right)^3 \right] \quad [3.21]$$

$$cp_{FC\&VM} = 0.83 * (1 + 0.08 * VM_{WAF,coal}) * (1 + 0.2 * 4 - 0.0033 * 4^3) + 0.00473 * (T_{coal} - 400)^{0.726} \quad [3.22]$$

where  $VM_{WAF,coal}$  is the mass fraction of volatile matter in coal, on a WAF basis. How this fraction is calculated is explained below.

Since, on any basis, the mass fractions in proximate analysis must sum to one, Equation 3.23 can be written for a WAF basis. Assuming that, on different bases, the ratio of FC to VM is equal, Equation 3.24 can be written. Manipulating Equation 3.24 and substituting this into Equation 3.23 allows the VM on a WAF basis to be calculated. A ratio for FC to VM is available for both AR and WF bases, where these differ slightly. As such, the average of the VM fractions on a WAF basis, calculated from each ratio, will be used in this model. This is shown in Equation 3.25.

$$FC_{WAF,coal} + VM_{WAF,coal} = 1 \quad [3.23]$$

$$\frac{FC_{AR,coal}}{VM_{AR,coal}} = \frac{FC_{WF,coal}}{VM_{WF,coal}} = \frac{FC_{WAF,coal}}{VM_{WAF,coal}} \quad [3.24]$$

$$VM_{WAF,coal} = \left[ \left( 1 + \frac{FC_{AR,coal}}{VM_{AR,coal}} \right)^{-1} + \left( 1 + \frac{FC_{WF,coal}}{VM_{WF,coal}} \right)^{-1} \right] / 2 \quad [3.25]$$

here subscripts are used to denote the bases for the various VM and FC mass fractions.

Using the same methodology as above, the mass fraction of moisture in coal on an AF basis can be calculated with Equation 3.29. This is derived by manipulating and substituting Equations 3.27 and 3.28 into 3.26.

$$FC_{AF,coal} + VM_{AF,coal} + Mst_{AF,coal} = 1 \quad [3.26]$$

$$\frac{FC_{AR,coal}}{Mst_{AR,coal}} = \frac{FC_{AF,coal}}{Mst_{AF,coal}} \quad [3.27]$$

$$\frac{VM_{AR,coal}}{Mst_{AR,coal}} = \frac{VM_{AF,coal}}{Mst_{AF,coal}} \quad [3.28]$$

$$Mst_{AF,coal} = \left( 1 + \frac{FC_{AR,coal}}{Mst_{AR,coal}} + \frac{VM_{AR,coal}}{Mst_{AR,coal}} \right)^{-1} \quad [3.29]$$

where subscripts are used to define the bases of the various moisture, FC, and VM mass fractions.

### 3.5.2 Steam and Oxygen

In this subsection, the calculation of enthalpy-flow for  $H_2O$  and  $O_2$  is discussed. Equations 3.30 and 3.31 are, respectively, applied to calculate enthalpy-flow and enthalpy for inlet  $H_2O$ . The related equations for calculating enthalpy-flow and enthalpy for inlet  $O_2$  are provided, in Appendix A.3, as Equations A.6 and A.7.

$$\dot{H}_{H_2O,in} = \dot{m}_{H_2O,in} * h_{H_2O,in} \quad [3.30]$$

$$h_{H_2O,in} = h_{H_2O,frm} + \Delta h_{H_2O,in}(T_{H_2O\&O_2}, P_{H_2O,in}, T_{frm}, P_{frm}) \quad [3.31]$$

here the enthalpy of  $H_2O$  at inlet conditions is  $h_{H_2O,in}$ . The enthalpy of formation for  $H_2O$  is  $h_{H_2O,frm}$ , and  $\Delta h_{H_2O,in}$  represents a function that calculates the difference in enthalpy, between reference and inlet conditions.

It should be noted that the enthalpy of formation introduced here for steam,  $h_{H_2O,frm}$ , is different from that specified in Equation 3.16 for liquid water,  $h_{liqH_2O,frm}$  (Borgnakke & Sonntag, 2017:634). The inlet temperature for  $H_2O$  and  $O_2$  is  $T_{H_2O\&O_2}$ , and  $P_{H_2O,in}$  represents the partial pressure of  $H_2O$ . The partial pressures of  $H_2O$  and  $O_2$  sum to the total pressure at the inlet,  $P_{H_2O\&O_2}$ , and the reference pressure for enthalpy of formation calculations is  $P_{frm}$ . Note, throughout this study, unless stated otherwise, pressures are in kPa.

Equations 3.32 through 3.34 are used to calculate the partial pressure of  $H_2O$  applied in Equation 3.31. The related equations for calculating the partial pressure of inlet  $O_2$  are provided, in Appendix A.3, as Equations A.8 and A.9. Equations for calculating partial

pressure are derived according to Dalton's model for gas mixtures, which was mentioned in Section 2.2.3 (Borgnakke & Sonntag, 2017:417,419).

$$P_{H_2O,in} = y_{H_2O,in} * P_{H_2O\&O_2} \quad [3.32]$$

$$y_{H_2O,in} = \frac{\dot{n}_{H_2O,in}}{\dot{n}_{H_2O\&O_2}} \quad [3.33]$$

$$\dot{n}_{H_2O\&O_2} = \dot{n}_{H_2O,in} + \dot{n}_{O_2,in} \quad [3.34]$$

where the mole fraction of  $H_2O$  in this mixture is represented by  $y_{H_2O,in}$ , and  $\dot{n}_{H_2O\&O_2}$  is the sum of the mole flow rates of  $H_2O$  and  $O_2$ .

### 3.5.3 Ash

This subsection describes how the heat absorption rate of ash is calculated. Since only the difference in thermal energy, between the inlet and outlet conditions of ash, needs to be accounted for, the term heat absorption rate is applied. This is due to the assumptions that ash does not undergo chemical reactions and is entirely comprised of MMA. Thus, the ash's mass flow rate and enthalpy of formation do not change.

To account for the change in thermal energy of the ash, Equations 3.35 and 3.36 are used.

$$\Delta\dot{Q}_{ash} = \dot{m}_{ash} * \Delta h_{ash} \quad [3.35]$$

$$\Delta h_{ash} = cp_{ash} * (T_{ash,in} - T_{ash,out}) \quad [3.36]$$

here  $\Delta h_{ash}$  is the change in enthalpy and  $cp_{ash}$  is the constant pressure specific heat of ash. The temperatures of ash at inlet and outlet conditions are, respectively,  $T_{ash,in}$  and  $T_{ash,out}$ .

Ash, of the coal that compositional data has been selected from, contains a greater quantity of aluminum oxide ( $Al_2O_3$ ) and silica ( $SiO_2$ ) than calcium oxide ( $CaO$ ) and ferric oxide ( $Fe_2O_3$ ) (Eisermann *et al.*, 1980:42; Pretorius, 2016:31). As such, constant pressure specific heat of ash is calculated using a correlation presented in Equation 3.37 (Gräbner, 2015:90).

$$cp_{ash} = 0.752 + 0.293 * \frac{T_{ash,avg} + 273.15}{1,000} \quad [3.37]$$

where  $T_{ash,avg}$  is the average of temperatures at the ash's inlet and outlet conditions.

### 3.5.4 Water Jacket

This subsection discusses the calculation of the heat absorption rate of steam in the water jacket. This is completed with Equation 3.38.

$$\Delta\dot{Q}_{wj} = \dot{m}_{wj} * (h_{wj,in} - h_{wj,out}) \quad [3.38]$$

here  $\dot{m}_{wj}$  is the mass flow rate of water in the jacket. The enthalpies of water at the jacket's inlet and outlet conditions are  $h_{wj,in}$  and  $h_{wj,out}$ , respectively, where inlet and outlet conditions are defined by temperature and pressure measurements. Since the chemical composition of the water does not change, it is not necessary here to account for enthalpy of formation.

### 3.5.5 Heat Loss

This subsection describes the calculation for the rate of heat loss. Heating values for coal are applied in this calculation, which are determined with empirical correlations based on the elemental composition of coal. These calculations are presented in Equations 3.39 through 3.41 (Gräbner, 2015:54-55,160).

$$\dot{Q}_{loss} = [(FC_{AR,coal} + VM_{AR,coal}) * \dot{m}_{coal}] * Ht_{loss} * LHV_{WAF} \quad [3.39]$$

$$LHV_{WAF} = HHV_{WAF} - 2,442 * (9 * H_{WAF,coal}) \quad [3.40]$$

$$\begin{aligned} HHV_{WAF} = & 35,200 * C_{WAF,coal} + 116,400 * H_{WAF,coal} - 11,100 * O_{WAF,coal} \\ & + 10,500 * S_{WAF,coal} + 6,290 * N_{WAF,coal} \end{aligned} \quad [3.41]$$

where  $Ht_{loss}$  is the fraction of heat lost from a gasifier. This fraction is reported to be between 2.5 and 6% of the lower heating value of coal for fixed-bed dry bottom gasifiers (Gräbner, 2015:160). In this study, the fraction of heat lost will be set when information regarding the model's accuracy is available. On a WAF basis, the lower and higher heating values of coal, in kJ/kg, are  $LHV_{WAF}$  and  $HHV_{WAF}$ , respectively. The mass fraction of nitrogen in coal, on a WAF basis, is  $N_{WAF,coal}$ .

### 3.5.6 Raw Syngas

This subsection discusses the calculation of enthalpy-flow for the gases comprising raw syngas. In Section 3.4, the necessity for differentiation between the three apparent outlet gas streams, drying, devolatilization, and gasification, was introduced. As such, in the equations presented, flow rates from these streams are summed together per gas, and the resulting total outlet flow rates are applied to calculate respective enthalpy-flow terms.

Equations 3.42 and 3.44 are applied to calculate the enthalpy-flow of outlet  $H_2$ . The enthalpy-flow of outlet  $H_2O$  is calculated by applying Equations 3.43 and 3.45. Related equations for calculating the enthalpy-flows of  $CO$ ,  $CO_2$ , and  $CH_4$  are provided in Appendix A.3 as Equations A.10 through A.15.

$$\dot{H}_{H_2,out} = (\dot{m}_{H_2,gas} + \dot{m}_{H_2,dvl}) * h_{H_2,out} \quad [3.42]$$

$$\dot{H}_{H_2O,out} = (\dot{m}_{H_2O,gas} + \dot{m}_{H_2O,dry}) * h_{H_2O,out} \quad [3.43]$$

$$h_{H_2,out} = h_{H_2,frm} + \Delta h_{H_2,out}(T_{sgs}, P_{H_2,out}, T_{frm}, P_{frm}) \quad [3.44]$$

$$h_{H_2O,out} = h_{H_2O,frm} + \Delta h_{H_2O,out}(T_{sgs}, P_{H_2O,out}, T_{frm}, P_{frm}) \quad [3.45]$$

where the enthalpy of  $H_2$  and  $H_2O$ , at outlet conditions, is  $h_{H_2,out}$  and  $h_{H_2O,out}$ , respectively. Enthalpy of formation for  $H_2$  is represented by  $h_{H_2,frm}$ . The differences in enthalpy, between outlet and reference conditions for  $H_2$  and  $H_2O$ , are calculated using functions represented by  $\Delta h_{H_2,out}$  and  $\Delta h_{H_2O,out}$ , respectively. The temperature of syngas at a gasifier's outlet is  $T_{sgs}$ , and  $P_{H_2,out}$  and  $P_{H_2O,out}$  are, respectively, partial pressures for  $H_2$  and  $H_2O$ . The mass flow rate of  $H_2$  released during coal devolatilization is  $\dot{m}_{H_2,dvl}$ , and the mass flow rate of  $H_2O$  released during coal drying is  $\dot{m}_{H_2O,dry}$ . Calculation of flow rates developed during devolatilization and drying is described in Section 3.5.7.1.

Equations 3.46 through 3.50 are used to calculate the partial pressures of  $H_2$  and  $H_2O$ . The related equations for calculating the partial pressures of  $CO$ ,  $CO_2$ , and  $CH_4$  are provided in Appendix A.3 as Equations A.16 through A.21.

$$P_{H_2,out} = y_{H_2,out} * P_{sgs} \quad [3.46]$$

$$P_{H_2O,out} = y_{H_2O,out} * P_{sgs} \quad [3.47]$$

$$y_{H_2,out} = \frac{\dot{n}_{H_2,gas} + \dot{n}_{H_2,dvl}}{\dot{n}_{sgs}} \quad [3.48]$$

$$y_{H_2O,out} = \frac{\dot{n}_{H_2O,gas} + \dot{n}_{H_2O,dry}}{\dot{n}_{sgs}} \quad [3.49]$$

$$\begin{aligned} \dot{n}_{sgs} = & (\dot{n}_{H_2,gas} + \dot{n}_{H_2,dvl}) + (\dot{n}_{CO,gas} + \dot{n}_{CO,dvl}) \\ & + (\dot{n}_{CO_2,gas} + \dot{n}_{CO_2,dvl}) + (\dot{n}_{CH_4,gas} + \dot{n}_{CH_4,dvl}) + (\dot{n}_{H_2O,gas} + \dot{n}_{H_2O,dry}) \end{aligned} \quad [3.50]$$

here  $P_{sgs}$  is the total pressure of syngas at a gasifier's outlet. Mole fractions of  $H_2$  and  $H_2O$  are represented by  $y_{H_2,out}$  and  $y_{H_2O,out}$ , respectively, and the sum of mole flow rates, for gases comprising syngas, is  $\dot{n}_{sgs}$ . The mole flow rates of  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  released during coal devolatilization are  $\dot{n}_{H_2,dvl}$ ,  $\dot{n}_{CO,dvl}$ ,  $\dot{n}_{CO_2,dvl}$ , and  $\dot{n}_{CH_4,dvl}$ , respectively, and  $\dot{n}_{H_2O,dry}$  is the mole flow rate of  $H_2O$  released during coal drying.

### 3.5.7 Drying and Devolatilization

#### 3.5.7.1 Gas

This subsection introduces flow rate calculations for gases released during the drying and devolatilization of coal. To determine the mass flow rate of  $H_2$  released during devolatilization, Equations 3.51 and 3.53 are employed. Related equations, for calculating the flow rates of  $CO$ ,  $CO_2$ , and  $CH_4$  released during devolatilization, are provided in Appendix A.3 as Equations A.22 through A.27. The mass flow rate of  $H_2O$  released during coal drying is calculated with Equation 3.53.

$$\dot{m}_{H_2,dvl} = \dot{V}_{H_2,dvl} * \rho_{H_2} \quad [3.51]$$

$$\dot{V}_{H_2,dvl} = H_{2WAF,coal} * [(FC_{AR,coal} + VM_{AR,coal}) * \dot{m}_{coal}] \quad [3.52]$$

$$\dot{m}_{H_2O,dry} = Mst_{AR,coal} * \dot{m}_{coal} \quad [3.53]$$

here  $H_{2WAF,coal}$  represents the Normal volume of  $H_2$ , per unit mass of coal, that is released during devolatilization, which is specified in  $m_n^3/kg$ , on a WAF basis. The Normal volume flow rate of  $H_2$  released during devolatilization is represented by  $\dot{V}_{H_2,dvl}$ , and  $\rho_{H_2}$  denotes the density of  $H_2$  at Normal conditions. The mass fractions of FC, VM, and moisture in coal, on an AR basis, are  $FC_{AR,coal}$ ,  $VM_{AR,coal}$ , and  $Mst_{AR,coal}$ , respectively.

The conversion between mass and mole flow rates, for gases released during devolatilization and drying, are provided as follows. Equation 3.54 is applied for the  $H_2$  released during devolatilization, Equation 3.55 is employed for the  $H_2O$  released during drying, and Equations A.28 through A.30 are provided, in Appendix A.3, for the  $CO$ ,  $CO_2$ , and  $CH_4$  released during devolatilization.

$$\dot{n}_{H_2,dvl} = \dot{m}_{H_2,dvl} * M_{H_2}^{-1} \quad [3.54]$$

$$\dot{n}_{H_2O,dry} = \dot{m}_{H_2O,dry} * M_{H_2O}^{-1} \quad [3.55]$$

### 3.5.7.2 Tar

This subsection discusses the calculation of enthalpy-flow for tar. It is assumed, for these calculations, that tar exits the gasifier at the same temperature as syngas. In addition, it is assumed that AD and AR coal bases are identical. This is since the quantity of surface moisture removed from coal during air drying is not known. As such, it is assumed that this is negligible compared to the total quantity of water in coal. Tar is also assumed to contain negligible quantities of ash and water, which defines that the mass flow rate of tar is specified on a WAF basis.

The enthalpy-flow of tar is calculated with Equations 3.56 through 3.59 (Eisermann *et al.*, 1980:41,43-44,48).

$$\dot{H}_{tar} = \dot{H}_{tar,frm} + \Delta\dot{H}_{tar} \quad [3.56]$$

$$\begin{aligned} \dot{H}_{tar,frm} = & -\dot{m}_{tar} * 30,890 + \dot{n}_{C,tar} * (h_{CO2,frm} * M_{CO2}) \\ & + \dot{n}_{S,tar} * (h_{SO2,frm} * M_{SO2}) + \frac{\dot{n}_{H,tar} * (h_{liqH2O,frm} * M_{H2O})}{2} \end{aligned} \quad [3.57]$$

$$\Delta\dot{H}_{tar} = \dot{m}_{tar} * \Delta h_{tar} \quad [3.58]$$

$$\Delta h_{tar} = 2.11 * 10^{-3} * (T_{tar}^2 - T_{frm}^2) \quad [3.59]$$

here  $\dot{H}_{tar,frm}$  is the tar enthalpy-flow calculated from its flow rate and enthalpy of formation. The difference in enthalpy, for tar between reference and outlet conditions, is  $\Delta h_{tar}$ , and the enthalpy-flow, due this difference and its flow rate, is  $\Delta\dot{H}_{tar}$ . The mass flow rate of tar is  $\dot{m}_{tar}$ , and  $\dot{n}_{C,tar}$ ,  $\dot{n}_{S,tar}$ , and  $\dot{n}_{H,tar}$  are, respectively, the mole flow rates of elemental carbon, sulfur, and hydrogen in tar. Enthalpies of formation for  $CO_2$  and  $SO_2$  are  $h_{CO2,frm}$  and  $h_{SO2,frm}$ , respectively. Molar masses for  $CO_2$  and  $SO_2$  are  $M_{CO2}$  and  $M_{SO2}$ , respectively. The temperature of the tar at the outlet of a gasifier,  $T_{tar}$ , and  $T_{frm}$  are specified here in K.

Equations 3.60 is used to calculate the mass flow rate of tar, and Equations 3.61 and 3.62 are employed to calculate the mole flow rate of elemental carbon in tar. Related equations for calculating the mole flow rates of elemental sulfur and hydrogen in tar are provided in Appendix A.3 as Equations A.31 through A.34.

$$\dot{m}_{tar} = Tar_{AD,coal} * \dot{m}_{coal} \quad [3.60]$$

$$\dot{m}_{C,tar} = C_{WAF,tar} * \dot{m}_{tar} \quad [3.61]$$

$$\dot{n}_{C,tar} = \dot{m}_{C,tar} * M_C^{-1} \quad [3.62]$$

here  $Tar_{AD,coal}$  is the mass fraction, on an AD basis, of tar released from coal during devolatilization. The elemental mass fraction of carbon in tar, on a WAF basis, is represented by  $C_{WAF,tar}$ , and the mass flow rate of elemental carbon in tar is  $\dot{m}_{C,tar}$ . The elemental molar mass of carbon is represented by  $M_C$ .

### 3.6 Chemical Equilibrium

In this section, an equation that defines chemical equilibrium for a reversible reaction, as is required for this model, will be introduced. As stated in Section 3.3, an equilibrium condition will be applied to either the water gas shift or the methanation reaction. In order to clarify the differences between these conditions, equations for both will be provided.

Equations 3.63 and 3.64 are used to define the chemical equilibrium condition for the water gas shift reaction (Borgnakke & Sonntag, 2017:554-555).

$$\ln(K_{wgs}) = -\frac{g_{CO2,out} + g_{H2,out} - g_{CO,out} - g_{H2O,out}}{R_{unv} * T_{sgs}} \quad [3.63]$$

$$K_{wgs} = \frac{y_{CO2,out} * y_{H2,out}}{y_{CO,out} * y_{H2O,out}} \quad [3.64]$$

Equations 3.65 and 3.66 are used to define the chemical equilibrium condition for the methanation reaction (Borgnakke & Sonntag, 2017:554-555).

$$\ln(K_{meth}) = -\frac{g_{CH4,out} + g_{H2O,out} - g_{CO,out} - 3 * g_{H2,out}}{R_{unv} * T_{sgs}} \quad [3.65]$$

$$K_{meth} = \frac{y_{CH4,out} * y_{H2O,out}}{y_{CO,out} * y_{H2,out}^3} * \left(\frac{P_{sgs}}{P_{frm}}\right)^{-2} \quad [3.66]$$

where  $K_{wgs}$  and  $K_{meth}$  are the dimensionless equilibrium constants for the water gas shift and methanation reactions, respectively. Gibbs functions, in kJ/kmol, for  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are  $g_{H2O,out}$ ,  $g_{H2,out}$ ,  $g_{CO,out}$ ,  $g_{CO2,out}$ , and  $g_{CH4,out}$ , respectively. The mole fractions of outlet  $CO$ ,  $CO_2$ , and  $CH_4$  are  $y_{CO,out}$ ,  $y_{CO2,out}$ , and  $y_{CH4,out}$ , respectively. The universal gas constant is  $R_{unv}$ , for which a value of 8.31451 kJ/kmol-K will be used in this model (Borgnakke & Sonntag, 2017:619). In the calculations for this section,  $T_{sgs}$  is specified in K.

Equation 3.67 is used to calculate the Gibbs function of  $H_2O$ , and the related equations for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are provided in Appendix A.4 as Equations A.35 through A.38 (Borgnakke & Sonntag, 2017:554).

$$g_{H_2O,out} = M_{H_2O} * [h_{H_2O,out,idl}(T_{sgs}) - T_{sgs} * s_{H_2O,out,idl}(T_{sgs}, P_{frm})] \quad [3.67]$$

here the enthalpies and entropies in this equation are determined using ideal gas properties. As such, the enthalpy function,  $h_{H_2O,out,idl}$ , is only provided with  $T_{sgs}$ , and the entropy function,  $s_{H_2O,out,idl}$ , is evaluated at  $T_{sgs}$  and  $P_{frm}$ . Throughout this study, unless otherwise stated, entropies are specified in kJ/kg-K, and as such,  $T_{sgs}$  must be specified in K for multiplication with entropy.

The equilibrium condition for the water gas shift reaction does not contain higher order exponents. Thus, this is simpler to apply in the model than the condition for methanation. For this reason, the equilibrium condition for the water gas shift reaction is preferred and will be pursued further. If this is proven unsuitable for use in the model, the equilibrium condition for methanation will be revisited, as stated in Section 3.3.

### 3.7 Summary of Thermal-Fluid Model Variables

This section presents a summary of the thermal-fluid model's input and output variables. This indicates whether the input variables are measured, assumed, or calculated from empirical data. The model's output variables, summations of respective flow rates developed during gasification, devolatilization, and drying, are also specified. Lastly, the variables for which initial values must be provided in EES, as described in Section 4.5, are indicated. This summary is presented in Table 3.4.

**Table 3.4: Summary of Thermal-Fluid Model Variables**

| Variable            | Measured input | Assumed input | Empirically calculated input | Output | EES initial value |
|---------------------|----------------|---------------|------------------------------|--------|-------------------|
| <b>Flow rates</b>   |                |               |                              |        |                   |
| $\dot{m}_{coal}$    | <b>x</b>       |               |                              |        |                   |
| $\dot{m}_{H_2O,in}$ | <b>x</b>       |               |                              |        |                   |

| Variable                                  | Measured input | Assumed input | Empirically calculated input | Output   | EES initial value |
|-------------------------------------------|----------------|---------------|------------------------------|----------|-------------------|
| $\dot{V}_{O_2,in}$                        | <b>x</b>       |               |                              |          |                   |
| $\dot{m}_{w,j}$                           | <b>x</b>       |               |                              |          |                   |
| $\dot{m}_{ash}$                           |                | <b>x</b>      |                              |          |                   |
| $\dot{m}_{tar}$                           |                |               | <b>x</b>                     |          |                   |
| $\dot{m}_{H_2O,dry}$                      |                |               | <b>x</b>                     |          |                   |
| $\dot{V}_{CO_2,dvl}$                      |                |               | <b>x</b>                     |          |                   |
| $\dot{V}_{CO,dvl}$                        |                |               | <b>x</b>                     |          |                   |
| $\dot{V}_{H_2,dvl}$                       |                |               | <b>x</b>                     |          |                   |
| $\dot{V}_{CH_4,dvl}$                      |                |               | <b>x</b>                     |          |                   |
| $\dot{m}_{CO_2,gas} + \dot{m}_{CO_2,dvl}$ |                |               |                              | <b>x</b> |                   |
| $\dot{m}_{CO,gas} + \dot{m}_{CO,dvl}$     |                |               |                              | <b>x</b> |                   |
| $\dot{m}_{H_2,gas} + \dot{m}_{H_2,dvl}$   |                |               |                              | <b>x</b> |                   |
| $\dot{m}_{CH_4,gas} + \dot{m}_{CH_4,dvl}$ |                |               |                              | <b>x</b> |                   |
| $\dot{m}_{H_2O,gas} + \dot{m}_{H_2O,dry}$ |                |               |                              | <b>x</b> |                   |
| <b>Temperatures</b>                       |                |               |                              |          |                   |
| $T_{H_2O\&O_2}$                           | <b>x</b>       |               |                              |          |                   |
| $T_{sgs}$                                 | <b>x</b>       |               |                              |          |                   |
| $T_{coal}$                                |                | <b>x</b>      |                              |          |                   |
| $T_{ash,in}$                              |                | <b>x</b>      |                              |          |                   |

| Variable              | Measured input | Assumed input | Empirically calculated input | Output | EES initial value |
|-----------------------|----------------|---------------|------------------------------|--------|-------------------|
| $T_{ash,out}$         |                | x             |                              |        |                   |
| <b>Pressures</b>      |                |               |                              |        |                   |
| $P_{H2O\&O2}$         |                | x             |                              |        |                   |
| $P_{sgs}$             | x              |               |                              |        |                   |
| <b>Enthalpies</b>     |                |               |                              |        |                   |
| $h_{wj,in}$           |                | x             |                              |        |                   |
| $h_{wj,out}$          |                | x             |                              |        |                   |
| $h_{CO2,out}$         |                |               |                              |        | x                 |
| $h_{CO,out}$          |                |               |                              |        | x                 |
| $h_{H2,out}$          |                |               |                              |        | x                 |
| $h_{CH4,out}$         |                |               |                              |        | x                 |
| $h_{H2O,out}$         |                |               |                              |        | x                 |
| <b>Mole fractions</b> |                |               |                              |        |                   |
| $y_{CO2,out}$         |                |               |                              |        | x                 |
| $y_{CO,out}$          |                |               |                              |        | x                 |
| $y_{H2,out}$          |                |               |                              |        | x                 |
| $y_{CH4,out}$         |                |               |                              |        | x                 |
| $y_{H2O,out}$         |                |               |                              |        | x                 |

### 3.8 Conclusions

In this chapter, the equation system that forms the fixed-bed dry bottom coal gasifier thermal-fluid model has been discussed and developed. This began with a discussion regarding the aim of the model, specifically the determination of the mass flow rates comprising syngas. This was defined within the study as the prediction of the mass flow rates of  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ , and  $H_2O$  exiting the gasifier. This led to the requirement that the model be based on five equations developed from fundamental thermodynamic principles.

The five equations were derived from the first law of thermodynamics, the law of mass conservation, and chemical equilibrium for a reversible reaction. The application of the first law of thermodynamics resulted in a single equation that equates the total energy entering a gasifier with that exiting it. The application of the law of mass conservation resulted in three equations. These, respectively, ensure that the moles of carbon, hydrogen, and oxygen that enter a gasifier exit it. The application of thermodynamic equilibrium to the water gas shift reaction provided the fifth equation.

The equations that are required to calculate the mole flow rates of carbon, hydrogen, and oxygen entering a gasifier were also discussed. This included equations for determining the quantity of FC in coal and ash. In addition, equations for unit conversions between mole, mass, and Normal volume flow rates were provided as required.

The equations that allow the calculation of the chemical and thermal energy available in the various streams entering and exiting a gasifier were also discussed in detail. This included equations for the enthalpy-flow of coal and the heat absorption rate of ash. In addition, equations for determining the enthalpy-flow of  $H_2O$  and  $O_2$  entering a gasifier were provided. Equations for determining the heat absorption rate of steam in the water jacket and the rate of heat loss were also discussed.

The equations for calculating enthalpy-flows of the  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ , and  $H_2O$ , which comprise raw syngas, were presented. These equations require the calculation of flow rates for gases developed during the drying and devolatilization of coal. As such, equations for this were provided and discussed. Equations for calculating the enthalpy-flow of tar were also supplied.

Equations were also provided for thermodynamic equilibrium applied to the reversible water gas shift and methanation reactions. From these chemical reactions, the equilibrium condition for the water gas shift reaction is preferred. Lastly, a summary of the thermal-fluid model's input and output variables was provided. The input variables were indicated for which process

measurements and empirical data are available, respectively. In addition, the input variables, for which assumptions must be made, were designated, and the model's output flow rates were indicated. Variables, which will be discussed in the following chapter, that are significant to the implementation of the thermal-fluid model in EES were also noted.

## CHAPTER 4: THERMAL-FLUID MODEL IMPLEMENTATION

### 4.1 Introduction

In the previous chapter, the five fundamental equations for the thermal-fluid model were developed and discussed. These are three mole balances, an energy balance, and chemical equilibrium for a reversible reaction. The equations that are applied to calculate the variables required by these fundamental equations were also provided and described.

In this chapter, the implementation of the thermal-fluid model will be discussed. Section 4.2 provides reference gasifier data for use in the model. Section 4.3 discusses the requirement that led to two versions of this model being developed, and Section 4.4 describes the operation of these models. As previously mentioned, the model will be developed in EES. A discussion is also provided regarding how the R Statistical Software (R) will be used in conjunction with EES. Versions 11.183 and 4.1.3 of EES and R, respectively, will be used in this study. R is included in preparation for the statistical modeling of Chapter 5 (R Core Team, 2022).

Section 4.5 describes the adjustment of the heat loss fraction. The process used to adjust the initial values of specific variables in EES, as required for the iterative numerical solution, is also discussed. This is necessary since, due to the complexity of the models, the numerical solutions will not converge to required tolerances, unless these variables are assigned accurate initial values. Section 4.6 describes the verification process, and the results thereof, from which the efficacy of the models must be judged. Section 4.7 concludes the chapter.

### 4.2 Inputs

Reference gasifier data, for use as thermal-fluid model inputs, are provided in this section. The input variables where assumptions must be made are also discussed in this section. These variables must be assumed, since while these are required in the thermal-fluid model, measurements for these are absent from the data. The reference data for a fixed-bed dry bottom coal gasifier is provided Table 4.1 (Bunt, 2006:51,54). Syngas compositional data, available in Table 4.1, will be required when tuning the model's heat loss parameter.

As is evident from Table 4.1, measurements of the steam and oxygen inlet pressure ( $P_{H_2O\&O_2}$ ), coal and ash temperatures ( $T_{coal}$ ,  $T_{ash,in}$ , and  $T_{ash,out}$ ), water jacket temperatures and pressures (used to calculate  $h_{wj,in}$  and  $h_{wj,out}$ ), and grate frequency ( $Gr t_{frq}$ ) are not available. Assumptions are made for these variables, from which representative values are provided. However, these do not conclusively capture the respective details of the gasification process.

As such, a sensitivity analysis could be performed to determine the contributions of these assumptions to model accuracy, but this is outside of the study's scope.

**Table 4.1: Fixed-Bed Dry Bottom Coal Gasifier Data from Literature – Bunt, 2006:51,54**

| Variable                                          | Symbol              | Measurement |
|---------------------------------------------------|---------------------|-------------|
| Coal mass flow rate [t/h]                         | $\dot{m}_{coal}$    | 50.0        |
| Steam mass flow rate [t/h]                        | $\dot{m}_{H_2O,in}$ | 52.5        |
| Oxygen Normal volume flow rate [ $10^3 m_n^3/h$ ] | $\dot{V}_{O_2,in}$  | 9.20        |
| Water jacket mass flow rate [t/h]                 | $\dot{m}_{wj}$      | 8.30        |
| Steam and oxygen inlet temperature [°C]           | $T_{H_2O\&O_2}$     | 344         |
| Syngas outlet temperature [°C]                    | $T_{sgs}$           | 550         |
| Syngas outlet pressure [kPa]                      | $P_{sgs}$           | 2,850       |
| <b>Syngas volume percentage</b>                   |                     |             |
| $H_2$ [%]                                         | $Ref_{H_2}$         | 39.7        |
| $CO$ [%]                                          | $Ref_{CO}$          | 23.0        |
| $CO_2$ [%]                                        | $Ref_{CO_2}$        | 28.3        |
| $CH_4$ [%]                                        | $Ref_{CH_4}$        | 8.80        |

A value of 3,500 kPa is assumed for the steam and oxygen inlet pressure ( $P_{H_2O\&O_2}$ ). Since this pressure has been applied in fixed-bed dry bottom coal gasifier models, the assumption is reasonable (He *et al.*, 2013:751). In addition, this value will be assumed for the pressure at the water jacket inlet and outlet. This requires an additional assumption, namely that there is a negligible pressure drop between the inlet and outlet of the water jacket. Given that there is limited information regarding the design of the water jacket, and these pressures are not available, such an assumption is necessary.

The water jacket inlet temperature is assumed to be 25°C. Since this temperature has been applied to this variable in fixed-bed dry bottom coal gasifier models, the assumption is acceptable (He *et al.*, 2013:751). The saturation temperature of steam at a pressure of 3,500 kPa is approximately 242°C (Borgnakke & Sonntag, 2017:649). As such, a value of 240°C is assumed for the water jacket outlet temperature. The assumption here is that the jacket heats liquid water to a temperature that is close to the respective boiling point. In Section 2.2.1, it was stated that the high pressure in the jacket limits the saturated steam quality. This assumption then, defining that the steam quality is near unity, does not completely align with this statement from literature. However, given that the measurements are not available, these assumptions are necessary when calculating the water jacket's enthalpies ( $h_{wj,in}$  and  $h_{wj,out}$ ).

A value of 190°C is assumed for the coal and ash inlet temperatures ( $T_{coal}$  and  $T_{ash,in}$ ). This is approximately the mean between the average temperature of the coal inside the gasifier, near the inlet, and 25°C. The former is reported to be 354°C (Bunt, 2006:vii). The latter temperature has been applied to this variable in fixed-bed dry bottom coal gasifier simulations (He *et al.*, 2013:751). As stated in Section 2.2.1, coal is supplied from a lock-hopper that is pressurized with syngas. It can thus be assumed that the temperature of the inlet coal, comprising FC, VM, moisture, and MMA, will be higher than 25°C but lower than the temperature it reaches upon entry into the gasifier.

The ash outlet temperature is assumed to be 350°C ( $T_{ash,out}$ ). This is reported to be the temperature at which ash is expected to exit a fixed-bed dry bottom coal gasifier (Bunt, 2006:145). The grate frequency ( $Grt_{frq}$ ) is assumed to be 4.5 rev/h. A value of 3 rev/h is reported to be the minimum frequency that the grate can be operated at (Bunt, 2006:49). Given then that a measurement for this is not available, a value 50% larger than the minimum is assumed.

### 4.3 EES Models A and B

In this section, an introduction is provided to the development of two versions of the thermal-fluid model. The first version, referred to as Model A, applies assumed positive values for the mole fractions of syngas, when calculating partial pressures with Equations 3.46, 3.47, and A.16 through A.18. Whereas, the second version, referred to as Model B, applies calculated values for these mole fractions. The assumed mole fractions, and the method by which these were arrived at, are discussed in Section 4.5.

The assumption of positive values for the mole fractions defines that the corresponding partial pressures are also positive values. These partial pressures are used in the respective

enthalpy functions of Equations 3.44, 3.45, and A.13 through A.15. The corresponding enthalpy functions in EES must be supplied with positive values for pressure, or the software will not generate a solution. Thus, this assumption allows solutions to be generated, for Model A, at observations where one or more negative values are obtained for mole fractions. In this study, the term observation is used to define a single complete set of input variables for the thermal-fluid model. Since Model B calculates partial pressures using the actual mole fractions, it is possible for one or more enthalpy functions to receive negative pressures. When this occurs, no viable solution exists for the input parameters.

Mole fractions cannot be negative in reality, referring to physical real-life situations, although negative quantities of mass are possible mathematically. As such, Model A is fundamentally more mathematical than Model B. This is since Model A allows negative mole fractions to be predicted within the confines of its system of equations, while Model B does not.

The enthalpy of a gas can be assumed to be a function of temperature only, under a specific ideal condition. This condition is high temperature accompanied by low pressure (Borgnakke & Sonntag, 2017:96-97). It cannot be claimed that the environment of a gasifier meets this condition entirely, given the gasifier's high internal pressure. For this reason, pressure is still accounted for in the enthalpy functions; it is simply calculated using the assumed values for mole fractions. The assumption is that, when calculating enthalpies at gasifier outlet conditions, approximate partial pressures can be utilized without significant loss of accuracy. In Section 4.6.1.4, this assumption will be evaluated by comparing the syngas volume percentages, calculated with Models A and B, and determining whether or not these correspond. As will be explained in the following section, Model A is faster to execute. What is significant regarding Model B, as will be explained in Chapter 5, is the knowledge of where this model cannot be solved.

#### **4.4 Operation in EES and R**

In this section, the operation of the completed models will be described. As previously stated, Models A and B have been developed in EES to allow the use of available thermodynamic property functions. Since data processing and statistical functions will be required in Chapter 5, software with access to these must be used in conjunction with the EES models. The decision has been made to use R, given that there are extensive statistical functions available for this software (James *et al.*, 2014:vii,8). For this reason, a program has been written in R that can transfer data to and from the EES models. The process is explained visually in Figures 4.1 through 4.4.

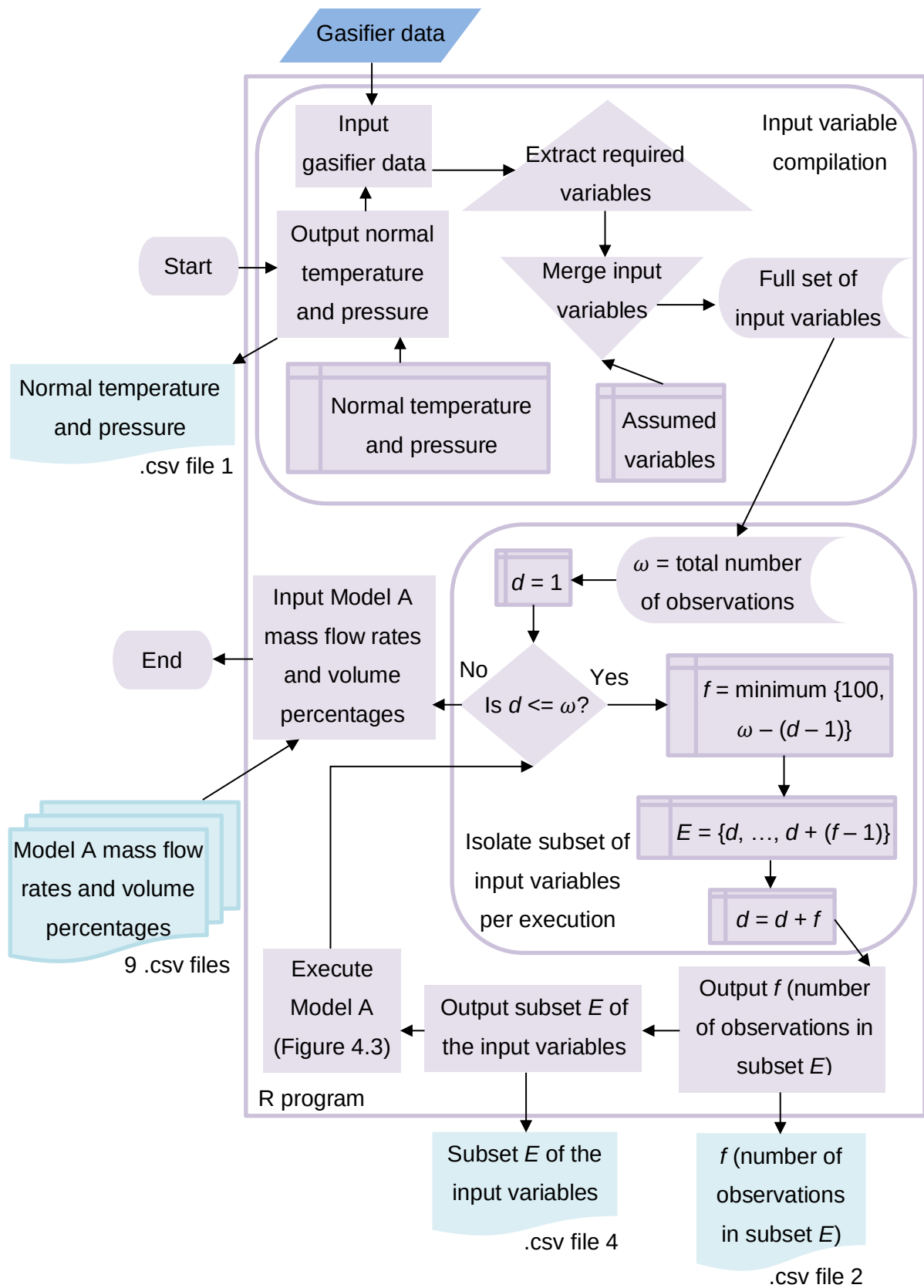


**Figure 4.1: Execution of Model A from R Program**

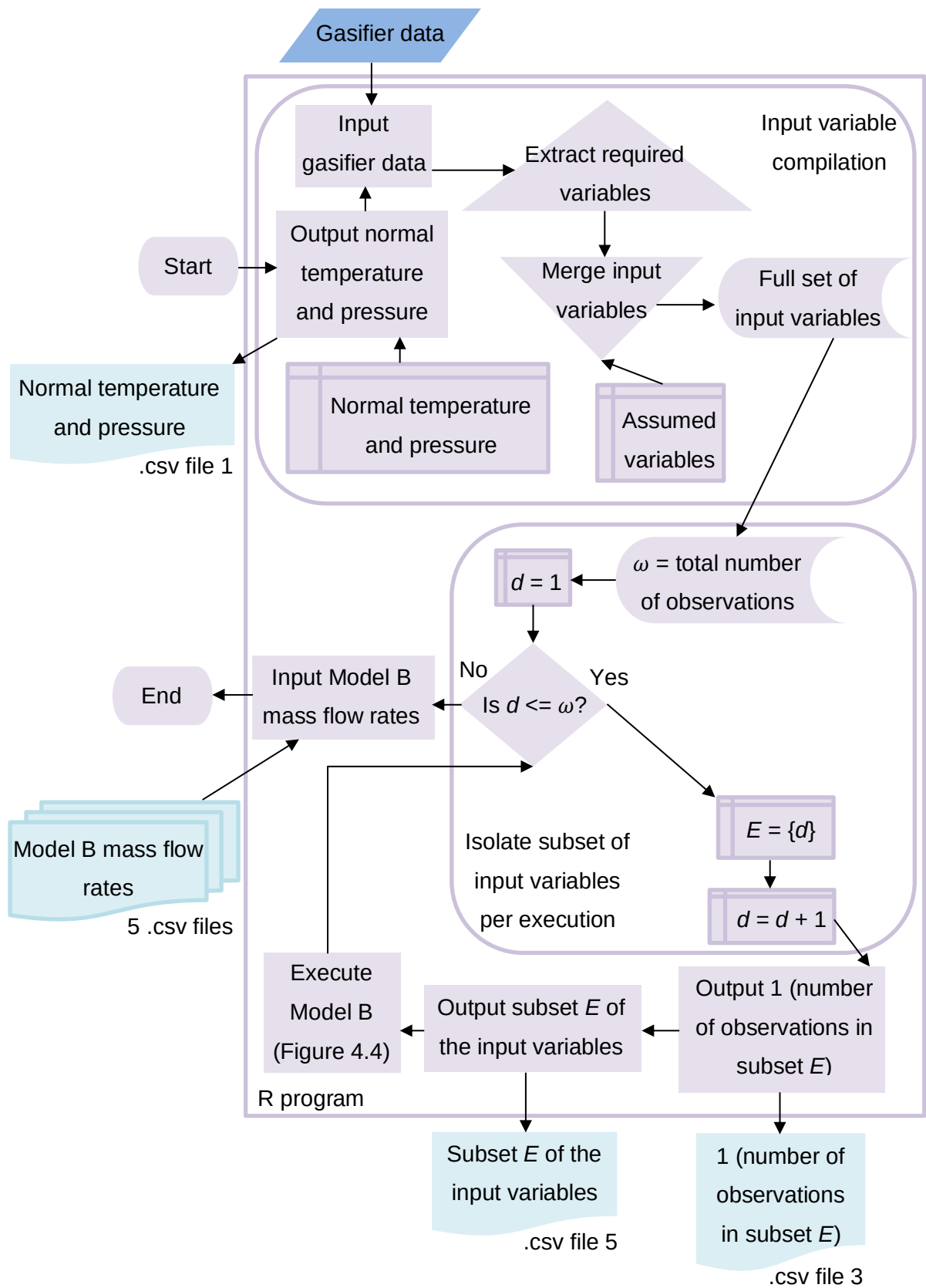


**Figure 4.2: Execution of Model B from R Program**

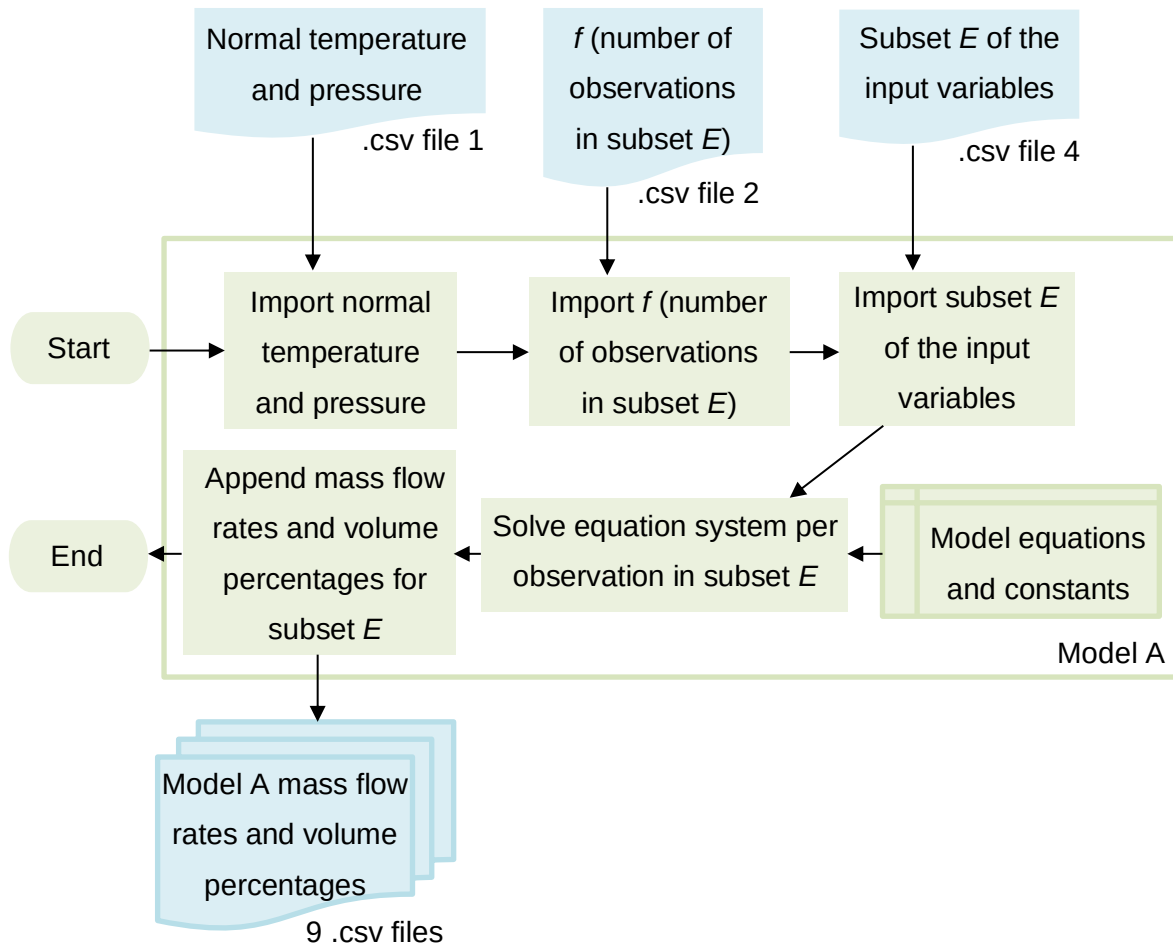


**Figure 4.3: Operation of Model A in EES**

The EES models and R program are operated as follows. Gasifier data are input from a .csv file into the R program, and the input variables for the model are compiled. This includes the addition of variables that are absent from the data, for which assumptions were made in Section 4.2. The compiled data is output to five .csv files as required.

The first .csv file contains Normal temperature and pressure, which are required when converting between volume and mass flow rates. The second and third files each contain a number, which defines the number of observations that will be input to Models A and B, respectively. The fourth and fifth files, respectively, contain the observations for Models A and B, for which solutions must be generated. The first file needs only be output once. However, the second and fourth files will need to be output each time Model A is executed. Correspondingly, the third and fifth files will need to be output each time Model B is executed.

The required EES model is executed directly from R. For Model A, up to 100 observations can be sent to EES per execution. This requires that the total number of observations be

divided into subsets of up to 100, and the model be executed once per subset. This is to accommodate for the maximum available memory in EES, which must store most of the variables required per observation. For Model B, only 1 observation can be sent to EES per execution. This is the case since solutions will not be generated if, for even 1 observation in a subset, negative values are obtained for mole fractions. The result of this difference is that Model A is significantly faster to execute. The set of operations for Models A and B are essentially identical. However, the number of observations used per execution and the output variables are different.

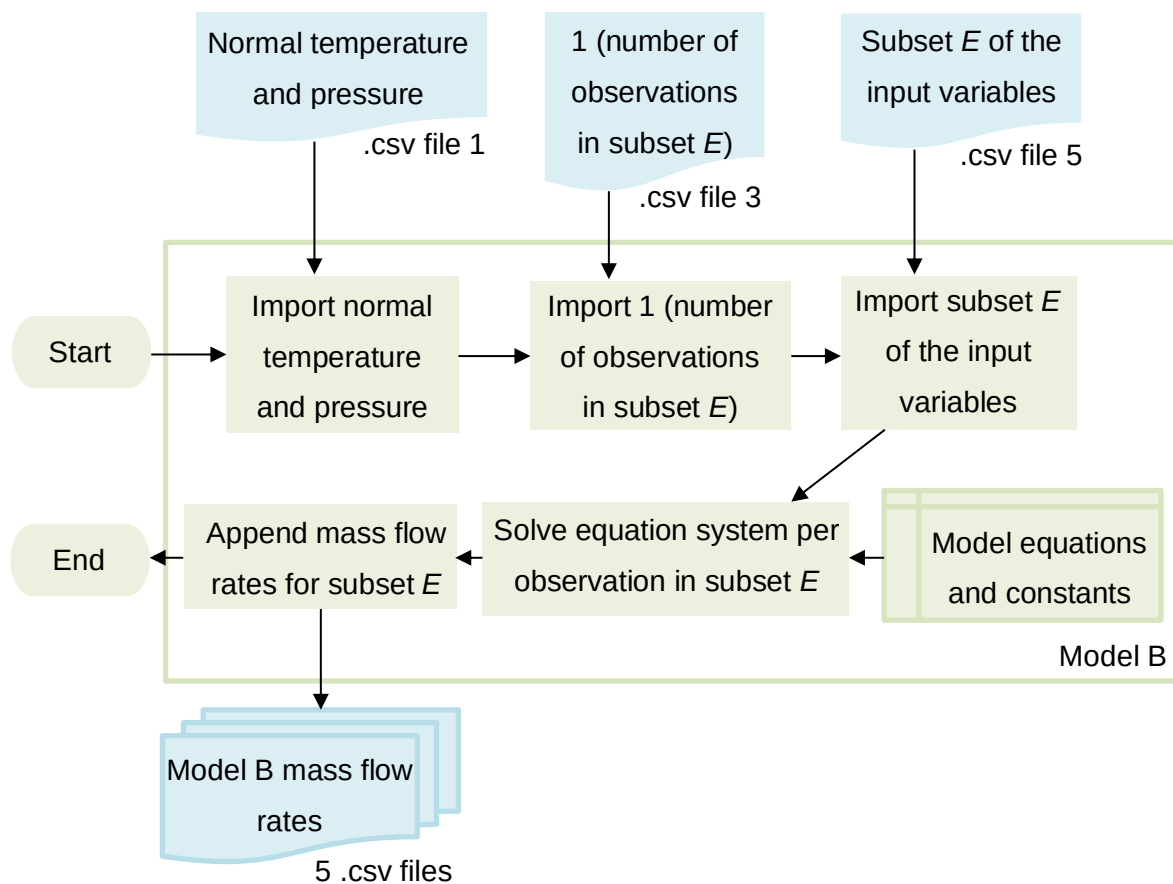


**Figure 4.4: Operation of Model B in EES**

EES opens automatically when executed from R, and the three respective .csv files are input. The model is solved, and the mass flow rates for  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$  and  $H_2O$  are, respectively, output to five .csv files. However, for Model A, the volume percentages for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are also output to four .csv files. EES closes automatically after the respective values have been output. As indicated by Figures 4.1 and 4.2, the EES models are repeatedly executed until all the observations have been processed. After this, the output variables are input into R from the respective .csv files.

## 4.5 Solvability and Convergence

In this section, the process followed to adjust the fraction of heat lost is described. As will be discussed, this is required in order to account for uncertainty in the model. The process followed to develop the EES models, so that numerical solutions for these could be generated, is also discussed.

Due to the difficulty in accounting for trace gases that may be present in raw syngas, uncertainty exists within the thermal-fluid model developed. These gases include hydrogen sulfide ( $H_2S$ ), ammonia ( $NH_3$ ), carbonyl sulfide ( $COS$ ), hydrogen cyanide ( $HCN$ ), and nitrogen ( $N_2$ ) (Gräbner, 2015:49,51; He *et al.*, 2013:752). In order to account for the VM released from coal, energy terms for the tar and  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  released during devolatilization are included in the thermal-fluid model. This leads to additional uncertainty in the thermal-fluid model, since the hydrocarbons released during devolatilization are not accounted for individually. In addition to tar, these hydrocarbons include phenol and naphtha (Aasberg-Peterson *et al.*, 2004:361; He *et al.*, 2013:752; Higman & van der Burgt, 2008:97).

To correct for these uncertainties, the heat loss fraction will be adjusted. Since there are gases and volatiles that are not included in the model, the enthalpy-flows of these are not accounted for. As such, this fraction will be adjusted to account for the total enthalpy-flow lost in these syngas contaminants. However, before this can be adjusted, it must be possible to generate solutions in EES for the thermal-fluid model.

In EES, before the software can converge to a solution, specific variables must be supplied with accurate initial values. This is due to the complexity of the non-linear thermal-fluid model. For Model A, this is partly accomplished with the assumed positive mole fractions for syngas that are applied in Equations 3.46 and 3.47. However, these mole fractions are also stored in EES as initial values for the respective variables. In EES, these are applied in a numerical solution method as initial values for respective variables.

In Model B, as discussed in Section 4.3, the assumed mole fractions are not applied in Equations 3.46 and 3.47. However, these mole fractions are stored in EES as initial values. In addition, initial values for the enthalpies of the gases that constitute raw syngas must be stored in EES.

In order to determine these initial values, the following methodology is applied. In Model A, an initial value of 0.2 is entered into Equations 3.46 and 3.47, as these appear in EES, for the mole fractions. This is also stored in EES as initial values for these variables, where these

must be stored per respective array. The reason for this is that these values are required to generate a solution at each observation, and the variables in EES are stored in arrays.

For heat loss fraction adjustment, an initial value of 5% will be applied in Model A, with the mole fractions set to 0.2. The fraction will be adjusted to minimize errors when solving for the data in Section 4.2. These errors will be calculated between the predicted and reference syngas volume percentages.

By applying Model A to the data in Section 4.2, the effect that the heat loss fraction has on errors could be determined. The results of this are visually presented in Figure 4.5. In this figure, the fraction varies from 5% to 25%. The sum of square errors is calculated, for the reference syngas volume percentages in Table 4.1, by applying Equation 4.1.

$$SSE = (V_{H_2} - Ref_{H_2})^2 + (V_{CO} - Ref_{CO})^2 + (V_{CO_2} - Ref_{CO_2})^2 + (V_{CH_4} - Ref_{CH_4})^2 \quad [4.1]$$

here  $SSE$  is the sum of square errors. The predicted volume percentages for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are represented by  $V_{H_2}$ ,  $V_{CO}$ ,  $V_{CO_2}$ , and  $V_{CH_4}$ , respectively. Reference volume percentages for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are, respectively,  $Ref_{H_2}$ ,  $Ref_{CO}$ ,  $Ref_{CO_2}$ , and  $Ref_{CH_4}$ .

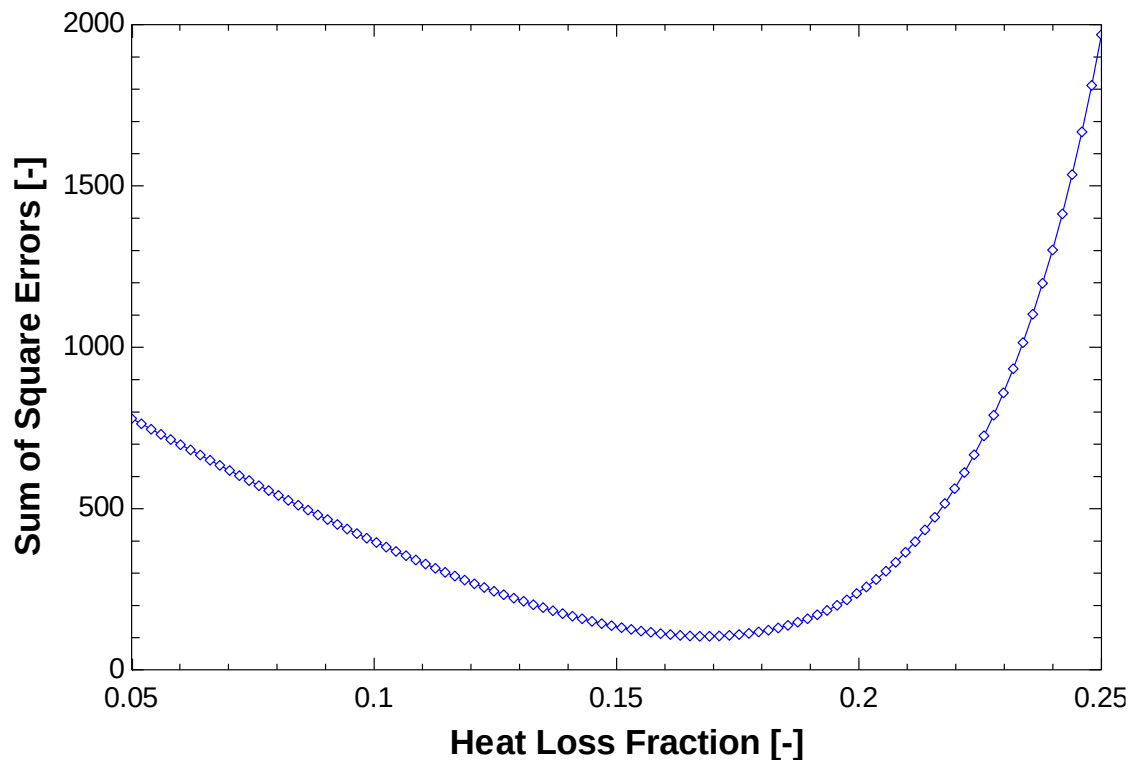


Figure 4.5: Sum of Square Errors vs Heat Loss Fraction

From Figure 4.5, it is evident that the heat loss fraction has a significant effect on the model's accuracy. In addition, it is clear that the minimum error falls between the fractions of 15% and 20%. As such, a minimization problem was setup and solved in EES. This determined that a 16.8% heat loss fraction results in the smallest sum of square errors. To further investigate the errors for the predicted syngas volume percentages, the values in Table 4.2 were calculated in EES. In this table, per heat loss fraction, relative errors for the individual syngas volume percentages are provided along with the respective sum of square errors. A relative error for the predicted  $H_2$  syngas volume percentage is calculated by applying Equation 4.2, and related equations for  $CO$ ,  $CO_2$ , and  $CH_4$  are provided, in Appendix A.7, as Equations A.53 through A.55.

$$RE_{H_2} = \frac{|V_{H_2} - Ref_{H_2}|}{Ref_{H_2}} * 100 \quad [4.2]$$

where  $RE_{H_2}$  represents the relative error for the predicted  $H_2$  syngas volume percentage.

**Table 4.2: Errors per Heat Loss Fraction**

| Heat loss fraction | Syngas volume percentage relative errors [%] |      |        |        | Sum of square errors [-] |
|--------------------|----------------------------------------------|------|--------|--------|--------------------------|
|                    | $H_2$                                        | $CO$ | $CO_2$ | $CH_4$ |                          |
| <b>15%</b>         | 23.5                                         | 25.0 | 1.25   | 42.5   | 134                      |
| <b>16%</b>         | 18.9                                         | 29.7 | 6.22   | 25.4   | 111                      |
| <b>17%</b>         | 13.8                                         | 34.6 | 11.5   | 6.88   | 104                      |
| <b>18%</b>         | 8.23                                         | 39.7 | 17.2   | 13.5   | 119                      |
| <b>19%</b>         | 1.97                                         | 45.1 | 23.4   | 36.0   | 162                      |
| <b>20%</b>         | 5.09                                         | 50.7 | 30.0   | 61.2   | 241                      |

From Table 4.2, it is evident that the relative errors present at heat loss fractions of 16% and 17% are similar in magnitude. However, the sum of square error is lowest at a 17% heat loss fraction. Table 4.3 provides the syngas volume percentages predicted at the heat loss fractions listed in Table 4.2.

**Table 4.3: Syngas Volume Percentages per Heat Loss Fraction**

|                    | Syngas volume percentages [%] |      |                 |                 |
|--------------------|-------------------------------|------|-----------------|-----------------|
| Heat loss fraction | $H_2$                         | CO   | CO <sub>2</sub> | CH <sub>4</sub> |
| 15%                | 49.0                          | 17.3 | 28.7            | 5.06            |
| 16%                | 47.2                          | 16.2 | 30.1            | 6.56            |
| 17%                | 45.2                          | 15.0 | 31.6            | 8.19            |
| 18%                | 43.0                          | 13.9 | 33.2            | 9.99            |
| 19%                | 40.5                          | 12.6 | 34.9            | 12.0            |
| 20%                | 37.7                          | 11.3 | 36.8            | 14.2            |

From Table 4.3, it is evident that as the heat loss increases, the percentages of  $H_2$  and CO decrease, while the percentages of  $CO_2$  and  $CH_4$  increase. This indicates that as more heat is lost the quantity of usable syngas,  $H_2$  and CO, decreases.

Given the available information, the fraction of heat lost will be set at 17% for the remainder of this study. Within the range of values investigated, it has been demonstrated that this fraction cannot be set so that the relative errors are all at local minimums. For this reason, the fraction has been selected where both the sum of square and percentage  $CH_4$  errors are close to local minimums. The thermal-fluid model's average prediction accuracy, at a 17% heat loss fraction, is 83.3%. This is comparable with the average prediction accuracy, which is reported as at least 90%, for a fixed-bed gasifier model developed by He *et al.* (2013:754).

The iterative process for determining the initial mole fraction values is as follows. In the R program, a random set of 100 observations is sampled from the data, and .csv files 1, 2, and 4 are output. These files are input into Model A, and solutions are generated for these 100 random observations.

The averages, per gas, of the 100 predicted mole fractions are accessed in EES. The respective averages are entered into Equations 3.46 and 3.47 and stored in EES as initial values. New solutions are generated for the same 100 observations, and the new average

mole fractions are accessed. These new averages are entered into the equations and stored in EES. New solutions are generated, and this process repeats until the respective averages, per iteration, converge to 3 decimal places.

The averages of the 100 enthalpies predicted, corresponding to respective gases that comprise syngas, are accessed in EES. These averages, along with those of the mole fractions, are entered into Model B as initial values for the respective variables. This process is explained visually in Figure 4.6. The average mole fractions and enthalpies that were determined, through an application of this process, are provided in Table 4.4.

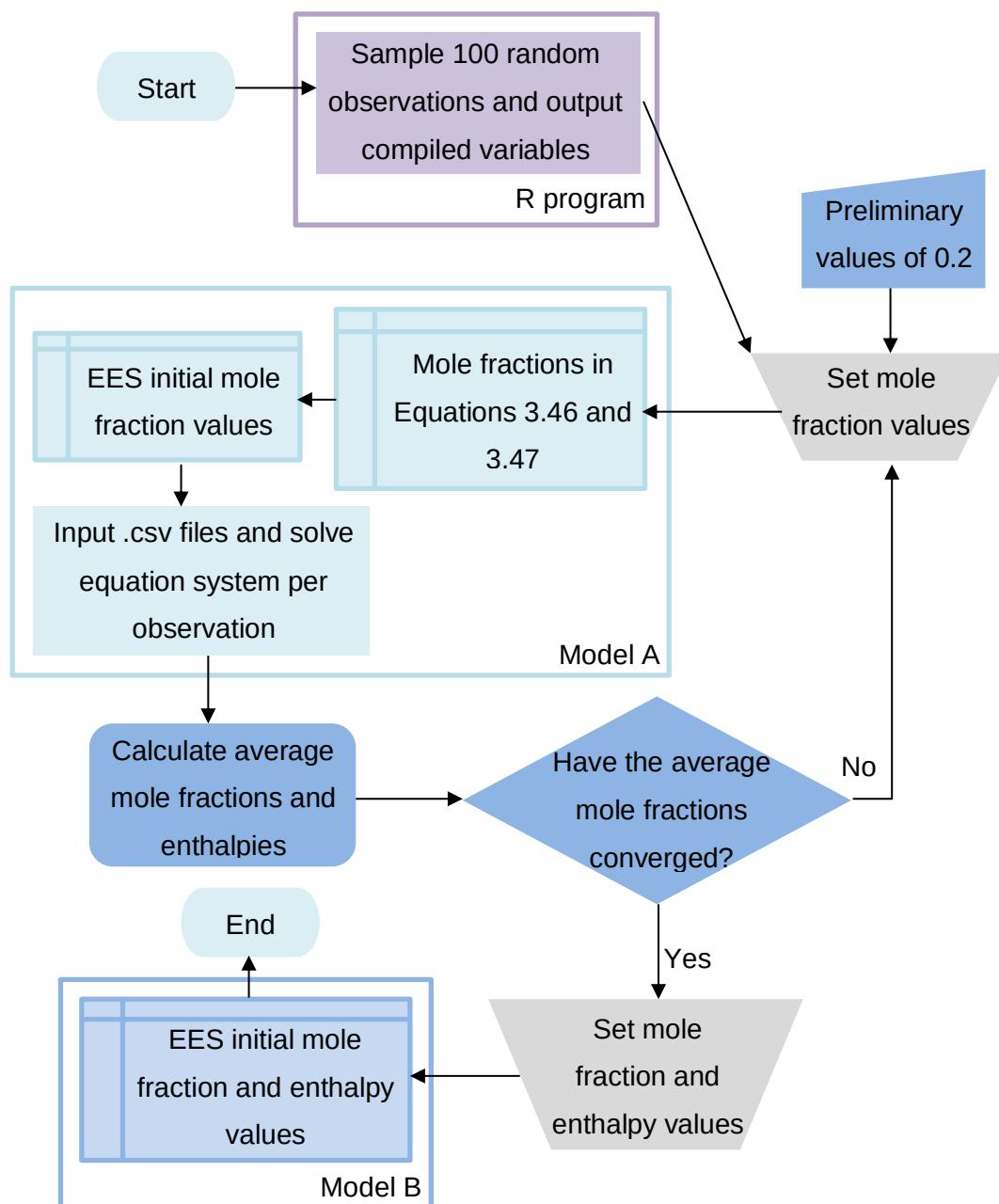


**Figure 4.6: Process for Ascertaining Initial Mole Fraction and Enthalpy Values**

**Table 4.4: Average Mole Fractions and Enthalpies for Syngas**

| Syngas component | Mole fraction [-] | Enthalpy [kJ/kg] |
|------------------|-------------------|------------------|
| $H_2$            | 0.362             | 6,540            |
| $CO$             | 0.132             | -3,470           |
| $CO_2$           | 0.281             | -8,480           |
| $CH_4$           | 0.103             | -3,330           |
| $H_2O$           | 0.123             | -10,100          |

#### 4.6 Verification

In this section, the verification process that is required to determine the efficacy of the thermal-fluid model is discussed. The syngas composition data, from Table 4.1, has already been applied to determine the required heat loss fraction. As such, the accuracy of the results developed, for this data, cannot be used to determine the model's efficacy. For this reason, the verification process will involve demonstrating the calculations that are performed by the model. It must be verified that the model's predictions satisfy the conditions imposed by the five fundamental thermodynamic equations.

For calculations presented in this section, the data discussed in Section 4.2 are input into Model B. This model is applied to determine the mole flow rates developed during gasification, as these values will be required to verify that the five equations have been met. Constants that are required for the calculations are also output from Model B. These are molar masses, densities at Normal conditions, and enthalpies of formation. Enthalpies and entropies for various fluids, calculated at respective temperatures and pressures, are also output from Model B. As applicable, numerical values are provided at the number of significant figures required by standard conventions governing these.

##### 4.6.1 Thermal-Fluid Model Calculations

The complete set of calculations for the thermal-fluid model are available in Appendix B. However, the following results are significant for the three mole balances, the energy balance, and chemical equilibrium for the water gas shift reaction.

#### 4.6.1.1 Mole Balances

In order to verify the three mole balances, the following mole flow rates are provided. The mole flow rates of FC entering and exiting the gasifier are:

- $\dot{n}_{C,in} = 0.545 \text{ kmol/s}$
- $\dot{n}_{C,out} = 0.025 \text{ kmol/s}$

The mole flow rates of steam and oxygen entering the gasifier are:

- $\dot{n}_{H_2O,in} = 0.810 \text{ kmol/s}$
- $\dot{n}_{O_2,in} = 0.108 \text{ kmol/s}$

The respective mole flow rates developed during gasification are:

- $\dot{n}_{H_2O,gas} = 0.249 \text{ kmol/s}$
- $\dot{n}_{H_2,gas} = 0.443 \text{ kmol/s}$
- $\dot{n}_{CO,gas} = 0.144 \text{ kmol/s}$
- $\dot{n}_{CO_2,gas} = 0.317 \text{ kmol/s}$
- $\dot{n}_{CH_4,gas} = 0.0588 \text{ kmol/s}$

Therefore, the following results for the left hand side (LHS) and right hand side (RHS) of the carbon balance, Equation 3.1:

- LHS = 0.545 kmol/s
- RHS = 0.025 kmol/s + 0.144 kmol/s + 0.317 kmol/s + 0.0588 kmol/s = 0.545 kmol/s

The following results for the LHS and RHS of the hydrogen balance, Equation 3.2:

- LHS = 2 \* 0.810 kmol/s = 1.619 kmol/s
- RHS = 2 \* 0.249 kmol/s + 2 \* 0.443 kmol/s + 4 \* 0.0588 kmol/s = 1.619 kmol/s

The following results for the LHS and RHS of the oxygen balance, Equation 3.3:

- LHS = 0.810 kmol/s + 2 \* 0.108 kmol/s = 1.026 kmol/s
- RHS = 0.249 kmol/s + 0.144 kmol/s + 2 \* 0.317 kmol/s = 1.026 kmol/s

Thus, the predicted flow rates, for the  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  developed during gasification, meet the requirements imposed by the equations for the three mole balances.

#### 4.6.1.2 Energy Balance

In order to verify the energy balance, the following variables are provided. The enthalpy-flows of steam and oxygen entering the gasifier are:

- $\dot{H}_{H_2O,in} = -152,000 \text{ kW}$
- $\dot{H}_{O_2,in} = 1,060 \text{ kW}$

The enthalpy-flows of the gases comprising syngas are:

- $\dot{H}_{H_2O,out} = -49,000 \text{ kW}$
- $\dot{H}_{H_2,out} = 7,400 \text{ kW}$
- $\dot{H}_{CO,out} = -15,000 \text{ kW}$
- $\dot{H}_{CO_2,out} = -125,000 \text{ kW}$
- $\dot{H}_{CH_4,out} = -4,300 \text{ kW}$

The heat absorption rates for ash and water jacket steam are:

- $\Delta\dot{Q}_{ash} = -550 \text{ kW}$
- $\Delta\dot{Q}_{wj} = -2,100 \text{ kW}$

The enthalpy-flows of coal and tar are:

- $\dot{H}_{coal} = -9,000 \text{ kW}$
- $\dot{H}_{tar} = -26,700 \text{ kW}$

The rate of heat loss is:

- $\dot{Q}_{loss} = 50,200 \text{ kW}$

Therefore, the following results for the LHS and RHS of the energy balance, Equation 3.12:

- $LHS = -9,000 \text{ kW} - 152,000 \text{ kW} + 1,060 \text{ kW} - 550 \text{ kW} - 2,100 \text{ kW} = -163,000 \text{ kW}$
- $RHS = 50,200 \text{ kW} - 49,000 \text{ kW} + 7,400 \text{ kW} - 15,000 \text{ kW} - 125,000 \text{ kW} - 4,300 \text{ kW} - 26,700 \text{ kW} = -163,000 \text{ kW}$

Thus, the predicted respective total flow rates for  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ , and  $H_2O$  meet the requirements imposed by the equation for the energy balance.

#### 4.6.1.3 Chemical Equilibrium

In order to verify the chemical equilibrium condition for the water gas shift reaction, the following values are provided. The Gibbs functions for the gases present in the water gas shift reaction are:

- $g_{H_2O,out} = -408,000 \text{ kJ/kmol}$
- $g_{H_2,out} = -117,000 \text{ kJ/kmol}$
- $g_{CO,out} = -282,000 \text{ kJ/kmol}$
- $g_{CO_2,out} = -583,000 \text{ kJ/kmol}$

The equilibrium constant from the water gas shift reaction is:

- $K_{wgs} = 3.66$

Therefore, the following results for the LHS and RHS of the equilibrium condition for the water gas shift reaction, Equation 3.63:

- $LHS = \ln(3.66) = 1.30$
- $RHS = -(-583,000 \text{ kJ/kmol} - 117,000 \text{ kJ/kmol} + 282,000 \text{ kJ/kmol} + 408,000 \text{ kJ/kmol}) / [8.31451 \text{ kJ/kmol-K} * (550^\circ\text{C} + 273.15)] = 1.30$

Thus, the predicted respective total flow rates for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $H_2O$  meet the requirements imposed by the equation for this chemical equilibrium condition.

#### 4.6.1.4 Syngas Volume Percentages

In order to determine whether or not the predictions produced by Models A and B are consistent, the following values are provided. The total Normal volume flow rates for the gases, excluding  $H_2O$ , that comprise syngas are:

- $\dot{V}_{H_2,out} = 11.4 \text{ m}_n^3/\text{s}$
- $\dot{V}_{CO,out} = 3.78 \text{ m}_n^3/\text{s}$
- $\dot{V}_{CO_2,out} = 7.93 \text{ m}_n^3/\text{s}$
- $\dot{V}_{CH_4,out} = 2.06 \text{ m}_n^3/\text{s}$

The total Normal volume flow rate of syngas is:

- $\dot{V}_{sgs} = 25.1 \text{ m}_n^3/\text{s}$

Therefore, the syngas volume percentages are:

- $V_{H_2} = [(11.4 \text{ m}_n^3/\text{s}) / (25.1 \text{ m}_n^3/\text{s})] * 100 = 45.2\%$
- $V_{CO} = [(3.78 \text{ m}_n^3/\text{s}) / (25.1 \text{ m}_n^3/\text{s})] * 100 = 15.0\%$
- $V_{CO_2} = [(7.93 \text{ m}_n^3/\text{s}) / (25.1 \text{ m}_n^3/\text{s})] * 100 = 31.6\%$
- $V_{CH_4} = [(2.06 \text{ m}_n^3/\text{s}) / (25.1 \text{ m}_n^3/\text{s})] * 100 = 8.19\%$

Thus, these results are essentially identical to those reported in Table 4.3, at the corresponding heat loss fraction of 17%. This implies that results produced with Model A correspond with those developed with Model B. Thus, applying assumed mole fractions in Equations 3.46 and 3.47 of Model A is reasonable.

#### 4.6.2 Results Discussion

The mass flow rates predicted by the thermal-fluid model satisfy the requirements imposed by the five fundamental thermodynamic equations. As such, the predictions have been verified within the context of the presented model.

Within the calculations that have been presented, in the previous section and Appendix B, the following results require discussion. It was demonstrated that inlet steam more significantly impacts the mole balances than oxygen. This is due to the fact that steam provides over 75% of the total moles of oxygen. It was also established that the flow rates developed during drying and devolatilization, except for methane, are relatively small compared to those developed during gasification.

It was evident that the heat absorption rate of ash has the lowest impact on the energy balance. This is since the magnitude of this variable is significantly smaller than those of the remaining energy terms. The enthalpy-flow of oxygen has the second smallest magnitude in the energy balance. In addition, it was established that the enthalpy-flow of inlet steam has the largest magnitude in the energy balance. The enthalpy-flow of outlet carbon dioxide has the second largest magnitude. The rate of heat loss and the enthalpy-flow of outlet steam are approximately equal in magnitude. For this reason, these are each defined as having the third largest magnitude in the energy balance. This provides an indication of the most significant variables in the balance. These are the variables that, when adjusted, affect the greatest changes in the energy balance.

In Section 4.5, adjustments were made to the rate of heat loss that resulted in large changes to the model's accuracy. Since it has been demonstrated that this parameter has the third largest magnitude in the energy balance, these large changes can thus be explained. It was

noted, in Section 3.5.5, that the fraction of heat lost to the environment is usually between 2.5 and 6% of the lower heating value of coal. In Section 4.5, it was stated that the heat loss fraction is adjusted to account for uncertainty in the model. This uncertainty was attributed to enthalpy-flows of trace gases and hydrocarbons, present in raw syngas, that are not accounted for directly in the model. Therefore, the difference between a 6% heat loss fraction, indicated in literature, and the 17% selected in this study is attributed to the total enthalpy-flow lost in those trace gases and hydrocarbons. As such, the heat loss fraction can no longer be compared with the 2.5 to 6% indicated in literature, since additional losses have been incorporated in this fraction's value.

The following must be noted regarding the accuracy of the predicted syngas volume percentages. As is discussed in Section 2.2.3, thermodynamic equilibrium in reversible chemical reactions cannot be assumed throughout the entire length of a fixed-bed dry bottom coal gasifier. Given that the model assumes chemical equilibrium for the water gas shift reaction at the gasifier's outlet, inaccuracies in the predictions might be attributed to this assumption.

As is evident when comparing the predicted percentages with the reference values from Table 4.1, more hydrogen and less carbon monoxide are predicted than were measured. This supports the hypothesis that the assumption of chemical equilibrium is a source of inaccuracy in the model. This is the case since the water gas shift reaction requires that increases in hydrogen coincide with decreases in carbon monoxide. Thus, it is hypothesized that the reference gasifier data represent non-equilibrium conditions.

In Section 3.6, the equilibrium condition for the water gas shift reaction was selected for application in the thermal-fluid model. However, it was stated that if the equilibrium condition for this reaction was demonstrated to be unsuitable, the methanation reaction would be considered. According to the verification results, the equilibrium condition for the water gas shift reaction has been applied successfully in the model. Thus, the methanation reaction does not need to be considered further. However, a brief analysis of the equilibrium condition for this reaction is provided, in Appendix C, for comparison with that of the water gas shift reaction. From the results presented in Appendix C, it is evident that the methanation equilibrium condition is not satisfied, at syngas outlet conditions, by the predicted mole fractions for the reference gasifier data. Therefore, it is clear that the equilibrium condition for the methanation reaction is unsuitable for implementation in the thermal-fluid model, either in place of or simultaneously along with the presently applied condition. This is given that the accuracy of the thermal-fluid model's predictions, with the present implementation of the equilibrium condition for the water gas shift reaction, has been established.

## 4.7 Conclusions

During this chapter, reference fixed-bed dry bottom coal gasifier data were introduced and discussed. Since not all of the thermal-model's input variables are available in this data, assumptions for these were provided and described. Reference measurements for syngas composition were also available in the data.

The requirement that led to the development of two versions of the thermal-fluid model was presented, and the differences between these versions were described. This requirement is that the model be solvable at data points where negative flow rates are predicted. To achieve this, Model A was developed with a fixed partial pressure calculation, based on assumed mole fractions. Model B was developed without this additional assumption, although it was demonstrated that the results do not differ significantly. In addition, the operation of these models and the integration with an R program was discussed and presented visually.

Some of the uncertainties that still exist in the model were described. These led to the application of an adjusted heat loss fraction in the model, for which a value of 17% was selected. The process followed to select this value required the reference gasifier data and involved calculating relative and absolute errors for predicted syngas volume percentages.

In addition, the application of initial values in EES was discussed. As such, the process for determining the initial values for syngas mole fractions and enthalpies was described and presented visually. This process involved iteratively updating the initial values in EES and re-solving until the mole fractions converged to 3 decimal places. The initial syngas mole fraction and enthalpy values that resulted from the application of this process were provided.

Values from some of the calculations that are performed automatically in EES were provided. This was in order to verify that the predicted mass flow rates satisfy the requirements imposed by the model's five fundamental thermodynamic equations. Within the framework of the thermal-fluid model, this was verified. In addition, a discussion regarding these calculations was provided, wherein the most significant variables were highlighted. A potential source of inaccuracy in the model, the assumed chemical equilibrium of the water gas shift reaction, was also discussed.

## CHAPTER 5: STATISTICAL MODELS

### 5.1 Introduction

In the previous chapter, the thermal-fluid model's implementation in EES was discussed. This included defining two versions of the model, referred to as Models A and B. It was established that the results of these models correspond, and the accuracies of those results were determined. In addition, outputs from the full set of calculations for Model B were provided to verify that the conditions imposed by the model's fundamental equations are satisfied.

In this chapter, statistical models will be developed to determine if, from the results of the thermal-fluid model, gasifier operations can be classified as normal and abnormal. Typical gasifier data were supplied to Models A and B, and the resulting mass flow rate predictions will be analyzed in this chapter. Models A and B were applied as described in Chapter 4.

A result of the differences between Models A and B is that solutions can be generated for the former at observations where the latter cannot be solved, as discussed in Section 4.3. It is hypothesized that unsolvable observations represent abnormal operating conditions that are not in line with scientific expectations from the thermal-fluid model. The reason for this is that negative flow rates, and the corresponding negative mole fractions that prevent Model B from solving, are indicative of abnormal operations. There are also no process measurements, available for use in this study, that indicate abnormal operations. As such, the statistical modeling will be aimed at predicting whether Model B will solve at a given observation.

Statistical models are developed in Section 5.2. As mentioned in Section 4.4, this modeling will be performed in R. The statistical methods and models that were discussed in Section 2.3 are principal component analysis, qualitative trend analysis, support vector machines, and classification trees. However, from these available methods and models, qualitative trend analysis will not be considered further, since the unsolvability of Model B will be used to indicate potential abnormal observations. Principal component analysis is undertaken in Section 5.2.1. Section 5.2.2 presents and discusses the classification models that are developed to predict where Model B can and cannot be solved. These are classification trees and support vector machines, in Sections 5.2.2.1 and 5.2.2.2, respectively. In Section 5.2.3, one statistical model is selected for further investigation.

The selected statistical model's results will be interpreted in Section 5.3. This will allow the validity of the thermal-fluid model to be ascertained. It will also be determined if the hypothesis that unsolvable observations represent abnormal operating conditions is invalid. Section 5.4

compares the selected model with conventional process monitoring for coal gasifiers, and Section 5.5 concludes the chapter.

## 5.2 Statistical Models

In this section, statistical models are developed and discussed to predict the points at which Model B can and cannot be solved. This model operates so that a 1 is recorded for any point where the mass flow rates of  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ , and  $H_2O$  cannot be predicted and recorded. Points, such as these, are where a solution cannot be generated for Model B, and the locations of these are known from the model's results. The results developed by Model A, along with the locations of these points from Model B, will be applied in the statistical models. In the next section, principal component analysis (PCA) is utilized to determine the mass flow rates that will be applied in the statistical classification models.

### 5.2.1 Principal Component Analysis

As mentioned in Section 2.3.1, PCA is significant when correlated variables are present. The major elements in syngas, carbon, hydrogen, and oxygen, are divided between  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ , and  $H_2O$  according to mole and energy balances. Therefore, the thermal-fluid model yields flow rates that are correlated. The application of chemical equilibrium for the water gas shift reaction also ensures correlation between flow rates.

PCA can allow more accurate models to be developed, than are produced when using the original variables (James *et al.*, 2014:234). This is since the latent variables generated by PCA usually explain a large percentage of the variability in the data (James *et al.*, 2014:234,374). Furthermore, applying this smaller set of predictors aids in the prevention of overfitting (James *et al.*, 2014:234). Overfitting causes patterns, which are only present due to random variations in the data, to become part of the model, which decreases prediction accuracy (James *et al.*, 2014:32).

In addition, PCA predicts the location of surfaces that are nearest to the data points. These provide simpler, lower dimensional, perspectives on the data (James *et al.*, 2014:374-375,379). Analyzing the predicted mass flow rates, when projected onto one of these surfaces, is the aim with PCA.

The first principal component is the direction along which data has the greatest variability. The second component is determined so that, from the available directions that are uncorrelated with the first component, the largest variability is evident along it (James *et al.*, 2014:376-377).

Thus, a surface spanned by the first two principal components provides a near-exhaustive two-dimensional perspective on the total variability of a data set.

Principal component directions are defined by loading vectors. Loadings, which comprise these vectors, are weights assigned to the respective variables. When observations are projected onto a principal component direction, the values that result are referred to as scores (James *et al.*, 2014:375-376,378).

Principal components are produced, from the five mass flow rates, with Equation 5.1. The principal component loadings, applied in Equation 5.1, are calculated according to the optimization problem defined in Equation 5.2 (James *et al.*, 2014:375-376).

$$\begin{aligned} \underline{pc}_x = & \phi_{x,1} * \underline{\dot{m}}_{H_2O,out} + \phi_{x,2} * \underline{\dot{m}}_{H_2,out} + \phi_{x,3} * \underline{\dot{m}}_{CO,out} \\ & + \phi_{x,4} * \underline{\dot{m}}_{CO_2,out} + \phi_{x,5} * \underline{\dot{m}}_{CH_4,out} \end{aligned} \quad [5.1]$$

$$maximize \left\{ \frac{1}{\omega} \sum_{w=1}^{\omega} (pc_{x,w})^2 \right\} \text{ subject to } \sum_{r=1}^5 (\phi_{x,r})^2 = 1 \quad [5.2]$$

here  $\underline{pc}_x$  represents an  $\omega$ -dimensional vector for principal component  $x$ , where  $\omega$  is the total number of observations and  $x$  is an element of  $\{1, 2, 3, 4, 5\}$ . Loading  $r$  for principal component  $x$  is represented by  $\phi_{x,r}$ , where loadings 1 through 5 are allocated to  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$ , respectively. The outlet mass flow rates of  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are contained in the  $\omega$ -dimensional vectors  $\underline{\dot{m}}_{H_2O,out}$ ,  $\underline{\dot{m}}_{H_2,out}$ ,  $\underline{\dot{m}}_{CO,out}$ ,  $\underline{\dot{m}}_{CO_2,out}$ , and  $\underline{\dot{m}}_{CH_4,out}$ , respectively. The flow rates in these vectors, calculated with Equations A.48 through A.52, are summations of the predicted dried, devolatilized, and gasified flow rates. Observation  $w$  of  $\underline{pc}_x$ , also referred to as score  $w$  of principal component  $x$ , is  $pc_{x,w}$ .

It is recommended that before PCA is undertaken, variables are centered and standardized. These ensure that the variables applied in PCA have respective means of zero and standard deviations of one (James *et al.*, 2014:380-381). The results of performing PCA on the five predicted mass flow rates are presented in Figure 5.1. This was performed on the outputs of Model A, for the observations where Model B is applicable, after centering and standardizing. The principal component scores are plotted, in black, along the lower horizontal and left vertical axes. The principal component loadings are plotted, in blue, along the upper horizontal and right vertical axes. A 95% confidence ellipse for the scores is drawn, in red, on Figure 5.1. Confidence ellipses are plotted using functions from the ellipse R package, version 0.4.3 (Murdoch & Chow, 2022).

The first principal component direction explains 60.8% of the variance in the data, and 25.2% is explained by the second direction. This implies that Figure 5.1 clarifies a significant portion, 86.0%, of the variability in the data. Thus, from the data projected onto this two-dimensional plane, the mass flow rates can be analyzed further.

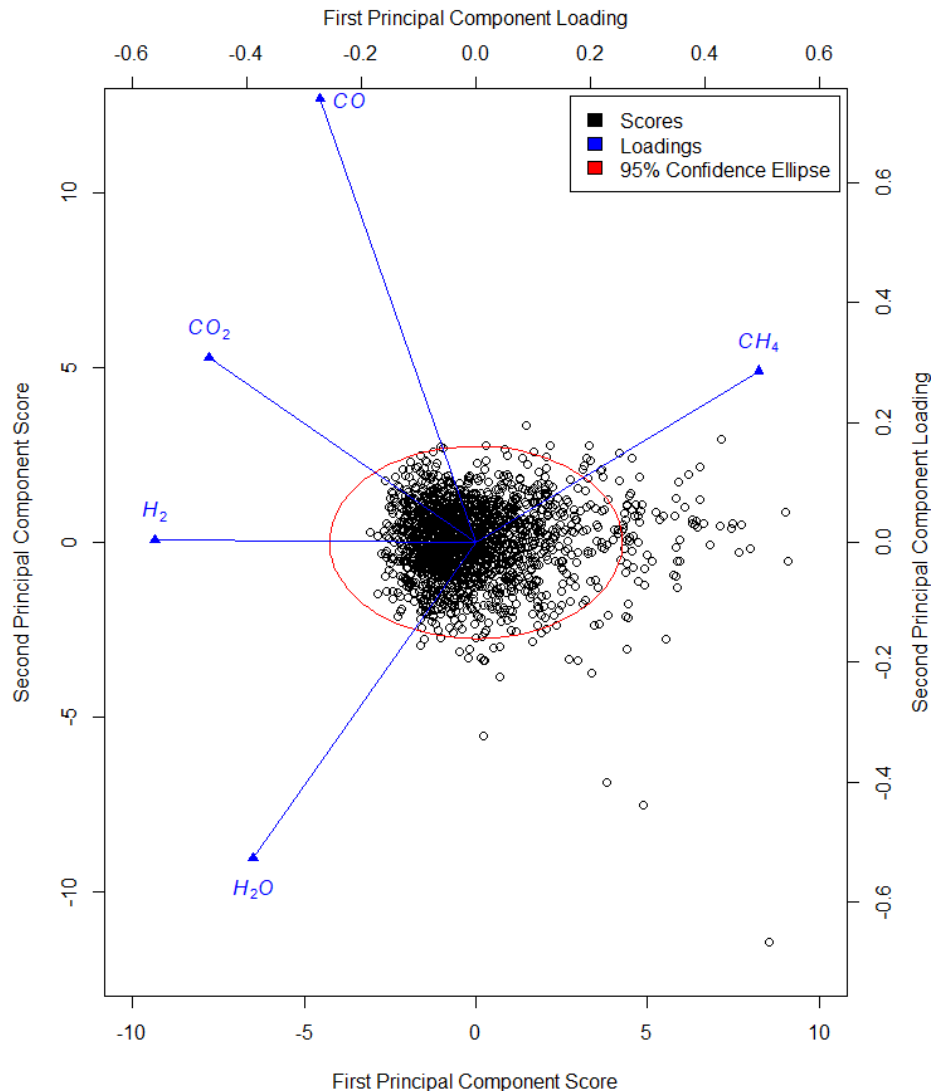


**Figure 5.1: PCA Biplot for Predicted Mass Flow Rates**

As is evident from Figure 5.1, the majority of scores outside the 95% confidence ellipse are present along the positive half of the first principal component direction. This is contrasted with the result that, along both halves of the second direction, points outside the ellipse are present in similar proportions. However, the furthest outlying points are evident along the negative half of this direction. Arguably, the gasifier's operations could be classified as normal and abnormal, solely based on the results of this PCA. The points inside and outside the ellipse would be defined as normal and abnormal, respectively (Coetzer *et al.*, 2014:60-61). However, this is reliant on the selection of an arbitrary confidence level.

In this study, principal component loadings for the flow rates are significant. This is since larger weights are assigned to variables that more significantly contribute to the respective principal component direction (James *et al.*, 2014:377-378,381). In addition, uncorrelated variables have loading directions that are orthogonal (James *et al.*, 2014:377-378).

From Figure 5.1, it is evident that the mass flow rate of  $H_2$  has a large negative first principal component loading. However, this flow rate has a negligible second component loading. The respective loadings for the first and second principal components are provided in Table 5.1. From this table, it is evident that  $H_2$  has the largest contribution to the first principal component direction. In addition, the flow rate with the smallest contribution to the first principal component and the largest contribution to the second principal component is  $CO$ .

**Table 5.1: First and Second Principal Component Loadings**

| Variable | First principal component loadings | Second principal component loadings |
|----------|------------------------------------|-------------------------------------|
| $H_2O$   | -0.389                             | -0.527                              |
| $H_2$    | -0.560                             | 0.003                               |
| $CO$     | -0.272                             | 0.739                               |
| $CO_2$   | -0.466                             | 0.307                               |
| $CH_4$   | 0.494                              | 0.286                               |

From Figure 5.1, the following pairs of mass flow rates demonstrate the least correlation,  $H_2$  and  $CO$ ,  $CO$  and  $CH_4$ ,  $CO_2$  and  $CH_4$ , and  $H_2O$  and  $CO_2$ . Therefore, in the following sections, statistical models will be developed for these pairs of flow rates and the first two principal components. Considering pairs of flow rates will reduce the potential for overfitting, while ensuring that the developed models are interpretable. PCA aids in the avoidance of overfitting, as previously mentioned, but lowers the interpretability of results. This is since the principal component directions, which are uncorrelated and explain a significant portion of the variability in the data, are predicted as linear combinations of the available variables (James *et al.*, 2014:375-376). As such, variables that demonstrate the least correlation, while still clarifying a significant portion of the variability in the data, should be applied to develop the statistical models. These models will be more interpretable, and overfitting is likely to be avoided.

### 5.2.2 Classification Models

As stated, classification trees and support vector machines have been selected for application in this chapter. Thus, these statistical models will be applied to predict where Model B can and cannot be solved. Investigation of the resulting models will indicate the suitability of these to the gasification process. However, it must be noted that the existence of gasifier faults at specific observations has yet to be ascertained.

Although the ratio of unsolvable to solvable observations is extremely low at 0.0222, class imbalance correction techniques will not be applied in this study. This is since it has been established that observations in the minority class, which are separated from the majority class, can be differentiated during unbalanced classification problems (Jegierski & Saganowski, 2020:125193). Furthermore, as is evident in the results presented in the following sections, essentially all the unsolvable observations are separate from those where Model B is solvable.

For the statistical modeling, observations will randomly be selected for use in a validation set, as mentioned in Section 1.4. During classification model training, a randomly selected set composed of 50% to 70% of the available observations should be applied (Xu & Goodacre, 2018:259). As such, 550 observations, or approximately 30% of the data, will be randomly selected for use in a validation set.

In addition, repeated random sampling, and subsequent model training and testing, is recommended (Xu & Goodacre, 2018:259). However, it is likely that the classification errors for the minority class will be indistinguishable within the results of a statistical resampling technique. This is given the extremely low ratio of unsolvable to solvable observations. As such, accuracy and classification errors will be compared, between statistical models, for one randomly selected pair of training and validation sets.

#### 5.2.2.1 Classification Trees

In this section, classification trees are developed and analyzed for the mass flow rate pairs, selected in Section 5.2.1, and the first two principal components. A classification tree (CT) functions by applying rules, per variable, to split the provided space into smaller regions (James *et al.*, 2014:303). Predictions are made based on the most frequently occurring class, evident during model training, in a respective terminal region (James *et al.*, 2014:311). The term tree refers to the frequent summarization of the model's splitting rules in the form of an upside-down tree (James *et al.*, 2014:303,305). Figure 5.2 presents the splitting rules and resulting regions for a two-dimensional CT (James *et al.*, 2014:308).

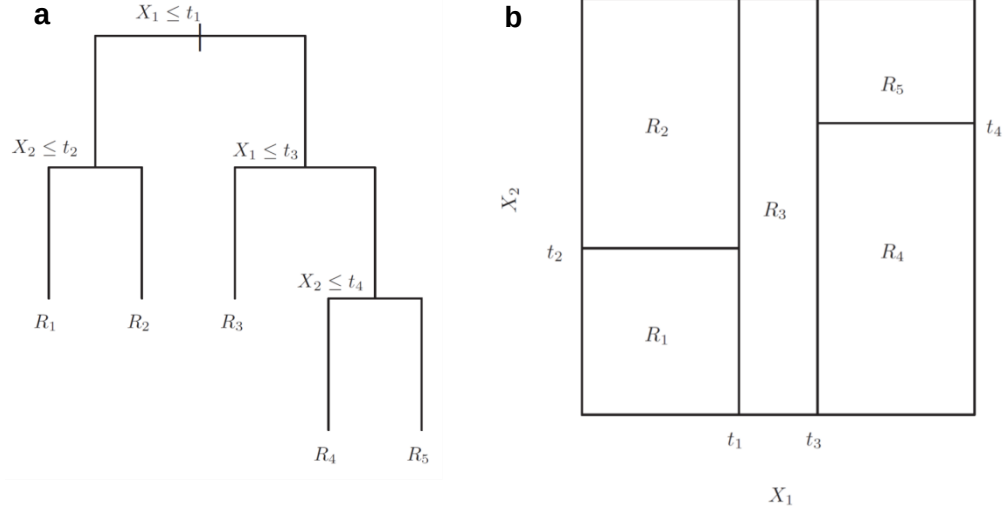


**Figure 5.2: CT (a) Splitting Rules and (b) Resulting Regions – James *et al.*, 2014:308**

The CT equations provided, in this section, are defined in terms of mass flow rates. Appendix A.8.1 provides equations for the principal component CT model, which are adapted from those provided in this section. Equation 5.3 defines the division of a given space into two regions. The space is split, for one of the variables, at a value selected according to the optimization problem in Equation 5.4. Equation 5.4 provides the classification model's Gini index, which is a measure of total variance. Binary splitting of the space continues until a minimum is reached for the Gini index, while excessive splitting is avoided in order to prevent overfitting (James *et al.*, 2014:307,312).

$$Rg_{2b-1} = \{\dot{m}_{j,out} \mid \dot{m}_{\zeta,out} < sp_b\} \text{ and } Rg_{2b} = \{\dot{m}_{j,out} \mid \dot{m}_{\zeta,out} \geq sp_b\} \quad [5.3]$$

$$\text{minimize } Gn = \sum_{a=1}^A \sum_{cl=1}^2 [pr_{a,cl} * (1 - pr_{a,cl})] \quad [5.4]$$

here  $\dot{m}_{j,out}$  represents the two-dimensional outlet mass flow rate space that the CT model is applied to. Regions corresponding to split  $b$ ,  $sp_b$ , of outlet mass flow rate  $\zeta$ ,  $\dot{m}_{\zeta,out}$ , are  $Rg_{2b-1}$  and  $Rg_{2b}$ . Therefore,  $\zeta$  is an element of  $j$  and represents  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , or  $CH_4$ . The number of terminal regions is  $A$ , and  $b$  is an element of the set of natural numbers  $\{1, 2, 3, 4, \dots\}$ . Along with  $sp_b$ ,  $\dot{m}_{\zeta,out}$  is selected from  $\dot{m}_{j,out}$  in order to minimize the Gini index,  $Gn$ , of the CT model. In the Gini index,  $pr_{a,cl}$  is the proportion of training observations, which are present in terminal region  $a$ , that belong to class  $cl$ .

Terminal regions are defined by the intersection of regions that precede these in the CT's decision structure (James *et al.*, 2014:305). This intersection is defined in Equation 5.5.

$$Tg_a = \bigcap_{tn \in TN} Rg_{tn} \quad [5.5]$$

where  $Tg_a$  is terminal region  $a$ . Subset  $TN$  defines the set of regions, represented by  $Rg_{tn}$ , in the CT that precede  $Tg_a$ .

After a CT is produced, it is pruned to improve interpretability. Pruning involves systematically removing splits in order to select a subtree with the lowest classification error rate. The classification error rate is the fraction of misclassified observations and is calculated with Equation 5.6 (James *et al.*, 2014:307-308,311-312).

$$CER = \frac{1}{\omega} \sum_{a=1}^A \min \left\{ \begin{matrix} num[\dot{m}_{j,out}(cl = 1) \in Tg_a] \\ num[\dot{m}_{j,out}(cl = 2) \in Tg_a] \end{matrix} \right\} \quad [5.6]$$

where  $CER$  is the classification error rate and  $\omega$  is the total number of observations. The functions  $\min$  and  $num$  are applied to select the number of observations in the smallest class of  $Tg_a$ , which represent misclassifications. Observations can either belong to class 1 or 2, which represent where Model B is or is not solvable, respectively.

From Section 5.2.1, the first pair of predicted mass flow rates that will be applied in a statistical model are  $H_2$  and  $CO$ . The regions that result from applying a CT model to the training set, for these flow rates, are presented in Figure 5.3. CT models are trained using functions from the tree R package, version 1.0-41 (Ripley, 2021). A 95% confidence ellipse, for the observations where Model B can be solved, is drawn on the figure in red. Observations are colored, and regions are shaded, blue and green to indicate where Model B can and cannot be solved, respectively. The boundaries, defined by the CT model, are drawn on the figure in black.

As is evident from Figure 5.3, the  $H_2$  and  $CO$  CT model defined three splitting rules, which divide the area into four regions. Of these, the region where Model B is predicted to solve is bounded in three directions. Thus, this tree model falsely predicts that Model B can be solved at observations where the mass flow rate of  $CO$  approaches infinity.

In addition, it is evident from Table 5.2 that only a few observations, 9 in total, are misclassified by the  $H_2$  and  $CO$  CT model. The number of observations that are misclassified, during model

training or validation, is presented in what is referred to as a confusion matrix (James *et al.*, 2014:145). This model's training and validation confusion matrices are provided in Table 5.2.

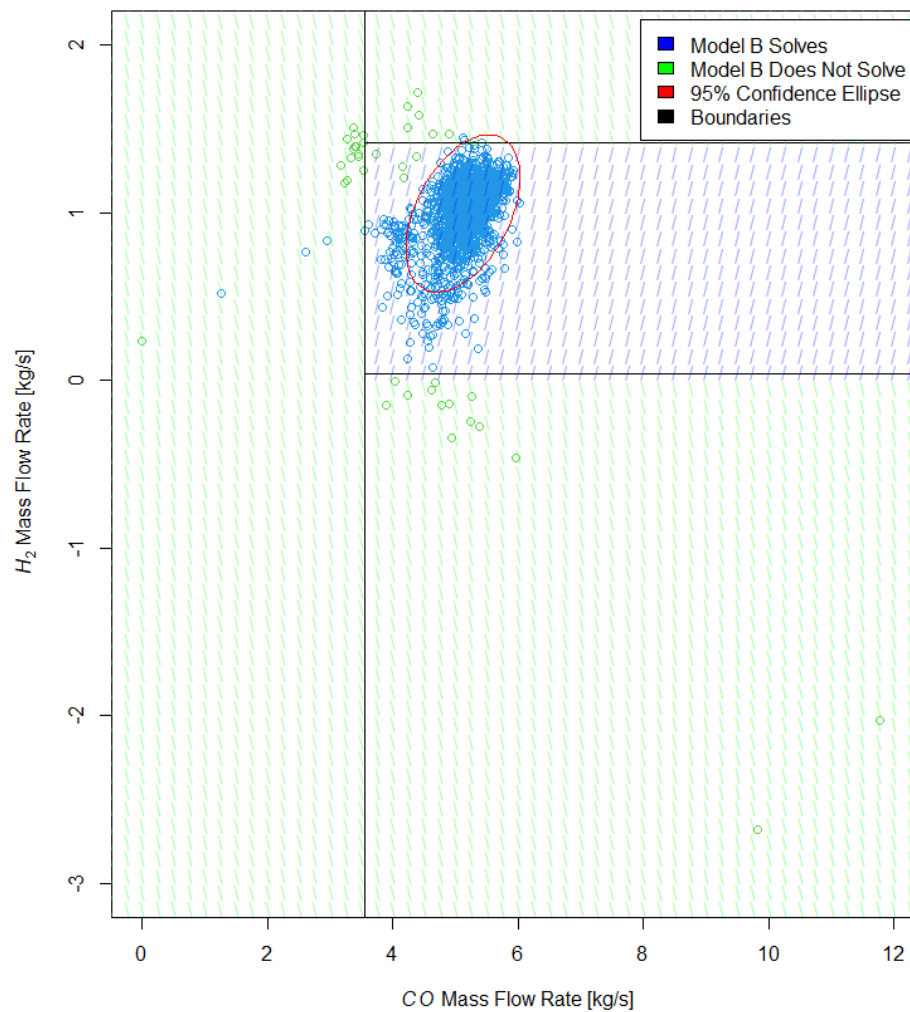


**Figure 5.3:  $H_2$  and CO CT Model Plot**

**Table 5.2:  $H_2$  and CO CT Model Confusion Matrices**

|                               | Training set   |        | Validation set |        |
|-------------------------------|----------------|--------|----------------|--------|
|                               | Does not solve | Solves | Does not solve | Solves |
| <b>Predicted not to solve</b> | 23             | 3      | 14             | 2      |
| <b>Predicted to solve</b>     | 3              | 1,311  | 1              | 533    |

In this study, model accuracy defines the percentage of observations that are correctly classified. This is calculated according to Equation 5.7, which is provided at the end of this section, using the values in a confusion matrix (James *et al.*, 2014:37,145). The  $H_2$  and CO CT model's training and validation accuracies, as provided in Table 5.7, are 99.6% and 99.5%, respectively. These values are relatively high, indicating that this CT model can differentiate between observations where Model B does and does not solve. However, having an unbounded region, where Model B is predicted to solve, indicates that the results of this CT model are not generalizable.

From Section 5.2.1, the second pair of predicted mass flow rates that will be applied in a statistical model are  $CH_4$  and CO. The regions that result from applying a CT model to the training set, for these flow rates, are presented in Figure 5.4. The confusion matrices for this model are provided in Table 5.3.

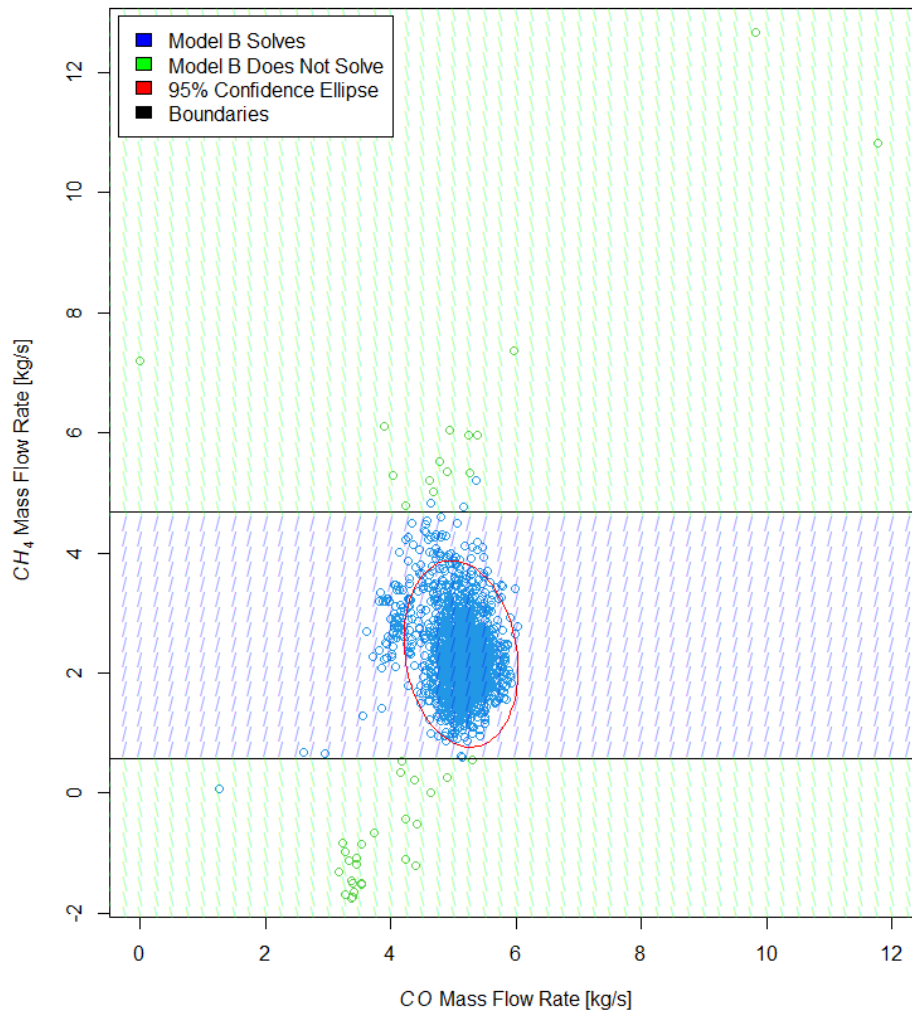


**Figure 5.4:  $CH_4$  and CO CT Model Plot**

**Table 5.3:  $CH_4$  and CO CT Model Confusion Matrices**

|                               | Training set   |        | Validation set |        |
|-------------------------------|----------------|--------|----------------|--------|
|                               | Does not solve | Solves | Does not solve | Solves |
| <b>Predicted not to solve</b> | 26             | 3      | 15             | 1      |
| <b>Predicted to solve</b>     | 0              | 1,311  | 0              | 534    |

It is evident from Figure 5.4 that the CT defined two splitting rules, which divide the area into three regions. In addition, those rules only depend on the  $CH_4$  mass flow rates, and the developed model is independent of CO. Therefore, this CT model implies that whenever the mass flow of  $CH_4$  is between the defined upper and lower limits, Model B will solve. It also is evident from Table 5.3 and Figure 5.4 that very few observations, 4 in total, are misclassified. Furthermore, the accuracies of this CT model, provided in Table 5.7, for the training and validation sets are both extremely high at 99.8%. Thus, the results of this CT model may be generalizable.

From Section 5.2.1, the third pair of predicted mass flow rates that will be applied in a statistical model are  $CH_4$  and  $CO_2$ . The regions that result from applying a CT model to the training set, for these flow rates, are presented in Figure 5.5. The confusion matrices for this model are provided in Table 5.4.

**Table 5.4:  $CH_4$  and  $CO_2$  CT Model Confusion Matrices**

|                               | Training set   |        | Validation set |        |
|-------------------------------|----------------|--------|----------------|--------|
|                               | Does not solve | Solves | Does not solve | Solves |
| <b>Predicted not to solve</b> | 26             | 3      | 15             | 1      |
| <b>Predicted to solve</b>     | 0              | 1,311  | 0              | 534    |

It is evident from Figure 5.5 that the CT defined two splitting rules, which divide the area into three regions. In addition, those rules only depend on the  $CH_4$  mass flow rates. Therefore,

the developed model is independent of  $CO_2$  and is identical to that developed for  $CH_4$  and  $CO$ . Table 5.4 further confirms that these CT models are identical, given that the presented results are equal to those in Table 5.3. Accordingly, these CT models' respective accuracies, provided in Table 5.7, are also equal.

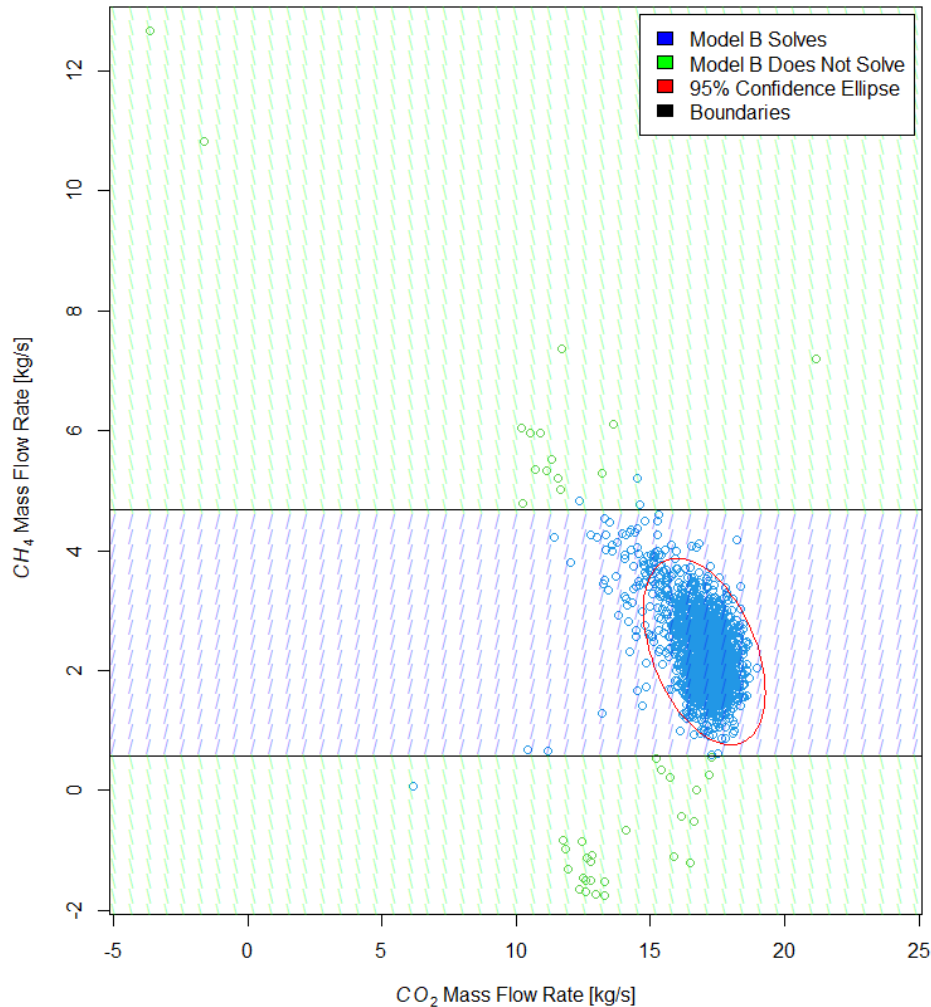


**Figure 5.5:  $CH_4$  and  $CO_2$  CT Model Plot**

From Section 5.2.1, the fourth pair of predicted mass flow rates that will be applied in a statistical model are  $H_2O$  and  $CO_2$ . The regions that result from applying a CT model to the training set, for these flow rates, are presented in Figure 5.6. The confusion matrices for this model are provided in Table 5.5.

From Figure 5.6, it is evident that the CT model defined three splitting rules, which divide the area into four regions. It is also evident, from Table 5.5, that a larger number of observations are misclassified, 13 in total, when compared with the results of the previous three CT models. This model's accuracies for the training and validation sets are, respectively, 99.4% and

99.1%, as provided in Table 5.7. Although slightly lower than those for the previous three CT models, these values represent high accuracy. In addition, it is evident from Figure 5.6 that the regions, where Model B is predicted to solve, are not bounded. Therefore, this CT model incorrectly predicts that Model B will solve at mass flow rates of  $\text{CO}_2$  and  $\text{H}_2\text{O}$  ranging from minus infinity to infinity. Thus, the results of this model are not generalizable.

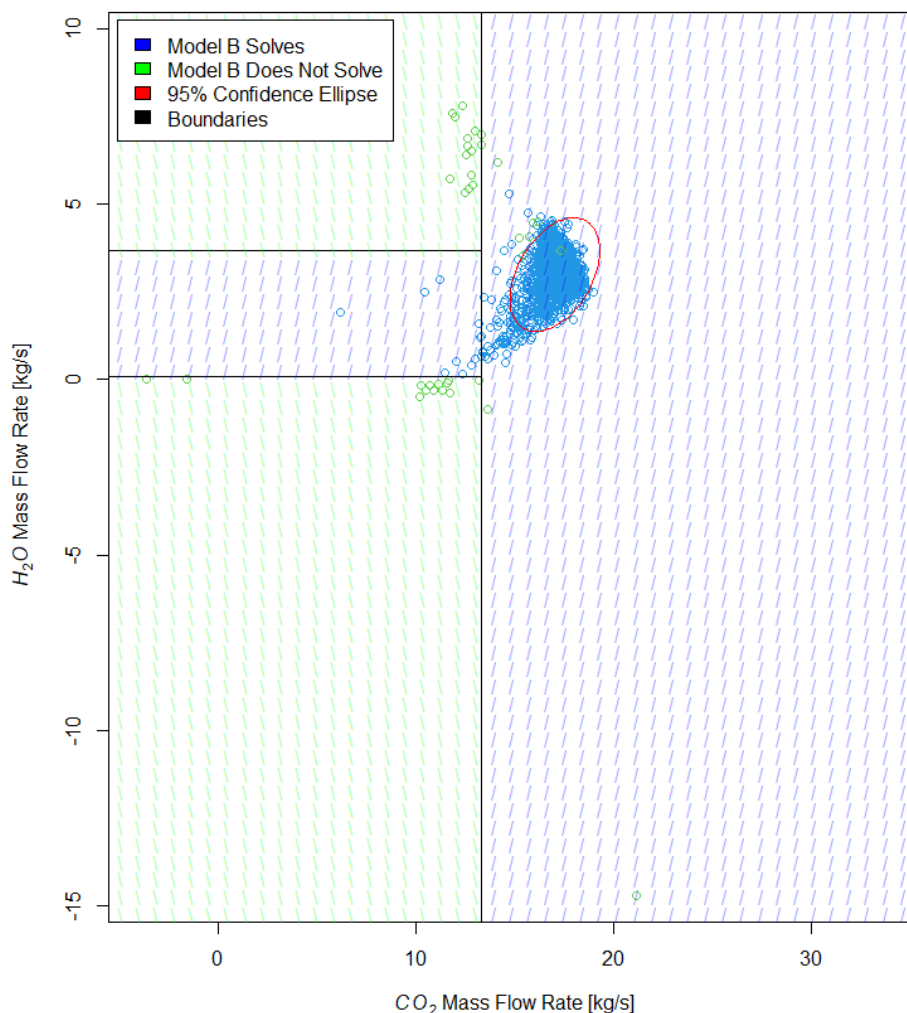


**Figure 5.6:  $\text{H}_2\text{O}$  and  $\text{CO}_2$  CT Model Plot**

It is informative to develop and present a CT model for the first two principal components, in order to compare this with the previous four CT models. Classification models developed for the first two principal component directions are expected to possess accuracies that are similar to those of the presented models. This is since the first two principal component directions are uncorrelated and explain a significant portion, 86.0%, of the variability in the mass flow rates.

**Table 5.5:  $H_2O$  and  $CO_2$  CT Model Confusion Matrices**

|                               | Training set   |        | Validation set |        |
|-------------------------------|----------------|--------|----------------|--------|
|                               | Does not solve | Solves | Does not solve | Solves |
| <b>Predicted not to solve</b> | 18             | 0      | 10             | 0      |
| <b>Predicted to solve</b>     | 8              | 1,314  | 5              | 535    |

Through the loadings in Table 5.1, observations where Model B can and cannot be solved are projected onto the first two principal component directions. The results of applying a CT model to the training set, for scores of the first two principal component directions, are presented in Figure 5.7. The confusion matrices for this model are provided in Table 5.6.

**Table 5.6: Principal Component CT Model Confusion Matrices**

|                               | Training set   |        | Validation set |        |
|-------------------------------|----------------|--------|----------------|--------|
|                               | Does not solve | Solves | Does not solve | Solves |
| <b>Predicted not to solve</b> | 22             | 0      | 14             | 0      |
| <b>Predicted to solve</b>     | 4              | 1,314  | 1              | 535    |

From Figure 5.7, it is evident that the CT model defined three splitting rules, which divide the area into four regions. It is also evident, from Figure 5.7 and Table 5.6, that very few observations, 5 in total, are misclassified. Furthermore, this model only misclassifies one more observation than the  $CH_4$  and  $CO$ , or equivalent  $CH_4$  and  $CO_2$ , CT model. This model's accuracies for the training and validation sets, as provided in Table 5.7, are 99.7% and 99.8%, respectively, which represent extremely high accuracy. However, it is evident from Figure 5.7 that the regions, where Model B is predicted to solve, are not bounded. Therefore, this CT model incorrectly predicts that Model B will solve for first and second principal component scores ranging from minus infinity to infinity. Thus, the results of this model are not generalizable.

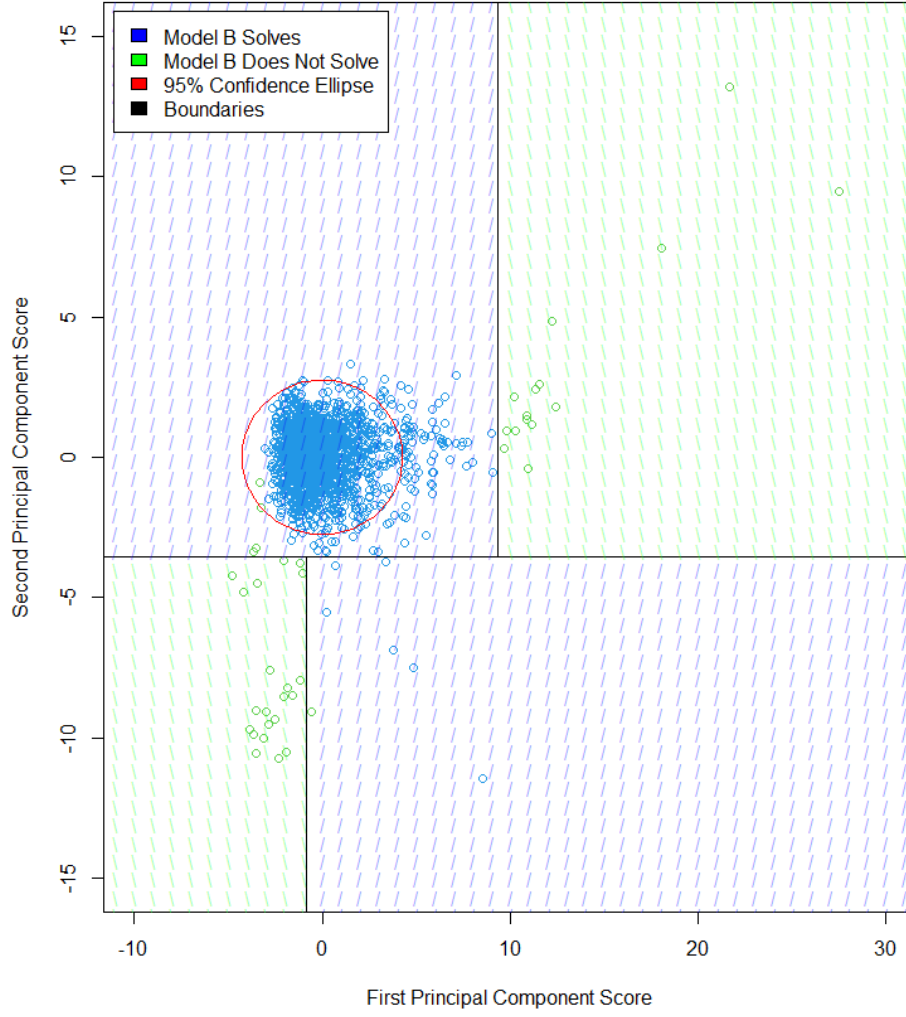


**Figure 5.7: Principal Component CT Model Plot**

The values presented in confusion matrices are applied to determine error and accuracy metrics for classification models (James *et al.*, 2014:145). The equations presented for these metrics, in this section, are defined in terms of mass flow rates. Equations defined in terms of principal components, for these error and accuracy metrics, are provided in Appendix A.8.3.

Model accuracy is calculated using Equation 5.7 and defines, as previously stated, the percentage of observations that are correctly classified (James *et al.*, 2014:37).

$$Ac = \frac{\text{num}[\dot{m}_{j,out}(cl' = cl)]}{\omega} * 100 \quad [5.7]$$

here  $Ac$  is accuracy, and predicted classes are represented by  $cl'$ .

Model errors are presented, in this study, through metrics referred to as the non-detection and false-alarm rates. Equations 5.8 and 5.9 are applied, respectively, to calculate the non-detection and false-alarm rates, which define the percentage of unsolvable and solvable observations that are misclassified (Jiang *et al.*, 2016:380).

$$NDR = \frac{\text{num}[\dot{m}_{j,out}(cl' = 1 | cl = 2)]}{\text{num}[\dot{m}_{j,out}(cl = 2)]} * 100 \quad [5.8]$$

$$FAR = \frac{\text{num}[\dot{m}_{j,out}(cl' = 2 | cl = 1)]}{\text{num}[\dot{m}_{j,out}(cl = 1)]} * 100 \quad [5.9]$$

where  $NDR$  and  $FAR$  are the non-detection and false-alarm rates, respectively. The classes where Model B is solvable and unsolvable are, respectively, indicated by  $cl = 1$  and  $cl = 2$ .

Table 5.7 contains the accuracies, non-detection rates, and false alarm rates of the CT models presented in this section. It is immediately evident that the false-alarm rates are considerably smaller than the non-detection rates. This is due to the extremely low ratio of unsolvable to solvable observations. As such, the non-detection and false-alarm rates cannot be compared with each other.

The inequality between the non-detection and false-alarm rates is evident between the principal component CT model and the  $CH_4$  and  $CO$ , or equivalent  $CH_4$  and  $CO_2$ , CT model. The principal component model has low non-detection rates of 15.4% and 6.67% for the training and validation sets, respectively. However, the  $CH_4$  and  $CO$  model has non-detection rates of 0% for both the training and validation sets. False-alarm rates of 0%, for both the training and validation sets, are evident for the principal component model. The  $CH_4$  and  $CO$  model has extremely low false-alarm rates of 0.228% and 0.187% for the training and validation sets, respectively.

Therefore, the non-detection and false-alarm rates erroneously indicate that the  $CH_4$  and  $CO$  model is more applicable to this classification problem than the principal component model. However, the  $CH_4$  and  $CO$  model misclassifies four observations in total, whereas the principal component model misclassifies five observations in total. Therefore, when judging between models during imbalanced classification problems, it is more informative to compare the total number of misclassifications, rather than the non-detection and false-alarm rates.

Table 5.7: Accuracy and Error Metrics for CT Models

|                                                        | Training set | Validation set |
|--------------------------------------------------------|--------------|----------------|
| <b><i>H<sub>2</sub></i> and <i>CO</i></b>              |              |                |
| <b>Accuracy</b>                                        | 99.6%        | 99.5%          |
| <b>Non-detection rate</b>                              | 11.5%        | 6.67%          |
| <b>False-alarm rate</b>                                | 0.228%       | 0.374%         |
| <b><i>CH<sub>4</sub></i> and <i>CO</i></b>             |              |                |
| <b>Accuracy</b>                                        | 99.8%        | 99.8%          |
| <b>Non-detection rate</b>                              | 0%           | 0%             |
| <b>False-alarm rate</b>                                | 0.228%       | 0.187%         |
| <b><i>CH<sub>4</sub></i> and <i>CO<sub>2</sub></i></b> |              |                |
| <b>Accuracy</b>                                        | 99.8%        | 99.8%          |
| <b>Non-detection rate</b>                              | 0%           | 0%             |
| <b>False-alarm rate</b>                                | 0.228%       | 0.187%         |
| <b><i>H<sub>2</sub>O</i> and <i>CO<sub>2</sub></i></b> |              |                |
| <b>Accuracy</b>                                        | 99.4%        | 99.1%          |
| <b>Non-detection rate</b>                              | 30.8%        | 33.3%          |
| <b>False-alarm rate</b>                                | 0%           | 0%             |
| <b>Principal components</b>                            |              |                |
| <b>Accuracy</b>                                        | 99.7%        | 99.8%          |
| <b>Non-detection rate</b>                              | 15.4%        | 6.67%          |
| <b>False-alarm rate</b>                                | 0%           | 0%             |

### 5.2.2.2 Support Vector Machines

In this section, support vector machines are developed for the mass flow rate pairs, selected in Section 5.2.1, and the first two principal components. A support vector machine (SVM) is applied to determine non-linear decision boundaries between classes, according to the constraints of an optimization problem and a specified kernel function (James *et al.*, 2014:346,350,352). This optimization problem defines that the width of a margin around the classification boundary must be maximized (James *et al.*, 2014:346).

Observations, from a respective class, that are located on or across that class's edge of the margin are referred to as support vectors (James *et al.*, 2014:347). The support vectors and kernel function, respectively, determine the location and shape of the decision boundary (James *et al.*, 2014:347,352). Types of kernels include linear, polynomial, and radial functions (James *et al.*, 2014:352). Figure 5.8 provides a visual representation of a polynomial SVM, in two dimensions, as applied to determine a decision boundary between two classes. These classes, and the respective regions determined by the SVM, are colored blue and pink in this figure. In addition, the boundaries and margins in this figure are represented by solid and dashed lines, respectively (James *et al.*, 2014:353).

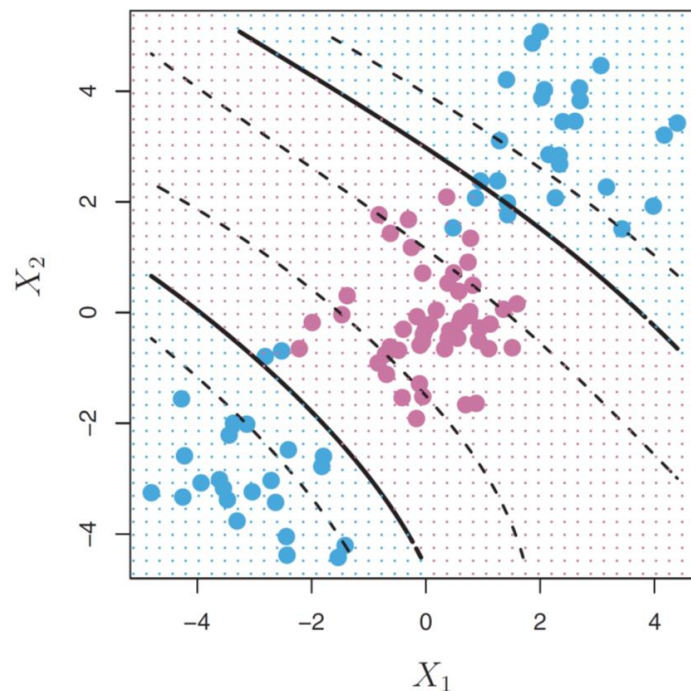


**Figure 5.8: Two Class Polynomial SVM – James *et al.*, 2014:353**

Second-degree polynomial kernels are applied, in the SVM models developed in this section, to produce non-linear decision boundaries. This is since linear boundaries are already present

in Figures 5.3 through 5.7. However, the validation accuracy of the SVM model is paramount. As such, higher degree polynomial and radial kernels are not applied in this study, in order to avoid the development of high variance models that have lower validation accuracy (James *et al.*, 2014:34-35,352-353).

The equations provided in this section for two-dimensional SVM models are defined in terms of mass flow rates. Appendix A.8.2 provides equations for principal component SVM models, which are adapted from the equations in this section. As applied in Equation 5.10, the SVM's margin width,  $Mi$ , must be maximized. This is accomplished by adjusting parameters  $\beta_0$ ,  $\varepsilon_w$ , and  $\alpha_{\varpi}$ , where indices  $w$  and  $\varpi$  define parameters that are specified per observation. A second-degree polynomial kernel, as required in Equation 5.10, is provided in Equation 5.11, and Equation 5.12 defines a constraint for the model's classification errors (James *et al.*, 2014:346,351-352).

$$f_w * \left\{ \beta_0 + \sum_{\varpi=1}^{\omega} [\alpha_{\varpi} * K(\dot{m}_{j,out,\varpi}, \dot{m}_{j,out,w})] \right\} \geq Mi * (1 - \varepsilon_w) \quad [5.10]$$

$$\forall w \in \{1, \dots, \omega\}$$

$$K(\dot{m}_{j,out,\varpi}, \dot{m}_{j,out,w}) = \left[ \delta_0 + \gamma * \sum_{j \in F} (\dot{m}_{j,out,\varpi} * \dot{m}_{j,out,w}) \right]^2 \quad [5.11]$$

$$\sum_{w=1}^{\omega} \varepsilon_w \leq Cs \quad [5.12]$$

here subset  $F$ , of which  $j$  is an element, defines the two mass flow rates specified for use in the SVM model. Training observations  $\varpi$  and  $w$ , for outlet mass flow rate  $j$ , are  $\dot{m}_{j,out,\varpi}$  and  $\dot{m}_{j,out,w}$ , respectively. The parameters  $\beta_0$  and  $\alpha_{\varpi}$  are estimated during model training, where the latter is only non-zero at indices,  $\varpi$ , that correspond with support vectors. Therefore, wherever  $\alpha_{\varpi}$  is not equal to 0,  $\dot{m}_{j,out,\varpi}$  is a support vector. A class specific factor for training observation  $w$  is  $f_w$ . This factor is assigned a value of either -1 or 1, respectively, based on whether an observation is from class 1 or 2. A non-negative margin violation variable,  $\varepsilon_w$ , is included to allow observation  $w$ , from a respective class, to be present across that class's edge of the margin. The kernel function is represented by  $K(\dot{m}_{j,out,\varpi}, \dot{m}_{j,out,w})$ , and additional constants that are specified before the SVM is trained are represented by  $\delta_0$  and  $\gamma$ . The total permissible margin violations, which is specified before model training, is  $Cs$ .

In this study, functions from the e1071 R package, version 1.7-9, will be applied to train SVM models (Meyer *et al.*, 2021). As such, the default values for  $\delta_0$ ,  $\gamma$ , and  $C_s$ , from the e1071 R package, can be specified. These are a value of 0 for  $\delta_0$ , half the number of input parameters for  $\gamma$ , and 1 for  $C_s$  (Meyer *et al.*, 2021). Since the input parameters will be either two flow rates or two principal components,  $\gamma$  has a value of 0.5.

The function for applying a trained two class SVM model, to a given observation, is provided in Equation 5.13. A corresponding second-degree polynomial kernel is defined in Equation 5.14 (James *et al.*, 2014:351-353).

$$f(\dot{m}_{j,out}^*) = \beta_0 + \sum_{\varpi \in SV} [\alpha_{\varpi} * K(\dot{m}_{j,out,\varpi}, \dot{m}_{j,out}^*)] \quad [5.13]$$

$$K(\dot{m}_{j,out,\varpi}, \dot{m}_{j,out}^*) = \left[ \delta_0 + \gamma * \sum_{j \in F} (\dot{m}_{j,out,\varpi} * \dot{m}_{j,out}^*) \right]^2 \quad [5.14]$$

here a new observation,  $\dot{m}_{j,out}^*$ , is classified based on the sign of function  $f(\dot{m}_{j,out}^*)$ , and a subset specifying the support vectors is  $SV$ .

The results of applying a SVM model with a second-degree polynomial kernel, to the training set of predicted  $H_2$  and  $CO$  mass flow rates, are presented in Figure 5.9. This SVM model is trained to classify observations where Model B can and cannot be solved. The confusion matrices for this model are provided in Table 5.8. A 95% confidence ellipse, for the observations where Model B can be solved, is drawn on Figure 5.9 in red. The observations, regions, and boundaries are colored as in Section 5.2.2.1.

**Table 5.8:  $H_2$  and  $CO$  SVM Model Confusion Matrices**

|                               | Training set   |        | Validation set |        |
|-------------------------------|----------------|--------|----------------|--------|
|                               | Does not solve | Solves | Does not solve | Solves |
| <b>Predicted not to solve</b> | 19             | 2      | 13             | 1      |
| <b>Predicted to solve</b>     | 7              | 1,312  | 2              | 534    |

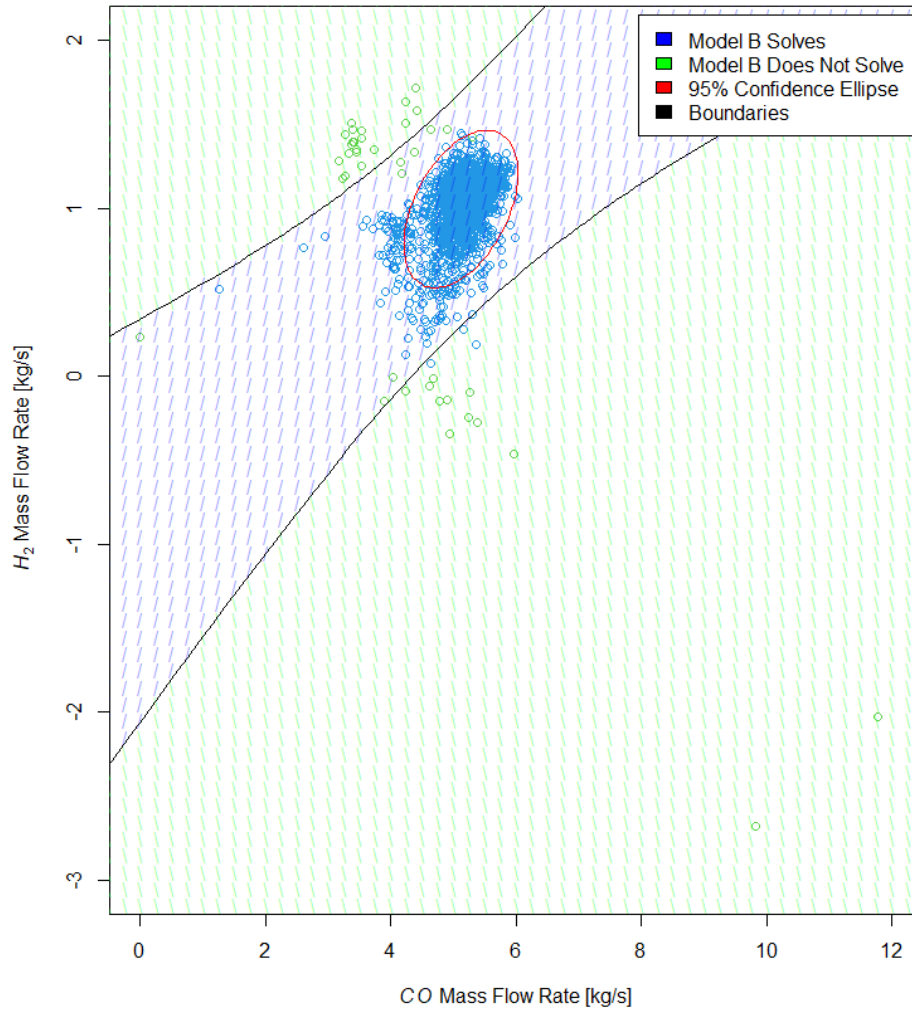


**Figure 5.9:  $H_2$  and CO SVM Model Plot**

From Figure 5.9 and Table 5.8, it is evident that the  $H_2$  and CO SVM model misclassifies a relatively large number of observations, 12 in total. This SVM model's training and validation accuracies, provided in Table 5.13, are 99.3% and 99.5%, respectively. These values represent relatively high accuracy. However, the  $H_2O$  and  $CO_2$  CT model, with a total of 13 misclassifications, is the only CT model with more misclassifications than this SVM model. In addition, this SVM model defined an unbounded region where Model B is predicted to solve. Therefore, this model incorrectly predicts that Model B will solve at CO and  $H_2$  flow rates ranging from minus infinity to infinity. Thus, the results of this SVM model are not generalizable.

The results of applying a SVM model, to the training set of predicted  $CH_4$  and CO mass flow rates, are presented in Figure 5.10. The confusion matrices for this model are provided in Table 5.9.

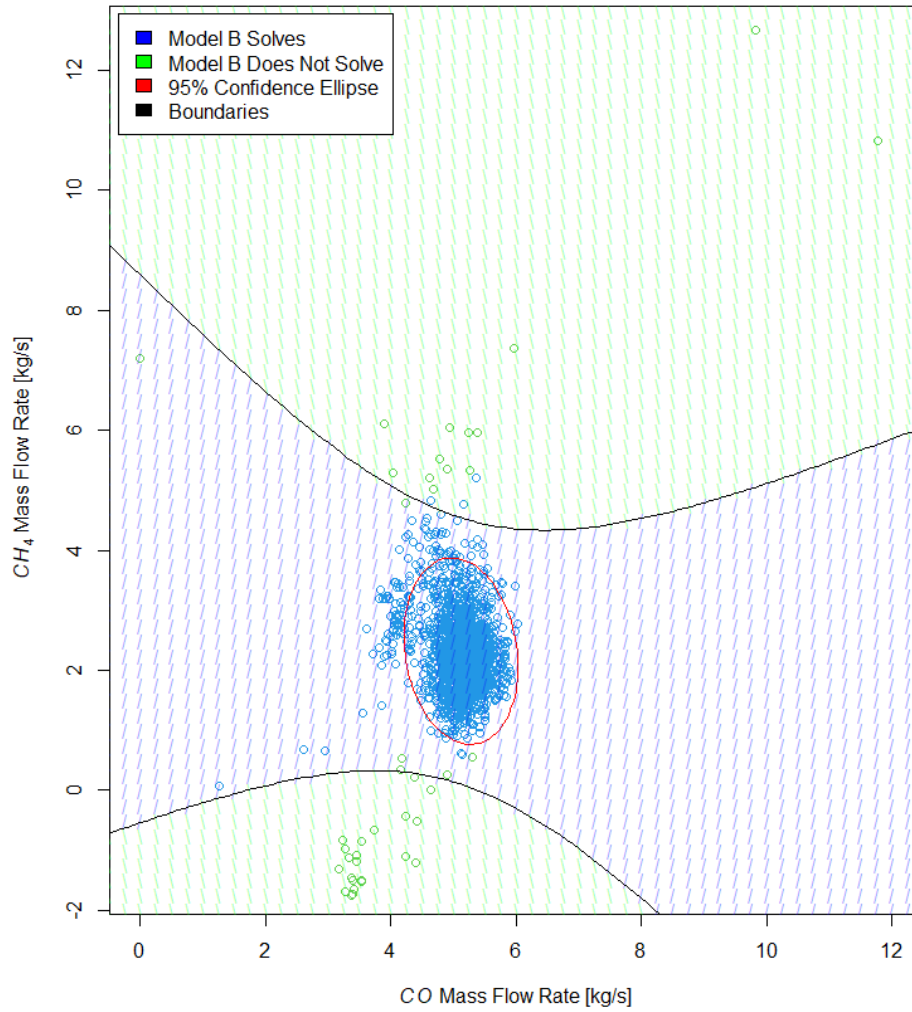


**Figure 5.10:  $CH_4$  and CO SVM Model Plot**

**Table 5.9:  $CH_4$  and CO SVM Model Confusion Matrices**

|                               | Training set   |        | Validation set |        |
|-------------------------------|----------------|--------|----------------|--------|
|                               | Does not solve | Solves | Does not solve | Solves |
| <b>Predicted not to solve</b> | 22             | 2      | 13             | 1      |
| <b>Predicted to solve</b>     | 4              | 1,312  | 2              | 534    |

From Figure 5.10 and Table 5.9, it is evident that the  $CH_4$  and CO SVM model misclassifies relatively few observations, 9 in total. This SVM model's training and validation accuracies,

provided in Table 5.13, are 99.6% and 99.5%, respectively. These values represent relatively high accuracy, where the total number of misclassified observations is equal to that of the  $H_2$  and  $CO$  CT model. In addition, this SVM model defined an unbounded region where Model B is predicted to solve. Therefore, this model incorrectly predicts that Model B will solve at  $CO$  and  $CH_4$  flow rates ranging from minus infinity to infinity. Thus, the results of this SVM model are not generalizable. This is in contrast with the  $CH_4$  and  $CO$ , or equivalent  $CH_4$  and  $CO_2$ , CT model. Therefore, the application of a SVM model to these flow rates, when compared with a CT model, results in overfitting.

The results of applying a SVM model, to the training set of predicted  $CH_4$  and  $CO_2$  mass flow rates, are presented in Figure 5.11. The confusion matrices for this model are provided in Table 5.10.

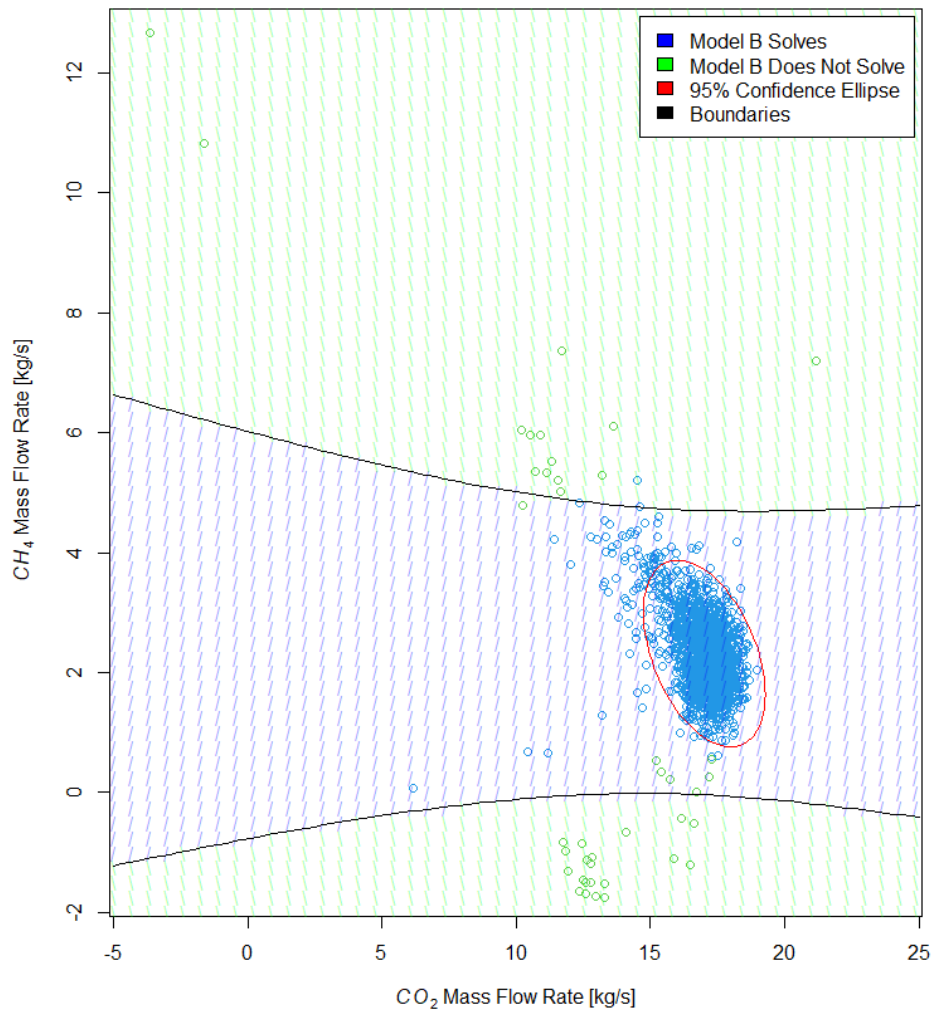


**Figure 5.11:  $CH_4$  and  $CO_2$  SVM Model Plot**

**Table 5.10:  $CH_4$  and  $CO_2$  SVM Model Confusion Matrices**

|                               | Training set   |        | Validation set |        |
|-------------------------------|----------------|--------|----------------|--------|
|                               | Does not solve | Solves | Does not solve | Solves |
| <b>Predicted not to solve</b> | 20             | 1      | 14             | 1      |
| <b>Predicted to solve</b>     | 6              | 1,313  | 1              | 534    |

From Figure 5.11 and Table 5.10, it is evident that the  $CH_4$  and  $CO_2$  SVM model misclassifies relatively few observations, 9 in total. This SVM model's accuracies for the training and validation sets, provided in Table 5.13, are 99.5% and 99.6%, respectively. These values represent high accuracy, where the total number of misclassifications is equal to that for the  $CH_4$  and  $CO$  SVM model. In addition, similarly to the  $CH_4$  and  $CO$  SVM model, the  $CH_4$  and  $CO_2$  SVM model defined an unbounded region where Model B is predicted to solve. Therefore, this SVM model's results are also not generalizable, given that Model B is predicted to solve at  $CH_4$  and  $CO_2$  flow rates ranging from minus infinity to infinity. Thus, it has been established that, when compared with a CT model, the application of a SVM model to the mass flow rates of  $CH_4$  and  $CO$  or  $CH_4$  and  $CO_2$  results in overfitting.

The results of applying a SVM model, to the training set of predicted  $H_2O$  and  $CO_2$  mass flow rates, are presented in Figure 5.12. The confusion matrices for this model are provided in Table 5.11.

**Table 5.11:  $H_2O$  and  $CO_2$  SVM Model Confusion Matrices**

|                               | Training set   |        | Validation set |        |
|-------------------------------|----------------|--------|----------------|--------|
|                               | Does not solve | Solves | Does not solve | Solves |
| <b>Predicted not to solve</b> | 20             | 2      | 11             | 1      |
| <b>Predicted to solve</b>     | 6              | 1,312  | 4              | 534    |

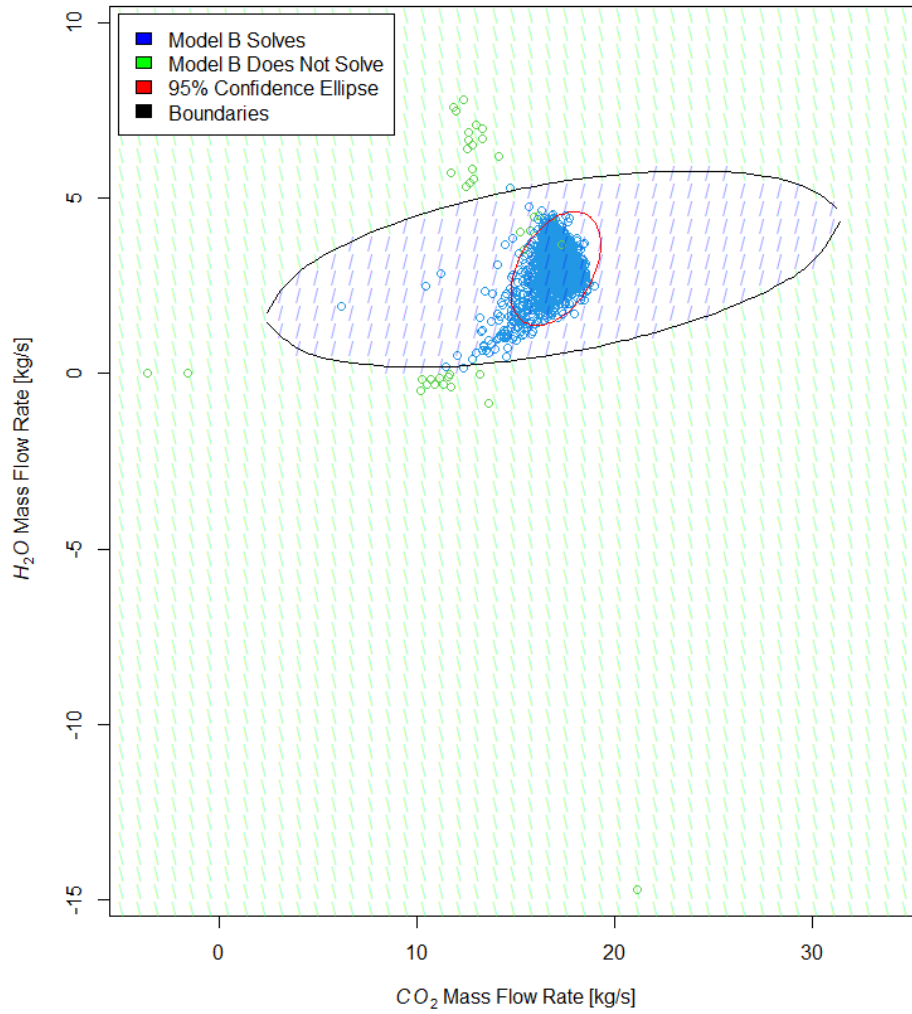


**Figure 5.12:  $H_2O$  and  $CO_2$  SVM Model Plot**

From Figure 5.12 and Table 5.11, it is evident that the SVM model for  $H_2O$  and  $CO_2$  misclassifies a relatively large number of observations, 13 in total. This SVM model's training and validation accuracies, provided in Table 5.13, are 99.4% and 99.1%, respectively. Although these values represent relatively high accuracy, the total number of misclassifications is equal to that of the  $H_2O$  and  $CO_2$  CT model. Therefore, these  $H_2O$  and  $CO_2$  models, when compared with the previous CT and SVM models, have the largest number of misclassifications. However, from Tables 5.5 and 5.11, the  $H_2O$  and  $CO_2$  SVM model's classification errors are more balanced between classes than those of the  $H_2O$  and  $CO_2$  CT model. In addition, it is evident from Figure 5.12 that the  $H_2O$  and  $CO_2$  SVM model defined a bounded region where Model B is predicted to solve. Therefore, the results of this SVM model may be generalizable.

Figure 5.13 presents the results of applying a SVM model to the training set of the first two principal component scores. The confusion matrices for this model are provided in Table 5.12.

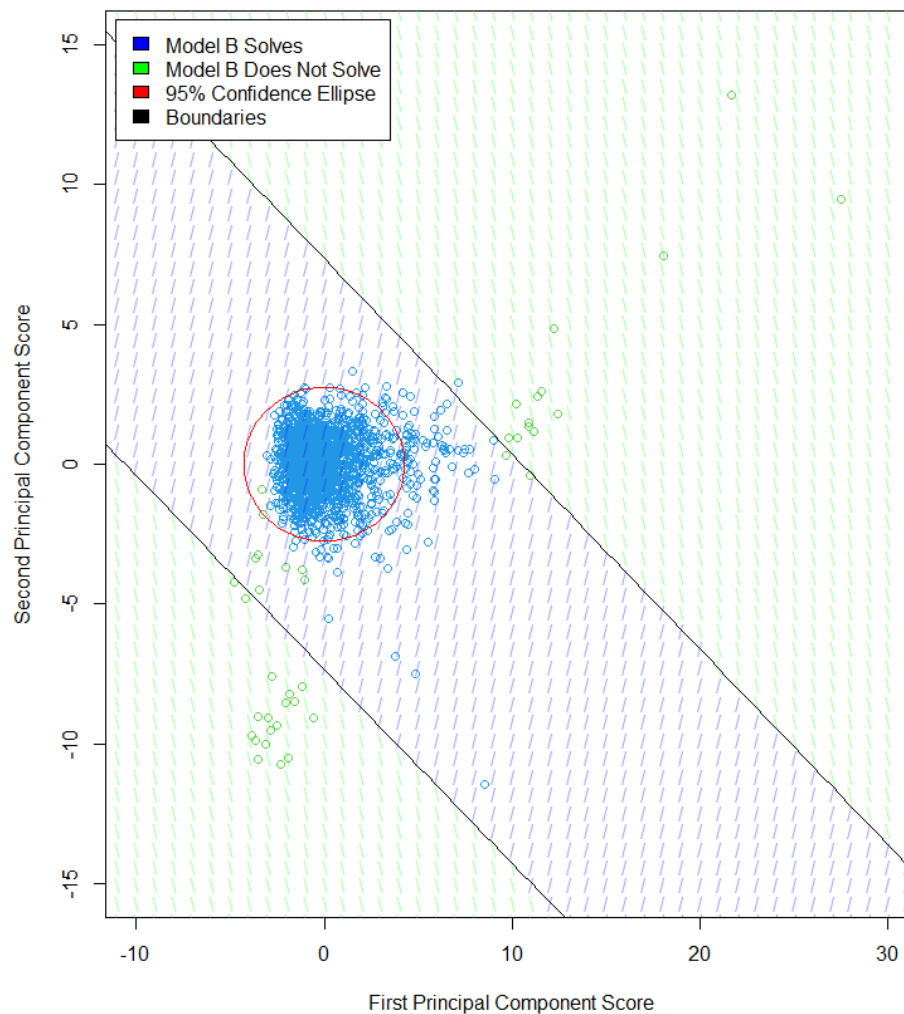


**Figure 5.13: Principal Component SVM Model Plot**

**Table 5.12: Principal Component SVM Model Confusion Matrices**

|                               | Training set   |        | Validation set |        |
|-------------------------------|----------------|--------|----------------|--------|
|                               | Does not solve | Solves | Does not solve | Solves |
| <b>Predicted not to solve</b> | 18             | 1      | 13             | 0      |
| <b>Predicted to solve</b>     | 8              | 1,313  | 2              | 535    |

From Figure 5.13 and Table 5.12, it is evident that the principal component SVM model misclassifies a relatively large number of observations, 11 in total. This SVM model's accuracies for the training and validation sets, provided in Table 5.13, are 99.3% and 99.6%, respectively. These values represent relatively high accuracy. However, the region defined by the principal component SVM model, where Model B is predicted to solve, is unbounded. Therefore, this SVM model incorrectly predicts that Model B will solve at scores, for the first two principal components, ranging from minus infinity to infinity. Thus, the results of this SVM model are not generalizable. From the SVM models, only the  $CH_4$  and  $CO$  and the  $CH_4$  and  $CO_2$  models have fewer misclassifications, 9 in total, than the principal component model, 11 in total. However, the misclassifications for the principal component CT model, 5 in total, are considerably fewer than those for the principal component SVM model.

It is noteworthy that, when compared with the respective SVM models, the CT models consistently had fewer total misclassifications. This was not true for the  $H_2O$  and  $CO_2$  models alone, with equal totals. In addition, only two classification models defined bounded regions where Model B is predicted to solve. These are the  $CH_4$  and  $CO$ , or equivalent  $CH_4$  and  $CO_2$ , CT model and the  $H_2O$  and  $CO_2$  SVM model. As such, these models will be compared in the following section, and one will be selected for further investigation. The accuracies, non-detection rates, and false alarm rates for the SVM models are provided in Table 5.13.

**Table 5.13: Accuracy and Error Metrics for SVM Models**

|                                              | Training set | Validation set |
|----------------------------------------------|--------------|----------------|
| <b><math>H_2</math> and <math>CO</math></b>  |              |                |
| <b>Accuracy</b>                              | 99.3%        | 99.5%          |
| <b>Non-detection rate</b>                    | 26.9%        | 13.3%          |
| <b>False-alarm rate</b>                      | 0.152%       | 0.187%         |
| <b><math>CH_4</math> and <math>CO</math></b> |              |                |
| <b>Accuracy</b>                              | 99.6%        | 99.5%          |
| <b>Non-detection rate</b>                    | 15.4%        | 13.3%          |
| <b>False-alarm rate</b>                      | 0.152%       | 0.187%         |

|                                                        | Training set | Validation set |
|--------------------------------------------------------|--------------|----------------|
| <b><i>CH<sub>4</sub></i> and <i>CO<sub>2</sub></i></b> |              |                |
| <b>Accuracy</b>                                        | 99.5%        | 99.6%          |
| <b>Non-detection rate</b>                              | 23.1%        | 6.67%          |
| <b>False-alarm rate</b>                                | 0.0761%      | 0.187%         |
| <b><i>H<sub>2</sub>O</i> and <i>CO<sub>2</sub></i></b> |              |                |
| <b>Accuracy</b>                                        | 99.4%        | 99.1%          |
| <b>Non-detection rate</b>                              | 23.1%        | 26.7%          |
| <b>False-alarm rate</b>                                | 0.152%       | 0.187%         |
| <b>Principal components</b>                            |              |                |
| <b>Accuracy</b>                                        | 99.3%        | 99.6%          |
| <b>Non-detection rate</b>                              | 30.8%        | 13.3%          |
| <b>False-alarm rate</b>                                | 0.0761%      | 0%             |

### 5.2.3 Model Selection

In this section, from the statistical classification models, which have been developed and discussed in Section 5.2.2, one is selected for further investigation. Firstly, the selected classification model must possess results that can be generalized. As such, the region where this classification model predicts that Model B is solvable must be bounded. This ensures that the selected model does not incorrectly predict that Model B is solvable at values approaching infinity. Therefore, as previously stated, selection will be made between the *CH<sub>4</sub>* and *CO*, or equivalent *CH<sub>4</sub>* and *CO<sub>2</sub>*, CT model and the *H<sub>2</sub>O* and *CO<sub>2</sub>* SVM model.

From Table 5.3, the *CH<sub>4</sub>* and *CO* CT model misclassified 1 validation and 3 training observations where Model B is solvable. Similarly, from Table 5.11, the *H<sub>2</sub>O* and *CO<sub>2</sub>* SVM model misclassified 1 validation and 2 training observations where Model B is solvable. In addition, this SVM model also misclassified 4 validation and 6 training observations where

Model B is unsolvable. However, the  $CH_4$  and  $CO$  CT model misclassified 0 validation and training observations where Model B is unsolvable. Therefore, these SVM and CT models misclassify essentially the same number of solvable observations, but the SVM model misclassifies more unsolvable observations.

As previously stated, predictions made by the  $CH_4$  and  $CO$ , or equivalent  $CH_4$  and  $CO_2$ , CT model are only based on  $CH_4$  mass flow rates. Therefore, this is a one-dimensional model that is applied to a classification problem in a five-dimensional space. However, as evident from Figure 5.1, the  $H_2O$  and  $CO_2$  mass flow rates are two uncorrelated dimensions that explain a significant portion of the variability in this five-dimensional space. Thus, the  $H_2O$  and  $CO_2$  SVM model predicts a more generalizable region where Model B is solvable, in this five-dimensional space, than the  $CH_4$  and  $CO$ , or equivalent  $CH_4$  and  $CO_2$ , CT model. This is since, from Figures 5.4 and 5.5, it is evident that variations in the flow rates of  $CO$  and  $CO_2$  do affect changes in the distribution of solvable and unsolvable observations.

Therefore, the one-dimensional  $CH_4$  and  $CO$  CT model, although highly accurate for the data set that it was trained on, is likely to possess poor accuracy when applied to new data sets. As such, the  $H_2O$  and  $CO_2$  SVM model is selected for further investigation, based on its more representative and generalizable prediction of the region, within this five-dimensional space, where Model B is solvable.

### 5.3 Interpretation and Validation

In this section, the  $H_2O$  and  $CO_2$  SVM classification model will be investigated, to determine the validity of the thermal-fluid model developed in this study. The relationship between the inlet flow rates, which govern the gasification process, and the results of the statistical model will also be investigated. This is to ascertain whether the trends predicted by the model can be attributed to process faults.

The thermal-fluid model will be validated by determining whether the causal relationships are correctly predicted for the gasification process. Specifically, the predicted trends as a function of changes in the input variables must conform with established fundamental knowledge of the coal gasification process.

Changes, due to variations in the mass flow rates of steam and coal, will be investigated for the model's input variables. These flow rates are the sole sources of carbon and hydrogen in the gasifier. In addition, as discussed in Section 4.6.2, steam accounts for over 75% of the total moles of oxygen in the gasifier. As such, variations in the flow rate of oxygen will not be

directly included in this analysis. Furthermore, it will be assumed that the steam to oxygen ratio is essentially kept constant during gasification.

To investigate the effects of steam and coal flow rate variations, a statistical technique referred to as k-means clustering will be applied. This technique is aimed at revealing trends present in data, by dividing observations into a predefined number of distinct groups or clusters (James *et al.*, 2014:385-386). K-means clustering functions such that observations are, respectively, assigned to a cluster with the smallest squared Euclidean distance between the observations and the cluster's center (James *et al.*, 2014:387-388).

K-means clustering will be applied separately to the inlet flow rates of steam and coal, and two clusters will be defined per variable. As such, both flow rates are divided into clusters that can be designated high and low, respectively. These designations are specified, per flow rate, given the relative magnitude of each cluster's centroid. Based on where the steam and coal clusters intersect, observations can be divided into four respective groups. This allows the thermal-fluid model's results to be analyzed for a set of four inlet flow rate conditions.

The high steam cluster is centered around the respective reference inlet flow rate (Bunt, 2006:54). However, the reference inlet flow rate for coal is centered between the respective high and low clusters (Bunt, 2006:54). As such, the reference syngas volume percentages, provided in Table 4.1, can be defined as respective outputs for a high steam and an average coal inlet condition.

Figure 5.14 presents the clusters in a plot for the predicted  $H_2O$  and  $CO_2$  mass flow rates. The boundaries defined by the  $H_2O$  and  $CO_2$  SVM model are overlaid, in black, on Figure 5.14. Additional plots, with these clusters, are provided in Appendix D.

In Figure 5.14, observations are colored to differentiate between respective inlet flow rate groups. The high steam and high coal group is colored green, and the low steam and low coal group is colored pink. The high steam and low coal group is colored dark blue, and the low steam and high coal group is colored light blue. A 95% confidence ellipse is also provided on the figure in red, for the observations where Model B is solvable.

It is evident, from Figure 5.14, that the high steam and high coal group mainly constitutes the observations within the 95% confidence ellipse. This group is also largely centered within the region, defined by the  $H_2O$  and  $CO_2$  SVM, where Model B is predicted to be solvable. However, the high steam and low coal group is mainly present outside this region. Thus,

Model B is solvable for essentially the entire high steam and high coal group, but it is effectively unsolvable for the high steam and low coal group.

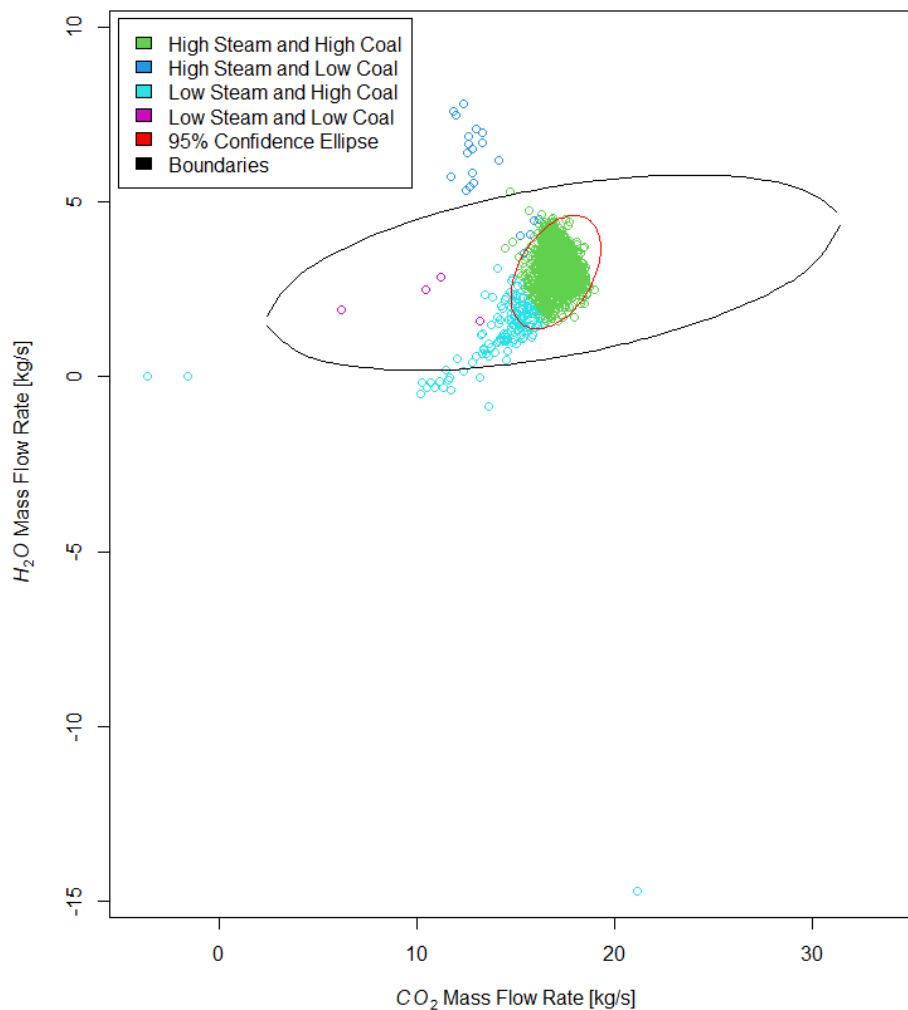


**Figure 5.14:  $H_2O$  and  $CO_2$  SVM Model Plot with Inlet Flow Rate Groups**

In addition, from Figure 5.14, it is apparent that the low steam and low coal group is entirely within the region where Model B is predicted to be solvable. The low steam and high coal group is, however, present in both predicted, solvable and unsolvable, regions for Model B. Furthermore, this group contains the predicted flow rates that are the most remote from the center.

Table 5.14 provides the minimum, maximum, and average syngas volume percentages, for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$ , predicted by Model A for each group. Thus, these values define the thermal-fluid model's outputs, for specific inlet flow rate conditions. As such, the differences between these must be analyzed, with respect to inlet flow rate conditions.

**Table 5.14: Syngas Volume Percentages per Inlet Flow Rate Group**

|                          | Minimum             | Maximum | Average | Minimum             | Maximum | Average |
|--------------------------|---------------------|---------|---------|---------------------|---------|---------|
| Group                    | $H_2$ [%]           |         |         | CO [%]              |         |         |
| High steam and high coal | 24.3                | 57.2    | 41.7    | 11.0                | 17.6    | 15.0    |
| High steam and low coal  | 53.3                | 70.6    | 64.9    | 11.4                | 13.2    | 12.3    |
| Low steam and high coal  | -2,060              | 484     | 12.3    | -127                | 859     | 22.5    |
| Low steam and low coal   | 46.8                | 57.6    | 51.4    | 10.1                | 13.4    | 12.2    |
| Group                    | CO <sub>2</sub> [%] |         |         | CH <sub>4</sub> [%] |         |         |
| High steam and high coal | 29.6                | 36.1    | 31.8    | 0.0659              | 24.2    | 11.4    |
| High steam and low coal  | 28.2                | 30.5    | 28.9    | -10.4               | 2.92    | -6.07   |
| Low steam and high coal  | -74.6               | 45.9    | 33.2    | -286                | 1,370   | 32.0    |
| Low steam and low coal   | 31.1                | 31.5    | 31.3    | 0.938               | 8.39    | 5.01    |

As previously stated, the syngas volume percentages in Table 4.1 are respective outputs for a high steam and an average coal inlet condition. However, it is more beneficial to apply the syngas volume percentages, predicted with Model B for the gasifier measurements in Table 4.1, as reference outputs for this condition. These values are available in Section 4.6.1.4 and can be more efficiently compared with those in Table 5.14. This is since these values are all predictions generated by the thermal-fluid model. As such, the thermal-fluid model's

inaccuracy, discussed in Section 4.5, can be accounted for by allowing direct comparison with a reference set of outputs for the model.

Comparing the syngas volume percentages, in Table 5.14, with the model's reference outputs, the following is evident. Only two groups exist where all four reference volume percentages are between the respective minimum and maximum values. These are the high steam and high coal group and the low steam and high coal group. Furthermore, the averages in the former group, per volume percentage, are closest to the reference values.

For the high steam and high coal group, the average volume percentages for  $CO$  and  $CO_2$  are essentially identical to the reference values. However, the average percentages for  $H_2$  and  $CH_4$  are, respectively, lower and higher than the reference values. These variations are explainable, given that the inlet coal flow rate, for this group, is higher than the reference. The greater supply of carbon inlet to the gasifier must result in higher flow rates of  $CO$ ,  $CO_2$ , and  $CH_4$ . According to the equilibrium condition for the water gas shift reaction, an increase in  $CO_2$  can be balanced with a corresponding increase in  $CO$ . Thus, the average percentages of  $CO$  and  $CO_2$  can remain constant, even for process variations leading to higher respective flow rates.

However, the higher flow rate of  $CH_4$  must be balanced with a decrease in  $H_2$  and  $H_2O$ . This is since the flow rate of steam, the supply of hydrogen, does not change. As such, the hydrogen present in this additional  $CH_4$  must result from a decrease in the flow rates of  $H_2$  and  $H_2O$ . Given that there is more available oxygen, from the decrease in  $H_2O$ , the assumed increase in  $CO$  and  $CO_2$  is reasonable. This is since additional oxygen is required to react with the extra carbon, and the flow rate of oxygen, inlet to the gasifier, has not increased. Under these conditions, corresponding decreases in  $H_2$  and  $H_2O$  are balanced, according to the equilibrium for the water gas shift reaction. From these results, the differences evident between the thermal-fluid model's predictions, for this group and the reference conditions, correspond with an understanding of the model's fundamental equations. Therefore, these predicted variations are valid.

Furthermore, the high steam and high coal group can be concluded as representing normal gasifier operations. This is evident from the location of this group within the center of the region, defined by the selected statistical model, where Model B is predicted to be solvable. It is also supported by the fact that this group mainly comprises the observations within the 95% confidence ellipse. Furthermore, the average volume percentages for this group are the closest to the reference outputs for the thermal-fluid model. The model, as discussed in Section 4.5, does produce accurate predictions for these reference gasifier measurements.

Therefore, the high steam and high coal group is the most clearly defined set of normal operating conditions, within this data.

For the high steam and low coal group, a larger average volume percentage for  $H_2$ , than the reference value, is predicted. In addition, slightly lower average percentages for  $CO$  and  $CO_2$  and a negative average for  $CH_4$  are predicted. This negative average provides an explanation for the fact that, at the observations in this group, Model B is largely unsolvable. These variations are explainable, given that the inlet coal flow rate, for this group, is lower than the reference. The lower supply of carbon must result in decreased flow rates of  $CO$ ,  $CO_2$ , and  $CH_4$ . The equilibrium condition for the water gas shift reaction allows corresponding decreases in  $CO$  and  $CO_2$ , which result in more available oxygen. As such, the flow rate of  $H_2O$  must increase. Under these conditions, this must be balanced with an increase in  $H_2$ , according to equilibrium for the water gas shift reaction. As such, there is less hydrogen available to react with carbon to form  $CH_4$ . Although negative flow rates are impossible realistically, this does explain the model's predicted negative average percentage for  $CH_4$ . As such, the differences predicted between this group and the reference conditions correspond with an understanding of the thermal-fluid model's fundamental equations.

In addition to Figure 5.14, figures in Appendix D provide evidence of the flow rate changes hypothesized to occur for variations in the coal flow rate. These are Figures D.1 through D.3. A qualitative summary of the changes per flow rate, for decreases in coal, is provided in Table D.1.

Although the average volume percentage for  $CH_4$ , for the high steam and low coal group, is negative, the maximum percentage is a small positive value. This indicates that there are some observations, within this group, that more closely resemble normal gasifier operations. However, given these results, it is reasonable to judge that most of the observations in this group represent abnormal operating conditions. This is since it is evident that, at these observations, the conditions do not comply with the model's underlying assumption of normal gasifier operations. There is, however, insufficient evidence to prove that these observations represent a specific gasifier fault.

For the low steam and high coal group, comparisons will not be made against the reference inlet conditions. This is since the reference conditions are high steam and average coal. As such, it is more valuable to draw comparisons against the high steam and high coal group. When compared, a considerably smaller average volume percentage for  $H_2$  is predicted for the low steam and high coal group. A slightly larger average percentage for  $CO_2$  is predicted, and significantly larger average percentages are predicted for  $CO$  and  $CH_4$ . As previously

stated, the furthest outlying observations are present in this group. This is evident in the minimum and maximum values for this group, per volume percentage, which have the largest respective magnitudes. Given the reduced inlet flow rate of steam, these variations are explainable. This reduced flow rate results in lower supplied quantities of hydrogen and oxygen. The obvious result is a lower outlet flow rate of  $H_2O$ . However, the explanation, for the corresponding changes that occur in the flow rates of  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$ , is more involved.

This analysis begins with a supposition that the total flow rate of  $CO$  and  $CO_2$  will be lower than for the high steam and high coal group. This is due to the lower flow rate of oxygen, since, as previously stated, the inlet steam to oxygen ratio is assumed to be relatively constant. The implication of this is that less combustion can occur. In addition, it implies that combustion will be incomplete, the result of which is a higher  $CO$  to  $CO_2$  ratio, than is predicted for the high steam and high coal group. As such, the average percentage of  $CO$  can increase significantly, while the average percentage of  $CO_2$  increases slightly. The reduction in hydrogen must result in a decreased flow rate of  $H_2$ . This corresponds to the water gas shift reaction, where less  $H_2O$  is available to react with  $CO$  to form  $H_2$  and  $CO_2$ . This supports the previous statement that there will be a larger  $CO$  to  $CO_2$  ratio.

The lower total flow rate of  $CO$  and  $CO_2$  must result in a larger flow rate of  $CH_4$ . This is since the inlet flow rate of coal has not been varied. Thus, the total quantity of carbon remains constant, although less reacts to form  $CO$  and  $CO_2$ . As such, the additional carbon must react with hydrogen to produce  $CH_4$ . This explains the significant increase in the average percentage of  $CH_4$  and results in a further reduction in  $H_2$ . Thus, the relatively large reduction in the average percentage of  $H_2$  can be rationalized, and the initial supposition is valid. As such, the differences predicted between this and the high steam and high coal group correspond with an understanding of the thermal-fluid model's fundamental equations.

Figures 5.14, D.1, D.2, and D.3 confirm the flow rate changes hypothesized to occur for a decrease in steam. A qualitative summary of these changes per flow rate is provided in Table D.2. From the hypothesized changes, the higher  $CO$  to  $CO_2$  ratio is evident from Figures D.2 and D.3. This is since, when compared to  $CO$  flow rates, a larger decrease in  $CO_2$  is present between the high steam and high coal and the low steam and high coal groups.

The low steam and high coal group contains observations that are present on both sides of the boundary defined by the SVM. Thus, this group represents a transition from normal to abnormal gasifier operations. In addition, this group's maximum percentage for  $CO_2$  is above 40%, while the maximum percentages for the other groups are below this limit. In Section

2.2.5, it was noted that raw syngas  $\text{CO}_2$  volume percentages of more than 40% are indicative of channel burning. Thus, it is possible that this fault is present in the low steam and high coal group, although this has not been proven. Some observations within this group are representative of normal gasifier operations, specifically those near the center of the region defined by the SVM. However, there are observations, which are near the region's boundary, that must be defined as abnormal.

The low steam and low coal group is most readily compared with the low steam and high coal group. From the discussion regarding the differences between the high steam and low coal group and the reference inlet conditions, the following is expected. The average percentage for  $\text{H}_2$  should be higher, and the average percentages for  $\text{CO}$ ,  $\text{CO}_2$ , and  $\text{CH}_4$  should be lower. The average percentage for  $\text{H}_2$  is significantly larger, the average percentages for  $\text{CO}$  and  $\text{CH}_4$  are considerably smaller, and the average percentage for  $\text{CO}_2$  is slightly lower. Therefore, the differences predicted, for variations in the inlet flow rate of coal, correspond between the low and high steam groups. This indicates that the thermal-fluid model develops predictions that are consistent, within the context of its fundamental equations.

Although more involved, the low steam and low coal group must also be compared with the high steam and low coal group. From the discussion regarding the differences between the low steam and high coal and the high steam and high coal groups, the following is expected. The average percentage for  $\text{H}_2$  should be smaller, and the average percentages for  $\text{CO}$ ,  $\text{CO}_2$ , and  $\text{CH}_4$  should be larger. When comparing the low steam and low coal and the high steam and low coal groups, this prediction is accurate for the  $\text{H}_2$ ,  $\text{CO}_2$ , and  $\text{CH}_4$  average volume percentages. The average percentage for  $\text{H}_2$  is slightly smaller, the average percentage for  $\text{CO}_2$  is slightly larger, and the average percentage for  $\text{CH}_4$  is significantly larger. However, the average volume percentage for  $\text{CO}$  is essentially identical to that from the high steam and low coal group. This deviation is explainable, since the difference in average percentages for  $\text{H}_2$ , between the low steam and high coal and the high steam and high coal groups, was considerably larger. However, the differences for  $\text{CO}_2$  and  $\text{CH}_4$ , between respective groups, are of similar magnitudes. As such, according to the water gas shift reaction, this smaller deviation between the average percentages for  $\text{H}_2$  cannot result in a significant difference between the average percentages for  $\text{CO}$ . Therefore, it has been demonstrated that the predictions are consistent with both the underlying thermodynamics of the coal gasifier and the model's fundamental equations.

Given that Model B is solvable for the low steam and low coal group, this is unlikely to represent abnormal operations. This is evident from the average syngas volume percentages in this group. These values are, after the high steam and high coal group, the closest to the

predicted values for reference inlet conditions. Thus, this group is likely to only represent a low load condition and not a gasifier fault.

It can now be judged whether the hypothesis that unsolvable observations represent abnormal operating conditions is invalid. According to the results of this section, it has been demonstrated that the outlying observations, where Model B is unsolvable, are indicative of abnormal operating conditions. There are, however, observations where the unsolvability of Model B cannot be attributed to abnormal operations. These are the ten unsolvable observations present within the solvable region of Figure 5.12. However, from a visual comparison of Figures 5.12 and 5.14, it is evident that five of these observations are present in the high steam and low coal group. As such, these are present in a group that has been indicated as mostly comprised of abnormal observations. Thus, there are five remaining observations, present in the high steam and high coal group, where it can be definitively stated that the unsolvability of Model B is not due to abnormal operating conditions. This is in contrast with the 31 unsolvable and outlying observations, which are present in the high steam and low coal and the low steam and high coal groups, that indicate abnormal operations according to the results of this section. Therefore, while the hypothesis is not indefectible or unassailable, it is sufficient for the purposes of statistical modeling in this study and cannot be rejected.

#### **5.4 Comparison with Conventional Process Monitoring**

In this section, the  $H_2O$  and  $CO_2$  SVM model is compared with conventional process monitoring for coal gasifiers. As mentioned in Section 5.2.1, observations can be designated normal or abnormal based solely on the location of these, relative to the boundaries of a confidence ellipse. This form of process monitoring is acceptable for fixed-bed dry bottom coal gasifiers (Coetzer *et al.*, 2014:61). Therefore, the difference between the  $H_2O$  and  $CO_2$  SVM model and this form of process monitoring is discussed in this section.

The  $H_2O$  and  $CO_2$  SVM model is applied to predict abnormal operating conditions as follows. A gasifier observation is input to thermal-fluid Model A, and the predicted mass flow rates of  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are output. From these, the  $H_2O$  and  $CO_2$  flow rates are input to the SVM model and are overlayed on a plot of the SVM model's decision boundary. The observation is predicted to be normal or abnormal by the SVM model, but the plot is used to judge the degree of normality or abnormality. This is since observations that are predicted to be normal may be near the center or the edge of the region defined by the SVM model. In addition, it must be judged whether observations near the edge of the SVM model's decision boundary are only suboptimal or indicate potential faults.

Conventional process monitoring with a confidence ellipse is applied to predict abnormal operating conditions as follows. A gasifier observation is input to thermal-fluid Model A, and the predicted mass flow rates of  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are output. Principal component scores, calculated from these flow rates, are overlayed on a plot of the respective confidence ellipse. Thus, the observation is predicted to be normal or abnormal based on its location inside or outside the principal component score confidence ellipse, respectively (Coetzer *et al.*, 2014:60-61).

It is evident that abnormal gasifier operating conditions are mainly present for observations outside the region defined by the  $H_2O$  and  $CO_2$  SVM model. This is given the discussion, in Section 5.3, regarding the results of this SVM model. Thus, between this model and the presented form of process monitoring, a difference is evident in an apparent misclassification of specific observations. In Figure 5.12, these observations are located outside the 95% confidence ellipse but are present within the region defined by the  $H_2O$  and  $CO_2$  SVM model. As such, whenever the gasification process is not abnormal but only suboptimal, observations are misclassified by this conventional form of process monitoring. Here, the term suboptimal is applied to refer to the low steam and low coal group and specific observations from the two high coal groups.

Confusion matrices, for this conventional form of process monitoring, are provided in Table 5.15. Here, 95% confidence ellipses are defined, respectively, for the first two principal components and the flow rates of  $H_2O$  and  $CO_2$ . The former is selected since coal gasifiers can be monitored, from a reference set of measurements, with a confidence ellipse for the first two principal components (Coetzer *et al.*, 2014:60-61). However, comparisons should be drawn against a confidence ellipse for the mass flow rates from the selected statistical model. These confidence ellipses are developed for the observations where Model B is solvable. This is since, according to the discussion in Section 5.3, these observations largely represent normal gasifier operations.

Model B is predicted to solve at observations within the 95% confidence ellipse for the conventional form of process monitoring. As such, the non-detection rate defines the percentage of observations where Model B solves, which are present outside the ellipse. In addition, the false-alarm rate defines the percentage of observations where Model B does not solve, which are present within the ellipse. The accuracies, the non-detection rates, and the false alarm rates for the 95% confidence ellipses are provided in Table 5.16.

**Table 5.15: Conventional Process Monitoring Confusion Matrices**

|                               | Principal components |        | $H_2O$ and $CO_2$ |        |
|-------------------------------|----------------------|--------|-------------------|--------|
|                               | Does not solve       | Solves | Does not solve    | Solves |
| <b>Predicted not to solve</b> | 39                   | 103    | 36                | 83     |
| <b>Predicted to solve</b>     | 2                    | 1,746  | 5                 | 1,766  |

**Table 5.16: Accuracy and Error Metrics for Conventional Process Monitoring**

|                           | Principal components | $H_2O$ and $CO_2$ |
|---------------------------|----------------------|-------------------|
| <b>Accuracy</b>           | 94.4%                | 95.3%             |
| <b>Non-detection rate</b> | 4.88%                | 12.2%             |
| <b>False-alarm rate</b>   | 5.57%                | 4.49%             |

From Table 5.15, it is evident that the confidence ellipse for  $H_2O$  and  $CO_2$  misclassifies fewer observations, 88 in total, than the ellipse for the first two principal components, 105 in total. In addition, in Table 5.16, the non-detection and false alarm rates are comparable in magnitude. This is unlike the non-detection and false alarm rates provided in Tables 5.7 and 5.13. The difference is due to the significantly larger number of observations where Model B solves, which are predicted to be unsolvable according to this form of conventional process monitoring.

From Table 5.16, the false-alarm rate for the 95% confidence ellipse on the first two principal components is approximately 5%. This confirms the use of a 95% confidence ellipse, since approximately 5% of the normal observations should be identified as abnormal operating conditions.

In addition, it is observed that the conventional process monitoring approach results in reduced non-detection and increased false-alarm rates, when compared to the  $H_2O$  and  $CO_2$  SVM model. From Table 5.16, the accuracies of the principal component and the  $H_2O$  and  $CO_2$  confidence ellipses are 94.4% and 95.3%, respectively. Therefore, these values are lower

than both the training and validation accuracies of the  $H_2O$  and  $CO_2$  SVM model, provided in Table 5.13 as 99.4% and 99.1%, respectively. This is since, from Table 5.11, it is evident that the SVM model developed in this study only misclassifies 13 observations in total. Thus, this total is considerably smaller than the 105 and 88 observations, from Table 5.15, misclassified by the principal component and the  $H_2O$  and  $CO_2$  confidence ellipses, respectively. As such, the approach for fixed-bed dry bottom coal gasifier fault prediction, followed in this study, is a valid alternative to the conventional process monitoring methodology.

## 5.5 Conclusions

In this chapter, statistical techniques were applied, to the predicted gasifier outlet mass flow rates, to ascertain whether or not faults could be isolated from within the data. Flow rates were predicted with Model A, and Model B was applied to determine the location of observations where the thermal-fluid model's underlying assumptions are not satisfied. Primary among these assumptions is the existence of five mass flow rates, as are present during normal gasifier operations. Where the assumptions are not satisfied, Model B is unsolvable.

Principal component analysis was applied to determine the least correlated pairs of flow rates. These pairs, and the first two principal components, were utilized in classification models that were developed to predict where Model B is unsolvable. For this purpose, CTs and SVMs were introduced and applied. The accuracies of the developed classification models were ascertained, and a model was selected for further investigation. The selected model is a second-degree polynomial kernel SVM for the predicted mass flow rates of  $H_2O$  and  $CO_2$ . This SVM is the only model which produced a fully bounded two-dimensional region where Model B is solvable.

A notable observation made, during the preliminary work undertaken for this chapter, was that increasing the number of flow rates utilized in the classification models resulted in reduced accuracy and increased complexity. It also became evident, from the PCA biplot, that the least accurate models utilized highly correlated flow rates. As such, the least correlated pairs of flow rates were selected, from the PCA biplot, for use in the classification models presented in this chapter.

Classification models, which predicted unbounded regions, were rejected with the purpose of developing a statistical model that predicts normal operating conditions more accurately than the criterion of thermal-fluid model solvability. This is since a bounded region provides a more accurate representation of the model's solvability, and normal gasifier operating conditions,

than an unbounded region. A bounded region will classify observations with outlying flow rates as abnormal, while ensuring that flow rates near the region's center are classified as normal. It should be noted that Model A will never predict extremely large positive or negative flow rates, since even this model would fail to solve at extreme outlying observations. In addition, an increase in one flow rate will result in corresponding decreases in the other flow rates, as evidenced in Section 5.3. Therefore, with two-dimensional statistical classification models for the five-dimensional flow rate space, it is paramount that there be ultimate upper and lower limits on the values for both axes. A lower limit indicates where the presented flow rate approaches negative values. An upper limit indicates where at least one flow rate from the remaining three dimensions approaches negative values, from decreases corresponding to the respective increase in the given flow rate. Therefore, lower and upper limits allow the higher dimensionality of this classification problem to be accounted for in a two-dimensional space.

The results of the selected model were investigated with the aid of k-means clustering. This allowed four groups to be isolated and analyzed with respect to the inlet steam and coal flow rate clusters. An extensive discussion was provided, which was aimed at explaining the syngas volume percentage variations present between groups. The reasoning applied in this discussion was consistent with established knowledge regarding the thermal-fluid model's fundamental equations and the coal gasification process.

Groups where abnormal operating conditions are present were also identified with the aid of k-means clustering. However, for these identified abnormal operating conditions, causal relationships to specific faults were not proven. It must be emphasized that various gasifier faults can occur, which cause deviation from normal operating conditions, as discussed in Section 2.2.5. As is also evident in Section 2.2.5, these deviations may coincide between faults. Therefore, as stated in Section 2.3.2, fault classification is more challenging than fault detection, since the latter requires only the prediction of abnormal conditions and not the additional identification of the causal fault. In Section 2.3.2, the decision to select between fault detection and classification could not yet be made. However, the use of classification models to differentiate between solvable and unsolvable observations, which have been established as, respectively, representing normal and abnormal operating conditions, can now be defined as fault detection. Therefore, the validity of the thermal-fluid model, for the prediction of mass flow rates during normal and abnormal gasifier operations, has been established.

The selected statistical model was compared with a conventional form of process monitoring that is applicable to coal gasification. This form of monitoring involves developing 95%

confidence ellipses for a reference set of process observations. When compared with the  $H_2O$  and  $CO_2$  SVM model, this conventional form of process monitoring was shown to misclassify more observations in total. Therefore, the validity of the selected SVM model for predicting abnormal gasifier operations was demonstrated.

## CHAPTER 6: CONCLUSIONS AND RECOMMENDATIONS

### 6.1 Conclusions

To conclude this study, it is necessary to review the work that has been completed in order to address the problem statement provided in Chapter 1. The problem statement noted that gasifier fault prediction requires data driven statistical techniques, since the relationships between faults and the thermal-fluid process are not defined sufficiently, in literature, for this. As such, the research aim was to develop an integrated thermal-fluid and statistical approach for fault prediction in fixed-bed dry bottom coal gasifiers.

The research began, in Chapter 2, with a literature study into the respective gasification process. This included material on the gasifier's operation, control, and chemical reactions. Literature for gasifier thermal-fluid modeling and statistical fault prediction were also investigated. Fixed-bed dry bottom gasifier faults were investigated, and the composition of coal was discussed, with respect to the gasification process.

The required thermal-fluid model, which predicts mass flow rates of the  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ , and  $H_2O$  comprising syngas, was developed in Chapter 3. This model is based on five fundamental thermodynamic equations, which are provided and described in that chapter. These equations are three mole balances, an energy balance, and a chemical reaction equilibrium condition. Equations for calculating the mole flow rates, enthalpies, Gibbs functions, and other variables required in these fundamental equations are also provided in Chapter 3.

Chapter 4 described the separate implementation of the thermal-fluid model as Models A and B. Model A applies assumed mole fractions when calculating syngas enthalpies, while Model B calculates these mole fractions. In Chapter 5, this difference in implementation allowed the isolation of abnormal operating conditions from typical gasifier data. This is since Model A is solvable at observations, representing abnormal operating conditions, where negative flow rates are predicted. However, Model B is unsolvable at these observations.

In addition, Chapter 4 provided reference gasifier data, which were applied when adjusting the heat loss fraction to account for model uncertainty. Accuracies for the predicted syngas volume percentages were determined at the selected heat loss fraction, as 86.2%, 65.4%, 88.5%, and 93.1% for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$ , respectively. As provided, the thermal-fluid model's average prediction accuracy of 83.3% is comparable with that of a fixed-bed gasifier model available in literature. The calculations for Model B were also verified, and it was

demonstrated that predicted syngas volume percentages correspond between Models A and B.

The statistical modeling undertaken in Chapter 5 resulted in the development and selection of a classification model that predicts abnormal gasifier operating conditions. In this modeling, PCA was applied, to the outputs of Model A, to determine uncorrelated mass flow rates pairs. These flow rate pairs, and scores for the first two principal component directions, were applied in CT and SVM models to predict where Model B is unsolvable. From these models, the  $H_2O$  and  $CO_2$  SVM model was selected due to its prediction of a bounded two-dimensional region where Model B is solvable.

In addition, K-means clustering was applied to determine four inlet steam and coal flow rate groups. These were analyzed, with respect to the predicted outlet syngas mass flow rates and volume percentages, in order to validate the thermal-fluid model. This demonstrated that the changes expected, from a fundamental understanding of the gasification reactions, are predicted by the model for variations in these inlet flow rates.

The existence of gasifier faults, within inlet flow rate groups containing abnormal observations, was also discussed. Therefore, as required to address the research problem, an integrated thermal-fluid and statistical modeling approach, for fault prediction in fixed-bed dry bottom coal gasifiers, has been developed and applied. In addition, when compared with the 94.4% accuracy of conventional principal component confidence ellipse-based process monitoring for fixed-bed dry bottom coal gasifiers, it was demonstrated that the  $H_2O$  and  $CO_2$  SVM model possesses a higher accuracy of 99.3%.

As evident, the integrated approach for predicting abnormal fixed-bed dry bottom coal gasifier operations has been verified and validated sufficiently to justify its ultimate utilization in process monitoring. However, prior to practical testing or implementation of this approach, additional research is required in order to improve the thermal-fluid and statistical models. As such, research recommendations are presented in the following section, as identified during the course of the study.

## **6.2 Recommendations**

From the research that has been completed in this study, the following are recommended topics for future work. Additional experimental research should be completed into the release of volatiles in fixed-bed dry bottom coal gasifiers. This is in order to establish more accurate correlations for predicting the mass flow rates of products released during coal devolatilization.

Experimental research should be undertaken to define more accurate correlations for the energy content of volatiles. Research into the prediction of trace gases present in raw syngas should also be accomplished. These research topics are significant, given that a heat loss fraction had to be adjusted to account for uncertainties in the thermal-fluid model, which included the flow rates of volatiles and trace gases.

Research should be completed into the prediction of FC in the ash exiting a gasifier. This is since, in the thermal-fluid model, the fraction of FC in ash is predefined. Changes that occur during gasification in ash's chemical composition, and associated thermal properties, should also be investigated. This research is required since, in the thermal-fluid model, ash is assumed to not undergo chemical reactions.

In addition, a sensitivity analysis for the thermal-fluid model should be completed. This is in order to assess the effect of the model's assumptions on its prediction accuracy. The relative contributions of the model's input variables, to the predicted mass flow rates, are also determinable from the results of a sensitivity analysis.

Research should be undertaken to establish error margins for the model's predictions, by determining accuracy across a range of values for the input variables. This will define the thermal-fluid model's accuracy for suboptimal and abnormal observations. This research is required since accuracy has only been calculated for the reference gasifier data, which was shown to represent normal operations.

Experimental research should be completed, wherein a fixed-bed dry bottom coal gasifier is operated under the abnormal conditions predicted by the  $H_2O$  and  $CO_2$  SVM model. In such research, the resulting syngas and ash would be analyzed to determine if faults are present in these abnormal operating conditions.

In addition, statistical techniques for correcting class imbalance should be applied to the predicted mass flow rates output by Model A. This is to determine if improvements, in the results of the statistical classification models, are attainable by reducing the class imbalance present between solvable and unsolvable observations. Repeated random sampling of the training and validation set, and subsequent statistical model training and testing, should also be undertaken. This is to further verify the results presented for the statistical models.

SVM models with radial kernel functions should also be applied to predict where Model B is solvable. This is since these SVM models are expected to either predict bounded solvable regions, with corresponding unbounded unsolvable regions, or unbounded solvable regions, with corresponding bounded unsolvable regions. In accordance with the previous

recommendation, these models should be developed before and after statistical techniques for class imbalance correction have been applied.

Statistical classification models, which utilize original gasifier process measurements to predict abnormal operating conditions, should also be developed and compared with those presented. These were excluded from the study in order to focus on predictions made with the thermal-fluid model's outputs. This is since the integrated thermal-fluid and statistical modeling approach was the focus of the study. Therefore, pure statistical modeling for fault prediction, applied to the original process measurements, has not yet been undertaken.

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## APPENDIX A – SUPPLEMENTARY EQUATIONS

### A.1 Introduction

This appendix contains the additional thermal-fluid model equations referred to in Chapters 3 and 4, along with supplemental equations for the statistical models in Chapter 5. Additional equations required to process the model's outputs are also provided.

Sections A.2 through A.4 contain the additional equations referred to in Sections 3.4 through 3.6, respectively. Equations for calculating the syngas volume percentages are provided in Section A.5. Section A.6 contains equations for calculating the total outlet mass flow rates for gases that comprise syngas. The additional equations referred to in Section 4.5 are provided in Section A.7. Supplemental equations for the principal component classification models are contained in Appendix A.8.

### A.2 Mole Balances

Equation A.1 provides the mass to mole flow rate conversion for inlet  $O_2$ .

$$\dot{n}_{O_2,in} = \dot{m}_{O_2,in} * M_{O_2}^{-1} \quad [A.1]$$

where the molar mass of  $O_2$  is represented by  $M_{O_2}$ .

Equations A.2 through A.5 provide mole to mass flow rate conversions, respectively, for the  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  produced during combustion and gasification reactions.

$$\dot{m}_{H_2,gas} = \dot{n}_{H_2,gas} * M_{H_2} \quad [A.2]$$

$$\dot{m}_{CO,gas} = \dot{n}_{CO,gas} * M_{CO} \quad [A.3]$$

$$\dot{m}_{CO_2,gas} = \dot{n}_{CO_2,gas} * M_{CO_2} \quad [A.4]$$

$$\dot{m}_{CH_4,gas} = \dot{n}_{CH_4,gas} * M_{CH_4} \quad [A.5]$$

here the mass flow rates of  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are represented by  $\dot{m}_{H_2,gas}$ ,  $\dot{m}_{CO,gas}$ ,  $\dot{m}_{CO_2,gas}$ , and  $\dot{m}_{CH_4,gas}$ , respectively. The respective molar masses for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are  $M_{H_2}$ ,  $M_{CO}$ ,  $M_{CO_2}$ , and  $M_{CH_4}$ .

### A.3 Energy Balance

Equations A.6 and A.7 are applied to calculate enthalpy and enthalpy-flow of inlet  $O_2$ .

$$\dot{H}_{O_2,in} = \dot{m}_{O_2,in} * h_{O_2,in} \quad [A.6]$$

$$h_{O_2,in} = h_{O_2,frm} + \Delta h_{O_2,in}(T_{H_2O\&O_2}, P_{O_2,in}, T_{frm}, P_{frm}) \quad [A.7]$$

here the enthalpy of  $O_2$  at inlet conditions is  $h_{O_2,in}$ . The enthalpy of formation for  $O_2$  is  $h_{O_2,frm}$ , and  $\Delta h_{O_2,in}$  represents a function that calculates the difference in enthalpy, between reference and inlet conditions.

Equations A.8 and A.9 are applied to calculate the mole fraction and partial pressure of inlet  $O_2$ .

$$P_{O_2,in} = y_{O_2,in} * P_{H_2O\&O_2} \quad [A.8]$$

$$y_{O_2,in} = \frac{\dot{n}_{O_2,in}}{\dot{n}_{H_2O\&O_2}} \quad [A.9]$$

where the mole fraction and partial pressure of  $O_2$  are represented by  $y_{O_2,in}$  and  $P_{O_2,in}$ , respectively.

Equations A.10 through A.15 are applied to calculate enthalpy and enthalpy-flows of outlet  $CO$ ,  $CO_2$ , and  $CH_4$ .

$$\dot{H}_{CO,out} = (\dot{m}_{CO,gas} + \dot{m}_{CO,dvl}) * h_{CO,out} \quad [A.10]$$

$$\dot{H}_{CO_2,out} = (\dot{m}_{CO_2,gas} + \dot{m}_{CO_2,dvl}) * h_{CO_2,out} \quad [A.11]$$

$$\dot{H}_{CH_4,out} = (\dot{m}_{CH_4,gas} + \dot{m}_{CH_4,dvl}) * h_{CH_4,out} \quad [A.12]$$

$$h_{CO,out} = h_{CO,frm} + \Delta h_{CO,out}(T_{sgs}, P_{CO,out}, T_{frm}, P_{frm}) \quad [A.13]$$

$$h_{CO_2,out} = h_{CO_2,frm} + \Delta h_{CO_2,out}(T_{sgs}, P_{CO_2,out}, T_{frm}, P_{frm}) \quad [A.14]$$

$$h_{CH_4,out} = h_{CH_4,frm} + \Delta h_{CH_4,out}(T_{sgs}, P_{CH_4,out}, T_{frm}, P_{frm}) \quad [A.15]$$

here the enthalpy at outlet conditions for  $CO$ ,  $CO_2$ , and  $CH_4$  are  $h_{CO,out}$ ,  $h_{CO_2,out}$ , and  $h_{CH_4,out}$ , respectively. The mass flow rates of  $CO$ ,  $CO_2$ , and  $CH_4$  released during coal devolatilization are represented by  $\dot{m}_{CO,dvl}$ ,  $\dot{m}_{CO_2,dvl}$ , and  $\dot{m}_{CH_4,dvl}$ , respectively. Enthalpies of formation for  $CO$ ,  $CO_2$ , and  $CH_4$  are, respectively,  $h_{CO,frm}$ ,  $h_{CO_2,frm}$ , and  $h_{CH_4,frm}$ . The differences in enthalpy, between outlet and reference conditions for  $CO$ ,  $CO_2$ , and  $CH_4$ , are calculated using

functions represented by  $\Delta h_{CO,out}$ ,  $\Delta h_{CO_2,out}$ , and  $\Delta h_{CH_4,out}$ , respectively. Partial pressures for CO, CO<sub>2</sub>, and CH<sub>4</sub> are, respectively, represented by  $P_{CO,out}$ ,  $P_{CO_2,out}$ , and  $P_{CH_4,out}$ .

Equations A.16 through A.21 are applied to calculate the mole fractions and partial pressures of the outlet CO, CO<sub>2</sub>, and CH<sub>4</sub>.

$$P_{CO,out} = y_{CO,out} * P_{sgs} \quad [A.16]$$

$$P_{CO_2,out} = y_{CO_2,out} * P_{sgs} \quad [A.17]$$

$$P_{CH_4,out} = y_{CH_4,out} * P_{sgs} \quad [A.18]$$

$$y_{CO,out} = \frac{\dot{n}_{CO,gas} + \dot{n}_{CO,dvl}}{\dot{n}_{sgs}} \quad [A.19]$$

$$y_{CO_2,out} = \frac{\dot{n}_{CO_2,gas} + \dot{n}_{CO_2,dvl}}{\dot{n}_{sgs}} \quad [A.20]$$

$$y_{CH_4,out} = \frac{\dot{n}_{CH_4,gas} + \dot{n}_{CH_4,dvl}}{\dot{n}_{sgs}} \quad [A.21]$$

where  $P_{CO,out}$ ,  $P_{CO_2,out}$ , and  $P_{CH_4,out}$  are the partial pressures of CO, CO<sub>2</sub>, and CH<sub>4</sub>, respectively. The respective mole fractions for the outlet CO, CO<sub>2</sub>, and CH<sub>4</sub> are  $y_{CO,out}$ ,  $y_{CO_2,out}$ , and  $y_{CH_4,out}$ .

Equations A.22 through A.27 are applied to calculate the Normal volume and mass flow rates of the CO, CO<sub>2</sub>, and CH<sub>4</sub> released during coal devolatilization.

$$\dot{m}_{CO,dvl} = \dot{V}_{CO,dvl} * \rho_{CO} \quad [A.22]$$

$$\dot{m}_{CO_2,dvl} = \dot{V}_{CO_2,dvl} * \rho_{CO_2} \quad [A.23]$$

$$\dot{m}_{CH_4,dvl} = \dot{V}_{CH_4,dvl} * \rho_{CH_4} \quad [A.24]$$

$$\dot{V}_{CO,dvl} = CO_{WAF,coal} * [(FC_{AR,coal} + VM_{AR,coal}) * \dot{m}_{coal}] \quad [A.25]$$

$$\dot{V}_{CO_2,dvl} = CO_{2,WAF,coal} * [(FC_{AR,coal} + VM_{AR,coal}) * \dot{m}_{coal}] \quad [A.26]$$

$$\dot{V}_{CH_4,dvl} = CH_{4,WAF,coal} * [(FC_{AR,coal} + VM_{AR,coal}) * \dot{m}_{coal}] \quad [A.27]$$

here the Normal volumes of  $CO$ ,  $CO_2$ , and  $CH_4$ , per unit mass of coal, that are released during devolatilization are  $CO_{WAF,coal}$ ,  $CO_{2,WAF,coal}$ , and  $CH_{4,WAF,coal}$ , respectively. The Normal volume flow rates of  $CO$ ,  $CO_2$ , and  $CH_4$  released during devolatilization are  $\dot{V}_{CO,dvl}$ ,  $\dot{V}_{CO_2,dvl}$ , and  $\dot{V}_{CH_4,dvl}$ , respectively. Densities, at Normal conditions, for  $CO$ ,  $CO_2$ , and  $CH_4$  are, respectively,  $\rho_{CO}$ ,  $\rho_{CO_2}$ , and  $\rho_{CH_4}$ .

Equations A.28 through A.30 provide mass to mole flow rate conversions for the  $CO$ ,  $CO_2$ , and  $CH_4$  released during devolatilization.

$$\dot{n}_{CO,dvl} = \dot{m}_{CO,dvl} * M_{CO}^{-1} \quad [A.28]$$

$$\dot{n}_{CO_2,dvl} = \dot{m}_{CO_2,dvl} * M_{CO_2}^{-1} \quad [A.29]$$

$$\dot{n}_{CH_4,dvl} = \dot{m}_{CH_4,dvl} * M_{CH_4}^{-1} \quad [A.30]$$

Equations A.31 through A.34 are applied to calculate the mass and mole flow rates of elemental sulfur and hydrogen in tar.

$$\dot{m}_{S,tar} = S_{WAF,tar} * \dot{m}_{tar} \quad [A.31]$$

$$\dot{m}_{H,tar} = H_{WAF,tar} * \dot{m}_{tar} \quad [A.32]$$

$$\dot{n}_{S,tar} = \dot{m}_{S,tar} * M_S^{-1} \quad [A.33]$$

$$\dot{n}_{H,tar} = \dot{m}_{H,tar} * M_H^{-1} \quad [A.34]$$

where the elemental mass fractions of sulfur and hydrogen in tar, on a WAF basis, are  $S_{WAF,tar}$  and  $H_{WAF,tar}$ , respectively. Mass flow rates for the sulfur and hydrogen in tar are, respectively,  $\dot{m}_{S,tar}$  and  $\dot{m}_{H,tar}$ , and elemental molar masses for sulfur and hydrogen are  $M_S$  and  $M_H$ , respectively.

#### A.4 Chemical Equilibrium

Equations A.35 through A.38 are applied to calculate the Gibbs functions for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$ .

$$g_{H_2,out} = M_{H_2} * [h_{H_2,out,idl}(T_{sgs}) - T_{sgs} * S_{H_2,out,idl}(T_{sgs}, P_{frm})] \quad [A.35]$$

$$g_{CO,out} = M_{CO} * [h_{CO,out,idl}(T_{sgs}) - T_{sgs} * S_{CO,out,idl}(T_{sgs}, P_{frm})] \quad [A.36]$$

$$g_{CO2,out} = M_{CO2} * [h_{CO2,out,idl}(T_{sgs}) - T_{sgs} * s_{CO2,out,idl}(T_{sgs}, P_{frm})] \quad [A.37]$$

$$g_{CH4,out} = M_{CH4} * [h_{CH4,out,idl}(T_{sgs}) - T_{sgs} * s_{CH4,out,idl}(T_{sgs}, P_{frm})] \quad [A.38]$$

here ideal gas enthalpy functions for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are  $h_{H2,out,idl}$ ,  $h_{CO,out,idl}$ ,  $h_{CO2,out,idl}$ , and  $h_{CH4,out,idl}$ , respectively. The ideal gas entropy functions for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are, respectively,  $s_{H2,out,idl}$ ,  $s_{CO,out,idl}$ ,  $s_{CO2,out,idl}$ , and  $s_{CH4,out,idl}$ .

## A.5 Volume Percentages

Equations A.39 through A.43 are applied to calculate outlet Normal volume flow rates.

$$\dot{V}_{H2,out} = \frac{\dot{m}_{H2,gas} + \dot{m}_{H2,dvl}}{\rho_{H2}} \quad [A.39]$$

$$\dot{V}_{CO,out} = \frac{\dot{m}_{CO,gas} + \dot{m}_{CO,dvl}}{\rho_{CO}} \quad [A.40]$$

$$\dot{V}_{CO2,out} = \frac{\dot{m}_{CO2,gas} + \dot{m}_{CO2,dvl}}{\rho_{CO2}} \quad [A.41]$$

$$\dot{V}_{CH4,out} = \frac{\dot{m}_{CH4,gas} + \dot{m}_{CH4,dvl}}{\rho_{CH4}} \quad [A.42]$$

$$\dot{V}_{sgs} = \dot{V}_{H2,out} + \dot{V}_{CO,out} + \dot{V}_{CO2,out} + \dot{V}_{CH4,out} \quad [A.43]$$

here  $\dot{V}_{H2,out}$ ,  $\dot{V}_{CO,out}$ ,  $\dot{V}_{CO2,out}$ , and  $\dot{V}_{CH4,out}$  are the outlet Normal volume flow rates for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$ , respectively. The total Normal volume flow rate for syngas, excluding  $H_2O$ , is  $\dot{V}_{sgs}$ .

Equations A.44 through A.47 are applied to calculate the syngas volume percentages.

$$V_{H2} = \frac{\dot{V}_{H2,out}}{\dot{V}_{sgs}} * 100 \quad [A.44]$$

$$V_{CO} = \frac{\dot{V}_{CO,out}}{\dot{V}_{sgs}} * 100 \quad [A.45]$$

$$V_{CO2} = \frac{\dot{V}_{CO2,out}}{\dot{V}_{sgs}} * 100 \quad [A.46]$$

$$V_{CH_4} = \frac{\dot{V}_{CH_4,out}}{\dot{V}_{sgs}} * 100 \quad [A.47]$$

## A.6 Outlet Syngas Mass Flow Rates

Equations A.48 through A.52 are applied to calculate the total outlet mass flow rates of the gases comprising syngas.

$$\dot{m}_{H_2O,out} = \dot{m}_{H_2O,gas} + \dot{m}_{H_2O,dvl} \quad [A.48]$$

$$\dot{m}_{H_2,out} = \dot{m}_{H_2,gas} + \dot{m}_{H_2,dvl} \quad [A.49]$$

$$\dot{m}_{CO,out} = \dot{m}_{CO,gas} + \dot{m}_{CO,dvl} \quad [A.50]$$

$$\dot{m}_{CO_2,out} = \dot{m}_{CO_2,gas} + \dot{m}_{CO_2,dvl} \quad [A.51]$$

$$\dot{m}_{CH_4,out} = \dot{m}_{CH_4,gas} + \dot{m}_{CH_4,dvl} \quad [A.52]$$

where  $\dot{m}_{H_2O,out}$ ,  $\dot{m}_{H_2,out}$ ,  $\dot{m}_{CO,out}$ ,  $\dot{m}_{CO_2,out}$ , and  $\dot{m}_{CH_4,out}$  are the outlet mass flow rates of  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$ , respectively.

## A.7 Errors

In order to calculate relative errors for the predicted syngas volume percentages for  $CO$ ,  $CO_2$ , and  $CH_4$ , Equations A.53 through A.55 are applied, respectively.

$$RE_{CO} = \frac{|V_{CO} - Ref_{CO}|}{Ref_{CO}} * 100 \quad [A.53]$$

$$RE_{CO_2} = \frac{|V_{CO_2} - Ref_{CO_2}|}{Ref_{CO_2}} * 100 \quad [A.54]$$

$$RE_{CH_4} = \frac{|V_{CH_4} - Ref_{CH_4}|}{Ref_{CH_4}} * 100 \quad [A.55]$$

here  $RE_{CO}$ ,  $RE_{CO_2}$ , and  $RE_{CH_4}$  represent relative errors for the predicted syngas volume percentages of  $CO$ ,  $CO_2$ , and  $CH_4$ , respectively.

## A.8 Principal Component Classification Models

### A.8.1 Classification Trees

Equation A.56 is adapted from Equation 5.3 and defines the division of a given two-dimensional principal component space into two regions. However, the Gini index for the principal component CT model is identical to that in Equation 5.4 for the mass flow rate CT model.

$$Rg_{2b-1} = \{pc_x \mid pc_\zeta < sp_b\} \text{ and } Rg_{2b} = \{pc_x \mid pc_\zeta \geq sp_b\} \quad [A.56]$$

here  $pc_x$  represents the space spanned by the first two principal components. The component selected from these, along with split  $sp_b$ , to minimize the Gini index is  $pc_\zeta$ , where  $b$  is an element of the set of natural numbers. Therefore,  $\zeta$  is an element of  $x$ .

Terminal regions in the principal component CT model are defined according to Equation 5.5. However, Equation A.57 is adapted from Equation 5.6 to provide the classification error rate for the principal component CT model.

$$CER = \frac{1}{\omega} \sum_{a=1}^A \min \left\{ \begin{matrix} num[pc_x(cl = 1) \in Tg_a] \\ num[pc_x(cl = 2) \in Tg_a] \end{matrix} \right\} \quad [A.57]$$

### A.8.2 Support Vector Machines

Equation A.58 is an adaptation of Equation 5.10 for the first two principal components. Thus, it is applicable, during SVM model training, to the margin maximization problem. The second-degree polynomial kernel that corresponds to Equation A.58 is provided in Equation A.59. However, the constraint for this model's classification errors is still defined by Equation 5.12.

$$f_w * \left\{ \beta_0 + \sum_{\varpi=1}^{\omega} [\alpha_{\varpi} * K(pc_{x,\varpi}, pc_{x,w})] \right\} \geq Mi * (1 - \varepsilon_w) \quad [A.58]$$

$$\forall w \in \{1, \dots, \omega\}$$

$$K(pc_{x,\varpi}, pc_{x,w}) = \left[ \delta_0 + \gamma * \sum_{x \in PC} (pc_{x,\varpi} * pc_{x,w}) \right]^2 \quad [A.59]$$

here training observations  $\varpi$  and  $w$ , for principal component  $x$ , are  $pc_{x,\varpi}$  and  $pc_{x,w}$ . Subset  $PC$ , which is  $\{1, 2\}$ , specifies the first two principal components. Wherever  $\alpha_{\varpi}$  is not equal to 0,  $pc_{x,\varpi}$  is a support vector.

Equation A.60 is adapted from Equation 5.13 and defines the application of the trained principal component SVM model. Equation A.61 provides the kernel function required in Equation A.60.

$$f(pc_x^*) = \beta_0 + \sum_{\varpi \in SV} [\alpha_{\varpi} * K(pc_{x,\varpi}, pc_x^*)] \quad [A.60]$$

$$K(pc_{x,\varpi}, pc_x^*) = \left[ \delta_0 + \gamma * \sum_{x \in PC} (pc_{x,\varpi} * pc_x^*) \right]^2 \quad [A.61]$$

where  $pc_x^*$  is a new observation, which is classified based on the sign of  $f(pc_x^*)$ , and  $SV$  defines the subset of support vectors.

### A.8.3 Error and Accuracy Metrics

The classification accuracy, non-detection rate, and false-alarm rate for the principal component models are, respectively, calculated with Equations A.62, A.63, and A.64. The equations for these metrics are adapted from Equations 5.7 through 5.9.

$$Ac = \frac{num[pc_x(cl' = cl)]}{\omega} * 100 \quad [A.62]$$

$$NDR = \frac{num[pc_x(cl' = 1 | cl = 2)]}{num[pc_x(cl = 2)]} * 100 \quad [A.63]$$

$$FAR = \frac{num[pc_x(cl' = 2 | cl = 1)]}{num[pc_x(cl = 1)]} * 100 \quad [A.64]$$

## APPENDIX B – VERIFICATION CALCULATIONS

### B.1 Introduction

This appendix contains the full set of calculations required for verification of the thermal-fluid model. Values are provided, as output from EES, for these calculations. Section B.2 provides the calculations required to verify the three mole balances. The calculations required to verify the energy balance are presented in Section B.3. Chemical equilibrium for the water gas shift reaction is verified with the calculations in Section B.4, and the syngas volume percentages are calculated in Section B.5.

### B.2 Mole Balances

#### B.2.1 Model Outputs

The three mole balances, for carbon, hydrogen, and oxygen, respectively, are assessed as follows. The values required from Model B are first output from EES. The values predicted for the mole flow rates developed during gasification are:

- $\dot{n}_{H_2O,gas} = 0.249 \text{ kmol/s}$
- $\dot{n}_{H_2,gas} = 0.443 \text{ kmol/s}$
- $\dot{n}_{CO,gas} = 0.144 \text{ kmol/s}$
- $\dot{n}_{CO_2,gas} = 0.317 \text{ kmol/s}$
- $\dot{n}_{CH_4,gas} = 0.0588 \text{ kmol/s}$

The required densities at Normal conditions are:

- $\rho_{O_2} = 1.35 \text{ kg/m}^3$
- $\rho_{H_2} = 0.0852 \text{ kg/m}^3$
- $\rho_{CO} = 1.18 \text{ kg/m}^3$
- $\rho_{CO_2} = 1.87 \text{ kg/m}^3$
- $\rho_{CH_4} = 0.680 \text{ kg/m}^3$

#### B.2.2 Flow Rate Calculations

Secondly, to calculate the remaining flow rates required for the mole balances, the following is completed. The flow rates of inlet steam, coal, and water jacket steam are converted from t/h to kg/s:

- $\dot{m}_{H_2O,in} = (52.5 \text{ t/h} * 1,000 \text{ kg/t}) / (60^2 \text{ s/h}) = 14.6 \text{ kg/s}$
- $\dot{m}_{coal} = (50.0 \text{ t/h} * 1,000 \text{ kg/t}) / (60^2 \text{ s/h}) = 13.9 \text{ kg/s}$
- $\dot{m}_{wj} = (8.30 \text{ t/h} * 1,000 \text{ kg/t}) / (60^2 \text{ s/h}) = 2.31 \text{ kg/s}$

The mass flow rate of inlet oxygen is calculated by converting from  $10^3 \text{ m}_n^3/\text{h}$  to  $\text{m}_n^3/\text{s}$  and applying Equation 3.10:

- $\dot{m}_{O_2,in} = [(9.20 \cdot 10^3 \text{ m}_n^3/\text{h}) / (60^2 \text{ s/h})] * 1.35 \text{ kg/m}^3 = 3.46 \text{ kg/s}$

The mole flow rates of inlet steam and oxygen are calculated by applying Equations 3.9 and A.1:

- $\dot{n}_{H_2O,in} = 14.6 \text{ kg/s} * (18.015 \text{ kg/kmol})^{-1} = 0.810 \text{ kmol/s}$
- $\dot{n}_{O_2,in} = 3.46 \text{ kg/s} * (31.999 \text{ kg/kmol})^{-1} = 0.108 \text{ kmol/s}$

The mass flow rate of ash is calculated by applying Equation 3.6 and converting from kg/h to kg/s:

- $\dot{m}_{ash} = (3,000 \text{ kg/rev} * 4.5 \text{ rev/h}) / (60^2 \text{ s/h}) = 3.8 \text{ kg/s}$

The mass flow rates of FC entering and exiting the gasifier are calculated by applying Equations 3.4 and 3.7:

- $\dot{m}_{C,in} = 0.471 * 13.9 \text{ kg/s} = 6.54 \text{ kg/s}$
- $\dot{m}_{C,out} = 0.081 * 3.8 \text{ kg/s} = 0.30 \text{ kg/s}$

Finally, the mole flow rates of FC entering and exiting the gasifier are calculated by applying Equations 3.5 and 3.8:

- $\dot{n}_{C,in} = 6.54 \text{ kg/s} * (12.01115 \text{ kg/kmol})^{-1} = 0.545 \text{ kmol/s}$
- $\dot{n}_{C,out} = 0.30 \text{ kg/s} * (12.01115 \text{ kg/kmol})^{-1} = 0.025 \text{ kmol/s}$

### B.2.3 Balance Calculations

Thus, the variables required by the mole balances, Equations 3.1 through 3.3, have been provided. For the carbon balance, the following results for the left hand side (LHS) and right hand side (RHS) of Equation 3.1:

- LHS = 0.545 kmol/s
- RHS = 0.025 kmol/s + 0.144 kmol/s + 0.317 kmol/s + 0.0588 kmol/s = 0.545 kmol/s

For the hydrogen balance, the following results for the LHS and RHS of Equation 3.2:

- $LHS = 2 * 0.810 \text{ kmol/s} = 1.619 \text{ kmol/s}$
- $RHS = 2 * 0.249 \text{ kmol/s} + 2 * 0.443 \text{ kmol/s} + 4 * 0.0588 \text{ kmol/s} = 1.619 \text{ kmol/s}$

For the oxygen balance, the following results for the LHS and RHS of Equation 3.3:

- $LHS = 0.810 \text{ kmol/s} + 2 * 0.108 \text{ kmol/s} = 1.026 \text{ kmol/s}$
- $RHS = 0.249 \text{ kmol/s} + 0.144 \text{ kmol/s} + 2 * 0.317 \text{ kmol/s} = 1.026 \text{ kmol/s}$

### B.3 Energy Balance

#### B.3.1 Model Outputs

The energy balance is assessed as follows. The required enthalpies of formation, as output from EES, are:

- $h_{liqH_2O,frm} = -15,900 \text{ kJ/kg}$
- $h_{H_2O,frm} = -13,400 \text{ kJ/kg}$
- $h_{O_2,frm} = 0 \text{ kJ/kg}$
- $h_{H_2,frm} = 0 \text{ kJ/kg}$
- $h_{CO,frm} = -3,950 \text{ kJ/kg}$
- $h_{CO_2,frm} = -8,940 \text{ kJ/kg}$
- $h_{CH_4,frm} = -4,650 \text{ kJ/kg}$
- $h_{SO_2,frm} = -4,630 \text{ kJ/kg}$

#### B.3.2 Flow Rate Calculations

The mass flow rates developed during gasification are calculated by applying Equations 3.11 and A.2 through A.5:

- $\dot{m}_{H_2O,gas} = 0.249 \text{ kmol/s} * 18.015 \text{ kg/kmol} = 4.48 \text{ kg/s}$
- $\dot{m}_{H_2,gas} = 0.443 \text{ kmol/s} * 2.0159 \text{ kg/kmol} = 0.894 \text{ kg/s}$
- $\dot{m}_{CO,gas} = 0.144 \text{ kmol/s} * 28.001 \text{ kg/kmol} = 4.02 \text{ kg/s}$
- $\dot{m}_{CO_2,gas} = 0.317 \text{ kmol/s} * 44.010 \text{ kg/kmol} = 13.9 \text{ kg/s}$
- $\dot{m}_{CH_4,gas} = 0.0588 \text{ kmol/s} * 16.043 \text{ kg/kmol} = 0.944 \text{ kg/s}$

The Normal volume flow rates developed during devolatilization are calculated by applying Equations 3.52 and A.25 through A.27:

- $\dot{V}_{H_2,dvl} = 0.09 \text{ m}_n^3/\text{kg} * [(0.471 + 0.218) * 13.9 \text{ kg/s}] = 0.9 \text{ m}_n^3/\text{s}$
- $\dot{V}_{CO,dvl} = 0.04 \text{ m}_n^3/\text{kg} * [(0.471 + 0.218) * 13.9 \text{ kg/s}] = 0.4 \text{ m}_n^3/\text{s}$
- $\dot{V}_{CO_2,dvl} = 0.05 \text{ m}_n^3/\text{kg} * [(0.471 + 0.218) * 13.9 \text{ kg/s}] = 0.5 \text{ m}_n^3/\text{s}$
- $\dot{V}_{CH_4,dvl} = 0.07 \text{ m}_n^3/\text{kg} * [(0.471 + 0.218) * 13.9 \text{ kg/s}] = 0.7 \text{ m}_n^3/\text{s}$

The mass flow rates developed during devolatilization are calculated by applying Equations 3.51 and A.22 through A.24:

- $\dot{m}_{H_2,dvl} = 0.9 \text{ m}_n^3/\text{s} * 0.0852 \text{ kg/m}^3 = 0.07 \text{ kg/s}$
- $\dot{m}_{CO,dvl} = 0.4 \text{ m}_n^3/\text{s} * 1.18 \text{ kg/m}^3 = 0.5 \text{ kg/s}$
- $\dot{m}_{CO_2,dvl} = 0.5 \text{ m}_n^3/\text{s} * 1.87 \text{ kg/m}^3 = 0.9 \text{ kg/s}$
- $\dot{m}_{CH_4,dvl} = 0.7 \text{ m}_n^3/\text{s} * 0.680 \text{ kg/m}^3 = 0.5 \text{ kg/s}$

The mass flow rate of steam developed during drying is calculated by applying Equation 3.53:

- $\dot{m}_{H_2O,dry} = 0.036 * 13.9 \text{ kg/s} = 0.50 \text{ kg/s}$

The mole flow rates developed during devolatilization are calculated by applying Equations 3.54 and A.28 through A.30:

- $\dot{n}_{H_2,dvl} = 0.07 \text{ kg/s} * (2.0159 \text{ kg/kmol})^{-1} = 0.04 \text{ kmol/s}$
- $\dot{n}_{CO,dvl} = 0.5 \text{ kg/s} * (28.001 \text{ kg/kmol})^{-1} = 0.02 \text{ kmol/s}$
- $\dot{n}_{CO_2,dvl} = 0.9 \text{ kg/s} * (44.010 \text{ kg/kmol})^{-1} = 0.02 \text{ kmol/s}$
- $\dot{n}_{CH_4,dvl} = 0.5 \text{ kg/s} * (16.043 \text{ kg/kmol})^{-1} = 0.03 \text{ kmol/s}$

The mole flow rate of steam developed during drying is calculated by applying Equation 3.55:

- $\dot{n}_{H_2O,dry} = 0.50 \text{ kg/s} * (18.015 \text{ kg/kmol})^{-1} = 0.028 \text{ kmol/s}$

The total mole flow rates entering and exiting the gasifier are calculated by applying Equations 3.34 and 3.50:

- $\dot{n}_{H_2O\&O_2} = 0.810 \text{ kmol/s} + 0.108 \text{ kmol/s} = 0.918 \text{ kmol/s}$
- $\dot{n}_{sgs} = (0.443 \text{ kmol/s} + 0.04 \text{ kmol/s}) + (0.144 \text{ kmol/s} + 0.02 \text{ kmol/s}) + (0.317 \text{ kmol/s} + 0.02 \text{ kmol/s}) + (0.0588 \text{ kmol/s} + 0.03 \text{ kmol/s}) + (0.249 \text{ kmol/s} + 0.028 \text{ kmol/s})$   
 $= 1.34 \text{ kmol/s}$

### B.3.3 Mole Fraction and Partial Pressure Calculations

The mole fractions of inlet steam and oxygen are calculated by applying Equations 3.33 and A.9:

- $y_{H_2O,in} = (0.810 \text{ kmol/s}) / (0.918 \text{ kmol/s}) = 0.882$
- $y_{O_2,in} = (0.108 \text{ kmol/s}) / (0.918 \text{ kmol/s}) = 0.118$

The mole fractions of the gases comprising syngas are calculated by applying Equations 3.48, 3.49, and A.19 through A.21:

- $y_{H_2O,out} = (0.249 \text{ kmol/s} + 0.028 \text{ kmol/s}) / (1.34 \text{ kmol/s}) = 0.21$
- $y_{H_2,out} = (0.443 \text{ kmol/s} + 0.04 \text{ kmol/s}) / (1.34 \text{ kmol/s}) = 0.36$
- $y_{CO,out} = (0.144 \text{ kmol/s} + 0.02 \text{ kmol/s}) / (1.34 \text{ kmol/s}) = 0.12$
- $y_{CO_2,out} = (0.317 \text{ kmol/s} + 0.02 \text{ kmol/s}) / (1.34 \text{ kmol/s}) = 0.25$
- $y_{CH_4,out} = (0.0588 \text{ kmol/s} + 0.03 \text{ kmol/s}) / (1.34 \text{ kmol/s}) = 0.07$

The partial pressures of inlet steam and oxygen are calculated by applying Equations 3.32 and A.8:

- $P_{H_2O,in} = 0.882 * 3,500 \text{ kPa} = 3,100 \text{ kPa}$
- $P_{O_2,in} = 0.118 * 3,500 \text{ kPa} = 410 \text{ kPa}$

The partial pressures of the gases comprising syngas are calculated by applying Equations 3.46, 3.47, and A.16 through A.18:

- $P_{H_2O,out} = 0.21 * 3,500 \text{ kPa} = 590 \text{ kPa}$
- $P_{H_2,out} = 0.36 * 3,500 \text{ kPa} = 1,000 \text{ kPa}$
- $P_{CO,out} = 0.12 * 3,500 \text{ kPa} = 340 \text{ kPa}$
- $P_{CO_2,out} = 0.25 * 3,500 \text{ kPa} = 720 \text{ kPa}$
- $P_{CH_4,out} = 0.07 * 3,500 \text{ kPa} = 190 \text{ kPa}$

### B.3.4 Steam, Oxygen, and Syngas Enthalpy-Flow Calculations

The differences in enthalpy for inlet steam and oxygen, calculated in EES according to Equations 3.31 and A.7, are:

- $\Delta h_{H_2O,in} = 3,000 \text{ kJ/kg}$
- $\Delta h_{O_2,in} = 310 \text{ kJ/kg}$

The differences in enthalpy for the gases comprising syngas, calculated in EES according to Equations 3.44, 3.45, and A.13 through A.15 are:

- $\Delta h_{H_2O,out} = 3,500 \text{ kJ/kg}$
- $\Delta h_{H_2,out} = 7,600 \text{ kJ/kg}$
- $\Delta h_{CO,out} = 560 \text{ kJ/kg}$
- $\Delta h_{CO_2,out} = 550 \text{ kJ/kg}$
- $\Delta h_{CH_4,out} = 1,600 \text{ kJ/kg}$

The enthalpies of inlet steam and oxygen are calculated by applying Equations 3.31 and A.7:

- $h_{H_2O,in} = -13,400 \text{ kJ/kg} + 3,000 \text{ kJ/kg} = -10,400 \text{ kJ/kg}$
- $h_{O_2,in} = 0 \text{ kJ/kg} + 310 \text{ kJ/kg} = 310 \text{ kJ/kg}$

The enthalpies of the gases comprising syngas are calculated by applying Equations 3.44, 3.45, and A.13 through A.15:

- $h_{H_2O,out} = -13,400 \text{ kJ/kg} + 3,500 \text{ kJ/kg} = -9,900 \text{ kJ/kg}$
- $h_{H_2,out} = 0 \text{ kJ/kg} + 7,600 \text{ kJ/kg} = 7,600 \text{ kJ/kg}$
- $h_{CO,out} = -3,950 \text{ kJ/kg} + 560 \text{ kJ/kg} = -3,390 \text{ kJ/kg}$
- $h_{CO_2,out} = -8,940 \text{ kJ/kg} + 550 \text{ kJ/kg} = -8,400 \text{ kJ/kg}$
- $h_{CH_4,out} = -4,650 \text{ kJ/kg} + 1,600 \text{ kJ/kg} = -3,100 \text{ kJ/kg}$

The enthalpy-flows of inlet steam and oxygen are calculated by applying Equations 3.30 and A.6:

- $\dot{H}_{H_2O,in} = 14.6 \text{ kg/s} * -10,400 \text{ kJ/kg} = -152,000 \text{ kW}$
- $\dot{H}_{O_2,in} = 3.46 \text{ kg/s} * 310 \text{ kJ/kg} = 1,060 \text{ kW}$

The enthalpy-flows of the gases comprising syngas are calculated by applying Equations 3.42, 3.43, and A.10 through A.12:

- $\dot{H}_{H_2O,out} = (4.48 \text{ kg/s} + 0.50 \text{ kg/s}) * -9,900 \text{ kJ/kg} = -49,000 \text{ kW}$
- $\dot{H}_{H_2,out} = (0.894 \text{ kg/s} + 0.07 \text{ kg/s}) * 7,600 \text{ kJ/kg} = 7,400 \text{ kW}$
- $\dot{H}_{CO,out} = (4.02 \text{ kg/s} + 0.5 \text{ kg/s}) * -3,390 \text{ kJ/kg} = -15,000 \text{ kW}$
- $\dot{H}_{CO_2,out} = (13.9 \text{ kg/s} + 0.9 \text{ kg/s}) * -8,400 \text{ kJ/kg} = -125,000 \text{ kW}$
- $\dot{H}_{CH_4,out} = (0.944 \text{ kg/s} + 0.5 \text{ kg/s}) * -3,100 \text{ kJ/kg} = -4,300 \text{ kW}$

### B.3.5 Heat Absorption Calculations

The enthalpies at the inlet and outlet of the water jacket, calculated in EES, are:

- $h_{wj,in} = 110 \text{ kJ/kg}$
- $h_{wj,out} = 1,000 \text{ kJ/kg}$

The rate of heat absorbed by the steam in the water jacket is calculated by applying Equation 3.38:

- $\Delta\dot{Q}_{wj} = 2.31 \text{ kg/s} * (110 \text{ kJ/kg} - 1,000 \text{ kJ/kg}) = -2,100 \text{ kW}$

The average temperature of ash is calculated:

- $T_{ash,avg} = (190^\circ\text{C} + 350^\circ\text{C}) / 2 = 270^\circ\text{C}$

The constant pressure specific heat of ash is calculated by applying Equation 3.37:

- $cp_{ash} = 0.752 + 0.293 * (270^\circ\text{C} + 273.15) / 1,000 = 0.91 \text{ kJ/kg-K}$

The change in enthalpy of ash is calculated by applying Equation 3.36:

- $\Delta h_{ash} = 0.91 \text{ kJ/kg-K} * (190^\circ\text{C} - 350^\circ\text{C}) = -150 \text{ kJ/kg}$

The heat absorption rate of ash is calculated by applying Equation 3.35:

- $\Delta\dot{Q}_{ash} = 3.8 \text{ kg/s} * -150 \text{ kJ/kg} = -550 \text{ kW}$

### B.3.6 Coal Enthalpy-Flow Calculations

The mass fraction of VM in coal on a WAF basis is calculated by applying Equation 3.25:

- $VM_{WAF,coal} = [(1 + 0.471 / 0.218)^{-1} + (1 + 0.489 / 0.227)^{-1}] / 2 = 0.317$

The mass fraction of moisture in coal on an AF basis is calculated by applying Equation 3.29:

- $Mst_{AF,coal} = (1 + 0.471 / 0.036 + 0.218 / 0.036)^{-1} = 0.050$

The constant pressure specific heat of FC and VM in coal is calculated by applying Equation 3.21:

- $cp_{FC\&VM} = 0.83 * (1 + 0.08 * 0.317) * [1 + 0.2 * (190^\circ\text{C} / 100) - 0.0033 * (190^\circ\text{C} / 100)^3]$   
 $= 1.16 \text{ kJ/kg-K}$

The constant pressure specific heat of coal is calculated by applying Equation 3.20:

- $cp_{coal} = (1 - 0.050) * 1.16 \text{ kJ/kg-K} + 0.050 * 4.187 \text{ kJ/kg-K} = 1.3 \text{ kJ/kg-K}$

The difference in enthalpy of coal is calculated by applying Equation 3.19:

- $\Delta h_{coal} = 1.3 \text{ kJ/kg-K} * (190^\circ\text{C} - 25^\circ\text{C}) = 220 \text{ kJ/kg}$

The combustion value of coal is calculated by applying Equation 3.18:

- $f_{com} = 0.01379 * \ln(0.127) - 0.01393 * 0.127^{0.75} + 1.02111 = 0.990$

The heat of combustion of coal is calculated by applying Equation 3.17:

- $h_{coal,com} = -32,763.3 * 0.795 - 141,789.2 * 0.042 - 9,276.8 * 0.016 = -32,200 \text{ kJ/kg}$

The enthalpy of formation of FC and VM in coal is calculated by applying Equation 3.16:

- $h_{FC\&VM,frm} = -32,200 \text{ kJ/kg} * (1 - 0.990) = -330 \text{ kJ/kg}$

The enthalpy of formation of coal is calculated by applying Equation 3.15:

- $h_{coal,frm} = (1 - 0.050) * -330 \text{ kJ/kg} + 0.050 * -15,900 \text{ kJ/kg} = -1,100 \text{ kJ/kg}$

The enthalpy of coal is calculated by applying Equation 3.14:

- $h_{coal} = -1,100 \text{ kJ/kg} + 220 \text{ kJ/kg} = -890 \text{ kJ/kg}$

The enthalpy-flow of coal is calculated by applying Equation 3.13:

- $\dot{H}_{coal} = (13.9 \text{ kg/s} - 3.8 \text{ kg/s}) * -890 \text{ kJ/kg} = -9,000 \text{ kW}$

### **B.3.7 Tar Enthalpy-Flow Calculations**

The mass flow rate of tar is calculated by applying Equation 3.60:

- $\dot{m}_{tar} = 0.031 * 13.9 \text{ kg/s} = 0.43 \text{ kg/s}$

The mass flow rates of C, S, and H in tar are calculated by applying Equations 3.61, A.31, and A.32:

- $\dot{m}_{C,tar} = 0.773 * 0.43 \text{ kg/s} = 0.33 \text{ kg/s}$
- $\dot{m}_{S,tar} = 0.014 * 0.43 \text{ kg/s} = 0.0060 \text{ kg/s}$

- $\dot{m}_{H,tar} = 0.049 * 0.43 \text{ kg/s} = 0.021 \text{ kg/s}$

The mole flow rates of C, S, and H in tar are calculated by applying Equations 3.62, A.33, and A.34:

- $\dot{n}_{C,tar} = 0.33 \text{ kg/s} * (12.01115 \text{ kg/kmol})^{-1} = 0.028 \text{ kmol/s}$
- $\dot{n}_{S,tar} = 0.0060 \text{ kg/s} * (32.064 \text{ kg/kmol})^{-1} = 0.00019 \text{ kmol/s}$
- $\dot{n}_{H,tar} = 0.021 \text{ kg/s} * (1.00797 \text{ kg/kmol})^{-1} = 0.021 \text{ kmol/s}$

The difference in enthalpy of tar is calculated by applying Equation 3.59:

- $\Delta h_{tar} = 2.11 * 10^{-3} * [(550^\circ\text{C} + 273.15)^2 - (25^\circ\text{C} + 273.15)^2] = 1,240 \text{ kJ/kg}$

The enthalpy-flow for the difference in enthalpy of tar is calculated by applying Equation 3.58:

- $\Delta \dot{H}_{tar} = 0.43 \text{ kg/s} * 1,240 \text{ kJ/kg} = 540 \text{ kW}$

The enthalpy-flow for the enthalpy of formation of tar is calculated by applying Equation 3.57:

- $\dot{H}_{tar,frm} = -0.43 \text{ kg/s} * 30,890 + 0.028 \text{ kmol/s} * (-8,940 \text{ kJ/kg} * 44.010 \text{ kg/kmol})$   
 $+ 0.00019 \text{ kmol/s} * (-4,630 \text{ kJ/kg} * 64.064 \text{ kg/kmol})$   
 $+ [0.021 \text{ kmol/s} * (-15,900 \text{ kJ/kg} * 18.015 \text{ kg/kmol})] / 2 = -27,000 \text{ kW}$

The enthalpy-flow of tar is calculated by applying Equation 3.56:

- $\dot{H}_{tar} = -27,000 \text{ kW} + 540 \text{ kW} = -26,700 \text{ kW}$

### B.3.8 Rate of Heat Loss Calculations

The higher heating value of coal is calculated by applying Equation 3.41:

- $HHV_{WAF} = 35,200 * 0.795 + 116,400 * 0.042 - 11,100 * 0.127 + 10,500 * 0.016$   
 $+ 6,290 * 0.021 = 32,000 \text{ kJ/kg}$

The lower heating value of coal is calculated by applying Equation 3.40:

- $LHV_{WAF} = 32,000 \text{ kJ/kg} - 2,442 * (9 * 0.042) = 31,000 \text{ kJ/kg}$

The rate of heat loss is calculated by applying Equation 3.39:

- $\dot{Q}_{loss} = [(0.471 + 0.218) * 13.9 \text{ kg/s}] * 0.17 * 31,000 \text{ kJ/kg} = 50,200 \text{ kW}$

### B.3.9 Balance Calculations

Thus, the variables required by the energy balance, Equation 3.12, have been provided. The following results for the LHS and RHS of this equation:

- $LHS = -9,000 \text{ kW} - 152,000 \text{ kW} + 1,060 \text{ kW} - 550 \text{ kW} - 2,100 \text{ kW} = -163,000 \text{ kW}$
- $RHS = 50,200 \text{ kW} - 49,000 \text{ kW} + 7,400 \text{ kW} - 15,000 \text{ kW} - 125,000 \text{ kW} - 4,300 \text{ kW} - 26,700 \text{ kW} = -163,000 \text{ kW}$

## B.4 Chemical Equilibrium

### B.4.1 Model Outputs

The chemical equilibrium condition for the water gas shift reaction is assessed as follows. The ideal gas enthalpies and entropies are output from EES. The required enthalpies are:

- $h_{H_2O,out,idl} = -12,400 \text{ kJ/kg}$
- $h_{H_2,out,idl} = 7,620 \text{ kJ/kg}$
- $h_{CO,out,idl} = -3,380 \text{ kJ/kg}$
- $h_{CO_2,out,idl} = -8,400 \text{ kJ/kg}$

The required entropies are:

- $s_{H_2O,out,idl} = 12.5 \text{ kJ/kg-K}$
- $s_{H_2,out,idl} = 79.5 \text{ kJ/kg-K}$
- $s_{CO,out,idl} = 8.15 \text{ kJ/kg-K}$
- $s_{CO_2,out,idl} = 5.88 \text{ kJ/kg-K}$

### B.4.2 Gibbs Function Calculations

The Gibbs functions are calculated by applying Equations 3.67 and A.35 through A.37:

- $g_{H_2O,out} = 18.015 \text{ kg/kmol} * [-12,400 \text{ kJ/kg} - (550^\circ\text{C} + 273.15) * 12.5 \text{ kJ/kg-K}]$   
 $= -408,000 \text{ kJ/kmol}$
- $g_{H_2,out} = 2.0159 \text{ kg/kmol} * [7,620 \text{ kJ/kg} - (550^\circ\text{C} + 273.15) * 79.5 \text{ kJ/kg-K}]$   
 $= -117,000 \text{ kJ/kmol}$
- $g_{CO,out} = 28.001 \text{ kg/kmol} * [-3,380 \text{ kJ/kg} - (550^\circ\text{C} + 273.15) * 8.15 \text{ kJ/kg-K}]$   
 $= -282,000 \text{ kJ/kmol}$

- $g_{CO_2,out} = 44.010 \text{ kg/kmol} * [-8,400 \text{ kJ/kg} - (550^\circ\text{C} + 273.15) * 5.88 \text{ kJ/kg-K}]$   
 $= -583,000 \text{ kJ/kmol}$

### B.4.3 Equilibrium Calculations

The equilibrium constant is calculated by applying Equation 3.64:

- $K_{wgs} = (0.25 * 0.36) / (0.12 * 0.21) = 3.66$

Thus, the variables required for the equilibrium condition, Equation 3.63, have been provided.

The following results for the LHS and RHS of this equation:

- $LHS = \ln(3.66) = 1.30$
- $RHS = -(-583,000 \text{ kJ/kmol} - 117,000 \text{ kJ/kmol} + 282,000 \text{ kJ/kmol} + 408,000 \text{ kJ/kmol}) / [8.31451 \text{ kJ/kmol-K} * (550^\circ\text{C} + 273.15)] = 1.30$

### B.5 Syngas Volume Percentages

Although the calculations for the syngas volume percentages are not discussed in Chapter 3, the demonstration of these is pertinent. This is accomplished with Equations A.39 through A.47, which are provided in Appendix A.5. The total outlet volume flow rates for  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are calculated by applying Equations A.39 through A.42:

- $\dot{V}_{H_2,out} = (0.894 \text{ kg/s} + 0.07 \text{ kg/s}) / (0.0852 \text{ kg/m}^3) = 11.4 \text{ m}_n^3/\text{s}$
- $\dot{V}_{CO,out} = (4.02 \text{ kg/s} + 0.5 \text{ kg/s}) / (1.18 \text{ kg/m}^3) = 3.78 \text{ m}_n^3/\text{s}$
- $\dot{V}_{CO_2,out} = (13.9 \text{ kg/s} + 0.9 \text{ kg/s}) / (1.87 \text{ kg/m}^3) = 7.93 \text{ m}_n^3/\text{s}$
- $\dot{V}_{CH_4,out} = (0.944 \text{ kg/s} + 0.5 \text{ kg/s}) / (0.680 \text{ kg/m}^3) = 2.06 \text{ m}_n^3/\text{s}$

The total volume flow rate of syngas is calculated by applying Equation A.43:

- $\dot{V}_{sgs} = 11.4 \text{ m}_n^3/\text{s} + 3.78 \text{ m}_n^3/\text{s} + 7.93 \text{ m}_n^3/\text{s} + 2.06 \text{ m}_n^3/\text{s} = 25.1 \text{ m}_n^3/\text{s}$

The respective volume percentages are calculated by applying Equations A.44 through A.47:

- $V_{H_2} = [(11.4 \text{ m}_n^3/\text{s}) / (25.1 \text{ m}_n^3/\text{s})] * 100 = 45.2\%$
- $V_{CO} = [(3.78 \text{ m}_n^3/\text{s}) / (25.1 \text{ m}_n^3/\text{s})] * 100 = 15.0\%$
- $V_{CO_2} = [(7.93 \text{ m}_n^3/\text{s}) / (25.1 \text{ m}_n^3/\text{s})] * 100 = 31.6\%$
- $V_{CH_4} = [(2.06 \text{ m}_n^3/\text{s}) / (25.1 \text{ m}_n^3/\text{s})] * 100 = 8.19\%$

## APPENDIX C – METHANATION EQUILIBRIUM ANALYSIS

This appendix provides an analysis of the equilibrium condition for the methanation reaction. Data from the verification calculations are applied in order to compare the results with those, in Section 4.6.1.3, for water gas shift equilibrium.

Ideal gas enthalpy and entropy for methane, as output from EES, are:

- $h_{CH_4,out,idl} = -3,000 \text{ kJ/kg}$
- $s_{CH_4,out,idl} = 14.6 \text{ kJ/kg-K}$

The Gibbs function for methane is calculated by applying Equation A.38:

- $g_{CH_4,out} = 16.043 \text{ kg/kmol} * [-3,000 \text{ kJ/kg} - (550^\circ\text{C} + 273.15) * 14.6 \text{ kJ/kg-K}]$   
 $= -242,000 \text{ kJ/kmol}$

The equilibrium constant for methanation is calculated by Applying Equation 3.66:

- $K_{meth} = [(0.07 * 0.21) / (0.12 * 0.36^3)] * (2,850 \text{ kPa} / 100 \text{ kPa})^{-2} = 0.00302$

Thus, the variables required for the methanation equilibrium condition, Equation 3.65, have been provided. The following results for the LHS and RHS of this equation:

- $LHS = \ln(0.00302) = -5.80$
- $RHS = -(-242,000 \text{ kJ/kmol} - 408,000 \text{ kJ/kmol} + 282,000 \text{ kJ/kmol} + 3 * 117,000 \text{ kJ/kmol}) / [8.31451 \text{ kJ/kmol-K} * (550^\circ\text{C} + 273.15)] = 2.55$

Therefore, the predicted respective mole fractions for  $H_2$ ,  $CO$ ,  $CH_4$ , and  $H_2O$  do not satisfy the methanation reaction's equilibrium condition. As such, it is evident that this equilibrium condition is unsuitable for application in the thermal-fluid model. This further validates the selection of the water gas shift reaction's equilibrium condition.

## APPENDIX D – SUPPLEMENTAL PLOTS WITH CLUSTERS

This appendix contains supplemental mass flow rate plots, wherein observations are colored according to the inlet flow rate groups discussed in Section 5.3. These plots are provided since, although the flow rates presented therein are discussed in Section 5.3, those flow rates are not available in that section. Figures D.1 through D.3 are these plots, where the  $H_2$  to  $CO$ ,  $CH_4$  to  $CO$ , and  $CH_4$  to  $CO_2$  outlet mass flow rates are graphed, respectively.

From Figure 5.14 and Figures D.1 through D.3, the relative differences between inlet flow rate groups are evident. As such, in Tables D.1 and D.2, the sign of the difference between average flow rates, for specific group pairs, is provided. In these tables, + represents that the respective average flow rate of the group listed first, in the heading, is larger than that of the second group. Correspondingly, - represents that the respective average flow of the group listed second is larger than that of the first group.

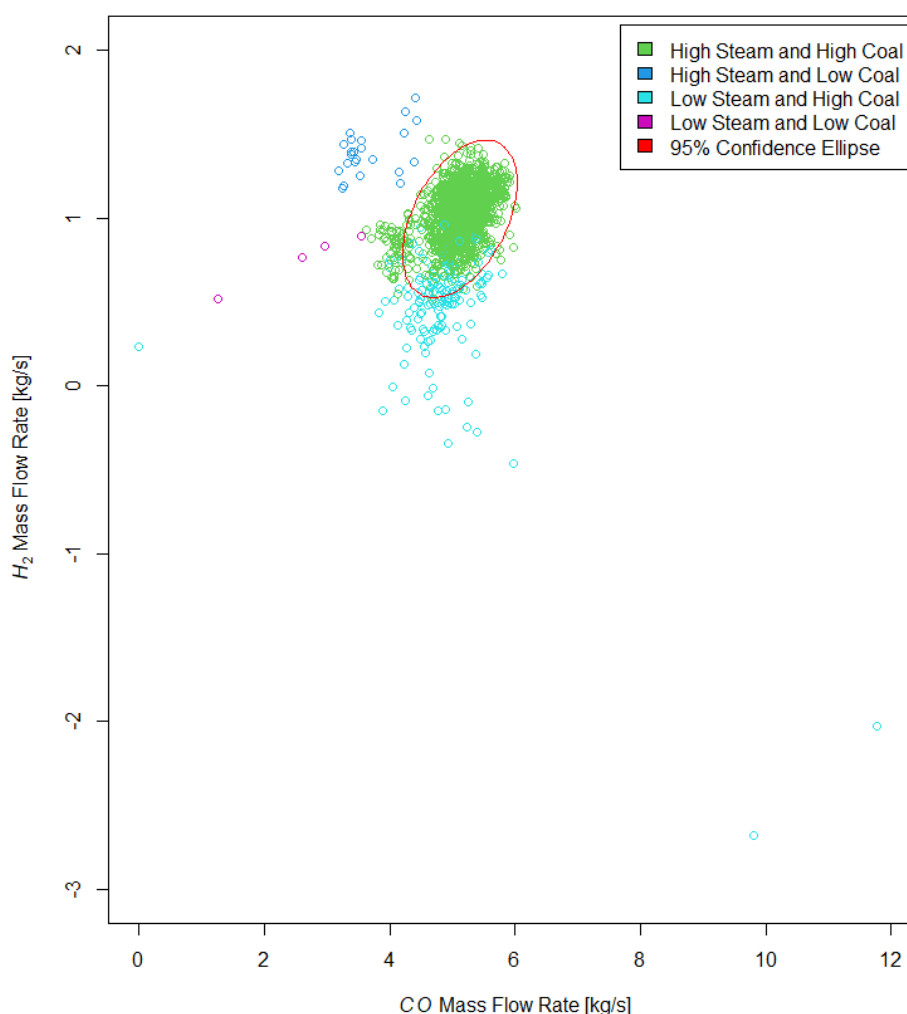


Figure D.1:  $H_2$  and  $CO$  Plot with Inlet Flow Rate Groups

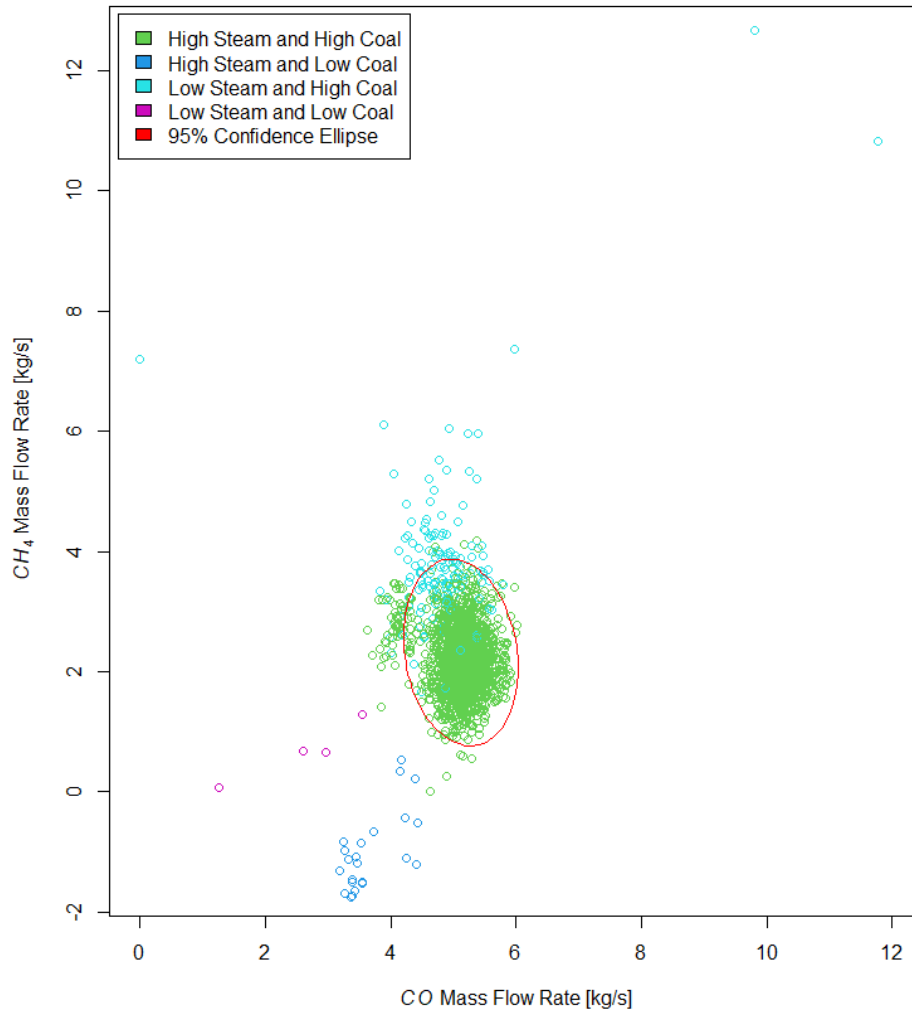


Figure D.2:  $CH_4$  and CO Plot with Inlet Flow Rate Groups

Table D.1: Relative Flow Rate Changes for Decreases in Coal

| $H_2O$                                                 | $H_2$ | CO | $CO_2$ | $CH_4$ |
|--------------------------------------------------------|-------|----|--------|--------|
| (High steam and high coal) – (High steam and low coal) |       |    |        |        |
| -                                                      | -     | +  | +      | +      |
| (Low steam and high coal) – (Low steam and low coal)   |       |    |        |        |
| -                                                      | -     | +  | +      | +      |

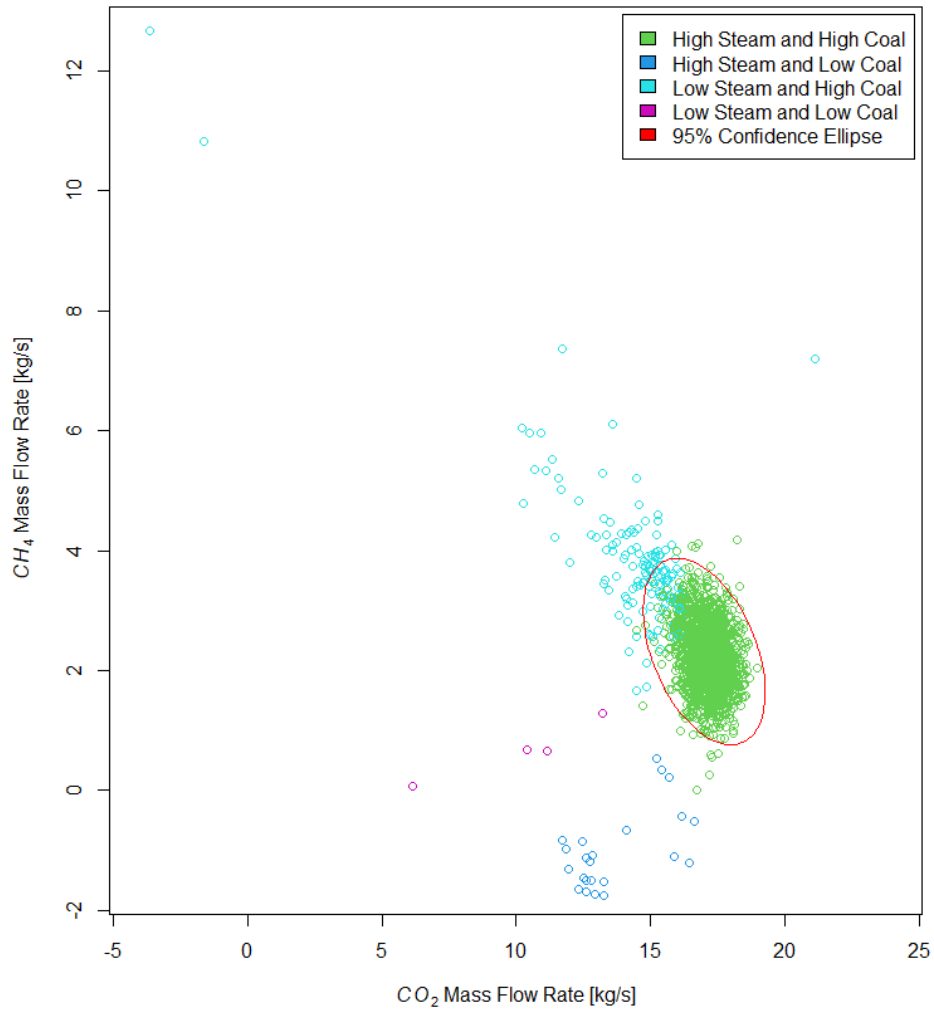


Figure D.3:  $CH_4$  and  $CO_2$  Plot with Inlet Flow Rate Groups

Table D.2: Relative Flow Rate Changes for Decreases in Steam

| $H_2O$                                                 | $H_2$ | $CO$ | $CO_2$ | $CH_4$ |
|--------------------------------------------------------|-------|------|--------|--------|
| (High steam and high coal) – (Low steam and high coal) |       |      |        |        |
| +                                                      | +     | +    | +      | -      |
| (High steam and low coal) – (Low steam and low coal)   |       |      |        |        |
| +                                                      | +     | +    | +      | -      |

## APPENDIX E – EES CODE FOR MODEL A

This appendix contains the EES code for thermal-fluid model A.

```
{EES code to calculate the mass flow rates in the raw syngas}
$UnitSystem SI C kPa kJ mass

{Read number of entries}
$Import 'Nm_Rows_A.csv' /skiplines=1 nm_rows

{Read normal temperature and pressure for volume flow rate conversions}
$Import 'Norm_TP.csv' /skiplines=1 N_TP[1..2]
{N_TP[1] normal temperature [C], N_TP[2] normal pressure [kPa]}

{Read input properties}
$Import 'Input_Properties_A.csv' /skiplines=1 Prop_in[1..nm_rows,1..14]
{Prop_in[i,1] inlet steam and oxygen temperature [C], Prop_in[i,2] inlet steam and oxygen pressure [kPa]}
{Prop_in[i,3] inlet coal temperature [C], Prop_in[i,4] outlet ash temperature [C]}
{Prop_in[i,5] inlet waterjacket feedwater temperature [C], Prop_in[i,6] outlet waterjacket steam temperature [C], Prop_in[i,7] waterjacket pressure [kPa]}
{Prop_in[i,8] outlet raw syngas temperature [C], Prop_in[i,9] outlet raw syngas pressure [kPa]}
{Prop_in[i,10] inlet steam mass flow rate [tonne/hr], Prop_in[i,11] inlet oxygen normal volume flow rate [kNm^3/hr]}
{Prop_in[i,12] inlet coal mass flow rate [tonne/hr]}
{Prop_in[i,13] waterjacket feedwater mass flow rate [tonne/hr]}
{Prop_in[i,14] grate rotational frequency [rev/hr]}

{Function for heat capacity of coal}
Function cp_coal(T_cl, VM_cl, WT_cl)
{Heat capacity of water}
cp_wt = 4.187

IF(T_cl < 400) Then
cp_waf = 0.83*(1 + 0.08*VM_cl)*(1 + 0.2*T_cl/100 - 0.0033*(T_cl/100)^3)
Else
cp_waf = 0.83*(1 + 0.08*VM_cl)*(1 + 0.2*4 - 0.0033*4^3) + 0.00473*(T_cl - 400)^0.726
Endif

cp_coal = cp_waf*(1 - WT_cl) + cp_wt*WT_cl
End

{Function for enthalpy of carbon monoxide}
Function h_en_co(T_r, P_r, h_r_co, T_co, P_t, y_co)
{Upper temperature limit of thermodynamic tables for carbon monoxide}
T_cr_co = 226.85

If(T_co <= T_cr_co) Then
h_en_co = h_r_co + enthalpy(CarbonMonoxide, T = T_co, P = y_co*P_t) -
enthalpy(CarbonMonoxide, T = T_r, P = P_r)
Else
h_en_co = h_r_co + enthalpy(CarbonMonoxide, T = T_cr_co, P = y_co*P_t) +
cp(CarbonMonoxide, T = T_cr_co, P = y_co*P_t)*(T_co - T_cr_co) - enthalpy(CarbonMonoxide, T =
T_r, P = P_r)
Endif
End
```

{Function for enthalpy of methane}

**Function** h\_en\_ch4(T\_r, P\_r, h\_r\_ch4, T\_ch4, P\_t, y\_ch4)

{Upper temperature limit of thermodynamic tables for methane}

T\_cr\_ch4 = 351.85

If(T\_ch4 <= T\_cr\_ch4) **Then**

**h\_en\_ch4** = h\_r\_ch4 + **enthalpy**(Methane, T = T\_ch4, P = y\_ch4\*P\_t) - **enthalpy**(Methane, T = T\_r, P = P\_r)

**Else**

**h\_en\_ch4** = h\_r\_ch4 + **enthalpy**(Methane, T = T\_cr\_ch4, P = y\_ch4\*P\_t) + **cp**(Methane, T = T\_cr\_ch4, P = y\_ch4\*P\_t)\*(T\_ch4 - T\_cr\_ch4) - **enthalpy**(Methane, T = T\_r, P = P\_r)

**Endif**

**End**

{Reference temperature [C] and pressure [kPa] for formation enthalpies and standard entropies}

T\_ref = 25

P\_ref = 100

{Universal gas constant [kJ/kmol-K]}

R\_ugc = 8.31451

{Conversion factor for ash grate rotational frequency to ash mass removal rate [tonne/rev]}

grt\_as\_rm = 3

{Normal densities for oxygen, hydrogen, carbon monoxide, carbon dioxide, and methane [kg/m^3]}

rho\_o2\_n = **density**(Oxygen, T = N\_TP[1], P = N\_TP[2])

rho\_h2\_n = **density**(Hydrogen, T = N\_TP[1], P = N\_TP[2])

rho\_co\_n = **density**(CarbonMonoxide, T = N\_TP[1], P = N\_TP[2])

rho\_co2\_n = **density**(CarbonDioxide, T = N\_TP[1], P = N\_TP[2])

rho\_ch4\_n = **density**(Methane, T = N\_TP[1], P = N\_TP[2])

{Molar masses [kg/kmol]}

Mol\_c = 12.01115

Mol\_s = 32.064

Mol\_h = 1.00797

Mol\_o2 = **molar mass**(Oxygen)

Mol\_h2 = **molar mass**(Hydrogen)

Mol\_h2o = **molar mass**(Water)

Mol\_co = **molar mass**(CarbonMonoxide)

Mol\_co2 = **molar mass**(CarbonDioxide)

Mol\_ch4 = **molar mass**(Methane)

Mol\_so2 = **molar mass**(SulfurDioxide)

{As received mass fraction fixed carbon, volatile matter, ash, and water in coal [-]}

ar\_fr\_fc\_coal = 0.471

ar\_fr\_vm\_coal = 0.218

ar\_fr\_as\_coal = 0.275

ar\_fr\_w\_coal = 0.036

{As received mass fraction fixed carbon in ash [-]}

ar\_fr\_fc\_ash = 0.081

{Water free mass fraction fixed carbon and volatile matter in coal [-]}

wf\_fr\_fc\_coal = 0.489

wf\_fr\_vm\_coal = 0.227

{Water and ash free mass fraction volatile matter in coal [-]}

waf\_fr\_vm\_coal = ((1 + ar\_fr\_fc\_coal/ar\_fr\_vm\_coal)^(-1) + (1 + wf\_fr\_fc\_coal/wf\_fr\_vm\_coal)^(-1))/2

{Ash free mass fraction water in coal [-]}

$$af\_fr\_w\_coal = (ar\_fr\_fc\_coal/ar\_fr\_w\_coal + ar\_fr\_vm\_coal/ar\_fr\_w\_coal + 1)^{-1}$$

{Air dried mass fraction tar after devolatilization at 720 C [-]}

$$ad\_fr\_720\_tr\_devl = 0.031$$

{Normal volume flows of gaseous volatile matter per kilogram of water and ash free coal after devolatilization at 720 C [Nm<sup>3</sup>/kg]}

$$vf\_720\_h2\_gs\_vm = 0.09$$

$$vf\_720\_co\_gs\_vm = 0.04$$

$$vf\_720\_co2\_gs\_vm = 0.05$$

$$vf\_720\_ch4\_gs\_vm = 0.07$$

{Water and ash free mass fraction carbon, sulfur, hydrogen, oxygen, and nitrogen in coal [-]}

$$waf\_fr\_c\_coal = 0.795$$

$$waf\_fr\_s\_coal = 0.016$$

$$waf\_fr\_h\_coal = 0.042$$

$$waf\_fr\_o\_coal = 0.127$$

$$waf\_fr\_n\_coal = 0.021$$

{Water and ash free mass fraction carbon, sulfur, hydrogen in tar [-]}

$$waf\_fr\_c\_tar = 0.773$$

$$waf\_fr\_s\_tar = 0.014$$

$$waf\_fr\_h\_tar = 0.049$$

{Coal higher and lower heating value [kJ/kg]}

$$HHV = 1000 \cdot (35.2 \cdot waf\_fr\_c\_coal + 116.4 \cdot waf\_fr\_h\_coal - 11.1 \cdot waf\_fr\_o\_coal + 10.5 \cdot waf\_fr\_s\_coal + 6.29 \cdot waf\_fr\_n\_coal)$$

$$LHV = HHV - 2442 \cdot (9 \cdot waf\_fr\_h\_coal)$$

{Enthalpies of formation [kJ/kg]}

$$h\_h2o\_L\_0 = \text{enthalpy\_formation}(H2O)$$

$$h\_h2o\_V\_0 = \text{enthalpy}(H2O, T = T\_ref)$$

$$h\_o2\_0 = \text{enthalpy}(O2, T = T\_ref)$$

$$h\_h2\_0 = \text{enthalpy}(H2, T = T\_ref)$$

$$h\_co\_0 = \text{enthalpy}(CO, T = T\_ref)$$

$$h\_co2\_0 = \text{enthalpy}(CO2, T = T\_ref)$$

$$h\_ch4\_0 = \text{enthalpy}(CH4, T = T\_ref)$$

$$h\_so2\_0 = \text{enthalpy}(SO2, T = T\_ref)$$

{Enthalpy of formation of coal}

$$h\_coal\_0 = h\_coal\_com \cdot (1 - f\_com) \cdot (1 - af\_fr\_w\_coal) + h\_h2o\_L\_0 \cdot af\_fr\_w\_coal$$

$$h\_coal\_com = 1000 \cdot (-32.7633 \cdot waf\_fr\_c\_coal - 141.7892 \cdot waf\_fr\_h\_coal - 9.2768 \cdot waf\_fr\_s\_coal)$$

$$f\_com = 0.01379 \cdot \ln(waf\_fr\_o\_coal) - 0.01393 \cdot waf\_fr\_o\_coal^{0.75} + 1.02111$$

{Gasifier heat loss}

$$Ht\_loss = 0.17$$

$$Q\_HV\_loss = Ht\_loss \cdot LHV$$

{Calculate enthalpy}

**Duplicate** i = 1,nm\_rows

{Inlet steam and oxygen mass flow rates [kg/s]}

$$m\_dot\_stm\_in[i] = \text{convert}(\text{tonne/hr, kg/s}) \cdot \text{Prop\_in}[i, 10]$$

$$m\_dot\_o2\_in[i] = 1000 \cdot \text{convert}(1/\text{hr}, 1/\text{s}) \cdot \text{Prop\_in}[i, 11] \cdot \rho_{o2\_n}$$

{Inlet steam and oxygen mole flow rates [kmol/s]}

$$mol\_dot\_stm\_in[i] = m\_dot\_stm\_in[i] \cdot Mol\_h2o^{(-1)}$$

$$mol\_dot\_o2\_in[i] = m\_dot\_o2\_in[i] \cdot Mol\_o2^{(-1)}$$

{Feedwater mass flow rate [kg/s]}

$m\_dot\_w[i] = \text{convert}(\text{tonne/hr, kg/s}) * \text{Prop\_in}[i, 13]$

{Coal and inlet fixed carbon mass flow rates [kg/s]}

$m\_dot\_coal[i] = \text{convert}(\text{tonne/hr, kg/s}) * \text{Prop\_in}[i, 12]$

$m\_dot\_fc\_in[i] = ar\_fr\_fc\_coal * m\_dot\_coal[i]$

{Ash and outlet fixed carbon mass flow rates [kg/s]}

$m\_dot\_ash[i] = \text{convert}(\text{tonne/hr, kg/s}) * grt\_as\_rm * \text{Prop\_in}[i, 14]$

$m\_dot\_fc\_out[i] = ar\_fr\_fc\_ash * m\_dot\_ash[i]$

{Inlet and outlet fixed carbon mole flow rates [kmol/s]}

$mol\_dot\_fc\_in[i] = (m\_dot\_fc\_in[i] * Mol\_c)^{-1}$

$mol\_dot\_fc\_out[i] = (m\_dot\_fc\_out[i] * Mol\_c)^{-1}$

{Volatile matter mass flow rates [kg/s]}

$m\_dot\_tar\_vm[i] = m\_dot\_coal[i] * ad\_fr\_720\_tr\_devl$

$m\_dot\_gs\_vm\_h2[i] = m\_dot\_coal[i] * (ar\_fr\_fc\_coal + ar\_fr\_vm\_coal) * vf\_720\_h2\_gs\_vm * rho\_h2\_n$

$m\_dot\_gs\_vm\_co[i] = m\_dot\_coal[i] * (ar\_fr\_fc\_coal + ar\_fr\_vm\_coal) * vf\_720\_co\_gs\_vm * rho\_co\_n$

$m\_dot\_gs\_vm\_co2[i] = m\_dot\_coal[i] * (ar\_fr\_fc\_coal + ar\_fr\_vm\_coal) * vf\_720\_co2\_gs\_vm * rho\_co2\_n$

$m\_dot\_gs\_vm\_ch4[i] = m\_dot\_coal[i] * (ar\_fr\_fc\_coal + ar\_fr\_vm\_coal) * vf\_720\_ch4\_gs\_vm * rho\_ch4\_n$

{Coal evaporated water mass flow rate [kg/s]}

$m\_dot\_coal\_w[i] = m\_dot\_coal[i] * ar\_fr\_w\_coal$

{Coal evaporated water mole flow rate [kmol/s]}

$mol\_dot\_coal\_w[i] = m\_dot\_coal\_w[i] * Mol\_h2o^{(-1)}$

{Tar carbon, sulfur, hydrogen mass flow rates [kg/s]}

$m\_dot\_c\_tar[i] = waf\_fr\_c\_tar * m\_dot\_tar\_vm[i]$

$m\_dot\_s\_tar[i] = waf\_fr\_s\_tar * m\_dot\_tar\_vm[i]$

$m\_dot\_h\_tar[i] = waf\_fr\_h\_tar * m\_dot\_tar\_vm[i]$

{Tar carbon, sulfur, hydrogen mole flow rates [kmol/s]}

$mol\_dot\_c\_tar[i] = m\_dot\_c\_tar[i] * Mol\_c^{(-1)}$

$mol\_dot\_s\_tar[i] = m\_dot\_s\_tar[i] * Mol\_s^{(-1)}$

$mol\_dot\_h\_tar[i] = m\_dot\_h\_tar[i] * Mol\_h^{(-1)}$

{Raw syngas mole flow rates [kmol/s]}

$mol\_dot\_rw\_gs\_h2o[i] = m\_dot\_rw\_gs\_h2o[i] * Mol\_h2o^{(-1)}$

$mol\_dot\_rw\_gs\_h2[i] = m\_dot\_rw\_gs\_h2[i] * Mol\_h2^{(-1)}$

$mol\_dot\_rw\_gs\_co[i] = m\_dot\_rw\_gs\_co[i] * Mol\_co^{(-1)}$

$mol\_dot\_rw\_gs\_co2[i] = m\_dot\_rw\_gs\_co2[i] * Mol\_co2^{(-1)}$

$mol\_dot\_rw\_gs\_ch4[i] = m\_dot\_rw\_gs\_ch4[i] * Mol\_ch4^{(-1)}$

{Gaseous volatile matter mole flow rates [kmol/s]}

$mol\_dot\_gs\_vm\_h2[i] = m\_dot\_gs\_vm\_h2[i] * Mol\_h2^{(-1)}$

$mol\_dot\_gs\_vm\_co[i] = m\_dot\_gs\_vm\_co[i] * Mol\_co^{(-1)}$

$mol\_dot\_gs\_vm\_co2[i] = m\_dot\_gs\_vm\_co2[i] * Mol\_co2^{(-1)}$

$mol\_dot\_gs\_vm\_ch4[i] = m\_dot\_gs\_vm\_ch4[i] * Mol\_ch4^{(-1)}$

{Input steam and oxygen mole fractions [-]}

$tot\_mol\_in[i] = mol\_dot\_stm\_in[i] + mol\_dot\_o2\_in[i]$

$y\_stm\_in[i] = mol\_dot\_stm\_in[i] / tot\_mol\_in[i]$

$y\_o2\_in[i] = mol\_dot\_o2\_in[i] / tot\_mol\_in[i]$

{Raw syngas mole fractions [-]}

```

tot_mol_rw_gs[i] = mol_dot_rw_gs_h2o[i] + mol_dot_rw_gs_h2[i] + mol_dot_rw_gs_co[i] +
mol_dot_rw_gs_co2[i] + mol_dot_rw_gs_ch4[i] + mol_dot_gs_vm_h2[i] + mol_dot_gs_vm_co[i] +
mol_dot_gs_vm_co2[i] + mol_dot_gs_vm_ch4[i] + mol_dot_coal_w[i]
y_rw_gs_h2o[i] = (mol_dot_rw_gs_h2o[i] + mol_dot_coal_w[i])/tot_mol_rw_gs[i]
y_rw_gs_h2[i] = (mol_dot_rw_gs_h2[i] + mol_dot_gs_vm_h2[i])/tot_mol_rw_gs[i]
y_rw_gs_co[i] = (mol_dot_rw_gs_co[i] + mol_dot_gs_vm_co[i])/tot_mol_rw_gs[i]
y_rw_gs_co2[i] = (mol_dot_rw_gs_co2[i] + mol_dot_gs_vm_co2[i])/tot_mol_rw_gs[i]
y_rw_gs_ch4[i] = (mol_dot_rw_gs_ch4[i] + mol_dot_gs_vm_ch4[i])/tot_mol_rw_gs[i]

```

#### {Raw syngas normal volume flow rates}

```

vf_rw_gs_h2[i] = (m_dot_rw_gs_h2[i] + m_dot_gs_vm_h2[i])*rho_h2_n^(-1)
vf_rw_gs_co[i] = (m_dot_rw_gs_co[i] + m_dot_gs_vm_co[i])*rho_co_n^(-1)
vf_rw_gs_co2[i] = (m_dot_rw_gs_co2[i] + m_dot_gs_vm_co2[i])*rho_co2_n^(-1)
vf_rw_gs_ch4[i] = (m_dot_rw_gs_ch4[i] + m_dot_gs_vm_ch4[i])*rho_ch4_n^(-1)

```

#### {Raw syngas volume flow rate percentages}

```

vf_per_h2[i] = convert(-, %)*vf_rw_gs_h2[i]/(vf_rw_gs_h2[i] + vf_rw_gs_co[i] + vf_rw_gs_co2[i] +
vf_rw_gs_ch4[i])
vf_per_co[i] = convert(-, %)*vf_rw_gs_co[i]/(vf_rw_gs_h2[i] + vf_rw_gs_co[i] + vf_rw_gs_co2[i] +
vf_rw_gs_ch4[i])
vf_per_co2[i] = convert(-, %)*vf_rw_gs_co2[i]/(vf_rw_gs_h2[i] + vf_rw_gs_co[i] + vf_rw_gs_co2[i] +
vf_rw_gs_ch4[i])
vf_per_ch4[i] = convert(-, %)*vf_rw_gs_ch4[i]/(vf_rw_gs_h2[i] + vf_rw_gs_co[i] + vf_rw_gs_co2[i] +
vf_rw_gs_ch4[i])

```

#### {Input steam and oxygen enthalpies [kJ/kg]}

```

h_stm_in[i] = h_h2o_V_0 + enthalpy(Water, T = Prop_in[i,1], P = y_stm_in[i]*Prop_in[i,2]) -
enthalpy(Water, T = T_ref, P = P_ref)
h_o2_in[i] = h_o2_0 + enthalpy(Oxygen, T = Prop_in[i,1], P = y_o2_in[i]*Prop_in[i,2]) -
enthalpy(Oxygen, T = T_ref, P = P_ref)

```

#### {Coal enthalpy for fixed carbon, volatile matter, and water}

```

h_coal[i] = h_coal_0 + cp_coal(Prop_in[i,3], waf_fr_vm_coal, af_fr_w_coal)*(Prop_in[i,3] - T_ref)

```

#### {Ash change in enthalpy}

```

delta_h_ash[i] = (0.752 + 0.293*((Prop_in[i,3] + Prop_in[i,4])/2 + 273.15)/1000)*(Prop_in[i,3] -
Prop_in[i,4])

```

#### {Tar power}

```

Q_tar[i] = m_dot_h_tar_0[i] + m_dot_cp_dT_tar[i]

```

```

m_dot_h_tar_0[i] = -m_dot_tar_vm[i]*30980 + mol_dot_c_tar[i]*h_co2_0*Mol_co2 +
mol_dot_s_tar[i]*h_so2_0*Mol_so2 + mol_dot_h_tar[i]*(h_h2o_L_0/2)*Mol_h2o
m_dot_cp_dT_tar[i] = m_dot_tar_vm[i]*cp_dT_tar[i]
cp_dT_tar[i] = 2.11*10^(-3)*(converttemp(C,K,Prop_in[i,8])^2 - converttemp(C,K,T_ref)^2)

```

#### {Water jacket feedwater and steam enthalpies [kJ/kg]}

```

h_wj_fw[i] = enthalpy(Water, T = Prop_in[i,5], P = Prop_in[i,7])
h_wj_stm[i] = enthalpy(Water, T = Prop_in[i,6], P = Prop_in[i,7])

```

#### {Raw syngas enthalpies [kJ/kg]}

```

h_rw_gs_h2o[i] = h_h2o_V_0 + enthalpy(Water, T = Prop_in[i,8], P = 0.123*Prop_in[i,9]) -
enthalpy(Water, T = T_ref, P = P_ref)
h_rw_gs_h2[i] = h_h2_0 + enthalpy(Hydrogen, T = Prop_in[i,8], P = 0.362*Prop_in[i,9]) -
enthalpy(Hydrogen, T = T_ref, P = P_ref)
h_rw_gs_co[i] = h_en_co(T_ref, P_ref, h_co_0, Prop_in[i,8], Prop_in[i,9], 0.132)
h_rw_gs_co2[i] = h_co2_0 + enthalpy(CarbonDioxide, T = Prop_in[i,8], P = 0.281*Prop_in[i,9]) -
enthalpy(CarbonDioxide, T = T_ref, P = P_ref)
h_rw_gs_ch4[i] = h_en_ch4(T_ref, P_ref, h_ch4_0, Prop_in[i,8], Prop_in[i,9], 0.103)

```

{Raw syngas ideal gas enthalpies [kJ/kg]}

$h_{ideal\_rw\_gs\_h2o[i]} = \text{enthalpy}(H2O, T = \text{Prop\_in}[i,8])$   
 $h_{ideal\_rw\_gs\_h2[i]} = \text{enthalpy}(H2, T = \text{Prop\_in}[i,8])$   
 $h_{ideal\_rw\_gs\_co[i]} = \text{enthalpy}(CO, T = \text{Prop\_in}[i,8])$   
 $h_{ideal\_rw\_gs\_co2[i]} = \text{enthalpy}(CO2, T = \text{Prop\_in}[i,8])$

{Raw syngas ideal gas entropies [kJ/kg-K]}

$s_{ideal\_rw\_gs\_h2o[i]} = \text{entropy}(H2O, T = \text{Prop\_in}[i,8], P = P\_ref)$   
 $s_{ideal\_rw\_gs\_h2[i]} = \text{entropy}(H2, T = \text{Prop\_in}[i,8], P = P\_ref)$   
 $s_{ideal\_rw\_gs\_co[i]} = \text{entropy}(CO, T = \text{Prop\_in}[i,8], P = P\_ref)$   
 $s_{ideal\_rw\_gs\_co2[i]} = \text{entropy}(CO2, T = \text{Prop\_in}[i,8], P = P\_ref)$

{Gibbs functions [kJ/kmol]}

$g_{h2o[i]} = (\text{Mol\_h2o}) * (h_{ideal\_rw\_gs\_h2o[i]} - \text{converttemp}(C,K,\text{Prop\_in}[i,8]) * s_{ideal\_rw\_gs\_h2o[i]})$   
 $g_{h2[i]} = (\text{Mol\_h2}) * (h_{ideal\_rw\_gs\_h2[i]} - \text{converttemp}(C,K,\text{Prop\_in}[i,8]) * s_{ideal\_rw\_gs\_h2[i]})$   
 $g_{co[i]} = (\text{Mol\_co}) * (h_{ideal\_rw\_gs\_co[i]} - \text{converttemp}(C,K,\text{Prop\_in}[i,8]) * s_{ideal\_rw\_gs\_co[i]})$   
 $g_{co2[i]} = (\text{Mol\_co2}) * (h_{ideal\_rw\_gs\_co2[i]} - \text{converttemp}(C,K,\text{Prop\_in}[i,8]) * s_{ideal\_rw\_gs\_co2[i]})$

{Energy balance}

$Q\_wj[i] = m\_dot\_wj[i] * (h\_wj\_fw[i] - h\_wj\_stm[i])$

$m\_dot\_stm\_in[i] * h\_stm\_in[i] + m\_dot\_o2\_in[i] * h\_o2\_in[i] + (m\_dot\_coal[i] - m\_dot\_ash[i]) * h\_coal[i] +$   
 $m\_dot\_ash[i] * \Delta h\_ash[i] + Q\_wj[i] = (m\_dot\_rw\_gs\_h2o[i] + m\_dot\_coal\_w[i]) * h\_rw\_gs\_h2o[i] +$   
 $(m\_dot\_rw\_gs\_h2[i] + m\_dot\_gs\_vm\_h2[i]) * h\_rw\_gs\_h2[i] + (m\_dot\_rw\_gs\_co[i] +$   
 $m\_dot\_gs\_vm\_co[i]) * h\_rw\_gs\_co[i] + (m\_dot\_rw\_gs\_co2[i] + m\_dot\_gs\_vm\_co2[i]) * h\_rw\_gs\_co2[i] +$   
 $(m\_dot\_rw\_gs\_ch4[i] + m\_dot\_gs\_vm\_ch4[i]) * h\_rw\_gs\_ch4[i] + Q\_tar[i] + (ar\_fr\_fc\_coal +$   
 $ar\_fr\_vm\_coal) * m\_dot\_coal[i] * Q\_HV\_loss$

{Hydrogen balance}

$2 * m\_dot\_stm\_in[i] = 2 * m\_dot\_rw\_gs\_h2o[i] + 2 * m\_dot\_rw\_gs\_h2[i] + 4 * m\_dot\_rw\_gs\_ch4[i]$

{Oxygen balance}

$1 * m\_dot\_stm\_in[i] + 2 * m\_dot\_o2\_in[i] = 1 * m\_dot\_rw\_gs\_h2o[i] + 1 * m\_dot\_rw\_gs\_co[i] +$   
 $2 * m\_dot\_rw\_gs\_co2[i]$

{Carbon balance}

$1 * m\_dot\_fc\_in[i] = 1 * m\_dot\_fc\_out[i] + 1 * m\_dot\_rw\_gs\_co[i] + 1 * m\_dot\_rw\_gs\_co2[i] +$   
 $1 * m\_dot\_rw\_gs\_ch4[i]$

{Equilibrium condition}

$\ln(K1\_eq[i]) = -(g\_co2[i] + g\_h2[i] - g\_co[i] - g\_h2o[i]) / (R\_ugc * \text{converttemp}(C,K,\text{Prop\_in}[i,8]))$   
 $K1\_eq\_x[i] = (y\_rw\_gs\_co2[i] * y\_rw\_gs\_h2[i]) / (y\_rw\_gs\_co[i] * y\_rw\_gs\_h2o[i])$   
 $K1\_eq[i] = K1\_eq\_x[i]$

{Total raw syngas mass flow rates for export [kg/s]}

$m\_dot\_tot\_rw\_gs\_h2o[i] = m\_dot\_rw\_gs\_h2o[i] + m\_dot\_coal\_w[i]$   
 $m\_dot\_tot\_rw\_gs\_h2[i] = m\_dot\_rw\_gs\_h2[i] + m\_dot\_gs\_vm\_h2[i]$   
 $m\_dot\_tot\_rw\_gs\_co[i] = m\_dot\_rw\_gs\_co[i] + m\_dot\_gs\_vm\_co[i]$   
 $m\_dot\_tot\_rw\_gs\_co2[i] = m\_dot\_rw\_gs\_co2[i] + m\_dot\_gs\_vm\_co2[i]$   
 $m\_dot\_tot\_rw\_gs\_ch4[i] = m\_dot\_rw\_gs\_ch4[i] + m\_dot\_gs\_vm\_ch4[i]$

**End**

{Sum of squared errors applied when varying the fraction of heat lost}

$\{SSE = (39.7 - vf\_per\_h2[1])^2 + (23 - vf\_per\_co[1])^2 + (28.3 - vf\_per\_co2[1])^2 + (8.8 -$   
 $vf\_per\_ch4[1])^2\}$

{Relative errors applied when adjusting the fraction of heat lost}

$RE\_h2 = \text{convert}(-, \%) * \text{abs}(39.7 - vf\_per\_h2[1]) / 39.7$   
 $RE\_co = \text{convert}(-, \%) * \text{abs}(23 - vf\_per\_co[1]) / 23$

```

RE_co2 = convert(-, %)*abs(28.3 - vf_per_co2[1])/28.3
RE_ch4 = convert(-, %)*abs(8.8 - vf_per_ch4[1])/8.8}

$Export /A /D 'STEAM_MASS_FLOW_RATE_A.csv' m_dot_tot_rw_gs_h2o[1..nm_rows]
$Export /A /D 'HYDROGEN_MASS_FLOW_RATE_A.csv' m_dot_tot_rw_gs_h2[1..nm_rows]
$Export /A /D 'CARBON_MONOXIDE_MASS_FLOW_RATE_A.csv'
m_dot_tot_rw_gs_co[1..nm_rows]
$Export /A /D 'CARBON_DIOXIDE_MASS_FLOW_RATE_A.csv' m_dot_tot_rw_gs_co2[1..nm_rows]
$Export /A /D 'METHANE_MASS_FLOW_RATE_A.csv' m_dot_tot_rw_gs_ch4[1..nm_rows]

$Export /A /D 'HYDROGEN_PERCENT_VOLUME_FLOW_RATE_A.csv' vf_per_h2[1..nm_rows]
$Export /A /D 'CARBON_MONOXIDE_PERCENT_VOLUME_FLOW_RATE_A.csv'
vf_per_co[1..nm_rows]
$Export /A /D 'CARBON_DIOXIDE_PERCENT_VOLUME_FLOW_RATE_A.csv'
vf_per_co2[1..nm_rows]
$Export /A /D 'METHANE_PERCENT_VOLUME_FLOW_RATE_A.csv' vf_per_ch4[1..nm_rows]

```

## APPENDIX F – EES CODE FOR MODEL B

This appendix contains the EES code for thermal-fluid model B.

```
{EES code to calculate the mass flow rates in the raw syngas}
$UnitSystem SI C kPa kJ mass

{Read number of entries}
$Import 'Nm_Rows_B.csv' /skiplines=1 nm_rows

{Read normal temperature and pressure for volume flow rate conversions}
$Import 'Norm_TP.csv' /skiplines=1 N_TP[1..2]
{N_TP[1] normal temperature [C], N_TP[2] normal pressure [kPa]}

{Read input properties}
$Import 'Input_Properties_B.csv' /skiplines=1 Prop_in[1..nm_rows,1..14]
{Prop_in[i,1] inlet steam and oxygen temperature [C], Prop_in[i,2] inlet steam and oxygen pressure [kPa]}
{Prop_in[i,3] inlet coal temperature [C], Prop_in[i,4] outlet ash temperature [C]}
{Prop_in[i,5] inlet waterjacket feedwater temperature [C], Prop_in[i,6] outlet waterjacket steam temperature [C], Prop_in[i,7] waterjacket pressure [kPa]}
{Prop_in[i,8] outlet raw syngas temperature [C], Prop_in[i,9] outlet raw syngas pressure [kPa]}
{Prop_in[i,10] inlet steam mass flow rate [tonne/hr], Prop_in[i,11] inlet oxygen normal volume flow rate [kNm^3/hr]}
{Prop_in[i,12] inlet coal mass flow rate [tonne/hr]}
{Prop_in[i,13] waterjacket feedwater mass flow rate [tonne/hr]}
{Prop_in[i,14] grate rotational frequency [rev/hr]}

{Function for heat capacity of coal}
Function cp_coal(T_cl, VM_cl, WT_cl)
{Heat capacity of water}
cp_wt = 4.187

IF(T_cl < 400) Then
cp_waf = 0.83*(1 + 0.08*VM_cl)*(1 + 0.2*T_cl/100 - 0.0033*(T_cl/100)^3)
Else
cp_waf = 0.83*(1 + 0.08*VM_cl)*(1 + 0.2*4 - 0.0033*4^3) + 0.00473*(T_cl - 400)^0.726
Endif

cp_coal = cp_waf*(1 - WT_cl) + cp_wt*WT_cl
End

{Function for enthalpy of carbon monoxide}
Function h_en_co(T_r, P_r, h_r_co, T_co, P_t, y_co)
{Upper temperature limit of thermodynamic tables for carbon monoxide}
T_cr_co = 226.85

If(T_co <= T_cr_co) Then
h_en_co = h_r_co + enthalpy(CarbonMonoxide, T = T_co, P = y_co*P_t) -
enthalpy(CarbonMonoxide, T = T_r, P = P_r)
Else
h_en_co = h_r_co + enthalpy(CarbonMonoxide, T = T_cr_co, P = y_co*P_t) +
cp(CarbonMonoxide, T = T_cr_co, P = y_co*P_t)*(T_co - T_cr_co) - enthalpy(CarbonMonoxide, T =
T_r, P = P_r)
Endif
End
```

{Function for enthalpy of methane}

**Function** h\_en\_ch4(T\_r, P\_r, h\_r\_ch4, T\_ch4, P\_t, y\_ch4)

{Upper temperature limit of thermodynamic tables for methane}

T\_cr\_ch4 = 351.85

If(T\_ch4 <= T\_cr\_ch4) **Then**

**h\_en\_ch4** = h\_r\_ch4 + **enthalpy**(*Methane*, T = T\_ch4, P = y\_ch4\*P\_t) - **enthalpy**(*Methane*, T = T\_r, P = P\_r)

**Else**

**h\_en\_ch4** = h\_r\_ch4 + **enthalpy**(*Methane*, T = T\_cr\_ch4, P = y\_ch4\*P\_t) + **cp**(*Methane*, T = T\_cr\_ch4, P = y\_ch4\*P\_t)\*(T\_ch4 - T\_cr\_ch4) - **enthalpy**(*Methane*, T = T\_r, P = P\_r)

**Endif**

**End**

{Reference temperature [C] and pressure [kPa] for formation enthalpies and standard entropies}

T\_ref = 25

P\_ref = 100

{Universal gas constant [kJ/kmol-K]}

R\_ugc = 8.31451

{Conversion factor for ash grate rotational frequency to ash mass removal rate [tonne/rev]}

grt\_as\_rm = 3

{Normal densities for oxygen, hydrogen, carbon monoxide, carbon dioxide, and methane [kg/m^3]}

rho\_o2\_n = **density**(*Oxygen*, T = N\_TP[1], P = N\_TP[2])

rho\_h2\_n = **density**(*Hydrogen*, T = N\_TP[1], P = N\_TP[2])

rho\_co\_n = **density**(*CarbonMonoxide*, T = N\_TP[1], P = N\_TP[2])

rho\_co2\_n = **density**(*CarbonDioxide*, T = N\_TP[1], P = N\_TP[2])

rho\_ch4\_n = **density**(*Methane*, T = N\_TP[1], P = N\_TP[2])

{Molar masses [kg/kmol]}

Mol\_c = 12.01115

Mol\_s = 32.064

Mol\_h = 1.00797

Mol\_o2 = **molar mass**(*Oxygen*)

Mol\_h2 = **molar mass**(*Hydrogen*)

Mol\_h2o = **molar mass**(*Water*)

Mol\_co = **molar mass**(*CarbonMonoxide*)

Mol\_co2 = **molar mass**(*CarbonDioxide*)

Mol\_ch4 = **molar mass**(*Methane*)

Mol\_so2 = **molar mass**(*SulfurDioxide*)

{As received mass fraction fixed carbon, volatile matter, ash, and water in coal [-]}

ar\_fr\_fc\_coal = 0.471

ar\_fr\_vm\_coal = 0.218

ar\_fr\_as\_coal = 0.275

ar\_fr\_w\_coal = 0.036

{As received mass fraction fixed carbon in ash [-]}

ar\_fr\_fc\_ash = 0.081

{Water free mass fraction fixed carbon and volatile matter in coal [-]}

wf\_fr\_fc\_coal = 0.489

wf\_fr\_vm\_coal = 0.227

{Water and ash free mass fraction volatile matter in coal [-]}

waf\_fr\_vm\_coal = ((1 + ar\_fr\_fc\_coal/ar\_fr\_vm\_coal)^(-1) + (1 + wf\_fr\_fc\_coal/wf\_fr\_vm\_coal)^(-1))/2

{Ash free mass fraction water in coal [-]}

$$af\_fr\_w\_coal = (ar\_fr\_fc\_coal/ar\_fr\_w\_coal + ar\_fr\_vm\_coal/ar\_fr\_w\_coal + 1)^{-1}$$

{Air dried mass fraction tar after devolatilization at 720 C [-]}

$$ad\_fr\_720\_tr\_devl = 0.031$$

{Normal volume flows of gaseous volatile matter per kilogram of water and ash free coal after devolatilization at 720 C [Nm<sup>3</sup>/kg]}

$$vf\_720\_h2\_gs\_vm = 0.09$$

$$vf\_720\_co\_gs\_vm = 0.04$$

$$vf\_720\_co2\_gs\_vm = 0.05$$

$$vf\_720\_ch4\_gs\_vm = 0.07$$

{Water and ash free mass fraction carbon, sulfur, hydrogen, oxygen, and nitrogen in coal [-]}

$$waf\_fr\_c\_coal = 0.795$$

$$waf\_fr\_s\_coal = 0.016$$

$$waf\_fr\_h\_coal = 0.042$$

$$waf\_fr\_o\_coal = 0.127$$

$$waf\_fr\_n\_coal = 0.021$$

{Water and ash free mass fraction carbon, sulfur, hydrogen in tar [-]}

$$waf\_fr\_c\_tar = 0.773$$

$$waf\_fr\_s\_tar = 0.014$$

$$waf\_fr\_h\_tar = 0.049$$

{Coal higher and lower heating value [kJ/kg]}

$$HHV = 1000 \cdot (35.2 \cdot waf\_fr\_c\_coal + 116.4 \cdot waf\_fr\_h\_coal - 11.1 \cdot waf\_fr\_o\_coal + 10.5 \cdot waf\_fr\_s\_coal + 6.29 \cdot waf\_fr\_n\_coal)$$

$$LHV = HHV - 2442 \cdot (9 \cdot waf\_fr\_h\_coal)$$

{Enthalpies of formation [kJ/kg]}

$$h\_h2o\_L\_0 = \text{enthalpy\_formation}(H2O)$$

$$h\_h2o\_V\_0 = \text{enthalpy}(H2O, T = T\_ref)$$

$$h\_o2\_0 = \text{enthalpy}(O2, T = T\_ref)$$

$$h\_h2\_0 = \text{enthalpy}(H2, T = T\_ref)$$

$$h\_co\_0 = \text{enthalpy}(CO, T = T\_ref)$$

$$h\_co2\_0 = \text{enthalpy}(CO2, T = T\_ref)$$

$$h\_ch4\_0 = \text{enthalpy}(CH4, T = T\_ref)$$

$$h\_so2\_0 = \text{enthalpy}(SO2, T = T\_ref)$$

{Enthalpy of formation of coal}

$$h\_coal\_0 = h\_coal\_com \cdot (1 - f\_com) \cdot (1 - af\_fr\_w\_coal) + h\_h2o\_L\_0 \cdot af\_fr\_w\_coal$$

$$h\_coal\_com = 1000 \cdot (-32.7633 \cdot waf\_fr\_c\_coal - 141.7892 \cdot waf\_fr\_h\_coal - 9.2768 \cdot waf\_fr\_s\_coal)$$

$$f\_com = 0.01379 \cdot \ln(waf\_fr\_o\_coal) - 0.01393 \cdot waf\_fr\_o\_coal^{0.75} + 1.02111$$

{Gasifier heat loss}

$$Ht\_loss = 0.17$$

$$Q\_HV\_loss = Ht\_loss \cdot LHV$$

{Calculate enthalpy}

**Duplicate** i = 1,nm\_rows

{Inlet steam and oxygen mass flow rates [kg/s]}

$$m\_dot\_stm\_in[i] = \text{convert}(\text{tonne/hr, kg/s}) \cdot \text{Prop\_in}[i, 10]$$

$$m\_dot\_o2\_in[i] = 1000 \cdot \text{convert}(1/\text{hr}, 1/\text{s}) \cdot \text{Prop\_in}[i, 11] \cdot \rho_{o2\_n}$$

{Inlet steam and oxygen mole flow rates [kmol/s]}

$$mol\_dot\_stm\_in[i] = m\_dot\_stm\_in[i] \cdot Mol\_h2o^{(-1)}$$

$$mol\_dot\_o2\_in[i] = m\_dot\_o2\_in[i] \cdot Mol\_o2^{(-1)}$$

{Feedwater mass flow rate [kg/s]}

$m\_dot\_w[i] = \text{convert}(\text{tonne/hr, kg/s}) * Prop\_in[i, 13]$

{Coal and inlet fixed carbon mass flow rates [kg/s]}

$m\_dot\_coal[i] = \text{convert}(\text{tonne/hr, kg/s}) * Prop\_in[i, 12]$

$m\_dot\_fc\_in[i] = ar\_fr\_fc\_coal * m\_dot\_coal[i]$

{Ash and outlet fixed carbon mass flow rates [kg/s]}

$m\_dot\_ash[i] = \text{convert}(\text{tonne/hr, kg/s}) * grt\_as\_rm * Prop\_in[i, 14]$

$m\_dot\_fc\_out[i] = ar\_fr\_fc\_ash * m\_dot\_ash[i]$

{Inlet and outlet fixed carbon mole flow rates [kmol/s]}

$mol\_dot\_fc\_in[i] = (m\_dot\_fc\_in[i] * Mol\_c^{(-1)})$

$mol\_dot\_fc\_out[i] = (m\_dot\_fc\_out[i] * Mol\_c^{(-1)})$

{Volatile matter mass flow rates [kg/s]}

$m\_dot\_tar\_vm[i] = m\_dot\_coal[i] * ad\_fr\_720\_tr\_devl$

$m\_dot\_gs\_vm\_h2[i] = m\_dot\_coal[i] * (ar\_fr\_fc\_coal + ar\_fr\_vm\_coal) * vf\_720\_h2\_gs\_vm * rho\_h2\_n$

$m\_dot\_gs\_vm\_co[i] = m\_dot\_coal[i] * (ar\_fr\_fc\_coal + ar\_fr\_vm\_coal) * vf\_720\_co\_gs\_vm * rho\_co\_n$

$m\_dot\_gs\_vm\_co2[i] = m\_dot\_coal[i] * (ar\_fr\_fc\_coal + ar\_fr\_vm\_coal) * vf\_720\_co2\_gs\_vm * rho\_co2\_n$

$m\_dot\_gs\_vm\_ch4[i] = m\_dot\_coal[i] * (ar\_fr\_fc\_coal + ar\_fr\_vm\_coal) * vf\_720\_ch4\_gs\_vm * rho\_ch4\_n$

{Coal evaporated water mass flow rate [kg/s]}

$m\_dot\_coal\_w[i] = m\_dot\_coal[i] * ar\_fr\_w\_coal$

{Coal evaporated water mole flow rate [kmol/s]}

$mol\_dot\_coal\_w[i] = m\_dot\_coal\_w[i] * Mol\_h2o^{(-1)}$

{Tar carbon, sulfur, hydrogen mass flow rates [kg/s]}

$m\_dot\_c\_tar[i] = waf\_fr\_c\_tar * m\_dot\_tar\_vm[i]$

$m\_dot\_s\_tar[i] = waf\_fr\_s\_tar * m\_dot\_tar\_vm[i]$

$m\_dot\_h\_tar[i] = waf\_fr\_h\_tar * m\_dot\_tar\_vm[i]$

{Tar carbon, sulfur, hydrogen mole flow rates [kmol/s]}

$mol\_dot\_c\_tar[i] = m\_dot\_c\_tar[i] * Mol\_c^{(-1)}$

$mol\_dot\_s\_tar[i] = m\_dot\_s\_tar[i] * Mol\_s^{(-1)}$

$mol\_dot\_h\_tar[i] = m\_dot\_h\_tar[i] * Mol\_h^{(-1)}$

{Raw syngas mole flow rates [kmol/s]}

$mol\_dot\_rw\_gs\_h2o[i] = m\_dot\_rw\_gs\_h2o[i] * Mol\_h2o^{(-1)}$

$mol\_dot\_rw\_gs\_h2[i] = m\_dot\_rw\_gs\_h2[i] * Mol\_h2^{(-1)}$

$mol\_dot\_rw\_gs\_co[i] = m\_dot\_rw\_gs\_co[i] * Mol\_co^{(-1)}$

$mol\_dot\_rw\_gs\_co2[i] = m\_dot\_rw\_gs\_co2[i] * Mol\_co2^{(-1)}$

$mol\_dot\_rw\_gs\_ch4[i] = m\_dot\_rw\_gs\_ch4[i] * Mol\_ch4^{(-1)}$

{Gaseous volatile matter mole flow rates [kmol/s]}

$mol\_dot\_gs\_vm\_h2[i] = m\_dot\_gs\_vm\_h2[i] * Mol\_h2^{(-1)}$

$mol\_dot\_gs\_vm\_co[i] = m\_dot\_gs\_vm\_co[i] * Mol\_co^{(-1)}$

$mol\_dot\_gs\_vm\_co2[i] = m\_dot\_gs\_vm\_co2[i] * Mol\_co2^{(-1)}$

$mol\_dot\_gs\_vm\_ch4[i] = m\_dot\_gs\_vm\_ch4[i] * Mol\_ch4^{(-1)}$

{Input steam and oxygen mole fractions [-]}

$tot\_mol\_in[i] = mol\_dot\_stm\_in[i] + mol\_dot\_o2\_in[i]$

$y\_stm\_in[i] = mol\_dot\_stm\_in[i] / tot\_mol\_in[i]$

$y\_o2\_in[i] = mol\_dot\_o2\_in[i] / tot\_mol\_in[i]$

{Raw syngas mole fractions [-]}

```

tot_mol_rw_gs[i] = mol_dot_rw_gs_h2o[i] + mol_dot_rw_gs_h2[i] + mol_dot_rw_gs_co[i] +
mol_dot_rw_gs_co2[i] + mol_dot_rw_gs_ch4[i] + mol_dot_gs_vm_h2[i] + mol_dot_gs_vm_co[i] +
mol_dot_gs_vm_co2[i] + mol_dot_gs_vm_ch4[i] + mol_dot_coal_w[i]
y_rw_gs_h2o[i] = (mol_dot_rw_gs_h2o[i] + mol_dot_coal_w[i])/tot_mol_rw_gs[i]
y_rw_gs_h2[i] = (mol_dot_rw_gs_h2[i] + mol_dot_gs_vm_h2[i])/tot_mol_rw_gs[i]
y_rw_gs_co[i] = (mol_dot_rw_gs_co[i] + mol_dot_gs_vm_co[i])/tot_mol_rw_gs[i]
y_rw_gs_co2[i] = (mol_dot_rw_gs_co2[i] + mol_dot_gs_vm_co2[i])/tot_mol_rw_gs[i]
y_rw_gs_ch4[i] = (mol_dot_rw_gs_ch4[i] + mol_dot_gs_vm_ch4[i])/tot_mol_rw_gs[i]

```

#### {Raw syngas normal volume flow rates}

```

vf_rw_gs_h2[i] = (m_dot_rw_gs_h2[i] + m_dot_gs_vm_h2[i])*rho_h2_n^(-1)
vf_rw_gs_co[i] = (m_dot_rw_gs_co[i] + m_dot_gs_vm_co[i])*rho_co_n^(-1)
vf_rw_gs_co2[i] = (m_dot_rw_gs_co2[i] + m_dot_gs_vm_co2[i])*rho_co2_n^(-1)
vf_rw_gs_ch4[i] = (m_dot_rw_gs_ch4[i] + m_dot_gs_vm_ch4[i])*rho_ch4_n^(-1)

```

#### {Raw syngas volume flow rate percentages}

```

vf_per_h2[i] = convert(-,%)*vf_rw_gs_h2[i]/(vf_rw_gs_h2[i] + vf_rw_gs_co[i] + vf_rw_gs_co2[i] +
vf_rw_gs_ch4[i])
vf_per_co[i] = convert(-,%)*vf_rw_gs_co[i]/(vf_rw_gs_h2[i] + vf_rw_gs_co[i] + vf_rw_gs_co2[i] +
vf_rw_gs_ch4[i])
vf_per_co2[i] = convert(-,%)*vf_rw_gs_co2[i]/(vf_rw_gs_h2[i] + vf_rw_gs_co[i] + vf_rw_gs_co2[i] +
vf_rw_gs_ch4[i])
vf_per_ch4[i] = convert(-,%)*vf_rw_gs_ch4[i]/(vf_rw_gs_h2[i] + vf_rw_gs_co[i] + vf_rw_gs_co2[i] +
vf_rw_gs_ch4[i])

```

#### {Input steam and oxygen enthalpies [kJ/kg]}

```

h_stm_in[i] = h_h2o_V_0 + enthalpy(Water, T = Prop_in[i,1], P = y_stm_in[i]*Prop_in[i,2]) -
enthalpy(Water, T = T_ref, P = P_ref)
h_o2_in[i] = h_o2_0 + enthalpy(Oxygen, T = Prop_in[i,1], P = y_o2_in[i]*Prop_in[i,2]) -
enthalpy(Oxygen, T = T_ref, P = P_ref)

```

#### {Coal enthalpy for fixed carbon, volatile matter, and water}

```

h_coal[i] = h_coal_0 + cp_coal(Prop_in[i,3], waf_fr_vm_coal, af_fr_w_coal)*(Prop_in[i,3] - T_ref)

```

#### {Ash change in enthalpy}

```

delta_h_ash[i] = (0.752 + 0.293*((Prop_in[i,3] + Prop_in[i,4])/2 + 273.15)/1000)*(Prop_in[i,3] -
Prop_in[i,4])

```

#### {Tar power}

```

Q_tar[i] = m_dot_h_tar_0[i] + m_dot_cp_dT_tar[i]

```

```

m_dot_h_tar_0[i] = -m_dot_tar_vm[i]*30980 + mol_dot_c_tar[i]*h_co2_0*Mol_co2 +
mol_dot_s_tar[i]*h_so2_0*Mol_so2 + mol_dot_h_tar[i]*(h_h2o_L_0/2)*Mol_h2o
m_dot_cp_dT_tar[i] = m_dot_tar_vm[i]*cp_dT_tar[i]
cp_dT_tar[i] = 2.11*10^(-3)*(converttemp(C,K,Prop_in[i,8])^2 - converttemp(C,K,T_ref)^2)

```

#### {Water jacket feedwater and steam enthalpies [kJ/kg]}

```

h_wj_fw[i] = enthalpy(Water, T = Prop_in[i,5], P = Prop_in[i,7])
h_wj_stm[i] = enthalpy(Water, T = Prop_in[i,6], P = Prop_in[i,7])

```

#### {Raw syngas enthalpies [kJ/kg]}

```

h_rw_gs_h2o[i] = h_h2o_V_0 + enthalpy(Water, T = Prop_in[i,8], P = y_rw_gs_h2o[i]*Prop_in[i,9]) -
enthalpy(Water, T = T_ref, P = P_ref)
h_rw_gs_h2[i] = h_h2_0 + enthalpy(Hydrogen, T = Prop_in[i,8], P = y_rw_gs_h2[i]*Prop_in[i,9]) -
enthalpy(Hydrogen, T = T_ref, P = P_ref)
h_rw_gs_co[i] = h_en_co(T_ref, P_ref, h_co_0, Prop_in[i,8], Prop_in[i,9], y_rw_gs_co[i])
h_rw_gs_co2[i] = h_co2_0 + enthalpy(CarbonDioxide, T = Prop_in[i,8], P =
y_rw_gs_co2[i]*Prop_in[i,9]) - enthalpy(CarbonDioxide, T = T_ref, P = P_ref)
h_rw_gs_ch4[i] = h_en_ch4(T_ref, P_ref, h_ch4_0, Prop_in[i,8], Prop_in[i,9], y_rw_gs_ch4[i])

```

{Raw syngas ideal gas enthalpies [kJ/kg]}

$h_{ideal\_rw\_gs\_h2o[i]} = \text{enthalpy}(H2O, T = \text{Prop\_in}[i,8])$

$h_{ideal\_rw\_gs\_h2[i]} = \text{enthalpy}(H2, T = \text{Prop\_in}[i,8])$

$h_{ideal\_rw\_gs\_co[i]} = \text{enthalpy}(CO, T = \text{Prop\_in}[i,8])$

$h_{ideal\_rw\_gs\_co2[i]} = \text{enthalpy}(CO2, T = \text{Prop\_in}[i,8])$

{Raw syngas ideal gas entropies [kJ/kg-K]}

$s_{ideal\_rw\_gs\_h2o[i]} = \text{entropy}(H2O, T = \text{Prop\_in}[i,8], P = P\_ref)$

$s_{ideal\_rw\_gs\_h2[i]} = \text{entropy}(H2, T = \text{Prop\_in}[i,8], P = P\_ref)$

$s_{ideal\_rw\_gs\_co[i]} = \text{entropy}(CO, T = \text{Prop\_in}[i,8], P = P\_ref)$

$s_{ideal\_rw\_gs\_co2[i]} = \text{entropy}(CO2, T = \text{Prop\_in}[i,8], P = P\_ref)$

{Gibbs functions [kJ/kmol]}

$g_{h2o[i]} = (\text{Mol}_{h2o}) * (h_{ideal\_rw\_gs\_h2o[i]} - \text{converttemp}(C,K,\text{Prop\_in}[i,8]) * s_{ideal\_rw\_gs\_h2o[i]})$

$g_{h2[i]} = (\text{Mol}_{h2}) * (h_{ideal\_rw\_gs\_h2[i]} - \text{converttemp}(C,K,\text{Prop\_in}[i,8]) * s_{ideal\_rw\_gs\_h2[i]})$

$g_{co[i]} = (\text{Mol}_{co}) * (h_{ideal\_rw\_gs\_co[i]} - \text{converttemp}(C,K,\text{Prop\_in}[i,8]) * s_{ideal\_rw\_gs\_co[i]})$

$g_{co2[i]} = (\text{Mol}_{co2}) * (h_{ideal\_rw\_gs\_co2[i]} - \text{converttemp}(C,K,\text{Prop\_in}[i,8]) * s_{ideal\_rw\_gs\_co2[i]})$

{Energy balance}

$Q_{wj[i]} = m_{dot\_wj[i]} * (h_{wj\_fw[i]} - h_{wj\_stm[i]})$

$m_{dot\_stm\_in[i]} * h_{stm\_in[i]} + m_{dot\_o2\_in[i]} * h_{o2\_in[i]} + (m_{dot\_coal[i]} - m_{dot\_ash[i]}) * h_{coal[i]} +$   
 $m_{dot\_ash[i]} * \Delta h_{ash[i]} + Q_{wj[i]} = (m_{dot\_rw\_gs\_h2o[i]} + m_{dot\_coal\_w[i]}) * h_{rw\_gs\_h2o[i]} +$   
 $(m_{dot\_rw\_gs\_h2[i]} + m_{dot\_gs\_vm\_h2[i]}) * h_{rw\_gs\_h2[i]} + (m_{dot\_rw\_gs\_co[i]} +$   
 $m_{dot\_gs\_vm\_co[i]}) * h_{rw\_gs\_co[i]} + (m_{dot\_rw\_gs\_co2[i]} + m_{dot\_gs\_vm\_co2[i]}) * h_{rw\_gs\_co2[i]} +$   
 $(m_{dot\_rw\_gs\_ch4[i]} + m_{dot\_gs\_vm\_ch4[i]}) * h_{rw\_gs\_ch4[i]} + Q_{tar[i]} + (ar\_fr\_fc\_coal +$   
 $ar\_fr\_vm\_coal) * m_{dot\_coal[i]} * Q_{HV\_loss}$

{Hydrogen balance}

$2 * m_{dot\_stm\_in[i]} = 2 * m_{dot\_rw\_gs\_h2o[i]} + 2 * m_{dot\_rw\_gs\_h2[i]} + 4 * m_{dot\_rw\_gs\_ch4[i]}$

{Oxygen balance}

$1 * m_{dot\_stm\_in[i]} + 2 * m_{dot\_o2\_in[i]} = 1 * m_{dot\_rw\_gs\_h2o[i]} + 1 * m_{dot\_rw\_gs\_co[i]} +$   
 $2 * m_{dot\_rw\_gs\_co2[i]}$

{Carbon balance}

$1 * m_{dot\_fc\_in[i]} = 1 * m_{dot\_fc\_out[i]} + 1 * m_{dot\_rw\_gs\_co[i]} + 1 * m_{dot\_rw\_gs\_co2[i]} +$   
 $1 * m_{dot\_rw\_gs\_ch4[i]}$

{Equilibrium condition}

$\ln(K1\_eq[i]) = -(g_{co2[i]} + g_{h2[i]} - g_{co[i]} - g_{h2o[i]}) / (R\_ugc * \text{converttemp}(C,K,\text{Prop\_in}[i,8]))$

$K1\_eq\_x[i] = (y_{rw\_gs\_co2[i]} * y_{rw\_gs\_h2[i]}) / (y_{rw\_gs\_co[i]} * y_{rw\_gs\_h2o[i]})$

$K1\_eq[i] = K1\_eq\_x[i]$

{Total raw syngas mass flow rates for export [kg/s]}

$m_{dot\_tot\_rw\_gs\_h2o[i]} = m_{dot\_rw\_gs\_h2o[i]} + m_{dot\_coal\_w[i]}$

$m_{dot\_tot\_rw\_gs\_h2[i]} = m_{dot\_rw\_gs\_h2[i]} + m_{dot\_gs\_vm\_h2[i]}$

$m_{dot\_tot\_rw\_gs\_co[i]} = m_{dot\_rw\_gs\_co[i]} + m_{dot\_gs\_vm\_co[i]}$

$m_{dot\_tot\_rw\_gs\_co2[i]} = m_{dot\_rw\_gs\_co2[i]} + m_{dot\_gs\_vm\_co2[i]}$

$m_{dot\_tot\_rw\_gs\_ch4[i]} = m_{dot\_rw\_gs\_ch4[i]} + m_{dot\_gs\_vm\_ch4[i]}$

**End**

\$Export /A /D 'STEAM\_MASS\_FLOW\_RATE\_B.csv' m\_dot\_tot\_rw\_gs\_h2o[1..nm\_rows]

\$Export /A /D 'HYDROGEN\_MASS\_FLOW\_RATE\_B.csv' m\_dot\_tot\_rw\_gs\_h2[1..nm\_rows]

\$Export /A /D 'CARBON\_MONOXIDE\_MASS\_FLOW\_RATE\_B.csv'

m\_dot\_tot\_rw\_gs\_co[1..nm\_rows]

\$Export /A /D 'CARBON\_DIOXIDE\_MASS\_FLOW\_RATE\_B.csv' m\_dot\_tot\_rw\_gs\_co2[1..nm\_rows]

\$Export /A /D 'METHANE\_MASS\_FLOW\_RATE\_B.csv' m\_dot\_tot\_rw\_gs\_ch4[1..nm\_rows]

## APPENDIX G – R PROGRAM

This appendix contains the R program code.

```
#Gasifier statistical modeling

#Clear previous data
rm(list = ls())

#Remove previously used headings
detach(Dat_pr)
detach(new_dat)
detach(new_dat2)

#Required libraries
library(tree)
library(e1071)
library(ellipse)

#Retrieve typical gasifier data
Dat_orl = read.csv(file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Gasifier_Data_Set1.csv", header = TRUE, sep = ",", dec = ".")

#Remove observations with missing entries
Dat_pr = na.omit(subset(Dat_orl))
attach(Dat_pr) #Attach headings
nm_ent = dim(Dat_pr)[1] #Number of measurements

#Input variable names

#Temperature
#Steam and oxygen inlet: stm_o2_tmp
#Coal inlet: coal_tmp
#Water jacket outlet: wj_out_tmp
#Ash outlet: ash_tmp
#Syngas outlet: sgs_tmp

#Flow Rates
#Steam inlet mass: m_stm
#Oxygen inlet volume: vol_o2
#Coal inlet mass: m_coal
#Water jacket mass: m_wj

#Syngas outlet pressure: sgs_pr
#Grate angular frequency:grt_frq

#Export normal temperature and pressure
tp_norm = matrix(nrow = 1, ncol = 2)
tp_norm[1,1] = 15
tp_norm[1,2] = 101.325

write.csv(tp_norm, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Norm_TP.csv", row.names = FALSE)

#Property matrix columns
#prop_mat[,1]: steam and oxygen inlet temperature
```

```

#prop_mat[,2]: steam and oxygen inlet pressure
#prop_mat[,3]: coal inlet temperature
#prop_mat[,4]: ash outlet temperature
#prop_mat[,5]: water jacket inlet temperature
#prop_mat[,6]: water jacket outlet temperature
#prop_mat[,7]: water jacket pressure
#prop_mat[,8]: syngas outlet temperature
#prop_mat[,9]: syngas outlet pressure
#prop_mat[,10]: steam inlet mass flow rate
#prop_mat[,11]: oxygen inlet volume flow rate
#prop_mat[,12]: coal inlet mass flow rate
#prop_mat[,13]: water jacket mass flow rate
#prop_mat[,14]: grate frequency

#Thermal-fluid model A

#Single data point used while tuning the heat loss
prop_mat = matrix(nrow = 1, ncol = 14) #Matrix for input properties
prop_mat[,1] = 344
prop_mat[,2] = 3500
prop_mat[,3] = 190
prop_mat[,4] = 350
prop_mat[,5] = 25
prop_mat[,6] = 240
prop_mat[,7] = 3500
prop_mat[,8] = 550
prop_mat[,9] = 2850
prop_mat[,10] = 52.5
prop_mat[,11] = 9.2
prop_mat[,12] = 50
prop_mat[,13] = 8.3
prop_mat[,14] = 4.5

#Export properties and number of observations
k = 1
write.csv(k, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Nm_Rows_A.csv", row.names = FALSE)
write.csv(prop_mat, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Input_Properties_A.csv", row.names = FALSE)

#Random set of 100 data points used while tuning the guess values
set.seed(2)
i = sample(x = nm_ent, size = 100) #Sample index
prop_mat = matrix(nrow = 100, ncol = 14) #Matrix for input properties
prop_mat[,1] = stm_o2_tmp[i]
prop_mat[,2] = 3500
prop_mat[,3] = coal_tmp[i]
prop_mat[,4] = ash_tmp[i]
prop_mat[,5] = 25
prop_mat[,6] = wj_out_tmp[i]
prop_mat[,7] = 3500
prop_mat[,8] = sgs_tmp[i]
prop_mat[,9] = sgs_pr[i]
prop_mat[,10] = m_stm[i]
prop_mat[,11] = vol_o2[i]
prop_mat[,12] = m_coal[i]
prop_mat[,13] = m_wj[i]
prop_mat[,14] = grt_frq[i]

#Export properties and number of observations

```

```

k = 100
write.csv(k, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Nm_Rows_A.csv", row.names = FALSE)
write.csv(prop_mat, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Input_Properties_A.csv", row.names = FALSE)

#Full data set

#Blank files for mass flow rates
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/STEAM_MASS_FLOW_RATE_A.csv", row.names = FALSE)
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/HYDROGEN_MASS_FLOW_RATE_A.csv", row.names = FALSE)
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/CARBON MONOXIDE_MASS_FLOW_RATE_A.csv", row.names = FALSE)
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/CARBON DIOXIDE_MASS_FLOW_RATE_A.csv", row.names = FALSE)
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/METHANE_MASS_FLOW_RATE_A.csv", row.names = FALSE)

#Blank files for percentage volume flow rates
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/HYDROGEN_PERCENT_VOLUME_FLOW_RATE_A.csv", row.names = FALSE)
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/CARBON MONOXIDE_PERCENT_VOLUME_FLOW_RATE_A.csv", row.names =
FALSE)
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/CARBON DIOXIDE_PERCENT_VOLUME_FLOW_RATE_A.csv", row.names =
FALSE)
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/METHANE_PERCENT_VOLUME_FLOW_RATE_A.csv", row.names = FALSE)

#Repeatedly execute EES code for up to 100 observations
i = 1
while (i <= nm_ent)
{
  if (nm_ent - i >= 100)
  {
    k = 100
  } else
  {
    k = nm_ent - (i - 1)
  }
  #Export number of observations
  write.csv(k, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Nm_Rows_A.csv", row.names = FALSE)

  #Matrix for input properties
  prop_mat = matrix(nrow = k, ncol = 14) #Matrix for input properties
  for (j in 1:k)
  {
    prop_mat[j,1] = stm_o2_tmp[i]
    prop_mat[j,2] = 3500
    prop_mat[j,3] = coal_tmp[i]
    prop_mat[j,4] = ash_tmp[i]
    prop_mat[j,5] = 25
    prop_mat[j,6] = wj_out_tmp[i]
    prop_mat[j,7] = 3500
    prop_mat[j,8] = sgs_tmp[i]
    prop_mat[j,9] = sgs_pr[i]

```

```

prop_mat[j,10] = m_stm[i]
prop_mat[j,11] = vol_o2[i]
prop_mat[j,12] = m_coal[i]
prop_mat[j,13] = m_wj[i]
prop_mat[j,14] = grt_frq[i]

i = i + 1
}
#Export properties
write.csv(prop_mat, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Input_Properties_A.csv", row.names = FALSE)

#Solve equation sets for inputs
system('C:/ees32/ees.exe F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Thermal_Fluid_Model_A_Rev7.EES /solve')
}

#Import and collate syngas mass flow rates and volume percentages
m_h2o_A = read.csv(file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/STEAM_MASS_FLOW_RATE_A.csv", header = FALSE, sep = ",", dec =
".")
m_h2_A = read.csv(file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/HYDROGEN_MASS_FLOW_RATE_A.csv", header = FALSE, sep = ",",
dec = ".")
m_co_A = read.csv(file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/CARBON_MONOXIDE_MASS_FLOW_RATE_A.csv", header = FALSE, sep =
",", dec = ".")
m_co2_A = read.csv(file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/CARBON_DIOXIDE_MASS_FLOW_RATE_A.csv", header = FALSE, sep =
",", dec = ".")
m_ch4_A = read.csv(file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/METHANE_MASS_FLOW_RATE_A.csv", header = FALSE, sep = ",", dec
= ".")

m_dot_rw_gs_h2o_A = matrix(nrow = nm_ent, ncol = 1)
m_dot_rw_gs_h2_A = matrix(nrow = nm_ent, ncol = 1)
m_dot_rw_gs_co_A = matrix(nrow = nm_ent, ncol = 1)
m_dot_rw_gs_co2_A = matrix(nrow = nm_ent, ncol = 1)
m_dot_rw_gs_ch4_A = matrix(nrow = nm_ent, ncol = 1)

vol_h2_A = read.csv(file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/HYDROGEN_PERCENT_VOLUME_FLOW_RATE_A.csv", header = FALSE, sep
= ",", dec = ".")
vol_co_A = read.csv(file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/CARBON_MONOXIDE_PERCENT_VOLUME_FLOW_RATE_A.csv", header =
FALSE, sep = ",", dec = ".")
vol_co2_A = read.csv(file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/CARBON_DIOXIDE_PERCENT_VOLUME_FLOW_RATE_A.csv", header =
FALSE, sep = ",", dec = ".")
vol_ch4_A = read.csv(file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/METHANE_PERCENT_VOLUME_FLOW_RATE_A.csv", header = FALSE, sep
= ",", dec = ".")

vol_per_h2_A = matrix(nrow = nm_ent, ncol = 1)
vol_per_co_A = matrix(nrow = nm_ent, ncol = 1)
vol_per_co2_A = matrix(nrow = nm_ent, ncol = 1)
vol_per_ch4_A = matrix(nrow = nm_ent, ncol = 1)

#For each variable, merge separate columns from individual EES executions
into single column
i = 1

```

```

j = 1
while (i <= ceiling(nm_ent/100))
{
  if (nm_ent - j >= 100)
  {
    m = 100
  } else
  {
    m = nm_ent - (j - 1)
  }
  for (k in 1:m)
  {
    m_dot_rw_gs_h2o_A[j,1] = m_h2o_A[i,k]
    m_dot_rw_gs_h2_A[j,1] = m_h2_A[i,k]
    m_dot_rw_gs_co_A[j,1] = m_co_A[i,k]
    m_dot_rw_gs_co2_A[j,1] = m_co2_A[i,k]
    m_dot_rw_gs_ch4_A[j,1] = m_ch4_A[i,k]

    vol_per_h2_A[j,1] = vol_h2_A[i,k]
    vol_per_co_A[j,1] = vol_co_A[i,k]
    vol_per_co2_A[j,1] = vol_co2_A[i,k]
    vol_per_ch4_A[j,1] = vol_ch4_A[i,k]

    j = j + 1
  }
  i = i + 1
}

#Thermal-fluid model B

#Single data point used for manual verification calculations
prop_mat = matrix(nrow = 1, ncol = 14) #Matrix for input properties
prop_mat[,1] = 344
prop_mat[,2] = 3500
prop_mat[,3] = 190
prop_mat[,4] = 350
prop_mat[,5] = 25
prop_mat[,6] = 240
prop_mat[,7] = 3500
prop_mat[,8] = 550
prop_mat[,9] = 2850
prop_mat[,10] = 52.5
prop_mat[,11] = 9.2
prop_mat[,12] = 50
prop_mat[,13] = 8.3
prop_mat[,14] = 4.5

#Export properties and number of observations
k = 1
write.csv(k, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Nm_Rows_B.csv", row.names = FALSE)
write.csv(prop_mat, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Input_Properties_B.csv", row.names = FALSE)

#Full data set

#Blank files for mass flow rates
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/STEAM_MASS_FLOW_RATE_B.csv", row.names = FALSE)

```

```

write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/HYDROGEN_MASS_FLOW_RATE_B.csv", row.names = FALSE)
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/CARBON_MONOXIDE_MASS_FLOW_RATE_B.csv", row.names = FALSE)
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/CARBON_DIOXIDE_MASS_FLOW_RATE_B.csv", row.names = FALSE)
write.csv(x = NULL, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/METHANE_MASS_FLOW_RATE_B.csv", row.names = FALSE)

#Export number of observations
k = 1
write.csv(k, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Nm_Rows_B.csv", row.names = FALSE)

prop_mat = matrix(nrow = 1, ncol = 14) #Matrix for input properties

#Execute EES code per observation
i = 1
while (i <= nm_ent)
{
  prop_mat[1,1] = stm_o2_tmp[i]
  prop_mat[1,2] = 3500
  prop_mat[1,3] = coal_tmp[i]
  prop_mat[1,4] = ash_tmp[i]
  prop_mat[1,5] = 25
  prop_mat[1,6] = wj_out_tmp[i]
  prop_mat[1,7] = 3500
  prop_mat[1,8] = sgs_tmp[i]
  prop_mat[1,9] = sgs_pr[i]
  prop_mat[1,10] = m_stm[i]
  prop_mat[1,11] = vol_o2[i]
  prop_mat[1,12] = m_coal[i]
  prop_mat[1,13] = m_wj[i]
  prop_mat[1,14] = grt_frq[i]

  i = i + 1

  #Export properties
  write.csv(prop_mat, file = "F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Input_Properties_B.csv", row.names = FALSE)

  #Solve equation sets for inputs
  system('C:/ees32/ees.exe F:/Aiden/Academic/Masters/Data
Analysis/Rev7/Thermal_Fluid_Model_B_Rev7.EES /solve')
}

#Import syngas mass flow rates
m_dot_rw_gs_h2o_B = as.matrix(read.csv(file =
"F:/Aiden/Academic/Masters/Data Analysis/Rev7/STEAM_MASS_FLOW_RATE_B.csv",
header = FALSE, sep = ",", dec = "."))
m_dot_rw_gs_h2_B = as.matrix(read.csv(file =
"F:/Aiden/Academic/Masters/Data
Analysis/Rev7/HYDROGEN_MASS_FLOW_RATE_B.csv", header = FALSE, sep = ",",
dec = "."))
m_dot_rw_gs_co_B = as.matrix(read.csv(file =
"F:/Aiden/Academic/Masters/Data
Analysis/Rev7/CARBON_MONOXIDE_MASS_FLOW_RATE_B.csv", header = FALSE, sep =
",", dec = "."))
m_dot_rw_gs_co2_B = as.matrix(read.csv(file =
"F:/Aiden/Academic/Masters/Data

```

```

Analysis/Rev7/CARBON_DIOXIDE_MASS_FLOW_RATE_B.csv", header = FALSE, sep =
",", dec = ".")
m_dot_rw_gs_ch4_B = as.matrix(read.csv(file =
"F:/Aiden/Academic/Masters/Data
Analysis/Rev7/METHANE_MASS_FLOW_RATE_B.csv", header = FALSE, sep = ",", dec
= "."))

#Index of points where thermal-fluid model B can and can't be solved
p_set = which(m_dot_rw_gs_h2o_B != 1) #Can be solved
n_set = which(m_dot_rw_gs_h2o_B == 1) #Can't be solved

#Ratio of class sizes
IR = length(n_set)/length(p_set)

#Factor matrix for where model B can (TRUE) and can't (FALSE) be solved
B_solvs = matrix(data = TRUE, nrow = nm_ent, ncol = 1)
B_solvs[n_set] = FALSE
B_solvs = as.factor(B_solvs)

#Statistical models to predict where model B can and can't be solved

#PCA to determine which combinations of variables to model

#Collate data set for PCA
new_pca_dat = data.frame(m_dot_rw_gs_h2o_A[p_set], m_dot_rw_gs_h2_A[p_set],
m_dot_rw_gs_co_A[p_set], m_dot_rw_gs_co2_A[p_set],
m_dot_rw_gs_ch4_A[p_set])

#PCA and variance explained
pca_gsf = prcomp(new_pca_dat, scale = TRUE)

pca_var = pca_gsf$sdev^2
pve = pca_var/sum(pca_var)*100 #Percentage of variance explained by
principal components

#Plot PCA scores
par(mar=c(5,5,5,5) + .1)
plot(pca_gsf$x[,1], pca_gsf$x[,2], xlim = c(-10, 10), ylim = c(-12, 12),
xlab = "First Principal Component Score", ylab = "Second Principal
Component Score")
title("Mass Flow Rate PCA Biplot", line = 4)

#Plot 95% confidence ellipse
elips_dat_pca = cbind(pca_gsf$x[,1], pca_gsf$x[,2])

cov_gsf_pca = cov(elips_dat_pca) #Covariance of 1st and 2nd principal
component
mean_pc1 = mean(elips_dat_pca[,1]) #Average and center of 1st principal
component
mean_pc2 = mean(elips_dat_pca[,2]) #Average and center of 2nd principal
component
center_gsf_pca = c(mean_pc1, mean_pc2)

lines(ellipse(cov_gsf_pca, centre = center_gsf_pca, level = 0.95), col =
"red")

#Plot PCA loadings
par(new = TRUE)

```

```

plot(pca_gsf$rotation[,1], pca_gsf$rotation[,2], xlim = c(-0.6, 0.6), ylim
= c(-0.7, 0.7), axes = FALSE, pch = 17, col = "blue", xlab = "", ylab = "")

lines(c(0, pca_gsf$rotation[1,1]), c(0, pca_gsf$rotation[1,2]), col =
"blue")
lines(c(0, pca_gsf$rotation[2,1]), c(0, pca_gsf$rotation[2,2]), col =
"blue")
lines(c(0, pca_gsf$rotation[3,1]), c(0, pca_gsf$rotation[3,2]), col =
"blue")
lines(c(0, pca_gsf$rotation[4,1]), c(0, pca_gsf$rotation[4,2]), col =
"blue")
lines(c(0, pca_gsf$rotation[5,1]), c(0, pca_gsf$rotation[5,2]), col =
"blue")

#Add secondary axes
axis(side = 3, at = pretty(range(pca_gsf$rotation[,1])))
mtext("First Principal Component Loading", side = 3, line = 2.5)

axis(side = 4, at = pretty(range(pca_gsf$rotation[,2])))
mtext("Second Principal Component Loading", side = 4, line = 2.5)

#Add loading variable labels
text(x = pca_gsf$rotation[1,1], y = pca_gsf$rotation[1,2] - 0.05,
substitute(paste(italic(H[2]*O))), col = "blue")
text(x = pca_gsf$rotation[2,1], y = pca_gsf$rotation[2,2] + 0.05,
substitute(paste(italic(H[2]))), col = "blue")
text(x = pca_gsf$rotation[3,1] + 0.05, y = pca_gsf$rotation[3,2],
substitute(paste(italic(CO))), col = "blue")
text(x = pca_gsf$rotation[4,1], y = pca_gsf$rotation[4,2] + 0.05,
substitute(paste(italic(CO[2]))), col = "blue")
text(x = pca_gsf$rotation[5,1], y = pca_gsf$rotation[5,2] + 0.05,
substitute(paste(italic(CH[4]))), col = "blue")

#Add legend
legend(0.164, 0.745, legend = c("Scores", "Loadings", "95% Confidence
Ellipse"), fill = c("black", "blue", "red"))

par(new = FALSE) #Allow new graphs to be plotted

#Collate data set for classification trees and support vector machines
new_dat = data.frame(B_solvs, m_dot_rw_gs_h2o_A, m_dot_rw_gs_h2_A,
m_dot_rw_gs_co_A, m_dot_rw_gs_co2_A, m_dot_rw_gs_ch4_A)

colnames(new_dat)[1] = "solvs"
colnames(new_dat)[2] = "m_h2o"
colnames(new_dat)[3] = "m_h2"
colnames(new_dat)[4] = "m_co"
colnames(new_dat)[5] = "m_co2"
colnames(new_dat)[6] = "m_ch4"

attach(new_dat)

#Training set
set.seed(7)
train = sample(x = nm_ent, size = nm_ent - 550)

#Classification trees

```

```

tree_gsf_h2_co = tree(solvs ~ m_h2 + m_co, data = new_dat, subset = train,
split = "gini")
tree_gsf_ch4_co = tree(solvs ~ m_ch4 + m_co, data = new_dat, subset =
train, split = "gini")
tree_gsf_ch4_co2 = tree(solvs ~ m_ch4 + m_co2, data = new_dat, subset =
train, split = "gini")
tree_gsf_h2o_co2 = tree(solvs ~ m_h2o + m_co2, data = new_dat, subset =
train, split = "gini")

#Function for determining tree paths for plotting decision boundaries
Path = function(j, indx_leaf, split, path_x)
{
  path_vect = matrix(data = 0, nrow = length(indx_leaf), ncol = 1) #Vector
for individual paths
  chng_drt = FALSE

  for(k in (indx_leaf[j] - j):1)
  {
    if(j == 1) #Condition for 1st path
    {
      path_vect[k,1] = split[k,1] - 0.01
    }else if(identical(path_x[k,1:j-1], rep(0, (j - 1))) == TRUE)
#Condition for new path on different branch
    {
      path_vect[k,1] = split[k,1] - 0.01
    }else if(chng_drt == TRUE) #Condition for change at end of a path
    {
      path_vect[k,1] = path_x[k,j-1]
    }else if(path_x[k,j-1] < split[k,1] && path_x[k,j-1] != 0) #Condition
for second path at terminal split
    {
      path_vect[k,1] = split[k,1] + 0.01
      chng_drt = TRUE
    }
  }

  return(path_vect[,1])
}

#Function to plot decision boundaries
plot_db = function(indx_split, var, m_1, m_2, split_1, split_2, path_x)
{
  j = 1
  k = 1
  for(i in 1:length(indx_split))
  {
    if(var[indx_split[i]] == 2) #Plotting perpendicular to y-axis for 1st
flow rate
    {
      if(k == 1) #Plot if no lines for the 2nd flow rate are present
      {
        lines(c(min(m_2) - 7, max(m_2) + 7), c(split_1[j], split_1[j]))
        j = j + 1
      }else
      {
        path_mat = as.matrix(path_x[1:i-1, which(path_x[i,] != 0)]) #Matrix
for path conditions that supersede present line
        dl_rw = 0
      }
    }
  }
}

```

```

    for(l in 1:(i - 1)) #Remove conditions that aren't applicable to
present line
    {
        if(var[indx_split[l]] == 2)
        {
            path_mat = as.matrix(path_mat[-(l-dl_rw),])
            dl_rw = dl_rw + 1
        }
    }
    path_mat = as.matrix(path_mat[apply(path_mat, 1,
function(x)all(x!=0)),]) #Remove 0's
    if(dim(path_mat)[2] == 1) #If single row becomes column, transpose
back to row
    {
        path_mat = t(path_mat)
    }
    rw = dim(path_mat)[1]

    comp_mat = matrix(data = NA, nrow = rw, ncol = 2) #Comparison
matrix for path conditions and splits
    comp_mat[,1] = path_mat[,1]
    comp_mat[,2] = split_2[1:rw]

    df_mat = matrix(data = NA, nrow =rw, ncol = 1) #Difference matrix
for path conditions and splits
    df_mat = as.matrix(round(comp_mat[,1] - comp_mat[,2], digits = 4))

    indx_pos_df = which(df_mat == 0.01) #Lower boundary conditions
    indx_neg_df = which(df_mat == -0.01) #Upper boundary conditions

    if(length(indx_pos_df) == 0 && length(indx_neg_df) != 0) #Only
upper boundary condition
    {
        upr_2 = min(comp_mat[indx_neg_df,1]) #Minimum upper boundary
        lines(c(min(m_2) - 5, upr_2), c(split_1[j], split_1[j]))
        j = j + 1
    }else if(length(indx_pos_df) != 0 && length(indx_neg_df) == 0)
#Only lower boundary condition
    {
        low_2 = max(comp_mat[indx_pos_df,1]) #Maximum lower boundary
        lines(c(low_2, max(m_2) + 5), c(split_1[j], split_1[j]))
        j = j + 1
    }

    }else if(length(indx_pos_df) != 0 && length(indx_neg_df) != 0)
#Upper and lower boundary condition
    {
        low_2 = max(comp_mat[indx_pos_df,1]) #Maximum lower boundary
        upr_2 = min(comp_mat[indx_neg_df,1]) #Minimum upper boundary
        lines(c(low_2, upr_2), c(split_1[j], split_1[j]))
        j = j + 1
    }
    }
}

if(var[indx_split[i]] == 3) #Plotting perpendicular to x-axis for 2nd
flow rate
{
    if(j == 1) #Plot if no lines for the 1st flow rate are present
    {
        lines(c(split_2[k], split_2[k]), c(min(m_1) - 7, max(m_1) + 7))
        k = k + 1
    }
}

```

```

    }else
    {
        path_mat = as.matrix(path_x[1:i-1, which(path_x[i,] != 0)]) #Matrix
for path conditions that supersede present line
        dl_rw = 0
        for(l in 1:(i - 1)) #Remove conditions that aren't applicable to
present line
        {
            if(var[indx_split[l]] == 3)
            {
                path_mat = as.matrix(path_mat[-(l-dl_rw),])
                dl_rw = dl_rw + 1
            }
        }
        path_mat = as.matrix(path_mat[apply(path_mat, 1,
function(x)all(x!=0)),]) #Remove 0's
        if(dim(path_mat)[2] == 1) #If single row becomes column, transpose
back to row
        {
            path_mat = t(path_mat)
        }
        rw = dim(path_mat)[1]

        comp_mat = matrix(data = NA, nrow = rw, ncol = 2) #Comparison
matrix for path conditions and splits
        comp_mat[,1] = path_mat[,1]
        comp_mat[,2] = split_1[1:rw]

        df_mat = matrix(data = NA, nrow =rw, ncol = 1) #Difference matrix
for path conditions and splits
        df_mat = as.matrix(round(comp_mat[,1] - comp_mat[,2], digits = 4))

        indx_pos_df = which(df_mat == 0.01) #Lower boundary conditions
        indx_neg_df = which(df_mat == -0.01) #Upper boundary conditions

        if(length(indx_pos_df) == 0 && length(indx_neg_df) != 0) #Only
upper boundary condition
        {
            upr_1 = min(comp_mat[indx_neg_df,1]) #Minimum upper boundary
            lines(c(split_2[k], split_2[k]), c(min(m_1) - 5, upr_1))
            k = k + 1
        }else if(length(indx_pos_df) != 0 && length(indx_neg_df) == 0)
#Only lower boundary condition
        {
            low_1 = max(comp_mat[indx_pos_df,1]) #Maximum lower boundary
            lines(c(split_2[k], split_2[k]), c(low_1, max(m_1) + 5))
            k = k + 1
        }else if(length(indx_pos_df) != 0 && length(indx_neg_df) != 0)
#Upper and lower boundary condition
        {
            low_1 = max(comp_mat[indx_pos_df,1]) #Maximum lower boundary
            upr_1 = min(comp_mat[indx_neg_df,1]) #Minimum upper boundary
            lines(c(split_2[k], split_2[k]), c(low_1, upr_1))
            k = k + 1
        }
    }
}
}
}
}
}

```

```

#Prune tree to smallest size and lowest 10-fold cross-validation error rate
set.seed(3)

#H2 and CO
cv_h2_co = cv.tree(tree_gsf_h2_co, FUN = prune.misclass, K = 10)
min_h2_co = which(cv_h2_co$dev == min(cv_h2_co$dev))
len_min_h2_co = length(min_h2_co)
prune_h2_co = prune.misclass(tree_gsf_h2_co, best =
cv_h2_co$size[min_h2_co[len_min_h2_co]])

#CH4 and CO
cv_ch4_co = cv.tree(tree_gsf_ch4_co, FUN = prune.misclass, K = 10)
min_ch4_co = which(cv_ch4_co$dev == min(cv_ch4_co$dev))
len_min_ch4_co = length(min_ch4_co)
prune_ch4_co = prune.misclass(tree_gsf_ch4_co, best =
cv_ch4_co$size[min_ch4_co[len_min_ch4_co]])

#CH4 and CO2
cv_ch4_co2 = cv.tree(tree_gsf_ch4_co2, FUN = prune.misclass, K = 10)
min_ch4_co2 = which(cv_ch4_co2$dev == min(cv_ch4_co2$dev))
len_min_ch4_co2 = length(min_ch4_co2)
prune_ch4_co2 = prune.misclass(tree_gsf_ch4_co2, best =
cv_ch4_co2$size[min_ch4_co2[len_min_ch4_co2]])

#H2O and CO2
cv_h2o_co2 = cv.tree(tree_gsf_h2o_co2, FUN = prune.misclass, K = 10)
min_h2o_co2 = which(cv_h2o_co2$dev == min(cv_h2o_co2$dev))
len_min_h2o_co2 = length(min_h2o_co2)
prune_h2o_co2 = prune.misclass(tree_gsf_h2o_co2, best =
cv_h2o_co2$size[min_h2o_co2[len_min_h2o_co2]])

#Plot classification trees
par(mar=c(5,5,5,2) + .1)

#H2 and CO
plot(m_co, m_h2, main = "Hydrogen to Carbon Monoxide", xlab =
substitute(paste(italic(CO), " Mass Flow Rate [kg/s]")), xlim = c(0, 12),
ylim = c(-3, 2),
ylab = substitute(paste(italic(H[2]), " Mass Flow Rate [kg/s]")), col
= (as.numeric(solvs) + 2))

#Classification tree decision structure
var_h2_co = as.numeric(prune_h2_co$frame$var) #Vector containing 1 for
leaves, 2 for H2 splits, and 3 for CO splits

indx_leaf_h2_co = which(var_h2_co == 1) #Leaf index

indx_split_h2_co = which(var_h2_co != 1) #Split index
split_h2_co = matrix(data = 0, nrow = length(indx_split_h2_co), ncol = 1)
#Matrix for H2 and CO split values

split_h2 = matrix(data = 0, nrow = length(which(var_h2_co == 2)), ncol = 1)
#Matrix for H2 split values

split_co = matrix(data = 0, nrow = length(which(var_h2_co == 3)), ncol = 1)
#Matrix for CO split values

#Extract split values

```

```

j = 1
k = 1
l = 1
for(i in 1:length(var_h2_co))
{
  if(var_h2_co[i] != 1) #Condition for H2 and CO splits
  {
    split_h2_co[l] = as.numeric(sub(".", "",
prune_h2_co$frame$splits[i,1]))
    l = l + 1
  }

  if(var_h2_co[i] == 2) #Condition for H2 splits
  {
    split_h2[j] = as.numeric(sub(".", "", prune_h2_co$frame$splits[i,1]))
    j = j + 1
  }

  if(var_h2_co[i] == 3) #Condition for CO splits
  {
    split_co[k] = as.numeric(sub(".", "", prune_h2_co$frame$splits[i,1]))
    k = k + 1
  }
}

#Tree path matrix
path_h2_co = matrix(data = 0, nrow = length(which(var_h2_co == 1)), ncol =
length(which(var_h2_co == 1)))

for(i in 1:length(indx_leaf_h2_co))
{
  path_h2_co[,i] = Path(i, indx_leaf_h2_co, split_h2_co, path_h2_co)
}

#Plot decision boundaries
plot_db(indx_split_h2_co, var_h2_co, m_h2, m_co, split_h2, split_co,
path_h2_co)

#Plot 95% confidence ellipse
elips_h2_co = cbind(m_co[p_set], m_h2[p_set])

cov_h2_co = cov(elips_h2_co) #Covariance of H2 and CO
mean_co = mean(elips_h2_co[,1]) #Average and center of CO
mean_h2 = mean(elips_h2_co[,2]) #Average and center of H2
center_h2_co = c(mean_co, mean_h2)

lines(ellipse(cov_h2_co, centre = center_h2_co, level = 0.95), col = "red")

#Shade regions determined by classification tree
co_plot = seq(from = floor(min(m_co)), to = ceiling(max(m_co)) + 0.5, by =
0.25) #CO values for grid
h2_plot = seq(from = floor(min(m_h2)) - 0.25, to = ceiling(max(m_h2)) +
0.25, by = 0.1) #H2 values for grid

for(i in 1:length(co_plot))
{
  for(j in 1:length(h2_plot))
  {
    temp_pred = predict(prune_h2_co, newdata = data.frame(m_h2 =
h2_plot[j], m_co = co_plot[i]), type = "class")

```

```

    if(temp_pred == TRUE) #Condition for grid color
    {
      points(co_plot[i], h2_plot[j], pch = '/', col = rgb(0, 0, 1, 0.4))
    } else
    {
      points(co_plot[i], h2_plot[j], pch = '\\', col = rgb(0, 1, 0, 0.4))
    }
  }
}

#Add legend
legend(7.95, 2.15, legend = c("Model B Solves", "Model B Does Not Solve",
"95% Confidence Ellipse", "Boundaries"), fill = c("blue", "green", "red",
"black"))

#CH4 and CO
plot(m_co, m_ch4, main = "Methane to Carbon Monoxide", xlab =
substitute(paste(italic(CO), " Mass Flow Rate [kg/s]")), xlim = c(0, 12),
ylim = c(-1.5, 12.5),
      ylab = substitute(paste(italic(CH[4]), " Mass Flow Rate [kg/s]")), col
= (as.numeric(solvs) + 2))

#Classification tree decision structure
var_ch4_co = as.numeric(prune_ch4_co$frame$var) #Vector containing 1 for
leaves, 2 for CH4 splits, and 3 for CO splits

indx_leaf_ch4_co = which(var_ch4_co == 1) #Leaf index

indx_split_ch4_co = which(var_ch4_co != 1) #Split index
split_ch4_co = matrix(data = 0, nrow = length(indx_split_ch4_co), ncol = 1)
#Matrix for CH4 and CO split values

split_ch4 = matrix(data = 0, nrow = length(which(var_ch4_co == 2)), ncol =
1) #Matrix for CH4 split values

split_co = matrix(data = 0, nrow = length(which(var_ch4_co == 3)), ncol =
1) #Matrix for CO split values

#Extract split values
j = 1
k = 1
l = 1
for(i in 1:length(var_ch4_co))
{
  if(var_ch4_co[i] != 1) #Condition for CH4 and CO splits
  {
    split_ch4_co[l] = as.numeric(sub(".", "",
prune_ch4_co$frame$splits[i,1]))
    l = l + 1
  }

  if(var_ch4_co[i] == 2) #Condition for CH4 splits
  {
    split_ch4[j] = as.numeric(sub(".", "", prune_ch4_co$frame$splits[i,1]))
    j = j + 1
  }

  if(var_ch4_co[i] == 3) #Condition for CO splits
  {
    split_co[k] = as.numeric(sub(".", "", prune_ch4_co$frame$splits[i,1]))

```

```

    k = k + 1
  }
}

#Tree path matrix
path_ch4_co = matrix(data = 0, nrow = length(which(var_ch4_co == 1)), ncol
= length(which(var_ch4_co == 1)))

for(i in 1:length(indx_leaf_ch4_co))
{
  path_ch4_co[,i] = Path(i, indx_leaf_ch4_co, split_ch4_co, path_ch4_co)
}

#Plot decision boundaries
plot_db(indx_split_ch4_co, var_ch4_co, m_ch4, m_co, split_ch4, split_co,
path_ch4_co)

#Plot 95% confidence ellipse
elips_ch4_co = cbind(m_co[p_set], m_ch4[p_set])

cov_ch4_co = cov(elips_ch4_co) #Covariance of CH4 and CO
mean_co = mean(elips_ch4_co[,1]) #Average and center of CO
mean_ch4 = mean(elips_ch4_co[,2]) #Average and center of CH4
center_ch4_co = c(mean_co, mean_ch4)

lines(ellipse(cov_ch4_co, centre = center_ch4_co, level = 0.95), col =
"red")

#Shade regions determined by classification tree
co_plot = seq(from = floor(min(m_co)), to = ceiling(max(m_co)) + 0.5, by =
0.25) #CO values for grid
ch4_plot = seq(from = floor(min(m_ch4)) - 0.25, to = ceiling(max(m_ch4)) +
0.25, by = 0.25) #CH4 values for grid

for(i in 1:length(co_plot))
{
  for(j in 1:length(ch4_plot))
  {
    temp_pred = predict(prune_ch4_co, newdata = data.frame(m_ch4 =
ch4_plot[j], m_co = co_plot[i]), type = "class")

    if(temp_pred == TRUE) #Condition for grid color
    {
      points(co_plot[i], ch4_plot[j], pch = '/', col = rgb(0, 0, 1, 0.4))
    } else
    {
      points(co_plot[i], ch4_plot[j], pch = '\\', col = rgb(0, 1, 0, 0.4))
    }
  }
}

#Add legend
legend(-0.35, 12.95, legend = c("Model B Solves", "Model B Does Not Solve",
"95% Confidence Ellipse", "Boundaries"), fill = c("blue", "green", "red",
"black"))

#CH4 and CO2
plot(m_co2, m_ch4, main = "Methane to Carbon Dioxide", xlab =
substitute(paste(italic(CO[2]), " Mass Flow Rate [kg/s]")), xlim = c(-4,
24), ylim = c(-1.5, 12.5),

```

```

    ylab = substitute(paste(italic(CH[4]), " Mass Flow Rate [kg/s]")), col
= (as.numeric(solvs) + 2))

#Classification tree decision structure
var_ch4_co2 = as.numeric(prune_ch4_co2$frame$var) #Vector containing 1 for
leaves, 2 for CH4 splits, and 3 for CO2 splits

indx_leaf_ch4_co2 = which(var_ch4_co2 == 1) #Leaf index

indx_split_ch4_co2 = which(var_ch4_co2 != 1) #Split index
split_ch4_co2 = matrix(data = 0, nrow = length(indx_split_ch4_co2), ncol =
1) #Matrix for CH4 and CO2 split values

split_ch4 = matrix(data = 0, nrow = length(which(var_ch4_co2 == 2)), ncol =
1) #Matrix for CH4 split values

split_co2 = matrix(data = 0, nrow = length(which(var_ch4_co2 == 3)), ncol =
1) #Matrix for CO2 split values

#Extract split values
j = 1
k = 1
l = 1
for(i in 1:length(var_ch4_co2))
{
  if(var_ch4_co2[i] != 1) #Condition for CH4 and CO2 splits
  {
    split_ch4_co2[l] = as.numeric(sub(".", "",
prune_ch4_co2$frame$splits[i,1]))
    l = l + 1
  }

  if(var_ch4_co2[i] == 2) #Condition for CH4 splits
  {
    split_ch4[j] = as.numeric(sub(".", "",
prune_ch4_co2$frame$splits[i,1]))
    j = j + 1
  }

  if(var_ch4_co2[i] == 3) #Condition for CO2 splits
  {
    split_co2[k] = as.numeric(sub(".", "",
prune_ch4_co2$frame$splits[i,1]))
    k = k + 1
  }
}

#Tree path matrix
path_ch4_co2 = matrix(data = 0, nrow = length(which(var_ch4_co2 == 1)),
ncol = length(which(var_ch4_co2 == 1)))

for(i in 1:length(indx_leaf_ch4_co2))
{
  path_ch4_co2[,i] = Path(i, indx_leaf_ch4_co2, split_ch4_co2,
path_ch4_co2)
}

#Plot decision boundaries
plot_db(indx_split_ch4_co2, var_ch4_co2, m_ch4, m_co2, split_ch4,
split_co2, path_ch4_co2)

```

```

#Plot 95% confidence ellipse
elips_ch4_co2 = cbind(m_co2[p_set], m_ch4[p_set])

cov_ch4_co2 = cov(elips_ch4_co2) #Covariance of CH4 and CO2
mean_co2 = mean(elips_ch4_co2[,1]) #Average and center of CO2
mean_ch4 = mean(elips_ch4_co2[,2]) #Average and center of CH4
center_ch4_co2 = c(mean_co2, mean_ch4)

lines(ellipse(cov_ch4_co2, centre = center_ch4_co2, level = 0.95), col =
"red")

#Shade regions determined by classification tree
co2_plot = seq(from = floor(min(m_co2)) - 1, to = ceiling(max(m_co2)) + 3,
by = 0.65) #CO2 values for grid
ch4_plot = seq(from = floor(min(m_ch4)) - 0.25, to = ceiling(max(m_ch4)) +
0.25, by = 0.25) #CH4 values for grid

for(i in 1:length(co2_plot))
{
  for(j in 1:length(ch4_plot))
  {
    temp_pred = predict(prune_ch4_co2, newdata = data.frame(m_ch4 =
ch4_plot[j], m_co2 = co2_plot[i]), type = "class")

    if(temp_pred == TRUE) #Condition for grid color
    {
      points(co2_plot[i], ch4_plot[j], pch = '/', col = rgb(0, 0, 1, 0.4))
    } else
    {
      points(co2_plot[i], ch4_plot[j], pch = '\\', col = rgb(0, 1, 0, 0.4))
    }
  }
}

#Add legend
legend(14.6, 12.95, legend = c("Model B Solves", "Model B Does Not Solve",
"95% Confidence Ellipse", "Boundaries"), fill = c("blue", "green", "red",
"black"))

#H2O and CO2
plot(m_co2, m_h2o, main = "Steam to Carbon Dioxide", xlab =
substitute(paste(italic(CO[2]), " Mass Flow Rate [kg/s]")), xlim = c(-4,
34), ylim = c(-14.5, 9.5),
ylab = substitute(paste(italic(H[2]), italic(O), " Mass Flow Rate
[kg/s]")), col = (as.numeric(solvs) + 2))

#Classification tree decision structure
var_h2o_co2 = as.numeric(prune_h2o_co2$frame$var) #Vector containing 1 for
leaves, 2 for H2O splits, and 3 for CO2 splits

indx_leaf_h2o_co2 = which(var_h2o_co2 == 1) #Leaf index

indx_split_h2o_co2 = which(var_h2o_co2 != 1) #Split index
split_h2o_co2 = matrix(data = 0, nrow = length(indx_split_h2o_co2), ncol =
1) #Matrix for H2O and CO2 split values

split_h2o = matrix(data = 0, nrow = length(which(var_h2o_co2 == 2)), ncol =
1) #Matrix for H2O split values

```

```

split_co2 = matrix(data = 0, nrow = length(which(var_h2o_co2 == 3)), ncol =
1) #Matrix for CO2 split values

#Extract split values
j = 1
k = 1
l = 1
for(i in 1:length(var_h2o_co2))
{
  if(var_h2o_co2[i] != 1) #Condition for H2O and CO2 splits
  {
    split_h2o_co2[l] = as.numeric(sub(".", "",
prune_h2o_co2$frame$splits[i,1]))
    l = l + 1
  }

  if(var_h2o_co2[i] == 2) #Condition for H2O splits
  {
    split_h2o[j] = as.numeric(sub(".", "",
prune_h2o_co2$frame$splits[i,1]))
    j = j + 1
  }

  if(var_h2o_co2[i] == 3) #Condition for CO2 splits
  {
    split_co2[k] = as.numeric(sub(".", "",
prune_h2o_co2$frame$splits[i,1]))
    k = k + 1
  }
}

#Tree path matrix
path_h2o_co2 = matrix(data = 0, nrow = length(which(var_h2o_co2 == 1)),
ncol = length(which(var_h2o_co2 == 1)))

for(i in 1:length(indx_leaf_h2o_co2))
{
  path_h2o_co2[,i] = Path(i, indx_leaf_h2o_co2, split_h2o_co2,
path_h2o_co2)
}

#Plot decision boundaries
plot_db(indx_split_h2o_co2, var_h2o_co2, m_h2o, m_co2, split_h2o,
split_co2, path_h2o_co2)

#Plot 95% confidence ellipse
elips_h2o_co2 = cbind(m_co2[p_set], m_h2o[p_set])

cov_h2o_co2 = cov(elips_h2o_co2) #Covariance of H2O and CO2
mean_co2 = mean(elips_h2o_co2[,1]) #Average and center of CO2
mean_h2o = mean(elips_h2o_co2[,2]) #Average and center of H2O
center_h2o_co2 = c(mean_co2, mean_h2o)

lines(ellipse(cov_h2o_co2, centre = center_h2o_co2, level = 0.95), col =
"red")

#Shade regions determined by classification tree
co2_plot = seq(from = floor(min(m_co2)) - 1, to = ceiling(max(m_co2)) + 15,
by = 0.9) #CO2 values for grid
h2o_plot = seq(from = floor(min(m_h2o)) - 0.25, to = ceiling(max(m_h2o)) +
3, by = 0.5) #H2O values for grid

```

```

for(i in 1:length(co2_plot))
{
  for(j in 1:length(h2o_plot))
  {
    temp_pred = predict(prune_h2o_co2, newdata = data.frame(m_h2o =
h2o_plot[j], m_co2 = co2_plot[i]), type = "class")

    if(temp_pred == TRUE) #Condition for grid color
    {
      points(co2_plot[i], h2o_plot[j], pch = '/', col = rgb(0, 0, 1, 0.4))
    } else
    {
      points(co2_plot[i], h2o_plot[j], pch = '\\', col = rgb(0, 1, 0, 0.4))
    }
  }
}

#Add legend
legend(-5.1, 10.25, legend = c("Model B Solves", "Model B Does Not Solve",
"95% Confidence Ellipse", "Boundaries"), fill = c("blue", "green", "red",
"black"))

#Determine classification tree train and test accuracy and inaccuracy
#H2 and CO
pred_train_h2_co = predict(prune_h2_co, new_dat[train,], type = "class")
#Training set predictions
tabl_train_h2_co = table(pred_train_h2_co, solvs[train]) #Confusion matrix
ndr_train_h2_co = tabl_train_h2_co[2,1]/length(which(B_solvs[train] ==
FALSE))*100 #Non-detection rate
far_train_h2_co = tabl_train_h2_co[1,2]/length(which(B_solvs[train] ==
TRUE))*100 #False-alarm rate
acc_train_h2_co = (tabl_train_h2_co[1,1] +
tabl_train_h2_co[2,2])/length(B_solvs[train])*100 #Accuracy

pred_test_h2_co = predict(prune_h2_co, new_dat[-train,], type = "class")
#Test set predictions
tabl_test_h2_co = table(pred_test_h2_co, solvs[-train]) #Confusion matrix
ndr_test_h2_co = tabl_test_h2_co[2,1]/length(which(B_solvs[-train] ==
FALSE))*100 #Non-detection rate
far_test_h2_co = tabl_test_h2_co[1,2]/length(which(B_solvs[-train] ==
TRUE))*100 #False-alarm rate
acc_test_h2_co = (tabl_test_h2_co[1,1] +
tabl_test_h2_co[2,2])/length(B_solvs[-train])*100 #Accuracy

#CH4 and CO
pred_train_ch4_co = predict(prune_ch4_co, new_dat[train,], type = "class")
#Training set predictions
tabl_train_ch4_co = table(pred_train_ch4_co, solvs[train]) #Confusion
matrix
ndr_train_ch4_co = tabl_train_ch4_co[2,1]/length(which(B_solvs[train] ==
FALSE))*100 #Non-detection rate
far_train_ch4_co = tabl_train_ch4_co[1,2]/length(which(B_solvs[train] ==
TRUE))*100 #False-alarm rate
acc_train_ch4_co = (tabl_train_ch4_co[1,1] +
tabl_train_ch4_co[2,2])/length(B_solvs[train])*100 #Accuracy

pred_test_ch4_co = predict(prune_ch4_co, new_dat[-train,], type = "class")
#Test set predictions
tabl_test_ch4_co = table(pred_test_ch4_co, solvs[-train]) #Confusion matrix

```

```

ndr_test_ch4_co = tabl_test_ch4_co[2,1]/length(which(B_solvs[-train] ==
FALSE))*100 #Non-detection rate
far_test_ch4_co = tabl_test_ch4_co[1,2]/length(which(B_solvs[-train] ==
TRUE))*100 #False-alarm rate
acc_test_ch4_co = (tabl_test_ch4_co[1,1] +
tabl_test_ch4_co[2,2])/length(B_solvs[-train])*100 #Accuracy

#CH4 and CO2
pred_train_ch4_co2 = predict(prune_ch4_co2, new_dat[train,], type =
"class") #Training set predictions
tabl_train_ch4_co2 = table(pred_train_ch4_co2, solvs[train]) #Confusion
matrix
ndr_train_ch4_co2 = tabl_train_ch4_co2[2,1]/length(which(B_solvs[train] ==
FALSE))*100 #Non-detection rate
far_train_ch4_co2 = tabl_train_ch4_co2[1,2]/length(which(B_solvs[train] ==
TRUE))*100 #False-alarm rate
acc_train_ch4_co2 = (tabl_train_ch4_co2[1,1] +
tabl_train_ch4_co2[2,2])/length(B_solvs[train])*100 #Accuracy

pred_test_ch4_co2 = predict(prune_ch4_co2, new_dat[-train,], type =
"class") #Test set predictions
tabl_test_ch4_co2 = table(pred_test_ch4_co2, solvs[-train]) #Confusion
matrix
ndr_test_ch4_co2 = tabl_test_ch4_co2[2,1]/length(which(B_solvs[-train] ==
FALSE))*100 #Non-detection rate
far_test_ch4_co2 = tabl_test_ch4_co2[1,2]/length(which(B_solvs[-train] ==
TRUE))*100 #False-alarm rate
acc_test_ch4_co2 = (tabl_test_ch4_co2[1,1] +
tabl_test_ch4_co2[2,2])/length(B_solvs[-train])*100 #accuracy

#H2O and CO2
pred_train_h2o_co2 = predict(prune_h2o_co2, new_dat[train,], type =
"class") #Training set predictions
tabl_train_h2o_co2 = table(pred_train_h2o_co2, solvs[train]) #Confusion
matrix
ndr_train_h2o_co2 = tabl_train_h2o_co2[2,1]/length(which(B_solvs[train] ==
FALSE))*100 #Non-detection rate
far_train_h2o_co2 = tabl_train_h2o_co2[1,2]/length(which(B_solvs[train] ==
TRUE))*100 #False-alarm rate
acc_train_h2o_co2 = (tabl_train_h2o_co2[1,1] +
tabl_train_h2o_co2[2,2])/length(B_solvs[train])*100 #Accuracy

pred_test_h2o_co2 = predict(prune_h2o_co2, new_dat[-train,], type =
"class") #Test set predictions
tabl_test_h2o_co2 = table(pred_test_h2o_co2, solvs[-train]) #Confusion
matrix
ndr_test_h2o_co2 = tabl_test_h2o_co2[2,1]/length(which(B_solvs[-train] ==
FALSE))*100 #Non-detection rate
far_test_h2o_co2 = tabl_test_h2o_co2[1,2]/length(which(B_solvs[-train] ==
TRUE))*100 #False-alarm rate
acc_test_h2o_co2 = (tabl_test_h2o_co2[1,1] +
tabl_test_h2o_co2[2,2])/length(B_solvs[-train])*100 #accuracy

#Training set
set.seed(7)
train = sample(x = nm_ent, size = nm_ent - 550)

#Support Vector machines

```

```

svm_gsf_h2_co = svm(solvs ~ m_h2 + m_co, data = new_dat, subset = train,
kernel = "polynomial", degree = 2)
svm_gsf_ch4_co = svm(solvs ~ m_ch4 + m_co, data = new_dat, subset = train,
kernel = "polynomial", degree = 2)
svm_gsf_ch4_co2 = svm(solvs ~ m_ch4 + m_co2, data = new_dat, subset =
train, kernel = "polynomial", degree = 2)
svm_gsf_h2o_co2 = svm(solvs ~ m_h2o + m_co2, data = new_dat, subset =
train, kernel = "polynomial", degree = 2)

#Plot support vector machines
par(mar=c(5,5,5,2) + .1)

#H2 and CO
plot(m_co, m_h2, main = "Hydrogen to Carbon Monoxide", xlab =
substitute(paste(italic(CO), " Mass Flow Rate [kg/s]")), xlim = c(0, 12),
ylim = c(-3, 2),
      ylab = substitute(paste(italic(H[2]), " Mass Flow Rate [kg/s]")), col
= (as.numeric(solvs) + 2))

#Plot 95% confidence ellipse
elips_h2_co = cbind(m_co[p_set], m_h2[p_set])

cov_h2_co = cov(elips_h2_co) #Covariance of H2 and CO
mean_co = mean(elips_h2_co[,1]) #Average and center of CO
mean_h2 = mean(elips_h2_co[,2]) #Average and center of H2
center_h2_co = c(mean_co, mean_h2)

lines(ellipse(cov_h2_co, centre = center_h2_co, level = 0.95), col = "red")

#Shade regions determined by support vector machine
co_plot = seq(from = floor(min(m_co)), to = ceiling(max(m_co)) + 0.5, by =
0.25) #CO values for grid
h2_plot = seq(from = floor(min(m_h2)) - 0.25, to = ceiling(max(m_h2)) +
0.25, by = 0.1) #H2 values for grid

for(i in 1:length(co_plot))
{
  for(j in 1:length(h2_plot))
  {
    temp_pred = predict(svm_gsf_h2_co, newdata = data.frame(m_h2 =
h2_plot[j], m_co = co_plot[i]), type = "class", decision.values = TRUE)

    if(temp_pred == TRUE) #Condition for grid color
    {
      points(co_plot[i], h2_plot[j], pch = '/', col = rgb(0, 0, 1, 0.4))
    } else
    {
      points(co_plot[i], h2_plot[j], pch = '\\', col = rgb(0, 1, 0, 0.4))
    }
  }
}

#Add boundary curves
co_plot = seq(from = floor(min(m_co)), to = ceiling(max(m_co)) + 0.5, by =
0.1) #CO values for grid
h2_plot = seq(from = floor(min(m_h2)) - 0.25, to = ceiling(max(m_h2)) +
0.25, by = 0.05) #H2 values for grid

```

```

bndr_co = matrix(data = NA, nrow = 9*length(co_plot), ncol = 1) #CO
boundary points
bndr_h2 = matrix(data = NA, nrow = 9*length(h2_plot), ncol = 1) #H2
boundary points

k = 1
for(i in 1:length(co_plot))
{
  for(j in 1:length(h2_plot))
  {
    temp_pred = predict(svm_gsf_h2_co, newdata = data.frame(m_h2 =
h2_plot[j], m_co = co_plot[i]), type = "class", decision.values = TRUE)

    if(round(as.numeric(attr(temp_pred, "decision.values")), digits = 0) ==
0) #Condition for boundary points
    {
      bndr_co[k,1] = co_plot[i]
      bndr_h2[k,1] = h2_plot[j]
      k = k + 1
    }
  }
}

bndr_co = na.omit(bndr_co)
bndr_h2 = na.omit(bndr_h2)
bndr = data.frame(bndr_co, bndr_h2)

bndr_pca = prcomp(bndr, scale = TRUE) #PCA to make two boundary lines
separable
bndr_clst = kmeans(bndr_pca$x[,2], 2, nstart = 50) #K-means clusters for
two boundary lines
clst_indx = which(bndr_clst$cluster == 1) #Cluster index

spl1 = smooth.spline(bndr_co[clst_indx], bndr_h2[clst_indx]) #Spline curve
for the first line
spl2 = smooth.spline(bndr_co[-clst_indx], bndr_h2[-clst_indx]) #Spline
curve for the second line
lines(spl1)
lines(spl2)

#Add legend
legend(7.95, 2.15, legend = c("Model B Solves", "Model B Does Not Solve",
"95% Confidence Ellipse", "Boundaries"), fill = c("blue", "green", "red",
"black"))

#CH4 and CO
plot(m_co, m_ch4, main = "Methane to Carbon Monoxide", xlab =
substitute(paste(italic(CO), " Mass Flow Rate [kg/s]")), xlim = c(0, 12),
ylim = c(-1.5, 12.5),
ylab = substitute(paste(italic(CH[4]), " Mass Flow Rate [kg/s]")), col
= (as.numeric(solvs) + 2))

#Plot 95% confidence ellipse
elips_ch4_co = cbind(m_co[p_set], m_ch4[p_set])

cov_ch4_co = cov(elips_ch4_co) #Covariance of CH4 and CO
mean_co = mean(elips_ch4_co[,1]) #Average and center of CO
mean_ch4 = mean(elips_ch4_co[,2]) #Average and center of CH4
center_ch4_co = c(mean_co, mean_ch4)

```

```

lines(ellipse(cov_ch4_co, centre = center_ch4_co, level = 0.95), col =
"red")

#Shade regions determined by support vector machine
co_plot = seq(from = floor(min(m_co)), to = ceiling(max(m_co)) + 0.5, by =
0.25) #CO values for grid
ch4_plot = seq(from = floor(min(m_ch4)) - 0.25, to = ceiling(max(m_ch4)) +
0.25, by = 0.25) #CH4 values for grid

for(i in 1:length(co_plot))
{
  for(j in 1:length(ch4_plot))
  {
    temp_pred = predict(svm_gsf_ch4_co, newdata = data.frame(m_ch4 =
ch4_plot[j], m_co = co_plot[i]), type = "class", decision.values = TRUE)

    if(temp_pred == TRUE) #Condition for grid color
    {
      points(co_plot[i], ch4_plot[j], pch = '/', col = rgb(0, 0, 1, 0.4))
    } else
    {
      points(co_plot[i], ch4_plot[j], pch = '\\', col = rgb(0, 1, 0, 0.4))
    }
  }
}

#Add boundary curves
co_plot = seq(from = floor(min(m_co)), to = ceiling(max(m_co)) + 0.5, by =
0.1) #CO values for grid
ch4_plot = seq(from = floor(min(m_ch4)) - 0.25, to = ceiling(max(m_ch4)) +
0.25, by = 0.1) #CH4 values for grid

bndr_co = matrix(data = NA, nrow = 9*length(co_plot), ncol = 1) #CO
boundary points
bndr_ch4 = matrix(data = NA, nrow = 9*length(ch4_plot), ncol = 1) #CH4
boundary points

k = 1
for(i in 1:length(co_plot))
{
  for(j in 1:length(ch4_plot))
  {
    temp_pred = predict(svm_gsf_ch4_co, newdata = data.frame(m_ch4 =
ch4_plot[j], m_co = co_plot[i]), type = "class", decision.values = TRUE)

    if(round(as.numeric(attr(temp_pred, "decision.values")), digits = 0) ==
0) #Condition for boundary points
    {
      bndr_co[k,1] = co_plot[i]
      bndr_ch4[k,1] = ch4_plot[j]
      k = k + 1
    }
  }
}

bndr_co = na.omit(bndr_co)
bndr_ch4 = na.omit(bndr_ch4)

bndr_clst = kmeans(bndr_ch4, 2, nstart = 50) #K-means clusters for two
boundary lines
clst_indx = which(bndr_clst$cluster == 1) #Cluster index

```

```

spl1 = smooth.spline(bndr_co[clst_indx], bndr_ch4[clst_indx]) #Spline curve
for the first line
spl2 = smooth.spline(bndr_co[-clst_indx], bndr_ch4[-clst_indx]) #Spline
curve for the second line
lines(spl1)
lines(spl2)

#Add legend
legend(-0.35, 12.95, legend = c("Model B Solves", "Model B Does Not Solve",
"95% Confidence Ellipse", "Boundaries"), fill = c("blue", "green", "red",
"black"))

#CH4 and CO2
plot(m_co2, m_ch4, main = "Methane to Carbon Dioxide", xlab =
substitute(paste(italic(CO[2]), " Mass Flow Rate [kg/s]")), xlim = c(-4,
24), ylim = c(-1.5, 12.5),
ylab = substitute(paste(italic(CH[4]), " Mass Flow Rate [kg/s]")), col
= (as.numeric(solvs) + 2))

#Plot 95% confidence ellipse
elips_ch4_co2 = cbind(m_co2[p_set], m_ch4[p_set])

cov_ch4_co2 = cov(elips_ch4_co2) #Covariance of CH4 and CO2
mean_co2 = mean(elips_ch4_co2[,1]) #Average and center of CO2
mean_ch4 = mean(elips_ch4_co2[,2]) #Average and center of CH4
center_ch4_co2 = c(mean_co2, mean_ch4)

lines(ellipse(cov_ch4_co2, centre = center_ch4_co2, level = 0.95), col =
"red")

#Shade regions determined by support vector machine
co2_plot = seq(from = floor(min(m_co2)) - 1, to = ceiling(max(m_co2)) + 3,
by = 0.65) #CO2 values for grid
ch4_plot = seq(from = floor(min(m_ch4)) - 0.25, to = ceiling(max(m_ch4)) +
0.25, by = 0.25) #CH4 values for grid

for(i in 1:length(co2_plot))
{
  for(j in 1:length(ch4_plot))
  {
    temp_pred = predict(svm_gsf_ch4_co2, newdata = data.frame(m_ch4 =
ch4_plot[j], m_co2 = co2_plot[i]), type = "class", decision.values = TRUE)

    if(temp_pred == TRUE) #Condition for grid color
    {
      points(co2_plot[i], ch4_plot[j], pch = '/', col = rgb(0, 0, 1, 0.4))
    } else
    {
      points(co2_plot[i], ch4_plot[j], pch = '\\', col = rgb(0, 1, 0, 0.4))
    }
  }
}

#Add boundary curves
co2_plot = seq(from = floor(min(m_co2)) - 1, to = ceiling(max(m_co2)) + 3,
by = 0.3) #CO2 values for grid
ch4_plot = seq(from = floor(min(m_ch4)) - 0.25, to = ceiling(max(m_ch4)) +
0.25, by = 0.1) #CH4 values for grid

```

```

bndr_co2 = matrix(data = NA, nrow = 9*length(co2_plot), ncol = 1) #CO2
boundary points
bndr_ch4 = matrix(data = NA, nrow = 9*length(ch4_plot), ncol = 1) #CH4
boundary points

k = 1
for(i in 1:length(co2_plot))
{
  for(j in 1:length(ch4_plot))
  {
    temp_pred = predict(svm_gsf_ch4_co2, newdata = data.frame(m_ch4 =
ch4_plot[j], m_co2 = co2_plot[i]), type = "class", decision.values = TRUE)

    if(round(as.numeric(attr(temp_pred, "decision.values")), digits = 0) ==
0) #Condition for boundary points
    {
      bndr_co2[k,1] = co2_plot[i]
      bndr_ch4[k,1] = ch4_plot[j]
      k = k + 1
    }
  }
}

bndr_co2 = na.omit(bndr_co2)
bndr_ch4 = na.omit(bndr_ch4)

bndr_clst = kmeans(bndr_ch4, 2, nstart = 50) #K-means clusters for two
boundary lines
clst_indx = which(bndr_clst$cluster == 1) #Cluster index

spl1 = smooth.spline(bndr_co2[clst_indx], bndr_ch4[clst_indx]) #Spline
curve for the first line
spl2 = smooth.spline(bndr_co2[-clst_indx], bndr_ch4[-clst_indx]) #Spline
curve for the second line
lines(spl1)
lines(spl2)

#Add legend
legend(14.6, 12.95, legend = c("Model B Solves", "Model B Does Not Solve",
"95% Confidence Ellipse", "Boundaries"), fill = c("blue", "green", "red",
"black"))

#H2O and CO2
plot(m_co2, m_h2o, main = "Steam to Carbon Dioxide", xlab =
substitute(paste(italic(CO[2]), " Mass Flow Rate [kg/s]")), xlim = c(-4,
34), ylim = c(-14.5, 9.5),
ylab = substitute(paste(italic(H[2]), italic(O), " Mass Flow Rate
[kg/s]")), col = (as.numeric(solvs) + 2))

#Plot 95% confidence ellipse
elips_h2o_co2 = cbind(m_co2[p_set], m_h2o[p_set])

cov_h2o_co2 = cov(elips_h2o_co2) #Covariance of CO2 and H2O
mean_co2 = mean(elips_h2o_co2[,1]) #Average and center of CO2
mean_h2o = mean(elips_h2o_co2[,2]) #Average and center of H2O
center_h2o_co2 = c(mean_co2, mean_h2o)

lines(ellipse(cov_h2o_co2, centre = center_h2o_co2, level = 0.95), col =
"red")

```

```

#Shade regions determined by support vector machine
co2_plot = seq(from = floor(min(m_co2)) - 1, to = ceiling(max(m_co2)) + 15,
by = 0.9) #CO2 values for grid
h2o_plot = seq(from = floor(min(m_h2o)) - 0.25, to = ceiling(max(m_h2o)) +
3, by = 0.5) #H2O values for grid

for(i in 1:length(co2_plot))
{
  for(j in 1:length(h2o_plot))
  {
    temp_pred = predict(svm_gsf_h2o_co2, newdata = data.frame(m_h2o =
h2o_plot[j], m_co2 = co2_plot[i]), type = "class", decision.values = TRUE)

    if(temp_pred == TRUE) #Condition for grid color
    {
      points(co2_plot[i], h2o_plot[j], pch = '/', col = rgb(0, 0, 1, 0.4))
    } else
    {
      points(co2_plot[i], h2o_plot[j], pch = '\\', col = rgb(0, 1, 0, 0.4))
    }
  }
}

#Add boundary curves
co2_plot = seq(from = floor(min(m_co2)) - 1, to = ceiling(max(m_co2)) + 15,
by = 0.2) #CO2 values for grid
h2o_plot = seq(from = floor(min(m_h2o)) - 0.25, to = ceiling(max(m_h2o)) +
3, by = 0.15) #H2O values for grid

bndr_co2 = matrix(data = NA, nrow = 9*length(co2_plot), ncol = 1) #CO2
boundary points
bndr_h2o = matrix(data = NA, nrow = 9*length(h2o_plot), ncol = 1) #H2O
boundary points

k = 1
for(i in 1:length(co2_plot))
{
  for(j in 1:length(h2o_plot))
  {
    temp_pred = predict(svm_gsf_h2o_co2, newdata = data.frame(m_h2o =
h2o_plot[j], m_co2 = co2_plot[i]), type = "class", decision.values = TRUE)

    if(round(as.numeric(attr(temp_pred, "decision.values")), digits = 1) ==
0.0) #Condition for boundary points
    {
      bndr_co2[k,1] = co2_plot[i]
      bndr_h2o[k,1] = h2o_plot[j]
      k = k + 1
    }
  }
}

bndr_co2 = na.omit(bndr_co2)
bndr_h2o = na.omit(bndr_h2o)
bndr = data.frame(bndr_co2, bndr_h2o)

axis = lm(bndr_h2o ~ bndr_co2) #Axis of boundary parabola
ang1 = atan(axis$coefficients[2]) #Angle between horizontal axis and
parabola axis

```

```

trans_mat = matrix(data = NA, nrow = 2, ncol = 2) #Rotational
transformation matrix
trans_mat[1,1] = cos(pi/2 - angl)
trans_mat[1,2] = -sin(pi/2 - angl)
trans_mat[2,1] = sin(pi/2 - angl)
trans_mat[2,2] = cos(pi/2 - angl)

bndr_trans = t(trans_mat%*%t(bndr)) #Rotate parabola to be vertical

indx1 = which(bndr_trans[,1] >= bndr_trans[which.min(bndr_trans[,2]),1])
#Index for the left half of the parabola
indx2 = which(bndr_trans[,1] <= bndr_trans[which.min(bndr_trans[,2]),1])
#Index for the right half of the parabola

spl1 = smooth.spline(bndr_co2[indx1], bndr_h2o[indx1]) #Spline curve for
the first half of the parabola
spl2 = smooth.spline(bndr_co2[indx2], bndr_h2o[indx2]) #Spline curve for
the second half of the parabola
lines(spl1)
lines(spl2)

#Add legend
legend(-5.1, 10.25, legend = c("Model B Solves", "Model B Does Not Solve",
"95% Confidence Ellipse", "Boundaries"), fill = c("blue", "green", "red",
"black"))

#Determine support vector machine train and test accuracy and inaccuracy
#H2 and CO
pred_train_h2_co = predict(svm_gsf_h2_co, new_dat[train,], type = "class")
#Training set predictions
tabl_train_h2_co = table(pred_train_h2_co, solvs[train]) #Confusion matrix
ndr_train_h2_co = tabl_train_h2_co[2,1]/length(which(B_solvs[train] ==
FALSE))*100 #Non-detection rate
far_train_h2_co = tabl_train_h2_co[1,2]/length(which(B_solvs[train] ==
TRUE))*100 #False-alarm rate
acc_train_h2_co = (tabl_train_h2_co[1,1] +
tabl_train_h2_co[2,2])/length(B_solvs[train])*100 #Accuracy

pred_test_h2_co = predict(svm_gsf_h2_co, new_dat[-train,], type = "class")
#Test set predictions
tabl_test_h2_co = table(pred_test_h2_co, solvs[-train]) #Confusion matrix
ndr_test_h2_co = tabl_test_h2_co[2,1]/length(which(B_solvs[-train] ==
FALSE))*100 #Non-detection rate
far_test_h2_co = tabl_test_h2_co[1,2]/length(which(B_solvs[-train] ==
TRUE))*100 #False-alarm rate
acc_test_h2_co = (tabl_test_h2_co[1,1] +
tabl_test_h2_co[2,2])/length(B_solvs[-train])*100 #Accuracy

#CH4 and CO
pred_train_ch4_co = predict(svm_gsf_ch4_co, new_dat[train,], type =
"class") #Training set predictions
tabl_train_ch4_co = table(pred_train_ch4_co, solvs[train]) #Confusion
matrix
ndr_train_ch4_co = tabl_train_ch4_co[2,1]/length(which(B_solvs[train] ==
FALSE))*100 #Non-detection rate
far_train_ch4_co = tabl_train_ch4_co[1,2]/length(which(B_solvs[train] ==
TRUE))*100 #False-alarm rate
acc_train_ch4_co = (tabl_train_ch4_co[1,1] +
tabl_train_ch4_co[2,2])/length(B_solvs[train])*100 #Accuracy

```

```

pred_test_ch4_co = predict(svm_gsf_ch4_co, new_dat[-train,], type =
"class") #Test set predictions
tabl_test_ch4_co = table(pred_test_ch4_co, solvs[-train]) #Confusion matrix
ndr_test_ch4_co = tabl_test_ch4_co[2,1]/length(which(B_solvs[-train] ==
FALSE))*100 #Non-detection rate
far_test_ch4_co = tabl_test_ch4_co[1,2]/length(which(B_solvs[-train] ==
TRUE))*100 #False-alarm rate
acc_test_ch4_co = (tabl_test_ch4_co[1,1] +
tabl_test_ch4_co[2,2])/length(B_solvs[-train])*100 #Accuracy

#CH4 and CO2
pred_train_ch4_co2 = predict(svm_gsf_ch4_co2, new_dat[train,], type =
"class") #Training set predictions
tabl_train_ch4_co2 = table(pred_train_ch4_co2, solvs[train]) #Confusion
matrix
ndr_train_ch4_co2 = tabl_train_ch4_co2[2,1]/length(which(B_solvs[train] ==
FALSE))*100 #Non-detection rate
far_train_ch4_co2 = tabl_train_ch4_co2[1,2]/length(which(B_solvs[train] ==
TRUE))*100 #False-alarm rate
acc_train_ch4_co2 = (tabl_train_ch4_co2[1,1] +
tabl_train_ch4_co2[2,2])/length(B_solvs[train])*100 #Accuracy

pred_test_ch4_co2 = predict(svm_gsf_ch4_co2, new_dat[-train,], type =
"class") #Test set predictions
tabl_test_ch4_co2 = table(pred_test_ch4_co2, solvs[-train]) #Confusion
matrix
ndr_test_ch4_co2 = tabl_test_ch4_co2[2,1]/length(which(B_solvs[-train] ==
FALSE))*100 #Non-detection rate
far_test_ch4_co2 = tabl_test_ch4_co2[1,2]/length(which(B_solvs[-train] ==
TRUE))*100 #False-alarm rate
acc_test_ch4_co2 = (tabl_test_ch4_co2[1,1] +
tabl_test_ch4_co2[2,2])/length(B_solvs[-train])*100 #Accuracy

#H2O and CO2
pred_train_h2o_co2 = predict(svm_gsf_h2o_co2, new_dat[train,], type =
"class") #Training set predictions
tabl_train_h2o_co2 = table(pred_train_h2o_co2, solvs[train]) #Confusion
matrix
ndr_train_h2o_co2 = tabl_train_h2o_co2[2,1]/length(which(B_solvs[train] ==
FALSE))*100 #Non-detection rate
far_train_h2o_co2 = tabl_train_h2o_co2[1,2]/length(which(B_solvs[train] ==
TRUE))*100 #False-alarm rate
acc_train_h2o_co2 = (tabl_train_h2o_co2[1,1] +
tabl_train_h2o_co2[2,2])/length(B_solvs[train])*100 #Accuracy

pred_test_h2o_co2 = predict(svm_gsf_h2o_co2, new_dat[-train,], type =
"class") #Test set predictions
tabl_test_h2o_co2 = table(pred_test_h2o_co2, solvs[-train]) #Confusion
matrix
ndr_test_h2o_co2 = tabl_test_h2o_co2[2,1]/length(which(B_solvs[-train] ==
FALSE))*100 #Non-detection rate
far_test_h2o_co2 = tabl_test_h2o_co2[1,2]/length(which(B_solvs[-train] ==
TRUE))*100 #False-alarm rate
acc_test_h2o_co2 = (tabl_test_h2o_co2[1,1] +
tabl_test_h2o_co2[2,2])/length(B_solvs[-train])*100 #Accuracy

#Classification tree and support vector machine for first two principal
component directions

#Center and scale all measurements

```

```

m_h2o_cnt_std = (m_dot_rw_gs_h2o_A - pca_gsf$center[1])/pca_gsf$scale[1]
m_h2_cnt_std = (m_dot_rw_gs_h2_A - pca_gsf$center[2])/pca_gsf$scale[2]
m_co_cnt_std = (m_dot_rw_gs_co_A - pca_gsf$center[3])/pca_gsf$scale[3]
m_co2_cnt_std = (m_dot_rw_gs_co2_A - pca_gsf$center[4])/pca_gsf$scale[4]
m_ch4_cnt_std = (m_dot_rw_gs_ch4_A - pca_gsf$center[5])/pca_gsf$scale[5]

#Project all measurements onto first two principal component directions
new_pca_dat2 = data.frame(m_h2o_cnt_std, m_h2_cnt_std, m_co_cnt_std,
m_co2_cnt_std, m_ch4_cnt_std)
pc1 = as.matrix(new_pca_dat2)%*%pca_gsf$rotation[,1]
pc2 = as.matrix(new_pca_dat2)%*%pca_gsf$rotation[,2]

#Collate data set for classification tree and support vector machine
new_dat2 = data.frame(B_solvs, pc1, pc2)

colnames(new_dat2)[1] = "solvs_2"
colnames(new_dat2)[2] = "pc_1"
colnames(new_dat2)[3] = "pc_2"

attach(new_dat2)

#Training set
set.seed(7)
train = sample(x = nm_ent, size = nm_ent - 550)

#Classification tree and support vector machine
tree_pca = tree(solvs_2 ~ pc_2 + pc_1, data = new_dat2, subset = train,
split = "gini")
svm_pca = svm(solvs_2 ~ pc_2 + pc_1, data = new_dat2, subset = train,
kernel = "polynomial", degree = 2)

#Prune tree to smallest size and lowest 10-fold cross-validation error rate
set.seed(3)
cv_pc2_pc1 = cv.tree(tree_pca, FUN = prune.misclass, K = 10)
min_pc2_pc1 = which(cv_pc2_pc1$dev == min(cv_pc2_pc1$dev))
len_min_pc2_pc1 = length(min_pc2_pc1)
prune_pc2_pc1 = prune.misclass(tree_pca, best =
cv_pc2_pc1$size[min_pc2_pc1[len_min_pc2_pc1]])

#Plot classification tree and support vector machine
par(mar=c(5,5,5,2) + .1)

#Classification tree
plot(pc1, pc2, main = "Mass Flow Rate Principal Components", xlim = c(-10,
30), ylim = c(-15, 15), xlab = "First Principal Component Score", ylab =
"Second Principal Component Score",
col = (as.numeric(B_solvs) + 2))

#Classification tree decision structure
var_pc2_pc1 = as.numeric(prune_pc2_pc1$frame$var) #Vector containing 1 for
leaves, 2 for PC2 splits, and 3 for PC1 splits

indx_leaf_pc2_pc1 = which(var_pc2_pc1 == 1) #Leaf index
indx_split_pc2_pc1 = which(var_pc2_pc1 != 1) #Split index

```

```

split_pc2_pc1 = matrix(data = 0, nrow = length(indx_split_pc2_pc1), ncol =
1) #Matrix for PC2 and PC1 split values

split_pc2 = matrix(data = 0, nrow = length(which(var_pc2_pc1 == 2)), ncol =
1) #Matrix for PC2 split values

split_pc1 = matrix(data = 0, nrow = length(which(var_pc2_pc1 == 3)), ncol =
1) #Matrix for PC1 split values

#Extract split values
j = 1
k = 1
l = 1
for(i in 1:length(var_pc2_pc1))
{
  if(var_pc2_pc1[i] != 1) #Condition for PC2 and PC1 splits
  {
    split_pc2_pc1[l] = as.numeric(sub(".", "",
prune_pc2_pc1$frame$splits[i,1]))
    l = l + 1
  }

  if(var_pc2_pc1[i] == 2) #Condition for PC2 splits
  {
    split_pc2[j] = as.numeric(sub(".", "",
prune_pc2_pc1$frame$splits[i,1]))
    j = j + 1
  }

  if(var_pc2_pc1[i] == 3) #Condition for PC1 splits
  {
    split_pc1[k] = as.numeric(sub(".", "",
prune_pc2_pc1$frame$splits[i,1]))
    k = k + 1
  }
}

#Tree path matrix
path_pc2_pc1 = matrix(data = 0, nrow = length(which(var_pc2_pc1 == 1)),
ncol = length(which(var_pc2_pc1 == 1)))

for(i in 1:length(indx_leaf_pc2_pc1))
{
  path_pc2_pc1[,i] = Path(i, indx_leaf_pc2_pc1, split_pc2_pc1,
path_pc2_pc1)
}

#Plot decision boundaries
plot_db(indx_split_pc2_pc1, var_pc2_pc1, pc_2, pc_1, split_pc2, split_pc1,
path_pc2_pc1)

#Plot 95% confidence ellipse
elips_dat_pca = cbind(pc1[p_set], pc2[p_set])

cov_gsf_pca = cov(elips_dat_pca) #Covariance of 1st and 2nd principal
component
mean_pc1 = mean(elips_dat_pca[,1]) #Average and center of 1st principal
component
mean_pc2 = mean(elips_dat_pca[,2]) #Average and center of 2nd principal
component
center_gsf_pca = c(mean_pc1, mean_pc2)

```

```

lines(ellipse(cov_gsf_pca, centre = center_gsf_pca, level = 0.95), col =
"red")

#Shade regions determined by classification tree
pc1_plot = seq(from = floor(min(pc_1)) - 7, to = ceiling(max(pc_1)) + 5, by
= 1.0) #PC1 values for grid
pc2_plot = seq(from = floor(min(pc_2)) - 5, to = ceiling(max(pc_2)) + 5, by
= 0.65) #PC2 values for grid

for(i in 1:length(pc1_plot))
{
  for(j in 1:length(pc2_plot))
  {
    temp_pred = predict(prune_pc2_pc1, newdata = data.frame(pc_2 =
pc2_plot[j], pc_1 = pc1_plot[i]), type = "class")

    if(temp_pred == TRUE) #Condition for grid color
    {
      points(pc1_plot[i], pc2_plot[j], pch = '/', col = rgb(0, 0, 1, 0.4))
    } else
    {
      points(pc1_plot[i], pc2_plot[j], pch = '\\', col = rgb(0, 1, 0, 0.4))
    }
  }
}

#Add legend
legend(-11.1, 15.9, legend = c("Model B Solves", "Model B Does Not Solve",
"95% Confidence Ellipse", "Boundaries"), fill = c("blue", "green", "red",
"black"))

#Support vector machine
plot(pc1, pc2, main = "Mass Flow Rate Principal Components", xlim = c(-10,
30), ylim = c(-15, 15), xlab = "First Principal Component Score", ylab =
"Second Principal Component Score",
      col = (as.numeric(B_solvs) + 2))

#Plot 95% confidence ellipse
elips_dat_pca = cbind(pc1[p_set], pc2[p_set])

cov_gsf_pca = cov(elips_dat_pca) #Covariance of 1st and 2nd principal
component
mean_pc1 = mean(elips_dat_pca[,1]) #Average and center of 1st principal
component
mean_pc2 = mean(elips_dat_pca[,2]) #Average and center of 2nd principal
component
center_gsf_pca = c(mean_pc1, mean_pc2)

lines(ellipse(cov_gsf_pca, centre = center_gsf_pca, level = 0.95), col =
"red")

#Shade regions determined by support vector machine
pc1_plot = seq(from = floor(min(pc_1)) - 7, to = ceiling(max(pc_1)) + 5, by
= 1.0) #PC1 values for grid
pc2_plot = seq(from = floor(min(pc_2)) - 5, to = ceiling(max(pc_2)) + 5, by
= 0.65) #PC2 values for grid

for(i in 1:length(pc1_plot))
{

```

```

for(j in 1:length(pc2_plot))
{
  temp_pred = predict(svm_pca, newdata = data.frame(pc_2 = pc2_plot[j],
pc_1 = pc1_plot[i]), type = "class", decision.values = TRUE)

  if(temp_pred == TRUE) #Condition for grid color
  {
    points(pc1_plot[i], pc2_plot[j], pch = '/', col = rgb(0, 0, 1, 0.4))
  } else
  {
    points(pc1_plot[i], pc2_plot[j], pch = '\\', col = rgb(0, 1, 0, 0.4))
  }
}
}

#Add boundary curves
pc1_plot = seq(from = floor(min(pc_1)) - 7, to = ceiling(max(pc_1)) + 5, by
= 0.5) #PC1 values for grid
pc2_plot = seq(from = floor(min(pc_2)) - 5, to = ceiling(max(pc_2)) + 5, by
= 0.3) #PC2 values for grid

bndr_pc1 = matrix(data = NA, nrow = 9*length(pc1_plot), ncol = 1) #PC1
boundary points
bndr_pc2 = matrix(data = NA, nrow = 9*length(pc2_plot), ncol = 1) #PC2
boundary points

k = 1
for(i in 1:length(pc1_plot))
{
  for(j in 1:length(pc2_plot))
  {
    temp_pred = predict(svm_pca, newdata = data.frame(pc_2 = pc2_plot[j],
pc_1 = pc1_plot[i]), type = "class", decision.values = TRUE)

    if(round(as.numeric(attr(temp_pred, "decision.values")), digits = 0) ==
0) #Condition for boundary points
    {
      bndr_pc1[k,1] = pc1_plot[i]
      bndr_pc2[k,1] = pc2_plot[j]
      k = k + 1
    }
  }
}

bndr_pc1 = na.omit(bndr_pc1)
bndr_pc2 = na.omit(bndr_pc2)
bndr = data.frame(bndr_pc1, bndr_pc2)

bndr_pca = prcomp(bndr, scale = TRUE) #PCA to make two boundary lines
separable
bndr_clst = kmeans(bndr_pca$x[,2], 2, nstart = 50) #K-means clusters for
two boundary lines
clst_indx = which(bndr_clst$cluster == 1) #Cluster index

spl1 = smooth.spline(bndr_pc1[clst_indx], bndr_pc2[clst_indx]) #Spline
curve for the first line
spl2 = smooth.spline(bndr_pc1[-clst_indx], bndr_pc2[-clst_indx]) #Spline
curve for the second line
lines(spl1)
lines(spl2)

```

```

#Add legend
legend(-11.1, 15.9, legend = c("Model B Solves", "Model B Does Not Solve",
"95% Confidence Ellipse", "Boundaries"), fill = c("blue", "green", "red",
"black"))

#Determine principal component model train and test accuracy and inaccuracy
#Classification tree
pred_train_tree = predict(prune_pc2_pcl, new_dat2[train,], type = "class")
#Training set predictions
tabl_train_tree = table(pred_train_tree, solvs_2[train]) #Confusion matrix
ndr_train_tree = tabl_train_tree[2,1]/length(which(B_solvs[train] ==
FALSE))*100 #Non-detection rate
far_train_tree = tabl_train_tree[1,2]/length(which(B_solvs[train] ==
TRUE))*100 #False-alarm rate
acc_train_tree = (tabl_train_tree[1,1] +
tabl_train_tree[2,2])/length(B_solvs[train])*100 #Accuracy

pred_test_tree = predict(prune_pc2_pcl, new_dat2[-train,], type = "class")
#Test set predictions
tabl_test_tree = table(pred_test_tree, solvs_2[-train]) #Confusion matrix
ndr_test_tree = tabl_test_tree[2,1]/length(which(B_solvs[-train] ==
FALSE))*100 #Non-detection rate
far_test_tree = tabl_test_tree[1,2]/length(which(B_solvs[-train] ==
TRUE))*100 #False-alarm rate
acc_test_tree = (tabl_test_tree[1,1] +
tabl_test_tree[2,2])/length(B_solvs[-train])*100 #Accuracy

#Support vector machine
pred_train_svm = predict(svm_pca, new_dat2[train,], type = "class")
#Training set predictions
tabl_train_svm = table(pred_train_svm, solvs_2[train]) #Confusion matrix
ndr_train_svm = tabl_train_svm[2,1]/length(which(B_solvs[train] ==
FALSE))*100 #Non-detection rate
far_train_svm = tabl_train_svm[1,2]/length(which(B_solvs[train] ==
TRUE))*100 #False-alarm rate
acc_train_svm = (tabl_train_svm[1,1] +
tabl_train_svm[2,2])/length(B_solvs[train])*100 #Accuracy

pred_test_svm = predict(svm_pca, new_dat2[-train,], type = "class") #Test
set predictions
tabl_test_svm = table(pred_test_svm, solvs_2[-train]) #Confusion matrix
ndr_test_svm = tabl_test_svm[2,1]/length(which(B_solvs[-train] ==
FALSE))*100 #Non-detection rate
far_test_svm = tabl_test_svm[1,2]/length(which(B_solvs[-train] ==
TRUE))*100 #False-alarm rate
acc_test_svm = (tabl_test_svm[1,1] + tabl_test_svm[2,2])/length(B_solvs[-
train])*100 #Accuracy

#Clusters for gasifier input conditions

set.seed(2)
km_stm = kmeans(m_stm, 2, nstart = 50) #Steam inlet mass flow rate
clusters
km_coal = kmeans(m_coal, 2, nstart = 50) #Coal inlet mass flow rate
clusters

#Combination cluster index
com_clust = matrix(data = NA, nrow = nm_ent, ncol = 1)

```

```

#Cluster metrics

#Maximum, minimum, and average volume percentage for H2
min_h2_per = matrix(data = NA, nrow = 4, ncol = 1)
max_h2_per = matrix(data = NA, nrow = 4, ncol = 1)
mean_h2_per = matrix(data = NA, nrow = 4, ncol = 1)

#Maximum, minimum, and average volume percentage for CO
min_co_per = matrix(data = NA, nrow = 4, ncol = 1)
max_co_per = matrix(data = NA, nrow = 4, ncol = 1)
mean_co_per = matrix(data = NA, nrow = 4, ncol = 1)

#Maximum, minimum, and average volume percentage for CO2
min_co2_per = matrix(data = NA, nrow = 4, ncol = 1)
max_co2_per = matrix(data = NA, nrow = 4, ncol = 1)
mean_co2_per = matrix(data = NA, nrow = 4, ncol = 1)

#Maximum, minimum, and average volume percentage for CH4
min_ch4_per = matrix(data = NA, nrow = 4, ncol = 1)
max_ch4_per = matrix(data = NA, nrow = 4, ncol = 1)
mean_ch4_per = matrix(data = NA, nrow = 4, ncol = 1)

l = 1
for(i in 1:2)
{
  for(j in 1:2)
  {
    com_clust[which(km_stm$cluster == i & km_coal$cluster == j),1] = 1

    min_h2_per[l,1] = min(vol_per_h2_A[which(com_clust == 1)])
    max_h2_per[l,1] = max(vol_per_h2_A[which(com_clust == 1)])
    mean_h2_per[l,1] = mean(vol_per_h2_A[which(com_clust == 1)])

    min_co_per[l,1] = min(vol_per_co_A[which(com_clust == 1)])
    max_co_per[l,1] = max(vol_per_co_A[which(com_clust == 1)])
    mean_co_per[l,1] = mean(vol_per_co_A[which(com_clust == 1)])

    min_co2_per[l,1] = min(vol_per_co2_A[which(com_clust == 1)])
    max_co2_per[l,1] = max(vol_per_co2_A[which(com_clust == 1)])
    mean_co2_per[l,1] = mean(vol_per_co2_A[which(com_clust == 1)])

    min_ch4_per[l,1] = min(vol_per_ch4_A[which(com_clust == 1)])
    max_ch4_per[l,1] = max(vol_per_ch4_A[which(com_clust == 1)])
    mean_ch4_per[l,1] = mean(vol_per_ch4_A[which(com_clust == 1)])

    l = l + 1
  }
}

#Plot selected graph with combination clusters
par(mar=c(5,5,5,2) + .1)

plot(m_co2, m_h2o, main = "Steam to Carbon Dioxide", xlab =
  substitute(paste(italic(CO[2]), " Mass Flow Rate [kg/s]")), xlim = c(-4,
34), ylim = c(-14.5, 9.5),
  ylab = substitute(paste(italic(H[2]), italic(O), " Mass Flow Rate
[kg/s]")), col = (com_clust + 2))

```

```

#Plot 95% confidence ellipse
elips_h2o_co2 = cbind(m_co2[p_set], m_h2o[p_set])

cov_h2o_co2 = cov(elips_h2o_co2) #Covariance of CO2 and H2O
mean_co2 = mean(elips_h2o_co2[,1]) #Average and center of CO2
mean_h2o = mean(elips_h2o_co2[,2]) #Average and center of H2O
center_h2o_co2 = c(mean_co2, mean_h2o)

lines(ellipse(cov_h2o_co2, centre = center_h2o_co2, level = 0.95), col =
"red")

#Add boundary curves
co2_plot = seq(from = floor(min(m_co2)) - 1, to = ceiling(max(m_co2)) + 15,
by = 0.2) #CO2 values for grid
h2o_plot = seq(from = floor(min(m_h2o)) - 0.25, to = ceiling(max(m_h2o)) +
3, by = 0.15) #H2O values for grid

bndr_co2 = matrix(data = NA, nrow = 4*length(co2_plot), ncol = 1) #CO2
boundary points
bndr_h2o = matrix(data = NA, nrow = 4*length(h2o_plot), ncol = 1) #H2O
boundary points

k = 1
for(i in 1:length(co2_plot))
{
  for(j in 1:length(h2o_plot))
  {
    temp_pred = predict(svm_gsf_h2o_co2, newdata = data.frame(m_h2o =
h2o_plot[j], m_co2 = co2_plot[i]), type = "class", decision.values = TRUE)

    if(round(as.numeric(attr(temp_pred, "decision.values")), digits = 1) ==
0.0) #Condition for boundary points
    {
      bndr_co2[k,1] = co2_plot[i]
      bndr_h2o[k,1] = h2o_plot[j]
      k = k + 1
    }
  }
}

bndr_co2 = na.omit(bndr_co2)
bndr_h2o = na.omit(bndr_h2o)
bndr = data.frame(bndr_co2, bndr_h2o)

axis = lm(bndr_h2o ~ bndr_co2) #Axis of boundary parabola
angl = atan(axis$coefficients[2]) #Angle between horizontal axis and
parabola axis

trans_mat = matrix(data = NA, nrow = 2, ncol = 2) #Rotational
transformation matrix
trans_mat[1,1] = cos(pi/2 - angl)
trans_mat[1,2] = -sin(pi/2 - angl)
trans_mat[2,1] = sin(pi/2 - angl)
trans_mat[2,2] = cos(pi/2 - angl)

bndr_trans = t(trans_mat%*%t(bndr)) #Rotate parabola to be vertical

indx1 = which(bndr_trans[,1] >= bndr_trans[which.min(bndr_trans[,2]),1])
#Index for the left half of the parabola
indx2 = which(bndr_trans[,1] <= bndr_trans[which.min(bndr_trans[,2]),1])
#Index for the right half of the parabola

```

```

spl1 = smooth.spline(bndr_co2[indx1], bndr_h2o[indx1]) #Spline curve for
the first half of the parabola
spl2 = smooth.spline(bndr_co2[indx2], bndr_h2o[indx2]) #Spline curve for
the second half of the parabola
lines(spl1)
lines(spl2)

#Add legend
legend(-5.1, 10.25, legend = c("High Steam and High Coal", "High Steam and
Low Coal", "Low Steam and High Coal", "Low Steam and Low Coal", "95%
Confidence Ellipse", "Boundaries"),
      fill = c("3", "4", "5", "6", "red", "1"))

#Additional flow rate graphs with clusters

#H2 and CO
plot(m_co, m_h2, main = "Hydrogen to Carbon Monoxide", xlab =
substitute(paste(italic(CO), " Mass Flow Rate [kg/s]")), xlim = c(0, 12),
ylim = c(-3, 2),
      ylab = substitute(paste(italic(H[2]), " Mass Flow Rate [kg/s]")), col
= (com_clust + 2))

#Plot 95% confidence ellipse
elips_h2_co = cbind(m_co[p_set], m_h2[p_set])

cov_h2_co = cov(elips_h2_co) #Covariance of H2 and CO
mean_co = mean(elips_h2_co[,1]) #Average and center of CO
mean_h2 = mean(elips_h2_co[,2]) #Average and center of H2
center_h2_co = c(mean_co, mean_h2)

lines(ellipse(cov_h2_co, centre = center_h2_co, level = 0.95), col = "red")

#Add legend
legend(7.67, 2.15, legend = c("High Steam and High Coal", "High Steam and
Low Coal", "Low Steam and High Coal", "Low Steam and Low Coal", "95%
Confidence Ellipse"),
      fill = c("3", "4", "5", "6", "red"))

#CH4 and CO
plot(m_co, m_ch4, main = "Methane to Carbon Monoxide", xlab =
substitute(paste(italic(CO), " Mass Flow Rate [kg/s]")), xlim = c(0, 12),
ylim = c(-1.5, 12.5),
      ylab = substitute(paste(italic(CH[4]), " Mass Flow Rate [kg/s]")), col
= (com_clust + 2))

#Plot 95% confidence ellipse
elips_ch4_co = cbind(m_co[p_set], m_ch4[p_set])

cov_ch4_co = cov(elips_ch4_co) #Covariance of CH4 and CO
mean_co = mean(elips_ch4_co[,1]) #Average and center of CO
mean_ch4 = mean(elips_ch4_co[,2]) #Average and center of CH4
center_ch4_co = c(mean_co, mean_ch4)

lines(ellipse(cov_ch4_co, centre = center_ch4_co, level = 0.95), col =
"red")

#Add legend

```

```

legend(-0.35, 12.95, legend = c("High Steam and High Coal", "High Steam and
Low Coal", "Low Steam and High Coal", "Low Steam and Low Coal", "95%
Confidence Ellipse"),
      fill = c("3", "4", "5", "6", "red"))

#CH4 and CO2
plot(m_co2, m_ch4, main = "Methane to Carbon Dioxide", xlab =
substitute(paste(italic(CO[2]), " Mass Flow Rate [kg/s]")), xlim = c(-4,
24), ylim = c(-1.5, 12.5),
      ylab = substitute(paste(italic(CH[4]), " Mass Flow Rate [kg/s]")), col
= (com_clust + 2))

#Plot 95% confidence ellipse
elips_ch4_co2 = cbind(m_co2[p_set], m_ch4[p_set])

cov_ch4_co2 = cov(elips_ch4_co2) #Covariance of CH4 and CO2
mean_co2 = mean(elips_ch4_co2[,1]) #Average and center of CO2
mean_ch4 = mean(elips_ch4_co2[,2]) #Average and center of CH4
center_ch4_co2 = c(mean_co2, mean_ch4)

lines(ellipse(cov_ch4_co2, centre = center_ch4_co2, level = 0.95), col =
"red")

#Add legend
legend(13.95, 12.95, legend = c("High Steam and High Coal", "High Steam and
Low Coal", "Low Steam and High Coal", "Low Steam and Low Coal", "95%
Confidence Ellipse"),
      fill = c("3", "4", "5", "6", "red"))

#Accuracy for 95% confidence ellipse predictions

#PCA confidence ellipse
mean_pc1 = mean(pc_1[p_set]) #Average of 1st principal component
mean_pc2 = mean(pc_2[p_set]) #Average of 2nd principal component

pc_cov_mat = cov(cbind(pc_1[p_set], pc_2[p_set])) #Covariance of 1st and
2nd principal components
pc_eig_val = eigen(pc_cov_mat)$values #Covariance eigenvalues
pc_eig_vec = eigen(pc_cov_mat)$vectors #Covariance eigenvectors
pc_phi = atan(pc_eig_vec[2,1]/pc_eig_vec[1,1]) #Angle to major axis of
ellipse

pc_cfd = sqrt(qchisq(0.95, 2)) #Confidence level
pc_elp_indx = matrix(data = NA, nrow = nm_ent, ncol = 1) #Ellipse index

for(i in 1:nm_ent)
{
  pc_rt = (pc_2[i] - mean_pc2)/(pc_1[i] - mean_pc1) #Ratio of 2nd to 1st
principal component
  pc_psi = atan(pc_rt) #Angle to observation

  pc_theta = pc_psi - pc_phi

  x_crt = pc_cfd*sqrt(pc_eig_val[1])*cos(pc_theta) #Distance to ellipse
boundary at angle in x-direction
  y_crt = pc_cfd*sqrt(pc_eig_val[2])*sin(pc_theta) #Distance to ellipse
boundary at angle in y-direction

```

```

    dist = sqrt((pc_1[i] - mean_pc1)^2 + (pc_2[i] - mean_pc2)^2) #Distance
from ellipse center to observation
    dist_crt = sqrt(x_crt^2 + y_crt^2) #Distance to ellipse boundary at
angle

    if(dist < dist_crt)
    {
        pc_elp_indx[i] = 1 #Index value for observation inside ellipse
    } else
    {
        pc_elp_indx[i] = 0 #Index value for observation outside ellipse
    }
}

pc_elp_class = matrix(data = NA, nrow = 2, ncol = 2)
pc_elp_class[1,1] = length(which(pc_elp_indx == 0 & B_solvs == FALSE))
#Number of observations outside the ellipse where model B does not solve
pc_elp_class[2,1] = length(which(pc_elp_indx == 1 & B_solvs == FALSE))
#Number of observations inside the ellipse where model B does not solve
pc_elp_class[1,2] = length(which(pc_elp_indx == 0 & B_solvs == TRUE))
#Number of observations outside the ellipse where model B solves
pc_elp_class[2,2] = length(which(pc_elp_indx == 1 & B_solvs == TRUE))
#Number of observations inside the ellipse where model B solves

pc_elp_nldr = pc_elp_class[2,1]/length(which(B_solvs == FALSE))*100 #Non-
detection rate
pc_elp_far = pc_elp_class[1,2]/length(which(B_solvs == TRUE))*100 #False-
alarm rate
pc_elp_acc = (pc_elp_class[1,1] + pc_elp_class[2,2])/length(B_solvs)*100
#Accuracy

#H2O and CO2 confidence ellipse
mean_co2 = mean(m_co2[p_set]) #Average of CO2
mean_h2o = mean(m_h2o[p_set]) #Average of H2O

co2_h2o_cov_mat = cov(cbind(m_co2[p_set], m_h2o[p_set])) #Covariance of
H2O and CO2
co2_h2o_eig_val = eigen(co2_h2o_cov_mat)$values #Covariance eigenvalues
co2_h2o_eig_vec = eigen(co2_h2o_cov_mat)$vectors #Covariance eigenvectors
co2_h2o_phi = atan(co2_h2o_eig_vec[2,1]/co2_h2o_eig_vec[1,1]) #Angle to
major axis of ellipse

co2_h2o_cfd = sqrt(qchisq(0.95, 2)) #Confidence level
co2_h2o_elp_indx = matrix(data = NA, nrow = nm_ent, ncol = 1) #Ellipse
index

for(i in 1:nm_ent)
{
    co2_h2o_rt = (m_h2o[i] - mean_h2o)/(m_co2[i] - mean_co2) #Ratio of H2O
to CO2
    co2_h2o_psi = atan(co2_h2o_rt) #Angle to observation

    co2_h2o_theta = co2_h2o_psi - co2_h2o_phi

    x_crt = co2_h2o_cfd*sqrt(co2_h2o_eig_val[1])*cos(co2_h2o_theta) #Distance
to ellipse boundary at angle in x-direction
    y_crt = co2_h2o_cfd*sqrt(co2_h2o_eig_val[2])*sin(co2_h2o_theta) #Distance
to ellipse boundary at angle in y-direction

```

```

    dist = sqrt((m_co2[i] - mean_co2)^2 + (m_h2o[i] - mean_h2o)^2) #Distance
from ellipse center to observation
    dist_crt = sqrt(x_crt^2 + y_crt^2) #Distance to ellipse boundary at
angle

    if(dist < dist_crt)
    {
        co2_h2o_elp_indx[i] = 1 #Index value for observation inside ellipse
    } else
    {
        co2_h2o_elp_indx[i] = 0 #Index value for observation outside ellipse
    }
}

co2_h2o_elp_class = matrix(data = NA, nrow = 2, ncol = 2)
co2_h2o_elp_class[1,1] = length(which(co2_h2o_elp_indx == 0 & B_solvs ==
FALSE)) #Number of observations outside the ellipse where model B does not
solve
co2_h2o_elp_class[2,1] = length(which(co2_h2o_elp_indx == 1 & B_solvs ==
FALSE)) #Number of observations inside the ellipse where model B does not
solve
co2_h2o_elp_class[1,2] = length(which(co2_h2o_elp_indx == 0 & B_solvs ==
TRUE)) #Number of observations outside the ellipse where model B solves
co2_h2o_elp_class[2,2] = length(which(co2_h2o_elp_indx == 1 & B_solvs ==
TRUE)) #Number of observations inside the ellipse where model B solves

co2_h2o_elp_ndr = co2_h2o_elp_class[2,1]/length(which(B_solvs ==
FALSE))*100 #Non-detection rate
co2_h2o_elp_far = co2_h2o_elp_class[1,2]/length(which(B_solvs == TRUE))*100
#False-alarm rate
co2_h2o_elp_acc = (co2_h2o_elp_class[1,1] +
co2_h2o_elp_class[2,2])/length(B_solvs)*100 #Accuracy

```

## APPENDIX H – JOURNAL ARTICLE

This appendix contains a copy of an article manuscript that has been submitted to the journal *Industrial and Engineering Chemistry Research*.

# Statistical Classification with Thermal-Fluid Modeling for Fault Prediction in Fixed-Bed Dry Bottom Coal Gasifiers

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## ABSTRACT

Gasifiers are utilized to convert carbonaceous material to syngas. In literature, limited material is available regarding modeling, which incorporates thermodynamic principles and statistical techniques, for the prediction of abnormal gasifier operations. In this paper, an integrated thermal-fluid and statistical approach is presented for predicting abnormal conditions in fixed-bed dry bottom coal gasifiers. The thermal-fluid model, based on the first law of thermodynamics, mass conservation, and chemical equilibrium, predicts flow rates for five of the gases comprising syngas and indicates potential abnormal conditions. When compared with reference data, the developed model has an average prediction accuracy of 83.3%. To differentiate between normal and abnormal conditions, classification tree (CT) and support vector machine (SVM) models are trained. From those, the SVM model for steam ( $H_2O$ ) and carbon dioxide ( $CO_2$ ) flow rates has superior generalizability, for predicting abnormal conditions, and a 4.9% higher accuracy than a conventional approach.

## 1. INTRODUCTION

The fixed-bed dry bottom coal gasifier is a technology that forms a significant part of various chemical processes.<sup>1-3</sup> It operates by gasifying the fixed carbon in coal, with steam ( $H_2O$ ) and oxygen ( $O_2$ ), to produce syngas.<sup>1-4</sup> Being mainly comprised of hydrogen ( $H_2$ ) and carbon monoxide ( $CO$ ), syngas can in turn be converted to various products that include diesel, petrol, ammonia, and synthetic natural gas.<sup>2-3,5-6</sup>

Coal, which is loaded at the top, travels down the fixed-bed dry bottom gasifier's length to form a bed on the grate at the bottom.<sup>1-2</sup> The  $H_2O$  and  $O_2$  are supplied from below this grate.<sup>1-</sup>

<sup>2</sup> Four reaction regions, which overlap to an extent, exist along the length of these gasifiers,

namely, from top to bottom, drying, devolatilization, gasification, and combustion.<sup>1</sup> Coal, which is heated by the exiting syngas, is converted to char, moisture, and various hydrocarbons in the top two regions.<sup>1,3,6</sup> Within the bottom region, char reacts with  $O_2$  to produce ash and carbon dioxide ( $CO_2$ ).<sup>1</sup> Between the combustion and devolatilization regions, reversible gasification reactions occur that result in the production of  $H_2$ ,  $CO$ , and methane ( $CH_4$ ).<sup>1,3</sup> The ash, which must be removed as a solid, is extracted as the gasifier's grate rotates.<sup>1,6</sup> In addition, heat produced in the reactor is partly absorbed by water, within a jacket that forms part of the gasifier's walls, and is partly lost to the environment.<sup>1,3</sup> A schematic diagram of this gasifier is provided in Figure 1.<sup>7</sup>

Process faults collectively refer, in this study, to deviations from normal gasifier operating conditions. Changes in gasifier temperature and pressure measurements, which are of greater magnitude than the fluctuations during normal operations, are indicative of faults.<sup>8</sup> An example of a fault that can occur in fixed-bed dry bottom coal gasifiers is channel burning.<sup>3,6</sup> It involves localized ash formation in the gasifier's coal bed and causes excessive variability in the composition of syngas.<sup>6,9</sup> In addition, faults can occur due to the use of coal with highly variable ash content, excessive fines, or a very wide particle size distribution.<sup>1,4,6</sup>

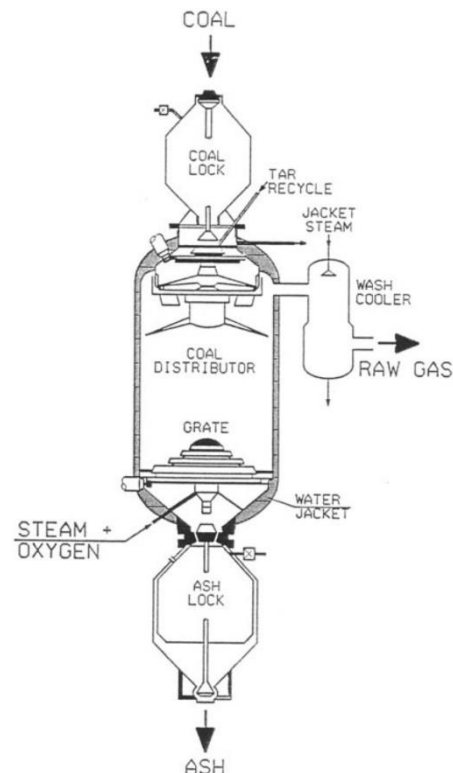


**Figure 1.** Fixed-Bed Dry Bottom Coal Gasifier<sup>7</sup>

Statistical and thermal-fluid approaches to gasifier modeling, for process prediction and monitoring, have been independently developed.<sup>1</sup> Examples of statistical approaches include Guo et al.<sup>8</sup> and Coetzer et al.,<sup>10</sup> while examples of thermal-fluid approaches include He et al.,<sup>11</sup> Luckos and Bunt,<sup>12</sup> and Ismail et al..<sup>13</sup>

Thermal-fluid approaches for gasifier modeling include calculations based on the first law of thermodynamics, the conservation of mass, and thermodynamic equilibrium.<sup>1,14-15</sup> Statistical approaches for gasifier modeling include those for the selection of reference sets and those for the detection of potential faults.<sup>8,10</sup> Reference sets define the nearest to optimal operation of a system and can be selected using a technique based on principal component analysis (PCA).<sup>10</sup> The detection of potential faults can involve analyzing trends present in gasifier measurements.<sup>8</sup> However, a statistical confidence region, for a selected reference set, can also be used to locate potential abnormal conditions.<sup>10</sup> Once the locations of abnormal conditions, which represent potential faults, are available in a given data set, statistical classification models can be trained for fault prediction.<sup>16</sup> Relevant classification models include the support vector machine (SVM) and the classification tree (CT).<sup>16</sup>

However, very little is available in literature on an integrated statistical and thermal-fluid approach to fault prediction in fixed-bed dry bottom coal gasifiers. Therefore, in this paper, the development and application of such an approach for the prediction of abnormal gasifier operating conditions is presented.

This paper is outlined as follows. In section 2, the methodology applied to predict abnormal operating conditions is described. The results from the application of this methodology are discussed in Section 3, after which conclusions are presented in Section 4.

## 2. METHODOLOGY

The integrated statistical and thermal-fluid approach to fault prediction, for fixed-bed dry bottom coal gasifiers, is applied as follows. Generic gasifier data,<sup>4</sup> including process measurements of temperature, pressure, and flow rates, are input to the thermal-fluid model. Per solvable observation, this model predicts mass flow rates for  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ , and  $H_2O$ , which are assumed to be the major components of syngas.<sup>3</sup> As such, five equations are required in the model, where these are three mole balances, one energy balance, and one chemical equilibrium condition.<sup>1,15</sup>

Observations for which a solution cannot be generated from the thermal-fluid model, due to the unrealistic prediction of at least one negative mass flow rate, indicate abnormal operations. Therefore, potential gasifier faults, where abnormal operating conditions exist, are obtained from the thermal-fluid model's outputs.

In this paper, PCA is used to create a two-dimensional representation for the five-dimensional space of flow rates.<sup>17</sup> In addition, flow rates that are the least correlated are identified from the PCA results.<sup>17</sup> Given that the predicted flow rates are correlated, considering pairs of these is likely to reduce the potential for overfitting.<sup>17</sup>

In order to differentiate between normal and abnormal conditions, statistical classification models are applied to the first two principal components and the approximately uncorrelated pairs of flow rates. The resulting regions where conditions are predicted to be normal, by the CT and SVM models, are interpreted. From these results, the most suitable statistical model is selected from which generalizable results may be drawn.

## 2.1 Thermal-Fluid Model

### 2.1.1 Conservation of Mass

The application of the conservation of mass to coal gasification results in the formulation of three mole balances.<sup>1,15</sup> Respectively, these are carbon, hydrogen, and oxygen balances.<sup>1</sup> Specifically, these define that the quantity of each element's atoms entering and exiting the gasifier, which is assumed to operate at steady-state conditions, is equal.<sup>15</sup> The operations of fixed-bed dry bottom coal gasifiers are assumed to be steady-state, since process measurements are recorded and averaged over a period of time.<sup>4,15</sup>

Equations 1 through 3, respectively, represent the mole balances for carbon, hydrogen, and oxygen.

$$\sum_{j \in C \cap N} n_{1,j} * \dot{n}_{j,1} = \sum_{j \in C \cap U} n_{1,j} * \dot{n}_{j,2} \quad (1)$$

$$\sum_{j \in H \cap N} n_{2,j} * \dot{n}_{j,1} = \sum_{j \in H \cap U} n_{2,j} * \dot{n}_{j,2} \quad (2)$$

$$\sum_{j \in O \cap N} n_{3,j} * \dot{n}_{j,1} = \sum_{j \in O \cap U} n_{3,j} * \dot{n}_{j,2} \quad (3)$$

where the variable  $n_{i,j}$  represents an element's number of kilomoles, per kilomole of a component. The mole flow rates of compounds are represented by  $\dot{n}_{j,k}$ , in kmol/s. The product of  $n_{i,j}$  and  $\dot{n}_{j,k}$  is the mole flow rate of element  $i$  available in compound  $j$  of stream  $k$ .

The term compound is applied to refer to both the groups of matter, which coals are comprised of according to proximate analysis, and molecules. Indices for the subscripts are provided in Table 1, where  $i \in I \subset \mathbb{R}^3$ ,  $j \in J \subset \mathbb{R}^7$ , and  $k \in K \subset \mathbb{R}^2$  define elements, compounds, and streams, respectively. The term stream is applied to differentiate between gasifier inlet and outlet flows.

Subsets  $C$ ,  $H$ , and  $O$  define the compounds that are comprised of carbon, hydrogen, and oxygen, respectively. These are  $\{1, 5, 6, 7\}$ ,  $\{2, 4, 7\}$ , and  $\{2, 3, 5, 6\}$ . Inlet and outlet flows are defined by subsets  $N$  and  $U$ , where these are  $\{1, 2, 3\}$  and  $\{1, 2, 4, 5, 6, 7\}$ , respectively.

**Table 1.** Thermal-Fluid Model Indices

| Index | $I$ : Element | $J$ : Compound | $K$ : Stream   |
|-------|---------------|----------------|----------------|
| 1     | $C$           | Fixed carbon   | Inlet ( $N$ )  |
| 2     | $H$           | $H_2O$         | Outlet ( $U$ ) |
| 3     | $O$           | $O_2$          |                |
| 4     |               | $H_2$          |                |
| 5     |               | $CO$           |                |
| 6     |               | $CO_2$         |                |
| 7     |               | $CH_4$         |                |

Proximate and ultimate analyses data, regarding the composition of coal and ash, are applied in this thermal-fluid model.<sup>4</sup> This in order to account for the percentage of fixed carbon in inlet coal and outlet ash. In addition, the moisture released during coal drying is also accounted for. Furthermore, flow rates developed during devolatilization,  $H_2$ ,  $CO$ ,  $CO_2$ ,  $CH_4$ , and tar, are accounted for in the model using Fischer assay and gas chromatography data.<sup>18</sup> As such, these are not included in the mole balances. In this study, the hydrocarbons released during devolatilization are collectively referred to as tar.<sup>1,3,6</sup>

### 2.1.2 First Law of Thermodynamics

The application of the first law of thermodynamics to coal gasification results in the formulation of an energy balance.<sup>1,15</sup> This defines that the respective energy flows entering and exiting the gasifier are equal.<sup>1,15</sup> The former is provided in the thermal and chemical energy

of the inlet flows, which are defined using enthalpy.<sup>1,15</sup> However, the latter includes the thermal and chemical energy in the outlet flows, as well as the heat transferred from the gasifier.<sup>1</sup>

In this study, the term power includes the relative chemical and thermal energy, per unit of time, that is available in a given flow. Chemical energy, defined by the respective compound's enthalpy of formation, is specified at a reference condition of 25°C and 100 kPa.<sup>15</sup> Thermal energy, defined by a difference in enthalpy, is calculated between this reference and the respective conditions.<sup>15</sup>

Equation 4 represents the energy balance.

$$\begin{aligned} & \dot{H}_{coal} + \dot{H}_{2,1} + \dot{H}_{3,1} \\ &= \Delta\dot{Q}_{ash} + \Delta\dot{Q}_{wj} + \dot{Q}_{loss} + \dot{H}_{2,2} \quad (4) \\ &+ \dot{H}_{4,2} + \dot{H}_{5,2} + \dot{H}_{6,2} + \dot{H}_{7,2} + \dot{H}_{tar} \end{aligned}$$

where  $\dot{H}_{coal}$  is the power inlet to the gasifier via the flow rate of coal. Power afforded by the flows of  $H_2O$  and  $O_2$  inlet to the gasifier are represented by  $\dot{H}_{2,1}$  and  $\dot{H}_{3,1}$ , respectively. The heat absorption rate of the ash in coal is  $\Delta\dot{Q}_{ash}$ , and the rate of heat transferred into the gasifier's water jacket is  $\Delta\dot{Q}_{wj}$ . The rate of heat loss to the environment is  $\dot{Q}_{loss}$ . Power available in the total outlet flows of  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  are, respectively,  $\dot{H}_{2,2}$ ,  $\dot{H}_{4,2}$ ,  $\dot{H}_{5,2}$ ,  $\dot{H}_{6,2}$ , and  $\dot{H}_{7,2}$ . The power conveyed by the flow of tar released during devolatilization is represented by  $\dot{H}_{tar}$ . In this study, power, heat absorption rates, and heat loss are in kW.

Empirical correlations, for enthalpy of formation and constant pressure specific heat, are applied to calculate the power available in coal.<sup>3</sup> The rate of heat absorbed by ash<sup>3</sup> and the power conveyed by tar<sup>19</sup> are also calculated with empirical correlations. Although the rate of heat loss is calculated using empirical correlations,<sup>3</sup> as described in Section 3, the heat loss fraction is adjusted to account for uncertainties in the thermal-fluid model.

### 2.1.3 Thermodynamic Equilibrium

In the thermal-fluid model, a thermodynamic equilibrium condition is applied to the reversible water gas shift reaction, which is  $CO + H_2O \rightleftharpoons CO_2 + H_2$ .<sup>2-3</sup> Equations 5 and 6 define the thermodynamic equilibrium condition for this reaction.<sup>15</sup>

$$\ln(K_{wgs}) = -\frac{g_{6,2} + g_{4,2} - g_{5,2} - g_{2,2}}{R_{unv} * T_{sgs}} \quad (5)$$

$$K_{wgs} = \frac{y_{6,2} * y_{4,2}}{y_{5,2} * y_{2,2}} \quad (6)$$

where  $K_{wgs}$  is the dimensionless equilibrium constant for the reaction,  $T_{sgs}$  is the temperature of syngas in K, and  $R_{unv}$  [8.31451 kJ/kmol-K] is the universal gas constant. The respective Gibbs functions, in kJ/kmol, for  $H_2O$ ,  $H_2$ ,  $CO$ , and  $CO_2$  at  $T_{sgs}$  and 100 kPa are  $g_{2,2}$ ,  $g_{4,2}$ ,  $g_{5,2}$ , and  $g_{6,2}$ , respectively. Mole fractions for the total  $H_2O$ ,  $H_2$ ,  $CO$ , and  $CO_2$  in syngas are  $y_{2,2}$ ,  $y_{4,2}$ ,  $y_{5,2}$ ,  $y_{6,2}$ , respectively.

## 2.2 Statistical Modeling

### 2.2.1 Principal Component Analysis

Principal components are uncorrelated directions that are selected during PCA, according to an optimization problem, in order to maximize the variability along these directions.<sup>17</sup> From these components, models can be developed with fewer predictors, instead of using all the variables in a data set.<sup>17</sup> Principal component directions are defined by loading vectors, where these contain the weights assigned to the respective variables.<sup>17</sup> Scores are the values that result when observations are projected onto the principal component directions.<sup>17</sup>

Equation 7 is used to calculate principal components, where the required loadings are determined according to Equation 8.<sup>17</sup>

$$\begin{aligned} \underline{pc}_x &= \phi_{x,1} * \underline{\dot{m}}_{2,2} + \phi_{x,2} * \underline{\dot{m}}_{4,2} \\ &+ \phi_{x,3} * \underline{\dot{m}}_{5,2} + \phi_{x,4} * \underline{\dot{m}}_{6,2} + \phi_{x,5} * \underline{\dot{m}}_{7,2} \end{aligned} \quad (7)$$

$$\begin{aligned} &\text{maximize } \left\{ \frac{1}{\omega} \sum_{w=1}^{\omega} (pc_{x,w})^2 \right\} \\ &\text{subject to } \sum_{r=1}^5 (\phi_{x,r})^2 = 1 \end{aligned} \quad (8)$$

where  $\underline{pc}_x$  is the  $\omega$ -dimensional vector for principal component  $x$ . The total number of observations is  $\omega$ , and  $x \in \{1, 2, 3, 4, 5\}$ . Total outlet mass flow rates, in kg/s, for the  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  comprising syngas, predicted by the thermal-fluid model per observation, are specified by the  $\underline{\dot{m}}_{2,2}$ ,  $\underline{\dot{m}}_{4,2}$ ,  $\underline{\dot{m}}_{5,2}$ ,  $\underline{\dot{m}}_{6,2}$ , and  $\underline{\dot{m}}_{7,2}$  vectors, respectively. Loading  $r$  for principal component  $x$  is represented by  $\phi_{x,r}$ , with loadings 1 through 5 corresponding to  $H_2O$ ,  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$ , respectively. Score  $w$  of principal component  $x$ , corresponding to observation  $w$ , is  $pc_{x,w}$ .

### 2.2.2 Classification Trees

A CT divides a variable space of given dimensions into smaller regions, by applying rules per variable, in order to differentiate between classes.<sup>17</sup> In a respective terminal region, observations are predicted to belong to the dominant class observed during model training.<sup>17</sup>

Equation 9 defines the division of a given two-dimensional space of outlet mass flow rates into two regions.<sup>17</sup> The space is split at a value,  $sp_b$ , for one of the two flow rates, that is selected according to the optimization problem in Equation 10.<sup>17</sup>

$$\begin{aligned} Rg_{2b-1} &= \{\dot{m}_{j,2} \mid \dot{m}_{\zeta,2} < sp_b\} \\ \text{and } Rg_{2b} &= \{\dot{m}_{j,2} \mid \dot{m}_{\zeta,2} \geq sp_b\} \end{aligned} \quad (9)$$

$$\begin{aligned}
& \text{minimize } \{Gn\} \\
Gn &= \sum_{a=1}^A \sum_{cl=1}^2 [pr_{a,cl} * (1 - pr_{a,cl})] \quad (10)
\end{aligned}$$

where  $Gn$  is the Gini index of the CT model, and the two-dimensional space of flow rates, which the model is applied to, is represented by  $\dot{m}_{j,2}$ . Regions corresponding to split  $b$ ,  $sp_b$ , of flow rate  $\zeta$ ,  $\dot{m}_{\zeta,2}$ , are  $Rg_{2b-1}$  and  $Rg_{2b}$ , where  $b$  is an element of the set of natural numbers  $\{1, 2, 3, 4, \dots\}$ . The proportion of training observations that belong to class  $cl$ , which are present in terminal region  $a$ , is represented by  $pr_{a,cl}$ , and  $A$  is the number of terminal regions.

### 2.2.3 Support Vector Machines

A SVM is applied to determine non-linear decision boundaries between classes.<sup>17</sup> SVM models are developed according to the constraints of an optimization problem, which defines that the width of a margin around the decision boundary must be maximized.<sup>17</sup> When differentiating between two classes, an observation is classified based on its predicted sign, which represents the side of the boundary that the observation is located on.<sup>17</sup>

Support vectors refer to observations, from a respective class, that are located on or across that class's edge of the decision boundary margin.<sup>17</sup> In SVM models, the shape and location of the decision boundary are, respectively, determined by the specified kernel function and the support vectors.<sup>17</sup>

Equations 11 and 12 are applied during SVM model training.<sup>17</sup>

$$f_w * \left\{ \beta_0 + \sum_{\varpi=1}^{\omega} [\alpha_{\varpi} * K(\dot{m}_{j,2,\varpi}, \dot{m}_{j,2,w})] \right\} \quad (11)$$

$$\geq Mi * (1 - \varepsilon_w) \forall w \in \{1, \dots, \omega\}$$

$$\sum_{w=1}^{\omega} \varepsilon_w \leq Cs \quad (12)$$

where  $M_i$  represents the margin width that must be maximized. The parameters that can be adjusted, while resolving the optimization problem, are  $\beta_0$ ,  $\varepsilon_w$ , and  $\alpha_{\varpi}$ . The indices  $w$  and  $\varpi$  refer to the number of observations in the data set. Training observations  $w$  and  $\varpi$ , of flow rate  $j$ , are represented by  $\dot{m}_{j,2,w}$  and  $\dot{m}_{j,2,\varpi}$ , respectively. A class specific factor is  $f_w$ , which is assigned a value of either -1 or 1 depending on whether an observation is from class 1 or 2, respectively. The specified kernel function is represented by  $K(\dot{m}_{j,2,\varpi}, \dot{m}_{j,2,w})$ . A non-negative margin violation variable,  $\varepsilon_w$ , allows observations, from a respective class, to be across that class's edge of the margin. The constant that is specified before model training, which defines the total permissible margin violations, is  $Cs$ .

Equation 13 defines the function that can be applied after SVM model training.<sup>17</sup>

$$f(\dot{m}_{j,2}^*) = \beta_0 + \sum_{\varpi \in SV} [\alpha_{\varpi} * K(\dot{m}_{j,2,\varpi}, \dot{m}_{j,2}^*)] \quad (13)$$

where  $f(\dot{m}_{j,2}^*)$  is the predicted value for a new observation  $\dot{m}_{j,2}^*$ , and  $SV$  is a subset that specifies the support vectors.

### 3. RESULTS AND DISCUSSION

A typical gasifier observation is provided in Table 2.<sup>4</sup> This observation is applied, while adjusting the heat loss fraction in the thermal-fluid model, in order to account for uncertainty. This uncertainty is, in part, due to the trace gases that may be present in syngas,<sup>3,11</sup> which are not accounted for in the model. Furthermore, since the various hydrocarbons released during coal devolatilization are not accounted for individually in the thermal-fluid model, additional uncertainty is present.

**Table 2.** Gasifier Data<sup>4</sup>

| Variable                                                  | Measurement |
|-----------------------------------------------------------|-------------|
| Coal inlet mass flow rate [t/h]                           | 50.0        |
| $H_2O$ inlet mass flow rate [t/h]                         | 52.5        |
| $O_2$ inlet Normal volume flow rate<br>[ $10^3 m_n^3/h$ ] | 9.20        |
| Water jacket mass flow rate [t/h]                         | 8.30        |
| $H_2O$ and $O_2$ inlet temperature [ $^{\circ}C$ ]        | 344         |
| Syngas outlet temperature [ $^{\circ}C$ ]                 | 550         |
| Syngas outlet pressure [kPa]                              | 2,850       |
| Syngas volume percentage                                  |             |
| $H_2$ [%]                                                 | 39.7        |
| $CO$ [%]                                                  | 23.0        |
| $CO_2$ [%]                                                | 28.3        |
| $CH_4$ [%]                                                | 8.80        |

Table 3 provides values for the variables that, although required in the thermal-fluid model, are not presented in Table 2 and must be assumed.

**Table 3.** Assumed Gasifier Variables

| Variable                                                           | Value |
|--------------------------------------------------------------------|-------|
| $H_2O$ and $O_2$ inlet pressure [kPa] <sup>11</sup>                | 3,500 |
| Water jacket inlet and outlet pressure [kPa] <sup>1,11</sup>       | 3,500 |
| Water jacket inlet temperature [ $^{\circ}C$ ] <sup>11</sup>       | 25    |
| Water jacket outlet temperature [ $^{\circ}C$ ] <sup>1,11,15</sup> | 240   |
| Coal inlet temperature [ $^{\circ}C$ ] <sup>4,11</sup>             | 190   |
| Ash outlet temperature [ $^{\circ}C$ ] <sup>4</sup>                | 350   |

| Variable                                 | Value |
|------------------------------------------|-------|
| Ash grate frequency [rev/h] <sup>4</sup> | 4.5   |

### 3.1 Thermal-Fluid Model Implementation

The thermal-fluid model is implemented in version 11.183 of the Engineering Equation Solver (EES) software. Furthermore, it is separately implemented as versions A and B, where the following is significant. Model A applies assumed mole fractions, when calculating syngas partial pressures according to Dalton's model for gas mixtures,<sup>15</sup> for use in the EES enthalpy functions. However, Model B applies calculated mole fractions when determining syngas partial pressures. The result of this is that Model A is solvable at observations where negative mole fractions, due to the prediction of negative flow rates, are present. Model B is, however, unsolvable at these observations. Therefore, the conditions for these abnormal observations are evident in the results of versions A and B of the thermal-fluid model.

When Model A is applied to the specified gasifier data in Tables 2 and 3, predicted syngas mass flow rates and volume percentages are generated. Therefore, the sum of squares errors can be calculated for predicted syngas volume percentages. This allows the heat loss fraction to be adjusted to account for uncertainty in the model. Figure 2 presents the results of varying the heat loss fraction, in order to reduce the sum of square errors for predicted syngas volume percentages.

From Figure 2, it is evident that a minimum sum of square errors lies between heat loss fractions of 15% and

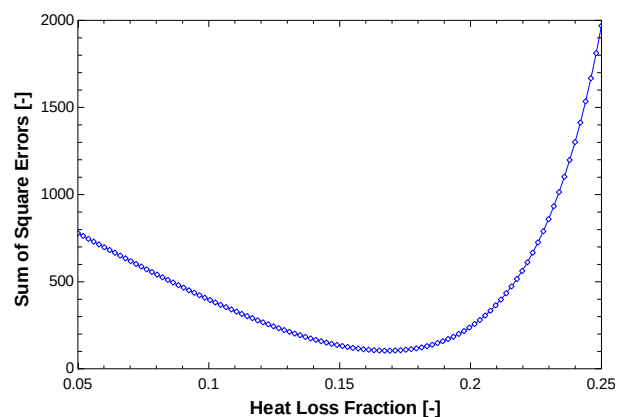


**Figure 2.** Sum of Square Errors vs Heat Loss Fraction

20%. Furthermore, the minimum sum of square errors is obtained at 16.8%, which is rounded off to 17% for application in this model. Table 4 provides the predicted syngas volume percentages for heat loss fractions between 15% and 20%. In addition, it has been determined that Model B generates predicted syngas volume percentages that correspond, with those predicted by Model A, to three significant figures. At the selected heat loss fraction, the prediction accuracies are 86.2%, 65.4%, 88.5%, and 93.1% for the  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  volume percentages, respectively.

**Table 4.** Syngas Volume Percentages per Heat Loss Fraction

| Heat loss fraction | Syngas volume percentages [%] |      |        |        |
|--------------------|-------------------------------|------|--------|--------|
|                    | $H_2$                         | $CO$ | $CO_2$ | $CH_4$ |
| 15%                | 49.0                          | 17.3 | 28.7   | 5.06   |
| 16%                | 47.2                          | 16.2 | 30.1   | 6.56   |
| 17%                | 45.2                          | 15.0 | 31.6   | 8.19   |
| 18%                | 43.0                          | 13.9 | 33.2   | 9.99   |
| 19%                | 40.5                          | 12.6 | 34.9   | 12.0   |
| 20%                | 37.7                          | 11.3 | 36.8   | 14.2   |

Given that the accuracy of the thermal-fluid model has been established, it can be applied to typical gasifier conditions.<sup>4</sup> The resulting mass flow rates, which are predicted with Model A, along with the locations of abnormal observations, from the outputs of Model B, are applied in the statistical models provided in the following sections. Statistical models are developed in version 4.1.3 of the R Statistical Software (R).<sup>20</sup>

### 3.2 Principal Component Analysis

Figure 3 is a biplot of the first two principal components that provides the scores and loadings, which were obtained from applying PCA to the syngas mass flow rates predicted by Model A. Furthermore, the flow rates provided are those for which Model B is solvable. As recommended, these flow rates are centered and standardized prior to the application of PCA.<sup>17</sup>

In Figure 3, principal component scores are plotted, in black, along the lower horizontal and left vertical axes. The loadings are plotted in blue, with the ranges indicated on the upper horizontal and right vertical axes. A 95% confidence ellipse for the scores, plotted using version 0.4.3 of the ellipse R package,<sup>21</sup> is also provided on the figure in red. Table 5 provides the loadings for the first two principal components.

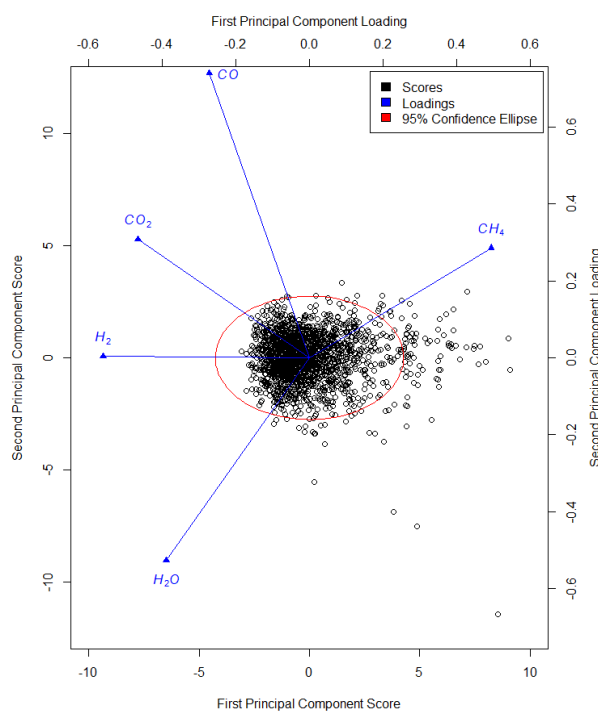


**Figure 3.** PCA Biplot

**Table 5.** Principal Component Loadings

| Variable | Component 1 loadings | Component 2 loadings |
|----------|----------------------|----------------------|
| $H_2O$   | -0.389               | -0.527               |
| $H_2$    | -0.560               | 0.003                |
| $CO$     | -0.272               | 0.739                |
| $CO_2$   | -0.466               | 0.307                |
| $CH_4$   | 0.494                | 0.286                |

The first two principal components explain 86.0% of the variability in the data. Therefore, from this two-dimensional representation, the existence of abnormal conditions can be analyzed further. However, interpretability of the developed statistical models is also significant. From Figure 3, the  $H_2$  and  $CO$ ,  $H_2O$  and  $CO_2$ ,  $CO$  and  $CH_4$ , and  $CO_2$  and  $CH_4$  mass flow rate pairs demonstrate the least correlation. This is since uncorrelated variables are indicated by loading vectors that are approximately orthogonal.<sup>17</sup> Therefore, the existence of abnormal conditions are also analyzed for these flow rate pairs.

### 3.3 Classification Tree Models

Although the ratio of unsolvable to solvable observations is relatively low at 0.022, class imbalance correction techniques are not applied in this study. This is since, as is evident in the figures provided later in this section, effectively all the observations in the minority class are separated from the majority class.<sup>22</sup> In addition, a randomly selected set comprising approximately 70% of the data is used for model training.<sup>23</sup> The remaining data are applied as a validation set.

Figure 4 presents the results from applying a CT model to the training set of  $H_2$  and  $CO$  mass flow rates predicted by Model A. CT models are trained, using version 1.0-41 of the tree R package,<sup>24</sup> to differentiate between observations where Model B is and is not solvable. Observations, and predicted regions, where Model B is

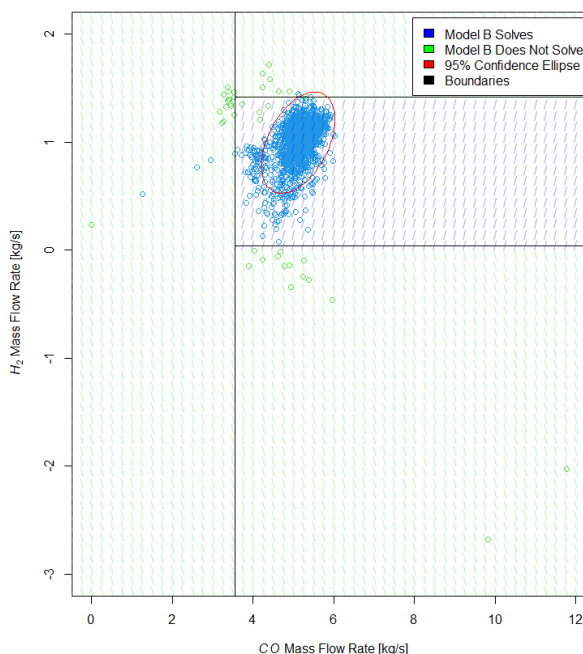


**Figure 4.**  $H_2$  and  $CO$  CT Model

solvable and unsolvable are colored blue and green, respectively.

From Figure 4, it is evident that the CT model defines three splitting rules that divide the space into four regions. In addition, five solvable and four unsolvable observations are misclassified. The training and validation accuracies for this model, and the remaining CT models presented in this section, are provided in Table 6. It is clear from Figure 4 that the region, where Model B is predicted to solve, is unbounded. Specifically, this model predicts that Model B is solvable at  $CO$  flow rates approaching infinity, and these results are not generalizable.

Figure 5 provides the results from applying a CT model to the predicted  $H_2O$  and  $CO_2$  flow rates. It is evident, from this figure, that the model defines three splitting rules that divide the space into four regions. It is also clear that 13 unsolvable observations are misclassified, which is relatively large when compared with the results of the previous model. In addition, the regions where Model B is predicted to be

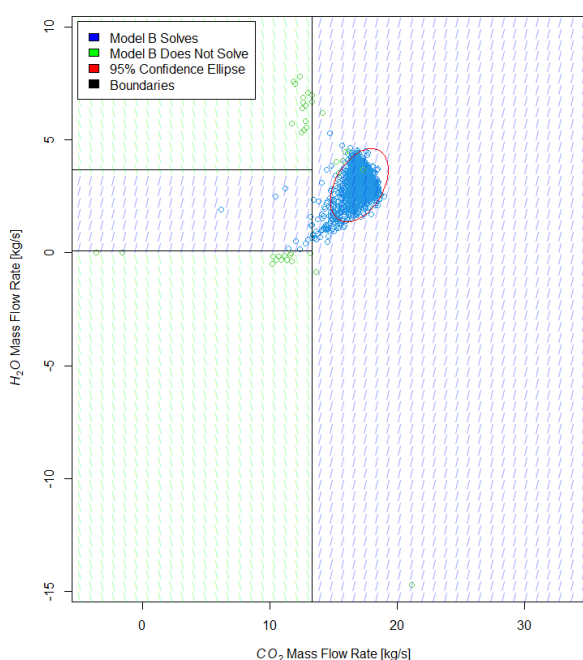


**Figure 5.**  $H_2O$  and  $CO_2$  CT Model

solvable are unbounded. Specifically, this model predicts that Model B will solve for flow rates ranging from minus infinity to infinity. Therefore, the results of this model are not generalizable.

The following results are evident when applying a CT model to the predicted  $CH_4$  and  $CO$  flow rates. The model defines two splitting rules to divide the space into three regions and only misclassifies four solvable observations. Furthermore, the splitting rules depend only on  $CH_4$

flow rates, and the model is, therefore, independent of  $CO$  flow rates. As such, the CT model predicts that Model B is solvable for  $CH_4$  flow rates between an upper and a lower limit. Therefore, the results of this CT model are generalizable.

When applying a CT model to the predicted  $CH_4$  and  $CO_2$  flow rates, it is evident that the developed model is independent of  $CO_2$  flow rates. Therefore, this model is essentially identical to the one developed for  $CH_4$  and  $CO$  flow rates, including the total number of misclassifications.

Figure 6 provides the results from applying a CT model to the first two principal components. From this figure, it is clear that the model defines three splitting rules that divide the space into four regions. This model misclassifies five unsolvable observations, and the regions where Model B is predicted to solve are unbounded. Specifically, this model predicts that Model B is solvable for principal component scores ranging from minus infinity to infinity. Therefore, these results are not generalizable.

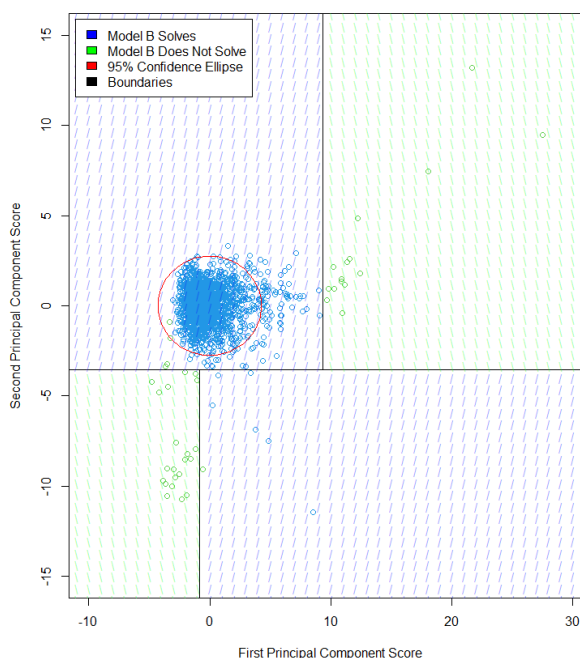


**Figure 6.** Principal Component CT Model

The results depicted in Table 6 indicate that the  $CH_4$  and  $CO$  CT model, or equivalently the  $CH_4$  and  $CO_2$  CT model, yields the highest training and validation accuracies, with both at 99.8%. The principal component CT model is the second most accurate, with training and validation accuracies of 99.7% and 99.8%, respectively. However, the  $H_2O$  and  $CO_2$  CT model

is the least accurate model presented in this section, with training and validation accuracies of 99.4% and 99.1%, respectively.

**Table 6.** CT Model Accuracies

|                                                 | Training set | Validation set |
|-------------------------------------------------|--------------|----------------|
| <i>H<sub>2</sub></i> and <i>CO</i>              |              |                |
| Accuracy                                        | 99.6%        | 99.5%          |
| <i>H<sub>2</sub>O</i> and <i>CO<sub>2</sub></i> |              |                |
| Accuracy                                        | 99.4%        | 99.1%          |
| <i>CH<sub>4</sub></i> and <i>CO</i>             |              |                |
| Accuracy                                        | 99.8%        | 99.8%          |
| <i>CH<sub>4</sub></i> and <i>CO<sub>2</sub></i> |              |                |
| Accuracy                                        | 99.8%        | 99.8%          |
| Principal components 1 and 2                    |              |                |
| Accuracy                                        | 99.7%        | 99.8%          |

### 3.4 Support Vector Machine Models

In the SVM models developed in this study, using version 1.7-9 of the e1071 R package,<sup>25</sup> second-degree polynomial kernels are applied to produce non-linear decision boundaries. This allows for comparisons between these and the linear boundaries evident for the CT models presented in the previous section. However, higher degree polynomial and radial kernel functions, which also result in non-linear boundaries, are not applied in this study. This is since the SVM models' validation accuracies are significant, and these kernels will likely result in higher variance models yielding lower validation accuracies.<sup>17</sup>

A SVM model is applied to the training set of *H<sub>2</sub>* and *CO* flow rates predicted by Model A. The aim is to differentiation between observations where Model B is and is not solvable. The

results from this SVM model are presented in Figure 7, where observations and regions are colored as in the previous section.

From Figure 7, it is clear that three solvable and nine unsolvable observations are misclassified. The training and validation accuracies for this model, and the remaining SVM models presented in this section, are provided in Table 7. It is also evident, from Figure 7, that the  $H_2$  and  $CO$  SVM model yields unbounded regions in which Model B is solvable. Therefore, the results of this model are not generalizable. Specifically, the model predicts that Model B solves for flow rates ranging from minus infinity to infinity.

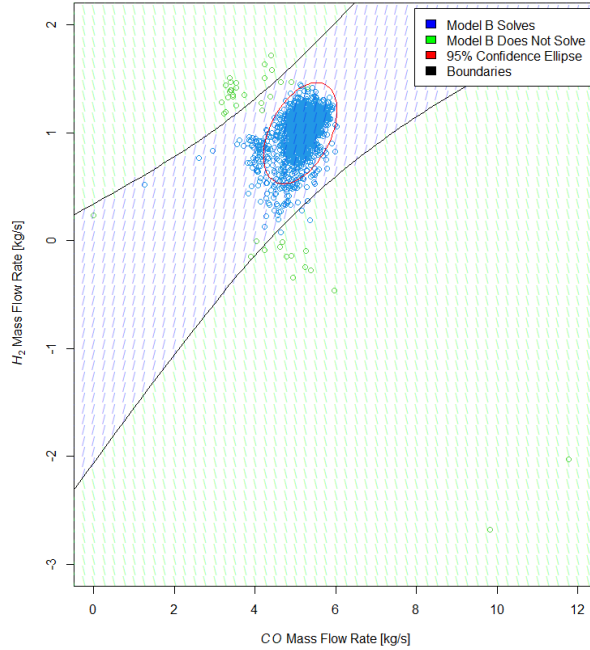


**Figure 7.**  $H_2$  and  $CO$  SVM Model

Figure 8 presents the results of applying a SVM model to the predicted flow rates of  $H_2O$  and  $CO_2$ . From this figure, it is clear that three solvable and ten unsolvable observations are misclassified. In addition, it is evident that a bounded two-dimensional region, where Model B is solvable, is predicted. In contrast, the  $H_2O$  and  $CO_2$  CT model

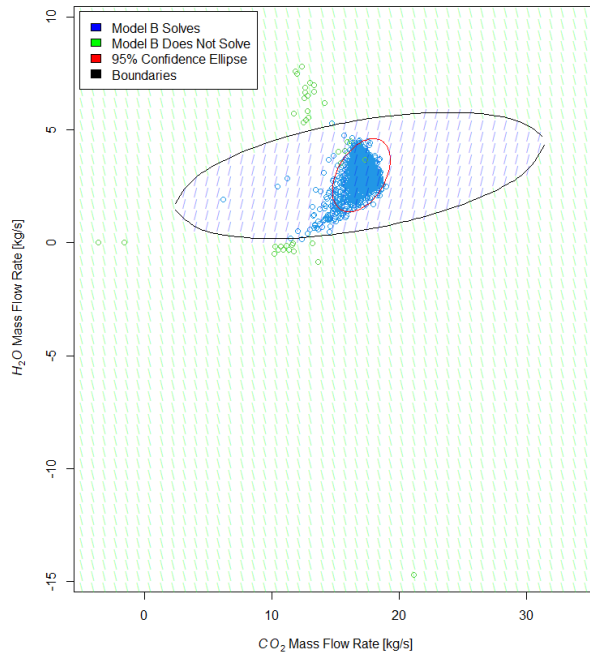


**Figure 8.**  $H_2O$  and  $CO_2$  SVM Model

predicts an unbounded two-dimensional region. Therefore, the results of the  $H_2O$  and  $CO_2$  SVM model are generalizable.

The following results are evident when applying an SVM model to the predicted  $CH_4$  and  $CO$  flow rates. This model misclassifies three solvable and six unsolvable observations, and it predicts an unbounded region where Model B is solvable. Thus, the results of the  $CH_4$  and  $CO$  SVM model are not generalizable. This is since the model incorrectly predicts that Model B will solve for flow rates ranging from minus infinity to infinity. In contrast, the  $CH_4$  and  $CO$  CT model predicts a bounded region where Model B is solvable.

When applying a SVM model to the predicted flow rates of  $CH_4$  and  $CO_2$ , the following results are clear. The developed model misclassifies two solvable and seven unsolvable observations, and the predicted region, where Model B is solvable, is unbounded. Therefore, the results of the  $CH_4$  and  $CO_2$  SVM model are not generalizable, since Model B is predicted to solve for flow rates varying from minus infinity to infinity.

The results of applying a SVM model to the first two principal components are presented in Figure 9. This model misclassifies one solvable and ten unsolvable observations. It is evident from Figure 9 that an unbounded region is developed, where Model B is predicted to solve. Therefore, since Model B is predicted to solve for principal component scores ranging

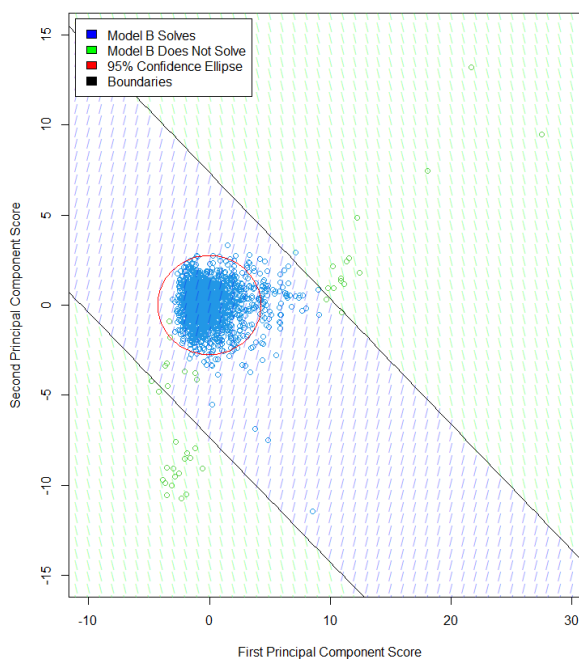


**Figure 9.** Principal Component SVM Model

from minus infinity to infinity, the results of this model are not generalizable.

The results depicted in Table 7 indicate that the  $CH_4$  and  $CO$  and the  $CH_4$  and  $CO_2$  SVM models yield the highest accuracies. The training and validation accuracies for the former model are 99.6% and 99.5%, respectively, and the training and validation accuracies for the latter model are 99.5% and 99.6%, respectively. In addition, the  $H_2O$  and  $CO_2$  SVM model yields the third highest training accuracy of 99.4% but the lowest validation accuracy of 99.1%. The principal component SVM model has a slightly higher validation accuracy of 99.6%, when compared with 99.5% for the  $H_2$  and  $CO$  SVM model. However, the training accuracies of these models are equal at 99.3%.

**Table 7.** SVM Model Accuracies

|                                                | Training set | Validation set |
|------------------------------------------------|--------------|----------------|
| <i><math>H_2</math> and <math>CO</math></i>    |              |                |
| Accuracy                                       | 99.3%        | 99.5%          |
| <i><math>H_2O</math> and <math>CO_2</math></i> |              |                |
| Accuracy                                       | 99.4%        | 99.1%          |
| <i><math>CH_4</math> and <math>CO</math></i>   |              |                |
| Accuracy                                       | 99.6%        | 99.5%          |
| <i><math>CH_4</math> and <math>CO_2</math></i> |              |                |
| Accuracy                                       | 99.5%        | 99.6%          |
| Principal components 1 and 2                   |              |                |
| Accuracy                                       | 99.3%        | 99.6%          |

It is clear, when comparing respective accuracies in Tables 6 and 7, that the CT models yield similar or slightly higher accuracies compared to the SVM models. Nevertheless, the values provided in both tables represent respectively high accuracy, where all are above 99%. Thus,

the developed CT and SVM models are all capable of differentiating between the two classes of observations.

However, only two classification models provide bounded regions where Model B is solvable. These are the  $CH_4$  and  $CO$  CT model, or the equivalent  $CH_4$  and  $CO_2$  CT model, and the  $H_2O$  and  $CO_2$  SVM model. The former model is only dependent on  $CH_4$  flow rates and is, therefore, a one-dimensional model. Although the accuracies of the  $H_2O$  and  $CO_2$  SVM model are slightly lower than those of the  $CH_4$  and  $CO$  CT model, the predicted region is two-dimensional and bounded. Therefore, a two-dimensional model, as a function of two approximately uncorrelated variables, is more representative of the region where Model B is solvable than a one-dimensional model. Therefore, the resulting region of normal gasifier operations, as predicted by the  $H_2O$  and  $CO_2$  SVM model, is selected for its generalizability.

Conventional process monitoring, for fixed-bed dry bottom coal gasifiers, involves utilizing a confidence ellipse to determine the location of abnormal observations.<sup>10</sup> This ellipse is applied to principal component scores, and observations that are inside and outside of this are judged to be normal and abnormal, respectively.<sup>10</sup>

The following is evident when the principal component confidence ellipse, in Figures 6 and 9, are applied to predict abnormal observations. The accuracy of the conventional approach is 94.4%, which is lower by 4.9% than the  $H_2O$  and  $CO_2$  SVM model's accuracy of 99.3%. This confirms that the presented approach to fault prediction in fixed-bed dry bottom coal gasifiers is a valid alternative to the established conventional approach.

### **3.5 Inlet Flow Rate Analysis**

To investigate and interpret the differences between the normal and abnormal observations identified in Sections 3.3 and 3.4, a statistical technique referred to as k-means clustering is

applied. This technique divides observations into a predefined number of clusters, according to the Euclidean distance between these and the clusters' centers, in order to reveal trends present in the data.<sup>17</sup> K-means clustering is applied separately to the inlet flow rates of steam and coal, with the purpose of analyzing the resulting groups of predicted flow rates.

Two clusters are defined per inlet flow rate, referred to as high and low respectively. Based on the intersection of these clusters, four inlet flow rate groups are identified. Figure 10 provides the predicted syngas  $H_2O$  and  $CO_2$  flow rates, which are colored according to the four inlet flow rate groups. Boundaries from the selected SVM model are also provided in this figure, along with a 95% confidence ellipse for the observations where Model B is solvable. The high steam and high coal group is colored green, while the low steam and low coal group is pink. The high steam and low coal group is colored dark blue, and the the low steam and high coal group is light blue.

It is evident from Figure 10 that the high steam and high coal group comprises most of the observations in the 95% confidence ellipse. Therefore, these observations represent normal gasifier operations.<sup>10</sup> This group is also approximately centered within the region where Model B is predicted to solve. This indicates that gasifier operations transition from normal to abnormal, as observations move from the edge of the confidence ellipse to the boundary of the SVM predicted region.

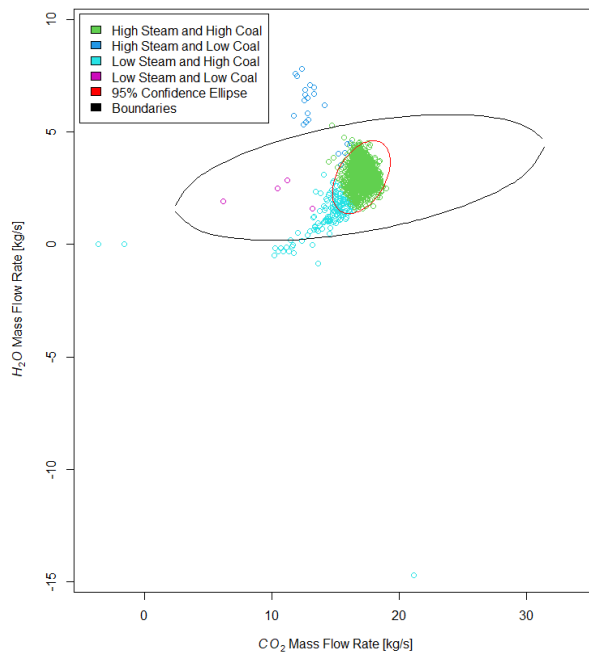


**Figure 10.**  $H_2O$  and  $CO_2$  SVM Model with Inlet Flow Rate Groups

There are two inlet flow rate groups that mainly comprise the abnormal observations present in this data. These are the high steam and low coal and the low steam and high coal groups. While the former is mainly present outside of the SVM predicted region for normal operations, the latter is largely present inside this region. Therefore, the high steam and low coal group represents abnormal gasifier operations. This is since Model B, which applies calculations based on an underlying assumption of normal operations, is largely unsolvable for this group. However, the low steam and high coal group represents a transition from normal to abnormal gasifier operations, since Model B is largely solvable for this group.

Average predicted syngas volume percentages, per inlet flow rate group, are provided in Table 8. From this table, it is evident that the closest respective average percentages, to those in Table 2, are present for the high steam and high coal group. In addition, the smallest differences in syngas composition, between the high steam and high coal group and the remaining groups, are evident for the low steam and low coal group. Therefore, given relatively normal syngas composition, this group likely represents a low gasifier load condition but not abnormal operations.

**Table 8.** Average Syngas Volume Percentages per Inlet Flow Rate Group

| Group                       | Syngas Volume Percentages [%] |      |        |        |
|-----------------------------|-------------------------------|------|--------|--------|
|                             | $H_2$                         | $CO$ | $CO_2$ | $CH_4$ |
| High steam<br>and high coal | 41.7                          | 15.0 | 31.8   | 11.4   |
| High steam<br>and low coal  | 64.9                          | 12.3 | 28.9   | -6.07  |
| Low steam<br>and high coal  | 12.3                          | 22.5 | 33.2   | 32.0   |
| Low steam<br>and low coal   | 51.4                          | 12.2 | 31.3   | 5.01   |

#### 4. CONCLUSIONS

In this study, a novel integrated thermal-fluid and statistical approach to fault prediction in fixed-bed dry bottom coal gasifiers has been developed and applied. This approach utilizes the thermodynamic principles that are established in gasifier modeling in order to predict abnormal conditions. These principles include the conservation of mass, the first law of thermodynamics, and chemical equilibrium. These are implemented, with the necessary supporting equations, in EES. The model's prediction accuracy, for normal gasifier operations, is 86.2%, 65.4%, 88.5%, and 93.1% for the  $H_2$ ,  $CO$ ,  $CO_2$ , and  $CH_4$  syngas volume percentages, respectively.

In order to determine if the results of the thermal-fluid model could be generalized, statistical models were applied. Principal component analysis was utilized to determine the pairs of mass flows rates that are the least correlated. These pairs are  $H_2$  and  $CO$ ,  $H_2O$  and  $CO_2$ ,  $CH_4$  and  $CO$ , and  $CH_4$  and  $CO_2$ . The principal components and these pairs of flow rates were applied, in statistical classification models, to predict generalizable regions where gasifier operations are normal.

Two generalizable normal operating regions were predicted. These are a one-dimensional region, comprised of an upper and a lower limit for the  $CH_4$  flow rates, and a two-dimensional region, in the form of an ellipse for the  $H_2O$  and  $CO_2$  flow rates. The two-dimensional region, predicted by a SVM model with a second-degree polynomial kernel, was selected for further analysis. Four different groups of gasifier load conditions were identified using k-means clustering. From these, two groups of abnormal conditions were identified, and the average syngas composition was provided per group.

To conclude, a fault prediction SVM model, which is a function of  $H_2O$  and  $CO_2$  mass flow rates, was trained and selected with this novel integrated statistical and thermal-fluid approach.

This SVM model yielded an accuracy of 99.3%, which is 4.9% higher than that of the conventional approach for fixed-bed dry bottom coal gasifiers.

## **NOMENCLATURE**

### **Abbreviations**

|                              |                                           |
|------------------------------|-------------------------------------------|
| °C                           | degrees Celsius                           |
| K                            | Kelvins                                   |
| kg/s                         | kilograms per second                      |
| kJ/kmol                      | kilojoules per kilomole                   |
| kJ/kmol-K                    | kilojoules per kilomole Kelvin            |
| kmol/s                       | kilomoles per second                      |
| kPa                          | kilopascals                               |
| kW                           | kilowatts                                 |
| rev/h                        | revolutions per hour                      |
| $10^3 \text{m}_n^3/\text{h}$ | thousand Normalized cubic meters per hour |
| t/h                          | tons per hour                             |

### **Acronyms**

|     |                              |
|-----|------------------------------|
| CT  | classification tree          |
| EES | Engineering Equation Solver  |
| PCA | principal component analysis |

|     |                        |
|-----|------------------------|
| R   | R Statistical Software |
| SVM | support vector machine |

## Symbols

|        |                 |
|--------|-----------------|
| $CO_2$ | carbon dioxide  |
| $CO$   | carbon monoxide |
| $H_2$  | hydrogen        |
| $CH_4$ | methane         |
| $H_2O$ | steam           |

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