

CHAPTER 1

1.0

GENERAL INTRODUCTION

1.1 Introduction

This chapter briefly introduces the research project and concisely answers the basic management questions of “what, why, and how”. Sections 1.1 - 1.2.1 give the background information and motivation for engaging in this study. The working hypotheses and objectives of the research work are presented in Sections 1.3 and 1.4, while the scope of the research is laid down in Section 1.5.

1.2 Background Information and Motivation

Coal is the most abundant fossil fuel and it will be available long after petroleum and natural gas wells are dry (Podolski *et al.*, 2008). It is a major contribution to the fossil fuel energy sources and resources. Currently, it is the most important primary energy source for power generation as well as for industrial processes and this will remain unchanged until at least 2030 (Cloke *et al.*, 2003). Coal is comparatively cheap relative to other fossil fuels; is safe to transport; and is easy to store. It thus remains vital in achieving a diverse and balanced energy mix. It is also the major source of energy for the developed and the developing economies, providing 26% of global primary energy needs and 41% of global electricity generation (WCI Coal Statistics, 2007). This is illustrated in Figure 1.1.

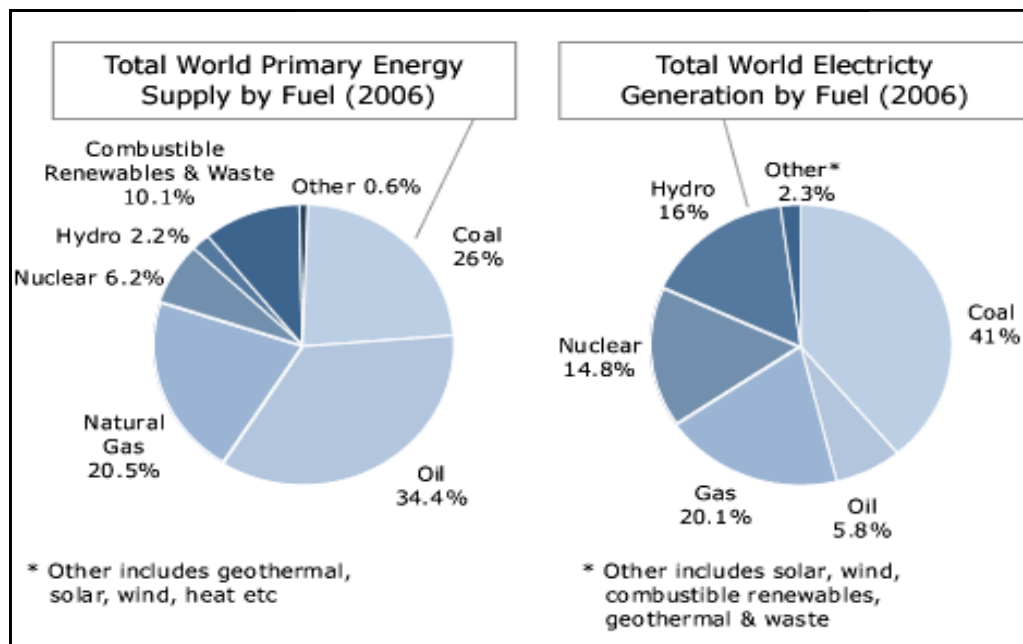


Figure 1.1: Total world energy supply and generation by fuel respectively in 2006 (WCI Coal Statistics, 2007)

South Africa is ranked 5th in the global coal export market and uses coal to generate 95% of its electricity (ESKOM Annual report, 2008), and to meet 77% of its primary energy needs (DME: Digest of South African Energy Statistics, 2006). ESKOM fact

Sheet, (2007) also noted that this trend is not likely to change significantly in the next decade due to the relative lack of suitable alternatives to coal as an energy source.

An average of 224 million tonnes of coal is produced annually in South Africa, 25% of which is exported. The remainder is used locally by various coal utilisation industries (DME Coal Statistics, 2006). Among these users are Eskom, which uses 53% for electricity generation and Sasol, which uses 33% for transport fuel and petrochemical production. A further 12% is consumed by the metallurgical industries and the remaining 2% is used for domestic cooking and heating. These figures show the energy intensity level of South Africa and its heavy dependency on coal, more so as it produces 45% of total electricity in the African continent (ESKOM Fact Sheet, 2007). The Eskom Annual Report (2008) estimates South Africa's coal reserves to be in the order of 53 billion tones. At the current production rate, these reserves should provide almost 200 years of coal supply.

South Africa's extractable coals are located in widely separated coal provinces stretching interruptedly from the border with Botswana in the North-West, through the Limpopo and Mpumalanga provinces and into Kwazulu-Natal in the east (Keaton Energy, 2009). These coal provinces are themselves divided into distinct coalfields and most of the commercially mineable resources are contained in the Permian-aged Vryheid formations of the Ecca Group (Highveld and Witbank coalfields) (Snyman, 1989; Falcon, 1989; Snyman and Botha, 1993; Keaton Energy, 2009). Other coalfields of emerging importance are the Waterberg, the Soutpansberg and the Ermelo coalfields (Keaton Energy, 2009).

Most of the country's coal (about 83%) is currently mined in the Highveld, Witbank and Ermelo coalfields located in the Mpumalanga province (Cairncross, 2001). Geology has determined that the Witbank and Highveld coalfields are by far the most important source of South Africa's mined coal at present. However, the Waterberg deposits, which extend into Botswana, are widely expected to become the country's principal future coal resource (Snyman and Botha, 1993, Cairncross, 2001), particularly as this is the region expected to be home to many of the new generation of thermal power stations (Eskom Annual Report, 2008; Keaton Energy, 2009). The Ultretch and Klip River coalfields in Kwazulu-Natal are comparatively small and production has been slowly declining overall. However, the area produces most of the

country's anthracites as well as a fair part of its coking coals (Snyman and Botha, 1993; Cairncross, 2001).

Falcon *et al.* (2010) reported that the concentrated coal mining operation in some of the coalfields (especially the Highveld coalfield), has had the effect that the current products from this area are of low quality, Grade D (Calorific value (CV) $< 25.5 \text{ kJ}\cdot\text{mol}^{-1}$) with ash content up to or greater than 40%. The application of these low grade coals in conversion and utilisation processes (combustion and gasification) in conventional existing commercialized facilities is, therefore, limited by its low efficiency; its poor burnout characteristics; and the increased equipment investment (greater than 30%) (Hu *et al.*, 2004). Furthermore, an indisputably huge amount of environmentally unfriendly emissions (SO_x and NO_x and particulate matter) that require expensive downstream processing are produced (Marban, *et al.*, 1995; Spalding-Fecher *et al.*, 2000; Hu *et al.*, 2004; Kaitano, 2007).

Current stringent legislation has imposed pollutant emission limits on fossil fuel utility plants. This puts immense pressure on coal utility plants and coal resource users to evolve technologies and techniques for drastically reducing emissions. One of these legislations is the United Nations Framework Convention on Climate Change (UNFCCC) Clean Development Mechanism (CDM), an offshoot of the Kyoto protocol (UNFCCC Website).

The coal utilisation industries and the government are continuously investigating, assessing and encouraging various technologies for the utilisation of these low-grade coals. In this regard, the government and major coal dependent industries offer their support to research into the efficient use of the vast reserves in the Highveld, Witbank, Waterberg, Soutpansberg, and Ermelo coalfields.

1.2.1 Clean Coal Technologies

The growing worldwide awareness on pollutants emanating from coal usage has had the result that processes reliant on coal as feedstock had to evolve new processes not as harmful to the environment as the older processes. Such processes are commonly referred to as Clean Coal Technologies (CCT) and are defined as technologies

designed to enhance both the efficiency and the environmental acceptability of coal extraction, preparation and utilisation (WCI, Ecoal, 2003). The main motive for developing these new coal conversion technologies is the need to achieve significant improvements in the important areas of fuel effectiveness, technical performance and environmental impact protection (Osborne *et al.*, 1996; Koornneef *et al.*, 2007). Of the various Clean Coal Technologies that have recently emerged, Fluidised Bed Combustion (FBC) and Fluidised Bed Gasification (FBG) are more suited to the utilisation of South African low grade coals. These technologies reduce emissions and waste and increase the amount of energy produced from each tonne of coal processed (Grainger and Gibson, 1981).

FBC and FBG have gained rapid acceptance and commercialization. They can be operated at atmospheric and pressurised conditions and have been shown to be a viable alternative to Pulverised Coal Combustion (PCC) (Koornneef *et al.*, 2007). One major advantage of the FBC is that most of the pollutants produced during the conversion processes as oxides of nitrogen- NO_x and N_2O can be removed by *in-situ* reduction, while sorbents such as dolomite can be added to capture SO_x in the process (Marban *et al.*, 1995). The disposal of the associated ash from the high ash feedstocks can also be done more effectively due to the fluidised state of the ash particles. FBC has two main types: Bubbling Fluidised Bed Combustion (BFBC) and Circulating Fluidised Bed Combustion (CFBC). Both types have been developed operating at pressures between atmospheric and 100 atmospheres and at temperatures in the range of 750 °C - 950 °C. Particle sizes <25mm are used in CFBC while bigger particle sizes <50mm may be used in BFBC (Kulasekaran *et al.*, 1998; Koornneef *et al.*, 2007). Koornneef *et al.*, (2007) also noted that the major difference between the BFBC and CFBC is the particle fluidising velocity which is higher in CFBC than in BFBC, but the BFBC has the advantage that its operation is much simpler and is more suitable for poor quality coals that could require longer burnout time (Kaitano, 2007).

Integrated Gasification Combined Cycle (IGCC) power generation involving both gas and steam turbines with fluidised bed gasification has been examined extensively and commercialised internationally for coal feed stocks very different (more reactive) to typical South African coals (Littlewood, 1977; Watkinson *et al.*, 1991; Marban *et al.*,

1995; Osborne *et al.*, 1996; Buskies, 1996; Campbell *et al.*, 2000; Koornneef *et al.*, 2007; Lee, 2007; GTC, 2008; Podolski *et al.*, 2008).

This research work was therefore motivated by the quest to contribute positively to the understanding of the inherent utilisation deficiencies of these low grade coals through a better knowledge of its gasification kinetics with respect to the coal and char properties. Until recently, coal processing, conversion and utilisation studies were concentrated on high quality northern hemisphere's coals, which have low ash and high vitrinite and volatile matter content. These coals are completely different to typical Highveld coals characterised by their low volatile matter and high ash and inertinite content (Falcon, 1989; Snyman, 1989; Snyman and Botha, 1993; Cloke and Lester, 1994); which caused Snyman and Botha (1993) to described South African coals as "abnormal" in their review. Evolution of solutions and designs to use these low grade coals will translate to an extended reserve as well as an improved environment.

1.3. Hypotheses of the Study

The working hypotheses of this investigation are:

- i. That the maceral composition of parent coals: vitrinites and inertinites, and their ratio (inertinite-vitrinite ratio) have some effect on the reactivity of the subsequent chars during gasification. Liptinites content in the coal samples were very low (< 4 vol. %, *mmb*) and thus were not considered in this study. Besides, the coal samples were found to be deficient in them. It should be noted that the ash contents of the parent coal samples selected for this study are approximately equal.
- ii. That the changes that occur in these coals during the charring process (both physical and chemical), influence the gasification reactivity of the chars.
- iii. That changes in chemical composition and structure of the carbon crystallite (carbon BSU) at a molecular level, during this coal to char transition, impacts on the resultant char gasification reactivity.

1.4 The Objectives of the Study

The overall objective of this research work is to investigate the effects of the chemical and physical properties imparted on chars during pyrolysis at 900 °C, on their subsequent gasification kinetics, and to identify coal and char properties that influence char reactivity. A suitable kinetic model that can best describe the gasification behaviour of these chars derived from low grade coals towards carbon dioxide is also evaluated. The findings can be used in the design of gasifiers or implemented on existing technologies as retrofits to effectively use these low quality coals with improved efficiency, economics and minimal environmental impact.

The specific objectives of this study are therefore as follows:

- i. To study the subsequent changes in chemical, physical and structural properties from coal to char, and to investigate the resultant char carbon forms along with the changes in crystallite properties of both coals and chars.
- ii. To conduct CO_2 gasification reactivity experiments on the chars on a laboratory scale under conditions similar to fluidised bed gasification conditions.
- iii. To correlate the parent coals petrographic properties: maceral compositions, vitrinite reflectance, inertinite-vitrinite ratio, maceral and modified reactive maceral indices with the char CO_2 reactivity result.
- iv. To correlate the consequent chemical, physical and structural changes, including changes in carbon crystallite properties, of the chars with their observed CO_2 gasification reactivity.
- v. To apply the resulting data from the experiments in the kinetic modelling and validation of the gasification reaction.

To achieve these objectives, the following were undertaken:

- The original coal samples were analysed for chemical, petrographic, and physical structural properties prior to intermediate char production.
- The subsequent chars were also analysed for chemical, physical, structural and petrographic properties to understand the changes imparted on them during the transition. Petrography was used here to identify the resultant char carbon forms.
- An advanced analytic technique, XRD, was used to analyse the carbon crystallite properties of both the parent coals and the chars to identify transitional changes in crystallite parameters such as: inter-layer spacing; crystallite height and diameter; aromaticity; fraction of amorphous carbon; and degree of disorder index.
- CO_2 gasification experiments were conducted on the chars on a laboratory scale using a Thermax 500, a high pressure thermogravimetric analyser (HPTGA). The reactivity experiments involved the gasification of the chars with a $CO_2 - N_2$ mixture isothermally at atmospheric pressure (0.875 bar in Potchefstroom).
- Char conversion and reactivity was determined using the experimental results.
- Correlation of the various parent coal and char properties was done to determine their contribution to the overall reactivity.
- The gasification reaction kinetic parameters were determined and the experimental results were fitted to a suitable kinetic model taking into account the rate controlling resistance observed during the experiments.
- The model was tested and validated against the experimental results and the evaluated kinetic parameters.

1.5 Scope of the Research Work

The research route followed in this study is as shown in the flow chart of Figure 2.

A brief introduction of the research work with some background information, motivation, hypotheses and objectives are dealt with in chapter 1.

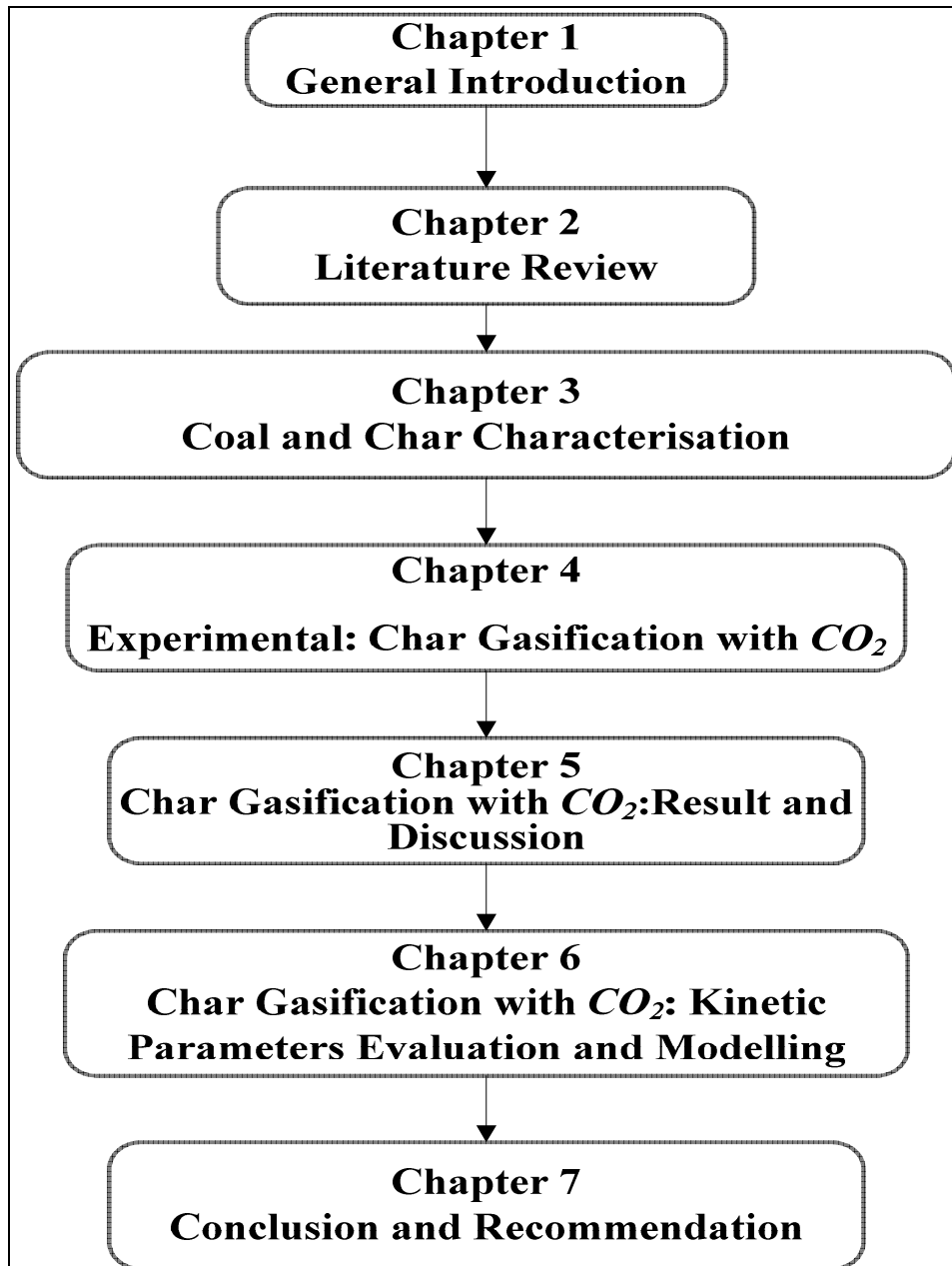


Figure 1.2: Scope of the research work.

In chapter 2, a detailed review of the relevant literatures pertaining to the coal gasification industry in general, and char- CO_2 gasification in particular is presented.

The methods used and the results of the various conventional and advanced analyses used to characterise the parent coal and the resulting chars are presented in chapter 3, while a description of the experimental apparatus, procedures and materials used are given in chapter 4.

The reactivity results from the char- CO_2 gasification experiments, including correlations with parent coal and char properties and discussions thereof, are shown in chapter 5. These results were compared with those of other investigators.

The kinetic modelling, validation and determination of the relevant kinetic parameters from the reactivity experimental data are presented in chapter 6. The results were also compared with those of other researchers.

Based on the preceding chapters, conclusions are drawn and recommendations for further studies made based on the findings of this study. These are presented in chapter 7.