Statistical modelling of an ammonium nitrate fluidised bed granulator for inference measurement

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Fluidised bed granulation is a widely used wet granulation technique. The operation of a fluidised bed granulator (FBG) as well as the quality of the product is strongly influenced by multiple process variables and disturbances. Controlling this process is difficult due to long lag times between sample analysis. Inference sensors are therefore an effective control solution for this complex process. A continuous industrial FBG was used to develop multiple linear regression (MLR) models that included two-way interaction effects. Elementary artificial neural network (ANN) models were developed to qualitatively assess the MLR models. The influences of the fluidizing air, the spray liquid and the seed particle size on the product quality were investigated and modelled. The spray liquid was found to have the largest correlation with the quality variables. Both modelling techniques produced accurate models, however undertraining of some ANN models resulted in a larger deviation between the model and validation data.

Keywords: Statistical methods, process modelling, artificial neural network, fluidised bed granulation, inference measurement

1. INTRODUCTION

Granulation is widely used in different agriculture and pharmaceutical industries for granule growth and for improvement of material properties such as bulk density, dissolution rate and shape (Zhang et al. 2000; Palis et al. 2012). Granulation is divided into (i) dry granulation, where dry powder is compressed into pellets and (ii) wet granulation, where a spray solution is used to bind granules together (Biswal 2011).

Fluidised bed granulation is a wet granulation technique comprising a single operating unit with the advantage of low operational cost and an improved mass and heat transfer rate. Wet granulation is a complex multidimensional process comprising mixing, granulation and drying, making modelling challenging. The control of an FBG is also difficult due to long sample analysing times. Inference sensors may be an efficient control solution allowing the prediction of the product quality in real-time (Burggraeve et al. 2013). A thorough understanding of the correlations between the operating and quality variables can improve the analysis, modelling and control of FBGs (Aleksic et al. 2014).

Wong et al. (2013) investigated the influences of the spray liquid flow rate, binder addition, and the distance between the spray nozzle and granulator bed on the product properties. The authors used quadratic MLR models and developed accurate models with reasonable prediction capabilities. Ziyani & Fatah (2014) used MLR models with two-way interactions to study the influences of the fluidising air flow rate, fluidising air temperature, spray liquid flow rate, and spray liquid pressure on the granule properties and obtained acceptable models.

Murtoniemi et al. (1994) compared ANN models with MLR models by investigating the influences of fluidising air temperature, atomising air pressure, and binder addition rate on the size and strength of the granules. The authors concluded that ANNs performed more accurately than the MLR models. Aleksic et al. (2014) developed ANNs with the back-propagation learning algorithm to model and predict the size and shape properties of the granules using the binder additions and granulation time as the inputs. The authors obtained high correlation values and concluded that the ANN models proved successful as a modelling technique for FBGs.

Most of the FBG models were developed on batch lab-scale FBGs with only a few operating variables being investigated. This study investigates the influences of the operating variables on the product quality of a continuous industrial FBG. The operating variables include the fluidising air, the spray liquid, and the seed particle variables, which have not received much attention yet.

The operation of an FBG and the influences of the operating variables are described in section 2 of this paper. The modelling approach is discussed in section 3 followed by the experimental description and evaluation criteria in section 4. The results and conclusions are presented and discussed in sections 5 and 6 respectively.

2.   FLUIDISED BED GRANULATION

Fluidised bed granulation comprises three main processes referred to as wetting, granule growth, and attrition. Figure 1 is a schematic representation of an FBG, displaying the different operating and quality variables.
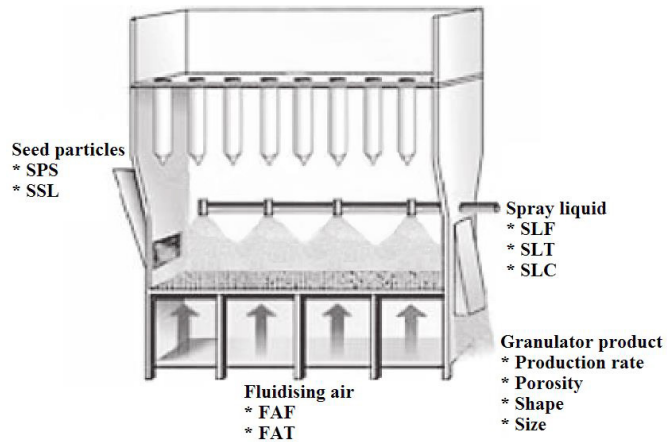

Figure 1: FBG adapted from Qiu et al. (2009, p.704)

The granulation process starts with fluidising the seed particles entering the granulator as observed in Figure 1. The fluidised seed particles are sprayed with a solution or melt of the product material. The wetted particles can follow either the agglomeration or layering growth mechanism. The growth mechanism is determined by the operating conditions of the FBG and the physicochemical properties of the material. Layering occurs when the liquid on the wetted particle dries before colliding with another particle. Collisions between wetted particles result in the formation of liquid bridges that solidifies, forming an agglomerate (Sahoo 2012; Srinivasakannan & Balasubramaniam 2003).

The rate of collision is larger than the rate of solidification, making the agglomeration the predominate growth mechanism. Attrition of granules occur during drying where agglomerates break due to weak solid bridges and collisions with other granules or the granulator wall, forming smaller granules. Some agglomerates withstand attrition and result in the production of larger granules (Iveson et al. 2001; Ziyani & Fatah 2014).

The final particles exit the granulator and are sieved where the oversize and undersized particles are recycled as seed material. The oversize particles first undergo crushing which tend to result in nonlinear oscillations of the particle size distribution (Palis et al. 2015).

FBGs are complex multidimensional process units with many influential process variables. These include the fluidisation and atomising spray conditions along with the physicochemical properties of the spray liquid (Burggraeve et al. 2013; Ziyani & Fatah 2014). The influence of some operating variables was investigated by various authors and are summarised in Table 1.

Table 1. Influences of FBG operating variables

Variable	Source	Observations
Fluidising air flow rate (FAF)	Fries et al. (2014) and Rambali et al. (2003)	Increasing FAF, increases attrition rate. Decrease of particle size.
Fluidising air temperature (FAT)	Becher & Schlünder (1998) and Ziyani & Fatah (2014)	Increasing FAT, increases evaporation rate. Decrease of particle size.
Spray liquid flow rate (SLF)	Becher & Schlünder (1998), Fries et al. (2014), and Wong et al. (2013)	Increasing SLF, increases droplet size. Increase in granule growth.
Spray liquid temperature (SLT)	Sahoo (2012)	Increase in SLT, decreases viscosity.
Spray liquid concentration (SLC)	Sahoo (2012) and Srinivasakannan & Balasubramaniam (2003)	Increase in SLC, increases growth rate.
Seed particle size (SPS)	Biswal (2011), Sahoo (2012), and Srinivasakannan & Balasubramaniam (2003)	Decrease in SPS, increases growth rate.

3.   MODELLING OF THE FLUIDISED BED GRANULATOR

A thorough understanding of the influences that the operating variables have on the process is required to develop effective inference models for control purposes (Burggraeve et al. 2013). Figure 2 represents a block diagram of a soft sensor being used for inference control. The inference model uses process variables to predict the output quality, Y, of the process in real-time, which can then be used for control purposes (Seborg et al. 2011, p.297). This study focusses on developing inference models for an FBG.
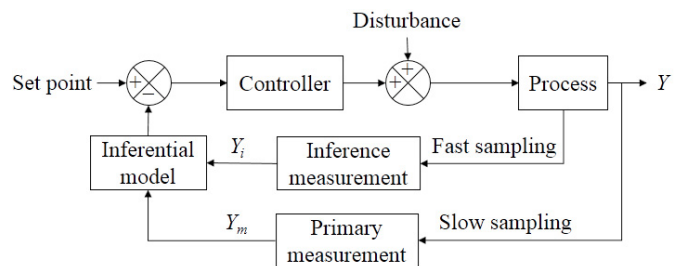

Figure 2: Block diagram for inference control adapted from Seborg et al. (2011, p.297)

Modelling approaches can be divided into (i) white-box, (ii) black-box, and (iii) grey-box. The white-box approach incorporates conservational aspects such as thermodynamics, mass and heat transfer, and particle growth into the models. Black-box models use arbitrary functions to fit experimental data. The grey-box approach combines both approaches by including the underlying chemical and physical aspects with

process data to develop models. The white-box and grey-box models are flexible but time consuming and expensive while the black-box approach develops models faster but is limited to the measurement range. The black-box approach is often considered, especially with complex processes such as granulation (Burggraeve et al. 2013; Cameron et al. 2005).

Predicting the product quality in real-time is essential for process control with common inference models following the black-box approach which includes statistical modelling techniques such as MLR and ANN (Zhu et al. 2011).

3.1 Muliple Linear Regression Models

MLR models are used to assess the strength of the relationships between independent variables with a dependent variable. The general equation for MLR models is given by (1), with $\hat{Y}$ representing the dependent variable, X_i the independent variables, b_0 the constant offset and b_i the regression coefficients. The regression coefficients are calculated using the least squares method which minimizes the sum squared error between the model and actual data (Azadi & Karimi-Jashni 2016; Berenson et al. 2012, pp.579–580).

$$\hat{Y} = b_0 + \sum_{i=1}^{k} b_i X_i \qquad (1)$$

The effect of one independent variable can change due to another independent variable and is referred to as an interaction effect. Equation (1) only includes the main effects but it can be expanded to include interactions as shown in (2) (Berenson et al. 2012, p.602).

$$\hat{Y} = b_0 + b_1 X_1 + b_2 X_2 + b_3 X_1 X_2 \qquad (2)$$

3.2 Artificial Neural Networks

ANNs is a powerful nonlinear technique that can be applied to any situation where there is a relationship between the dependent and independent variables (Hill & Lewicki 2006, p.420). ANNs are constructed with an input layer, an output layer, and hidden layers with hidden nodes. The nodes are connected to the adjacent layer with a weight value assigned to each connection, representing the strength of that connection. The values from the independent variables enter the ANN from the input layer and is multiplied by their respective weight values. The sum of the results pass through an activation function to determine the node values. This process is repeated until all the values for each node are calculated. This process is referred to as the feed forward phase (Azadi & Karimi-Jashni 2016; Hill & Lewicki 2006, pp.421–422).

The error is minimized during the training phase where the weights for each connection are adjusted. Different learning algorithms have different attributes, with the most common algorithm being the back-propagation algorithm. The algorithm takes small step changes making it stable but slow. The Gauss-Newton algorithm has faster convergence but is unstable due to a first order derivative. The Levenberg-Marquardt learning algorithm is a combination of both

algorithms, having the stability of the back-propagation algorithm and the speed of the Gauss-Newton algorithm (Hill & Lewicki 2006, p.437; Wilamowski & Irwin 2011, p.7).

4. EXPERIMENTAL PROCEDURE

4.1 Materials

Experiments were conducted on an industrial Prill Granule Ammonium Nitrate FBG with tangential spraying. Ammonium nitrate (AN) granules with an average mean particle size of 1.2 mm were used as seed particles. The spray solution consisted of AN, water, and an internal additive as binder. Atmospheric air was used as process air for fluidisation and atomisation.

4.2 Experimental Design

The independent variables are the FAF, FAT, SLF, SLT, SLC, SPS, and seed particle size slope (SSL). Five operating levels are used for each operating variable. Each variable is randomly changed to ensure discrete data. The data set consists of 73 data samples that are divided into 80% for modelling and 20% for validation of the models.

The dependent variables include the normalised run rate (RUR), normalised recycle rate (RER), spray efficiency (EFF), product mean particle size (PPS), granulator particle size (GPS), product porosity (POR), product circularity (PCR), product particle slope (PSL), and granulator particle slope (GSL).

4.3 Product Quality Analysis

The RUR is the amount of final product produced per hour. Both the RUR and RER values are calculated using a steady state mass balance. The EFF of the plant is calculated as the ratio between the fresh AN entering the granulator to the amount of AN sprayed.

The particle size information (average mean, first- and third quartile particle sizes) and circularities are determined using a Haver CPA 2-1 laser diffraction particle size analyser. The particle slope (m) is calculated using the linearized Rosin-Rammler equation given in (3) using the first- (d_{25}) and third quartile particle size (d_{75}). The POR is the ratio of the void volume over the particle volume. The values are obtained using an oil absorption technique.

$$m = \ln\left(\frac{\ln[1 - 0.25]}{\ln[1 - 0.75]}\right) / \ln\left(\frac{d_{25}}{d_{75}}\right) \qquad (3)$$

4.4 Multiple Linear Regression Model Developement

Two-way interaction MLR models were developed using IMB SPSS (2013) with the forced entry method (Landau & Everitt 2004). The p-values, obtained from a t-test, of the independent variables were evaluated to determine the statistically significant variables. The Shapiro-Wilk test of normality was used to assess the normality of the dependent variables (Field 2009, p.144). Standardised residual box plots were used to identify and remove outliers.

4.5 Artificial Neural Network Model Development

The Neural Network Toolbox™ of MATLAB® was used to develop ANN models (Beale et al. 2014). One hidden layer was chosen with a rule of thumb for the number of hidden nodes, given by (4) with N_H, N_I, and N_O representing the number of hidden nodes, inputs, and outputs respectively (Azadi & Karimi-Jashni 2016). The tan-sigmoid and pure-linear activation functions were used for the hidden and outputs nodes respectively. The Levenberg-Marquardt learning algorithm was used with the default training parameters. The maximum validation failure was set to 10 to ensure efficient training. The mean square error was used as error performance evaluation. All the independent variables were used as inputs to the neural network.

$$N_H = \frac{2}{3} \times N_I + N_O \qquad (4)$$

4.6 Model Performance Evaluation

The relationship between two variables can be determined using the Spearman's rho's non-parametric value. The Spearman's rho value determines the strength to which a monotonic function can describe the relationship between two variables. Correlation values close to 1 indicate a strong positive relationship, values close to -1 indicate a strong negative relationship, and values close to 0 indicate no relationship (Hauke & Kossowski 2011). A two-tailed test was used to assess the confidence of the Spearman values by assessing the p-values.

There is no universally applicable parameter to measure the performance and accuracy of models. The standard error of estimate (SEE) calculates the deviation of the actual data points from the predicted line. The coefficient of multiple determination (CMD) describes the amount of variation of the dependent variable that can be explained by the independent variables. The mean absolute error (MAE) is a dimensional parameter that evaluates the error between the actual and predicted values. This parameter can be used to compare models predicting the same dependent variable.

5. RESULTS AND DISCUSSION

The Spearman's rho results are summarised in Table 2. The SLF has a large positive correlation with the RUR variable. An increase in the spray flow rate will result in an increase in the production of granules. This correlation result agrees with the findings obtained from Fries et al. (2014).

A higher spray temperature increases the evaporation rate and consequently increases the concentration which promotes the layering mechanism. Layered granules are denser and stronger resulting in a better spray efficiency and lower recycle rate. The Spearman's rho results indicate a strong positive correlation between the SLT, SLC and EFF variable and a negative correlation with the RER variable.

Sahoo (2012) observed that an increase in SLT lowers the viscosity of the spray liquid. A lower viscosity and increase in evaporation rate favour the layering mechanism, resulting in an increase in production rate and the production of more spherical particles. The strong negative correlation found between the SLT and PCR is contradictory to this literature observations and might be due to the presence of an adhesive.

The FAF and FAT do not have significant correlations with any of the quality variables. These variables are important for FBG operation and might only have a small contribution towards the final product quality.

The GPS and POR quality variables have low correlations with all operating variables.

Table 2: Spearman's rho correlation matrix

	FAF	FAT	SLF	SLT	SLC	SPS	SSL
RUR	-.06	-.14	.74**	.17	.12	.30*	-.05
RER	-.00	.21	-.26*	-.55**	-.41**	.13	.04
EFF	-.05	-.09	-.04	.71**	.72**	-.26	-.08
PPS	-.16	-.27*	.10	.42**	.28*	-12	-.37**
GPS	-.08	-.09	.16	.12	-.09	.26*	-.13
POR	-.03	.22	.14	-.23	.03	-.15	.17
PCR	-.10	.01	.01	-.63**	-.49**	.17	-.09
PSL	-.28*	-.04	-.39**	-.15	-.21	-.14	.17
GSL	.01	.55**	-.39**	.35**	.20	-.36**	.47**

Note: *p<.05, **p<.01

Table 3: Performance results for the multiple linear regression and artificial neural network models

	Parameter	RUR	RER	EFF	PPS	GPS	POR	PCR	PSL	GSL
MLR	CMD$_{model}$	0.90	0.73	0.71	0.35	0.43	0.74	0.49	0.36	0.73
	CMD$_{validation}$	0.96	0.85	0.70	0.33	0.66	0.16	0.43	0.60	0.78
	SEE	0.40	1.71	5.88	0.13	0.10	1.56	1.20	0.58	0.36
	MAE	0.28	1.31	4.06	0.09	0.07	1.05	0.80	0.42	0.27
ANN	CMD$_{model}$	0.92	0.85	0.76	0.36	0.27	0.31	0.61	0.59	0.78
	CMD$_{validation}$	0.94	0.80	0.51	0.55	0.23	0.20	0.66	0.72	0.89
	SEE	0.39	1.32	5.65	0.13	0.11	2.33	1.06	0.49	0.33
	MAE	0.28	0.98	4.08	0.10	0.08	1.79	0.75	0.37	0.26

The MLR models that include interaction effects were compared to the ANN models and the performance evaluated. The results for both models are summarised in Table 3.

The RUR, RER, EFF, and GSL models show high CMD values. The results from the Spearman's rho correlation values indicated that there were significant correlations between these variables and the operating variables, resulting in these high accuracy models.

Both the MLR- and ANN RUR models fit the data with high accuracy (low MAE values) as shown in Figure 3 and Figure 4. The deviation between the data points is low, as indicated from the low SEE values. The ANN model fits the validation data points with higher accuracy, despite its lower validation CMD value.
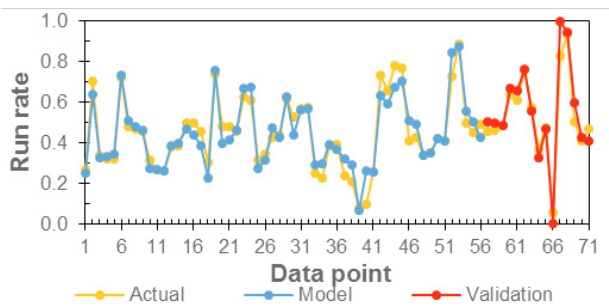

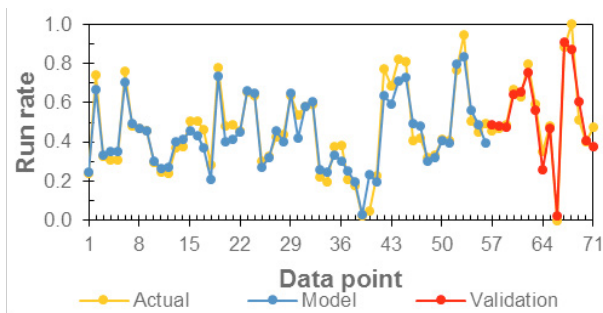Figure 3: RUR MLR model performance

Figure 4: RUR ANN model performance

There is a smaller difference in the CMD values for the ANN RER model which indicates a more reliable prediction performance from the model. The same conclusion is obtained for the MLR EFF model.

The PPS and GPS models have low CMD values indicating a low accuracy. The values are distributed almost evenly around the developed model resulting in the low SEE and MAE values. These models are inaccurate and cannot be used, despite their low error values.

The MLR POR model fits the modelling data with a high CMD value, but fails the validation fit. The ANN model fits both the modelling and validation data with low accuracies. The validation check performed during training forced the ANN to stop when the validation error started to increase. This was done to prevent overtraining but resulted in the undertraining of the ANN with a large MAE value and a large deviation between the actual and predicted values.

The PCR models have modelling and validation CMD values with small differences. The ANN model's CMD values are higher compared to the MLR model, however the MLR model performed better as seen in Figure 5. The ANN gave near linear predicted PCR values between data points 25 to 49, which indicates undertraining. The validation data contain high and low extreme values which caused a large increase in the validation error as training continued, resulting in prematurely stopping the training process of the ANN.
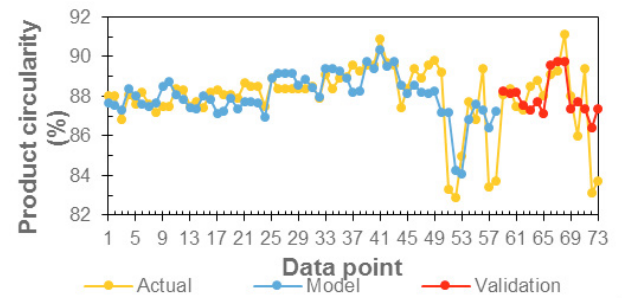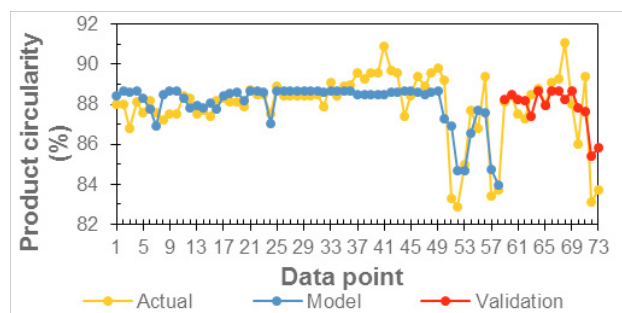

Figure 5: PCR MLR model performance

Figure 6: PCR ANN model performance

The PSL models have low CMD model values but increased CMD validation values. This indicates that the validation data were only partly representative of the modelling data set. Outliers in the modelling data set resulted in an inaccurate fit and lower CMD modelling values.

Both GSL models show high accuracies with low deviation as indicated by the low SSE and MEA values. The ANN GSL model has a CMD validation value of 90% compared to a 80% model value. The large difference in the CMD values indicates that the validation data were not representative of the modelling data. The MLR GSL model is therefore preferred due to the smaller difference in the CMD values.

6. CONCLUSIONS

The Spearman's rho matrix gave a good indication of the correlations between the operating and quality variables. Most of the correlation results obtained agreed with literature findings. It is concluded from the Spearman's matrix that the spray conditions have the strongest correlations with the quality variables. The correlations serve as a good initial step in determining the possible independent variables for each model.

Accurate models were developed using both the MLR and ANN techniques, especially with regards to the RUR, RER, EFF, and GSL variables. Both techniques struggled to obtain accurate models for the PPS, GPS, POR, PCR, and PSL quality variables. The ANN models had a better data fit compared to

the MLR models. However, undertraining occurred in some models due to early termination in training.

The ANN models were only used as a first order evaluation and were therefore not optimised. Fixed initial weight values were used which limited the training performance. Using randomised weights reduces the risk of getting stuck in local minima of the error and increases the training performance.

Larger data sets will increase the accuracy and reduce the risk of undertraining. It is also important to randomise the data to ensure that the validation data are a true representation of the full data set.

Future work includes the optimisation of the ANN models with the implementation and evaluation of the final models on-site for control purposes. Additional process variables could also be considered for inference measurement, i.e. the fluidised bed height and bed density.

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