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A Regression Model for Estimating Sugar Crystal Size in a Fed-Batch Vacuum Evaporative Crystalliser

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Crystallisation occurs in a large group of biotechnological, food, pharmaceutical and chemical processes. These processes are usually carried out in a batch or fed-batch mode. Traditionally, in sugar industry, the crystals quality is examined at the end of the process. Consequently, lack of real time measurement of sugar crystal size in a fed-batch vacuum evaporative crystalliser hinders the feedback control and optimisation of the crystallisation process. A mathematical model can be used for online estimation of the sugar crystal size. Unfortunately, the existing sugar crystallisation models are not in the form suitable for online implementation. Therefore, based on these existing models and seven process variables namely temperature (T), vacuum pressure (P_{vac}), feed flowrate (F_f), steam flowrate (Fs), crystallisation time (t), initial super-saturation (S_0) and initial crystal size (L_0), 128 data sets which were obtained from a 2-level factorial experimental design using MINITAB 14 were used to obtain a simple but online-implementable 6-input regression model for estimating crystal size. The initial crystal size (L_0) was found to play no significant role within the range of the studied process conditions. The performance of the model was evaluated. The coefficient of determination, R^2 was obtained as 0.994 and the maximum absolute relative error (MARE) was obtained as 4.6%. The high R^2 (~1.0) and the reasonably low MARE values are an indication that the proposed model can be used online for accurate estimation of sugar crystal size in a fed-batch vacuum evaporative crystalliser.

1. Introduction

Crystallisation is common in pharmaceutical, chemical and food processing industries (e.g. sugar refining). Industrially, these processes are carried out in a batch or fed-batch mode. In the sugar refining industry, the aim of sugar crystallisation is to separate sucrose from the syrup/molasses and to obtain quality sugar crystals (White et al, 1998; Anabel, 2001). The crystal quality, traditionally, is examined by crystal size distribution (CSD) at the end of the process. The CSD is quantified by the average crystal size of the distribution in the mean aperture (MA) and the width of the distribution in the coefficient of crystal size variation (CV) (Adrian, 1983). The CSD of sugar products affect its acceptability in the market. "Customers' acceptance requires individual crystals to be strong, non-aggregated, uniform in size and non-caking in the package. While for industrial purposes, reasonable size and size uniformity are desirable for filtering, washing and reacting with other chemicals" (Umo and Alabi, 2016). Large variation of CSD from market specification usually results in final product recycling which requires extra cost, energy and time. Thus, there is the need to determine the size of the sugar crystals real-time.

Georgieva et al (2003) stated that there are no techniques for the real time (online) measurement of CSD in the sugar industries and that generally, data are limited to measurements made at the end of each batch by laboratory (sieve) analysis. As good as laboratory analysis may be, there is a challenge of deviation from the required values due to the fact that during the crystallisation process, samples taken from the pan (crystalliser) for analysis do not wholly represent the nature of the crystals at the time the analysis is completed. Although Schoolnees-Muir et al (2008) reported that online measurement techniques are being developed for CSD during crystallisation processes, but none of these techniques have been commercially applied in the sugar industries (in South Africa). A review of these techniques is presented in the works of Meenesh et al (2012).

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These new methods can provide certain information on particle shapes and sizes; however, they are limited by large time delay and data-processing requirements. Thus, there is an urgent need for better techniques to estimate crystal size and related properties online (Zhang et al, 2015; Presles et al, 2009).
Sliskovic et al (2011) suggested that, to guarantee final product quality, process safety and efficiency, real time monitoring and control systems should be installed in the industrial plants. It is therefore paramount that a model for real-time estimation of sugar crystal size be developed to assist in addressing the issue of sugar crystal quality. Garcia (2001) developed a dynamic model to simulate the process units of a sugar factory. This dynamic model which is theoretical in nature consists of differential and algebraic equations which are difficult to solve. Georgieva et al (2003) compared the white, black and grey box modelling strategies which were applied to a fed-batch evaporative sugar crystallisation process. The grey box model was found to be most promising as it offers a compromise between the extensive efforts required in obtaining fully parameterized mechanistic models and the poor generalization of the data-based models. However, the model was considerably complex and, requires sophisticated software tools and computational power. Luis (2011) applied the classical model-based predictive control and the neural network model predictive control to a fed-batch crystallization process.
Unfortunately, the control loops were characterized by strong nonlinearities, difficult dynamics and large delay. A common limitation of all the existing crystallisation process models is that they contain variables that are difficult to measure online. Thus, these models are not in the form which can be directly utilised as soft sensor for real time estimation of CSD in the sugar crystallisation unit. Hence, the main contribution of this study is the development of a regression model for online estimation of sugar crystal size. The developed model focuses only on the input parameters that can easily be measured online. Moreover, it eliminates the computational burden associated with the existing theoretical models while retaining their good generalization capability.

2. Methods

2.1 Modelling approach

To estimate the crystal size (L) of sugar at any time (t) in a crystallisation unit, Anabel (2001) postulated that change in the size of the crystal is directly proportional to the time the sugar seed spent in the crystalliser and other factors that affect the change in the crystal size are lumped in the crystal growth rate (G). This relationship is expressed in Eq(1). For the statement of Anabel (2001) to hold, the crystals must have a Common History (CH). CH crystals are crystals nucleated at the same instant of time and then grown under the same temperature and super-saturation for the same length of time without any further nucleation or crystal breakage (Iswanto et al, 2007). Therefore, with proper process control in a fed-batch vacuum evaporative crystallizer, the factors that affect G are maintained at specified set-point values and consequently, G does not vary with time under these conditions. Consequently, by integrating Eq (1) between the limits (L, L_o) and (t, 0), the resulting equation is Eq(2) which was used in this work to obtain an online-implementable sugar crystal size model.

$$\frac{dL}{dt} = G \tag{1}$$

$$L = L_0 + Gt \tag{2}$$

2.2 Regression model for sugar crystal growth rate

Several researchers including Wright and White (1969), Lauret et al (2000), Georgieva et al (2003) and Iswanto et al (2006) have developed models for predicting sugar crystal growth rate (G). The limitations in modelling sugar crystal growth rate are expounded in the work of Lauret et al (2000).The crystal growth phenomenon is complex because of the large number of interacting variables and the effects of some of these variables on the kinetics are nonlinear in nature and/or even unpredictable. Because of the difficulty in formulating the physical-based mathematical models, the empirical correlations have a long tradition (Georgieva et al, 2003). The challenge with the existing empirical models is due to the difficulty of estimating the parameters of the empirical expressions through nonlinear programming (NLP) optimization technique which gives poor results, in that the convergence to the optimum parameters are not guaranteed, especially when the optimized parameters are many (more than two). This motivated the choice of the growth rate model developed by Georgieva et al (2003) in this work, as given in Eq(3),

$$G = K_g \exp\left[\frac{-57000}{R(T+273)}\right](S-1)\exp[-13.863(1-P_{sol})](1+2V_c) \tag{3}$$

where K_g, R, T, S, P_{sol} and V_c are the kinetic constant, gas constant, temperature, super-saturation, purity of solution and the volume fraction of crystals, respectively. Unfortunately, volume fraction of crystal (V_c) and the purity of the solution (P_{sol}) cannot easily be measured online. Thus, this model (Eq (3)) cannot be directly substituted into Eq(2) for online estimation of crystal size. Consequently, Eq(3) was used in this work to simulate growth rate in the design of experiment (DOE) to develop a new model with the capability for online estimation of linear growth rate as a function of temperature (T) and super-saturation (S) which can be easily measured and/or estimated online. The resulting regression model is in the form expressed in Eq(4), where $x_1$and x_2 are the independent variables (temperature and super-saturation); a_0 is the offset term (intercept); a_1 and $a_2$are the linear effects, a_3 and a_4 are the quadratic effects while a_{12} is the interaction effect.

$$G = a_0 + a_1x_1 + a_2x_2 + a_3x_1^2 + a_4x_2^2 + a_{12}x_1x_2 \tag{4}$$

The DOE, based on response surface central composite method on Minitab 14 statistical software package, was used to fit the required data to Eq(4). Table 1 shows the range of data used in the DOE for the regression model development. The data are within the typical range of operating conditions of a fed-batch evaporative sugar crystallisation unit.

Table 1: Low and high values of the input parameters for linear growth rate

S/N	Symbol	Parameter	Low	High
1	T	Temperature (^oC)	65	75
2	S	Super-saturation ()	1.15	1.25
3	P_{sol}	Purity of solution (%)	88	90
4	V_c	Volume fraction of sugar crystals ()	0.4	0.5

The model described in Eq(4) for online prediction of sugar crystal growth rate is a function of temperature and super-saturation. Although super-saturation is one of the difficult-to-measure parameters during sugar crystallisation process, the model developed by Umo and Alabi (2016) can be used for real time post-seeding super-saturation estimation. Their model is given in Eq(5),

$$\begin{aligned} S = {} & 0.349080 - 2.12210P_{vac} - 2.85042F_f + 0.166238F_s + 1.2360S_0 - 6.45838E - 05t \\ & + 33.2521P_{vac}F_f - 2.72190F_fF_s - 10.2033F_fS_0 + 0.00873790F_ft \\ & - 0.0106366F_fS_0t \end{aligned} \tag{5}$$

where: P_{vac} is the vacuum pressure, F_f is the feed flowrate, *Fs* is the steam flowrate, t is the crystallisation time, S_0 is the initial super-saturation.

2.3 Regression model for sugar crystal size

To derive the sugar crystal size model, the super-saturation model (Eq(5)) was combined with the proposed linear growth rate model (Eq(4)) and substituted into Eq(2). The model obtained is given in Eq(6).

$$\begin{aligned} L = L_0 + \Big\{ & 12.5321 - 4.38199[0.349080 - 2.12210P_{vac} - 2.85042F_f + 0.166238F_s + 1.2360S_0 \\ & - 6.45838\times10^{-5}t + 33.2521P_{vac}F_f - 2.72190F_fF_s - 10.2033F_fS_0 \\ & + 0.00873790F_ft - 0.0106366F_fS_0t] - 0.0610348T \\ & - 0.0172929[0.349080 - 2.12210P_{vac} - 2.85042F_f + 0.166238F_s + 1.2360S_0 \\ & - 6.45838\times10^{-5}t + 33.2521P_{vac}F_f - 2.72190F_fF_s - 10.2033F_fS_0 \\ & + 0.00873790F_ft - 0.0106366F_fS_0t]^2 + 6.91967\times10^{-5}T^2 \\ & + 0.0135844T[0.349080 - 2.12210P_{vac} - 2.85042F_f + 0.166238F_s + 1.2360S_0 \\ & - 6.45838\times10^{-5}t + 33.2521P_{vac}F_f - 2.72190F_fF_s - 10.2033F_fS_0 \\ & + 0.00873790F_ft - 0.0106366F_fS_0t]\Big\}t \end{aligned} \tag{6}$$

The factors that affect sugar crystal size, that can be easily measured online are identified in Eq(6). Using these factors, a two-level factorial experimental design was carried out based on the typical operating range of these factors in a fed-batch evaporative sugar crystallization unit (see Table 2).
With the aid of a pareto chart analysis, factors and their interactions with low statistical significance on the response (crystal size) were eliminated to produce a simpler and final model presented in section 3.

Table 2: Upper and lower values of input variables for sugar crystal size model

S/N	Symbol	Variables	Lower limit	Upper limit
1	T	Temperature (^oC)	65	75
2	P_{vac}	Vacuum pressure (bar)	0.2	0.3
3	F_f	Feed flowrate (m^3/s)	0	0.0275
4	F_s	Steam flowrate (kg/s)	1.4	2.75
5	S_0	Initial super-saturation ()	1.15	1.25
6	t	Crystallisation time (s)	1200	5400
7	L_0	Initial crystal size (microns)	10	12

3. Results and discussion

3.1 Regression model for sugar crystal growth rate

Response surface central composite method on Minitab 14 statistical software was used to fit the DOE data reported in Table 1. The resulting sugar crystal growth rate model is given in Eq(7).

$$G = 12.5321 - 4.38199S - 0.0610348T - 0.0172929S^2 + 6.91967 \times 10^{-5}T^2 + 0.0135844TS \quad (7)$$

The performance of the developed regression model (Eq(7)) was compared with the crystal growth rate data obtained from the growth rate model developed by Georgieva et al (2003) (i.e. Eq(3)) under the same input conditions. The results show that the model reasonably fits the data with high R^2 of 0.86. The experimental matrix for the prediction of sugar crystal growth rate consists of 31 runs in which the average and maximum relative errors in the predictions are 9.23% and 23.3%, respectively. The maximum relative error of 23.3% is as a result of non-inclusion of P_{sol} and V_c in Eq(7) because they are not easily measured online. Hence, as crystallization proceeds, V_c increases while P_{sol} decreases.
Thus, towards the end of the crystallization batch, the predictions from Eq(7) will gradually deviate from the predictions from the model of Georgieva et al (2003) (Eq(3)). However, the advantage of Eq(7) is that it can be used for online prediction of sugar crystal growth rate as opposed to the existing models.

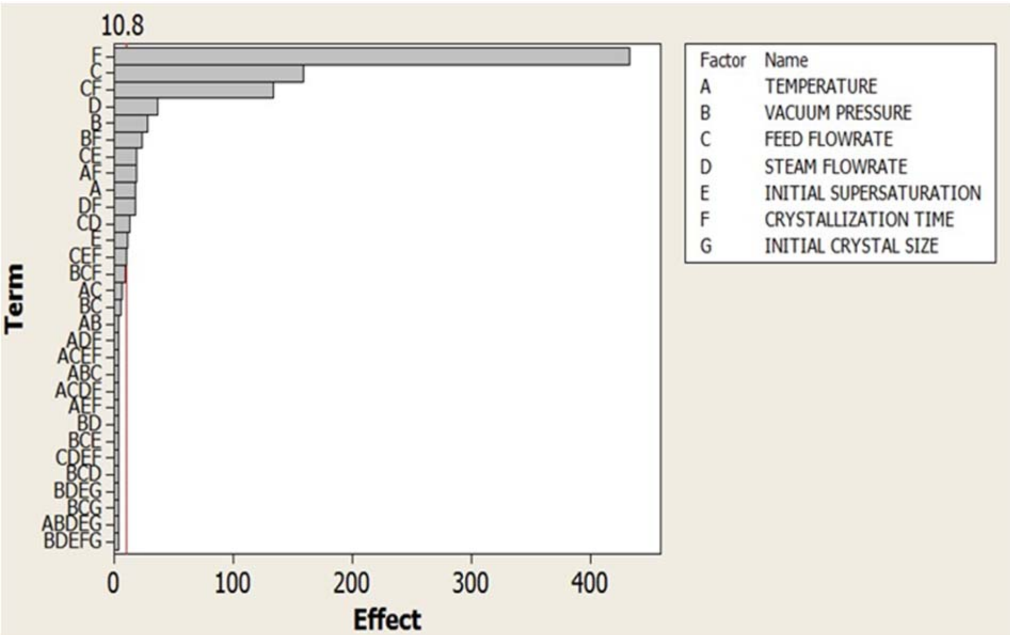

Figure 1: Pareto chart showing the effects of input factors on sugar crystal size

3.2 Regression model for sugar crystal size

The final reduced model developed in this paper for the prediction of sugar crystal size is given in Eq(8). The Pareto chart (Fgure 1) shows each of the estimated effects and the interaction of each effect. The interaction effects that had no statistical significance as shown in the Pareto chart (that is, those that are below the line running across the bars in the Pareto chart) are not included in Eq(8).

$$\begin{aligned} L = 242.688 + 0.087247T + 77.4442P - 6791.58F_f - 6.93352F_s - 222.285S_0 - 0.109252t - 6.20908 \\ \times 10^{-5}Tt - 4865.77PF_f + 0.0873013Pt + 466.376F_fF_s + 4653.64F_fS_0 \\ - 0.732568F_ft - 0.00714577F_st + 2.45876F_fS_0t \end{aligned} \quad (8)$$

On comparing the performance of the model derived in Eq(6) with that of the final reduced model (Eq(8)),the regression analysis gives the value of the determination coefficient, R^2 as 99.39% which indicates that only 0.61% of the total variations are not explained by the model. Moreover, the model predictions have a maximum relative error of 4.6% which is deemed accurate enough for practical applications. In addition, analysis of variance (ANOVA) of the Eq(8) shows that the model is significant as reflected in the very low p-value in the main effects (see Table 3).

Table 3: Analysis of Variance for Sugar Crystal Size Regression Model

SOURCE	DF	Seq SS	Adj SS	Adj MS	F	P
Main Effects	6	6903945	6903945	1150658	2812.68	0.000
2-Way interactions	6	625024	625024	104171	254.64	0.000
3-Way interactions	2	5799	5799	2899	7.09	0.001

4. Conclusions

Hitherto, the existing sugar crystallisation process models contain variables that are difficult to measure online. Thus, these models are not in the form which can be directly utilised as soft sensor for real time estimation of CSD in the sugar crystallisation unit. In this study, a regression model for online estimation of sugar crystal size was developed as a function of easy-to-measure input variables. The performance evaluation of the model show that the coefficient of determination, R^2 is 0.994 and the maximum absolute relative error (MARE) is 4.6%. It is concluded that the high R^2 (~1.0) and the reasonably low MARE values are an indication that the proposed model can be used online for accurate estimation of sugar crystal size in a fed-batch vacuum evaporative crystalliser.

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