

Predictive Model for Post-Seeding Super-Saturation of Sugar Masseccuite in a Fed-Batch Evaporative Crystalliser

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Abstract—The conflicting reports on the performances of the online probes for super-saturation of sugar masseccuite necessitate the application of soft-sensor to complement or replace them. Unfortunately, the available sugar crystallisation models which are theoretical and semi-empirical in nature are not in the form which can be directly utilised as soft sensor for real time estimation of the masseccuite super-saturation. Therefore, in this study, easy-to-measure online variables that can be correlated with the super-saturation were identified and used to develop a regression model for online estimation of the super-saturation value of sugar masseccuite after seeding. The post-seeding regression model gave coefficient of determination and maximum relative error of 0.994 and 4.7%, respectively. It is therefore concluded that the resulting model has the potential of being used for real time estimation of post-seeding super-saturation of sugar masseccuite, as opposed to the existing complex fundamental and semi-empirical sugar crystallisation models.

Index Terms—predictive model, super-saturation, post-seeding, sugar crystallization

I. INTRODUCTION

Super-saturation is the main driving force of crystallisation; it increases the rate of diffusion of sucrose molecules to the crystal surface [1]. Increase in super-saturation increases the rate of crystallisation; however, its effect on sugar crystal quality cannot be ignored. High super-saturation (labile zone) will result in poor crystal size distribution and will make centrifuging of the product difficult. Moreover, conglomerates (twin or multiple crystals) will be formed. This will have serious consequences, not only on the crystal size distribution, but the product colour because removal of the mother liquor during centrifuging from the multiple crystals is less than perfect [2]. The online probes currently in use for crystallisation control measure one or two parameters of the masseccuite and not the super-saturation directly. At the moment, accuracy of these hardware sensors is an issue, as the variable to be measured is a multivariable function with many unknowns [1], [2]. Moreover, there are conflicting reports on their performances (see the results presented in [2]-[5]. Richardson and Co-workers

[6] recommended that when there is difficulty in the assessment of super-saturation when measuring a concentration-dependent property of the system, a component mass balance can be used to predict the super-saturation. This approach can be referred to as soft-sensor, based on the definition given in [7]. However, the available sugar crystallisation models which are theoretical and semi-empirical in nature focused on different variables and not super-saturation. Moreover, they contain variables that are difficult to measure online they consist of several nonlinear algebraic-differential equations which must be solved numerically. Thus, these models are not in the form which can be directly utilised as soft sensor for real time estimation of masseccuite super-saturation in the sugar crystallisation unit. Umo and Alabi [8] proposed a regression model for explicit prediction of super-saturation of masseccuite as a function of variables that can be measured online. Unfortunately, this model is limited to pre-seeding situation, i.e. before the crystals formation begin. In this current study, easy-to-measure online variables that can be correlated with the masseccuite super-saturation after seeding were identified. Consequently, a regression model for predicting post-seeding super-saturation of masseccuite as a function of the identified easy-to-measure online variables was developed. This model has the potential for online application, as opposed to the existing complex fundamental and semi-empirical models. The remaining sections of this paper are organised as follow: Section 2 discusses the theories of modeling evaporative sugar crystallisation process; Section 3 presents the methods utilised in the data generation and post-seeding super-saturation model development; Section 4 presents and discusses the obtained results while in Section 5, relevant conclusions based on the findings are drawn.

II. THEORY OF MODELLING EVAPORATIVE SUGAR CRYSTALLISATION PROCESS

Modelling of a crystallisation process is conceptually obtained by appropriate mass and energy balances together with a mathematical representation of the crystallisation rate. The rate of crystallisation can be computed through basic mass transfer considerations or by a population balance represented by its moment [9], [10]. The use of population balance to account for the

rate of crystallisation is common in models that involve the prediction of crystal size distribution [5], [9], [11]-[13]. Although this makes the model very complex, it accounts for the initial experimental distributions and considers complex mechanisms such as those of size dispersion and/or particle agglomeration/aggregation [9]. To carry out material balance in an evaporative sugar crystalliser, the mass balance equations for water (M_w), impurities (M_i), dissolved sucrose (M_s) and crystals (M_c) are involved. Thus, the material balance consists of four set of algebraic-differential equations (see Refs. [9], [10], [13], [14]). In order to compute the rate of crystallisation ($\frac{dM_c}{dt}$) for the material balance to be complete, the population balance is derived (see Refs. [9], [10], [13]-[15]) and is given by the derivative of the first four moments $\frac{d\mu_0}{dt}$, $\frac{d\mu_1}{dt}$, $\frac{d\mu_2}{dt}$ and $\frac{d\mu_3}{dt}$. Petia and Co-workers [9] identified the zeroth and the first particles. They related the crystallisation rate to the derivative of the first moment as given by (1).

$$J_{cris} = \rho_c \frac{d\mu_1}{dt} \quad (1)$$

This derivation (1) given in [9], is not in agreement with normal physical meaning attached to moment equations, where the zeroth, first, second and third moments represent the total number, length, surface and mass of crystals per unit volume of suspension (see [5], [14], [15]). The mathematical representation for the physical meaning of these moments is provided in [15], as given in (2)-(5).

$$N_c = \int_0^\infty n(L) dL = \mu_0 \quad (2)$$

$$L_c = \int_0^\infty L n(L) dL = \mu_1 \quad (3)$$

$$A_c = k_a \int_0^\infty L^2 n(L) dL = k_a \mu_2 \quad (4)$$

$$W_c = k_v \rho_c \int_0^\infty L^3 n(L) dL = k_v \rho_c \mu_3 \quad (5)$$

where k_a is the surface area shape factor and k_v is the volume shape factor.

The expression, given in (1), used by Petia and Co-workers [9] for the rate of crystallisation, is justified since the volume growth rate G_v was used in place of linear growth rate (G) of crystal; hence, the reason the μ_2 (m^4) and μ_3 (m^5) have no physical meanings. By applying linear growth rate (G) in the population balance equations, the rate of crystallisation is given as a derivative of the third moment (6).

$$J_{cris} = \rho_c \frac{d\mu_3}{dt} \quad (6)$$

A mathematical representation of the crystallisation rate given in (7) was applied in Ref. [14]. Here, R_G is the mass growth rate while A_T is the total surface area of crystal.

$$\frac{dM_c}{dt} = R_G A_T \quad (7)$$

Growth rate is contained in (1), (6) and (7). In order to calculate growth rate, the value of super-saturation must be known. Cedric and Co-workers [10] proposed a mathematical representation for crystallisation rate that

omit growth rate and does not involve the complexity of population balance. This representation is given by (8).

$$\frac{dM_c}{dt} = cc(\rho_f F_f - J_{vap}) + \alpha_{cris} \quad (8)$$

Equation (8) is preferred in this current work since the aim is the prediction of super-saturation of massecuite and not the crystal size distribution. In addition, to account for the rate of evaporation in the material balance equation, energy balance must be carried out in the crystallisation unit. The energy balance in an evaporative crystallisation unit was presented in Refs. [10] and [14]. The challenge in implementing their models were the difficulties in determining the enthalpy terms and specific heat capacities derived as time dependent functions of physical and thermodynamic properties. The model, applied by Georgieva and Co-workers [12], Petia and Co-workers [9] and Luis and Co-workers [13], as shown in (9) does not suffer from these challenges, as it incorporates the enthalpy terms and specific heat capacities.

$$\frac{dT_m}{dt} = aJ_{cris} + bF_f + cJ_{vap} + d \quad (9)$$

III. METHODS

A. Factorial Design of Experiment for Post-Seeding Super-Saturation Prediction

The factors affecting the post-seeding super-saturation of massecuite during sugar crystallisation process were studied using factorial experimental design. Preliminary simulation experiments on sugar crystallisation process indicate that the most important factors for the online estimation of post seeding super-saturation are feed flow rate (F_f), steam flow rate (F_s), pressure of vacuum (P_{vac}), initial super-saturation (S_0) and the change in time (t). Hence, these factors were chosen as the independent variables while super-saturation (S) is the dependent (output) variable. Thirty two simulations, based on 2-level full factorial experimental design, were carried out. The low and high values of the input factors used in the factorial experimental design are presented in Table I.

TABLE I. LOW AND HIGH VALUES OF THE INPUT FACTORS

factor	Name	Low	High
P_{vac}	pressure of vacuum (kPa)	0.2	0.3
F_f	feed flow rate (m^3/h)	0.0000	0.0275
F_s	steam flow rate (m^3/h)	1.1	2.1
S_0	initial super-saturation value	1.02	1.25
t	Change in time (sec)	500	2000

B. Numerical Solution of the Fundamental Crystallisation Model

The model of the crystalliser which was obtained from Refs. [10], [12]-[14] and [16] involves appropriate mass balance, energy and population balances/mathematical representation of the crystallisation rate. The mathematical representation of crystallisation rate utilised in this study is that which was applied by Cedric and Co-workers [10] to an industrial scale fed-batch evaporative 'C' sugar crystallisation process; this is given in (13). The mass balance equations for water (M_w), impurities (M_i), dissolved sucrose (M_s), and crystals (M_c)

in an evaporative sugar crystallisation unit consist of the following set of algebraic-differential equations.

$$\frac{dM_w}{dt} = F_f \rho_f (1 - B_f) + F_w \rho_w - J_{vap} \quad (10)$$

$$\frac{dM_i}{dt} = F_f \rho_f B_f (1 - Pur_f) \quad (11)$$

$$\frac{dM_s}{dt} = F_f \rho_f B_f Pur_f - \frac{dM_c}{dt} \quad (12)$$

$$\frac{dM_c}{dt} = cc(\rho_f F_f - J_{vap}) + \alpha_{cris} \quad (13)$$

The energy balance in an evaporative crystallisation unit is given in (14).

$$\frac{dT_m}{dt} = aJ_{cris} + bF_f + cJ_{vap} + d \quad (14)$$

where parameters a, b, c and d incorporate the enthalpy terms and specific heat capacities derived as time dependent functions of physical and thermodynamic properties as given in (15), (16)-(18) where W and Q are the stirrer power and heat input, respectively.

$$a = \frac{1}{M_{sol}Cp_{sol} + M_cCp_c} \left[H_{sol} - H_c + M_c + (1 - B_{sol}) \frac{dH_{sol}}{dB_{sol}} + \frac{1 - Pur_{sol}}{B_{sol}} \frac{dH_{sol}}{dPur_{sol}} \right] \quad (15)$$

$$b = \frac{\rho_f}{M_{sol}Cp_{sol} + M_cCp_c} \left[H_f - H_{sol} + (B_f - B_{sol}) \frac{dH_{sol}}{dB_{sol}} + \frac{B_f}{B_{sol}} (Pur_f - Pur_{sol}) \frac{dH_{sol}}{dPur_{sol}} \right] \quad (16)$$

$$c = \frac{1}{M_{sol}Cp_{sol} + M_cCp_c} \left[H_f - H_{vap} - B_{sol} \frac{dH_{sol}}{dB_{sol}} \right] \quad (17)$$

$$d = \frac{1}{M_{sol}Cp_{sol} + M_cCp_c} \left[W + Q + F_w \rho_w (H_w - H_{sol} + B_{sol}) \frac{dH_{sol}}{dB_{sol}} \right] \quad (18)$$

$$Q = \alpha_s F_s \Delta H_s \quad (19)$$

The rate of evaporation is given in (20). The correlations for physical properties used in the model are adapted from [12] and [13].

$$J_{vap} = \frac{W+Q}{\lambda_w(vac)} + K_{vap}(T_m - T_w(vac) + BPE) \quad (20)$$

Based on the solution of the mass and energy balance (10)-(20), the super-saturation level of the masseccite is obtained using (21).

$$S = \frac{\left(\frac{Bx_{sol}}{100 - Bx_{sol}} \right)}{\left(\frac{Bx_{sat}}{100 - Bx_{sat}} \right) \times C_{sat}} \quad (21)$$

The correlations for physical properties used in (21) are given in (22)-(25). All other correlations for physical properties used in the evaporative sugar model are adapted from [12] and [13].

$$Bx_{sol} = \frac{M_s + M_i}{M_{sol}} \quad (22)$$

$$Bx_{sat} = 64.447 + 8.22 \times 10^{-2} T_m + 1.66169 \times 10^{-3} T_m^2 - 1.558 \times 10^{-6} T_m^3 - 4.63 \times 10^{-8} T_m^4 \quad (23)$$

$$C_{sat} = 0.1 \frac{Bx_{sol}}{100 - Bx_{sol}} (1 - Pur_{sol}) + 0.4 + 0.6 \exp \left(-0.24 \frac{Bx_{sol}}{100 - Bx_{sol}} (1 - Pur_{sol}) \right) \quad (24)$$

$$Pur_{sol} = \frac{M_s}{M_s + M_i} \quad (25)$$

TABLE II. INITIAL INPUT PARAMETERS FOR SIMULATION OF POST-SEEDING SUPER-SATURATION OF SUGAR MASSECCITE

Input variables	Value
K_{vap} (kg/s.°C)	0.03
W (J/s)	15000
∞_s	0.0076
F_s (kg/s)	2
P_s (bar)	2
T_s (°C)	150
P_{vac} (bar)	0.3
F_f (m ³ /s)	0.0001
ρ_f (kg/m ³)	1356.235
ρ_c (kg/m ³)	1580
B_f	0.72
M_c (kg)	5000
F_w (m ³ /s)	0
ρ_w (kg/m ³)	1000
Pur_f	0.9
T_f (°C)	70
T_w (°C)	70
R	8.314
∞_{cryst}	0.9217
S_0	1.25

Although the above equations are elegant, they are bulky and cannot be directly utilised as soft sensor for online estimation of super-saturation of the sugar masseccite. The novelty in this work involves the conversion of these hitherto complex models to a single simple model that has the potentials of being used for real time estimation of the masseccite super-saturation. To achieve this, first, these algebraic-differential equations are solved numerically over the practical ranges of operating conditions (see Table I and Table II) of sugar evaporative crystalliser. The numerical solution was carried out using explicit Euler method in Microsoft Excel spreadsheet. Second, a portion of the data (numerical solutions) obtained was used to estimate the parameters: $b_0, b_1, b_2, \dots, b_5$ and $b_{12}, b_{13}, \dots, b_{12345}$ of the proposed regression model given in (26) using Minitab 14 statistical software.

$$Y = b_0 + b_1 x_1 + b_2 x_2 + b_3 x_3 + b_4 x_4 + b_5 x_5 + b_{12} x_1 x_2 + b_{13} x_1 x_3 + b_{14} x_1 x_4 + \dots \quad (26)$$

where Y is the predicted response variable (post-seeding super-saturation of masseccite); x_1, x_2, x_3, x_4 and x_5 are the independent variables (feed flow rate, steam flow rate, pressure of vacuum, initial super-saturation and the change in time, respectively); b_0 is the offset term (intercept); b_1, b_2, \dots, b_5 are the linear effects while $b_{12}, b_{13}, \dots, b_{12345}$ are the interaction effects. The operating conditions used in the simulations are based on the average of the available industrial conditions and input constraints as reported in Refs. [1], [10], [13], [16] and [17]. The initial input parameters used in the simulation are as shown in Table II.

IV. RESULTS AND DISCUSSION

Following the steps outlined in Section (3), a regression model (given in (27) for predicting the post-seeding super-saturation of masseccite was obtained. The

dependent variable (super-saturation (S)) was obtained as the sum of the contributions of the independent variables ($P_{vac}, F_f, F_s, S_0, t$) and the interaction terms in the regression model, as given by (27). The Pareto chart Fig.

$$S = 0.349080 - 2.12210P_{vac} - 2.85042F_f + 0.166238F_s + 1.23607S_0 - 6.45838E - 05t + 33.2521P_{vac}F_f - 2.72190F_fF_s - 10.2033F_fS_0 + 0.00873790F_ft - 0.0106366F_fS_0t \quad (27)$$

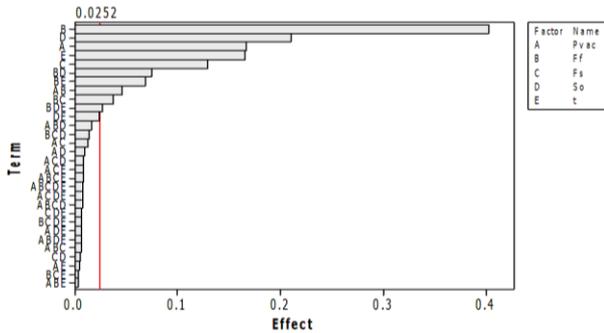


Figure 1. Pareto chart of the effect of input factors on post-seeding super-saturation of masecuite

TABLE III. ANALYSIS OF VARIANCE FOR POST-SEEDING SUPER-SATURATION REGRESSION MODEL

Source	DF	Seq SS	Adj SS	Adj MS	F	P
Main Effects	5	2.22371	2.22371	0.444741	607.91	0
2-Way Interactions	4	0.10995	0.10995	0.027489	37.57	0
3-Way Interactions	1	0.00583	0.00583	0.005826	7.96	0.01
R ² = 99.35%						
Max. %relative error=4.67						

The regression analysis gives the value of the determination coefficient, R² as 99.35% which indicate that only 0.65% of the total variations are not explained by the model. Moreover, the model predictions have a maximum relative error of 4.7% which is deemed accurate enough for practical applications. In addition, analysis of variance (ANOVA) of the regression model for post-seeding super-saturation demonstrates that the model is significant as reflected in the very low p-value in the main effect (see Table III).

V. CONCLUSION

The existing complex theoretical and semi-empirical sugar crystallisation models are not in the form which can be utilised as soft sensors for real time estimation of post-seeding super-saturation of sugar masecuite. In this study, feed flow rate, steam flow rate, pressure of vacuum, initial super-saturation and change in time were identified as easy-to-measure online variables that can be correlated with the super-saturation of sugar masecuite. Consequently, a regression model for predicting post-seeding super-saturation of sugar masecuite as a function of the identified easy-to-measure online variables was developed. The evaluation of the predictive ability of the model was found to be satisfactory, as it gave the coefficient of determination (R²) and the maximum relative error of 0.994 and 4.7%, respectively.

1 shows each of the estimated effects and interactions of each of the effects. Thus, interaction effect that has no statistical significance as shown in the Pareto chart are not included in (27).

It is therefore concluded that the resulting model has the potential of being used for real time estimation of post-seeding super-saturation of sugar masecuite, as opposed to the existing complex fundamental and semi-empirical sugar crystallisation models.

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