SUCROSE COOLING CRYSTALLIZATION MODELLING

Valeriy MYRONCHUK[1], Oxana YESHCHENKO[2], *Maryna SAMILYK[3]

[1]National University of Food technologies, Kyiv, Ukraine, mironcuk@nuft.edu.ua
[2]National University of Food technologies, Kyiv, Ukraine, oxayes@mail.ru
[3]National University of Food technologies, Kyiv, Ukraine, m.samilyk@ukr.net
* Corresponding author
Received April 8th 2013, accepted May 15th 2013

Abstract: *Based on the material balance equations and understanding of the final-grade massecuite cooling crystallization process as the technology object, a simulation model of the process has been built by which the computational experiments have been conducted. By results of these experiments, analytical exponential dependences of the massecuite characteristics change during cooling crystallization have been obtained, namely, grain content, weight, purity and dry solids weight ratio of massecuite syrup. The constructed model has been used to study the industrial cooling crystallization process. It is proved that the results of the experiment of the developed simulation model fully reflect the nature of the industrial process of final-grade massecuite cooling crystallization of sucrose. Typical scheme industrial cooling crystallization with massecuite water or impure sugar solution dilution and authors' scheme with an intermediate heat massecuite have been considered. It's experimentally shown that the use of final-grade massecuite intermediate heating after cooling to 50-52 ºC by 7-10 ºC increases the effect of crystallization to 8.4% by reducing the viscosity of massecuite syrup, the surface tension and alignment of the massecuite cooling rate at sucrose crystalization rate. Consequently, the exclusion of dilution of massecuite by water or impure sugar solution and its replacement by intermediate heating provides a more complete depletion of molasses and increases the amount sugar grains in the massecuite.*

Keywords: *massecuite, syrup, grains, dilution, mixer-crystallizer, the intermediate heat.*

1. Introduction

Additional sucrose crystallization process by cooling has complex dynamics and depends on many factors [1] – [7]. That is why there are many difficulties in regulating the cooling of the massecuite [8]. In addition, the long duration of the process generates complex physical and chemical transformations of the system.

Despite numerous studies in this area, today there are gaps in the optimal mode of heat and mass transfer in the crystallization of the massecuite in the mixer-crystallizers. The study of this process using physical modelling is difficult in large part due to its duration (more than 30 hours). This can be eliminated by the use of simulation [9] – [11].

The objective of our research was to create a mathematical model of the process of crystallization by cooling, the ability to regulate it, to bring the best mode in which the crystallization rate corresponds to the cooling rate.

2. Experimental

We presented the crystallization process as a technology object (Fig. 1) with the input, control and output parameters.

The basis of the mathematical description of the sucrose cooling crystallization

process accepts the material balance of the process.
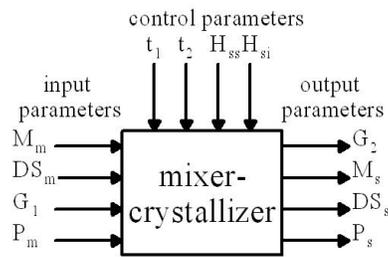
Figure 1. The sucrose cooling crystallization process in mixers-crystallizers as a technology object: input parameters: M_m – massecuite mass, DS_m – massecuite dry solids weight ratio, G_1 – grain content at the beginning of crystallization, P_m – massecuite purity; control parameters: t_1 – the initial temperature of the process, t_2 – the final temperature of the process, H_{ss} – supersaturation coefficient, H_{si} – solubility index; output parameters: G_2 – grain content at the end of crystallization, M_s – syrup mass, DS_s – syrup solids weight ratio, P_s – syrup purity.

General material balance equation:

$$M_m = M_{g_1} + M_{s_1} = M_{g_2} + M_{s_2}, \quad (1)$$

balance equation by sucrose:

$$\begin{aligned} M_m Sc_m &= M_{s_1} Sc_{s_1} + M_{g_1} = \\ &= M_{s_2} Sc_{s_2} + M_{g_2} \end{aligned}, (2)$$

balance equation by nonsugar:

$$M_m Ns_m = M_{s_1} Ns_{s_1} = M_{s_2} Ns_{s_2}, \quad (3)$$

balance equation by dry solids:

$$\begin{aligned} M_m DS_m &= M_{s_1} DS_{s_1} + M_{g_1} = \\ &= M_{s_2} DS_{s_2} + M_{g_2} \end{aligned}, \quad (4)$$

balance equation by water:

$$M_m W_m = M_{s_1} W_{s_1} = M_{s_2} W_{s_2}, \quad (5)$$

Where M_m, M_{g_1}, M_{g_2}, M_{s_1}, M_{s_2} – massecuite mass, the mass of crystals in

the massecuite and syrup at the beginning and at the end of the cooling crystallization process, kg; Sc_m, Sc_{s_1}, Sc_{s_2}, Ns_m, Ns_{s_1}, Ns_{s_2}, DS_m, DS_{s_1}, DS_{s_2}, W_m, W_{s_1}, W_{s_2} – sucrose, non-sugars, dry solids and water weight fractions in the massecuite and molasses at the beginning and at the end of process, %/

The mathematical description of the sucrose cooling crystallization is necessary to determine the sucrose amount in the syrup. It is determined from equation

$$\frac{M_s Sc_s}{100\%} = M_{w_m} H_{si}(t,P) H_{ss}, \quad (6)$$

Then

$$Sc_s = \frac{M_{w_m} H_{si}(t,P) H_{ss}}{M_s} 100\%, \quad (7)$$

where M_s – the syrup mass, kg, Sc_s – sucrose weight ratio in syrup,%, M_{w_m} – water mass in the massecuite, kg, $H_{si}(t,P)$ – solubility index of sucrose as a function of temperature and purity, H_{ss} – supersaturation coefficient of the massecuite.

Variation of sucrose solubility index with temperature and the solution is determined by the regression equation of the third order, we have received the least square method:

$$\begin{aligned} H_{si}(t,P) = {}& 1{,}0502 + 0{,}1903t - \\ &- 0{,}01827P + \\ &+ 6{,}084 \cdot 10^{-5} t^2 + \\ &+ 6{,}926 \cdot 10^{-3} P^2 - \\ &- 4{,}1505 \cdot 10^{-3} tP + \\ &+ 6{,}429 \cdot 10^{-6} t^3 - \\ &- 5{,}244 \cdot 10^{-6} P^3 - \\ &- 8{,}317 \cdot 10^{-6} t^2 P + \\ &+ 2{,}821 \cdot 10^{-5} tP^2 \end{aligned}, \quad (8)$$

Where t – the product temperature, ºC, P – sugar solution purity, %.

We believe that in the cooling crystallization process water and non-sugars content in syrup remains unchanged. Then at the beginning of cooling crystallization, we have

Syrup mass:

$$M_{s_1} = M_{w_m} H_{si}(t_1, P_1) H_{ss} + \frac{M_m Ns_m}{100\%} + \frac{M_m W_m}{100\%} \quad , \quad (9)$$

Grain mass:

$$M_{g_1} = M_m - M_{s_1} , \quad (10)$$

Grain content in the massecuite:

$$G_1 = \frac{M_{g_1}}{M_m} 100\% . \quad (11)$$

Similarly, we find the syrup and crystals masses at the end of the cooling crystallization process:

$$M_{s_2} = M_{w_m} H_{si}(t_2, P_2) H_{ss} + \frac{M_m Ns_m}{100\%} + \frac{M_m W_m}{100\%} \quad , \quad (12)$$

$$M_{g_2} = M_m - M_{s_2} , \quad (13)$$

$$G_2 = \frac{M_{g_2}}{M_m} 100\% . \quad (14)$$

In general, for any temperature in the cooling process massecuite $t_i = t_1 \ldots t_2$ we have

$$M_{s_i} = M_{w_m} H_{si}(t_i, P_i) H_{ss} + \frac{M_m Ns_m}{100\%} + \frac{M_m W_m}{100\%} \quad , \quad (15)$$

$$M_{g_i} = M_m - M_{s_i} , \quad (16)$$

$$G_i = \frac{M_{g_i}}{M_m} 100\% . \quad (17)$$

The sucrose, non-sugars, dry solids and water mass fractions in the syrup, and its purity were determined by the equations:

$$Sc_{s_i} = \frac{M_{w_m} H_{si}(t_i, P_i) H_{ss}}{M_{s_i}} 100\% , \quad (18)$$

$$Ns_{s_i} = \frac{M_m Ns_m}{M_{s_i}} 100\% , \quad (19)$$

$$DS_{s_i} = \frac{M_{w_m} H_{si}(t_i, P_i) H_{ss} + M_m Ns_m}{M_{s_i}} 100\% , \quad (20)$$

$$W_{s_i} = \frac{M_m M_{w_m}}{M_{s_i}} 100\% , \quad (21)$$

$$P_{s_i} = \frac{Sc_{s_i}}{DS_{s_i}} 100\% , \quad (22)$$

Equations (15)–(22) is a mathematical description of the model of sucrose cooling crystallization process.

On the basis of computational experiments on the model (15)–(22) we have obtained analytical dependence of the massecuite characteristics (grain content, mass, purity, and the dry solids weight ratio of syrup) on time and massecuite purity:

$$G(\tau, P_m) = a_G(P_m)\left(b_G(P_m) - e^{-c_G(P_m)\tau}\right), \quad (23)$$

$$M_s(\tau, P_m) = \frac{a_{M_м}(P_m)}{1 + b_{M_м}(P_m) e^{-c_{M_м}(P_m)\tau}} , \quad (24)$$

$$P_s(\tau, P_m) = \frac{a_{P_s}(P_m)}{1 + b_{P_s}(P_m) e^{-c_{P_s}(P_m)\tau}} , \quad (25)$$

$$DS_s(\tau, P_m) = \frac{a_{DS_s}(P_m)}{1 + b_{DS_s}(P_m) e^{-c_{DS_s}(P_m)\tau}} , \quad (26)$$

Where $\tau = \dfrac{\tau_i}{\tau_c}$ – the relative time, τ_i - current time, τ_c - the total cycle time, P_m – massecuite purity.

3. Results and Discussion

The above model we used to simulate the industrial sucrose cooling crystallization process in the mixer-crystallizers, which requires water or impure sugar solution dilution of massecuite. Although this method to some extent, can improve the crystallization conditions, the water addition in the massecuite violates isohydric conditions of crystallization. This reduces the final crystallization effect, increases the molasses amount, and hence the sucrose content in it, as well as energy costs in sugar house. For this reason, it is advisable to carry out massecuite heated to a definite temperature instead of water dilution.

The constructed sucrose cooling crystallization model is used to study the process of industrial sucrose crystallization in two modes, flow graphs of which are presented in Fig. 2.

The amount of water or impure sugar solution for dilution of massecuite is calculated as:

$$M_d = \frac{M_m\left(DS_{dm} - DS_m\right)}{DS_d - DS_{dm}}, \qquad (27)$$

Where M_d – mass of water or impure sugar solution for dilution of massecuite, M_m – masscuite mass, DS_d, DS_m, DS_{dm} – dry solids weight ratio of sugar solution for the dilution, of massecuite before and after dilution, respectively; if massecuite is diluted with water, then $DS_d = 0$.
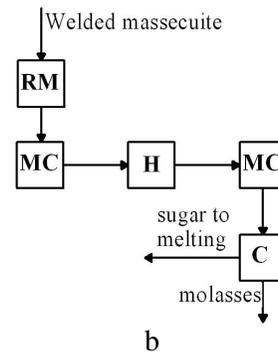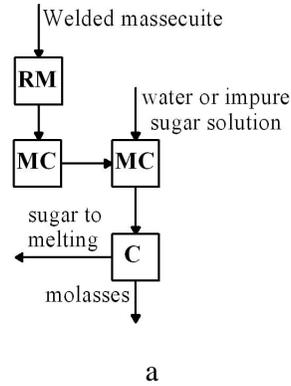

Figure 2. Flow graph of the massecuite cooling crystallization process: a – with water or impure sugar solution dilution, b – with an intermediate heat, RM – reception mixer, MC – mixer-crystallizer, H – heater, C – centrifuge.

Temperature range of the cooling crystallization process is described by dependencies:

When massecuite is diluted with water or impure sugar solution:

$$t(\tau) = \frac{a_1}{1 + b_1 e^{-c_1 \tau}} \qquad (28)$$

When intermediate heating is used

$$t(\tau) = \begin{cases} \dfrac{a_1}{1 + b_1 e^{-c_1 \tau}}, \text{ befor heating} \\[2mm] \dfrac{a_2}{1 + b_2 e^{-c_2 \tau}}, \text{ after heating} \end{cases} . \qquad (29)$$
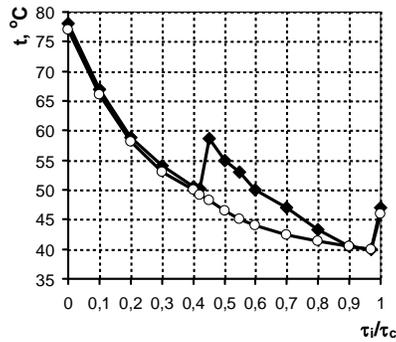
Figure 3. Temperature range of final massecuite cooling crystallization:
◆ – with an intermediate heat, ○ – to the standard mode.

It stands to reason that the water dilution not only reduces the grain content in the massecuite at the end of crystallization, but also increases syrup purity (Fig. 4).

For bringing of massecuite to a given dry solids weight ratio impure sugar solution for dilution of its must be much more than water. This may explain the smallest content of the grains in the massecuite after dilution and at the end of crystallization for the scheme with the impure sugar solution dilution. Besides that decrease the purity syrup in this case is not achieved at the expense of its desugarization, but due to high content of non-sugars which were added during dilution (Fig. 4).
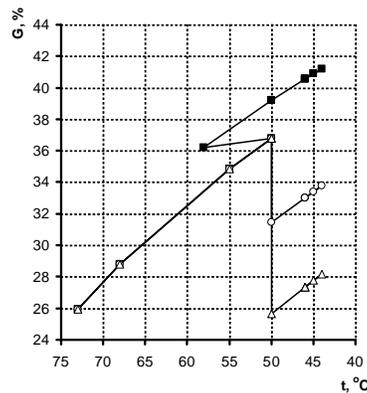
a.
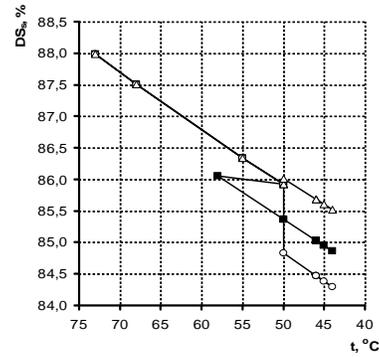
b.
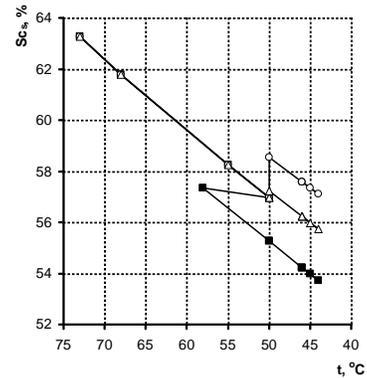
c.
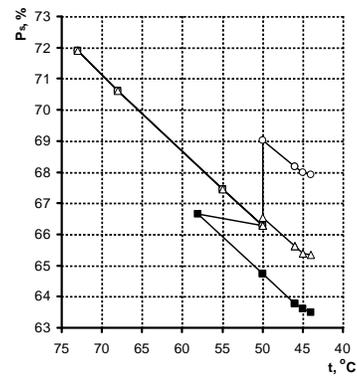
d.

Figure 4. Changing massecuite technological characteristics during the cooling crystallization process:
○ – with water dilution; △ – with impure sugar solution dilution; ■ – with an intermediate heat; a – grain content in the massecuite; b – dry solid weight ratio in the syrup; c – sucrose weight ratio in the syrup; d – syrup purity

To analyze the effectiveness of intermediate heating final massecuite in the mixer-crystallizers we carried out a series of research in which besides the addition of

Valeriy MYRONCHUK, Oxana YESHCHENKO, Maryna SAMILYK, *Sucrose cooling crystallization modelling,* Food and Environment Safety, Volume XII, Issue 2 – 2013, pag. 109 - 114

113

dilution water massecuite or molasses, massecuite intermediate heating by 5, 7, 10 and 12 °C has been used. The results suggest that the best effect is got when the temperature of the intermediate heating of massecuite in the mixer-crystallizers after cooling to 50-52 ℃ is 7-10 ℃ (Fig. 4). In this case, the viscosity of the syrup is reduced almost by half, the surface tension is also reduced. Decreases the viscosities of syrup increases the sucrose molecules diffusing from solution to the crystal surface, and reduce the surface tension increases the rate of crystallochemical reaction at the phase interface during the transition of sucrose dissolved in the crystal. Crystallization effect in this case is 8.4%. In addition, we found that if the purity of the initial massecuite decreases, the temperature of its intermediate heating must be increase.

Also a significant improvement in the sugar grains size moves up in fractions of 0.63-1.0 mm and greater than 1.0 mm

4. Conclusion

Alternative water dilutions of final massecuite in the cooling crystallization process in mixers-crystallizers is to use an massecuite intermediate heating by 8 ℃-10 ℃ after reducing its temperature by 50-52 ℃. The use of massecuite intermediate heating reduces the syrup viscosity and the surface tension at phase interface "solution" – "solid", which increases the intensification of sucrose crystallization. In this case greater molasses desugarization and better grain size of sugar crystals in the final massecuite are achieved.

5. Acknowledgments

6. References

[1] S. OUIAZZANE, B. MESSNAOUI, S. ABDERAFI, J. WOUTERS, T. BOUNAHMIDI. Estimation of sucrose crystallization kinetics from batch crystallizer data // Journal of Crystal Growth, Volume 310, Issue 4, 15 February 2008, Pages 798-803

[2] Lie-Ding SHIAU. The distribution of dislocation activities among crystals in sucrose crystallization // Chemical Engineering Science, Volume 58, Issues 23–24, December 2003, Pages 5299-5304

[3] A. FERREIRA, N. FARIA, F. ROCHA. Roughness effect on the overall growth rate of sucrose crystals // Journal of Crystal Growth, Volume 310, Issue 2, 15 January 2008, Pages 442-451

[4] Håkan GROS, Teuvo KILPIÖ, Juha NURMI. Continuous cooling crystallization from solution // Powder Technology, Volume 121, Issue 1, 5 November 2001, Pages 106-115

[5] Issam A. Khaddour, Luís S.M. Bento, António M.A. Ferreira, Fernando A.N. Rocha Kinetics and thermodynamics of sucrose crystallization from pure solution at different initial supersaturations // Surface Science, Volume 604, Issues 13–14, 15 July 2010, Pages 1208-1214

[6] Giuseppe Vaccari, Giulio Sgualdino, Elena Tamburini, Giorgio Pezzi, Piercarlo Citterio, Roberto Verardi, Krzysztof Urbaniec. New eco-friendly proposal for the crystallization of beet raw juice // Journal of Cleaner Production, Volume 13, Issue 15, December 2005, Pages 1447-1460

[7] M. QUINTAS, T.R.S. BRANDÃO, C.L.M. SILVA, R.L. CUNHA. Rheology of supersaturated sucrose solutions // Journal of Food Engineering, Volume 77, Issue 4, December 2006, Pages 844-852

[8] P.M. MARTINS, F. ROCHA. New developments on size-dependent growth applied to the crystallization of sucrose // Surface Science, Volume 601, Issue 23, 1 December 2007, Pages 5466-5472

[9] N. FARIA, S. FEYO DE AZEVEDO, F.A. ROCHA, M.N. PONS. Modelling agglomeration degree in sucrose crystallisation // Chemical Engineering and Processing: Process Intensification, Volume 47, Issues 9–10, September 2008, Pages 1666-1677

[10] C. CAMPAÑÁ CUÉ, A.R. RUIZ SALVADOR, S. AGUILERA MORALES, F.L. FALCON RODRIGUEZ, P. PÉREZ GONZÁLEZ. Raffinose–sucrose crystal interaction modelling // Journal of Crystal Growth, Volume 231, Issues 1–2, September 2001, Pages 280-289

[11] N. A. F. A. SAMAD, Ravendra SINGH, Gürkan SIN, Krist V. GERNAEY, Rafiqul GANI. A generic multi-dimensional model-based system for batch cooling crystallization processes // Computers & Chemical Engineering, Volume 35, Issue 5, 11 May 2011, Pages 828-843.