

# Model based Soft-Sensor for Industrial Crystallization: On-line Mass of Crystals and Solubility Measurement

Cédric Damour, Michel Benne, Brigitte Grondin-Perez, and Jean-Pierre Chabriat

**Abstract**—Monitoring and control of cane sugar crystallization processes depend on the stability of the supersaturation ( $\sigma$ ) state. The most widely used information to represent  $\sigma$  is the electrical conductivity  $\kappa$  of the solutions. Nevertheless, previous studies point out the shortcomings of this approach:  $\kappa$  may be regarded as inappropriate to guarantee an accurate estimation of  $\sigma$  in impure solutions. To improve the process control efficiency, additional information is necessary. The mass of crystals in the solution ( $m_c$ ) and the solubility (mass ratio of sugar to water  $m_s/m_w$ ) are relevant to complete information. Indeed,  $m_c$  inherently contains information about the mass balance and  $m_s/m_w$  contains information about the supersaturation state of the solution. The main problem is that  $m_c$  and  $m_s/m_w$  are not available on-line. In this paper, a model based soft-sensor is presented for a final crystallization stage (C sugar). Simulation results obtained on industrial data show the reliability of this approach,  $m_c$  and the crystal content ( $cc$ ) being estimated with a sufficient accuracy for achieving on-line monitoring in industry.

**Keywords**—Soft-sensor, on-line monitoring, cane sugar crystallization.

## I. INTRODUCTION

CRYSTALLIZATION is an example of particulate processes that are widely used in industry for the production of many products, including cane sugar extracted from sucrose solutions. The presence of a continuous phase and a dispersed phase gives rise to physicochemical phenomena such as nucleation, growth, agglomeration and dissolution. Specifically, growth occurs in specific conditions defined by the supersaturation state. In order to improve exhaustion, crystallization is achieved grade wise, inside vacuum boiling pans, through three stages named A, B and C. As the purity of the solutions decreases from A to C stages, the efficiency of the C-grade crystallization is of high interest, because it is the last step where sugar could be extracted.

The aim of this paper is to develop a reliable soft-sensor for on-line  $m_c$  and  $m_s/m_w$  measurement, applied to C crystallization. The rest of the paper is organized as follows.

C. Damour, M. Benne, B. Grondin-Perez & J.-P. Chabriat are with the Laboratory of Energetic, Electronics and Processes (EA 4079), University of La Reunion, France (E-mail: {cdamour | benne | bgrondin | chabriat}@univ-reunion.fr)

The industrial problem is described in section II. Section III presents the structure of the soft-sensor, based on 5 ordinary differential equations to describe heat and mass transfer with phase change. In section IV, validation is based upon industrial data. Simulation results show the reliability of this approach for achieving on-line monitoring of an industrial process.

## II. THE INDUSTRIAL PROBLEM

In cane sugar industry, this process monitoring and control is based on an electrical conductivity setpoint, the most usual approach to control the supersaturation state of solutions and keep it in the appropriate range.

The observation of C crystallization operations reveals that the electrical conductivity  $\kappa$  of impure sucrose solutions is affected by the change of concentration, material's quality and other technical conditions such as the reduced pressure in the vacuum pan. In other words,  $\kappa$  may be regarded as inappropriate to guarantee an accurate estimation of the supersaturation state in impure solutions [1].

Experimental investigations point out the shortcomings of control strategies following a conductivity setpoint. Indeed, observation of growth phase operations in solutions containing a high level of impurities reveals  $m_c$  may not increase even if  $\kappa$  tracks the desired setpoint (Fig. 1 & Fig. 2).

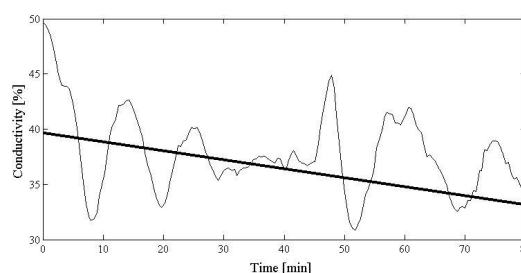


Fig. 1 Conductivity variations (setpoint in bold)

Without mention of the effect of impurities on both measurements ( $\kappa$  and  $m_c$ , same figures) and performance, the usual control strategy based on a conductivity setpoint tracking has proved to be unsatisfactory, especially in terms of global productivity (time and energy loss).

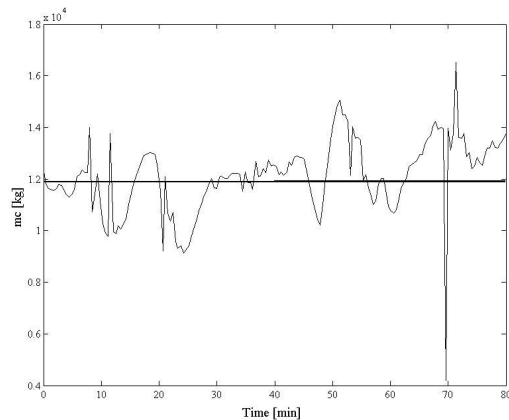


Fig. 2 Mass of crystal (rather constant)

As the crystallization performance is measured by the crystal growth, quantified by the increase in the mass of crystals,  $m_c$  appears to be a suitable performance criterion. Indeed,  $m_c$  inherently contains information about the mass balance and the crystal content ( $cc$ ). Better than the changes in  $\kappa$ , the variations in  $m_c$  tend to describe accurately the complex physicochemical reactions occurring during the phase change. Besides, the solubility ( $m_s/m_w$ ) gives information about the supersaturation state of the solution. According to the solubility diagram, the on-line measurement of the solubility can be used to ensure that the solution remains in the metastable region (Fig. 3).

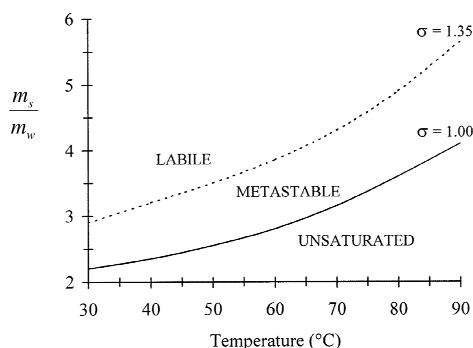


Fig. 3 Solubility diagram

The problem is that  $m_c$  and  $m_s/m_w$  are not available on-line [2]. These considerations lead to the concept of soft-sensors, first investigated a couple of years ago to control both supersaturation of liquors and crystal content ( $cc$ ) of massecuites, by measuring di-electric constant and electrical conductivity at radiofrequencies [3]. Recently, on-line applications have been successfully implemented in cane sugar industry [4, 5, 6], which allows to consider alternative strategies for monitoring and process control of industrial crystallization processes.

### III. MODEL BASED SOFT SENSOR FOR MONITORING AND CONTROL

This section considers the development of a model based soft-sensor for  $m_c$  and  $m_s/m_w$  estimation.

#### A. Crystal Growth Modeling: The Case of Impure Solutions

In cane sugar industry, to increase performance with minimum energy consumption, crystallization is achieved in three steps, named A-grade, B-grade and C-grade:

- crystallization of high purity liquors allows the extraction of A sugar from A molasses;
- crystallization of A molasses allows the extraction of B sugar from B molasses;
- and crystallization of B molasses allows the extraction of C sugar from C molasses, called blackstrap.

Since it is important to maximize the yield at each stage of the process, the fact remains that a key step in sugar recovery is C-grade crystallization (no more sugar can be extracted from blackstrap molasses in a cost-effective fashion). Consequently, global quality and productivity are very dependent on an efficient control of the C-grade crystallization.

The most widely approaches investigated to describe sugar extraction take into account three physical-chemical phenomena: nucleation, growth and dissolution. During the last 10 years, these investigations led to the development of models of A-grade processes (high purity liquors).

Little information is available about C-grade in the industrial context. There, low-grade sucrose solutions (impure solutions) contain a high proportion of impurities, which considerably changes their properties. It makes it difficult to adapt A-grade models to C-grade modeling. To overcome this difficulty, the most usual approach consists of considering nucleation and crystal agglomeration negligible in super-saturated conditions [7, 8].

In other words, crystal growth is assumed to be preponderant, and this assumption underlies the development of a model dedicated to process control simulation.

#### B. Model based Soft Sensor

Sugar crystallization consists in the exhaustion of a liquor to produce crystals. Extraction is induced by seeding the liquor in vacuum pans. During this operation, both the liquor and growing crystals are mixed together to obtain a homogeneous supersaturated magma. Super-saturation of the magma is obtained by vacuum evaporation.

The three steps process is performed through a series of continuous and semi batch vacuum pans. At each step, several semi batch vacuum pans, operating as continuously stirred tank reactors (CSTR, Fig. 4), are fed with different purity liquors.

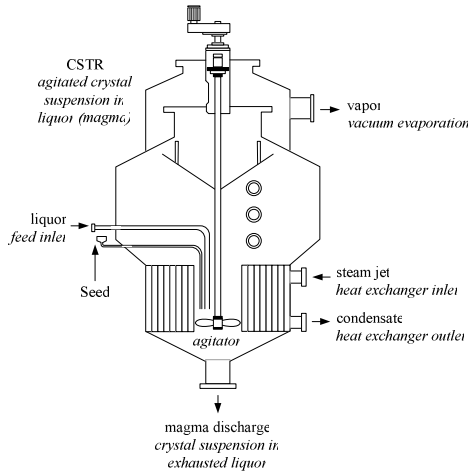


Fig. 4 CSTR used for sugar crystallization

Because of the impact of many variables in terms of quality (liquors, molasses, heating steam, vacuum), it is important to develop a mathematical model that incorporates enough knowledge to describe the involved heat and mass transfer (HMT). A wide variety of mathematical models can be found in the scientific literature to describe these HMT, both steady state [9] and dynamic models [10, 11].

The main difficulty consists in the estimation of the crystallization rate, usually calculated by solving the population balance represented by moment equations [12, 13]. Under the aforementioned assumptions, a dynamic model can be derived from five ordinary differential equations [14]:

$$\frac{dm_w}{dt} = \rho_f F_f \left( 1 - \frac{Bx_f}{100} \right) + \rho_w F_w - \dot{m}_{vap} \quad (1)$$

$$\frac{dm_i}{dt} = \rho_f F_f \frac{Bx_f}{100} \left( 1 - \frac{Pte_f}{100} \right) \quad (2)$$

$$\frac{dm_s}{dt} = \rho_f F_f \frac{Bx_f}{100} \frac{Pte_f}{100} - \frac{dm_c}{dt} \quad (3)$$

$$\frac{dm_c}{dt} = cc \left( \rho_f F_f - \dot{m}_{vap} \right) + \alpha_{Crys} \quad (4)$$

$$\frac{dT_{mg}}{dt} = \frac{1}{m_{mg} C_{p_{mg}}} \left( \dot{W} + \dot{Q} + \rho_f F_f (h_f - h_{ml}) + \rho_w F_w (h_w - h_{ml}) - \dot{m}_{vap} L_{vap} + \frac{dm_c}{dt} L_c \right) \quad (5)$$

with  $\dot{m}_{vap} = \rho_{cw} F_{vap}$ : mass flow rate of emitted vapor;

$\dot{Q} = \rho_{hs} F_{hs} L_{cw}$ : heating power brought by heating steam condensation;

and  $cc = m_c / m_{mg}$ : crystal content, ratio of  $m_c$  to the total mass.

## NOMENCLATURE

## Thermodynamic variables

$Bx\%$	Brix (mass fraction of dry substance)	%
$cc$	crystal content	%
$C_p$	specific heat capacity	$J.K^{-1}.kg^{-1}$
$F$	flow rate	$m^3.s^{-1}$
$h$	specific enthalpy	$J.kg^{-1}$
$L$	specific latent heat	$J.kg^{-1}$
$m$	mass	$kg$
$\dot{m}$	mass flow rate	$kg.s^{-1}$
$Pte\%$	purity (mass fraction of sugar)	%
$\dot{Q}$	heating power	$W$
$T$	temperature	$^{\circ}C$
$\dot{W}$	stirring power	$W$
$\rho$	density	$kg.m^{-3}$
$\alpha$	adjusted parameter	

## Subscripts

$i$	impurities
$c, Crys$	crystal
$s$	dissolved sucrose
$w$	water
$cw$	condensate (from heating steam)
$f$	feed
$hs$	heating steam
$mc$	mass of crystals
$ml$	mother liquor
$mg$	magma (mother liquor & crystals)
$vap$	emitted vapor

## IV. IDENTIFICATION AND EXPERIMENTAL VALIDATION

Taking into account the aforementioned hypothesis and further assumptions based on experimental considerations, two thermo-dynamic equilibrium relationships complete the description of HMT during the growth phase:

- the time delay between the heating steam ( $hs$ ) supply and the condensed water ( $cw$ ) outlet is inconsiderable (with regard to the process kinetics); the heating power is calculated using the measured flow rate of the condensate:

$$F_{hs} = \alpha_Q F_{cw \leftarrow hs} \quad (6)$$

- to evaporate 1 kg of water about 1 kg of heating steam is needed; the evaporation capacity ( $F_{vap}$ ) is calculated using:

$$F_{vap} = \alpha_{vap} F_{cw \leftarrow hs} \quad (7)$$

The model identification has been performed using 4 databases randomly selected from one year industrial

measurements. Three parameters had to be estimated to make the model fit the data:  $\alpha_{Crys}$ ,  $\alpha_Q$  &  $\alpha_{vap}$  (cf. eq. 4, 6 and 7). An iterative fitting procedure has been implemented. The algorithm is based on the minimization of a 3D quadratic criterion expressing the mean squared difference between simulated and experimental variables.

In the present approach, the optimization led to the following adjusted parameters:

$$[\alpha_Q = 1.8177; \alpha_{vap} = 1.6455; \alpha_{Crys} = 0.9217]$$

The validation step is performed using industrial data obtained using two on-line Brix measurements ( $Bx_{mg}^{\%}$  given by a microwave sensor and  $Bx_{ml}^{\%}$  given by a refractometer). Prior to each run, the initial conditions for the state variables are determined from Brix, purity and level off-line measurements at time  $t = 0$  s.

Fig. 5 & Fig. 6 show simulated versus measured results for  $m_c$  (database 1 & 2).

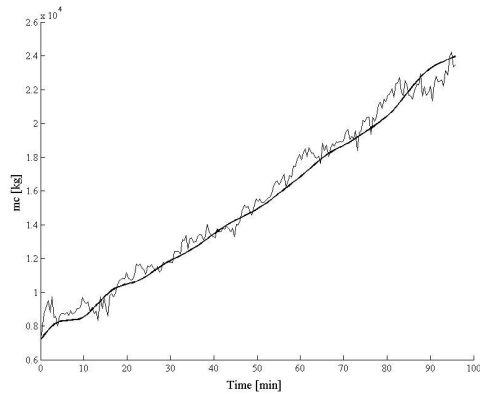


Fig. 5 Simulated  $m_c$  (bold) compared with industrial measurements (database 1)

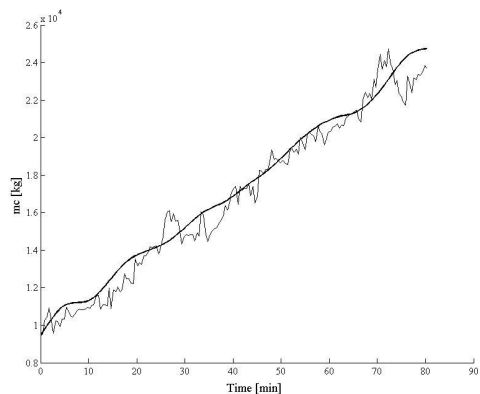


Fig. 6 Simulated  $m_c$  (bold) compared with industrial measurements (database 2)

Fig. 7 shows simulated versus measured results for  $cc$ .

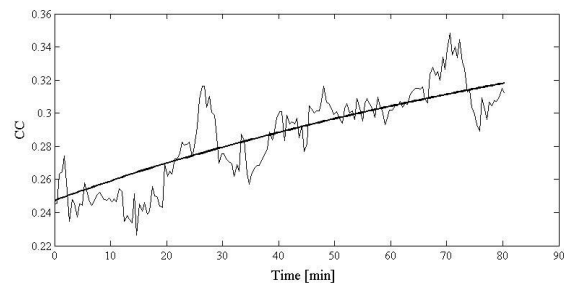


Fig. 7 Simulated  $cc$  (bold) compared with industrial measurements (database 2)

In comparison with industrial data, the simulator has proved to be able to calculate  $m_c$  and  $cc$  with a relative error less than 4 % and 0.8 %, respectively.

Fig. 8 shows the evolution of the solubility of the solution during a crystallization operation at 65 °C.

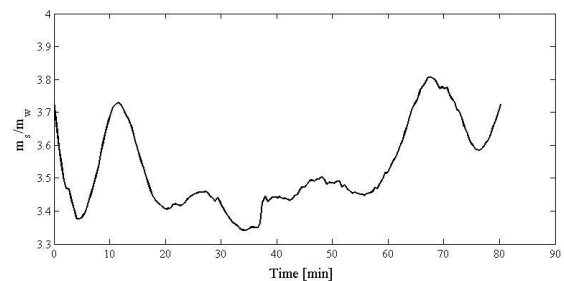


Fig. 8 Simulated  $m_s / m_w$  at 65 °C (database 2)

According to the solubility diagram (Fig. 3), the evolution of the experimental solubility shows that the solution remains in the metastable zone during all the crystallization.

The accuracy of the simulated  $m_c$ ,  $cc$  and  $m_s / m_w$  is sufficient for achieving soft-sensor objectives, without using on-line Brix measurements (given by microwave or refractometric sensors), the software being initialized with offline measurements.

## V. CONCLUSION

In this paper, a model based soft-sensor for on-line measurement of the mass of crystals, the crystal content and the solubility have been developed and applied to an industrial C crystallization process. Simulation results show the efficiency of this approach for achieving monitoring tasks. Associated with the widely used electrical conductivity  $\kappa$ ,  $m_c$  and appears to be a suitable performance criterion, carrying additional information about the mass balance and the crystal content ( $cc$ ) and  $m_s / m_w$  carrying additional information about the supersaturation state of the solution. In terms of process control, because of the large number of parameters and the multiple interactions between each other, the traditional strategies have proved to be unsatisfactory [15]. These results and the relevant experimental observations allow drawing

some perspectives about the use of  $m_c$  and  $cc$  for achieving alternative process control strategies.

#### ACKNOWLEDGMENT

The authors gratefully acknowledge the help of Mr. Jean-Claude Pony, Dir. of the BR sugar mill ([www.bois-rouge.fr](http://www.bois-rouge.fr)), and its technical staff, and Mr. Jean-Paul Dijoux, networks engineer ([www.cerf.re](http://www.cerf.re)).

#### REFERENCES

- [1] Grondin-Perez B., M. Benne, C. Bonnecaze & J.-P. Chabriot, 2005, Industrial multi-step forward predictor of mother liquor purity of the final stage of a cane sugar crystallization plant, *J. of Food Engineering*, 66, pp. 361-367
- [2] Grondin-Perez B., M. Benne & J.-P. Chabriot, 2006, Supervision of C crystallization in Bois Rouge sugar mill using on-line crystal content estimation using synchronous microwave and refractometric Brix measurements, *J. of Food Engineering*, 76, pp. 639-645
- [3] Radford D. J., 1985, EP0162580: massecuite supersaturation monitor, TONGAAT HULETT GROUP LIMITED (ZA), Nov. 1985, <http://www.freepatentsonline.com/EP0162580A1.html>
- [4] Devogelaere D., M. Rijckaert, O.G. Leon & G.C. Lemus, 2002, Application of feed forward neural networks for soft sensors in the sugar industry, VIIth Brazilian Symp. on Neural Networks, pp. 2-6
- [5] Bakir T., S. Othman, F. Puel & H. Hammouri, 2005, Continuous-discrete observer for crystal size distribution of batch crystallization process, 44th IEEE Conf. on Decision and Control 2005 & 2005 European Control Conf.), 12-15 Dec. 2005, pp. 6240-6244
- [6] Simoglou A., P. Georgieva, E. B. Martin, A. J. Morris & S. Feyo de Azevedo, 2005, On-line monitoring of a sugar crystallization process, *Computers & Chemical Engineering*, Vol. 29, Issue 6, pp. 1411-1422
- [7] Pautrat C., J. Génotelle & M. Mathlouthi, 1997, Sucrose crystal growth: effect of supersaturation, size and macromolecular impurities, in *Sucrose crystallization, science and technology*, VanHook et al., Bartens, pp. 57-70
- [8] Barth S., 2006, Utilization of FBRM in the Control of CSD in a Batch Cooled Crystallizer, PhD thesis, Georgia Institute of Technology, 121 p.
- [9] Semlali Aouragh Hassani N., K. Saidi & T. Bounahmidi, 2001, Steady state modeling and simulation of an industrial sugar continuous crystallizer, *Computers & Chemical Engineering*, Vol. 25, Issues 9-10, pp. 1351-1370
- [10] Feyo de Azevedo S., J. Chorão, M. J. Gonçalves & L. Bento, 1993, On-line monitoring of white sugar crystallization through software sensors. Part I, *International Sugar J.*, 95, pp. 483-488
- [11] Feyo de Azevedo S., J. Chorão, M. J. Gonçalves & L. Bento, 1994, On-line monitoring of white sugar crystallization through software sensors. Part II, *International Sugar J.*, 96, pp. 18-26
- [12] Hulburt H. M. & S. Katz, 1964, Some problems in particle technology. A statistical mechanical formulation, *Chemical Engineering Science*, Vol. 19, pp. 555-574
- [13] Ramkrishna D., 1985, The status of population balances, *Reviews in chemical engineering*, 3 (1), pp. 49-95
- [14] Benne M., B. Grondin-Perez & J.-P. Chabriot, 2008, Estimation of two parameters to fit a tendency model for dynamic simulation of an industrial crystallization process, *FoodSim 08*, 26-28 June 2008, University College Dublin, Ireland
- [15] Beyou S., 2008, Proposition d'un algorithme PID à paramètres variables pour l'amélioration de la conduite d'un procédé de cristallisation industriel, PhD thesis, University of La Réunion (EA 4079), 181 p.