Advances in Super-Saturation Measurement and Estimation Methods for Sugar Crystallisation Process

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Abstract—The super-saturation level of the massecuite is an important quality parameter for sugar crystallisation process, as it determines the seeding point, contributes to the quality of crystals and the cost of production. This paper critically appraises the current measurement and estimation methods for super-saturation level of sugar massecuite. On the one hand, the review shows that the current online hardware sensors lack the necessary accuracy, as the variable to be measured is a multivariable function of many unknowns. Moreover, the sensors require regular maintenance and recalibration to be able to obtain reliable readings. On the other hand, the review shows that soft (model or software-based) sensors are capable of offering solutions to some of the challenges of the online hardware sensors. However, their predictions depend on the hardware sensors for some input data and the available sugar crystallisation models are not in the form suitable for online estimation of super-saturation level of the sugar massecuite. It is concluded that the effective measurement/estimation and control of super-saturation of sugar massecuite is still a challenge in the sugar processing industry. It is therefore recommended that soft sensors should be introduced to complement the online hardware sensors.

Index Terms—sugar, crystallisation, super-saturation, soft sensor, hardware sensor

I. INTRODUCTION

Sugar factory is classified into raw sugar factory and the refined sugar factory where further processing such as purification and decolourisation are carried out on the raw sugar [1], [2]. There are two main sources of sugar production: sugar cane and sugar beet. Cane sugar is mainly produced in the developing countries; it accounts for 60-65% of crystal sugar production while the remaining being the share of the beet sugar produced in industrialized countries [1], [3]. The raw materials are harvested, transported to the factory and weighed on a platform weighing scale. The raw materials can be stored for several weeks if harvesting disruption occurs. However, US EPA [2] reported that "when the cane is cut, rapid deterioration of the cane begins. Therefore, unlike sugar beets, sugarcane cannot be stored for later processing without excessive deterioration of the sucrose

content". The raw materials are washed and the surface area increased through a milling process, breaking its hard structure and grinding it. The tissues of the raw materials are disintegrated and the juice removed by compression and squeezing either by pressure between rollers of mills, extraction by lixiviation or leaching in a diffuser [4]. The raw juice is turbid and contains undesirable impurities which have to be eliminated so that the concentrated clear juice or syrup is suitable for sucrose crystallisation in the boiling strikes from which white sugar is produced. The juice is clarified by adding calcium hydroxide (lime) to neutralize the organic acid in the liquor; the mixture is heated and a heavy precipitate is observed [1]. This precipitate is separated from the juice by centrifugation. Kulkarni [3] observed that the clear juice contains about 83% to 85% of water. When the clear juice is evaporated in a typical evaporator station in a sugar plant, syrup with about 65% solids and 35% water is produced. The evaporation stage is to prepare the juice for crystallisation. Crystallisation involves growing sugar crystals under controlled conditions. The massecuite discharged from the crystallisation stage undergoes centrifugation, where the refined sugar is separated from the (mother) liquor and dried. The process of sugar manufacture essentially consists in isolating sucrose crystals from water as well as the impurities. Therefore, crystallisation of sugar is the heart of sugar production [3].

Super-saturation is the driving force for crystallisation. Kulkarni [3] pointed out that sugar crystal growth in a super-saturated sugar solution takes place as a result of the transfer of sucrose molecules from the bulk of solution to the surface of the crystal by diffusion. He stated further that super-saturation is the main driving force which increases the rate of diffusion of sucrose molecules to the crystal surface and thus the growth of crystals is determined mainly by the degree of supersaturation of the sugar solution. Increase in supersaturation increases the rate of crystallization. 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

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liquor during centrifuging from the multiple crystals is less than perfect [5]. There is a growing awareness in the sugar industry regarding the importance of product quality and the cost of production. Both are closely related to the sensors and the control methods used during sugar crystallisation process. Therefore, the aim of this paper is to critically review the current measurement and estimation methods of super-saturation for sugar massecuite. The findings are expected to aid in the decision making to improve super-saturation measurement and control during sugar crystallisation.

II. PROCESS MEASUREMENT AND CONTROL WITH HARDWARE SENSORS IN SUGAR PROCESSING PLANT

The sensors currently in use for measurement in crystallisation process measure one or two parameters of the syrup or massecuite, which are refractive index, viscosity or consistency, attenuation of nuclear radiation, attenuation and phase shift of microwave radiation, boiling point rise, or electrical parameters. Fig. 1 is a flow diagram of a typical sugar manufacturing plant where online refractometers are utilised in measurement of sucrose concentration in sugar production process. The subsequent sections discuss various hardware sensors being used in sugar crystallization unit.

A. Sensors That Measure Electrical Parameters

The electrical parameters that could be measured include resistance, conductance and capacitance. Transducers which measure the electrical properties of a massecuite include conductivity probes, resistance probe and microwave devices [6]. Rozsa [5] observed that measurement of any of these electrical properties is strongly affected by the changes in composition of the non-sugars (ash content) which are unknown. Hence, the value of super-saturation measured from this sensor is not reliable.

Radio Frequancy (RF) probe: The RF or resistance probe consists of a tuning circuit with a radio frequency in the range 10 to 45MHz. This is connected with a Teflon-coated probe in the vacuum pan. The probe produces a signal proportional to series resistance with the value of resistance being ranged to represent dielectric loss of the massecuite which is proportional to both the Brix and crystal content of the massecuite [6], [7]. According to Taylor and Getaz [6], the monotrac RF probe is cost-saving since it does not require the complication of microprocessor; it is simple to operate and calibrate, not susceptible to scaling and can tolerate some level of encrustation without their reading being affected as a result of its high frequency signal. However, they noted that the monotrac probe can only be reliably used on massecuites with high impurity; thus, it cannot be used for refinery white sugar massecuites. Successful advances were made to develop duotrac RF probe to measure high quality massecuite making use of microprocessor [7], [8]. However, Taylor and Getaz [6] reported that the duotrac RF probe was largely replaced by microwave probes due to measurement inaccuracy.

Microwave-Based sensors: Microwaves are electromagnetic waves. The microwave devices used for measurement in the sugar industry have a frequency of approximately 2.5GHz [9]. Ulrich [9] explained that microwave from a transmitter irradiate the product as a wave and are detected by a receiver. The sugar solutions are polarised at varying strengths during microwave transmission, resulting in the microwave signal, losing speed and energy. This gives a correlation between water content and dry substance that enables a precise measurement of the concentration, brix and density in all stages of sugar production. He pointed out that the major advantage of microwave measurement technology is that it measures both in the solution phase up to the seeding point and in the magma phase up to product discharge. However, Rozsa [5] noted that one can rarely find data on the specified accuracy of microwave measurement and that they are a relatively new entry to the field with reported conflicting experiences on their use. In addition, it is susceptible to scaling and cannot tolerate some level of encrustation; hence, it requires frequent removal for cleaning as gradual accumulation of sugar encrustation on the sensing tip of the probes which unless periodically removed and cleaned will affect the probe reading [6].



Figure 1. Flow diagram of a sugar refining plant where online refractometers are in use [10].

Conductivity probe: Conductivity probes measure the electrical properties of sugar solution at low frequency between 1kHz and 2kHz [6].Conductivity is the ability of a solution to pass an electric current. The amount of current flowing is roughly proportional to the number and mobility of the ions present in the solution as a result of increased crystal content [11]. Thus, the conductivity of a massecuite is dependent on the mother liquor concentration. Conductivity probe has been widely used for measurement in vacuum pan boiling B-massecuite and C-massecuite where the ash content is sufficiently high and comparatively constant. Taylor and Getaz [6] enumerated the limitations of conductivity probe to include its frequent recalibration due to unpredicted changes in ash content in the massecuite, susceptibility to scaling and encrustation because of its low frequency signal used for measurement. Moreover, the probes corrode due to electrolytic action, and it cannot be

applied in the measurement of high purity massecuite such as A-massecuite and white sugar massecuite.

B. Viscosity- or Consistency-Based Sensor

The basic principle of consistency probes operation is to measure the power consumption of a small electric motor driving a propeller intruding into the pan. Viscosity of solution is closely correlated to their concentration. In the boiling pan, as seeding continues, the crystal content increases, and thereby leading to a corresponding increase in viscosity. Since viscosity has a retarding effect on the mobility of ions in solution [11], it is therefore natural to try to use viscosity data to give some information on super-saturation. Kulkarni [3] reported that in India, process measurement in sugar crystallisation using sensor that measures viscosity is widely used. Kulkarni [3] reported that in India, process measurement in sugar crystallisation using sensor that measures viscosity is widely used. However, the viscosity of molasses is reduced by 50% by raising temperature by $5 \, \mathbb{C}$ and that for a particular massecuite, the higher the viscosity the lower will be the purity of the molasses, as certain impurities like gums, dextran and other colloidal matters contribute to viscosity. Therefore, Rozsa [5] concluded that though viscosity is correlated to liquid concentration and therefore to super-saturation, viscosity is a multivariable function and accurate measurement of super-saturation cannot be expected from it.

C. Nuclear Radiation-Based Sensors

Usually, attenuation of nuclear radiation is translated into density or solids content of the massecuite. Density data can provide reliable and accurate data on the concentration of the solution only until there are no crystals present; that is, up to seeding of the pan. Having completed seeding, density depends on the concentration of the mother liquor and the crystal content. This indicates that any type of density measurement, if accurate enough, can be used only to determine the seeding point, but as the contribution from the crystals increases, density data will become increasingly useless for the monitoring of super-saturation. Another major cause for concern is the possibility of using radioactive material for terrorist purposes. There are already quite a few countries, where their use is already banned. No wonder, Managements of sugar plants all over the world are in a rush to replace their nuclear density probes with different types of probes [5].

D. Sensors Based on the Boiling Point Rise (BPR) of Solutions

Boiling point rise is the rise in boiling temperature of concentrated syrup or molasses over that of pure water at the same pressure or vacuum. In a boiling solution, the BPR is given by the difference in the temperature of the boiling solution and the vapors leaving the solution. This boiling point rise is proportional to the concentration of the solution. Therefore, it could be used to provide an online estimate of sugar super-saturation [3], [5]. Rozsa [5] gives reasons why devices for monitoring supersaturation based on BPR do not find wide acceptance. BPR depends not only on the concentration of the syrup or mother liquor, but also on its purity and on the composition of its non-sugars which are difficult, or even impossible to compensate for, and that BPR of highpurity syrups is rather small, which makes its accurate measurement very difficult.

E. Sensors Based on Refractive Indexes of Solutions

The refractive indexes of solutions depend on their concentration and temperature. According to Rozsa [5], refractometers measure refractive index by using a single photo element to provide an analogue signal (voltage) proportional to the illumination of the optical image. He observed that the coefficient of determination between the experimentally determined super-saturation and the online refractometer reading as 0.952 while for RF capacitance and resistance probe prediction of supersaturation, the coefficients of determination were 0.6015 and 0.1822, respectively. However, Rozsa [5] results conflict with Radford and Cox [8] observations in which the coefficient of determination for the predicted supersaturation using the RF capacitance and RF resistance probe were reported as 0.93 and 0.87, respectively. Though Rozsa [5] insisted that the most successful and popular transducers are those which measure the refractive index of a massecuite, Taylor and Getaz [6] recommended those that measures the electrical properties of massecuite. Thus, the issue of which online measuring probe is the best is not established.

III. PROCESS MEASUREMENT AND CONTROL WITH SOFT SENSOR IN SUGAR PROCESSING PLANT

The term, 'soft sensor,' is a combination of the words, software and sensor. Software is used because the model is usually a computer program and sensor is used because the model is delivering similar information as its hardware counterpart. Other common terms for soft sensors in the process industry are predictive sensors, inferential sensors, virtual online analysers and observerbased sensors [12]. According to Huang [13], soft sensor is a mathematical model, correlating difficult-to-measure quality variables with the frequently and easily measured process variables. It can also be defined as, 'a model which estimates unmeasurable process states based on easy-to-measure input and output variables' [14]. There are basically two types of models for soft sensor development namely, the first principle (theoretical or fundamental) models and the data-based models also known as data-driven or empirical models [14], [15]. Huang [13] included a third model, the hybrid model or grey-box model, which combines the strength of both the first principle and data-driven models.

A. First Principle Models

The first principle models are based on phenomenological equations, obtained from fundamental process knowledge. It captures the physical behavior of the process and as such it has the potential to extrapolate beyond the regions for which the model was constructed [16]. The first principle modeling approach was applied by Georgieva and Co-workers [17] to a fed batch evaporative sugar crystallisation process. The model consists of three parts. The first part includes mass balances of water, impurities, dissolved sucrose and mass of crystals derived from five ordinary differential equations. The second part of the model is the energy balance which incorporates the enthalpy terms and specific heat capacities derived as a function of physical and thermodynamic properties. The third part of the model is the population balance expressed by the leading moment of crystal size distribution in the volume coordinates. The population balance defined the kinetic mechanisms of nucleation, crystal growth and particle agglomeration. In addition, Adrian [18] also developed a theoretical model for calculation of the development of the crystal size distribution in a batch crystalliser. The model was based on moment equation, to derive the kinetic equation for nucleation and growth rate needed for crystal development in a batch crystalliser. The challenges with these models are that they contain variables that are difficult to measure online and also consist of several nonlinear algebraic-differential equations which must be solved numerically. None of them is able to explicitly account for super-saturation level of sugar massecuite. Hence, they are not in the forms that are suitable for online estimation of the masecuite super-saturation.

B. Data-Based Models

Data-based model (data-driven or empirical model) is also called the black-box model. Tian-hong [19] prefers data-driven model since the cost of data acquisition and storage has continuously decreased owing to advances in information technology. Many process variables can be easily measured, stored and accessed in real time from distributed control systems. Huang [13] pointed out that poor instrument readings, missing data, outliers and noise during signal transmission in the measuring instrument may affect the quality of the historical data; hence, the performance of the developed model. A classical empirical model for sugar crystal growth rate (RG) was developed by Lauret and Co-workers [20]. The constants used in the model include the overall growth rate constant and the growth rate order. The parameters values in the model were obtained by applying nonlinear programing technique to the data in the sugar crystalliser. Lauret and Co-workers [20] concluded that this model structure overcame the problem of convergence (slow and difficult to obtain numerical solution) but does not resolve the problem of accuracy outside the region where the parameters were fitted. In addition, RG is a function of super-saturation and crystal content and the accurate online measurement of these variables is still an issue. Hence, the accuracy of the sugar crystal growth rate model developed by Lauret and Co-workers [20] depend on the accuracy of crystal content and super-saturation measured by the existing instruments. Umo and Alabi [21] developed a predictive model to estimate the supersaturation of massecuite based on mass balance in a sugar crystallization unit. The input variables were easy to measure online. They are mass composition of sucrose in

the feed, the feed flow rate and distillate rate. Performance evaluation of the model indicated that it is capable of providing accurate estimates of the massecuite super-saturation values. However, the model is limited to the prediction of pre-seeding super-saturation and its performance on real system is yet to be validated.

C. Hybrid Model

Hybrid model (grey-box model) is also known as Knowledge-Based Hybrid Modeling (KBHM). According to Azevedo [16], the idea of KBHM is to complement the analytical model with the data-driven approach. In the development of such models, it is possible to combine theoretical and experimental knowledge as well as information from different sources: theoretical knowledge from physical and mass conservation laws, experimental data from laboratory experiments, experimental data from real plant experiments, data from regular process operation and knowledge and experience from qualified process operators. The clear advantages of KBHM over the data-based modeling are that there is more physical transparency of the model parameters and less training data is required. Cédric and Co-workers [22] applied an empirical representation for crystallisation rate with mass and energy balances to obtain the mass of crystals and crystal content. The maximum relative errors associated with the predictions of the mass of crystal and crystal content were reported as 4% and 0.8%, respectively. Georgieva and Co-workers [17] carried out a research to compare the three modeling approaches applied to a fedbatch evaporative sugar crystallisation process to predict crystal size and crystal size variation of sugar. Their findings showed that the hybrid modeling approach outperformed other approaches. The hybrid model is a combination of first principle modeling method based on mass, energy and population balances, with Artificial Neural Networks (ANN) to approximate the crystal growth rate, nucleation rate and the agglomeration kernel. However, Georgieva and his Co-workers [17] observed that the hybrid model was considerably complex, required sophisticated software tools and numerical solution. In addition, it contains variables that are not easily measured online. Hence, it cannot be applied online in its current form. Moreover, the focus of the model was not for explicit estimation of super-saturation of sugar massecuite.

IV. CONCLUSIONS

The super-saturation level of the massecuite is an important quality parameter during sugar crystallisation process, as it determines the seeding point, contributes to the quality of crystals and the cost of production. In this paper, the current measurement and estimation methods for super-saturation level of sugar massecuite were reviewed. The existing online hardware sensors were found to lack the necessary accuracy, as the variable to be measured is a multivariable function of many unknowns. Moreover, the sensors require regular maintenance and recalibration to be able to obtain reliable readings. The review shows further that soft (model or software-based) sensors are capable of offering solutions to some of the challenges of the online hardware sensors. However, their predictions depend on the hardware sensors for some input data. Moreover, the available sugar crystallisation models are not in the form suitable for online estimation of super-saturation level of the sugar massecuite. The study therefore concludes that the effective measurement/estimation and control of super-saturation of sugar massecuite is still a challenge in the sugar processing industry. It is therefore recommended that the instrument manufacturers for sugar industry should strive to develop more accurate and reliable hardware-based super-saturation sensors for effective super-saturation measurement and control of sugar crystallisation unit. Moreover, soft sensor should be introduced to complement the online hardware sensors.

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