POSTER SUMMARY

THE USE OF ULTRASOUND FOR ON-LINE CRYSTAL SIZE DISTRIBUTION (CSD) ANALYSES

SCHOONEES-MUIR BM and GOOCH MA

Sugar Milling Research Institute, c/o University of KwaZulu-Natal, Durban 4041, South Africa
bmuir@smri.org

Abstract

The use of ultrasound to determine the solids content and particle size distribution of a suspension of solid particles (such as crystals) in a liquid medium has specific advantages over other techniques. The measurement is not affected by vibrations or changes in the flow velocity and can be applied under extreme pH, pressure and abrasive conditions. The technique is non-destructive and non-invasive, there is no sample preparation and measurements are quick. The instruments that are commercially available can handle high viscosity and opaque and dark suspensions.

In general, the measurement occurs in two steps: the actual measurement using a range of frequencies and the interpretation of the results using mathematical modelling. The crystal size distribution (CSD) can then be calculated in terms of the solids content or density distribution. Parameters such as the mean crystal size and the percentage of crystals by mass that falls within certain cumulative percentage fractions in the distribution can be determined.

Samples of A- and C-massecuite were analysed using one of the commercially available instruments. While results did not correlate to those obtained using the conventional Sugar Milling Research Institute CSD analysis, the potential of using this technique in the sugar industry is unmistakable. Further work is needed to develop the equations specifically for a massecuite environment. However, the cost of the instruments may discourage its application.

Keywords: sugar crystals, crystal size distribution, analyses, ultrasound, massecuite, laboratory

Introduction

Crystallisation is used in the sugar industry at the end of the raw sugar manufacturing process for purification and recovery of sucrose from the mother liquor. This is done through an evaporative crystallisation process under vacuum using slurry seeding. One of the parameters determining the quality of the final product is the size distribution of the forming crystals. In general, a narrow crystal size distribution (CSD) is considered more valuable than a broad CSD, as it has bearing on the centrifugation recovery, storage and impurity characteristics, as well as the sugar's processability in the refinery. One of the main aims of the pan boiling control system is to obtain an acceptable CSD.

Analysis of the average crystal sizes and CSD of the crystals in a massecuite in the vacuum pans of a typical cane sugar factory is still being done off-line and only on a weekly basis by the Sugar Milling Research Institute (SMRI) laboratory. The visual technique requires the preparation of a 50% (m/m) suspension in glycerol, which is spread thinly on a microscope slide. A software program that was developed locally is then used to select the correct crystals

and to measure the b-axis and c-axis of each crystal (assuming the a- and b-axes to be equal). Air bubbles are eliminated by discrimination on aspect ratios, and crystals touching each other are separated by contracting or dilation processes. Histograms and various statistics are then produced (Lionnet, 1999).

Factory staff working on the pan floor have overcome any delay in CSD results from the SMRI by developing an ability to determine the correct crystal sizes by visual observation and touch when striking the pans. Many factories take microscope pictures of the crystals for group evaluation to eliminate individual bias.

Over the years, many techniques have been considered in an attempt to standardise and automate a CSD analysis for control during pan boiling. Direct image analysis (Dalziel *et al*, 1999) was used by Ingram and Steindl (2001) to develop an at-line CSD system. Working on a similar technique, Wagner *et al* (1999) actually commercialised a system through Schmidt and Haensch (Germany) in late 2000. Other off-line techniques that have been investigated include tomography (Jones *et al*, 2000), NIR spectroscopy (Schäffler, 1999), laser diffraction (Miller and Beath, 2000), and the use of commercial size distribution instruments calibrated for spherical particles (Iswanto *et al*, 2007). In terms of real time imaging Mhlongo and Alport (2002) demonstrated the potential use of neural networks on images of crystals splashing onto a port window of a crystallisation pan captured by a closed circuit video camera, or from a crystalloscope immersed in a sugar solution. None of these techniques have yet found commercial application in the South African sugar industry.

The use of ultrasound for CSD analysis is used in the pharmaceutical and other chemical industries. This approach has specific advantages over other techniques, which include:

- the measurement is not affected by vibrations or changes in the flow velocity and can be applied under extreme pH, pressure and abrasive conditions (Neidhardt and Behrens, 2004),
- the technique can be applied on-line and in-line since there is no sample preparation and measurements are quick (typically 30 seconds) (Hartman and Behrens, 2006),
- instruments can handle high viscosity and opaque and dark suspensions; the material of construction can be adapted to the specific operating conditions (e.g. where resistance is required) (Pankewitz and Behrens, 2002), and
- the measurement is non-destructive and non-invasive (McClements, 2000).

One particular commercial instrument developed by Sympatec GmbH is described by a number of authors (Pankewitz and Behrens, 2002; Geers and Witt, 2003; Neidhardt and Behrens, 2004; Hartman and Behrens, 2006). This unit sends a range of 31 frequencies from a transducer to a receiver over a fixed path length in a sample in order to divide the particle sizes into 31 classes.

The attenuation coefficient (α) is determined using the following formula:

$$\alpha = \frac{1}{x} \ln \frac{A_x}{A_0}$$

where $x \equiv$ distance travelled
$A_0 \equiv$ initial amplitude
$A_x \equiv$ amplitude after distance x

The attenuation coefficient in a suspension containing solid particles is dependent on the frequency of the ultrasonic wave. The change in attenuation is due to a number of mechanisms and is mainly dependent on the composition of the suspension and the size of the particles.

Ultrasonic attenuation spectra are converted into particle size distributions using an appropriate theory, based on a mathematical treatment of the physical processes present during the ultrasound interaction with the particles suspended in the fluid. The particle size distribution is determined by finding the size distribution that gives the best fit between the predictions of the ultrasonic scattering theory and experimental ultrasound spectra. This has proved to give highly accurate data in on-line crystallisation systems (McClements, 2000).

Experimental

One sample each of A-massecuite and C-massecuite were sent to Germany for evaluation on the Sympatec online CSD instrument. These two samples represented typical minimum and maximum size ranges encountered in the cane sugar industry and the tests were intended as an initial screening to establish whether this instrument could obtain usable data.

Each sample was first analysed at room temperature. The samples were then heated to 60°C and analysed. The samples were then diluted at a 1:1 ratio with a supersaturated refined sugar solution and again analysed at 60°C.

The samples were also analysed on the SMRI CSD system as described by Jullienne (1985) and semi-automated in 1999 (Lionnet, 2001).

For comparison, the mean aperture (MA) and coefficient of variation (CV) results for both the ultrasound data and the SMRI data were calculated (personal communication[1]) on an Excel spreadsheet using the Butler method (Anon, 2007).

Results

The samples at room temperature were too viscous to analyse and no results were reported.

The samples at 60°C could be fed into the system, but analyses were hampered by the presence of entrained air bubbles and no results were reported.

When the samples were diluted with a melted refined sugar, the air bubbles were eliminated and results could be obtained.

A-massecuite:	SMRI MA = 0.61 mm	Sympatec MA = 1.37 mm
	SMRI CV = 32%	Sympatec CV = 74%
C-massecuite:	SMRI MA = 0.22 mm	Sympatec MA = 0.07 mm
	SMRI CV = 30%	Sympatec CV = 47%

[1] SB Davis, Sugar Milling Research Institute, c/o University of KwaZulu-Natal, Durban, 4041, RSA (2008).

Discussion

Results confirmed that the Sympatec instrument was able to obtain data in bell-shaped size distributions typical of both A- and C-massecuite samples. As expected the MA and CV results for the two analyses were different. This will be remedied when the equations associated with the indirect ultrasound measurement can be adapted and optimised for a massecuite environment. The ultrasound data for the C-massecuite sample showed a bimodal distribution due to the difference in the length and width of each crystal. The SMRI analysis takes this into account by measuring length and width separately.

Conditions during the ultrasound analysis were not representative of actual conditions during pan boiling, but rather of an off-line evaluation. In this regard, the measurement of diluted and undiluted samples under various conditions needs to be investigated.

Further evaluation of the instrument by the SMRI was suspended due to concerns about the cost of the instrument.

Acknowledgements

The authors would like to acknowledge Set Point Instruments (Durban, South Africa) and Sympatec GmbH (Germany) for shipment of samples and analyses performed using the ultrasound instrument.

REFERENCES

Anon (2007). Particle size distribution of white sugar. International Commission for Uniform Methods of Sugar Analysis (ICUMSA) Methods Book, GS2-37.

Dalziel SM, Tan S-Y, White ET and Broadfoot R (1999). An image analysis system for sugar crystal sizing. *Proc Aust Soc Sug Cane Technol* 21: 366-372.

Geers H and Witt W (2003). Ultrasonic extinction for in-line measurement of particle size and concentration of suspensions and emulsions. *Particulate Systems Analysis:* 5 pp.

Hartman and Behrens (2006). Simultaneous on-line analysis of solid concentration and particle size distribution of gypsum slurries applying ultrasonic extinction. *ZKG International* 4(59): 17-21.

Ingram GD and Steindl RJ (2001). Rapid crystal sizing on the pan stage by digital image analysis. *Proc Aust Soc Sug Cane Technol* 23: 361-368.

Iswanto NUR, White ET, Hardin MT and Broadfoot R (2007). Measurement of crystal size spread through the high grade pan stage for an Indonesian and an Australian factory. *Proc Aust Soc Sug Cane Technol* 29: 420-430.

Jones OD, White ET and Butler BK (2000). Estimating crystal growth rates using computed tomography. *Inverse Problems* 16: 1477-1485.

Jullienne LMSA (1985). South African C-massecuites: crystal size distribution and its effect on centrifugal losses. *Proc S Afr Sug Technol Ass* 59: 79-82

Lionnet GRE (1999). Pan Boiling I. *Sugar Technology for Students*, Study Text 7, GRE Lionnet, Durban: 19.

Lionnet GRE (2001). The measurement of the C-crystal size and its impact on sucrose losses. *Proc Int Soc Sug Cane Technol* 24: 387-387.

McClements DJ (2000). Ultrasonic measurements in particle size analysis. pp 1-8 In: Meyers RA (Ed.), *Encyclopedia of Analytical Chemistry*. John Wiley and Sons, Chichester, England.

Mhlongo AZ and Alport MJ (2002). Application of artificial neural network techniques for measuring grain sizes during sugar crystallization. *Proc S Afr Sug Technol Ass* 76: 460-468.

Miller KF and Beath AC (2000). The measurement of raw sugar crystal size by sieving and by laser diffraction. *Proc Aust Soc Sug Cane Technol* 22: 393-398.

Neidhardt R and Behrens C (2004). Inline particle size analyses. *Instrumentation, Control and Automation* 2: 48-49.

Pankewitz A and Behrens C (2002). In-line crystal analysis with a highly adaptable and industrially approved sensor based on ultrasonic extinction. Paper presented at the 15th International Symposium on Industrial Crystallization. 7 pp.

Schäffler KJ (1999). NIR estimation of crystal size distribution: a technical step too far? Sugar Milling Research Institute Technical Report No 1819. 6 pp.

Wagner G, Schick R, Austmeyer KE and Fleischer L (1999). Optimization of an image analysis system for the determination of the crystal quality in sucrose crystal suspensions. *Zuckerind* 126: 34-41.