

CFD MODELLING OF SUCROSE CRYSTALLISATION

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ABSTRACT

In this paper one- and three-dimensional numerical models for the crystallisation of sucrose from low grade massecuite are presented. Crystal growth is simulated by solving population balance equations for the moments of the crystal size distribution together with a transport equation describing sucrose mass balance. The models require the flow and temperature field to be known, and these are either specified for the one-dimensional model or obtained from solution of the mass, momentum and energy equations for the two-dimensional model. The models are applied to study crystal growth in a vertical continuous cooling crystalliser. The simulations show that the vessel studied does not perform satisfactorily as the predicted crystal growth is less than desired. The results illustrate some of the problems associated with cooling crystallisation in highly viscous fluids, demonstrate the feasibility of simulating crystallisation processes using CFD, and display the enormous potential that CFD provides to improve crystalliser design.

NOMENCLATURE

C_{sat}	saturation concentration of sucrose in solution
CV	coefficient of variation of CSD
D/Dt	substantial time derivative ($\frac{\partial}{\partial t} + \mathbf{v} \cdot \nabla$)
D_G	crystal growth rate diffusivity
G	linear crystal growth rate
I	mass ratio of impurities to water
k_v	volume shape factor ($\pi/6$)
k_a	area shape factor (π)
K_t	total kinetic growth coefficient
K_s	kinetic coefficient for surface incorporation
K_d	kinetic coefficient for diffusion (mass transfer)
L	crystal size
\bar{L}	mean crystal size
m_j	j^{th} moment of the crystal size distribution
n	crystal population density distribution
P	molasses purity ($100 S/(I+S)$)
r	radial coordinate
r_i	radius of inner shaft (0.381 m)
r_o	radius of outer wall (2.625 m)
S	mass ratio of sucrose to water
S_S	supersaturation (S/S_{sat})
S_{lim}	limiting supersaturation
t	time
T	temperature
u_z	component of velocity in axial direction
\mathbf{v}	velocity vector of both crystal and molasses
x_c	crystal content of massecuite by mass
x_w	water content of massecuite by mass
z	axial coordinate

ϕ	general variable
$\dot{\gamma}$	shear rate
μ_{mo}	apparent molasses viscosity
ρ_c	density of sucrose crystal (1587.9 kg/m ³)
σ	variance of CSD

Subscripts

sat	saturation or equilibrium conditions
0	conditions at inlet of crystalliser

INTRODUCTION

The Australian raw sugar industry utilises both evaporative and cooling crystallisers to extract sucrose from solution. In evaporative crystallisers, crystal growth occurs in a supersaturated sucrose solution (molasses) which is produced by evaporating water under vacuum. This process concentrates impurities in the sucrose solution and eventually becomes uneconomical as the increased concentration of impurities slows the growth kinetics. The supersaturated solution is given time to crystallise in a cooling crystalliser where the molasses and crystal mixture, known as massecuite, is cooled to decrease sucrose solubility and further drive crystal growth. Due to the slow growth kinetics a cooling crystalliser requires a longer residence time compared with an evaporative crystalliser (typically 10-20 h compared with 2-3 h for a batch vacuum pan). After leaving the cooling crystalliser the massecuite is separated into crystals, which are recycled to the vacuum pans, and final molasses, which is a low value by-product.

Approximately 6% of the sucrose entering in the cane supply to an Australian sugar factory leaves in the final molasses, and this represents the largest loss of sucrose in the processing of cane to crystal sugar (Broadfoot and Steindl, 1992). Even a small increase in the recovery of sucrose in the cooling crystallisers would represent a significant benefit to the industry. For example, if the average purity (a measure of sucrose concentration in the molasses) of the final molasses could be reduced by 2%, the net revenue gain for the industry would be approximately \$5 million per annum.

One of the challenges faced by designers of cooling crystallisers is how to achieve the high shear rates necessary for effective crystallisation in a very viscous fluid. This problem is coupled with the inherent difficulties associated with cooling viscous fluids; namely, that cooling is confined to extremely narrow thermal boundary layers adjacent to the cooling surfaces, and that the high viscosity hinders mixing of cold and hot fluid. Controlling the flow of massecuite in a cooling crystalliser

is also difficult as there is a strong tendency to form preferential flow paths of hot, less viscous fluid.

Computational fluid dynamics (CFD) holds great promise to help overcome these design challenges and achieve a more efficient crystalliser design. For example, Sima and Harris (1999) demonstrated that simple design modifications, arrived at with the assistance of CFD, made a large improvement in the residence time distribution of a commercial design of vertical cooling crystalliser.

This paper considers the same crystalliser design studied by Sima and Harris (1999) but extends their CFD model to include prediction of crystal growth. The crystal growth model developed here computes the moments of the crystal size distribution at all points in the vessel, from which quantities of interest, such as purity, crystal content, and crystal size, can be computed.

CRYSTALLISER GEOMETRY

The crystalliser studied here is a vertical continuous crystalliser consisting of a steel cylindrical vessel, 5.25 m in diameter and 13.84 m in height. The vessel contains 8 horizontal rows of 150 NB cooling pipes (only 7 rows are modelled) and 15 rows of 100 NB cooling pipes in the upper and lower sections, respectively. A central shaft and 22 rows of radial stirrer arms rotating at about 0.25 rpm provides a stirring action to facilitate crystal growth.

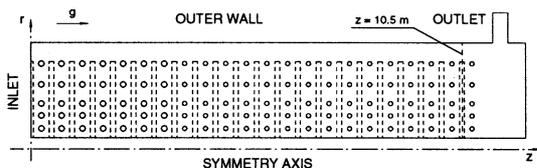


Figure 1: Geometry of axisymmetric vertical crystalliser model. The dashed lines show where the tangential body forces are applied.

The actual crystalliser is inherently three-dimensional, but, due to computational limitations, a rotationally symmetric numerical model of the crystalliser was constructed by Sima and Harris (1999). Figure 1 shows the simplified axisymmetric crystalliser. In this model the rotating stirrer arms are replaced by tangential body forces which produce the equivalent drag of the stirrer arms. Their model solves for the temperature, pressure and the axial, radial and tangential velocity components. As the simplified geometry introduces artefacts that are not present in the actual vessel (e.g., the outlet is a radial slot rather than a single pipe, and the cooling pipes are rings) the results from their simulations have to be interpreted carefully. Despite this limitation their predictions were consistent with the measured residence time distribution on the actual vessel. The reader is referred to Sima and Harris (1999) for full details of the numerical model as the present work makes use of their results.

MATHEMATICAL MODEL OF CRYSTAL GROWTH

The total growth of a crystal is determined by its growth rate integrated along its path through the vessel. The growth rate can vary greatly from one region of the vessel to another, depending on the spatial distribution of temperature and shear rate. The path of each crystal through the vessel can be determined from the velocity field. Thus, the calculation of crystal growth cannot

proceed without a knowledge of the flow field and temperature field. It is assumed that the crystal growth has minimal effect on the flow field, so that an uncoupled calculation of crystal growth is possible. As demonstrated by Sima (1998), this is a valid assumption for the low grade massecuite considered here.

Growth of an individual crystal is a two-step process consisting of mass transfer through a volume with a concentration gradient surrounding the crystal, followed by surface incorporation into the crystal structure. The mass transfer depends on the hydrodynamic conditions while the surface incorporation depends on the supersaturation at the crystal-solution interface, the temperature and the presence and composition of impurities (i.e., materials other than the sucrose and water). A consequence of the two-step nature of crystal growth is that the slowest of the two processes limits the net growth of sucrose crystals. A detailed account of models for crystallisation from solution is given by Ohara and Reid (1973).

Low grade molasses is characterised by a very high concentration of impurities. In the Australian raw sugar industry the composition of the impurities tends not to differ much from a “standard” composition so that the total impurity concentration (impurity to water ratio) is used to characterise the amount of impurities. The effect of impurities on crystal growth is discussed by Maurandi *et al.* (1988) and Broadfoot and Steindl (1992). Some impurities, termed reducing sugars, bind water, thereby decreasing sucrose solubility. Others, termed ash, bind sucrose thereby increasing sucrose solubility. Certain impurities affect the surface incorporation, thereby decreasing the growth rate and changing the crystal shape. The mass transfer to the crystals is also decreased by the impurities, mainly through the considerable increase in molasses viscosity. The overall effect on the total growth rate is an exponential decrease with increasing concentration of impurities. According to Maurandi (1981), the impurities also increase the supersaturation required for nucleation.

Pure sucrose solutions are Newtonian (Gibbs *et al.*, 1996). However, the impurities make the molasses non-Newtonian and pseudoplastic (Broadfoot and Steindl, 1992).

For a dilute suspension of crystals in a pure sucrose solution the mass transfer to a crystal may be determined from Sherwood number correlations for a single sphere. However, the low-grade sucrose solutions considered here are dense suspensions containing high levels of impurities, so that the Sherwood number correlations are inappropriate. Instead, use is made of correlations for the mass transfer and associated crystal growth based on an experimental study performed under the conditions encountered in a low grade crystalliser.

Population balance

The growth rate of a crystal is usually described by the rate of change of a length that represents the size of the crystal, since this has been observed to be independent of the size itself. Since the addition of crystal mass depends on the existing crystal area, some information on the total crystal area is required to determine the amount of solute

crystallised and the total crystal volume or mass. It is also of interest to predict the sizes of the crystals that are produced. This information is given by the crystal size distribution (CSD). The evolution of the CSD is described by a population balance equation (Randolph and Larson, 1988).

For the purposes of modelling, it is assumed that (1) there is no nucleation, agglomeration or crystal breakage; (2) the crystals flow with the same velocity as the molasses; and (3) the change in density due to the phase change of sucrose is negligible. The very high impurity concentrations in low grade molasses makes the crystallisation kinetics very slow and the molasses very viscous. The high viscosities and low velocities encountered in a cooling crystalliser make it unlikely that crystal breakage occurs. Also, it is considered that agglomeration (the joining of crystals), which commonly occurs at high supersaturation and high growth rates, is unlikely to occur. The assumption of no nucleation is probably the weakest, as contact nucleation could occur. However, it is uncertain whether the contact or shear forces are large enough to cause contact nucleation, due to the very high viscosities and low velocities (both relative and absolute) encountered in a cooling crystalliser. Since the crystal settling velocity (8×10^{-8} m/s for a 0.32 mm crystal in the molasses considered here) is almost four orders of magnitude smaller than the characteristic molasses axial velocity (0.28 mm/s) the second assumption is justified. The third assumption was confirmed by calculating the density change of the mixture using the one-dimensional model. It was found to be less than 1% during a run with a mean residence time of 16 h.

Given the above assumptions the population balance equation can be written as

$$\frac{Dn}{Dt} = -\frac{\partial}{\partial L} \left(G_n - D_G \frac{\partial n}{\partial L} \right) \quad (1)$$

where D/Dt is the substantial time derivative and n is the population density distribution (per unit mass of mixture). The j^{th} moment of the population density distribution is defined as

$$m_j = \int_0^{\infty} L^j n(L) dL \quad (2)$$

The zeroth moment is thus the total number of crystals (per unit mass of massecuite), the first moment is the total length of the crystals, the second moment multiplied by the area shape factor (k_a) is the total surface area, and the third moment multiplied by the volume shape factor (k_v) is the total volume.

The population balance equation can be transformed into equations for the moments by multiplying by L^j and integrating from zero to infinity with respect to L . By partial integration, under the assumption that D_G is independent of L and using the boundary conditions of zero n and $\partial n/\partial L$ at $L = 0$ and at $L = \infty$, the following moment equations are obtained (Sima, 1998)

$$\frac{Dm_j}{Dt} = jGm_{j-1} + j(j-1)D_G m_{j-2} \quad (3)$$

for $j = 0, 1, 2$ and 3 . The boundary conditions result from the assumptions that no nucleation occurs and that the crystal size distribution is limited and does not contain any

nuclei. In the above equation D_G accounts for crystal growth rate dispersion (i.e., for the same conditions not all crystals grow at the same rate). Growth rate dispersion is important if a precise prediction of the crystal size distribution is required, or when interpreting growth kinetics from experiments. On the other hand, growth rate dispersion is not a significant feature when determining the design or operation of a crystalliser that will produce the maximum growth of the mean crystal size. For the simulations presented here the growth rate diffusivity is set to zero.

When describing the CSD the mean size and coefficient of variation is preferred to the moments. The mean crystal size is given by

$$\bar{L} = m_1 / m_0 \quad (4)$$

and the coefficient of variation is defined as

$$CV = \sigma / \bar{L} \quad (5)$$

where the variance is

$$\sigma^2 = \frac{1}{m_0} \int_0^{\infty} (L - \bar{L})^2 n(L) dL = \frac{m_2}{m_0} - \left(\frac{m_1}{m_0} \right)^2 \quad (6)$$

Mass balance

A knowledge of the sucrose concentration is required to determine the growth rate, and an equation for sucrose concentration may be derived from a mass balance. When crystallisation takes place, sucrose from the molasses is incorporated into crystals. Since the sucrose crystals consist of more than 99.5% sucrose, the crystals are assumed to consist only of sucrose so that all the impurities remain in the molasses. That is, the increase of crystal mass equals the rate of decrease of sucrose mass in the molasses, which is written as

$$\frac{DS}{Dt} = -\frac{1}{x_w} \frac{Dx_c}{Dt} \quad (7)$$

Noting that $x_c = \rho_c k_v m_{3,0}$, and that the mass of water per unit mass of massecuite (a constant) is

$$x_w = \frac{1 - \rho_c k_v m_{3,0}}{S_0 + I + 1} \quad (8)$$

the sucrose mass balance can be written as

$$\frac{DS}{Dt} = \rho_c k_v \frac{S_0 + I + 1}{1 - \rho_c k_v m_{3,0}} \frac{Dm_3}{Dt} \quad (9)$$

Growth rate model

The linear growth rate of a crystal is given by the supersaturation driving force multiplied by a kinetic growth coefficient (Broadfoot and Steindl, 1992)

$$G = K_t C_{sat} (S_s - S_{lim}) \quad (10)$$

The limiting supersaturation is taken to be the value at which no further crystal growth is observed. Theoretically the limiting supersaturation is unity, but in this model is slightly greater than one for reasons discussed by Sima (1998). The expression for the total kinetic growth coefficient, K_t , is

$$1/K_t = 1/K_s + 1/K_d \quad (11)$$

where $K_s(T, I)$ and $K_d(T, \mu_{m_0}, \dot{\gamma})$ are the kinetic coefficients for surface incorporation and mass transfer to

the crystal, respectively. As can be seen from equation (11), the process which has the smallest kinetic coefficient will limit the overall growth rate. Empirical correlations for the kinetic coefficients, limiting supersaturation and other quantities as functions of the independent variables were determined experimentally by Broadfoot and Steindl (1992) for parameter ranges that were assumed to be representative of the average conditions in a low grade cooling crystalliser. In the absence of more general results, these expressions have been used in this study, even though it was necessary to extrapolate outside their range of validity to cover the large range of conditions encountered in the CFD crystal growth model.

NUMERICAL SOLUTION

Equation (3) represents a closed set in terms of the moments of the CSD once the growth rate relationship is specified. The growth rate depends on the sucrose concentration, given by equation (9), and the kinetic growth coefficients, which are functions of the shear rate, viscosity, temperature and other variables obtained from a solution of the mass, momentum and energy transport equations. In this paper these solutions are from Sima and Harris (1999), using their model with unmodified geometry and adiabatic external walls.

The equations that describe crystal growth (equations (3) and (9)) were solved using three general species transport equations in the general purpose Fluid Dynamics Analysis Package (FIDAP), making use of a user-defined subroutine to define the source terms. The existing solution for the velocity and temperature fields was read in using the ICNODE(READ) option and used in the computation of parameters such as the kinetic growth coefficients. The transport equations do not contain any diffusion terms, which causes numerical difficulties in FIDAP. For this reason small diffusion coefficients (of order 10^{-9} m²/s) were introduced into each equation to facilitate the solution. The FIDAP solution method was validated against an analytical two-dimensional channel flow model that included crystal growth by Sima (1998).

One-dimensional model

A simplified model, analogous to that developed by Broadfoot and Steindl (1992) was also prepared. This model integrates the one-dimensional form of the moment equations with respect to time. The model requires the initial (inlet) conditions and residence time to be specified, along with temperature and shear rate profiles. Numerical integration of the governing ODEs is carried out using Engineering Equation Solver (EES). Since it runs extremely quickly, this model is useful for carrying out parametric studies to investigate the effect of global parameters such as the temperature profile and shear rate. For example, Broadfoot and Steindl used their model to determine optimum cooling profiles for maximum exhaustion (i.e., extraction of sucrose).

RESULTS AND DISCUSSION

Many of the features of crystal growth are determined by the residence time distribution, which is related to the velocity field. The present vessel has a poor residence time distribution, with major short circuiting of virtually uncooled massecuite down the region adjacent to the outer wall for reasons discussed by Sima and Harris (1999). This hot, fast moving massecuite has insufficient residence

time for much crystal growth to occur. Conversely, there is an area of cool, very viscous massecuite in the region surrounding the inner three columns of cooling pipes. This massecuite has a long residence time (2–3 days) compared to the mean (11 h) and so has ample time for crystal growth.

An important parameter that measures sucrose extraction is molasses purity, P , which is a measure of sucrose concentration in the molasses. A shaded contour plot of the computed purity distribution is shown in Fig. 2. The lowest purity level (i.e., highest sucrose extraction) occurs in the region where the velocity is slowest and the temperature is the lowest. However, these low purity values make an almost insignificant contribution to the mass-averaged purity leaving the outlet. The major contribution to the outlet purity is from the fast moving, high temperature, high purity region adjacent to the outer wall. The net result is that the predicted overall purity decrease is not as large as would be expected in a properly functioning cooling crystalliser.

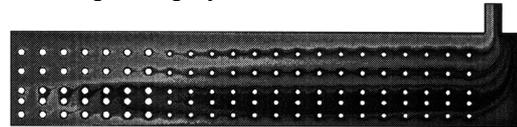


Figure 2: Molasses purity distribution. The lightest shade represents the inlet purity (47%) and the darkest shade represents the minimum purity (36%).

Radial profiles from the CFD model at an axial distance of 10.5 m from the inlet (refer to Fig. 1) are presented in Fig. 3. This vertical location was selected as it is close to the outlet, but is not unduly influenced by the changed flow direction at the outlet. The corresponding residence time at this vertical location is 10.35 h. Figs. 3 (a) and (b) illustrate that a significant fraction of the massecuite moves rapidly down the outer part of the vessel with minimal cooling (i.e., the temperature at $r = 2.5$ m is still virtually equal to the inlet temperature). This undesirable behaviour results in a poor residence time distribution with a peak at around 3.2 h, significantly lower than the mean residence time of 10.35 h at this location. The region of the vessel around $r = 1$ m receives excessive cooling which leads to a large region of very viscous, virtually stagnant massecuite. In this essentially stagnant region, which has a residence time of 2–3 days, the crystal content is very high and the molasses purity is very low, since crystal growth has a long time to occur. The opposite is true in the fast moving outer region, in which there is insufficient time to achieve the desired amount of sucrose extraction. Since it occupies a large part of the cylindrical crystalliser volume and moves with high axial velocity the outer region makes the major contribution to the overall mass-averaged crystal growth.

The overall performance of the crystalliser is illustrated in Fig. 4. The open symbols are radially mass-averaged results computed from the FIDAP CFD model using

$$\bar{\phi} = \frac{\int_{r_i}^{r_o} \phi u_z r dr}{\int_{r_i}^{r_o} u_z r dr} \quad (12)$$

where ϕ represents the quantity to be mass-averaged (e.g., temperature). The three curves shown in Fig. 4 represent different runs of the one-dimensional crystal growth model. In each case the prescribed inlet conditions are a

temperature of 60°C, a crystal content of 35%, molasses purity of 47%, a mean crystal size of 0.32 mm, and an impurity to water ratio of 3.7. Except in case (iii), the prescribed temperature profile was set to match the mass-averaged temperature profile computed from the CFD model. The prescribed shear rate was set to 0.014 s⁻¹ (except in case (ii)) which matches the global mean value from the CFD simulation.

Fig. 4 (a) shows that the section of 150 NB cooling pipes produces a faster cooling rate than the 100 NB pipe section, as it was designed to. However, the overall temperature drop is less than 4 K, which is significantly less than the ‘optimum’ cooling rate of about 1 K/h recommended by Broadfoot and Steindl (1992). The low overall cooling rate is caused by significant short-circuiting of virtually uncooled massecuite down the outer walls of the vessel. Despite the poor residence time distribution of this (unmodified) vessel, it is evident that crystal growth occurs. However, the outlet purity of 44.4% represents a drop of only 2.6 units through the crystalliser, and a significant portion of this will be lost when the massecuite is reheated (to lower the viscosity) before the final centrifuge process. The target outlet purity is less than 42% if possible, so a large improvement is needed to achieve this level.

The base-line case, with temperature profile and shear rate matching the CFD simulations, shows very good agreement with the mass-averaged CFD results.

For the low mean shear rate predicted by the CFD simulation, the kinetic coefficient for mass transfer is about half that for surface incorporation, so that the crystal growth is mass transfer limited. This suggests that one way to improve crystal growth (in this particular case) is to provide higher shear rates through increased stirring or some other means. The crystal growth predicted by the one-dimensional model for an increased shear rate of 0.1 s⁻¹ (holding everything else constant) is shown as curve (ii) in Fig. 4. As can be seen, there is a marked improvement in overall crystal growth. At this shear rate and temperature the kinetic coefficients for mass transfer and surface incorporation are approximately equal. Further increases in shear rate (up to about 1–2 s⁻¹, where the crystal growth becomes limited by surface incorporation) also produces improved crystal growth. Another way to increase overall crystal growth is to provide a longer residence time (e.g., reduce the flow rate through the vessel), but this is not desirable from an operational viewpoint, where the goal is to increase throughput.

As mentioned above, the massecuite temperature only drops about 4 K after 10.35 h, which is less than recommended. Case (iii) shows the one-dimensional model results for the hypothetical cooling profile shown in Fig. 4 (a), which provides a 10 K temperature drop over 10.35 h (approximately 1 K/h). Interestingly, the predicted crystal growth is practically the same as the base-line case, showing that crystal growth is virtually independent of the cooling profile (at this low shear rate). The reason for this finding is that the beneficial effect of a decrease in sucrose solubility with decreasing temperature (causing an increase in the driving force $S_s - S_{lim}$) is offset by the detrimental effect of a decrease in the total kinetic growth coefficient, K_t , with the net result that the total growth rate is only marginally altered. For higher mean shear rates

(e.g., 0.1–1 s⁻¹), increased cooling does result in higher growth rates since the crystal growth driving force increases faster than the total kinetic growth coefficient decreases.

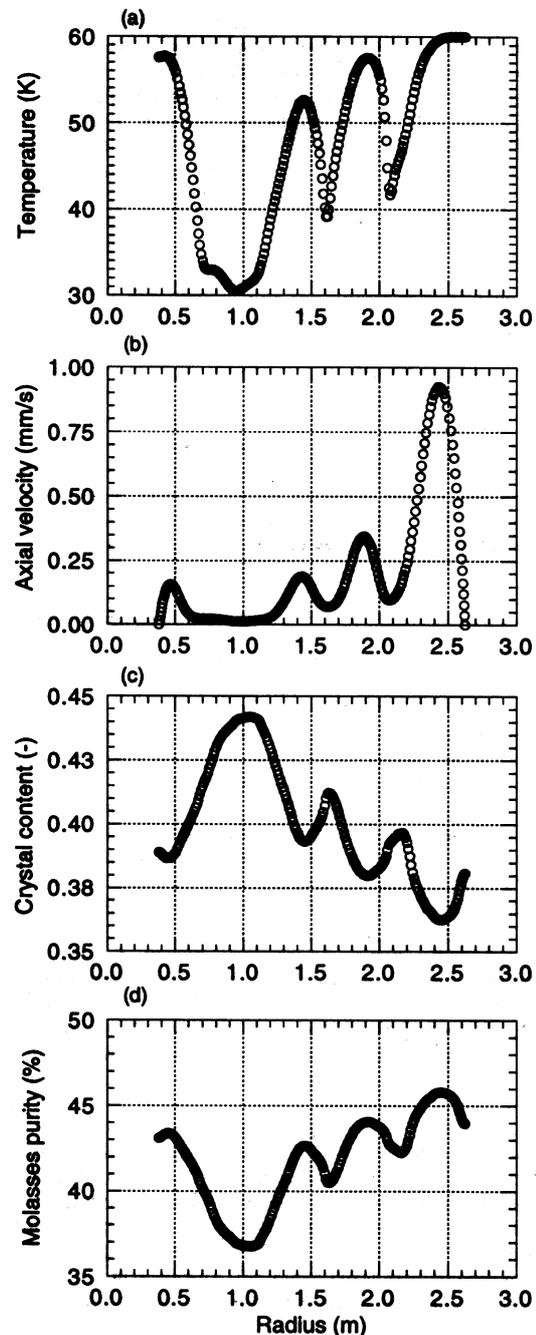


Figure 3: Radial profiles from the CFD model at a height of $z = 10.5$ m. (a) temperature; (b) axial velocity; (c) crystal content; (d) molasses purity.

The quantitative predictions of crystal growth from both the one-dimensional model and the CFD model depend strongly on the correlations for the kinetic growth coefficients and massecuite viscosity. As these correlations required extrapolation for the low shear rates encountered in the CFD model, the quantitative values should be treated with some caution. Work is currently under way at James Cook University and the Sugar Research Institute to extend these correlations to low shear rates.

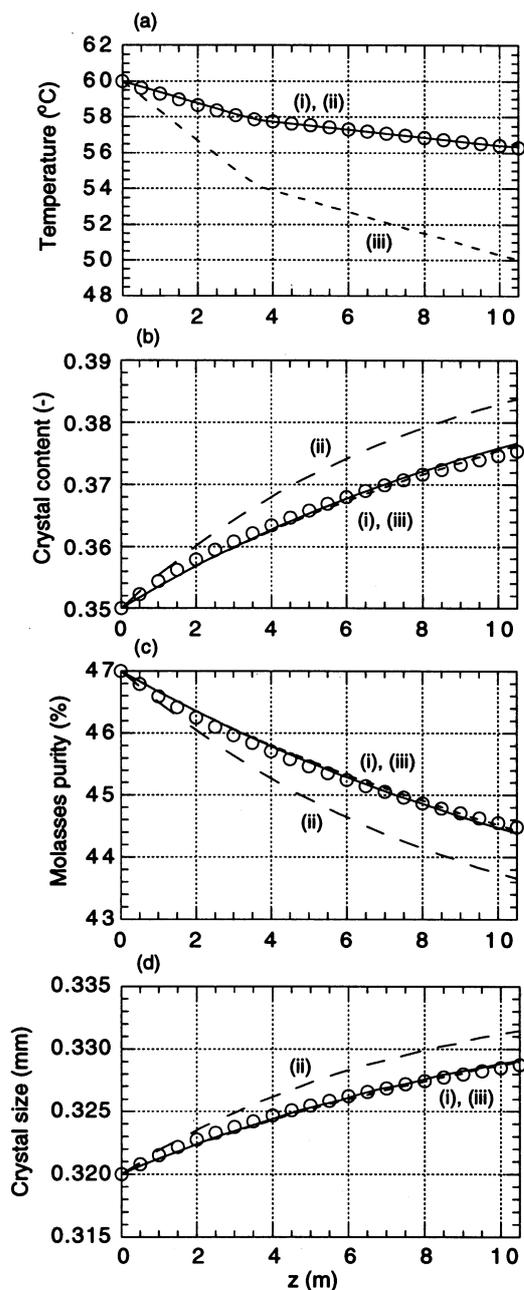


Figure 4: Crystal growth as function of position: (a) temperature; (b) crystal content, x_c ; (c) molasses purity; (d) mean crystal size. Open symbols are radially mass-averaged results from the FIDAP CFD model. Curve (i): baseline case; (ii) increased shear rate; (iii) increased cooling rate.

The results also demonstrate the importance of accurate predictions of the shear rate in a crystalliser, since it has a major influence on the crystal growth rate. The present model is an axisymmetric simplification of the true three-dimensional geometry and so may yield shear rates that are somewhat different to those in the actual vessel. If quantitative crystal growth predictions are required it may be necessary to perform three-dimensional simulations to correctly predict the shear rate. However for comparative studies of different crystalliser designs, one-dimensional or simplified CFD models are probably adequate.

CONCLUSION

A method for simulating sucrose crystal growth in low-grade massecuite has been presented. The method can be solved in the form of a one-dimensional model or can be incorporated into a CFD model using user-defined scalar transport equations. In the former case the model requires the cooling profile and shear rate to be specified, whereas in the latter case these are obtained from a separate solution of the equations of motion and energy conservation. The one-dimensional model is simple and executes rapidly, but only provides global averaged quantities. On the other hand, the CFD model is computationally intensive but provides a wealth of detail about the spatial distribution of variables relating to crystal growth. The CFD model is an ideal tool for revealing problematic regions in the vessel, so should find considerable application in the design of improved crystallisers, or the modification of existing vessels.

In the cooling crystalliser studied the overall crystal growth was less than desirable, primarily due to a poor residence time distribution and a low mean shear rate. To improve the performance of this vessel the residence time distribution would have to be addressed (e.g., through the use of baffles to prevent the short circuiting) and, ideally, the mean shear rate increased so that mass transfer does not limit the growth rate.

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