A PROCESS DESIGN STUDY FOR A RAW SUGAR CRYSTALLIZATION PROCESS

by

Rinly Ruiz Gecosala

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APPROVAL BY THESIS DIRECTOR

This thesis has been approved on the date shown below:

A. D. Randolph
A. D. RANDOLPH
Professor of Chemical Engineering

7/3/79
Date

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ABSTRACT

Process designs for batch and continuous sugar crystallizers were undertaken to demonstrate use of the population balance with growth dispersion for this task. Both the batch and continuous units were assumed to be seeded with a continuous nucleator/ripener unit. A simplified technique using moment relations and constant growth rate was used for design of the batch pans. A classified recycle configuration was adopted in the design of the high-grade continuous unit. A previously developed numerical program, RECYC, was used to simulate the continuous classified recycle configuration.

All designs had detailed specifications which justified the choice of conditions, design constraints, and calculations. Product weight distributions were determined. When possible, an approximate final design was summarized and commented on.

Finally, continuous pans were compared to batch pans. The continuous classified recycle configuration appears to require perhaps as little as 50% of the batch pan working volume, based on equal production rates. However, a narrower CSD could be produced with batch operation.

CHAPTER 1

INTRODUCTION

The production of sugar is a large-scale industry in many countries. The annual world production approaches 100 million tons.

A key step in the process is the crystallization of sugar from solution. Traditionally, this has been carried out as a batch operation, but in the last few years there has been increasing interest in continuous crystallization.

Possible advantages claimed (Broadfoot and White, 1978) for continuous crystallization are:

1. Easier automatic control.
2. Less fluctuation in product quality.
3. Full utilization of vessel capacity.
4. Improved energy usage.
5. More efficient operation.
6. Less manual supervision.

The disadvantages of continuous operation would be:

1. A possibly wider product size range.
2. Fouling of heat transfer surfaces, requiring periodic shutdown for cleaning.
3. Lack of experience with continuous operation.

A number of prototype continuous crystallizers (pans) have been built and installed commercially (Windal, 1971; Barkow and Witte, 1971; Riviere and Pithois, 1976; Langreney, 1977; Steindel et al., 1975). It is clear from the variations in design that the optimum design may not yet have evolved. Also, there are techniques such as product classification and fines recyle, common in other crystallization fields, which might be applicable to continuous sugar crystallization.

The purpose of this thesis is to detail the steps in a process design method for sugar crystallizers. This will be done by specific example, illustrating the necessary calculations for a raw sugar factory producing 800 tons per day of sugar. A batch system and a continuous system will be considered.

CHAPTER 2

REVIEW OF CURRENT SUGAR CRYSTALLIZATION TECHNOLOGY

Sugar (sucrose) is produced commercially primarily from sugarcane and cane sugar beet. The review which follows gives specific attention to sugarcane, as this is the source in the author's own country, the Philippines.

Sugar is synthesized and stored in the cane plant with contents up to 15% by weight. The manufacturing process is concerned mainly with separating the sucrose, in highly purified form, from all other constituents that occur in the cane.

The basic manufacturing process, as shown in Figure 1, consists of a sequence of unit operations. The process is described in detail in a number of books. Jenkins (1966) discusses the technology for raw cane sugar, while Baikow (1967) describes both raw sugar manufacture and the refined sugar process. Spencer, Meade, and Chen (1977) include recent advances in continuous crystallization.

A brief description of the total process is now given.

Raw Cane Sugar Manufacture

Cane is brought to the factory from the fields (Figure 1). It is first identified and weighed (for payment to suppliers or for control) and is then shredded through a hammer mill before passing to the mills. In the mills, the juice is squeezed from the fiber and the fiber is

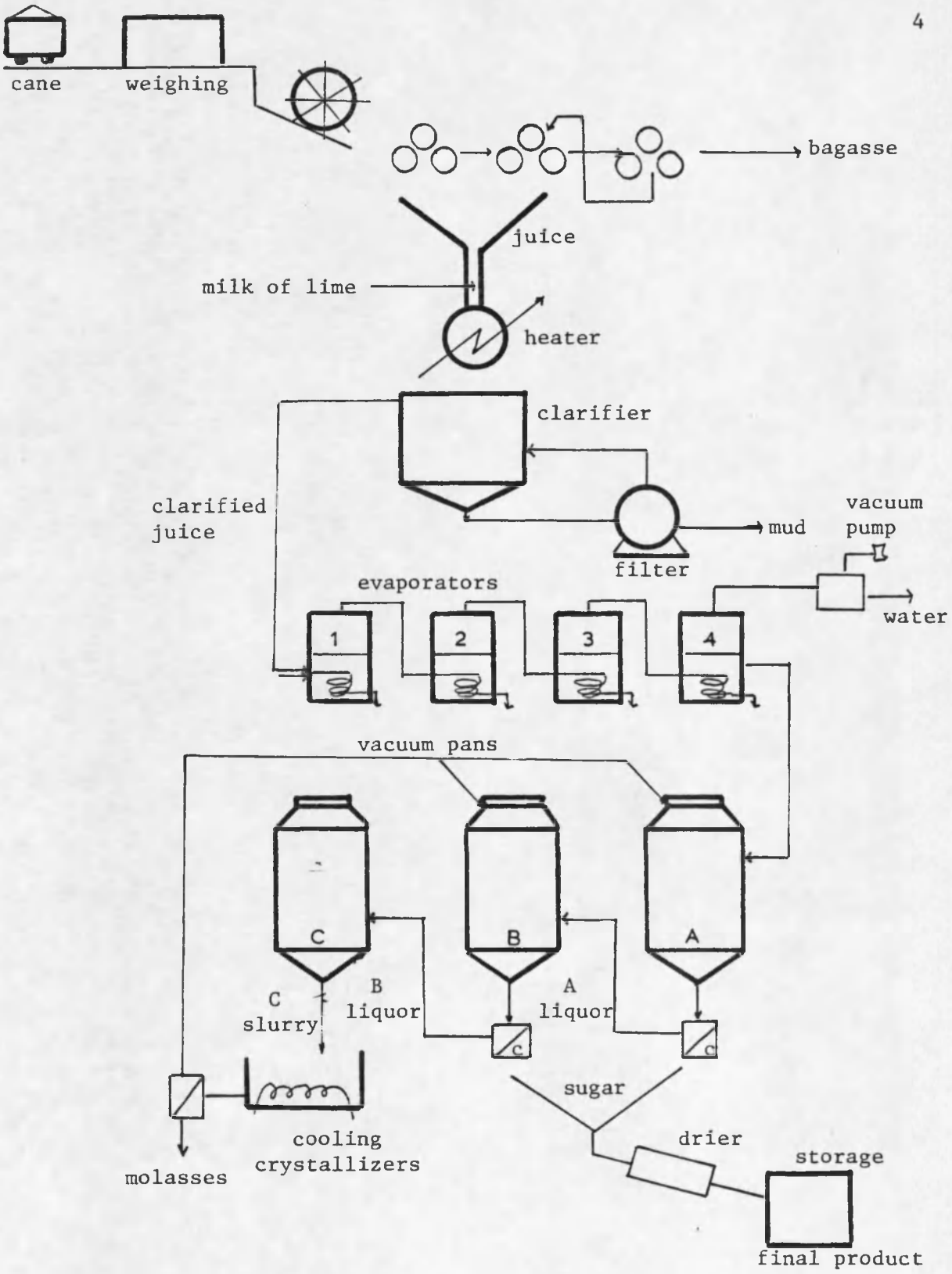


Figure 1. Raw Sugar Process Flow Diagram.

washed counter-currently (imbibition) in subsequent mills to recover up to 95% of the sugar. The moisture content of the residual fiber (bagasse) can be reduced to 45%. This material can then be used as a fuel, which makes the whole process energy self-sufficient.

As an alternative to milling, a "diffusion" (extraction) process can be used (Baikow, 1967).

The clarification step comes next. This consists of juice screening, liming (with milk of lime) to a controlled pH, heating and clarification to give a clear overflow juice. Sucrose loss in the clarifier mud is reduced by means of washing in decanters or on a rotary filter. Clarification using heat and lime has been found to be the simplest technique even though other chemical reagents may be used. The purpose of the clarification step is to obtain the maximum efficiency of removal of suspended impurities and the adjustment of pH. This minimizes degradation of sucrose in subsequent operations.

Removal of an enormous quantity of water from the clarified juice is required before crystallization can be carried out. This is done in multiple effect evaporators where the vapor leaving one vessel is used in heating the next. From the evaporators, a slightly under-saturated "syrup" emerges, containing 55-65% dissolved solids.

The next stage involves crystallization, whereby the further evaporation of water occurs, the syrup becomes supersaturated, and crystals grow. Eventually, a large bulk of crystals is produced, leaving a small quantity of mother liquor (molasses). The bulk of crystallization occurs in vacuum pans operated at 24 to 26 in. Hg vacuum so that

boiling occurs at a reduced temperature, e.g., 60-65°C. Higher temperatures cause sucrose degradation. Crystallization is usually carried out in the absence of nucleation (the generation of further fine crystals) and, thus, seed crystals must be fed to the pans.

Because the amount of residual liquor (molasses) becomes quite small compared to the quantity of crystal produced, crystallization has to be carried out in several stages (or boilings) with the molasses and crystals being separated at the end of each boiling. Separation of molasses from sugar is usually carried out in high-speed centrifuges.

Two boiling, three boiling, and sometimes four boiling schemes have been used. In each case, for the last (low-grade) boiling with the lowest quality molasses, fine seed crystals are used. The crystal product from this boiling is used as seed crystals for the earlier (purer liquor), high-grade boilings. The crystal products produced from these boilings are mixed and become the final raw sugar product.

In Figure 1, a three boiling scheme (A,B,C) is indicated diagrammatically. The syrup enters the A pan together with the seed crystals from the C boiling. After evaporation of water and crystal growth, the crystals are separated in a centrifuge and pass to product while the liquor (termed A molasses) passes to the B boiling which is seeded from the C boiling pan. Product crystals from the B pan are again produced. The liquor from the centrifuge after the B boiling (B molasses) is used as the feed to the C pan, together with fine seed crystals produced elsewhere in the factory, e.g., by grinding sugar crystals in an alcohol slurry.

After the C boiling, the molasses-crystal mixture (massecuite) is usually cooled to effect a final recovery of sugar (exhaustion of molasses) before centrifuging. Such cooling crystallization is always a slow process.

The above description of the crystallization process applies equally well to batch and continuous operation. The vast majority of sugar crystallization at present is batch operated. Both batch and continuous crystallizer operation will be discussed in greater detail in a later section.

Product crystals from the crystallization step are then dried (by heated air, usually in a rotary drum dryer) and cooled. Product sugar is then shipped (usually by bulk handling) to outside markets.

There are by-products produced. As indicated, the bagasse is usually used in the factory as a fuel, although it may also be used as a source of fiber for pulp or building board. The molasses contains residual sugars and is a suitable feedstock for a fermentation process. It can also be used as livestock food supplement. The clarifier mud is sometimes treated to recover waxes but is usually distributed back to the fields as it contains considerable mineral nutrients. The process also produces a considerable quantity of water from the evaporation. Such process water has a high BOD level and can be a disposal problem.

Refined Sugar Production

The refining process starts with raw sugar. After being weighed, the raw sugar is washed (affinated) to remove adhering molasses from the surface. This washing liquor is treated separately. The affinated

sugar is then completely dissolved (melted). The resulting solution (syrup) is treated similarly as in the raw sugar process by further clarification and filtration, although the solutions involved are now much purer. The solutions are then decolorized by adsorption of impurities on bone char or ion exchange resins. These clarified syrups are boiled and crystallized under vacuum in a manner similar to raw sugar. The dried crystals are screened and packaged for consumption.

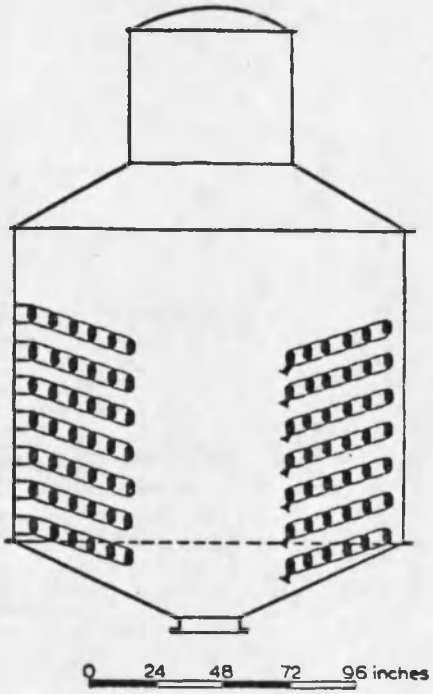
Washwater (sweetwater) resulting, for example, from filter and adsorbent washings, is not discarded. Instead, it is concentrated in multiple-effect evaporators, purified, and recrystallized.

In general, the refining process is similar to the raw sugar process, except for the affination and decolorizing stages. The removal of colorants is an important factor in refinery operation.

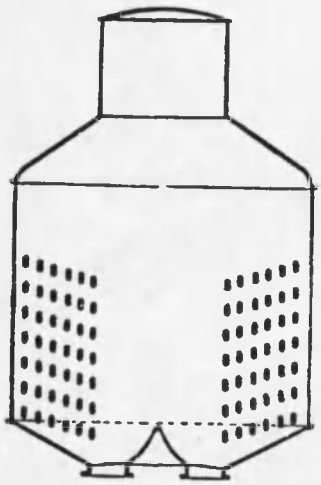
Batch Pan Operation

Batch crystallization takes place in large vacuum vessels (pans) of volumes up to 100 m^3 (25,000 gallons), usually cylindrical in shape. The pans usually contain a steam calandria with a large number of short (3' to 4'), large diameter (4" to 5") tubes. Other designs use steam coils. Figure 2 illustrates several designs. The evaporated water is condensed in a surface or direct contact condenser backed up by a mechanical vacuum pump or steamjet ejector.

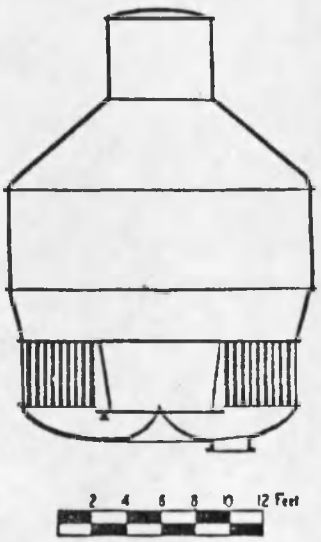
To initiate a batch, a "footing" of seed crystals is added to the pan. This must be sufficient to cover the heating surface, so that on heating the pan contents will circulate and avoid overheating, local dissolution, or "false grain" (nuclei) formation. The footing needs to



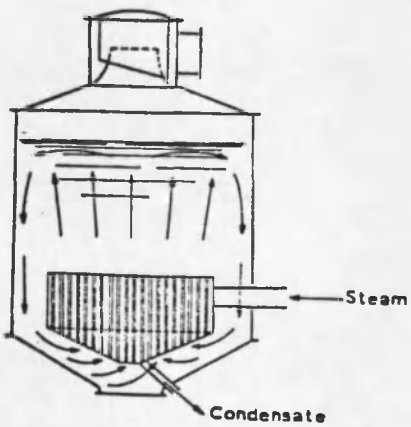
Halpin coil pan



Ditmar Jansse coil pan



Pan with fixed calandria



Pan with floating calandria

Figure 2. Types of Pans. -- After Jenkins (1966).

contain sufficient crystal content so that the crystals do not dissolve completely when liquor is added or with temperature changes due to steam usage and vacuum fluctuations. After boiling and circulation of the footing is established, further liquor is added as evaporation proceeds. Typically, a reasonably constant steam rate is used and the liquor (syrup or molasses) is adjusted to maintain a suitable supersaturation for good crystal growth without nucleation. This supersaturation is an important variable in control of the batch operation and is measured by conductivity, boiling point elevation, or refractive index devices.

As the batch proceeds, crystals grow and the level in the vessel increases. When the batch vessel is full, the contents are "heavied up," i.e., evaporation continues without liquor addition to raise the crystal content as high as practicable. This value depends on the boiling, but is limited by the need to maintain circulation. Boiling must be stopped before the contents become so viscous that they will not circulate. At the end of the batch, the vacuum is broken, and the contents of the vessel are discharged through a large valve at the bottom of the vessel. The pan is washed, cleaned, and readied for the next batch.

Factors desired in a good batch operation are:

1. To obtain the correct footing initially, i.e., correct number and size of seed crystals and crystal content.
2. In operation, to maintain high crystal growth rates without false grain (nucleation).
3. To heavy up to a high consistency, yet avoid loss of circulation.

The contents of a pan must always be kept circulating, so that if, because of scheduling problems, a pan cannot be emptied immediately, evaporation is continued, and "movement" water has to be added to balance the evaporation loss.

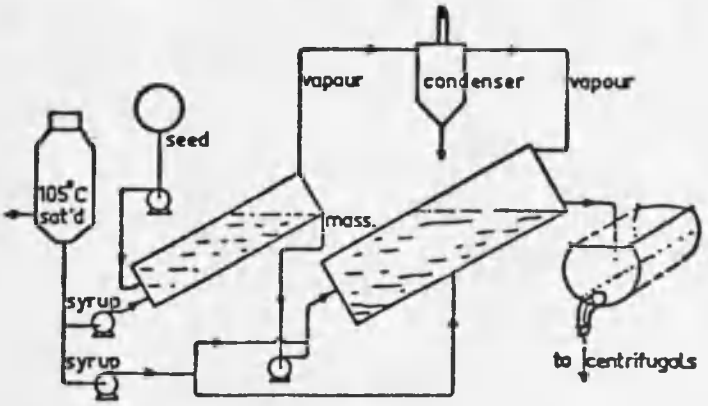
The seed crystals for high-grade boilings are the product crystals from the low-grade boilings (typically 0.3 to 0.35 mm). Seed crystals for low-grade boilings are prepared elsewhere in the factory, usually in special pans (graining pans). Such seeds are produced by supersaturating syrup and adding a quantity of finely ground sugar (ca 10 μm) which is slowly grown to a suitable size for the low-grade pan (typically 0.1 to 0.15 mm). Alternately, self-nucleation initiated by air bubbles, mechanical shocks, etc., can be used in the graining pan, but the results are less reproducible.

Raw sugar crystals tend to be in the size range 0.6 to 1.0 mm, with a mean size of about 0.8 mm.

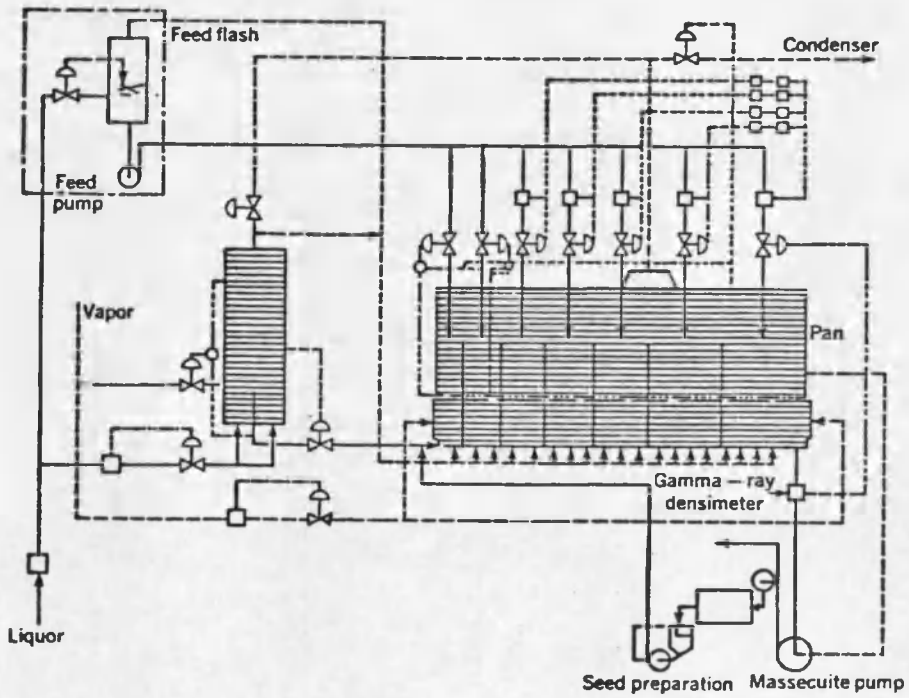
Continuous Pans

Continuous pans have been considered and occasionally implemented in sugar refinery operation. Only a few have been installed for raw sugar production, usually for low-grade boilings. The work to date is summarized by Spencer et al. (1977). Continuous pans have been tested in France (Fives-Lille: Barkow and Witte, 1971); in South Africa (De Vries: Riviere and Pithois, 1976); and in Australia (Steindel et al., 1975). A selection of designs is shown in Figure 3.

A continuous pan is a multicompartmented vacuum vessel. Each compartment is steam heated. Seed and liquor are fed into the first



S.W.S. Continuous Crystallization System. (Dintelood)



Continuous Boiling Plant with Control System. (Fives Lille-Cail patents)

Figure 3. A Selection of Systems Having Continuous Pans. -- After Spencer et al. (1977).

compartment and the mixture flows from compartment to compartment with further evaporation and crystal growth in each. Liquor is added separately to each of the early compartments to maintain a suitable supersaturation and crystal content. "Heavying up" occurs in the last few compartments before discharge from the vacuum vessel through a suitable pump. The product is then centrifuged.

The pan runs under steady conditions. Automatic control is necessary to maintain the feed and steam rates to each compartment. A source of seed crystal is required initially. Periodically, the pan needs to be removed from service to wash fouling from the heat transfer surfaces.

Continuous pans are expected to give a wider crystal size distribution than batch because of the spread of residence times involved. A recent evaluation of an Australian continuous pan design is given by Broadfoot and White (1978).

Although industrial experience with continuous pans is limited, they are reported to work efficiently and to be simpler to operate than batch pans. Further developmental work can be expected.

CHAPTER 3

BASIC CALCULATION METHODS

To design a sugar crystallizer, it is necessary to be able to predict:

1. The recovery of sugar (exhaustion of molasses).
2. The product size distribution.
3. The purity of the crystal product.
4. The material quantities.
5. Steam and energy consumptions.
6. Conditions at various stages throughout the processing.

The basic calculation tools to perform these predictions are:

1. Material conservation relations.
2. Energy balances.
3. The population balance.
4. Rate relations for crystallization, heat transfer, and fluid flow.

These will be discussed in sequence.

Material Balances

The feed liquor entering the crystallization stage consists not only of sucrose and water but a large variety of other compounds also present in the cane plant. These include reducing sugars, inorganics

such as potash and phosphates, amino acids and polysaccharides of various types. A partial list with typical concentrations is given in Spencer et al. (1977, Table 15.1). For calculation purposes, these are all lumped together as "impurities." Certain impurity constituents, however, can have a marked effect on crystallization; for example, dextrose in cane sugar and raffinose in beet sugar. The constituents of the "impurities" will vary from country to country, from factory to factory, and from season to season.

In sugar processing, a liquor is analyzed in terms of:

1. Its dry substance (DS). This is the fraction by weight of dissolved material in the solution.
2. Its purity (true purity, TP). This is the fraction of the dissolved material that is sucrose.

Dry substance and true purity are hard to measure quickly and in the factory the Brix of a solution (based on density or refractive index measurements) is often used in place of DS. This leads to an apparent purity instead of TP. These values approximate to, but differ slightly from, DS and TP. The trend in the sugar industry is to move to DS and TP in reporting results.

For our design purposes, we shall use dry substance (DS) and true purity (TP) throughout.

The syrup coming from the evaporators typically has a DS of 60-68% and a TP of approximately 90%. At the end of the crystallization, the molasses can have a DS of over 90% with a TP of below 40%. Washed product sugar crystals typically have a TP of 97.5%.

To carry out material calculations, balances may be done on sugar (sucrose, S), water (W), and impurities (I). Sugar will be present both in solution and as crystals.

The quantity of crystalline material in the mixture may be expressed as the crystal content, i.e., the fraction of crystal by mass in the slurry. Because of difficulties in separating crystals from molasses, it is usually measured by adding excess water to dissolve the crystals, measure the DS, and back calculate.

Energy Balance

During sugar boiling, the temperature usually does not change significantly. It depends mainly on the vacuum employed. Boiling point elevation changes and superheat variations are usually minor compared to vacuum changes.

For this reason, energy balances are usually only involved at the end of a design to evaluate steam requirements.

The Population Balance

A population balance gives the time and spatial dependence of the size distribution for a group of particles. In crystallization, it is the relationship which allows the crystal size distribution to be predicted.

It may be applied to areas other than crystallization. For example, Pulvermacher and Ruckenstein (1974) said that probably the field where such balances were first formulated was in the coagulation of colloids in 1917. Other examples are: size distribution in the

granulation process (Kapur, 1972), size distribution of crystallites of supported metal (Ruckenstein and Pulvermacher, 1973), continuous emulsion polymerization (Thompson and Stevens, 1977), and solid phase reaction (Bhatia and Perlmutter, 1979).

However, in crystallization, the pioneering work was done by Randolph and Larson (1962) and Hulburt and Katz (1964). They formulated the general population relation which may be written as:

$$\frac{\partial n}{\partial t} + \frac{\partial}{\partial L}(Gn) - D\frac{\partial^2 n}{\partial L^2} + n\frac{d(\log V)}{dt} = -\sum \frac{\vec{Q}_k n_k}{V} + B' - D' \qquad [1]$$

The first term gives the accumulation term of crystal numbers in a given size range, the second and third are the internal "convective" terms due to growth and growth dispersion, while the fourth and fifth are the external "convective" terms due to changes in slurry volume and bulk flow, and the last two are generation terms due to birth and death of particles. Further details on the population balance and its application may be found in the text by Randolph and Larson (1971).

This relation is applicable to batch or continuous crystallizers for transient or steady-state conditions.

For the idealized unseeded MSMPR (mixed suspension mixed product removal) crystallizer, this becomes:

$$\frac{\partial n}{\partial t} + \frac{\partial}{\partial L}(Gn) - D\frac{\partial^2 n}{\partial L^2} + \frac{n}{\tau} = 0 \qquad [2]$$

for the dynamic case, and for the steady state:

$$\frac{d(Gn)}{dL} - D\frac{\partial^2 n}{\partial L^2} + \frac{n}{\tau} = 0 \qquad [3]$$

For a seeded MSMPR (which is relevant in continuous sugar crystallization), the relationship becomes:

$$\frac{\partial}{\partial L}(Gn) - D\frac{\partial^2 n}{\partial L^2} + \frac{n}{\tau} = \frac{Q_i n_i}{V} \qquad [4]$$

For batch vessels with no inflow and outflow of crystals, the relation becomes:

$$\frac{\partial n}{\partial t} + \frac{\partial}{\partial L}(Gn) - D\frac{\partial^2 n}{\partial L^2} = 0 \qquad [5]$$

Where the growth rate, G, is not a function of crystal size, L, i.e., McCabe's delta law applies, G may be taken outside of the parentheses in the above relationships. Under these conditions, the population balance may be converted to a "moment" form. The moments of a distribution with a population density function n(L) are defined by:

$$m_j = \int_0^\infty L^j n(L)\,dL \qquad [6]$$

In its moments form, the general population balance (Eq. 1) becomes:

$$\frac{dm_j}{dt} + m_j\frac{d(\ln V)}{dt} + \sum\frac{\vec{Q}m_j}{V} = jGm_{j-1} + 0^j B^0 + j(j-1)Dm_{j-2} + \bar{B} - \bar{D} \qquad [7]$$

and Equations [4] and [5] become, respectively:

$$\frac{m_j}{\tau} = \frac{Q_i}{V} m_{j,i} + jGm_{j-1} + j(j-1)Dm_{j-2} \qquad [8]$$

$$\frac{dm_j}{dt} = jGm_{j-1} + j(j-1)Dm_{j-2} \qquad [9]$$

These moments are the usual statistical moments about size zero and can be related to physical properties of the distribution such as the mean, standard deviation, surface area, volume, etc. In particular, the third moment can be related to the mass of the crystal by:

$$M_c = \rho_c \frac{\pi}{6} m_3 \qquad [10]$$

provided size L is measured as the volume equivalent size. Thus, the moment relations are convenient forms of the population balance in doing calculations around crystallizers to satisfy material balances and to predict mean sizes and the spread of the sizes. Most of the design methods which follow will use these moment relations solely or at least initially. It should be emphasized again that "size independent growth" is assumed when moment equations are used.

Rate Relations for Crystallization

Use of the population balance requires knowledge of crystallization kinetics concerning growth, growth dispersion, and nucleation.

Growth Rate

The rate of growth, G, of a crystal is defined as the rate of change of linear size, L, i.e.:

$$G \equiv \frac{dL}{dt} \qquad [11]$$

The linear size best suited for crystallization work is the volume equivalent size, i.e., the diameter of a sphere with the same volume as the crystal. Sugar crystal sizes may be measured by sieving (giving a sieve size), by microscopic counting (giving some particle crystal dimension), by individual weighing (White and Wright, 1972), or electro-zone sensing. The latter two methods give the volume equivalent size directly. Other measures of size should be converted to a volume equivalent size.

The growth rate of sugar crystals will depend upon:

1. Supersaturation driving force.
2. Temperature.
3. Amount and nature of impurities.
4. Degree of agitation.
5. Perfection of seed crystals.

In sugar work, supersaturation, which is a measure of solution concentration above saturation, is defined as

$$OS = \frac{(S/W)}{(S/W)^*} - 1 \qquad [12]$$

where OS is the relative supersaturation (termed oversaturation in sugar work); S and W are the mass fractions of sucrose and water in solution, respectively; and the asterisk indicates equilibrium (saturation) conditions.

Before supersaturation can be evaluated, it is necessary first to know solubility data. The solubility of pure sucrose in water is well-established and the data of Charles (1960) is widely used. The effect of impurities depends on the nature of the impurities. The form of equations given in Table 1 (from Wright and White, 1974) is widely used. Taking typical values for cane sugar, solubility curves as shown in Figure 4 are obtained.

Also shown in Table 1 are the equations from Wright and White (1974) which correlate available growth rate data for industrial size crystallizers under industrial agitation conditions. The effects of temperature, supersaturation, and impurity level are included. Figure 4, from Broadfoot and White (1975), shows these data plotted for 65°C.

The correlation of Wright and White (1974) includes the data of VanHook (1969), Smythe (1971), and other workers, together with extensive data obtained industrially in Australia. This shows that the rate of growth is approximately linear with supersaturation. At high temperatures, growth appears to be diffusion controlled, while at lower temperatures it is surface kinetics controlled.

For the design calculations in this thesis, these data will be used.

Table 1. Growth Expressions for Industrial Crystallizers. -- After Wright and White (1974).

Effect	Range	Expressions
Oversaturation (OS)	a) $OS > 1.5\ P_2$ b) $0 < OS < 1.5\ P_2$ c) $OS > 0$	$G = \begin{cases} k(OS - P_2) \\ (k/3)OS \\ 5k(OS) \end{cases}$
Temperature	45 to 75°C	$k = k_{60} \exp\left[\frac{-E_{act}}{R}\left(\frac{1}{T + 273.2} - \frac{1}{333.2}\right)\right]$
		$E_{act} = 11.0 - 0.02\left(T - 60\right) + 8.0\left(\frac{I}{W}\right)$
Impurity (I/W Ratio)	0 to 4	$k_{60} = P_1 \exp\left(P_3 \frac{I}{W}\right)$

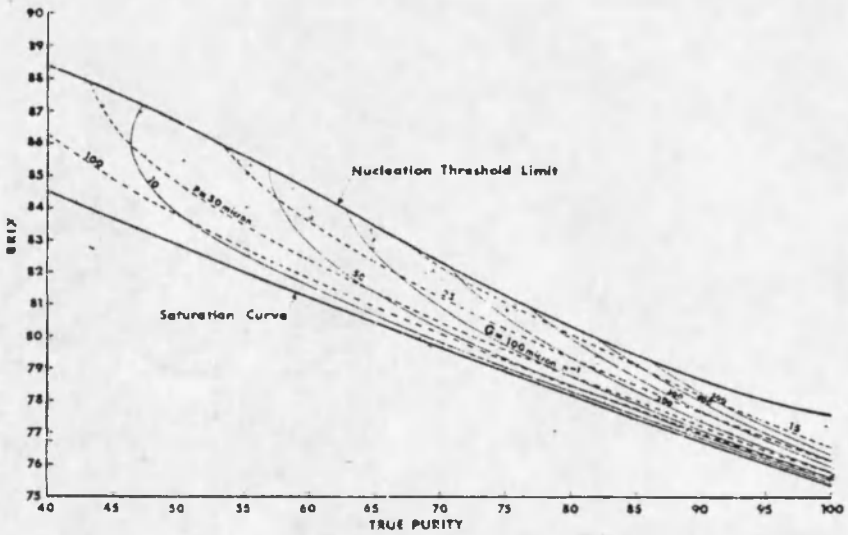


Figure 4. Growth Rate and Size Dispersion Data for Cane Sugar Crystallization at 65°C. -- From Broadfoot and White (1975).

Bennett and Petri (1973) reported results for a pilot plant MSMPR using high purity sugar solutions which gave growth rates much lower than from the above correlation. It may be that their results were affected by considerable crystal breakage in the circulation pump.

A recent laboratory MSMPR study by Hartel et al. (1979), also using pure sugar-water solutions, gave growth rates closer to the correlation used here.

Growth Dispersion

The population balance (Equations 1 to 9) contains a term for growth dispersion. This is a phenomenon resulting from crystals of the same size having randomly different growth rates and thus exhibiting a spread of sizes when grown. It is an important mechanism which limits the ultimate narrowness of CSD in batch processes. The matter was first discussed by Wright and White (1968) and White and Wright (1971), while Randolph and White (1977) adopted this early work in the conventional form of a diffusivity to give the terms as shown in the population balances given above.

There are few data available other than those of White and Wright (1971) and, as recalculated by Randolph and White (1977), the sugar data in the latter paper can be correlated by:

$$\frac{D}{G} = 50 \ \mu m \qquad [13]$$

where D is the growth dispersion parameter as used in Equations 1 to 9 and G is the linear growth rate.

Nucleation

As the concentration of a sugar solution is raised above the saturation level, crystals will grow, but no nuclei will form. This is the metastable region (Miers and Isaac, 1906). As the concentration is raised higher, a value is reached (in the order of OS = 0.3) where substantial nucleation occurs (rates up to 10^5/cc-min). This is the nucleation threshold (Broadfoot and White, 1975) in the presence of crystals. It is considerably lower than the spontaneous nucleation threshold (nucleation in the absence of crystals) reported in early crystallization texts (OS up to 0.5).

That there is a large metastable region where substantial growth rates can be attained and where no nuclei will form is a great convenience which is exploited to obtain a narrow-sized product. All industrial sugar crystallization is carried out in this metastable region. MSMPR studies, such as those by Hartel et al. (1979) must, by their very nature, be conducted outside this region as continuous nucleation was observed and correlated. In batch pan sugar boiling, transgression beyond the nucleation threshold results in massive nucleation ("false grain"). If this occurs, water is immediately added to the batch to undersaturate it to dissolve the nuclei.

If operation is required to be carried out in the metastable region, the values for the nucleation threshold need to be known. Wright and White (1974) give an equation for this limit under industrial conditions as quoted in Table 1. This limit is also in Figure 4 of this report.

Wright and White (1974) and Broadfoot and White (1975) suggest that, in practical designs, operating values be taken at 60-70% of the nucleation threshold supersaturation to give a factor of safety in operation. Crystal growth rate at 65°C at this 70% value, as a function of solution purity, is shown in Figure 5. These will be taken as growth rate values for batch and continuous pans and used in subsequent calculations.

Rates of Heat Transfer and Fluid Flow

To calculate heat transfer rates and thus to determine heating surface area, heat transfer data are required. A considerable amount of data is available (Jenkins, 1966; Spencer et al., 1977). Values depend on the detailed mechanical design. For our present purposes of process design, this aspect is of secondary importance.

It is worth noting, however, that in batch pans it has been traditional to design with a heat transfer surface area per unit volume of vessel of 6 m^{-1} for high-grade and 5 m^{-1} for low-grade liquor. This can be increased to 9.0 m^{-1} in continuous pans but larger values are difficult to achieve.

Rates of fluid flow can be predicted from the usual flow equations. Molasses and molasses-crystal mixtures are somewhat pseudo-plastic. Values of apparent viscosities as a function of temperature, crystal content, DS, and TP are given by Awang and White (1976). This also is a secondary problem for present process design purposes. It is usually taken that vacuum pan circulation will be limited if mixture viscosities exceed 500 P_{as} (Maudarbocus and White, 1978).

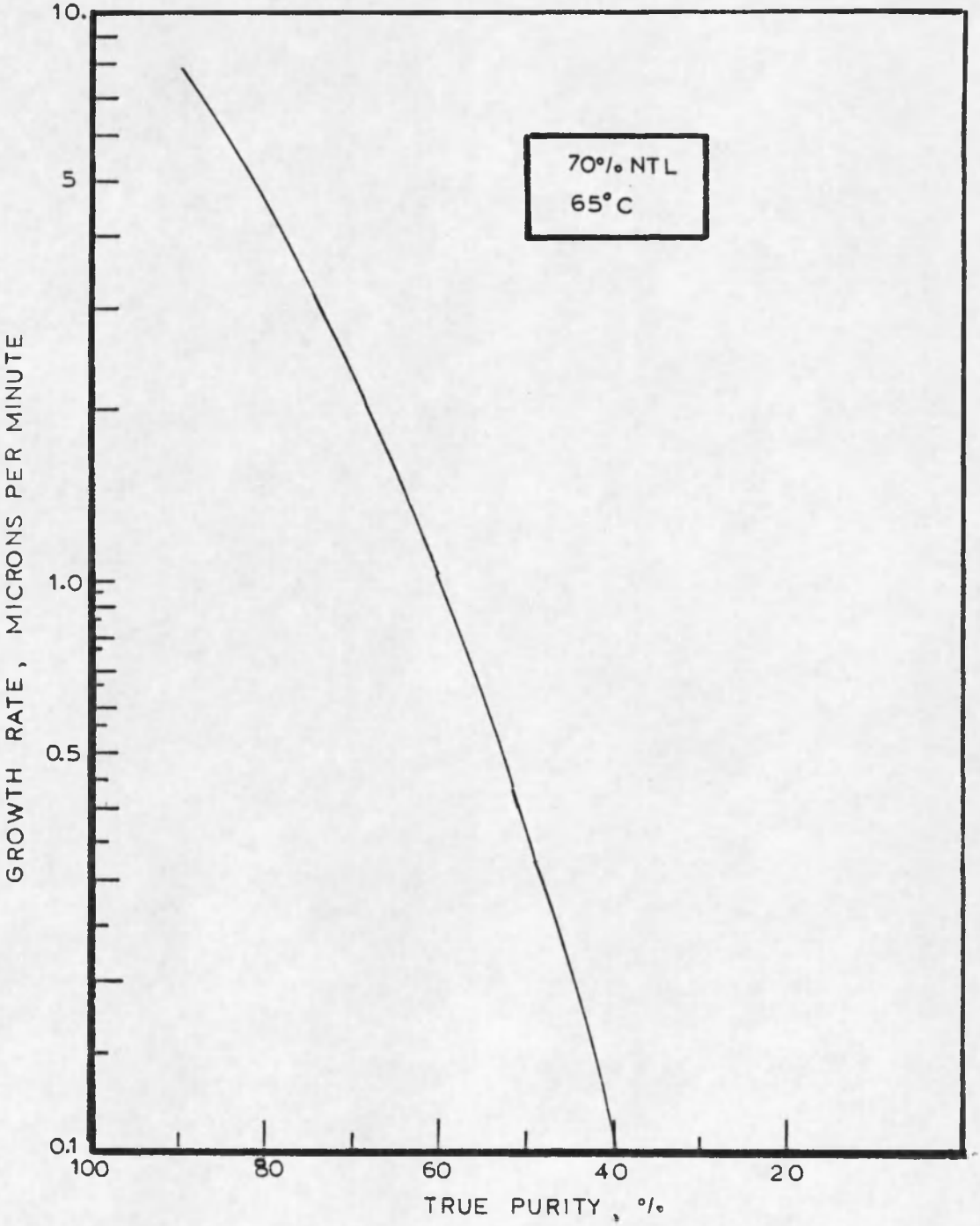


Figure 5. Growth Rate Data. -- After Wright and White (1974).

Design Packages

The combination of the mass, energy, and population balance equations and the rate equations gives a package for designing a crystallizer or for modelling (simulating) performance. Several packages have been developed, usually for a particular type of crystallizer, e.g., batch, continuous, or seeded. In some applications or with certain simplifications, the equations can be solved analytically. In general, however, they have to be solved numerically as a computer program. A number of packages (models, simulators) have been written and will be discussed in the next chapter.

CHAPTER 4

DESIGN METHODS FOR SUGAR CRYSTALLIZERS

The equations given in Chapter 3 can be solved for given crystallizer arrangements. Which population balance relation is used will depend on the particular arrangement involved.

Before discussing particular cases, it is worth noting that the following assumptions are made in all cases:

1. Vessels are well-mixed.
2. The linear growth of a sugar crystal is independent of its size (McCabe's ΔL law holds).

It is not known how well these assumptions hold, but they have been used in all models described in the literature, and will be used here.

Crystallization Station Performance

Also before discussing individual arrangements, it is worth stating explicitly what the requirements are from a sugar crystallization station. These are:

1. Product raw sugar crystals with a mean size about 0.8 mm.
2. A narrow size distribution.

Product with a narrow size distribution separates better from the molasses, has a reduced tendency to cake on storage, and has improved bulk handling properties.

The narrowness of a distribution is expressed by its CV (coefficient of variation), defined as the ratio of σ, the standard deviation of the distribution, to the mean size, $\bar{L}$, i.e.:

$$CV = \frac{\sigma}{\bar{L}} \qquad [14]$$

Typically, raw sugar produced for export has a CV (based on the mass size distribution) of 0.20.

This criterion of a narrow size distribution is an important one when considering continuous crystallization. Avoiding further nucleation, using equipment with a near plug flow residence time distribution, and introducing size classifiers into the process are methods which will be considered for minimizing the size spread.

3. High recovery -- As sugar is a valuable product ($300/ton), as much sugar should be recovered as economically possible. With a syrup of true purity (TP) of 90% and a final molasses of TP = 40%, the recovery of sugar can be calculated to be over 92%. This is typical of modern crystallization stations.

Batch Pans -- General Method

For a batch pan, Wright and White (1968, 1974) used the moments form of the unsteady population balance (Equation 9) and solved these (for four moments) numerically over small time intervals in conjunction with the material balance. This gave the growth rate, the mean sizes, and the spread of sizes as a function of time. For design purposes, it allowed maximum growth rates as shown in Figure 5 to be used and the

liquor and steam rates to achieve these conditions to be calculated. A digital solution was necessary with up to 50 time increments for a complete batch boiling. They report little expected round-off error with the stepwise procedure.

Their simulation agreed well with factory trials, and the model (Batterham, Frew, and Wright, 1974) has been used:

1. For simulation of a factory batch pan.
2. For parameter estimation to determine kinetic data for both factory and laboratory experiments.
3. For the optimum design of a batch pan.
4. For devising optimal operating procedures for pan operation.

Batch Pans -- Simplified Method

The general method used by Wright and White requires digital computer solution. If, however, the growth rate, G, is constant during a batch, the step-by-step procedure is not necessary since the overall equations can be solved between initial and final conditions. This may be simply done using hand calculations.

This is the method used in this thesis. To the author's knowledge, it has not been used previously for sugar crystallizer design.

In actual practice, the growth rate will decrease during the batch as the concentration of impurities in solutions increases. During boiling, both water and sugar are removed from solution. For optimal design, the growth rates should follow the curve given in Figure 5.

The problem with this method is to pick an average growth rate to use. As a first estimate for optimal design, the log mean of the

growth rates at the initial and final impurity concentrations might be used. As a service to users, the general model could be solved to determine how accurate this estimate is.

If the time variation of the growth rate is known, the average is given by:

$$G_{ave} = \frac{1}{t} \int_0^t G(t)dt \qquad [15]$$

Continuous -- The Unseeded Well-Mixed Tank

The unseeded self-nucleating MSMPR without growth dispersion is the classic example in crystallization studies (Randolph and Larson, 1971). The product size distribution on a number basis is exponential and on a mass basis is a third-order gamma distribution. The mean size depends on the product of the growth rate and the residence time. The size spread is very large (CV = 0.50 on a mass distribution basis). An extensive analysis of this crystallizer is given in the text quoted.

To operate, an unseeded MSMPR must generate nuclei, that is, must operate beyond the metastable region (above the nucleation threshold). This is not necessary if a substantial number of nuclei are generated by crystal attrition, as perhaps was the case with the experiments by Bennett and Petri (1973). The results of recent laboratory MSMPR studies (Hartel et al., 1979) all required concentrations beyond the nucleation threshold.

The self-nucleating MSMPR could be used as a nuclei generator, as has been proposed by Randolph and Ziebold (1974) and the design for

this purpose will be considered in Chapter 6. Because the nuclei generated will cause a wide size distribution, this development would not be used for the main crystallizer operation.

Continuous -- The Seeded Well-Mixed Tank

The continuous well-mixed tank crystallizer can be operated in the metastable region if it is seeded. This unit is at the heart of continuous sugar pans. Seeds are continuously added and conditions are adjusted to remain in the metastable region.

The design calculations are most simply performed using the moment relations (Equation 8). Once the exit concentration is chosen, the growth rate is known, for the crystallizer contents are the same as the exit conditions. Broadfoot and White (1975) and Randolph and White (1977) provide charts based on a solution of the moment equations which show how CV will change for a single seeded crystallizer, as a function of growth rate and growth dispersion. If the full size distribution is required, this can be obtained by numerical solution of Equations 3 and 4. The Mark 1 model of The University of Arizona is a computer package which performs these calculations.

There are two weaknesses with the seeded well-mixed tank crystallizer. It still gives an excessively wide size distribution, and also growth rates will be very low and crystallizer volume large.

Methods of Narrowing the Size Distribution from Continuous Crystallizers

The product size distribution will be narrowed if:

1. There is no further nucleation in the crystallizer (i.e., stay in the metastable zone with seeded crystallizers).
2. Multi-tank or multi-compartment vessels are used. The residence time distribution for tanks in series is much narrower than for a single tank, and this should cause a somewhat similar reduction in the size distribution.
3. The product crystal is sieved and the fines are either recycled for further growth or are destroyed.

Continuous Multi-Tank

Randolph, Deepak, and Iskander (1968) showed the improvement in narrowing of CSD which would result from using crystallizers in series. With no growth dispersion, with equal increments of size in each vessel, and with a fine seed fed (or nucleation occurring) in the first tank, the CV of the product distribution drops as the inverse of the square root of the number of tanks. With growth rate dispersion, the distributions are not as narrow (Randolph and White, 1977).

To model a series of tanks is merely a matter of applying the seeded single tank model several times in succession. For a large number of tanks or compartments, this is usually done using the moment relations to minimize computation time (Randolph and White, 1977; Broadfoot and White, 1975). Broadfoot and White give a series of charts by which the calculations for each tank can be solved graphically.

The problem with the multi-unit crystallizers is to choose the intermediate conditions. This could be done by allocating equal increments in size for each vessel. In this case, much more mass and, therefore, evaporation would be occurring in the last vessel under conditions where the growth rate is lowest. An alternative is to undertake equal increase in crystal mass in each vessel.

Another alternative is to define an objective function and to find the optimum choice which minimizes the objective function. Larson and Wolff (1971) show the technique for two vessels in series with nucleation. Broadfoot and White (1978) have performed such calculations for up to 10 seeded tanks in series for sugar. Their objective function was to minimize the total crystallizer volume required. As well as selecting the cut point between vessels, they also optimized on the distribution of the addition of liquor to each unit and the number of units for heavying up. Their design used a single vessel with internal compartments, and the final crystallizer volume was of the same order as that of the corresponding batch vessels.

Continuous-Product Classification

Screening the product (and if necessary intermediate streams also) is a direct way of narrowing the size distribution. Returning the fines to the crystallizer, however, does have certain consequences on crystal content which are discussed by Randolph and Larson (1971).

The application of product classification to continuous sugar crystallization was studied by Randolph and White (1976). To apply classifier efficiency curves, it is necessary to determine the full

crystal size of the distribution, rather than the moments. The size-distribution population balance relation (Equations 3 and 4) must be used in general. Further, a trial-and-error procedure is required if crystals are recycled to an earlier tank. Randolph and White used Program SHUG which performed these calculations and the iterations.

Because of considerable computation time involved, they limited the calculations to two crystallizers in series with return of the classified fines to the first vessel. They showed that a significant improvement in the spread of the size distribution could be obtained by classification with only a modest amount of recycle.

To allow a larger number of vessels in series to be analyzed, Randolph and Tan (1978) solved the moment equations for the crystallizers in series and reconstructed the full distribution before the classifier by assuming it was a gamma distribution with the same moments as the product. This is an approximation but a reasonable one and it does speed up the calculations considerably. This program, entitled RECYC, is available at The University of Arizona.

Randolph and Tan (1978) applied the model to continuous sugar crystallization and showed the effect of the number of stages and the classifier cut size on the product CV and the recycle ratio.

This program will be used for the design of a continuous crystallizer described in Chapter 8.

Of considerable difficulty in practice is to find an economic means of classifying sugar crystals, particularly raw sugar. This is an area for fruitful research.

Cooling Crystallizer

All the above modelling applies to evaporative crystallizers which operate at substantially constant temperature (usually about 65°C). These are the major crystallizers in the sugar process (see Chapter 2). However, cooling crystallizers are used in a minor role to extract the last remaining sugar from a thickened crystal-final molasses mixture.

To model a cooling crystallizer, the moment form of the population balance is appropriate together with material and energy balances. Kinetic data over a range of temperatures is also required. Maudarbocus and White (1978) have performed calculations for such cooling crystallizers for sugar and have determined the optimum procedures for such cases.

Conclusion

Models exist for most of the arrangements likely to be considered for sugar crystallization. For the purposes of illustrating and comparing design methods, the following arrangements will be considered in later sections:

1. The unseeded MSMPR, in conjunction with mixed vessels for the nucleator/ripener (Chapter 6).
2. Batch pans using the simplified moment method (Chapter 7).
3. Continuous pans with product classification (using Program RECYC, Chapter 8).

Specific examples will be considered.

CHAPTER 5

OVERALL DESIGN CRITERIA AND OVERALL BALANCES

The object of the present study is to detail the method of calculation for the design of a sugar crystallization station. The calculations will be performed for two designs:

1. Batch pans with a continuous seeder.
2. Continuous seeded pans with external classification and recycle.

Particular Case

As a basis for further calculation, the following design specifications are given:

1. Production rate = 800 tons per day of sugar.
2. Product crystals must have a mass mean size of 800 μm.
3. Syrup is available from the evaporator station at 68% DS and 90% TP.
4. The recovery of sugar should exceed 90%.
5. Seed crystals are to be produced by alcohol precipitation in a continuous seeder of the type described by Randolph and Ziebold (1974). The seeds will be grown to a mass mean size of about 160 μm.
6. Crystallization will be done under vacuum at a temperature of 65°C.

Design Data

The correlation for solubility, growth rates, and nucleation thresholds discussed in Chapter 3 will be used.

Organization

A design for the continuous seeder will be given in Chapter 6, for the batch pan in Chapter 7, and for the continuous pan with classification in Chapter 8.

The same continuous seeder will be used for both the batch and continuous pans.

Overall Material Quantities

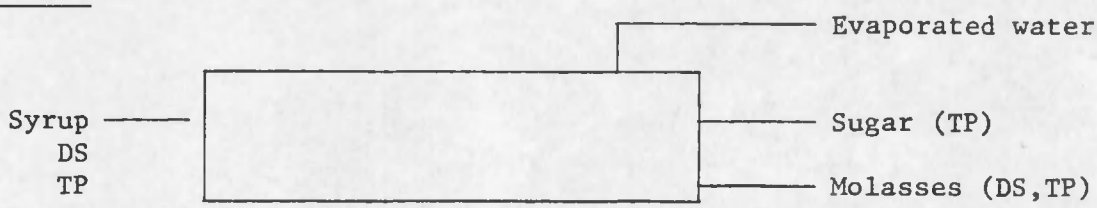
An overall material balance can be performed as shown in Figure 6. Balances are done on water, sugar, and impurity. In the same figure, it can be seen that, for every ton of sugar produced, 1.72 tons of syrup are required, 0.184 ton of molasses is produced, and 0.521 ton of water has to be evaporated in the crystallizer station. Thus, up to one ton of steam will be required for evaporation and 30 tons of cooling water to condense it.

Number of Crystals

An important aspect in the design of crystallizers is to control the number of crystals. A given mass is to be deposited. If too many nuclei are provided, the crystals will be small; if too few, large.

The crystal number rate is evaluated on Figure 6. This number is in the order of 2 trillion per day. If the seeds are to be 100 μm ($\bar{L}_{3,0}$), then the mass rate of production will be 1.9 tons per day.

Flowsheet



Assumptions

a. Final molasses (40% TP, 86% DS).
b. Recovery of sucrose (92.5%).
c. Raw sugar (97.5% TP).
d. Evaporator syrup (90% TP, 68% DS).

Material Quantities

Basis = 1 day; unit = tons.

	In				Out				
	Total	S	I	W	W	I	S	Total	
Syrup	1.72	1.05	0.12	0.55	0.0	0.025	0.975	1.00	Sugar
					0.026	0.095	0.063	0.184	Molasses
					0.536	0.0	0.0	0.536	Water

Approximate Number Rate

Tentatively assume the volume number mean size, $\bar{L}_{3,0}$ of the distribution is about 750 microns. To this volume equivalent size, the mass of one crystal

$$= 1.57 \times \frac{\pi}{6} \times (0.075)^3 = 3.5 \times 10^{-4} \text{ grams}$$

$$\therefore \text{ approximate daily number rate} = \frac{800 \times 10^6}{3.5 \times 10^{-4}} = 2.3 \times 10^{12} \ \#/\text{day}$$

Seed Rate

If the seeds have an $\bar{L}_{3,0}$ = 100 μm, the seed mass rate will be

$$2.3 \times 10^{12} \times 1.57 \times \frac{\pi}{6} \times (0.01)^3 \times 10^{-6} = 1.9 \text{ tons/day}$$

Figure 6. Overall Material Balances.

The exact value of the number rate will depend on the shape of the product size distribution. If mass mean size $\bar{L}_{4,3}$ is specified, the calculations are based on a volume-number mean size $\bar{L}_{3,0}$ and these will differ for different distributions.

However, the present values will be used for the initial design of the continuous nucleator. The design may then be adjusted to the exact final number rate when the final product size distribution is known.

CHAPTER 6

CONTINUOUS SEEDER

A source of nuclei is necessary. Traditionally, processes have used ball milled sugar or icing sugar.

For the present design, however, the continuous seeded developed by Randolph and Ziebold (1974) will be used for both batch and continuous pans. It has the advantage of continuously producing reproducible seed of good crystal shape. For the continuous pan, the seeds will be fed continuously into the first crystallizer. For the batch pan, the seed will need to be stored for use as required.

The continuous seeder will consist of three well-mixed tank crystallizers (Figure 7). In the first tank (the nucleator), the nuclei are formed by salting out of syrup with alcohol. From the work of Randolph and Ziebold (1974), the mean size (population basis) of the product crystals will be about 20 μm. Since seed crystals are required with a mean size of about 160 μm (see Chapter 5), further growth is required. This will be carried out in the two remaining vessels (ripeners). In these vessels, further syrup is added, and evaporation occurs under vacuum, causing further growth. The bulk of the alcohol is removed in the first evaporation, and could be recovered from the condensate.

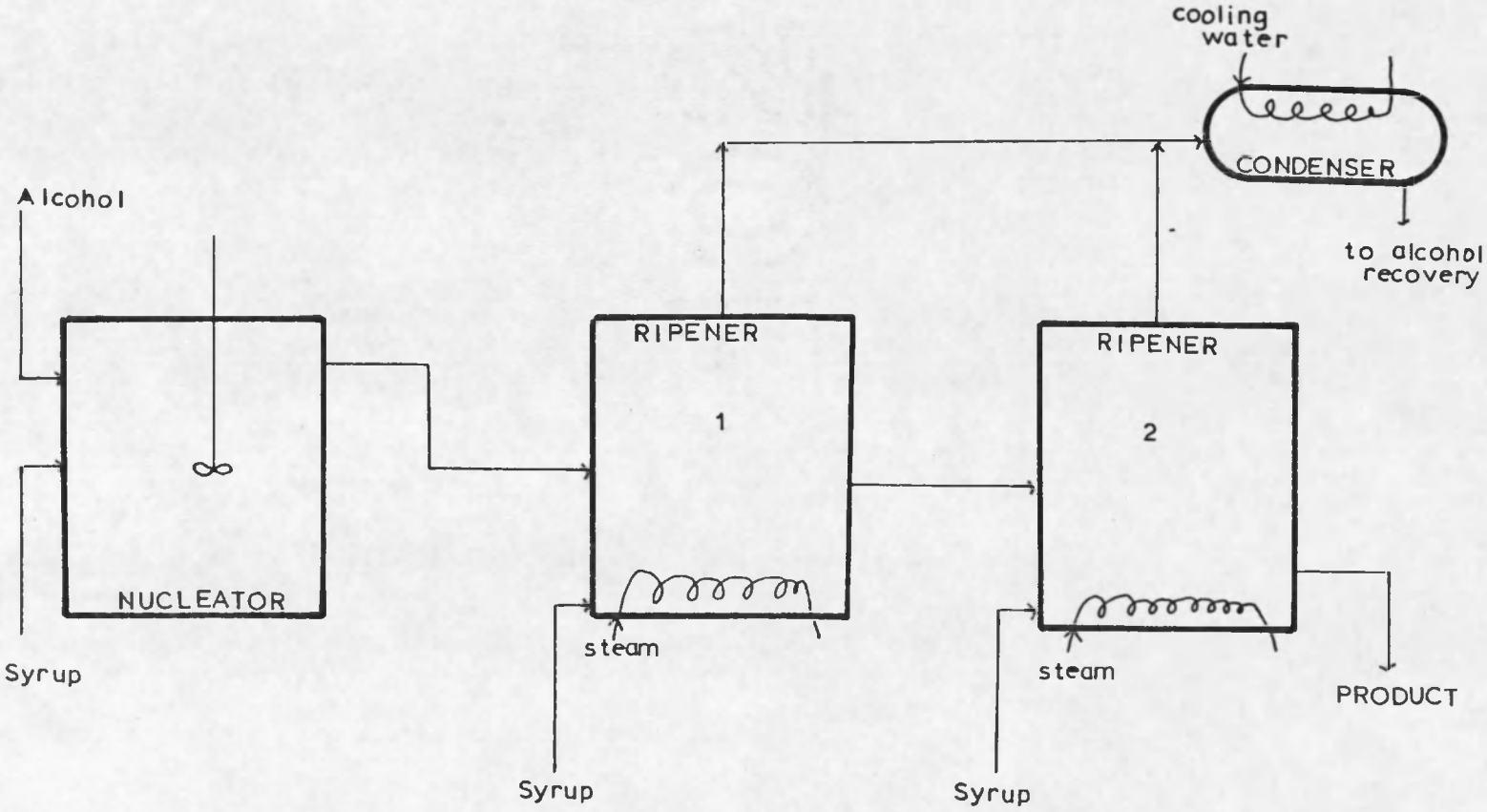


Figure 7. Process Flow Diagram for the Continuous Seeder.

Nucleator

Specification

The nucleator must produce crystals at the prescribed rate of 1.6×10^9 #/min (see Chapter 5).

The following input variables must be determined:

1. Size of vessel.
2. Operating conditions, e.g., P, T, agitation.
3. Feeds (flow rates and conditions).

From these follow the dependent variables:

1. Production rate.
2. Product size distribution.

Choice of Conditions

The amount of design data available on the continuous precipitation of sugar by alcohol addition is limited (Randolph and Ziebold, 1974; Crawford, 1970). Conditions used by these investigators will be used in this design; thus:

1. Temperature -- ambient, 27 ± 1°C.
2. Pressure -- ambient, 1 atm.
3. Alcohol -- industrial spirit (~ 95% by weight), at approximately the same rate (volumetric) as the syrup.
4. Syrup -- ex., evaporator (68% DS, 90% TP).
5. Residence time -- approximately 50 minutes.

The syrup used will be that available from the evaporators. This differs a little from the higher purity syrup used by the above

investigators. It will be assumed, however, that this does not affect the mean size of the nuclei produced to a significant extent.

Design Method

The nucleator is an unseeded self-nucleating continuous tank crystallizer (MSMPR). The size distribution will be exponential on a number basis with a mean size $G\tau$. where G is the growth rate and τ is the mean residence time. Randolph and White (1977) have shown that with growth dispersion the above is still true, though an apparent G allowing for growth dispersion must be used.

The measurements reported above were performed under MSMPR conditions, so the growth rates are "apparent" values. Thus, the MSMPR design procedure may be used. As discussed in Chapter 4, this is quite straightforward and the equations are amply described in the text by Randolph and Larson (1971). The moment equations (derived from the population balance) are repeated in part A of Table 2.

Since the conditions are chosen to be identical to those used by Crawford (1970), his results may be scaled up directly, i.e., the mean crystal size, $\bar{L}_{1,0}$, will be 20.5 μm and the nucleation rate will be 3.2×10^4 #/cc slurry-min.

Calculations

1. Nucleator working volume $= \dfrac{\text{Required nucleation rate}}{\text{Crawford's rate}}$

$$= \frac{1.6 \times 10^9}{3.2 \times 10^4} = 5 \times 10^4 \text{ cc}$$

$$= 13.2 \text{ gallons} \simeq 49.9 \text{ liters.}$$

Table 2. Moment Equations and Other CSD Design Expressions for N/R Model.

Moment Equations		CSD
(A) Nucleator: $G\tau = a$	Mean Size:	$\bar{L}_{1,0} = a$
$m_{1,0} = 1.0$		$\bar{L}_{3,0} = (6a^3)^{1/3} = 1.817\ a$
$m_{1,1} = a$		$\bar{L}_{4,3} = 4a$
$m_{1,2} = 2a^2$	Mode:	$\bar{L}_d = 3a$
$m_{1,3} = 6a^3$	CV:	1.0
⋮ ⋮		
(B) Ripener 1: $G\tau = b$	Mean Size:	$\bar{L}_{1,0} = a+b$
$m_{2,0} = 1.0$		$\bar{L}_{3,0} = 1.817 \left[(a+b)(a^2+b^2) \right]^{1/3}$
$m_{2,1} = a+b$		$\bar{L}_{4,3} = 4 \left[\frac{a^3+b^3}{a^2+b^2} + \frac{b^2a^2}{(a+b)(a^2+b^2)} \right]$
$m_{2,2} = 2(a^2+ab+b^2)$		
$m_{2,3} = 6(a^3+a^2b+ab^2+b^3)$	Mode:	$\bar{L}_d = 3\left(\frac{a^3+a^2b+ab+b^2}{a^2+ab+b^2} \right)$
⋮ ⋮		
	CV:	$\left\{ \frac{2(a^2+ab+b^2)}{a^2+2ab+b^2} - 1 \right\}^{1/2}$

Table 2, Continued.

Moment Equations		CSD	
(C) Ripener 2: $G\tau = c$	Mean Size:	$\bar{L}_{1,0}$	$= a+b+c$
$m_{3,0} = 1.0$		$\bar{L}_{3,0}$	$= (a^3+a^2b+ab^2+b^3+a^2c+abc+b^2c+ac^2+bc^2+c^3)^{1/3}$
$m_{3,1} = a+b+c$		$\bar{L}_{4,3}$	$= 4\left(\frac{(a^2+b^2+c^2)(a+b+c) + (abc)}{(a^3+b^3+c^3)(a+b+c) + (abc)(b+c) + a^2(b^2+bc+b^2+b^2+c^2)}\right)$
$m_{3,2} = 2(a^2+b^2+c^2+ab+ac+bc)$			
⋮ ⋮	Mode:	$\bar{L}_d$	$= 3\left(\frac{a^3+a^2b + \ldots}{a^2+ab + \ldots}\right)$
	CV:		$\left\{ \frac{2(a^2+b^2+c^2+ab+ac+bc)}{(a^2+b^2+c^2) + 2(ac+ab+bc)} - 1 \right\}^{1/2}$

2. Exit flow rate = $\frac{13.2}{48.8} = 0.27 \approx 1.02$ liters/min.

3. Crystal size distribution -- using part A of Table 1 with a = 20.5 μm, the results are shown in Table 3.

4. Crystal mass rate = (Crawford's yield) $\left(\frac{\text{cc slurry}}{\text{min}}\right)$

$$= (0.18)\left(\frac{5 \times 10^4}{48.8}\right)$$

$$= 1.84 \times 10^2 \text{ gm/min}$$

$$= 0.29 \text{ ton/day} \approx 263.3 \text{ kg/day}.$$

5. Crystal content = 0.29/(0.27)(6) = 0.18 gm/cc slurry.

6. Material balance -- basis = 1 hr; unit = kg.

	In					Out					
	Total	A	S	I	W	A	S	I	W	Total	
(F_2) alcohol	23.9	22.7	0	0	1.2	22.7	13.9	2.8	14	53.6	Liquor (L) } Prod.
(F_1) syrup	40.7		24.9	2.8	13	0	11.0	0	11	11.0	Crystal (X) } Prod.

These two flow rates are not exactly equal as suggested by Crawford (1970), possibly because of differences in the solubility data.

These calculations were performed using the alcohol-sucrose-water solubility diagram at 27°C shown in Figure 8. It is assumed that the residual supersaturation will be small, i.e., this is a high yield (or Class II) system, as shown by Randolph and Ziebold (1974).

Table 3. Design Data and Results for Continuous Seeder.

Parameter	Nucleator	Ripener 2	Ripener 2
Chosen Variables			
Growth rate, μm/min	0.42	2.00	3.00
Characteristic size, $G\tau$, μm	20.50	23.00	33.00
Crystal content, gm/cc slurry	--	0.12	0.12
Temperature, °C	27±1	65	65
Feed supersaturation, S_F, gm/cc	0.27	--	--
Calculated Results			
CSD			
$\bar{L}_{4,3}$, μm	82.0	108.5	158.3
$\bar{L}_{3,0}$, μm	37.3	62.8	100.0
$\bar{L}_{2,0}$, μm	29.0	37.7	88.8
$\bar{L}_{1,0}$, μm	20.5	43.5	76.5
CV { wt basis	0.50	0.48	0.43
CV { pop basis	1.00	0.71	0.59
Mass Rates			
Production, kg/day	263.3	426.8	1716.1
Solid concentration, M_T, gm/cc slurry	0.18	0.12	0.12
Exit rate, liters/min	1.02	2.46	9.84
Syrup rate, liters/min	0.51	2.66	10.70
Evaporation rate, liters/min	--	1.22	3.32
Dissolved sucrose (clear liquor), %	--	70	76
Oversupersaturation, OS	--	1.16	1.18
Volume			
Residence time, min	48.8	11.5	11.0
Slurry volume, liters	49.9	28.4	108.3

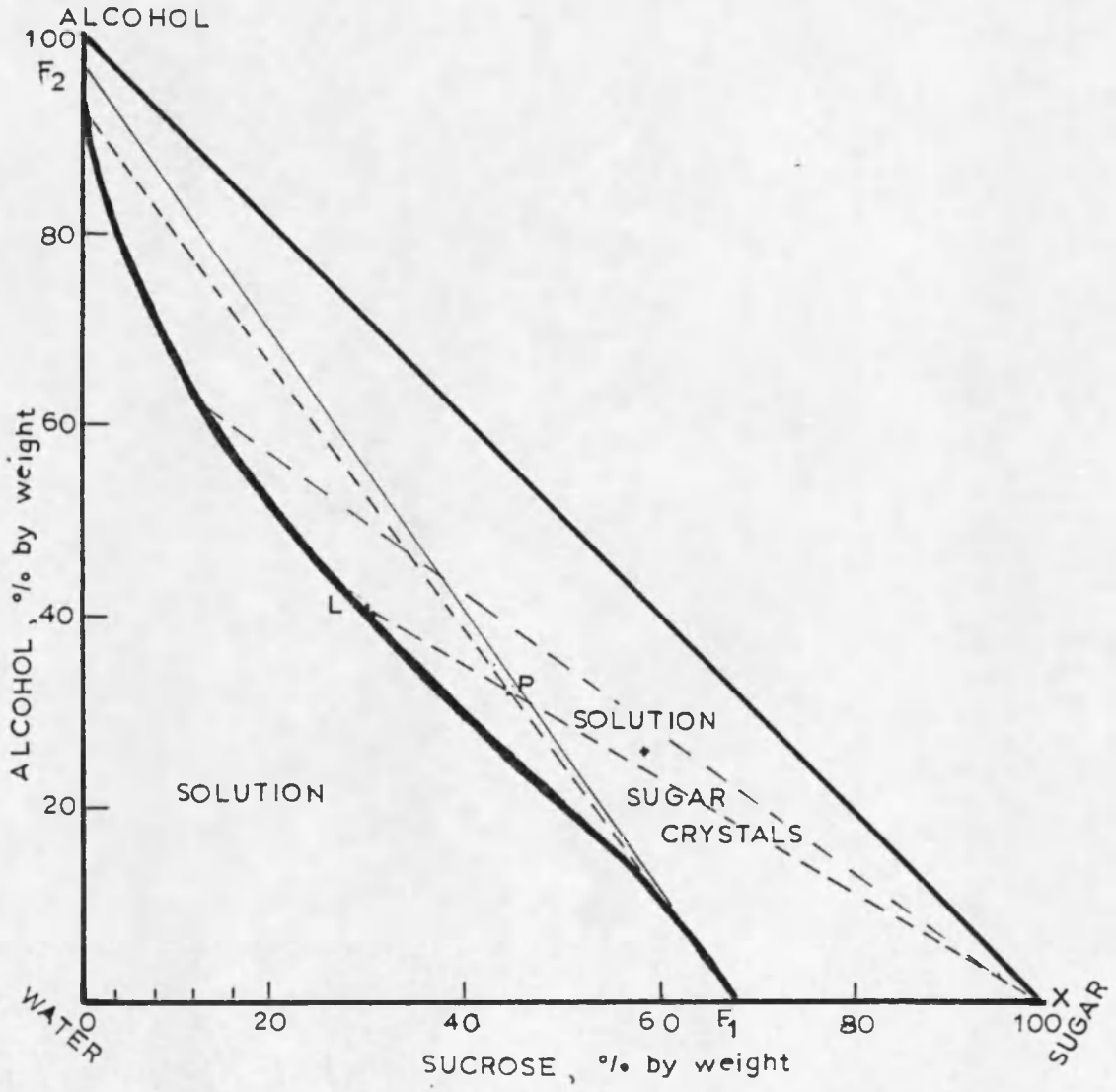


Figure 8. Solubility Diagram. -- After Spencer et al. (1977).

The symbols F_1, F_2, L, X, and P shown refer to the streams identified in the tabulated material balance.

Agitation Rate

Using the Zwietering equation for the suspension of solid particles in a stirred tank (Perry and Chilton, 1973), the power input required can be estimated to be 0.022 hp.

A summary of the design data and results for continuous seeder are shown in Table 3.

Ripeners

Specification

Both ripeners will be operated as vacuum evaporative crystallizers. It is assumed that they will be operated under conditions giving no further nucleation; thus, the crystal number production rate will still be 1.6×10^9 #/min.

The design procedure must evaluate the following:

1. Size of vessels.
2. Operating conditions, i.e., P, T, agitation.
3. Syrup and steam rates.

From which the following can be determined:

1. Production rates.
2. Crystal size distributions.

Choice of Conditions

Temperature. Boiling temperature should be kept below 70°C to avoid caramelization of the sugar. In practice, 65°C is readily maintained and this value will be used here. A vacuum of 24 in. Hg would be sufficient to maintain this temperature (the elevation of boiling point is usually less than 10°C).

Feeds. The feed slurry comes from the previous vessel. Syrup, if required, is available from the evaporators (68% DS, 90% TP). Low pressure steam (< 50 psig) is also available.

Growth Rates. Most of the alcohol will evaporate in the first ripener and the amount left in solution should not greatly affect growth conditions. The growth rate correlation described in Chapter 3 is applicable.

For efficient design, the growth rate should be as high as possible without causing nucleation. A design value of G = 2 μm/min is chosen for the first ripener and G = 3 μm/min for the second. The smaller value in the first ripener is to allow a factor of safety for the effects of any residual alcohol.

Amount of Growth. The final product seed should have a mass mean size of about 160 μm. Two ripening vessels have been chosen instead of one to give a narrower size distribution and a smaller total crystallizer volume. The results given by Broadfoot and White (1975) and Randolph and White (1977) indicate that, with growth dispersion, the

distribution will be narrowest if the increase in number mean size is about the same as that of the crystal entering. Thus, for the first ripener, an increase in $\bar{L}_{1,0}$ of 23 μm is chosen. The increase in the second ripener will be such to give the desired product size.

Crystal Content. The crystal content should not be too low or a rise in temperature (e.g., due to a loss of vacuum) will dissolve all the seeds. A 10°C rise in temperature will dissolve 0.02 g of crystal per cc of solution. To provide some safeguard against such occasional variations, a crystal content of at least 0.12 g/cc of slurry will be used in each ripener.

Design Method

Each ripener is a continuous seeded, well-mixed tank crystallizer, operated without nucleation. The methods of modelling such crystallizers were described in Chapter 4.

The simplest approach is to solve the moment equations first of all, together with the material balances. Then, if required, the full size distribution can be found using the full population balance.

Initial calculations were carried out including the growth dispersion effects. Growth dispersion caused very large changes to the size distribution, and these were considered not to be reasonable. The cause of this was the very small crystal sizes involved in this seeder. The experimental investigations of White and Wright (1971) used much larger crystals, and it may not be appropriate to extrapolate it to small sizes.

All further ripener calculations were carried out omitting the growth dispersion effects. The moments equations used are given in Table 2.

Calculations for First Ripener

1. Resident time -- for the first ripener, $\Delta\bar{L}_{1,0} = G\tau = 23$ μm, $\therefore \tau = 23/2 = 11.5$ min.
2. Moments -- using the relations of part B of Table 2, where a = 20.5 and b = 23, the moments of the product size distribution can be calculated. These results are shown in Table 3.
3. Crystal mass rate -- from $\bar{L}_{3,0} = 62.8$ μm, the mass rate of crystals $= 2.3 \times 10^{12} \times 1.57 \times \frac{\pi}{6} (0.00628)^3 \times 10^{-6}$

   $\simeq 0.47$ ton/day $\simeq 426.8$ kg/day.
4. Exit flow rate -- for a crystal content of 0.12 gm/cc of slurry, the exit flow rate $= \frac{0.47}{(6)(0.12)} = 0.65$ gpm $\simeq 2.5$ liters/min.
5. Crystallizer working volume = (0.65)(11.5) = 7.5 gallons $\simeq$ 28.4 liters.
6. Material balance -- basis = 1 hr; units = kg.

		In				Out				
		Total	S	I	W+A	W+A	I	S	Total	
Nuc.	Liquor	53.6	13.9	2.8	36.9	43.5	17.2	136.9	197.6	Liquor } Product
	Crystal	11.0	11.0	0	0	0	0	17.8	17.8	Crystal }
	Syrup	212.1	129.8	14.4	67.9	61.3	0	0	61.3	Evaporation

The supersaturation of the product liquor = 1.15 is taken from the growth rate correlation given in Chapter 4. Hence:

syrup feed rate = 0.7 gpm ≃ 2.66 liters/min;

evaporation rate = 65.4 kg/hr ≃ 0.32 gpm; and

steam rate = 72.17 kg/hr.

7. Heat transfer area (based on U = 100 Bh/hr-ft^2-°F and ΔT_L = 50°F):

$$q = UA\Delta T_L$$

$$A = \frac{(72.17)(2.2)(954)}{(100)(50)} = 7 \text{ ft}^2 .$$

Heat transfer area/volume ratio = $\frac{7}{(7.5)(7.48)} = 7 \text{ ft}^{-1}$.

8. Summary of results -- refer to Table 3.

Calculations for Second Ripener

1. Amount of growth -- since $\bar{L}_{4,3} \simeq 160$ μm, the moment equations for $\bar{L}_{4,3}$ (part C, Table 2) can be solved by trial and error to obtain C (=Gτ), the amount of growth in the second ripener. Thus, C = 33 μm.

2. Crystal size moments -- using the other moments relation of Table 2, CV and other distribution properties shown in Table 3 can be calculated.

3. Crystal mass rate = $2.3 \times 10^{12} \times 1.57 \times \frac{\pi}{6} \times (.01)^3 \times 10^{-6}$

= 1.89 tons/day = 1716.1 kg/day.

4. Exit flow rate (for crystal content of 0.120 gm/cc slurry) = $\frac{1.89}{6 \times 0.120}$ = 2.6 gpm ≃ 9.8 liters/min.

5. Ripener volume:

$$\text{Residence time} = \frac{G_T}{G} = \frac{33}{3} = 11 \text{ min.}$$

$$\text{Working volume (slurry)} = (2.6)(11) = 28.6 \text{ gallons}$$

$$\simeq 108.3 \text{ liters.}$$

6. Material balance -- basis = 1 hr; unit = kg.

		In				Out				
		Total	S	I	W	W	I	S	Total	
Ripener 1	Liquor	197.6	136.9	17.2	43.5	114.6	75.3	605.9	795.8	Liquor } Prod.
	Crystal	17.8	17.8	0	0	0	0	71.5	71.5	Crystal } Prod.
	Syrup	854.1	522.7	58.1	273.3	202.2	0	0	202.2	Evaporation

Hence, syrup addition rate = 2.8 gpm and steam rate = 213 kg/hr.

7. Heat transfer area ($U = 100$, $\Delta T_L = 50°F$):

$$A = \frac{(213.0)(2.2)(954)}{(100)(50)} = 22.7 \text{ ft}^2.$$

$$\text{Heat transfer area to volume ratio} = \frac{22.7}{(28.6)(7.48)} = 5.9 \text{ ft}^{-1}.$$

Size Distribution

The crystal size distribution can be readily calculated using the full population balance. For unseeded crystallizer in series without growth dispersion, the mass distribution is the sum of a number of gamma distributions. Table 4 gives the appropriate equation for each vessel. The distributions are shown in Figure 9.

Table 4. Expressions for Crystal Mass Distributions.

Population Density	Dimensionless Weight Fraction
A. Nucleator	
$n_1(L) = n_1^\circ \exp\left\{-\frac{L}{a}\right\}$	$w(x_1) = \frac{x_1^3 \exp(-x_1)}{6}$, $x_1 = \frac{L}{a}$
B. Ripener 1	
$n_2(L) = n_1^\circ \left(\frac{a}{a-b}\right)\left\{\exp\left[-\frac{L}{a}\right] - \exp\left[-\frac{L}{b}\right]\right\}$	$w(x_2) = \frac{x_2^3\left\{\exp\left[-\frac{x_2 b}{a}\right] - \exp\left[-x_2\right]\right\}}{6\left\{\left[\frac{a}{b}\right]^4 - 1\right\}}$, $x_2 = \frac{L}{b}$
C. Ripener 2	
$n_3(L) = n_1^\circ \left(\frac{a}{a-b}\right)\left\{\frac{a}{a-c}\exp\left[-\frac{L}{a}\right] - \frac{b}{b-c}\exp\left[-\frac{L}{b}\right] - \left[\frac{a}{a-c} - \frac{b}{b-c}\right]\exp\left[-\frac{L}{c}\right]\right\}$	$w(x_3) = \frac{x^3\left\{\frac{a}{a-c}\exp\left[-\frac{c}{a}x_3\right] - \frac{b}{b-c}\exp\left[-\frac{c}{b}x_3\right] - \left[\frac{a}{a-c} - \frac{b}{b-c}\right]\exp\left[-x_3\right]\right\}}{6\left\{\left[\frac{a}{a-c}\right]\left[\frac{a}{c}\right]^4 - \left[\frac{b}{b-c}\right]\left[\frac{b}{c}\right]^4 - \left[\frac{a}{a-c} - \frac{b}{b-c}\right]\right\}}$, $x_3 = \frac{L}{c}$

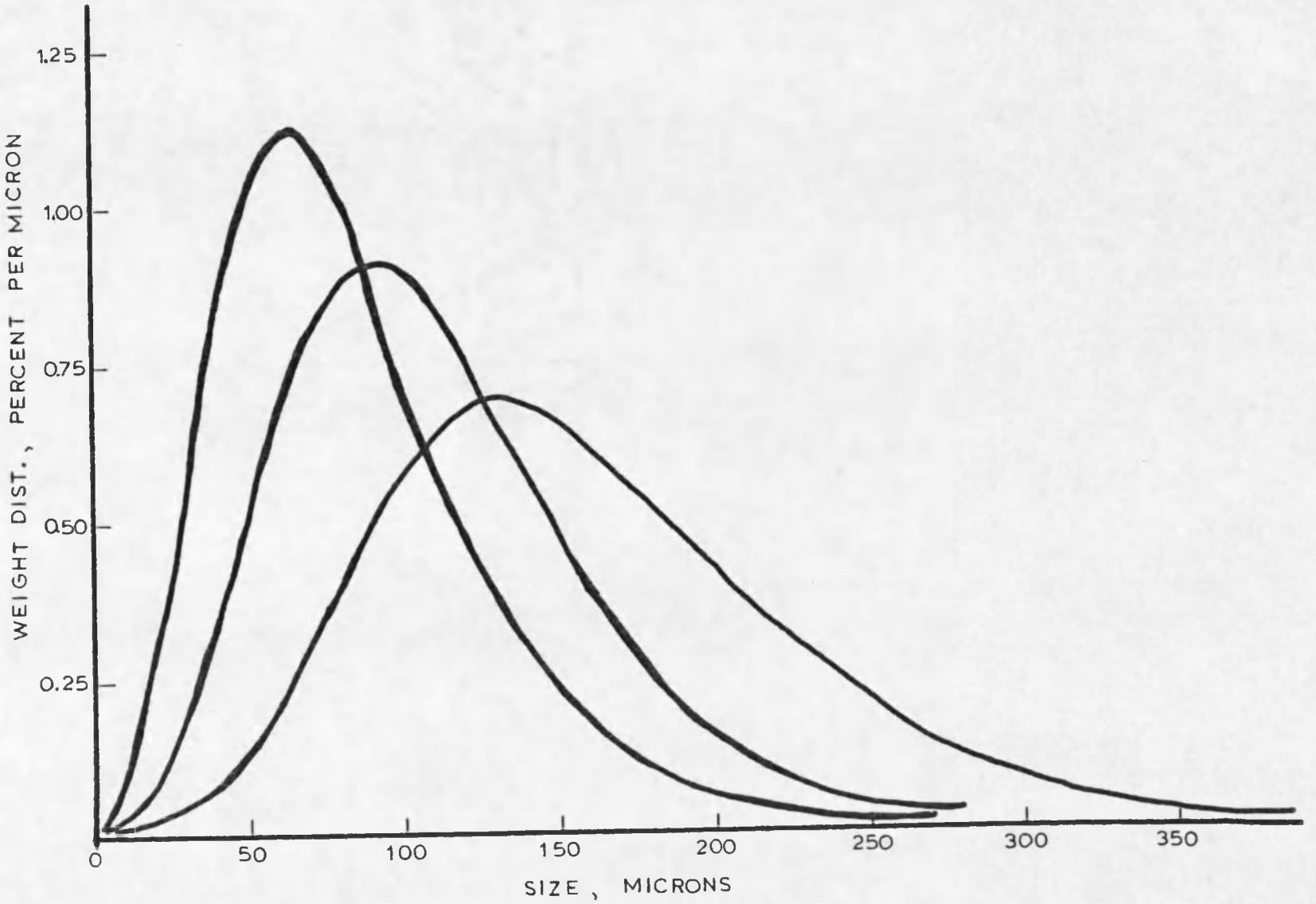


Figure 9. Seeder Crystal Size Distribution Diagram.

Overall Design

The purpose of this thesis was to establish process design conditions for the crystallization steps and to demonstrate design methods. The next stage of full flowsheet design has not been attempted in any great detail. A tentative flowsheet design is given in Figure 10, which shows vessels approximately to scale.

For the nucleator, the volume was 125% of working volume to allow free-board. The evaporative ripeners were made 250% of the working volume to give sufficient vapor disengaging and calandria heating space. Since both ripeners work under the same vacuum, it is more economical to include both in the same vessel with a single baffle separating them. Agitation of the ripeners is probably unnecessary as the boiling action will cause circulation and suspension of the crystals.

From the continuous seeder, crystals are available which can be used for either continuous or batch pan operation.

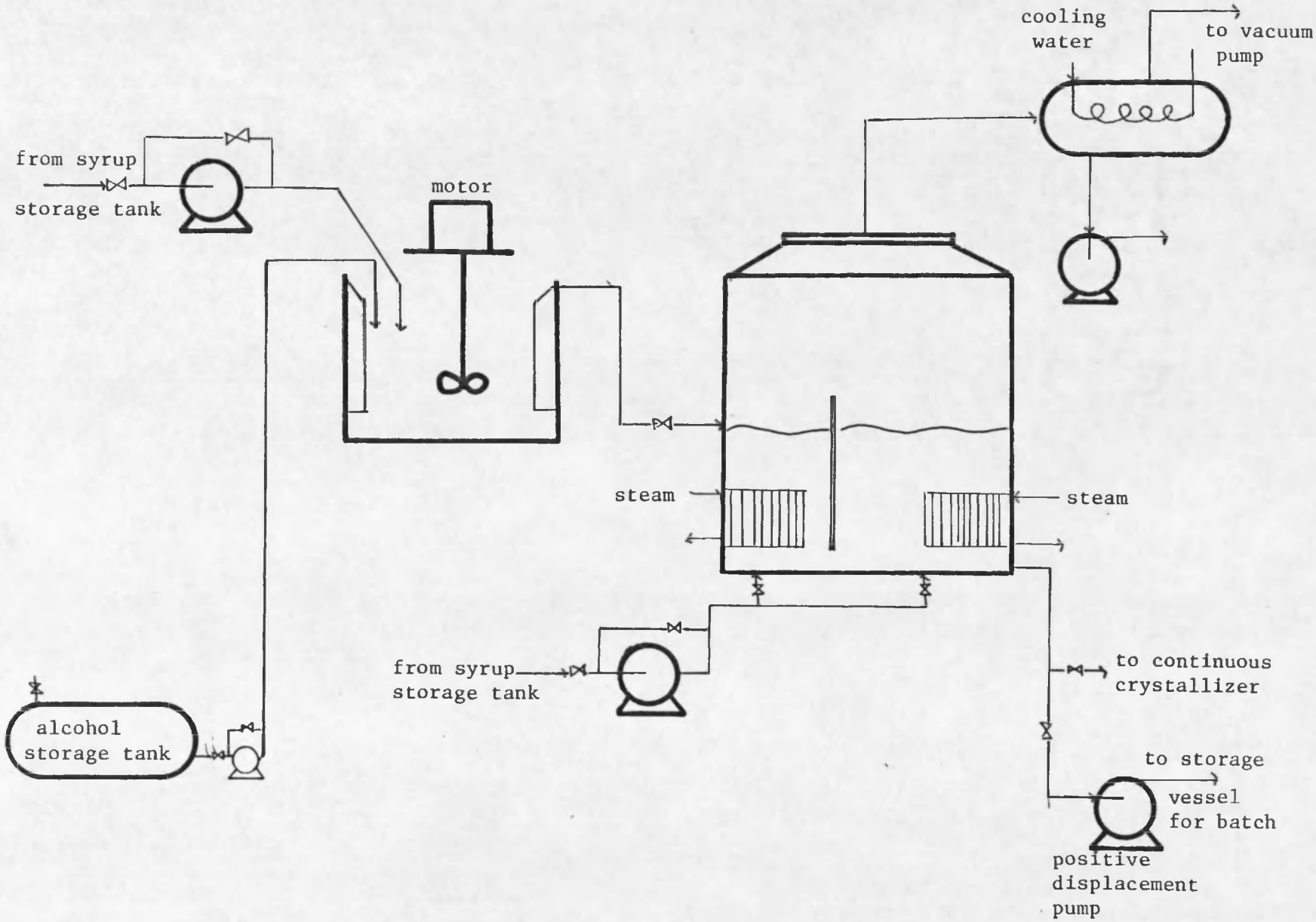


Figure 10. Design Flow Sheet.

CHAPTER 7

BATCH PANS

The majority of the world's sugar crystallization is carried out in batch crystallizers. A batch design will be considered here first as a basis for comparison with continuous pan design and, secondly, to demonstrate a simplified design procedure.

As mentioned in Chapter 4, there are several different boiling schemes that may be used in batch crystallization. For this design, the popular three-stage boiling scheme will be adopted.

As shown in Figure 11, this scheme features three crystallizations from the liquor before it leaves as final molasses. After each crystallization, the crystals are separated from the liquor by centrifuging and go separate ways. The crystals start as grain to the C boiling and after two crystallizations are of the required product size. The scheme thus involves three stages of recovery of sugar from solution and two stages of crystal growth.

In the last boiling stage (C), fine seed crystals (with a large surface area) are used to attempt high recovery from the most impure (and thus slowest growth rate) solutions. The crystals from this boiling are used for the "earlier" (A and B) boilings.

As discussed earlier, a general method of modelling a batch pan has been presented by Wright and White (1974). The method used in this

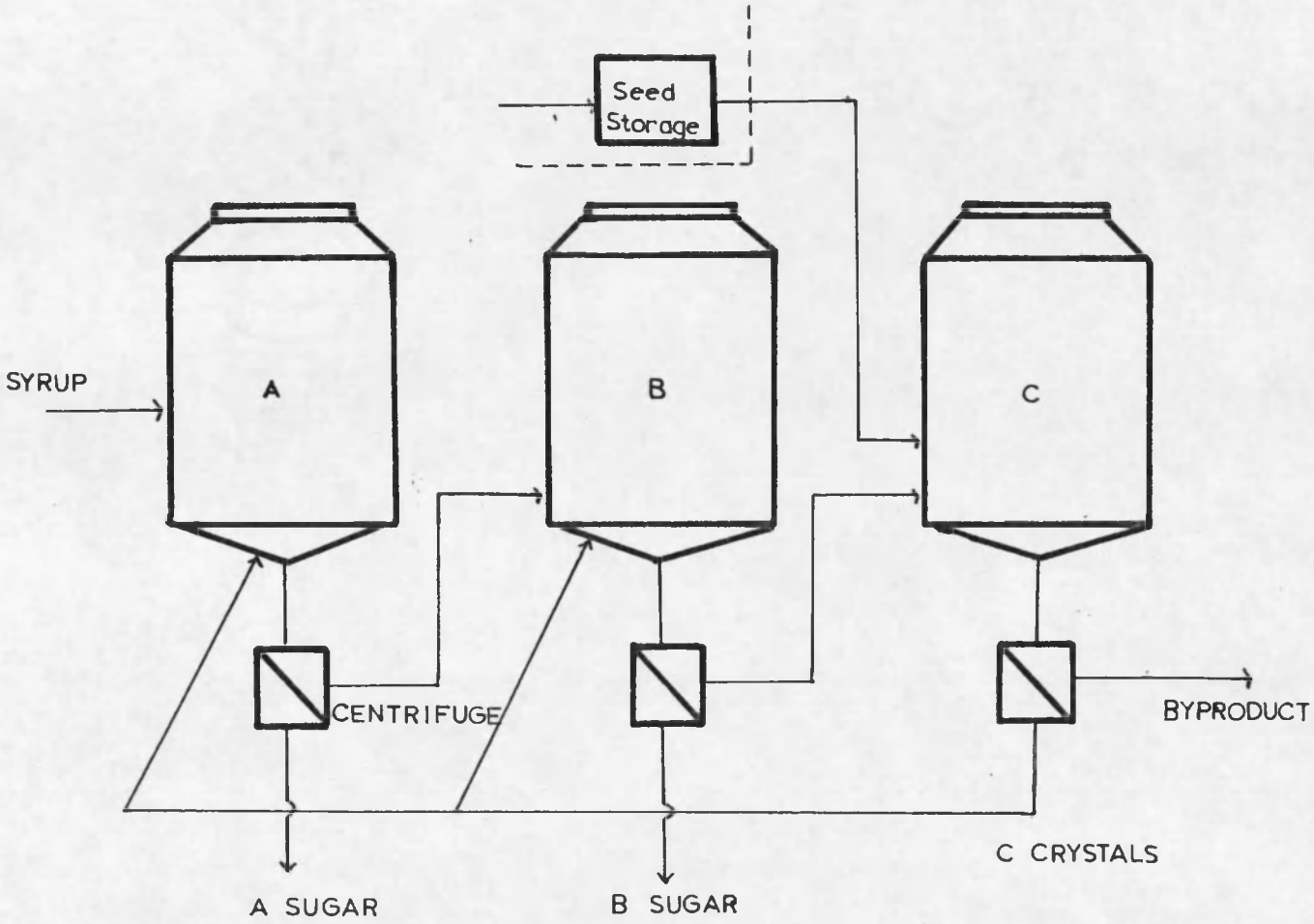


Figure 11. Basic Diagram of Three Boiling Schemes for Batch Sugar Crystallization.

thesis is a simplification of their method, using a constant average growth rate.

Specification

The crystallizer station has to make 800 tons/day of raw sugar of mass mean size 800 μm. It is seeded with the product from the continuous seeder (Chapter 6). A recovery of 92.5% of the sugar from the syrup is required.

It is necessary to determine:

1. The number of vessels.
2. The size of the vessels.
3. The operating conditions, i.e., temperature, pressure, agitation.
4. The batch time.
5. The syrup addition rate and evaporation rate.

From these follow the dependent variables:

1. Production rate.
2. Product crystal size distribution.
3. Recovery of sugar.

Selection of Conditions

Temperature

To avoid deterioration of the sugar, a temperature of 65°C will be used. A vacuum about 24 in. Hg is necessary and this can readily be maintained in practice. Agitation is not necessary, as the boiling action causes circulation keeping the crystals in suspension.

Recovery of Sugar

As indicated in the specifications of Chapter 5, a recovery of more than 90% is required. In the crystallization process, water is evaporated to crystallize the sugar. So both the amount of water and of sugar in solution change; however, the amount of impurities does not change through the process. Hence they may be used as a "tie" substance in material balance calculations. Thus, this recovery will correspond to a final molasses of 40% TP. In plant operation, it is not practical to boil sugar down to this low purity as the viscosity becomes too high and circulation in the pan fails. It is usual to stop at about 50% TP and achieve the final crystallization in a cooling crystallizer designed to handle the highly viscous mixture.

For the present thesis, where the aim is to demonstrate the design method for batch pans, calculations will be performed assuming material of this consistency can be handled in the C pan.

Amount of Growth

Each pan must contain internal heating surface to give the heat transfer required. Boiling cannot begin until the heating surface (usually a calandria) is fully covered with the initial seed slurry (footing), otherwise there will be no circulation and this leads to local overheating, dissolving, or nucleation. The aim of the pan designer is to keep the footing volume as small as possible compared to the final volume. There is also a limitation on the height of the fluid in a pan. If it is too high, the hydrostatic head will prevent boiling in the tubes and circulation will fail. For these reasons, even for the

best designed pans, the footing volume is seldom less than one-third of the final volume (Spencer et al., 1977).

The amount of the crystals that can be grown in a single batch will depend on the allowable increase in the volume of slurry and the permissible increase in the crystal content. The final crystal content is limited. If it becomes too high, the slurry becomes near solid and will not circulate. Likewise, the slurry content of the initial footing cannot be too small. As mentioned in Chapter 6, this is necessary to give a certain "robustness" to the slurry to prevent crystals dissolving away due to inopportune temperature changes. The range of crystal content seldom exceeds a factor of 5. Thus, the crystal mass cannot increase by more than a factor of 15, i.e., the crystal size by more than 2.5-fold.

This decides the number of stages of crystal growth. If the product from the A or B pans is to be about 800 μm (expressed as the volume mean, $\bar{L}_{3,0}$), then the C crystal product cannot be smaller than 320 μm. A value of 350 μm is typical, and this value will be used here.

Likewise, the seed (grain) to the C pan cannot be smaller than about 150 μm. The crystal from the C pan (with the impurest and most viscous liquor) must be reasonably large if they are to be separated by centrifuging, and thus a grain seed size smaller than 150 μm cannot be used. Since the continuous seeder of Chapter 6 produced seed with a volume mean size of about 100 μm, an extra graining pan will be necessary to grow the crystals to 150 μm. If the continuous seeder had been designed for batch use alone it would have been a simpler design to

increase its product size to this value. However, it is also used for the continuous design.

For the present design, one vessel will be used for each operation and will be designed exclusively for that operation. In practice the vessels are often used for several operations (A, B, or C boiling) depending on scheduling needs.

The vessels necessary, shown with the crystal size increases required and the approximate feed liquor purities, are given in Table 5.

The true purities of the B and C pans will be calculated from the material balances in following sections.

Two "high grade" (A and B) boilings are necessary. If all the crystals were put in a single pan, the crystal content would reach a limiting value when only about 60% of the sugar is recovered, so a second boiling is necessary.

Table 5. Batch Vessels Required.

Name	Crystal Size Increase (μm)	Feed Liquor (% TP)
Graining pan	100-150	90
C pan	150-350	60*
B pan	350-800	75*
A pan	350-800	90

*Approximate

Limiting Crystal Contents

The minimum crystal content for a footing will be taken as 9%, the value used in the continuous seeder. The crystal content of the material from the graining pan will be taken about 1.5 times this value. The maximum crystal content for the A, B, and C pans will be taken as 48, 40, and 30% by wt, respectively (Jenkins, 1966).

Design Method

The design method was outlined in Chapter 4. The moments of the population balance will be used. Rather than solving the population balance and material balances iteratively over small time increments as was done by Wright and White (1974), a constant average growth rate will be assumed. Then, the relations can be solved in one step from beginning to end. The population balance relations are given in Table 6, first as a general differential equation, then as the integrated equation for constant G, as used here.

Since the size increments have been chosen, it is possible to apply these equations in turn from the A to the C pans. Knowing the increase in size (number mean) and the average growth rate, its growth time can be calculated.

Calculations

Mass of Crystal

Since

$$\frac{\text{mass}_1}{\text{mass}_2} = \left\{ \frac{\left(\bar{L}_{3,0}\right)_1}{\left(\bar{L}_{3,0}\right)_2} \right\}^3$$

Table 6. Moment Equations for Batch Sugar Boiling Model.

Moment	Differential Equations	Solution (for constant G)
1	$\frac{dm_o}{dt} = 0$ (unseeded)	$m_o = m_o^o$
2	$\frac{dm_1}{dt} = Gm_o$	$m_1 = Gm_o^o t + m_1^o$
3	$\frac{dm_2}{dt} = 2Gm_1 + 2Dm_o$	$m_2 = G^2t^2m_o^o + 2Gtm_1^o + 2Dtm_o^o + m_2^o$
4	$\frac{dm_3}{dt} = 3Gm_2 + 6Dm_1$	$m_3 = (G^3t^3 + 6GDt^2)m_o^o + (3G^2t^2 + 6Dt)m_1^o + 3Gtm_2^o + m_3^o$
5	$\frac{dm_4}{dt} = 4Gm_3 + 12Dm_2$	$m_4 = (G^4t^4 + 12G^2Dt^3 + 12D^2t^2)m_o^o + (4G^3t^3 + 24GDt^2)m_1^o$ $+ (6G^2t^2 + 12Dt)m_2^o + (4Gt)m_3^o + m_4^o$
	. . .	

Table 6, Continued.

Mean Sizes (for constant G)

$$\bar{L}_{1,0} = Gt + (\bar{L}_{1,0})_i$$

$$\bar{L}_{3,0} = \left\{(G^3t^3 + 6GDt^2) + (3G^2t^2 + 6Dt)(\bar{L}_{1,0})_i + 3Gt(\bar{L}_{2,0})_i^2 + (\bar{L}_{3,0})_i^3\right\}^{1/3}$$

$$CV = \left\{\frac{2Dt + (CV_iL_i)^2}{(Gt + L_i)^2}\right\}^{1/2} \quad \text{or} \quad \frac{\left[\frac{2Dt}{L_i^2} + CV_i^2\right]^{1/2}}{\left[\frac{Gt}{L_i} + 1\right]}$$

$$\bar{L}_{4,3} = \frac{m_4}{m_3}$$

if no nuclei are formed, then the mass of crystal can be evaluated for each pan as shown in Table 7a.

Overall Material Balance

These are also shown in Table 7. For material balance purposes only, it is assumed that the exit liquor is saturated, i.e., solubility relations can be used. These are given in Chapter 3 (Figure 4).

Growth Rates

This is the crucial choice in the design. For present purposes, values from the maximum (70% of nucleation threshold limit) growth rate curve (Chapter 3, Figure 5) will be used, at the log mean value between the inlet and exit conditions (Chapter 4). These values are given in Table 8, together with values of D, the growth dispersion. The final supersaturations are also given.

Moment Equations

The moment equations can now be solved to give the various mean sizes as shown in Table 9.

Batch Time

The batch growth time is calculated from the increase in number mean size divided by its growth rate. The actual batch time is larger than this to allow for starting, emptying, cleaning, and idle time (increased by 30%).

Table 7. Material Balances for Batch Pans. -- Basis = 1 day; units = tons.

a. Crystal Rates

Continuous	Grain	C	A / B Total
1.9 21.0	6.4 73.6*	81.4	100

	In				Out				
	Total	S	I	W	W	I	S	Total	
b. Grain Pan									
Crystal	1.9	1.9					6.4	6.4	Crystal
Liquor	21.0	16.5	2.0	2.5	2.0	12.3	26.0	40.3	Liquor
B-Liquor	30	14	10.3	5.7	6.2			6.2	Evap. Water
c. C Pan									
Grain { Crystal	6.4	6.4					81.4	81.4	Crystal
Grain { Liquor	40.3	26.	12.3	2.0	29.5	96.	64.9	190.4	Liquor
B-Liquor	241.7	113.9	83.7	44.1	16.6			16.6	Evap. Water
d. B Pan									
Seed Crystal	25.3	25.3					250.	250.	Crystal
A-Liquor	572.6	352.6	94.	126.	67.1	130.6	175.	372.7	Liquor
Recyc. Liquor	101.	47.1	36.6	17.3	76.2			76.2	Evap. Water
e. A Pan									
Crystal	56.1	56.1					550.	550.	Crystal
					126.	94.	352.6	572.6	Liquor
Syrup	1383.5	846.5	94.0	442.7	316.7			316.7	Evap. Water

*Based on crystal content of 8% by wt.

Table 8. Selected Growth Rates for Batch Pans.

	Graining Pan	A	B	C
G (μm/min)	2.0	5.5	2.0	0.25
Growth dispersion coefficient, D (μm^3/min)	100	275	200	18
Final supersaturation (OS)	1.04	1.06	1.04	1.02

Table 9. Summary of Results for Batch Pans.

	Grain	C	B	A
a. CSD				
Mean sizes, microns				
$L_{3,0}$	150	350	750	750
$L_{2,0}$	134	324	711	711
$L_{1,0}$	112	287	667	667
$L_{4,3}$	158	401	810	810
CV	0.67	0.46	0.35	0.35
CV^{pop}_{wt}	0.51	0.32	0.24	0.24
b. Batch Time				
Growth, hours	0.29	8.3	3.2	1.2
Batch, hours	0.38	10.8	4.2	1.6
c. Volume and Rates				
Volume				
Vessel working, ft^3	19	3,000	2,600	2,400
Vessel total, ft^3	48	6,000	5,200	4,800
Mass rates, tons/batch				
Syrup				98.8
A -- slurry				80.2
A -- liquor			95.4	
B -- slurry			103.8	
B -- liquor			14.0	
C -- slurry		135.9		
Seeds		3.2	4.3	4.0
A -- sugar				39.3
B -- sugar			41.2	
Crystal content, % by wt.	13.7	30	40	49
d. Heat Transfer Area, ft^2	28	4500	3,900	3,600

Vessel Volumes

From the total throughput and the batch cycle time, the quantity to be handled in each batch can be calculated and thus the vessel working volumes.

Heating Surface

The heat transfer areas of Table 9 can be evaluated. Heat transfer area per unit working volume is of the order of 1.5 ft^{-1}, which is the typical figure used in most batch pans.

Holding Tank

Because of the differing batch times for each pan, intermediate holding tanks are necessary. Holding tanks for the grain, the C crystals, and B molasses are chosen to have volumes sufficient for two C pan batches. The tank for the A molasses likewise holds two batches required for the B pan.

Liquor Feed Rates during Batch

A batch pan would be operated to give the maximum growth rate which still avoids false grain (nucleation). This would be monitored by supersaturation meters (conductivity, boiling point elevation or refractive index measurements). Control is either by the steam rate or the liquor feed rate (usually the latter). The liquor feed rate should be as high as possible while yet maintaining the crystal content above the minimum value. Once the pan is full, liquid is added just to balance evaporation until the crystal content reaches the final limiting ("heavied up") value.

Knowing the policy during the batch (e.g., following the maximum allowable liquor feed rate or following constant crystal content), it is possible to calculate the rate of feed liquor addition at any time using the same method described earlier. This has not been done here, as in practice control is from meter measurements.

Complete Size Distribution

The complete product size distribution can be evaluated using the population balance. A numerical method of solution is necessary. Details are given in Appendix A. The weight distribution obtained in this way are shown in Figure 12.

Final Design

The final designs are shown in Figure 13.

Comments

The values obtained from the design are in reasonable agreement with batch conditions reported in the text (Jenkins, 1966). The simplified design method is very simple to use. To check on the growth rate selected, the calculation could be performed for the selected pan volume for intermediate time (actually intermediate mean sizes), and the variation of G with time determined. This would check the log mean value used.

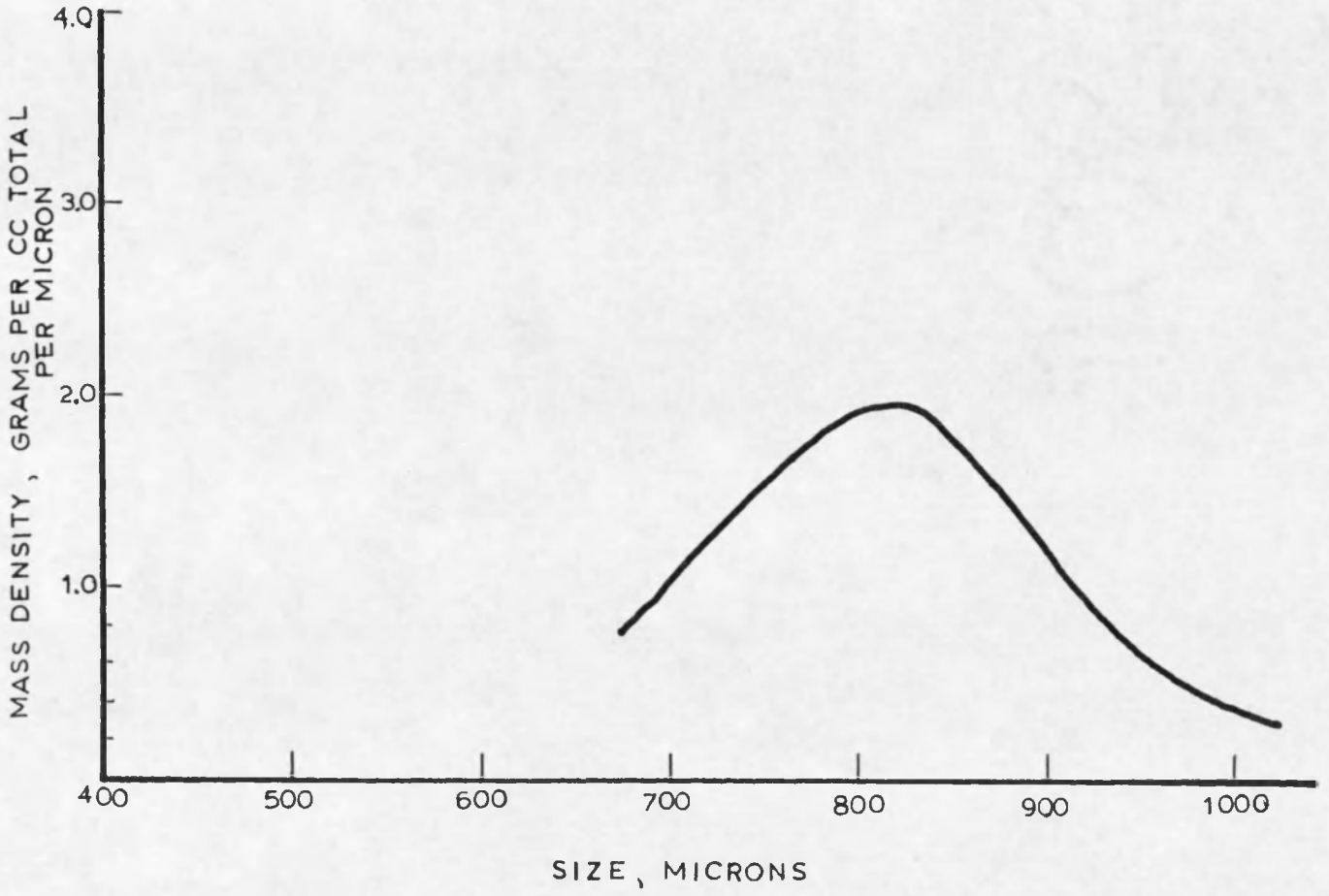


Figure 12. Product Weight Distribution Diagram for Batch Pans.

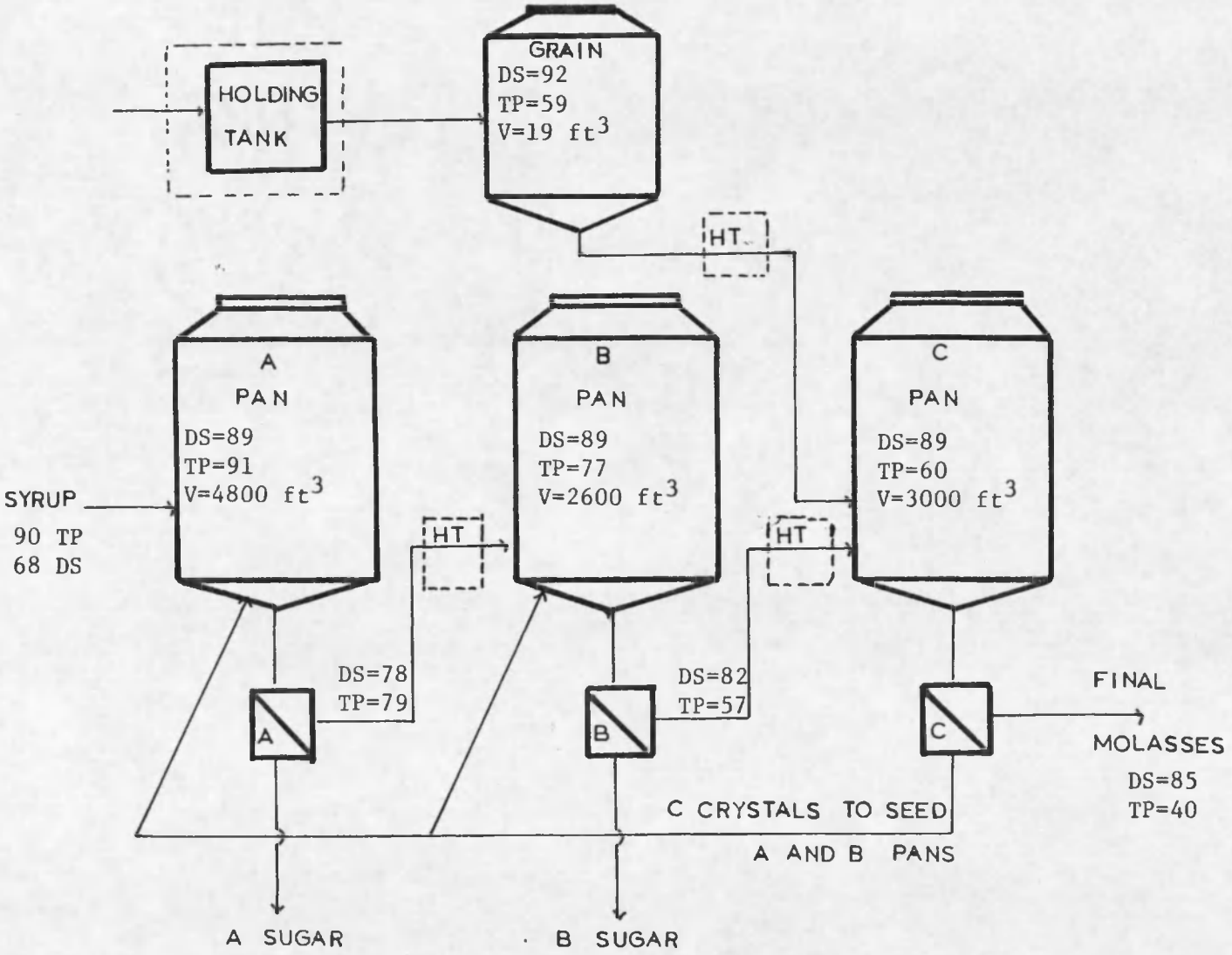


Figure 13. Diagram of a Seeded Batch Process Showing Design Values.

CHAPTER 8

CONTINUOUS PANS WITH CLASSIFIED RECYCLE

The technology on continuous pan process design is still developing. To date, several prototype pans have been designed and tested. The most optimum design probably has not yet appeared. Large-scale plant evaluations are necessary.

For the present design, the continuous sugar crystallization unit will consist of a low-grade station and a high-grade boiling station (Figure 14). Following the path of the crystals, the process starts with growing the product from the continuous seeder in a low-grade continuous pan. After centrifuging, these crystals are mixed with a stream of syrup and fed into the high-grade station where they are grown into product crystals. The product crystals will be classified and the fines fraction recycled for further growth. This will give a narrower size distribution. Following the path of the liquor, syrup feeds into the high-grade station and after centrifuging the molasses passes into the low-grade station where the final sugar is recovered, giving the final molasses.

The high-grade station will consist of several stages of pans. The crystal from the last stage is centrifuged and classified to give the product crystals. The crystal content increases with each stage. Because no intermediate centrifugation is used, molasses from the last stage may have to be recycled back to "earlier" stages.

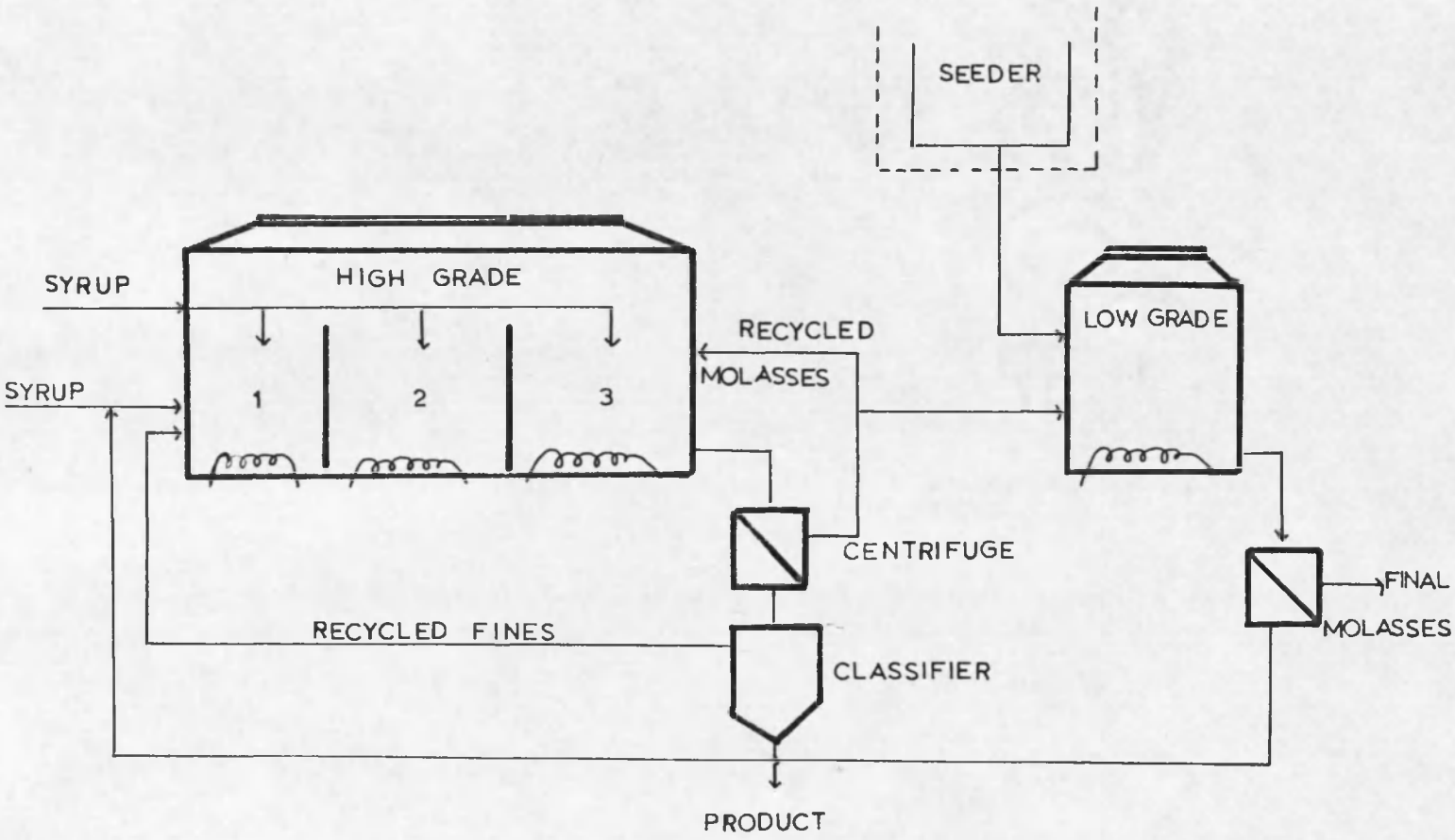


Figure 14. Schematic Diagram of a Continuous Sugar Crystallization Unit.

As previously mentioned (Chapter 4), program RECYC, developed by Randolph and Tan (1978), will be used in this design. This will be necessary only for the high-grade station.

Specification

The vessels will be operated to give a production of 800 tons/day with no further nucleation. Thus, the only nuclei source is the seed crystals at a rate of 2.3×10^{12} #/day. The crystal product must have a mass mean size of about 800 μm, and the recovered sugar about 92%.

The design input variables must specify the following:

1. Number and volume of vessels.
2. Operating conditions, i.e., P, T, agitation.
3. Distribution of feed syrup (68% DS, 90% TP) to various vessels.
4. Classifier performance (crystal size recycled).

From which the following can be determined:

1. Residence time.
2. Rate of recycled crystals and molasses recirculation.
3. Product crystal mean size and product size distribution.

Choice of Conditions

Temperature

All the vessels will be operated under vacuum to give a temperature of about 65°C to avoid degradation of the sugar. Agitation will be by natural convection through the calandria heaters.

Number of Stages

The problem with continuous pans is that a wide size distribution in the final product is caused by the wide spread of residence time. As discussed in Chapter 4, one method of narrowing the size distribution from continuous crystallizers is by staging. Three high-grade stations were chosen for this design because it requires less computation time, yet results in a considerable reduction in the working volumes compared to two stages. With the classifier in the design, more vessels may not give a great advantage in further narrowing the CSD.

For the low-grade station, a single stage was used since the amount of crystal produced is small.

Growth Rates and Growth Dispersion

The increase in mean size ($G\tau$) for the low-grade station (single seeded MSMPR) will depend on the choice of mass mean size of crystals leaving the station. A typical value of 350 μm will be used here.

The true purities of the exit liquor in each stage of a high-grade station will determine the growth rate (using the correlation in Chapter 3, Figure 5). Since the vessel is well-mixed, the growth rate will be that corresponding to the exit liquor concentration. The exit liquor concentration depends on the amount of crystals produced in each vessel and has to be selected. Equal increments in the volume mean size ($\bar{L}_{3,0}$) will be the basis of choice, i.e., $\Delta\bar{L}_{3,0}$ = 125 μm in each vessel; thus, the volume mean crystal size ($\bar{L}_{3,0}$) in the three stages will be 500, 625, and 750 μm. The 750 μm product is chosen as compared with the final size in the batch (Chapter 7).

The growth diffusivity correlation of Randolph and White (1977) discussed in Chapter 3 (Equation 13) is used.

Product Classification

A cut size of 500 μm will be used in the classifier. This is a compromise choice. If a larger size were used, a much larger mass of crystals would have to be recycled; if a small value is chosen, difficulty would be experienced in obtaining a mass mean size of 800 μm. In work by Tan (1977), a similar classifier cut size was used.

A classifier efficiency curve of the form shown in Figure 15 will be used where 80% of the very fine crystals is removed. There are as yet no raw sugar classifiers available, so the figures are speculative, but the shape of this efficiency curve is consistent with screen operation and pneumatic classifiers (Perry and Chilton, 1973).

A single stage of classification is shown and has been used in the present calculations. If required, a second stage could be used, e.g., to take off the very fine material and dissolve it completely.

The recycled fines should be returned to the vessel containing crystal nearest in size to the recycled material. It is expected this will be the first high-grade stage; this choice has been used in the design.

Crystal Content

The crystal content for the low-grade C pan will be taken as ca. 15% by wt. For high-grade pans, the first stage is about 38%, i.e.,

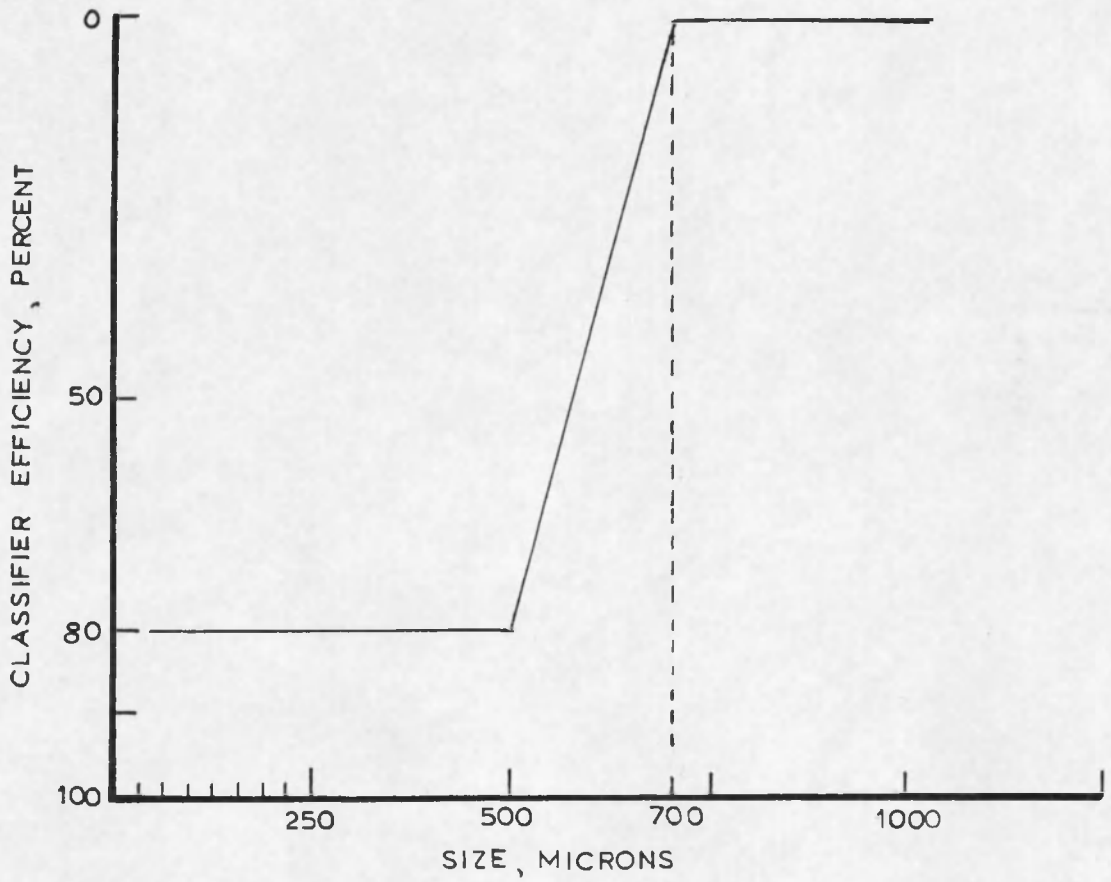


Figure 15. Classification Function for the High-Grade Continuous Pans.

twice the value used by Broadfoot and White (1978), the second stage is maximum (about 45%), and the third stage is 40% by wt.

Design Method

The high-grade station utilizes recycle of fine crystals. This increases the complexities of the calculations considerably. A computer program to handle such calculations was available to the author. This was entitled RECYC and is described in detail by Randolph and Tan (1978).

In principle, the calculations for each vessel in series are handled using the moment equations. An initial value is taken for the moments of the recycled seed. For the last stage before the classifier, an approximate size distribution is constructed. This is assumed to be a gamma distribution with the same first, second, and third moments as the product in the last stage. Classifier efficiency calculations are performed on this approximate distribution to give the approximate distributions of the product and the recycled fines. The moments of the recycled fines distribution are calculated and are used. Iterations are continued until:

$$\left| \frac{m_{R,j}^{New} - m_{R,j}^{Old}}{m_{R,j}^{New}} \right| < 2.0 \times 10^{-3}$$

Should convergence not occur in thirty iterations, the program stops.

Several options are allowable depending on the nature of the unknowns and the information available. Ideally for this design, it

would be useful if the growth rate and the exit crystal content could have been read in for each vessel, together with its make-up feed rates. The program would then solve for stage volumes, crystal product size and production, and recycle rates. The closest option available was to specify the first crystallizer volume (later ones were taken to be inversely proportional to the growth rate) as well as the individual growth rates and crystal contents. The flow rate from each vessel was then calculated. Manually, new volumes were then found to give the desired flow rates and these were put into the program for recalculation. Conditions were adjusted at the end of each run of RECYC to obtain a mass mean size, $\bar{L}_{4,3}$, of 800 μm for the product. The successful set of conditions gives the final design.

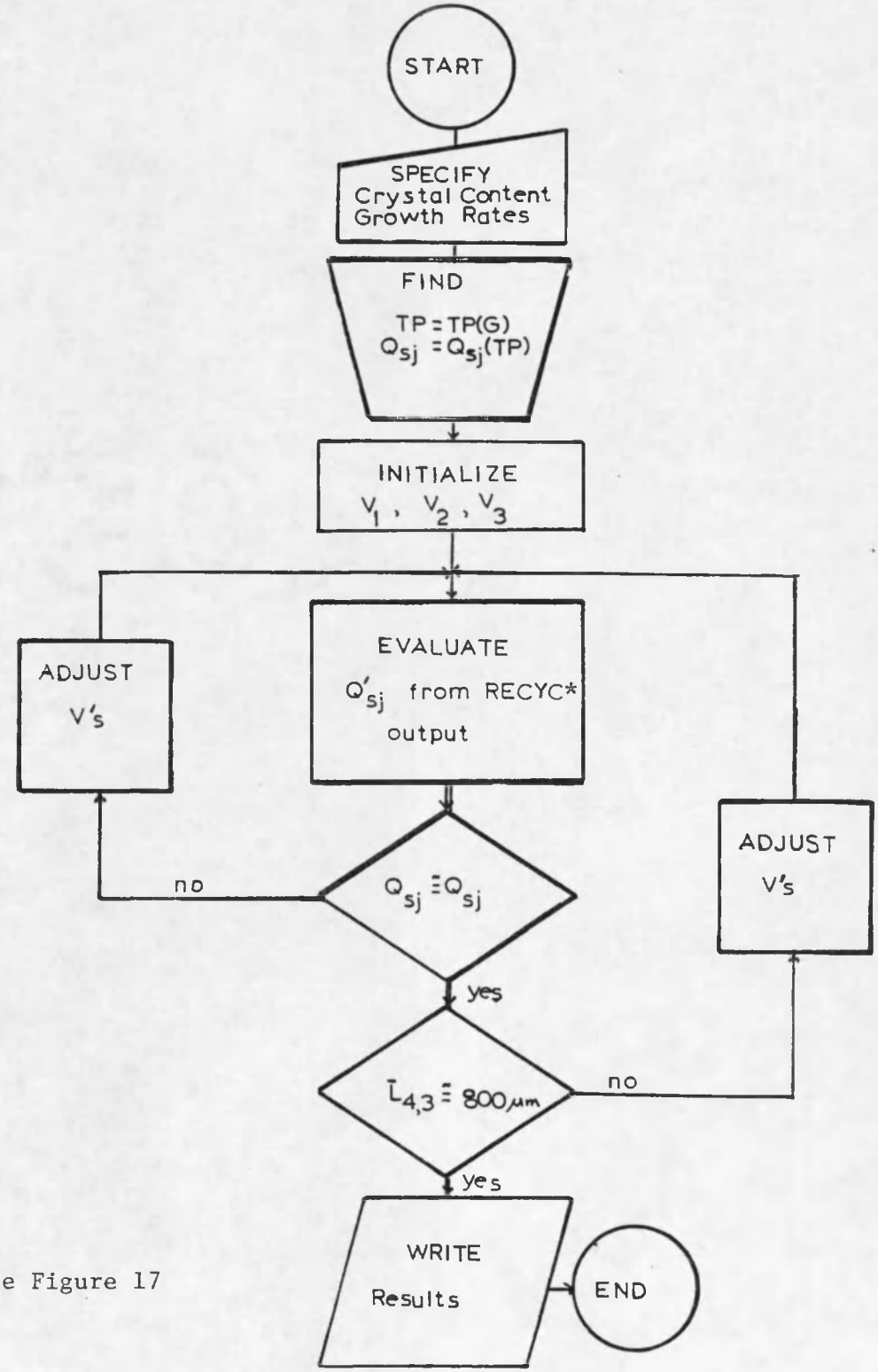
Flow sheets of the calculation procedures are given in Figures 16 and 17. The method of selecting a gamma distribution having some of the moments of the real distribution is given in Appendix B.

Table 10 lists the data fed into the program for the last run of RECYC. Table 11 shows the output values obtained.

Overall Balance

The overall mass balance is shown as a flow diagram in Figure 18. This aids tracing the flow by which each component (sucrose, impurity, water) changes in quantity as sugar is formed from stage to stage.

Steam consumption can be easily obtained by doing an energy balance around each stage.



*See Figure 17

Figure 16. Computational Algorithm for Continuous Pans.

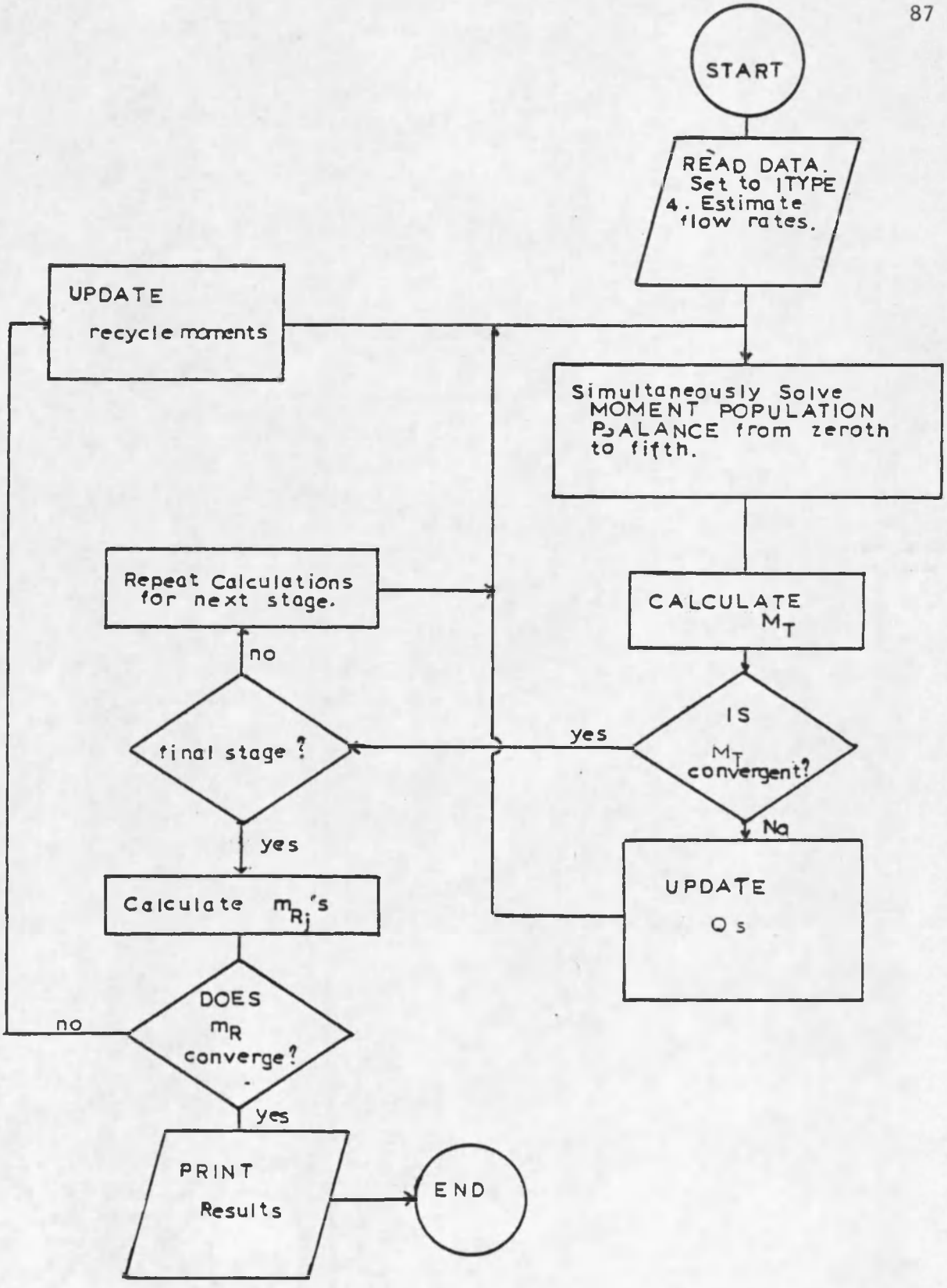


Figure 17. Computational Algorithm of RECYC. -- After Tan (1977).

Table 10. Input Data for the Last RECYC Run.

Data	Stage 1	Stage 2	Stage 3
Seeds			
Rate, gpm	10.1	—	—
Gamma constants: α	22.5	—	—
β	126.0	—	—
Solid concentration, gm/cc	0.70	—	—
Volume (working), gallons	1,263.0	2,052.0	23,670.0
Solid concentration (liquor basis), gm/cc	0.90	1.30	0.95
Growth rates, μm/min	5.30	4.00	0.80

Table 11. Output from Last RECYC Run.

Variable	Stage 1	Stage 2	Stage 3	Product
CSD				
$\bar{L}_{1,0}$, μm	343	394	457	636
$\bar{L}_{4,3}$, μm	559	635	729	806
CV_{pop}	0.52	0.51	0.49	0.32
CV_{wt}	0.29	0.30	0.31	0.25
Residence time, min	9.66	12.5	79.1	—
Exit flow rate (total slurry), gpm	130.7	164.2	299.3	—
Production, tons/day	—	—	—	800
Recycled fines, tons/day	—	—	—	201.3

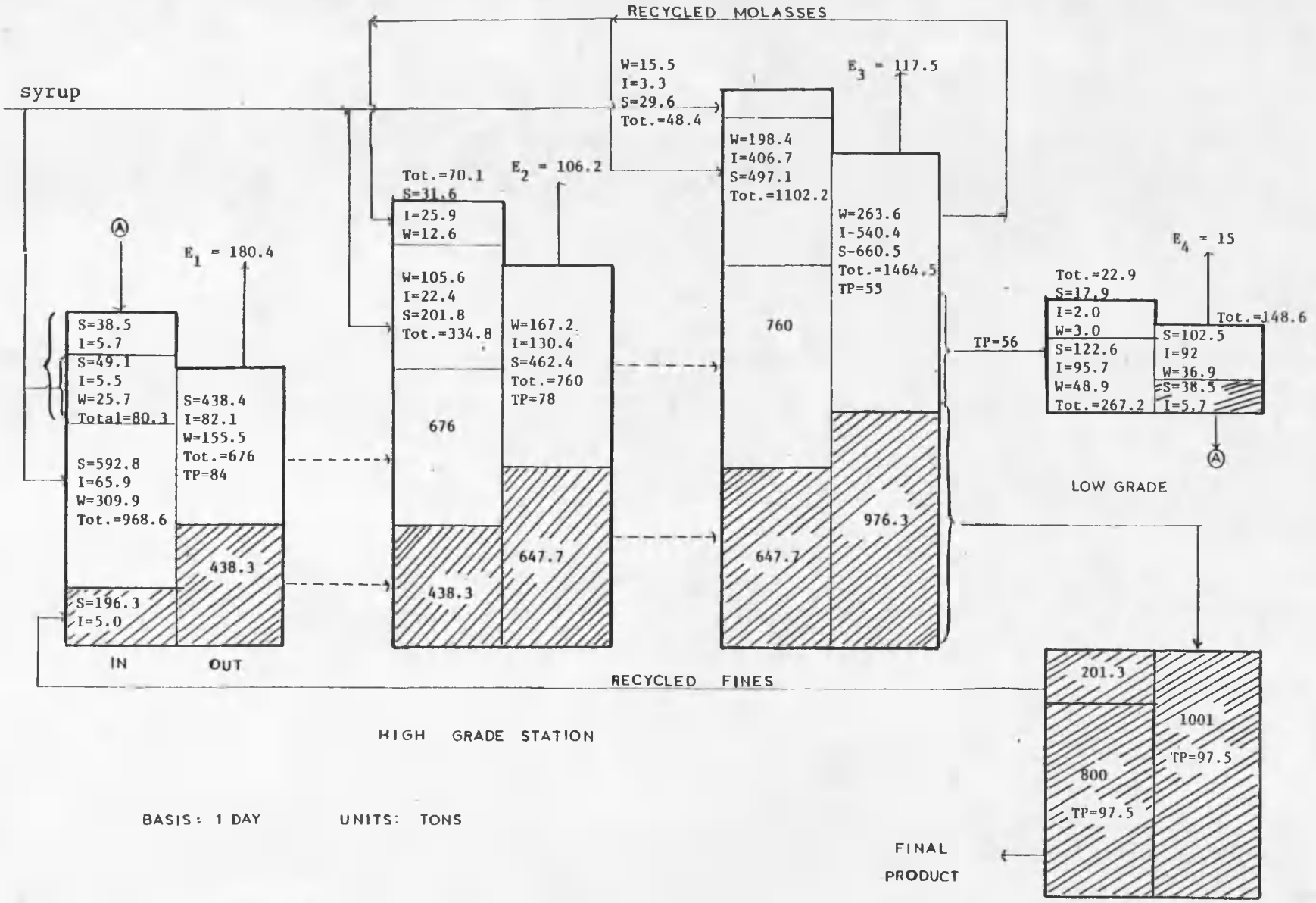


Figure 18. Mass Flow Diagram for Continuous Pans.

Calculations for Low-Grade Vessel

1. Amount of growth -- by trial and error, $G\tau$ for the low-grade pan is obtained until $\bar{L}_{4,3} = 350$ μm.

   $\therefore G\tau = 50$

2. Residence time -- from Figure 5, Chapter 3 (TP = 52):

   $G = 0.44$ μm/min

   $$\therefore \tau = \frac{G\tau}{G} = \frac{50}{0.45} = 111 \text{ minutes}$$

3. Crystal rate:

   $$\left(\frac{275}{750}\right)^3 (780) = 38.5 \text{ tons/day (TP = 100\%)}$$

4. Flow rate -- taking the total slurry mass from Figure 18:

   $$\text{flow rate} = \frac{(230.5 + 44.2)(2000)(454)}{(1.45)(1440)(3785.43)} = 31.6 \text{ gpm}$$

5. Vessel volume = (31.6)(111) = 3510 gallons.

Product Size Distribution

The complete product size distribution can be plotted from the output of the last RECYC run. This is given in Figure 19.

Comments

Since the high-grade unit operates under the same vacuum with no intermediate centrifugation, it is economical to include the three stages in the same huge vessel, separated by baffles. Only the size of

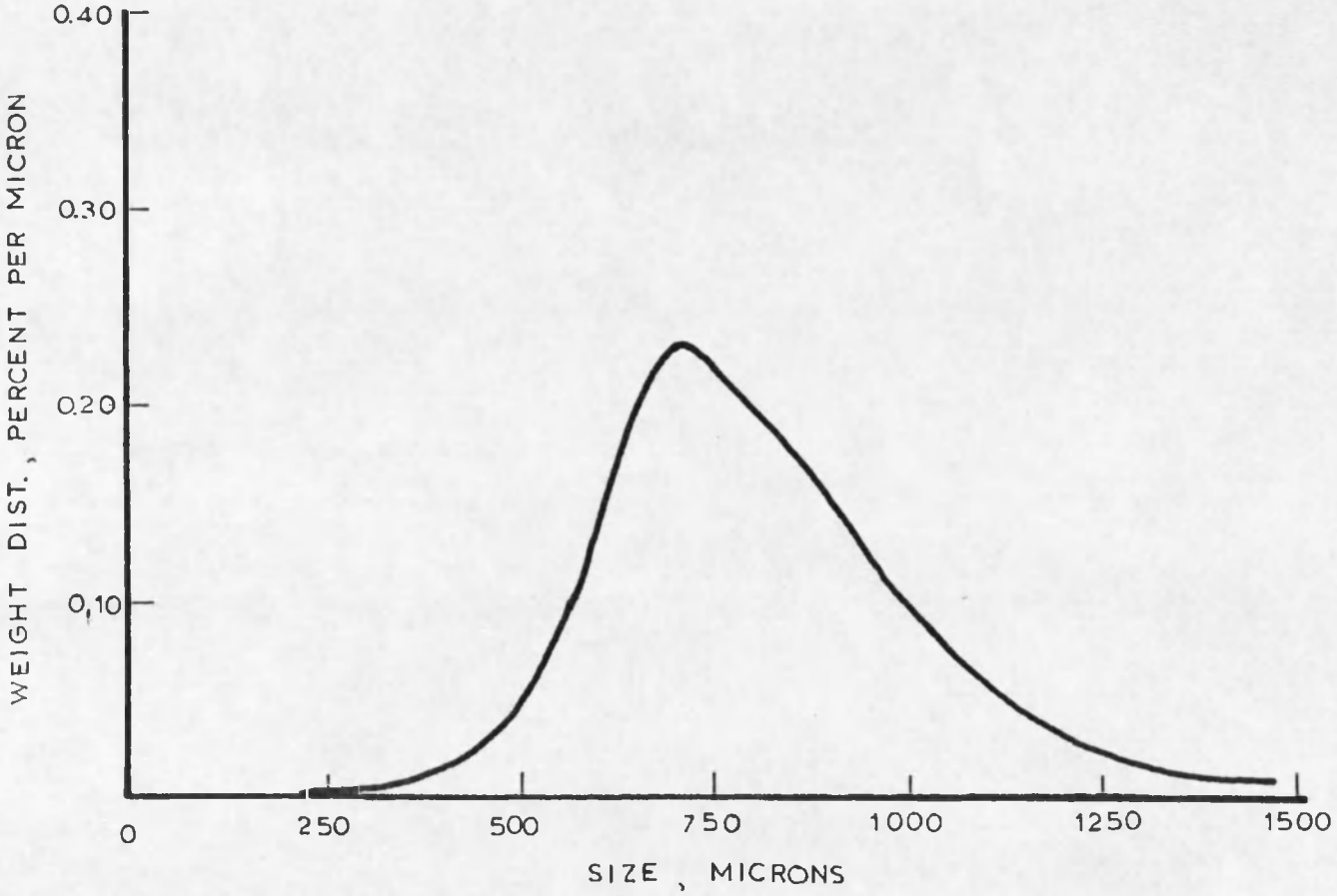


Figure 19. Product Weight Distribution Diagram for Continuous High-Grade Pans.

each compartment has to be different because either molasses has been recycled or syrup has been further added in some stages.

Continuous centrifuges have to be installed for the last stages of the high-grade unit as well as the low-grade vessel.

There is no classifier that has been available for raw sugar as of now. This part could be a good area for further research. Certainly a classifier would have to operate on dried centrifuged product because of the high molasses viscosity, unless the centrifuge could be modified to act as a classifying centrifuge.

The recycling of molasses to maintain the operation at the limiting crystal content makes the design non-optimum. This could have been avoided by installing intermediate centrifuges between stages. In summary, this will give several advantages:

1. Avoidance of the large amount of recycled molasses.
2. The size of the working volumes of the three vessels could be significantly reduced with the inverse ratio of volume approaching the ratio of allowable growth rate.

CHAPTER 9

SUMMARY AND CONCLUSIONS

Population balance together with material balance, energy balance, and rate relations were used to determine the process design of a sugar crystallization station. Design for both batch and continuous crystallizer stations were performed as well as the design for a continuous seeder. In this section, the batch design and continuous design are discussed together with a comparative summary.

Batch Pans

The dynamic population balance equations were solved by the moment technique on the assumption of a constant growth rate. This resulted in a simpler design method which was easy to use. The choice of the average growth rate to use was a crucial part of the design method because growth rate is very dependent on several design factors such as the impurity and boiling rate. A log mean value of growth between the exit and inlet conditions was chosen. There was close agreement of this design reported in texts.

Continuous Sugar Crystallizers

The continuous crystallizer design method was used for two applications: the continuous seeder and the continuous pans. In the design of a continuous seeder, Crawford's nucleation studies were scaled

to generate the required number of nuclei. These nuclei were "ripened" in two evaporative crystallizers (ripeners). This resulted in a nucleator whose working volume (13.2 gallons) is nearly half the sum of two ripener working volumes (7.5 gallons and 28.6 gallons, respectively). In this design, crystal content was the crucial design choice for when it becomes too low, dissolution of seeds may occur.

The continuous pans consisted of a high-grade station and a single low-grade vessel. Classified recycle of fines was included in the high-grade station in order to narrow the size distribution of the product. An available computer program, RECYC, was used in search for a final design. The problem encountered with the program RECYC was the presumption that an indefinitely available amount of clear liquor feed was entering every stage, to which the previous stage flow rates were added. To remedy the situation, volumes entered to the program were adjusted manually until the clear liquor feed tallied with those computed from the program at specified crystal content and growth rates. A good deal of further iterations was necessary then if the program had allowed this type of calculations.

For the resulting final design, a larger volume of vessel is required for the last stage because recycling of molasses was necessary to maintain the operation at the limiting crystal content. This made the design nonoptimum. Were it possible to install an intermediate centrifuge between the stages, this problem could have been avoided. However, the program RECYC could not handle this situation.

There are advantages that interstage removal (through intermediate centrifuge) may amount to. They are:

1. Avoiding the recycling of large amounts of molasses, thus decreasing the volume of the third stage.
2. The relative sizes of the vessels will be in small increasing differences (inverse to the ratio of growth rates).

Comparative Summary

The results for the batch pan design can be compared with those for the continuous pans. On the basis of the same production (800 tons/day) and product mean size (= 800 μm), Table 12 lists comparative figures.

As shown in Table 12, it appeared that continuous pans with classified recycle required as little as 50% of the batch pan working volume. Perhaps recycled fines (having large surface area) had size (= 500 μm) close enough to the first stage final crystal size and thus required less volume to grow a little further.

The classifier included in continuous pans had nearly solved the long-standing problem of wider size distribution as shown in the tabulation. This had required severe classification (an 80% efficient classifier), however.

There was a little difference in steam required to evaporate the mass of water from the mass of syrup fed into both stations. Certainly these showed that since final molasses has approximately equal DS (= 85%) for both processes, the feed material would lose the same amount

Table 12. Comparative Results. -- Units in tons.

Variable Compared	Batch	Continuous
Production/100 gallons of total working volume	1.34	2.58
Production/tons of steam consumption for water evaporation	1.83	1.81
CV obtainable	0.24	0.25

of water. However, idling, starting, and cleaning for batch pans may consume steam as much as 25% more than those listed in Table 12.

Suggestions for Further Work

During the course of this design, various aspects were encountered where sufficient information is not available. These included:

1. Prediction of solubility data for impure solutions -- the solubility correlation is limited. Broadfoot and White's (1975) plot of S/W versus I/W is linear but it is suggested somewhere else (Maudarbocus and White, 1978) that this plot is curved.
2. Growth rates, growth dispersion rate, and nucleation threshold data -- like the solubility data, this information is also limited. So far, the only available correlation is from Wright and White (1974) and recalculated by Randolph and White (1977).
3. Assumption of size independent growth -- for sugar work, size independent growth is not always true because growth rate as a fraction of clear liquor true purity (TP) decreases with TP, while the size of crystals increases with boiling time.
4. Nucleator -- the nucleation rate used in the design for estimating the nucleator working volume is quite low. The correlation given by Crawford (1970) on ethanol-sucrose-water systems could further be checked or his nucleation rate may not be scaled directly to give the desired nucleator working volume.
5. Ripeners -- heat transfer area to volume ratio for both ripeners is very high (e.g., 7 ft^{-1} and 5.9 ft^{-1}, respectively) compared

to the value (1.5 ft^{-1}) existing in good vacuum pan design. This would suggest that such ripeners, whose volumes are small, have heating calandrias which occupy a space approximately of 1:2 ratio of slurry to heating tubes. Should the working volume be increased, the growth rate decreases (for an increase in τ at fixed $G\tau$), and the supersaturation drops dramatically, such that perhaps the seeds would dissolve. Thus, it is suggested that other kinds of evaporators be adopted for both ripeners which may not require calandria heaters, e.g., wiped film evaporators. Utilizing forced circulation evaporators would almost certainly result in destruction of the seed crystals.

6. Test of procedure for estimating constant growth rate for batch design methods -- if a time variation function for growth rate can be developed, an average constant growth rate could be solved from zero to any time t. It may be checked for intermediate time (actually intermediate mean size).
7. Development of raw sugar classifier -- raw sugar carries with it some adhering molasses that affords classification somewhat difficult to achieve by screening. Some classification, like elutriation or fluidized bed drying, could be an area for further work.
8. Modification of program RECYC -- ideally for sugar work, it would be useful if the growth rate and exit liquor crystal content could have been read in for each vessel together with its make-up feed rates. The program would then solve for stage

volumes, production, and recycle rates. This could be a good modification for program RECYC.

9. Optimization of the design -- the recycling of molasses in the second and last stages is necessary in order to operate within the limiting crystal content. This renders the design non-optimum. Avoiding such recycling by intermediate centrifugation between stages could be a good area to look into.

APPENDIX A

NUMERICAL METHOD FOR CALCULATION OF BATCH SIZE DISTRIBUTION

A batch pan is fed with crystals with a size distribution $W_i(L)$, and is grown so the number mean size increases by Δ (see Figure A-1).

The population balance (Chapter 3) gives

$$\frac{\partial n'}{\partial t} = - G \frac{\partial n'}{\partial L} + D \frac{\partial^2 n'}{\partial L^2}$$

where n' is the population density (based on the total vessel contents rather than per volume of slurry). This may be solved numerically starting with n_i' calculated from the seed distribution.

Alternatively, the problem can be solved from first principle and this is perhaps easier. If the seed crystals were initially of one size only (L_i), the final size distribution would be a normal one, displaced to larger sizes by an increment Δ, i.e., with mean $(L_i + \Delta)$ and with standard deviation $(2D\Delta/G)$ (refer to Randolph and White, 1977). Thus, a computational method would be to split the size distribution in a number of discrete size ranges and to "grow" each monosize range into its displaced normal distribution result and sum the lot.

Let N_i be the number of particles in a population size range = $n_i \delta L$, where δL is the size interval chosen. After growth, let the

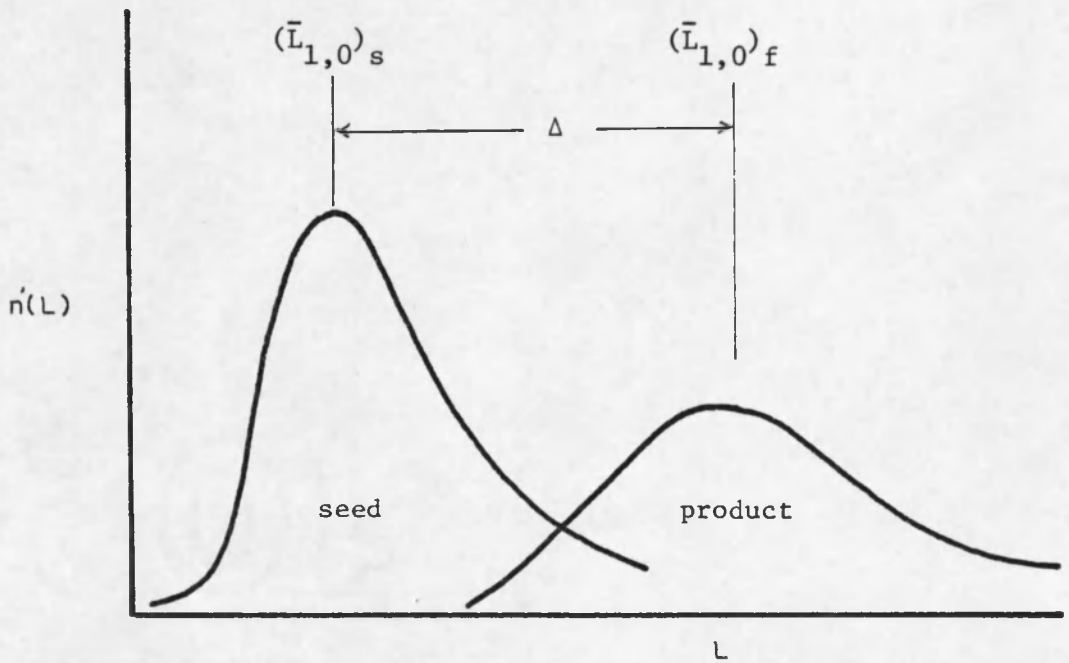


Figure A-1. Seed and Product Population Distribution Showing Δ.

fraction in the size range Δ larger be α_o (see Figure A-2) and in the size range either side of this be α_1, and the size range two intervals from the mean be α_2, etc.

Then, after growth and dispersion, the size distribution will be given by

$$N(L) = \alpha_o N_i(L - \Delta) + \alpha_1 [N_i(L - \Delta - 1) + N_i(L - \Delta + 1)]$$

$$+ \alpha_2[N_i(L - \Delta - 2) + N_i(L - \Delta + 2)] + \alpha_3 \ldots$$

This is a simple algorithm and the solution can be obtained easily. The size distribution so obtained can be converted to a mass distribution in the usual manner.

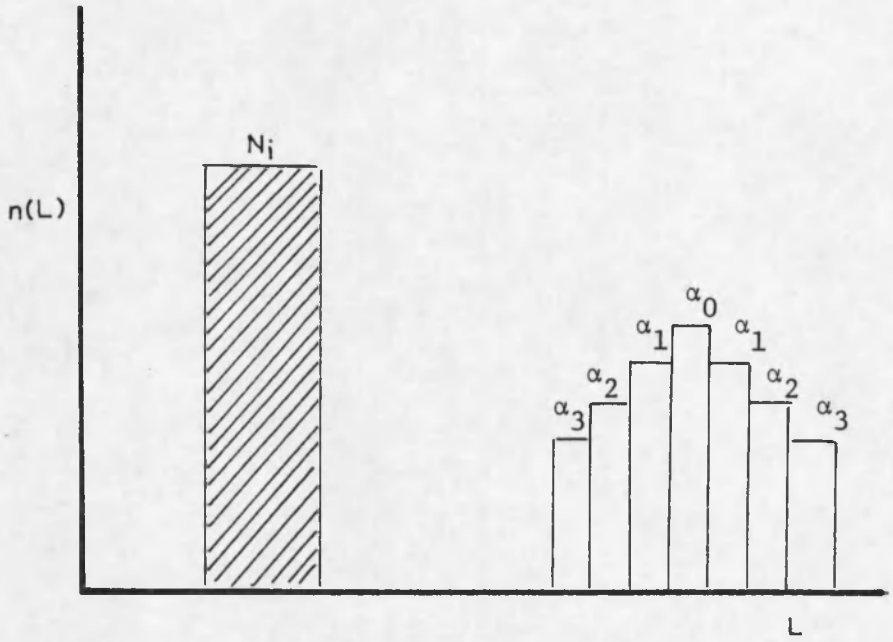


Figure A-2. Number Population Distribution Showing Increments α_0, α_1, α_2.

APPENDIX B

METHOD OF OBTAINING GAMMA PARAMETERS

The gamma parameters are obtained from appropriate mean size and coefficient of variation, all on weight basis, as follows. Given a gamma distribution fraction

$$f(L) = \frac{1}{\Gamma(\alpha + 1)\left(\frac{\beta}{\alpha}\right)^{\alpha+1}} L^{\alpha} \exp\left(-\frac{\alpha}{\beta} L\right)$$

the jth moment can be obtained from the definition in Equation 6 (Chapter 3) as

$$m_j = \frac{\beta^j \Gamma(j + \alpha + 1)}{\alpha^j \Gamma(\alpha + 1)}$$

Moments can be generated from j = 1 to j = 5 from which, by definition of normalized mean size and coefficient of variation

$$CV_{wt} = \frac{1}{(\alpha + 4)^{1/2}}$$

and

$$\bar{L}_{wt} = \frac{\beta(\alpha + 4)}{\alpha}$$

Once CV and $\bar{L}$ are known, α and β can be determined.

NOMENCLATURE

a	Population mean size increase in the nucleator.
A	Recycling surface area.
b	Population mean size increase in ripener 1.
$\bar{B}$	Birth function, #/μm-cc-time.
c	Population mean size increase in ripener 2.
CV	Coefficient of variation: CV_{pop} [=] population basis; CV_{wt} [=] weight basis.
D	Growth dispersion coefficient, μm^2/min.
$\bar{D}$	Death function, #/μm-cc-time.
DS	Dissolved solids.
E_{act}	Energy of activation of the crystallization reaction.
f(L)	Distribution fraction.
F_1	Stream of syrup in nucleator.
F_2	Stream of alcohol in nucleator.
G	Growth rate, μm/min.
G_τ	Characteristic size, microns.
hp	Horsepower.
I	Mass of impurity components in the pan.
L	Particle size, μm.
$\bar{L}$	Mean size: $\bar{L}_{pop}$ [=] population basis; $\bar{L}_{wt}$ [=] weight basis.
$\bar{L}_{1,0}$	Population or number mean size = L_i.
$\bar{L}_{3,0}$	Volume mean size.

$\bar{L}_{4,3}$ Mass mean size.

m_j The jth moment: $m_{j,1}$ [=] seed moment; $m_{j,k}$ [=] kth stage.

M_c Mass of crystal, gm.

M_T Solid concentration, gm/cc.

n Population density: n_i [=] seed population density;

n_k [=] kth stream.

n^o Nuclei density, #/cc-μm.

OS Oversupersaturation coefficient of the solution.

P_1-P_4 Parameters in the growth and solubility expression.

q Heat transfer rate.

Q Flow rate: Q_i [=] seed; Q_{sj} [=] syrup; Q_k [=] kth stream.

R Gas constant.

S Mass of sucrose in the pan.

(S/W)* Ratio of sucrose and water at saturation.

T Temperature.

TP True purity.

ΔT_L Delta log mean temperature difference.

U Overall heat transfer coefficient.

V Clear liquor volume, gallons.

w(x) Dimensionless weight fraction.

W Mass of water in the pan.

x Dimensionless size: $x_1 = L/a$ for nucleator; $x_2 = L/b$; $x_3 = L/c$.

α Gamma parameter, microns.

β Gamma parameter, dimensionless.

β^o Nucleation rate, nuclei/cc-time.

Δ	Increment.
π	3.1416, a constant.
ρ_c	Density of crystal, gm/cc.
σ	Standard deviation.
τ	Residence time.

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