# A Population Balance Approach to Direct-Contact Secondary Refrigerant Freezing

A population balance approach is used in developing an analysis of homogeneous freezing in a brine-ice slurry by direct-contact heat transfer to a dispersed secondary refrigerant. Exponential and beta distributions are assumed for the ice crystals and refrigerant droplets, respectively. The effects of freezer pressure (refrigerant temperature), inlet refrigerant drop size, and refrigerant and brine flow rates on well-mixed freezer characteristics are presented.

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## Introduction

An analytical study of freezing in a well-mixed liquid-ice slurry due to contact with evaporating refrigerant droplets is presented. Such a process has been studied in relation to freeze desalination and thermal energy storage, and is important in other applications involving crystallization. The method of analysis presented has generic significance, although the techique is applied to freeze desalination in the present paper. A mixture of crystal, solute, and evaporating liquid droplets exists in a steady state freezer fed separately by liquid and refrigerant spray. The product crystals exit the freezer in a slurry of liquid and carryover refrigerant. The analysis attempts to relate freezer pressure (which controls refrigerant saturation temperature), inlet liquid flow rate, and inlet refrigerant flow rate to the ice fraction and liquid refrigerant carryover, which is essential to understanding the behavior and stability of such a unit.

A population balance approach is presented wherein both the number of solid particles and the number of evaporating liquid droplets in the liquid are accounted for by use of separate population balances. An exponential size distribution is used for the ice particle distribution, as presented by Randolph and Larson (1971) and as used extensively in prior crystallization studies. A scaled, two-parameter beta distribution is used for the refrigerant droplets. Such a distribution is particularly effective in representing physical processes that extend over a finite size range and that often display skew. Both include particle radius as the independent variable. The differential equations describing the population balances are then transformed into nonlinear algebraic moment equations that are solved numerically in conjunction with other equations describing the freezer. The objectives of the study were to investigate the feasibility of coupling a beta distribution of dispersed refrigerant droplets to an exponential distribution of ice crystals, within a population balance analysis, and to evaluate the effectiveness of the procedure in describing freezer behavior for the specific application of seawater desalination with butane refrigerant.

A schematic illustration of a steady state, well-mixed, direct-contact, secondary refrigerant freezer with no mechanical agitation or separation of refrigerant from the exit slurry is shown in Figure 1. Entering the freezer at one end, the brine is thoroughly mixed with refrigerant droplets pumped into the bottom of the freezer. These droplets partially evaporate and leave the freezer as vapor through the compressor and as liquid carryover with the exit brine-ice slurry. Thus, at any given instant there exists in the active volume of the freezer a mixture of brine, ice, and composite liquid-vapor droplets of refrigerant. The freezer is assumed to be well mixed, that is, any reasonably sized sub-

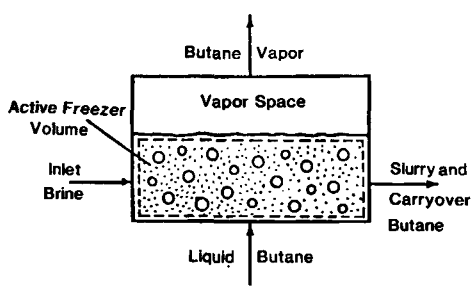


**Figure 1. Freezer diagram showing multicomponent and multiphase working medium.**

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volume of slurry contains the same particle size distribution as the bulk freezer, and the exit flow is assumed to be the same except that the vapor component exits separately through a compressor.

The simple mixed-suspension, mixed-product removal concept that results in an exponential crystal size distribution has been used to describe industrial crystallization kinetics for some time. By introducing an additional size distribution, the method can be extended to study the mixing of dissimilar particles.

## Mathematical Formulation

Generally, the mathematical formulation of the freezer presented here is based on:

- A population balance for the refrigerant
- The Kane et al. (1974) crystal distribution parameters for ice
- A heat and mass balance between the refrigerant and slurry
- A salt balance
- Two equations describing well-mixed behavior
- A volume relation
- Brine properties.

An integral transformation of the number conservation equations (population balance) reduces the differential equations to nonlinear algebraic moment equations. By assuming that the butane size distribution can be modeled as a scaled beta distribution, an unknown can be eliminated to form a closed system of equations. The heat and mass balance equates the energy absorbed by the butane to the energy lost by the brine plus energy dissipated by mixing. The salt balance provides a simple equation relating the mass ice fraction to the bulk salinity in the freezer. Well-mixed behavior is described mathematically by setting the ratio of the exit ice-butane mass flow rates equal to the ratio of the ice-butane masses in the freezer. Also, the size distributions in the exit flow are set equal to the distributions in the freezer. The volume relation states that the volume of refrigerant plus the volume of slurry is equal to the active freezer volume. The brine property relations are taken from the Office of Saline Water *Technical Data Book*. They provide equations for the freezer salinity and brine density in terms of the equilibrium freezing temperature.

## Analysis

### *Refrigerant number conservation equation*

Let $f(r)$ be defined as the number fraction of refrigerant droplets per unit freezer volume with a bubble radius between $r$ and $r + dr$ at any instant in time. The rate at which drops accumulate in this size range is given by Orcutt et al. (1971) as

$$N\frac{\partial}{\partial r}[g_o(r)f(r)] = Q^o f^o(r) - Q^{co} f^{co}(r) - Q^v f^v(r) + \Lambda(r) - \Omega(r) \quad (1)$$

The terms in Eq. 1 can be explained as follows. The lefthand side represents the net growth into the size range $r$ to $r + dr$ due to heat transfer. The first term on the right gives the number of droplets that enter the size range due to the inlet droplets. The second term is the number of droplets that leave as carryover with the exit slurry. The third term is the number of drops leaving as vapor. The final two terms, which will be neglected in the calculations of this work, respectively represent droplets that enter and leave the size range due to agglomeration of the bubbles. Since it is assumed that the refrigerant droplets enter the freezer as liquid with radius $r = r_1$, the inlet distribution can be described by the delta function $f^o(r) = \delta(r - r_1)$, which has the following properties

$$\delta(r - r_1) = \begin{cases} 0 & r \neq r_1 \\ \infty & r = r_1 \end{cases} \quad (2a)$$

$$\int_o^\infty F(r)\delta(r - r_1)dr = F(r_1) \quad (2b)$$

Similarly, since the vapor flow consists of completely evaporated droplets, they all have the radius $r = r_2$ if agglomeration is neglected. Thus, the vapor distribution is given by $f^v(r) = \delta(r - r_2)$. The well-mixed assumption implies the carryover distribution is the same as that in the freezer, that is, $f^{co}(r) = f(r)$.

### *Droplet growth term $g_o(r)$*

The droplet growth term $g_o(r)$ is given by Byrd (1984) as

$$g_o(r) = \frac{dr}{dt} = h\Delta T_r(\phi_o - 1)/\rho_\ell\lambda_r \quad (3)$$

where the heat transfer coefficient is approximated as

$$h = c_o + c_1 r (\mathrm{W/m^2 \cdot {}^\circ C}) \quad (4)$$

By assuming that the droplets are spherical, the growth can be characterized by a single variable, the radius. The empirical relation used for the heat transfer coefficient is based on the data of Simpson et al. (1974), which use an equivalent spherical diameter although the actual shape is much more complex.

### *Refrigerant moment equations*

The moment equations are developed by multiplying both sides of Eq. 1 by $r^n (n = 0, 1, 2, 3, \ldots)$ and integrating from $r = 0$ to $\infty$. The moments, $\nu_n$, are defined as

$$\nu_n = \int_o^\infty r^n f(r) dr \quad (5)$$

The physical significance of the first four moments are

$$\nu_o = 1 \text{ for a normalized distribution} \quad (6a)$$

$$\nu_1 = \text{mean bubble radius} \quad (6b)$$

$$\nu_2 \sim \text{total bubble area in freezer,} \quad A = \int_o^\infty 4\pi r^2 N f(r) dr \quad (6c)$$

$\nu_3 \sim$ total bubble volume in freezer,

$$V = 4/3\pi \int_o^\infty r^3 N f(r) dr \quad (6d)$$

In the present analysis, the refrigerant droplets entering the freezer have uniform size and composition. Since no agglomeration or breakup occurs, the droplets have the same mass at all times. Thus it can be shown that $\rho_o \nu_3^o = \rho_v \nu_3^v = \rho\, \nu_3$ and $\nu_n^v = \phi_o^{n/3} \nu_n^o$ where:

$\nu_3^o$ = third moment of the inlet distribution, m$^3$

$\nu_3^v$ = third moment of the vapor distribution, m$^3$

$\rho$ = mean density of an evaporating droplet, kg/m$^3$.

Using these relations and writing the mass flow rates in terms of the appropriate moments gives:

n moment equation

$$(1)\quad -k_1\Delta T_r N(C_o + C_1\nu_1) = Q^o(\nu_1^o - \nu_1) + Q^v(\nu_1 - \nu_1^o\phi_o^{1/3}) \quad (7a)$$

$$(2)\quad -2k_1\Delta T_r N(C_o\nu_1 + C_1\nu_2) = Q^o(\nu_2^o - \nu_2) + Q^v(\nu_2 - \nu_2^o\phi_o^{2/3}) \quad (7b)$$

$$(3)\quad -3k_1\Delta T_r N(C_o\nu_2 + C_1\nu_3) = Q^o(\nu_3^o - \nu_3) + Q^v(\nu_3 - \nu_3^o\phi_o) \quad (7c)$$

where $k_1 = (\phi_o - 1)/(\rho_\ell\lambda_r)$ and the integral of the growth term is evaluated using integration by parts.

### *Freezer heat and mass balance*

The energy lost by the brine as it is cooled and freezes, and the work dissipated in mixing of the slurry are absorbed by the refrigerant. The heat removed from the brine is the sum of the following:

1. Sensible cooling to the equilibrium freezing point corresponding to the inlet salinity.
2. Cooling of the brine that does not freeze to the subcooled bulk temperature.
3. The latent heat of fusion for the fraction of the brine that does freeze.

Written as a positive quantity using a salinity-dependent specific heat for the brine, this heat flux is

$$\dot{Q} = \dot{m}_{br}\left[\int_{T_{fi}}^{T_{in}} C_{pi}\,dT + (1-\epsilon_m)\int_{T_f}^{T_{fi}} C_p(X_s)\,dT_f + (1-\epsilon_m)\int_T^{T_f} C_p(X_s)\,dT + \epsilon_m\lambda_{br}\right] + W_m \quad (8)$$

The specific heat as a function of salinity for a sodium chloride-water system can be approximated as

$$C_p = 4.187 - 0.0565X_s \quad (9)$$

For this equation to be used the salinity must be known as a function of the temperature. This can be found from the phase diagram for a sodium chloride-water system as

$$T_f = 0.2911 - 0.61511X_s \quad (10)$$

Both of these brine property values are only approximations for ocean water, which contains several dissolved salts along with the principal solute, sodium chloride. However, since sea water varies in composition and concentration of salts at different locations, measurements at a specific site would have to be taken if more accuracy is desired. The brine is assumed to enter the freezer at its equilibrium freezing temperature. The power dissipated by mixing as the refrigerant is sparged into the bottom of the freezer is difficult to determine for different operating conditions. In most cases this is neglected compared to the heat transfer that occurs in the freezer. However, certain values of power dissipation are used in the determination of the ice crystal nucleation, so this term is included to be consistent in the analysis. Using the relations given by Eqs. 9 and 10, Eq. 8 can be written as

$$\dot{Q} = \dot{m}_{br}\{C_{pi}(T_{in} - T_{fi}) + (1-\epsilon_m)[a_1(T_{fi} - T_f) + a_2(T_{fi}^2 - T_f^2) + (4.187 - 0.0565X_s)(T_f - T)] + \epsilon_m\lambda_{br}\} + W_m \quad (11)$$

where $a_1$ and $a_2$ result from using the equations for the brine properties.

The heat transferred to the refrigerant, calculated using the convective heat transfer coefficient and total bubble area, is

$$\dot{Q} = \int_o^\infty 4\pi r^2 N h\Delta T_r f(r)\,dr \quad (12)$$

Using Eqs. 3, 11, and 12, the heat and mass balance can be written as

$$4\pi k_1 N\Delta T_r(C_o\nu_2 + C_1\nu_3) = \dot{m}_{br}\{\epsilon_m\lambda_{br} + (1-\epsilon_m)[a_1(T_{fi} - T_f) + a_2(T_{fi}^2 - T_f^2) + (4.187 - 0.0565X_s)(T_f - T)]\} + W_m \quad (13)$$

### *Salt balance*

A salt balance can be written as

$$\dot{m}_{br}X_{si} = \dot{m}_{s1}(1-\epsilon_m)X_s \quad (14)$$

where

$X_{si}$ = mass fraction of salt in inlet brine

$\dot{m}_{s1}$ = mass flow rate of exit slurry/unit volume, kg/s · m$^3$.

For steady state $\dot{m}_{br} = \dot{m}_{s1}$, which gives

$$\epsilon_m = 1 - X_{si}/X_s \quad (15)$$

### *Well-mixed behavior relations*

Well-mixed behavior has already been incorporated into several of the equations previously derived. The refrigerant moment equations, the heat and mass balance, and the salt balance reflect the assumption that the composition of the carryover stream is the same as the uniform composition in the freezer. The mass ice fraction, $\epsilon_m$, in the exit slurry is the same as that calculated from Eq. 15. Writing the mass flow rate of the ice leaving the freezer in terms of crystal parameters and rearranging gives

$$Q_c' = \frac{\dot{m}_{br}\epsilon_m}{(4/3)\pi\rho_i\zeta_3} \quad (16)$$

Similarly, for the refrigerant

$$\frac{\dot{m}_{co}}{\dot{m}_{s\ell} + \dot{m}_{co}} = \frac{M_r}{M_r + M_{s1}} \quad (17)$$

where

$$M_r = (4/3)\pi\rho_r N\nu_3$$

and

$$M_{sl} = M_i/\epsilon_m = \frac{(4/3)\pi\rho_i N_c \zeta_3}{\epsilon_m}$$

Writing Eq. 17 in terms of refrigerant moments and rearranging gives

$$N_c/N = Q_c'/(Q^o - Q^v) \tag{18}$$

### *Freezer volume relation*

The active volume of the freezer consists of slurry and refrigerant. The volume of the components can be written on a unit volume basis as

$$(4/3)\pi\nu_3 N + \frac{(4/3)\pi\rho_i N_c \zeta_3}{\rho_{sl}\epsilon_m} = 1 \tag{19}$$

$\rho_{sl}$ is the average density of the slurry and can be written in terms of brine and ice density as

$$\rho_{sl} = \frac{\rho_{br}\rho_i}{\rho_i + \epsilon_m(\rho_{br} - \rho_i)} \text{ kg/m}^3 \tag{20}$$

The brine density is approximated as a linear function of the salinity as

$$\rho_{br} = 999.5 + 7.897X_s \text{ kg}^3/\text{m}^3 \tag{21}$$

### *Ice crystal size distribution*

The crystal size distribution has been formulated assuming size-independent crystal growth and nucleation at zero size by Randolph and Larson (1971) as

$$f_c(r_c) = f_o \exp(-\beta r_c/G) \tag{22}$$

where $f_o = \beta/G$, and $\beta$ is the nucleation rate per crystal (1/s). The moments, $\zeta_n$, can be calculated in a straightforward manner by integration; they have the same physical significance for the ice distribution that the refrigerant moments, $\nu_n$, have for the refrigerant distribution. A population balance for the crystals gives

$$\beta N_c = Q_c' \tag{23}$$

where $Q_c'$ is the rate at which crystals are removed from the freezer.

### *Crystal nucleation and growth relations*

Kane et al. (1975) studied the effect of brine subcooling, $\Delta T_f$, salt concentration, $X_s$, and power dissipation, $P_v$, on $\beta$ for small values of $\epsilon_m$. Their results can be approximated as

$$\beta = [0.8472 + 6.04(10^{-4})P_v - 1.032(10^{-7})P_v^2] \\ (1.557 - 0.02566X_s) \cdot (\Delta T_f)(1.4 + 0.066X_s) \tag{24}$$

They also published expressions giving $G$ for fresh water and 5.3% brine as a function of $P_v$ and $\Delta T_f$. Assuming a linear dependence on the salinity results in the following equation for the growth rate

$$G = [1.704(10^{-5}) - 0.2087(10^{-5})X_s]P_v^{0.25}\Delta T_f \tag{25}$$

Thus, using Eqs. 23, 24, and 25, the ice crystal moments can be calculated for a given $P_v$ if $T$ and $T_f$ are known.

### *Beta distribution*

The beta distribution is given by Mood et al. (1974) on an interval of $(0, \infty)$ as

$$f_b(x) = x^{a-1}(1 - x)^{b-1}I_o(x)/B(a, b) \tag{26}$$

where $a$ and $b$ are variable parameters greater than zero,

$$B(a, b) = \Gamma(a)\Gamma(b)/\Gamma(a + b) \tag{27}$$

and $\Gamma(x)$ is the gamma function. As shown in Figure 2, this distribution has a wide range of shapes depending on the values of $a$ and $b$. The moments, $\mu_n$, are given by

$$\mu_n = B(n + a, b)/B(a, b) \tag{28}$$

By using the transformation $x = (r - r_1)/(r_2 - r_1)$ and normalizing by dividing by $(r_2 - r_1)$, the nonzero part of this distribution can be extended over any finite interval of interest. The scaled distribution can be written as

$$f(r) = f_b[(r - r_1)/r_{21}]/r_{21} \tag{29}$$

where $r_{21} = r_2 - r_1$. The moments of the scaled function are

$$\nu_o = 1 \tag{30a}$$

$$\nu_1 = r_1 + r_{21}\mu_1 \tag{30b}$$

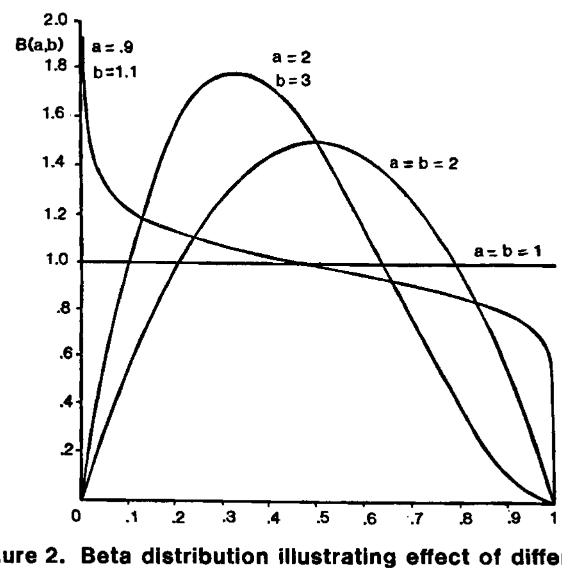


**Figure 2. Beta distribution illustrating effect of different values of parameters *a* and *b*.**

$$\nu_2 = r_1^2 + 2r_1r_{21}\mu_1 + r_{21}^2\mu_2 \quad (30c)$$

$$\nu_3 = r_1^3 + 3r_1^2r_{21}\mu_1 + 3r_1r_{21}^2\mu_2 + r_{21}^3\mu_3 \quad (30d)$$

$$\nu_3 = r_1^3 + 3r_1^2r_{21}\mu_1 + 3r_1r_{21}^2\mu_2 + r_{21}^3\mu_3 \quad (30d)$$

## Numerical Solution

The specific problem solved in this paper uses butane as the refrigerant, freezing a 3.498% brine solution. The brine enters at its equilibrium freezing temperature while the butane enters as saturated liquid. The model can now be written in terms of Eqs. 7a–c, 13, 16, 18, 19, and 23 with $N$, $Q^o$, $a$, $b$, $T$, $T_f$, $N_c$, and $Q_c'$ as unknowns. $\nu_n$ can be calculated using Eqs. 28 and 30. $X_s$ is determined from Eq. 10, allowing $\epsilon_m$ and $\rho_{br}$ to be calculated from Eqs. 15 and 21. The slurry density is taken from Eq. 20. The system of equations can be reduced by using Eq. 23 to eliminate $N_c$, Eq. 16 to eliminte $Q_c'$, and this result to produce the following equations with $a$, $b$, $T$, and $T_f$ as unknowns:

$$k_1\Delta T_r\eta_2(C_o/\nu_2^o + C_1\bar{\nu}_1) + Q^o(1 - \bar{\nu}_1) + \eta_1(\bar{\nu}_1 - \phi_o^{1/3}) = 0 \quad (31)$$

$$2k_1\Delta T_r\eta_2(C_o\nu_1/\nu_2^o + C_1\bar{\nu}_2) + Q^o(1 - \bar{\nu}_2) + \eta_1(\bar{\nu}_2 - \phi_o^{2/3}) = 0 \quad (32)$$

$$\dot{Q} - 4\pi\eta_2\Delta T_r(C_o\nu_2 + C_1\nu_3) = 0 \quad (33)$$

$$(4/3)\pi N\nu_3 + \frac{\dot{m}_{br}}{\beta\rho_{s1}} - 1 = 0 \quad (34)$$

$$\eta_1 = \frac{Q^o[3k_1\Delta T_r(C_o\nu_2 + C_1\nu_3) + \beta(\nu_3^o - \nu_3)]}{\beta(\nu_3^o\phi_o - \nu_3) + 3k_1\Delta T_r(C_o\nu_2 + C_1\nu_3)} \quad (35)$$

$$\eta_2 \equiv \frac{Q^o - \eta_1}{\beta} \quad (36)$$

where $\bar{\nu} = \nu/\nu^o$ and $\bar{\nu} = \nu/\nu^o$. These equations were solved by using the IMSL subroutine ZSPOW, which uses a form of M. J. D. Powell's algorithm.

## Results and Discussion

In Figures 3 through 6 various output variables are plotted against the butane saturation temperature, $T_{sat}$, as a function of two parameters. The first is the brine flow rate, $\dot{m}_{br}$, and the other is either the refrigerant flow rate, $\dot{m}_r$, or the inlet drop radius, $r_1$. One set of curves with $\dot{m}_r = 0.1602$ kg/s · m$^3$, $r_1 = 1.676 \times 10^{-3}$ m, and $P_v = 1{,}000$ W/m$^3$, is the same for each plot. The inlet brine temperature is $\simeq -1.83$°C.

### *Freezer response to changes in $T_{sat}$, $\dot{m}_r$ and $\dot{m}_{br}$*

The percent liquid butane carryover is seen in Figure 3 to increase with saturation temperature. This is expected because the bubble growth rate is proportional to $\Delta T_r$, which decreases as $T_{sat}$ increases. Campbell and Duvall (1978) reported excessive carryover for $\Delta T_r = 1.67$°C in pilot plant operations using Freon 114 as the refrigerant.

Increasing the butane flow rate increases the number density in the freezer, resulting in more carryover. A larger brine flow rate has the same effect because the residence time of the slurry decreases. Thus, even though the number density may be smaller, more slurry leaves the freezer per unit time, increasing the carryover.

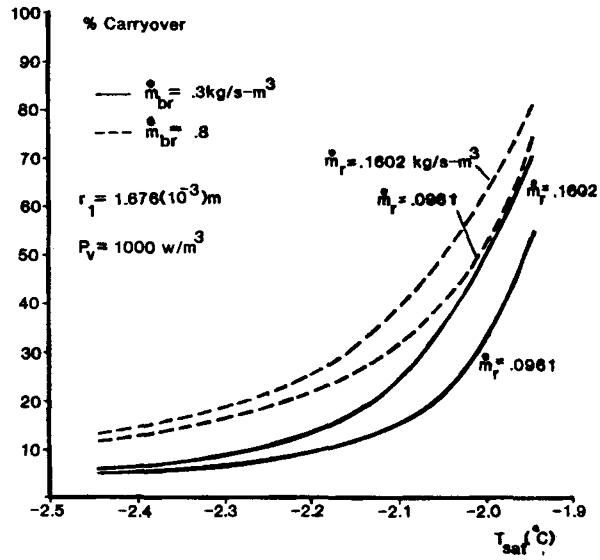


**Figure 3. Liquid-refrigerant carryover as a function of refrigerant flow rate, brine flow rate, and saturation temperature of refrigerant (or freezer pressure).**

The mass ice fraction, $\epsilon_m$, approaches a maximum as $T_{sat}$ decreases, as can be seen in Figure 4. If there is no carryover and brine subcooling is neglected, a heat and mass balance on the

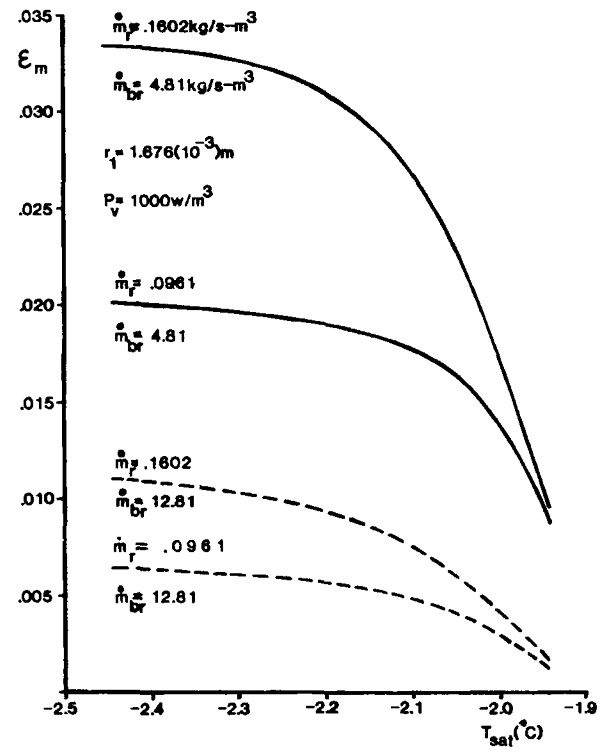


**Figure 4. Slurry ice fraction on a weight basis as a function of refrigerant and brine flow rate, and refrigerant saturation temperature (or freezer pressure).**

freezer gives

$$\dot{m}_{br}\epsilon_m\lambda_{br} = \dot{m}_r\lambda_r \rightarrow \epsilon_m = (\dot{m}_r/\dot{m}_{br})(\lambda_r/\lambda_{br}). \qquad (37)$$

For the operating conditions of $\dot{m}_r = 0.1602$ kg/s · m³, $\dot{m}_{br} = 4.81$ kg/s · m³ at $T_{sat} = -2.44$°C, the mass ice fraction is $\simeq 0.0377$. The model predicts $\epsilon_m = 0.0335$ with approximately 6% carryover. Operation at a lower temperature will not greatly increase the ice production but may be important in reducing carryover. Increasing $\dot{m}_r$ or decreasing $\dot{m}_{br}$ results in a larger ice fraction, as expected. The values of $\epsilon_m$ are relatively low, less than 4% compared to a commercial freezer, which may operate up to approximately 20%. However, because the ice crystal kinetics were claimed to be more accurate for low ice fractions, this range of parameters was emphasized in the calculations.

### *Freezer Response to Changes in Inlet Drop Radius*

The butane carryover increase with $r_1$ shown in Figure 5 results from two factors. First, the droplets have more mass and take longer to evaporate. Second, increasing the drop size decreases the total bubble area entering the freezer for the same mass flow rate. The proper inlet drop size was found by Orcutt et al. (1971) to be essential for practical pilot plant operation.

The ice fraction increases slightly as $r_1$ decreases due to the lower carryover, as seen in Figure 6. It can be seen that this is more effective at higher values of $T_{sat}$ where carryover is more important.

Figure 7 is a plot taken from the final report of Ganiaris et al. (1969) on a pilot plant of Struther. This graph represents their best approximation to crystal size as a function of the square root of the residence time when the refrigerant was introduced into the freezer through fog nozzles. Due to the lack of data, the slopes of the solid segments of the lines were taken to be the same as those found when the refrigerant was introduced into the freezer through 1/8 in (3.2 mm) holes. The dashed segments of the line represents extrapolation to lower residence times. The straight lines from the origin were believed to represent minimum freezer performance as suggested by the mathematical analysis of Orcutt et al. (1971). The mean diameters calculated from $\zeta_1$ are included for comparison.

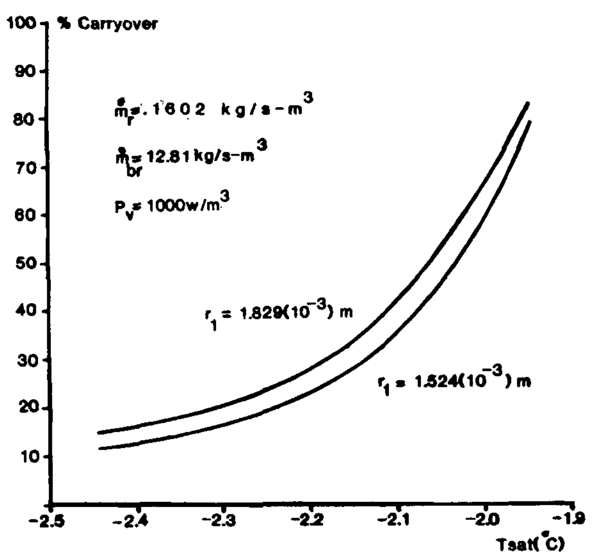


**Figure 5. Effect of entering liquid-refrigerant drop size on liquid-refrigerant carryover.**

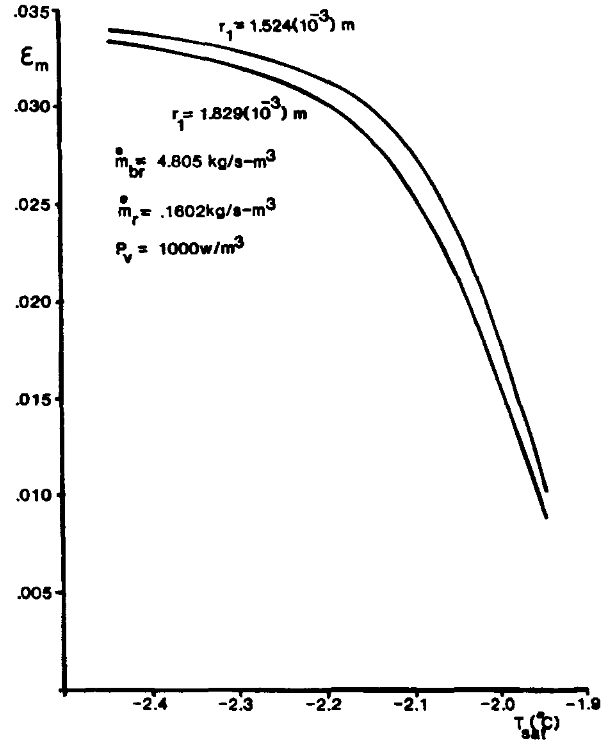


**Figure 6. Effect of entering liquid-refrigerant drop size on product ice fraction.**

The experimental values of $D_e$ were calculated from permeability measurements assuming a spherical shape. They were found to be considerably lower than the mean diameter estimated from photographs of the crystals. This may have been because the crystals were not truly spherical. For this work values of the freezer input parameters were estimated from averages of data for representative runs tabulated in the final report of Ganiaris et al. A paddle wheel was used to provide additional mixing. The power consumed, approximately 2,290 W/m³, was used for $P_v$. This is much larger than in the bench-scale experimental studies used to establish the ice crystal kinetics, so the

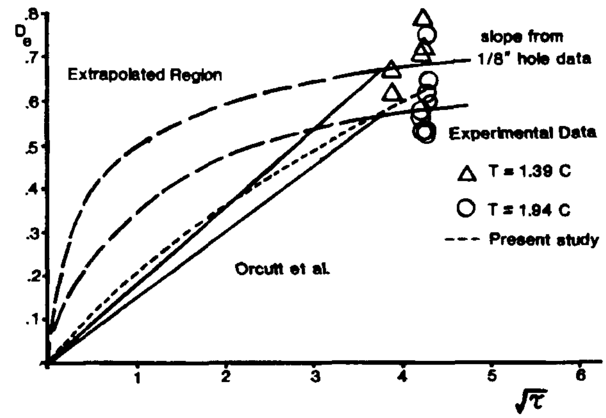


**Figure 7. Equivalent diameter of product ice particles as a function of residence time of slurry.**

risk of error in extrapolating a nonlinear relation is unavoidable. Butane-1 and butane were used as the refrigerant on different runs and the experimental residence time was approximated as $\tau = V_{sl}/q_{sl}$. This neglects the presence of the refrigerant in the slurry. The value of $T_{sat}$ used was $-3.89°C$, which resulted in a $\Delta T_r$ between 1.67 and 1.94°C, depending on the brine flow rate.

As can be seen, the mean diameter calculated in the present study gives reasonable agreement with the actual recorded experimental data. Orcutt et al. predicted $D_e$ to be approximately a linear function of the square root of the residence time, which is also similar to the present study. They also suggest that the mean size decreases as $\Delta T_r$ increases from 1.39 to 1.94°C due to increased nucleation. This is not seen in the present model except for larger values of $T_{sat}$, possibly because of hydrostatic effects. The mass ice fractions predicted by the model are approximately 0.197, which is only slightly lower than the experimental values of approximately 0.209. This is not surprising in view of the low amount of carryover when operating at this value of $T_{sat}$.

## Conclusions

The use of a population balance approach in analyzing the behavior of a direct-contact, secondary refrigerant crystallizer in which the refrigerant is dispersed as liquid droplets gives physically realistic trends for the ice fraction and liquid refrigerant carryover in the product outflow. A two-parameter beta distribution was found to work well for the refrigerant droplets and to couple conveniently with an exponential crystal size distribution. Using this technique it was possible to show the effects of variations in inlet liquid flow rate, inlet refrigerant flow rate and droplet size, and freezer pressure. In the special case of seawater and butane, it was found that the most important parameter reducing butane carryover is the freezer pressure (refrigerant saturation temperature), although decreasing the inlet drop size produces a similar effect. However, for saturation temperatures below approximately $-3.5°C$ there is little improvement. Potentially, the most significant aspect of the mathematical model is the ability to predict transient start-up behavior as well as provide data necessary to show optimum freezer performance. While neither of these are attempted in the present paper, it appears that with the proper extensions of the method such applications could be successful.

## Notation

$a$ = beta distribution parameter
$a_1$ = constant = 4.16026
$a_2$ = constant = 0.045927
$b$ = beta distribution parameter
$B(a, b)$ = beta function with parameters $a$ and $b$
$c_p$ = specific heat, J/kg°C
$c_{pi}$ = specific heat of the inlet brine, J/kg°C
$C_o$ = constant = 0.5956 W/m$^{2o}$C
$C_1$ = constant = 0.04652 W/m$^{3o}$C
$D_e$ = equivalent spherical diameter of ice crystals, m
$f^o(r)$ = inlet refrigerant size distribution, 1/m
$f^{co}(r)$ = carryover refrigerant size distribution, 1/m
$f^v(r)$ = vapor refrigerant size distribution, 1/m
$f(r)$ = freezer refrigerant size distribution, 1/m
$f_o$ = population density of embryo size ice crystals, 1/m
$f_b(x)$ = beta distribution nonzero on (0, 1)
$f_c(r_c)$ = ice crystal size distribution, 1/m
$g_o(r)$ = $dr/dt$, butane droplet growth rate, m/s
$G$ = ice crystal growth rate, m/s
$h(r)$ = refrigerant droplet heat transfer coefficient, W/m$^{2o}$C
$\dot{m}_{br}$ = brine mass flow rate/freezer volume, kg/s · m$^3$
$\dot{m}_r^o$ = refrigerant mass flow rate/freezer volume, kg/s · m$^3$
$M_i$ = total mass of ice in freezer at any given instant, kg
$M_r$ = total mass of butane in freezer at any given instant, kg
$M_{sl}$ = total mass of ice plus brine in freezer at any given instant, kg
$n$ = integer equal to 0, 1, 2, 3, . . .
$N$ = total number of butane drops/freezer volume, 1/m$^3$
$N_c$ = total number of ice crystals/freezer volume, 1/m$^3$
$P_v$ = power dissipated/freezer volume, W/m$^3$
$\dot{Q}$ = energy withdrawn from the brine/freezer volume-time, W/m$^3$
$Q^o$ = number of butane droplets/freezer volume-time fed into freezer, 1/m$^3$ s
$Q^{co}$ = number of droplets/freezer volume-time leaving as carryover, 1/m$^3$ s
$Q^v$ = number of droplets/freezer volume-time leaving as vapor, 1/m$^3$ s
$Q_c$ = number of ice crystals/freezer volume-time leaving in exit flow, 1/m$^3$ s
$r$ = drop radius, m
$r_1$ = initial drop radius, m
$r_2$ = radius of a totally evaporated drop, m
$r_c$ = ice crystal radius, m
$T$ = bulk brine temperature in freezer, °C
$T_f$ = equilibrium freezing temperature corresponding to bulk freezer salinity, °C
$T_{fi}$ = equilibrium freezing temperature at inlet brine salinity, °C
$T_{in}$ = temperature of inlet brine, °C
$T_{sat}$ = saturation temperature of refrigerant at mean freezer pressure, °C
$\Delta T_f = T - T_f$, °C
$\Delta T_r = T - T_{sat}$, °C
$V_f$ = freezer volume, m$^3$
$V_{tot}$ = total refrigerant volume per unit freezer volume
$W_m$ = work done by mixing per unit freezer volume and time, W/m$^3$
$X_s$ = bulk freezer salinity on a mass basis, % salt
$X_{si}$ = bulk salinity of inlet brine on a mass basis, % salt

### *Greek letters*

$\beta$ = nucleation rate per crystal, 1/s
$\delta(r)$ = delta function
$\epsilon_m$ = mass ice fraction
$\lambda_{br}$ = latent heat of fusion for brine, J/kg
$\lambda_r$ = latent heat of vaporization for butane, J/kg
$\Lambda(r)$ = number of droplets that enter size range $r$ to $r + dr$ due to agglomeration per unit time and volume, 1/s · m$^3$
$\Omega(r)$ = number of droplets that leave size range $r$ to $r + dr$ per unit time and volume due to agglomeration, 1/s · m$^3$
$\phi_o$ = ratio of liquid to vapor refrigerant density
$\eta_1$ = relation for $Q^v$
$\eta_2$ = relation for $N$
$\rho_{br}$ = brine density, kg/m$^3$
$\rho_i$ = ice density, kg/m$^3$
$\rho_\ell$ = liquid density, kg/m$^3$
$\rho_r$ = liquid refrigerant density, kg/m$^3$
$\rho_v$ = vapor refrigerant density, kg/m$^3$
$\zeta_n$ = $n$th moment of ice crystal size distribution, m$^n$
$\nu_n$ = $n$th moment of refrigerant size distribution, m$^n$
$\mu_n$ = $n$th moment of beta distribution

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*Manuscript received Oct. 23, 1985, and revision received Mar. 11, 1986.*