Recent Improvements to the
Freeze Crystallization Method of Water Purification

Dr. William M. Conlon, P.E.
Polar Spring Corporation
3501 Edison Way
Menlo Park, California 94025 USA
415/368-2852

ABSTRACT

Improvements to the freeze crystallization method of water purification make possible several novel residential and commercial appliance concepts. Operating in a batch process, source water enters a purifying chamber, a fraction of the water is purified via freeze crystallization and the unfrozen portion is discarded. The purified ice fraction is then melted and stored in a holding tank and the cycle is repeated. Careful optimization of temperature, size, and refrigeration parameters are required to maximize yield, energy and volumetric efficiency. This paper summarizes the state of the art of water treatment by freeze crystallization, describes the improved process and presents curves and equations relating process optimization. Several revolutionary appliance concepts are presented for residential and commercial water treatment products.

INTRODUCTION

Freeze crystallization has been used to separate a wide variety of contaminants from water, such as dissolved minerals, organic chemicals and particulates. Similar in principle to the methods used to purify silicon for semiconductor chips, freezing methods have also been used for seawater desalination. Continuous crystallizers have been the preferred approach, due to the availability of design methods and proven industrial process equipment. However mechanical complexity and high installed costs make continuous freezing processes too expensive for small-scale water treatment systems.

Previously, we reported on the application of alternative water treatment technologies to refrigerator water dispensers (1), and suggested that a batch freeze crystallization method would be preferable to conventional techniques such as activated carbon adsorption, ion exchange, distillation and reverse osmosis. Batch freeze crystallization technology is simpler and more compact than continuous crystallizers. It can be applied on small scales, and can potentially lead to a new class of drinking water appliances to serve both residential and commercial markets.

Batch freeze crystallization has several potential advantages over alternative water treatment processes:
- Performance — crystallization can remove high percentages of a wide variety of contaminants without the need for expendable filters or membranes;
- Reliability — cooling can be provided by proven household type refrigeration systems;
- Efficiency — freezing uses only 1/7 as much energy as boiling, and heat can be recovered to melt the crystals;
- Water Temperature — the melted crystals are prechilled and drinking water can be readily kept cold within an insulated appliance.

This paper presents an overview of freeze crystallization technology and reports on progress toward realizing the potential of batch crystallization for appliances.

SEPARATION BY FREEZING

Freeze crystallization is based on the principle that impurities are generally one to two orders of magnitude more soluble in the liquid phase of a solvent than in the solid phase. Freeze crystallization can be applied in separation processes to either purify or concentrate a material, depending on the desired end product. As the solvent crystallizes, the solute is concentrated in the remaining liquid solvent. The purified crystals are removed

from the impurity laden liquid (for example by gravity, filtration or centrifuging) and may be used in the solid phase or melted for use as a refined liquid. In some applications, the concentrated liquid may be the desired end product.

A wide variety of materials may be separated by freeze crystallization, for example water from seawater, milk solids from raw milk, or pure silicon from raw ore. Moreover, materials can be repeatedly crystallized to achieve very high degrees of purification. For example, in the production of purified silicon, each successive crystallization pass reduces the aluminum content by about two orders of magnitude. The remainder of this section reviews work by other investigators related to removal of contaminants from water.

Desalination

Freezing methods were of considerable interest for large scale desalination during the 1960s because of the potential energy savings compared to distillation processes. Research and development projects and several pilot plants were supported by the U.S. Office of Saline Water in the Department of the Interior. Results from a pilot plant operated in Israel are reported by Speigler (10). As with other dissolved species, water purification by freeze crystallization is based on the difference in component concentration between liquid and solid phases that are in equilibrium. The component concentrations are depicted in phase diagrams, such as Figure 1 for sodium chloride in water.
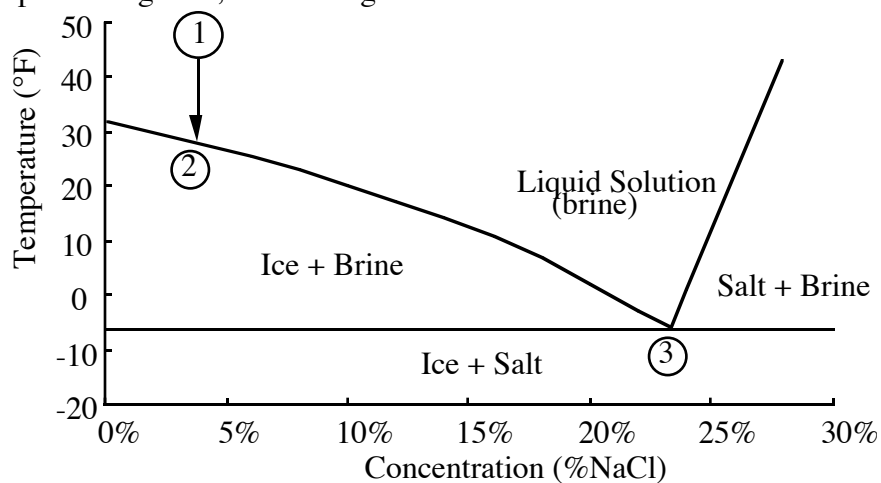

Figure 1. Seawater at point 1 is first cooled to point 2, where water ice crystals begin to form. Further cooling results in increasing salt concentration until the eutectic at point 3 is reached. Further cooling from 3 results in a solid solution of ice and salt.

The diagram shows four distinct phases as the temperature and salt concentration vary: a liquid solution or brine, a solid solution of water and salt, a mixture of ice and brine, and a mixture of salt and brine. Selective purification or concentration can be achieved, depending on the starting conditions. The figure depicts the cooling of seawater to form essentially pure ice crystals followed by increasing salt concentration until the eutectic, at a concentration of 23.3% NaCl. Further cooling from the eutectic would cause the formation of a solid solution. The maximum water extraction at the eutectic would be 85% [100 (1 - (3.5 ÷ 23.3))], but in practice, only about 20% of the water can be extracted. Also note that if the initial salt concentration had been 25%, salt crystals would have precipitated upon cooling, instead of ice crystals.

Organic Molecules

A simple experiment with common food coloring (propylene glycol and dye) mixed in distilled water demonstrates separation of organic chemicals by freeze crystallization. Freezing the mixture into ice cubes, where crystals grow from the outside to the inside, vividly concentrates the food coloring in the center of the cube.

The molecular diffusivity is an important factor in separation of organic chemicals from water by freezing. To be separated, the molecule must diffuse as fast or faster than the rate at which the ice front grows. Taylor (2) studied the separation of TNT and RDX from ice by placing a cold plate in contact with an aqueous solution of these large organic molecules. A high degree of separation was found when the ice velocity was less than about 0.125 inch per hour (90 x 10^{-6} cm/s). However in a region of higher ice velocity (a 0.25 inch thick transition layer next to the cold plate) separation was found to be only about 85%. Since the diffusivity of molecules is inversely proportional to the square root of molecular weight, small organic molecules, such as the trihalomethanes, would be expected to be readily separated. Our results confirm this effect.

Microorganisms
Freeze crystallization may remove microbial contaminants from water. We inoculated water samples with E. coli bacteria, and found no E. coli in the melted water. Further work is needed to determine the extent to which bacteria are separated or killed. Interestingly, in a literature review, Martel (3) cited work by Sedgwick (4) concerning bacterial removal from ice:

> "At the turn of the century, all of the ice consumed by the general public was gathered from lakes and ponds during the winter and stored in icehouses. Sedgwick (1903) noted that very few epidemics could be attributed to infected ice. He concluded that under the quiescent conditions beneath the ice, bacteria and other foreign particles were eliminated by the growing ice front."

Particulates
Several investigators (5, 6, 7) have found a critical ice front velocity below which particles are rejected by growing ice. If the growth rate of the ice front were controlled to less than a critical velocity, almost complete particulate separation would be expected.

Radionuclides
Since there is no chemical difference between radioactive isotopes and their stable counterparts, the separation of radioactive isotopes is expected to be identical to the corresponding stable isotope. Indeed this principle was used by Cox and Weeks to study brine drainage from ice using radioactive sodium as a tag (8).

CONTINUOUS FREEZE CRYSTALLIZATION
Freeze crystallization has been used in a variety of chemical separation processes (9) and is especially used to recover salts from aqueous media in the inorganic chemical industry. As a result, there are design methods and operating experience to support the development of continuous crystallization apparatus for new applications. Freeze crystallization was once believed to be the preferred method of desalination because of its favorable energy consumption compared to distillation. Most applications of freeze crystallization for water treatment have used variants of the vacuum freezing method, depicted schematically in Figure 2, and consisting of the following principal components:
- freezer, where heat is removed to crystallize part of the fluid;
- separator, where the crystals are separated from the unfrozen concentrated fluid and washed with a few per cent of the melted water;
- melter, where the separated crystals are melted, usually by means of the waste heat from a heat pump;
- heat exchanger, where the incoming feedwater is cooled using the melted water and concentrated brine from the freezer.

Vacuum freezing is accomplished by flashing a fraction of the feedwater, using the large latent heat of evaporation (1075 Btu per pound) to cool and crystallize a larger fraction of ice with a smaller latent heat of fusion (144 Btu per pound). Vacuum freezing methods are potentially quite energy efficient, but have the following drawbacks:
- Freezers. The vacuum freezer operates at the triple point of the fluid (the tempera-

ture at which vapor, liquid and solid phases are in equilibrium). The vacuum can be induced by a mechanical pump, steam ejector or absorber. Mechanical vacuum pumps require large multi-stage centrifugal fans or blowers which have size, efficiency and maintenance drawbacks. The absorption methods will not work if there are non-absorbable volatiles, which build up and inhibit heat removal. An alternative to the vacuum freezer is the indirect scraped surface heat exchanger. This alternative uses a refrigerant and operates at atmospheric pressure, but has a higher installed cost because of the complexity of the scraper.
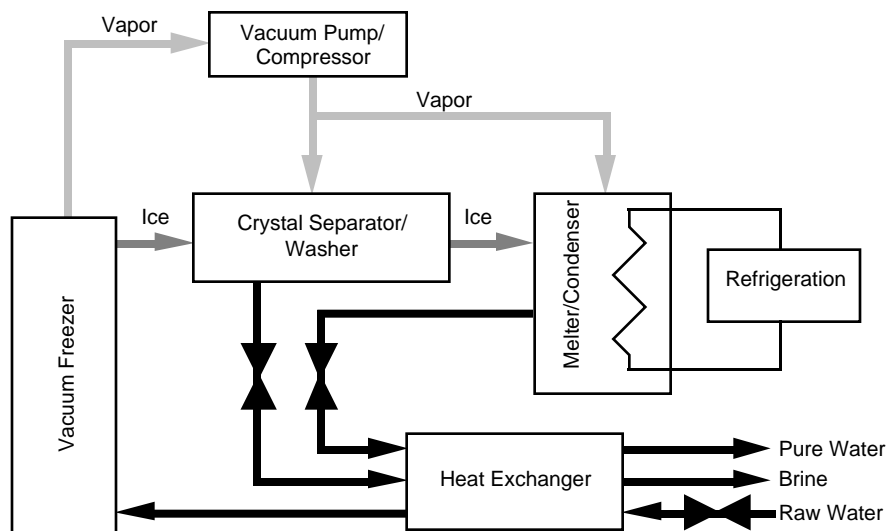

Figure 2.   Most continuous crystallizers for water treatment have used vacuum freezing, which operates at the triple point of water. In addition to the vacuum pump, the typical system requires the transport and washing of purified ice crystals.

• Separators.   The vacuum freezing method produces a multitude of small ice crystals which must be separated from the liquid. Also, the high surface-to-volume ratio of the small crystals means that the crystals must be washed to remove contaminated water adhering to the surface. The most effective method has been the wash column, but a large volume of liquid must also be transported along with the ice to facilitate pumping. In one desalination application 8% of the purified water was used for rinsing the crystals (10).

• Melters.   Vapor removed by the vacuum pump from the freezer is condensed onto the ice crystals to melt them. Usually the crystals are transported to a separate melt chamber, which increases the expense and complexity.

As described above, continuous crystallization requires complex process equipment, and very large scales are needed to achieve reasonable economy. For the foreseeable future, desalination will likely remain the province of technologies such as reverse osmosis and distillation.

BATCH CRYSTALLIZATION
Batch crystallization has been confined to small scale applications because of higher energy costs than continuous crystallizers and difficulties with reproducing the crystal size distribution from batch to batch. According to Moyers and Rousseau (11), "the development of methods for design and analysis of batch crystallizers lags those for continuous systems." This point is reinforced by the relative coverage of batch crystallization they provide — less than one fourth of a page in their 65 page survey article.

An improved batch freeze crystallization method is potentially simpler and more reliable than continuous crystallization methods (12). This improved method uses conventional refrigeration equipment in an optimized batch process. It can be implemented in a variety of sizes, ranging from small consumer appliances to unit equipment suitable for separating temperature sensitive chemical or biological products. The method operates as a batch process in a repeating sequence of steps, as illustrated in Figure 3.

In this batch process, water is treated entirely within a "Freeze Engine," which includes cooling and heating coils, and water valves. Since both freezing and melting occur within the Freeze Engine, ice transport is eliminated. The process operates at atmospheric pressure and cooling is provided by a conventional household-type vapor compression refrigeration system. The water valves admit source water for treatment, drain concentrated impurity laden water away, and direct the treated melt water to a holding tank.
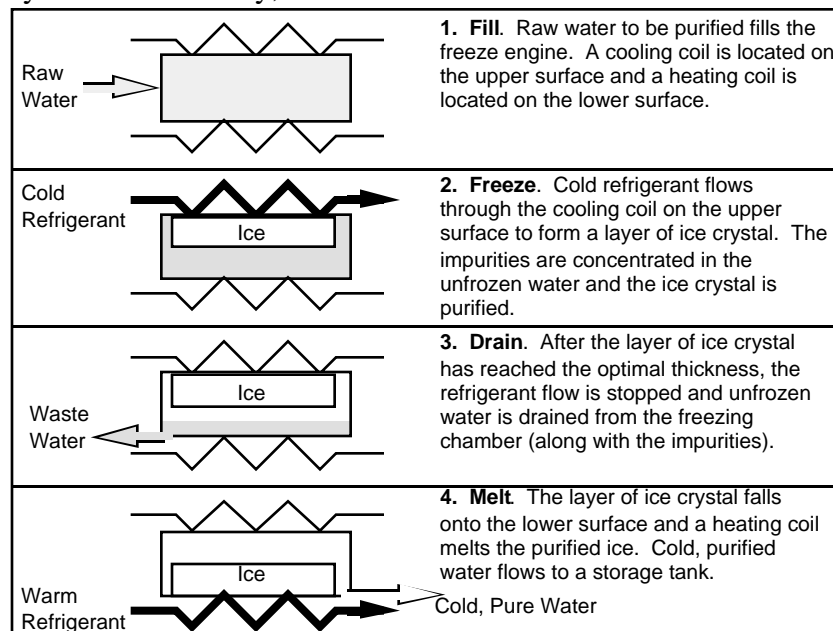

Figure 3.   The batch freeze crystallization process employs a repeating sequence of four steps (Fill, Freeze, Drain, Melt) to separate impurities from water.

Process Characteristics
The batch freeze crystallization process has the following distinguishing characteristics compared to continuous crystallizers.
   • Indirect freezing. Refrigerant is circulated through a freeze plate on which the pure crystals are grown. Also, the process operates at atmospheric pressure, unlike the many vacuum freezing methods used previously.
   • Simplified Ice Transport. The purified crystal is melted in the same chamber in which it is formed, and is transported to the melter by gravity (i.e., the ice sheet falls off the freeze plate). No complex transport mechanisms are needed.
   • Large Crystals. A polycrystalline ice mass with a much smaller surface to volume ratio is formed. This eliminates the need for washing the multitude of tiny crystals found in vacuum methods. Moreover, since any contaminating liquid adheres to only one side of the large crystal, the film could be readily washed off if desired.
   • Quiescent crystallization. Unlike the continuous processes, the crystal forms in a quiet state, thereby promoting high quality crystal growth and minimizing the entrapment of impurities in the crystal lattice.

Separation Performance
We have investigated the separation performance of this batch freeze crystallization process using prototype Freeze Engines. The results are illustrated in Figure 4 for three

major categories of contaminants: General Minerals, Harmful Minerals, and Organic Chemicals. The source water for the general mineral test was obtained by blending two municipal supplies from snow run-off (Hetch-Hetchy Reservoir, Calif.) and groundwater (Los Altos, Calif.). The other source waters were obtained by dissolving contaminants in distilled water. These test results indicate that batch freeze crystallization can be highly effective at separating general minerals, harmful minerals and organic chemicals that are found in tap water.

	Contaminant	Source Concentration	Product Concentration	Rejection
General Minerals	Bicarbonate	120 ppm	6 ppm	95.00%
	Calcium	42 ppm	4 ppm	90.50%
	Chloride	26 ppm	0.95 ppm	96.30%
	Magnesium	15 ppm	2.7 ppm	82.00%
	Sodium	17 ppm	0.65 ppm	96.20%
	Sulfate	13 ppm	0.62 ppm	95.20%
Harmful Minerals	Nitrate	190 ppm	0.66 ppm	99.65%
	Lead	190 ppm	0.88 ppm	99.54%
	Lead	82 ppb	Not Detected	100%
	Lead	55 ppb	Not Detected	100%
	Lead	49 ppb	Not Detected	100%
Organic Chemicals	Chloroform	34.188 ppb	0.094 ppb	99.73%
	BrCl2Methane	8.029 ppb	0.005 ppb	99.94%
	Br2ClMethane	15.418 ppb	0.0025 ppb	99.98%
	Bromoform	11.28 ppb	0.0 ppb	100.00%

Figure 4. Test results indicate that batch crystallization is highly effective at separating general minerals, harmful minerals and organic chemicals from tap water.

Process Implementation
In the simplest configuration, the batch freeze crystallization process can be implemented with three major assemblies:
• Freeze Engine. Water enters the Freeze Engine and the uppermost fraction is frozen. The concentrated liquid is drained away, and then the ice is melted and drained to a storage tank.
• Refrigeration System. A conventional household type refrigeration system including a highly reliable vapor compressor and condenser.
• Microcontroller. Performs sequencing and optimization of the Refrigeration System and Freeze Engine.

In a single Freeze Engine configuration, melting of the crystallized water can be achieved by using thermal energy stored during the freezing step, or by electric heating. We estimate that water can be treated for approximately 800 watt-hours per gallon using electric heat for melting (8¢ per gallon with 10¢ per kilowatt-hour electricity). Energy consumption can be reduced by one-third if stored thermal energy is used for melting.

More complex arrangements of multiple Freeze Engines can provide higher capacity (if operated in parallel) or higher separation (if operated in series). Conventional staged separations can also be arranged with the concentrate from one stage feeding back to the input of a previous stage. When multiple Freeze Engines are used, they may be operated in an alternating sequence, as shown schematically in Figure 5. The heat removed during ice formation in one bank of Freeze Engines is used to melt the ice in an opposite bank (13). Calculations indicate that an energy consumption of between 100 and 40 watt-hours per gallon is achievable (1¢ to 0.4¢ per gallon with 10¢ per kilowatt-hour electricity). This range of energy consumption is believed to be practical for production capacities as low as 10 gallons per day.
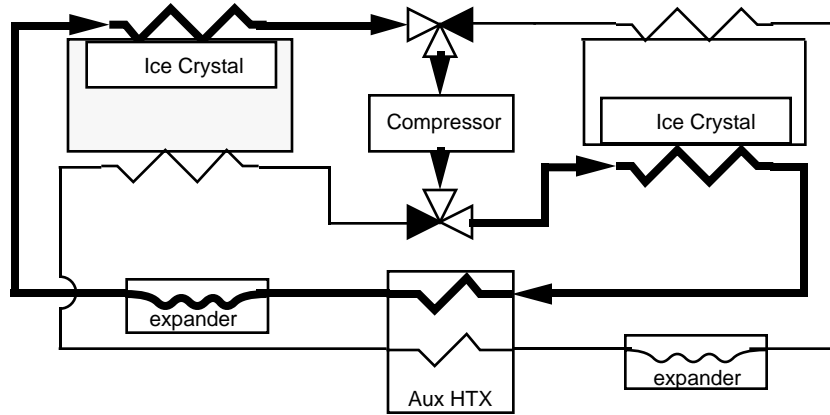
Figure 5. Cold refrigerant flows through the left hand Freeze Engine forming a purified ice crystal. Hot refrigerant is discharged from the compressor to melt the ice crystal in the right hand Freeze Engine. Excess heat is exhausted from the auxiliary heat exchanger.

FREEZE ENGINE OPTIMIZATION

A critical relationship exists between the cycle time, freeze plate surface area and refrigeration system capacity (13). To maximize capacity and minimize equipment size, the Freeze Engine and refrigeration compressor must be optimized. The refrigeration system is coupled to the Freeze Engine via the rate of heat removal through the layer of ice that forms on the Freeze Plate. This relationship is described below.

The heat flow through the ice layer at any instant is proportional to the temperature difference across the ice and the heat transfer area, and is inversely proportional to the thickness of the ice layer, as given by equation (1) below.

$$q = k \, A \, (T_{cold} - T_{water}) \div \Delta x, \tag{1}$$

where,

k is the thermal conductivity of ice, 1.28 Btu/sq.ft.-hr.-°F

A is the area of the Freeze Plate (sq. ft.)

T_{cold} is the temperature of the colder surface (approximately the evaporation temperature)

T_{water} is the temperature at the ice/water interface (32°F)

Δx is the ice thickness.

One can assume that the temperature difference between the evaporating refrigerant and the ice/cold plate interface is negligible (ie., the cold plate is a good heat conductor). For a given Freeze Plate area, Equation (1) can be plotted versus evaporation temperature for various ice thicknesses as shown below in Figure 6. For comparison, the heat removal capacity of a small refrigerant compressor is plotted on the same chart. As the evaporation temperature increases, the compressor capacity also increases. At a given evaporation temperature, a critical ice thickness occurs where the compressor capacity meets the heat transfer rate through the ice. To the left of this intersection, the ice growth rate is limited by the compressor capacity. To the right of this intersection, the ice growth rate is limited by the diffusion of heat through the ice. The maximum capacity is obtained when ice is grown to the critical thickness.

The critical ice thickness occurs when the heat removed by the refrigeration system through the freeze plate balances the heat that can be transferred through the ice layer. At the critical condition, equation (1) becomes
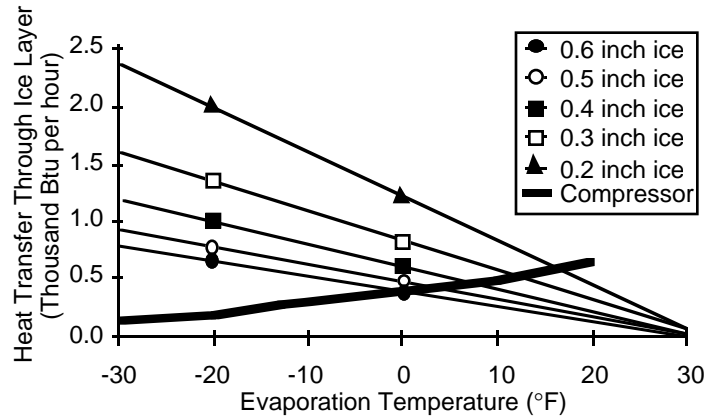
Figure 6. Heat transfer rate through ice layers of various thickness to a 0.75 sq. ft. Freeze Plate. For any evaporation temperature, there is a critical ice thickness at which the compressor capacity equals the heat transfer rate through the ice.

$$q_c = [k \bullet A \bullet \Delta T \div \Delta x]_c \tag{2}$$

The left hand side is the compressor heat removal capacity (which is a function of evaporation temperature), while the right hand side is the heat transfer through an ice layer of thickness Δx and cross-sectional area A. This equation can be re-arranged in dimensionless groups as follows:

$$(\Delta x \div \sqrt{A})_c = [k \bullet \sqrt{A} \bullet \Delta T \div q]_c \tag{3}$$

In cylindrical geometry, the equivalent heat balance is:

$$q_c = [2\pi \bullet k \bullet l \bullet \Delta T \div \ln(r_o \div r_i)]_c \tag{4}$$

where,

l is the length of the cylinder,
r_o is the outside radius of the cylinder,
r_i is the inside radius of the ice.

Re-arranging equation (4) in dimensionless groups gives:

$$(r_o \div r_i)_c = \exp [2\pi \bullet k \bullet l \bullet \Delta T \div q]_c \tag{5}$$

If the ice thickness is again represented by Δx, then $r_i = r_o - \Delta x$, and the optimal dimensionless ice thickness in cylindrical geometry is given by

$$(\Delta x \div r_o)_c = 1 - \exp [- 2\pi \bullet k \bullet l \bullet \Delta T \div q]_c \tag{6}$$

VOLUMETRIC CAPACITY

The volumetric capacity of a freeze crystallization apparatus is equal to the volume of purified water produced during a batch divided by the cycle time required to prepare the batch. The volume of water produced in a batch is

$$Q_{batch} = \Delta x \bullet A \bullet [\rho_s \div \rho_l] \div [\theta_s + \theta_f + \theta_m + \theta_o] \tag{7}$$

where,

Δx is the ice thickness described above,
A is the area of the freeze plate,

ρ_s is the density of the solid ice, and

ρ_l is the density of the liquid water

The cycle time is composed of the following components:

θ_s the time required to remove sensible heat from the water;

θ_f the time required to freeze the ice;

θ_m the time required to melt the ice;

θ_o the time required for overhead, eg., filling and draining the apparatus.

If one assumes that the entire batch of water must be cooled from the incoming feedwater temperature to the freezing point, then the sensible cooling, q_s is,

$$q_s = \rho_l \bullet A \bullet D \bullet \Delta T_{fw} \bullet c_p \div q \tag{8}$$

where,

D   is the depth of the freeze tray;

ΔT_{fw} is the difference between the feedwater temperature and the freezing,temperature;

c_p is the specific heat of the water to be purified;

q   is the compressor heat removal capacity.

The time required to freeze the purified water is

$$\theta_f = \rho_s \bullet A \bullet \Delta x \bullet h_{fs} \div q \tag{9}$$

where h_{fs} is the latent heat of fusion of the ice. The time to melt the purified ice is:

$$\theta_m = \rho_s \bullet A \bullet \Delta x \bullet h_{fs} \div q_{melt} \tag{10}$$

where,

q_{melt} is the heat addition capacity of the melting device.

The overhead time, θ_o, is composed of the time to fill the apparatus, the time to drain the impure water from the freeze tray, and the time to drain the melted ice from the Freeze Engine. These times should be proportional to the volume of fluid to be filled or drained, and to the pressure difference between the source of the water and the tray (for filling) or between the tray and the drain location. For a horizontal Freeze Engine, the drainage flow rate may be slow, and the overhead time may be as much as 10% of the overall cycle time. The overhead time must be empirically determined.

Thus the volumetric capacity is:

$$Q = \Delta x \bullet A \bullet [\, \rho_s \div \rho_l \,] \div \{\, [\rho_l \bullet A \bullet D \bullet \Delta T_{fw} \bullet c_p \div q\,] +$$
$$[\, \rho_s \bullet A \bullet \Delta x \bullet h_{fs} \div q\,] + [\, \rho_s \bullet A \bullet \Delta x \bullet h_{fs} \div q_{melt}\,] + \theta_o\,\} \tag{11}$$

If the overhead time is neglected, equation (11) can be re-arranged in dimensionless groups as follows:

$$Q \bullet \rho_f \bullet h_{fs} \div q = \{\, [\, D \bullet \rho_l \bullet \Delta T_{fw} \bullet c_p \div (\Delta x \bullet \rho_s \bullet h_{fs}\,)\,] + 1$$
$$+ \qquad\qquad\qquad\qquad\qquad\qquad [\qquad\qquad\qquad q \qquad\qquad\qquad \div$$

$$q_{melt} \,] \, \}^{-1} \tag{12}$$

This expression can be simplified by defining the Conversion Ratio, R, that is the ratio of purified water produced to feedwater required, as follows:

$$R = (\rho_l \div \rho_s) \bullet (D \div \Delta x) \tag{13}$$

Equation (12) is then simplified to:

$$Q \bullet \rho_l \bullet h_{fs} \div q = \{ \, [\, R \bullet \Delta T_{fw} \bullet c_p \div h_{fs} \,] + 1 + [\, q \div q_{melt} \,] \, \}^{-1}$$

The first term on the right hand side shows the influence of feedwater temperature and recovery ratio on the volumetric capacity, and the last term shows the influence of melter capacity. In the case of cylindrical geometry, the same equation above can be applied by defining the Conversion Ratio for cylindrical geometry to be:

$$R = (\rho_f \div \rho_s) \bullet [\, 1 - (r_i \div r_o)^2 \,], \text{ or } (\rho_l \div \rho_s) \bullet \{ \, 1 - [1 - (\Delta x \div r_o)^2 \,] \, \} \tag{14}$$

PERFORMANCE PROJECTIONS

With the above groundwork in place, it is possible to estimate the performance for a variety of Freeze Engines and product configurations. Figure 7 shows the estimated water production of a Freeze Engine as a function of the Freeze Time, θ_f. For a given evaporation temperature, as the Freeze Time increases, the water production increases to a maximum. Further operation of the refrigeration system is counterproductive since the insulating ice layer limits heat removal to less than the compressor capacity.
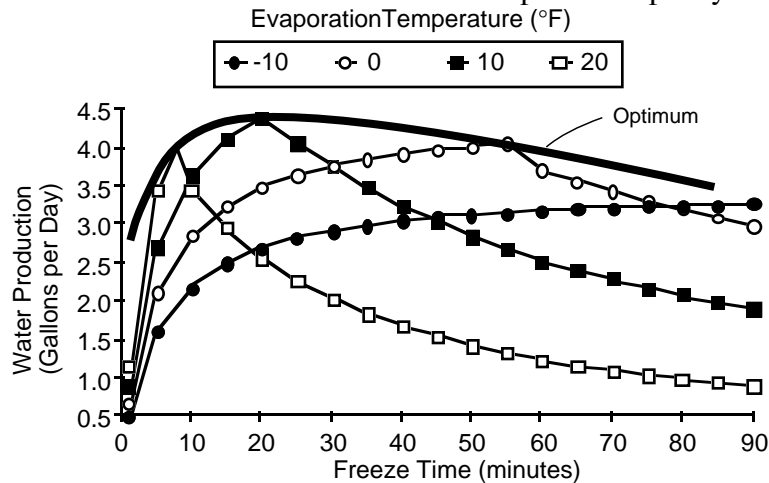

Figure 7.   Predicted water production for a Freeze Engine. The maximum production is obtained when ice is grown to the critical thickness.

This figure was prepared for a small appliance employing the compressor of Figure 6 and a 0.75 sq.ft. Freeze Plate. A Conversion Ratio of 50% was also assumed. Prototype Freeze Engines fitting these guidelines have been developed and fit within a 12 inch square form factor, including insulation. Water production in this form factor depends on the choice of compressor, but would typically be three to four gallons per day per Freeze Engine.

APPLICATIONS

Several potential applications of batch crystallization employing one or more Freeze Engines are illustrated in Figure 8. On a kitchen countertop, a small electric appliance might contain a single Freeze Engine and a gravity holding tank to dispense chilled water. Raw water could be provided by a diverter valve on the faucet and wastewater might drain into the sink.
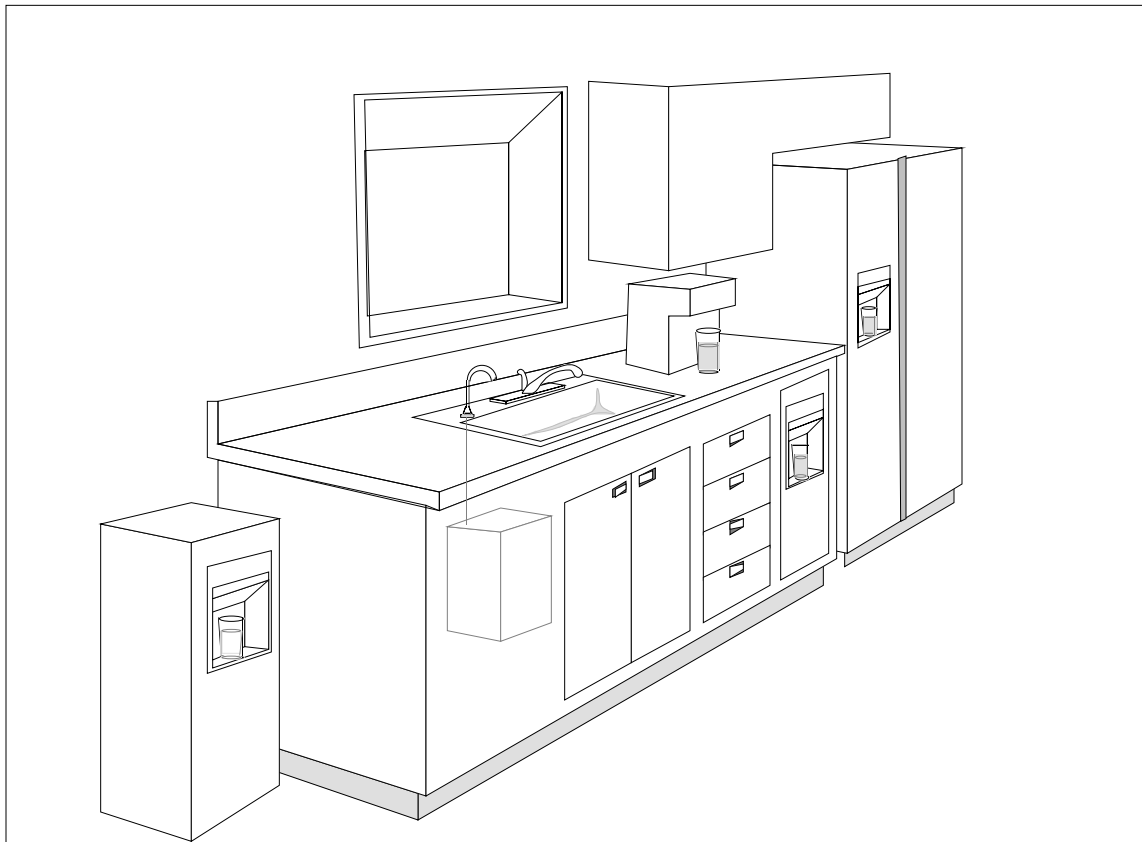

Figure 8. Freeze Engines could be integrated into existing appliances (refrigerators, ice-makers, bottle coolers) and could be designed into new appliance concepts (countertop, built-in, under-sink).

Two Freeze Engines and a three gallon holding tank could be placed within the envelope of an existing bottled water cooler. Water would be dispensed by gravity, just as in existing bottle coolers, and elimination of the bottle allows a coffee maker to be placed on top. The need for plumbing to supply raw water and dispose of wastewater might complicate installation in offices without plumbing available.

A built-in water appliance might use several Freeze Engines, and dispense water through a front panel. This appliance might also be integrated with an ice-maker.

Undersink products would have ready access to plumbing connections, and a repressurization pump would be included to make water available at the faucet. To minimize the ventilation requirements some additional water might be used for cooling.

Refrigerators already include many of the components needed to implement batch crystallization. Freeze Engines might be added to refrigerators that already have raw water connections for water dispensers and ice-makers. A pump would be added to the refrigerator to transfer wastewater through a return line back to the drain.

CONCLUSION

Freeze crystallization has been used as an industrial separation technology and has been applied in the past for seawater desalination. Prior approaches have used continuous crystallizers, but these are large, expensive and complex. Batch crystallization is simpler and can be implemented cost-effectively on a small scale. A "Freeze Engine" and a sequential process have been developed to treat drinking water, with the following potential advantages:

- freeze crystallization can treat and separate a wide variety of water contaminants;
- freeze crystallization does not use expendable filters or membranes;
- batch crystallization can be implemented using reliable household type refrigeration systems;
- chilled water is available as a by-product of the treatment process.

The batch crystallization process optimizes refrigerant evaporation temperature, heat pumping capacity and heat transfer area to maximize throughput. Performance projections suggest that a Freeze Engine could produce three to four gallons per day in a one square foot form factor. Possible applications for batch crystallization technology in drinking water appliances include countertop and undersink units, built-ins, ice-makers and refrigerators, and bottle-less water coolers.

REFERENCES AND NOTES

1. W. M. Conlon, C. N. Chang and D. M. Hendricks. Purified Water Dispensers for Refrigerators, 40th International Appliance Technical Conference. (May 1989).
2. S.Taylor. Ice—Water partition coefficients for RDX and TNT. USA Cold Regions Research and Engineering Laboratory, CRREL Report 89-8. ADA 209 248. (1989).
3. G.F.N. Cox and W.F. Weeks. Brine drainage and initial salt entrapment in sodium chloride ice. USA Cold Regions Research and Engineering Laboratory, Research Report 345. ADA 021 765. (1975).
4. C. J. Martel. Development and design of sludge freezing beds. USA Cold Regions Research and Engineering Laboratory, CRREL Report 88-20. ADA 205 955. (1988).
5. W.T. Sedgwick. Principles of Sanitary Science and the Public Health. New York: MacMillan, p. 251-262. (1903).
6. A. E. Corte. (1962) Vertical migration of particles in front of a moving freezing plane. Journal of Geophysical Research, 67(3): 1085-1090. (1962).
7. D.R. Uhlmann, B. Chalmers and K.A. Jackson. Interaction between particles and a solid/liquid interface. Journal of Applied Physics, 35(10):2986-2992. (1964).
8. V.H.S. Kuo and W. R. Wilcox. Removal of particles by solidification. Industrial and Engineering Chemistry Process Design and Development. 12(3): 376-379. (1973).
9. J.A. Heist. Freeze Crystallization, Chemical Engineering, May 7, 1979: 348 - 358. (1979).
10. K. S. Speigler, Saline Water Conversion Research in Israel, Advances in Chemistry Series 38; (American Chemical Society, Washington, D. C. 1963).
11. C. G. Moyers, and R. W. Rousseau. Crystallization Operations. Handbook of Separation Process Technology, R. W. Rousseau, ed. Wiley-Interscience. 578-643. (1987).
12. The batch freeze crystallization process is known commercially as the Crystallix™ process, a trademark of Polar Spring Corporation.
13. C. N. Chang, Dual Freezing Chamber System and Method for Water Purification. U. S. Patent Number 4,799,945, (1989).
14. C. N. Chang, W. M. Conlon and D. M. Hendricks, Method and Means for Optimizing Batch Crystallization for Purifying Water. U.S. Patent Number 4,954,151, (1990).