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### B. SYMBOLS

$A$	area, $m^2$	$\mathcal{P}$	shape coefficient in Plank's equation
$c$	specific heat, $J/kg\ K$	$q$	heat flux, $J/m^2s$
$d$	diameter	$r$	radius, $m$
$h_{ts}$	latent heat of fusion (melting or freezing), $J/kg\ K$	$R$	radial position of the freeze-front, $m$
$h$	enthalpy, $h$	$\mathcal{R}$	shape coefficient in Plank's equation
$\bar{h}$	convective heat transfer coefficient, $W/m^2s$	$t$	time, $s$
$k$	thermal conductivity, $W/m\ K$	$T$	temperature, $K$ or $^{\circ}C$
$L$	characteristic length	$u$	velocity of the freeze-front due to density change, $m/s$
$m$	the time-variation constant of the ambient temperature $T_a$ , $K/s$	$v$	specific volume, $m^3/kg$
$N_{Bi}$	Biot number, $\equiv \bar{h}L/k$	$V$	volume of the body, $m^3$
$N_{Fo}$	Fourier number, $\alpha/L^2$	$x$	coordinate
$N_{Ste}$	Stefan number, $\equiv c(T_0 - T_f)/h_{ts}$	$X$	position of the freezing front along the x-direction, $m$
$N_t$	dimensionless time parameter	$y$	coordinate
$P$	pressure, $Pa$	$z$	coordinate

**Greek Symbols**

$\alpha$	thermal diffusivity, $m^2/s$
$\beta$	thermal "boundary layer" growth constant
$\delta$	thermal "boundary layer"
$\Delta v_{\ell s}$	(specific volume of the solid phase) - (specific volume of the liquid phase), $m^3/kg$
$\kappa$	$\equiv \sqrt{\frac{\alpha_s}{\alpha_\ell}}$
$\lambda$	freezing rate parameter, dimensionless
$\mu$	$\equiv \rho_s/\rho_\ell$
$\xi$	dimensionless position of the freezing front, $\equiv X/L$
$\rho$	density, $kg/m^3$
$\theta$	dimensionless temperature, Eq. (8-51)
$\chi$	concentration, $kg/kg$ ; or dimensionless distance $x/L$
$\omega$	shape factor [Eq. (8-38)]

**Subscripts**

0	at the surface ( $x=0$ )
a	ambient (fluid surrounding the freezing object)
c	coolant
f	fusion (melting or freezing)
i	inner; or initial (at $t=0$ )
$\ell$	liquid
$\ell s$	phase-change from liquid to solid
o	outer
s	solid
w	at wall

**C. REFERENCES**

There has always been an intense interest in predicting freezing phenomena as related to such applications as food preservation, climate and its control, navigation, and materials processing. This interest expanded with time into new areas such as power generation and medicine. Increasingly rigorous quantitative predictions started in the 19th century, and the number of published papers is now in the tens of thousands. The main books and reviews on the topic, representative key general papers, as well as some of the references on appropriate thermophysical and transport properties, are listed in references [1]-[273] below. A further rather extensive, yet not complete, list of references is given in Subsection V, "Applications Bibliography" below under the specific topics in which freezing plays an important role. In addition to the identification of past work on specific topics, this extensive list of references also helps identify various applications in which freezing plays a role, and the journals that typically cover the field. While encompassing sources from many countries, practically all of the references listed here were selected from the archival refereed literature published in English. Many pertinent publications on this topic also exist in other languages.

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**D. INTRODUCTION, APPLICATIONS, AND BASIC PHYSICAL CONCEPTS**

Freezing occurs naturally, such as with environmental ice in the atmosphere (hail, icing on aircraft), on water bodies and ground regions at the earth's surface, and in the magma at the envelope of the molten earth core. It is also a part of many technological processes, such as preservation of foodstuffs, refrigeration and air-conditioning, snow and ice making, manufacturing (such as casting, molding, sintering, combustion synthesis, coating and electro-deposition, soldering, welding, high energy beam cutting and forming, crystal growth, electro-discharge machining, printing), organ preservation and cryosurgery, and thermal energy storage using solid/liquid phase-changing materials. A bibliography for these applications, with a brief introduction, is given in Subsection V, "Applications Bibliography." Freezing is often accompanied by melting, and the thermodynamics as well as transport principles—and thus the mathematical treatment—of the two processes are very similar. Specific discussion of melting is given in Section 507.9 of this *Databook*.

In simple thermodynamic systems (i.e., without external fields, surface tension, etc.) of a pure material, freezing occurs at certain combinations of temperature and pressure. Since pressure typically has a relatively smaller influence, only the freezing temperature is often used to identify this phase transition.

It is noteworthy that many liquids can be cooled to temperatures significantly below the freezing temperature without solidification taking place. This phenomenon is known as supercooling. Water, for example, is known to reach temperatures of about -40 C without freezing, and silicates and polymers can sustain supercooling levels of hundreds of degrees. To initiate freezing, it is necessary to form or introduce a solid-phase nucleus into the liquid. Once this nucleus is introduced, freezing proceeds rapidly. Further information can be found in Alexiades and Solomon [1] - [3], Knight [40], Perepezko and Utomark [53], and Pounder [55].

The conditions for freezing are strongly dependent on the concentration when the material contains more than a single species. Furthermore, freezing is also influenced by external effects, such as electric and magnetic fields, in more complex thermodynamic systems.

The equilibrium thermodynamic system parameters during phase transition can be calculated from the knowledge that the partial molar Gibbs free energies (chemical potentials) of each component in the two phases must be equal (cf. Alexiades and Solomon [3], Hultgren *et al* [35], Kurz and Fisher [41], Lior [45], and Poulikakos [54]). One important result of using this principle for simple single-component systems is the Clapeyron equation relating the temperature

(T) and pressure (P) during the transition from the liquid to the solid phase, viz

$$\frac{dP}{dT} = \frac{h_{\ell s}}{T\Delta v_{\ell s}} \quad \text{Eq. (8-1)}$$

where  $h_{\ell s}$  is the enthalpy difference between phases ( $= h_s - h_\ell < 0$ , the latent heat of freezing), and  $\Delta v_{\ell s}$  is the specific volume difference between the phases ( $= v_s - v_\ell$ ). Examination of Eq. (8-1) shows that increasing the pressure will result in an increase of the freezing temperature if  $\Delta v_{\ell s} < 0$  (i.e., when the specific volume of the liquid is higher than that of the solid, which is a property of tin, for example), but will result in a decrease of the freezing temperature when  $\Delta v_{\ell s} > 0$  (for water, for example). The latter case explains why ice may melt under the pressure of a skate blade.

In some materials, called glassy, the phase change between the liquid and solid occurs with a gradual transition of the physical properties, from those of one phase to those of the other. When the liquid phase flows during the process, the flow is strongly affected because the viscosity increases greatly as the liquid changes to solid. Other materials, such as pure metals and ice, and eutectic alloys, have a definite line of demarcation between the liquid and the solid, and the transition is abrupt. This situation is easier to analyze and is therefore more rigorously addressed in the literature.

To illustrate the above-described gradual transition, most distinctly observed in mixtures, consider the equilibrium phase diagram for a binary mixture (or alloy) composed of species *a* and *b*, shown in Fig. 8-1. Phase diagrams, or equations describing them, become increasingly complicated as the number of components increases).  $\chi$  is the concentration of species *b* in the mixture,  $\ell$  denotes the liquid, *s* the solid,  $s_a$  a solid with a lattice structure of species *a* in its solid phase but containing some molecules of species *b* in that lattice, and  $s_b$  a solid with a lattice structure of species *b* in its solid phase but containing some molecules of species *a* in that lattice. "Liquidus" denotes the boundary above which the mixture is just liquid, and "solidus" is the boundary separating the final solid mixture of species *a* and *b* from the solid-liquid mixture zones and from the other zones of solid  $s_a$  and solid  $s_b$ .

For illustration, assume that a liquid mixture is at point 1, characterized by concentration  $\chi_1$  and temperature  $T_1$  (Fig. 8-1), and is cooled (descending along the dashed line) while maintaining the concentration constant. When the temperature drops below the liquidus line, solidification starts, creating a mixture of liquid and of solid  $s_a$ . Such a two-phase mixture is called the **mushy zone**. At point 2 in that mushy zone; for example, the solid phase ( $s_a$ ) portion contains a concentration  $\chi_{2,s_a}$  of component *b*, and the liquid phase portion contains a concentration  $\chi_{2,\ell}$  of component *b*. The ratio of the mass of the solid  $s_a$  to that of the liquid is determined by the lever rule, and is

$(\chi_{2,\ell} - \chi_2)/(\chi_2 - \chi_{2,s_a})$  at point 2. Further cooling to below the solidus line, say to point 3, results in a solid mixture (or alloy) of  $s_a$  and  $s_b$ , containing concentrations  $\chi_{3,s_a}$  and  $\chi_{3,s_b}$  of species  $b$ , respectively. The ratio of the mass of the solid  $s_a$  to that of  $s_b$  is again determined by the lever rule, and is  $(\chi_{3,s_b} - \chi_3)/(\chi_3 - \chi_{3,s_a})$  at point 3.

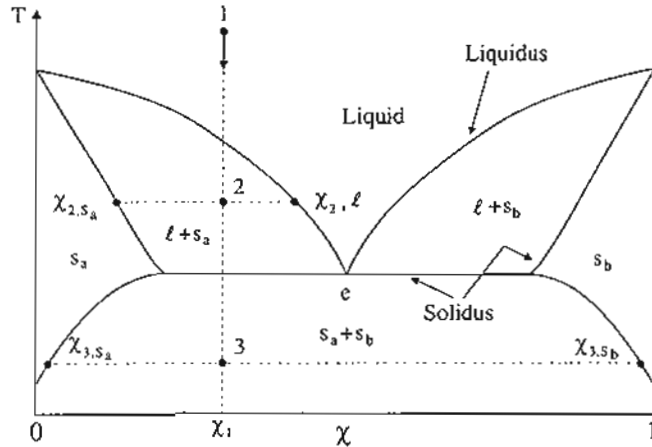


Figure 8-1. A Liquid-Solid Phase Diagram of a Binary Mixture

A unique situation occurs if the initial concentration of the liquid is  $\chi_e$ : upon constant concentration cooling, the liquid forms the solid mixture  $s_a + s_b$  having the same concentration  $\chi_e$  and without the formation of a two-phase zone.  $\chi_e$  is called the *eutectic concentration*, and the resulting solid mixture (or alloy) is called a *eutectic*.

It is obvious from the above that the concentration distribution changes among the phases, which accompany the freezing process (Fig. 8-1), are an important factor in the composition of alloys, and are the basis for freeze-separation processes.

The presence of a two-phase mixture zone with temperature-dependent concentration and phase-proportion obviously complicates heat transfer analysis, and requires the simultaneous solution of both the heat and mass transfer equations. Furthermore, the liquid usually does not solidify on a simple planar surface between the phases. Crystals of the solid phase are formed at some preferred locations in the liquid, or on colder solid surfaces immersed in the liquid, and as freezing progresses the crystals grow in the form of intricately-shaped fingers, called dendrites. This complicates the geometry significantly and makes mathematical modeling of the process very difficult. An introduction to these phenomena and further references are available in Chalmers [13], Colucci-Mizenko *et al* [20], Flemings [27], Kurz and Fisher [41], Murray *et al* [49], Poulidakos, [54], Trivedi and Kurz [65], Gilpin [98]-[100], Prescott *et al* [124] and Glicksman *et al* and Hayashi and Kunimine (both cited in Subsection V. B.).

Flow of the liquid phase often has an important role in the

inception of—and during—melting and freezing (cf. Cheung and Epstein [17], Yao and Prusa [72] and references [79]-[152]). The flow may be forced, such as in the freezing of a liquid flowing through or across a cooled pipe, and/or may be due to natural convection that arises whenever there are density gradients in the liquid, here generated by temperature and possibly concentration gradients. It is noteworthy that the change in phase usually affects the original flow, such as when the liquid flowing in a cooled pipe gradually freezes, and the frozen solid thus reduces the flow passage, or when the evolving dendritic structure gradually changes the geometry of the solid surfaces that are in contact with the liquid (cf. Cheng and Wong [85], Cho and Özisik [88], Epstein and Cheung [91, 92], Epstein and Hauzer [93], Gilpin [101,102], Hirata and Hanaoka [106], Hirata and Ishihara [107], Hwang and Tsai [107], Kikuchi *et al* [112], Kuzay and Epstein [115], Lee and Hwang [116], Madejski [119], Sampson and Gibson [129, 130], Seki *et al* [135], Thomason *et al* [144], Weigand *et al* [147], Zerkle and Sunderland [152]). Under such circumstances, strong coupling may exist between the heat transfer and fluid mechanics, and also with mass transfer when more than a single species is present, and the process must be modeled by an appropriate set of continuity, momentum, energy, mass conservation, and state equations, which need to be solved simultaneously.

## E. PREDICTIVE METHODS

The mathematical description of the freezing process is characterized by non-linear partial differential equations, which have analytical (closed-form) solutions for only a few simplified cases. As explained above, the problem becomes even less tractable when flow accompanies the process, or when more than a single species is present. A very large amount of work has been done in developing solution methods for the freezing problem (sometimes also called the Stefan problem, after the seminal paper by Stefan [62]), published both as monographs and papers, and included in the list of references to this Section (Alexiades and Solomon [3], Bankoff [6], Chadam and Rasmussen [12], Cheng and Seki [16], Crank [22], Fasano and Primicerio [26], Hill [33], Özisik [51], Tanasawa and Lior [64], Yao and Prusa [72], and references [152] - [273], with emphasis on the reviews by Fox [175], Friedman [178], Fukusako and Seki [179], Meirmanov [209], Ockendon and Hodgkins [219], Rubinshtein [233], and Wilson *et al* [266]). Generically, solutions are obtained either by 1) linearizing the original equations (e.g., perturbation methods) where appropriate, and solving these linear equations, or 2) simplifying the original equations by neglecting terms, such as the neglect of thermal capacity in the "quasi-static method" described in Subsection IV below, or 3) using the "integral method," which satisfies energy conservation over the entire body of interest, as well as the boundary condi-

tions, but is only approximately correct locally inside the body (Subsection IV. A. below), or 4) employing a numerical method.

Many numerical methods have been successfully employed in the solution of freezing problems, both of the finite difference and element types, and many well-tested software programs exist that include solutions for that purpose. A significant difficulty in the formulation of the numerical methods is the fact that the liquid-solid interface moves and perhaps changes shape as freezing progresses (making this a "moving boundary" or "free boundary" problem). This requires continuous monitoring of the interface position during the solution sequence, and adjustment of the numerical model cell or element properties to those of the particular phase present in them at the time-step being considered (cf. Alexiades and Solomon [3], Crank [22], Dilley and Lior [171], Fasano and Primicerio [26], Fox [175], Friedman [178], Furzeland [180], Gupta and Kumar [185], Hsu *et al* [189], Hyman [192], Kim *et al* [197], Meyer [212, 213], Mori and Araki [218], Ockendon and Hodgkiss [219], Rubinsky and Shitzer [234], Saunders [239], Siegel *et al* [245], Tsai and Rubinsky [255], Udaykumar and Shyy [256], Vick and Nelson [258], Wilson *et al* [266], Yoo and Rubinsky [268], Yu *et al* [269], Zabaras and Mukherjee [270], Zhang *et al* [273]). Several formulations of the original equations were developed to simplify their numerical solution. One of them is the popular "enthalpy method" discussed in more detail in Subsection IV.B. below.

The predictive equations provided below are all for materials whose behavior can be characterized as being pure. This would also apply to multi-component material where changes of the freezing temperature and of the composition during the freezing process can be ignored. General solutions for cases where these can not be ignored are much more difficult to obtain, and the readers are referred to the literature; some of the key citations are provided in references [3], [27], [41], [77], [265] and Subsection V.B.

Furthermore, the solutions presented here by closed-form equations are only for simple geometries, since no such solutions are available for complex geometries. Simplified expressions, however, are presented for freezing times also in arbitrary geometries.

## II. PREDICTIVE EQUATIONS FOR FREEZING WITHOUT FLOW

### A. ONE-DIMENSIONAL FREEZING WITHOUT DENSITY CHANGE

Examination of the simplified one-dimensional case provides some important insights into the phenomena, identifies the key parameters, and allows analytical solutions and thus qualitative predictive capability for at least

this class of problems. In this Section we deal with cases in which the densities of both phases are the same, and in which the freezing liquid does not flow, thus also ignoring, for simplification, the effects of buoyancy-driven convection that accompanies the freezing process when a temperature gradient exists in the liquid phase. As stated in Subsection I.E., the effects of natural convection may sometimes be significant, and information about this topic can be found in the references quoted in that Section. Freezing of non-opaque media may also include internal radiative heat transfer, which is ignored in the equations presented below. Information about such problems is contained in references [14] and [31].

The solutions presented below can be found in many books and reviews that deal with melting and freezing (cf. refs. [3], [6], [22], [33], [41], [45], [72], and [179], [209], [233]) and in textbooks dealing with heat conduction (cf. [51] and [54]).

#### 1. Solutions for Materials that are Initially at the Freezing Temperature

If the liquid to be frozen is initially at the freezing temperature throughout its extent, as shown in Fig. 8-2, heat transfer occurs in the solid phase only. This somewhat simplifies the solution and is presented first.

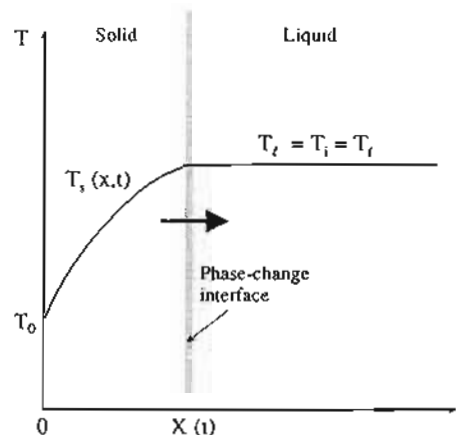


Figure 8-2. Freezing of a Semi-infinite Liquid Initially at the Fusion Temperature. Heat conduction takes place consequently in one of the phases only.

Consider a liquid of infinite extent is to the right ( $x > 0$ ) of the infinite surface at  $x = 0$  (i.e., semi-infinite), described in Fig. 8-2, initially at the freezing temperature  $T_f$ . For time  $t > 0$  the temperature of the surface (at  $x = 0$ ) is lowered to  $T_0 < T_f$ , and the solid consequently starts to freeze there. In this case the temperature in the liquid remains constant,  $T_s = T_f$ , so the temperature distribution needs to be calculated only in the solid phase. It is assumed that the liquid remains motionless and in place. The initial condition in the solid is

$$T_s(x, t) = T_f \text{ in } x > 0, \text{ at } t = 0, \quad \text{Eq. (8-2)}$$

the boundary condition is

$$T_s(0, t) = T_0 \text{ for } t > 0, \quad \text{Eq. (8-3)}$$

and the liquid-solid interfacial temperature and heat flux continuity conditions, respectively, are

$$T_s[X(t)] = T_f \text{ for } t > 0, \quad \text{Eq. (8-4)}$$

$$-k_s \left( \frac{\partial T_s}{\partial x} \right)_{[X(t)]} = \rho_s h_{fs} \frac{dX(t)}{dt} \text{ for } t > 0, \quad \text{Eq. (8-5)}$$

The analytical solution of this problem yields the temperature distribution in the solid as

$$T_s(x, t) = T_0 + (T_f - T_0) \frac{\operatorname{erf}\left(\frac{x}{2\sqrt{\alpha_s t}}\right)}{\operatorname{erf} \lambda'} \text{ for } t > 0, \quad \text{Eq. (8-6)}$$

where erf stands for the *error function* (described and tabulated in mathematics handbooks), and  $\lambda'$  is the solution of the equation

$$\lambda' e^{\lambda'^2} \operatorname{erf}(\lambda') = \frac{N_{Ste_s}}{\sqrt{\pi}}, \quad \text{Eq. (8-7)}$$

with

$N_{Ste_s}$  being the Stefan Number, here defined for the solid as

$$N_{Ste_s} \equiv \frac{c_s(T_0 - T_f)}{h_{fs}}, \quad \text{Eq. (8-8)}$$

recalling that the latent heat of freezing,  $h_{fs}$ , must be entered into the equations as a negative value. Equation (8-7) can be solved to find the value of  $\lambda'$  for the magnitude of  $N_{Ste_s}$ , which is calculated for the problem at hand by using Eq. (8-8). The solution of Eq. (8-7), yielding the values of  $\lambda'$  as a function of  $N_{Ste_s}$  for  $0 \leq N_{Ste_s} \leq 5$ , is given in Fig. 8-3.

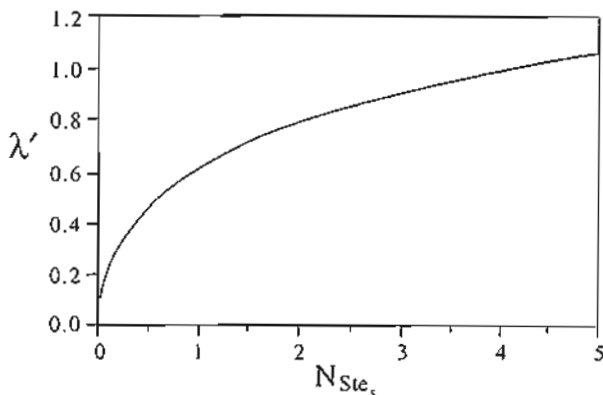


Figure 8-3. The Root  $\lambda'$  of Eq. (8-7)

The interface position (which also is the freezing front progress) is defined by Eq. (8-9)

$$X(t) = 2\lambda'(\alpha_s t)^{1/2}. \quad \text{Eq. (8-9)}$$

### EXAMPLE

The temperature of the vertical surface of a large volume of liquid paraffin used for heat storage, initially at the freezing temperature,  $T_i = T_f = 60^\circ\text{C}$ , is suddenly lowered to  $30^\circ\text{C}$ . Any motion in the melt may be neglected. How long would it take for the paraffin to solidify to a depth of 0.1 m? Given properties:  $\alpha_s = (1.09)10^{-7} \text{ m}^2/\text{s}$ ,  $\rho_s = \rho_l = 814 \text{ kg/m}^3$ ,  $h_{fs} = -241 \text{ kJ/kg}$ ,  $c_s = 2.14 \text{ kJ/kg }^\circ\text{C}$ . To find the required time we use Eq. (8-9), in which the value of  $\lambda'$  needs to be determined.  $\lambda'$  is calculated from Eq. (8-7) or determined from Fig. 8-3, which requires knowledge of  $N_{Ste_s}$ . From Eq. (8-8)

$$N_{Ste_s} = \frac{(2.14 \text{ kJ/kg }^\circ\text{C})(30^\circ\text{C} - 60^\circ\text{C})}{-241.2 \text{ kJ/kg}} = 0.266.$$

The solution of Eq. (8-7) as a function of  $N_{Ste_s}$  is given in Fig. 8-3, yielding  $\lambda' \approx 0.4$ . Using Eq. (8-9), the time of interest is calculated by

$$t = \frac{[X(t)]^2}{4\lambda'^2 \alpha_s} = \frac{(0.1 \text{ m})^2}{4(0.4)^2 [(1.09)10^{-7} \text{ m}^2/\text{s}]} = (1.43)10^5 \text{ s} = 39.8 \text{ h}.$$

### 2. Solutions for Materials that are Initially not at the Freezing Temperature

If, initially, the liquid to be frozen is above the freezing temperature, conductive heat transfer takes place in both phases. Consider a semi-infinite liquid initially at a temperature  $T_i$  higher than the freezing temperature  $T_f$  (Fig. 8-4). At time  $t = 0$  at the liquid surface temperature at  $x = 0$  is suddenly lowered to a temperature  $T_0 < T_f$ , and maintained at that temperature for  $t > 0$ . Consequently, the liquid starts to freeze at  $x = 0$ , and the freezing interface (separating in Fig. 8-4 the solid to its left from the liquid on its right) located at the position  $x = X(t)$  moves gradually to the right (in the positive  $x$  direction).

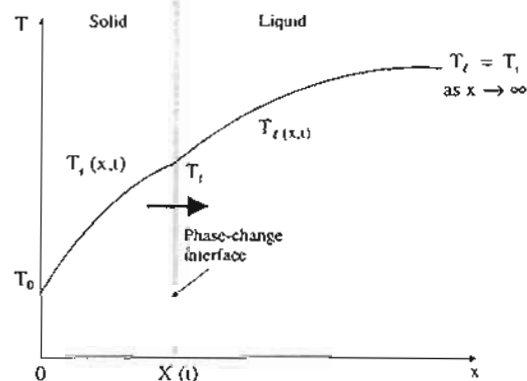


Figure 8-4. Freezing of a Semi-infinite Liquid Initially at an Above-freezing Temperature. Heat conduction takes place in both phases.

The analytical solution of this problem yields the temperature distributions in the liquid and solid phases, respectively, as

$$T_l(x,t) = T_i - (T_i - T_f) \frac{\operatorname{erfc}\left(\frac{x}{2\sqrt{\alpha_l t}}\right)}{\operatorname{erfc}\left(\frac{\lambda\sqrt{\alpha_s}}{\alpha_l}\right)}, \quad \text{Eq. (8-10)}$$

and

$$T_s(x,t) = T_0 + (T_f - T_0) \frac{\operatorname{erf}\left(\frac{x}{2\sqrt{\alpha_s t}}\right)}{\operatorname{erf}\lambda}, \quad \text{Eq. (8-11)}$$

where  $\operatorname{erfc}$  is the complementary error function;  $\lambda$  is a constant, obtained from the solution of the equation

$$\frac{e^{-\lambda^2}}{\operatorname{erf}\lambda} - \frac{k_l}{k_s} \sqrt{\frac{\alpha_s}{\alpha_l}} \frac{T_i - T_f}{T_f - T_0} \frac{e^{-(\alpha_s/\alpha_l)\lambda^2}}{\operatorname{erfc}(\lambda\sqrt{\alpha_s/\alpha_l})} = \frac{\lambda\sqrt{\pi}}{N_{Ste_s}}, \quad \text{Eq. (8-12)}$$

where  $N_{Ste_s}$  is the Stefan number defined by Eq. (8-8).

Solutions of Eq. (8-12) are available for some specific cases in several of the references (cf. [3] and [51]), and can, in general, be obtained relatively easily by a variety of commonly-used software packages used for the solution of nonlinear algebraic equations.

The transient position of the freezing interface is

$$X(t) = 2\lambda(\alpha_s t)^{1/2}, \quad \text{Eq. (8-13)}$$

where  $\lambda$  is the solution of Eq. (8-12), and thus the expression for the rate of freezing, i.e. the velocity of the motion of the solid-liquid interface, is

$$\frac{dX(t)}{dt} = \lambda\alpha_s^{1/2} t^{-1/2}. \quad \text{Eq. (8-14)}$$

## B. ONE-DIMENSIONAL FREEZING WITH DENSITY CHANGE

For most materials the density of the liquid and solid phases is somewhat different, usually by up to about 10% and in some cases up to 30%. Usually the density of the liquid phase is smaller than that of the solid one, causing volume expansion upon melting and shrinkage upon freezing. This phenomenon causes, for example, a manufacturing problem in that metals and plastic materials filling a mold in their liquid phase shrink when solidified, forming voids in the solid and a poorly-conducting gas layer between the mold (or container) wall. Water is one of the materials in which the density of the liquid phase is higher than that of the solid one, and thus ice floats on water, and pipes tend to burst when water confined in them freezes. If the densities

of the liquid and solid phases differ, motion of the phase-change interface is not only due to the phase change process, but also due to the associated volume (density) change.

A reasonably good analytical solution for small ( $\sim \pm 10\%$ ) solid-liquid density differences is available (Alexiades and Solomon [3]) for the semi-infinite slab at  $x \geq 0$ , initially solid at  $T_i > T_f$ , frozen by imposing a constant temperature  $T_0 < T_f$  at the surface  $x = 0$  (Fig. 8-4). It is assumed that  $\rho_l > \rho_s$ ,  $c_l, c_s, k_l, k_s, h_{ls}$ , and  $T_f$  are constants and positive. The freeze front  $X(t)$  starts at  $x = X(0) = 0$  and advances to the right. Buoyancy-driven convection is ignored, but the volume expansion of the solid upon freezing is considered, in that it pushes the entire liquid volume rightward (Fig. 8-4) without friction, at uniform speed  $u(t)$  without motion inside the liquid itself. The temperature distributions are, in the solid and liquid phases

$$T_l(x,t) = T_0 + (T_f - T_0) \frac{\operatorname{erf}\left(\frac{x}{2\sqrt{\alpha_s t}}\right)}{\operatorname{erf}\lambda'''} \quad \text{Eq. (8-15)}$$

in  $0 \leq x \leq X(t)$  for  $t > 0$ ,

$$T_s(x,t) = T_i - (T_i - T_m) \frac{\operatorname{erfc}\left[\frac{x}{2\sqrt{\alpha_l t}} - (1-\mu)\kappa\lambda'''\right]}{\operatorname{erfc}(\mu\kappa\lambda''')} \quad \text{Eq. (8-16)}$$

in  $x \geq X(t)$  for  $t > 0$ .

The location of the freezing front is

$$X(t) = 2\lambda'''(\alpha_s t)^{1/2}, \quad \text{Eq. (8-17)}$$

and the speed of the liquid body motion due to the expansion is

$$v(t) = (1-\mu)\lambda''' \sqrt{\alpha_s/t}. \quad \text{Eq. (8-18)}$$

In Eqs. (8-15) - (8-18)  $\lambda'''$  is the root of the equation

$$\frac{N_{Ste_s}}{\lambda''' e^{\lambda'^{2}} \operatorname{erf}\lambda'''} - \frac{N_{Ste_l}}{(\mu\kappa\lambda''') e^{(\mu\kappa\lambda''')^2} \operatorname{erfc}(\mu\kappa\lambda''')} = \sqrt{\pi}, \quad \text{Eq. (8-19)}$$

which can, for the specific problem parameters, be solved numerically or by using one of the many software packages for solving nonlinear algebraic equations. The remaining parameters are defined as

$$N_{Ste_s} \equiv \frac{c_s(T_0 - T_f)}{h_{ls}}, \quad N_{Ste_l} \equiv \frac{c_l(T_f - T_i)}{h_{ls}}, \quad \text{Eq. (8-20)}$$

$$\kappa \equiv \sqrt{\frac{\alpha_s}{\alpha_l}}, \quad \mu \equiv \frac{\rho_s}{\rho_l}$$

with  $h_{fs}$  taking a negative value in this freezing problem.

Because of the approximate nature of this analytical solution it is expected that Eq. (8-17) slightly overestimates the melt front position. If  $\rho_l < \rho_s$ , freezing would cause the solid to shrink, moving the liquid leftward, in a direction opposite to that of the freezing interface, and the solution represented by Eqs. (8-15) - (8-20) would not be valid. Approximate but less accurate solutions for this and other cases are described by Alexiades and Solomon [3].

### C. THE QUASI-STATIC APPROXIMATION

To obtain rough estimates of melting and freezing processes quickly, in cases where heat transfer takes place in only one phase, it is assumed that effects of sensible heat are negligible relative to those of latent heat ( $Ste \rightarrow 0$ ). This is a significant simplification, since the energy equation then becomes independent of time, and solutions to the steady state heat conduction problem are much easier to obtain. At the same time, the transient phase-change interface condition [such as Eq. (8-5)] is retained, allowing the estimation of the transient interface position and velocity. This is hence a quasi-static approximation, and its use is shown below. The simplification allows solution of freezing problems in more complicated geometries. Some solutions for the cylindrical geometry are presented below. More details can be found in refs. [3], [22], [33], [166], [179], and [248].

It is important to emphasize that these are just approximations, without full information on the effect of specific problem conditions on the magnitude of the error incurred when using them. In fact, in some cases, especially with a convective boundary condition, they may produce very wrong results. It is thus necessary to examine the physical viability of the results, such as overall energy balances, when using these approximations.

It is assumed here that the problems are one-dimensional, and that the material is initially at the freezing temperature  $T_f$ .

#### 1. Examples of the Quasi-Static Approximation for a Slab

Given a semi-infinite liquid on which a *time-dependent temperature*  $T_0(t) < T_f$  is imposed at  $x = 0$ , (Fig. 8-2), the above-described quasi-static approximation yields the solution for the position of the phase-change front and of the temperature distribution in the solid as

$$X(t) = \left[ 2 \frac{k_s}{\rho h_{fs}} \int_0^t [T_0(t) - T_f] dt \right]^{1/2} \text{ for } t \geq 0, \quad \text{Eq. (8-21)}$$

$$T_s(x, t) = T_0(t) + \left[ T_f - T_0(t) \right] \frac{x}{X(t)} \text{ in } 0 \leq x \leq X(t) \text{ for } t \geq 0, \quad \text{Eq. (8-22)}$$

respectively.

The heat flux released during freezing,  $q(x, t)$ , can easily be determined from the temperature distribution in the liquid [Eq. (8-22)], viz.

$$q(x, t) = k_s \frac{T_f - T_0(t)}{X(t)}. \quad \text{Eq. (8-23)}$$

This approximate solution is exact when  $Ste_s \rightarrow 0$ , and it otherwise overestimates the values of both  $X(t)$  and  $T(x, t)$ . While the errors depend on the specific problem, they are confined to about 10% in the above-described case (Alexiades and Solomon [3]).

For the same freezing problem but with the **boundary condition of an imposed time-dependent negative heat flux (cooling)**  $-q_0(t)$ ,

$$-k_s \left( \frac{dT_s}{dx} \right)_{0,t} = q_0(t) \text{ for } t > 0, \quad \text{Eq. (8-24)}$$

the quasi-static approximate solution is

$$X(t) = \frac{1}{\rho h_{fs}} \int_0^t q_0(t) dt \text{ for } t > 0, \quad \text{Eq. (8-25)}$$

$$T_s(x, t) = T_f + \frac{q_0}{k_s} \left( \frac{q_0}{\rho h_{fs}} t - x \right) \text{ in } 0 \leq x \leq X(t) \text{ for } t > 0. \quad \text{Eq. (8-26)}$$

Note that both  $h_{fs}$  and  $q_0$  must be entered into the equations as negative values.

#### 2. Examples of the Quasi-Static Approximation for Cylinder

It is assumed in these examples that the cylinders are very long and that the problems are axisymmetric. Just as in the slab case, the energy equation is reduced by the approximation to its steady state form.

Consider the **outward-directed freezing** of a hollow cylinder of liquid with internal radius  $r_i$  and outer radius  $r_o$  (Fig. 8-5) due to a low temperature imposed at the internal radius  $r_i$ ,

$$T_s(r_i, t) = T_0(t) < T_f \text{ for } t > 0. \quad \text{Eq. (8-27)}$$

The solution is

$$T_s(x, t) = T_f - [T_f - T_0(t)] \frac{\ln[r/R(t)]}{\ln[r_i/R(t)]} \quad \text{Eq. (8-28)}$$

in  $r_i \leq r \leq R(t)$  for  $t > 0$ ,

and the transient position of the freezing front,  $R(t)$ , can be calculated from the transcendental equation

$$2R(t)^2 \ln \frac{R(t)}{r_i} = R(t)^2 - r_i^2 + \frac{4k_s}{\rho h_{fs}} \int_0^t [T_0(t) - T_f] dt. \quad \text{Eq. (8-29)}$$

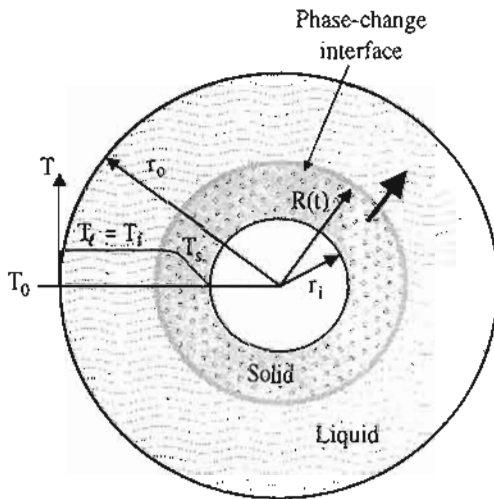


Figure 8-5. Outward Freezing in a Hollow Cylinder Containing Liquid Initially at the Freezing Temperature ( $T_f$ ), Subjected at its Inner Radius ( $r_i$ ) to a Temperature  $T_0 < T_f$

If the freezing for the same case occurs due to the imposition of a negative heat flux  $q_0$  at  $r_i$ ,

$$-k_s \left( \frac{dT_s}{dr} \right)_{r_i, t} = q_0(t) > 0 \quad \text{for } t > 0. \quad \text{Eq. (8-30)}$$

the solution is

$$T_s(r, t) = T_f - \frac{q_0(t)r_i}{k_s} \ln \frac{r}{R(t)} \quad \text{Eq. (8-31)}$$

in  $r_i \leq r \leq R(t)$  for  $t > 0$ ,

$$R(t) = \left[ r_i^2 + 2 \frac{r_i}{\rho h_{fs}} \int_0^t q_0(t) dt \right]^{1/2} \quad \text{for } t > 0, \quad \text{Eq. (8-32)}$$

where  $q_0(t)$  and  $h_{fs}$  must be entered as negative values.

If the freezing for the same case occurs due to the imposition of a convective heat flux to a fluid at the transient

lower temperature  $T_a(t)$ , with a heat transfer coefficient  $\bar{h}$  at  $r_i$ , the heat flux boundary condition there is

$$-k_s \left( \frac{dT_s}{dr} \right)_{r_i, t} = \bar{h} [T_a(t) - T_s(r_i, t)] > 0 \quad \text{for } t > 0, \quad \text{Eq. (8-33)}$$

and the solution is

$$T_s(r, t) = T_f - [T_f - T_a(t)] \frac{\ln[r/R(t)]}{\ln[r_i/R(t)] - k_s/\bar{h}r_i}$$

in  $r_i \leq r \leq R(t)$  for  $t > 0$ , Eq. (8-34)

with  $R(t)$  calculated from the transcendental equation

$$2R(t)^2 \ln \frac{R(t)}{r_i} = \left( 1 - \frac{2k_s}{\bar{h}r_i} \right) [R(t)^2 - r_i^2] - \frac{4k_s}{\rho h_{fs}} \int_0^t [T_f - T_a(t)] dt. \quad \text{Eq. (8-35)}$$

The solutions for inward freezing of a cylinder, where cooling is applied at the outer radius  $r_o$ , are the same as the above-described ones for the outward-freezing cylinder, if the replacements  $r_i \rightarrow r_o$ ,  $q_0 \rightarrow -q_0$ , and  $\bar{h} \rightarrow -\bar{h}$  are made. If such a cylinder is not hollow then  $r_i = 0$  is used.

#### D. ESTIMATION OF FREEZING TIME

There are a number of approximate formulas for estimating the freezing and melting times of different materials having a variety of shapes.

##### 1. Freezing Time of Foodstuff

The American Society of Heating, Refrigerating, and Air-Conditioning Engineers (ASHRAE) provides a number of approximations for estimating the freezing and thawing times of foods (ASHRAE [4]). For example, if it can be assumed that the freezing or thawing occur at a single temperature, the time to freeze or thaw,  $t_f$ , for a body that has shape parameters  $\mathcal{P}$  and  $R$  (described below) and thermal conductivity  $k$ , initially at the fusion temperature  $T_f$ , and which is exchanging heat via heat transfer coefficient  $\bar{h}$  with an ambient at the constant temperature  $T_a$ , can be approximated by Plank's equation

$$t_f = \frac{h_{fs}\mathcal{P}}{[T_f - T_a]} \left( \frac{\mathcal{P}d}{\bar{h}} + \frac{Rd^2}{k} \right). \quad \text{Eq. (8-36)}$$

where  $d$  is the diameter of the body if it is a cylinder or a sphere, or the thickness when it is an infinite slab, and where the shape coefficients  $\mathcal{P}$  and  $R$  for a number of body forms are given in Table 8-1 below.



**Table 8-1.**  
**Shape Factors for Eq. (8-36), ASHRAE [4]**

Forms	$\mathcal{P}$	R
Slab	1/2	1/8
Cylinder	1/4	1/16
Sphere	1/6	1/24

Shape coefficients for other body forms are also available. To use Eq. (8-37) for freezing,  $k$  and  $\rho$  should be the values for the food in its frozen state.

If the initial temperature ( $T_i$ ) of the material to be frozen is higher than  $T_f$  and the surface temperature  $T_0$  is given (or  $N_{Bi_s} \rightarrow \infty$ ), the following simple formula for estimating the freezing time  $t_f$  of an **infinitely-long cylinder** of diameter  $d$  and radius  $r_0$  was proposed:

$$N_{Fo,f} = (0.14 + 0.085 Y_0) + (0.252 - 0.0025 Y_0) h_{ts} d. \quad \text{Eq. (8-37)}$$

where

$$N_{Fo,f} \quad \text{Fourier number, } \equiv \frac{\alpha_s t_f}{r_0^2},$$

$$Y_0 \quad \text{Dimensionless Temperature, } \frac{T_i - T_f}{T_f - T_a}.$$

In fact, freezing or melting of food typically takes place over a range of temperatures, and approximate Plank-type formulas have been developed for various specific food-stuffs and shapes to represent reality more closely than Eq. (8-36) (Cleland *et al* [167], ASHRAE [4]).

**EXAMPLE**

Using Plank's Equation (8-36) for estimating freezing time estimate the time needed to freeze a fish, the shape of which can be approximated by a cylinder 0.5 m long having a diameter of 0.1 m. The fish is initially at its freezing temperature, and during the freezing process it is surrounded by air at  $T_a = -25^\circ\text{C}$ , with the cooling performed with a convective heat transfer coefficient  $\bar{h} = 68 \text{ W/m}^2\text{K}$ . For the fish,  $T_f = -1^\circ\text{C}$ ,  $h_{sl} = 200 \text{ kJ/kg}$ ,  $\rho_s = 992 \text{ kg/m}^3$ , and  $k_s = 1.35 \text{ W/m K}$ .

Using Table 8-1, the geometric coefficients for the cylindrical shape of the fish are  $\mathcal{P} = 1/4$  and  $R = 1/16$ , while  $d$  is the cylinder diameter,  $= 0.1\text{m}$ . Substituting these values into Eq. (8-36) gives

$$t_f = \frac{200000 \cdot 992}{-1 - (-25)} \left( \frac{1/4(0.1)}{68} + \frac{1/16(0.1)^2}{1.35} \right) = 6866 \text{ s} = 1.9\text{h}.$$

**2. Other Approximations for Freezing Time**

Alexiades and Solomon [3] provide an easily-computable approximate equation for estimating the time needed to freeze a simple-shaped liquid volume initially at the freezing temperature  $T_f$ . It is assumed that conduction occurs in one phase (the solid) only, that the problems are axi- and spherically-symmetric for cylindrical and spherical bodies, respectively, and that the freezing process for differently shaped bodies can be characterized by a single geometric parameter,  $r$ , in the body domain  $0 \leq r \leq L$ , using a shape factor,  $\omega$ , defined by

$$\omega = \frac{LA}{V} - 1, \quad \text{Eq. (8-38)}$$

where  $A$  is the surface area across which the heat is removed from the body, and  $V$  is the body volume, to account for the specific body shape, viz.

$$\omega = \begin{cases} 0 & \text{for a slab insulated at one end} \\ 1 & \text{for a cylinder} \\ 2 & \text{for a sphere.} \end{cases} \quad \text{(Eq. (8-39))}$$

$0 \leq \omega \leq 2$  always, and  $\omega$  may be assigned appropriate values for shapes intermediate between the slab, cylinder, and sphere. For example, a football-shaped body, somewhere between a cylinder and sphere, may be assigned  $\omega = 1.5$ , and a short cylinder with a large diameter-to-height ratio may have  $\omega = 0.5$ .

For the case where the temperature  $T_0 < T_f$  is imposed on the boundary at  $t = 0$ , the time required for complete freezing,  $t_m$  can be estimated by the equation

$$t_f = \frac{L^2}{2\alpha_s(1+\omega)N_{Ste}} \left[ 1 + (0.25 + 0.17\omega^{0.7})N_{Ste} \right], \quad \text{Eq. (8-40)}$$

claimed to be valid with an accuracy within 10% for  $0 \leq N_{Ste} \leq 4$ .

Validating by comparison to the results of an experimentally-verified two-dimensional numerical model of **freezing of lake-shore water (with a mildly-sloped adiabatic lake bottom) initially at the freezing temperature**, Dilley and Lior [170] have shown that freezing progress can be estimated well by the following simple equations.

For a **constant heat flux**  $q_0$  from the top surface to the ambient, the relationship between the depth of freezing  $X(t)$  and time can be expressed as

$$X(t) = \frac{q_0}{\rho_s h_{ts}} t - \left( \frac{q_0}{\rho_s h_{ts}} \right)^3 \frac{t^2}{2\alpha_s} + \frac{5}{6} \left( \frac{q_0}{\rho_s h_{ts}} \right)^5 \frac{t^3}{2} - \dots \quad \text{Eq. (8-41)}$$

with an error < 0.02% for the first 100 hours.

For **convective cooling at the surface** by air at temperature  $T_a$  and with a convective heat transfer coefficient  $\bar{h}$ , the relationship between the depth of freezing  $X(t)$  and time can be expressed as

$$N_{Bi_s}(t) = N_r(t) - \frac{1}{2} N_r^2(t), \quad \text{Eq. (8-42)}$$

where

$$N_{Bi_s} \quad \text{Biot number for the solid, } \equiv \frac{\bar{h}X(t)}{k_s}$$

$$N_t \quad \text{Dimensionless time parameter, } \equiv \frac{\bar{h}^2 N_{Ste_s} t}{k_s \rho_s c_s}$$

valid for small values of  $N_{Ste_s}$  (the quasi-static approximation).

When the top surface is subject to a **combination of constant and of a convective heat flux** so that the total heat flux,  $q_{tot}$ , there is

$$q_{tot}(0, t) = q_0 + \bar{h}[T_a - T(0, t)], \quad \text{Eq. (8-43)}$$

Eq. (8-43) is applicable also for this case, if  $N_t$  is expressed by

$$N_t(t) = \frac{\bar{h}}{k} \left( \frac{q_0 + \bar{h}T_a}{\rho_s h_{fs}} \right) t. \quad \text{Eq. (8-44)}$$

In the lake-freezing simulation, this expression was found to represent the data well up to the time when the ice depth became 2 m.

If freezing started when the ice layer already had a thickness  $X_i$  (at time  $t_i$ ), the approximate solution becomes

$$N_{Bi_s}(t) = -1 + \left[ 1 + Bi_{s,X_i}^2 + 2Bi_{s,X_i} + 2N_t(t) \right]^{\frac{1}{2}}, \quad \text{Eq. (8-45)}$$

where  $N_t$  is defined by Eq. (8-44).

When the top surface is subjected to a **combination of constant and of a convective heat flux, where the air temperature varies linearly with time** as

$$T_a(t) = T_{a,i} + mt, \quad \text{Eq. (8-46)}$$

where

$m$  the time-variation constant of the temperature  $T_a$ ,

$T_{a,i}$  the initial air temperature,

the total heat flux there,  $q_{tot}$ , is

$$q_{tot}(0, t) = \bar{h}[T_a(t) - T(0, t)] + q_0. \quad \text{Eq. (8-47)}$$

The approximate expression relating  $X(t)$  to time is still Eq. (8-42), but with  $N_t$  defined as

$$N_r(T) = \frac{\bar{h}}{k_s \rho_s h_{fs}} \left[ (q_0 + \bar{h}T_{a,i})t + \frac{m\bar{h}}{2} t^2 \right]. \quad \text{Eq. (8-48)}$$

### III. PREDICTIVE EQUATIONS FOR FREEZING WITH FLOW

Freezing may occur when a liquid flows through a cooled conduit or along a cooled wall where the conduit/wall temperature ( $T_w$ ) is below the freezing temperature of the liquid ( $T_w < T_f$ , Fig. 8-6). The heat balance at the phase change interface can be expressed as

$$q[x, R(x, t)] + k_s \left( \frac{\partial T_s}{\partial r} \right)_{[x, R(x, t)]} = \rho_s h_{fs} \frac{\partial R(x, t)}{\partial t}, \quad \text{Eq. (8-49)}$$

where the first and second terms on the left-hand side of the equation account for the heat flow from the flowing liquid and the frozen solid, respectively, to the phase-change interface, and the term on the right-hand side expresses the rate of latent heat release due to the increase in the frozen layer thickness. The first term in Eq. (8-49), i.e. the heat transfer from the flowing liquid, can be expressed as

$$q[x, R(x, t)] = \bar{h}[T_c(x, t) - T_f] \quad \text{Eq. (8-50)}$$

for convection from a liquid at temperature  $T_c(x, t)$  with a convective heat transfer coefficient  $\bar{h}$ .

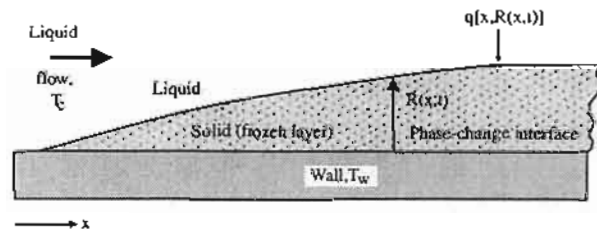


Figure 8-6. Freezing During Liquid Flow Over a Cold Wall

Freezing occurs when the the term on the right-hand side of Eq. (8-49) is negative, happening if at least one of the terms on the left hand side of the equation is negative and larger in its absolute value than the other. Obviously, freezing can thus occur if the interface is cooled on both sides (when  $T_c < T_f$  and thus  $q < 0$ , and also  $T_w < T_f$  and thus the gradient in the conduction term is negative). It can, however, occur even when the flowing fluid temperature is higher than  $T_f$ , if the cooling rate through the frozen layer is high enough, or when the tube wall temperature is higher than  $T_f$  if the cooling rate  $q$  into the flowing fluid (when  $T_c < T_f$ ) is large enough.

Since the flow boundary and cross section keep varying during phase-change, the nature of the flow, including its

velocity, as well as the consequent effects on heat transfer, also vary. For example, inward freezing in a cooled tube would progressively diminish the flow cross section and increase the flow pressure drop (Fig. 8-7). If, as often found in practice, the given flow head is constant, freezing would result in a gradual decrease of the flow rate. Figure 8-8 also shows the experimentally-observed fact that some remelting of the frozen layer occurs at the exit from the partially-frozen region, due to the flow expansion there. Some references on the effects of freezing on flow, pressure drop, and conditions leading to complete flow stoppage in conduits due to freezing, are cited in Subsection I. D. above.

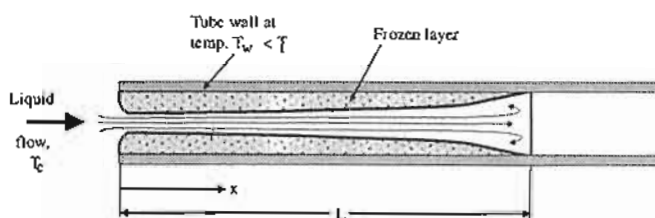
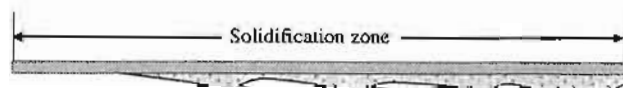


Figure 8-7. A Freezing During Liquid Flow in a Tube



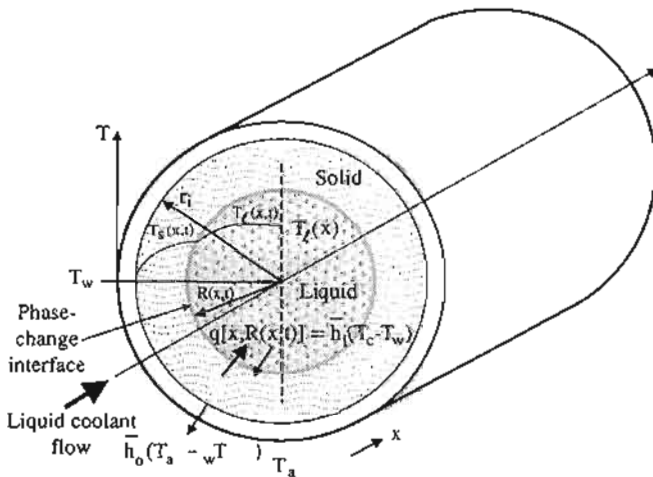
decreasing  $\bar{h}_i$ , and stops when the convective heat transfer at the interface is equal to the conductive one in the ice layer. The flow thus becomes laminar again, which brings another such freezing-melting cycle about, generates another ice band downstream of the first one, and so on. In addition to such changes in the interface shape, dendritic growth of the solid phase—especially prominent when the liquid is subcooled—will create interface roughness on a smaller scale.

Even when the above-described interface shape variations are not taken into account, no analytical solutions for the complete flow-accompanied melting/freezing problem are available. Many numerical, experimental and approximate results have, however, been reported in the literature and listed in the above mentioned reviews (especially see citations [79]-[152] in Subsection I. C.). One useful solution is shown here, for the case of a fluid at the radially-average entrance temperature  $T_c > T_f$  flowing along a flat plate, or in a tube of internal radius  $r_i$ , which are convectively-cooled on their exterior surface by a fluid at temperature  $T_a < T_f$  with a convective heat transfer coefficient  $\bar{h}_o$  (Fig. 8-9). Neglecting heat conduction in the axial direction, an approximate collocation-type transient solution, which accounts for the motion of the phase change interface and for the heat conduction in the frozen layer was

- $n$  geometry index,  $n = 0$  for flow along a flat wall,  $n = 1$  for flow in a cylindrical tube
- $Bi$  Biot number for internal heat transfer to the phase-change interface,  $= \bar{h}_i r_i / k_s$ , dimensionless
- $\eta$  dimensionless length parameter,

$$R^*(x^*, \tau) = \int_1^{X^*} \frac{dX}{X^n}$$

- $q^*[R^*(x^*, \tau)]$  dimensionless heat flux from the liquid stream to the interface,  
 $= q[R(x, t)] r_i / (\rho_s h_{fs} \alpha_s)$
- $q[R(x, t)]$  heat flux from the liquid stream to the interface,  $= \bar{h}_i [T_c(x, t) - T_f]$ ,  $W / m^2$ .



**Figure 8-9.** Sketch of the Phase-Change Problem of a Flowing Liquid on a Cooled Planar Wall or Inside a Cooled Tube, with Notations for Stephan's [141] Solution [Eq. (8-51)]

In this solution, the internal heat flux term  $q[R(x, t)]$  must be specified by the user, since the solution here does not include consideration of the flow momentum equations. Representative constant values of  $q$  may be used for rough assessment.

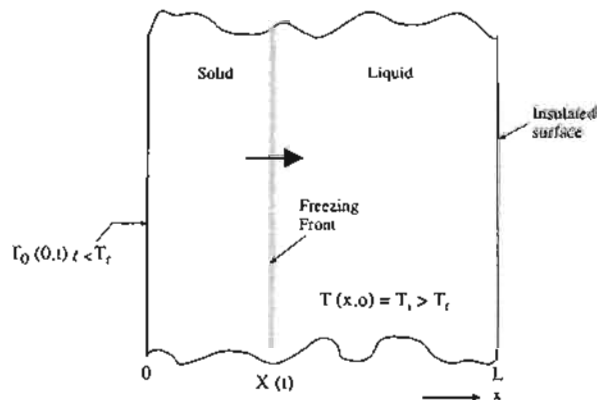
Equation (8-52) can be solved by numerical methods, most easily by using one of the available ordinary differential equation solution software programs. Once  $R^*(x^*, \tau)$  is thus determined, all necessary information about this flow-freezing problem becomes available.

#### IV. SOME METHODS FOR SIMPLIFYING SOLUTION

##### A. THE INTEGRAL METHOD, WITH SAMPLE SOLUTION FOR FREEZING OF A SLAB

A simple approximate technique for solving melting and freezing problems is the *heat balance integral method* (Goodman [181]), which was found to give good results in many cases. The advantage of this method is that it reduces the second-order partial differential equations describing the problem to ordinary differential equations that are much easier to solve. This is accomplished by guessing a temperature distribution shape inside the phase-change media, but making them satisfy the boundary conditions. These temperature distributions are then substituted into the partial differential energy equations in the liquid and solid, which are then integrated over the spatial parameter(s) (here just  $x$ ) in the respective liquid and solid domains. This results in ordinary differential equations having time ( $t$ ) as the independent variable. The disadvantage of the method is clearly the uncertainty in the temperature distribution within the media. This technique is introduced here by applying it to a useful case, and the reader can thus also learn to apply it to other cases.

Consider, as shown in Fig. 8-10, a liquid initially at an above-freezing temperature ( $T_i > T_f$ ) confined in a space  $0 \leq x \leq L$ , with the surface at  $x = 0$  subjected for time  $t > 0$  to a below-freezing temperature  $T_0 < T_f$ , and the surface at  $x = L$  is perfectly insulated,  $(\partial T_i / \partial x)_{L, t} = 0$ . Freezing thus starts at  $x = 0$ , and the freezing front, as shown in Fig. 8-9, is moving rightward. This problem has no exact solution, and is thus a good example for the application of the integral method described in this section.



**Figure 8-10.** Sketch for the Problem of Freezing of a Liquid Slab Initially at an Above-Freezing Temperature

In the derivation and discussion, the following dimensionless parameters are used:

$$\theta_j \equiv \frac{T_j - T_0}{T_i - T_0}, \text{ where } j = s, \ell, \text{ or } f, \quad \text{Eq. (8-52)}$$

$$\chi \equiv \frac{x}{L}, \quad \xi \equiv \frac{X}{l}, \quad N_{Fo_s} \equiv \frac{\alpha_s t}{L^2}, \quad \text{Eq. (8-53)}$$

and the problem is re-sketched in terms of these dimensionless parameters in Fig. 8-11. For the example at hand, the partial differential equations describing the problem are, for the solid

$$\frac{\partial \theta_s(\chi, N_{Fo_s})}{\partial N_{Fo_s}} = \frac{\partial^2 \theta_s}{\partial \chi^2} \text{ in } 0 < \chi < \xi(t), \text{ for } N_{Fo_s} > 0, \quad \text{Eq. (8-54)}$$

with the boundary condition

$$\theta_s(0, N_{Fo_s}) = 0 \text{ for } N_{Fo_s} > 0, \quad \text{Eq. (8-55)}$$

for the liquid

$$\frac{\partial \theta_\ell(\chi, N_{Fo_s})}{\partial N_{Fo_s}} = \frac{\partial^2 \theta_\ell}{\partial \chi^2} \text{ in } 0 < \chi < \xi(t), \text{ for } N_{Fo_s} > 0, \quad \text{Eq. (8-56)}$$

with the boundary condition

$$\frac{\partial \theta_\ell}{\partial \chi} = 0 \text{ at } X = 1, \text{ for } N_{Fo_s} > 0, \quad \text{Eq. (8-57)}$$

and the initial condition

$$\theta_\ell = 1 \text{ in } 0 < \chi < 1 \text{ for } N_{Fo_s} = 0, \quad \text{Eq. (8-58)}$$

and at the phase-change interface

$$\theta_s[\xi(t), N_{Fo_s}] = \theta_\ell[\xi(t), N_{Fo_s}] = \theta_f, \quad \text{Eq. (8-59)}$$

$$\frac{\partial \theta_s}{\partial \chi} - \frac{k_\ell}{k_s} \frac{\partial \theta_\ell}{\partial \chi} = \frac{h_{fs}}{c_s(T_i - T_0)} \frac{d\xi(N_{Fo_s})}{dN_{Fo_s}} \quad \text{Eq. (8-60)}$$

$$\text{at } \chi = \xi(N_{Fo_s}), \text{ for } N_{Fo_s} > 0.$$

The next step, as explained above, is to choose temperature distributions in the two phases. Obviously, the closer the chosen distributions are to the actual (but unknown) ones, the better the solution would be. A reasonable guess (although other ones can be tried) in the solid phase is the exact solution obtained for freezing a semi-infinite liquid initially at an above-freezing temperature, shown in Eq. (8-11), which is here, in its dimensionless form,

$$\frac{\theta_s(\chi, N_{Fo_s})}{\theta_f} = \frac{\text{erf}\left(\frac{\chi}{2\sqrt{N_{Fo_s}}}\right)}{\text{erf}\lambda}, \text{ for } N_{Fo_s} > 0, \quad \text{Eq. (8-61)}$$

where  $\lambda$  is a parameter yet to be determined. The reader can easily prove that this is indeed the solution of Eqs. (8-55), (8-56).

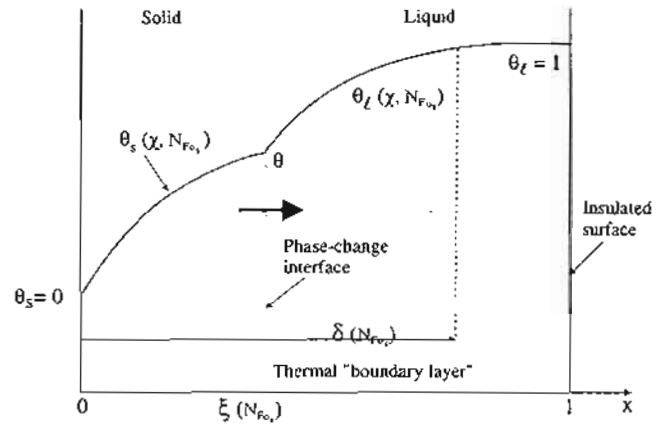


Figure 8-11. Sketch for the Energy Integral Method Solution for Freezing of a Liquid Slab Initially at an Above-Freezing Temperature, with Dimensionless Variables

It is also assumed that the position of the phase change interface is defined by an expression similar to Eq. (8-13),

$$\xi(N_{Fo_s}) = 2\lambda\sqrt{N_{Fo_s}}, \quad \text{Eq. (8-62)}$$

The value of  $\lambda$  as well as the temperature distribution in the liquid phase are now determined by the integral method, as follows. A thermal "boundary layer"  $\delta(N_{Fo_s})$  is defined at an  $x$ -location where the liquid temperature is still at its initial value (the cooling effect has not penetrated to that location yet. See Fig. 8-10.), and the heat flux is 0, viz.

$$\theta_\ell[\delta(N_{Fo_s}), N_{Fo_s}] = 1, \quad \text{Eq. (8-63)}$$

and

$$\left(\frac{\partial \theta_\ell}{\partial x}\right)_{\delta(N_{Fo_s})} = 0, \quad \text{Eq. (8-64)}$$

respectively. Note that a solution is valid only if  $\delta(N_{Fo_s}) \leq 1$ . Now the differential energy equation for the liquid, Eq. (8-57), is integrated in the liquid phase domain from  $\xi(N_{Fo_s})$  to  $\delta(N_{Fo_s})$ , and the boundary conditions represented by Eqs. (8-58), (8-64) and (8-65), giving the expression

$$\frac{\alpha_\ell}{\alpha_s} \left(\frac{\partial \theta_\ell}{\partial x}\right)_{\xi(N_{Fo_s})} - \frac{\alpha_\ell}{\alpha_s} \left(\frac{\partial \theta_\ell}{\partial x}\right)_{\delta(N_{Fo_s})} + \frac{d\delta}{dN_{Fo_s}} - \theta_f \frac{d\xi(N_{Fo_s})}{dN_{Fo_s}} = \frac{d}{dN_{Fo_s}} \left\{ \int_{\xi(N_{Fo_s})}^{\delta(N_{Fo_s})} \theta_\ell(\chi, N_{Fo_s}) d\chi \right\} \quad \text{Eq. (8-65)}$$

This is the *energy-integral equation* for this problem. An appropriate temperature distribution must be chosen for completing the integration. For example, the polynomial distribution

$$\theta_t(x, N_{Fo_s}) = 1 - (1 - \theta_f) \left( \frac{\delta - \chi}{\delta - \xi} \right)^n, \quad \text{Eq. (8-66)}$$

where  $n \geq 2$  is the power of the polynomial, satisfies the boundary conditions, Eqs. (8-58), (8-63) and (8-64). It is also assumed that  $\delta(N_{Fo_s})$  is related to  $N_{Fo_s}$  through the relation

$$\delta(N_{Fo_s}) = 2\beta \sqrt{N_{Fo_s}}, \quad \text{Eq. (8-67)}$$

with the parameter  $\beta$  to be determined.

The temperature distribution [Eq. (8-66)] is substituted into the energy integral equation (8-65), which is integrated using Eq. (8-67) to yield

$$\beta - \lambda = \frac{n+1}{2} \left[ -\lambda + \sqrt{\lambda^2 + \frac{2n}{n+1} \frac{\alpha_t}{\alpha_s}} \right], \quad \text{Eq. (8-68)}$$

Next, the temperature distributions in the solid and liquid, Eqs. (8-61) and (8-66), respectively, are introduced into the interfacial condition Eqs. (8-59) and (8-60), to yield the following transcendental equation for the unknown parameter  $\lambda$

$$\frac{e^{-\lambda^2}}{\text{erf}(\lambda)} + \frac{k_t}{k_s} \left( \frac{\alpha_s}{\alpha_t} \right)^{1/2} \frac{\theta_f - 1}{\theta_f} \frac{1}{Z_n} = \frac{\lambda h_{ts} \sqrt{\pi}}{c_s (T_f - T_0)}, \quad \text{Eq. (8-69)}$$

where

$$Z_n \equiv \frac{n+1}{n\sqrt{\pi}} \left( -\gamma + \sqrt{\gamma^2 + \frac{2n}{n+1}} \right), \quad \text{Eq. (8-70)}$$

$$\gamma \equiv \lambda \left( \frac{\alpha_s}{\alpha_t} \right)^{1/2}, \quad \text{Eq. (8-71)}$$

$$\frac{\theta_f - 1}{\theta_f} \equiv \frac{T_f - T_i}{T_f - T_0}, \quad \text{Eq. (8-72)}$$

Solution of Eq. (8-69) manually, or easily done by one of many software packages available for solving nonlinear algebraic equations, yields the value of  $\lambda$ . This and Eq. (8-68) yield the value of  $\beta$ , and thus the transient position of the freezing front,  $\xi(N_{Fo_s})$ , can be calculated from Eq. (8-62), and the temperature distributions in the solid and liquid can be calculated from Eqs. (8-61) and (8-66), respectively.

Inspection of Eq. (8-62) also indicates that the slab would be completely frozen when the dimensionless time reaches

the value  $N_{Fo_s} = 1/(4\lambda^2)$ , and [with Eq. (8-67)] that the validity of this particular integral solution is confined to dimensionless times for which  $\delta \leq 1$ , corresponding to  $N_{Fo_s} \leq 1/(4\beta^2)$ .

Many integral solutions yield good results, with errors within a few percent. The accuracy, as mentioned above, depends on the closeness of the chosen temperature distributions to the real ones. Experience from previous successful solutions or experimental results naturally improves this choice. Additional information about this method can be found in refs. [181], [51], and [210].

## B. THE ENTHALPY METHOD

It is noteworthy that one of the biggest difficulties in numerical solution techniques for such problems is the need to track the location of the phase-change interface continuously during the solution process, so that the interfacial conditions could be applied there. One popular technique that alleviates this difficulty is the enthalpy method (refs. [3], [72], [154], [1723 174], [179], [190], [228], [240], [259] and [260]), in which a single partial differential equation, using the material enthalpy instead of the temperature, is used to represent the entire domain, including both phases and the interface. Based on the energy equation, just as Eqs. (8-55) and (8-57), the one-dimensional melting problem is thus described by

$$\rho \frac{\partial h}{\partial t} = k \frac{\partial^2 h}{\partial x^2} \quad \text{in } x \geq 0, \text{ for } t > 0, \quad \text{Eq. (8-73)}$$

where the temperature-enthalpy relationship is expressed by

$$\begin{aligned} \frac{h}{c} & \quad h \leq cT_f & \text{(solid)} \\ T = T_f & \quad cT_f < h < cT_f + h_{ts} & \text{(interface)} \\ \frac{h - h_{ts}}{c} & \quad h \geq cT_f + h_{ts} & \text{(liquid)} \end{aligned} \quad \text{Eq. (8-74)}$$

The numerical computation scheme is rather straightforward: knowing the temperature, enthalpy and thus from Eq. (8-74) the phase of a cell at time step  $j$ , the enthalpy at time step  $(j+1)$  is computed from the discretized version of Eq. (8-73), and then Eq. (8-74) is used to determine the temperature and phase at that new time. If a computational cell  $i$  is in the mushy zone, the liquid fraction is simply  $h_i / h_{ts}$ .

Care must be exercised in the use of the enthalpy method when the phase change occurs over a very narrow range of temperatures. Oscillating non-realistic solutions were obtained in such cases, but several modifications (see above references) to the numerical formulation were found to be reasonably successful.

**V. APPLICATIONS BIBLIOGRAPHY**

An extensive—yet by no means complete—bibliography identifying papers and books that treat freezing in the main areas in which it takes place, is presented below. The classification is by application, and the internal order is alphabetical by author.

**A. CASTING, MOLDING, SINTERING**

Freezing (solidification) is a key component in casting, molding, and production of solid shapes from powders by processes such as sintering and combustion synthesis. The materials include metals, polymers, glass, ceramics, and superconductors. Flow of the molten material, the course of its solidification (including volume changes due to phase transition, and internal stress creation), and the evolving surface and interior quality, are all of significant industrial importance. In production of parts from powders, the conditions necessary for bonding of the particles by melting and resolidification are of importance. Such diverse processes as spinning and wire-making are included. Much attention has lately been focused on the manufacturing of materials for superconductors.

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#### B. MULTI-COMPONENT SYSTEMS, FREEZE-SEPARATION, AND CRYSTAL-GROWTH

The bibliography in this section primarily focuses on freezing of multi component system, but crystal growth also includes pure crystals. As compared with the freezing of single component systems, multi-component system freezing is accompanied by change of composition as discussed in section 5XX.1, a phenomenon of great significance in the formation of the solid material. The analysis and prediction of the process are thus also made more complex, in that the species diffusion process and the effect of the concentration on the freezing point and other properties, must be considered.

One of the most prominent applications is alloy-making, and the last several decades have seen large and increasing involvement with crystal growth, primarily for the electronics and optical industries. Crystals are typically grown by melting the feedstock and letting it solidify in the form of a crystal. Crystals may be made of either pure or multi-component materials, but even when pure crystals are made, much research has been done on the effect



of impurities introduced during the manufacturing process. This in effect renders even the pure crystal to be considered as a multi-component system. Crystal growth is accomplished by a variety of processes, including Czochralski, Bridgman, Float-Zone, and thin film deposition.

The change of composition of multi-component systems during freezing is used in various freeze-separation processes, in which components are separated for some useful purpose. One example is the process of freeze-desalination, in which saline water is frozen, thereby separating the water from the salt. The latter migrates to the ice crystal surface, from which it is washed by fresh water. The ice is made of pure water, which can then be used. Another well-known process is freeze-drying, in which water is separated from a solid, such as coffee.

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The bibliography in this section is related to processes associated with solidification which are used for joining, cutting, shaping and property-modification of solids. Among other applications, laser energy is used for cutting and surface property modification, a melting process followed by solidification. In the electro-discharge machining process, sparks are generated by the application of a voltage between an electrode (the tool) and the workpiece to be machined. The closely controlled sparks melt small craters in the workpiece, and the melt is continuously removed. Some of the melt solidifies at the crater rim, and should, for best surface finish, also be removed.

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The bibliography in this section is of various coating processes in which a vapor or liquid are deposited on a surface at a temperature below the freezing point, on which they solidify to form a coating. The process can also be continued to build up desired shapes. Some of the processes covered in the bibliography include plasma and thermal spray deposition, vapor deposition, dipping in a melt, and coating by liquid films.

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## F. MEDICAL APPLICATIONS AND FOOD PRESERVATION

The bibliography in this section covers biological applications of freezing, such as those used in medicine and in food preservation. The medical applications include organ preservation, preservation of tissue cultures, cryosurgery, and freezing damage to live tissue, such as in frost-bite.

Food preservation by freezing, an ancient practice, still attracts much R&D attention, in attempts to shorten freezing times, reduce energy consumption, prolong the life of foods, and minimize damage to their nutritional value, taste, odor and appearance.

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## E. FROST FORMATION

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### G. NUCLEAR POWER SAFETY

Solidification play a role in several areas related to nuclear power plant safety and operation. The primary one is the risk of the reactor core meltdown upon failure of the cooling system to limit the temperature rise, such as the accident at the Three Mile Island reactor. Melting, and the subsequent solidification due to cooling, have been the subject of numerous publications, some of which are listed in the bibliography in this section. Further, interaction of the molten metal with coolant, which promotes solidification, will also call vapor explosions and chemical reactions which may generate large quantities of gas, some of which (such as hydrogen) are explosive.

Liquid-metal cooled reactors use coolants, such as potassium, sodium and their alloys, which solidify a room temperature or somewhat above it. A number of publications on this topic are available. Another nuclear application employing freezing is vitrification of nuclear waste for longer term storage.

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### H. THERMAL ENERGY STORAGE

Storage of heat in a material by melting, and subsequent release of the heat by re-freezing the material allows a large amount of heat to be stored in a relatively-small mass of material and at a theoretically constant temperature (that of fusion). An issue which has received much attention is the way to exchange with such a phase-change storage material at a high rate and low investment in heat transfer equipment. A very broad range of materials have been considered and used, including water, inorganic salts, hydrocarbons, polymers, and metals.

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