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AN APPLIED OVERVIEW OF MOVING BOUNDARY PROBLEMS

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August 1977

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AN APPLIED OVERVIEW OF MOVING BOUNDARY PROBLEMS

Bruno A. Boley

An overview of moving-boundary problems is sketched in a general way with emphasis on some types of problems in which further work appears to be desirable from an applied viewpoint. Areas touched are those of the mechanical behavior of melting or solidifying bodies, the establishment of suitable constitutive equation, the use of approximate methods, the state of multidimensional problems, and the application of continuum approaches to the study of crystal growth.

1. INTRODUCTION

Under the three headings of Theory, Methods and Applications, the present conference spans the remarkably broad body of current research in problems involving moving boundaries. A glance at the program will convince anyone of the impossibility of summarizing in a single paper even the principal features of the work being done in any one of the three categories, without degenerating into a tedious, although possibly useful, extensive bibliography. Moreover, an attempt at such a summary at the beginning of the conference cannot possibly include adequate reference to the work to be presented during the next succeeding days, and thus ipso facto must fall short of an up-to date description of the state of the field.

The panel discussion on the last day of the conference will nevertheless endeavor to formulate some general observations on the state of current knowledge and current needs. I thought it would be more appropriate at this time to indicate certain areas of research which, in my opinion, have a distinct practical importance, but are nevertheless insufficiently emphasized or absent at the present conference. The principal such areas are the following:

- Studies of the mechanical response accompanying changes of phase
- Determinations of material behavior and constitutive relations in regimes encountered in change of phase problems
- The use of approximations and of approximate methods
- Analytical work on multidimensional problems
- Correlations between approaches at the microscopic ("material science") and continuum("mechanics") scales

Each of these will be discussed in what follows, together with some general observations which may on occasion be appropriately introduced. In all cases, no effort will be made at a comprehensive bibliography, and only typical researches will be indicated; these and the references cited therein should nevertheless form a good introduction to the work in any one of the areas under discussion. In addition, it may be well to mention here an earlier conference [1.1] which also brought together--to the author's knowledge, for the first time--mathematicians and practicing engineers active in this field.

2. MECHANICAL BEHAVIOR DURING CHANGES OF PHASE

The deformations and stresses accompanying problems of change of phase are of great importance in a variety of applications. As examples one might mention the stresses arising during casting solidification [2.10,18] the deformations responsible for air-gap formation between casting skin and mold [2.12,14,17] and the accompanying phenomenon of interface instability [2.11,19], the deformation ablating shields [2.9,15] and solid propellants [2.13] and those of nuclear reactor fuel elements [2.5].

The first question that must be raised with regard to problems of this kind concerns the permissibility of uncoupling the thermal and mechanical effects, as indeed was done in all the works mentioned above. Such uncoupling is permissible in ordinary thermoelastic problems, when the coupling parameter.

$$\delta = \frac{(3\lambda + 2\mu)^2 \alpha^2 T^*}{(\lambda + 2\mu) \rho c_v} \quad (2.1)$$

is small compared to unity, and dynamic effects are small [2.7]. In this equation λ and μ are the Lamé elastic constants, α is the coefficient of linear expansion, ρ the density, c_v the specific heat at constant volume and T^* a reference (absolute) temperature at which the material is stress-free. The latter effect depends principally on the magnitude of a thermal velocity (speed of interface advance, or of isotherm motion) relative to the speed of stress waves in the solid; this is normally sufficiently small to cause mechanical effects to be passed (other than after reflections) by the time the thermal ones become significant. An elastic analysis of the dynamic stresses accompanying a melting process may be found in [2.16]; the stresses in the Neumann problem in the direction of solidification may be the same order of magnitude as those arising in the other directions under static conditions [cf. eq. (2.6)], but can be expected to decrease rapidly if the actual temperature does not experience instantaneous jumps (cf. 2.7)).

An estimate of the effect of δ may be obtained by recalling [2.14] that in simple thermoelastic problems it is equivalent to a replacement of the diffusivity κ by the effective diffusivity

$$\kappa^* = \frac{\kappa}{1 + \delta} \quad (2.2)$$

and by noting that this statement carries over to moving-boundary solutions of the Neumann type. This is best seen by regarding such solutions as arising from linear combinations of elementary heat-conduction solutions for a half-space with arbitrary conditions at the surface and at the far end, and initially (cf. [2.4]); then use of the effective diffusivity κ^* applies to each elementary solution, provided that in each the surface tractions are required to vanish. This follows from integration of the one-dimensional equation of equilibrium in the absence of inertia effects, namely

$$(\lambda + 2\mu)u_{x,xx} - (3\lambda + 2\mu)\alpha T_{,x} (\equiv \sigma_{xx,x}) = 0 \quad (2.3)$$

where u_x and σ_{xx} represent displacement and stress components respectively, and commas indicate differentiation. Substitution into the linearized coupled heat equation.

$$KT_{,xx} = \rho c_v \dot{T} + (3\lambda + 2\mu)\alpha T^* \dot{u}_{x,x} \quad (2.4)$$

(where K is the thermal conductivity and dots indicate differentiation with respect to time) gives

$$\kappa T^*_{,xx} = \dot{T} \quad (2.5)$$

The stress component σ_{xx} is, as has been noted, identically zero, and so are, of course, the shears. The only non-zero stress components are (in the melting case)

$$\sigma_{yy} = \sigma_{zz} = -\frac{\alpha ET}{1 - \nu} \quad (2.6)$$

where E is Young's modulus and ν is Poisson's ratio.

The above results have been obtained for the purely elastic one-dimensional Neumann problem, but may be practically useful in providing a preliminary estimate of the magnitude of this effect even in more complicated problems where its neglect may be questioned. In any case, certain parts of the solution (e.g., the coefficient λ in the Neumann expression for the interface positions, i.e.,

$$s(t) = 2\lambda \sqrt{\kappa t} \quad (2.7)$$

(where t is time) are not likely to be very sensitive to changes in diffusivity; λ in fact is independent of κ for equal properties in the solid and liquid [2.8,11]. Extension of these results to problems other than the ones for which they are strictly valid may be exemplified by an examination of (still one-dimensional) cases in which the moduli are temperature dependent, in which (with $\sigma_{xx} \equiv 0$ and $\sigma_{yy} = \sigma_{zz} \equiv \sigma$)

$$u_{x,x} = f_1(\sigma, T) ; u_{y,y} = u_{z,z} \equiv 0 = f_2(\sigma, T) \quad (2.8)$$

or

$$u_{x,x} = F(T) \quad (2.8a)$$

where the effective diffusivity is

$$\kappa^* = \frac{\kappa}{1 + \delta \frac{dF}{dT}} \quad (2.8b)$$

since similarity still holds in problems with temperature-dependent properties [2.4]. The last statement is no longer valid if the solid is viscoelastic, since in that case (even on the normal assumption of elastic behavior under hydrostatic conditions [2.7])

$$(\lambda + 2\mu)\bar{u}_{x,x} = (3\lambda + 2\mu) f(p)\alpha\bar{T} \quad (2.9)$$

where bars indicate Laplace transforms and p is the Laplace transform parameter. Thus the transformed heat equation becomes (since dissipative effects vanish in the one-dimensional problem [2.7]):

$$\kappa\bar{T}_{,xx} = p\bar{T}[1 + \delta f(p)] \quad (2.9a)$$

This somewhat detailed discussion of coupling should not be taken to imply that this effect is often likely to assume major importance, nor is it an open invitation to mathematicians to plunge heedlessly into a relatively unexplored field. It is rather an expression of the feelings that this very lack of experience makes it prudent to proceed with caution, and to give some thought to the validity of the inevitable underlying assumption of the physical model under study. Some rules of thumb permitting some preliminary estimates of the validity of such assumptions are therefore particularly useful.

Whether coupling is important or not, the mechanical response itself of course is. In particular, the stresses of eq. (2.6) are present regardless of coupling. In the solidification case, these stresses are

$$\sigma_{yy} = \sigma_{zz} = - \frac{\alpha E(T - T_m)}{1 - \nu} \quad (2.10)$$

on the assumption that no bending occurs during the formation of the solidified skin. Should such bending occur, they may at times be estimated as in [2.6]; in particular if the skin is free of bending and axial restraints, they cannot exceed $(4/3) |\alpha E \Delta T / (1-\nu)|$, where ΔT is the maximum temperature excursion in the skin. No studies of stresses and deformation in multidimensional problems have yet been made to the author's knowledge.

The calculation of the mechanical response requires, first of all, an accurate knowledge of temperature distributions, gradients, and history (some discussions of the errors which may arise even in non-moving-boundary problems because of inaccuracies in the temperature may be found in [2.3,6]). It cannot be overemphasized that analyses which are sufficiently accurate for "thermal" purposes may not be so for stress calculations, a fact that may be easily overlooked particularly when dealing with purely numerical solutions. In fact the development of some information regarding the accuracy required in the temperature for desired accuracy of stresses and deformations would seem to be a useful subject of future mathematical research.

It should be clear from the preceding discussion, however, the type of material behavior assumed in the analysis will greatly influence the results; thus, before proceeding much further, it is necessary that more data on this subject be developed experimentally.

3. MATERIAL PROPERTIES AND CONSTITUTIVE EQUATIONS

Considerable data have been gathered by various investigators on the variation of material properties with temperature, in some cases approaching the melting temperature. Much more is known regarding the thermophysical properties (e.g., [3.1]) than mechanical ones, although many investigations for specific materials have been undertaken over the years (see, for example, the 21 studies of low-carbon steels listed in [3.3]). Information of

this type is of course extremely useful, but falls somewhat short of the more general goal of establishing constitutive relations appropriate to a given material under given conditions. For example, the studies cited in [2.18], [2.17] and [2.12], treat the solidifying ingot as temperature-dependent elasto-plastic, elastic, and non-linearly viscoelastic respectively. There is some conceptual difficulty with the first of these, in that uniqueness of solution is not assured (the uniqueness theorems are incremental [2.7], and uniqueness of the initial stress state in a strip of zero initial thickness is difficult to establish; no such difficulty arises in analogous melting problems, e.g. [2.9] or [2.5]. Elastic behavior is almost certainly not to be found in reality in the regimes encountered in change-of-phase problems, but that does not mean that such studies are without value: they may provide, at the very least, an extremely useful initial understanding of the problem, and, in some instances, a useful approximation. Thus, it was noted in [2.9] that the deformations of a melting elasto-plastic plate would be well approximated (from above) by those calculated elastically. When melting occurs as a result of internal heat generation, the approximation still holds, but from below [2.5].

A combination of temperature-dependent plastic and non-linear viscoelastic behavior will undoubtedly describe most desired conditions. However, even aside from the fact that the material properties in such a model are quantitatively not known, what is needed is some knowledge of the simplest model that may properly be used. For example, a simple work-hardening behavior was found to be exhibited by the austenitic steel in [3.2], but not by the delta-ferritic iron of [3.3], the latter however lending itself to a simple relation for the stress (under conditions of constant strain), namely

$$\sigma = A(\dot{\epsilon})^m \exp(B/T) \quad (3.1)$$

where A, B and m are constants, while the former does not.

Progress in the field would probably be best aided by a joint effort--whether in the format of a workshop or a task-force study--of experimental investigators, mechanics experts, and practicing engineers to formulate the needs for various conditions and the corresponding necessary tests. Such an activity would not, of course, immediately provide all the desired answers, but would probably focus future research in the most useful directions.

Attention has been centered here on the importance of the subject from the standpoint of the calculation of the mechanical response; one should however not ignore the problems presented by the temperature-dependence of properties in the simple diffusion problem itself. The prevailing feeling probably is that (once the variation of the various properties with temperature has been determined) such problems can only be dealt with by means of numerical solutions. Even so, most numerical analyses ignore the temperature dependence because of the difficulties introduced by the additional non-linearity, and certainly more research towards rendering solutions of the type practicable would be useful.

It may be noted, in the meantime, that work is lacking even in the simple Neumann case, in which, as has been mentioned, similarity holds even if the properties are temperature-dependent. In that case [2.4] the construction of the solutions as linear combinations of elementary ones with stationary boundaries should prove useful for the numerical analysis of many simple but important change-of-phase problems.

4. APPROXIMATIONS

Two types of approximations should be distinguished: the first are in the nature of basic assumptions underlying the physical model adopted for the solution, the second refers to the method of solution itself. A few words about each of these types may be of interest.

Basic assumptions, introduced as a matter of course in order

to bring complex actual problems within reach of solution, are normally justified by ad hoc considerations of the particular problem at hand. As a consequence, their validity cannot be judged except by someone with considerable acquaintance and expertise with the field. Furthermore, they are not always very explicitly stated in most papers, since these are as a rule addressed to other experts in the same field, namely to people whose familiarity with them makes their repetition superfluous. Thus, for example, papers describing moving-boundary problems in astrophysics (e.g., [4.6,21]) or in combustion (e.g., [4.16]) are difficult to assess by workers with primary interest, say, in metal castings. What is more serious is the fact that this makes effective communications between different groups difficult; such communication as does occur in fact takes place at the next level of the investigation, namely after the physical model formulation has been accepted. At this level, the second type of approximation earlier mentioned comes into play.

Before proceeding to a discussion of methods of solution, however, one additional observation might be appropriate. The physical model must be chosen so as to represent with reasonable accuracy the aspects of the actual behavior which are of particular interest, and may slight others of lesser immediate interest. Thus, for example, it is permissible to neglect the change in density during change of phase if the progress of the solid-liquid interface is of paramount interest, but this effect cannot be ignored in determining the details of the flow in the liquid [4.13] or the grain structure of the solid metal [4.14]. One particular example can illustrate this point rather dramatically: the rate of melting of the interior of nuclear reactor fuel rod can be described rather accurately [4.17] without consideration of the density increase upon melting, but if this effect is ignored, the catastrophic collapse caused by the accompanying high internal pressure will not appear [4.19,20].

Once the basic model to be studied has been established, one

is ready to consider the method of solution. It goes without saying that this should be as accurate as possible, so as to insure that a faithful representation of the model's behavior has been obtained. The few cases in which an exact solution is possible require no further comment; for the others, a choice of numerical or analytical techniques, or a combination of them, must be made. It may seem inappropriate, especially in view of the separation of Methods and Applications in the format of this Conference, that in a survey of the latter a discussion of the former should be included, but I did think it was useful to mention the perhaps obvious fact that the method of solution must be fitted (just as the physical model was) to the practical goals of the analysis.

Numerical methods for Stefan-type problems will be discussed in detail by several other participants during this Conference. From the standpoint of applications, there is little doubt that numerical methods are the most attractive, and often the only ones, which can accommodate in any reasonable manner the inevitable practical complexities of the problem. This implies that emphasis should be placed on the development of numerical methods capable of incorporating the additional non-linearity introduced by the temperature dependence of material properties. Cooperation between analytical and numerical solutions is however extremely important; an example of this is the necessity of developing in many cases analytical short-time solutions to be extended numerically [4.20].

Approaches based on various finite difference schemes have been generally more popular in heat and mass transfer problems than those based on the finite element approach. The usefulness of the latter should however be further examined, particularly in problems in which the mechanical response is also desired.

Not to be overlooked among numerical methods are those which are based on essentially analytical developments, such as the numerical solution of integral equations (e.g., [4.11,18]).

We finally turn to approximate methods proper, namely techniques for developing approximate analytical solutions. These should play an important role in applied problems, since they can provide, at their best, rapid and hopefully reasonable estimates of the principal characteristics of the sought-for solutions. Best known among such methods is the "heat balance" approach [4.10], normally used in conjunction with a "penetration-depth" concept, namely an unknown distance from the exposed surface beyond which the body is unaffected by the conditions to which the surface is exposed.

The heat balance method is a special case of the "weighting-function" approach [2.6]; alternatively, it can be thought as a special case of a variational approach. The latter have received considerable attention in the literature either in direct form (e.g., [4.5]) or in Biot's Lagrangian formulation [4.1].

These methods have been rather successful in the solution of several one-dimensional problems. From the applied standpoint, more work seems to be necessary in three principal directions, namely the development of techniques for obtaining improvements on a given approximate solution, the adaptation to cases of variable properties, and their use in multidimensional problems. Some work on the first two of these is being done (e.g., [4.2,4]); application to moving-boundary problems of the Stefan-type was carried out in [4.7] (see also [2.2]) based on the use [4.4] of multiple penetration depth, and shows the expected improvement. An approximate method of solution of multidimensional problems based on a separation of variables approach was given in [4.12].

The accuracy of approximate solutions obtained by methods such as the ones mentioned above is always difficult to assess. When two or more successive approximations are available, one may be satisfied (or may have to be) in an engineering sense with a comparison between them. In the case of temperature dependent properties, it was suggested in [4.4] that comparison of two separate solutions of the problem (one obtained directly from the

heat conduction equation, and the other after application of the Kirchhoff transformation) would yield a means of estimating the accuracy of either solution.

Finally, methods of determining strict but easily calculated bounds have been explored as a possible answer to this problem (e.g., [4.3, 9, 15]), but clearly more work in this direction is necessary. Published work in this area has mainly been based on theorems comparing the behavior under different prescribed surface conditions, or different rates of ablation, (cf. [2.6]), or on some special characteristics of the solution (e.g., [4.15]). The development of further such theorems and their applications should be further examined; at the same time comparison theorems based on different variations of material properties should be devised. Such theorems would permit the replacement of the actual temperature dependence of the properties by simpler ones for which the solution is more tractable, and would thus provide useful and hopefully easily obtainable bounds to the desired solution.

Unfortunately, theorems of this type are difficult to develop, and in fact, to the author's knowledge, now have appeared in the literature. To indicate what might be done, the following proof, although allowing only limited variations of properties, may be an interesting first step illustrating both the opportunities and the difficulties of the problem.

Consider the one-phase Stefan problem in a slab $0 < x < L$, initially solid at zero temperature, heated by a non-decreasing heat flux $Q_0(t)$ at $x = 0$ and insulated at $x = L$. The problem is thus described by the system

$$[K(T)T']' = (\rho c)(T)\dot{T} \quad s(t) < x < L \quad (4.1)$$

$$T(x, 0) = 0 \quad 0 < x < L \quad (4.2)$$

$$T'(L, t) = 0 \quad 0 < t < t_L \quad (4.3)$$

$$-K(T_m)T'(s, t) = Q_0(t) - \rho l \dot{s} \quad 0 < t < t_L \quad (4.4)$$

$$T[s(t), t] = T_m \quad t_m < t < t_L \quad (4.5)$$

$$s(t) = 0 \quad 0 < t \leq t_m \quad (4.6)$$

where the times t_m and t_L are defined by

$$T(0, t_m) = T_m \quad \text{and} \quad s(t_L) = L \quad (4.7)$$

The solution of this problem is unique [4.12, 2.6]; furthermore $\dot{T} \geq 0$ and $\dot{s} \geq 0$ throughout.

Consider now two problems, denoted by the subscripts 1 and 2 respectively, in which

$$(\rho c)_2(T) \geq (\rho c)_1(T) \quad (4.8)$$

Then, from the difference in the two heat balances

$$\int_0^t Q_o(t) dt = \int_{s_{1,2}(t)}^L H_{1,2} dx + (\rho l + H_{m_{1,2}}) s_{1,2} \quad (4.9)$$

where the heat content is

$$H(T) = \int_0^T \rho c dT \quad ; \quad H_m = H(T_m) \quad (4.9a)$$

we have (cf. [4.12])

$$t_{L1} \leq t_{L2} \quad (4.9b)$$

Assume now that the difference in the physical properties characterizing the two problems is such that the solutions of problem (4.1-7) satisfies

$$T_1 \geq T_2 \quad (4.10)$$

if the boundary conditions and the initial conditions satisfy inequalities of the type

$$T_1(s^*, t) \geq T_2(s^*, t) \quad \text{or} \quad T_1'(s^*, t) \leq T_2'(s^*, t) \quad (4.11)$$

$$T_1(0, t) \geq T_2(0, t)$$

for any curve $0 < x = s^*(t) < L$. The development of useful and fairly general conditions for (4.10) to hold is however one of the

principal difficulties of the problem, and no attempt to resolve it here is presented; we shall devote our attention to proving that, if it holds, then

$$s_1(t) \geq s_2(t) \quad (4.12)$$

Some simple conditions under which (4.10) is valid may be listed first. For example, if K is constant and equal in the two solutions, then the heat equation is

$$KT''_{1,2} = (\rho c)_{1,2} (T_{1,2}) \dot{T}_{1,2} \quad (4.13)$$

and (4.10) follows from subtraction of one of (4.13) from the other, i.e.,

$$K(T_2 - T_1)'' - (\rho c)_2(T_2 - T_1)' + [\rho c_1(T_1) - \rho c_2(T_2)] \dot{T}_1 = 0 \quad (4.14)$$

provided that one can insure that

$$\rho c_2(T_2) \geq \rho c_1(T_1) \quad (4.14a)$$

This can be done if the specific heat is constant (or dependent on time and position but not temperature), or if c_2 is replaced by the maximum possible value of c_1 (which may be possible in change-of-phase problems because the extremes of temperature can often be estimated a priori).

As another example, let now

$$K_1(T) \geq K_2(T) \quad \text{and} \quad \kappa_1(T) \geq \kappa_2(T) \quad (4.15)$$

Then, with

$$T^*(T) = \frac{1}{K_0} \int_0^T K(T) dT \quad (4.16)$$

we have

$$\kappa(T^*) T^{*\prime\prime} = \dot{T}^* \quad (4.16a)$$

and so

$$\begin{aligned} & \kappa_2 (T_2^* - T_1^*)'' - (T_2 - T_1)' + \\ & [\kappa_2 (T_2^*) - \kappa_1 (T_1^*)] T_1^{*''} = 0 \end{aligned} \quad (4.17)$$

whence (4.10) again follows, if $T^{*''} \geq 0$, under conditions similar to those earlier considered, both for the effective temperatures T^* and the actual temperatures T .

As has been mentioned, we shall assume (4.10) to hold; in particular, it does so before change of phase starts, or

$$t_{m1} \leq t_{m2} \quad (4.18)$$

In other words, (4.12) holds at $t = t_{m1}$ and at $t = t_{L1}$. It is obviously possible to devise functions $(\rho c)_2^*(T)$ or $K_1^*(T)$ sufficiently large, in the previous two examples respectively, in which (4.12) holds for all $t_{m1} < t < t_{L1}$

If (4.12) is to fail, there must exist a period where $s_1 < s_2$, and therefore a first time at which $s_1 = s_2$. Consider now a continuous sequence of functions bounded by $(\rho c)_2$ and $(\rho c)_2^*$, or by K_1 and K_1^* ; There must exist one such function for which the period just mentioned shrinks to a point $t = t^*$ at which

$$s_1 = s_2 \quad \text{and} \quad \dot{s}_1 = \dot{s}_2 \quad (4.16)$$

At that point (in either example, with T^* replacing T in the second),

$$T_1' = T_2' ; \quad \dot{T}_1 = \dot{T}_2 ; \quad T_1'' < T_2'' \quad (4.16a)$$

respectively from (4.4), from the differential form $T's + \dot{T} = 0$ of (4.5), and from either (4.13) or (4.16a). But then, near $x = s$ we have

$$T_{1,2} = T_m + T_{1,2}'(x - s) + T_{1,2}''[(x - s)^2/2] + \dots \quad (4.17)$$

or

$$T_1 - T_2 = (T_1'' - T_2'')[(x - s)^2/2] + \dots < 0 \quad (4.17a)$$

which contradicts (4.10); hence (4.12) must hold at all times. ●

5. MULTIDIMENSIONAL PROBLEMS

The major portion of the published work on moving boundaries is concerned with one-dimensional solutions, and indeed the mathematical complexities of the subject are such that many unsolved problems remain even with this restriction. Nevertheless, many practical problems are not, and cannot be, reasonably approximated by one-dimensional models. Many numerical methods are readily applied to multidimensional problems, but there appears to be considerable scope for analytical work, both approximate and exact, in this area.

It may be useful to recall the classification of multidimensional problems presented in [5.13], in which a distinction was made according to the type of motion characterizing the spread of the change-of-phase interface *along the surface*. Class I includes problems in which the change of phase starts simultaneously at all points of the surface. One-dimensional problems are of this class, as are all solutions in which the initial conditions are ignored or in which a steady-state is assumed to prevail. This class includes also the important two-dimensional solutions of [5.8,9] pertaining to dendritic growth and the work of [5.11] on the effect of imperfect mold-ingot contact.

Class II comprises problems in which change of phase starts simultaneously at all points in some portion of, but not the entire surface, and Class III those in which it starts at a point. In these two classes the intercept between the phase interface and the surface moves along the surface, at the same time as the interface itself progresses into the medium; as a consequence these problems are of a higher order of magnitude of difficulty than those of Class I (cf. [5.1,3,10] and the references cited there). Nevertheless, problems of Class III are some of the physically most important ones, particularly at the early stages of the various processes being examined under the general heading of moving boundaries, and should be attracting more attention on

on the part of researchers. Particularly lacking are works in which more than one phase is considered, and those with more complicated interface conditions. Among the latter, a particularly interesting effect often neglected is that of curvature on the interface equilibrium temperature [5.5,12] and on concentration [5.10], usually expressed by the Gibbs-Thompson relations

$$T_r = T_\infty - \frac{2\sigma T_m}{\ell r} ; C_r = C_\infty \left(1 + \frac{2\sigma V}{RT_r} \right) \quad (5.1)$$

or its generalizations [5.4,6,10]. In these equations the subscripts r and ∞ indicate equilibrium values corresponding to surfaces with radius of curvature r and plane respectively, σ is the surface energy, V the molar volume of the solute or crystal, and R the gas constant. If the coupled heat and mass transfer problem is analyzed, the effects on both temperature and concentration must of course be included simultaneously.

The extension of the standard analytical approximate techniques--heat balance, Biot's method and the like--into multidimensional problems has hardly been explored, and perhaps not found feasible. The same can be said of the construction of practical ways of devising bounds for the solution of such problems. Some promising approximate methods for multidimensional problems were presented in [4.7], based either on the concept of a thermal layer or on simplification in the heat equation.

Development of bounds on the basis of theorems comparing the behavior of one or two phase problems of the type considered for the one-dimensional case in [5.2,14,15] would be useful. This would particularly be true if the analytical solution were to be used as a starting solution for further numerical work (i.e., for very short times), since it may be conjectured that the one-dimensional conclusion that the starting solution is the same in these problems will carry over to multidimensional problems. If that is true, then one may obtain a valid starting solution for all cases from the obviously simpler one-phase case.

6. MACROSCOPIC AND MICROSCOPIC APPROACHES

The preceding discussion has been exclusively concerned with solutions based on the continuum approach; at the same time considerable research is being pursued on such topics as the structure of growing crystals, which clearly require analyses at the atomic level of the material (e.g., [6.4-6,12]). Consideration of both approaches and of their relation is evident in some works (e.g., [6.10]), and leads to a formulation more complete than has formally been considered in most available analyses. Such a formulation requires the temperatures T_S and T_L , and the concentrations C_S and C_L , in the solid and liquid respectively, to satisfy the heat and diffusion equations under interface conditions of the following type during solidification from the melt:

$$C_S = kC_L ; D_S \frac{\partial C_S}{\partial n} - D_L \frac{\partial C_L}{\partial n} = V_n (1 - k) C_L \quad (6.1)$$

$$T_S = T_L ; K_S \frac{\partial T_S}{\partial n} - K_L \frac{\partial T_L}{\partial n} = V_n \rho \ell \quad (6.2)$$

where V_n is the normal component of the interface velocity and k the partition coefficient. The usual way of determining V_n [6.6] simplifies the appropriate expression by means of fact that the temperatures at the interface are constant and equal to the melting temperature. It has already been seen, however, that this condition is not strictly correct for curved interfaces: the equilibrium temperature there must satisfy a thermodynamic relation of the form

$$T_r = T_E(C_L) - \frac{\sigma}{S} \quad (6.3)$$

where S is the entropy of melting and where T_E is the equilibrium freezing temperature of a plane surface at concentration C_L , the functional relation between T_E and C_L being determined by the liquidus line of the phase diagram for the material in question. The additional relation required to complete the formulation is

then the so-called kinetic condition, which relates the velocity V of freezing to the difference between the interface and the equilibrium temperatures, or

$$V = f(T - T_r) \quad (6.4)$$

The form of this relation depends on the details of the atomic mechanism of crystal formation [5.5] and is usually taken to be linear (e.g., [5.12], [6.10], or

$$V = \mu(T - T_r) \quad (6.5)$$

where μ is the atomic kinetic coefficient.

Solution of the complete problem outlined above has not yet been attempted, and it should in fact be noted that it is not expected to be unique on physical grounds. In other words, several interface shapes are thought to exist which satisfy the formulation given (the initial and fixed-boundary conditions must of course be added); the actual one is the one which corresponds to the maximum velocity of growth, or [6.10]

$$\delta V_{\text{ext}} < 0 \quad (6.6)$$

where V_{ext} is the velocity of the extremities of the formed crystal, and δ indicates the difference in the velocities corresponding to the actual and any other interface morphologies.

It would seem desirable to perform some careful studies of uniqueness of solution of the stated problem; in particular, such studies might shed some light on the types of functional relations (6.4) which are theoretically admissible, thus simplifying (or at least providing some direction for) the task of the experimentalists. Of course, if the simple one-dimensional problem is considered, the curvature dependence and thus most of the foregoing difficulties are absent; uniqueness in this case has been considered [6.2]. In that work, the coupled heat and mass transfer problem is considered; similarity solutions for this problem have been examined by a number of authors (e.g., 6.8-10), and a more general case has been discussed in the literature [6.1].

One additional difficulty should be added to the foregoing formulation, namely that required to describe the generally anisotropic character of the formed crystals [5.5, 6.7]. This effect has not been considered before, but it can be safely expected to complicate matters a great deal, because the orientation of the crystals is not known a priori, but must be determined by a condition similar to (6.6).

It might be worth while to consider here the modification which anisotropy would introduce in the known exact short-time solution for the half-space $z > 0$ under arbitrary heating rates [5.5]; this is easy to do if one assumes z to be a principal direction of the conductivity tensor. Consider for simplicity the two-dimensional special case of the problem [6.3] and let the x and z directions be principal with conductivities K_1 and K_3 respectively. It is then easy to show that the transformation

$$x' = x\sqrt{K_3/K_1} \quad ; \quad z' = z \quad ; \quad s' = s \quad (6.7)$$

reduces the entire problem to its original form, since the interface heat-balance condition

$$- \left[K_3 + K_1 \left(\frac{\partial s}{\partial x} \right)^2 \right] \frac{\partial T}{\partial x} = Q_o(x, t) - \rho l \frac{\partial s}{\partial t} \quad (6.8a)$$

becomes

$$- K_3 \left[1 + \left(\frac{\partial s'}{\partial x'} \right)^2 \right] \frac{\partial T}{\partial z'} = Q_o'(x', t) - \rho l \frac{\partial s'}{\partial t} \quad (6.8b)$$

Hence the solution is obtained simply by replacing the quantity

$C_{2x} = (\partial^2 T / \partial x^2)$ by $C_{2x}' = - (\partial^2 T / \partial x'^2)$, evaluated at the instant of start of melting. Thus

$$C_{2x}' = C_{2x} (K_1/K_3) \quad (6.9)$$

Thus the maximum penetration is

$$\frac{s_{\max}(t)}{2\sqrt{\mu_3 t_m}} = \frac{4MC_1}{3\pi} \left[1 + \frac{1}{8} \left(\frac{C_{2x} K_1}{C_1 K_3} \right) + \dots \right] \tau^{3/2} + \dots \quad (6.10a)$$

and the maximum surface extent of melting is

$$\frac{x_o(\tau)}{2\sqrt{\kappa_3 t_m}} = \sqrt{\frac{2C_1\tau}{C_{2x}}} \left[1 - \frac{1}{8} \left(\frac{C_{2x}K_1}{C_1K_3} \right) + \frac{11}{128} \left(\frac{C_{2x}K_1}{C_1K_3} \right)^2 + \dots \right] + \dots \quad (6.11)$$

where

$$M = \frac{\sqrt{\pi c} T_m}{2\ell} ; \quad \kappa_3 = \frac{K_3}{\rho c} ; \quad \tau = \frac{t - t_m}{t_m} ; \quad C_1 = \left(\frac{\partial T}{\partial t} \right)_{\substack{t=t_m \\ x=z=0}} \quad (6.12)$$

It is of interest to calculate the ratio of the two distances given in eqs. (6.10), or

$$\frac{s_{\max}(\tau)}{x_o(\tau)} = \frac{4M\sqrt{C_1C_{2x}}}{3\pi\sqrt{2}} \tau \left[1 + \frac{1}{4} \left(\frac{C_{2x}K_1}{C_1K_3} \right) + \dots \right] + \dots \quad (6.13)$$

This appears at first sight to predict that the penetration will be greater (relative to the surface spread) the greater the ratio of conductivities (K_1/K_3) in the directions x_o and s respectively. This would be intuitively disturbing; in reality, a true assessment of the effect of variations in conductivity, is obtained only if the accompanying variation in the pre-melting solution is included. A meaningful comparison on the basis of eq. (6.13) therefore results only if it is assumed that the temperature distribution at the instant of incipient melting is unchanged. Thus, consider two cases corresponding to conductivities K_1' and K_1'' (with K_3 unaltered for conceptual simplicity); then it is necessary that

$$K_1' C_{2x}' = K_1'' C_{2x}'' \quad (6.13)$$

or that C_{2x} be inversely proportional to K_1 . Substitution in (6.13) will then give the more acceptable result that s_{\max}/x_o decreases with increasing values of (K_1/K_3) .

7. CONCLUDING REMARKS

The foregoing discussion has attempted to indicate--although in an admittedly sketchy and far from comprehensive manner--a number of problems connected with moving boundaries which have not received great emphasis during this conference. In the presentation an effort has been made to identify areas in which further cooperation between mathematicians and engineers is most desirable and can most readily be expected to bear fruit. It is hoped that this brief exposition will be helpful in furthering that cooperation.

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