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The steady-state solidification scenario of ternary systems: Exact analytical solution of nonlinear model

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ABSTRACT

A mathematical model describing the steady-state solidification of ternary systems with mushy layers (primary and cotectic) is formulated: solidification along a liquidus surface is characterized by a primary mushy layer, and solidification along a cotectic line is characterized by a secondary (cotectic) mushy layer. Exact analytical solutions of the model under consideration are found in a parametric form (thicknesses of mushy layers, growth rate of their boundaries, temperature and composition fields, solid fractions are determined in an explicit form). The velocity of solidification is completely determined by temperature gradients in the solid and liquid phases. This velocity coincides with similar expressions describing binary melt solidification with a planar front or a mushy layer. It is shown that the liquid composition of the main component decreases in the cotectic and primary layers, whereas the second (cotectic) composition increases in the cotectic layer, attains a maximum point and decreases in the primary layer.

1. Introduction

The constitutional supercooling arising under certain circumstances ahead of the planar phase transition interface [1] and its morphological instability [2–4] cause a system of elements of the solid phase in the form of dendrites, columnar and uniaxial crystals to appear in the supercooled melt [5–7]. The development of this system reduces the supercooling and leads to formation of a new stable solidification mode characterized by the presence of a mushy (two-phase) layer that separates the solid phase and the melt. The study of relationships governing solidification in the presence of a mushy region is rather complicated. This happens because it is necessary to investigate the interaction of nonlinear heat and mass transfer in the case of moving phase transition boundaries. For binary alloys, an exact analytical description of the steady-state solidification scenario with a mushy layer has been given for the first time in Refs. [8–10] by means of a new approach. This method of integration of the nonlinear heat and mass transfer equations connected with the transition to a new variable–solid fraction in the mushy layer will be developed in the present study for ternary systems. Hereafter, the theory of Refs. [8–10] has been further refined in order to take into account thermodiffusion and temperature-dependent diffusivity effects [11,12] as well as the influence of weak convection [13] and nonlinear liquidus equation [14]. Also note that approximate analytical approaches have been developed for the description of the self-similar [15,16] and unsteady-state [17,18] solidification regimes of binary systems.

It is well-known that a wide variety of processes met in geophysics and metallurgy involve the solidification of multicomponent melts (e.g., solidification of magmas [19], casting of metals [20] and crystal growth processes [21]). Directional and bulk solidification of different components of the system leads to variations in both the composition and temperature gradients near the phase transition interface. As in the case of binary melts, this gives rise to the formation of one (in the case of a binary melt) or more (in the case of a multicomponent melt) mushy layers between the pure solid and liquid phases [22]. Let us emphasize that as the number of components in a system increases, the range of possible behaviors increases as well [23] (e.g., compare dynamics of concentration profiles in binary and ternary melts [4,24]). The complexity of nonlinear interactions between the heat and mass transfer processes in mushy layers increases with the number of components of a multicomponent melt. Hence, a ternary melt or solution of three chemically distinct components represents a test system showing the main features of a multicomponent solidification in comparison with a binary system.

A recent investigation of the crystallization of a three-component solution begun in Ref. [25]. In that study, a three-component system was cooled from below and all convection was suppressed due to the fact that the buoyancy of the fluid released on solidification always increased (a possible influence of convection on solidification of ternary systems has been discussed in

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Nomenclature

B and C liquid compositions of components B and C
B_E and C_E liquid compositions of components B and C in the ternary eutectic point
B^C and C^C cotectic compositions of components B and C
B_∞ and C_∞ initial values of B and C in the liquid phase far from the primary mushy layer
D_B and D_C solute diffusivities of components B and C
G_L temperature gradient in the liquid phase
G_S temperature gradient in the solid phase
h_C cotectic mushy layer–primary mushy layer boundary
h_E solid phase–cotectic mushy layer boundary
h_P primary mushy layer–liquid-phase boundary
k_L thermal conductivity in the liquid
k_S thermal conductivity in the solid
$\bar{k} = k_L\chi + k_S(1-\chi)$ thermal conductivity in a mushy layer
L_V latent heat of solidification
m_B and m_C liquidus slopes corresponding to components B and C
m_B^C and m_C^C cotectic slopes corresponding to components B and C
t time
T temperature
T^L liquidus temperature
T_E temperature in the ternary eutectic point
T_E^{AB} temperature in the binary eutectic point
T_M phase transition temperature of pure component A
V solidification velocity
x and z spatial coordinates

Greek symbols
$\delta = \delta_C + \delta_P$ thickness of the phase transition layer
δ_C thickness of the cotectic mushy layer
δ_P thickness of the primary mushy layer
κ temperature diffusivity coefficient
φ_A, φ_B and φ_C solid fractions of components A, B and C
φ_{ACP}^+ solid fraction φ_A on the right side of the cotectic layer–primary layer interface
φ_{ACP}^- solid fraction φ_A on the left side of the cotectic layer–primary layer interface
φ_{APL}^- solid fraction φ_A on the left side of the primary layer–liquid interface
φ_{ASC}^+ solid fraction φ_A on the right side of the solid phase–cotectic layer interface
φ_{BSC}^+ solid fraction φ_B on the right side of the solid phase–cotectic layer interface
φ_{BCP}^- solid fraction φ_B on the left side of the cotectic layer–primary layer interface
φ_{BSC}^+ solid fraction φ_B on the right side of the solid phase–cotectic layer interface
φ_{BSC}^- solid fraction φ_B on the left side of the solid phase–cotectic layer interface
φ_{CSC}^+ solid fraction φ_C on the right side of the solid phase–cotectic layer interface
φ_{CSC}^- solid fraction φ_C on the left side of the solid phase–cotectic layer interface
χ liquid fraction
χ_{CP}^- liquid fraction on the left side of the cotectic layer–primary layer interface
χ_{SC}^+ liquid fraction on the right side of the solid phase–cotectic layer interface

Ref. [26]). A mathematical model for non-convecting diffusion-controlled solidification of a ternary solution cooled from below has been described and discussed in Ref. [27]. This model contains the heat and mass transfer equations in solid, liquid phases and two mushy layers (primary and cotectic), where all transfer coefficients are dependent on the liquid fraction. These four regions (solid, cotectic, primary and liquid) are connected by the corresponding boundary conditions imposed at three moving boundaries. Because an analytical technique for the solution of this highly nonlinear problem of unsteady-state solidification with two mushy layers does not exist, analytical solutions have been obtained for the case of zero solute diffusion and zero latent heat [27]. More general solutions of the model developed in [27] have been constructed for the self-similar solidification scenario on the basis of Scheil equations for the impurity distribution in mushy layers (this theory is developed in Refs. [28,29] and [30] for the linear and nonlinear liquidus and cotectic equations).

The present study is devoted to the theory of the steady-state solidification of a ternary system on the basis of model equations obtained in Ref. [27]. The steady-state growth, in which the interfaces are supposed to advance at a prescribed constant velocity, corresponds to many processes met in metallurgy and geophysics (e.g. the crystal pulling (Czochralski growth) and the freezing of a thick sea ice). Below we discuss how to integrate highly nonlinear heat and mass transfer problem in the presence of three moving boundaries of the phase transition when crystallization occurs with a constant velocity. The outline of this paper is as follows: Section 2 represents a theoretical description of the steady-state solidification scenario; exact analytical solutions are given in Section 3; results are reported and discussed in Section 4.

2. The steady-state solidification model of a ternary system

Let us consider a unidirectional solidification process of a ternary system in thermodynamic equilibrium illustrated in Fig. 1. A sketch of the ternary phase diagram under consideration in the spirit of Ref. [27] is shown in Fig. 2. We denote the liquid compositions of components A, B and C by A, B and C ($A + B + C = 1$). Each of the three sides of the phase diagram describes the binary phase diagram (the binary eutectic point E_{AB} has temperature T_E^{AB} and composition B_E^{AB} of the B component). Three liquidus surfaces are formed by the binary liquidus curves along each of the three sides of the ternary phase diagram and cotectic curves extend from the binary eutectic points into the interior of the diagram (these curves are the boundaries of the liquidus surfaces). The ternary eutectic point E is located at the intersection of these curves, where the temperature is T_E and the compositions are A_E, B_E and C_E. Let a liquid-phase ternary alloy be at the point P on a liquidus surface. After cooling, component A begins to solidify out, and the components B and C are rejected into the liquid. The latter leads the system to point S on the cotectic curve. At this time, the system has a single phase transition region of the A component–primary mushy layer. When the cotectic curve is reached (point S), solidification continues and two components A and B undergo transformations in the solid state. At this time, the system goes from point S to point E along the cotectic curve. Here we have two phase transition regions–primary and cotectic mushy layers. Thus, the curves P–S–E and S–E respectively correspond to the primary and primary–cotectic mushy layer solidification scenarios. Once the eutectic point E is reached, the remaining liquid solidifies to form a eutectic solid layer composed of solid A, B and C.