

CALCULATION OF DENDRITIC LIQUATION (MICROSEGREGATION) IN A SOLIDIFYING STEEL INGOT WITH ALLOWANCE FOR IONIC MIGRATION

Yu. A. Samoilovich,^a V. I. Timoshpol'skii,^b
and S. M. Kabishov^b

UDC 621.746

Based on the mathematical model, the analytical solution of the problem of formation of dendritic liquation for different configurations of a dendritic cell has been obtained; the use of this solution makes it possible to optimize melting processes from the viewpoint of the chemical inhomogeneity of ingots and billets.

Keywords: *crystallization of steel, dendritic liquation, diffusion, mathematical modeling, electric field, ionic migration.*

Introduction. Crystallization of steel consisting of the basic component (iron), alloying additions (carbon, silicon, manganese, and others), and inevitable impurities (sulfur, phosphorus, and others) is accompanied by the enrichment of the liquid phase with low-melting components. The process of separation diffusion occurs at the phase boundary, i.e., on the surface of growing dendrites, and gives rise to chemical inhomogeneity within each dendritic cell. In the literature [1–3], it is the practice to call this phenomenon dendritic liquation, although the term "microsegregation" is admittedly more correct. The prefix "micro-" is introduced to distinguish between chemical inhomogeneity within one dendritic cell and the inhomogeneity of the chemical composition of steel on individual portions of an ingot or a continuously cast slab (on the axis of the ingot or in its surface layer) — the so-called "macrosegregation."

Review of the Methods of Modelling of the Process of Dendritic Liquation. When dendritic liquation is theoretically described [1–6], emphasis is given to the process of separation of the alloy components at the phase boundary (it is allowed for by the equilibrium distribution coefficient k_0) and of diffusion of the mixture in the liquid phase (it is allowed for by the coefficient D_{liq}). In the present work, in formulating the problem, we take into account the process of ionic migration in a solidifying melt under the action of certain external factors (e.g., when an electric current is passed through the melt). The practical significance of allowance for the ionic migration in a melt can be illustrated with two examples. High-purity metals and semiconductors are used in many currently available technologies. Impurities are cleaned from the metals in zone melting in an electric field [7, 8]. In so doing, they seek to ensure the maximum separation of the impurities from the alloy's basic component, which is attained by selection of the corresponding polarity and density of the current. The separation is the most intense in the case of selection of such a polarity of the current where the direction of migration of impurity ions is coincident with the direction of motion of the phase boundary. In this case the flow of the low-melting impurity from the phase boundary grows and they can separate the alloy into the basic component and the low-melting impurity to the maximum possible degree in combination with intense mixing of the liquid phase. The other example belongs to attempts to diminish the chemical inhomogeneity of steel ingots by passing a constant or pulsating electric current [9–12].

It has been shown in [10] that the isolation of sulfur in a solidifying steel ingot is difficult for the inverse polarity of the electric current (the solid phase is the cathode); the sulfide inclusions in the solidified ingot are of high dispersity. Thus, in this case they must use such a polarity of the electric field for which the motion of migrating ions is in opposition to the motion of the phase boundary.

As is shown below, in formulating the corresponding problem of diffusion of impurities in a solidifying melt, we allow for the ionic migration by introduction of the parameter $V_m = u_j\rho$; the polarity of the electric current is determined by the selection of the corresponding sign of V_m .

^a"Platan" Science and Production Enterprise, Ekaterinburg, Russia; ^bBelarusian National Technical University, 65 Nezavisimost' Ave., Minsk, 220013, Belarus. Translated from *Inzhenerno-Fizicheskii Zhurnal*, Vol. 82, No. 1, pp. 98–105, January–February, 2009. Original article submitted December 7, 2007.

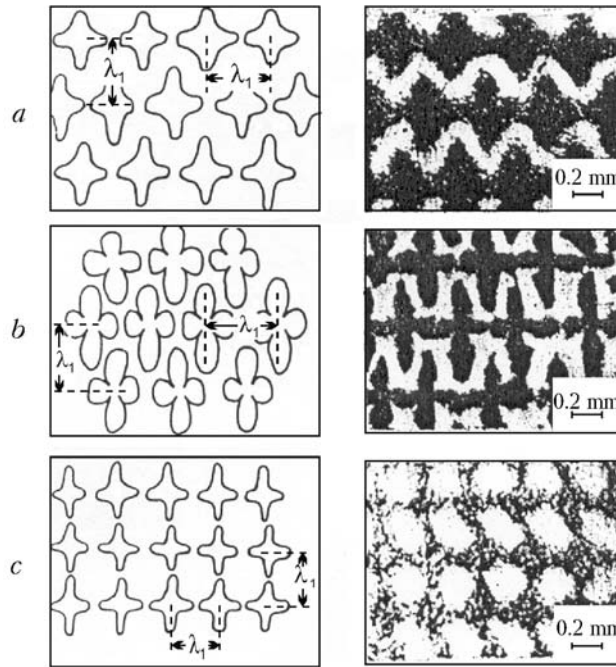


Fig. 1. Morphology of columnar crystallites in steel containing 0.5% C and 1.1% Mn for the rate of growth $R = 1$ mm/min (a), 4.17 mm/min (b), and 2 mm/min (c) according to the data of [13].

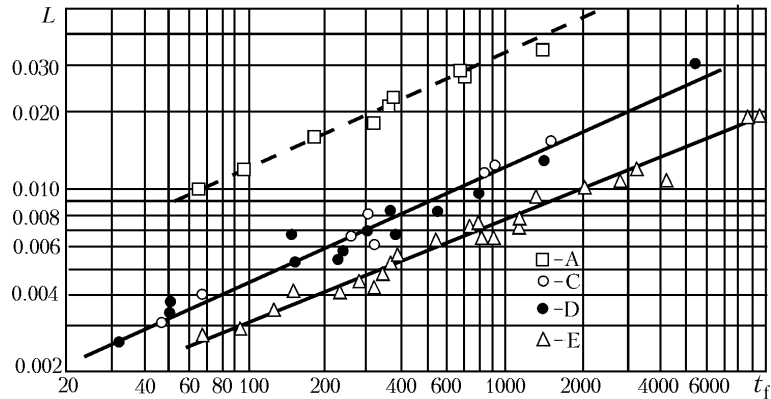


Fig. 2. Distances between secondary dendritic branches L vs. duration of local solidification t_f for different steels according to the data of [13]: steel A: 0.59% C and 1.1% Mn; steel C: 0.64% C and 27.7% Mn; steel D: 0.63% C and 10% Mn; steel E: 0.68% C and 28.3% Mn. L , m; t_f , sec.

In calculating dendritic liquation in solidifying ingots, we must allow for the dimensions and configuration of a dendritic cell within which diffusion processes occurs. This makes it necessary to determine the structural characteristics (morphology) of dendritic crystallites forming a two-phase zone in the solidifying ingot. The crystallites growing in solidifying steel ingots are characterized by the presence of the main trunks (axes of first order) and secondary dendritic branches growing at a right angle to the axes of first order. The regularities of dendritic growth are studied by special laboratory tests during which such indices of the process of solidification as the rate of growth of dendritic crystallites R and the total duration of local solidification of steel t_f are adjusted within required limits. Such investigations can be exemplified by [13] where the results of investigations of the morphology of dendritic crystallites and the distance between the axes of second order L as a function of the duration of local solidification t_f have been presented (see Figs. 1 and 2).

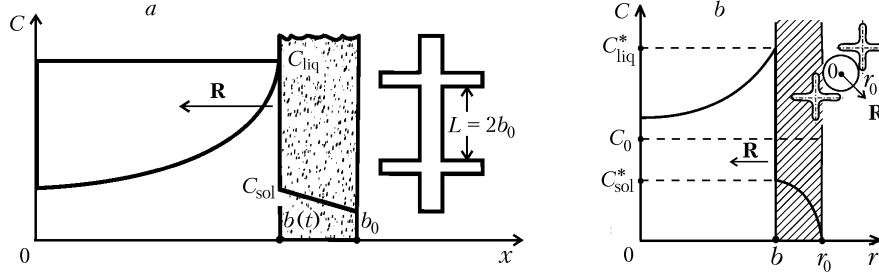


Fig. 3. Diagram of distribution of the impurity concentration within the dendritic cell, when the liquid-phase layer is assigned in the form of a plane layer (a) or a cylindrical layer (b).

In theoretical analysis of dendritic liquation, Schwerdtfeger [14] took the melt volume between two neighboring branches of second order of thickness L as the dendritic cell, as is shown in Fig. 3a; the phase boundary moves with velocity \mathbf{R} in opposition to the x axis.

At the same time, it may be inferred from consideration of the dendritic morphology of steel (Fig. 1) that low-melting impurities can collect between the dendritic branches of first order. The necessity of refining the parameters of the dendritic cell selected for subsequent theoretical analysis is determined by the fact that the interaxial distances differ by many times for the axes of first and second orders.

Analytical Solution of the Problem of Formation of Dendritic Liquation. Solution is carried out for two different configurations of a dendritic cell in the form of a plane layer of thickness $L = 2b_0$ (between secondary branches of the dendrites) and a cylindrical layer of radius r_0 (between primary trunks of the dendritic crystallites).

Diagram No. 1 for the Dendritic Cell in the Form of a Plane Layer of Thickness L . We consider diffusion processes in a plane layer of a melt which is located between two adjacent plane dendritic branches of second order. Assuming the symmetry of the concentration field of the impurity, we restrict ourselves to an analysis of the process of diffusion in the plane layer whose thickness decreases with time with rate $\mathbf{R} = db/dt$ (the vector of the rate \mathbf{R} is directed toward decreasing values of the coordinate x , as is shown in Fig. 3a).

A mathematical formulation of the problem includes the equation of nonstationary diffusion

$$\frac{\partial C}{\partial t} = D_{\text{liq}} \frac{\partial^2 C}{\partial x^2} \quad (1)$$

and the boundary conditions

$$\frac{\partial C}{\partial x} = 0 \quad \text{for } x = 0, \quad (2)$$

$$D_{\text{liq}} \frac{\partial C}{\partial x} = V_m C_{\text{liq}}^* - (C_{\text{liq}}^* - C_{\text{sol}}^*) \frac{db}{dt} \quad \text{for } x = b(t). \quad (3)$$

Introducing the equilibrium distribution coefficients $k_0 = C_{\text{sol}}^*/C_{\text{liq}}^*$, we represent boundary condition (3) as

$$D_{\text{liq}} \frac{\partial C}{\partial x} = V_m C_{\text{liq}}^* - (1 - k_0) C_{\text{liq}}^* \frac{db}{dt} \quad \text{for } x = b(t). \quad (4)$$

Integrating both sides of Eq. (4) along the coordinate x between the limits from zero to the variable $b(t)$ and using boundary conditions (2) and (3), we arrive at the ordinary differential equation

$$\frac{d}{dt} \int_0^b C dx = V_m C_{\text{liq}}^* + k_0 C_{\text{liq}}^* \frac{db}{dt}, \quad (5)$$

whose solution is obtained, when we assign the initial impurity distribution in the melt

$$C(x, 0) = C_0(x). \quad (6)$$

Further analysis is made in a quasistationary approximation, when the intensity of the external action causing ionic migration in the melt is assumed to be constant with time. If the external action is achieved by passing an electric current through a solidifying melt, the quasistationary approximation is reduced to assignment of a time-constant current density and accordingly a constant value of the parameter V_m .

We are able to find a simplified expression for the impurity-concentration distribution over the cross section of the liquid phase for the quasistationary approximation. As has been shown in [15], in this case the total duration of the process of solidification of the dendritic cell is subdivided into two periods: the initial period (t_0) and the regular one (t_r); for the latter, the impurity-concentration distribution over the cross section of the liquid phase can be represented by the parabolic equation

$$C(x, t) = a_0 + a_2 x^2. \quad (7)$$

The distribution (7) yields the relation

$$D \left. \frac{\partial C}{\partial x} \right|_{x=b} = 2a_2 D b(t). \quad (8)$$

Setting the right-hand sides of relations (4) and (8), equal to each other, we obtain an expression for the coefficient a_2 :

$$a_2 = \frac{V_m C_{\text{liq}}^*}{2bD} - \frac{1-k_0}{2D} C_{\text{liq}}^* \frac{1}{b} \frac{db}{dt}. \quad (9)$$

We express the coefficient a_0 from the distribution (7) with account for relation (9) and the formula $C(b, t) = C_{\text{liq}}^*$:

$$a_0 = C_{\text{liq}}^* + \frac{1-k_0}{2D} b \frac{db}{dt} C_{\text{liq}}^* - b \frac{V_m C_{\text{liq}}^*}{2D}. \quad (10)$$

We simplify the formation of the problem, using the assumption that the total duration of solidification for the local portion of the two-phase zone of the solidifying ingots t_f is known; the thickening of the dendritic branch with time obeys the linear law

$$b(t) = b_0 \left(1 - \frac{t}{t_f} \right). \quad (11)$$

We introduce the dimensionless variables

$$K = \frac{1-k_0}{3}, \quad P = \frac{b_0 \mathbf{R}}{D_{\text{liq}}}, \quad V = \frac{V_m}{\mathbf{R}}, \quad F = \frac{t}{t_f}, \quad (12)$$

where the rate of thickening of dendritic branches is $\mathbf{R} = -db/dt = b_0/t_f$.

Substituting the distribution (7), which is true for the regular regime under the integration sign in Eq. (5), and using the dimensionless numbers (12), we obtain an ordinary differential equation for the variable C_{liq}^* :

$$\frac{dC_{\text{liq}}^*}{dF} = G(t) C_{\text{liq}}^*, \quad (13)$$

where the time function is

$$G(t) = \frac{[3 - 2P(1 - F)] \left(K + \frac{V}{3} \right)}{1 - F - P(1 - F)^2 \left(K + \frac{V}{3} \right)}. \quad (14)$$

Integrating Eq. (13) with the initial condition $C_{\text{liq}}^* = C_0$, we obtain the time change in the maximum concentration of the impurity at the phase boundary $C_{\text{liq}}^*(t)$. Knowing the function C_{liq}^* , we can determine, using (7), the time change in the impurity concentration average over the cross section of the liquid phase

$$\bar{C} = C_{\text{liq}}^* \left[1 - P(1 - F) \left(K + \frac{V}{3} \right) \right] \quad (15)$$

and the difference in the impurity concentration over the liquid-phase cross section

$$\Delta C = 0.5 C_{\text{liq}}^* P(1 - F)(V + 3K). \quad (16)$$

Diagram No. 2 for the Dendritic Cell in the Form of a Cylindrical (Circular) Layer of Radius r_0 . A mathematical formulation of the problem for a cylindrical layer is determined by the system of equations

$$\frac{\partial C}{\partial t} = D_{\text{liq}} \left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r} \frac{\partial C}{\partial r} \right), \quad (17)$$

$$\frac{\partial C}{\partial r} = 0 \quad \text{for } r = 0, \quad (18)$$

$$D_{\text{liq}} \frac{\partial C}{\partial r} = V_m C_{\text{liq}}^* - (1 - k_0) C_{\text{liq}}^* \frac{db}{dt} \quad \text{for } r = b(t). \quad (19)$$

Integrating both sides of Eq. (17) along the radius r between the limits from zero to $b(t)$, we obtain

$$\frac{d}{dt} \int_0^b C dr - C(b, t) \frac{db}{dt} = D_{\text{liq}} \left. \frac{\partial C}{\partial r} \right|_{r=b} - D_{\text{liq}} \int_0^b \frac{1}{r} \frac{\partial C}{\partial r} dr. \quad (20)$$

We take $C(r, t) = a_0 + a_2 r^2$, where the coefficients a_0 and a_2 are determined, as previously, by relations (9) and (10), and substitute the resulting distribution of the impurity concentration into the balance equation (20). After simple manipulations, we reduce Eq. (20) to the form

$$\frac{dC_{\text{liq}}^*}{dF} = H(t) C_{\text{liq}}^*, \quad (21)$$

where the function $H(t)$ is determined by the relation

$$H(t) = \frac{3K + V - P(1 - F) \left(K + \frac{V}{3} \right)}{(1 - F) [1 - P(1 - F) (K + V)]} \quad (22)$$

on retention of the notation of (12) for dimensionless variables K , V , P , and F .

The differential equation (21) is integrated, when the initial condition

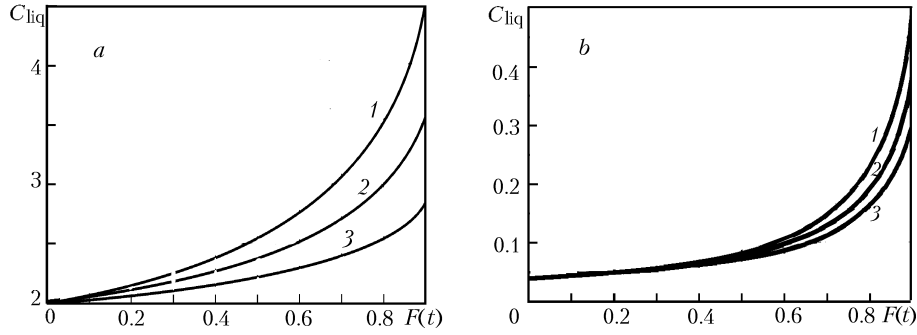


Fig. 4. Distribution of manganese (a) and sulfur (b) over the cross section of the dendritic cell by the instant of solidification of 90% of the molten steel containing 2.0% Mn and 0.04% S on assignment of different k_0 and D_{liq} (from the data of [14, 16]) and for several values of the rate of ionic migration: 1) $V_m = +0.1$, 2) 0, and 3) -0.1 (a) $k_0 = 0.75$ and $D_{\text{liq}} = 3.3 \cdot 10^{-5}$ cm²/sec; b) $k_0 = 0.015$ and $D_{\text{liq}} = 3.8 \cdot 10^{-5}$ cm²/sec). C_{liq} , %.

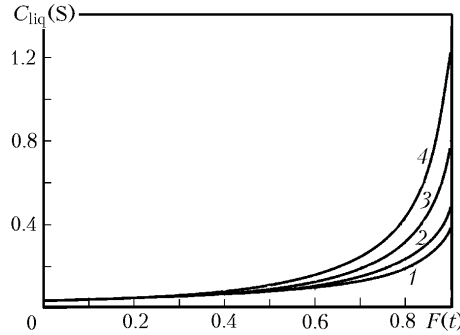


Fig. 5. Influence of the rate of ionic migration V_m on the distribution of the sulfur concentration $C_{\text{liq}}(\text{S})$ along the radius of the dendritic cell on assignment of $C_0(\text{S}) = 0.04\%$, $k_0 = 0.015$, and $D_{\text{liq}}(\text{S}) = 3.8 \cdot 10^{-4}$ cm²/sec: 1) $V_m = 0$, 2) 0.1, 3) 0.3, and 4) 0.5. $C_{\text{liq}}(\text{S})$, %.

$$C_{\text{liq}}^* = C_0 \text{ for } F = 0$$

is specified. We give results of integration of Eqs. (13) and (21) for dendritic cells of plane and cylindrical shapes as a time change in the concentration of two elements: manganese and sulfur.

Figure 4a gives the change in the concentration of manganese in a plane molten layer between two dendritic branches of thickness 0.006 cm as a function of the relative time F for three values of the relative rate of ionic migration: $V = 0$, -0.1 , and $+0.1$. The coefficient of diffusion of manganese in the iron melt was taken to be $3.3 \cdot 10^{-5}$ cm²/sec according to the data of [14, 16]; the equilibrium distribution coefficient was $k_0 = 0.75$. The initial concentration of manganese was 2.0%; the local solidification time was 120 sec. It follows from the figure that the maximum concentration of manganese at the phase boundary grows from 2.0 to 3.52% at $t = 0.9t_f$ in the absence of ionic migration. In the case of the migration of manganese ions in the direction of the solid crust (for $V > 0$), the maximum concentration C_{liq}^* increases by nearly 28% at $t = 0.9t_f$; in reverse ionic motion (from the phase boundary toward the axis of symmetry of the cell), the manganese concentration diminishes by the same degree.

Figure 4b gives the change in the concentration of sulfur in a plane layer of thickness 0.006 cm from an initial concentration of 0.04% with growth in the t/t_f ratio from zero to 0.9, when the characteristic properties of the solution of sulfur in the iron melt ($D_{\text{liq}} = 3.9 \cdot 10^{-5}$ cm²/sec, according to the data of [14, 16]) are assigned. As is seen from the figure, the concentration of sulfur grows from 0.04 to 0.38% i.e., nearly 10 times, with time in the absence of ionic migration, which is a consequence of the low value of the equilibrium distribution coefficient.

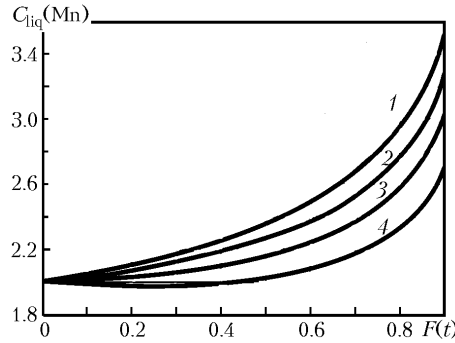


Fig. 6. Influence of the initial thickness of a dendritic branch b_0 and the solidification rate R on the distribution of the manganese concentration along the radius of the cylindrical cell on assignment of $C_0(\text{Mn}) = 2.0\%$, $k_0 = 0.75$, and $D_{\text{liq}}(\text{Mn}) = 3.3 \cdot 10^{-4} \text{ cm}^2/\text{sec}$: 1) $b_0 = 0.030$ and $R = 2.5 \cdot 10^{-4}$, 2) 0.075 and $6.3 \cdot 10^{-4}$, 3) 0.100 and $8.3 \cdot 10^{-4}$, and 4) 0.125 cm and $10.4 \cdot 10^{-4} \text{ cm}/\text{sec}$. $C_{\text{liq}}(\text{Mn})$, %.

The migration of sulfur ions in the positive direction (when $V = 0.1$ is assigned) is accompanied by a growth in the impurity concentration at the phase boundary from 0.38 to 0.49%, i.e., of nearly 28%, as in the case of manganese (see above); in ionic migration in the opposite direction, we observe a reduction in the maximum concentration of sulfur to the same degree.

The character of growth in the concentration at the phase boundary with the rate of sulfur-ion migration is exemplified by the solidification of a cylindrically shaped dendritic cell with a change in the parameter V_m from zero to 0.5, when $b_0 = 0.003$ cm and $t_f = 120$ sec are assigned (see Fig. 5).

As has been noted above, the cell of a cylindrical shape, according to the diagram in Fig. 3b, corresponds to the formation of dendritic liquation in the space between the axes of first order to a greater extent than the plane cell. The dimension of the cell ($2r_0$) may be one or two orders of magnitude larger than the distance L between the dendritic branches of second order.

Figure 6 shows the influence of the dimension of the cylindrical cell (within 0.03–0.125 cm) on the character of change in the dendritic liquation of manganese (when $D_{\text{liq}} = 3.3 \cdot 10^{-5} \text{ cm}^2/\text{sec}$, $k_0 = 0.75$, and $t_f = 120$ sec are assigned.)

The degree of dendritic liquation decreases with increase in the dimension of the cell (or in the rate of thickening of dendritic trunks of first order), which is attributed to the reduction in the intensity of diffusion processes in the dendritic cell).

CONCLUSIONS

1. We have posed and mathematically formulated the problem on the development of dendritic liquation in the space between the axes of first and second orders with allowance for ionic migration under the influence of the external force, e.g., when an electric current is passed through a solidifying melt.

2. We have obtained the analytical solution of the problem for a dendritic cell of the two simplest configurations under the assumption of quasistationarity of external actions (when the electric current of constant density is used). The local melt-solidification time determined during special experiments or on the basis of numerical solution of the problem on the dynamics of solidification of an ingot is assumed to be known.

3. The proposed solutions of the problem make it possible to evaluate the degree of development of the dendritic liquation (microsegregation) of the impurity in the steel ingot in the presence of external actions contributing to the migration of ions of the alloy components or the impurities and may work for optimization of the regimes of cleaning of metals by the method of zone melting or with an electric current to reduce the level of chemical inhomogeneity of commercial steel ingots.

NOTATION

a_i , dimensionless coefficients; b_0 and b , initial and running thickness of a dendritic branch, m; C , concentration of the impurity, %; D , coefficient of diffusion of the impurity in the liquid phase, m^2/sec ; $H(t)$, dimensionless variable; j , electric-current density, A/m^2 ; k_0 , equilibrium coefficient of distribution of the impurity; L , thickness of the plane layer between secondary dendritic branches, m; \mathbf{R} , rate of growth of dendritic crystallites, m/sec ; r , radius of the cylindrical layer, m; t , running time, sec; t_f , total duration of local solidification of steel, sec; u , ionic mobility, $\text{V}/(\text{m}^2 \cdot \text{sec})$; V_m , parameter allowing for ionic migration; x , running coordinate, m; ρ , electrical resistance of the melt, $\Omega \cdot \text{m}$. Subscripts: 0, initial; f, final; r, regular; *, equilibrium; liq, liquid phase; sol, solid phase; m, migration.

REFERENCES

1. I. N. Golikov, *Dendritic Liquefaction in Steel* [in Russian], Metallurgizdat, Moscow (1963).
2. I. I. Novikov and V. S. Zolotarevskii, *Dendritic Liquefaction in Alloys* [in Russian], Nauka, Moscow (1966).
3. I. N. Golikov and S. B. Maslennikov, *Dendrite Liquefaction in Steels and Alloys* [in Russian], Metallurgiya, Moscow (1977).
4. W. A. Tiller., K. A. Jackson., J. W. Rutter, and B. Chalmers, *Acta Metallurgica*, **1**, 428 (1953).
5. V. G. Smith, W. A. Tiller, and J. W. Rutter, *Canad. J. Phys.*, **33**, 723 (1955).
6. A. I. Landau, *Growth of Crystals* [in Russian], Vol. 1, Nauka, Moscow (1957).
7. W. Pfann, *Zone Melting* [Russian translation], Mir, Moscow (1970).
8. V. A. Mikhailov and D. D. Bogdanova, *Electrotransfer in Liquid Metals* [in Russian], Nauka, Novosibirsk (1978).
9. D. A. Dudko, R. G. Krutikov, and K. K. Prokhorenko, *Complex Improvement of the Quality of Steel Ingots* [in Russian], Naukova Dumka, Kiev (1969).
10. V. S. Dub, V. I. Bulatnikov, L. I. Berman, et al., *Special Problems of Electrothermics* [in Russian], Cheboksary (1980), pp. 16–22.
11. Yu. A. Samoilovich, *Crystallization of an Ingot in an Electromagnetic Field* [in Russian], Metallurgiya, Moscow (1986).
12. A. I. Manokhin and G. T. Mal'tsev, *Stal'*, No. 9, 65–67 (1990).
13. K. Schwerdtfeger, *Chernye Metally* [Russian translation], No. 5, 32–39 (1979).
14. K. Schwerdtfeger, *Archiv f.d. Eisenhüttenwesen*, **41**, No. 9, 923–937 (1970).
15. Yu. A. Samoilovich, *Microcomputer in Solution of Problems of Ingot Crystallization* [in Russian], Metallurgiya, Moscow (1988).
16. T. Matsumiya, H. Kajioka, S. Mizoguchi, Y. Ueshima, and H. Esaka, Mathematical analysis of segregations in continuously cast slabs, *Trans. Iron Steel Inst. Japan*, **24**, No. 11, 873–882 (1984).