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THE MIGRATION OF A LIQUID ZONE THROUGH A SOLID: PART II

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Abstract

↓  
The migration of slab, cylindrical and spherical zones through a block of solid under the influence of an electric field gradient has been analyzed theoretically. It was determined that the zone could migrate either up or down the field gradient,  $E^*$ , depending upon the magnitude and sign of (i) the effective ionic mobility of the solvent atoms,  $U$ , (ii) the Peltier coefficient at the solid-melt interface,  $\pi$  and (iii) the imposed temperature gradient,  $G^*$ .

~~$G^*$~~   
 ~~$E^*$~~



## THE MIGRATION OF A LIQUID ZONE THROUGH A SOLID: PART II

by

W. A. Tiller

In a companion paper<sup>(1)</sup> on this subject, a theoretical analysis of zone migration under the influence of a temperature gradient as the driving force was treated. It was found that the zone migration rate depended upon the diffusional transport across the zone at the atomic kinetics of both the freezing and the melting processes. The stability of zone shape was found to depend strongly on the crystallographic orientation of the zone, the anisotropy of the atomic kinetics and the orientation of the temperature gradient relative to the zone. In the present paper, the theoretical analysis of zone migration under the influence of an electric field is presented.

### Theory

The starting geometrical and thermal configurations for the zones in a block of solid is identical with that given in the first paper.<sup>(1)</sup> The same assumptions will be made here as there and the same three zone shapes will be considered.<sup>(1)</sup> Only isotropic atomic kinetics will be considered for the quantitative calculations. To best illustrate the origin of new effects associated with the application of an electric field, we shall proceed in three steps: (i) isothermal, zero current ( $G^* = 0, I^* = 0$ ), (ii) isothermal, non-zero current system ( $G^* = 0,$

$I^* \neq 0$ ) and (iii) non-isothermal, non-zero current system ( $G^* \neq 0$ ,  $I^* \neq 0$ ). Here  $G^*$  and  $I^*$  are the bulk temperature gradient and current respectively.

Case I:  $G^* = 0$ ,  $I^* = 0$

Consider an electric field  $E^*$  applied to the system along the Z-axis such that the potential increases with Z. Let the effective mobility of the solvent ions be U; i.e. if a field of E volts/cm is applied to the liquid alloy, the net rate of movement of each solvent atom (whether ionized or not) is given by U E cm/sec. The effect of the electric field is to force the solvent atoms in the positive Z direction if U is positive and in the negative Z-direction if U is negative. This additional force alters the conventional diffusion equation in the zone so that it takes the form

$$\nabla^2 c - \frac{UE}{D} \frac{\partial c}{\partial Z} = \frac{1}{D} \frac{\partial c}{\partial t} \quad (1)$$

where E is the electric field gradient inside the zone and D is the diffusion coefficient of the solvent atoms in the melt. Assuming the same dielectric constant outside the zone and constant parameters in the two phases, the electric field gradient, E, may be related to the bulk electric field gradient,  $E^*$ , by the following relationships:

$$(i) \text{ Slab} \quad E = \frac{\zeta_S}{\zeta_L} E^* \quad (2)$$

$$(ii) \text{ Cylinder} \quad E = \frac{2\zeta_S}{(\zeta_S + \zeta_L)} E^* \quad (3)$$

$$(iii) \text{ Sphere} \quad E = \frac{3\zeta_S}{(2\zeta_S + \zeta_L)} E^* \quad (4)$$

where  $\zeta$  is the dielectric constant.

Since the migration of the zone occurs at such a slow rate, the solute field in the zone may be described, at any moment of time, by the steady-state solution of eq. 1, i.e.,

$$C = A \exp (UEZ/D) + B \quad (5)$$

where A and B are constants (slowly varying functions of time) whose time-dependence may be determined by substitution in eq. 1. For positive U, the solvent concentration increases with Z and produces zone movement down the field gradient. For negative U, the solvent concentration decreases with Z and produces zone movement up the field gradient. In general,  $UE/D \ll 1$  and eq. 5 may be used in its linear approximation.

The motion of the zone destroys the complete isothermality of the system since heat is evolved at the freezing interface and absorbed at the melting interface. The temperature distribution in the system

will be similar to that illustrated in Fig. 1. Since the temperature distribution in the solid can be shown to fall off extremely slowly with distance from the zone surface, the temperature gradient created across the zone to provide the necessary heat transport is many orders of magnitude greater than that outside of the zone and is almost exactly given by setting  $G^* = 0$  in eqs. 5, 6 and 7 of reference 1; i.e.,

$$(i) \text{ Slab} \quad G = - \frac{V_{\pi} \Delta H}{K_L} \quad (6)$$

$$(ii) \text{ Cylinder} \quad G = - \frac{V_{\pi} \Delta H}{(K_S + K_L)} \quad (7)$$

$$(iii) \text{ Sphere} \quad G = - \frac{V_{\pi} \Delta H}{(2K_S + K_L)} \quad (8)$$

where  $V_{\pi}$ ,  $\Delta H$  and  $K$  are the velocity of the  $\theta = \pi$  interface, the latent heat of fusion per unit volume and the thermal conductivity respectively. If the zone moves in the positive Z-direction,  $V_{\pi}$  is positive and  $G$  is negative tending to impede the zone motion. If the zone moves in the negative Z-direction,  $V_{\pi}$  should be used in place of  $-V_{\pi}$  and  $G$  is positive again impeding the zone motion.

The conservation of solute conditions at the zone interfaces, for positive  $U$ , become

$$V_{\pi} (C_{\pi} - C^1) = UEC_{\pi} - \frac{D}{\delta} (C_0 - C_{\pi}) \quad (9)$$

and

$$V_o (1 - k_o) C_o = - [UEC_o - \frac{D}{l} (C_o - C_\pi)] \quad (10)$$

Since  $C_o - C_\pi$  has the same sign as UE and, since the R.H.S. of eqs. 9 and 19 have the same and opposite sign as UE respectively,  $V_o$  and  $V_\pi$  always have the same and opposite sign to UE respectively, i.e., if UE is positive,  $V_o$  is positive and  $V_\pi$  is negative. For UE positive,  $C_o > C_o^*$  and  $C_\pi < C_\pi^*$ .

Assuming that the atomic kinetics remain unaltered by the presence and magnitude of E, eqs. 1, 2 and 3 of reference 1 may be used for the three predominant growth mechanisms considered. For the uniform interface advance mechanism we have, for positive UE, the zone migration rate

$$V_o = - \frac{[UE/(1 - k_o)]}{\left[ 1 - \frac{\epsilon' \Delta H}{cm} - \frac{\epsilon'}{lm} \left( \frac{1}{\mu_o} + \frac{1}{\mu_\pi} \right) \right]} \quad (11)$$

where  $\epsilon' = - D/(1 - k_o) C_o$ ,  $\mu_\pi = - V_o \mu_\pi / V_\pi$  and  $\alpha = K_L, K_S + K_L$  and  $2K_S + K_L$  for a slab, cylinder and sphere respectively. For positive UE,  $V_o$  is positive and for negative UE,  $V_\pi$  is positive. In the denominator of eq. 11, the term in  $\alpha$  arises as a consequence of the temperature gradient within the zone tending to slow the motion of the zone. In some systems  $\Delta H/\alpha \sim 10^5 \text{ sec-}^\circ\text{C/cm}^2$  and may be as large as the atomic kinetic

term except for very small values of  $l$ . The appropriate equations for the other mechanisms may be readily developed from the comparison of eq. 11 with eqs. 12, 13 and 14 of reference 1. We can see from eq. 11 that, for positive  $E$ , the sign of  $U$  determines the direction of zone motion.

Case II:  $G^* = 0, I^* \neq 0$

When a current of density  $I^*$  is passed through the sample, a quantity of heat  $Q$  watts/cm<sup>2</sup> is absorbed at one interface and generated at the other by the Peltier effect. The Peltier coefficient  $S_L^{P_L} = Q/J$  where  $J$  is the current density flowing through the interface from solid to liquid and of magnitude given by  $|J| = \sigma_L |E|$ .<sup>†</sup> Here,  $\sigma_L$  is the electrical conductivity of the liquid and positive  $S_L^{P_L}$  signifies that absorption of heat occurs when current flows from solid to liquid. In this case, a different temperature gradient is developed across the zone than that given by eqs. 6, 7 and 8. A term of magnitude  $+ Q/\alpha$  must be added to the R.H.S. of these equations ( $Q = \sigma_L E S_L^{P_L}$ ). Any effects due to Thomson heat generation in the body of the solid and the zone have been neglected. Incorporating the Peltier heat generation effect, eq. 11 becomes

<sup>†</sup> The value of  $E$  is given by eqs. 5, 6 and 7 in reference 1 by replacing  $G, G^*$  and  $K$  by  $E, E^*$  and  $\sigma$  respectively with  $\Delta H = 0$ .

$$V_o = \frac{-E \left[ \frac{U}{(1-k_o)} - \frac{\epsilon' \sigma_L S_L^P}{\alpha m} \right]}{\left[ 1 - \frac{\epsilon' \Delta H}{\alpha m} - \frac{\epsilon'}{\ell m} \left( \frac{1}{\mu_o} + \frac{1}{\mu' \pi} \right) \right]} \quad (12)$$

When  $S_L^P$  is of the same sign as  $U$ , the Peltier heating tends to impede the motion of the zone; when they are of opposite sign, the rate of zone motion is enhanced. Depending upon the magnitude and sign of  $S_L^P$ , the zone may move in the opposite direction to that expected from the sign of  $UE$  alone.

Case III:  $G^* \neq 0, I^* \neq 0$

In this case, we have for the three different atomic mechanisms,

$$V_o = \frac{-E \left[ \frac{U}{(1-k_o)} - \frac{\epsilon'}{\alpha m} \left( \sigma_{LS}^P L + \frac{pG^*}{E} \right) \right]}{\left[ 1 - \frac{\epsilon' \Delta H}{\alpha m} - \frac{\epsilon'}{\ell m} \Gamma \right]} \quad (13)$$

with (i) Uniform interface advance  $\Gamma = \left( \frac{1}{\mu_o} - \frac{1}{\mu' \pi} \right)$  (14)

(ii) Screw dislocation  $\Gamma = V_o^{-1/2} \left( \sqrt{\frac{1}{\mu_o}} + \sqrt{\frac{1}{\mu' \pi}} \right)$  (15)

(iii) Two-dimensional nucleation

$$\Gamma = V_o^{-1} \left( \frac{A_o}{\ln(\mu_o/V_o)} - \frac{A_\pi}{\ln(\mu'/\pi V_o)} \right) \quad (16)$$

and with  $p = K_S, 2K_S,$  and  $3K_S$  for slab, cylinder and sphere respectively.

In general  $\epsilon' \Delta H / \alpha m < -1$  and the atomic kinetic effects are again seen

to depend upon  $l$  as in the first paper.<sup>(1)</sup> In figure 2,  $V_0/E$  is plotted versus  $U/(1-k_0)$  for several values of the parameter  $\phi = -(\epsilon'/\alpha m)$  ( $\sigma_L S_L^P + pG^*/E$ ) and we can see the additive effect of varying  $G^*$  and different  $S_L^P$  on  $V_0/E$ . We can see that the direction of zone motion depends not only on the sign of  $E$  and  $U$ , but also on the magnitude and sign of  $\phi$ .

#### Interface Stability

We can expect similar anisotropic kinetic effects as discussed earlier for the zero electric field case.<sup>(1)</sup> However, it is likely that, in some materials, the growth of perturbations of surface contour will be enhanced while in others it will be retarded by the presence of an electric field. For a ruffled surface contour, the equipotential lines will no longer be planar in the zone and the current density will also be a function of position. The differential equations of heat and mass transport and the interface boundary conditions will be much more complex than those considered thus far due to this positional variation of  $E$  and  $J$ . The solution to this stability question will, in itself, be extremely complex and will not be considered further in this paper.

#### Discussion

For some materials, as the zone approaches a surface between two phases, it will begin to be influenced by the field due to the electric double layer of this surface or some other charge excess located

there. Such electrostatic fields combine to either enhance or retard the motion of the zone depending upon the strength and sign of the field and the sign of the ionic mobility. This is a case of  $J^* = 0$  and will be more dominant the smaller are the electrical conductivity of the solid outside the zone and of the liquid in the zone. For semiconducting and insulating solids, the width of the normal space charge region at a surface varies from about  $10^{-5}$  cm to  $10^{-1}$  cm. It seems possible that such an effect as this may be responsible for the enhanced migration rate of Wernick's<sup>(2)</sup> Ge-Al wire zones through a block of Ge as the zone approached the upper surface. He found that  $V$  increased by a factor of almost 10 as the zone moved from the lower to the upper surface of the block (temperature difference =  $30^{\circ}\text{C}$ ). A similar type of surface field effect has been observed for the migration of spheres of aqueous solution through ice as they approached the ice-water interface.<sup>(3)</sup>

One difficulty with this type of explanation for the Ge-Al wire is that we normally expect the surface space charge region in Ge to be considerably smaller than the wire sizes used. Thus, for this system, such an effect should be observed only much closer to the surface than observed by Wernick<sup>(2)</sup> unless our estimate of the space charge region is small by an order of magnitude. An alternative explanation for this effect is that, for cylindrical and spherical zones, the value of  $G$  increases considerably as the zone becomes close to the upper surface provided  $K_S \ll K_L$  ( $G$  decreases for  $K_S \gg K_L$ ). The greater is the

ratio  $K_L/K_S$ , the further from the surface will this effect of increasing  $G$  begin to be felt. This effect is illustrated in figure 3 for  $K_L/K_S > 2$ . The isotherms distort around the zone to decrease the thermal gradient in the zone. However, as this isotherm distortion region touches the surface, the radiation sees a lowered temperature and greater heat flux enters this region of surface to maintain it at constant temperature. Thus, the quantity of heat flowing into the zone increases and  $G$  increases. The magnitude of this effect can be readily determined by considering the electrostatic analogue.

The potential outside of a conducting sphere or cylinder imbedded in a uniform field,  $E_0$ , is given by that due to a dipole or line dipole respectively of moment proportional to  $E_0$  and the volume of the sphere or cross-sectional area of the cylinder respectively.<sup>(4)</sup> The field due to a pair of oppositely signed dipoles aligned in the Z-direction and separated by a distance  $2R$  produces a constant potential on the median plane, distant  $R$  from each. Likewise, the temperature is constant on the median plane separating two point doublets or line doublets of opposite sign. The thermal field due to such a pair of doublets immersed in a constant temperature gradient and each situated distance  $R$  from the external surface should be the same as that due to a spherical or cylindrical zone placed distance  $R$  from the surface. The temperature gradient,  $G$ , in the zone is given, for these two cases, by:

(i) Sphere

$$G = \frac{K_S}{K_L} G^* \left\{ 1 + 2 \left[ 1 + \frac{1}{\left(\frac{4R}{l} - 1\right)} \right] 3 \left[ \frac{K_L - K_S}{K_L + 2K_S} \right] \right\} \quad (17)$$

(ii) Cylinder

$$G = \frac{K_S}{K_L} G^* \left\{ 1 + \left[ 1 + \frac{1}{\left(\frac{4R}{l} - 1\right)} \right] 2 \left[ \frac{K_L - K_S}{K_L + K_S} \right] \right\} \quad (18)$$

We can see that, as  $R/l \rightarrow \infty$ , eqs. 17 and 18 become eqs. 7 and 6 of reference 1 with  $V$  set equal to zero. However, even for the case  $K_L \gg K_S$ ,  $R = l$  gives an increase in  $G$  of only 5% and 2-1/2% for the cylinder and sphere respectively. Even at  $R$  only slightly greater than  $l/2$ ,  $G$  increases only 50% and 25% for the cylinder and sphere respectively.

Perhaps the combination of the increase of  $G$ , increase of  $l$ , faster atomic kinetics and a nonzero value of  $E$ , combine to give this enhanced value of  $V$ .

When an external electric field is applied to the solvent block such that current  $J^*$  flows, the zone may move either up or down the electric field depending upon the parameters of the system presented in eq. 13. A change in sign of the ionic mobility,  $U$ , can cause a change in the direction of zone motion provided the first term in the numerator of eq. 13 dominates. Similarly, if the second term dominates, a change in sign of  $S_L^P$  can cause a change in the direction of zone motion. One of these

two effects is undoubtedly responsible for the observations of Pfann et al<sup>(5)</sup> who showed that, although  $J^*$  was constant and  $S_L^P$  was expected to be of the same sign for a system of binary alloys with Ge, the zones containing either Pt, Pd, Au, or Ni moved in one direction and the zones containing either Ag, Al, Bi, Cu, Ga, Sn, Mg, Pb, Sb, or Te moved in the opposite direction. In this experiment  $G^* = 0$  so that if  $U$  is unchanged from alloy to alloy,  $S_L^P$  must be of the same sign as  $U$  but with the former four alloys having a larger value of  $S_L^P$  than the latter. However, it is likely that  $U$  will change from alloy to alloy so that no clear-cut conclusion can be made from these observations.

From eq. 13, we can see that by varying  $G^*/E$  in an experiment we can determine the value  $(G^*/E)_c$  that makes  $V_o = 0$  and thus can determine a relationship between the constants in the numerator, i.e.

$$\left(\frac{G^*}{E}\right)_c = \frac{1}{p} \left[ \frac{\alpha m U}{\epsilon'(1-k_o)} - \sigma_L S_L^P \right] \quad (19)$$

From the slope of the  $V_o/E$  vs  $G^*/E$  curve, at large  $l$  so that the denominator in eq. 13 is unity, the value of  $\epsilon'p/\alpha m$  may also be determined. In any electric field experiment with  $J^* \neq 0$  we are unable to separate  $U$  and  $S_L^P$ . Only if a  $J = 0$  experiment can be designed can the term containing  $S_L^P$  be set to zero and a value for  $U$  determined.

Since  $U$  is the effective mobility for the solvent, it may be produced by the field-induced migration of solute ions since the displacement of solute in one direction requires a displacement of solvent

in the opposite direction. If a second solute species is added to the melt,  $U$  will probably change in magnitude and may even change in sign.

#### Conclusions

The migration of slab, cylindrical and spherical zones under the influence of an electric field has been treated. It was determined that the zone could migrate both up or down the applied field gradient,  $E^*$ , depending upon the magnitude and sign of the effective ionic mobility of the solvent atoms,  $U$ , the sign and magnitude of the Peltier coefficient at the solid-melt interface,  $S_{S/L}^P$ , and the sign and magnitude of the imposed temperature gradient,  $G^*$ . It was further determined that, in some systems, electrostatic effects could affect the zone migration rate. Finally, it was shown that an enhanced migration rate of cylindrical and spherical zones in the immediate vicinity of a surface will occur when  $K_S/K_L \ll 1$  due to an increase in  $G$  across the zone.

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FIGURE CAPTIONS

Figure 1. Temperature distribution developed in an initially isothermal system due to zone migration under the influence of an internal electric field.

Figure 2. Plot of  $V_0/E$  versus  $U/(1-k_0)$  for several values of the parameter  $\phi = -\frac{\epsilon_1}{\alpha m} (\sigma_L S^2 L + pG^*/E)$ .

Figure 3. Isotherm distortion due to a spherical or cylindrical zone of greater thermal conductivity than the matrix when the zone is either imbedded in the bulk or is near a surface.

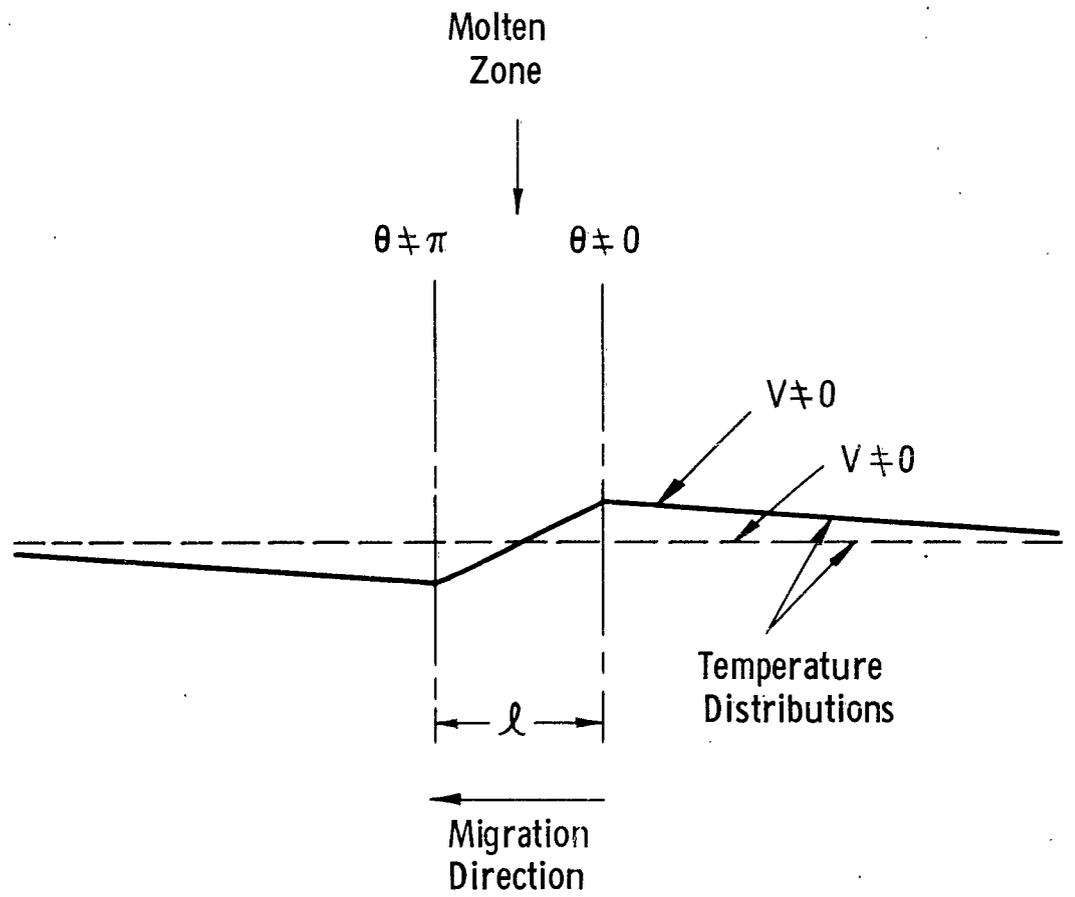


Fig. 1

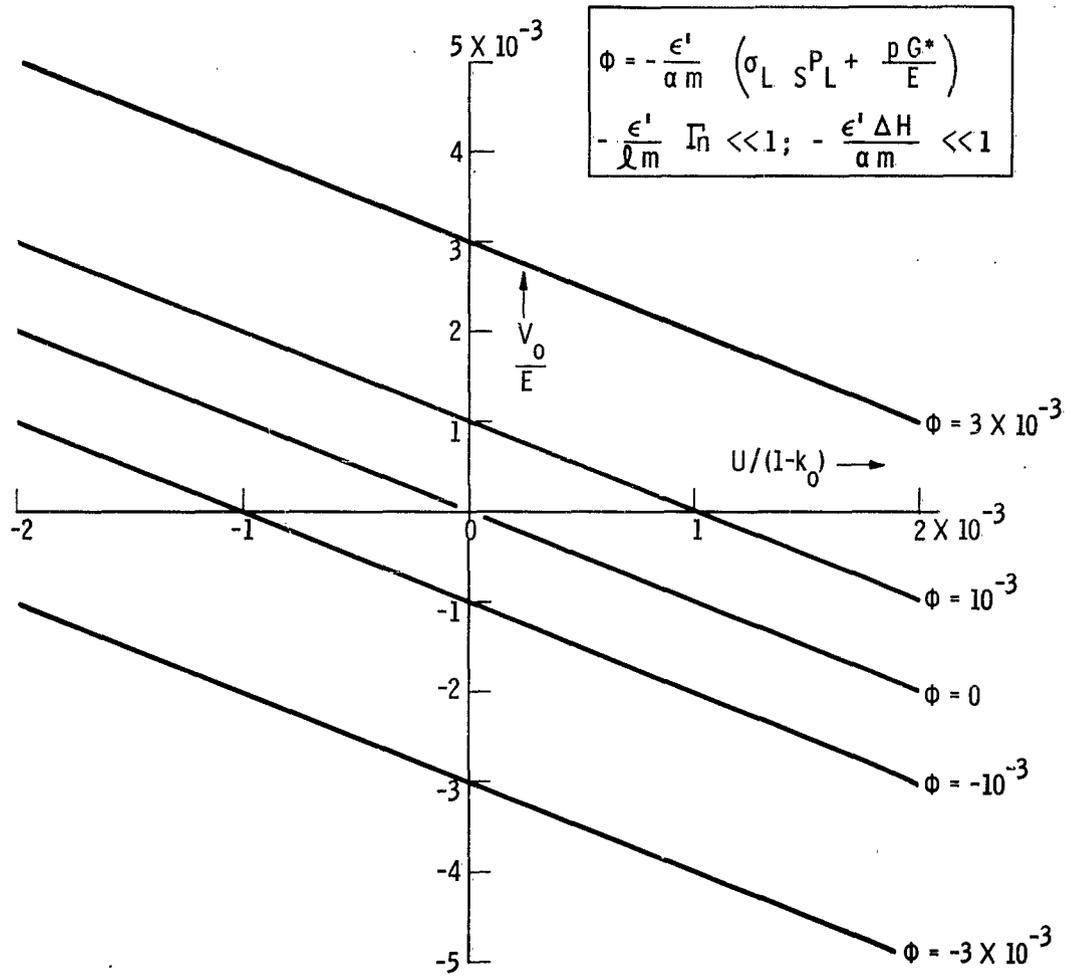


Fig. 2

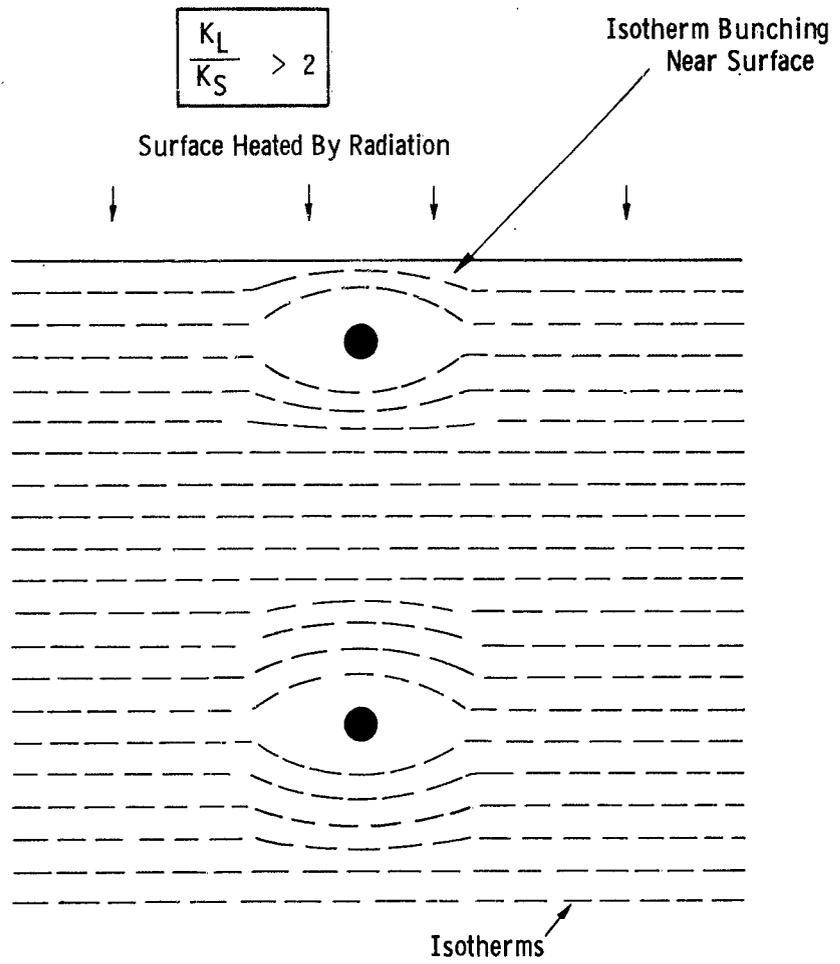


Fig. 3

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Lists 5 references.

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Note to secretaries - Same information  
must be typed three times on each mat.