THE KINETICS OF LIQUID-PHASE EPITAXY OF MOVING-TEMPERATURE-GRADIENT ZONE REFINING

V. N. Lozovskii, V. P. Popov, and I. P. Papkov

The authors analyze the dependence of the growth rate of layers during moving-temperature-gradient zone refining on the zone thickness, the process temperature, and the mechanism of atomic-kinetic processes at interfaces. The results of experimental investigations of the growth kinetics in the silicon-aluminum system are in agreement with theoretical analyses.

Moving-temperature-gradient zone refining (MTGZR) makes it possible to grow epitaxial layers of complex geometry and composition both on the surface and in the bulk of the crystal [1]. The principal advantage of this technique over stationary-temperature-gradient zone refining (STGZR) is that the conditions of absolute dynamic stability can be realized. This paper discusses the results of theoretical and experimental investigations of the kinetics of MTGZR under the conditions of absolute dynamic stability in the silicon-aluminum system.

Absolute dynamic stability of the interface between the liquid zones (MTGZR) is attained when the zone-dissolution front is in the region of zero thermal-field gradient of the rod heaters [1]. In this case a linear distribution of concentration and temperature is established in the crystal-melt-crystal composition. The conditions of material balance of solute atoms in a binary system on the crystallization and dissolution-interfaces are of the form:

\[ V_c (C_c' - C_d) = D \frac{C_d - C_c}{l} \]
\[ V_d (C_d' - C_d) = D \frac{C_d - C_s}{l} \]

where \( V_c \) and \( V_d \) are the crystallization and dissolution front velocities, respectively; \( C_s \) is the concentration of the solute in the growing crystal; \( C_c \) is the concentration of solute in the zone at the crystallization interface; \( D \) is the diffusion coefficient of the solute in the liquid phase; \( l \) is the thickness of the zone; \( C_d' \) and \( C_d \) are the concentrations of the solute in the solid phase and in the zone at the dissolution interface.

For each possible mechanism of atomic-kinetic processes at the interface the velocity is related to the supercooling by a functional relation. For example, with a uniform growth (dissolution) this relation is linear:

\[ V_c = \mu_{tc} \frac{dT}{dC} (C_c - C_d), \quad V_d = \mu_{td} \frac{dT}{dC} (C_c' - C_d) \]

where \( \mu_{tc} \) and \( \mu_{td} \) are atomic-kinetic coefficients; \( C_c \) and \( C_d \) are the equilibrium values in the zone at the crystallization-dissolution interface; \( dT/dC \) is the slope of the liquidus on the state diagram of the system. Using the relation between the velocity and the quantities \( C_d \) and \( C_c \) for different mechanisms of atomic-kinetic processes from Eqs. (1) and (2), we can find expressions for the rate of crystalline growth (dissolution). For example, for normal growth we find:
\[ V_c = \left( \frac{C_d^i - C_c^i}{C_d^e - C_c^e} \right) \cdot \frac{dC}{dT} \cdot \frac{dT}{dC} \cdot D \cdot \frac{1}{\varphi_1 c} + \frac{1}{\varphi_1 d}, \tag{3} \]

where \( \mu_{id} = \mu_{id}(V_d/V_c) \). With allowance for the approximate solution

\[ \frac{dT}{dC} = \frac{T_d - T_c}{C_d^e - C_c^e}, \]

where \( T_d \) and \( T_c \) are the temperatures at the dissolution–crystallization interface of the zone, Eq. \( (3) \) goes over into

\[ V_c = \left( \frac{T_d - T_c}{C_d^e - C_c^e} \right) \cdot \frac{dC}{dT} \cdot D \cdot \frac{1}{\varphi_1 c} + \frac{1}{\varphi_1 d}. \tag{4} \]

The actual jump in temperature in the liquid zone is found by using a function describing the temperature distribution in the field of the rod heaters \( [1] \):

\[ T_d - T_c = \frac{2}{\eta} \ln \left( \frac{1}{1 + \frac{\eta^2}{h^2}} \right), \tag{5} \]

where \( h \) is the half-distance between the rod heaters; \( \eta \) is the thermal power of a rod per unit length; \( \eta \) is a coefficient taking account of the field reduction in the crystal.

Substitution of Eq. \( (5) \) into Eq. \( (4) \) yields the following expression for the rate of crystalline growth under the conditions of absolute dynamic stability:

\[ V_c = \frac{2}{\eta} \ln \left( \frac{1}{1 + \frac{\eta^2}{h^2}} \right) \cdot \frac{dC}{dT} \cdot D \cdot \frac{1}{\varphi_1 c} + \frac{1}{\varphi_1 d}. \tag{6} \]

Similarly, we find the growth rate on screw dislocations:

\[ V_c = \frac{2}{\eta} \ln \left( \frac{1}{1 + \frac{\eta^2}{h^2}} \right) \cdot \frac{dC}{dT} \cdot D \cdot \frac{1}{V_c} \cdot \frac{\varphi_{2c}}{\varphi_{1c}} \cdot \frac{\varphi_{2d}}{\varphi_{1d}}. \tag{7} \]

and in the case of growth on two-dimensional nuclei:

\[ V_c = \frac{2}{\eta} \ln \left( \frac{1}{1 + \frac{\eta^2}{h^2}} \right) \cdot \frac{dC}{dT} \cdot D \cdot \frac{1}{V_c} \cdot \frac{\varphi_{2c}}{\varphi_{1c}} \cdot \frac{\varphi_{2d}}{\varphi_{1d}}. \tag{8} \]

The dissolution rate is found in similar fashion. Equations \( (6)-(8) \) describe the process of crystalline growth in the general case. For values of \( l \) at which the first term in their denominator is much greater than the second, Eqs. \( (6)-(8) \) become

\[ V_c = \frac{D}{C_d^e - C_c^e} \cdot \frac{2}{\eta} \cdot \frac{l}{V_c} \cdot \frac{dC}{dT} \ln \left( 1 + \frac{\eta^2}{h^2} \right). \tag{9} \]

Equation \( (9) \) describes the zone refining process in the diffusion regime. With the condition \( l^2/h^2 < 1 \), which is realized in experiments, Eq. \( (9) \) goes over into

\[ V_c = \frac{D}{C_d^e - C_c^e} \cdot \frac{2}{\eta} \cdot \frac{l}{h^2} \cdot \frac{dC}{dT}. \tag{10} \]