

# ON THE THEORY OF ISLAND FILMS GROWTH FROM EUTECTIC MELT AT THE LATE EVOLUTION STAGE

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**Abstract.** The theory of late-stage evolution of island films from the eutectic composition binary melt is developed. The complete system of equations describing the island film evolution on the substrate surface at the stage of Ostwald ripening is suggested. The asymptotic solution of that system of equations reveals a strong correlation in the evolution of the islands of different compositions at the Ostwald ripening stage which is associated with some specific features of eutectic systems. All main characteristics of island films at the late evolution stage such as islands size distribution function, critical sizes and density of the islands have been obtained. Relations between the individual agents constants causing predisposition of the eutectic system to one of the specific mechanisms of heat mass transfer have been found. The processes accompanying growth of the films from the eutectic composition melts in the presence of heat sink are discussed.

## 1. INTRODUCTION

Films and coatings with the eutectic-type state diagram (Fig.1) are widely exploited in metallurgy, machine industry and microelectronics [1]. The film growth from a liquid phase of eutectic composition is as widely used as growth from a gas phase. Particularly, this technique is applied for producing of protective coatings by rapid cooling of a liquid layer of eutectic melt [2].

According to the experimental data [1,2], the structures of the eutectic films are quite variable. They are fine-dispersed films consisting of both phase islands mixed in random way, single phase islands inside the other phase matrix, etc. Sometimes, amorphous films of the eutectic composition are formed. A formation of solid films on the solid surface (a substrate) is a typical first-order phase transition that can be divided into several stages. At the first stage, the new phase appears on the substrate surface. Then the islands formed grow without a change in their number. The last stage is the Ostwald ripening or coalescence of the ensemble of new phase islands followed by their merging into a continuous film. In this paper, we present a quantitative approach to the theory of late-stage evolution of island ensemble on the solid surface from the eutectic composition melt.

## 2. THE EVOLUTION OF ISLANDS ENSEMBLE IN CONSERVATIVE SYSTEMS. FORMULATION OF THE PROBLEM AND PHYSICAL ESSENCE OF THE PROCESS

First, let us consider the possible ways of nucleation process for islands ensemble on a solid surface. For instance, if the concentration of the component *A* is less than eutectic one, the cooling of the system results in change of its composition following liquidus line until the system reaches the eutectic point and the component *B* starts to precipitate. There is, however, another possibility. The system may reach this point from the right, from the side of the component *B* (see Fig. 1). When this way is used to synthesize the island film, by the moment of reaching the eutectic point the formed islands of one of the compositions exist on the surface already, and the closer is the initial composition of the melt to that of the pure component, the more islands of a new phase *A* will occupy the substrate surface. Thus, the formation of those secondary islands may occur not only on the substrate surface, but also on the surfaces of already existing and growing crystals.

Another way to obtain the islands ensemble on the substrate surface is to cool the eutectic composition melt to a temperature below the eutectic point, where new

phase islands of compositions *A* and *B* are formed simultaneously. Then the islands of both phases may not be in contact with each other directly, and growth of islands of one phase will not be distorted by the stress field of the other phase island. An interaction between the islands can occur by means of the heat and diffusion fields [3-7]. Hence, the structure of the eutectic depends both on the composition of the melt from which it originated and the way of the new phase formation.

We will restrict our consideration here only to growth of an island film from melts of strictly eutectic composition, using as a case in point the so-called "normal" eutectics. "Normal" eutectic means that the melting temperatures of the components in pure forms are close to each other [2]. A study of other ways of the film formation as well as growth of island films from "anomalous" eutectics is in progress.

Let us assume that there is an island film on the solid surface, characterized by initial size distribution function for each phase  $f_{0i}$ , where index *i* denotes the type of the chemical composition of an island. We assume, for the sake of simplicity, that the islands are either spherical segments of radius *R* or cylinders of radius *R* and height *h*. It should be noted that, according to [3,4,6,7], the process of Ostwald ripening occurs in the system under consideration, if the thickness *l* of liquid (melt) layer is of order of *h*, height of islands, that is,  $l \approx h$ . As it is demonstrated in [3], this condition can be achieved, for example, when a substrate is rotated in the melt. As with any system undergoing a first-order transition, the system under consideration will eventually reach the stage of Ostwald ripening [3-7]. This process is accompanied by creation of a generalized diffusion and thermal field induced by the whole ensemble of islands.

Let us consider an evolution of spherically symmetric islands. According to [3-7], during the stage of Ostwald ripening the islands with radii  $R < R_k$  dissolve, while those with radii above the critical value grow. Some time ago, the theory of Ostwald ripening for the compositions far from the eutectic one was developed [5,6]. To account for the effect of simultaneous growth of the islands of both compositions in the eutectic point, let us suggest the following system of equations:

$$\frac{\partial f_i(R, t)}{\partial t} + \frac{\partial}{\partial R} [f_i(R, t) V_{Ri}] = 0, \quad (1)$$

$$f_i|_{t=0}(R, t) = f_{0i},$$

$$\frac{d\bar{\xi}_i}{dt} + \chi \int_0^\infty f_i(R, t) J_{D,R,i} R^2 dR = 0, \quad (2)$$

$$\frac{d(\bar{T} c_{pi} \rho_i)}{dt} - \chi \int_0^\infty f_i(R, t) J_{T,R,i} R^2 dR = 0, \quad (3)$$

$$L_i J_{D,i} = J_{T,R,i}, \quad (4)$$

$$T_{Ri} = \varphi_i(\xi_{Ri}), \quad (5)$$

where (1) is the equation of continuity in the size space for the distribution function of phase *i* islands in terms of sizes  $f_i(R, t)$ ; (2)-(3) describe respectively the variation of the average component concentration and temperature in the melt; (4) connects the heat and matter flows on the boundary of an island of radius *R*; (5) relates the equilibrium temperature at the boundary of an island of radius *R* to concentration  $\xi_{Ri}$ ;  $J_{D,R,i}$  and  $J_{T,R,i}$  are respectively the flow of the component to a new-phase island and the heat flow released at growth of an island of radius *R* [6];  $c_{pi}$  and  $\rho_{Li}$  are the heat capacity and the density of the melt at constant pressure (in case when the process runs due to heat mass transfer in the bulk phase) or the heat capacity and the density of the substrate when heat mass transfer runs on the substrate;  $\chi = 2\pi(1 - \cos\Theta)$  is the shape factor;  $\varphi_i(\xi_{Ri})$  is determined by a given phase diagram of state and relates the equilibrium concentration at the boundary of the island of radius *R* and temperature.

The subscript *i* identifies the part of the diagram under consideration and, accordingly, the composition of new-phase islands. For the left-hand part of the diagram, where islands of the composition *A* grow, we can set  $i=1$ , and, for its right-hand part, where the islands have composition *B*,  $i=2$ . The specific feature of the eutectic point of the diagram of state is simultaneous growth of the islands of both types, in which case the derivative  $\partial T_0 / \partial \xi_0$  (where  $T_0$  and  $\xi_0$  are respectively the equilibrium temperature and equilibrium concentration) reverses its sign. We will demonstrate that at the late evolution stage the feature in question should result in self-correlated growth of an island ensemble of both phases.

Let us consider the diffusion fluxes to the spherical symmetry island  $J_{D,i}$ . To do so, we write the following diffusion equations for the phases, taking into account that spatial region occupied by the islands expands in space with the velocity  $W_p$ :

$$\frac{\partial \xi_i}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left[ 2r^2 \left( D_i \frac{\partial \xi_i}{\partial r} - W_p \xi_i \right) \right]. \quad (6)$$

From (6) we have

$$\frac{\partial}{\partial t} (\xi_1 + \xi_2) = \frac{1}{r^2} \frac{\partial}{\partial r} \left[ 2r^2 \left( D_1 \frac{\partial \xi_1}{\partial r} + \right. \right.$$

$$D_2 \frac{\partial \xi_2}{\partial r} - W_p (\xi_1 + \xi_2) \Bigg] \Bigg] .$$

Then, with the following equations taken into consideration:

$$\xi_1 + \xi_2 = 1, \quad \frac{\partial \xi_1}{\partial r} = - \frac{\partial \xi_2}{\partial r},$$

we get the equation for  $W_p$ :

$$W_p = (D_1 - D_2) \frac{\partial \xi_1}{\partial r} = (D_2 - D_1) \frac{\partial \xi_2}{\partial r}.$$

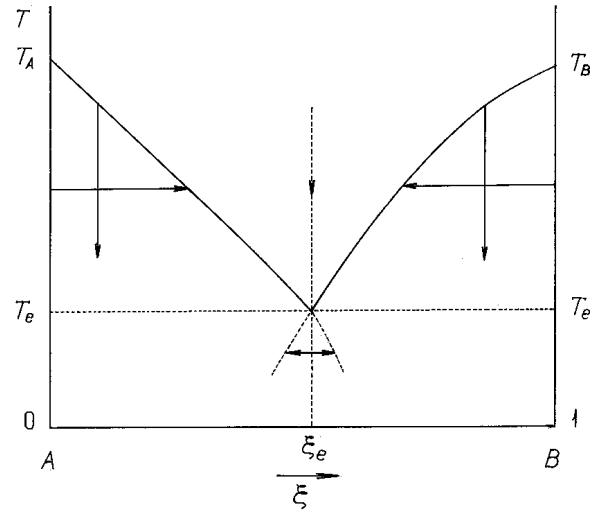
Then, turning back to (6), we can write down:

$$\frac{\partial \xi_i}{\partial t} = \frac{1}{r^2} \frac{\partial}{\partial r} \left[ 2r^2 (\xi_2 D_1 + \xi_1 D_2) \frac{\partial \xi_i}{\partial r} \right], \quad (7)$$

As it can be seen from the analysis of formula (7), the diffusion process kinetics is caused by the interdiffusion coefficient  $\tilde{D}$  which is as follows:

$$\tilde{D} = D_1 \xi_2 + D_2 \xi_1.$$

If equilibrium concentration lines are symmetric relative to the eutectic composition line (see Fig.1), and taking into account that supersaturation at the Ostwald ripening stage tends to zero and changes very slowly,  $\tilde{D}$  can be assumed constant. In the situation discussed, the fluxes of the components will be equal in magnitude, but opposite in direction:  $J_{D,R,1} = -J_{D,R,2}$ . This means that in the process of its growth an island of each phase changes concentration of its component in the melt, and, as a corollary, that of the other component. For example, the growing islands of composition A raise the concentration of component B, thus accelerating the rate of its growth. At the same time, an increase in the growth rate of island of composition B reduces the concentration of component B in the melt, which, in its turn, accelerates the growth of islands A, making the growth of islands correlated, in which case the average concentration in the system tends to the eutectic one,  $\xi_e$ . It should be noted, however, that in case when the metastable concentration lines are not strictly symmetric relative to the eutectic composition line (Fig.1) compositional fluctuations may set at the initial stage, and the individual component supersaturation will be "removed" independently. In going so, such system evolution should be considered as similar to that of multicomponent ensembles [7]. Nevertheless, at the Ostwald ripening stage, where supersaturations are low, the configuration of metastable lines is close to symmetrical one at any arrangement of the equilibrium concentration lines [2].



**Fig. 1.** Typical diagram of state of eutectic system of A and B components.  $T_A$  and  $T_B$  are the melting temperatures of pure components,  $\xi_e$  is the composition at the eutectic point,  $T_e$  is the crystallization temperature of eutectic mixture, arrows  $\rightarrow$  denote possible ways of crystallization. The lines of metastable states are indicated under the eutectic point as dotted lines.

Thus, it has been stated that the average concentration at the Ostwald ripening stage remains constant for the system as a whole, and equation (2) becomes identically equal to zero. It should be noted that it is certainly not true for the initial stage of film evolution.

### 3. BASIC SYSTEM OF EQUATIONS FOR THE ISLAND FILM EVOLUTION AT THE OSTWALD RIPENING STAGE

The above reasoning allows us to re-write the system (1)-(5) as follows:

$$\frac{\partial f_i(R, t)}{\partial t} + \frac{\partial}{\partial R} [f_i(R, t) V_{Ri}] = 0, \quad (8)$$

$$f_i|_{t=0}(R, t) = f_{0i},$$

$$\frac{d(\bar{T} c_{pe} \rho_e)}{dt} - 4\chi \sum_{i=1}^2 \int_0^\infty f_i(R, t) J_{T,R,i} R^2 dR = 0, \quad (9)$$

$$\sum_{i=1}^2 L_i J_{D,i} = \sum_{i=1}^2 J_{T,R,i}. \quad (10)$$

The system (8)-(10) should be supplemented by the dependence of island growth rate on its radius. To do so, let us consider the expression for island growth rate in the domain far from the eutectic point [5-7]:

$$V_i = \frac{\left[ 2\sigma_{SLi} \omega_i^2 \xi_{L\infty i} D_{Li} N_0 (K_{Li} R \alpha(\theta_i) \ln\left(\frac{H}{R_i}\right) + K_{si} l \right] \alpha(\theta) \psi_i(\theta_i)}{\left[ L_i^2 D_{Li} N_0 \xi_{L\infty i} \alpha(\theta_i) R \ln\left(\frac{H}{R_i}\right) + k T_{0i}^2 (K_{Li} R \alpha(\theta_i) \ln\left(\frac{H}{R_i}\right) + K_{si} l \right] R^2} \left( \frac{R}{R_{ki}} - 1 \right), \quad (11)$$

where  $\sigma_{SLi}$  denotes the surface tension on the boundary of a new-phase island,  $N_0$  the total number of molecules per unit volume,  $H=(\pi N(t))^{-1/2}$  is the characteristic dimension of the substrate area per one island,  $N(t)$  is a number of islands per the substrate unit surface,  $\alpha(\Theta) = (1 - \cos \Theta)/2$ ;  $\varphi_1(\Theta) = 4(2 - 3\cos\Theta + \cos^3\Theta)^{-1}$ ;  $\Theta$  is the contact angle;  $l$  is the substrate thickness;  $k$  is Boltzmann constant;  $D_{Li}^0$ ,  $K_{Li}^0$ ,  $K_{si}^0$  are respectively the generalized coefficients of diffusion and heat conductivity of the melt and the new phase [5,6]. Following [3], the coefficients are as follows:

$$D_{Li}^0 = \frac{D_{Li} \beta_i \omega_i R}{D_{Li} + \beta_i \omega_i R}; \quad K_{Li}^0 = \frac{(K_{Li} \beta_i L_i R)}{T_{0i}} \frac{1}{K_{Li} + \frac{\beta_i L_i R}{T_{0i}}}; \quad K_{si}^0 = \frac{K_{si} \beta_i L_i R}{T_0} \frac{1}{K_{si} + \frac{\beta_i L_i R}{T_0}},$$

where  $\beta$  is the specific flux of atoms from the melt to an island at equilibrium conditions.

In the case where the system composition is far from eutectic one, the equilibrium concentration  $\xi_{\infty i}$  can be found as the time asymptotic solution of the system (8)-(10). The concentration  $\xi_{\infty i}$  is real thermodynamic quantity, at reaching which Ostwald ripening comes to its finish, and a new-phase equilibrates with the melt. We now write down the expression for the growth rate  $V_i$  at the eutectic point using the approach developed in [5,6]. Let  $\xi_{Ri}$  be the metastable equilibrium concentration. The difference between the average concentration (which is equal to the eutectic concentration at the stage of Ostwald ripening) and the metastable equilibrium concentration causes the driving force for the growth of islands of the new phase. Having the system (1)-(5) solved in the situation with time asymptotic, using the method [5,6], one can find the concentration  $\xi_{\infty i}$ . It is obvious that, as with [5,6], the metastable equilibrium concentrations of components at the Ostwald ripening stage should be equal to one another, and as a corollary, have the value equal to eutectic one:  $\xi_{R1} = \xi_{R2} = \xi_e$ . The equilibrium temperatures at asymptotic should be equal to the eutectic temperatures:  $T_{01} = T_{02} = T_e$ .

Now let us write the expression for the island growth rate for the case of interest:

$$V_i = \frac{\left[ 2\sigma_{SLi} \omega_i^2 \xi_e \tilde{D} N_0 (K_{Li} R \alpha(\theta_i) \ln\left(\frac{H}{R_i}\right) + K_{si} l \right] \alpha(\theta) \psi_i(\theta_i)}{\left[ L_i^2 \tilde{D} N_0 \xi_e \alpha(\theta_i) R \ln\left(\frac{H}{R_i}\right) + k T_e^2 (K_{Li} R \alpha(\theta_i) \ln\left(\frac{H}{R_i}\right) + K_{si} l \right] R^2} \left( \frac{R}{R_{ki}} - 1 \right). \quad (12)$$

As seen above, in spite of the fact that the evolution of an ensemble of islands is driven by both thermal and diffusion processes, during film growth from the eutectic composition melts the supercooling relative to  $T_e$  becomes unique for the entire system, and it is this supercooling that is observable in a physical experiment. With this taken into account, let us consider the ratio between the critical radii of islands of different phases  $R_{ki}$ . According to the relation by Gibson-Tompson, the dependence of critical radius on supercooling has the form:

$$R_{ki} = \frac{2\sigma_{SLi} T_{0i} \omega_i}{L_i \Delta_i T},$$

where  $\Delta_i T$  is the melt supercooling in terms of the phase  $i$ . Since the supercooling in the eutectic composition melt becomes unique, the critical radii of islands have the form  $R_{ki} = (2\sigma_{SLi} T_e \omega_i) / (L_i \Delta T)$ . It leads to a very important conclusion that the critical radii of islands of both phases become similar, i.e.,

$$R_{k1} = \gamma R_{k2}, \quad \text{where} \quad \gamma = \frac{\sigma_{SL1} \omega_1 L_1}{\sigma_{SL2} \omega_2 L_2}. \quad (13)$$

From our above consideration it follows that, during evolution of an island ensemble on solid surface from the eutectic composition melt the critical radii of islands of different kind become similitude relate. This implies that

the critical radii of islands of different kind,  $R_{ki}$ , vary in time by the same law either. The whole equation system (8)-(10) looks similar to that [3] derived for island ensemble growth on the surface from single-component melt. Therefore, the system can be solved at  $t \rightarrow \infty$  in the same way as with [3]. This allows us to write below the solutions of (8)-(10) as those obtained in book [3].

In doing so, the distribution function for islands of each kind is as follows:

$$f_i(R, t) = \frac{N_i(t)}{R_{ki}} P_p(u). \quad (14)$$

Here  $p=2, 3$  or  $4$ , depending on heat mass transfer mechanism in the system [5,6], and  $P_p(u)$  is the density of probability for an island to have a dimension between  $u$  and  $u+du$ , where  $u=R/R_k$ . The form of the distribution function  $P_p(u)$  will be given in the following discussion of evolution in open systems.

The density of islands on the surface varies with time as

$$N_i(t) = N_i(0) \left( 1 + \frac{(A_{pi} t)}{R_{k0i}^{3/p}} \right)^{-3/p}, \quad (15)$$

where  $N_i(0)$  is the number of islands on the surface at the onset of the Ostwald ripening process;  $R_{k0i}^p$  is the critical radius of the islands of the new-phase at the onset of the Ostwald ripening process. The critical radii in closed systems are equal to the average ones and vary at  $t \rightarrow \infty$  as follows:

$$R_{ki}^p \sim A_{pi} t \quad (16)$$

with  $A_{pi}$  being are the kinetic coefficients whose values are available for different mechanisms of heat mass transfer from [5].

Since  $R_{k1}(t) = R_{k2}(t)$ , the function  $P_p(u)$  is identical for both phases, and, as a corollary, the distribution function  $f_1(R, t)$  is similar to  $f_2(R, t)$  (see the Eq. (14) and Fig. 2). From (13) and (16) it follows:

$$\frac{\sigma_{SLi} \omega_i T_e}{L_i \Delta T} = (A_{pi} t)^{1/p}.$$

The ratio between the critical radii of islands belonging to different phases can be written as:

$$\frac{L_2}{L_1} = \frac{\sigma_{SL2} \omega_2}{\sigma_{SL1} \omega_1} \left( \frac{A_{p1}}{A_{p2}} \right)^{1/p}. \quad (17)$$

This allows one to receive a very interesting relation between the coefficients of individual matters. We

consider the case where heat mass transfer is caused by the diffusion and heat conduction in the bulk phase, and the conditions [5,6] are met.

$$\tilde{D} \ll \tilde{\beta} \omega_i R;$$

$$K_L \ll \frac{\tilde{\beta} L_i R}{T_e},$$

$$A_{3i} = \frac{8}{9} \frac{\tilde{D} K_L \omega_i T_e N_0 \xi_e \psi_1(\Theta_i) \alpha(\Theta_i)}{\tilde{D} N_0 \xi_e L_i^2 + K_L k T_e^2}.$$

It shows clearly that, if  $\tilde{D} N \xi_e L_i^2 \gg K_L k T_e^2$ , the relation (17) takes the form:

$$\frac{L_2}{L_1} = \left( \frac{\omega_2 \sigma_{SL2}^2 \psi_1(\Theta_1) \alpha(\Theta_1)}{\omega_1 \sigma_{SL1}^2 \psi_1(\Theta_2) \alpha(\Theta_2)} \right)^{1/2}.$$

The ratios of the coefficients for most widespread mechanisms of heat mass transfer are shown in Table 1. The ratios for other mechanisms can be easily obtained in the same way, using [6]. The existence of such ratios allows to define which type of heat mass transfer is characteristic of the eutectic system under consideration. It should be noted that, if the islands are shaped as cylinders and their height and radius vary in a correlate way, the general nature of the process and its description do not change. In this case we have to take into account variations of the coefficient of shape  $\chi$  in the system (1)–(5) (as was done in [6]) and solve it by the above method.

#### 4. OPEN SYSTEMS

Apparently, the heat sink from the system influence evolution of the islands ensemble at the stage of Ostwald ripening. According to [3], at that stage, the heat sinks can be described by the polynomial  $g_T n T^{n-1}$ , where  $g_T$  is the heat sink power; and  $n$  is a damping index of heat sink, which is not necessarily to be an integer. It was stated in paper [3] that, for  $n < 0 < 3/p$ , the sinks are damped, while for  $n \geq 3/p$ , they are undamped. In this study we will consider the processes occurring during the growth of an island film from eutectic composition melt in the presence of damped heat sinks.

Let us write the equations (8)-(10) for an open system. The equation of continuity (8) in this case will remain unchanged, whereas the equation of balance (9) should be supplemented by the heat sink  $g_T n t^{n-1}$  as was done in [3-7]:

$$\frac{d(\bar{T} c_{pe} \rho_e)}{dt} + g_T n t^{n-1} = \quad (18)$$

**Table. 1.** The ratios between individual matters constants for some mechanisms of heat mass transfer.

<i>Mechanisms of mass transfer at Ostwald ripening</i>	<i>Mechanisms of heat transfer at Ostwald ripening</i>	<i>Particular cases</i>	<i>p</i>	<i>Relations <math>L_2/L_1</math></i>
Diffusion transfer via bulk phase	Heat transfer via bulk phase	$\tilde{D} \ll \beta_L^{0s} V_m^s R;$ $K_L \ll \frac{\beta_L^{0s} L^s R}{T_0^s};$ $\tilde{D} N_0 \xi_e L_i \gg K_L k T_e^2$	3	$\frac{L_2}{L_1} = \left( \frac{\omega_2 \sigma_{SL2}^2 \psi_1(\Theta_1) \alpha(\Theta_1)}{\omega_1 \sigma_{SL1}^2 \psi_1(\Theta_2) \alpha(\Theta_2)} \right)^{1/2}$
		$\tilde{D} N_0 \xi_e L_i \ll K_L k T_e^2$	3	$\frac{L_2}{L_1} = \left( \frac{\omega_2 \sigma_{SL2}^2 \psi_1(\Theta_1) \alpha(\Theta_1)}{\omega_1 \sigma_{SL1}^2 \psi_1(\Theta_2) \alpha(\Theta_2)} \right)^{1/3}$
Diffusion transfer along the substrate surface	Heat transfer via bulk phase	$\lambda \gg R;$ $K_L \ll \frac{\beta_i L_i}{T_e}$	3	$\frac{L_2}{L_1} = \left( \frac{\omega_2 \sigma_{SL2}^2 \psi_1(\Theta_1) \alpha(\Theta_1)}{\omega_1 \sigma_{SL1}^2 \psi_1(\Theta_2) \alpha(\Theta_2)} \right)$
		$\lambda \ll R;$ $K_L \ll \frac{\beta_i L_i}{T_e};$ $\tilde{D} N_0 \ll \beta \lambda_s;$ $\tilde{D} N_0 \xi_e L_i \varphi(\Theta) \gg K_L k T_e \psi_1(\Theta) \alpha(\Theta)$	3	$\frac{L_2}{L_1} = \left( \frac{\omega_2 \sigma_{SL2}^2 \psi_1(\Theta_1) \alpha(\Theta_1) \varphi(\Theta_2)}{\omega_1 \sigma_{SL1}^2 \psi_1(\Theta_2) \alpha(\Theta_2) \varphi(\Theta_1)} \right)$
		$\tilde{D} N_0 \xi_e L_i^2 \varphi(\Theta) \ll K_L k T_e \psi_1(\Theta) \alpha(\Theta)$	3	$\frac{L_2}{L_1} = \left( \frac{\omega_2 \sigma_{SL2}^2 \varphi(\Theta_2)}{\omega_1 \sigma_{SL1}^2 \varphi(\Theta_1)} \right)^{1/3}$
Diffusion along steps	Heat conductivity along the surface	$M = 2L / l;$ $2\tilde{D} \xi_e k \varphi L_i^2 \gg K_L k T \lambda \pi l \psi_1(\Theta)$	3	$\frac{L_2}{L_1} = \left( \frac{\omega_2 \sigma_{SL2}^2 \psi_1(\Theta_2) \varphi(\Theta_2)}{\omega_1 \sigma_{SL1}^2 \psi_1(\Theta_1) \varphi(\Theta_1)} \right)$
		$2\tilde{D} \xi_e k \varphi L_i^2 \ll K_L k T \lambda \pi l \psi_1(\Theta)$	3	$\frac{L_2}{L_1} = \left( \frac{\omega_2 \sigma_{SL2}^2 \varphi(\Theta_2)}{\omega_1 \sigma_{SL1}^2 \varphi(\Theta_1)} \right)^{1/3}$

$$4\chi \sum_{i=1}^2 \int_0^\infty f_i(R, t) J_{T, R, i} R^2 dR = 0.$$

When considering asymptotic region  $t \rightarrow \infty$ , we may neglect the term  $d(\bar{T}_{pe} \rho_e)/dt$ , since it is small compared to the heat sink. It is obvious that the system thus obtained can be solved as was done early in this paper and book [3].

In doing so, let us write the solution for the asymptotic size distribution function of new-phase islands  $i$  for those mechanisms of heat mass transfer corresponding to  $p=2, 3$  or  $4$  [6]:

$$f_i(R, t) = \frac{N_i(t)}{R_{ki}} P_p(u), \quad (19)$$

where:

$$P_2(U) = \begin{cases} \frac{(2e)^{3-2n} (3-2n) U \exp\left(-\frac{3-2n}{1-U/2}\right)}{(2-U)^{2+2\left(\frac{3}{2}-n\right)}}, & U < 2 \\ 0, & U \geq 2 \end{cases} \quad (20)$$

$$P_3(U) = \begin{cases} \frac{\left(\frac{3^3 e}{2^{5/3}}\right)^{1-n} 3(1-n) U^2 \exp\left[-\frac{1-n}{1-2U/3}\right]}{(U+3)^{1+\frac{4}{3}(1-n)} \left(\frac{3}{2}-U\right)^{2+\frac{5}{3}(1-n)}}, & U < \frac{3}{2} \\ 0, & U \geq \frac{3}{2} \end{cases} \quad (21)$$

$$P_4(U) = \begin{cases} C_4 \left(\frac{3-n}{4}\right) U^3 \exp\left\{\frac{2}{3} \frac{n-3/4}{1-3U/4} - \frac{\sqrt{2}}{9} \left(\frac{3}{4}-n\right)\right\} \times \\ \times \arctg\left[\frac{1}{\sqrt{2}} \left(1 + \frac{3}{4} U\right)\right] \Big\} (3-4n) \times \\ \left(U^2 + \frac{8}{3} U + \frac{16}{3}\right)^{\frac{11}{9} \left(n-\frac{3}{4}\right)-1} \left(\frac{4}{3}-U\right)^{\frac{14}{9} \left(n-\frac{3}{4}\right)-2}, & U < 4/3 \\ C_4 = \frac{4^4}{3^{25/9}} \exp\left(\frac{2}{3} + \frac{\sqrt{2}}{9} \arctg \frac{1}{\sqrt{2}}\right), & U \geq 4/3 \end{cases} \quad (22)$$

Here  $U = R/R_{ki}$ . For closed systems, in the expressions (20)-(22) a dumping index should be set as  $n=0$ .

The density of islands  $N_{i(t)}$  varies as:

$$N_i(t) = \frac{N_i(0)}{(3/p-n)(A_{pi} t)^{(3/p-n)}}. \quad (23)$$

The critical and average radii obey the law:

$$\begin{aligned} R_{ki}^p &\sim A_{pi} t; \\ \bar{R}_i &= R_{ki} C_{pn}, \end{aligned} \quad (24)$$

where  $C_{pn}$  is the coefficient depending on the heat mass transfer mechanism and the heat sink power. For definition of  $C_{pn}$ , see book [3].) The equations (19)-(24) coincide formally with the corresponding expressions obtained in [3]. Thus we can see that the basic features of the Ostwald ripening of island film persist in the case of the eutectic melt as well.

Now let us discuss briefly the processes which occur in the system of interest when its heat sinks are

intensive. Since, as was shown above, the system of equations for open systems in our case coincides with that examined in paper [5], the conclusions drawn there for the evolution at intensive heat sinks should be valid as well. It should be noted that the Ostwald ripening process can be disturbed when heat sink power exceeds a limiting value. This limiting value was determined in paper [5]. In this case the film may be amorphized.

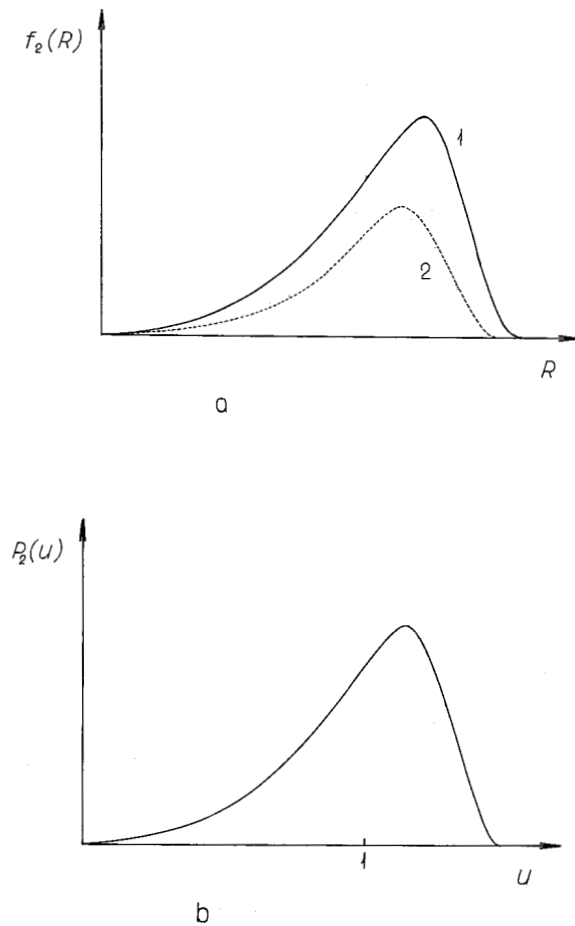
## 5. DISCUSSION OF RESULTS

Thus we have shown that a kind of symmetry is established in evolution of the ensemble of both phase islands when the island film is grown from the eutectic composition melts. The matter fluxes to an island became of equal values but opposite in sign, and growth of islands of both phases is caused by a reduction of the supercooling,  $\Delta T$ , relative to the eutectic temperature  $T_e$ . At the same time, the diffusion processes can limit the evolution. At the early evolution stages the mutual influence of the ensembles is utterly small. It is confirmed by the experimental studies [2] which show that the process of precipitation is usually initiated by one of the phase called as "leading phase". The evolution becomes correlated only at the stage of Ostwald ripening.

We have shown that in a description of the process of Ostwald ripening at the eutectic point one should use generalized coefficients of diffusion and boundary flux. The critical radii of islands of both phases are mutually related and similar. It was proved that certain relations exist between the constants of individual matters forming the eutectic system (see Table 1), which characterize predisposition of the system to the specific type of heat mass transfer. Also, it is evident from our study that a generalized distribution function  $P_p(u)$  does not depend on the initial size distribution of islands and is unique for both phases in the space of non-dimensional variables  $u = R/R_k$ . As for dimensional variables, the distribution functions are different for each phase (see Fig.2).

Most of papers dealing with experimental investigations of the eutectic melts disregard the size distribution of islands by restricting themselves studying the average radius change. As shown here, as well as in other works devoted to Ostwald ripening, interpretation of the processes involved requires to ascertain a probable mechanism of heat mass transfer using the analysis by Table 1.

From our theoretical consideration it follows that the structure of the film formed is affected to a considerable extent by the power of the heat sink from the system. In particular, in the case with  $p = 3$ , which corresponds, for instance, to the mechanism of heat



**Fig. 2.** The form of island size distribution function. (a) The functions of island distribution in dimensional variables (19). Curve lines 1 and 2 correspond to the phases 1 and 2, respectively. (b) The functions of island distribution in non-dimensional variables (20). The distribution function becomes common for both phases.

transfer through the bulk phase and diffusion transfer over the surface, it will imply an undamped sink, in which case the film structure, according to [3,6] is fine-dispersed.

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