# Distribution of Components between Immiscible Melts of a System under Nonisothermal Conditions

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**Abstract**—The influence of the temperature difference at the interface on the redistribution of components between coexisting liquid phase is analyzed using the U–Zr–O system as an example. It is demonstrated that, in this system, there can arise new dynamic effects in boundary regions of the coexisting phases. These effects are of considerable practical importance, for example, for the prediction of the behavior of the system in severe accidents at nuclear power plants.

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## INTRODUCTION

In real systems, there can arise a situation where two or several coexisting phases have different temperatures for a long time. If this time interval is longer than the relaxation time of mass transfer of the components, there can occur a stationary distribution of components between phases in the system. This situation can arise in a system formed by the coexisting liquid or liquid and gaseous phases (i.e., phases with a high mobility of components) in the case where the rates of heat release in the coexisting phases are different, whereas the rate of heat removal through the boundary of the system is constant.

The existence of hypothetical or real systems with a similar behavior can be illustrated by the following examples. Severe accidents at nuclear power plants with melting of the core of the nuclear reactor can be accompanied by the formation of a melt pool that has a stratified structure. A lighter layer, for example, enriched in metal components, such as zirconium and iron, is located at the top, and a melt containing uranium, zirconium, and iron oxides is located below. After a time, the temperature difference at the interface of liquid phases reaches a steady-state value (which can be as large as several hundred degrees) due to the energy release in the liquid phases, heat removal through the walls of the reactor vessel, and active stirring within each stratified phase [1]. A similar situation arises with a material in which layers of immiscible liquid phases are formed upon induction melting in a cold crucible [2]. In this case, the difference between the temperatures of the coexisting liquid phases is a function of the rate of heat release in each phase and the rate of heat removal through the interfaces.

The appearance of a constant temperature difference between the coexisting phases can affect the redistribution of components between them. Information on the distribution of components between stratified melts with different temperatures is of practical interest. In particular, in the case of a severe accident with melting of the core of the nuclear reactor, this information makes it possible to predict more reliably the accident scenario, as well as the intermediate and final structures of the melt pool [3]. When studying phase equilibria with the use of cold-crucible induction melting [4], the inclusion of nonisothermality of the system allows one to interpret more correctly the experimental results.

The thermodynamic approach to calculation of the distribution of components between coexisting phases under conditions of incomplete thermodynamic equilibrium in a system has long been in use. For example, the thermodynamic method of similar pseudoequilibrium states was developed and used in [5–7] for calculating the distributions of components between coexisting phases with a limited diffusion mobility of one or several components. This method has found wide application in analyzing the distribution of components between coexisting minerals in rocks. Pervukhin [8] analyzed the state of systems in which the mechanical, thermal, and phase equilibria are attained, but the chemical equilibrium has no time to be established (chemical affinity $A \neq 0$). In our previous paper [9], we demonstrated the possibility of calculating the redistribution of components between phases under temperature-gradient conditions. It should be noted that, as was shown in [8, 9], the results obtained in analysis of similar pseudoequilibria within the approach based on the methods of classical thermodynamics are virtually

identical to those determined with use of nonequilibrium thermodynamic approaches [10]. The above examples indicate that the methods of classical thermodynamics in a number of cases can be used to analyze pseudoequilibrium states in systems. The currently developed apparatus of the thermodynamics of irreversible processes [10, 11] permits one to examine more correctly (with a minimum number of model assumptions) the problem regarding the distribution of components in coexisting phases when a stationary state with a temperature difference between phases is attained in the system.

Therefore, the analysis of the influence of the temperature difference between stratified melts on the redistribution of components in these melts is an important problem. In this paper, the distribution of components in oxide and metal melts in the absence and presence of the temperature difference between these phases is analyzed using the U–Zr–O system as an example.

## SIMULATION AND CALCULATIONS

Since the aim of this study is to reveal the contribution of the nonisothermality of the system to the distribution of components between coexisting phases rather than to develop a thermodynamic model that can exactly describe the distribution of components between coexisting phases in a real system, as a first approximation, we consider the U–Zr–O system consisting of two melts, namely, the $(U,Zr)O_{2-x}^{ox}$ oxide and $(U,Zr)O_y^m$ metal melts. Hereafter, the superscript will indicate the correspondence of a particular quantity to the oxide (ox) or metal (m) melt. This representation of the formulas is explained by the nonstoichiometry of the oxide melt with respect to oxygen and by the presence of dissolved oxygen in the metal melt. These phases can coexist at a specific ratio of the components in the U–Zr–O system (Fig. 1). The redistribution of the components between the coexisting phases in the system is associated with the heterophase chemical reaction

$$UO_{2-x}^{ox} + ZrO_y^m = UO_y^m + ZrO_{2-x}^{ox}. \quad (1)$$
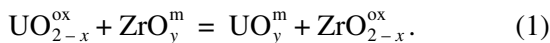
Under thermodynamically equilibrium conditions, the distribution of the components between the oxide and metal melts can be described by the equation

$$R\ln\frac{a_{UO_{2-x}}^{ox} \cdot a_{ZrO_y}^m}{a_{UO_y}^m \cdot a_{ZrO_{2-x}}^{ox}}$$

$$= -\frac{\mu_{UO_{2-x}}^{0^{ox}}(T) - \mu_{ZrO_{2-x}}^{0^{ox}}(T) - \mu_{UO_y}^{0^m}(T) + \mu_{ZrO_y}^{0^m}(T)}{T} \quad (2)$$

$$= -\frac{\Delta G_1^0(T)}{T},$$
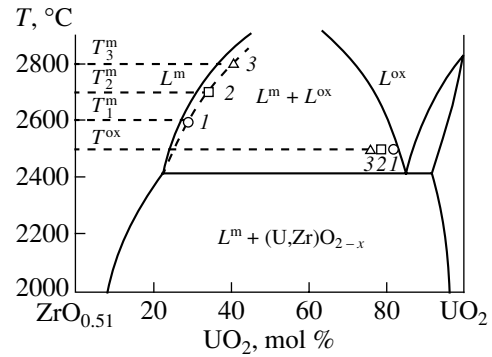


**Fig. 1.** Phase relationships in the $ZrO_{0.51}$–$UO_2$ system. Solid lines indicate the high-temperature region of the equilibrium phase diagram according to the data taken from [12]. Points *1–3* represent the calculated compositions in the case of the temperature difference at the interface. Designations: $T_i^m$ ($i = 1$–$3$) and $T^{ox}$ are the temperatures of the coexisting metal and oxide melts, respectively.
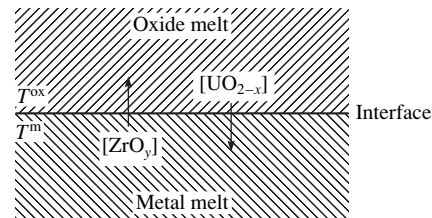


**Fig. 2.** Redistribution of the components between the oxide and metal melts due to the temperature difference at the interface ($T^m > T^{ox}$). Designations: $[ZrO_y]$ and $[UO_{2-x}]$ are the concentrations of the corresponding components in the metal and oxide melts. Arrows indicate the direction of mass transfer in the coexisting phases.

where $a_{UO_{2-x}(ZrO_{2-x})}^{ox}$ is the activity of $UO_{2-x}(ZrO_{2-x})$ in the oxide melt, $a_{UO_y(ZrO_y)}^m$ is the activity of $UO_y(ZrO_y)$ in the metal melt, $\mu_{UO_{2-x}(ZrO_{2-x})}^{0^{ox}}(T)$ is the standard value of the chemical potential of $UO_{2-x}(ZrO_{2-x})$ in the oxide melt, $\mu_{UO_y(ZrO_y)}^{0^m}(T)$ is the standard value of the chemical potential of $UO_y(ZrO_y)$ in the metal melt, $T$ is the temperature (K), $R$ is the universal gas constant, and $\Delta G_1^0(T)$ is the change in the standard Gibbs energy of reaction (1).

Now, we consider the case where the coexisting phases have different temperatures so that the temperature within each phase is constant (Fig. 2) and the temperature difference occurs at the interface. Let us assume that processes responsible for the maintenance of the temperature difference between the phases, i.e.,

irreversible thermal processes, proceed in the system. Moreover, mass transfer processes between the coexisting phases and chemical reactions can also occur in the system. We also assume that the system is in a state close to equilibrium. This implies that it is possible to use the linear nonequilibrium thermodynamic approximation when the generalized thermodynamic fluxes are proportional to the thermodynamic forces and the Onsager reciprocal relations [13] are satisfied. By using the theorem of minimum entropy production [14] with due regard for the Curie symmetry principle (which suggests the absence of reciprocity between chemical reactions and heat or mass transfer processes) and ignoring the reciprocity coefficients describing the interference between diffusion processes of different components, it can be easily shown that the condition for the stationary state of the system takes the form

$$
\left( \frac{\hat{a}^{\mathrm{ox}}_{\mathrm{UO}_{2-x}}(T^{\mathrm{ox}}) \cdot \hat{a}^{\mathrm{m}}_{\mathrm{ZrO}_y}(T^{\mathrm{m}})}{\hat{a}^{\mathrm{m}}_{\mathrm{UO}_y}(T^{\mathrm{m}}) \cdot \hat{a}^{\mathrm{ox}}_{\mathrm{ZrO}_{2-x}}(T^{\mathrm{ox}})} \right)_{\mathrm{noneq}}
$$

$$
= \left( \frac{a^{\mathrm{ox}}_{\mathrm{UO}_{2-x}} \cdot a^{\mathrm{m}}_{\mathrm{ZrO}_y}}{a^{\mathrm{m}}_{\mathrm{UO}_y} \cdot a^{\mathrm{ox}}_{\mathrm{ZrO}_{2-x}}} \right)_{\mathrm{eq}(T=T^{\mathrm{m}})} \cdot K(T^{\mathrm{ox}}, T^{\mathrm{m}}), \tag{3}
$$

where

$$
K(T^{\mathrm{ox}}, T^{\mathrm{m}})
$$

$$
= e^{\frac{\delta_{\mathrm{UO}_y} - \delta_{\mathrm{ZrO}_y}}{R}} \times e^{-\frac{1}{R}\left(\mu^{0^{\mathrm{ox}}}_{\mathrm{UO}_{2-x}}(T^{\mathrm{m}}) - \mu^{0^{\mathrm{ox}}}_{\mathrm{ZrO}_{2-x}}(T^{\mathrm{m}})\right)\left(\frac{1}{T^{\mathrm{ox}}} - \frac{1}{T^{\mathrm{m}}}\right)}
$$

$$
\times e^{-\frac{\int_{T^{\mathrm{m}}}^{T^{\mathrm{ox}}} \left(\bar{c}^{\mathrm{ox}}_{p_{\mathrm{ZrO}_{2-x}}} - \bar{c}^{\mathrm{ox}}_{p_{\mathrm{UO}_{2-x}}}\right)\left(1 - \frac{T^{\mathrm{m}}}{T}\right)dT}{RT^{\mathrm{ox}}}}. \tag{4}
$$

Here, $\hat{a}^{\mathrm{ox}}_{\mathrm{UO}_{2-x}(\mathrm{ZrO}_{2-x})}(T^{\mathrm{ox}})$ is the activity of $\mathrm{UO}_{2-x}(\mathrm{ZrO}_{2-x})$ in the oxide melt, $\hat{a}^{m}_{\mathrm{UO}_y(\mathrm{ZrO}_y)}(T^{\mathrm{m}})$ is the activity of $\mathrm{UO}_y(\mathrm{ZrO}_y)$ in the metal melt under nonequilibrium conditions, $\bar{c}^{\mathrm{ox}}_{p_{\mathrm{UO}_{2-x}(\mathrm{ZrO}_{2-x})}}$ is the partial heat capacity at a constant pressure for $\mathrm{UO}_{2-x}(\mathrm{ZrO}_{2-x})$ in the oxide melt, and $\delta_{\mathrm{UO}_y}$ and $\delta_{\mathrm{ZrO}_y}$ are parameters dependent on the reciprocity coefficients and proper coefficients [10].

As follows from relationships (3) and (4), the difference between the equilibrium (at $T = T^{\mathrm{m}}$) and stationary distributions of the components can be associated with two groups of factors. Factors of the first group are governed by the Soret effect [15] in the course of heat and mass transfer [the first factor in relationship (4)]. The contribution of this effect to the difference in the distribution of the components between the coexisting

phases under equilibrium and temperature-gradient conditions is determined by the expression

$$
\delta_{\mathrm{UO}_y} - \delta_{\mathrm{ZrO}_y}
$$

$$
= -\left( \frac{1}{T^{\mathrm{ox}}} - \frac{1}{T^{\mathrm{m}}} \right)\left( \frac{L_{\mathrm{th,\,dif}(\mathrm{UO}_y)}}{L_{\mathrm{dif}(\mathrm{UO}_y)}} - \frac{L_{\mathrm{th,\,dif}(\mathrm{ZrO}_y)}}{L_{\mathrm{dif}(\mathrm{ZrO}_y)}} \right). \tag{5}
$$

Here, $L_{\mathrm{th,\,dif}(\mathrm{UO}_y)}$ and $L_{\mathrm{th,\,dif}(\mathrm{ZrO}_y)}$ are the reciprocity coefficients and $L_{\mathrm{dif}(\mathrm{UO}_y)}$ and $L_{\mathrm{dif}(\mathrm{ZrO}_y)}$ are the proper phenomenological coefficients, which relate the mass transfer $J_{\mathrm{dif}}$ to the temperature and chemical potential gradients [10]:

$$
J_{\mathrm{dif}(\mathrm{UO}_y,\,\mathrm{ZrO}_y)} = L_{\mathrm{th,\,dif}(\mathrm{UO}_y,\,\mathrm{ZrO}_y)} \frac{\partial T^{-1}}{\partial h}
$$

$$
- L_{\mathrm{dif}(\mathrm{UO}_y,\,\mathrm{ZrO}_y)} \frac{\partial(\mu_{(\mathrm{UO}_y,\,\mathrm{ZrO}_y)} T^{-1})}{\partial h}, \tag{6}
$$

where $h$ is the spatial coordinate. This effect, as a rule, is insignificant. As was shown by Shewmon [16] and Sawatzky [17], the magnitude of this effect, at least, for the iron–carbon and uranium–hydrogen systems, i.e., the systems similar in chemical nature to the systems under consideration, is so small that, to a first approximation, it can be disregarded.

Factors of the second group are associated with the difference between the thermodynamic properties of the components at the temperatures $T^{\mathrm{ox}}$ and $T^{\mathrm{m}}$. It should be noted that the contribution represented by the second and third factors in relationship (4) can be obtained in terms of the concept of attainment of a pseudoequilibrium in the system. Furthermore, that the differences between the equilibrium and stationary distributions of the components under temperature-gradient conditions are also implicitly associated with the differences between the activities of the components at different temperatures. By explicitly separating this contribution to the difference between the equilibrium and temperature-gradient distributions of the components in the coexisting phases and assuming that the activity coefficients ($\gamma_i$) under equilibrium conditions $a_i = \gamma_i \cdot x_i$ and under nonequilibrium conditions $\hat{a}_i = \hat{\gamma}_i \cdot \hat{x}_i$ have the same form $[\hat{\gamma}_i(T, \{x_j\}) = \gamma_i(T, \{x_j\})]$, expressions (3) and (4) can be represented as follows:

$$
\left( \frac{\hat{x}^{\mathrm{ox}}_{\mathrm{UO}_{2-x}} \cdot \hat{x}^{\mathrm{m}}_{\mathrm{ZrO}_y}}{\hat{x}^{\mathrm{m}}_{\mathrm{UO}_y} \cdot \hat{x}^{\mathrm{ox}}_{\mathrm{ZrO}_{2-x}}} \right)_{\mathrm{noneq}}
$$

$$
= \left( \frac{x^{\mathrm{ox}}_{\mathrm{UO}_{2-x}} \cdot x^{\mathrm{m}}_{\mathrm{ZrO}_y}}{x^{\mathrm{m}}_{\mathrm{UO}_y} \cdot x^{\mathrm{ox}}_{\mathrm{ZrO}_{2-x}}} \right)_{\mathrm{eq}(T=T^{\mathrm{m}})} \cdot K_\gamma \cdot K(T^{\mathrm{ox}}, T^{\mathrm{m}}), \tag{7}
$$

where

$$K_{\gamma} = \frac{\gamma^{ox}_{UO_{2-x}}(T^m, x^{ox}_{UO_{2-x}}) \cdot \gamma^m_{ZrO_y}(T^m, x^m_{UO_y})}{\gamma^m_{UO_y}(T^m, x^m_{UO_y}) \cdot \gamma^{ox}_{ZrO_{2-x}}(T^m, x^{ox}_{UO_{2-x}})}$$

$$\times \frac{\gamma^m_{UO_y}(T^m, \hat{x}^{ox}_{UO_y}) \cdot \gamma^{ox}_{ZrO_{2-x}}(T^{ox}, \hat{x}^{ox}_{UO_{2-x}})}{\gamma^{ox}_{UO_{2-x}}(T^{ox}, \hat{x}^{ox}_{UO_{2-x}}) \cdot \gamma^m_{ZrO_y}(T^m, \hat{x}^m_{UO_y})}. \quad (8)$$

In the case where the influence of the difference in the phase composition under equilibrium and nonequilibrium conditions on the quantities $\gamma^{ox(m)}$ can be ignored, relationship (8) takes the form

$$K_{\gamma} \approx \frac{\gamma^{ox}_{UO_{2-x}}(T^m, x^{ox}_{UO_{2-x}})}{\gamma^{ox}_{UO_{2-x}}(T^{ox}, \hat{x}^{ox}_{UO_{2-x}})} \cdot \frac{\gamma^{ox}_{ZrO_{2-x}}(T^{ox}, \hat{x}^{ox}_{UO_{2-x}})}{\gamma^{ox}_{ZrO_{2-x}}(T^m, x^{ox}_{UO_{2-x}})}. \quad (9)$$

The quantity $K_{\gamma}$ can make a considerable contribution to the difference between the equilibrium and temperature-gradient distributions of the components only at a sufficiently large difference in the temperatures of the coexisting phases. This contribution can appear to be especially large if the structure of the melt undergoes substantial transformations with a change in the temperature. As follows from numerous investigations [18–20], similar transformations occur in the vicinity of the melting temperature.

The calculation of the distribution of the components between the coexisting phases (the oxide and metal melts) in the U–Zr–O system with the use of relationship (7) in the case where there is a temperature difference at the interface requires data on the distribution of the components under thermal equilibrium conditions in the system. The data on the equilibrium distribution of the components were taken from [12]. The uranium distribution between the metal and oxide melts in the case where the temperatures of the phases are different was calculated using the derived expressions. The results of the calculations are presented in Fig. 3.

## RESULTS AND DISCUSSION

The analysis of the results obtained demonstrates that the temperature difference between the coexisting phases can substantially affect the redistribution of the components between them (Fig. 3).

As follows from the analysis of the calculated distribution of the components in the oxide and metal melts, an increase or a decrease in the temperature of the metal melt with respect to the temperature of the oxide melt leads to an increase in the content of a particular component in the metal and oxide melts as compared to the corresponding content in the melts under thermal equilibrium conditions (Fig. 3). The calculations show (Fig. 3) that, although the change in the concentration of the components in the coexisting phases in the presence of the temperature difference at the interface is insignificant as compared to the corresponding concen-
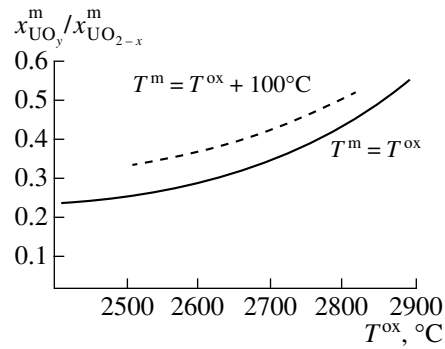
**Fig. 3.** Distributions of the components between the coexisting phases under thermal equilibrium conditions at $T^m = T^{ox}$ (solid line) and in the case of the temperature difference at the interface $T^m = T^{ox} + 100°C$ (dashed line).
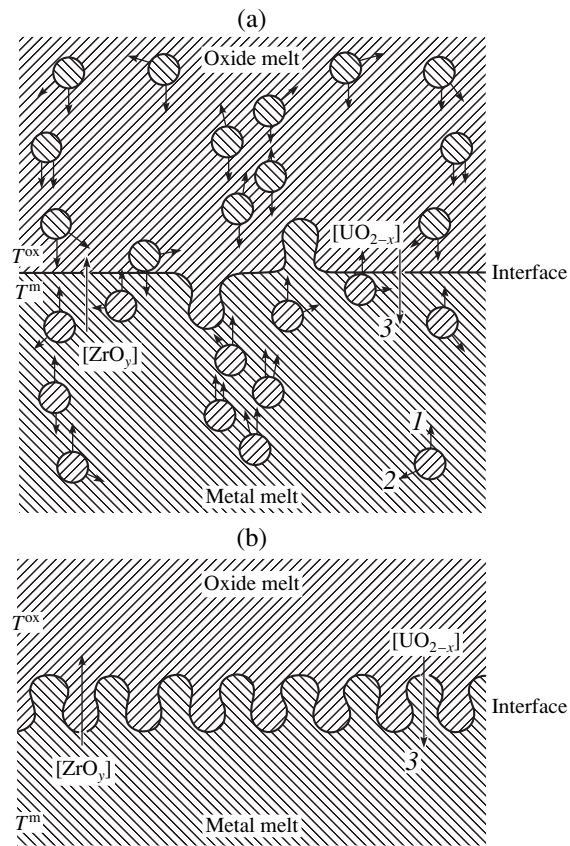
**Fig. 4.** Different configurations of phase inhomogeneities in the coexisting melts as a function of the ratio between the rates of diffusion of the components through the interface, the rates of formation and growth of nuclei of new phases in the interface region, and the velocities of motion of local domains of these phases: (a) strong convective flows and phase formation in regions both adjacent to the interface of the immiscible melts and far from it and (b) the high rate of phase formation in the region immediately adjacent to the melt interface. Arrows *1*, *2*, and *3* indicate the direction of the buoyancy and surface tension forces, the direction of convective flows, and the direction of mass transfer of the components in the coexisting phases, respectively.

trations under thermal equilibrium conditions, a number of new dynamic effects can be observed in the system. First and foremost, it should be noted that the phase separation can occur in the interface region due to the supersaturation of the melt with one of the components (Fig. 3). The dynamics of the phase separation can be described using different models, for example, the model proposed by Kovalenko [21]. In the present paper, we will not describe quantitatively the dynamic processes in the coexisting phases and dwell only on the qualitative effects that can be observed in the system under consideration. In the coexisting melts, there can arise different configurations of phase inhomogeneities depending on the ratio between the rates of diffusion of the components through the interface, the rates of formation and growth of nuclei of new phases in the interface region, and the velocities of motion of local domains formed by these phases (Fig. 4). The local domains of the newly formed phases can be transferred either toward the interface under the buoyancy and surface tension forces or in the direction of convective flows inside the liquid phases (Fig. 4a). A specific configuration of the dynamic phase distribution is formed upon spinodal (fast) phase decomposition in the vicinity of the interface as a result of the possible considerable supersaturation in the interface region. In this case, there arises a new type of instability of the planar interface due to the phase instability of the boundary regions (Fig. 4b).

## CONCLUSIONS

Thus, it has been demonstrated that a long-term temperature difference at the interface of two immiscible liquids brings about the exchange of their components. This leads to the phase separation in the coexisting melts in the interface region and, under specific conditions, to the formation of a dynamic nonplanar interface.

## ACKNOWLEDGMENTS

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