

Diffusion Behavior and Brown Motion in Many Elements System

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Abstract

In a series of diffusion studies, some new diffusion theories different from the existing ones have been reported. One of their theories revealed that the concept of intrinsic diffusion, which is conceived to understand the well-known Kirkendall effect relevant to the interdiffusion, is an illusion. Based on the theory, a new interdiffusion theory is established as a Brown motion in many elements system, regardless of the diffusion field (solvent) in a crystal material or an amorphous one. At the same time, the vacancy diffusion equation of interdiffusion problems in crystal is also presented. The present analytical system is useful for analyzing actual interdiffusion problems in the many elements system. By means of investigating vacancy behavior in the diffusion field composed of many elements, a more accurate analysis of interdiffusion problems will be possible.

Keywords: interdiffusion, many elements system, Brown motion, diffusion equation

1. Introduction

Recently, some new diffusion theories have been proposed in accordance with the fundamental theory in mathematics and physics (Okino, 2011, 2012a, 2012b, 2013a, 2013b). One of them revealed that the well-known Darken theory is wrong in relation to the fundamental theory in mathematics and physics (Darken, 1948; Okino, 2012b). Based on the new theory, we thus proposed a new interdiffusion mechanism in the binary system and also the theoretical equation relevant to the well-known Kirkendall effect (K-effect) was then reasonably obtained (Smigelskas & Kirkendall, 1947; Okino, 2012b, 2013b). In other words, it was revealed that the intrinsic diffusion concept is an illusion caused by the incompleteness of the Fickian first law. Nevertheless, the Darken equation is still used for analyzing interdiffusion problems. For example, it is reported that its equation is not valid in an analysis of interdiffusion problems of mass in liquid metals and alloys (Meyer & Kargl, 2013).

In the present study, the interdiffusion problems containing N (≥ 2) elements in the diffusion region are investigated. It was found that a diffusion particle moves in a common diffusion field as a Brownian particle in many elements system, where the diffusion field is not concerned in a crystal or an amorphous material. The K-effect in a metal alloy shows that the vacancy concentration yields the supersaturated section and the unsaturated one in the diffusion region, although it has never been explicitly investigated. The diffusion equation of vacancy concentration to investigate its behavior is derived in the present work.

The interdiffusion study is indispensable for fundamental understanding of mesoscopic systems such as nanoparticles, nanostructured materials, multilayered thin films and so on. Further, understanding of interdiffusion behavior is also important to the actual fabrication process of a compound semiconductor and a metal alloy composed of many elements. The diffusion equation of N elements system and that of vacancy concentration obtained here are applicable to various interdiffusion problems. From the new point of view, it is thus considered that the present results are useful for the fundamental diffusion study of various compound materials, even if they are in a solid state, a liquid state or a gas state.

Here, we briefly summarize the previous works. The Fickian diffusion equation denotes the behavior of diffusion materials in the diffusion field (solvent). The diffusivity depends on an interaction between a diffusing particle and the diffusion field. The coordinate system of the Fickian diffusion equation is thus set in the diffusion field. For example, it is expressed as

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x} \left\{ D \frac{\partial C}{\partial x} \right\} \quad (1)$$

in the time and space (t, x) for one dimensional space, where C and D are the concentration and diffusivity. When the diffusion field moves, it is apparent that the coordinate system (t, x) becomes the expression of the moving coordinate system. On the other hand, experimental results are usually obtained as ones of the fixed coordinate system (τ, ξ) in the outside of diffusion system. Therefore, there is a difference between analytical results in the moving coordinate system and experimental results.

In the initial state, the origin of fixed coordinate system $(\tau, \xi) = (0, 0)$ coincides with that of moving coordinate system $(t, x) = (0, 0)$. When the velocity of $(t, x) = (0, 0)$ is v with respect to $(\tau, \xi) = (0, 0)$ at an arbitrary time t , the relation of

$$t = \tau, \quad x = \xi - \Delta x_{\text{eff}} \quad \text{for} \quad \Delta x_{\text{eff}} = \int_0^t v dt \quad (2)$$

is valid. The Fickian diffusion equation and the diffusion flux in the fixed coordinate system is written as

$$\partial_{\tau} C = \partial_{\xi} (D \partial_{\xi} C - vC) \quad \text{and} \quad J_{\text{R}} = -D \partial_{\xi} C + vC, \quad (3)$$

where $\partial_{\tau} = \partial / \partial \tau$ and $\partial_{\xi} = \partial / \partial \xi$. The diffusion flux J_{R} obtained from the divergence theorem is not proportional to the concentration gradient. It is thus apparent that the Fickian first law is not valid in the fixed coordinate system, although it is generally invalid also in the moving coordinate system in accordance with the initial and/or boundary condition (Okino, 2012b).

In the interdiffusion problem, all researchers believe that the interdiffusion coefficient \tilde{D} relevant to the diffusivities D^{I} and D^{II} of equations for the concentrations C^{I} and C^{II} in the binary system is expressed as $\tilde{D}(t, x) = D^{\text{I}}(t, x) = D^{\text{II}}(t, x)$ in the moving coordinate system. In relation to the K-effect, however, they have also believed the well-known Darken equation in the moving coordinate system since 1948. On the contrary, it was directly revealed in the previous works that the Darken equation is invalid in mathematics (Okino, 2013b). Here, we can also understand the wrong of the Darken equation through a simple procedure. In the coordinate transformation theory, that is, the relation between $D^{\text{I}}(\tau, \xi)$ and $D^{\text{II}}(\tau, \xi)$ in the fixed coordinate system is obtained as $\tilde{D}(\tau, \xi) = D^{\text{I}}(\tau, \xi) = D^{\text{II}}(\tau, \xi)$ only by substituting Equation (2) into $\tilde{D}(t, x) = D^{\text{I}}(t, x) = D^{\text{II}}(t, x)$. There is thus no room to conceive the intrinsic diffusivity as a new material property in the diffusion system. At the same time, it is apparent that the K-effect is equivalent to Δx_{eff} of Equation (2). In mathematics, the K-effect is a mere shift between the coordinate systems (t, x) and (τ, ξ) , regardless of the composition and structure of materials. In physics, it depends on the difference between the diffusion fluxes caused by initial and/or boundary values of C^{I} and C^{II} at the diffusion region interface (Okino, 2012b).

2. Diffusion Problems of N Elements System in Amorphous Materials

In this section, the diffusion specimen A and B are composed of amorphous materials. The specimen A contains L ($L \leq N$) elements and the specimen B contains M ($M \leq N$) elements. The specimens A and B are used as a diffusion couple and there are N elements in the diffusion region. We set the space coordinate $x = 0$ at the interface between the specimens A and B in the initial state at $t = 0$. In the diffusion region of $x_{\text{A}} \leq x \leq x_{\text{B}}$ for the diffusion time $t > 0$, the concentrations of diffusion elements are defined as $C^{\text{I}}, C^{\text{II}}, \dots, C^{\text{N}}$ in the moving coordinate system (t, x) .

In the present diffusion system, we assume that the atomic sizes of diffusion particles in the diffusion region are roughly equal to each other. We then estimate a specimen cross section perpendicular to x axis where each cross section interval corresponds to the averaged jump distance in the solid state or the averaged collision distance in the gas or liquid state. If the cross section of diffusion couple is uniform and the shape variation of diffusion couple is negligible during the thermal diffusion, the normalized concentration yields

$$\sum_{j=1}^N C^j = 1, \quad (4)$$

on each cross section. The diffusion equation for an element $j = k$ is

$$\partial_t C^k = \partial_x (D^k \partial_x C^k), \quad (5)$$

where D^k is a diffusivity of the element $j = k$.

Equations (4) and (5) yield

$$\partial_x \left(\sum_{j=1}^N D^j \partial_x C^j \right) = \partial_x \left(\sum_{j=1}^N (D^j - D^k) \partial_x C^j \right) = 0 \quad (6)$$

because of $\sum_{j=1}^N \partial_t C^j = \partial_t \sum_{j=1}^N C^j = 0$. Since it is apparent that $\partial_x C^j$ depends on x , the identical Equation (6)

shows $D^j - D^k = 0$ for an arbitrary j . The interdiffusion coefficient \tilde{D} is thus given by

$$\tilde{D} = D^1 = D^2 = \dots = D^N. \quad (7)$$

Equation (7) shows that each of the diffusion particles randomly moves as a Brownian particle in the common diffusion field. Equation (5) is thus rewritten as:

$$\partial_t C^k = \partial_x (\tilde{D} \partial_x C^k). \quad (8)$$

Using Equations (4) and (7) for the diffusion flux, the relation of

$$\sum_{j=1}^N J^j = - \sum_{j=1}^N D^j \partial_x C^j = - \tilde{D} \partial_x \sum_{j=1}^N C^j = 0 \quad (9)$$

is obtained in the moving coordinate system.

Based on the previous study, the new diffusion flux J_R^k for $j = k$ is defined as

$$J_R^k = J^k + J_S^k + J_{eq}^k, \quad (10)$$

in the fixed coordinate system (τ, ξ) which is set in the outside of diffusion system (Okino, 2012b). The diffusion region $\xi_A \leq \xi \leq \xi_B$ in the fixed coordinate system corresponds to $x_A \leq x \leq x_B$. In Equation (10),

J^k is the diffusion flux in the moving coordinate system and corresponds to the Fickian first law, and J_{eq}^k is

the intrinsic flux relevant to the Brown motion in the thermal equilibrium state. The relations of $\sum_{j=1}^N J_{eq}^j = 0$ is

similarly valid to the previous work (Okino, 2012b). Here, J_S^k is the diffusion flux relevant to the source of C^k generated at the diffusion region interface $\xi = \xi_A$ and $\xi = \xi_B$. The J_S^k corresponds to the incompleteness of the Fickian first law and further it is independent of the space coordinate x and/or ξ .

Using the boundary values of $C^j = C_A^j$ ($\xi \leq \xi_A$), $C^j = C_B^j$ ($\xi \geq \xi_B$) and $\tilde{D} = \tilde{D}_A$ ($\xi \leq \xi_A$), $\tilde{D} = \tilde{D}_B$ ($\xi \geq \xi_B$) at $\xi = \xi_A$ or $\xi = \xi_B$, the J_S^k is defined as

$$J_S^k = \sqrt{\tilde{D}_\gamma} \left\{ \sum_{j=1, j \neq k}^N [C_A^j - C_B^j] \right\} / 2\sqrt{t}, \quad (11)$$

and it becomes a binary system if $N=2$ (Okino, 2012b). In Equation (11), the suffix γ of \tilde{D}_γ means $\gamma \rightarrow A$ for $C_A^j > C_B^j$, $\gamma \rightarrow B$ for $C_A^j < C_B^j$.

From the above relations, the equation of

$$\sum_{k=1}^N J_R^k = (N-1) \sqrt{\tilde{D}_\gamma} \sum_{j=1}^N [C_A^j - C_B^j] / 2\sqrt{t} \quad (12)$$

is valid between diffusion fluxes because of Equation (9) and $\sum_{j=1}^N J_{eq}^j = 0$. Further, Equations (2), (3), (9), (10)

and (12) yield

$$v = (N-1)\sqrt{\bar{D}_\gamma} \sum_{j=1}^N [C_A^j - C_B^j] / 2\sqrt{t}. \quad (13)$$

The shift $\Delta\xi_{\text{sft}}$ caused by the coordinate transformation between (t, x) and (τ, ξ) is thus expressed as

$$\begin{aligned} \Delta\xi_{\text{sft}} &= \int_0^t |v| dt = (N-1)\sqrt{\bar{D}_\gamma} \int_0^t \left| \sum_{j=1}^N [C_A^j - C_B^j] \right| / 2\sqrt{t} dt \\ &= (N-1)\sqrt{\bar{D}_\gamma} \left| \sum_{j=1}^N [C_A^j - C_B^j] \right| \sqrt{t}. \end{aligned} \quad (14)$$

Hereinbefore, we discussed the interdiffusion problems of N elements system in amorphous materials. In general, the diffusion field moves and each of the diffusion particles randomly moves in the common diffusion field as if each of the gas molecules randomly moves in a common space as a Brownian particle in accordance with the well-known Boyle Charles law.

3. Diffusion Problems of N Elements System in Crystal Materials

When we replace the amorphous diffusion specimen in the previous section with a crystal one, it is considered that the vacancy behavior plays an important role in diffusion problems. In general, there is a difference $\Delta C^V = -(C_A^V - C_B^V)$ between vacancy thermal equilibrium concentration C_A^V ($\xi \leq \xi_A$) in the specimen A and C_B^V ($\xi \geq \xi_B$) in the specimen B. However, the influence of ΔC^V on the interdiffusion problems has been neglected in the existing investigations.

The K-effect reveals that the diffusing particles depend on the vacancy concentration and that the vacancy concentration yields the supersaturated section and the unsaturated one in the diffusion region. It is thus essentially important to understand the behavior of vacancies in crystal materials for diffusion problems.

In the same manner as Equation (11), the vacancy flux in the fixed coordinate system is given by

$$J_R^V = J^V - D_\gamma^V \frac{\Delta C^V}{2\sqrt{D_\gamma^V t}} = -D^V \partial_x C^V + \sqrt{D_\gamma^V} (C_A^V - C_B^V) / 2\sqrt{t}, \quad (15)$$

where D^V is the vacancy diffusivity in the diffusion region of $\xi_A \leq \xi \leq \xi_B$ (Okino, 2012b). The moved vacancy quantity caused by $\Delta J = J_R^V - J^V$ is obtained as:

$$\Delta\xi_{\text{eff}}^V = \int \Delta J dt = \sqrt{\bar{D}_\gamma^V} (C_A^V - C_B^V) \sqrt{t}. \quad (16)$$

The influence of $\Delta\xi_{\text{eff}}^V$ on the K-effect in the N elements system as well as the binary system may be negligible in the usual case. However, the influence is worthy to investigate itself.

If the vacancy thermal equilibrium concentration is written as C_0^V in the diffusion region, the vacancy generation/annihilation term given by

$$k_V (C_0^V - C^V)$$

contributes to the vacancy diffusion equation, where k_V is a chemical reaction constant relevant to the vacancy generation/annihilation. Thus, the vacancy diffusion equation is obtained as:

$$\partial_t C^V = \partial_x (D^V \partial_x C^V) + k_V (C_0^V - C^V). \quad (17)$$

The same diffusion equation of Equation (5) is valid also for the diffusion particles in the crystal materials.

Under the condition where the shape variation of the diffusion couple is negligible, the relation of

$$C^V + \sum_{j=1}^N C^j = 1 \quad (18)$$

is valid for the normalized concentration on each crystal cross section, including vacant lattice points. Since the

K-effect reveals that the vacancy concentration yields the supersaturated section and the unsaturated one in the diffusion region, the absolute value of diffusion flux,

$$\int_{x_A}^{x_B} k_V (C_0^V - C^V) dx,$$

is considered as a negligible one. We thus assume that the averaged contribution of the vacancy generation/annihilation term to the total diffusion flux in the diffusion system is negligible.

Using Equations (5), (17) and (18) for the diffusion flux, therefore, the relation of

$$J^V + \sum_{j=1}^N J^j = D^V \partial_x C^V + \sum_{j=1}^N D^j \partial_x C^j = \sum_{j=1}^N (D^j - D^V) \partial_x C^j = 0, \quad (19)$$

is approximately valid in the moving coordinate system because of neglecting the contribution of $k_V (C_0^V - C^V)$.

The identical Equation (19) gives

$$\tilde{D} = D^V = D^1 = D^2 = \dots = D^N, \quad (20)$$

because of $\partial_x C^j \neq 0$. Equation (20) shows that we can accept a vacancy in crystal as a virtual diffusion particle and that the suffix V in Equations (18) ~ (20) is accepted as $V \rightarrow N+1$.

The diffusion equations of $(N+1)$ dimensions system is thus established as

$$\partial_t C^j = \partial_x (\tilde{D} \partial_x C^j) \quad \text{for } j=1, 2, \dots, k, \dots, N. \quad (21)$$

$$\partial_t C^V = \partial_x (\tilde{D} \partial_x C^V) + k_V (C_0^V - C^V), \quad (22)$$

under the condition of Equation (18). Understanding of the vacancy behavior is thus possible for the diffusion problems in a crystal material.

In the same manner as Equation (14) derivation, the K-effect is expressed as

$$\begin{aligned} \Delta \xi_{\text{eff}} &= \int_0^t \left| \sum_{k=1}^{N+1} J_R^k \right| dt = N \sqrt{\tilde{D}_\gamma} \int_0^t \left| \sum_{j=1}^{N+1} [C_A^j - C_B^j] \right| / 2\sqrt{t} dt \\ &= N \sqrt{\tilde{D}_\gamma} \left| \sum_{j=1}^{N+1} [C_A^j - C_B^j] \right| \sqrt{t}, \end{aligned} \quad (23)$$

because of $V \rightarrow N+1$. It is apparent that the obtained $\Delta \xi_{\text{eff}}$ corresponds to the previous $\Delta \xi_{\text{stf}}$. The analytical solutions, $C(t, x)$ and $D(t, x)$ of Equation (21) are already obtained as a problem in the parabolic space (Okino, 2011, 2012a). They are

$$C^j(t, x) = C_m^j - C_\Delta^j \operatorname{erf} \left(\frac{x}{2\sqrt{D_{\text{int}}^j t}} - \frac{x_{\text{IN}}}{2\sqrt{D_{\text{int}}^j t}} + \operatorname{erf}^{-1} \left(\frac{C_m^j - C_{\text{IN}}^j}{C_\Delta^j} \right) \right) \quad (24)$$

and

$$\tilde{D}(t, x) = \tilde{D}_m - \tilde{D}_\square \operatorname{erf} \left(\frac{x}{2\sqrt{D_{\text{int}}^j t}} + \operatorname{erf}^{-1} \left(\frac{\tilde{D}_m - \tilde{D}_{\text{IF}}}{\tilde{D}_\square} \right) \right). \quad (25)$$

Using the boundary values of C_A^j , \tilde{D}_A and C_B^j , \tilde{D}_B at $x = x_A$ and $x = x_B$, the notations in Equations (24) and (25) are as follows (Okino, 2011).

$$\begin{aligned} C_m^j &= (C_A^j + C_B^j)/2, \quad C_\Delta^j = (C_A^j - C_B^j)/2, \quad C_{\text{IN}}^j = C_m^j - C_\Delta^j (\tilde{D}_m - \tilde{D}_{\text{IF}})/\tilde{D}_\Delta, \\ x_{\text{IN}} &= 2\sqrt{\tilde{D}_A \tilde{D}_B t} (\sqrt{\tilde{D}_A} - \sqrt{\tilde{D}_B}) / (\sqrt{\tilde{D}_A} + \sqrt{\tilde{D}_B}), \\ \tilde{D}_m &= (\tilde{D}_A + \tilde{D}_B)/2, \quad \tilde{D}_\Delta = (\tilde{D}_A - \tilde{D}_B)/2, \quad \tilde{D}_{\text{IF}} = (\tilde{D}_A - \tilde{D}_B) / (\ln \tilde{D}_A - \ln \tilde{D}_B), \end{aligned}$$

$$D_{\text{int}} = \tilde{D}_m \text{ for } x \geq 0, \quad D_{\text{int}} = D_{\text{int-}} = \sqrt{\tilde{D}_A \tilde{D}_B} \text{ for } x < 0.$$

Further, if we substitute Equation (2) into Equations (24) and (25), the solutions in the fixed coordinate system are obtained. After substituting Equation (25) into Equation (22), the vacancy behavior is also obtained by its numerical analysis.

4. Conclusion

In the interdiffusion problems, Equation (9) or (19) shows that the diffusion behavior of an element is considered as a motion in the diffusion field composed of the other elements in the moving coordinate system. Under the condition of Equation (4), Equation (7) shows in the diffusion region that an interaction between an arbitrary diffusion particle and the others is equivalent to each other. The only difference between their partial differential equations for each of the elements is one between their initial and/or boundary values. This gives an image so the relative speed between 2 bodies is equal to each other in the moving coordinate system regardless of their velocities in the fixed coordinate system. If we accept a vacancy as a virtual diffusion particle, the same discussion is also valid for Equation (20) under the condition of Equation (18). Equation (4) or (18) is thus strong condition for the diffusion system.

The above discussion shows that a diffusing particle in the moving coordinate system moves in the common interdiffusion field as if the individual property of gas molecules is negligible in the well-known Boyle Charles law, satisfying the energy equipartition law. If we image the well-known Brown motion, the particle motion in the interdiffusion system is reasonably accepted. The dependence of a crystal structure on diffusion behavior should be incorporated into the diffusivity as a configuration space effect and/or an entropy effect around a diffusing particle in the given diffusion system. In the actual interdiffusion problems, however, the detail investigation may be necessary for the interaction between a diffusing particle and the diffusion field so the Boyle Charles law corresponds to the van der Waals theory.

Equation (8) or (21) is independent of each other for an arbitrary element. It does not depend on the diffusion mechanism in the moving coordinate system. In other words, the present theory is applicable to not only the vacancy type mechanism but also other type mechanisms, only if the condition of Equation (4) or (18) is valid in the N elements system. The present analytical method is thus simply applicable to actual diffusion problems using the common diffusivity \tilde{D} , compared with the existing analytical method. For example, the interdiffusion behavior in a ternary gas mixtures system is investigated still using the complicated interdiffusion coefficient expressions (Kosso, Kulzhanov, Poyarkov, & Fedorenko, 2013).

The vacancy influence of Equation (16) on interdiffusion elements may be negligible in the usual case. The vacancy effect has never been thus considered in the existing analytical method of interdiffusion problems. However, the vacancy behavior will be necessary for the detail investigation of interdiffusion problems. The interdiffusion problem of Ni-Al multilayers is investigated by the method of molecular dynamic simulations (Xu, Falk, & Weihs, 2013). In the work, the temperature dependence of interdiffusion coefficients is reported, although generally the interdiffusion coefficient depends on a temperature, a diffusion time and a space coordinate. It is thus difficult to solve the interdiffusion problems in accordance with the previous diffusion concept. However, if we use the new relation of Equation (7) or (20) for interdiffusion problems and analyze them in the parabolic space, their solutions will be possible (Okino, 2011, 2012a).

It is, therefore, considered that the present analytical system will be useful for various interdiffusion problems of a many elements system. Further, the new fundamental theory of diffusion problems has been discussed in a series of diffusion studies (Okino, 2011, 2012a, 2012b, 2013a, 2013b). They will be useful for the diffusion study in future.

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