The calculation of the diffusion coefficients in ternary multiphase Ti-NiAl system

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Abstract. In this paper the problem of determination of the diffusion coefficients in a ternary multiphase system is discussed. In the literature there is a lack of data concerning the approximation method which allows to determine diffusion coefficients in such complicated systems. We propose the mass conservation method (MCA) for calculation the diffusivities in phases where the differences in composition is linear. Only one experimental results is needed for determination of the diffusion data. The method is implemented and checked under ternary Ti-NiAl system - this system characterize occurrence three intermetallic phases in diffusion couple Ti-NiAl.

1. Introduction. The most important parameter describing the diffusion process is diffusion coefficient, \( D_i \). First time it was defined by Fick in his famous laws for diffusion. The first Fick law describes the process with constant mass in time - the flux, Eq.1.

\[
j_i = -D_i \frac{\partial N_i}{\partial x}
\]  \hspace{1cm} (1)

where: \( j_i \) and \( N_i \) is the diffusion flux and molar ratio of the i-th component, respectively.

The second one describe how the mass changes with time during the diffusion process, Eq. 2.

\[
\frac{\partial N_i}{\partial t} + \frac{\partial j_i}{\partial x} = 0
\]  \hspace{1cm} (2)

Diffusion coefficient helps to describe the diffusion related phenomena like mechanical properties of materials - stress induced diffusion [1], changes in morphology - phase growth [2] and others. In many products like e.g. coatings, the diffusion controls the growth of intermetallic compounds. The diffusion coefficient can be determined experimentally, namely with use of the tracer and diffusion coupling method [3]. However, this methods are very difficult to use in multicomponent [4]. It is rather straightforward to use this method in binary systems. The methods are modified by introducing additional parameters like position of the Kirkendall plane - thus only one experiment is required for
determination of diffusion coefficient [4, 5, 6, 7, 8, 9, 10, 11, 12, 13]. Ternary and higher order systems are much more complicated for experimental studies. The only main and cross diffusion coefficient can be determined at the point of intersection of the composition profiles [14]. The main disadvantages of the above mention methods is that they can determine diffusion coefficient only inside one phase with full solubility. There is a lack of reasonable method which allows for experimental determination of diffusion coefficient in multicomponent and multiphase systems. Moreover, this experimental methods are time and costs consuming.

At the present times, when computer revolution occurs, the diffusion coefficients are try to be calculated by the numerical treatment. The numerical methods are much more effective and efficient compared with experimental ones. Moreover, for better accuracy the diffusion coefficient should be composition dependent [15, 13]. Thus, effective methodologies which can provide the reliable diffusion coefficients in alloys are needed.

The composition dependency of interdiffusion coefficients in binary and ternary alloys are extensively studied from Fujita and Gosting works [16] and later by other authors [17, 18, 19, 20, 21, 22]. Cermak and Rothova [23] tried to compute the composition dependent diffusion coefficient by divide the volume into small parts and integrate. Bouchet and Mevrel [19] proposed the inverse method to calculate the composition dependent intrinsic diffusion coefficients. These methods based on the Fick's second law. All of above presented methods bases on the mass conservation law, thus they are deterministic ones. However, stochastic methods based mainly on the genetic algorithms or monte carlo simulations can also be found in the literature. These methods are much more effective and can be used for calculating the diffusion coefficients in the whole range.

The situation dramatically changes when diffusion coefficients should be found in multiphase systems. In such case, literature is very limited. One of the method to calculate the diffusion coefficient in binary multiphase system was proposed by Wagner [24, 25]. The method allowed for calculating the average intrinsic diffusion coefficient over the whole range. The integral diffusion coefficient can be calculated from the following expression:

\[
\tilde{D}(N^*_A) = \frac{(N^+_A - N^-_A)V_B}{2t(\partial N_A/\partial x)_{x=x}} \left[ (1-Y^*) \int_{-\infty}^{\infty} \frac{y}{V} dy + Y^* \int_{-\infty}^{\infty} \frac{1-y}{V} dy \right]
\]

(3)

where: \( N^-_A \) and \( N^+_A \) are the molar ratio of A and B component at the boundary of the system, \( V = \frac{1}{V_c} \) is the volume of the system. The proportionality variable Y can be expressed as:
The Wagner's integral coefficient can be used in binary multiphase systems.

In the literature there is a lack of the methods for calculation of the diffusion coefficients in multicomponent (even ternary) multiphase systems. Thus in this paper the deterministic method of approximating the intrinsic diffusion coefficients based on the Leibnitz rule at the phase boundaries is presented. The equations can be solved by the iteration method. Thus the initial guess will be used to generate the sequence of improving approximate solutions. The stop criterion of the minimal error can be used. Thus the obtained intrinsic diffusion coefficients can be appropriate in multiphase systems. Moreover, the method will be checked in ternary Ti-NiCu system. The diffusion coefficients will be approximated.

2. Modeling

2.1. Pseudobinary approach (PBA) in a ternary system

As it was already mentioned, in the literature there is lack of methods, that enable to calculate diffusion coefficients in ternary systems. However a few attempts have been made to estimate those coefficients by implementing some simplifying assumptions. A. Paul proposed a method of calculating diffusion coefficient, which based on Wagner’s equations [4]. He considered a ternary A-B-C system and a diffusion couple, in which component C does not diffuse, but replaces B component in the alloy – concentration of C remains constant in diffusion process (table 1). Paul assumed further, that new phase \( \gamma \) grows in the diffusion process in the middle of initial blocks – composition of the components changes linearly in the interdiffusion zone within a range of \( \Delta N'_{Y} = 0.2 - 0.3 \) and such change does not cause a change in molar volume (Fig. 1). Thus Eq. 3 for calculation the interdiffusion coefficient may be reformulated as follows:

\[
\tilde{D} = \frac{1}{2t} \left( \frac{dX}{dY_A} \right) \left[ \left(1 - Y_A^K\right) Y_A^{x} dx + Y_A^K \left(1 - Y_A\right) dx \right]
\]

(5)

where \( K \) denotes the marker plane position. Considering the diffusion time of 25 hours and thickness of \( \gamma \) phase as 90 \( \mu \)m, the marker plane had been found at 40.5 \( \mu \)m from Alloy 1– \( \gamma \) phase interface of the diffusion couple (Fig. 2). Concentration of A component at the marker plane is 24.5 at. %.
Table 1. Initial composition of the components

|                | A concentration, at. % | B concentration, at. % | C concentration, at. % |
|----------------|------------------------|------------------------|------------------------|
| Alloy 1        | 15                     | 75                     | 10                     |
| Alloy 2        | 35                     | 55                     | 10                     |

Fig. 1. Diffusion couple of two alloys: a) before diffusion process, b) after annealing for 25h

Fig. 2. Composition N of elements: a) A, b) B and C, c) normalized composition of A element $Y_A$
Paul assumed that component B replaces the component C, so that the composition profile of A component does not change by addition of component C to the alloy. In case of calculation interdiffusion coefficient (Eq. 5), concentrations of B and C components have no influence on the result –composition range of the components of the couple is normalized (Eq. 4). For calculation of intrinsic diffusion coefficients however, the composition of components B and C is of significant importance when the relationship between the interdiffusion and intrinsic diffusion coefficients is taken into account:

\[ D^K = N_AD^K_B + N_BD^K_A \]  

(6)

In such a case, when concentration of C component is neglected, the equation of the total atomic fraction for ternary system: \( N_A + N_B + N_C = 1 \) is not valid. To avoid this problem, for calculation of intrinsic diffusion coefficients, following concentration profiles has been taken into account: \( N_A \) and \( N_{B+C} \) (or in a normalized form: \( Y_A \) and \( Y_{B+C} \)) in the function of displacement (Fig. 2b). Thus, values of \( D_A \) and \( D_B \) may be calculated as follows:

\[
D_A = \frac{1}{2t} \left( \frac{dx}{dN_A} \right)_K \left[ N_A^{x_k} Y_A^{x_k} dx - N_A^{x_1} \left( 1 - Y_A \right) dx \right] 
\]

(7)

\[
D_B = -\frac{1}{2t} \left( \frac{dx}{dN_A} \right)_K \left[ N_{B+C}^{x_k} Y_A^{x_k} dx - N_{B+C}^{x_1} \left( 1 - Y_A \right) dx \right] 
\]

(8)

Putting together all the Paul’s assumptions and equations, calculated interdiffusion and intrinsic diffusion coefficients equaled: \( \tilde{D} = 1.68 \cdot 10^{-14} \), \( D_A = 1.13 \cdot 10^{-14} \), \( D_B = 3.38 \cdot 10^{-14} \) m²/s.

2.2. Interdiffusion in forming intermetallic compounds by average atom movement (AAM)

In the Paul’s example, all the experimental data is known – concentration of all elements, thickness of diffusion region, time, and position of marker plane. Thus, the interdiffusion coefficient of γ phase may be calculated by simple diffusion equation:

\[
(\Delta x)^2 = 4 \tilde{D} t
\]

(9)

Transformation of equation (Eq. (9)) and substitution of data from section 2.1 from this paper, enable to evaluate value of interdiffusion coefficient in γ phase, which equaled: \( \tilde{D} = 2.25 \cdot 10^{-14} \) m²/s.

Assuming further, that atomic volume does not vary, following relation can be written:

\[
\frac{J_B}{\beta J_A} = \frac{x_k - x_L}{x_R - x_K}
\]

(10)
where fluxes $J_A$ and $J_B$ are calculated from the Fick’s law (Eq. 11) and $\beta$ is a stoichiometric subscript, that indicates growing phase ($A_\beta B$, $\beta=1,2,\ldots$). Putting it together, for two diffusion elements A and B, following equation is obtained:

$$\frac{D_B}{D_A} = \frac{\beta(x_K - x_L)}{x_R - x_K} \tag{11}$$

Knowing the value of interdiffusion coefficient, assumption that $\beta = 2$ and connection of (Eq. (6)) with (Eq. (11)) make possible the calculation of intrinsic diffusion coefficient of elements A and B by following equations:

$$D_B = \frac{\bar{D}}{N_A + N_B} \cdot \frac{x_R - x_K}{\beta(x_K - x_L)}$$

$$D_A = \frac{D_B(x_R - x_K)}{\beta(x_K - x_L)} \tag{12}$$

Calculated values equal: $D_A = 1.95 \cdot 10^{-14}$ and $D_B = 3.19 \cdot 10^{-14}$ m$^2$/s.

\textbf{2.3. Approach by mass conservation law (MCA)}

Calculation of interdiffusion and intrinsic diffusion coefficients can be also approached in a different manner. Taking into account, that mass conservation law is obeyed in diffusion process, reactive diffusion between two alloys, where only one intermetallic phase $\gamma$ grows, can be characterized by following set of equations:

$$\left( N_{L,i}^{(1)} - N_{L,i}^{(0)} \right) \frac{dx_L}{dt} = J_i^{(1)} - J_i^{(0)}$$

$$\left( N_{R,i}^{(2)} - N_{R,i}^{(1)} \right) \frac{dx_R}{dt} = J_i^{(2)} - J_i^{(1)} \tag{13}$$

where: $N_{L,i}^{(1)}, N_{L,i}^{(0)}, N_{R,j}^{(2)}$ - molar fractions of i-th (i = A, B+C) component on the left (L) and right (R) boundary in Alloy 1, $\gamma$ phase and Alloy 2, respectively; $J_i^{(1)}$, $J_i^{(0)}$, $J_i^{(2)}$ - diffusion fluxes of i-th component, calculated by the Fick’s law, $x_L$ and $x_R$ - location of the left and the right boundary after the diffusion process (fig. 3).
Assuming further that the fluxes of \( i \)-th component in Alloy 1 and Alloy 2 are equal to zero: 
\[ J_i^{(1)} = J_i^{(2)} = 0 \] 
\[ \Delta x = x_R - x_L \] 
Eq. 9 may be rewritten as:

\[ \frac{d \Delta x}{dt} = \frac{-J_i^\gamma}{\left( N_{R,i}^{(2)} - N_{R,i}^{\gamma} \right) - \left( N_{L,i}^{\gamma} - N_{L,i}^{(1)} \right)} \]  
(14)

Because the flux of \( i \)-th component may be defined by Fick’s law as:

\[ J_i^\gamma = -D_i \frac{N_{R,i}^{\gamma} - N_{L,i}^{\gamma}}{\Delta x} \]  
(15)

it is possible, after some mathematical transformations, to calculate the intrinsic diffusion coefficient of \( i \)-th component:

\[ D_i = \Delta x \frac{d \Delta x}{dt} \left\{ \frac{\left( N_{R,i}^{(2)} - N_{R,i}^{\gamma} \right) \left( N_{L,i}^{\gamma} - N_{L,i}^{(1)} \right)}{\left( N_{R,i}^{(2)} - N_{R,i}^{\gamma} \right) \left( N_{L,i}^{\gamma} - N_{L,i}^{(1)} \right) + \left( N_{R,i}^{(2)} - N_{R,i}^{\gamma} \right) \left( N_{L,i}^{(2)} - N_{L,i}^{\gamma} \right)} \right\} \]  
(16)

Considering Paul’s assumed data, intrinsic diffusion coefficients (Eq. (16)) have been calculated and following values were obtained: \( D_A = 2.25 \cdot 10^{-14} \) and \( D_B = 2.25 \cdot 10^{-14} \) m\(^2\)/s. Both coefficients equal to the same value, as the differences in concentrations of elements A and B also equal. Taking that into account, it is obvious, that interdiffusion coefficient calculated in marker plane (Eq. (6)) gives exactly the same value: \( \bar{D} = 2.25 \cdot 10^{-14} \) m\(^2\)/s.

To sum up, in section 2 of this paper, interdiffusion and intrinsic diffusion coefficients have been calculated for Paul’s data using three methods – PBA, AAM, MCA. Obtained values are presented in Table 2.
Table 2. Calculated values of interdiffusion and intrinsic diffusion coefficients.

|                | Interdiffusion coefficient $\tilde{D}$, m$^2$/s | Intrinsic diffusion coefficient of element A - $D_A$, m$^2$/s | Intrinsic diffusion coefficient of element B - $D_B$, m$^2$/s | Calculated thickness of the intermetallic phase, µm |
|----------------|-----------------------------------------------|-------------------------------------------------|-------------------------------------------------|-----------------------------------------------|
| PBA            | 1.68⋅10^{-14}                                | 1.13⋅10^{-14}                                   | 3.38⋅10^{-14}                                   | 67.88                                         |
| MCA            | 2.25⋅10^{-14}                                | 2.25⋅10^{-14}                                   | 2.25⋅10^{-14}                                   | 79.76                                         |
| AAM            | 2.25⋅10^{-14}                                | 1.95⋅10^{-14}                                   | 3.19⋅10^{-14}                                   | 81.60                                         |

In case of pseudobinary alloy when only one intermetallic phase is formed the growth of the intermetallic phase can be calculated directly from the mass conservation law at the boundaries, eq. (13). Thus, the following expression should be solved for one component (e.g. 1):

$$\frac{dx_L}{dt} = \frac{J_i^\gamma}{N_{i,i}^L - N_{i,i}^{(1)}} \Rightarrow \frac{dx_R - x_L}{dt} = \frac{-J_i^\gamma (N_{i,i}^L - N_{i,i}^{(1)}) - J_i^\gamma (N_{i,i}^{(2)} - N_{i,i}^R)}{(N_{i,i}^L - N_{i,i}^{(1)})(N_{i,i}^{(2)} - N_{i,i}^R)}$$

(17)

In table 2, the results of calculations of the intermetallic phase thickness generated during the diffusion process - calculated by diffusion coefficients obtained by different methods are presented.

2.4. The diffusion coefficients in ternary Ti-NiAl system by MCA method. In this paragraph the MCA method will be used to approximate the intrinsic diffusion coefficients in ternary multiphase Ti-NiAl system at 1173K. In this system between pure Ti and NiAl three intermetallic phases are formed, mainly $\text{AlTi}_3$, $\text{Al}_3\text{NiTi}_2$ and $\text{AlNi}_2\text{Ti}$ [26]. The equilibrium molar fractions of each component in each phase as well as integral diffusion coefficients and thickness of each phase after annealing for 100 h at 1173 K are presented in figure 4 and Table 3.
Figure 4. The experimental concentration profile in Ti-NiAl diffusion couple after diffusion at 1173K for 100h.

Table 3. The initial kinetic data – equilibrium molar fractions in Ni-Al-Ti system at 1173K [26]

| Phase      | Phase thickness, µm | Equilibrium molar fractions |
|------------|---------------------|-----------------------------|
|            |                     | Ti  | Al     | Ni     |
| (αTi)      | 16.7                | 0.98-0.85 | 0.01-0.14 | 0.01-0.01 |
| Al Ti₃     | 8.7                 | 0.82-0.70 | 0.17-0.29 | 0.01-0.01 |
| Al₃NiTi₂   | 6.8                 | 0.44-0.36 | 0.33-0.35 | 0.23-0.29 |
| AlNi₂Ti    | 6.2                 | 0.28-0.22 | 0.22-0.28 | 0.5-0.5   |
| AlNi       | ----                | 0.01-0.01 | 0.49-0.49 | 0.5-0.5   |

The intrinsic diffusion coefficients may be predicted by the MCA method – it is sufficient to write down Eq. (13) at each phase boundary and each element and solve obtained set of
equations by iterative method. In case of Ti-NiAl system, where three different intermetallic phases form, the following set of equations should be solved:

$$\left\{ \begin{align*}
\left( N_{L,j}^{Ti} - N_{R,j}^{Ti} \right) \frac{dx_{Ti}^{AITi}}{dt} &= J_{i}^{AITi} - J_{i}^{Ti} \\
\left( N_{L,j}^{AlNi} - N_{R,j}^{AlNi} \right) \frac{dx_{AlNi}^{AlNi}}{dt} &= J_{i}^{AlNi} - J_{i}^{AlNi} \\
\left( N_{L,j}^{AlNiTi} - N_{R,j}^{AlNiTi} \right) \frac{dx_{AlNiTi}^{AlNiTi}}{dt} &= J_{i}^{AlNiTi} - J_{i}^{AlNiTi} \\
\left( N_{L,j}^{Al} - N_{R,j}^{Al} \right) \frac{dx_{Al}^{Al}}{dt} &= J_{i}^{Al} - J_{i}^{Al} \\
\left( N_{L,j}^{Ni} - N_{R,j}^{Ni} \right) \frac{dx_{Ni}^{Ni}}{dt} &= J_{i}^{Ni} - J_{i}^{Ni} \\
\left( N_{L,j}^{Ti} - N_{R,j}^{Ti} \right) \frac{dx_{Ti}^{Ti}}{dt} &= J_{i}^{Ti} - J_{i}^{Ti}
\end{align*} \right. \quad (18)$$

where $N_{L,j}^{i}$ and $N_{R,j}^{i}$ are molar fractions of i-th element in j-th phase on the left and right boundary, respectively. The examples of presented notation for concentration of Ti and Al in particular phases of Ti-Ni-Al system are depicted in fig. 5. The fluxes that appear on the left-hand side in the Eq. (18) are calculated by the Fick’s law – thicknesses of the phases used for calculations are the thicknesses presented in Table 3. Moreover, as the differences in equilibrium molar fractions in AlNi phase on the left and right boundary of its occurrence equal to 0, also $J_{i}^{AlNi} = 0$.

![Figure 5. Indication of concentrations of a) Ti and b) Al in particular phases](image)

Solution of Eq. (18) enables to establish intrinsic diffusion coefficients of each element in each phase, except the elements that did not present changes in molar concentrations on particular boundaries. The results of calculations are presented in Table 4.
Table 4. Calculated values of intrinsic diffusion coefficients

| Phase        | Intrinsic diffusion coefficients, cm²·s⁻¹ |
|--------------|------------------------------------------|
|              | Ti           | Al           | Ni           |
| (αTi)        | 6.4⋅10⁻¹¹    | 1.9⋅10⁻¹¹    | ---          |
| AlTi₂        | 3.5⋅10⁻¹¹    | 9.9⋅10⁻¹²    | ---          |
| Al₃NiTi₂     | 2.5⋅10⁻¹¹    | 3.7⋅10⁻¹¹    | 2.1⋅10⁻¹¹    |
| Al₃Ni₄Ti     | 2.3⋅10⁻¹¹    | 2.3⋅10⁻¹¹    | ---          |
| AlNi         | ---          | ---          | ---          |

Similarly as in binary system the eq. (13) has been transformed into eq. (17), so that the thickness of growing intermetallic phase could be calculated. Also the eq. (18) may be used for calculation of thicknesses of growing phases. In order to solve such inverse problem, Eq. (18) must be reformulated with the assumption that intrinsic diffusion coefficients in particular phases are known and phase thicknesses – are unknowns. The results of such calculations led to obtaining exactly the same values of thickness of the growing phases as it is presented in experimental results (Table 3). This in turn proves the correctness of mathematical calculations.

3. Conclusions. In the present paper the mass conservation law (MCA method) was used to approximate the intrinsic diffusion coefficients in multiphase ternary system. In the literature until now there were a lack of method (even numerical - inverse methods) how to approximate the intrinsic diffusion coefficients in complicated ternary systems. We have shown, that the MCA method can be used to approximate the diffusion data even after one experimental results. This data can be used at first approximation of the diffusivities characterizing process. We have shown, that MCA method approximate diffusion coefficients where the difference in chemical composition is known. In Ti-NiAl diffusion couple the diffusion coefficients were fond in four phases, namely: Al₃Ni₄Ti, Al₃NiTi₂, AlTi₃ and (αTi). We have found nine diffusion coefficients of the components. The method can be further supplemented in Gibbs energy calculations instead of simplifying assumption of chemical activity equate to the molar concentration of component.
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Data Availability. The data required to reproduce the work reported in the manuscript can be found in Bartek Wierzba - email: bwierzba@prz.edu.pl

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