



## Interdiffusivities and atomic mobilities in fcc Cu–Al–Fe alloys

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### ABSTRACT

Using solid/solid diffusion couples and the electron probe microanalysis (EPMA) technique, the interdiffusion coefficients in fcc Cu–Al–Fe alloys at 1273 K were determined by means of Whittle and Green's method. Based on the atomic mobilities of three sub-binary systems available in the literature and the interdiffusivities determined, the atomic mobilities in fcc Cu–Al–Fe alloys were assessed by means of the DICTRA (Diffusion Controlled TRAnsformation) software package. The calculated interdiffusivities agree well with the experimental ones. Further verification of the atomic mobilities obtained was carried out through comprehensive comparisons between the model-predicted concentration profiles/diffusion paths of several diffusion couples and the experimental data. The results indicate that the atomic mobilities obtained can reproduce the experiment data reasonably well.

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### 1. Introduction

Cu–Al–Fe ternary alloys attract world-wide interest due to their potential industrial applications, such as thermal barrier coatings [1], low-friction wear-resistant coatings, composite biomaterials and catalysts, as well as reinforcement phases/fillers for composite materials [2]. In addition, many investigations have been devoted to the Cu–Al–Fe system [3] owing to the formation of quasicrystalline phases. Apart from the thermodynamic information, the kinetic information, such as atomic mobility and activation energy, is also of significance to get an insight into phase transformation including selection of phases in solidification and formation of metastable and quasicrystalline phases during various metallurgical processes. So far, the atomic mobilities in fcc Cu–Al, Cu–Fe, and Al–Fe alloys have been studied by Liu et al. [4], Wang et al. [5] and Zhang et al. [6], respectively. However, to the best of our knowledge, there exists no information with respect to interdiffusivity or atomic mobilities in fcc Cu–Al–Fe alloys in the literature.

Consequently, the main objectives of the present work are: (i) to determine the interdiffusion coefficients in fcc Cu–Al–Fe alloys at 1273 K; (ii) to evaluate the atomic mobilities of fcc Cu–Al–Fe alloys by means of DICTRA based on the available atomic mobilities for the three subsystems in the literature and

our experimental results; and (iii) to verify the atomic mobilities obtained via the simulation of concentration profiles and diffusion paths from several diffusion couples.

### 2. Experiments

Copper (purity: 99.99 wt%), aluminum (purity: 99.99 wt%), and iron (purity: 99.99 wt%) were used as starting materials. Button samples of Cu–Al, Cu–Fe, and Cu–Al–Fe alloys were prepared by arc melting of the pure elements under a high-purity argon atmosphere using a non-consumable tungsten electrode. The corresponding nominal compositions of the alloys are listed in Table 1. The buttons were remelted five times to improve their homogeneity. After that, the buttons were cut into blocks of approximate dimensions 4 mm × 4 mm × 6 mm. The alloy blocks were then sealed in quartz tubes under vacuum atmosphere, and homogenized at 1273 K for 864 ks (10 days) in an L4514-type diffusion furnace (Qingdao Instrument & Equipment Co. Ltd., China), followed by water quenching. After being ground, polished, and cleaned, the blocks were bound together by molybdenum wires to make six diffusion couples according to the assembly listed in Table 1. These six diffusion couples were then sealed in quartz tubes under vacuum atmosphere, and annealed at 1273 K for 28.8 ks (8 h) in the L4514-type diffusion furnace. Subsequently, the diffusion couples were quenched into cold water. The quenched diffusion couples were cut parallel to the diffusion direction. After standard metallographic preparation, the concentration profiles were measured by the

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**Table 1**  
Diffusion couples and experimental conditions in the present work.

Couple designation	Nominal composition of the diffusion couples (at.%)	Diffusion temperature (K)	Time (ks)
A1/B1	Cu–8.2Al/Cu–1.0Al–2.0Fe	1273	28.8
A2/B2	Cu–12.5Al/Cu–1.0Al–1.2Fe	1273	28.8
A3/B3	Cu–1.0Al/Cu–5.5Al–1.2Fe	1273	28.8
A4/B4	Cu–0.5Al/Cu–8.9Al–1.4Fe	1273	28.8
A4/B5	Cu–0.5Al/Cu–13.5Al–2.0Fe	1273	28.8
A4/B6	Cu–0.5Al/Cu–14.0Al–3.0Fe	1273	28.8

electron probe microanalysis (EPMA) technique (JXA-8230, JEPL, Japan) on polished sections, parallel to the diffusion direction. The concentrations were obtained by comparison with standards of pure elements Cu, Al, and Fe, after absorption and fluorescence corrections (ZAF corrections).

### 3. Calculation of interdiffusivity in fcc Cu–Al–Fe alloys

Kirkaldy successfully extended the Boltzmann–Matano method into ternary and even higher-order systems [7]. According to Kirkaldy, it is possible to design two diffusion couples, the diffusion paths of which intersect at one common composition. The main and cross interdiffusivities of this common composition can be determined. Kirkaldy [7] presented Fick's second law in a 1–2–3 ternary system as

$$\frac{\partial C_i}{\partial t} = \frac{\partial}{\partial X} \left( \tilde{D}_{i1}^3 \frac{\partial C_1}{\partial X} \right) + \frac{\partial}{\partial X} \left( \tilde{D}_{i2}^3 \frac{\partial C_2}{\partial X} \right) \quad (i = 1, 2), \quad (1)$$

where  $i$  is the solute,  $C_1$  and  $C_2$  are the concentrations of solutes 1 and 2 respectively,  $t$  is the diffusion time, and  $X$  is the distance from the Matano interface. Solvent 3 is treated as the dependent variable.  $\tilde{D}_{11}^3$  and  $\tilde{D}_{22}^3$  are two main interdiffusion coefficients, while  $\tilde{D}_{12}^3$  and  $\tilde{D}_{21}^3$  two cross ones. In this work, solutes 1 and 2 are Al and Fe, with solvent 3 being Cu. The initial and boundary conditions for the semi-infinite diffusion couples are

$$C_i(\pm X, 0) = C_i(\pm \infty, t) = C_i^{\pm \infty} \quad (i = 1, 2). \quad (2)$$

With the aid of Boltzmann–Matano method [8,9], Eq. (1) can be extended to

$$\int_{C_i^{-\infty}}^{C_i} x dC_i = -2t \left[ \tilde{D}_{i1}^3 \frac{\partial C_1}{\partial x} + \tilde{D}_{i2}^3 \frac{\partial C_2}{\partial x} \right]_{C_i} \quad (i = 1, 2). \quad (3)$$

Assuming that the volume change is negligible, the position of the Matano plane should be the same for concentration profiles of solutes 1 and 2 in theory. In actual calculations, however, the difference between the locations of Matano plane for solutes 1 and 2 will exist for experimental measurements of concentration profiles and subsequent calculations. In order to avoid such differences, Whittle and Green [10] introduced normalized concentration variables  $Y_i$  ( $i = 1$  or  $2$ ) in the ternary system:

$$Y_i = (C_i - C_i^-)/(C_i^+ - C_i^-), \quad (4)$$

and the ternary interdiffusion coefficients can then be determined by solving the following equations:

$$\begin{aligned} \tilde{D}_{11}^3 + (dC_2/dC_1) \tilde{D}_{12}^3 &= (1/2t) (dx/dY_1) \\ &\times \left[ (1 - Y_1) \int_{-\infty}^x Y_1 dx + Y_1 \int_x^{+\infty} (1 - Y_1) dx \right], \end{aligned} \quad (5)$$

$$\begin{aligned} \tilde{D}_{22}^3 + (dC_1/dC_2) \tilde{D}_{21}^3 &= (1/2t) (dx/dY_2) \\ &\times \left[ (1 - Y_2) \int_{-\infty}^x Y_2 dx + Y_2 \int_x^{+\infty} (1 - Y_2) dx \right]. \end{aligned} \quad (6)$$

With four functions in Eqs. (5) and (6) from two diffusion couples, the four main and cross interdiffusivities in Eq. (1) for the intersection point can then be determined.

### 4. Modeling of atomic mobility

According to Andersson et al. [11], the atomic mobility of species  $B$ ,  $M_B$ , can be expressed as

$$M_B = M_B^0 \exp \left( \frac{-Q_B}{RT} \right) \frac{1}{RT} {}^{mg} \Omega, \quad (7)$$

where  $M_B^0$  is the frequency factor,  $Q_B$  is the activation enthalpy,  $R$  is the gas constant,  $T$  is the temperature in kelvin, and  ${}^{mg} \Omega$  is a factor taking account the effect of ferromagnetic contribution to diffusivity. For the fcc phase, the ferromagnetic contribution can be neglected [12]. In this case, the atomic mobility parameter  $\Delta G_B$  is defined by  $-Q_B + RT \ln(M_B^0)$ , which is dependent on composition and temperature. Thus,  $\Delta G_B$  for disordered solid solution fcc phase in the Cu–Al–Fe system can be represented by the Redlich–Kister polynomial [13]:

$$\begin{aligned} \Delta G_B &= \sum_i x_i \Delta G_B^i + \sum_i \sum_{j>i} x_i x_j \left[ \sum_{r=0}^n {}^r \Delta G_B^{i,j} (x_i - x_j)^r \right] \\ &+ \sum_i \sum_{j>i} \sum_{k>j} x_i x_j x_k \left[ {}^s \Delta G_B^{i,j,k} \right], \quad (s = i, j, k), \end{aligned} \quad (8)$$

where  $x_i$  is the mole fraction of species  $i$ ,  $\Delta G_B^i$  is the value of  $\Delta G_B$  for  $B$  in pure  $i$ , and  ${}^r \Delta G_B^{i,j}$  and  ${}^s \Delta G_B^{i,j,k}$  are the binary and ternary interaction parameters respectively. In addition, the parameter  ${}^s \nu_{ijk}^s$  can be expressed as

$$\nu_{ijk}^s = x_s + (1 - x_i - x_j - x_k) / 3. \quad (9)$$

The tracer diffusion coefficient  $D_B^*$  is correlated with its atomic mobility by the Einstein relation [14]:

$$D_B^* = RTM_B. \quad (10)$$

The interdiffusion coefficient  $\tilde{D}_{kj}^n$  is correlated to the atomic mobilities [11]:

$$\tilde{D}_{kj}^n = \sum_i (\delta_{ik} - x_k) x_i M_i \left( \frac{\partial \mu_i}{\partial x_j} - \frac{\partial \mu_i}{\partial x_n} \right), \quad (11)$$

where  $\delta_{ik}$  is the Kronecker delta ( $\delta_{ik} = 1$  if  $i = k$ ; otherwise,  $\delta_{ik} = 0$ ), and  $x_i$ ,  $\mu_i$ , and  $M_i$  are the mole fraction, chemical potential, and mobility of element  $i$ , respectively.

In the number-fixed frame of reference, for a substitutional solution, the diffusional flux of the species,  $\tilde{J}_k^N$ , in a multicomponent system is given by the Fick–Onsager law [14]:

$$\tilde{J}_k^N = - \sum_{j=1}^{n-1} \tilde{D}_{kj}^n \nabla C_j, \quad (12)$$

where  $\nabla C_j$  is the concentration gradient of element  $j$ , and the superscript  $N$  stands for the number-fixed frame of reference. When  $\tilde{J}_k^N = 0$ , the location of a zero-flux plane (ZFP) for solute  $k$  is determined within the diffusion zone [15]:

$$\int_{C_k(+\infty)}^{C_k(ZFP)} x dC_k = \int_{C_k(ZFP)}^{C_k(-\infty)} x dC_k = 0. \quad (13)$$

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