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Composition-dependent interdiffusivity matrices in face centered cubic Ni–Al–X (X = Rh and W) alloys at 1423, 1473 and 1523 K: A high-throughput experimental measurement

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ABSTRACT

Based on 18 face centered cubic (fcc) single-phase diffusion couples in ternary Ni–Al–X (X = Rh and W) systems together with the recently developed numerical inverse method, high-throughput measurements of the composition-dependent interdiffusivity matrices in fcc Ni–Al–X (X = Rh and W) alloys at 1423, 1473 and 1523 K were performed in the present work. Their reliability was comprehensively validated through comparison between the model-predicted composition/interdiffusion flux profiles for each diffusion couple and the corresponding experimental data. Moreover, the direct comparison with the interdiffusivities evaluated from traditional Matano-Kirkaldy method as well as those from the literature and in the boundary binary systems was also made. The errors for the determined interdiffusivities were evaluated by a scientific method considering the error propagation. The three-dimensional main interdiffusivity planes for fcc Ni–Al–X (X = Rh and W) systems over the investigated concentration ranges at 1423, 1473 and 1523 K were subsequently constructed. It was then found that $\widetilde{D}^{Ni}_{AlAl}$ is generally larger than $\widetilde{D}^{Ni}_{RhRh}$, while $\widetilde{D}^{Ni}_{WW}$ is the smallest.

1. Introduction

Ni-based superalloys are widely used in aviation and land-based gas turbine environments [1,2] due to their excellent mechanical properties and high resistance to creep and fatigue at elevated temperatures. To promote their performance, Al and different transition metal elements, including Rh, W, etc., are usually added in the Ni-based superalloys [3]. Addition of Al leads to precipitation of $L1_2$ (Ni_3Al) phase from the fcc-Ni matrix, resulting in superior high temperature strength for Ni-based superalloys [4]. Addition of Rh depresses efficiently the formation of topologically packed phases (TCPs) [3]. While the addition of W, of which diffusion coefficients are relatively low [5], can improve the creep resistance of Ni-based superalloys [6]. It is well known that accurate thermodynamic and diffusion information is critical for quantitative description of microstructure evolution in the target alloys during various materials processes using i.e., the phase-field simulation [7–10], and even for novel alloy design [1,11]. In contrast with the mature thermodynamic databases of Ni-based superalloys, the reports on diffusivities in Ni-based superalloys majorly focus on unaries [12–14] and sub-binaries [15–17]. As for ternary and even higher-order sub-systems of Ni-based superalloys, very limited experimental reports on their diffusivities are available in the literature, not to mention the composition- and temperature-dependent ones. For instance, no any experimental information about interdiffusivities for face-centered cubic (fcc) Ni–Al–Rh alloys exists in the literature. While for fcc Ni-Al-W system, only diffusivities at several specific concentration points are available [18,19]. Therefore, there is a necessity to improve this situation.

For a ternary system, single-phase diffusion couple technique together with the traditional Matano-Kirkaldy method is the most widely used method for calculating its interdiffusivities [20,21], but the traditional Matano-Kirkaldy method is considered to be of low efficiency [22]. Using the Matano-Kirkaldy method, only four independent interdiffusivities at the intersection point along two diffusion paths from two diffusion couples can be obtained in a ternary system. In order to circumvent the situation, Chen et al. [23] developed a pragmatic numerical inverse method, which can be used to measure efficiently the composition-dependent interdiffusivities along the entire diffusion path of a single diffusion couple. Such a numerical inverse method has been successfully applied in several ternary systems later [24–27]. Very recently, the numerical inverse method has been augmented, and validated in binary, ternary and even high-order systems [28]. Thus, the

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Table 1
List of terminal compositions of the diffusion couples in the present work.

Couple No.	Composition (wt%)	Temperature (K)	Diffusion time (ks)
RC1	Ni–2.20 Al/Ni–9.45 Rh	1423	499.5
RC2	Ni/Ni–1.60 Al–9.93 Rh	1423	499.5
RC3	Ni/Ni–2.20 Al–8.60 Rh	1423	499.5
RC4	Ni–2.35 Al/Ni–9.48 Rh	1473	500.4
RC5	Ni/Ni–2.23 Al–8.55 Rh	1473	500.4
RC6	Ni/Ni–1.70 Al–10.08 Rh	1473	500.4
RC7	Ni–2.44 Al/Ni–9.82 Rh	1523	172.8
RC8	Ni/Ni–1.76 Al–10.30 Rh	1523	172.8
RC9	Ni/Ni–2.21 Al–8.43 Rh	1523	172.8
WC1	Ni–4.90 Al/Ni–24.8 W	1423	499.5
WC2	Ni/Ni–2.14 Al–11.0 W	1423	499.5
WC3	Ni/Ni–1.62 Al–18.4 W	1423	499.5
WC4	Ni–4.86 Al/Ni–24.8 W	1473	500.4
WC5	Ni/Ni–2.08 Al–12.0 W	1473	500.4
WC6	Ni/Ni–1.62 Al–18.4 W	1473	500.4
WC7	Ni–4.69 Al/Ni–23.6 W	1523	172.8
WC8	Ni/Ni–2.10 Al–11.3 W	1523	172.8
WC9	Ni/Ni–1.60 Al–18.5 W	1523	172.8

numerical inverse method in combination with the single-phase diffusion couple technique can be utilized for high-throughput measurements of composition-dependent interdiffusivities in fcc Ni–Al–Rh and Ni-Al-W systems.

The major objectives of the present work are, (*i*) to perform high-throughput measurements of the composition-dependent interdiffusivities in Ni-rich fcc Ni–Al–X (X = Rh and W) alloys at 1423, 1473 and 1523 K by using the numerical inverse method together with single-phase Ni–Al–X (X = Rh and W) ternary diffusion couples, (*ii*) to comprehensively validate the resulting composition-dependent interdiffusivities by comparing the data with those obtained by using the traditional Matano-Kirkaldy method, as well as the simulated concentration profiles and interdiffusion fluxes with experimental data for each diffusion couple. The comparisons also include the boundary binary diffusivities available in the literature, (*iii*) to plot the three-dimensional (3-D) diffusivity planes for main interdiffusivities at different temperatures.

2. Experimental procedure

Alloys used for later eighteen groups of diffusion couples, as listed in Table 1, were prepared by arc-melting method. Pure elements Ni, Al, Rh and W were used as starting materials, and their purity is 99.99 wt%. In order to make the refractory metals, i.e., Rh or W, to be completely dissolved in the Ni matrix, the same melting processes as described in our recent work [16,17,25] were applied in the present. Then all the prepared alloy ingots and high-purity Ni bar were sealed in the vacuum quartz tubes and placed in a high-temperature furnace at 1573 ± 2 K for 288 ks to facilitate homogenization, followed by water quenching. Thick slices with the size of 5 × 5 × 1.5 mm^3 were cut from the ingots. All surfaces were ground and one plane with the size of 5 × 5 m^2 was polished. Two polished surfaces were bound together with Mo clamps to form diffusion-couples assembly, which were then encapsulated in vacuum quartz tubes. After annealing treatments as listed in Table 1, all assemblies were quenched in cold water. The concentration-distance profiles for all the 18 diffusion couples were examined by electron probe microanalyzer (EPMA, JXA-8100, JEOL, Japan) on the polished section. The error for concentration measurements is within 1%.

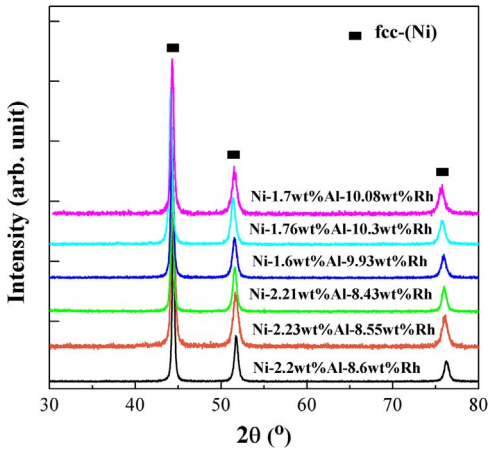

Fig. 2. XRD patterns of ternary Ni–Al–Rh alloys listed in Table 1.

3. Results and discussion

The measured diffusion paths for the eighteen groups of diffusion couples at 1423, 1473 and 1523 K listed in Table 1 are plotted in Fig. 1. For Ni – Al – Rh system, one intersection point exists along the diffusion paths for every two diffusion couples at one temperature. While for Ni – Al – W system, no any intersection points locate along the diffusion paths for all the diffusion couples at one temperature. Based on the thermodynamic descriptions of the Ni–Al–W system [29], all the diffusion couples (i.e., WC1∼WC9) locate in the single fcc phase region at

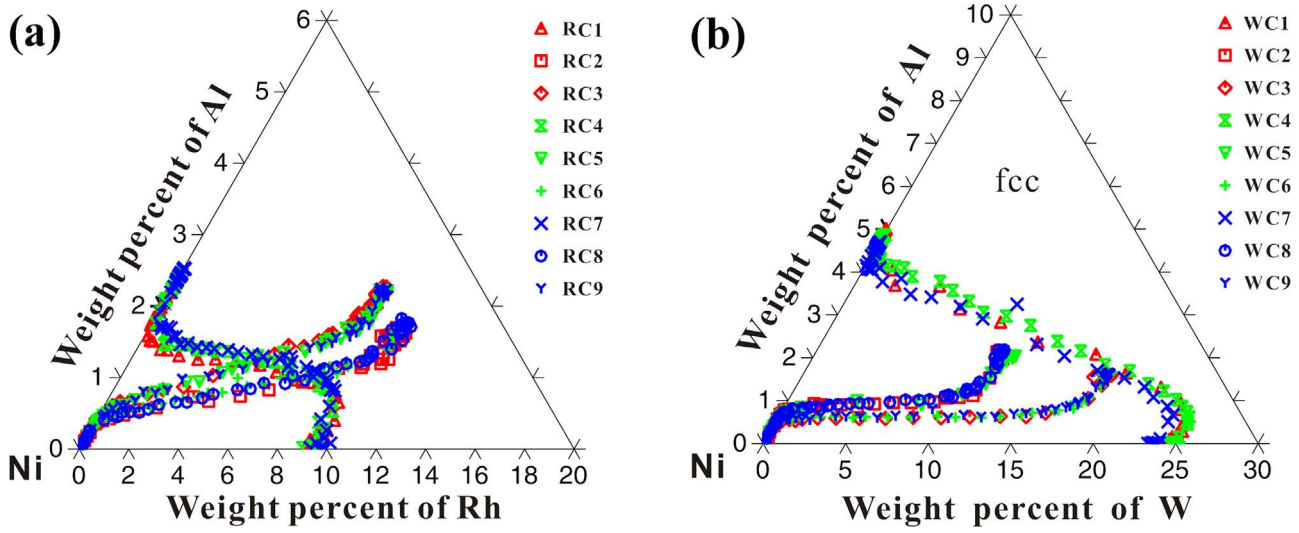

Fig. 1. Measured diffusion paths of the diffusion couples in (a) Ni–Al–Rh and (b) Ni–Al–W systems at 1423, 1473 and 1523 K.