Application of two-step diffusion couple technique in high-throughput screening of optimal composition and aging temperatures for alloys design: A demonstration in binary Ni-Al system

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Abstract
In this paper, four binary Ni-13.4 at.% Al/Ni-17.7 at.% Al diffusion couples were first prepared and subjected to homogenization at 1573 K for 10800 s, from which a continuous concentration profile forms. The three diffusion couples were then cooled down for aging at respective temperatures, i.e., 1173, 1123 and 1073 K, for 14400 s. The effect of composition and aging temperature on the aging microstructure was studied in detail by means of different experimental techniques and statistical analysis. The volume fraction, grain size and shape factor of γ′ precipitates in the three diffusion couples were plotted as a function of alloy composition and annealing temperatures. Together with the previously proposed evaluation function in which the phase fraction, grain size and shape factor of γ′ precipitates were chosen as the evaluation indicators, the optimal alloy composition and aging temperature for binary Ni-Al alloys with the best mechanical properties were evaluated, and finally validated by the measured hardness values. The successful demonstration of alloy design in the present binary Ni-Al alloys indicates that the two-step diffusion couple together with the evaluation function for mechanic properties should be of generality for high-throughput screening of optimal alloy composition and heat treatment process in different alloys.

Keywords: Two-step diffusion couple; Ni-Al alloys; Microstructure; Hardness; Alloys design
1. Introduction

Ni-based superalloys, mainly consisting of chemically disordered γ matrix and chemically ordered γ' precipitates, are widely used as engine blades and turbine discs in aerospace fields because of their excellent mechanical performance at high temperatures [1-4]. The performances of alloys are closely related to their microstructures, such as volume fraction, grain size, and morphology of the γ' precipitates, which strongly depend on alloy composition and heat-treatment processes. Therefore, a large number of experiments have been devoted to studying the effects of alloy composition and heat treatment process on the microstructure and properties of Ni-based superalloys [5-12] in order to improve the high-temperature strength and creep resistance of Ni-based superalloys. Qiu [8] observed the anomalous coarsening behavior of γ' precipitates in Ni-based superalloys after aging process. Nathal [9] investigated the influence of size and shape of γ' grains on the high temperature creep properties of Ni-based superalloys. Ges et al. [10] studied the effect of heat treatments on the γ' precipitate sizes and distribution in a CMSX-2 superalloy. Besides, the influence of Al content on the microstructure and creep behavior of Ni-based superalloy was also investigated by Fu et al. [11]. However, significant disadvantages exist in the traditional ‘trial-and-error’ method because of its low efficiency, and time/money-consuming process. Thus, the theoretical investigations may serve the supplement. In fact, there also exist certain amounts of modeling work on the microstructure evolution of Ni-based superalloys during different processes using, for instance, the powerful phase-field simulations [13-17]. Though the theoretical modeling can save time and money, its reliability should be validated by key experiments.

In recent years, the Materials Genome Initiative (MGI) project has promoted the emergence of various high-throughput technologies [18] in combination with micrometer-scale property measurement tools, which can accelerate the study on the properties of alloys, such as thermal conductivity, diffusivity, hardness, and elastic modulus, and so on [19, 20]. In the spirit of MGI, a series of methods utilizing the macro-component gradient of samples have been originally proposed to determine the phase diagrams at relatively low temperatures [21, 22], to evaluate the interfacial energy [23-25], and recently employed to obtain the abundant information on microstructure and different properties within the specified alloy composition range in a high-throughput way. One of such methods is the so-called two-step diffusion couple technique [19, 26, 27]. Taking the binary Ni-Al system for example, the sample preparation of a two-step diffusion couple can be illustrated in Fig. 1: (i) two single-phase alloys with different compositions (xA and xB) are combined together and annealed at a high temperature (i.e., T1, at which both alloys with composition xA and xB locate in the single-phase region) for a certain period to obtain a continuous composition region; (ii) the diffusion couple is then cooled down from T1 to a relatively lower temperature (i.e., T2, at which precipitates appear in the single-phase diffusion couple) and annealed for certain time to obtain the microstructure of the continuous composition at T2. Only at this point can a two-step diffusion couple be prepared successfully. Furthermore, the two-step diffusion couple technique in combination with different micrometer-scale property measurement tools can improve the screening
efficiency of the alloy composition and the heat treatment process with optimum properties.

Consequently, the Ni-Al binary system is taken as the target in this paper. Three Ni-Al two-step diffusion couples are to be prepared, and the continuous composition-dependent microstructure forming during annealing at three different temperatures will be measured. After that, the optimal alloy composition and aging temperature corresponding to the optimal mechanical properties for binary Ni-Al alloys can be then screened efficiently based on the previously proposed property evaluation function [15] together with the measured microstructure information. Finally, the screened alloy composition and aging temperature with the optimal mechanical property are validated by the experimentally measured hardness in the three diffusion couples.

2. Experimental procedure

Ni-13.4 at.% Al and Ni-17.7 at.% Al alloy ingots were prepared in an argon atmosphere by using an arc-melting furnace (WKDHL-1, Optoelectronics Co., Ltd., Beijing, China). Ni ingots (purity: 99.99 wt.%) and Al pieces (purity: 99.99 wt.%) were used as the raw materials. Subsequently, the Ni-13.4 at.% Al and Ni-17.7 at.% Al alloy ingots were cut into blocks of approximate 5 mm×5 mm×2.5 mm in size and annealed in vacuum-sealed quartz tubes at 1573 ± 3 K for 18000 s (5 hours) in a high-temperature diffusion furnace (GSL1700X, Hefei Kejing Materials Technology Co., Ltd., Hefei, China) to improve their homogeneity. All the homogenized alloys were subjected to X-ray diffraction detection (XRD, D8 Advance, Bruker, Germany) to confirm that only the γ phase exists in these samples. As shown in Fig. 2, the XRD patterns suggest that the two alloys locate in the single γ phase region. After being polished and cleaned, the polished surfaces of the alloys were stucked together and then bound together to prepare
diffusion couples by using two pieces of Mo plates, which are fastened by screws. Four Ni-13.4 at.% Al/Ni-17.7 at.% Al diffusion couples were annealed at 1573 ± 3 K for 10800 s (3 hours) in vacuum sealed quartz tubes to form the continuous composition profiles. After that, one of four diffusion couples was taken out and quenched in water, and then its concentration profile was measured by means of electron probe microanalysis (EPMA, JXA-8230, JEOL, Japan). Meanwhile, the other three diffusion couples were cooled to 1073 ± 3 K, 1123 ± 3 K, and 1173 ± 3 K at a rate of 10 K min⁻¹ respectively in the high-temperature diffusion furnace and then aged for 14400 s (4 hours), followed by water quenching.

![X-ray diffraction patterns](image)

**Fig. 2** X-ray diffraction patterns of Ni-13.4 at.% Al and Ni-17.7 at.% Al alloys after annealing at 1573K for 18000s.

The microstructure and hardness of the three diffusion couples after aging at 1073, 1123, and 1173 K for 14400 s were tested by different techniques: the microstructure along diffusion paths was characterized using scanning electron microscopy (SEM); then, the grain size and the volume fraction of the γ′ phase in SEM images were measured by using Image-Pro plus 6.0 metallographic analyzer. Later, the Vickers hardness of the three two-step diffusion couples was determined by nanoindentation (NHT2, Anton Paar, Austria). The nanoindentation tests were performed with the maximum indentation load of 20 mN, loading rate of 40 mN min⁻¹, and loading time of 10 s in the maximum loading, and the hardness was measured every 100 μm along diffusion paths of the diffusion couples. Each point was tested 3 times, and the mean value was accepted.

### 3. Results and discussion

#### 3.1 Experimental results

The measured Al concentration profile of the Ni-Al diffusion couple annealed at 1573 K for 10800 s is displayed in Fig. 3 (denoted in red symbol). The model-predicted composition profile (denoted in curve) according to Zhang et al. [28] is also
superimposed for a comparison. As can be seen, the model-predicted composition profile is in excellent agreement with the present experimental data. According to Zhao [23] and Miyazaki [25], the effect of aging treatments at lower temperatures (i.e., 1073, 1123, and 1173 K) on the composition gradient created during the higher temperature annealing (1573 K) can be neglected. It is because that: (i) the difference between the diffusing and aging temperatures is over 400 K, which results in the diffusion coefficient of solute atoms at diffusing temperature is larger than that at the aging temperature by $10^3$ times [28, 29]; (ii) the aging time (14400 s in this work) is too short for the composition profile to change as a function of distance at lower temperatures; and (iii) compared with the chemical driving force for the formation of γ', the driving force for interdiffusion is too small. Therefore, it can be assumed that the average composition profile of the two-step diffusion couples is the same as that of the diffusion couple only annealed at 1573 K. Moreover, the microstructure of the Ni-13.4 at.% Al/Ni-17.4 at.% Al diffusion couples aging at 1073 K, 1123 K, and 1173 K are shown in Fig. 4. It can be seen that both the size and volume fraction of γ' precipitates increase as Al content increases, which can be attributed to that as Al content increases, (i) the equilibrium fraction of γ' precipitates increases according to the level rule of the Ni-Al phase diagram (see Fig. 1), and (ii) the driving force for the formation of γ' precipitates [30] and the interdiffusion coefficients of the γ matrix [28] both increase.

![Graph](image-url)

**Fig. 3** Measured Al concentration profile of the Ni-13.4 at.% Al/Ni-17.7 at.% Al diffusion couple annealed at 1573 K for 10800s (denoted in red hollow diamond), compared with the model-predicted results by Zhang et al. [28] (denoted in black solid line).

Moreover, it also can be observed that with the increase of Al content and the growth of precipitates from left side to right side of Fig. 4, the small square γ' precipitates (cube in 3-D) gradually change into the composed square (octocube in 3-D)/butterfly shape (octodendrites in 3-D crystals), and then to irregular shape. This change in morphology should be attributed to the competition between the elastic strain...
energy associated with the lattice mismatch between $\gamma$ and $\gamma'$ phases and the interface energy [31-33]. During the evolution of $\gamma'$ precipitates, at very initial stage, the shape of precipitates tends to be spherical since the contribution of interface energy is predominant when the precipitates are very small. Subsequently, the elastic contribution increases as the precipitates grow, and the shape of precipitates transforms into the cubic one since the elastic strain energy in an octagonal cube should be lower than that in a cube. Since the elastic strain energy in an octagonal cube should be lower than that in a cube [32, 34], the decrease of elastic energy is higher than the increase of interface energy caused by splitting when the precipitates exceed a critical size, and the cubic precipitates thus split into octocube. Then, the further growth of the precipitates leads to the formation of octo-dendrites. While with the increase of $\gamma'$ phase volume fraction, the elastic interaction between precipitates will increase dramatically, resulting in a decrease of the driving force for splitting [34]. It has been found that cubic $\gamma'$ phase is the required structure of Ni-based superalloy since it is stable during service life and can better prevent dislocation migration during the creep stage because of its larger lattice misfit [35, 36]. Moreover, a preferred orientation of $\gamma'$ phase can be usually observed in superalloys, but it is not apparent in this work. That may be due to the difference in heat treatment. In this work, after homogenization, the samples were cooled at a low rate (i.e., 10 K min$^{-1}$). Such a low cooling rate could cause a low nucleus density, and the interaction among $\gamma'$ phases can be neglected in the early stage of precipitating. Besides, the relatively high aging temperatures (1173 K, 1123 K, and 1073 K) and the short aging time (i.e., 4h in this work) also lead to the random arrangement of $\gamma'$ precipitates. Furthermore, the shape factor $\xi$ can be used to describe the geometric shape of the $\gamma'$ precipitate [37]:

$$\xi = \frac{4\pi A}{P^2}$$  

(3-1)

where $P$ is the perimeter of $\gamma'$ precipitates and $A$ is the area of $\gamma'$ precipitate measured by the quantitative image analyzer. In this work, all the values of the calculated shape factor are below 0.785. In Equation (3-1), the closer to 0.785 the shape factor is, the closer to cube the shape is, and the closer to 0 the shape factor of irregular shape precipitation is. The evaluated shape factors according to the experimental data (denoted in solid spheres) are presented in Fig. 5. Moreover, the fitted results (denoted in solid lines) corresponding to each set of experimental data are also superimposed in the figure. It is indicated in Fig. 5 that the increase of the aging temperature decreases the shape factor. It is because that the increase of the temperature leads to the decrease of mismatch between the $\gamma$ phase and $\gamma'$ phase [38], and thus the cubic degree of the morphology of $\gamma'$ precipitate reduces. Meanwhile, as the increase of Al content, the shape factor increases and reaches the maximum value (0.71) at around Ni-16 at.% Al. Subsequently, it decreases to the minimum and then increases slightly, which corresponds to the change in shape described above.
**Fig. 4** Backscatter electron images of the Ni-13.4 at. % Al/Ni-17.4 at. % Al diffusion couples after aging at 1073 K, 1123 K and 1173 K for 14400 s.

**Fig. 5** Evolution of shape factors of $\gamma'$ precipitates evaluated from the experiment microstructures (denoted in solid spheres) as a function of Al composition and aging temperature, together with the fitted results using polynomial functions (denoted in solid lines).

The relationship among the size of $\gamma'$ precipitates, the aging temperature and Al content is shown in Fig. 6. The variation of $\gamma'$ precipitate size with the Al composition for each temperature is fitted, and the results (denoted in solid lines) are also added in Fig. 6. Since the shape of $\gamma'$ precipitates is irregular (as shown in Fig. 4), it is difficult
to obtain the size of $\gamma'$ precipitates by measuring the diameter along a certain direction. Therefore, the shape of the precipitated phase is assumed to be circular with almost the same area to gain an average diameter, which can be calculated by the following expression [31]:

$$\bar{d} = \left(\frac{4A}{\pi}\right)^{0.5} \quad (3-2)$$

The average diameter of each $\gamma'$ precipitate is calculated, and the mean value of these $\gamma'$ precipitates can be then obtained. The error bars represent the standard deviation from the mean value. It can be seen in Fig. 6 that the average size of $\gamma'$ precipitates increases with the increase of the aging temperature, while increases as the increase of Al content at first and then reaches the maximum value, which is due to that the growth of $\gamma'$ precipitates depends on the transport of the solute. The diffusivities increase with the increase of temperature and also the increase of Al content, and the large diffusivities can stimulate the growth of precipitates, which is controlled by diffusion. According to Refs. [39-41], the larger the grain size of $\gamma'$ precipitates, the lower the strength of alloys.

**Fig. 6** Evolution of diameter of $\gamma'$ particles evaluated from the experiment microstructures (denoted in solid spheres) as a function of Al composition and aging temperature, together with the fitted results using polynomial functions (denoted in solid lines).

Fig. 7 displays the volume fraction of $\gamma'$ precipitates, and the error bars represent the standard deviation from the mean value of the volume fraction. The variation of the volume fraction of $\gamma'$ precipitates with Al composition for each temperature is fitted, and the results (denoted in solid lines) are added in Fig. 7. Since the SEM results are typically two-dimensional images, one cannot directly obtain the volume fraction of $\gamma'$ phases. Therefore, the following expression is utilized to evaluate the volume fraction
\[ f_V = \frac{\pi}{6} N_A \bar{d}^2 \]  
(3-3)

where \( \bar{d} \) is average \( \gamma' \) precipitates diameter and \( N_A \) is particle density. As can be seen in Fig. 7, the volume fraction of \( \gamma' \) precipitates increases almost linearly with the increase of Al content, while it slightly decreases as the aging temperature increases, which conforms to the lever rule. According to the lever rule in binary alloys, the equilibrium phase fraction can be calculated based on the Ni-Al phase diagram (Fig. 1 (a)), in which the volume fraction of \( \gamma' \) precipitates is proportional to Al content and increases when the temperature decreases.

![Graph showing volume fraction of \( \gamma' \) precipitates as a function of Al composition and aging temperature.](image)

**Fig. 7** Evolution of volume fractions of \( \gamma' \) precipitates evaluated from the experiment microstructures (denoted in solid spheres) as a function of Al composition and aging temperature, together with the fitted results using polynomial functions (denoted in solid lines).

### 3.2 Evaluation of optimal alloys composition/aging temperature and its validation

As mentioned above, the mechanical properties of Ni-based alloys strongly depend on the microstructure of the material, including the volume fraction, grain size, and geometric shape of \( \gamma' \) precipitates. Therefore, based on the concept of Fuzzy Mathematics, a composite indicator, in which volume fraction, grain size and shape factor of \( \gamma' \) precipitates are chosen as evaluation indicators, has been proposed in our research group [15] to evaluate the optimal alloy composition and aging process. The
composite indicator can be established by assigning empowered weights $w_1$, $w_2$, and $w_3$, for the different indicators,

$$Composite \ Indicator = w_1 \times f_v - w_2 \times \overline{d} + w_3 \times \xi$$  \hspace{1cm} (3-4)$$

where the composite indicator increases with the increase of volume fraction $f_v$ and shape factor $\xi$, while decreases with the increase of diameter $\overline{d}$. With the maximum value of “Composite Indicator”, a point that balances the three indicators at the best degree is achieved. The empowered weights $w_1$, $w_2$, and $w_3$ can be calculated by the following equations:

$$w_j = \frac{w_j^*}{\sum n_i w_i^*} \hspace{1cm} (3-5)$$

$$w_i^* = \sin(Indicator \ membership) \hspace{1cm} (3-6)$$

$$Indicator \ membership = \frac{Indicator - Indicator_{min}}{Indicator_{max} - Indicator_{min}} \hspace{1cm} (3-7)$$

where “Indicator” represents volume fraction, grain size, and shape factor.

The composite indicator of the present experimental data is shown in Fig. 8. The alloy composition and the aging temperature corresponding to the maximum value of the composite indicator (0.295) are Ni-16.2 at.% Al alloy and 1073 K, at which the optimal mechanical property can be achieved in Ni-16.2 at.% Al aging at 1073 K.

![Fig. 8 Evaluated composite indicators for binary Ni-Al alloys as a function of Al composition and aging temperature.](image-url)

Furthermore, the measured hardness of the three two-step diffusion couples is shown in Fig. 9. The hardness of alloys decreases as the aging temperature increases. Meanwhile, it increases first as the increase of Al content and reaches the maximum value at the composition around Ni-16.2 at.% Al. Then, the hardness decreases as the
Al content increases. That is probably because of the increase of $\gamma'$ phase size. The alloy with the highest hardness is Ni-16.2 at. % Al, and the aging temperature is 1073 K, which is consistent with the result predicted by the linear synthesizing evaluation function. The successful prediction of the alloy composition and heat treatment process for binary Ni-Al alloys with optimal mechanical properties indicates that the two-step diffusion couple technique can be used for high-throughput screening of optimal composition and aging temperatures for alloy design.

**Fig. 9** Measured hardness values of the Ni-13.4 at. % Al/Ni-17.4 at. % Al diffusion couples aging at 1073 K, 1123 K and 1173 K for 14400 s.

### 4. Conclusion

- Based on the prepared Ni-Al two-step diffusion couples, the effect of composition and aging temperature on the microstructure during aging, including the volume fraction, grain size, and shape factor of $\gamma'$ precipitates, as well as the hardness in $\gamma+\gamma'$ two-phase region was comprehensively studied and analyzed.
- By combining the microstructure information measured in this work and the previously proposed evaluation function of mechanical properties, the optimal alloy composition and aging temperature for binary Ni-Al alloys with the best mechanical properties were efficiently screened to be Ni-16.2 at. % Al aging at 1073 K, which was then validated by the experimental hardness values.
- The successful demonstration in binary Ni-Al system indicates that the two-step diffusion couple technique in combination with the evaluation function(s) for key properties can be used to perform the high-throughput screening of the optimal alloy composition and heat treatment process in the target alloys for the best mechanical properties.
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Figure captions

Fig. 1 (a) Ni-rich Ni-Al phase diagram; (b) Schematic diagram for preparation of a two-step diffusion couple.

Fig. 2 X-ray diffraction patterns of Ni-13.4 at.% Al and Ni-17.7 at.% Al alloys after annealing at 1573K for 18000s.

Fig. 3 Measured Al concentration profile of the Ni-13.4 at.% Al/Ni-17.7 at.% Al diffusion couple annealed at 1573 K for 10800s (denoted in red hollow diamond), compared with the model-predicted results by Zhang et al. [28] (denoted in black solid line).

Fig. 4 Backscatter electron images of the Ni-13.4 at.% Al/Ni-17.4 at.% Al diffusion couples after aging at 1073 K, 1123 K, and 1173 K for 14400 s.

Fig. 5 Evolution of shape factors of $\gamma'$ precipitates evaluated from the experiment microstructures (denoted in solid spheres) as a function of Al composition and aging temperature, together with the fitted results using polynomial functions (denoted in solid lines).

Fig. 6 Evolution of diameter of $\gamma'$ particles evaluated from the experiment microstructures (denoted in solid spheres) as a function of Al composition and aging temperature, together with the fitted results using polynomial functions (denoted in solid lines).

Fig. 7 Evolution of volume fractions of $\gamma'$ precipitates evaluated from the experiment microstructures (denoted in solid spheres) as a function of Al composition and aging temperature, together with the fitted results using polynomial functions (denoted in solid lines).

Fig. 8 Evaluated composite indicators for binary Ni-Al alloys as a function of Al composition and aging temperature.

Fig. 9 Measured hardness values of the Ni-13.4 at.% Al/Ni-17.4 at.% Al diffusion couples aging at 1073 K, 1123 K and 1173 K for 14400 s.