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Microstructure development and helium behavior in nickel and vanadium base alloys

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Abstract

Transmission electron microscopy and thermal helium desorption spectrometry (TDS) have been used to investigate the influence of alloying elements on helium behavior and bubble microstructure evolution in FCC (Ni + 1 . . . 7.5 wt%Al) and BCC (V + 10 . . . 40 wt%Ti) metals. The samples were irradiated by 40-keV He^+ ions at room temperature up to a fluence of 5×10^{20} m^{-2}. Post-irradiation annealings were performed at 1023 K (Ni–Al) and 1075 K (V–Ti) for 1 h. It was shown that alloying elements reduced the bubble size ($\bar{d}_b$) and increased their density (ρ_b) in both types of alloys. In the Ni–Al alloys the TDS peaks are displaced to higher temperatures with increasing Al concentration in contrast to V–Ti alloys where the TDS peaks are displaced to lower temperatures with increasing Ti content. However in both systems of alloys the effective activation energy for helium desorption grows with alloying element concentration. The results are discussed in terms of alloying element influence on the mechanisms of bubble growth and migration.

1. Introduction

Helium can have a pronounced effect on the radiation damage of materials and often may be an important reason for degradation of their properties and shortening of the useful life of reactor structural elements. However, in spite of significant research on the influence of alloying elements on radiation damage of materials [1,2], the data on the influence of alloying elements on helium behavior and gas bubbles evolution are inadequate [3–5].

The aim of this paper is to investigate the ion-implanted helium behavior and gas bubble evolution in model alloys of Ni–Al and V–Ti as a function of alloying element concentration.

2. Experimental procedure

The Ni and V-based alloys were prepared from high-purity components. The Ni–Al alloys up to Al concentration $N_{Al} \leqslant 5\%$ and V–Ti alloys up to $N_{Ti} \leqslant 30\%$ were identified as solid solutions; and at higher contents of elements, as supersaturated solid solutions (Ni–Al) or ($\alpha + \beta$) solid solutions (of V–Ti) according to the corresponding equilibrium diagrams.

The samples were irradiated by 40-keV He^+ ions up to a fluence of 5×10^{20} m^{-2} at room temperature. The bubble microstructures were formed during post-irradiation annealings at 1023 K (Ni–Al alloys) and 1073 K (V–Ti alloys) for 1 h. Microstructural investigations were performed on a JEM-2000EX microscope. The details of implanted helium desorption were studied by means of a helium partial pressure meter (sensitivity 10^8–10^{10} at.% He/s). The effective activation energies of helium release (E) for the TDS peaks were calculated by means of a 'tempering' method [3,6].

3. Results and discussion

Figs. 1 and 2 show the typical microstructures of irradiated and annealed alloys As can be seen from Table 1, bubble size (d_b) decreases (by one half) and bubble density (ρ_b) increases (about one order of magnitude) with increasing Al concentration in Ni (from 0 up to 7.5%Al). Contrary to the Ni–Al alloys, a sharp decrease

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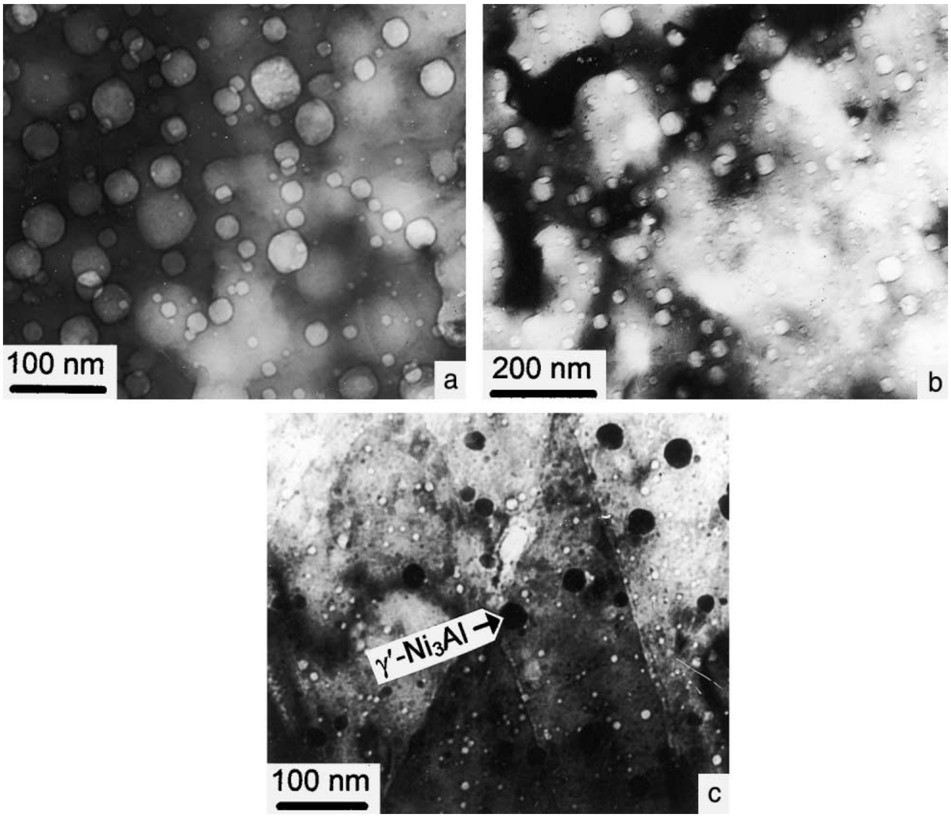

Fig. 1. Typical microstructure of (a) Ni, (b) Ni–4.6%Al and (c) Ni–7.5%Al alloys after irradiation by He^+ ions and post-irradiation annealing at 1023 K for 1 h.

in $\bar{d}_b$ and increase in ρ_b by three orders of magnitude take place in the case of addition of Ti in vanadium. One other difference is that the bubble parameters are independent of Ti content in V (for $10 \leqslant N_{Ti} \leqslant 40\%$). In this case a partially ordered distribution of bubbles is observed in alloys (Fig. 2(b)). The distance between ordered bubbles measured in transmission electron microscopy (TEM) and calculated from extra spots near the central reflection (Fig. 2(c)) is about 4–6 nm. This suggests that the bubble evolution in V–Ti alloys takes place through a stage of ordered structure formation during post-irradiation annealing.

The values of the thermal helium desorption spectrometry (TDS) peak temperatures (T_m) and E are presented in Table 2. As can be seen in Table 2, T_m increases with Al content in the Ni–Al alloys, while in V–Ti alloys T_m decreases with increase of Ti concentration. These data correlate with atom diffusivity change by alloying: Al suppresses the mobility of nickel atoms (Fig. 3(a)) and Ti increases the mobility of vanadium atoms (Fig. 3(b)). The value of E significantly grows with increasing of Al content in Ni (by a factor of two with Al content change between 0 and 7.5 wt%). By doping of Ti in V the value of E also increases but not too significantly. An increase in Ti content from 10 up to 40 wt% did not influence E.

The dependence $E = f(T_m)$ is linear for the solid solutions (Fig. 4). However, the linear dependence is disrupted in supersaturated solid solution Ni–7.5%Al, as well as in alloy V–40%Ti, where the Ti content corresponds to the two-phase ($\alpha + \beta$) alloys.

Helium accumulated via transmutation reactions or implantation at low temperatures can create complexes with vacancies [2,12]. Simple complexes of the He_mV_n type are formed in pure metals, of which the most stable ones dissociate in nickel in the 893–923 K temperature range [13]. As was shown for the Ni–Al system [3,4], the most thermally stable complexes of the $He_mMe_kV_n$ type (where He and Me are helium and alloying element atoms, V is a vacancy) are formed in the alloys along with these complexes. The decomposition temperature of these complexes is higher by about 140°. During post-irradiation annealing a breakdown of the complexes results in formation of bubbles. Two mechanisms of bubble growth were observed – via Ostwald ripening (OR) and bubble migration and coalescence (MC).

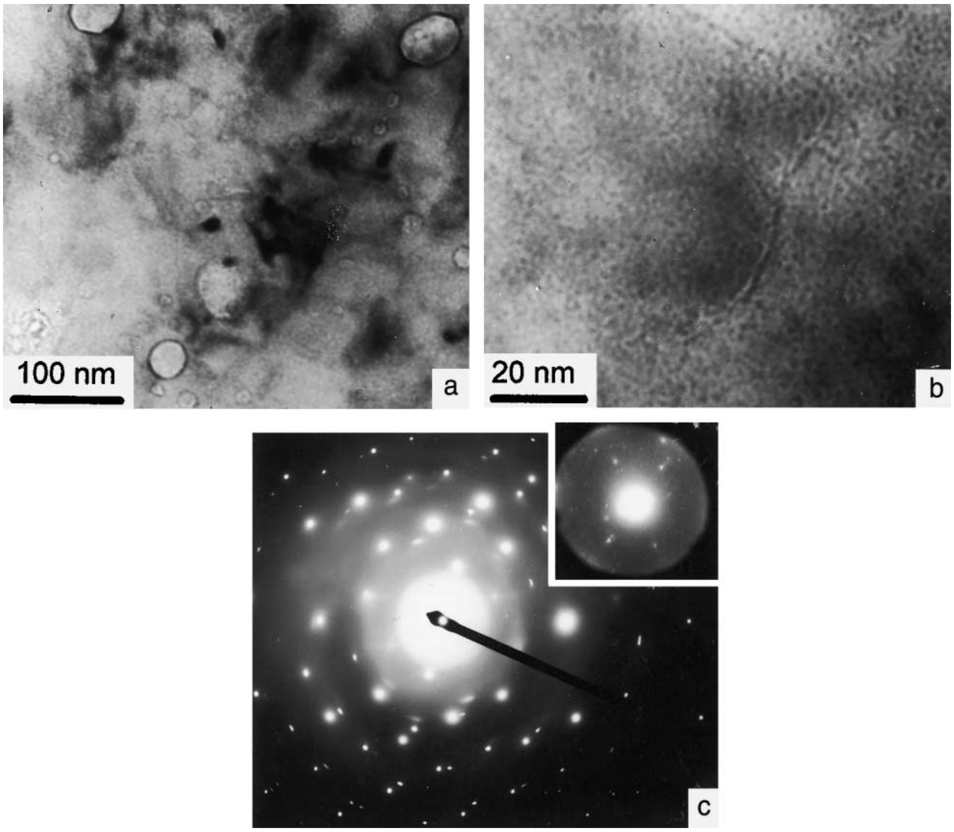

Fig. 2. Typical microstructure of (a) V and (b) V–30%Ti alloy after irradiation by He^+ ions and post-irradiation annealing at 1073 K for 1 h and microdifraction picture (c) from (b).

Table 1
The bubble parameters and their distribution character in the alloys after irradiation by He^+ ions and annealing at 1023 K (Ni–Al) and 1073 K (V–Ti)

Alloy (wt%)	$\bar{d}_b$ (nm)	ρ_b (m^{-3})	Distribution character
Ni	30	$(8.0 \pm 2.4) \times 10^{20}$	Random
Ni–1Al	23	$(6.0 \pm 1.8) \times 10^{21}$	Random
Ni–2Al	21	$(6.2 \pm 1.9) \times 10^{21}$	Random
Ni–4.6Al	17	$(8.5 \pm 2.6) \times 10^{21}$	Random
Ni–7.5Al	14	$(1.1 \pm 0.3) \times 10^{22}$	Random
V	13	$(1.0 \pm 0.3) \times 10^{21}$	Random
V + (10 ...40)Ti	~2	$\sim 10^{24}$	Partial ordered
V + 10Ti + 6Cr + 0.05Zr+0.1	~2	$\sim 10^{24}$	Partial ordered

Coarsening of bubbles occurred through the OR mechanism for the initial stage and the process of bubble growth includes the MC mechanism at the later stage [2,14].

The transport of helium and vacancies released from decomposing complexes is required for bubble growth by the OR mechanism and the changes in bubble parameters depending on annealing exposure time are given as [14]

$$\bar{r} \sim (D_{\mathrm{He}})^{1/n}, \quad \rho_b \sim 1/D_{\mathrm{He}}, \tag{1}$$

for bubble growth by MC mechanism

$$\bar{r} \sim D_b^{1/n}, \quad \rho_b \sim D_s^{1/2}, \tag{2}$$

where D_{He} – the diffusive mobility of helium; D_b – the bubble diffusion coefficient depending on the main diffusion mechanism; D_s – self-diffusion coefficient; n – indicator of degree depending on bubble growth

Table 2
The TDS peak temperatures for uniform annealing rates of 2.5 K/s (Ni–Al) and 4 K/s (V–Ti) and effective activation energies of gas release

Alloy (wt%)	Ni	Ni–1Al	Ni–2Al	Ni–4.6Al	Ni–7.5Al
T_m (K)	1183	1210	1239	1288	1340
E (eV)	1.5 ± 0.2	1.7 ± 0.2	1.8 ± 0.2	2.1 ± 0.2	3.0 ± 0.3
	V	V–10Ti	V–20Ti	V–30Ti	V–40Ti
T_m (K)	1573	1535	1526	1514	1495
E (eV)	2.5 ± 0.3	3.0 ± 0.3	3.1 ± 0.3	3.2 ± 0.3	3.2 ± 0.3

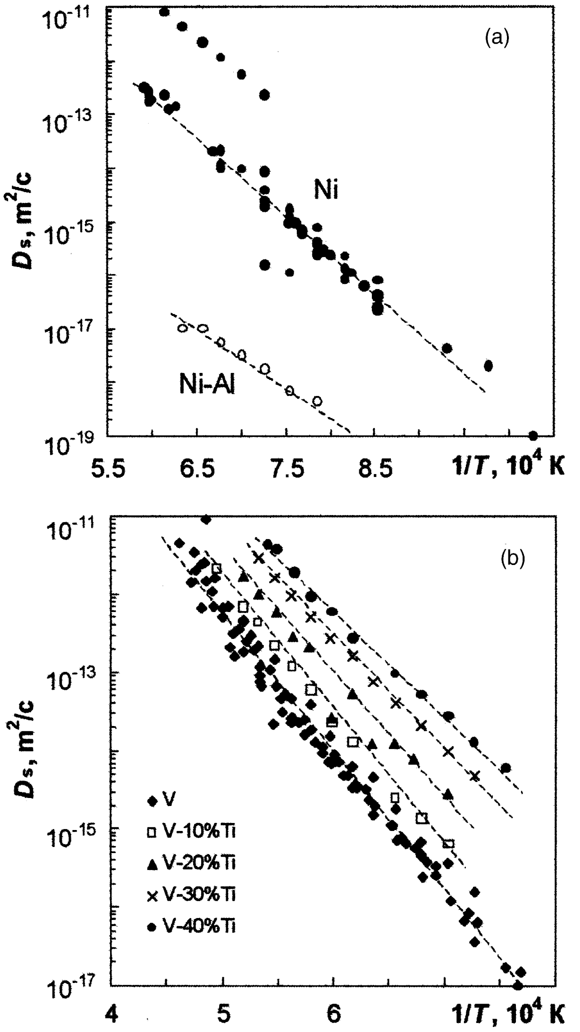

Fig. 3. Influence of Al (5.9…9.4 wt%) on self-diffusion coefficient of (a) Ni [7,8] and influence of Ti on self-diffusion coefficient of (b) V [9–11].

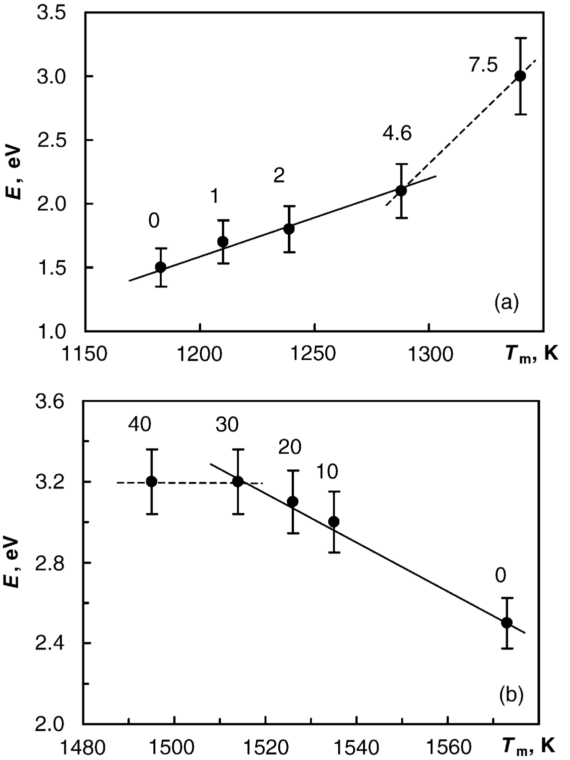

Fig. 4. Effective activation energies of gas release versus TDS peak temperatures for (a) Ni–Al and (b) V–Ti alloys. The numbers at the points are concentration of alloying elements.

mechanism ($n = 2$–6 [3]). As can be seen from expressions (1) and (2), for bubble growth by the OR mechanism they will have a smaller size and a high density with decreasing D_{He}. In the case of the MC mechanism, small bubbles are formed under suppression of their mobility.

Resolvable bubbles are formed in Ni and its alloys even at 773 K (~0.45 T_M were T_M is melting temperature) whereas they are not observed in iron-based alloys [3]. Therefore, we suggested that after nucleation bubble growth in Ni–Al alloys predominantly occurred at 1023 K (~$0.6T_M$ by the MC mechanism. Considering that aluminum significantly suppresses the diffusive mobility of nickel atoms (Fig. 3(a)), which decreases D_b the bubble diameters could be reduced and ρ_b could rise, as can be seen in Table 1. In addition, formation of complexes of the $He_mMe_kV_n$ type [4] also could give rise to an increase in bubble nucleation density.

The post-irradiation annealing temperature for V–Ti alloys (1073 K) is about $0.5T_M$ which is less than for Ni–Al alloys. And so we suggest, that during TEM-investigation of V–Ti alloys we fixed the most initial stage of bubble growth, when the OR mechanism is prevalent and diffusive mobility of helium in the matrix is of major importance. Obviously, Ti atoms in vanadium decrease D_{He} therefore according to Eq. (1), the smallest bubbles with high density were formed in V–Ti alloys (Table 1).

The helium release during uniform heating occurs through the migration and coalescence of bubbles, which results in the bubbles intersecting the sample surface [2,6]. Surface diffusion is prevalent in pure metals and the role of volume diffusion increases in alloys [3]. It is apparent that a decrease of the matrix atom self-diffusion coefficient in Ni–Al alloys with increasing N_{Al} can result in a decrease in bubble migration rate and shift of TDS peaks to higher temperatures. On the other hand, the increase of D_s in V–Ti alloys can lead to an increase of bubble migration rate and shift TDS peaks to lower temperatures (Table 2).

The increase of E for both crystal structures (Table 2) is due to the change of bubble migration mechanism from surface diffusion (for pure metals) to an increasing role of volume diffusion (for alloys) because the volume diffusion coefficient is greater than that for surface diffusion. The activation energy of volume diffusion for nickel is 2.6–2.9 eV [6–8], which is close to the E values for Ni–Al alloys with high N_{Al} (Table 2). This suggests that bubble migration follows the volume diffusion mechanism with increasing Al content in nickel.

The linear relationship between E and T_m (Fig. 4) indicates that the obtained results are correct. However, this is probably right only for solid solutions ($\leqslant$5%Al in Ni and $\leqslant$30%Ti in V). The deviation of the function $E = f(T_m)$ from linearity in the supersaturated Ni–Al alloys (Fig. 4(a)) can be connected with the formation of the segregation or precipitation of secondary γ'-Ni_3Al phase in the range of 873–1073 K and their reverse dissociation at the higher temperatures in the process of heating. Transition of atoms from precipitates to the matrix, consumes vacancies required for bubble migration. Supply of the required vacancies is possible only by the flow of thermal vacancies from free surfaces at higher temperatures [3,13] or climbing dislocations. It seems reasonable to say that this produces the deviation of the function $E = f(T_m)$ from linearity in the direction of increase of E for the Ni–7.5%Al alloy.

The $\alpha \rightarrow \beta$ polymorphic transformation in the V–40%Ti during heating results in the discontinuous increase of D_s reaching up to three orders of magnitude [15]. This significantly facilitates the bubble migration process and for V–40%Ti alloy the linear dependence of the function $E = f(T_m)$ deviates in the direction of E decrease.

4. Conclusion

The obtained experimental results have shown that the substitution alloying elements influence the helium behavior and gas bubble microstructure evolution in metals. In many instances it is determined by changes in matrix atoms diffusion parameters with alloying. At the same time, in spite of increase or decrease of diffusion coefficients by alloying, the bubble migration in pure metals takes place by surface diffusion, and in alloys with an additional contribution from volume diffusion.

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