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Letter to the Editors

Recovery of electron irradiated V–Ga alloys

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Abstract

The recovery characteristics of electron-irradiated V–Ga alloys with 1.2 and 4.6 at.% Ga have been investigated by positron annihilation spectroscopy (PAS). It is found that vacancies created by electron irradiation become mobile in these alloys at ~293 K. This temperature is noticeably lower than that in pure V and V–Ti alloys. The vacancies aggregate into microvoids in V–4.6Ga, but do not in V–1.2Ga. The results indicate that vacancies are bound to Ga–interstitial impurity pairs.

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1. Introduction

At present, V-based alloys are among the primary low-activation materials being investigated for structural applications in fusion reactors [1]. In particular, V–Ti–Cr ternary alloys have been developed having an optimal combination of physical and mechanical properties and resistance to swelling [2–4]. In these alloys, Ti is the alloying element inhibiting the swelling. Recently, it has been suggested that the decay rate of the induced activation in these materials can be improved if Ti is substituted by Ga and so avoiding the production of the long-lived radionucleide ^{39}Ar [5]. It has been demonstrated that V–Ga-based alloys can be produced with tensile, thermal and elastic properties comparable to those for V–Ti–Cr alloys, even with superior ductility [5]. Moreover, their corrosion resistance in liquid Li is acceptable [6]. The neutron irradiation effect on the properties of the V–Ga alloys has been investigated recently [7]. The results indicate that their elastic properties do not change significantly after neutron irradiation at 673 K up to a fluence of 5×10^{25} n/m^2. It was found that Ga reduces the swelling noticeably, but does not suppress it completely. Only when Ce or Cr is added can the swelling in the V–Ga alloys be suppressed.

The capability of oversized solute atoms to inhibit the swelling of the matrix appears to be due to their efficiency to trap radiation-induced vacancies [4,8,9]. Ti and Ga in V are oversized solutes; their atomic radii are 0.162 and 0.167 nm, respectively, versus 0.149 nm for V. However, Ti in V is a much better swelling inhibitory impurity than Ga [7,8,10,11]. To account for this fact and establish the Ga efficiency for vacancy binding we have investigated the recovery of the damage induced by electron irradiation in V–Ga alloys using positron annihilation spectroscopy (PAS).

2. Experimental

V–Ga alloys were prepared by arc-melting in a He atmosphere using as starting materials 99.9% pure V and 99.999% pure Ga. Prior to melting the starting materials, a Ti getter was melted to purify the atmosphere. The alloy buttons were melted several times to ensure a homogeneous composition. Slices cut from the buttons were solution annealed at 1600 K for 5 h in an oil-free vacuum of $\leqslant 10^{-3}$ Pa. After this treatment, the homogeneity and composition of alloy slices were checked by scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDXS). It was found that

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this treatment produces Ga depletion in the surface of the samples because of out diffusion; these data will be reported elsewhere [12]. After removing the Ga depleted layer from the surface by mechanical polishing and chemical etching, the composition of the alloys was determined by EDXS. One pair of identical samples with composition 1.2 Ga at.% and other pair with 4.6 Ga at.% were electron-irradiated with 1.8 MeV electrons up to a dose of 6.0×10^{21} m^{-2} and 4.7×10^{21} m^{-2}, respectively. The irradiations were carried out in a Van de Graaff accelerator with the samples mounted on a cold finger in vacuum. During irradiation the samples temperature remained constant at (263 ± 5) K. After irradiation the samples were stored at 77 K.

The samples were mounted inside a closed-He-cycle cryostat for positron lifetime measurements over the range 10–300 K. During handling the samples were warmed up to room temperature for about 15 min. Afterwards, the measurements were made at room temperature after isochronal annealing for 30 min in steps of 40 K. A ^{22}Na positron source sealed inside kapton foils was used. The positron lifetime spectra were obtained using a spectrometer with a time resolution of 240 ps (FWHM). The spectra were analyzed in terms of one or two exponential components after subtrating background and the source contribution to the spectrum.

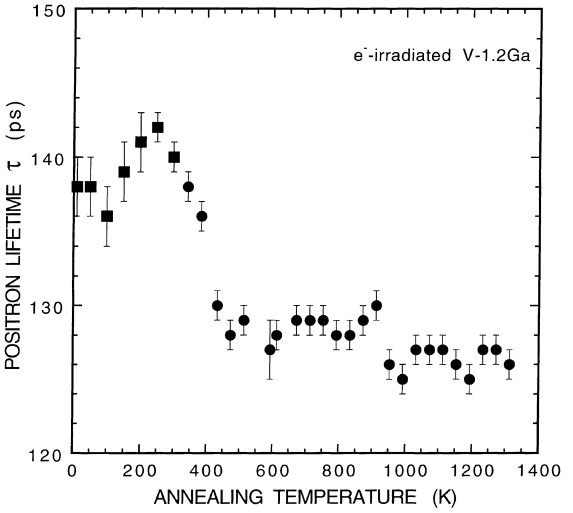

Fig. 1. Positron lifetime recovery for electron-irradiated V–1.2Ga: (●) positron lifetime as a function of annealing temperature; (■) positron lifetime versus temperature after post-irradiation aging at room temperature for 15 min.

3. Results and discussion

Electron irradiation under the conditions described produces changes in the positron lifetime spectrum of the samples revealing the presence of vacancy-type defects. The lifetime spectra for V–1.2Ga present a single component characterized by a value, τ, over the whole temperature range 10–300 K and during the recovery process induced by isochronal annealing. In contrast, V–4.6Ga exhibits a two-component spectrum for post-irradiation annealing below 500 K, characterized by the mean positron lifetime given by

$$\langle \tau \rangle = I_1 \tau_1 + I_2 \tau_2, \quad (1)$$

where I_i and τ_i are the intensity and the lifetime of the spectral components. However for post-irradiation annealing above 500 K, V–4.6Ga also exhibits a single lifetime spectrum. Figs. 1 and 2 show the results from measurements in equilibrium at $T \leqslant 300$ K, along with those from the isochronal annealing experiments.

The results from the measurements in equilibrium over the range 10–300 K differ qualitatively. The τ value for V–1.2Ga shows a trend to increase with increasing temperature up to 250 K, see Fig. 1. However, the $\langle \tau \rangle$ value for V–4.6Ga stays constant at 143 ps for $T \leqslant 300$ K, as shown in Fig. 2(a). The two-component spectra for V–4.6Ga exhibit a constant second-lifetime component of (330 ± 20) ps, see Fig. 2(b). This is a typical lifetime value for positrons trapped in microvoids in metals [13]. The intensity of this lifetime component increases significantly after annealing at 340 K, see Fig. 2(c), but then decreases for annealing temperatures above 400 K. This produces the $\langle \tau \rangle$ increase observed for V–4.6Ga prior its recovery. The above results indicate that the irradiation-induced vacancies coalesce into microvoids at $T < 340$ K in V–4.6Ga being unstable for annealing temperatures above 400 K. On the contrary, in V–1.2Ga there is no evidence for vacancy clustering into microvoids even though the results indicate that vacancies migrate at $T \leqslant 293$ K. From the present experiments we cannot determine precisely the migration temperature for vacancies in these alloys. However, from the results is clear that vacancies must be mobile at 293K, or even below.

It is well known that residual interstitial impurities in V shift the temperature of the recovery stage III, i.e., the migration temperature for free vacancies. In ultra pure V, this temperature is 170 K [14,15]. Residual impurities in V, such as O, N and C, shift the migration temperature for vacancies to temperatures above 400 K because of the capability of these impurities to bind vacancies [16,17]. Annealing out of vacancies released from interstitial impurities, in electron-irradiated pure V, is revealed by a steep recovery stage of the mean positron lifetime at $T \sim 400$ K [18]. The addition of Ti to V shifts this recovery stage to higher temperatures, i.e., ~460 K [18]. Since the present results reveal that vacancies are mobile at 293 K in V–Ga alloys, it has to be accepted

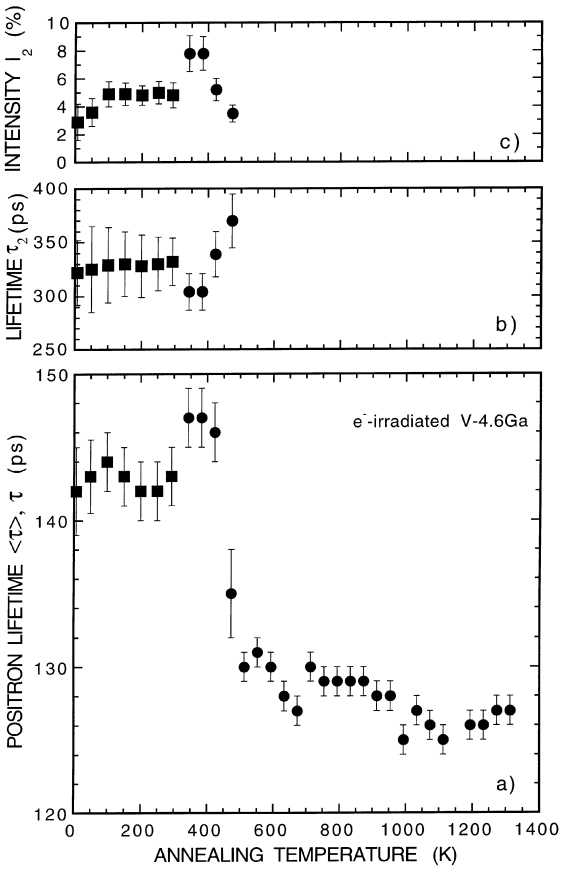

Fig. 2. Recovery of the positron annihilation parameters for electron-irradiated V–4.6Ga: (●) annihilation parameters as a function of annealing temperature; (■) annihilation parameters versus temperature after post-irradiation aging at room temperature for 15 min.

that solute Ga in V reduces the binding energy between vacancies and interstitial impurities. This effect would be explained assuming that the solute Ga in V, like Ti, is a sink for interstitial impurities. Therefore, the defects responsible for the positron trapping in electron-irradiated V–1.2Ga would be vacancy–Ga–interstitial impurity complexes. The lifetime for positrons trapped at these complex defects is expected to be shorter than the one for a bound vacancy–interstitial impurity pair, i.e., 182 ps [18], preventing a reliable decomposition of the lifetime spectra in the case of the V–1.2Ga samples.

Vacancies released from the vacancy–Ga–interstitial impurity complexes at ~293 K do not form three-dimensional clusters in the V–1.2Ga samples, but appear to sink into interstitial type defects created by the electron irradiation. The fact that microvoids are formed in the V–4.6Ga samples and not in V–1.2Ga, indicates that a threshold Ga concentration is required to promote the microvoid formation. Moreover, the temperature at which the microvoids start to be unstable in V–4.6Ga, i.e., ~400 K against 500 K in cold-rolled pure V [19] and 780 K in electron-irradiated V–Ti alloys [18], suggests that the nucleation sites are related to the Ga content. The nucleation sites could be complex defects formed by bound Ga–interstitial impurities.

It should be noted that the bulk positron lifetime, i.e., (126 ± 1) ps, is attained for annealing temperatures above 900 K in the both alloys. The recovery curves shown in Figs. 1 and 2(a) suggest the existence of a second recovery stage at ~ 900 K. A second recovery stage at 900 K has been found for the positron lifetime of cold-rolled pure V [19]. The origin of this recovery stage is uncertain, although it has been suggested that it could be due to the disappearance of the positron traps associated with precipitates [19].

4. Conclusions

The positron lifetime recovery in electron-irradiated V–1.2Ga and V–4.6Ga alloys indicates that solute Ga reduces the temperature at which vacancies become mobile in V. The results are consistent with the formation of vacancy–Ga–interstitial impurity complex defects by electron irradiation. These defects appear to be unstable at temperatures below ~293 K. For V–4.6Ga, vacancies released from these defects coalesce into microvoids which are stable up to temperatures around 420 K. Mobile vacancies do not coalesce into microvoids in V–1.2Ga irradiated under the present conditions.

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