Effect of Fast Neutron Irradiation on the Microstructure in Particle Dispersed Ultra-fine Grained V-Y Alloys

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An alloy having ultra-fine grains with dispersed particles is expected to have the resistance of irradiation embrittlement. We examine the microstructures of V-1.6 and 2.6Y alloys, having ultra-fine grains with dispersed particles, after neutron irradiation at temperatures between 290 and 800°C. The grain size was the level of a few hundreds of nanometers, and the size in diameter of V-2.6Y alloy was much smaller than that of V-1.6Y alloy, where the irradiation did not change the size. The number density of particles in V-2.6Y alloy was higher than that in V-1.6Y alloy. Voids were formed only at 290°C, where the number density of voids in V-2.6Y alloy was smaller than that in V-1.6Y alloy. The void formation was efficiently suppressed due to fine-grains and dispersed particles.

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1. Introduction

Vanadium alloys have been recognized as a candidate of structural material for fusion reactors¹-⁴ because of its low-induced radioactivity.³ They will be seriously embrittled by neutron irradiation at temperatures below 400°C.⁵,⁶,⁷ Since such embrittlement is caused by irradiation-induced lattice defects, it is effective to introduce sinks of the defects in the material to suppress the embrittlement. Grain boundary and interphase boundary act as sinks for irradiation-induced defects. An alloy possessing ultra-fine grains with dispersed particles is, therefore, expected to have the resistance of such irradiation embrittlement.⁸-¹⁰ An alloy having such the microstructures can be produced by mechanical alloying (MA) method. MA-treated vanadium powder, however, has large affinity with interstitial impurities such as oxygen and nitrogen, yielding a lack of ductility of vanadium alloys produced by MA without irradiation.¹¹,¹² One of our authors had successfully developed vanadium alloys having ultra-fine grains with dispersed particles containing extremely little interstitial impurities by using MA method.¹³ Three-point bend impact tests showed that the alloys exhibited good ductility even at 77 K.¹³ To the authors’ knowledge, no work has focused on the microstructural evolutions of such alloys after irradiation. The purpose of this study is to examine the microstructures in the alloys after neutron irradiation at temperatures between 290 and 800°C.

2. Experimental Procedures

Both V-1.6Y and V-2.6Y alloys (mass%), which were made by MA and hot isostatic pressing (HIP), were machined into disks of 3 mm in diameter and 0.1 mm thick. Detail procedures for preparation of the alloys have been reported in the previous study.¹³ The disks wrapped with Ta foil were encapsulated into evacuated quartz tubes. Dehydrogenation was done at 1000°C for 3.6 ks. They were irradiated with fluence of 1.3 × 10²⁴ n/m² (about 0.25 dpa) at 290°C and of 3.7 × 10²⁴ n/m² (0.7 dpa) at 600 or 800°C in the Japan Materials Testing Reactor (JMTR). Microstructures were analyzed by transmission electron microscopy (TEM). Thin foils for TEM were prepared by means of twin-jet electropolishing using a solution of 5 vol% H₂SO₄ and 95 vol% CH₃OH at around 5°C at 20 V, and examined in both analytical JEM2000FX microscope operating at 200 kV and JEM4010 microscope operating at 400 kV.

3. Results

3.1 Microstructures before irradiation

Figure 1(a) shows the histogram of grain size in both V-1.6Y (white) and V-2.6Y (black) alloys before irradiation. The V-1.6Y alloy contained fine grains of about a few hundred nanometers in diameter mixed with a small amount of coarse grains of about a few micrometers in diameter. On the other hand, V-2.6Y alloy had fine grains of about a few hundred nanometers in diameter. It should be noted that the alloys used in this study had quite small grains than that in melting-fabricated vanadium alloys. A small amount of coarsened grain was probably introduced due to unsatisfied MA treatment. Mean diameters of grains in the fine grain region (less than 1 μm in diameter) in V-1.6Y and V-2.6Y were 303 and 170 nm, respectively.

Size distributions of particles in V-1.6Y (white) and V-2.6Y (black) alloys before irradiation are shown in Fig. 1(b). The particles were formed only in the region of fine grains. In the region of coarse grains, the particles were not formed, indicating concentration of yttrium and interstitial impurities such as oxygen and nitrogen was assumed to be quite low in coarse grains. Mean diameters of the particles in V-1.6Y and V-2.6Y alloys were 25 and 21 nm, respectively.

Although the mean diameters of the particles in both alloys were almost same, the number density of particles in V-2.6Y alloy was much larger than that in V-1.6Y alloy, which led to a suppression of grain growth resulting in miniaturization of grains as can be seen in Fig. 1(a).
3.2 Microstructural evolutions after irradiation

The grain size distributions before and after irradiation were almost same. Coarse and fine grain regions also coexisted in irradiated V-1.6Y alloy. In the region of coarse grains, no particles were observed. Figure 2 shows the TEM micrographs for V-1.6Y alloy irradiated at 290°C; (a) bright field image and (b) dark field image by (420) Y₂O₃, respectively. Grain boundary is indicated by dash line in Fig. 2(a). Y₂O₃ particles and large voids are indicated by longitudinal and transverse arrows in (a), respectively. Magnified images of (a) are shown in Figs. 2(c) and (d), which were taken under the condition of under- and over-focus of objective lens, respectively.

V-2.6Y alloys were 22.2 and 28.9 nm, respectively. The number density of larger voids in V-2.6Y alloy was much smaller than that in V-1.6Y alloy. Figure 5 shows size distributions of particles before (white) and after (black) irradiation in (a) V-1.6Y and (b) V-2.6Y alloys, respectively. After irradiation, particle density decreased and its distribu-
tion shifted to smaller side in both alloys. This means dissolution of particles occurred by irradiation. It should be noted that twin formation could be detected in the alloys irradiated at 290°C. The detail of it is now under investigation and will be reported in future.

Bright field image of V-1.6Y alloy irradiated at 600°C is shown in (a). Twins are clearly observed in some grains as shown in (b).

Fig. 6 Bright field image of V-1.6Y alloy irradiated at 600°C is shown in (a). Twins are clearly observed in some grains as shown in (b).

Fig. 4 Size distribution of voids in both V-1.6Y (white) and V-2.6Y (black) alloys for; (a) small voids under 5 nm in diameter and (b) large voids over 5 nm in diameter.

Fig. 5 Comparisons of size distributions of particles before (white) and after (black) irradiation at 290°C in (a) V-1.6Y and (b) V-2.6Y alloys, respectively.

Fig. 7 Comparisons of size distributions of particles in V-1.6Y alloy before (white) and after (black) irradiation at (a) 600°C and (b) 800°C, respectively.

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coarse grain regions. Magnified images of both the regions are shown in Figs. 8(b) and (c), respectively. Particles were only observed in the region of fine grains. Voids and twins were not observed in both the grain regions. In the region of coarse grain, however, vacancy loops with stacking fault fringes were observed, as seen in Fig. 8(c). Composition analysis of these loops by energy dispersive X-ray spectroscopy equipped in the TEM revealed no yttrium enrichment into loops. Extra spots in diffraction pattern from these loops were hardly obtained in the present study. Loop free zone of about 500 nm in width was clearly observed in the vicinity of grain boundary in the coarse grains.

4. Discussion

4.1 Void formation by irradiation

voids were formed in the alloys irradiated at 290°C, which indicates that vacancies are mobile efficiently at this temperature. The migration of vacancy is not predominate in melting-fabricated vanadium alloys irradiated at temperatures below 350°C, therefore, the impurity level of this MA alloys is assumed to be low.

Small voids were formed homogeneously in the matrix of V-1.6Y alloy. Interstitials, on the other hand, migrate and annihilate into sinks and/or aggregate resulting in formation of large dislocation loops. Vacancies due to cascade damage form the small voids as seen in V-1.6Y alloy. In V-2.6Y alloy, however, smaller amount of small voids in the matrix was observed. Since the grain size of V-2.6Y alloy is much smaller than that of V-1.6Y alloy, most vacancies in V-2.6Y alloy might sink at grain boundaries, resulting in small amount of voids formation.

Large voids were formed only close to the grain boundaries or particles. Since grain boundaries and particles are sinks for interstitials, substantial excess vacancies would exist close to grain boundaries and particles resulting in formation of large voids.

4.2 Evolution of dispersed particle under irradiation

Although grain size was not changed by irradiation, particle size distribution was depended on irradiation temperature. The dissolution of Y$_2$O$_3$ particle was observed after the irradiation at 290°C. The results indicate that the particle can be dissolved by displacements of its constituent atoms into matrix. If there is no constraint for migration of yttrium-interstitials and oxygen atoms, they could return to the particles. However, there are many voids that could trap yttrium-interstitials and oxygen atoms in the alloy. Yttrium-interstitials and oxygen atoms, therefore, have little chance to return to the particles resulting in shrinkage of the particles.

Coarsening of Y$_2$O$_3$ particles occurred by irradiation at 600°C is clearly confirmed in Fig. 7, i.e., the number density of larger particles increased. Although Y$_2$O$_3$ particles might dissolve during irradiation, the recombination of yttrium and oxygen would quickly take place at high temperature.

Coarsening and nucleation of Y$_2$O$_3$ particles in the region of fine grains were detected during irradiation at 800°C. Although coarsening of the particles in the alloy irradiated at 800°C was observed, the number density of small particles did not decrease so much. This means that additional formation of Y$_2$O$_3$ particles occurred during irradiation. Volume fraction of Y$_2$O$_3$ particles was increased by irradiation at 800°C. Assuming all oxygen atoms in the alloy before irradiation, 0.450 at%, are combined with yttrium atoms, the amount of yttrium atoms combined with oxygen should be 0.3 at%. Since the amount of yttrium atoms combined with oxygen before irradiation is smaller than a total amount of yttrium in the alloy, 0.895 at%, free yttrium still exist in the matrix. Additional oxygen, which might come from the environment during irradiation, induces nucleation of Y$_2$O$_3$ particles. In the region of coarse grains, on the other hand, plate-shaped vacancy loops with fringes were formed. However, stacking fault energy in bcc structure is generally too high to form stacking fault. It is then possible that stacking fault energy is reduced by the Suzuki-effect due to impurity accumulation to the stacking fault. The stacking fault, therefore, probably is stabilized with light elements such as oxygen, nitrogen and/or carbon.
5. Conclusions

Microstructural evolutions of V-1.6 and 2.6Y alloys by fast neutron irradiation at temperatures between 290 and 800°C are summarized as follows.

(1) Small and large voids of about 2 and 25 nm in diameter, respectively, were formed by irradiation at 290°C, while no voids were observed at 600 and 800°C. Vacancy could migrate well at 290°C, indicating the concentration of impurity in the alloy is probably quite low.

(2) The number density of voids in V-2.6Y alloy was much smaller than that in V-1.6Y alloy. The alloy with fine grains and dispersed particles can suppress the formation of irradiation induced lattice defects.

(3) The grain size of the alloys was not changed by irradiation.

(4) Shrinkage of the particles occurred by irradiation at 290°C. Irradiation induced yttrium-interstitials and oxygen atoms from Y₂O₃ particles at 290°C were probably trapped by voids and/or vacancy clusters.

(5) Coarsening of Y₂O₃ particles was observed in V-1.6Y alloy irradiated at 600 and 800°C.

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