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Journal of Nuclear Materials 283–287 (2000) 492–497

journal of nuclear materials

www.elsevier.nl/locate/jnucmat

Mechanical behavior and microstructural evolution of vanadium alloys irradiated in ATR-A1

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Abstract

An irradiation experiment has been done in the ATR-A1 to investigate irradiation behavior of vanadium alloys in the low temperature regime from 200°C to 300°C with damage levels of 3 to 4 dpa. In creep measurements, creep tubes of V–3Fe–4Ti–0.1Si with inner pressures up to 165 MPa did not rupture during irradiation. The effective strain rate of creep was below 0.2% dpa^{-1} and it showed the same tendency as V–4Cr–4Ti alloys. In Charpy impact tests, all specimens of V–4Cr–4Ti–0.1Si and V–3Fe–4Ti–0.1Si showed brittle behavior at room temperature and the DBTT increased to 60–150°C. The fracture surface showed cleavage. Tensile tests conducted both at room temperature and at the irradiation temperature showed significant irradiation hardening and brittle responses. TEM showed that high densities of tiny defect clusters were formed in V–Cr–Ti and V–Fe–Ti alloys. Precipitates could not be seen in specimens irradiated below 300°C, however, fine defect clusters are considered to be the origin of brittle behavior in V–Cr–Ti alloys irradiated at low temperatures. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Vanadium-based alloys are being developed as candidate materials for structural materials of fusion power reactors because of their potential for low activation and attractive high-temperature properties [1]. While some properties, e.g., swelling behavior, have been well studied, mechanical properties, such as creep and Charpy impact behavior have not. They are important in estimating the lifetime of structural materials, and make it necessary to optimize the composition of vanadium-based alloys.

Some irradiation creep tests for vanadium alloys have been reported, but the data are extremely limited. Vitek et al. [2] have reported a creep rate of 10^{-5} dpa^{-1} MPa^{-1} for V–20Ti creep tubes irradiated in EBR-II at 700°C to a damage level of 22 dpa^{-1}. It was concluded that the irradiation creep did not affect creep behavior significantly without helium implantation. On the other hand, Troyanov et al. [3] have indicated that there are two regions in the stress dependence of creep rate, and the creep rate is significantly greater in the higher stress region during irradiation at 445°C. Tsai et al. [4,5] have investigated the creep rate of V–4Cr–4Ti irradiated in ATR-A1. They suggested that there was no significantly high creep rate below 300°C.

In the case of ductile–brittle transition behavior, there have been many studies. In fast reactor irradiation studies in a temperature range of 425–600°C good resistance of V–4Cr–4Ti alloys to irradiation-induced embrittlement has been reported. However, several recent studies for 100–400°C irradiation have reported significant irradiation hardening and embrittlement at low doses of 0.1–5 dpa [6–8].

To investigate various mechanical properties under irradiation of vanadium-based alloys, the ATR-A1 neutron irradiation experiment was conducted under the US/Japan JUPITER collaboration program on fusion structural materials. Since the ATR-A1 irradiation was performed at 200–300°C to 5 dpa, it offered an opportunity to investigate the degradation in properties of vanadium-based alloys. In this paper, experimental results on vanadium-based alloys irradiated in ATR-A1 are reported. An important factor is identified for

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PII: S0022-3115(00)00310-X

mechanical property degradation under neutron irradiation.

2. Experimental procedure

Various kinds of specimens were prepared for the ATR-A1 experiments: creep tubes, 1.5 mm Charpy V-notched specimens, miniaturized tensile specimens and transmission electron microscopy (TEM) specimens. The tubes were prepared from one kind of material; V–3Fe–4Ti–0.1Si. Details of creep tube specimen fabrication are described in [4]. Charpy and tensile specimens and TEM discs were prepared from V–4Cr–4Ti–0.1Si (IMR/Tohoku-heat, VM9401R-heat), V–4Cr–4Ti (USA heat, #832665-heat) and V–3Fe–4Ti–0.1Si (IMR/Tohoku-heat, VM9502-heat; in [4] labeled as VM9407). The size of the Charpy specimens was $1.5 \times 1.5 \times 26$ mm^3 with a V-notch (1.5CVN). The size of the miniaturized tensile specimens (SSJ specimens) was $16 \times 4 \times 0.25$ mm^3. The list of impurity level of the specimens is given in Table 1. VM9401-heat and VM9502-heat had a higher oxygen content than #832665-heat, but less carbon and nitrogen content. Two sets of heat treatment for materials were chosen: one annealing treatment was at 950°C for 2 h in vacuum of $< 2 \times 10^4$ Pa with specimens wrapped in a protective Ta foil and a Zr getter foil, the other annealing treatment was at 1100°C for 2 h in the same environmental condition. Irradiation was performed in ATR and lithium-bonded subcapsules were used. The calculated damage level and irradiation temperature are shown in Table 2. After irradiation, these capsules were disassembled and dimensional measurements of creep tubes and Charpy impact tests were performed. Tensile tests, TEM observation and positron annihilation lifetime measurements were conducted.

3. Results

3.1. Creep tube measurement

All pressurized specimens were intact after ATR neutron irradiation. Diameter measurements of two creep tubes of VM9502 (V–3Fe–4Ti–0.1Si) were completed. The gamma activity of VM9502 was 40 times higher than #832665-heat after two years of cooling. Fig. 1 shows results of dimensional measurements of unirradiated and irradiated VM9502 samples with a laser profilometer. The cross-section of the unirradiated sample is elliptic, especially at the end of the tube. This must have been caused by gripping when the end plugs were attached to the tube by welding. However, it

Table 1
Chemical composition of the three alloys investigated

Heat number	Nominal composition	Interstitial content (wppm)			
		O	N	C	Si
832665-heat	V–3.8Cr–3.9Ti	310	85	80	780
VM9401-heat	V–4.3Cr–4.3Ti–0.12Si	528	7	–	1200
VM9502-heat	V–3.6Fe–4.3Ti–0.08Si	482	6	33	800

Table 2
The summary of irradiation condition

Material	Type of specimen	Temperature (°C)	Damage (dpa)
VM9401 V–4Cr–4Ti–0.1Si	1.5CVN	286	4.3
		259 (950°C anneal)	3.9
		234 (1100°C anneal)	3.5
	Tensile (SSJ)	286	4.3
		234	3.5
	TEM disk	300	4.1
		259	3.9
VM9502 V–3Fe–4Ti–0.1Si	Pressurized tube, 1.5CVN, tensile (SSJ)	300	4.1
		212	3.0
	TEM disk	300	4.1
		259	3.9
ANL heat-#832665 V–4Cr–4Ti	1.5CVN	295	4.5
		213	1.5

became rounded by irradiation as shown in Fig. 1(b) due to irradiation swelling and creep. Billone has reported the effective stresses of the creep tubes [4,9]; they were 163 and 165 MPa, for specimens irradiated at 300°C and 212°C, respectively. The measured diametral strains were $0.75 \pm 0.10\%$ and $0.91 \pm 0.11\%$ for 212°C and 300°C samples, respectively. Combined with the previously obtained data, these strains are converted into effective strain rate and strain rate per unit stress, i.e., 2.2×10^{-3} dpa^{-1} and 1.3×10^{-5} dpa^{-1} MPa^{-1} for 212°C irradiation and 2.2×10^{-3} dpa^{-1} and 1.5×10^{-5} dpa^{-1} MPa^{-1} for 300°C irradiation, respectively. The measured strain rates were similar to the data on the #832665-heat alloy, reported by Tsai et al. [4,5]. These creep rates are lower than those reported by Troyanov et al. [3]. No abrupt acceleration of creep rates at high stress levels, as reported by Troyanov, was found below 300°C. Based on the data obtained in the present study, the creep strength of vanadium alloys at temperatures below 300°C should not be an issue problem during neutron irradiation and during operation of a fusion reactor with a liquid lithium blanket system. More testing is required to obtain the irradiation creep strength of vanadium alloys at high temperatures using neutron irradiation facilities.

3.2. Charpy impact tests

Charpy impact tests were conducted in air with a Dynatup drop-weight tester with an impact speed of 5 m s^{-1}. The results of the Charpy tests are summarized in Figs. 2(a) and (b). The data of unirradiated VM9401 annealed at 950°C and 1100°C are given in the same figure as a solid line. Unirradiated VM9401 annealed at 950°C showed ductile behavior over the entire temperature range. The data obtained by previous work show

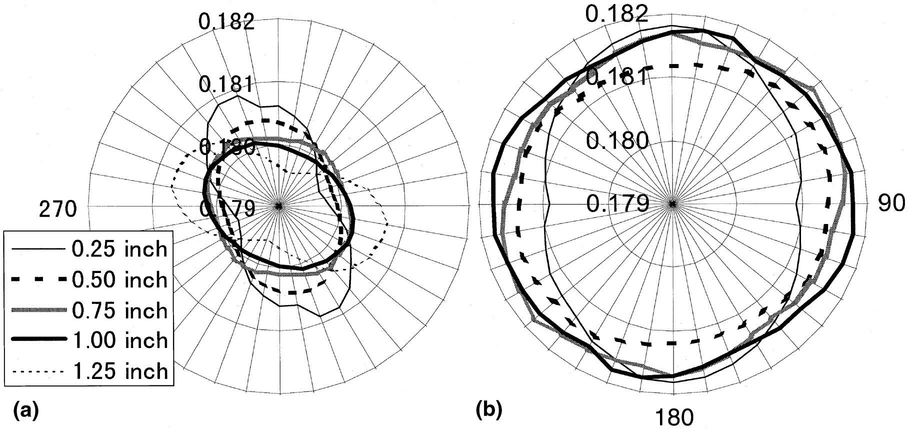

Fig. 1. The cross-section plots of dimensional measurement of (a) unirradiated and (b) irradiated VM9502 samples with a laser profilometer. Irradiation temperature is 212°C and damage level is 3 dpa. The indexes of lines indicate the distance from tube edge. The distance from an edge of 0.5 in. is at the center of the length of the tube. The scale on a lane shows the radius of the tube, from 0.179 to 0.182 in.

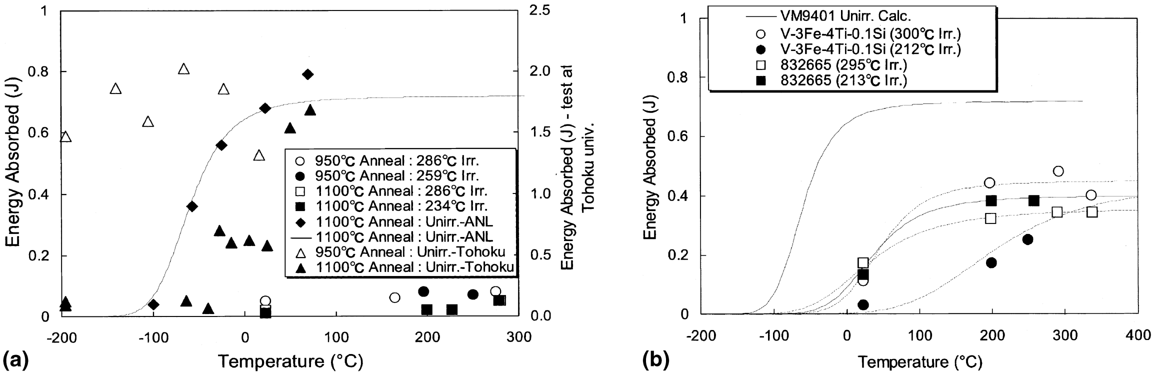

Fig. 2. The temperature dependence of the absorbed energy in impact tests (a) shows the results of unirradiated- and irradiated-VM9401-heat specimens. The solid line shows the DBTT curve of the unirradiated VM9401-heat annealed at 1100°C for 2 h, as a reference (b) shows the results of irradiated VM9502-heat and #832665-heat specimens.

that the DBTT increases up to near room temperature when it is annealed at 1100°C [10] as shown in Fig. 2(a). However, all of the VM9401 specimens irradiated between 200°C and 300°C showed fully brittle behavior up to 300°C. The fracture surfaces of VM9401 showed cleavage and brittle fracture. The VM9502-heat and #832665-heat also show same cleavage fracture mode at 60–130°C, which is the transient temperature regime in ductility. The upper shelf energy is reduced to half of the unirradiated value.

3.3. Tensile tests

Tensile tests were performed in a vacuum of $<10^{-4}$ Pa at both room temperature and irradiation temperature (200°C or 300°C). The strain rate was 6.67×10^{4} s^{-1} (corresponding to a cross-head speed of 0.2 mm/min). Fig. 3 shows the tensile behavior of irradiated and unirradiated VM9401-heat specimens. The unirradiated sample shows uniform elongation up to 40% and a yield stress of about 310 MPa at room temperature. Irradiated samples show significant irradiation hardening and loss of ductility in room temperature tests as well as in tests at the irradiation temperature. The yield stress increases to over 900 MPa and uniform elongation is below 1%. River patterns and cleavage fracture modes can be seen around the edge of the fracture surface by SEM observation.

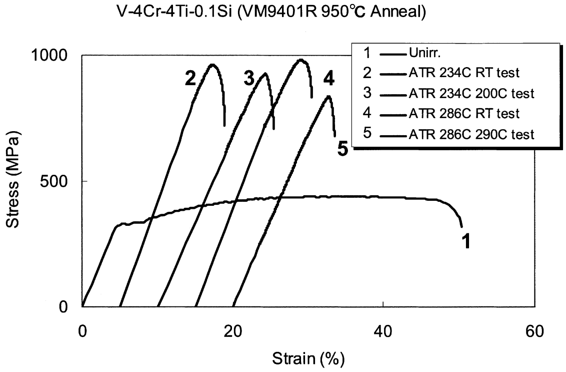

Fig. 3. Stress–strain curves of SSJ tensile tests for VM9401-heat annealed at 950°C. Tensile test temperatures are both room temperature and near the irradiation temperature (200°C and 290°C).

3.4. TEM observation

Fig. 4 shows a set of TEM micrographs of VM9401-heat irradiated at 259°C. In the bright field micrograph of Fig. 4(a), a lot of fine dot images can be seen inhomogeneously around the tangled dislocations. The dislocation density and the density of the feature dot are quite high about 4×10^{14} m^{-2} and 2×10^{23} m^{-3}, respectively. From the weak-beam dark field image in Fig. 4(b), the fine dot images show features of a dislocation loop structure. In order to distinguish dislocation loops from titanium oxide precipitates, the images obtained from the reflection at 3/4[2 0 0] and 2/3[2 2 2] were taken [11]. But no contrast from precipitation spots appeared on micrographs, as shown in Fig. 4(c). These features indicate that the fine dot images must be dislocation loops, and not titanium oxides precipitates. No precipitates can be seen in VM9401-heat irradiated at 300°C, where the dislocation network has coarsened and the dot features have grown into dislocation loops. The same tendency appeared in the irradiated VM9502-heats.

4. Discussion

Significant irradiation hardening is the origin of degradation for some mechanical properties at

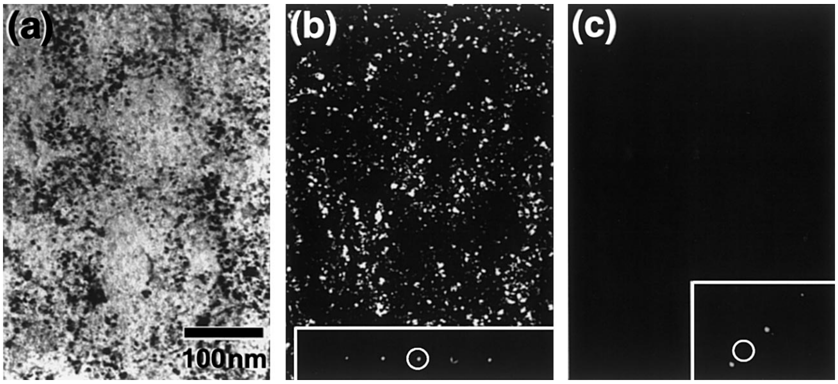

Fig. 4. A set of TEM micrographs of VM9401-heat (V–4Cr–4Ti–0.1Si) irradiated at 212°C. (a) Bright field image taken with $g = 110$, (b) weak-beam dark-field image taken with $g = 110$ and g-$5g$ s deviation condition, and (c) precipitates' spot image taken by 3/4[2 0 0] position (bottom figure shows the position taken from diffraction pattern).

temperatures below 400°C. Rice and Zinkle [12] have reported that lots of small defect clusters on $\{1\,0\,0\}$ planes with the Burgers vector of 1/2[1 1 0] should be the main source of irradiation hardening. In this work, a lot of fine defects can be detected but no precipitates. The nature of the fine defect, however, is still not clear. From previous work, radiation-induced precipitation of titanium oxides is one of the significant contributing factors to the increase of yield stress in vanadium alloys irradiated in JMTR in a temperature regime from 200°C to 350°C [13]. In the current work, however, no irradiation-induced precipitates could be detected by TEM.

An estimation of the yield stress increase from microstructural analysis has been done using the well-known dispersed barrier hardening equation [12],

$$\Delta\sigma_y = M\alpha\mu b(Nd)^{1/2}, \tag{1}$$

where $\Delta\sigma_y$ is the increase in strength compared to the unirradiated value, μ the shear modulus, b the magnitude of the $a/2[1\,1\,1]$ Burgers vector of the dislocations, N and d are the density and size of the defect clusters. M is the Taylor factor, which is set to $M = 3$ as suggested by Rice and Zinkle [12]. The calculated value of the barrier strength for the dislocations was 0.35 from the previous data revised by using the $M = 3$ estimation. The value of $\alpha \sim 0.2$ for precipitates was also derived in the other work for neutron irradiated V–Cr–Ti and V–Fe–Ti alloys in JMTR [13,14]. From these calculations, a yield stress increase of 675 and 465 MPa, for 259°C and 300°C irradiation are obtained, respectively. The measured yield stress increases are as 580 and 470 MPa for the 259°C and 300°C irradiation, respectively. Hence, the calculation from microstructural analysis is in good agreement with the experimental value of yield stress increase in irradiated vanadium alloys. The percentage of tangled dislocation contribution to irradiation hardening is 75%. Therefore, the yield stress increase is mainly due to tangled dislocations. The tangled dislocations form by the growth of dislocation loops nucleated at the initial stage of irradiation, while the fine defects form close to well-grown dislocation lines. It is likely that the fine defect clusters decorate and pin dislocation lines. Satou [15] has provided another result of microstructure and tensile behavior of V–4Cr–4Ti–SiAlY alloys irradiated in ATR-A1, which also shows significant irradiation hardening but rather good tensile ductility. Satou also observed a lower density of dislocation loops than found in this work. The difference in irradiation performance between Satou's results and this work is the addition of minor elements such as Si, Al and Y. The impurity level of both specimens is similar but the morphology of the microstructure might be different since less oxygen and carbon are contained in the matrix in vanadium alloys containing Si, Al and Y. The minor elements, such as Si, Al and Y, bind tightly with interstitial impurities and form precipitates such as oxides and nitrides. Moreover, these minor elements should work as scavengers for oxygen and nitrogen during neutron irradiation, while nucleation of dislocation loops should be delayed and growth of dislocation loops is suppressed.

5. Conclusions

Various types of mechanical test and microstructure investigation were performed on vanadium alloys irradiated in ATR at 200–300°C with damage levels of 3 to 4 dpa. The features found are as follows:

1. No significant creep acceleration appeared in V–Cr–Ti or V–Fe–Ti alloys during neutron irradiation below 300°C.
2. Neutron irradiation below 300°C leads to significant hardening and embrittlement of vanadium alloys in tensile and Charpy tests.
3. Irradiation hardening is caused by a high density of fine defects and tangled dislocations.

Impurity control and addition of minor element can improve irradiation-induced degradation of properties of vanadium alloys for fusion reactor applications.

Acknowledgements

The author is grateful in particular to L. Nowicki and D. Pushies for their skillful work and good suggestion in these experiments. This work was supported by JUPITER program (Japan–USA Program of Irradiation Test for Fusion Research) based on Monbsho US-DOE Annex-I.

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