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Modeling of microstructure evolution and mechanical property change of reduced-activation martensitic steel during varying-temperature irradiation

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Abstract

The effects of varying-temperature irradiation on mechanical properties and microstructure of reduced-activation 9Cr martensitic steels (RAMS) were investigated by means of micro-Vickers hardness tests and positron annihilation lifetime spectrometry. In case of stepwise increasing-temperature irradiation of 473/623 K, irradiation hardening accumulated at the lower temperature still existed after the elevation in temperature, while in case of the 493/693 K varying irradiation, the low temperature irradiation hardening disappeared after the elevation in irradiation temperature. In both the varying-temperature irradiations, it was observed that microvoids disappeared after the elevation in irradiation temperature. Result of a computer simulation of the evolution of defect clusters using rate theory were in good agreement with the experimental results when I-clusters were considered to be a factor controlling irradiation hardening. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Reduced-activation 9Cr–2W martensitic steels (RAMS) have been considered to be the prime candidates for structural material in the DEMO reactor and beyond in Japan [1]. These steels irradiated in fission reactors have been showing excellent resistance to void swelling and irradiation-induced embrittlement at around 693 K up to 70 dpa because of the good stability of the martensitic structure at the irradiation conditions [1–3]. Significant irradiation hardening, however, has been observed at lower irradiation temperatures even at low doses and with no observable changes in the microstructure [4–6].

The effect of varying-temperature irradiation on the hardening is considered to be complex, but very important to the practical use of these materials in fusion [7,8]. Recently, a varying-temperature irradiation study using temperature-controlled capsules in JMTR was performed, and it was shown that irradiation hardening of RAMS appeared not because of microvoids (V-clusters) but due to small interstitial loops (I-clusters) [9].

In this study, the effects of two sorts of step mode varying-temperature irradiations on the hardening of RAMS were investigated and a rate theory model was applied to simulate the experimental results.

2. Experimental

The chemical compositions and the heat treatment conditions of the Japanese low-activation martensitic steel (JLM-2) and modified JLF-1 used in this study are shown in Table 1. Varying-temperature irradiations were carried out in the JMTR with the multi-division and multi-section irradiation rig, which had a well-controlled temperature during irradiation [9]. Two sorts of step mode irradiations were performed, where the irradiation temperature was increased from 473 to 623 K (473/623 K) and from 493 to 693 K (493/693 K). In each varying-temperature irradiation, three capsules were irradiated; after the removal of one of three capsules from the reactor at the end of lower temperature irradiation, the irradiation temperature was elevated and the second

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Table 1
The chemical compositions and heat treatment conditions of RAMS used in this study; Chemical compositions (wt%)

	C	Si	Mn	P	S	Ni	Cr	V	Ti	W	Ta	B	Al
Mod. JLF-1/LSM[a]	0.12	0.01	0.01	0.003	0.0017	0.02	8.91	0.26	0.05	2.03	0.069	–	–
JLM-2[b]	0.12	0.050	0.53	0.002	0.0020	–	8.98	0.25	0.018	2.01	0.059	0.0029	0.028

[a] Normalized at 1323 K for 30 min and then tempered at 973 K for 1 h, followed by air cooling.

[b] Normalized at 1323 K for 30 min and then tempered at 1033 K for 1 h, followed by air cooling.

capsule was removed. Finally, the third capsule was removed at the end of the higher temperature irradiation. The irradiation histories are described in Fig. 1 in more detail.

Micro-Vickers hardness (Hv) was measured at room temperature with a load of 0.2 kg. Positron annihilation lifetime measurements (PA) were carried out at room temperature, and spectra were decomposed into two components, i.e., a matrix component (τ_1, I_1) and a microvoid component (τ_2, I_2), where τ_i and I_i are the lifetime and intensity of the ith component, respectively.

3. Results

Subsequent changes in Hv during varying-temperature irradiations are shown in Figs. 2(a) and (b) for 473/623 and 493/693 K irradiations [9], respectively. In both cases, the lower temperature irradiation-induced hardening. A significant difference was observed in the hardening immediately after the elevation of irradiation temperatures; a small increase in the hardening was observed in the 473/623 K irradiation, while a significant reduction of the hardening was observed in the 493/693 K irradiation [9]. At the end of the higher temperature irradiation, some residual hardening was found in the 473/623 K experiment, but none was found in the 493/693 K experiment.

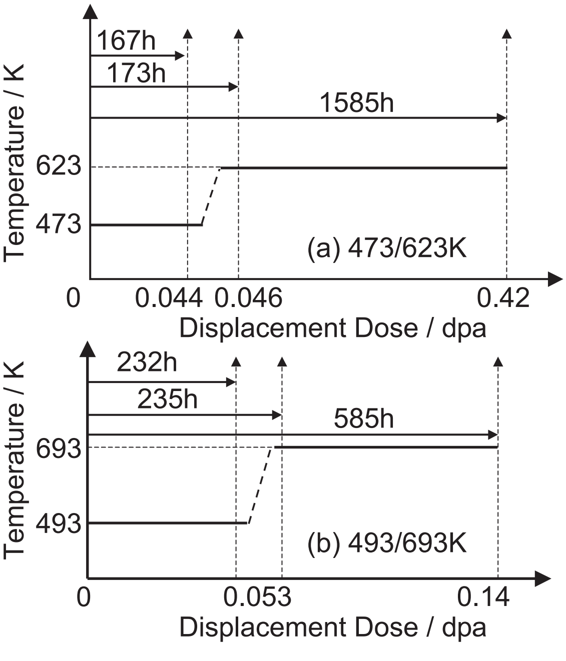

Fig. 1. Irradiation histories of stepwise varying-temperature irradiations.

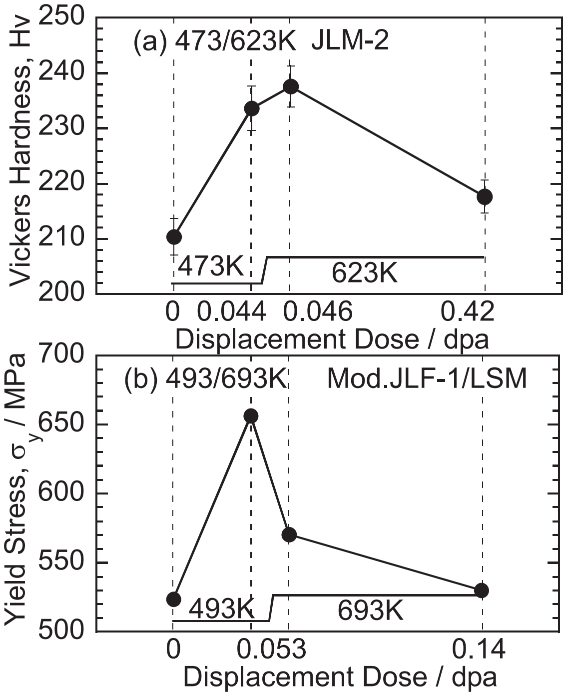

Fig. 2. Change in Hv under varying-temperature irradiation of: (a) 473/623 K and (b) 493/693 K.

As shown in Fig. 3(a), PA results indicated that microvoids formed at 473 K disappeared immediately after the elevation of temperature to 623 K; but they again appeared in the following irradiation at 623 K. This was also observed in the 493/693 K irradiation.

4. Discussion

4.1. Comparison with post-irradiation annealing experiment

Although the microvoid evolution under the 473/623 K irradiation was similar to that under the 493/693 K irradiation, irradiation hardening was completely

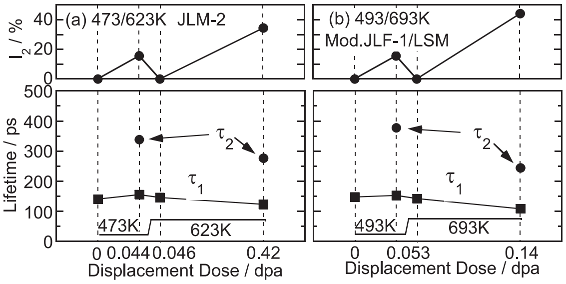

Fig. 3. PA parameters determined by two component analysis, following varying-temperature irradiation of: (a) 473/623 K and (b) 493/693 K.

different in the two irradiations. This is consistent with a previous study of recovery of irradiation hardening and positron annihilation lifetime during isochronal annealing of neutron-irradiated RAMS [10]; in this study, microvoids completely disappeared after increasing temperatures to 623 and 693 K, while irradiation hardening disappeared only at 673 K. In other words, the disappearance of microvoids at 623 K was not always accompanied by disappearance of the hardening [10]. This suggests that the factor controlling irradiation hardening is not microvoids but be other effects which are considered to be interstitial loops (I-loop) and possibly small precipitates [10].

4.2. Modeling I-cluster-induced hardening with a rate theory

The results obtained were evaluated in terms of defect cluster evolution using rate theory. This has not been well-established for RAMS because of the difficulty of direct observation of defect clusters by transmission electron microscopy. It was helpful for understanding the evolution of defect clusters under the varying-temperature irradiation even if it was a conventional model [11]. The parameters concerning defect properties and irradiation conditions are summarized in Table 2. The model used in this study consists of the evolution of interstitial clusters, which were formed as I-loops, and vacancy clusters, which were formed as microvoids, through absorption and emission of freely migrating interstitial atoms and vacancies. From the calculation results, the density and size of each defect cluster were evaluated and the increase in yield stress, $\Delta\sigma_y$, was calculated from a barrier hardening model,

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

where μ is the shear modulus (8.54×10^4 MPa), b the Burgers vector (0.248 nm), N the number density of defect clusters and d is the size of the defect clusters [12]. In order to evaluate the contribution of I-and V-clusters to irradiation hardening, the coefficient, α, giving the barrier strength of defect clusters against dislocation motion, was selected to be 0.23 for each size of I-loops and microvoids [13].

The results of calculations of defect cluster evolution during irradiation with a stepwise increasing-temperature from 493 to 693 K are shown in Figs. 4(a) and (c), which show densities of both clusters ($5 \leqslant n \leqslant 500$) and those of interstitial atoms and vacancies. In the same way, Fig. 4(b) shows the changes in those densities when the higher temperature was 623 K. During irradiation at

Table 2
Parameters of defect properties and irradiation conditions used in this calculation

Displacement rate (dpa/s)	6.6×10^{-8}
Cascade efficiency	0.1 [11]
Interstitial clustering parameters	
Total interstitial clustering fraction	0.3 [11]
di, tri, and tetra interstitial	
Clustering fractions	0.15, 0.1, 0.05 [11]
Cluster binding energies (eV)	0.5, 0.75, 1.25 [11][a]
Vacancy clustering parameters	
Total vacancy clustering fraction	0.6 [11]
di, tri, and tetra vacancy	
Clustering fractions	0.3, 0.2, 0.1[b]
Binding energy for V_n-cluster (eV)	$1.6 \times ((n-1)^{0.8} - n^{0.8} + 1)$[b]
Interstitial migration energy (eV)	0.35 [14]
Vacancy migration energy (eV)	1.3 [11]
Vacancy formation energy (eV)	1.6 [15]
Sink strength of dislocation (/atom)	10^{-6}[c]

[a] I-clusters larger than tetra clusters are assumed to be not decomposed.

[b] Vacancy clustering fractions and binding energies are assumed.

[c] Calculated from dislocation density of $1 \times 10^{15}/\mathrm{m}^2$.

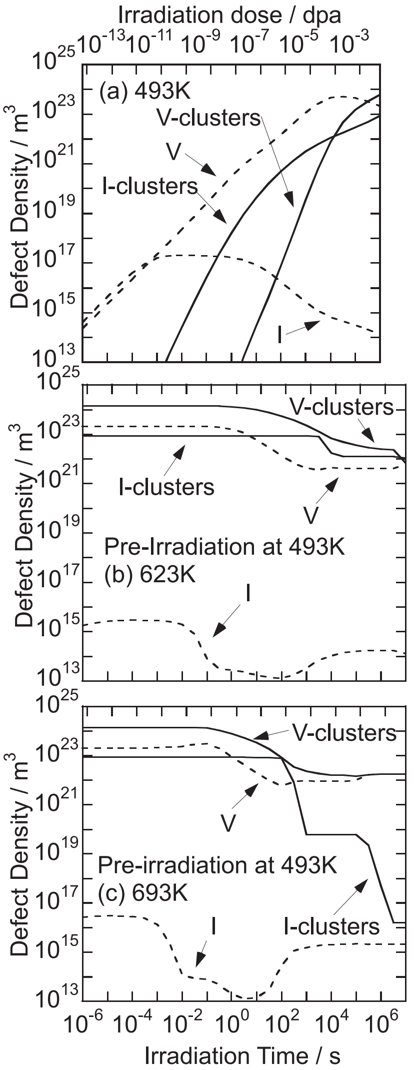

Fig. 4. Simulation of changes in the densities of V-, I-clusters ($5 < n < 500$) and V, I point defects during irradiation, (a) at 493 K, after elevation to (b) 623 K and (c) 693 K.

493 K, each type of cluster increased in density. After elevation of the temperature, the density of both types of cluster was reduced. The reduction of densities of I-loops is much larger than that of microvoids in the case of the 493/693 K irradiation, while such a significant difference was not observed in case of the 493/623 K irradiation. Also, the density of I-loops is much larger at 623 K than at 693 K, which is consistent with hardening being controlled by interstitial loops. The densities of microvoids at doses immediately after the temperature elevation are of the order of $10^{22}/\mathrm{m}^3$ in both the case of the 473/623 K and the 493/693 K irradiation. This is too low to be detected by PA and cannot explain the difference in hardening between 623 and 693 K. It is shown that the decrease in cluster densities under the 623 K irradiation occurs at a larger dose compared with that during the 693 K irradiation. The incubation time for the decomposition of microvoids at 623 K is longer than that at 693 K. The decomposition of microvoids increases the number of supersaturated vacancies flowing to I-loops, resulting in a decrease in the density of I-loops.

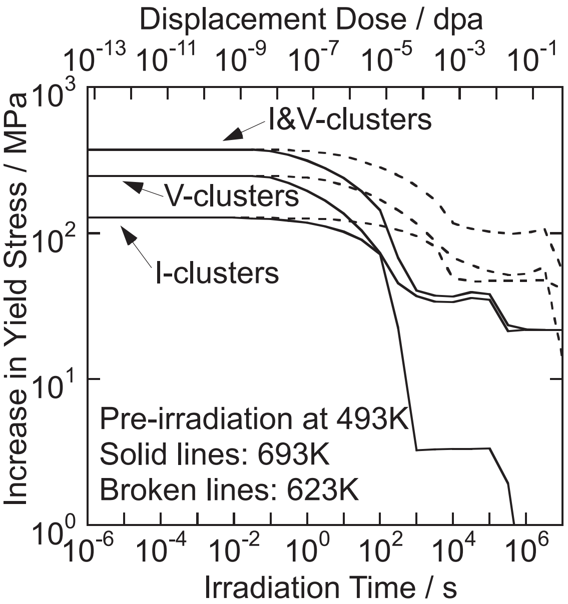

Fig. 5. Simulation of changes in the hardening after the elevation of irradiation temperatures from 473 to 693 and 623 K.

A simulation study was conducted to estimate yield stress changes during varying-temperature irradiation, and the results are shown in Fig. 5. This indicates that hardening by microvoids is about 20 and 60 MPa at 693 and 623 K, respectively, while hardening by I-loops is almost null and 60 MPa at 693 and 623 K, respectively. Hence, the I-loop hardening model is consistent with the experimental result showing irradiation hardening only in the case of an elevation of irradiation-temperature to 623 K.

In this calculation, the appearance of microvoids at the end of the higher temperature irradiation was not successfully simulated, although it was actually observed in PA measurements. It is expected that the sink strength for vacancies and/or thermal stability of microvoids will change with increasing irradiation dose, which could be important factors to determine swelling behavior under higher irradiation doses around 673 K [3]. Further studies are needed.

5. Conclusions

Effects of two types of step mode increasing-temperature irradiations, 473/623 and 493/693 K, on irradiation hardening and microstructure evolution were investigated through experiments and calculations. The main conclusions are as follows:

1. In the case of the 473/623 K irradiation, irradiation hardening at 473 K still remained after the elevation of temperature; while in the case of the 493/693 K irradiation, the hardening at 493 K completely disappeared after the elevation of temperature.
2. In both the varying-temperature irradiations, microvoids were extinguished by the elevation of temperature.
3. Hardening computed from a rate theory based calculation of microvoid and I-loop evolution suggests that I-loops were the factor controlling irradiation hardening of RAMS.

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