EFFECT OF PERIODIC TEMPERATURE VARIATIONS ON THE MICROSTRUCTURE OF NEUTRON-IRRADIATED METALS- S.J. Zinkle, N. Hashimoto, D.T. Hoelzer, A.L. Qualls (Oak Ridge National Laboratory) and T. Muroga (National Institute for Fusion Science)

OBJECTIVE

The objective of this report is to investigate the effect of periodic variations in irradiation temperature on the microstructural evolution of several structural materials.

SUMMARY

Specimens of pure copper, a high-purity austenitic stainless steel, and V-4Cr-4Ti were exposed to eight cycles of either constant temperature or periodic temperature variations during neutron irradiation in the High Flux Isotopes Reactor to a cumulative damage level of 4 to 5 displacements per atom. Specimens were exposed to a low temperature during the initial 10% of accrued dose in each of the eight cycles, and were exposed to a higher temperature during the remaining 90% of accrued dose in each cycle. Different specimens were exposed to low/high irradiation temperatures of 225/340˚C and 350/520˚C. The microstructure was compared with that of companion specimens that were continuously maintained at 340 ˚C and 520˚C, respectively during the entire irradiation. The low-temperature excursions produced enhanced nucleation and growth of radiation-induced defects (precipitates, dislocation loops) in V-4Cr-4Ti and stainless steel.

PROGRESS AND STATUS

INTRODUCTION

It is well known that the irradiation temperature can have a profound impact on the microstructure that develops in materials [1]. In qualitative terms, nucleation of defect clusters is maximized at lower temperatures whereas growth and coarsening of clusters are maximized at higher temperatures. Modest temperature excursions are expected to be a common occurrence in any commercial nuclear system due to scheduled startup and shutdown events. These varying-temperature excursions allow the possibility of enhanced nucleation and growth of radiation-induced defect clusters compared to a constant-temperature irradiation. Several previous ion irradiation and low-dose neutron irradiation studies [2-10] have found that these temperature excursions may exert a significant influence on the microstructural evolution, particularly if the temperature excursion transcends the recovery Stage V temperature regime (which corresponds to thermal dissociation of small vacancy clusters). The recovery Stage V occurs at ~325˚C in austenitic stainless steel [11-13] and ~375˚C in V-4Cr-4Ti [14]. Void formation was generally enhanced and loop formation suppressed for the cyclic temperature irradiation [4] or preirradiation at low temperatures [2].

Several studies have investigated the effect of constant versus varying temperature on the microstructure of irradiated austenitic stainless steel [4-6,8,15], vanadium [7,10], and vanadium alloys [7]. In some cases, rather spectacular differences were observed. For example, Yoshida and coworkers [5] observed that varying temperature (either 200/400˚C or 300/500˚C) neutron irradiation of Fe-16Cr-17Ni-0.25Ti austenitic stainless steel produced dramatically higher void swelling levels compared to constant temperature irradiation at 400˚C. Similarly, enhanced void nucleation has been observed in numerous binary vanadium alloys which were preirradiated at low temperature, compared to constant temperature irradiation [7]. Low temperature preirradiation has been observed to cause an enhancement in the defect cluster density in many cases [5,6], but in some cases either no effect [8] or a decrease in loop density [10] has been observed compared to the constant temperature irradiation condition.

In the present paper, results will be presented for V-4Cr-4Ti and stainless steel irradiated at 520˚C (constant temperature) vs. 350/520˚C (varying temperature), and for pure copper irradiated at a constant temperature of 340˚C. This irradiation experiment was performed as part of the JUPITER Japan-USA collaboration on fusion materials.

EXPERIMENTAL PROCEDURE

The materials used for this study were “P7” austenitic stainless steel (Fe-17Cr-16.7Ni-2.5Mo), V-4Cr-4Ti (Teledyne Wah Chang heat 832665), and Johnson-Matthey “Puratronic” 99.999% copper. All of the specimens were irradiated in the annealed condition as 3 mm diameter by 0.5 mm thick transmission electron microscope disks. The annealing conditions were 1050˚C for 0.5 h (stainless steel), 1000˚C for 2 h (V-4Cr-4Ti), and 550˚C for 2 h (Cu). The specimens were irradiated for a total of 8 irradiation cycles in the High Flux Isotopes Reactor at ORNL, which resulted in a fast neutron fluence (E>0.1 MeV) of ~$8x10^{21}$ n/m^2. This corresponds to a damage level of ~4 dpa in the stainless steel and vanadium specimens, and ~5 dpa in copper. Electrical heaters and a mixture of helium and argon gases were used to control the irradiation temperatures. The temperatures were continuously monitored and controlled during the irradiation. The capsule was divided into four independently-controlled zones in order to achieve four different irradiation temperature profiles [16]. The temperature profiles for each of the 8 HFIR irradiation cycles were as follows: Zone A, constant temperature of 340˚C; Zone B, constant temperature of 520˚C; Zone C, first 10% of dose at 350˚C and remaining 90% at 520˚C; Zone D, first 10% of dose at 225˚C and remaining 90% at 340˚C. Further experimental details are described elsewhere [16]. Following irradiation, the TEM specimens were jet electropolished and examined in a JEOL 2000FX or Philips CM30 electron microscope.

RESULTS

The dominant microstructural feature in V-4Cr-4Ti irradiated at both a constant temperature of 520˚C and the variable (350/520˚C) condition was a high density of disk-shaped precipitates on {001} matrix habit planes. Figure 1 shows an example of the precipitates observed in V-4Cr-4Ti irradiated at a constant temperature of 520˚C. Void or dislocation loop formation was not observed in either specimen. A very low density of network dislocations was observed in both specimens.

The microstructure of V-4Cr-4Ti irradiated at constant (520˚C) vs. varying (350/520˚C) temperature was qualitatively similar. However, as shown in Fig. 2, the varying temperature irradiation produced precipitates of a finer size and higher density compared to the constant temperature irradiation condition. According to preliminary measurements, the constant temperature irradiation produced precipitates with a mean diameter of 24 nm and a density of ~$4x10^{21}$/m^3. The varying temperature irradiation produced precipitates with a mean diameter of 11 nm and a density of ~$1.2x10^{22}$/m^3. Additional quantitative measurements of the precipitate size and density are in progress.

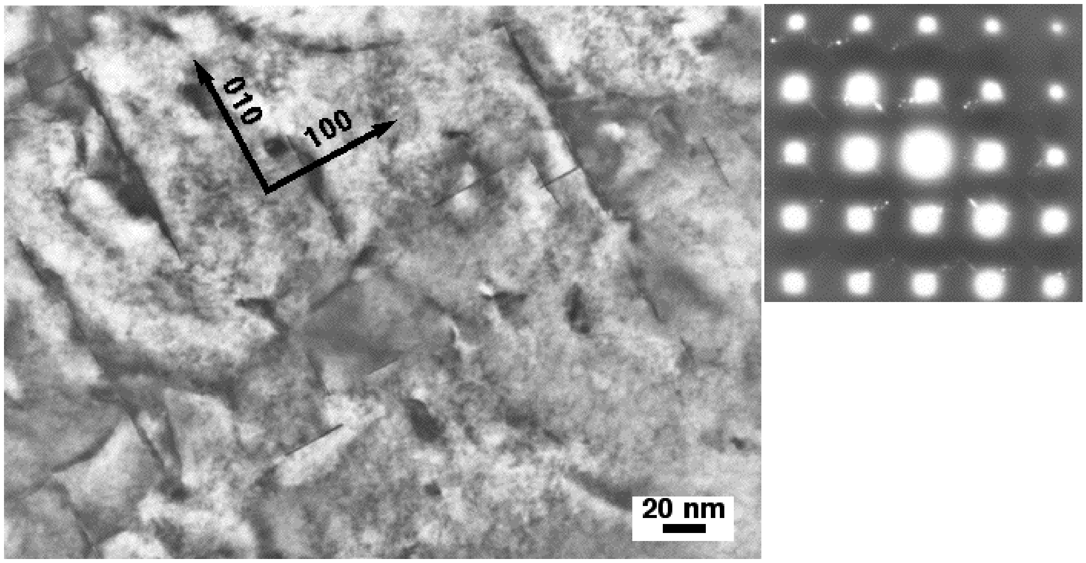

Fig. 1. Precipitates observed on {001} matrix habit planes in V-4Cr-4Ti irradiated to 4 dpa at 520˚C. Streaks associated with the precipitates are visible in the diffraction pattern.

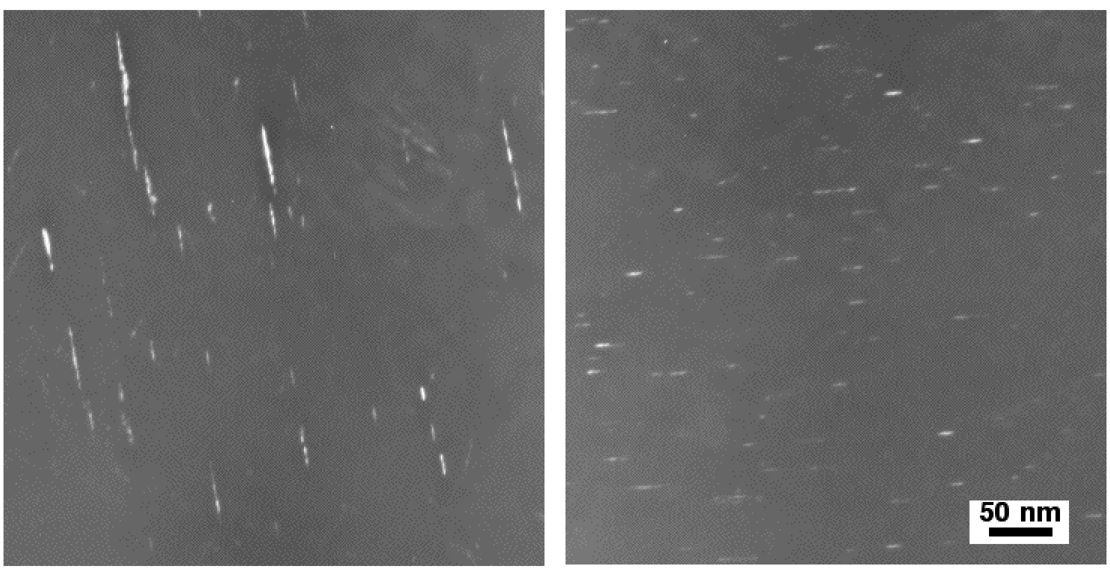

Fig. 2. Comparison of the precipitates observed in V-4Cr-4Ti irradiated to 4 dpa at 520˚C (left fig., constant temperature) vs. 350/520˚C (right fig., varying temperature). Centered dark field image.

A moderate density of small defect clusters (dislocation loops, stacking fault tetrahedra) and voids were produced in stainless steel during irradiation at either constant (520˚C) or varying (350/520˚C) irradiation conditions. The general microstructural features were qualitatively similar for the two irradiation conditions, although there were some quantitative differences. Figure 3 compares the general microstructure of stainless steel for the two irradiation conditions. The varying temperature irradiation produced enhanced growth of defect clusters compared to the constant irradiation condition. Although the defect cluster density was $\sim 3x10^{21}/m^3$ for both irradiation conditions, the mean defect cluster diameter was significantly larger for the varying temperature condition (6 nm) compared to the constant temperature condition (2.5 nm).

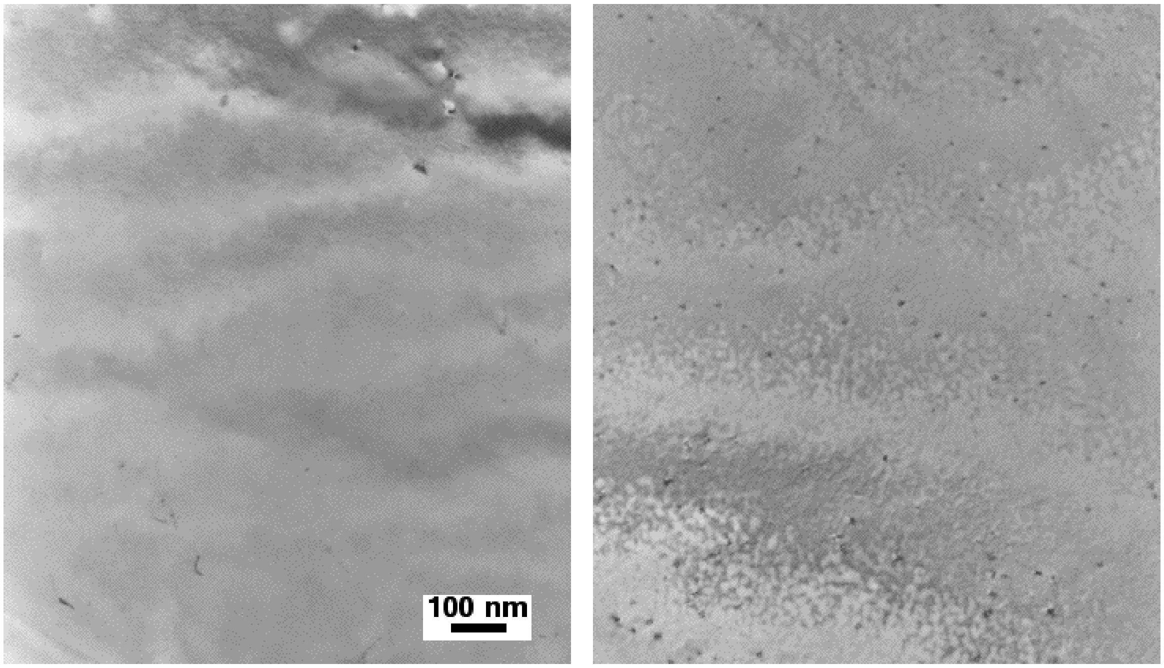

Fig. 3. Comparison of the defect clusters observed in stainless steel irradiated to 4 dpa at 520˚C (left fig., constant temperature) vs. 350/520˚C (right fig., varying temperature).

As shown in Fig. 4, a low density of small cavities was observed in stainless steel for both irradiation conditions. Although the number density appeared to be slightly higher for the varying temperature condition, the amount of void swelling was very small (<0.01%) for both conditions. Higher dose irradiations would be needed to examine the possibility of significant differences in the void swelling behavior for constant vs. varying temperature irradiation conditions.

Irradiation of pure copper at a constant temperature of 340˚C produced a large amount of cavity swelling. The mean void diameter and density were 80 nm and $8x10^{19}/m^3$, which yields a volume swelling of ~2.2%. The void density is about a factor of two larger than previously reported for copper irradiated to ~1 dpa at a constant temperature of 350˚C [17,18]. There was no evidence for crystallographic ordering of the cavities in the present study. A very low density of small stacking fault tetrahedra was also observed. Figure 5 shows an example of cavity formation adjacent to a grain boundary. The width of the zone denuded of cavities adjacent to the grain boundaries was ~0.45 µm. The slightly higher cavity density and smaller grain boundary denuded zone width in the present study compared to previous constant temperature irradiation studies at 350 ˚C [17,18] may be in part attributable to the lower irradiation temperature and higher damage rate in this study.

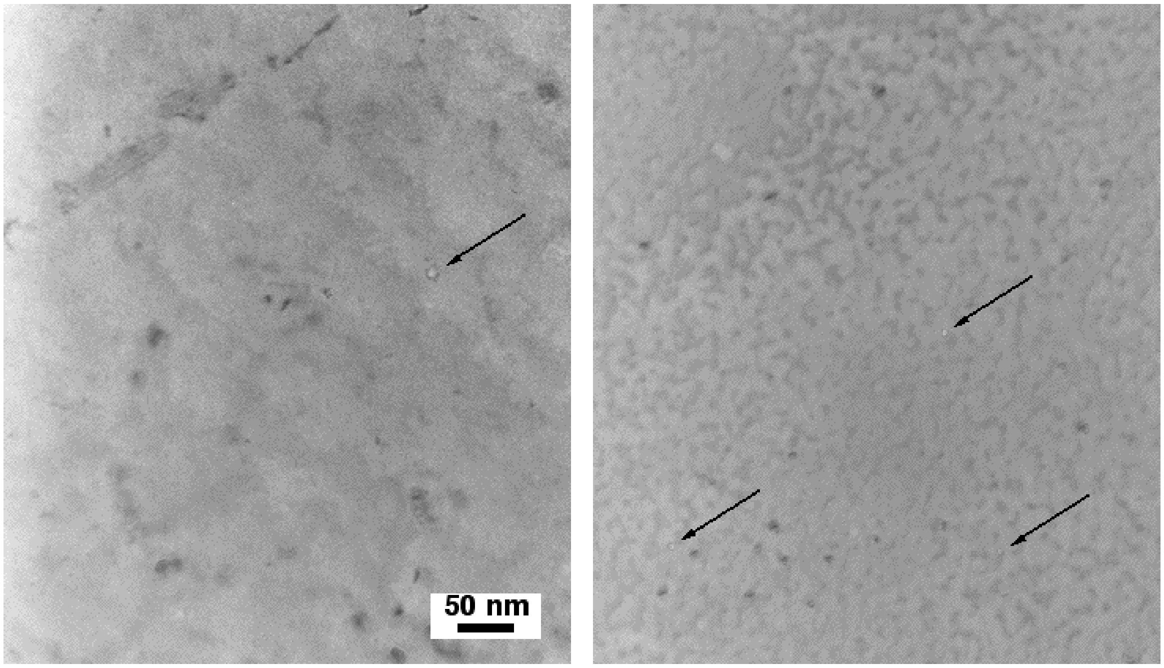

Fig. 4. Examples of the small cavities observed in stainless steel irradiated to 4 dpa at 520˚C (left fig., constant temperature) vs. 350/520˚C (right fig., varying temperature).

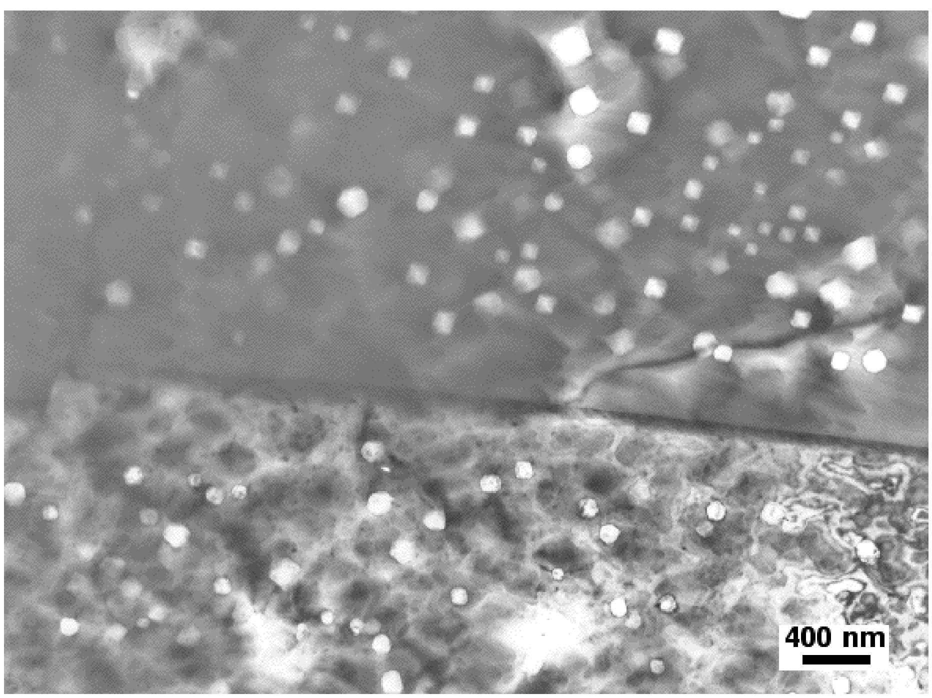

Fig. 5. Cavity swelling adjacent to a grain boundary in copper irradiated to 5 dpa at 340˚C

DISCUSSION

Qualitatively similar microstructures were observed in V-4Cr-4Ti and stainless steel for the constant and varying temperature irradiation conditions investigated in this study. In quantitative terms, the varying temperature irradiation generated a higher density of smaller precipitates in V-4Cr-4Ti and a larger average defect cluster size in stainless steel. The precipitate density observed for V-4Cr-4Ti (4×10^{21} and $12 \times 10^{21}/m^3$) was slightly higher than that observed in the same alloy heat following a constant temperature neutron irradiation to 0.1 dpa at 505˚C ($2.6 \times 10^{21}/m^3$) [14].

CONCLUSIONS

Low temperature excursions can produce enhanced nucleation and growth of radiation-induced defects (precipitates, dislocation loops) in V-4Cr-4Ti and stainless steel. The quantitative effect of low-temperature excursions was relatively small for the materials and conditions investigated in the present study (8 cycles of varying 350/520˚C temperature vs. constant 520˚C temperature, with 0.05 dpa at the lower temperature and 0.45 dpa at the higher temperature for each cycle). Further analysis is needed to investigate the quantitative influence of varying temperatures under other irradiation conditions (different temperatures, different doses at low temperature, etc.).

Acknowledgements

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