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ABSTRACT

High-energy electrons were used to study tensile property changes in simple Fe-Cu and Fe-Cu-Mn alloys irradiated at 288°C. A comparison was made with neutron irradiation data on the same alloys. An apparent effect of alloy chemistry was observed in which the presence of Mn affected embrittlement differently for electron and neutron irradiation. Comparison of previous experimental studies with the present experimental results indicates that electrons may be more efficient than fast neutrons at producing embrittlement.

INTRODUCTION

Copper-rich precipitates (CRPs) have been identified as key embrittling microstructural features which form during fast neutron irradiation of commercial US light-water nuclear reactor (LWR) pressure vessel (PV) steels. Understanding the kinetics of this CRP phase formation is therefore important for understanding radiation-induced embrittlement rates. The evolution of these precipitates is usually viewed in the context of radiation enhanced diffusion, in which freely-migrating defects (FMDs) produced during irradiation accelerate the transport of Cu, in turn accelerating the precipitate nucleation and growth process over that normally occurring at the temperature of interest (i.e., 288°C in commercial LWR PVs) [1].

In neutron irradiation studies, the relationship between precipitation kinetics and embrittlement rates is complicated by the cascade nature of defect production. Fast neutrons typically initiate high energy recoils which produce defects in dense cascades in which substantial recombination and defect (vacancy and/or interstitial) clustering can occur. Recent work has also suggested that diffuse Cu clusters may also be nucleated directly within cascades [2]. In addition to their direct hardening contribution, these cascade clusters (CC), whether defect or solute in nature, may mediate a number of kinetic processes impacting copper precipitation [3].

Our approach to study the kinetics of CRP phase formation, and probe the role that CC have on its kinetics and subsequent embrittlement, involves comparative experiments using electron irradiation and fast neutron irradiation. Unlike fast neutrons, high-energy electrons produce displacement damage at relatively low primary recoil energies. The median recoil damage energies, $T_{1/2}$, (i.e. that recoil energy above and below which one-half the displacement damage is produced) are 300 eV and 40 keV for electron and reactor neutron irradiations in Fe, respectively. The lower energy recoils generated during electron irradiation produce fewer, more isolated defects, and thus minimize clustering during the defect production process relative to that occurring in energetic neutron-induced cascades. Comparison of embrittlement resulting from electron irradiation, in which CC production is negligible, versus neutron irradiation can thus provide some insight into the role that the CC have on CRP formation kinetics and embrittlement.

Electron irradiations have been previously used to study microstructural and property changes (e.g. surface hardness) in various model PV alloys [4-11]. In the present study, tensile property changes in model PV alloys induced by 10-MeV electron irradiation at a nominal temperature of 288°C are examined and compared with neutron irradiation data from the same materials. Unlike previous work, the use of high-energy electrons in this study is advantageous because a greater volume of material may be irradiated, allowing the study of bulk mechanical property effects such as yield stress.

EXPERIMENTAL

Two model PV alloys were used: material VH was a binary Fe-0.9 wt. % Cu alloy and material VD was a ternary Fe-0.9 wt. % Cu-1.0 wt. % Mn alloy. Sheet mini-tensile specimens with a rectangular gage section 9 mm x 2 mm were precision die punched for use in the tensile tests.

Electron irradiations were performed with 10-MeV electrons from a pulsed linac operating at 15 Hz. The beam was incident on samples in a vacuum chamber after passing through a thin aluminum foil window and a 12 mm diameter collimator. The sample chamber was backfilled with He gas held at a slight vacuum between -130 mm Hg and -510 mm Hg relative to atmospheric pressure. Sample temperature was controlled by varying the electron beam current incident on the samples, which varied between 14 μA and 23 μA. Assuming a damage cross section of 86 barns for 10-MeV electrons in a medium Z material [12], these beam currents corresponded to respective damage rates of 6.6×10^{-9} dpa s^{-1} and 1.1×10^{-8} dpa s^{-1} (dpa=displacements per atom).

Temperature was monitored using a thermocouple spot-welded to the middle of the gage section of a sample located near the center of the irradiated area defined by the collimator. In addition, thermocouples were used to monitor the temperature on the same sample's shoulders away from the gage section, which lay outside of the electron beam. Heat sinking at the clamped sample end resulted in a slightly asymmetric temperature distribution along the gage section. With the middle of the gage section held at 288°C, it was estimated, using the shoulder temperature measurements, that the temperatures at the extremities of the 9.0 mm long gage section were 281°C and 250°C, with the lower temperature at the clamped end of the sample.

Neutron irradiation data on the VH and VD alloys for a single fluence were obtained from experiments performed in the University of Michigan Ford Nuclear Reactor (UM FNR) [13]. Samples were sealed in He-backfilled aluminum capsules and irradiated at 288°C. The neutron flux (E>1 MeV) was 7×10^{11} cm^{-2} s^{-1} and the neutron fluence achieved was 1×10^{19} cm^{-2}. Assuming a displacement cross section of 1500 barns, these values translate to a damage rate of 1.1×10^{-9} dpa s^{-1} and a damage level of 15 mdpa.

Static tensile tests were carried out at room temperature at a strain rate of 10^{-3} s^{-1} on an automated tensile tester. A minimum redundancy of 2 tests was used and the average precision of the yield stress measurements was better than ±15 MPa.

RESULTS AND DISCUSSION

Fig. 1 summarizes the changes in (a) yield stress (ΔYS) and (b) ultimate tensile stress (ΔUTS) induced by both electron and fast neutron irradiation in the two alloys. Tensile properties of the corresponding unirradiated control samples are listed in Table I.

The embrittlement behavior observed in Fig. 1, can be attributed nearly exclusively to CRP phase formation, as supported by previous irradiation studies. In the case of electrons, for example, hardness experiments in an Fe-1.5% Cu alloy showed irradiation at 300°C produced nearly the same increase in Vickers hardness as that produced by thermal annealing at 500°C [8]. Furthermore, the annealing formed essentially the same microstructure (i.e. the same CRP sizes and number densities) as electron irradiation, as evidenced by small angle neutron scattering (SANS). The slightly higher (~15%) hardness observed in the electron-irradiated alloy was attributed to an additional smaller component of embrittlement produced by Cu-vacancy defect complexes formed during irradiation, but absent during thermal annealing. Additional evidence for the direct formation of CRP during electron irradiation was obtained from high-resolution electron microscopy of an Fe-1.5% Cu model alloy [14]. The structure of CRP was observed to evolve in the same manner during irradiation as during thermal annealing. Taken as a whole, the results from these previous experiments suggest that any embrittlement rate differences observed between electron and fast neutron irradiation can be understood simply as difference in rates of radiation-enhanced Cu diffusion.

From Fig. 1 we see that addition of Mn decreases the hardening for a given damage level during electron irradiation relative to that in the binary alloy. The opposite effect appears to occur during neutron irradiation, where the ternary alloy hardens more than the Fe-Cu alloy. However, rather than an effect of the type of irradiation (electron vs. neutron), previous work suggests this observed embrittlement behavior is related to a chemistry effect that is a function of the damage level 15]. A similar effect is sometimes observed with Ni additions, i.e. inhibition of embrittlement at low damage levels and an increase at higher damage levels [16]. SANS and hardening neutron irradiation data have shown that Mn has a strong effect on the formation of CRPs and a systematic but weaker hardening effect on matrix defects (e.g. CC produced by fast neutron irradiation). Thus, under electron irradiation, the same, thermodynamically-driven, Mn effect is expected. The mechanistic effect of Mn may be similar to that of Ni which co-partions to the CRPs with Mn. However, the overall effect of these elements is complex since they affect the size, hence hardening efficiency, of the precipitates as well as their growth kinetics. Additional irradiation experiments in conjunction with microstructural characterization are required to clarify this Mn effect.

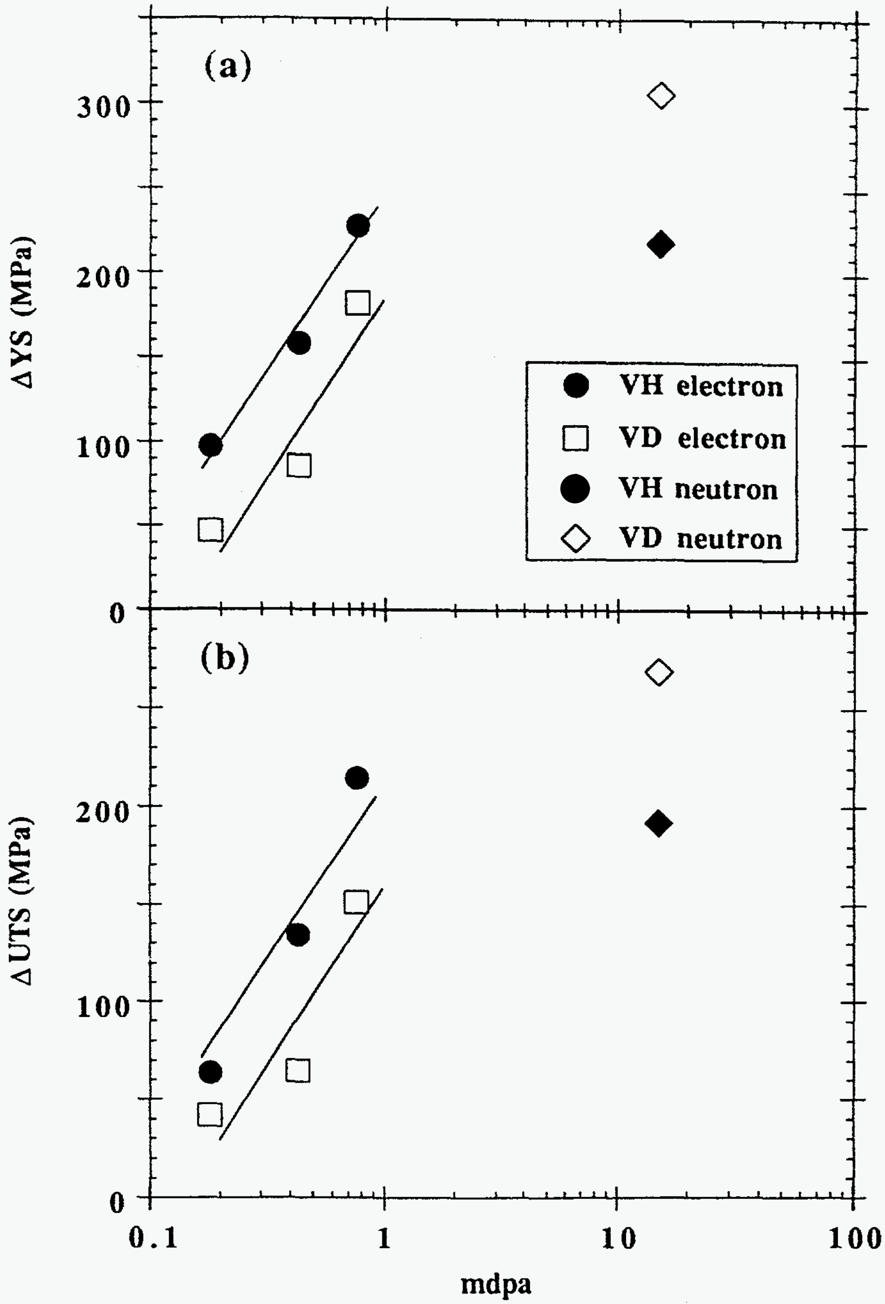

Fig. 1. Changes in tensile properties (a) yield stress and (b) ultimate tensile stress induced by irradiation with electrons and neutrons in model PV alloys at 288°C. VH=Fe-0.9Cu, VD=Fe-0.9Cu-1.0 Mn. Lines provided only as visual aids.

Table I. Tensile properties of unirradiated model alloys

Sample Designation	Composition (weight %)	Number Tested	YS ± std.deviation (MPa)	UTS ± std. deviation (MPa)
VD	Fe-0.9% Cu-1.0% Mn	4	216 ± 6	310 ± 7
VH	Fe-0.9% Cu	4	175 ± 12	252 ± 14

Examining a single alloy, definitive comparisons of the embrittling effects of electrons and neutrons based on the results in Fig. 1 are restricted by the absence of any overlapping doses for electrons and neutrons. In the VH alloy, the equivalent changes in YS and UTS found at about a factor of ten lower dpa during electron irradiation may indicate accelerated embrittlement during electron irradiation. However, these data may also reflect a saturation in embrittlement behavior which is achieved near the highest electron dpa level, and which extends to the VH neutron damage level.

Experimental support for a slight acceleration due to electron irradiation is further supported by comparison with recent neutron irradiation work by Odette et al. [17]. SANS studies of Cu precipitation were performed in neutron irradiated model alloys including one (Fe-0.7 wt.% Cu) similar to the VH alloy used in the present work. The results showed that nearly 80% of the available Cu in solution had precipitated by 3 mdpa, reaching 90% by 40 mdpa. This suggests that saturation in the hardness contribution due to CRPs is not reached in simple alloys until neutron damage levels slightly higher than those probed in the present electron irradiation studies. While matrix damage also contributes to embrittlement during neutron irradiation, in this previous work it was estimated to account for less than 10% of the observed hardening at the lowest dpa level.

CONCLUSION

A comparative study of embrittlement induced by high-energy electron and fast neutron irradiation was undertaken to investigate the role of cascade effects on the formation of Cu-rich precipitates in model PV alloys. An apparent chemistry effect was observed in which additions of Mn alternately inhibited and enhanced embrittlement at low and high damage levels, respectively, relative to that observed in a binary Fe-0.8 wt.% Cu alloy. Comparisons with previous work suggest a slight acceleration in embrittlement during electron irradiation in the binary alloy, which may be attributable to greater radiation-enhanced Cu diffusion relative to that induced during neutron irradiation. This results in faster Cu precipitation and, hence, an acceleration in the formation of the embrittling microstructure.

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