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EXPERIMENTS TO EXAMINE THE CONTRIBUTION
OF GAS ATOMS TO VOID FORMATION IN
IRRADIATED METALS

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Experiments to Examine the Contribution of Gas Atoms to Void Formation in Irradiated Metals

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Abstract

Vacuum-melting has been employed to demonstrate that residual gases, especially hydrogen, strongly influence void nucleation of copper, copper binary alloys and various Fe-Cr-Ni base alloys during either neutron or electron irradiation. Void nucleation in nickel appears not to be strongly affected by residual gases, however. Solute-free and solute-bearing Fe-Cr-Ni alloys appear to respond differently to differences in gas content. When contamination of specimens with sodium occurs during neutron irradiation, void nucleation in both as-fabricated and vacuum-melted specimens is similar, suggesting that gas atoms re-enter the specimens during irradiation.

Key Words: Copper, copper binary alloys, nickel, FeCrNi alloys, neutron irradiation, electron irradiation, gas influence

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1. Introduction

It is recognized that gas atoms play an important role in void nucleation and growth processes during radiation damage of fusion materials. It was reported previously that the density of voids formed during neutron irradiation decreased strongly when residual gas atoms were removed from either pure Cu and Cu-5Al.[1] Computer simulations using molecular dynamics show that voids can only nucleate in pure copper upon introduction of gas atoms into small vacancy clusters. [2] The objective of the present work is to examine the effects of gas atoms on the formation of voids in 1) pure copper and various copper binary alloys after high neutron exposure in the Fast Flux Test Facility (FFTF) and 2) in other alloys or metals after irradiation with either high voltage electrons or fission neutrons to low exposure in the Japan Materials Test Reactor (JMTR).

2. Experimental procedures

Pure copper and nickel, each of 99.999% nominal purity, and Cu-Al and Cu-Ni binary alloys with 5at.% solute were prepared. Pure copper and nickel were used in the as-received condition. The binary alloys were prepared by arc-melting the constituents in an argon atmosphere. In addition to these simple metals, the ternary alloys Fe-15Cr-20Ni and Fe-16Cr-17Ni, the Japan Primary Candidate Alloy (JPCA) and SUS316L were also used. Specimens of each metal or alloy were prepared in two ways, one in the as-received or as-prepared condition, and the second after vacuum-melting at $\sim 10^{-5}$ Pa. The gas evaporated from the specimens during vacuum-melting was analyzed using a quadrupole mass spectrometer.

In table I, the major features of the pressure history and the main components of the

outgas are shown. All specimens included hydrogen atoms or molecules as the major component. All oxygen in the outgas appears to have combined with carbon. After fabricating or vacuum-melting, the specimens were then cold-rolled and punched to produce standard microscopy disks. All specimens were then annealed under high vacuum.

Neutron irradiation was conducted in either FFTF at higher neutron flux or JMTR at low flux. In FFTF, the specimens were irradiated at temperatures of 365° to 460°C, reaching a maximum fluence of $8.4 \times 10^{26} \text{ n/m}^2$ ($E > 0.1 \text{ MeV}$). In JMTR, the specimens were irradiated at 400 and 500°C using a method involving improved temperature control [3] to fluences of 3.8×10^{23} and $8.9 \times 10^{23} \text{ n/m}^2$ ($E > 0.1 \text{ MeV}$), respectively. After irradiation, specimens were electropolished and observed at room temperature using an electron microscope operating at 200 kV. Electron irradiation was carried out on selected specimens at Nagoya University in a 1 MeV HVEM at $\sim 10^{-5} \text{ Pa}$ at 350 and 400°C. The peak electron flux was $\sim 4 \times 10^{24} \text{ m}^{-2}$.

3. Results

3.1. Cu and Cu alloys in FFTF

Specimens were observed from four different FFTF capsules. Several packets suffered leakage of sodium and the specimens were seriously contaminated. While a remarkable difference in void density was observed between the as-received and vacuum-melted specimens of pure copper in the non-leaking packets, no significant difference was observed in specimens from leaking packages. Examples of microstructures from sodium contaminated specimens are shown in fig. 1. A clear difference of void formation between as-fabricated and vacuum-melted Cu-5Al in non-leaking packets is shown in fig. 2. Many

large voids were observed in the as-fabricated specimens, while no voids were observed in vacuum-melted specimens, even when thicker areas were observed using a 400 kV electron microscope. In Cu-5Ni, no difference in void formation occurred in the two specimen types when irradiated to $7.0 \times 10^{26} \text{ n/m}^2$ ($E > 0.1 \text{ MeV}$) at 390°C concurrent with sodium leakage, as shown in fig. 3. Large voids and many tangled dislocations were observed in both specimen conditions. The only significant difference was that dislocation loops were observed in vacuum-melted Cu-5Ni, while no loops were found in the as-fabricated condition.

In the FFTF irradiation to $8.4 \times 10^{26} \text{ n/m}^2$ at 420°C , the surfaces of as-fabricated Cu-5Al specimens were covered with a white aluminum-rich material. Large voids were found in the center of the specimen that are similar to those in the left side of fig. 2. The composition in the center of the specimen was analyzed using EDX; the aluminum level had dropped to 1.2at%, suggesting that the decrease of Al via segregation to the specimen surface led to void nucleation in the center of the specimen.

3.2 Irradiation in JMTR.

Damage structures of as-fabricated and vacuum-melted pure Ni, Fe-16Cr-17Ni and SUS316L irradiated at 400°C in JMTR are shown in fig. 4. In pure Ni, the void density and size distribution were similar for both specimen conditions; almost all voids were isolated from dislocations. The void density was $\sim 1 \times 10^{20} \text{ m}^{-3}$ for both specimen conditions. However, void structures in the other two alloys were quite different. In Fe-16Cr-17Ni, there were tiny voids with sizes of $\sim 5 \text{ nm}$ in the as-fabricated specimen, while there were no voids in the vacuum-melted condition. In the as-fabricated SUS316L, large voids were

locally formed, but small needle-like precipitates were formed instead of voids in the vacuum-melted specimen.

Damage structures of these materials after irradiation at 500° C in JMTR are shown in fig. 5. In pure Ni, the void density and size distribution were similar for both types of specimens, although the mean void size was larger and the density smaller than observed after irradiation at 400° C. In as-fabricated Fe-16Cr-17Ni and SUS316L, there were tiny voids with sizes of ~10 nm in the as-fabricated specimen, while there were no voids in the vacuum-melted specimen.

3.3 Fe-Cr-Ni alloys irradiated in the HVEM

Fig. 6 shows damage structures after HVEM irradiation for Fe-15Cr-20Ni, SUS316L and JPCA. The irradiation temperature was 350° C for Fe-15Cr-20Ni, and 400° C for both SUS316L and JPCA. In Fe-15Cr-20Ni, there was a remarkable difference between the microstructures of as-fabricated specimens and vacuum-melted specimens, with the density of voids much larger in the vacuum-melted specimens. In SUS316L and JPCA, the opposite behavior was observed. Voids were observed in as-fabricated specimens, while no voids were observed in the vacuum-melted ones. Small dislocation loops were observed in these specimens, however.

4. Discussion

In the FFTF irradiation, the role of gas in nucleating voids is clear when the specimens are free of sodium contamination. When the contents of the packet were exposed to sodium, however, the microstructures of the two types of specimens were similar.

It appears that sodium exposure reintroduces gas atoms into the specimen. It is not certain that the reintroduced gases are the same as the original gases, however.

In the JMTR irradiation, the difference between the behavior of as-fabricated and vacuum-melted specimens was large at both temperatures, but this was not the case for pure nickel. Since most of the residual gas atoms were found to be hydrogen for both alloys, it is suggested that hydrogen atoms are responsible for void nucleation in Fe-Cr-Ni base alloys. In pure Ni, the different behavior cannot be explained in terms of the amount of hydrogen and helium created by transmutation, calculated for the JMTR irradiation to be about 1 appm and 0.007 appm, respectively. [4]

During HVEM irradiation, void swelling was suppressed by vacuum-melting in SUS316L and JPCA, but not in vacuum-melted Fe-15Cr-20Ni where the void density actually increased. This suggests that gas interactions with solutes such as silicon and carbon may also play some role. Easier movement of dislocations in solute-free alloys may also exert some influence, possibly by sweeping up gas atoms.

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Pressure (Pa)				
Specimen	before melting	max. at melting	ultimate after melting	main component of gaseous atoms
Cu	1.2×10^{-5}	2.0×10^{-4}	1.5×10^{-5}	H ₂ , C, CO
Cu-5Al	3.5×10^{-5}	2.0×10^{-4}	4.3×10^{-5}	H ₂ , C, CO
Cu-5Ni	3.0×10^{-5}	4.0×10^{-4}	5.5×10^{-5}	H ₂ , C, CO
Ni	0.8×10^{-6}	8.6×10^{-5}	0.8×10^{-6}	H ₂
Fe-16Cr-17Ni	1.7×10^{-6}	7.0×10^{-5}	1.5×10^{-5}	H ₂
Fe-15Cr-20Ni	1.6×10^{-6}	6.0×10^{-3}	3.2×10^{-5}	H ₂ , C, N ₂
SUS316L	0.8×10^{-6}	$> 1 \times 10^{-2}$	0.8×10^{-5}	H ₂ , C, N ₂
JPCA	0.8×10^{-6}	$> 1 \times 10^{-2}$	3.5×10^{-5}	H ₂ , C, N ₂

Table 1. Pressure history and major gas components observed during melting of the various specimens, as measured with a quadrupole mass spectrometer. For Cu and the Cu binary alloys, an electric furnace was used to melt the specimens. An induction furnace was used for the other materials.

Figure Captions

- Fig. 1. Damage structures of pure Cu specimens irradiated in FFTF to 2.1×10^{26} n/m² at 365°C. Sodium leakage occurred and contaminated the specimens.
- Fig. 2. Damage structures of Cu-5Al irradiated in FFTF without sodium contamination to 2.1×10^{26} n/m² at 410°C.
- Fig. 3. Damage structures of Cu-5Ni irradiated in FFTF to 7.0×10^{26} n/m² at 390°C. The specimens were contaminated with sodium.
- Fig. 4. Damage structures of pure Ni, Fe-16Cr-17Ni and SUS316L after neutron irradiation in JMTR to 3.8×10^{23} n/m² at 400°C, using improved temperature control.
- Fig. 5. Damage structures of pure Ni, Fe-16Cr-17Ni and SUS316L after irradiation in JMTR to 8.9×10^{23} n/m² at 500°C, using improved temperature control.
- Fig. 6. Damage structure of Fe-15Cr-20Ni, SUS316L and JPCA after irradiation in the HVEM at 350°C for Fe-15Cr-20Ni, and at 400°C for SUS316L and JPCA.

Fig. 1. Y. Shimomura et al.

1210

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Cu-5Ni As fabricated

Vacuum melted

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15-13

Fig. 5. Y. Shimomura et al.

16 14

Fig. 6. Y. Shimomura et al.

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