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Effects of solid transmutants and helium in copper studied by mixed-spectrum neutron irradiation

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Abstract

Microstructures of pure Cu and Cu–Ni–Zn alloys irradiated in High Flux Isotope Reactor (HFIR) at 573 K to 9.2 dpa and 673 K to 10.4 dpa have been observed with TEM. Transmutant Ni and Zn of ~3 wt% were produced during the irradiation. The effect of Ni and Zn production during irradiation was interpreted based on the knowledge obtained from Fast Flux Test Facility (FFTF) irradiated Cu and Cu–Ni–Zn alloys. The effect of He produced from Ni in Cu–5Ni during irradiation in HFIR was consistent with that produced from ^{10}B in Cu–5Ni–^{10}B during irradiation in FFTF.

1. Introduction

Since neutronics calculations showed that Ni and Zn are produced in Cu during fusion neutron irradiation [1,2], the role of those elements in Cu has attracted attention. Response of Cu–Ni, Cu–Zn and Cu–Ni–Zn alloys to fast neutron [3,4], electron [5] and ion [6] irradiations was investigated for this purpose.

In consequence, the understanding of the role of Ni and Zn on defect behavior, dislocation evolution, void nucleation and growth has been enhanced significantly in condition that those elements are doped before irradiation. However, the effect of increasing concentration of Ni and Zn during irradiation, as is the case of fusion neutron irradiation, has not been studied yet.

Since Ni and Zn are generated in Cu also by interactions with thermal neutrons, irradiation with mixed-spectrum neutrons would make it possible to study the transmutation effects in fusion-relevant conditions.

In the present study, microstructures of Cu and Cu–Ni–Zn alloys were observed after irradiation with mixed spectrum neutrons in HFIR. The production rates of transmutant Ni and Zn from Cu in the HFIR condition were higher than those in the fusion condition. This experiment is, thus, considered to be an acceleration simulation of the transmutation effect.

The results were compared with those by fast neutron irradiations, in which the transmutation rate was lower than that in the fusion condition.

2. Experimental

The alloys used are pure Cu, Cu–5Ni, Cu–3.5Zn and Cu–5Ni–2Zn. They were produced from high purity base metals by arc melting in pure Ar gas atmosphere. After the melting, the specimens were annealed at 873 K for 2 h. These specimens were from the same batches used in our previous experiments [4–6]. Some pure Cu specimens were also cold worked to study the role of pre-introduced dislocations.

Irradiation was carried out at the PTP position of HFIR at 573 K to 9.2 dpa and 673 K to 10.4 dpa. The temperature is considered to be accurate within ±25 K [7]. The chemical composition and He concentration were measured after irradiation. The results are summarized in Table 1. The procedure of the analysis is reported in another paper of these proceedings [8].

Table 1 shows that about 3 wt% Ni and Zn were produced in Cu at ~10 dpa. The production rates

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Table 1
Summary of chemical analysis of Cu and Cu alloys irradiated in HFIR-PTP [8]

	Ni (wt%)	Zn (wt%)	He (appm)
Cu			
Unirradiated	–	–	–
573 K, 9.2 dpa	2.8	3.1	3.8
673 K, 10.4 dpa	3.2	3.4	4.6
Cu–5Ni			
Unirradiated	5.5	–	–
573 K, 9.2 dpa	7.2	3.0	141
673 K, 10.4 dpa	8.1	3.7	179
Cu–3.5Zn			
Unirradiated	–	3.8	–
573 K, 9.2 dpa	2.7	6.3	8.2
673 K, 10.4 dpa	3.0	4.4	8.8
Cu–5Ni–2Zn			
Unirradiated	5.7	2.6	–
573 K, 9.2 dpa	NA	NA	146
673 K, 10.4 dpa	NA	NA	182

–: Less than dectection limit.
NA: Not available.

were much higher than those calculated in fusion conditions (0.1–0.2 wt% at 10 dpa [2]). He in Cu–5Ni and Cu–5Ni–2Zn was produced mostly through ^{58}Ni(n,γ) ^{59}Ni(n,α)^{56}Fe reactions.

After irradiation, specimens were electropolished for TEM examination. Microstructural observation and EDS microchemical analysis were carried out with JEM-2000FX electron microscope at Pacific Northwest National Laboratory.

3. Results

Fig. 1 shows void images for all alloys irradiated at 573 and 673 K. Voids were formed in all specimens except Cu–5Ni and Cu–5Ni–2Zn irradiated at 573 K. In these alloys, high density of small bubbles were observed in the matrix at 673 K. The significant difference in microstructures observed in the two alloys from those in other alloys is considered to be the influence of transmutant He produced from Ni.

Similar difference was observed for dislocation structure. In contrast to low density of tangled dislocation observed in Cu and Cu–3.5Zn, dislocations in Cu–5Ni and Cu–5Ni–2Zn were composed of interstitial loops at the both temperatures.

No remarkable difference was derived between microstructures in annealed Cu and cold worked Cu. Segregation of the transmutant Ni was detected at grain boundaries and voids by EDS. The distribution of Zn was, however, not detected by EDS because of heavy overlapping of the signal from Zn with radiation background.

Microstructural parameters and microchemical data will be compared with those derived by fast neutron irradiations in Section 4.

4. Discussion

4.1. Comparison with fast neutron irradiation data

The present microstructures at 673 K have been compared with those derived by irradiation in FFTF

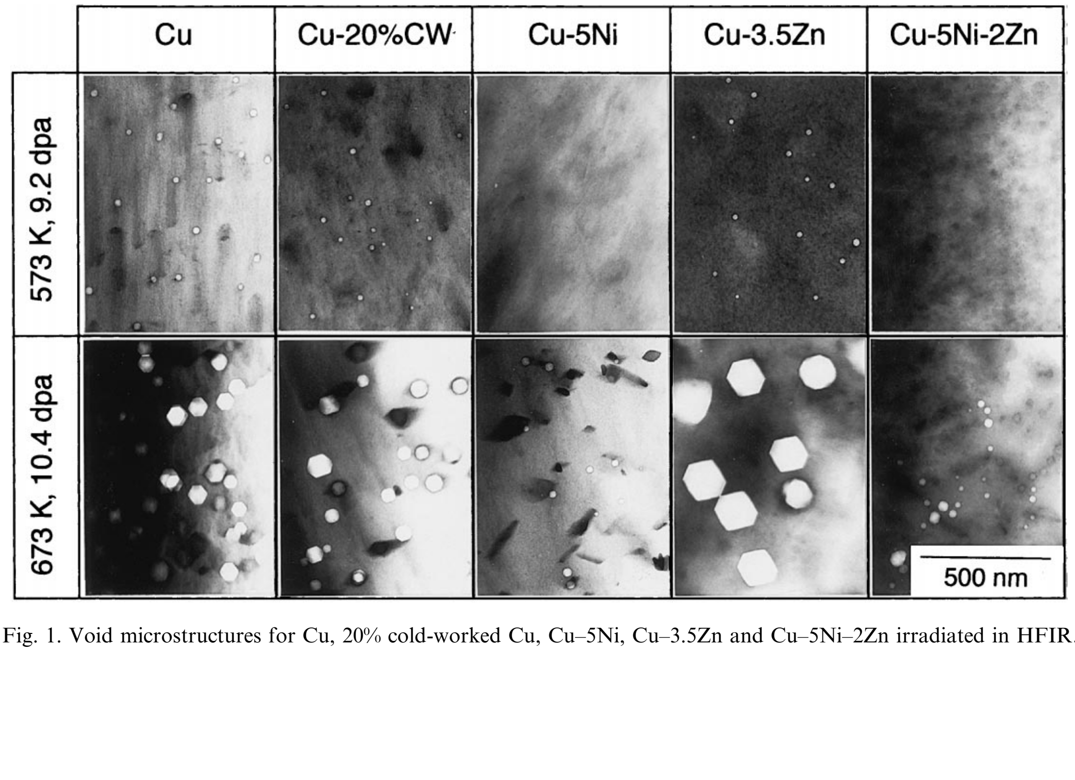

Fig. 1. Void microstructures for Cu, 20% cold-worked Cu, Cu–5Ni, Cu–3.5Zn and Cu–5Ni–2Zn irradiated in HFIR.

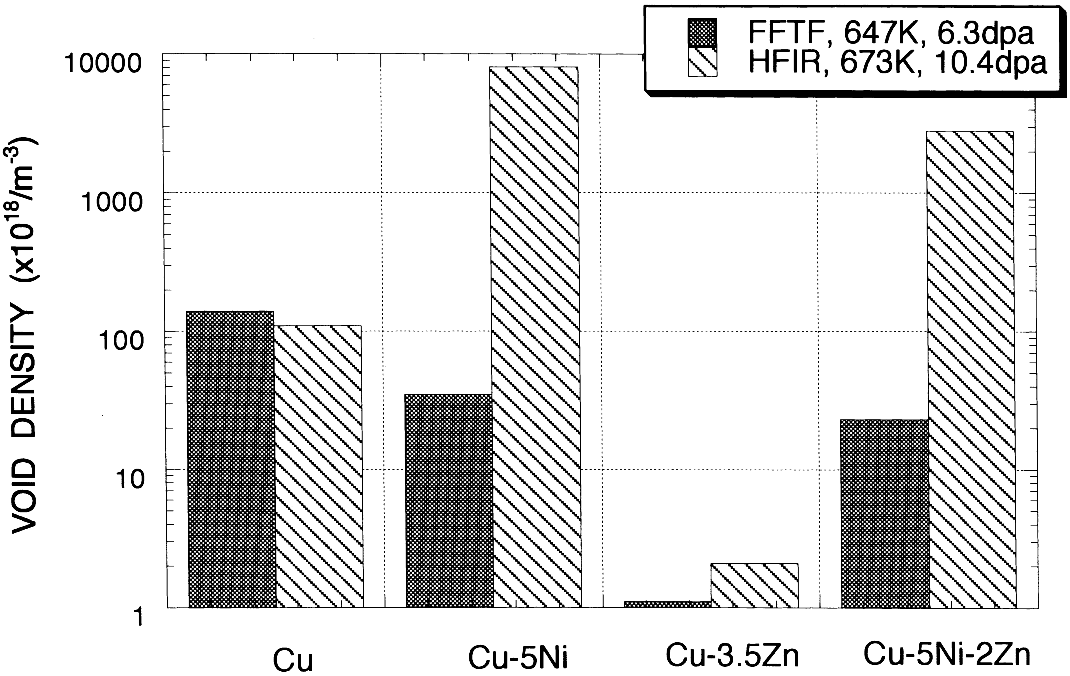

Fig. 2. Void density of Cu, Cu–5Ni, Cu–3.5Zn and Cu–5Ni–2Zn irradiated in FFTF [4] and HFIR (present study).

Below Core Canisters (BCC) at 647 K and 6.3 dpa [4]. Calculations show that the concentration of Ni and Zn after irradiation in FFTF to this fluence is only ~0.1 wt% [2].

Figs. 2 and 3 show void density and swelling for the four alloys irradiated in FFTF and HFIR. The enormous difference in void density of Cu–5Ni and Cu–5Ni–2Zn between FFTF and HFIR is attributed to that in He generation rate.

In Cu–3.5Zn, the void density was high and the swelling was low in the case of HFIR irradiation relative to those in the case of FFTF irradiation. In pure Cu, in

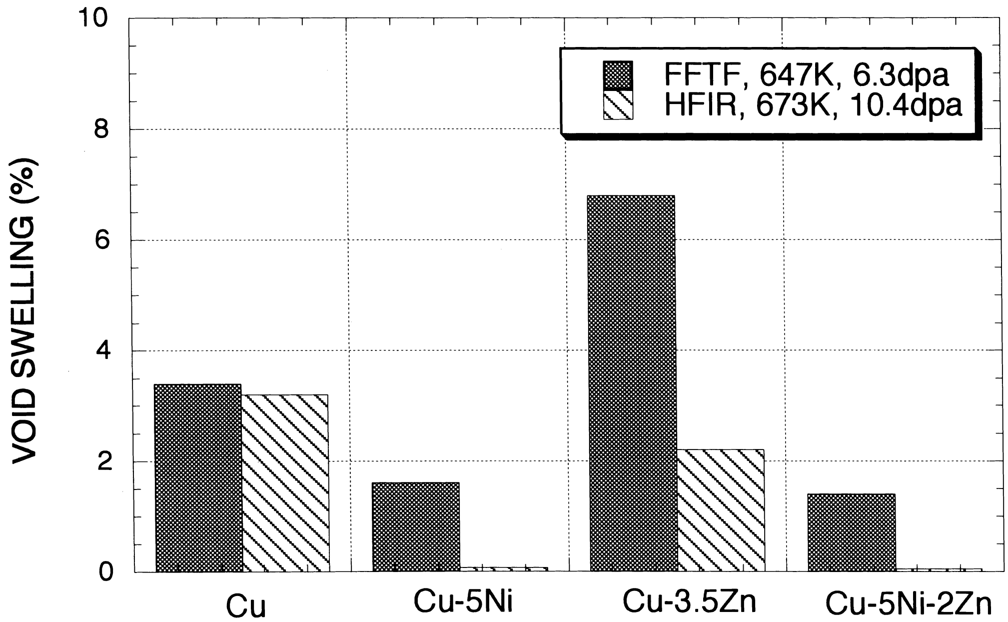

Fig. 3. Swelling of Cu, Cu–5Ni, Cu–3.5Zn and Cu–5Ni–2Zn irradiated in FFTF [4] and HFIR (present study).

contrast, significant difference in the void microstructure was not observed between the two cases.

The effect of Ni and Zn on void nucleation and growth during irradiation was qualitatively understood by fast neutron irradiations [4]. Part of the results were shown in Figs. 2 and 3. The comparison of Cu and Cu–5Ni–2Zn irradiated in FFTF implies that the addition of both Ni and Zn on Cu decreases the void density. The similar void density of FFTF and HFIR irradiated pure Cu, seen in Fig. 2, suggests that the transmutant Ni and Zn affected only weakly the void density of Cu during the HFIR irradiation. This is understandable, because the void density would reach its saturation level during the initial irradiation period (<1 dpa) [9], when the concentration of the solid transmutant was still low. If difference in doses is considered, however, the void swelling in Cu irradiated in FFTF is higher than that in HFIR on the dpa basis. The moderate effect of the solid transmutants and the irradiation history may be possible reasons of the difference.

The difference in void parameters of Cu–3.5Zn irradiated in FFTF and HFIR will be attributed to transmutant Ni generated during irradiation in HFIR. The void density and swelling of Cu–3.5Zn irradiated in HFIR are between those of Cu–3.5Zn and Cu–5Ni–2Zn irradiated in FFTF. During the later period of the irradiation in HFIR, the accumulation of Ni probably induced additional void nucleation and suppressed void growth in Cu–3.5Zn.

Fig. 4 compares the traverse of Ni concentration across an area containing a void for Cu–5Ni and Cu–5Ni–2Zn irradiated in FFTF and pure Cu irradiated in HFIR. The FFTF data imply that addition of Zn suppresses the segregation of Ni during irradiation. The segregation level of Ni at the void surface in Cu irradiated in HFIR is similar to that in Cu–5Ni–2Zn irradiated in FFTF. This is understandable according to the fact, shown in Table 1, that about 3 wt% Zn was produced in addition to Ni during irradiation in HFIR.

4.2. Helium effect on microstructure

He effects on microstructural evolution of Cu–5Ni during irradiation in FFTF were investigated by the ^{10}B-doping technique [10]. Fig. 5 compares void average size and density as a function of He/dpa ratio in the case of FFTF and HFIR irradiations. The figure shows that the void density once increases and then slowly decreases with the increase in He/dpa. The void average size changed in an opposite manner with He/dpa. The FFTF and the HFIR data are consistent with each other.

The He generation rate slowly decreased during irradiation in the case of FFTF because of burning of ^{10}B.

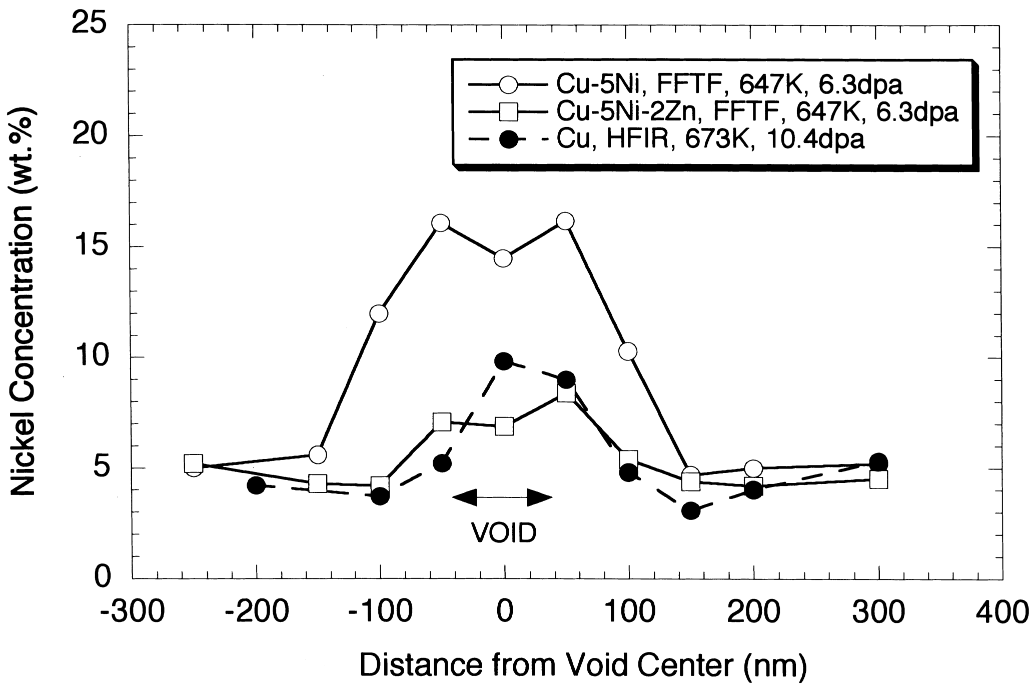

Fig. 4. Comparison of the Ni concentration traverse across an area containing a void for Cu–5Ni and Cu–5Ni–2Zn irradiated in FFTF [4] and pure Cu irradiated in HFIR (present study). The thickness of specimen area analyzed were ~200 nm.

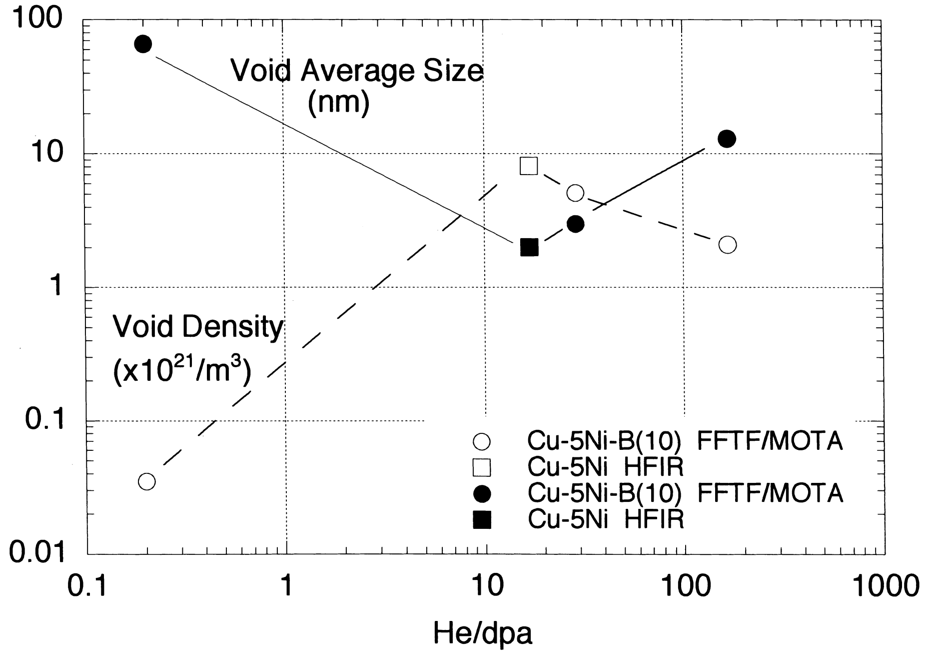

Fig. 5. Void average side and density as a function of He/dpa ratio for Cu–5Ni–^{10}B irradiated in FFTF [10] and Cu–5Ni irradiated in HFIR (present study).

The He generation rate in HFIR, on the other hand, increased gradually corresponding to the accumulation of ^{59}Ni. Fig. 5 shows, however, that the historical difference in He generation does not affect strongly the void microstructures.

The present result may imply that He effect overwhelm the solid transmutant effect. However, the comparison of Cu–^{10}B and Cu–5Ni–^{10}B irradiated in FFTF showed that the Ni effect is even stronger under continuous generation of He [10]. Thus, the effect of accumulation of both solid and gaseous transmutants should be investigated further.

4.3. Use of mixed spectrum neutrons for transmutation studies

The present study demonstrated that an acceleration simulation of solid transmutation by fusion neutron irradiation is possible using mixed spectrum neutrons. Also shown in this study was that the knowledge obtained from fast neutron irradiation of materials doped with transmutant elements can be usefully applied to elucidate the transmutation effects.

In Japan–USA fusion collaboration program (JUPITER project), irradiation in HFIR-RB* position with an Eu shield is now in progress. With the shield, the generation rates of Ni and Zn in Cu and Cr in V are reduced and approach to the fusion conditions [11].

5. Conclusions

An acceleration simulation of Ni and Zn generation in Cu during fusion neutron irradiation is possible using mixed spectrum neutrons. The fast neutron irradiation study with materials doped with expected transmutant elements can be applied to the interpretation of the mixed spectrum irradiation data. Tailoring of the transmutation rate and simultaneous generation of both solid and gaseous elements are very important to study the combined transmutation effects in fusion conditions [11].

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