EFFECT OF FISSION NEUTRON IRRADIATION ON THE TENSILE AND ELECTRICAL PROPERTIES OF COPPER AND COPPER ALLOYS

– S.A. Fabritsiev (D.V. Efremov Institute, St. Petersburg, Russia), A.S. Pokrovsky (SRIAR, Dimitrovgrad, Russia), S.J. Zinkle and A.F. Rowcliffe (Oak Ridge National Laboratory), B.N. Singh (Risø National Laboratory, Roskilde, Denmark), F.A. Garner and D.J. Edwards (Pacific Northwest Laboratory)

OBJECTIVE

The objective of this study is to evaluate the properties of several copper alloys following fission reactor irradiation at ITER-relevant temperatures of 80 to 200°C. This study provides some of the data needed for the ITER research and development Task T213.

SUMMARY

The tensile and electrical properties of several different copper alloys have been measured following fission neutron irradiation to ~1 and 5 dpa at temperatures between ~90 and 200°C in the SM-2 reactor. These low temperature irradiations caused significant radiation hardening and a dramatic decrease in the work hardening ability of copper and copper alloys. The uniform elongation was higher at 200°C compared to 100°C, but still remained below 1% for most of the copper alloys. As expected, specimens shielded from the thermal neutrons (which produced fusion-relevant solid transmutation rates) exhibited a lower increase in their electrical resistivity compared to unshielded specimens. A somewhat surprising observation was that the radiation hardening was significantly higher in unshielded copper specimens compared to spectrally-shielded specimens.

STATUS AND PROGRESS

1. Introduction

Copper alloys have been proposed for the first wall and divertor structure of the International Thermonuclear Experimental Reactor (ITER). Unfortunately, there is a lack of data on copper and copper alloys irradiated to damage levels greater than 1 dpa at ITER-relevant temperatures of 100 to 350°C [1]. In order to improve the existing data base on radiation effects on copper alloys at ITER-relevant conditions, planning for a collaborative irradiation experiment in the SM-2 reactor in Dimitrovgrad, Russia was initiated in 1992. Sheet tensile specimens and electron microscopy disks were prepared by US and Russian scientists from a wide range of copper and copper alloys [2], and the specimens were irradiated in several capsules between December, 1993 and June, 1994. A total of 74 copper alloys have been irradiated in the SM-2 reactor at temperatures between ~90 and 330°C in the Channel 4 and Core positions of the reactor, resulting in displacement damage levels of ~1 and 5 dpa, respectively [2]. The initial results obtained from postirradiation examination of the specimens are summarized in this report.

2. Experimental Procedure

The list of specimens and design irradiation conditions have been summarized elsewhere [2]. Specimens were irradiated in the form of transmission electron microscope (TEM) disks and sheet tensile specimens designated small tensile specimens (STS, 34 mm long and 0.25 mm thick) and large tensile specimens (LTS, 56 mm long and 1 mm thick). Electrical resistivity measurements were made at room temperature using the 4-point probe technique on the STS and LTS specimens prior to tensile testing. The irradiations were conducted in the Channel 4 and Core positions of the SM-2 reactor. Specimens were irradiated for 42.3 and 49.5 effective full power days in two different capsules in the Channel 4 position at design temperatures of 120 and 240°C (designated SMT-1 and SMT-2, respectively). The Channel 4 capsules consisted of 10 subcapsules in the form of flattened stainless steel tubes 12 to 14 mm in diameter and 170 mm long, with the copper specimens contained therein. The subcapsules were filled with helium at a pressure of 0.1 MPa and sealed. All subcapsules were surrounded by 50 to 60°C reactor coolant water.

Three complete sets of fluence monitors (based on ^{58}Fe, ^{54}Fe, ^{63}Cu, ^{58}Ni, ^{46}Ti, and ^{93}Nb) were used to determine the fast and thermal neutron fluxes as a function of position. Two sets of fluence monitors were positioned at the bottom and horizontal midplane of the reactor in order to determine the axial variation in the neutron flux. Duplicate sets of monitors were positioned at the inner and outer periphery of the capsule at the horizontal midplane in order to examine the radial flux gradient across the capsule. The average fast neutron fluence (E>0.1 MeV) in these capsules was 1.1×10^{25} n/m^2, which corresponds to a calculated damage level in copper of ~0.9 displacements per atom (dpa). The fast fluence (E>0.1 MeV) ranged from 0.73 to 2.3×10^{25} n/m^2 (~0.56 to 1.8 dpa) depending on specimen position in the capsule. The subcapsules in the bottom half of the Channel 4 capsules (containing all of the US specimens) were enclosed in a 1.5 mm Cd shroud in order to shield the specimens from thermal neutrons. The specimen irradiation temperatures in the Channel 4 capsules were determined from three different chromel-alumel thermocouples installed in the unshielded portion of the capsule. Heat transfer analysis codes were used to calculate the specimen temperatures in the shielded portion of the capsules. The average specimen temperature in the unshielded portion of the SMT-2 capsule was determined to be 160 and 210°C for the TEM specimens and LTS specimens, respectively. The corresponding temperatures in the shielded portion of the capsule were 200 and 220°C, respectively.

Four capsules were used to irradiate specimens in the Core position at design temperatures of 120, 120, 240 and 360°C (designated SMT 3-1, 3-2, 3-3 and 3-4, respectively). All of the Core position capsules were fabricated as a flattened stainless steel tube initially 9 mm in diameter and 350 mm long. The SMT 3-1 and SMT 3-2 capsules utilized Al spacers whereas the SMT 3-3 and SMT 3-4 capsules included strips of Mo and Ta, respectively in order to increase the nuclear heating in the capsule and thereby achieve higher temperatures. All of the capsules were irradiated for a total of 44.5 effective full power days. This produced average fast neutron fluences (E>0.1 MeV) of 5.8×10^{25} n/m^2 (~4.5 dpa)in the SMT 3-1 and 3-2 capsules and 7.1×10^{25} n/m^2 (~5.5 dpa) in the SMT 3-3 and 3-4 capsules. The corresponding thermal neutron fluences (E<0.67 eV) in these capsules were 6.5×10^{24} n/m^2 and 7.9×10^{24} n/m^2, respectively. Two sets of fluence monitors were installed in the SMT 3-1 capsule at the top and horizontal midplane of the core. The remaining Core capsules were furnished with one set of fluence monitors, located near the horizontal midplane. The temperature in the Core capsules was determined from postirradiation examination of small packets containing material with different melting temperatures. The SMT 3-1 and 3-2 capsules contained Sn alloy (T_M=156°C) and Pb-Bi alloy (T_M=125°C) packets near the horizontal midplane of the capsule. Melting was not observed in either material, indicating that the temperature in these capsules was <125°C. The SMT 3-3 capsule contained Sn (T_M=232°C) and Bi (T_M=271°C) packets near the top and horizontal midplane of the capsule. Examination of the packets indicated that the irradiation temperature was $\leq$ 270°C at the capsule midplane and $\leq$ 230°C at the top and bottom of the capsule. The SMT 3-4 capsule contained Pb (T_M=327°C) and Zn (T_M=419°C) packets near the midplane and top part of the capsule. The irradiation temperature was determined to be ~410°C at the capsule midplane and ~325°C near the top and bottom of the capsule.

Postirradiation examination of the irradiated specimens is in progress. The present report summarizes some of the measurements which have been made following irradiation near 90 and 200°C. All of the tensile measurements were performed at temperatures of either 100 or 200°C, which is approximately equal to the irradiation temperature.

3. Results and Discussion

3.1 Effect of Cd shielding

One of the important physical properties for copper alloys in high heat flux applications is the thermal conductivity. The effect of irradiation on the thermal conductivity (k_{th}) of copper and copper alloys can be determined from electrical resistivity (ρ_e) measurements (which can be measured more easily and accurately than k_{th}) by utilizing the well-known Wiedemann-Franz relation $k_{th}\,\rho_e = L\,T$, where the Lorentz number (L) equals 2.23×10^{-8} for copper at 0°C [1]. Table 1 compares the electrical resistivity measured in specimens in the Cd-shielded and unshielded regions of the SMT-1 capsule irradiated in the Channel 4 position to a nominal damage level of ~0.9 dpa at ~90°C. The typical increase in the room temperature electrical resistivity was ~10% for the specimens irradiated in the Cd-shielded positions, whereas the typical resistivity increase was ~30% for the unshielded specimens. The large resistivity increase in the unshielded specimens is due to the high transmutation rate of Cu to Ni and Zn in the presence of thermal neutrons [1]. The transmutation rate for Cu in the Channel 4 position of the SM-2 reactor in the absence of Cd shielding is ~3000 appm Ni/dpa and ~1700 appm Zn/dpa. The transmutation rates in the Cd-shielded regions of the SMT-1 capsule were ~570 appm Ni/dpa and ~320 appm Zn/dpa, which is closer to the fusion first wall transmutation rates [1] of ~190 appm Ni/dpa and ~90 appm Zn/dpa. Table 2 summarizes the corresponding tensile property measurements for Cd-shielded and unshielded specimens in the SMT-1 capsule.

Table 1. Room Temperature Electric Resistivity ρ (μΩ-cm) of Small Tensile Specimens (STS) Irradiated in the SMT-1 Capsule at ~88°C

Alloy	Fluence (10^{25}n/m^2)		$\rho_{nonirr.}$	$\rho_{irr.}$	$\Delta\rho_{nonirr.}$ (%)
	fast	thermal			
Cd shielded					
Cu + B3 cold worked	1.03	0.227	1.942	2.111	8.7
Cu + B1 cold worked	1.21	0.227	1.858	2.046	10.1
Cu + B1 annealed	–	–	1.742	2.099	20.5
Cu + B2 annealed	–	–	1.719	2.085	21.3
Cu pure 1 annealed	1.21	0.227	1.681	1.905	13.3
Cu pure 11 cold worked	1.78	0.292	1.779	1.989	11.8
Cu pure 1 cold worked	1.78	0.292	1.751	1.969	12.5
Cu-Mo-B as wrought	–	–	1.915	2.179	13.8
MAGT-0.2 as wrought	1.53	0.292	2.043	2.282	11.7
Cu + B2 coldworked	–	–	1.801	2.108	17.0
GlidCop Al25 (no B) 20% cold worked	1.27	0.292	1.948	2.266	16.3
GlidCop Al25 (no B) as wrought	–	–	1.981	2.264	14.3
GlidCop Al25 + B as wrought	–	–	1.991	2.144	15.4
Cu 99.999% oxyg. 950°C, 0.5 h	1.21	0.227	1.699	1.918	12.9
Cu 99.999% anneal 400°C, 1 h	1.21	0.227	1.692	1.904	12.5
GlidCop Al15 + B as wrought	0.86	0.227	2.018	2.308	14.4
GlidCop Al15 + B ind. braze	0.86	0.227	2.243	2.435	8.5
GlidCop Al15 + B 70% cold worked	1.27	0.292	1.941	2.218	14.3
Cu 99.999% 80% cold worked	1.27	0.292	1.727	1.943	12.5
GlidCop Al25 (no B) 20% cold worked	1.27	0.292	1.933	2.203	13.9
Cu 99.999% ann. 550°C, 2h	1.21	0.227	1.686	1.880	11.5
without Cd					
Cu + B3 cold worked	1.2	1.24	1.911	2.500	30.8
Cu + B1 cold worked	0.82	0.97	1.819	2.359	29.7
Cu + B1 annealed	1.85	1.4	1.736	2.461	41.8
Cu + B2 annealed	1.42	1.4	1.762	2.455	39.3
Cu pure 1 annealed	1.42	1.4	1.700	2.336	40.0
Cu pure 11 cold worked	–	–	1.776	2.287	28.7
Cu pure 1 cold worked	2.27	1.4	1.750	2.434	39.1
Cu-Mo-B	–	–	2.026	2.579	27.3
MAGT-0.2 as wrought	1.3	0.97	2.048	2.692	31.4
Cu + B2 cold worked	–	–	1.789	2.412	34.8

Table 2. Mechanical Properties of Copper and Alloys Irradiated in the SMT-1 Capsule at ~88°C and Tested at 100°C (LTS Geometry)

Alloy	Fluence (10^{25}n/m^2) fast	Fluence (10^{25}n/m^2) thermal	σ_y (MPa)	σ_{UTS} (MPa)	δ_u (%)	δ_{tot} (%)
Cd shielded						
Cu pure 350°C, 1h	0.86	0.227	268	269	0.6	4.4
Cu + B1 350°C, 1h	1.21	0.227	381	394	0.25	0.6
Cu + B2 350°C, 1h	1.78	0.292	411	413	0.16	1.8
MAGT-0.2 as wrought	1.27	0.298	538	544	0.25	0.62
Cu 99.999% as wrought	1.53	0.292	380	388	0.42	1.9
Cu 99.999% 550°C as wrought	1.78	0.292	187	216	12.0	16.4
GlidCop Al15 + B as wrought	1.78	0.292	385	391	0.38	0.5
GlidCop Al15 + B induction brazed	1.27	0.292	322	322	0	0
GlidCop Al25 (no B) as wrought	1.21	0.227	520	538	0.4	2.5
GlidCop Al25 + B as wrought	0.86	0.227	519	519	0.24	0.24
GlidCop Al25 (no B) 20% cold worked	1.03	0.227	458	469	0.41	3.9
GlidCop Al25 + B 20% cold worked	1.21	0.227	420	447	0.5	0.67
without Cd						
Cu pure cold worked	1.85	1.4	444	444	0.2	1.5
Cu pure 350°C, 1h	1.3	0.97	302	319	0.5	3.5
Cu pure 350°C, 1h	1.42	1.4	319	319	0.16	3.8
Cu + B1 350°C, 1 h	1.3	0.97	418	419	0.3	2.4
Cu + B1 350°C, 1 h	2.27	1.4	393	413	0.33	2.7
Cu + B2 350°C, 1h	0.82	0.97	494	497	0.1	1.0
Cu + B1 cold worked	1.9	1.24	506	506	0.16	0.16
Cu-Cr-Zr	2.27	1.4	413	413	0.1	0.1
Cu-Mo as wrought	1.2	1.24	199	206	0.33	1.4
Cu-Be	1.2	1.24	766	894	4.1	6.5
MAGT-0.2 as wrought	1.42	1.4	549	550	0.3	0.6
MAGT-0.2 as wrought	0.82	0.97	519	519	0.1	0.1

Figure 1 shows the measured and calculated electrical resistivity in pure copper as a function of thermal neutron fluence. The measurements were made on copper irradiated in the Core position (capsule SMT 3-1) and in the Cd-shielded and unshielded regions of the SMT-1 capsule irradiated in the Channel 4 position. The calculation assumed that the resistivity increase was mainly due to three contributions: radiation-induced defects (stacking fault tetrahedra and small dislocation loops) and Ni and Zn formed by solid transmutation reactions. The Ni and Zn contributions were calculated from their specific resistivities [1] and their thermal neutron production cross sections. The resistivity increase from radiation defects was taken to be constant (0.13 μΩ-cm) for damage levels greater than 0.1 dpa in accordance with previous studies on copper irradiated near room temperature [e.g., 3,4]. From Fig. 1 it can be seen that the dominant contribution to the resistivity increase at high thermal neutron fluences (> 5×10^{24} n/m^2) is due to Ni produced by transmutation reactions. The resistivity increase at low fluence (< 5×10^{24} n/m^2) is mainly due to defect clusters.

The effect of solid and gaseous (He) transmutations on the strength and electrical resistivity of copper was investigated by comparing the response of pure copper and two Cu-B alloys. High amounts of helium can be produced in boron-doped copper as a result of the thermal neutron $^{10}B(n,\alpha)^{7}Li$ reaction. Figure 2 shows the electrical resistivity increase in copper as a function of boron content for specimens irradiated in shielded and unshielded regions of the SMT-1 capsule (~0.9 dpa, 90°C). The boron additions had a

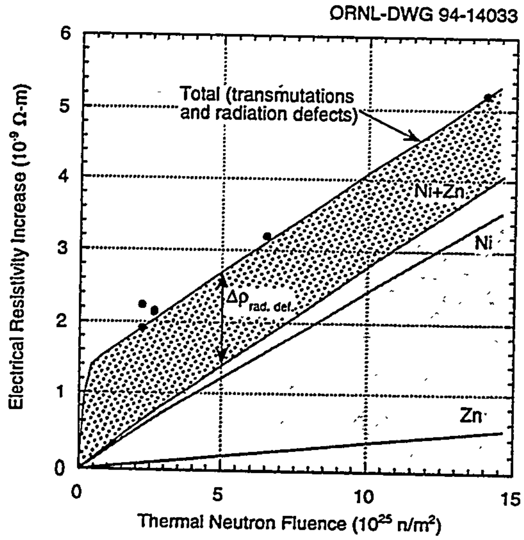

Fig. 1. Comparison of the calculated and experimentally measured effect of solid transmutations and radiation defects on the electrical resistivity of copper irradiated in the SM-2 reactor at 90°C.

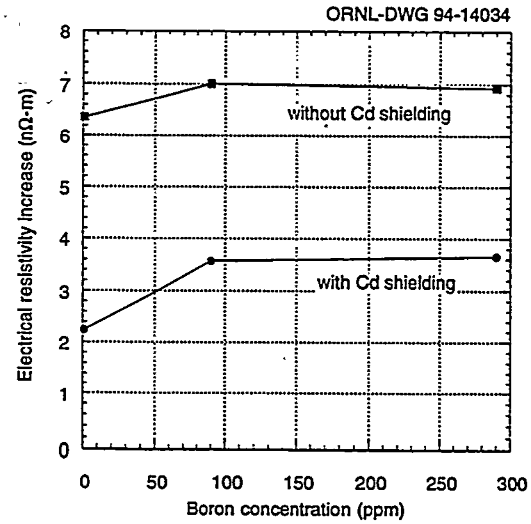

Fig. 2. Electrical resistivity change in copper and Cu-B alloys irradiated in the SMT-1 capsule (~90°C, 1 dpa).

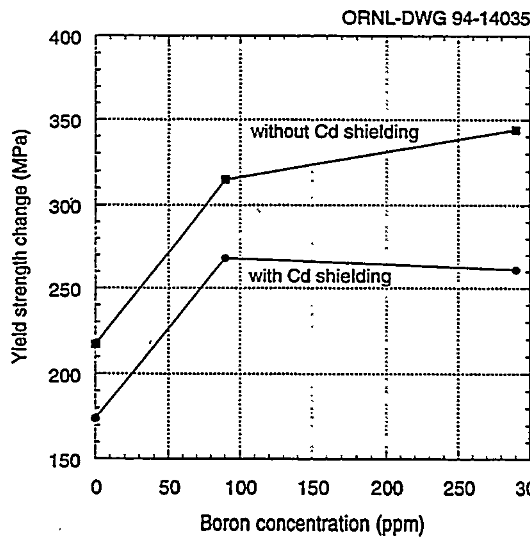

Fig. 3. Effect of Cd shielding and boron concentration on the yield strength of copper irradiated in the SMT-1 capsule (~90°C, 1 dpa).

relatively weak effect on the electrical resistivity increase, particularly in the unshielded specimens. It is obvious from Fig. 2 that the electrical resistivity increase is dominated by solid transmutation effects.

Figure 3 shows the yield strength change in copper as a function of boron content for specimens irradiated in shielded and unshielded regions of the SMT-1 capsule. The irradiated yield strength was relatively insensitive to the boron content. One surprising result was the large difference (>50 MPa) between the yield strength of the Cd-shielded and unshielded specimens. The major transmutation products in the unshielded copper specimens irradiated in SMT-1 capsule to a damage level of ~1 dpa would be ~0.3% Ni and ~0.17% Zn. Both of these elements are soluble in copper, and the resulting solution hardening for these solute concentrations would be expected to be only ~1 MPa [5]. This suggests that the small additions of Ni and Zn are somehow causing the radiation-induced defect structure to become more resistant to dislocation motion, perhaps by modifying the quenching behavior of the displacement cascade [6]. Recent studies have shown that addition of Ni to copper can greatly increase the dislocation loop density that forms during low temperature irradiation [7]. Further work, including TEM analysis, is needed to comprehend this unexpected pronounced dependence of radiation hardening on thermal neutron fluence.

3.2 Tensile Properties of Copper and High Strength Copper Alloys

Irradiation at 90 to 200°C to damage levels of ~0.9 dpa produced significant radiation hardening and an accompanying ductility reduction in all of the irradiated materials. Figure 4 shows the dose-dependent radiation hardening behavior of copper irradiated at temperatures between 20 and 100°C [8-12]. The yield strength in copper approaches an apparent "saturation" value of ~300 MPa after a dose of about 0.1 dpa. Higher dose studies (e.g., the SMT 3-1 capsule specimens that are still awaiting testing) are needed to determine if the yield strength has indeed reached a saturation level. According to the results presented in Fig. 3, it might be anticipated that solid transmutation effects might cause a gradual increase in the irradiated yield strength up high fluences.

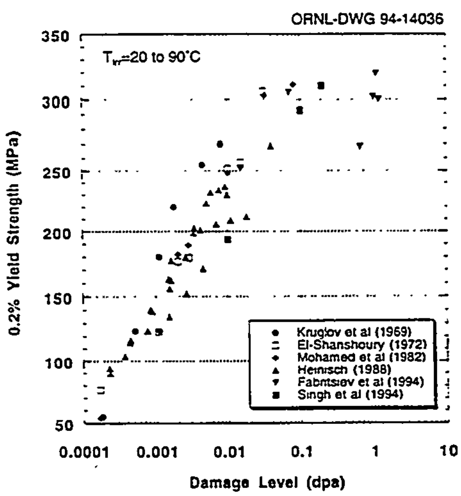

Fig. 4. Dose dependence of the yield strength of copper irradiated at low temperatures [8-12, this study].

Figure 5 shows the effect of irradiation (and test) temperature on the yield strength of GlidCop Al25 oxide dispersion strengthened copper and CuCrZr following irradiation in the Cd-shielded Channel 4 capsules to a damage level of ~1 dpa. The yield strength of all of the alloys was approximately 100 to 200 MPa lower at 200°C compared to 100°C. The two as-wrought GlidCop alloys (with and without ~100 ppm B) had the highest strength following irradiation near 100°C. The 20% cold-worked GlidCop alloy (without B) had the highest strength following irradiation near 200°C. The ultimate tensile strengths of the 5 alloys showed a similar temperature-dependent trend.

Figure 6 compares the temperature-dependent yield strength of as-wrought GlidCop Al-25 (containing ~100 ppm B as a deoxidant) before and after neutron irradiation to ~1 dpa in the Cd-shielded Channel 4 capsules. Irradiation at ~100°C produced ~170 MPa increase in the yield strength compared to the unirradiated material. However, the yield strength of GlidCop irradiated at ~200°C was comparable to the unirradiated material. The strong temperature dependence of radiation hardening in GlidCop is in agreement with previous studies performed on pure copper [13], and may be attributable in large part to thermally activated bypassing of the small defect clusters. At temperatures above 200°C a further rapid drop in the radiation hardening of copper would be expected due to the thermal instability of the radiation-produced defect clusters.

Radiation hardening in the temperature range of ≤200°C produces an increase in the strength of copper and copper alloys, which is generally considered to be a desirable feature. However, this strength increase is accompanied by a dramatic decrease in the work hardening ability of copper and copper alloys. Figure 7 compares the uniform and total elongation of pure copper before and after neutron irradiation at ~100°C to a damage level of ~1 dpa. The uniform elongation decreased from ~29% before irradiation to ~0.6% following irradiation. Similar pronounced decreases in the uniform and total elongation have been observed in all copper alloys examined to date. The smallest elongations occurred in alloys irradiated at 100°C, with significantly higher (although still low) elongations measured after irradiation near 200°C. Figure 8 shows examples of the total elongation in GlidCop oxide dispersion strengthened copper and CuCrZr following irradiation to ~1 dpa at ~100 and 200°C. The total elongation in most of the alloys was approximately doubled as the irradiation temperature increased from 100 to 200°C. A similar improvement in the uniform elongation was observed in all of the alloys as the irradiation temperature increased from 100 to 200°C, although the measured uniform elongation at 200°C was still generally <1%.

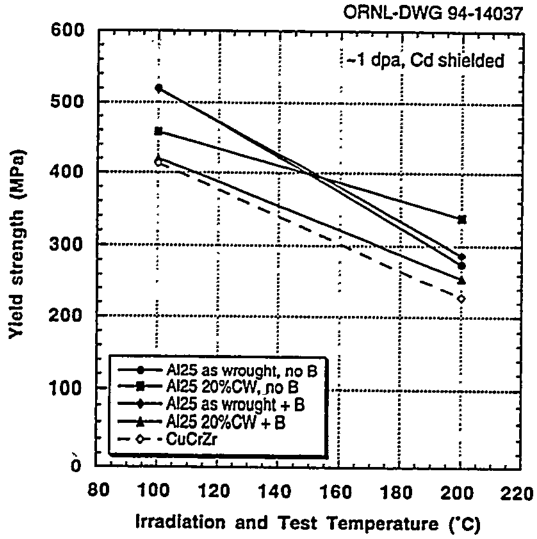

Fig. 5. Effect of temperature on the yield strength of copper alloys irradiated in Cd-shielded capsules to a dose of ~1 dpa.

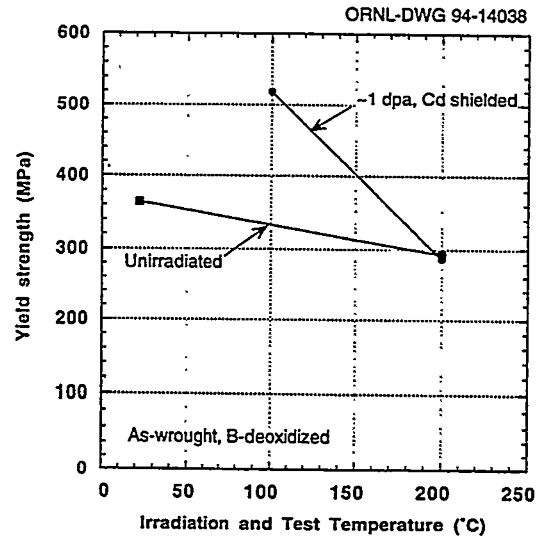

Fig. 6. Comparison of the temperature-dependent yield strength of GlidCop Al-25 before and after neutron irradiation.

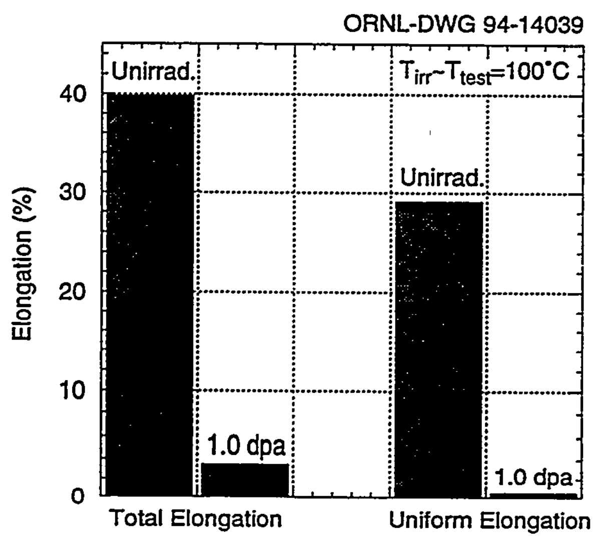

Fig. 7. Low temperature embrittlement of copper irradiated in the SMT-1 capsule. LTS specimens annealed at 350°C before irradiation.

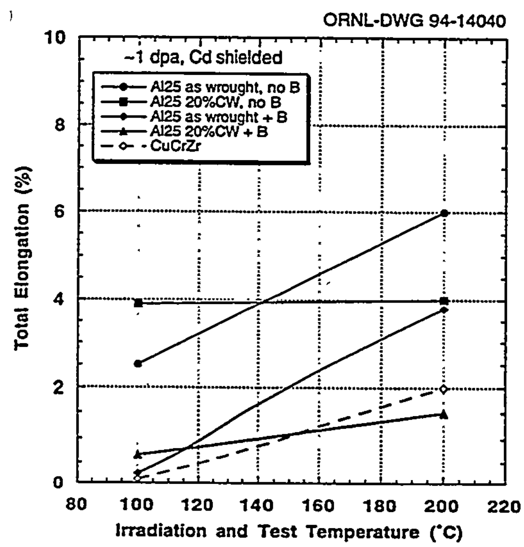

Fig. 8. Effect of temperature on the total elongation of copper alloys irradiated in Cd-shielded capsules to a dose of ~1 dpa.

FUTURE WORK

During the next reporting period, tensile and electrical property measurements will be completed on specimens irradiated in the Channel 4 and Core capsules. Transmission electron microscopy disks of the US specimens will be transported to Risø National Laboratory in Denmark and to PNL and ORNL in the US for immersion density measurements and TEM analysis.

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