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LOW-TEMPERATURE RADIATION EMBRITTLEMENT OF COPPER ALLOYS

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ABSTRACT

The effect of low-temperature (T_{irr} less than 0.3 T_{melt}) irradiation on the tensile properties of copper and precipitation-hardened (PH) and dispersion-strengthened (DS) copper alloys was investigated. Samples were irradiated with fission neutrons at temperatures of 80 to 200°C to doses of 0.6 to 5 dpa. Irradiation at temperatures <150°C resulted in significant hardening and accompanying embrittlement in all of the materials. By comparing the present results with literature data, it is concluded that severe radiation embrittlement occurs in copper alloys irradiated at temperatures $\leq$100°C for doses above ~0.01 to 0.1 dpa. On the other hand, irradiation at temperatures above 150°C causes only moderate embrittlement for doses up to ~5 dpa. It is recommended that the minimum operating temperature for copper alloys intended for structural applications in fusion energy systems should be 150°C, unless uniform elongations <1% can be accomodated in the design.

Key words: yield strength, uniform elongation, total elongation, radiation hardening

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1. Introduction

The use of copper alloys as a structural material for the ITER first wall and divertor poses before materials scientists a large variety of challenging problems. In many respects these problems arise from a lack of knowledge about the mechanical properties of copper alloys irradiated at ITER relevant conditions (50 to 350°C, doses up to ~10 dpa). Previous work on pure copper [1,2] and copper alloys [3-9] has shown that significant radiation hardening and loss of ductility occurs in these materials following neutron irradiation at temperatures below 250°C. These studies suggest that low-temperature irradiation to doses as low as ~0.1 dpa may cause substantial embrittlement of copper alloys. However, the data base on precipitation-hardened (PH) and dispersion strengthened (DS) copper alloys at irradiation temperatures below 250°C is very limited. There are only a few known studies of the tensile properties of these materials at damage levels >1 dpa at irradiation temperatures <200°C, where radiation hardening and embrittlement effects are most pronounced [3-6,8]. The present study summarizes recent results obtained on copper and copper alloys irradiated at 80-200°C to doses between 0.6 and 5 dpa. These data represent a subset of a collaborative irradiation program that investigated the electrical and mechanical properties of copper and copper alloys irradiated at temperatures between 80 and 400°C and doses of 0.6 to 5 dpa [3].

2. Experimental Procedure

Sheet tensile specimens from copper (Russian Federation (RF) Cu, 99.97% purity and A.D. Mackay Cu, 99.999% purity), DS copper alloys (GlidCop Al25, GlidCop Al15, MAGT-0.2, Cu-Mo), and a PH copper alloy (Cu-Cr-Zr) were irradiated in the Channel 4 and Core positions of the SM-2 reactor. Most of the materials were irradiated in both annealed and cold worked conditions. Two types of sheet tensile specimens were irradiated and tested. The large tensile specimens (LTS) had an overall length of 56 mm and a thickness of 1 mm, with a gage length and width of 30 and 4 mm, respectively. The small tensile specimens (STS) had an overall length of 34 mm and a thickness of 0.25 mm, with a gage length and width of 11 and 3.5 mm, respectively. The irradiated and control samples were tensile tested in vacuum (~10^{-4} Pa) at a crosshead speed of ~ 1 mm/min in the temperature range of 20-400°C, which corresponds to an initial strain rate of 5.6×10^{-4}/s and 1.5×10^{-3}/s for the LTS and STS specimens, respectively. For the elevated temperature tests, the tensile specimens were held at temperature for 0.5 hour prior to testing. The fracture surfaces were investigated by scanning electron microscopy (SEM).

The specimens were irradiated in the Channel 4 position to doses of 0.6 to 1.4 dpa (0.8 to 2×10^{25} n/m^2, E>0.1 MeV) at T_{irr} ~ 100 and 200°C, as well as to doses of ~ 3 to 5 dpa at T_{irr} ~ 100 and

250°C in the Core position. The average fast (E>0.1 MeV) and thermal neutron fluxes were $3x10^{18}$ and $2.5x10^{18}$ n/m^2-s, respectively, in the Channel 4 position, and $1.7x10^{19}$ and $1.9x10^{18}$ n/m^2-s, respectively, in the Core position. In order to reduce the solid transmutation production rate to fusion-relevant levels, some of the samples in the Channel 4 region were irradiated in Cd-shielded capsules, thus making it possible to considerably suppress the thermal neutron flux. The thermal neutron flux in the Cd-shielded Channel 4 capsules was ~$3.6x10^{17}$ n/m^2-s. Further details on the irradiation technique and capsule design are presented elsewhere [3,10].

3. Experimental results.

Fig. 1 shows the measured yield strength and total elongation in annealed and cold-worked pure copper after irradiation in the Channel 4 and Core positions at ~100°C. Also included in Fig. 1 are low-dose data by Makin [1] on cold-worked and annealed copper for the sake of comparison. It is apparent that low-temperature irradiation hardens pure copper, with an accompanying decrease in ductility. The data suggest that the radiation hardening in annealed copper has saturated at a dose of ~0.1 dpa. One interesting feature is that annealed and cold worked copper samples demonstrate nearly the same increase in yield strength following irradiation to ~1 dpa, i.e. $\Delta\sigma_y \approx 200$ MPa, though the final result for cold worked copper is $\sigma_y \approx 420$ MPa, and for annealed Cu it is $\sigma_y \approx 250$ MPa. The corresponding ductility for annealed copper is reduced by irradiation at ~100°C, with a typical irradiated uniform elongation of ~5-10% and total elongation of ~10-20%, respectively. For cold worked copper the embrittlement is more dramatic and the uniform elongation decreases to 0.2-1.1% after irradiation to ~1 dpa. The data in Fig. 1 suggest that radiation hardening of cold-worked copper is maximized at a damage levels of ~1 dpa. Irradiation of a cold-worked copper specimen to a damage level of ~5 dpa resulted in a lower strength and higher ductilty compared to 1 dpa, suggesting that radiation-enhanced dislocation recovery may be occuring at the higher dose. Additional data are needed to confirm the apparent high-dose (~5 dpa) softening behavior in cold-worked copper. Overall, these data suggest that the yield strengths of cold-worked and annealed copper irradiated at ~100°C are significantly different for damage levels below ~1 dpa, and tend to converge toward a common value at high damage levels ($\geq$ 5 dpa). Specimens of annealed and cold-worked copper irradiated in the Core position to a dose of 3 to 5 dpa at ~100°C exhibited good ductility, with uniform elongations of 9.5 to 19.5% and total elongations of 12 to 21%.

It is worth noting that a different tendency toward embrittlement was observed in the RF copper LTS and STS specimens irradiated in the annealed condition, most likely due to different procedures for specimen fabrication. The RF Cu STS specimens were made from a 1 mm sheet (from which the LTS samples were also cut out) that was cold rolled to a thickness of 0.2 mm with several intermediate anneals at 350°C for 1 h. It seems likely that the RF copper LTS specimens may not have fully recrystallized during preirradiation annealing at 350°C for 1 h, due to the low amount of cold-work in the as-wrought 1 mm sheet. As a result of the differences in processing history, the LTS samples of annealed RF copper hardened and embrittled more than the STS specimens. The 3 data points for annealed RF copper that are located outside of the shaded data bands in Figs. 1a and 1b correspond to the LTS specimens, and they exhibit tensile behavior that is intermediate between the annealed and cold-worked copper trend lines. It is also worth noting that the yield strength was slightly higher (and the uniform and total elongations were lower) for specimens irradiated in the unshielded Channel 4 capsule regions compared to Cd-shielded specimens (cf. Fig. 1). As discussed elsewhere [11], this implies that the higher concentrations of transmutation solutes in the unshielded capsules may enhance the residual radiation defect cluster density.

As demonstrated in Fig. 2a, the yield strength of Glidcop Al25 and MAGT-0.2 alloys irradiated at ~100°C increased by ~150 MPa at the lowest dose investigated (~0.8 dpa). Figure 2 also includes low-dose data on GlidCop Al25 obtained by Heinisch and coworkers [7,12]. The data in Fig. 2 indicate that radiation hardening in DS copper approaches a saturation level during irradiation at 36-100°C for damage levels above ~0.1 dpa.

The behavior of the uniform elongation of the irradiated DS copper alloys was similar to that of cold worked copper. The value of the uniform elongation decreased to 0.1 to 1% in specimens irradiated at ~100°C, with corresponding total elongations in the range of 0.2 to 4%. However, it should be noted that the relative degree of radiation embrittlement is considerably less in DS copper alloys; the degree of reduction in uniform elongation is about ten-fold for cold worked copper, whereas it is only a factor of 3 to 4 reduction for DS copper alloys. Fig. 2b summarizes the data base on dose-dependent uniform elongation of DS copper alloys irradiated at temperatures of 36-100°C [7,12]. It can be seen that severe embrittlement (δ_u~0.5%) occurs in DS copper for doses above 0.01 dpa at these low irradiation temperatures. The Cu-Cr-Zr radiation hardening behavior was very similar to that observed in the DS copper alloys. For example, irradiation of CuCrZr at 88°C to a dose of 1.5 dpa increased the yield strength to ~410 MPa, and the uniform and total elongation decreased to ~0.1%.

The load-elongation curves of the irradiated specimens exhibited varying behavior, depending on the specimen geometry and irradiation conditions. Yield drops were observed in the engineering stress-strain curves of the irradiated DS copper alloys, followed by low levels of work hardening. A slight yield drop was also observed in the annealed RF copper LTS specimens and cold-worked copper STS specimens irradiated in Channel 4 to ~1 dpa. On the other hand, yield drops were not observed in specimens which exhibited higher ductility values, in particular the annealed copper STS specimens irradiated in Channel 4 to ~1 dpa and both annealed and cold-worked copper irradiated in the Core position to 3-5 dpa. The post-yield work hardening rates were very low in all of the specimens irradiated at temperatures <150°C, which resulted in ultimate tensile strengths that were similar to the 0.2% yield strength.

The SEM investigations of the fracture character in unirradiated and irradiated samples revealed that annealed copper fractures in a transgranular manner (dimpled fracture surface) both in the initial and irradiated state after an appreciable local deformation (Fig. 3a,3b). The fracture surface of the irradited DS alloys appears more brittle, in particular necking in the irradiated samples is nearly absent (Fig. 3c). However, high magnification SEM observations on the irradiated DS copper fracture surfaces clearly indicate the prevalence of dimpled transgranular fracture (Fig. 3d).

4. Discussion.

The present results confirm previous observations that low temperature (~100°C) irradiation causes severe embrittlement in cold-worked copper [1] and high strength PH and DS copper alloys [4,6,7]. It is particularly noteworthy that cold-worked copper exhibits increased radiation hardening and embrittlement compared to annealed copper (Fig. 1). The presence of a yield drop followed by a low work hardening rate in the engineering stress-strain curves indicates that the low-temperature embrittlement results from localization of the intragranular deformation in the samples (i.e., "dislocation channeling") [13-16]. The extreme localization of the deformation associated with dislocation channeling would explain the low values of uniform and total elongations observed in the irradiated specimens. TEM investigations of the irradiated samples microstructure are in progress, and should provide additional information about the embrittlement mechanism.

Of special interest is the effect of irradiation temperature on the magnitude of radiation embrittlement. When plotting the generalized curves for the dependence of total and uniform elongations in DS and PH alloys on the irradiation temperature in Figs. 4, 5 (and considering previously published data [4,7,9]), it is apparent that irradiation of the PH and DS copper alloys at temperatures of 80-120°C produces uniform elongations of only 0.1 to 1.1% and corresponding total elongations of 0.1 to 3.9%. As demonstrated in Figs. 4, 5 and elsewhere [11], the radiation hardening decreases rapidly for irradiation temperatures >150°C, with a corresponding improvement in the uniform and total elongations. Previous work [17] has shown that the microstructure of copper does not vary to a significant degreee for irradiation temperatures between 50 and ~150°C, which agrees with the relative independence of the tensile properties on irradiation temperature in this range. Irradiation of copper at temperatures above ~150°C produces a rapid decrease in defect cluster density with increasing temperature [17], and therefore the radiation hardening and embrittlement effects become small. Since materials with uniform elongations <1% are classified as "brittle", the present results indicate that the operating temperature for the copper alloys in ITER should be maintained above 150°C unless the engineering design can accomodate brittle materials.

When using our data and those obtained in [5-7], it is possible to determine the generalized influence of dose and irradiation temperature on the embrittlement of PH and DS copper alloys. As seen in Fig 6, both the PH and DS copper alloys experience a significant loss in their total elongation at T_{irr}~100°C for doses above ~0.01 dpa, and the embrittlement appears to be nearly independent of dose in the range of ~0.01 to 10 dpa. At T_{irr}~150°C in the dose range of 0.1 to 10 dpa, the total elongation of irradiated PH and DS alloys was ~50% of the initial value. At T_{irr}>200°C (not shown in Fig. 6), the total elongation of the irradiated CuCrZr and DS alloys was more than 60% of the initial elongation up to doses of 10 dpa. The decrease in the uniform elongation following low temperature irradiation was even more pronounced than the decrease in the total elongation. Significant scatter is evident in the data plotted in Fig. 6, which implies that the radiation embrittlement has increased the sensitivity of the irradiated specimens to surface imperfections (notch acuity) and/or slight misalignments in the tensile specimen grips. The higher surface-to-volume ratio of the STS specimen geometry compared to the LTS geometry in the present study may have accentuated this sensitivity to surface imperfections; as demonstrated in Fig. 6b, the total elongations of GlidCop Al25 specimens irradiated at ~90°C were significantly lower for the STS geometry compared to the LTS geometry. However, there was no noticeable elongation difference between the STS and LTS geometries for specimens irradiated at ~150°C.

5. Conclusions

The present tensile results demonstrate that low temperature radiation embrittlement may have a considerable impact on the use of high-strength copper alloys for ITER structural applications. Low temperature radiation embrittlement is found to be very sensitive to the irradiation temperature. For PH and DS copper alloys, 150°C appears to be a critical temperature for manifestation of embrittlement. The high-strength alloys embrittle dramatically at T_{irr} <150°C, with uniform and total elongations close to zero. The plastic flow at the deformation is unstable under these irradiation conditions. At T_{irr}>150°C the PH and DS alloys have a satisfactory level of ductility, with irradiated elongations in the range of 50 to 90% of the unirradiated values.

For irradiation temperatures below 100°C, the radiation hardening and embrittlement becomes observable in high strength copper alloys at doses as low as ~0.001 dpa. The radiation hardening and embrittlement effects reach an apparent saturation condition for doses above ~0.1 dpa. These results suggest that high-strength copper alloys proposed for use in the ITER first wall and divertor should be operated at temperatures above 150°C in order to maintain satisfactory levels of ductility.

Acknowledgements

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Figure captions

Fig. 1. Effect of low temperature neutron irradiation on (a) yield strength and (b) total elongation of copper determined in this study and by Makin [1]. The open and filled symbols denote specimens that were annealed and cold-worked, respectively, prior to irradiation. The 3 open data points lying outside of the data bands are annealed RF Cu LTS specimens that did not recrystallize during annealing (see text).

Fig. 2. Effect of low temperature irradiation on the (a) yield strength and (b) uniform elongation of DS and PH copper alloys that were tensile tested at 100°C in the present study. Low-dose data by Heinisch and coworkers [7,12] that were tensile tested at 20°C are included for the sake of comparison.

Fig. 3. Fracture surfaces of (a,b) pure annealed (350°C) RF copper and (c,d) MAGT 0.2 dispersion strengthened copper, following irradiation at 85°C to ~ 1 dpa and tensile testing at 100°C. The width of the micrographs in Figs. 3b and 3c corresponds to ~40 µm.

Fig. 4. Effect of irradiation temperature (dose ~1 dpa) on the uniform elongation of Cu-Cr-Zr, according to data from the present study and refs. [4,6,8].

Fig. 5. Effect of irradiation temperature (dose ~1 dpa) on the uniform elongation of GlidCop Al25 and MAGT 0.2 (DS) alloys obtained from the present study and ref. [7].

Fig. 6. Dose dependence of the ratio of irradiated to unirradiated total elongation $\varepsilon_{irr}/\varepsilon_{unirr}$ of (a) CuCrZr and (b) DS copper alloys. The open symbols refer to irradiation and test temperatures below ~110°C, and the closed symbols correspond to temperatures of 150-200°C. The data were compiled from the present study and literature results [5-7].

Fabritsiev et al.
Fig. 1a

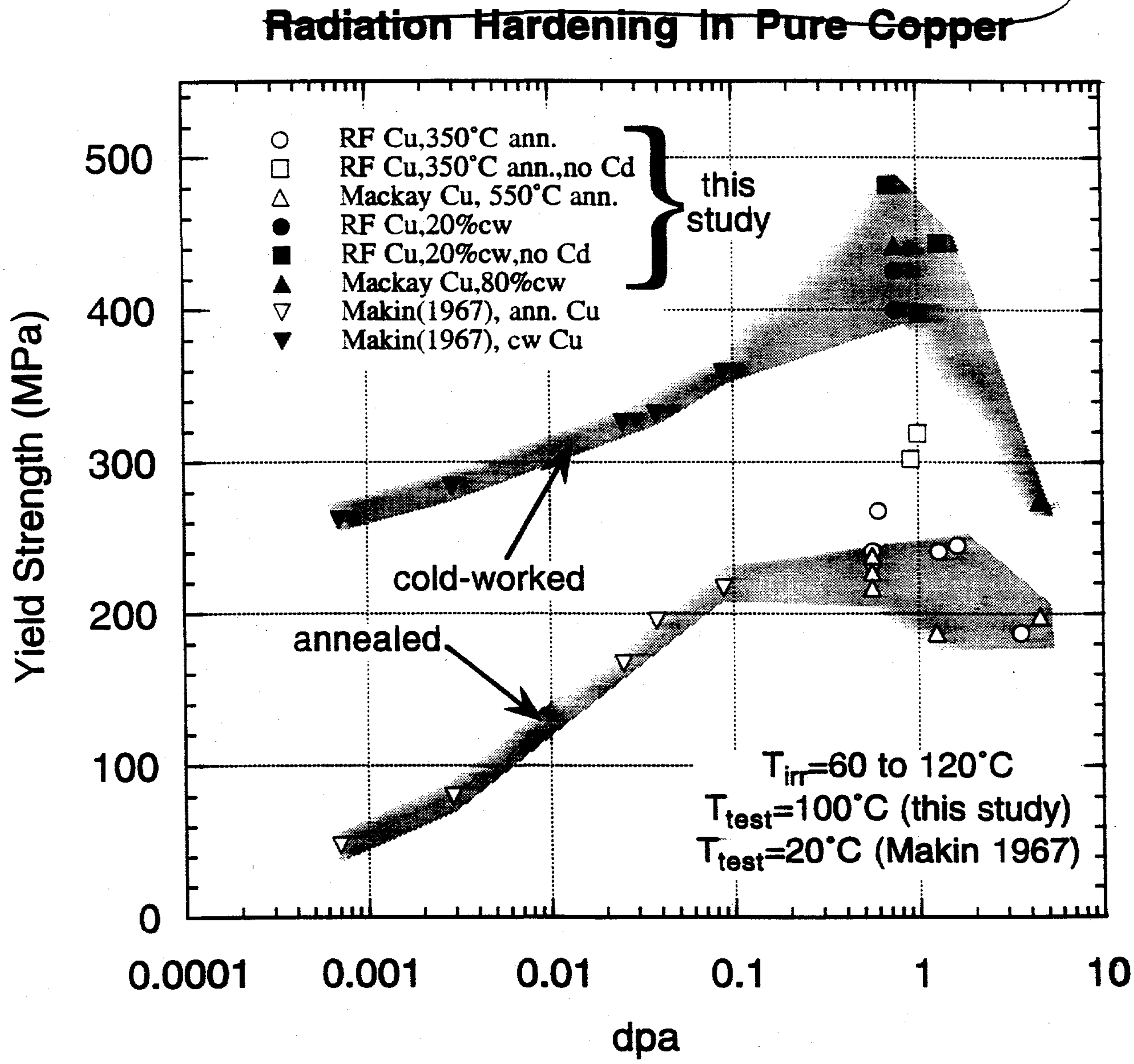

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Fig. 1b

~~Radiation Embrittlement in Pure Copper~~

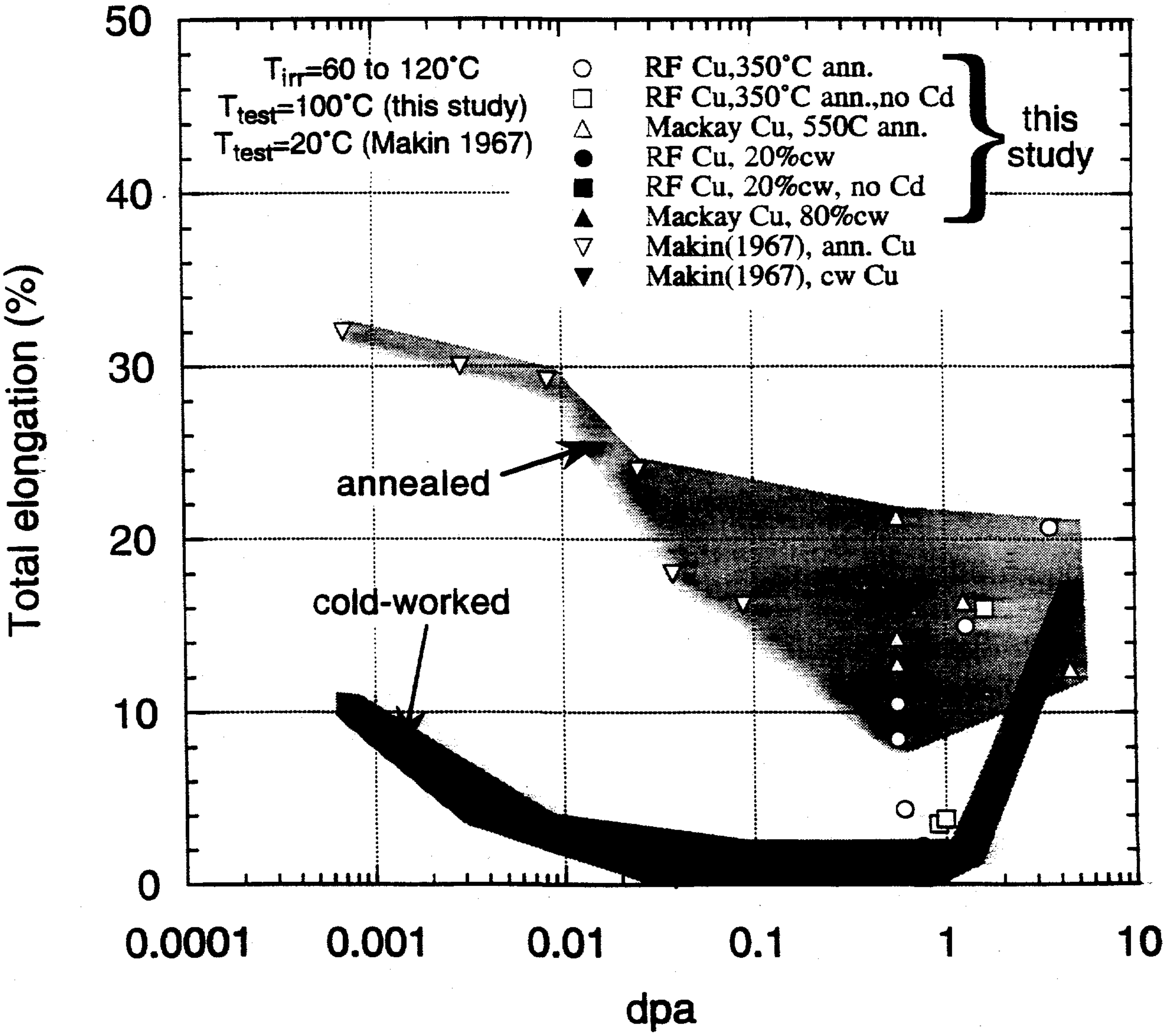

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Fig. 2a

~~RADIATION HARDENING IN NEUTRON-IRRADIATED OXIDE DISPERSION-STRENGTHENED COPPER ALLOYS~~

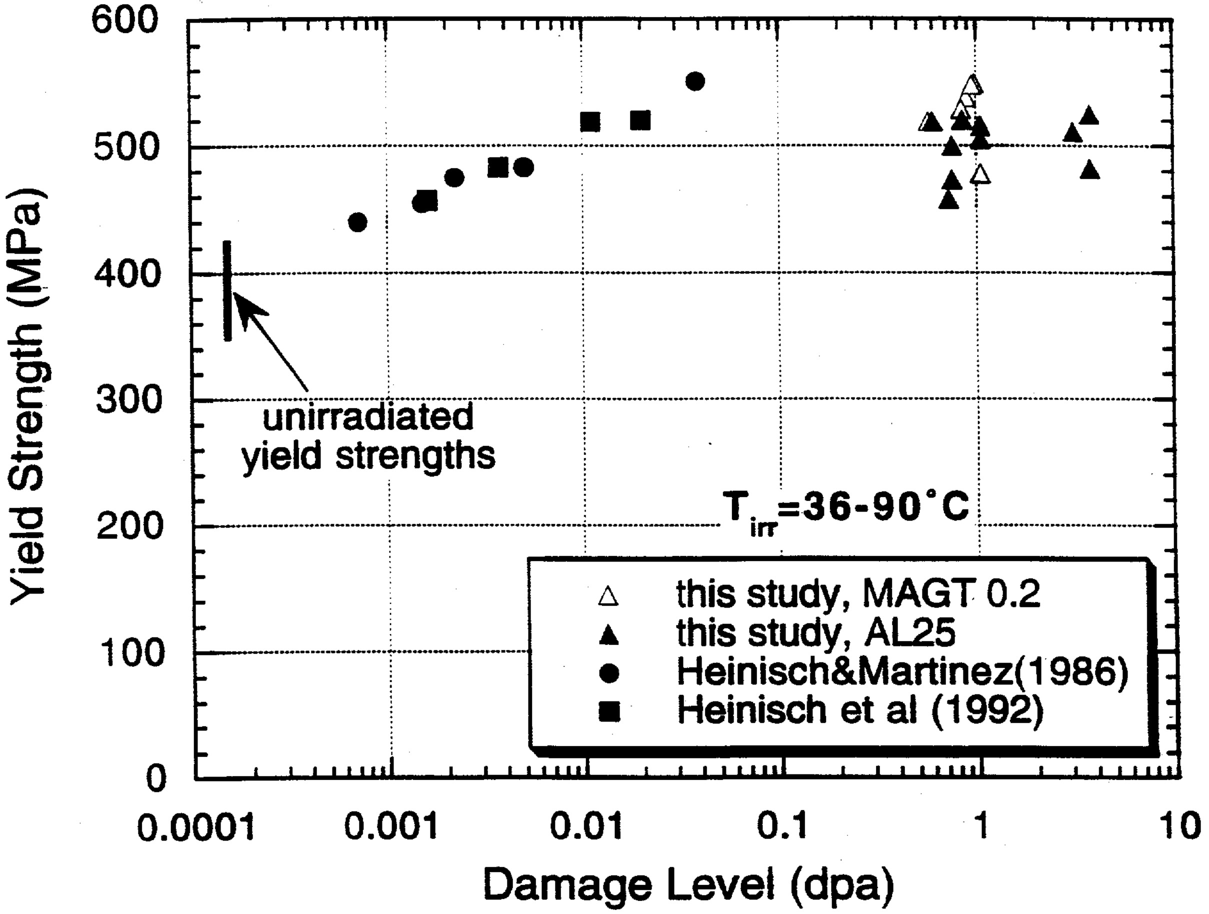

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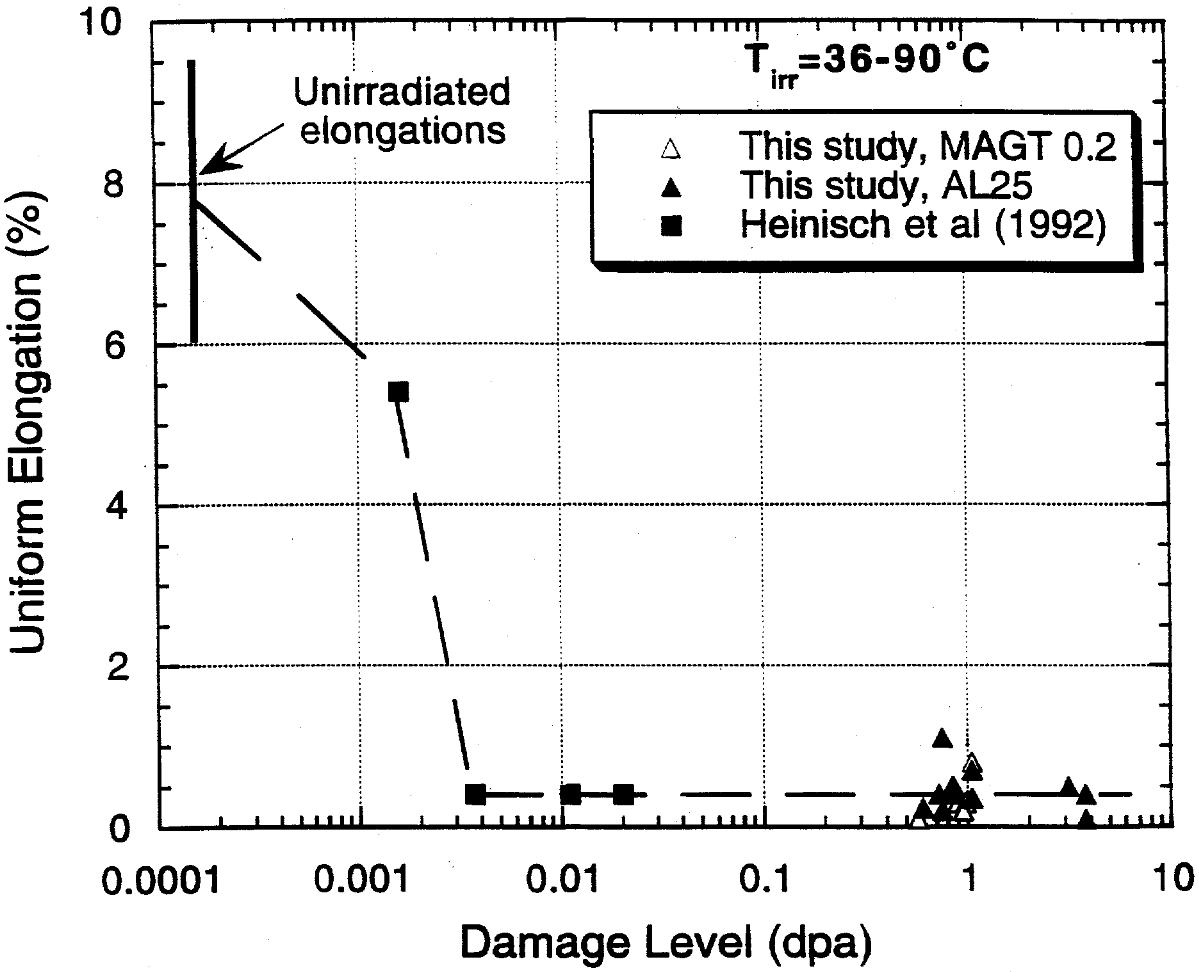
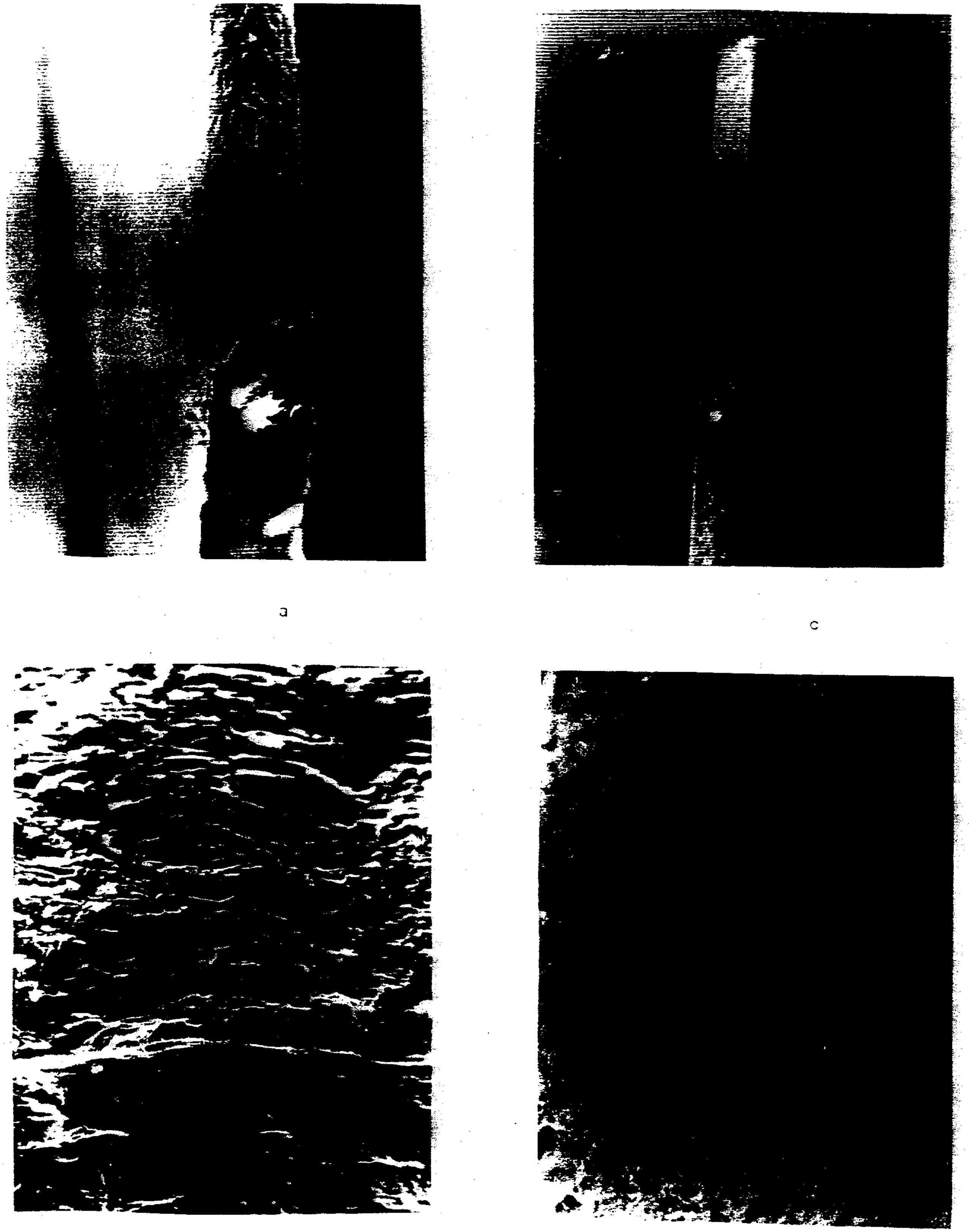
Fig. 2b

~~RADIATION EMBRITTLEMENT OF NEUTRON IRRADIATED OXIDE DISPERSION STRENGTHENED COPPER ALLOYS~~





a

c

b

d

Fig. 3

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Fig. 4

~~EFECT OF IRRADIATION TEMPERATURE~~
~~ON THE UNIFORM ELONGATION OF CuCrZr~~

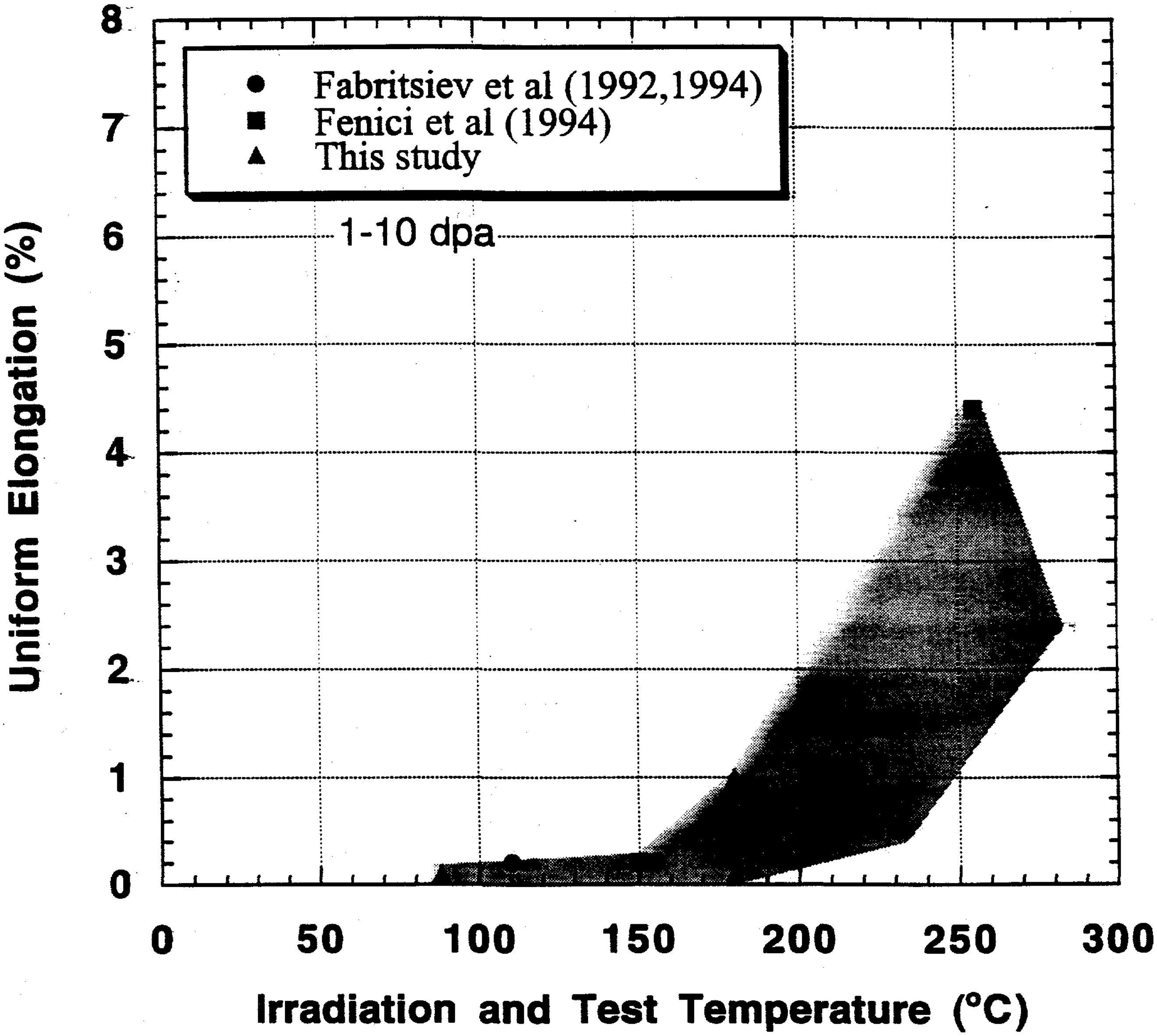

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Fig. 5

~~TEMPERATURE-DEPENDENT UNIFORM ELONGATION IN NEUTRON-IRRADIATED DISPERSION STRENGTHENED COPPER~~

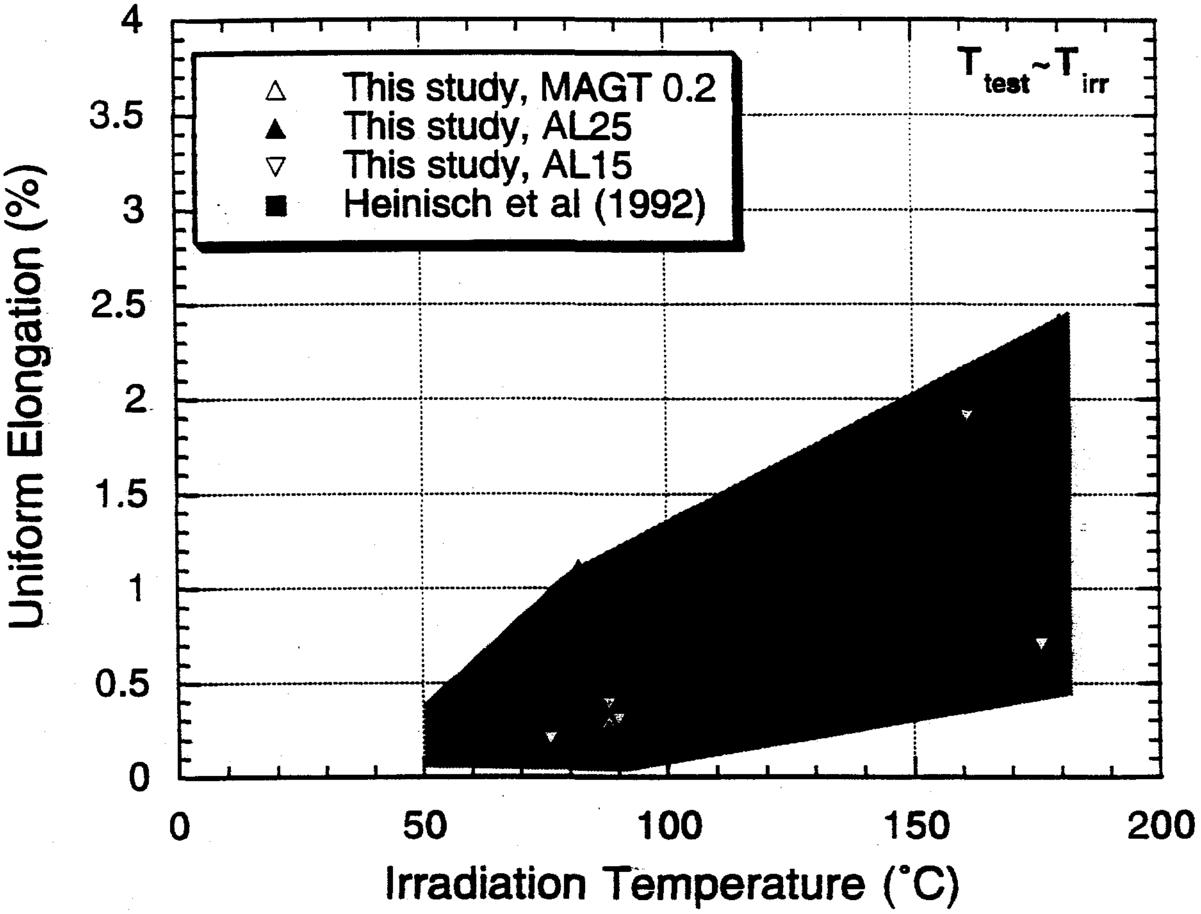

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Fig. 6a

~~EFFECT OF LOW TEMPERATURE IRRADIATION ON THE DUCTILITY OF CuCrZr ALLOYS~~

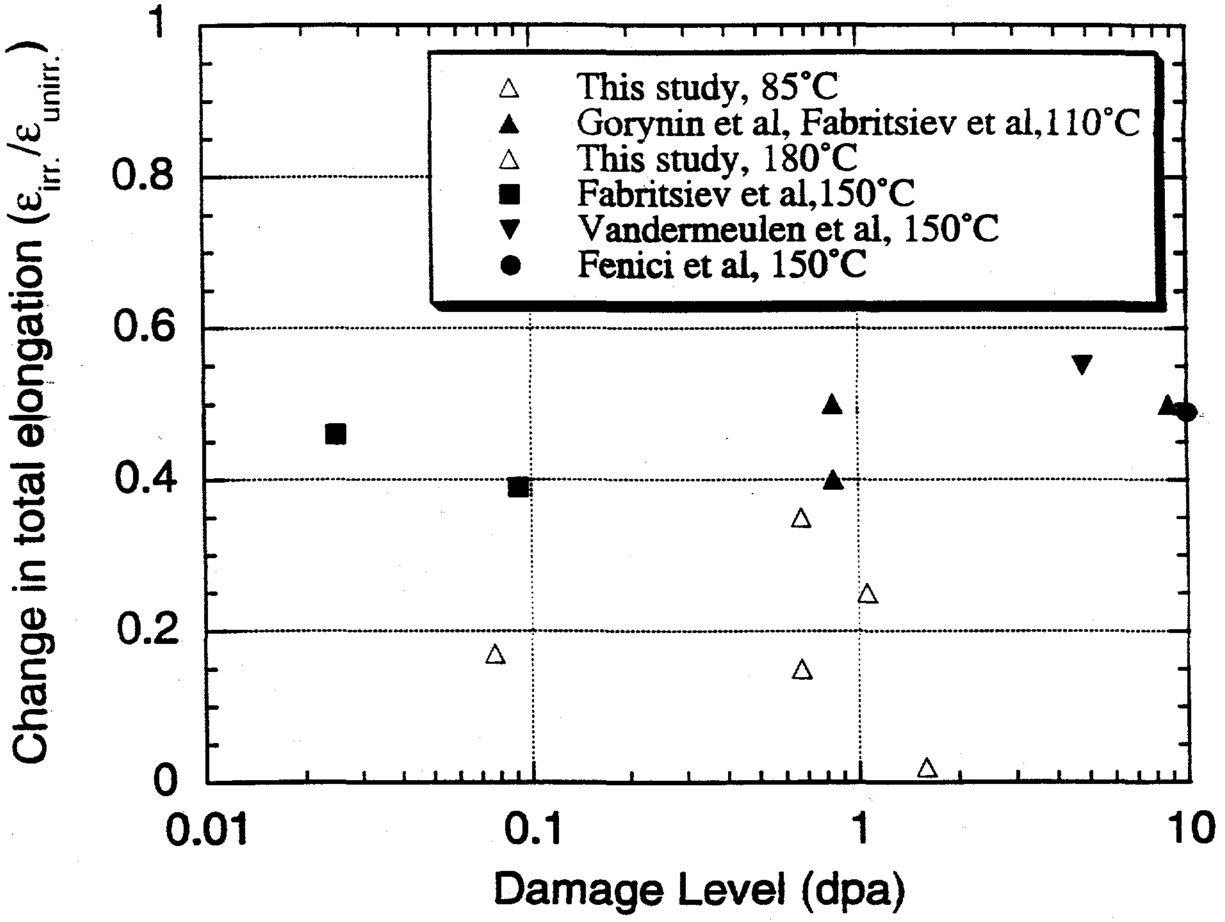

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Fig. 6b

~~EFFECT OF LOW TEMPERATURE IRRADIATION ON THE DUCTILITY OF DS COPPER ALLOYS~~