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A Brief Review of Cavity Swelling and Hardening in Irradiated Copper and Copper Alloys

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A BRIEF REVIEW OF CAVITY SWELLING AND HARDENING IN IRRADIATED COPPER AND COPPER ALLOYS*

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ABSTRACT: The literature on radiation-induced swelling and hardening in copper and its alloys is reviewed. Void formation does not occur during irradiation of copper unless suitable impurity atoms such as oxygen or helium are present. Void formation occurs for neutron irradiation temperatures of 180 to 550°C, with peak swelling occurring at ~320°C for irradiation at a damage rate of 2×10^7 dpa/s. The post-transient swelling rate has been measured to be ~0.5%/dpa at temperatures near 400°C. Dispersion-strengthened copper has been found to be very resistant to void swelling due to the high sink density associated with the dispersion-stabilized dislocation structure.

Irradiation of copper at temperatures below 400°C generally causes an increase in strength due to the formation of defect clusters which inhibit dislocation motion. The radiation hardening can be adequately described by Seeger's dispersed barrier model, with a barrier strength for small defect clusters of $\alpha \approx 0.2$. The radiation hardening apparently saturates for fluences greater than $\sim 10^{24}$ n/m² (~0.1 dpa) during irradiation at room temperature due to a saturation of the defect cluster density. Grain boundaries can modify the hardening behavior by blocking the transmission of dislocation slip bands, leading to a radiation-modified Hall-Petch relation between yield strength and grain size. Radiation-enhanced recrystallization can lead to softening of cold-worked copper alloys at temperatures above 300°C.

KEY WORDS: neutron, ion, irradiation, microstructure, tensile properties, void formation, radiation hardening, copper, copper alloys

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INTRODUCTION

Copper and its alloys have been the subject of numerous radiation damage studies, dating back to the 1950s [1,2]. Most of these studies were fundamental in nature and employed low damage levels. A large amount of our present understanding of the properties of point defects in metals, along with more applied aspects such as dispersed barrier strengthening in metals (radiation hardening), can be attributed to these early copper irradiation studies. Radiation effects in copper are also of technological interest because its high thermal and electrical conductivity make it useful for applications such as fusion reactors.

This paper reviews the current state of knowledge on radiation-induced swelling and hardening in copper and its alloys. The extensive literature on radiation-modified solute segregation and precipitation [1-4] will not be addressed directly. The effects of irradiation on the electrical properties of copper are also well studied [1,2], but will not be covered here. There have been numerous fundamental studies on radiation hardening conducted on copper and its alloys. This paper will concentrate on irradiation-induced changes in the tensile properties. Very little is known about the effects of irradiation on creep [5-7] or fatigue of copper.

There have been relatively few investigations on the performance of copper and copper alloys in high neutron irradiation environments. Figure 1 summarizes the temperatures and doses investigated for copper irradiated to neutron damage levels greater than 1 displacement per atom (dpa) [8-20]. It is worth noting that essentially all of these high fluence studies have been performed within the last five years. The most extensive investigation of copper at high damage levels was conducted by Brager and Garner and coworkers [14-18], where the microstructure, density changes, electrical resistivity, and tensile properties of copper and a range of solid solution, precipitation-strengthened, and dispersion-strengthened copper alloys were measured at temperatures $>400^{\circ}\text{C}$. The temperature dependence of void swelling and defect cluster formation over the temperature range of 182 through 500°C has been determined by Zinkle and coworkers [19,20]. There is a strong need for higher fluence data at irradiation

temperatures $\leq 350^\circ\text{C}$, where most of the applications occur for copper in high irradiation environments.

VOID SWELLING IN COPPER AND COPPER ALLOYS

It was noticed shortly after the discovery of void swelling in irradiated metals by Cawthorne and Fulton [21] that impurity gases, in particular oxygen and helium, could exert a significant influence on void formation [22,23].

Oxygen effects on void formation

A series of neutron, ion and electron irradiation experiments by a group of French scientists [23-26] demonstrated that preimplanted or residual impurity oxygen could have a dramatic effect on void swelling in copper. Void formation was not observed in "degassed" copper foils subjected to ion [24,26] or electron [25] irradiation at 450°C , and void annealing studies on oxygen preimplanted specimens showed that the surface energy could be reduced to nearly half of the pure copper value for oxygen contents of ~ 100 wt ppm [27].

Several recent studies have confirmed and quantified the influence of small concentrations of oxygen on void formation in copper [28-30]. In a high fluence ion irradiation study [28], void formation was not observed in high-purity, low-oxygen copper over the broad temperature range of 100 through 500°C . Figure 2 shows the low-magnification microstructures of some of the irradiated copper specimens from this study. Conversely, ion irradiation of copper containing 360 appm O (introduced by annealing high-purity copper in inert gas containing a trace amount of impurity oxygen) resulted in copious void formation at 375 and 475°C [29]. Figure 3 shows an example of the void formation observed in ion irradiated oxygen-bearing copper. Theoretical analyses [30-32] have found that the void is generally not thermodynamically stable in pure copper in comparison with the vacancy loop or stacking fault tetrahedron (SFT). A model based on oxygen chemisorption on void nuclei surfaces (which causes a reduction in surface energy and hence can stabilize void formation) is in reasonable agreement with

experimental observations on the effect of oxygen on void formation [29]. Figure 4 shows a quantitative comparison of the predicted and observed effect of oxygen on void formation (open symbols indicate void formation was observed, and filled symbols indicate no void formation occurred).

The charged particle studies have proved to be important for understanding the importance of impurity atoms on void formation. It is clear that impurities such as oxygen need to be controlled in order to minimize the propensity of copper for void swelling. The strong role of impurity oxygen has also been recently demonstrated in a neutron irradiation experiment [13] on electrolytic tough pitch (ETP) copper, which was of nominally high purity but contained about 300 wt ppm O (1200 appm O). The oxygen-bearing ETP copper swelled at a rate that was four times larger than that of oxygen-free copper (34% vs. 8% swelling after 13.5 dpa at 400°C).

Helium effects on void formation

There have been several experimental studies on the effects of helium on cavity formation in copper [20,24,26,33-41], but further information is needed at fusion-relevant conditions (~7 appm He/dpa). Most of these studies either preinjected helium [24,26,33,34,41] or produced helium via thermal reactor irradiation of boron-doped copper using very large He/dpa ratios [20,36-40]. Both methods are unsatisfactory for determining the effect of helium in fusion reactor environments. Figure 5 shows an example of substantial cavity swelling that occurred during simultaneous irradiation of copper with helium and heavy ion beams [35]. Void formation was not observed in specimens that were not exposed to the helium implantation. According to experimental results and theoretical calculations, the amount of helium needed to stabilize cavity formation in pure oxygen-free copper is very small--on the order of <1 appm He [26,42].

Several researchers have irradiated dilute Cu-B alloys in order to probe helium bubble formation in neutron-irradiated copper [20,36-40]. The reaction $^{10}\text{B}(n,\alpha)^7\text{Li}$ produces both helium and lithium during irradiation in a thermal neutron environment, both of which are essentially insoluble in copper. It has been observed that the cavity formation in boron-doped copper is due to the helium produced by the (n,α) reaction

(as opposed to the lithium) [37]. These studies have provided useful information on the thermal stability of helium bubbles and on the conversion of bubbles to voids.

Preinjection of small amounts of helium (up to 100 appm) enhances void formation in pure copper during irradiation to moderate damage levels (~20 dpa) [33]. However, preinjection of large amounts of helium (2000 appm) apparently overstimulates void nucleation and suppresses observable cavity swelling [41]. As discussed later, there is relatively little information available on helium effects in swelling-resistant copper alloys.

Effect of temperature and fluence on swelling

Copper has been found to contain voids following neutron irradiation at temperatures between 180 and 550°C (0.33 to 0.60 T_m) [1,8,9,11-20,23,43-52]. The temperature dependence of the cavity swelling of copper following low dose neutron irradiation [19,23,45,46] is similar to that observed in many other metals: The void size increases gradually with increasing temperature while the cavity density decreases rapidly, which results in a swelling peak at intermediate temperatures (~300 to 325°C for copper). Figure 6 shows the measured swelling as a function of irradiation temperature for copper irradiated to a damage level of ~1 dpa [19,20]. The measured temperature-dependent cavity swelling was similar for both pure copper and a helium-containing dilute Cu-B alloy. This is interpreted as evidence for a very small incubation fluence for void swelling due to the presence of large amounts of impurity oxygen in both materials (according to conventional radiation damage theory, helium has only a minor effect on the post-transient cavity swelling rate).

The primary effect of a variation in the damage rate on swelling is to shift the peak swelling temperature. An order of magnitude decrease in the neutron flux (from 2×10^7 to 2×10^8 dpa/s) decreased the location of the peak swelling temperature by ~20°C [45,46]. The peak swelling temperature shift between neutron (2×10^7 dpa/s) and ion (1×10^3 dpa/s) irradiated copper is ~165°C [24,45].

Voids have been detected in copper following irradiation to fluences as low as 1×10^{21} n/m² [48,49]. The swelling behavior at low doses (<0.2 dpa) is often nonlinear, and transient swelling rates in excess of 1%/dpa have been observed in some studies of

neutron-irradiated copper [44,49]. The void formation is often very heterogeneous for these low dose irradiations [48,49,52]. Higher dose neutron and electron irradiation studies have found that a steady state void swelling rate of 0.4 to 0.9%/dpa eventually occurs, with no evidence for saturation up to swelling levels in excess of 60% [11-16,33,53,54]. Some of the electron irradiation studies suggest that the steady state swelling rate is temperature-dependent [33,54], but these observations might also be attributable to thin foil effects. Figure 7 shows the fluence-dependent swelling observed in copper following neutron irradiation at $\sim 420^{\circ}\text{C}$ [14,15]. The transition fluence for void swelling in irradiated pure copper is apparently very small, ~ 1 dpa [14,15,33,41,53].

Influence of nongaseous solutes on void swelling

With the notable exceptions of Ag, Al, and Cd [33,41,55], the addition of alloying elements to copper generally results in a reduction of void swelling. The primary influence of solute additions appears to be an extension of the transient regime of void swelling (as opposed to a reduction in the steady state swelling rate) [14,15], although further studies are needed to confirm this finding. Reduced swelling compared to pure copper has been reported for solute additions of Au, Be, C, Fe, Ge, In, Mg, Ni, Pt, Sb, and Si [14,15,23,26,33,34,41,45,48,54-59]. Figure 7 compares the fluence-dependent swelling behavior of Cu-5%Ni with pure copper following neutron irradiation near 420°C [14-16]. It can be seen that the addition of nickel suppressed swelling at low doses (16 dpa), but had little effect on the swelling behavior at high doses.

Several potential physical mechanisms have been proposed to explain the observed suppression or enhancement of void swelling in solid solution copper alloys. Radiation-induced segregation of the solute atoms toward or away from void embryos could modify the surface energy of the void nuclei, and thereby enhance or suppress void swelling. Solute trapping effects could enhance the recombination rate of point defects and thereby suppress void swelling, but most substitutional solutes are not effective interstitial traps at temperatures above 100 K [60,61]. On the other hand, interstitial solutes could impede the migration of irradiation-produced interstitials and thereby increase the probability of vacancy-interstitial recombination. Reductions in

thereby increase the probability of vacancy-interstitial recombination. Reductions in stacking fault energy due to solute additions should in principle make other vacancy cluster morphologies favorable compared to the void [23,30,45,48]. However, experimental results on copper alloys of low stacking fault energy do not show a direct correspondence with a reduction in void swelling [59]. Further study in this area would be obviously worthwhile, but solid solution alloying does not appear to be useful for preventing cavity swelling at high doses in copper.

Void swelling in precipitation- and dispersion-strengthened Cu

The presence of a second phase in copper results in better swelling resistance compared to pure copper and solid solution copper alloys. Irradiation studies have been performed on aged Cu-Co, Cu-Be, Cu-Fe, Cu-Cr, Cu-Ti, and Cu-Zr binary alloys along with more complex alloys such as Cu-Cr-Zr, Cu-Be-Ni, Cu-Ni-Sn, and Cu-Ni-Ti [11-18,41,53,62-66]. Good swelling resistance (<2% swelling) has been observed in Cu-2%Be and Cu-3.5%Ti during fast reactor irradiation near 420°C to doses of 34 to 50 dpa [14-16]. All four of the ternary alloys studied to date show reasonable swelling resistance during high-fluence fast reactor [11-16,18] or dual-ion [64] irradiation, although the results are dependent on the initial thermomechanical treatment. For example, Cu-Ni-Be has been shown to be very swelling resistant during dual-ion [64] irradiation to 20 dpa at 250 to 500°C (30 appm He/dpa) and during fast reactor irradiation at 410°C to 34 dpa [16-18]. Conversely, fast reactor irradiation to 48 dpa at 430°C produced swelling in excess of 12% for cold-worked and aged Cu-Ni-Be [14,15]. Lower swelling levels were observed in an annealed and aged Cu-Ni-Be alloy. Tensile [15] and TEM [66] measurements revealed that recrystallization along with precipitate coarsening had occurred in the cold-worked alloy during irradiation at 430°C, and this led to the swelling instability.

Several studies have demonstrated that oxide dispersion-strengthened copper alloys containing inert particles of Al_2O_3 , TiO_2 , HfO_2 or ZrO_2 are swelling resistant [11-16,18]. Dispersion-strengthened Cu-Cr₂O₃ has poor swelling resistance relative to Cu-Al₂O₃, etc. [16], and has poor strength at elevated temperatures [13]. The Cu-Al₂O₃ dispersion-strengthened alloys have received particular attention due to their excellent

530°C [11-16,18] and during dual ion beam irradiation [64]. Substantial void swelling has been observed in one series of commercial oxide dispersion-strengthened copper alloys following high-fluence neutron irradiation due to the presence of excess oxygen in the matrix [16,18].

It appears that the swelling suppression in precipitation- and dispersion-strengthened copper is partly due to point defect trapping at the precipitate-matrix interface, but is primarily due to the presence of a high dislocation density. The swelling resistance is generally best in alloys that contain a stabilized cold-worked structure due to the additional point defect sink strength supplied by the dislocations. During high fluence ion [64] or neutron irradiation [12,66], void formation is initiated in regions of the matrix that have recrystallized (leaving a low defect sink density). A large part of the swelling resistance of dispersion-strengthened alloys can be attributed to their superior resistance to recrystallization (precipitate overaging does not occur since the dispersion is inert in the copper matrix). Figure 8 summarizes the swelling behavior of three grades of a commercial dispersion-strengthened copper, GlidCop, following neutron irradiation near 410°C [15]. The swelling resistance of the GlidCop alloys improved with increasing dispersoid content and cold-work level.

There is a strong need for irradiation data on swelling-resistant copper alloys at fusion-relevant He/dpa ratios. Fission reactor irradiation of copper generates only ~0.2 appm He/dpa, which is more than one order of magnitude smaller than the DT fusion first wall generation rate for copper of ~7 appm He/dpa. Spitznagel et al. [64] found that dispersion-strengthened copper had about five times higher swelling following dual-beam irradiation at 30 appm He/dpa as compared to specimens irradiated without helium coimplantation. On the other hand, Cu-Ni-Be specimens did not exhibit observable cavity swelling after either dual-beam or single ion-beam irradiation to 20 dpa. The relatively high swelling that was observed in the GlidCop Al-15 alloy compared to the Al-20 and Al-25 alloys following neutron irradiation (Fig. 8) may have been partially enhanced by the presence of a small amount of residual boron (<200 wt ppm) remaining from a preirradiation deoxidation treatment. A boron content of 100 wt ppm would transmute

during high-fluence fast reactor irradiation to produce ~100 appm He when fully burned up.

RADIATION HARDENING

Radiation hardening in pure copper

Numerous fundamental studies on radiation hardening have been conducted on pure copper over the past 40 years [67-75]. Significant amounts of hardening occur in annealed copper for irradiation temperatures $\leq 350^\circ\text{C}$. It has been demonstrated [72,73] that the strength increase is due to "black spot" formation (small dislocation loops and SFT). The yield strength increase is described by Seeger's [67] dispersed barrier model,

$$\Delta\tau = \alpha Gb\sqrt{Nd} \quad (1)$$

$$\Delta\sigma_y = \bar{M}\Delta\tau \quad (2)$$

where G is the shear modulus, b is the magnitude of the dislocation Burgers vector ($a_c/\sqrt{2}$ for copper), N and d are the density and mean diameter of the defect clusters, α is the barrier strength coefficient, and \bar{M} is the Taylor factor ($\bar{M} = 3.06$ for fcc polycrystals) which describes the relationship between shear stress ($\Delta\tau$) and polycrystalline yield stress ($\Delta\sigma_y$) [75]. Various experimental and theoretical studies have found that the barrier strength coefficient for defect clusters formed during room temperature irradiation of copper is $\alpha \sim 0.2$ to 0.25 [73,75,76].

The fluence dependence of radiation hardening in copper was the subject of a long-standing controversy [73], with different research groups claiming that the strength increase was proportional to the one-third power of fluence [68] or the square root of fluence with a saturation term [71,72]. It was also recently suggested that the hardening was best described by a one-fourth root dependence over an extended fluence range [77-80].

Part of the cause of the controversy can be resolved by examination of recent TEM studies on the fluence dependence of the defect cluster density in neutron-irradiated copper. Figure 9 shows that the defect cluster density in copper irradiated near room temperature [80-86] is initially proportional to the fluence up to $\phi t \sim 5 \times 10^{20} \text{ n/m}^2$. This produces a strength increase proportional to $\sqrt{\phi t}$ in this fluence regime since the defect cluster size was essentially independent of fluence [Eq. (1)]. The fluence dependence of the cluster density at higher fluences is strongly dependent on the purity content of the irradiated copper (in particular interstitial impurities) [84]. Several studies on high-purity copper and other fcc metals have demonstrated that there is a substantial fluence range where the defect cluster density is proportional to $(\phi t)^{1/2}$ [77,80,87,88], which would produce a one-fourth root dependence of hardening on fluence [Eq. (1)]. This has been interpreted as evidence for the interaction of freely migrating defects with defect clusters from surrounding displacement cascades, causing shrinkage and annihilation of some clusters. Simple theoretical models predict a square root dependence on fluence for the defect cluster density when migrating defect interactions with defect clusters are occurring [60,77,80,87]. Copper containing moderate amounts of interstitial impurities does not show a deviation from the linear accumulation of defect clusters until $\phi t > 5 \times 10^{21} \text{ n/m}^2$. This suggests that the impurities are blocking migration of the freely migrating interstitials. Hence, irradiation of nominally pure copper specimens containing differing amounts of interstitial impurities will produce differing radiation hardening rates over certain fluence ranges.

For defect cluster densities $> 10^{23}/\text{m}^3$, there is an increasing probability of cascade overlap events, i.e., a displacement cascade occurs in the vicinity of an existing defect cluster and causes its annihilation with the result that there is no net increase in defect cluster density. The TEM measurements (Fig. 9) suggest that the maximum defect cluster density in copper irradiated with neutrons near room temperature is $\sim 10^{24}/\text{m}^3$. Higher fluence studies are needed to confirm the saturation density of defect clusters. A saturation in the radiation hardening on neutron-irradiated copper has similarly been observed [89-92] at fluences $\sim 10^{24} \text{ n/m}^2$ ($\sim 0.1 \text{ dpa}$). Figure 10 shows the yield strength of neutron irradiated copper as a function of fluence [89]. In this study [89] an apparent

saturation in hardening already occurred at a fluence of 10^{23} n/m² ($E > 1$ MeV). A similar saturation in hardening also has been observed in ion-irradiated copper [93].

An interesting observation that is deserving of further study concerns the effect of grain size on the radiation hardening rate. Several studies have shown that the strength increase as a function of fluence is higher for small-grained compared to large-grained copper [70,94]. Figure 11 shows an example of the dependence of yield strength on grain size for neutron-irradiated copper [70]. It has been suggested that this effect is due to grain boundaries blocking the transmission of slip bands [75]. It is not clear whether this grain size-dependent radiation hardening rate continues at higher fluences.

Temperature dependence of radiation hardening

There have been extensive studies on the temperature dependence of radiation hardening in pure copper utilizing isochronal and isothermal annealing [70,71,73]. The flow stress in irradiated copper decreases with increasing irradiation temperature due to two factors. First, the barrier for dislocations to shear defect clusters is thermally activated. In addition, the defect clusters responsible for the radiation hardening of copper are thermally unstable at elevated temperatures. Figure 12 shows the temperature-dependent defect cluster density observed in copper following ion irradiation to a damage level of 10 dpa [28]. Similar results have been obtained in other studies of neutron- [48,95] and ion-irradiated [96] copper. The room temperature strength of ion-irradiated copper also shows a similar dependence on irradiation temperature [93].

The defect cluster density that is produced during irradiation at elevated temperatures is shown in more detail in Figure 13 [20]. The defect cluster density decreases by over 3 orders of magnitude between the irradiation temperatures of 182 and 450°C. The results demonstrate that the defect clusters responsible for radiation hardening are highly sensitive to irradiation temperatures $> 182^\circ\text{C}$. The decreased cluster density at elevated temperatures is a further indication that the defect clusters are thermally unstable, although additional factors such as the temperature dependence of the production of defect clusters directly in displacement cascades may also be playing a role.

At high fluences, some hardening from cavity formation is expected to occur [97]. However, experimental results indicate that the yield strength of irradiated copper decreases when void swelling becomes very high, due to a lack of load-bearing area [98].

A loss of ductility is associated with radiation hardening. Figure 14 shows the total elongation of annealed copper is rapidly reduced by neutron irradiation [70], and that cold work shortens the fluence required to reach the embrittled state ($\epsilon_{tot} < 2\%$). Confirmation of this cold-work effect on radiation effects is needed.

Other studies have shown that copper becomes severely embrittled in the presence of large cavities, introduced by the tritium trick [99,100] or neutron irradiation [98]. Helium embrittlement effects have also been suggested in some Cu-B irradiation studies [38], but more data are needed.

Alloying effects on radiation hardening

High-strength copper alloys are desirable for several applications in fusion reactors. Their strength is generally achieved by classical precipitation hardening, or by pinning a dislocation structure with precipitates or an inert dispersed phase such as Al_2O_3 particles. Radiation-enhanced recrystallization is an important phenomenon in cold worked copper alloys at irradiation temperatures $\geq 300^\circ C$ [10,12,64,65,101,102]. The enhanced diffusion associated with irradiation can accelerate the dislocation recovery and grain recrystallization processes, particularly if the dislocation structure is not pinned effectively. A substantial loss of strength associated with the recrystallization generally occurs in alloy systems whose strength is derived from a cold-worked structure that is stabilized by precipitates [103]. In addition, established phenomenon such as radiation-enhanced precipitate overaging or loss of coherency [1,63] in classic precipitation-hardened systems such as Cu-Be should cause a large strength decrease. These softening effects are offset to a certain degree at low irradiation temperatures ($< 300^\circ C$) by the increased hardening associated with defect cluster formation [10,65,101,102].

The most promising radiation-resistant copper alloy system from both a cavity swelling and retained strength perspective is dispersion-strengthened copper. The

commercial dispersion-strengthened alloys GlidCop Cu-Al₂O₃ have been found to be resistant to radiation-enhanced recrystallization for irradiation temperatures as high as 420 to 530°C and doses as high as 100 dpa [13,14,17,18,66]. (However, one study [17] reported that neutron irradiation at 410°C to 50 dpa caused substantial recrystallization in two GlidCop Cu-Al₂O₃ alloys.) The resistance to recrystallization may be attributed in large part to the chemical inertness of the Al₂O₃ particles, along with a high dislocation barrier strength for the particles that inhibits particle shearing. One concern is the possibility of radiation-induced dissolution of the Al₂O₃ particles during high-fluence irradiation due to displacement cascade effects (ballistic dissolution) as observed by Spitznagel et al. [54]. However, several experimental [12,63,66,104] and theoretical [105,106] studies on Cu-Al₂O₃ and other copper alloys have found that particles of sizes appropriate for dispersion strengthening ($d \geq 10$ nm) are stable during irradiation. Since the particle dissolution rate is dependent on temperature, dose rate and displacement cascade size, further study is warranted.

CONCLUDING REMARKS

Examination of the existing data base for irradiated copper shows that the irradiation temperature determines whether cavity swelling or radiation hardening effects will occur. Figure 15 shows in schematic form the temperature regimes that are important for cavity swelling and radiation hardening of neutron-irradiated copper. Radiation hardening effects are significant for temperatures <300°C, and the saturated defect cluster density is nearly constant for temperatures <180°C. The decrease in the defect cluster density at temperatures >180°C allows cavity growth to become appreciable due to the decrease in sink density. The amount of void swelling becomes small at temperatures >500°C due to thermal annealing of voids.

The strength of copper and copper alloys can be either increased or decreased by irradiation, depending on the cold-work level of the material and the irradiation temperature. Strengthening due to defect cluster formation is appreciable for irradiation temperatures <300°C. The strength increase associated with defect cluster formation in copper apparently saturates during neutron irradiation near room temperature at a dose

of ~ 0.1 dpa. Radiation-enhanced recrystallization and accelerated precipitate overaging can lead to a decrease in strength in cold-worked or precipitation-hardened systems for irradiation temperatures $> 300^\circ\text{C}$.

Cavity swelling during irradiation is of concern chiefly for irradiation temperatures of 200 to 500°C . Small amounts of swelling may occur at temperatures as low as 180°C and as high as 550°C . The swelling peak at low doses occurs at $\sim 320^\circ\text{C}$. It is apparent that small amounts of impurities (either gaseous or solid) are essential for the formation of cavities during irradiation. Only a small amount of oxygen (~ 10 appm) or helium (~ 1 appm He) is needed to stabilize cavities in pure copper. Deleterious matrix solutes such as oxygen should be minimized through careful control of the manufacturing process in order to produce materials suitable for high radiation environments.

There is a strong need for high fluence studies on copper alloys at irradiation temperatures $\leq 350^\circ\text{C}$ in order to generate data on tensile property changes, cavity swelling resistance, and microstructural changes. Additional low dose studies would be useful for understanding the details of the dose dependence of the defect cluster density (including impurity effects) and other matters of fundamental interest, including the effect of grain size on radiation hardening. In addition, irradiation data are required on fatigue and creep behavior, and the effects of irradiation on brazed or welded joints. Data are needed on the influence of helium on cavity swelling in swelling-resistant materials such as dispersion-strengthened Cu- Al_2O_3 alloys. From a design standpoint, it is not particularly useful to study He effects in pure copper, since He should have little influence on the swelling behavior of materials that swell readily in the absence of helium.

Dispersion-strengthened alloys such as GlidCop Cu- Al_2O_3 are the most promising class of commercially available copper alloys in terms of radiation resistance. The radiation resistance of this material can be attributed to its very high sink density (dislocations plus Al_2O_3 particles). Unlike precipitates, the high density ($> 10^{22}/\text{m}^3$ of small ($d \sim 10$ nm) inert Al_2O_3 particles) are resistant to overaging and thermal dissolution effects during irradiation. Additional high-dose studies are needed to confirm the absence of appreciable displacement cascade dissolution of particles in dispersion-strengthened

copper. Similar dispersion-strengthened alloys based on other alloy systems (e.g. Fe or Ni matrix) would be worthy of further investigation for fusion reactor structural applications. Precipitation-strengthened alloys such as Cu-Cr-Zr or Cu-Ni-Be may be suitable for high-fluence applications at temperatures where radiation-enhanced recrystallization is not of concern (<300 to 350°C).

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FIGURE CAPTIONS

Fig. 1. Summary of high dose neutron irradiation experiments on copper and copper alloys [8-20].

Fig. 2. Cross-section microstructure of pure copper following 14-MeV Cu^{3+} ion irradiation to a peak damage level of 40 dpa at 100 through 400°C [28]. The original surface lies to the left in all of the micrographs, and the measured ion damage range was about 3.5 μm .

Fig. 3. Cross-section microstructure of copper containing 360 appm O following 3.6 MeV Fe^{++} ion irradiation to a peak damage level of 17 dpa at 475°C [29]. The ion damage range was about 1.3 μm . The very small circular and large triangular features are due to surface contamination of the TEM foil.

Fig. 4. Comparison of predicted [29] and observed [25,26,28,29] void formation in oxygen-bearing copper. Filled symbols indicate no void formation was observed, and open symbols indicate voids were observed. Arrows indicate the possibility of higher or lower oxygen concentrations due to uncertainties in the oxygen measurement.

Fig. 5. Cross-section microstructure of copper following simultaneous dual beam irradiation at 440°C with 4 MeV Fe^{++} ions and 200 to 400 keV He^+ ions. The peak irradiation damage level was 16 dpa at a depth of about 1 μm and a uniform He concentration of 48 appm was injected at depths of 0.5 to 1.1 μm [35]. The calculated damage level at a depth of 0.5 μm was ~10 dpa, and the cavity swelling was ~3.5%.

Fig. 6. Temperature-dependent cavity swelling in copper and Cu-18 wt ppm ^{10}B following fission neutron irradiation to a damage level of $\sim 2 \times 10^7$ dpa/s [19,20]. The irradiation produced ~100 appm He in the Cu-B alloy.

Fig. 7. Comparison of the swelling behavior of copper and a solid solution Cu-5%Ni alloy following fast reactor irradiation at temperatures near 400°C [14-16].

Fig. 8. Swelling behavior of three commercial dispersion-strengthened copper alloys following fast reactor irradiation at 411 to 414°C [15]. The alloys contained 0.15, 0.20, and 0.25 wt % Al in the form of Al_2O_3 particles. The Cu-Al15 alloy contained <200 wt ppm B due to a preirradiation boron deoxidation treatment.

Fig. 9. Summary of recent high-resolution TEM studies on defect cluster formation in copper irradiated near room temperature. Data are shown for 14 MeV neutrons [80,82,84-86], fission neutrons [81,83,86], and 800 MeV protons [86].

Fig. 10. Measured yield strength of polycrystalline copper following fission reactor irradiation near room temperature [89].

Fig. 11. Yield strength of polycrystalline copper versus the epithermal fission reactor fluence, showing enhanced hardening rates for small-grained material [70]. The fast ($E > 1$ MeV) neutron fluence was ~10% of the epithermal fluence.

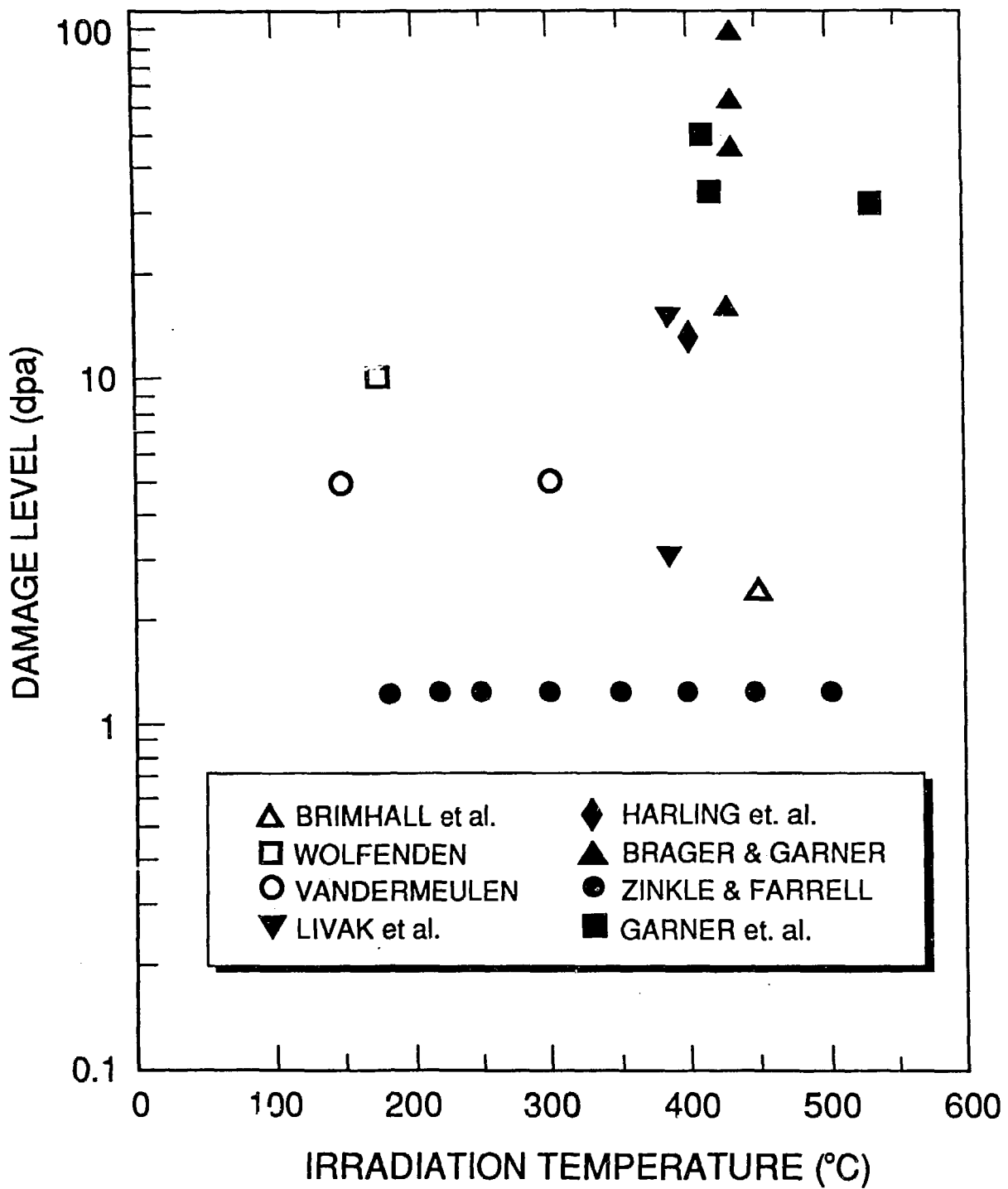
Fig. 12. Measured defect cluster density in 14-MeV Cu^{3+} ion-irradiated copper as a function of irradiation temperature [28].

Fig. 13. Temperature-dependent defect cluster density observed in Cu-18 wt ppm ^{10}B following fission neutron irradiation to a damage level of ~1.2 dpa [20].

Fig. 14. The effect of extension prior to irradiation on the dose dependence of elongation to fracture of fission neutron-irradiated copper tested at 20°C [70].

Fig. 15. Schematic temperature dependence of radiation hardening and void swelling regimes in neutron-irradiated copper.

SUMMARY OF HIGH-DOSE NEUTRON IRRADIATION EXPERIMENTS ON COPPER ALLOYS



CROSS-SECTION MICROSTRUCTURE OF ION-IRRADIATED COPPER

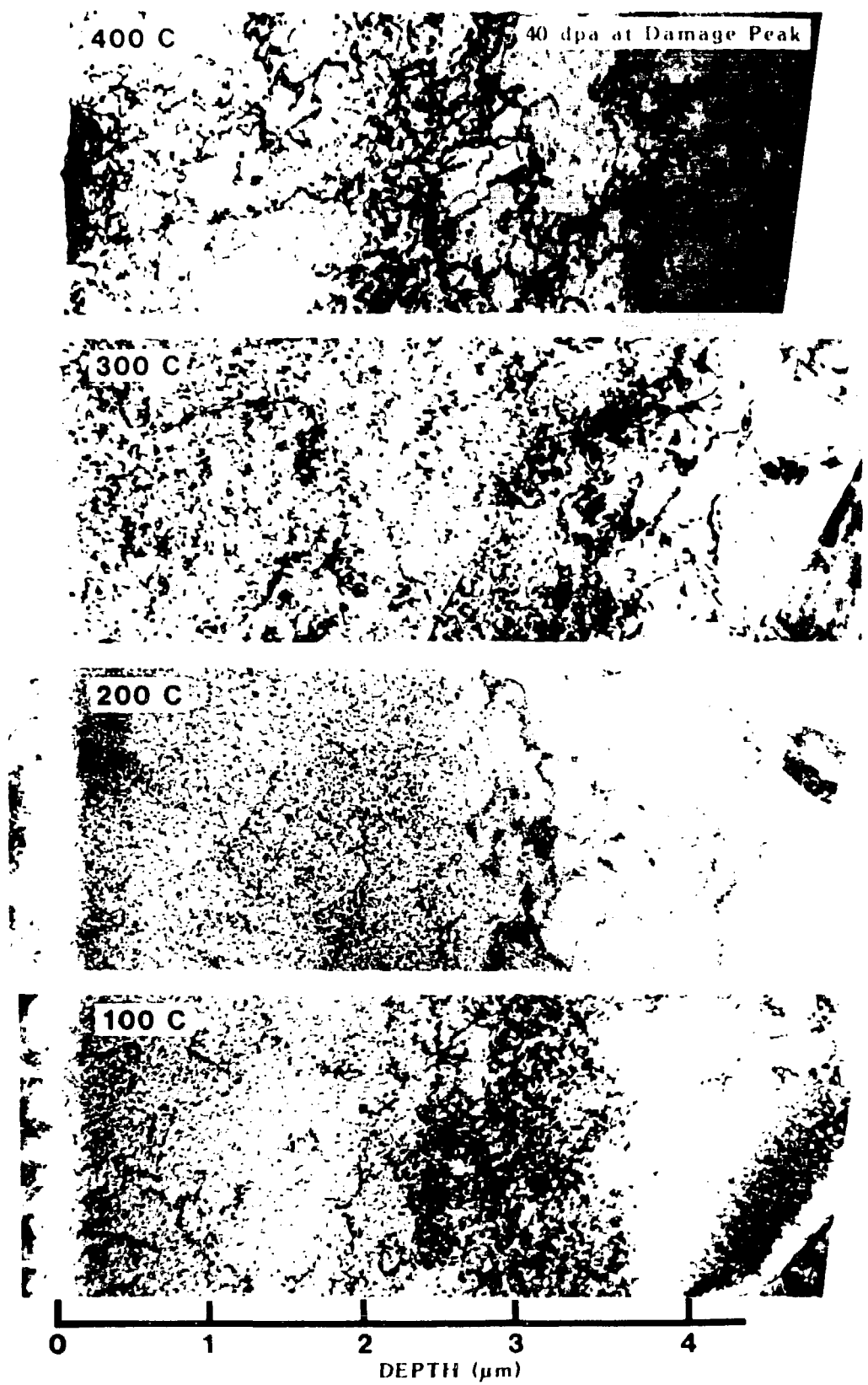


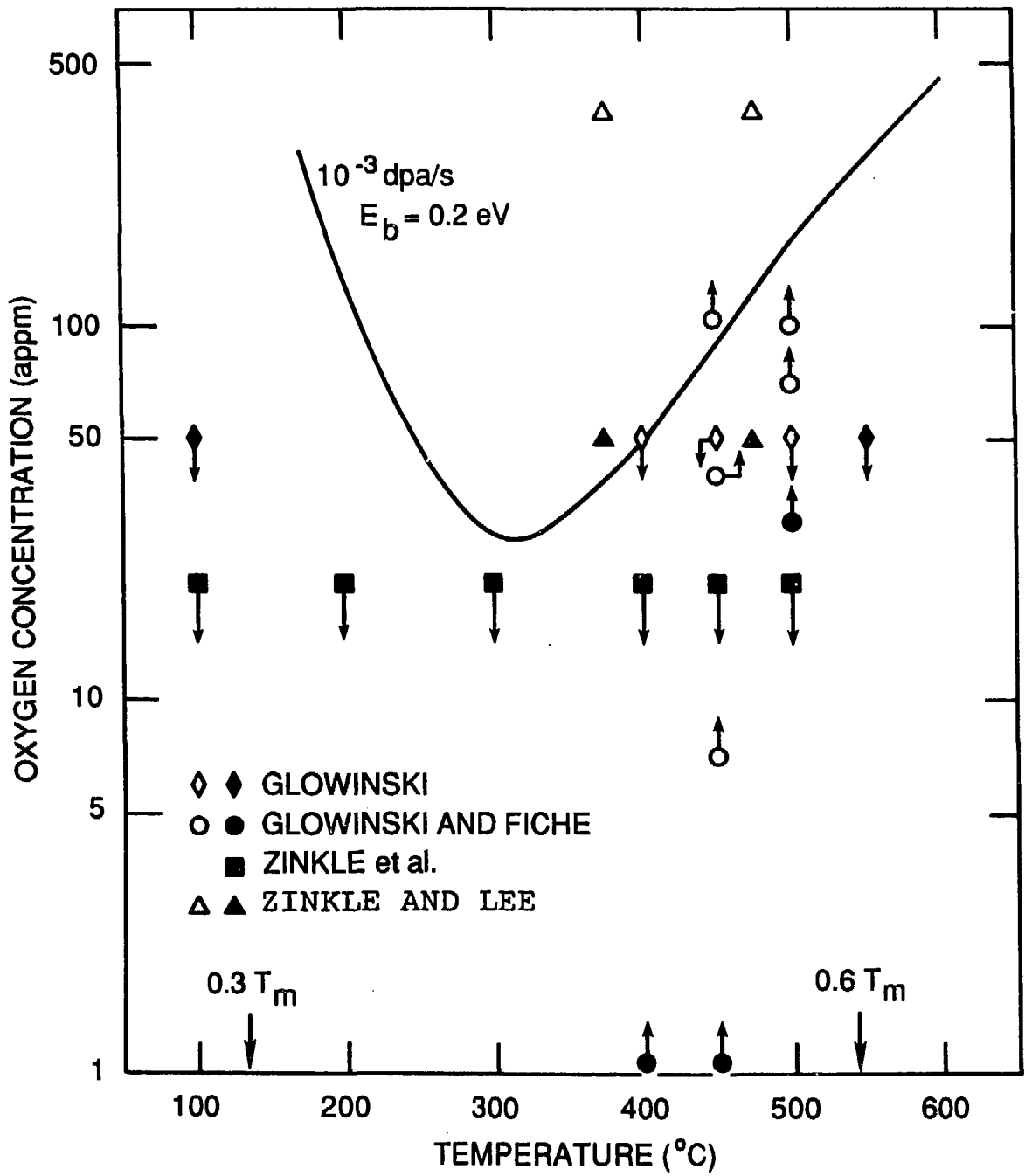
Fig. 7.32. Cross-section microstructure of copper following ion irradiation to a peak damage level of 40 dpa at 100-400 C.

HIGH-OXYGEN COPPER IRRADIATED AT 475 C



0 0.5 1.0 1.5 2.0
DEPTH (μm)

COMPARISON OF PREDICTED AND OBSERVED VOID FORMATION IN COPPER



DEPTH-DEPENDENT SWELLING IN ION-IRRADIATED COPPER

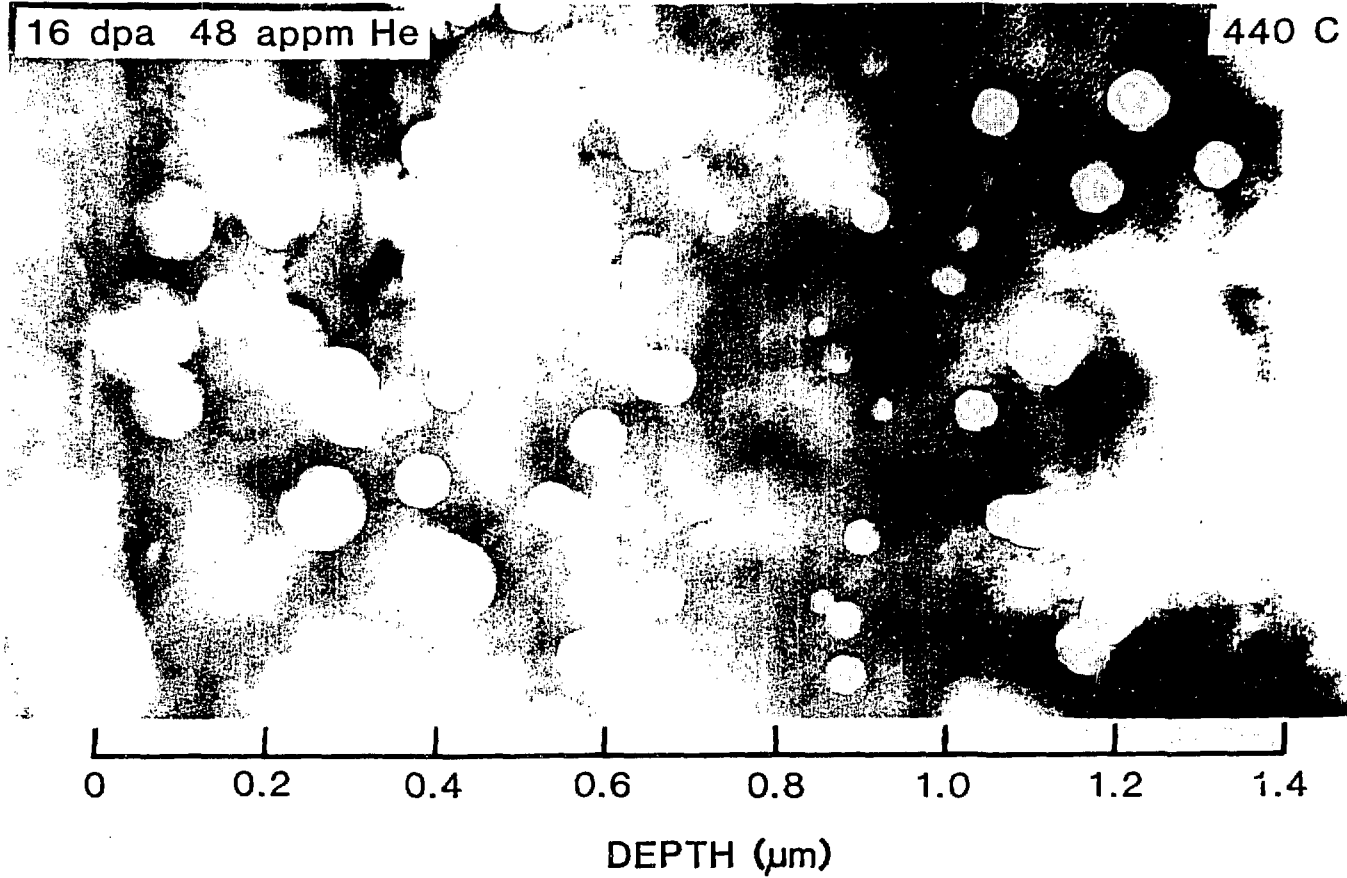
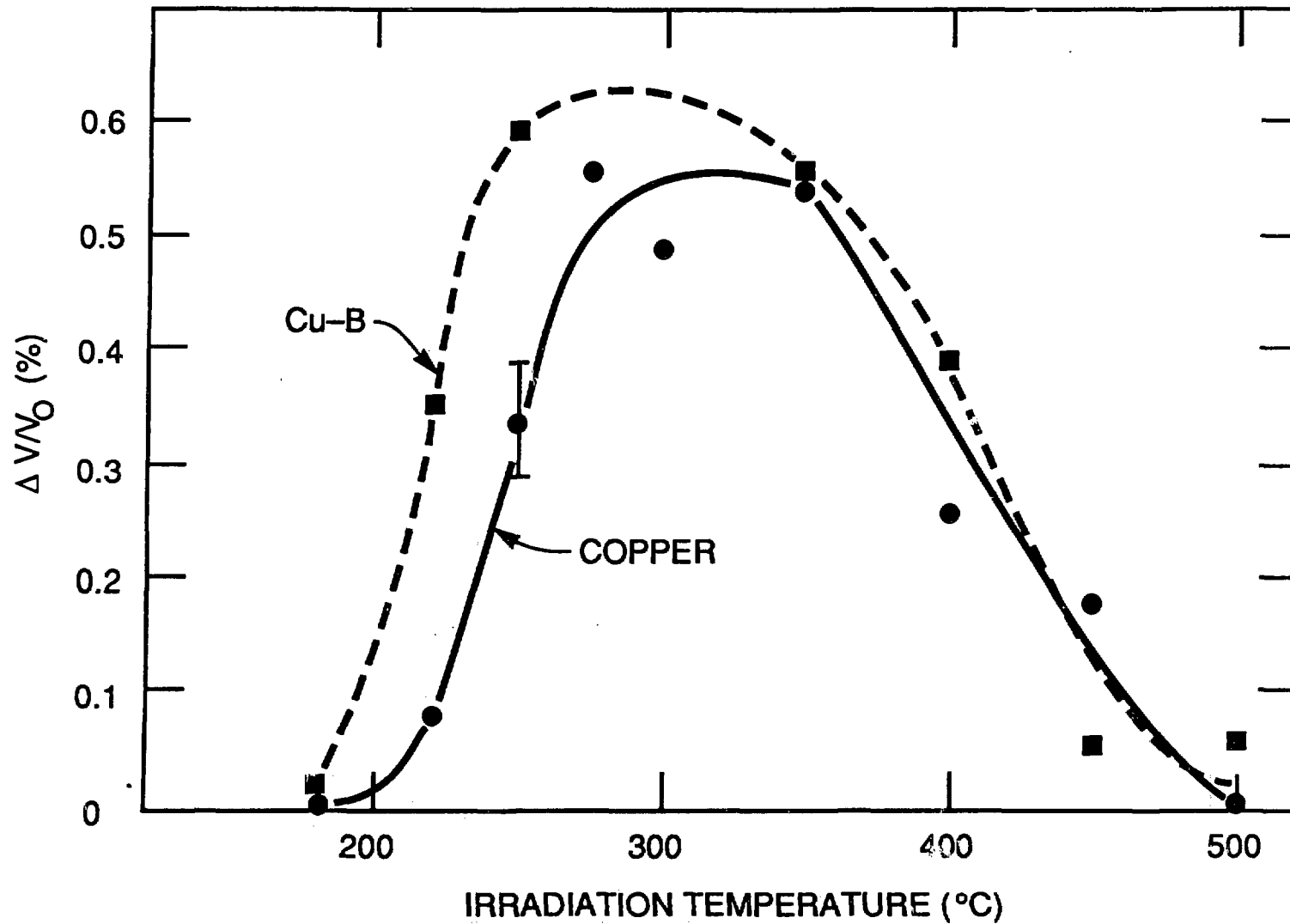
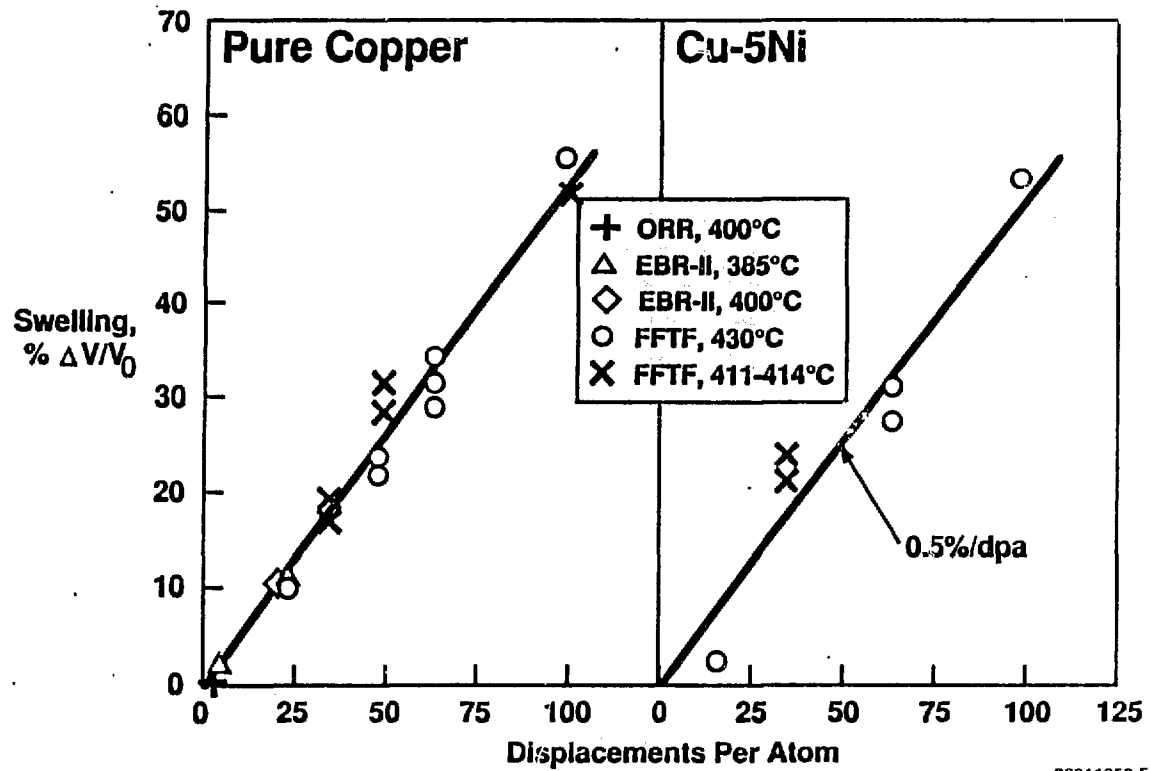


Fig. 5

COMPARISON OF VOID SWELLING IN NEUTRON IRRADIATED COPPER AND Cu-B

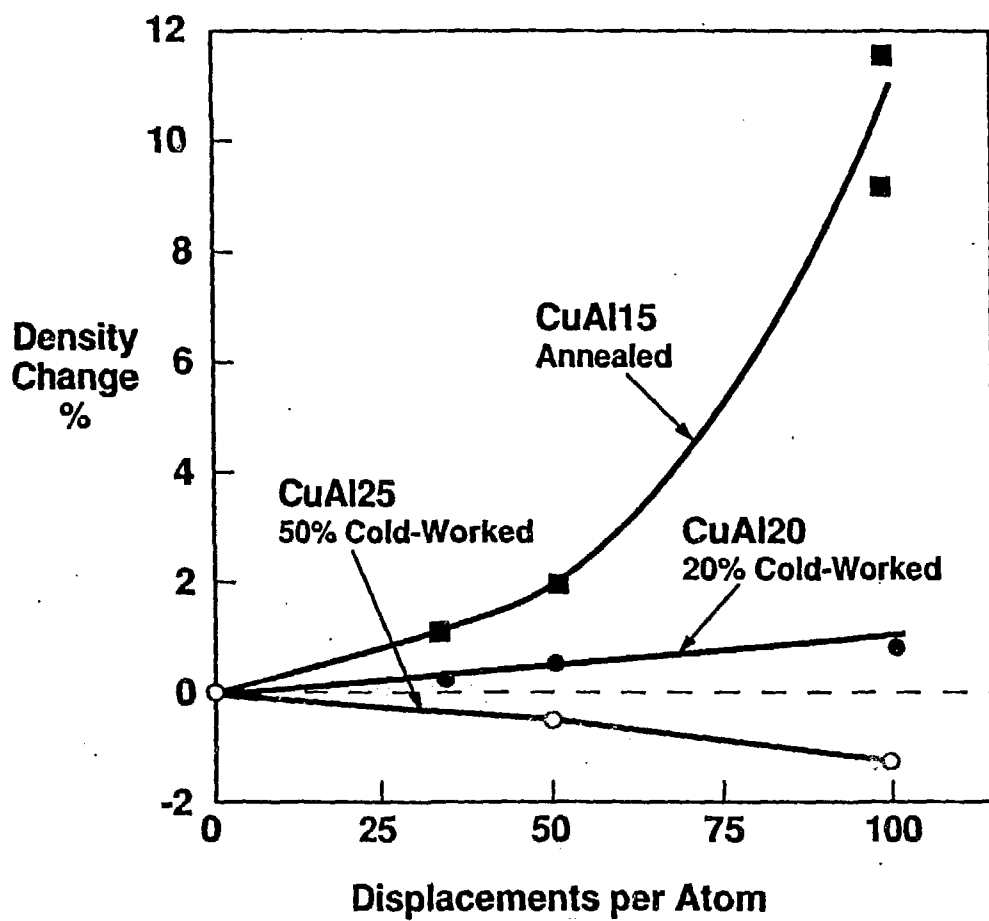


Garner, Brager and Anderson

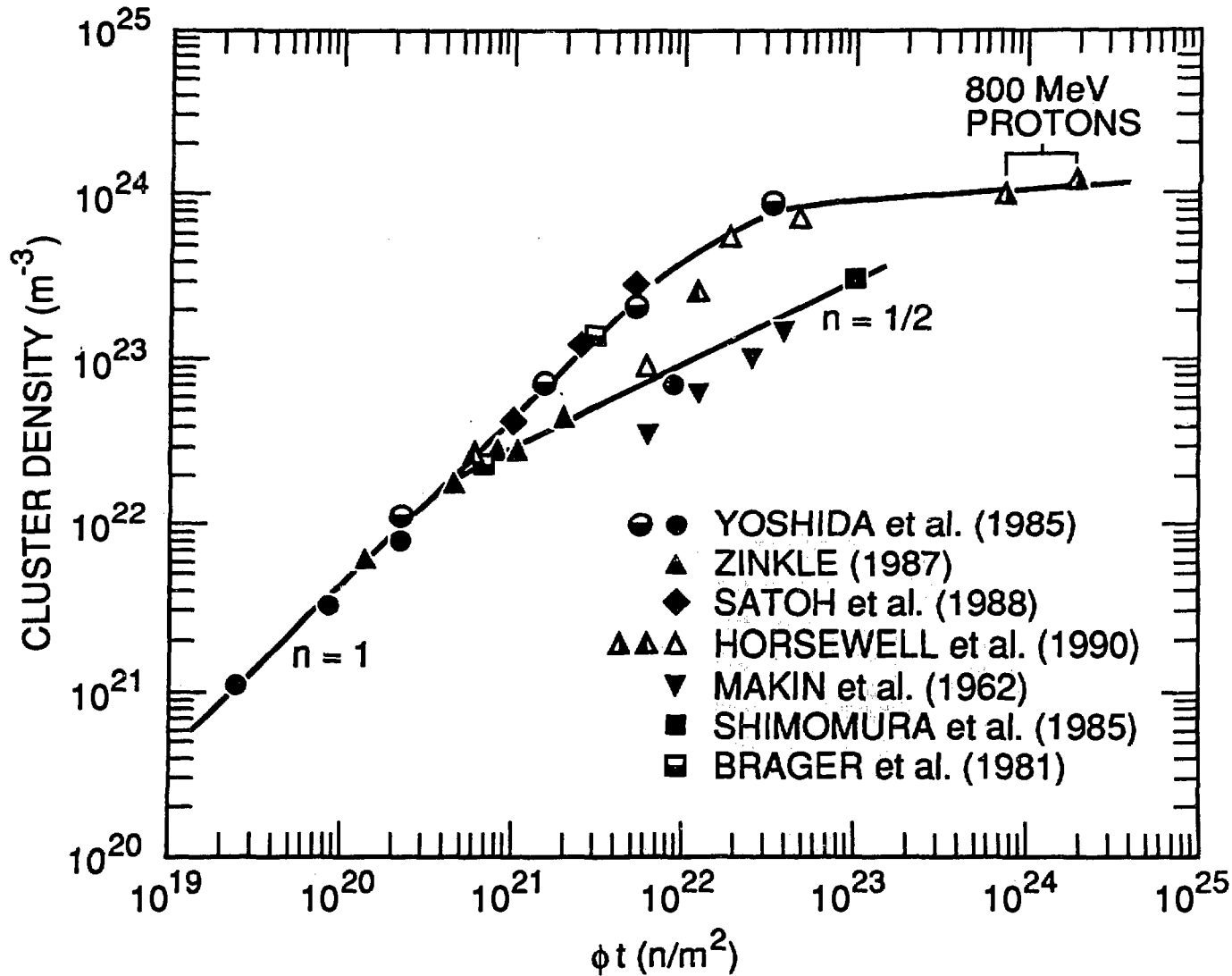


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F.A. Garner and H.R. Brager
FFTF, 411-414°C



Fluence-dependent Defect Cluster Formation in Copper Irradiated Near Room Temperature



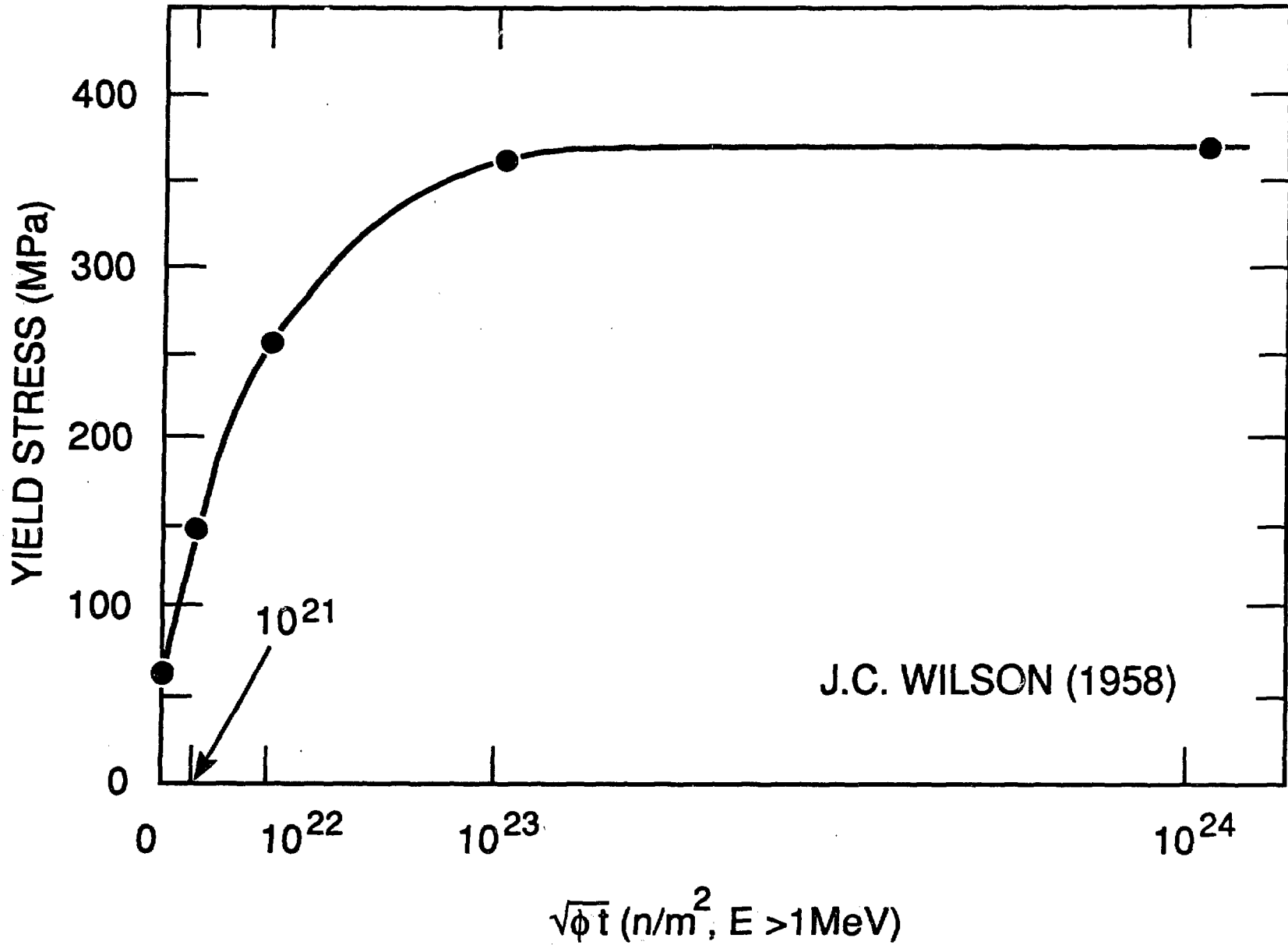


Fig. 10

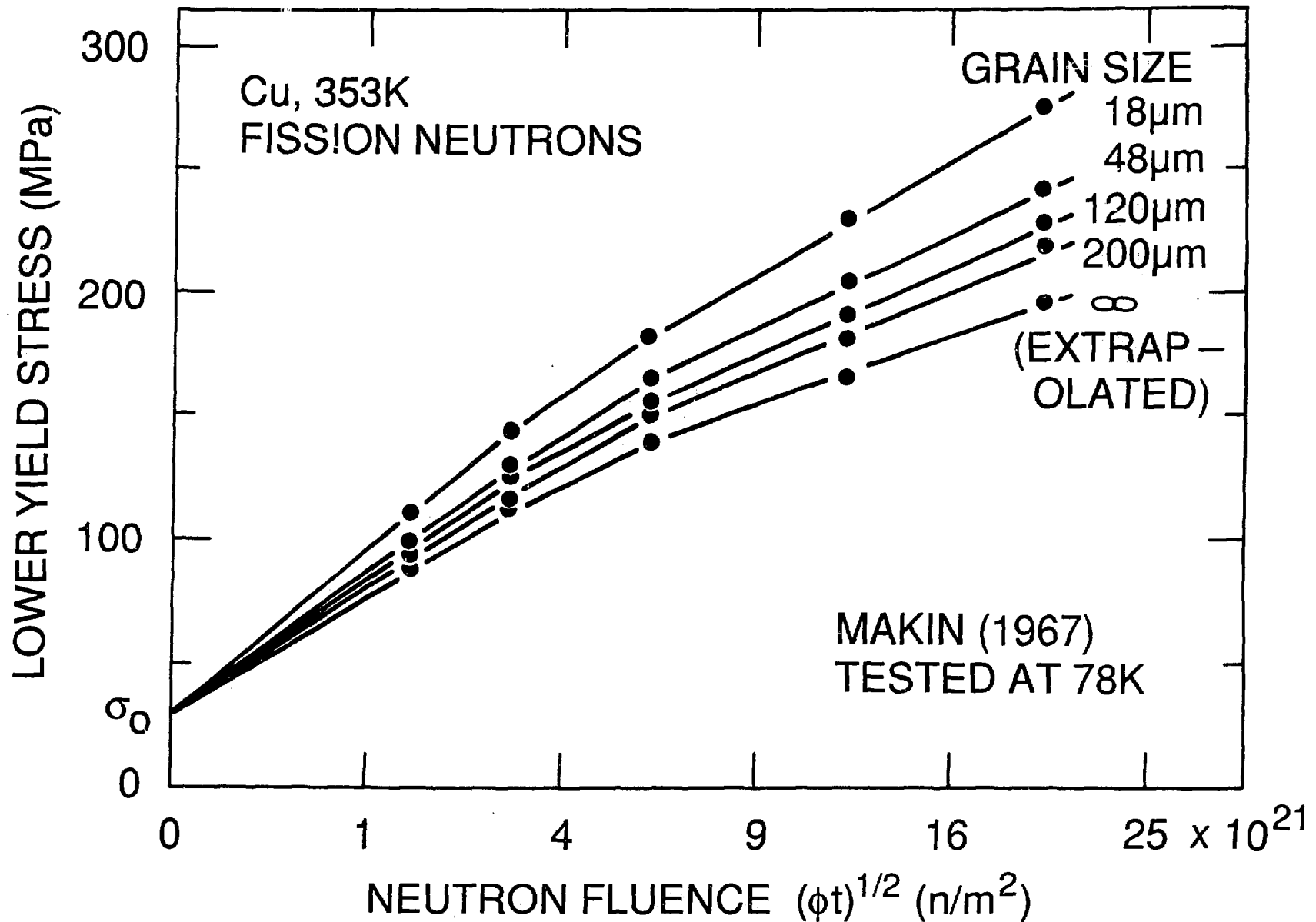
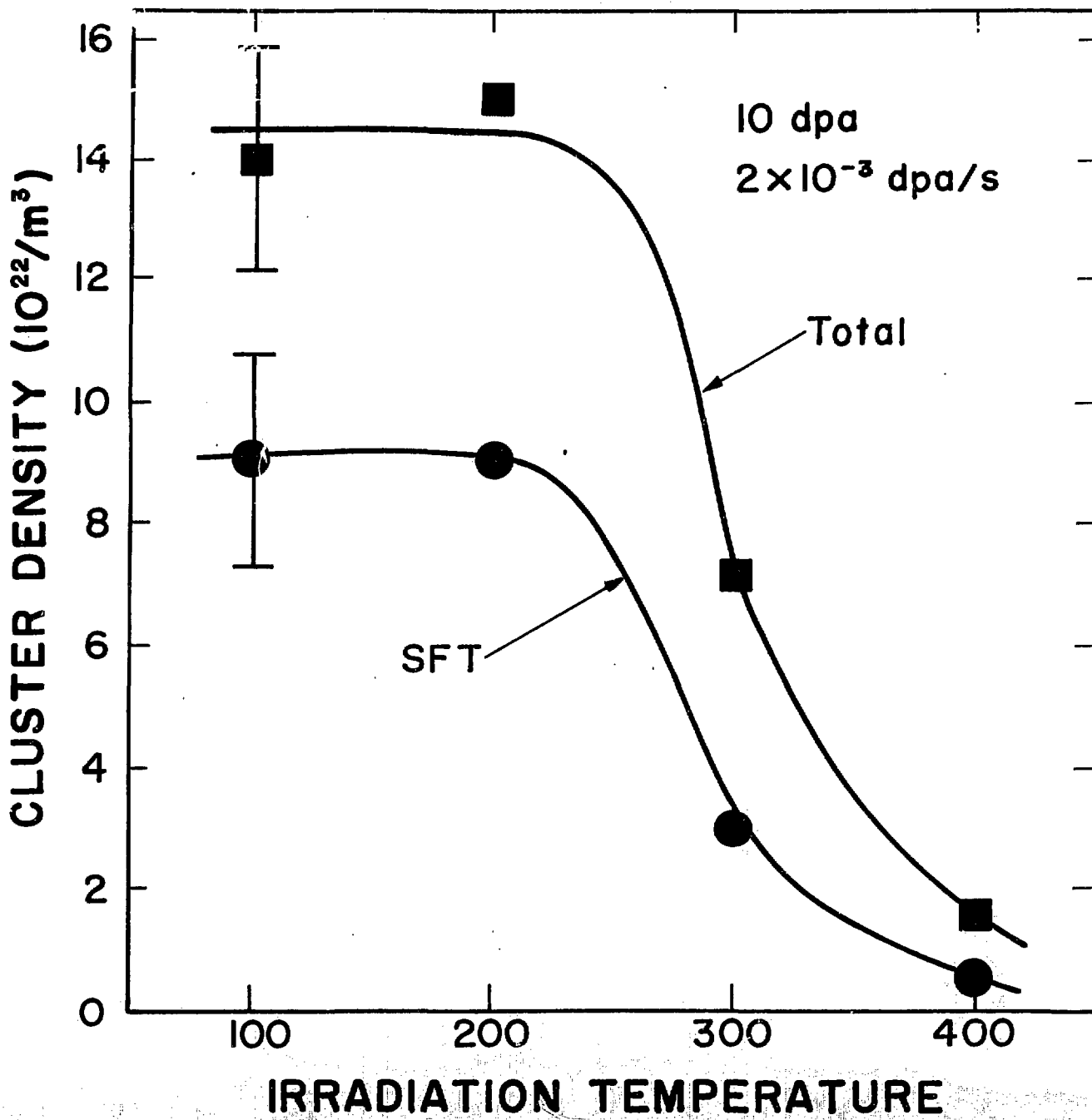


Fig. 11

DEFECT CLUSTER DENSITY IN ION IRRADIATED COPPER



ORNL-DWG 90M-14434

Planar Defect Cluster Density in Neutron-Irradiated Cu-B

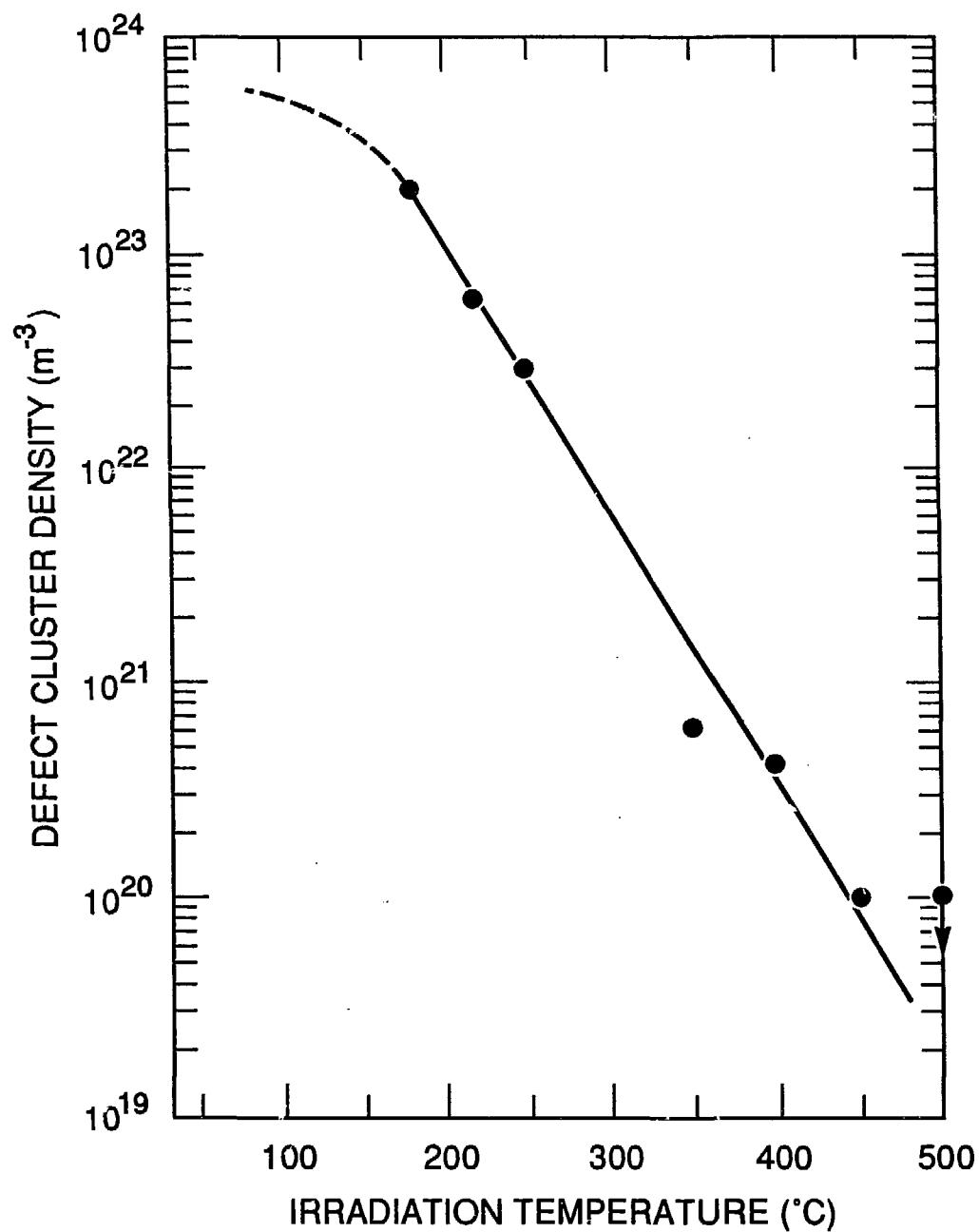


Fig. 14

