75-113

15 MeV NEUTRON DAMAGE IN Cu AND Nb

J. B. Roberto, J. Narayan, and M. J. Saltmarsh
Oak Ridge National Laboratory
Oak Ridge, Tenn. 37830

ABSTRACT

We have investigated high energy neutron damage in Cu and Nb irradiated with ~15 MeV neutrons at the Oak Ridge Isochronous Cyclotron. The neutrons were generated by bombarding a thick Be target with 40 MeV deuterons resulting in a high energy neutron spectrum broadly peaked at 15 MeV. Single crystals of Cu and Nb were irradiated at room temperature to fluences of ~2×10^{17} n/cm^2. The resulting loop-type defect clusters in the crystals were characterized using x-ray diffuse scattering and transmission electron microscopy. The cluster size distributions were found to be generally similar to those characteristic of fission neutron irradiations in these materials and no multiple clusters or sub-clusters were observed. Additional comparisons with fission reactor irradiations in Cu and Nb indicate that the retained displacement damage in these crystals is approximately 3 times greater for the high energy neutrons than for an equivalent fluence of fission neutrons. This result is consistent with detailed damage energy calculations for the Be(d,n) neutron spectrum.

INTRODUCTION

Recent experimental studies[1] and theoretical calculations[2] have suggested that high energy neutrons (E ≈ 15 MeV) are substantially more effective in producing displacement damage than fission neutrons. Such implications are important to the fusion reactor designer who must consider the effects of significant fluences of high energy neutrons on reactor materials. In this work, we have attempted to quantify some of the differences between high energy and fission neutron damage in Cu and Nb and to correlate the experimental results with theoretical calculations. The experiments have involved irradiations near room temperature followed by characterization of the retained damage in the resulting loop-type defect clusters.

This study is unique in several respects. First, high purity nearly perfect single crystals from the same source material were prepared identically for the high energy and fission neutron irradiations. As a result, spurious effects due to impurities or specimen differences were held to a practical minimum. Secondly, for Cu, both x-ray diffuse scattering and transmission electron microscopy were used to characterize the radiation-induced defects. Finally, the experimental results were compared with very detailed damage energy calculations which explicitly treated the various nuclear reactions which occur in the range of neutron energies present in these experiments.

This work also represents the first utilization of high energy neutrons from the deuteron breakup or "stripping" reaction to perform radiation damage experiments. The deuteron-breakup concept is the subject of considerable interest as a possible mechanism for producing a high-flux large volume source of high energy neutrons for CTR radiation effects research. Several proposals have been made[3,4] and our results provide direct evidence of the usefulness of such a source.

Be(d,n) NEUTRONS

The high energy neutrons were generated at the Oak Ridge Isochronous Cyclotron by stopping a 40 MeV deuteron beam in a thick Be target. The associated deuteron-breakup (d,n) reaction[5] results in a neutron spectrum which is broadly distributed in energy about a maximum at approximately 15 MeV with some neutrons above 30 MeV. The neutrons are strongly forward-peaked and are degraded in both energy and intensity with increasing angle from the deuteron beam axis. The ORIC source[6] produces a maximum flux of $2 \times 10^{12} n/cm^2$-sec at the specimen chamber with a full-width-half-maximum of approximately 7mm. The results of a recent time-of-flight measurement[4] of the Be(d,n) spectrum are shown in Fig. 1.

The broad energy distribution of the Be(d,n) spectrum presents somewhat of an interpretive problem for radiation damage experiments. In particular, the effects of the high energy tail of the spectrum must be considered. We have approached this problem by computing the displacement damage energy in Cu and Nb as a function of incident neutron energy. The damage energy is that part of the primary recoil energy which is ultimately available for producing atomic displacements. The damage energy concept is therefore useful for comparing displacement damage at various neutron energies. Our calculations are based on the electronic stopping theory of Lindhard[7] and are generally similar to earlier calculations of the energy dependence of neutron damage by Robinson.[2]

For the Be(d,n) spectrum, it is necessary to consider neutron interactions which occur at energies of 30 MeV and higher. Neutron cross-sections are generally unavailable above 15 MeV and we have used theoretical cross-sections[8] based on optical and pre-compound nuclear models for our damage energy calculations. These cross-sections were computed by C.Y.Fu and F.G. Perey of Oak Ridge National Laboratory and include elastic scattering as well as the principle classes of nonelastic interactions. The results of damage energy calculations based on these cross-sections will be published in more detail elsewhere,[9] but we include the damage energy curves for Cu as an example in Fig. 2.

EXPERIMENTAL PROCEDURE

Specimen Preparation

The Cu samples were high purity single crystal platelets approximately 1 x 1 x 0.1 cm which were acid cut from single crystal ingots and lightly polished in 10% H_3PO_4. The Nb crystals were grown from .05 cm thick sheets of zone-refined starting material using the strain-anneal technique. Large single crystal regions up to several square cm in area and with orientations predominately near (110) could be identified in the resulting grain structure. Small platelets 0.5 x 1 x 0.05 cm were spark cut from these single crystal regions and polished using 3:2 HNO_3 and HF.

The resulting Cu and Nb specimens were essentially perfect from the standpoint of Borrmann topography and exhibited near intrinsic Bragg diffraction widths. The Cu starting material was 99.999% pure. The predominate impurities in the Nb starting material were C, O, Ta and W, all at concentrations of 50 wt. ppm or less. The results of residual gas analysis by the vacuum fusion technique for H, N, and O in the Cu and Nb samples after polishing are shown in Table 1. Also shown in Table 1 are the concentrations of C, Ta, and W in the annealed Nb as determined by chemical analysis.

Irradiation Technique

The high energy Be(d,n) neutron irradiations were carried out in an open-ended aluminum capsule with the samples separated by aluminum spacer rings. Two Cu and two Nb specimens were mounted one behind the other along the beam axis. Ni and Co dosimetry foils were placed before and after the sample capsule and the neutron fluence decreased approximately 13% from the first to last sample. The details of the dosimetry have been described elsewhere.[6] The irradiations were carried out at room temperature on a continuous basis over a period of 30 hrs. with a total spectrum dose of 2.0 x $10^{17} n/cm^2$ at the center of the first sample.

The fission reactor irradiations were preformed at the CP-15 facility of the Solid State Division's bulk shielding reactor. The neutron flux and energy spectrum at the CP-15 position has been carefully measured[10] over the range from 0.6 to 3 MeV and is very similar to a pure fission spectrum. For our fluence and damage energy calculations we have assumed such a neutron distribution. This pure fission neutron spectrum can be compared with the Be(d,n) spectrum in Fig. 1. The fission reactor irradiations were carried out

Table 1. Impurity Analysis of Cu and Nb Samples

(wt. ppm)	H	C	N	O	Ta	W
Cu	30	-	<5	10	-	-
Nb	<5	50	10	50	50	15

at 43°C in a pressurized He-cooled vessel. The total fluences were 1.0×10^{18} and $5.0 \times 10^{17} n/cm^2$ ($E > 0.1$ MeV) respectively for the Cu and Nb samples. Following both the high energy and fission neutron irradiations, the samples were allowed to decay for approximately two months and then lightly polished using the techniques described above to remove any surface contamination.

TEM Measurements

Transmission electron microscopy measurements were carried out on both the Nb and Cu crystals using a Hitachi 200 keV electron microscope. The TEM samples were prepared by conventional electropolishing techniques. For the Be(d,n) irradiations, the samples were cut from the high-fluence center region of the crystals. High resolution weak beam (dark field), higher order diffraction (3g), and conventional bright and dark field microscopy were used to determine the size distributions of defect clusters which were present in the form of dislocation loops. Sample thicknesses were determined by stereomicroscopy.

X-ray Measurements

The radiation-induced defects in the Cu crystals were also characterized using x-ray diffuse scattering. These measurements involved collecting the total diffracted intensity into a wide open detector as a function of deviation from the Bragg angle. Care was taken to insure that the measurements corresponded to the high-fluence center region of the Be(d,n) irradiated crystals. Intensity vs. angle curves about the (111) and (222) reflections were measured both before and after irradiation. The difference in the curves represents the diffuse scattering from the defects themselves and was interpreted using a computed program developed by B. C. Larson.[11] The computer technique uses a least-squares fitting procedure to obtain a size distribution from the scattering data for the loop-type defect clusters. For Cu we have assumed loops on (111) planes with (110) Burgers vectors. Efforts to measure the diffuse scattering from Nb were unsuccessful due to the low retained defect density and high background associated with the Nb samples.

RESULTS AND DISCUSSION

The results of our measurements of the retained defect clusters in Be(d,n) and fission neutron irradiated Cu and Nb are summarized in Figs. 3-6. In Fig. 3, bright field electronmicrographs are shown for ~15 MeV and fission neutron damage in Cu. The micrographs correspond to doses of $2.0 \times 10^{17} n/cm^2$ and $1.0 \times 10^{18} n/cm^2$ ($E > 0.1$ MeV) respectively for the 15 MeV and fission neutron irradiations. Loop size distributions in Cu as-determined by TEM and x-ray measurements are compared in Fig. 4. These size distributions represent independent measurements with no adjustable parameters. The absolute agreement within about a factor of two between the TEM and x-ray results is encouraging, but more important is the close agreement on the relative effects of high energy and fission neutron irradiations as determined independently by the two experimental techniques.

Micrographs for the high energy and fission neutron irradiations in Nb are shown in Fig. 5. These micrographs correspond to doses of $1.8 \times 10^{17} n/cm^2$ over the Be(d,n) spectrum and $5.0 \times 10^{17} n/cm^2$ ($E > 0.1$ MeV) for fission neutrons. Loop size distributions from TEM measurements in Nb for both 15 MeV and fission neutron damage are shown in Fig. 6. The average loop size is much smaller for Nb than Cu.

General similarities between the retained damage from high energy and fission neutron irradiations in both Cu and Nb are apparent in the micrographs and size distributions of Figs. 3-6. Further study will be required to determine whether or not some of the small differences in the observed size distributions for 15 MeV and fission damage are significant. The possible existence of multiple defect clusters associated with the 15 MeV neutron irradiations was also investigated. Careful stereomicroscopy (both weak beam and higher order diffraction techniques) revealed no apparent indications of multiple clusters in either Cu or Nb.

In order to compare the Be(d,n) and fission neutron damage in Nb and Cu on an absolute basis, we have integrated the size distribution curves of Figs. 4 and 6 to determine the retained point defect densities. When adjusted for differences in fluence, these point defect densities give a relative indication of the damage effectiveness of the high energy and fission neutrons. Ratios of the damage effectiveness of Be(d,n) neutrons in terms of fission neutrons as determined from the experimentally observed point defect densities are shown in Table 2. The ratios derived from the TEM and x-ray measurements in Cu are in fairly good agreement and indicate that these independent techniques sample comparable aspects of the damage. Also shown in the table are theoretical ratios based on the damage energy calculations described above.

The agreement between theory and experiment in Table 2 is quite remarkable and indicates that the damage effectiveness of a Be(d,n) neutron with a mean energy of 15 MeV is approximately 3 times greater than a fission neutron in Cu and Nb. Of course, the irradiations were carried out near room temperature where annealing effects are important while the damage energy calculations are appropriate for low temperatures. Nevertheless, the good correlation between theory and experiment for both Cu and Nb suggests that the annealing characteristics of high energy and fission neutron damage are similar and that differences in displacement cascade structure are not striking.

Table 2. Damage Effectiveness of Be(d,n)[a] Neutrons as Compared with Fission Reactor Neutrons

Material	Experiment Retained Damage Be(d,n)/Fission	Theory Damage Energy Be(d,n)/Fission
Cu	3.3 (X-ray) 4.0 (TEM)	3.4
Nb	2.5 (TEM)	2.6

[a]Deuteron energy, 40 MeV.

SUMMARY AND CONCLUSIONS

We have described an experimental investigation of high energy ($E \simeq 15$ MeV) neutron damage in Cu and Nb. Both TEM and x-ray measurements have been used to characterize the surviving loop-type defect clusters in near room temperature irradiations using high energy Be(d,n) neutrons as well as fission neutrons. The resulting cluster size distributions have been compared for high energy and fission neutrons and the defect densities correlated with detailed damage energy calculations. We are led to the following conclusions:

1) The damage effectiveness of a Be(d,n) neutron with a mean energy of 15 MeV is ~3 times that of a fission reactor neutron in Cu and Nb. This result is supported by both experiment and theory.

2) There are general similarities in loop size distributions for the high energy and fission neutron irradiations. Additional studies are required to compare the size distributions in detail.

3) There are no apparent indications for the existence of multiple defect clusters resulting from the high energy neutron irradiations in either Cu or Nb.

4) The deuteron-breakup reaction has proven to be a useful technique for generating high energy neutrons for radiation damage studies. We have shown that we can computationally handle the broad energy spectrum of the neutron source and that the interpretation of the experimental results for Cu and Nb is not particularly complicated by the source spectrum.

The overall results suggest similarities between high energy and fission neutron radiation damage when compared using the damage energy concept. We are currently expanding our experiments and calculations to include Al and Au to represent a broader range in atomic species. Low temperature damage rate measurements are also planned as a complement to our damage effectiveness results. In addition, a detailed analysis of the nature of the defects produced in the high energy and fission neutron irradiations is underway.

ACKNOWLEDGEMENTS

The authors wish to acknowledge many of their ORNL colleagues for their assistance during the course of these experiments. In particular, we wish to thank F. W. Young, Jr. and R. E. Reed for respectively providing the Cu and Nb crystals, C. Y. Fu and F. G. Perey for computing neutron cross-sections and G. J. Smith for performing neutron dosimetry. The authors also acknowledge valuable discussions with M. T. Robinson, B. C. Larson, L. H. Jenkins, and T. S. Noggle as well as the experimental assistance of F. A. Sherrill. This work was supported by the USERDA under contract with Union Carbide Corporation.

REFERENCES

1. J. B. Mitchell, C. M. Logan and C. J. Echer, J. Nucl. Mater. 48, 139 (1973); J. B. Mitchell, R. A. VanKonynenburg, M. W. Guinan and C. J. Echer, Phil. Mag. 31, 919 (1975).

2. M. T. Robinson in Nuclear Fusion Reactors, p. 364, Brit. Nuclear Energy Soc. (1970).

3. A. N. Goland, D. H. Gurinsky, J. Hendrie, J. Kukkonen, T. Sheehan and C. L. Snead, Jr. in Proc. of Int. Conf. on Radiation Test Facilities for CTR Surface and Materials Program, Argonne National Laboratory, July 1975, in press.

4. M. J. Saltmarsh, A. P. Fraas, J. A. Horak and J. A. Martin in ref. 3.

5. R. Serber, Phys. Rev. 72, 1008 (1947).

6. L. H. Jenkins, T. S. Noggle, R. E. Reed, M. J. Saltmarsh and G. J. Smith, Appl. Phys. Lett. 26, 426 (1975).

7. J. Lindhard, V. Nielsen, M. Scharff and P. V. Thomsen, Kgl. Danske Videnskab. Selskab, Mat. - Fys. Medd. 33, No. 10 (1963).

8. C. Y. Fu and F. G. Perey, to be published.

9. J. B. Roberto and M. T. Robinson, to be published.

10. J. M. Williams, private communication.

11. B. C. Larson, J. Appl. Cryst. 8, 150 (1975).

FIGURE CAPTIONS

Fig. 1. High Energy Be(d,n) Neutron Spectrum Compared with a Pure Fission Spectrum.

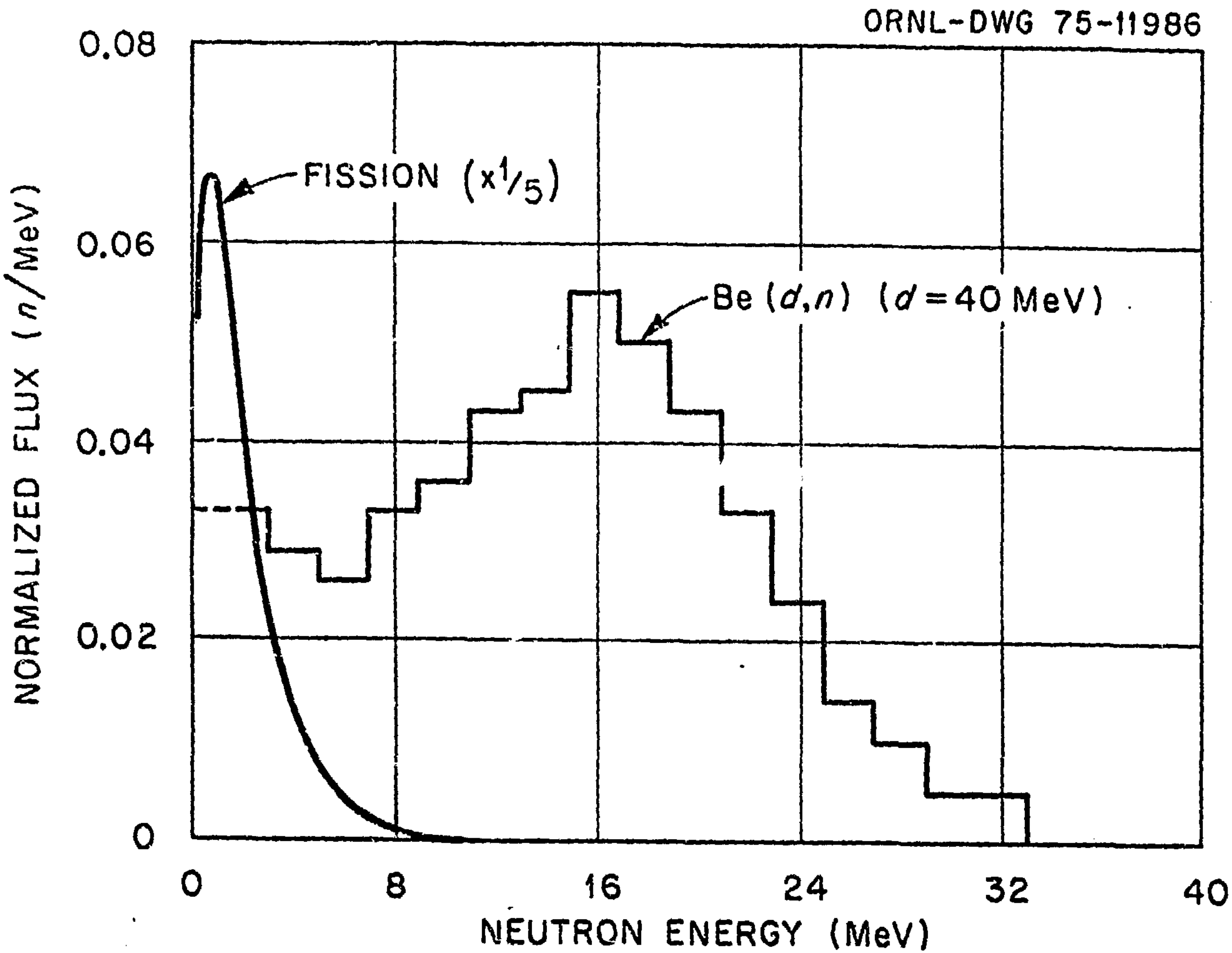
Fig. 2. Specific Damage Energy vs. Neutron Energy in Cu.

Fig. 3. TEM Micrographs of Cu Irradiated with (a) Fission Neutrons ($1.0 \times 10^{18} n/cm^2$ $E > 0.1$ MeV) and (b) ~15 MeV Neutrons ($2.0 \times 10^{17} n/cm^2$ over the Be(d,n) Spectrum). The arrow indicates the direction of the diffraction vector $[\bar{2}20]$ and corresponds to a length of 0.2 microns.

Fig. 4. Loop Size Distributions in Cu for Fission and ~15 MeV Neutron Irradiations as Determined by TEM and X-ray Techniques. The x-ray data for fission neutrons is from ref. 11.

Fig. 5. TEM Micrographs of Nb Irradiated with (a) Fission Neutrons ($5.0 \times 10^{17} n/cm^2$ $E > 0.1$ MeV) and (b) ~15 MeV Neutrons ($1.8 \times 10^{17} n/cm^2$ over the Be(d,n) Spectrum). The arrow indicates the diffraction vector $[3\bar{3}0]$ and corresponds to a length of 0.2 microns.

Fig. 6. Loop Size Distributions in Nb for Fission and ~15 MeV Neutron Irradiations as Determined by TEM Measurements.


ORNL-DWG 75-11986
FISSION (x1/5)
Be (d,n) (d=40 MeV)
NORMALIZED FLUX (n/MeV)
0.08
0.06
0.04
0.02
0
NEUTRON ENERGY (MeV)
0
8
16
24
32
40

ORNL-DWG 75-9720

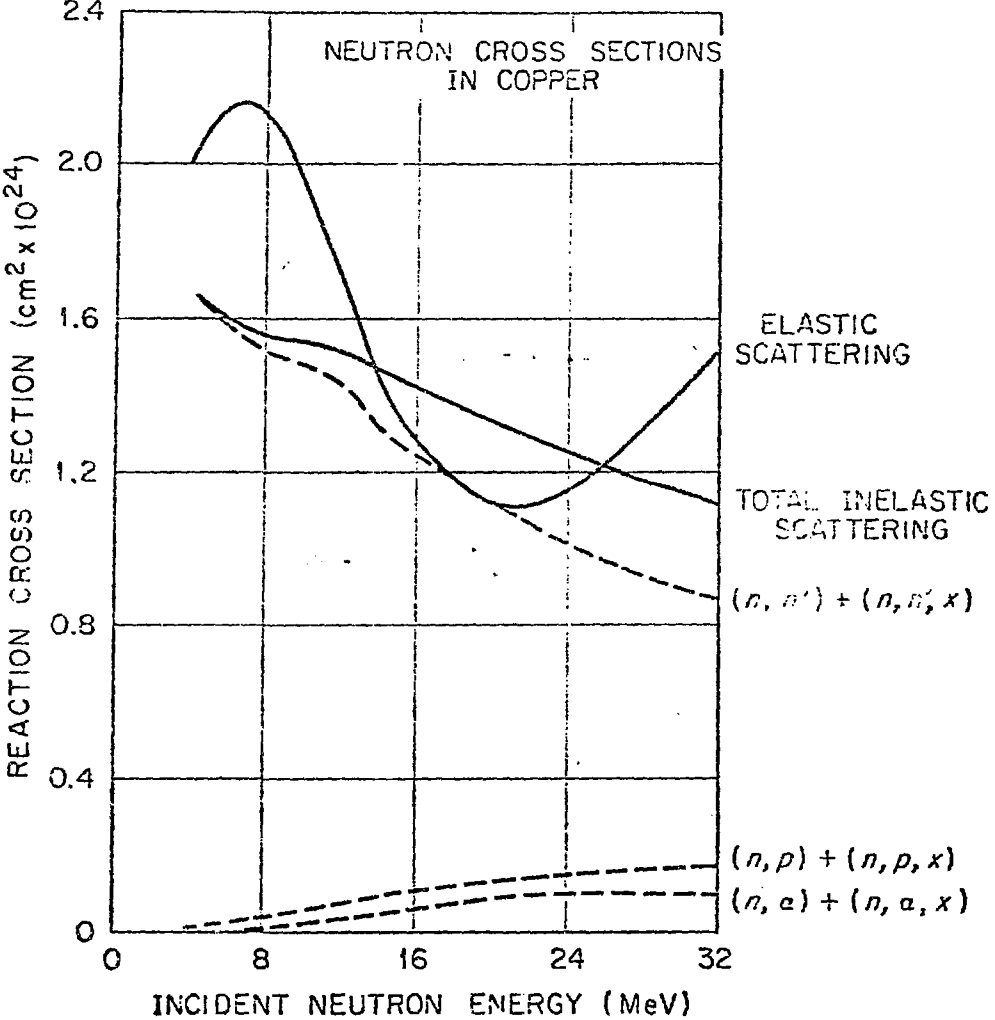
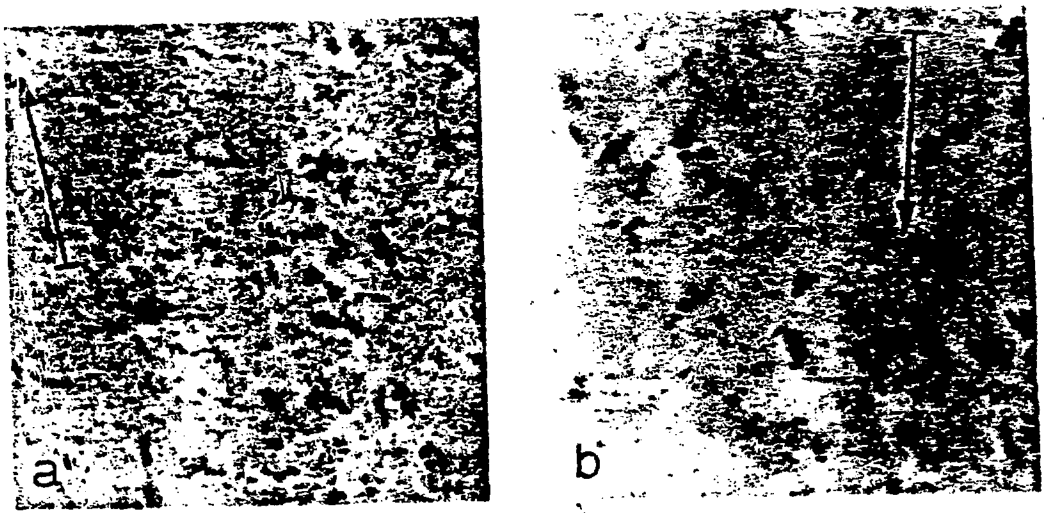
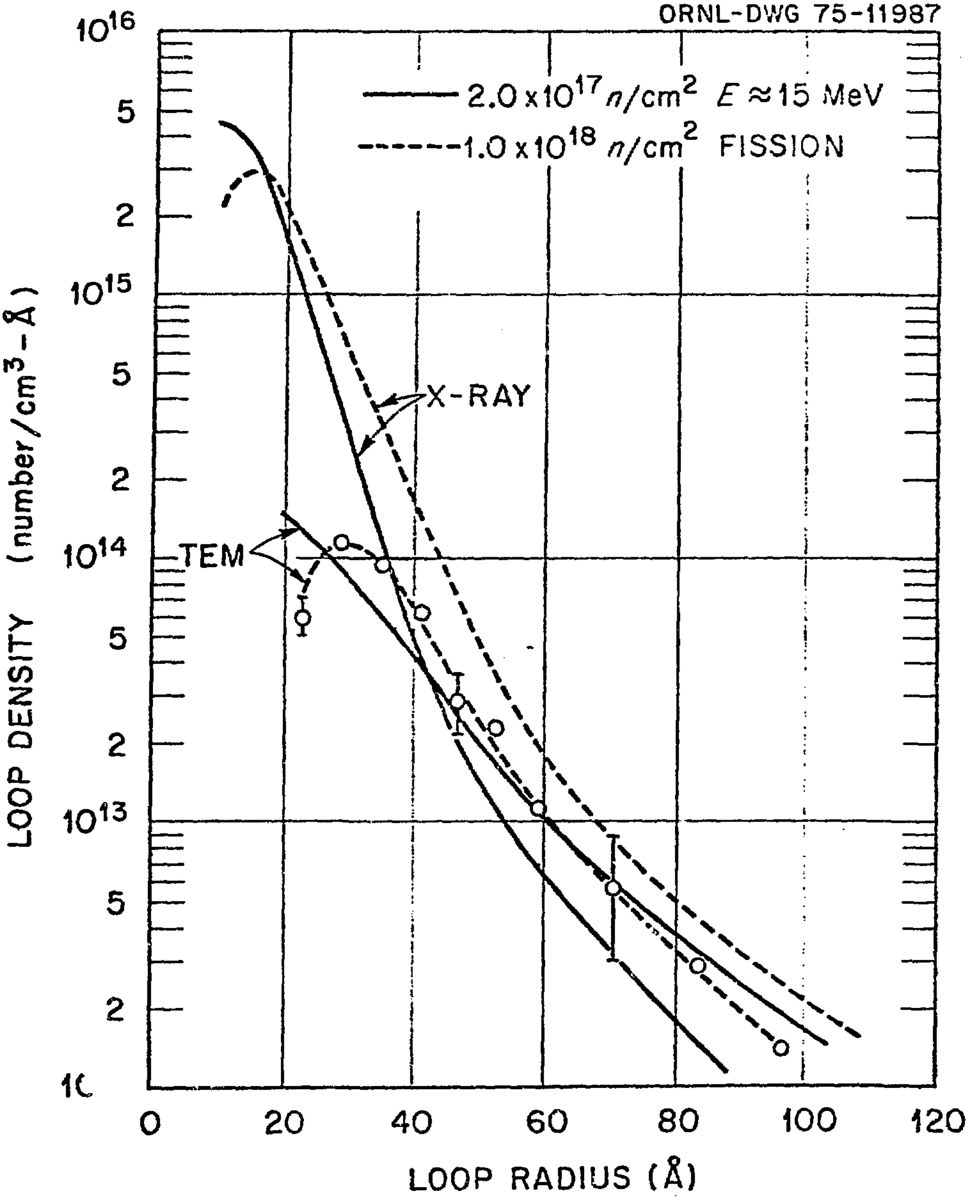


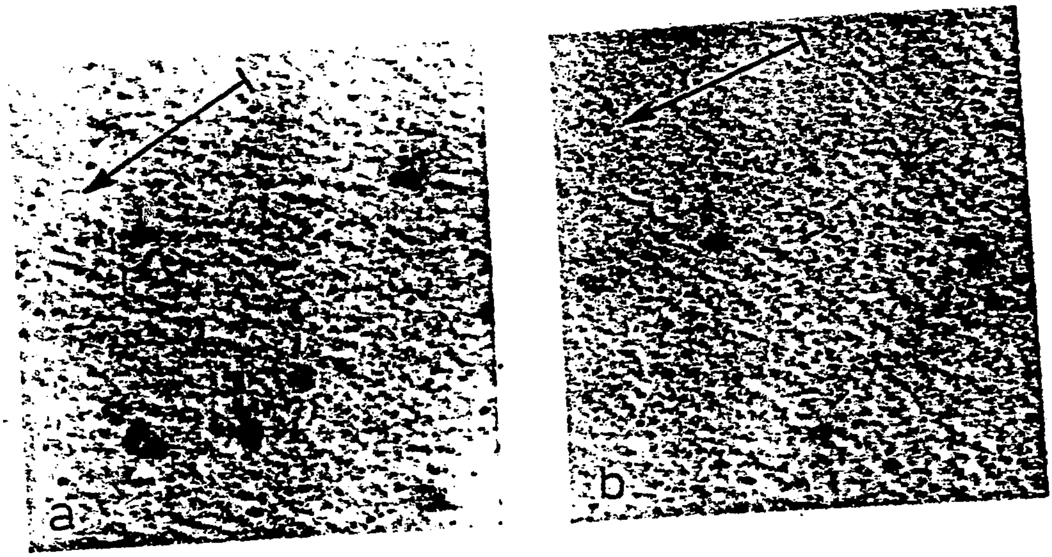
FIG 2


a
b

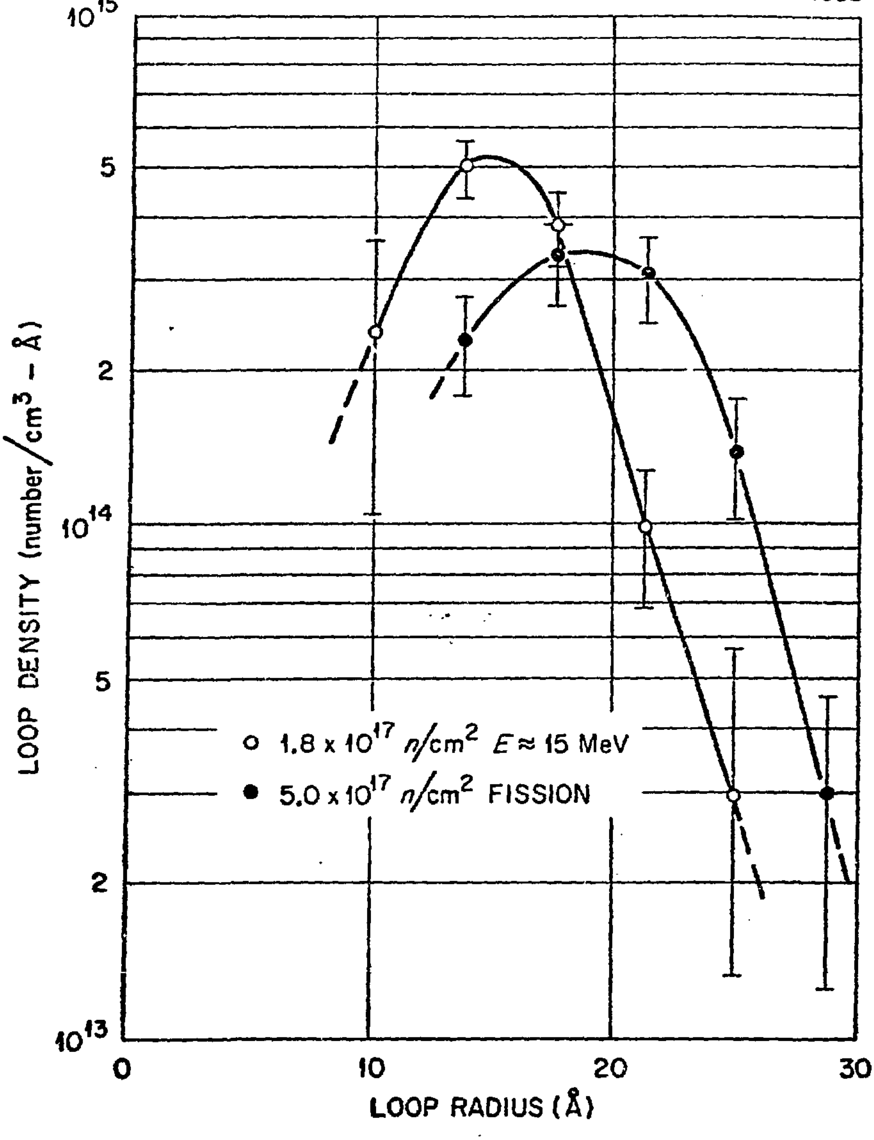


Loop Size Distributions in Cu.

FIG. 4


a
b

ORNL-DWG 75-11982



Loop Size Distributions in Nb.

FIG 6