CONF-741040--8

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

Radiation Damage Studies with Accelerators*

by

M. J. Saltmarsh

Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830

MASTER

ABSTRACT

Radiation damage is one of the major technological problems associated with future reactor development programs. In the hostile environment of a reactor core, drastic changes can occur in the properties of materials in ways which are generally complex and poorly understood. Direct measurement of these effects is difficult. Fission reactors have limited space, and experiments are often long and costly. No fusion reactor is yet available. As a result many laboratories are using accelerators to explore the various facets of radiation damage. The work at Oak Ridge is based mainly on the CN Van de Graaff and the Oak Ridge Isochronous Cyclotron. The program includes the simulation of high fluence neutron damage using heavy ion beams, examination of the effect of 14 MeV neutron bombardment on surfaces, the injection of He into materials to be subsequently irradiated, and various analytical techniques which involve accelerator beams. The experiments are directed not only toward an understanding of the fundamental mechanisms of radiation damage, but also toward development of techniques for rapidly testing the damage resistance of different alloys.

Introduction

The events of recent years have emphasized the need to develop nuclear power as a major source of energy as quickly as possible. Within the next few decades, we may expect to see the large-scale introduction of fast breeder reactors (FBR's) and possible controlled thermonuclear reactors (CTR's). Leaving aside the question of CTR feasibility, there still remain some formidable technical problems to be solved. In particular, there is an urgent need to develop materials which are resistant to the damage associated with the very high neutron fluxes to be encountered in such reactor cores. In this talk I should like to cover some of the general aspects of neutron-induced radiation damage, particularly those related to void production, and then discuss some of the current accelerator-based work at ORNL relating to both FBR and CTR materials research.

One of the major obstacles to materials research for the FBR program is the time-scale involved in reactor irradiations to the high fluences ($\sim 10^{23} n/cm^2$) which are expected in commercial reactors. It takes years to

*Research sponsored by U. S. Atomic Energy Commission under contract with Union Carbide Corporation.

achieve such doses in the available test facilities. Experiments are costly ($\approx \$10^6$) and difficult to instrument, and reactor space is severely limited. Until the FFTF goes critical the U. S. reactor studies must depend essentially on EBR-II and whatever space can be rented in, for example, the British Prototype Fast Reactor. The situation for CTR studies is even worse as there are no high flux neutron sources with the required energy spectrum. For these reasons there has been an upsurge of interest in accelerator-based research which, despite certain drawbacks, offers the enticing prospect of simulating years of reactor irradiation in a few hours of charged-particle bombardment. For some CTR studies accelerators are also used to generate high energy neutrons via the d-T reaction, the Be(d,n) reaction or spallation caused by high energy protons.

Radiation Damage

There are two principal mechanisms by which neutrons produce radiation damage in metals. Atoms are displaced from their normal lattice sites due to nuclear collisions, producing vacant sites and interstitials, and nuclear interactions cause a build-up of transmutation products, particularly Helium, inside the lattice. The evolution of this defect structure, and its interaction with the existing microstructure, result in the property changes which we class as radiation damage. These property changes may include drastic loss of ductility, enhanced creep rates and significant volume swelling.

This last effect was only recognized as a major problem comparatively recently. In 1966 Cawthorne and Fulton[1] reported the formation of voids which were not gas bubbles in fuel cladding removed from the Dounray Fast Reactor after two years of operation. These voids are clusters of vacancies which appear in most metals after high fluence irradiations (10^{20}-10^{22}n/cm^2) in the temperature range (0.35 - 0.55)Tm (Tm = absolute melting point). They are the underlying cause of significant volume swelling and other macroscopic property changes. Figure 1 shows the growth of such voids in high priority aluminum irradiated to various fluences in the HFIR.[2] The final picture corresponds to a volume swelling of about 7%. The 316 stainless steels chosen for use as fuel cladding and duct materials in FBR's are very susceptible to swelling, as much as 30% for their lifetime doses. This necessitates very conservative engineering to accomodate dimensional changes, leading to long breeding times and poor economics.

The vacancies which cluster to produce voids result from atomic displacements. These also occur when any charged particle passing through matter undergoes a collision which transfers sufficient kinetic energy (> 25 eV) to a lattice atom. Current studies of displacement damage use 1 MeV electrons from high voltage electron microscopes, protons, deuterons, alphas, and beams of heavier ions such as Ni from both Van de Graaffs and cyclotrons. It has been shown that the displacement damage caused by charged-particle bombardment also leads to void production, the damage being qualitatively similar to neutron-induced damage. At the end of the range of a heavy ion beam, where the nuclear stopping power dominates, displacement damage can occur at a rate 10^4 times that achieved in a reactor irradiation.

There are, however, complications in unravelling the swelling process. First, a means of comparing neutron and ion doses is required. The unit that is employed (dpa) is the average number of displacements each lattice atom has undergone during the bombardment. This must be calculated for both neutron and ion irradiations, requiring theoretical models for the stopping processes of very low energy recoil atoms. As this is one of the least well understood aspects of charged particle interactions in matter the dose calculations are rather uncertain.

The very fact that ion irradiations produce damage much faster than neutron irradiations has an effect. Figure 2 shows results of Johnston et al.[3] on the temperature dependence of swelling in stainless steel for 5 MeV Ni irradiations compared to fast reactor data. The shift toward higher temperatures can be qualitatively explained on the basis of the higher displacement rate for the ion irradiations.[4]

The existence of He as a transmutation product is extremely important as it encourages void nucleation. Unfortunately, the injection of He to some given concentration before irradiation does not give the same results as continuous injection to the same final concentration during an irradiation.[5] The group at Argonne are currently installing a dual accelerator system to permit simultaneous Ni and He bombardments.[6]

In the case of CTR neutron spectra the transmutation products appear in much greater quantities. Significant fractions of the first wall material will be transmuted during the reactor lifetime, and the effect on void production is not known.

The precise composition of an alloy is an important factor in swelling, as is the initial microstructure of the specimen. Most irradiations use ion species that are one of the major alloy constituents of the sample in order to avoid introducing impurities into the material. However, quite high concentrations of the incident ions may accumulate near the damage region, significantly altering the effective alloy constituents.

Finally, it should be noted that the damage is often produced in a thin (0.1 - 0.2μ) layer within 1μ or so of the surface of the sample. This region may not be representative of the bulk material. In any case, interpreting results from such a restricted region in terms of macroscopic property changes is not a trivial task.

Current Work at ORNL

Ion Bombardments

The accelerator facilities at ORNL are finding increased use in this area of research. The programs range from studies of fundamental damage mechanisms to developing methods of rapidly testing the swelling resistance of different alloys. Apart from the He injection program of Wiffen using a 50 MeV α beam from the ORIC, most current ion bombardments use the 4 MeV Ni beam at the CN Van de Graaff or the 50 MeV Ni beam from the ORIC.

Narayan, Noggle and Oen have used these Ni beams to produce displacement damage in Cu crystals. Using their sectioning technique they are able to examine the damage as a function of depth in the sample, estimating the number of point defects produced as well as the range of the incident ions. Figure 3 shows an example of their work with 1 MeV protons in Cu compared to the calculations of Oen. Their results for 57.8 MeV Ni ions in Cu indicate that existing range-energy calculations are correct at this energy to within a few percent, however, using 4.45 MeV ions their preliminary data indicate a range 25% greater than the accepted value. If this result proves to be correct, a possible explanation lies in fluctuations of stopping power as a function of target or projectile atomic number. This might be expected at low energies due to effects like level crossing which cause ionization cross-sections to be strong functions of target and projectile atomic numbers.[7] A good understanding of the stopping process is of course crucial to many damage studies in order to define not only the irradiation dose in terms of dpa, but also the region in which the damage occurs.

Packan has made initial irradiations with the 4 MeV Ni ions using high purity Nickel samples at dose levels ≈ 1-10 dpa in the temperature range 300° - 500°C. These were designed to simulate similar neutron irradiations of pure Ni done at 300°C in the ORR. The damage microstructure most evident in the reactor irradiations was aligned systems of dislocation loops. They were only evident in the ion irradiations performed at 400°C, illustrating the same type of shift in the temperature dependence as previously seen in swelling studies.

Other experiments are being done by Bloom, Farrell, and Packan to examine the effects of various surface preparation techniques on void production. Previous differences noted by a group from the CEGB[8] have been explained by them as due to differing amounts of hydrogen introduced by the various surface preparation techniques. The work at ORNL is designed to define acceptable surface preparation techniques to be used in other studies.

Bloom, Ullmaier, and I have begun a program to compare the results of precisely similar irradiations done at fairly high (70 dpa) dose levels on the ORIC, using 30-60 MeV beams and the Van de Graaff using 4 MeV beams. The object is to confirm that no differences exist in such irradiations that might be ascribed to the surface being rather close to the damage region.

Bloom and others are developing a method of quickly assessing the relative swelling resistance of many alloys, employing the sensitive profilometry technique used by Johnston et al.[9] This technique requires a portion of the sample to be masked from the beam. When swelling occurs in the damaged region the sample surface will be forced outward, except under the mask where no damage occurred. The surface then shows a step, the height of which is proportional to the volume swelling induced by the beam. By stacking eight samples in a small (5 mm x 9 mm) array it should be possible to survey many alloys rather quickly. The weakness of such a procedure is that one has to assume that the connection between neutron and ion-induced damage is fairly direct, and that it can be firmly established. In view of the known complexity of void production this is not an easy assumption to defend. However, in the case of the FBR program the time-scale in which material decisions must be made is rather short. This may be seen from the schedule listed in Table I.

Neutron Irradiations

The only neutron sources with spectra that approximate that incident on the first wall of a CTR are based on accelerators. The rotating target facility at Livermore[10] produces 14 MeV neutrons via the d-T reaction with fluxes up to $7 \times 10^{11} n/cm^2/sec$: The beam stop facility at LAMPF will produce an evaporation spectrum with a usable flux > $10^{13} n/cm^2$ when the accelerator is running at full intensity. Sources based on the $^9Be(d,n)$ reaction are in use at ORNL, NRL and the University of California at Davis, the NRL source being used exclusively for medical purposes. All these facilities use virtually identical cyclotrons and could produce neutron fluxes ≈ $2 \times 10^{12} n/cm^2/sec$:

Development of the ORNL neutron source was prompted by recent work of Kaminsky et al.[11] who reported very high yields for particle emission from cold-rolled Niobium surfaces which had been bombarded by 14 MeV neutrons to a total dose of 4.6×10^{15} neutrons/cm^2. "Chunk" emission, seen as micron-sized particles on the catcher foils, was estimated to give 0.25 ± 0.1 Nb atoms/neutron, while more uniform particle emission amounted to $(8.7 \pm 3) \times 10^{-3}$ Nb atoms/neutron. Both these figures are far higher than theoretical estimates, and if substantiated would imply serious erosion problems for the first wall of a CTR, and rapid poisoning of the plasma due to impurities from the wall.

The ORNL group, Jenkins, Noggle, Smith, Reed, and myself, have attempted to reproduce these results with neutron fluences up to 4 x 10^{16}n/cm^2. We used samples of both Nb and Au, employing a variety of surface finishing techniques. We saw no evidence for chunk emission, and overall yields were < 10^{-4} atoms/neutron. For the neutron source a beam of 40 MeV deuterons from the ORIC was used to bombard a thick (0.25 inch) water-cooled Beryllium target. The neutron energy spectrum for such a system has been measured at 0° by Schweimer,[12] and his data are shown in Figure 4. The yield is strongly peaked in the forward direction, resulting in a neutron beam with a divergence of approximately ± 10°. At the highest deuteron beam current available from the cyclotron the maximum neutron flux was ≈ 2 x 10^{12}n/cm^2/sec over a spot diameter ≈ 7 mm (full width half maximum). Samples were placed in a separate high vacuum system at distances between 6 mm and 21 mm from the Be target.

Foil activation dosimetry measurements were made using the ^{59}Co(n,2n) reaction, whose excitation function is also shown in Figure 4 as calculated from the formula of Pearlstein.[13] Figure 5 illustrates the method used to measure the spatial distribution of the neutrons. Small discs (1/16" in diameter) were punched from each detector foil in a radial pattern, and the activity of each disc measured using a calibrated Ge(Li) detector. These activities are shown in the figure arbitrarily normalized to 100 at the maximum. The data were then used to construct contour lines for the neutron beam spot as shown. It can be seen that the neutron spot was about 7 mm in diameter (full width half maximum) and slightly off center. The maximum fluence at the center was 4.0 x 10^{16}n/cm^2/sec: at the first sample position. The sample stack was basically a number of 1-inch-diameter discs separated by 0.010 inch thick annular spacers. The samples of Au and Nb were arranged, with high-purity Si catcher foils on either side. The Si surface adjacent to a sample was chemically polished. The five Nb and three Au samples were prepared in differing ways, the surfaces varying from single crystal to heavily worked (by discharge machining). The base pressure of the vacuum system before and after the irradiation was measured to be ≈ 10^{-10} Torr. During the irradiation a vacuum less than a few x 10^{-9} Torr could not be reliably measured, although we have no reason to believe that the pressure did rise during this period.

Initial examination of the catcher foils by both optical and electron microscopy failed to reveal any evidence for large particle emission. Subsequently the foils were analyzed in an effort to detect lower levels of ejected particles. Those catcher foils associated with the Au samples were all analyzed using neutron activation techniques. The limit on sensitivity was defined by the Au impurity levels already present in the Si foils, which was estimated from unused foils from the same batch. The measured yields and their uncertainties are listed in Table II. The error in the integrated fluence estimates has not been included, and would tend to reduce the quoted yields.

Some of the catcher foils associated with the Nb samples were analyzed by mass spectroscopy. No Nb was detected, and estimates of the sensitivity for the technique were made by introducing known quantities of neighboring isotopes into the system until a detectable limit was reached. An upper limit of 3 x 10^{12} atoms was established for the Nb content of these foils. Another catcher foil was analyzed using backscattering of 36.7 MeV Argon ions from the ORIC. Only a 3 mm diameter region in the center of the foil was examined. No Nb was detected by this method either. The detectable limit was established by comparison with a known standard. Table II shows the upper limits for the Nb sputtering yields.

TABLE I

Fast Breeder Schedule

(September 1974)

1976	Select 3-4 new duct materials
Dec. 1977	Fast Flux Test Facility (FFTF) goes critical
1978-1979	Select 3-4 new cladding materials
1980	Select final duct material for partial loading in FFTF or Demo
1982	Select cladding material for the reference fuel
1983	CRBRP goes critical (350 M Watt)
1984	Select cladding and duct materials for advance fuel
1993 ?	1000 M Watt commercial unit

TABLE II. Sputtering Yields

Sample Finish	Distance from front of array (mm)	Integrated Fluence ($\times 10^{15}$ neutrons)	Yield on catcher foil ($\times 10^{12}$ atoms)		Yield ($\times 10^{-4}$ atoms/neutron)	
			Forward	Backward	Forward	Backward
Au Electro-polished	1.8	4.0	< 2.9		< 0.7	
				< 1.4		< 0.3
Au Electro-polished	10.2	2.3	< 7		< 3.0	
(annular sample)				< 4		< 1.7
AuCold-rolled	13.2	2.5	< 1.9		< 0.7	
				< 3.2		< 1.3
Nb Single crystal	3.3	3.5	< 3		< 1	
Nb Discharge machine	4.8	3.3	< 3		< 1	
						< 1
Nb Single crystal	3.3	$2.4 \times 10^{16} n/cm^2$		0 ± 2		0 ± 0.8

REFERENCES

1) C. Cawthorne and E. J. Fulton, The Nature of Small Defect Structures, Vol. II, p. 446, Ed. by M. J. Makin, AERE-R-5269 (1966).

2) N. H. Packan, ORNL TM-3109 (1971).

3) W. G. Johnston, J. H. Rosolowski, A. M. Turkalo, J. Nuc. Mat. 48, 330 (1973).

4) A. D. Brailsford and R. Bullough, J. Nuc. Mat. 44, 121 (1972).

5) K. Farrel, Private communication.

6) A. Taylor, Private communication.

7) U. Fano and W. Lichten, Phys. Rev. Lett. 14, 627 (1965).
Werner Brandt and Roman Laubert, Phys. Rev. Lett. 24, 1037 (1970).

8) J. T. Buswell, S. B. Fisher, J. E. Harbottle, and D. I. Norris, Proceedings of the International Conference on Physical Metallurgy of Reactor Fuel Elements (1973), Berkeley Nuc. Lab., Gloucestershire, England.

9) W. G. Johnston, J. H. Rosolowski, A. M. Turkalo, and K. D. Challenger, Scripta Met. 6, 999 (1972).

10) R. Booth and H. H. Barschall, Nuc. Inst. Meth. 99, 1 (1972).

11) M. Kaminsky, J. H. Pearey, and S. K. Das, Phys. Rev. Lett. 32, 599 (1974).

12) G. Schweimer, Nuc. Phys. A 100, 537 (1967).

13) S. Pearlstein, Nuc. Data A3, 327 (1967).

PHOTO 98079

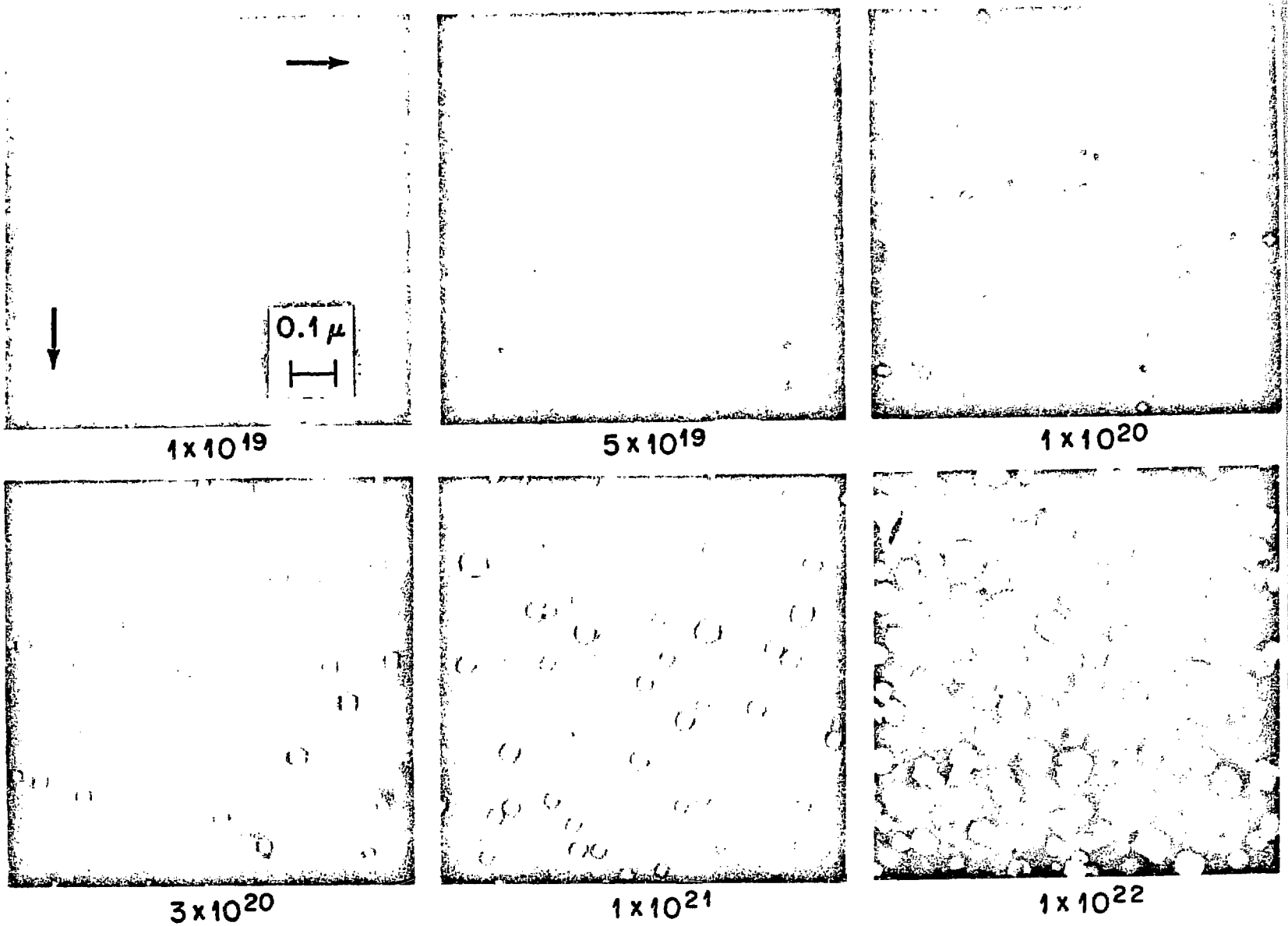


Figure 1

Examples of void production in high purity Al irradiated in HFIR. Fluences are in n/cm^2.

Fig. 2

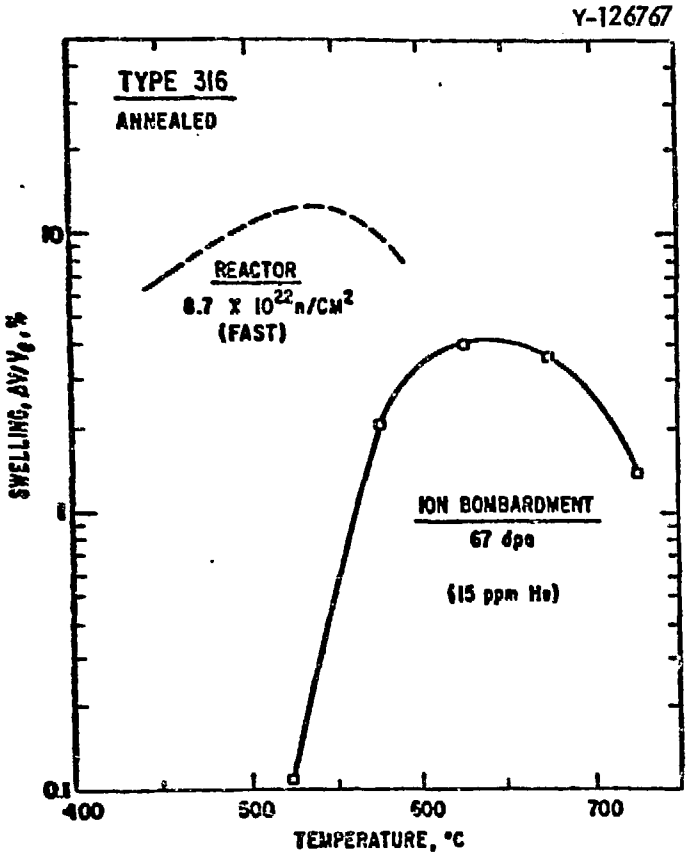
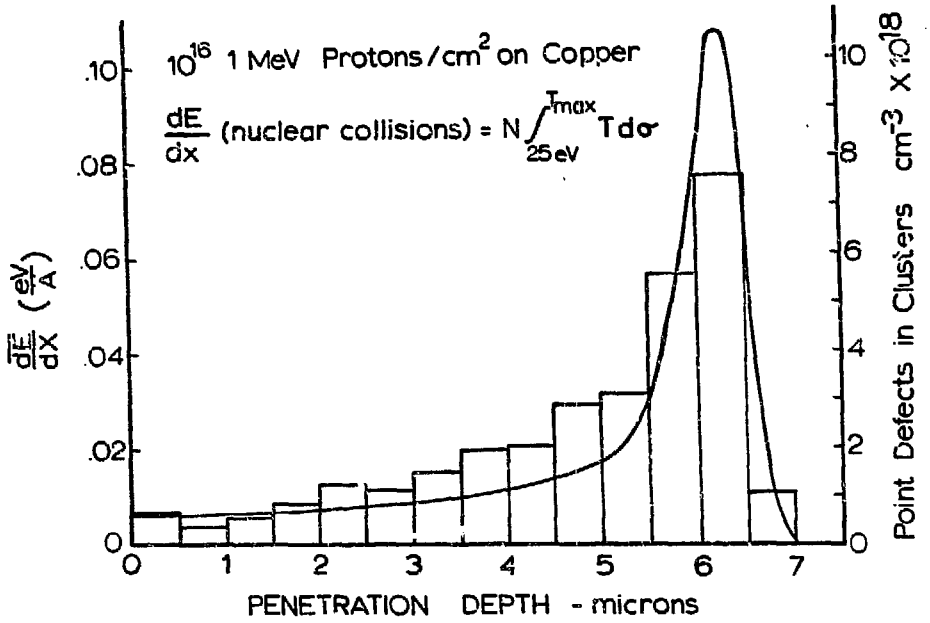

Y-126767
TYPE 316
ANNEALED
REACTOR
8.7 X 10^22 n/CM^2
(FAST)
ION BOMBARDMENT
67 dpa
(15 ppm He)
SWELLING, ΔV/V0, %
10
1
0.1
400
500
600
700
TEMPERATURE, °C

Figure 2

Temperature dependence of swelling in annealed type 316 stainless steel bombarded with 5 MeV nickel to a damage level of 67 dpa. The steel contained 15 ppm of cyclotron-injected helium. The temperature dependence of EBR-II core swelling is shown by the dashed curve. The displacement rates were $\sim 10^{-6}$ dpa/sec in EBR-II and 2×10^{-2} for the ion bombardment.

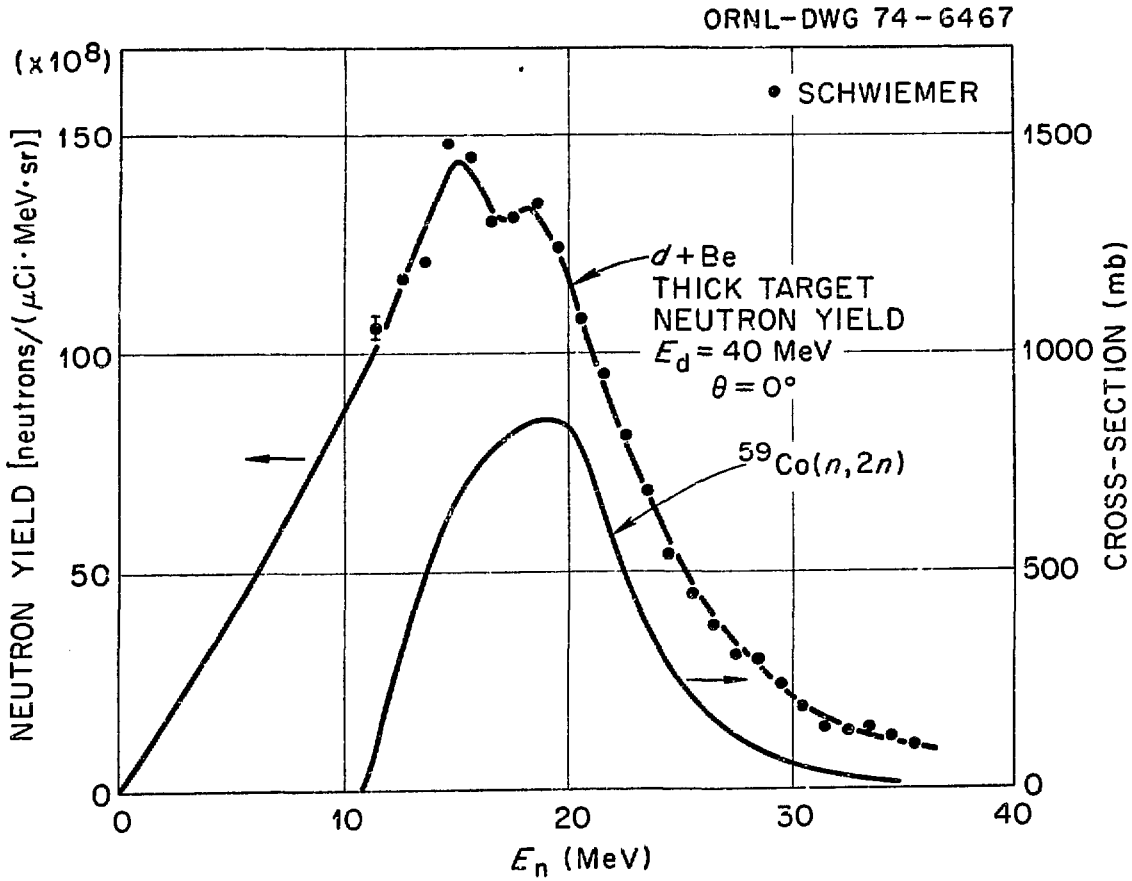
Fig. 3


10^{16} 1 MeV Protons/cm^2 on Copper
$\frac{dE}{dx}$ (nuclear collisions) = $N\int_{25eV}^{T_{max}} T d\sigma$
$\frac{dE}{dx}$ ($\frac{eV}{A}$)
.10
.08
.06
.04
.02
0
0 1 2 3 4 5 6 7
PENETRATION DEPTH - microns
10
8
6
4
2
0
Point Defects in Clusters cm^{-3} X 10^{18}

10μ

Figure 3

Damage induced by 1 MeV protons in Cu. The histogram represents data obtained by counting defects on electron microscope pictures. The smooth curve is from a calculation by Oen.

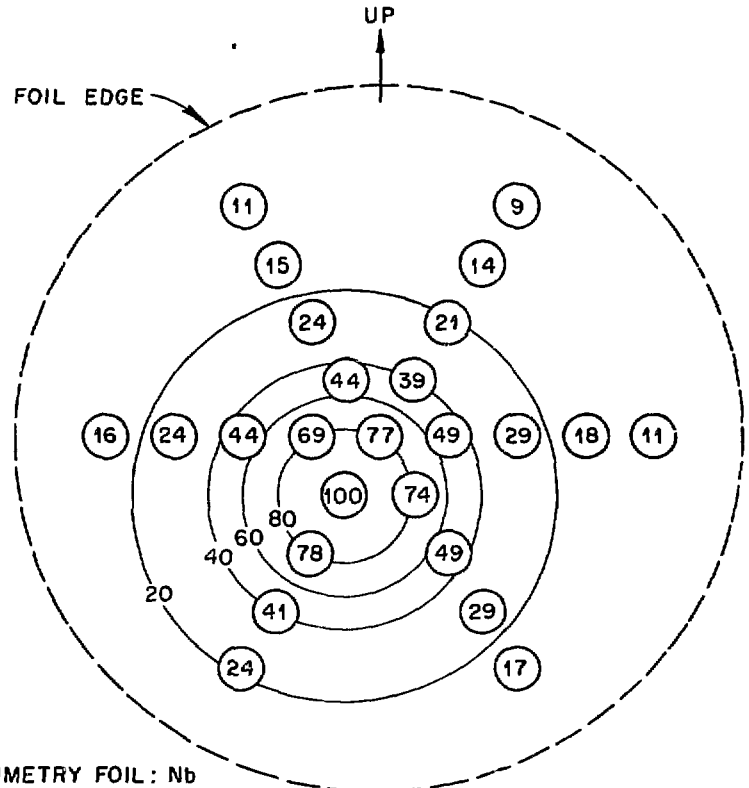


Neutron Energy Spectrum Compared to $^{59}Co\ (n,2n)$ Excitation Function.

Figure 4

The Be(d,n) neutron energy spectrum compared to the $^{59}Co(n,2n)$ excitation function.

ORNL-DWG 74-6468



DOSIMETRY FOIL: Nb
DIAMETER: 1.00 in.
THICKNESS: 0.002 in.
ACTIVITY: $E_\gamma = 934$ keV
NORMALIZATION: 100 = 241 c/sec

Figure 5

Measurements taken to establish the neutron beam spot profile on a 1 inch diameter dosimetry foil.