NEUTRON-SOURCE CHARACTERIZATION AND RADIATION-DAMAGE
CALCULATIONS FOR MATERIAL STUDIES*

CONF-811145-16
DE83 007857

L. R. Greenwood

Chemical Engineering Division
Argonne National Laboratory
9700 South Cass Avenue
Argonne, Illinois 60439

November 9, 1981

Proceedings of the International Conference on
Neutron Irradiation Effects at Argonne National
Laboratory, November 9-12, 1981

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of 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. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

*Work performed under the auspices of the U.S. Department of Energy

MASTER

WHP
DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Neutron-Source Characterization and Radiation-Damage Calculations for Material Studies

L. R. Greenwood
Argonne National Laboratory

Abstract

In our quest to understand radiation damage in materials, it is vital that we characterize radiation sources in terms of neutron flux and spectra as well as the more-fundamental displacement damage, gas production, and transmutation rates. Such data are crucial to correlations of materials-property changes in different environments and to predictions of materials performance in inaccessible environments, such as fusion reactors. Dosimetry techniques have been developed to measure the neutron flux and spectra in diverse facilities including thermal, fast, and mixed reactors, T (d,n) and Be (d,n) accelerator sources, and high-energy spallation sources. Displacement-damage cross sections have been calculated for 36 elements spanning the periodic table. All of these exposure parameters can now be routinely measured with 10-15% relative accuracy at all existing radiation-effect facilities.

Introduction

Since no fusion reactor currently exists for materials testing, experiments are currently in progress at a wide variety of facilities including fission reactors, particle accelerators, and high-energy spallation sources. The primary goal of our program is to characterize all of these facilities in terms of neutron flux and energy spectra as well as displacement damage, gas production, and transmutation. It is then hoped that these exposure parameters can be used to correlate property changes between facilities and to predict material properties in fusion reactors.

Figures 1 and 2 show an overview of some of the facilities currently in use for material studies. The weekly displacement and helium production rates are shown for iron in the highest flux regions of each facility. As can be seen, reactors currently produce the most displacement damage. However, accelerators can produce more helium with their high fluxes of fast neutrons, more closely simulating a fusion neutron energy spectrum, as shown in Figure 2. Fortunately, helium production in reactors can be greatly enhanced in nickel using the $^{58}\text{Ni}(n, \gamma)^{59}\text{Ni}(n, \alpha)$ thermal reaction. Clearly, accurate exposure data is crucial to any correlation of materials property changes between such diverse facilities. Furthermore, since no existing facility simulates the neutron environment expected in a fusion reactor, spectral effects must be understood before we can provide engineering data for the design of FED, INTOR, or other future fusion devices.

Neutron Dosimetry Techniques

The technique used to characterize these facilities involves multiple-material activation. In the best cases, more than 30 different nuclear reactions can be measured simultaneously. Cadmium covers and resonance detectors can be used to define the thermal and epithermal flux-spectrum.

Figure 1. Comparison of weekly DPA and He production rates for various irradiation facilities.

Figure 2. Integral recoil fractions for various neutron sources. The NSF and REF of the IPNS are compared to the Oak Ridge Reactor, a fission spectrum, a Be (d,n) source, and RTNS.

Threshold reactions can then be used to measure the fast neutron spectrum. Small packages of foils or wires are included with specimens during each irradiation. However, limitations of space, temperature, and decay time often necessitate separate, short irradiations to employ all of the reactions at our disposal. Following irradiation, samples are gamma counted with Ge(Li) spectrometers and activities are calculated correcting for the irradiation history and decay time, gamma and neutron self-shielding, burn-up or burn-in of materials, and flux and spectral gradients.

The saturated activities determined as above are equal to the energy integral of the neutron flux times the neutron activation cross section. A least squares technique is then used to minimize chi-square using the STAYSL¹ computer code. Variance-covariance matrices are defined for all input activities, the neutron cross sections, and the input flux spectrum. These data are then adjusted to predict the most probable neutron flux spectrum compatible with the data and stated uncertainties. Typical results are illustrated in Figure 3. The dotted and dashed lines represent standard deviations for each flux group. However, one must remember that these data are highly correlated. Hence, the uncertainties in integral fluxes or computed damage parameters are more typically 10-15%.

The reliability of this technique is crucially dependent on the accuracy of neutron cross section data taken primarily from ENDF/B-V,² extrapolated to 44 MeV for accelerator measurements.^{3,4} Integral data testing has thus been performed in a variety of fields and the results are listed in Table I. The T(d,n) results at the Rotating Target Neutron Source II at Lawrence Livermore Laboratory are in fact nearly monoenergetic cross section measurements near 14.9 MeV. The Be(d,n) measurements were performed at the Argonne Tandem Accelerator, the University of California at Davis Cyclotron,

Figure 3. Neutron spectra unfolded at the Oak Ridge Reactor (top), Experimental Breeder Reactor II (middle), and the newly-commissioned Intense Pulsed Neutron Source (bottom). The dotted and dashed lines represent one standard deviation. At least 28 activation reactions were measured in each case.

Table I. Results of Integral Cross Section Testing

Differential cross sections from ENDF/B-V, unless noted.
Estimated cross section errors are listed in percent.

Reaction	ORR ^a	RTNS II ^b	Be(d,n) ^c	
	Mixed-Spectrum	0°, 14.9 MeV	14-16 MeV	40 MeV
²⁷ Al(n,α) ²⁴ Na	+2	+7	+6	-1
⁴⁵ Sc(n,2n) ^{44m} Sc ^d	-	-3	-15	+3
Ti(n,x) ⁴⁶ Sc	-1	+10	-1	+24
Ti(n,x) ⁴⁷ Sc	+25	-30	+6	-56
⁴⁸ Ti(n,p) ⁴⁸ Sc	-2	-8	-1	+4
Fe(n,x) ⁵⁴ Mn	-3	-2	-3	+4
⁵⁴ Fe(n,α) ⁵¹ Cr ^d	-17	+2	+1	-20
⁵⁶ Fe(n,p) ⁵⁶ Mn	-	-	-2	-4
⁵⁵ Mn(n,2n) ⁵⁴ Mn	-2	-5	-	-
⁵⁹ Co(n,p) ⁵⁹ Fe ^d	-	+26	-4	+5
⁵⁹ Co(n,2n) ⁵⁸ Co	-	+4	+6	-3
⁵⁹ Co(n,α) ⁵⁶ Mn	-	-	-2	-5
⁵⁸ Ni(n,p) ⁵⁸ Co	-3	+13	-3	+3
⁵⁸ Ni(n,2n) ⁵⁷ Ni	-	-14	+1	+14
⁶⁰ Ni(n,p) ⁶⁰ Co	+13	-21	-2	+3
⁶³ Cu(n,α) ⁶⁰ Co	-4	-1	-	-
Zr(n,xn) ⁸⁹ Zr ^d	+7	-4	+9	-1
⁹³ Nb(n,2n) ^{92m} Nb ^d	-4	(+6) ^b	+6	+7
¹⁰⁷ Ag(n,2n) ^{106m} Ag ^d	-	+9	-	-
¹⁶⁹ Tm(n,2n) ¹⁶⁸ Tm ^d	-	-	-	+10
¹⁶⁹ Tm(n,3n) ¹⁶⁷ Tm ^d	-	-	-	-8
¹⁹⁷ Au(n,2n) ¹⁹⁶ Au ^d	-1	0	-8	+1
¹⁹⁷ Au(n,3n) ¹⁹⁵ Au ^d	-	-	-	+12
¹⁹⁷ Au(n,4n) ¹⁹⁴ Au ^d	-	-	-	+1
¹¹⁵ In(n,n') ^{115m} In	-	-	-2	-2
²³⁵ U(n,f)	-1	-	+8	+1
²³⁸ U(n,f)	-2	-	+4	-1
²³⁸ U(n,2n) ²³⁷ U ^d	-	-	+1	-11

^aSpectrum from neutronics calculation; values relative.

^bT(d,n) source; values normalized to ⁹³Nb(n,2n), 463 mb ± 7%.

^cSee References 5 and 6.

^dCross section not in ENDF; see Reference 2.

and the Oak Ridge Isochronous Cyclotron. At these facilities, time-of-flight spectrometry was used to measure the neutron energy spectrum, then foils were irradiated in a well-defined geometry.^{5,6} the reactor measurements are from the Oak Ridge Research Reactor and these data are only relative since the spectrum is taken from neutronics calculations. However, extensive testing in benchmark fission fields gives similar results.⁷ Of course, there are many other reactions, especially (n,γ) , which can be used in fission reactors, as illustrated in Figure 3.

The accuracy of unfolded neutron spectra, as shown in Figure 3, varies with neutron energy. Thermal and resonance detectors can be used to measure group fluxes to about 10-20%. Threshold detectors are also well-known and result in about 30% accuracy above 1 MeV. In the intermediate region, no reactions are very helpful and much larger errors are common from about 10-500 keV. At accelerator sources, no data is available above 28 MeV,³ although spallation reactions can in principle be used.⁸ In spite of these deficiencies, exposure parameters can usually be determined with 10-15% accuracy thanks to the strong correlations in the output flux spectrum. This can be easily understood since all of the exposure parameters are integral quantities as are the basic activation measurements which are typically accurate to $\pm 2\%$. Of course, improved cross sections are needed to reduce the uncertainties in the technique. In particular, the discrepancies in Table I should be corrected, cross sections should be extended to 50 MeV for accelerator sources (FMIT), and spallation reactions should be developed for spallation sources like IPNS. In addition, lower threshold reactions like $^{93}\text{Nb}(n,n^1)^{93\text{m}}\text{Nb}(16\text{ y})$ would improve the method below 1 MeV and more long-lived reaction products are needed for multi-year reactor experiments. Stable product monitors (e.g. - helium) are particularly useful for these lengthy experiments.

Displacement Damage Calculations

Neutron damage in conductors is primarily due to the displacement of atoms (DPA) from their lattice sites. These displacements are due to neutron-nucleus interactions, leading to a primary knock-on atom (PKA) spectrum, followed by recoil atom interactions, leading to secondary displacements. The fraction of the total recoil atom energy available for causing displacements is simply the nuclear stopping power fraction given by the Lindhard model.⁹ Electronic losses (as well as gamma irradiation) are assumed to produce no stable defects in conductors. This is of course not true for insulators and the total dose must be calculated in rads.

Displacement cross sections have been calculated for 30 elements up to 20 MeV using the DISCS¹⁰ computer code and neutron nuclear data from ENDF/B-V.² Exact calculations are performed for elastic scattering and an evaporation model is used for most other reactions assuming isotropic center-of-mass angular distributions. These assumptions are quite good for fission reactors and spallation sources, but are questionable at high neutron energies.

Typical damage-energy cross sections are illustrated in Figure 4. Partial cross sections are shown in Figure 5. As can be seen the elastic and inelastic reactions dominate damage below 10 MeV and are thus most important for fission reactors. The (n, xn) reactions are the most important above 14 MeV for accelerator neutron sources. Charged particle reactions are generally much weaker and contribute less than 10% of the total damage. Of course, (n, p) and (n, α) reactions are interesting in their own right since they lead to hydrogen and helium production in materials. Helium production is particularly important in fusion spectra since there may be a synergistic effect with the evolution of microstructure.

Figure 4. Damage energy cross sections are compared for Cu, Nb, and Au.

Figure 5. Partial damage cross sections are shown for various reactions for iron. Note that the elastic, inelastic, and (n, xn) reactions dominate at all energies.

Spectral-averaged displacement cross sections are listed for a variety of spectra in Table II. The fission values are also listed for neutron data from Version IV of ENDF.² As can be seen, all values agree within 10% (this is also true at 14 MeV). The thermal (n,γ) values were computed assuming that all of the capture gamma rays are emitted independently, as is true when nuclear lifetimes are longer than stopping times for the recoil atoms. Capture

Table II. Spectral-Averaged Damage Energy Cross Sections

To obtain displacement cross sections, multiply by 0.8/2E_d (Table III).

Element	$\langle\sigma_D\rangle$, keV - barn			
	Thermal (n,γ) ^a (2200 m/s)	14.5 MeV	²³⁵ U Fission	
			v ^b	IV ^b
Be	0.018	23.1	35.3	35.9
C	0.0030	39.0	52.0	51.3
Na	0.300	139.9	97.1	-
Mg	0.047	159.8	92.8	91.1
Al	0.187	176.9	96.3	98.3
Si	0.117	191.4	96.0	94.1
K	0.867	256.7	89.3	-
Ca	0.201	273.1	95.4	-
Ti	2.93	243.7	92.8	95.7
V	2.15	269.9	100.9	97.1
Cr	2.08	278.4	94.8	91.6
Mn	5.12	259.6	94.6	98.2
Fe	1.21	290.1	84.4	80.3
Co	13.47	294.1	81.7	85.7
Ni	2.62	300.1	85.0	82.2
Cu	1.57	296.0	79.2	81.3
Zr	0.030	258.9	86.0	73.1
Nb	0.144	270.9	79.5	79.5
Mo	0.315	259.2	83.5	83.4
Ag	8.65	229.6	71.1	71.2
Ta	0.094	215.5	53.5	-
W	0.259	197.2	50.8	-
Au	6.72	217.7	50.2	50.5
Pb	0.024	203.4	59.6	59.1

^aNeutron self-shielding must be included in most applications.

^bValues are compared for ENDF/B-V and -IV.

gamma energies and yields were taken from Orphan.¹¹ Table II lists displacement energies (below which atoms can't escape from their lattice sites) as well as true and effective average (n,γ) recoil energies. The true recoil energy is the average PKA energy. The effective value also includes the gamma multiplicity. For light elements, the multiplicity is high. For heavy elements, only a few gamma rays results in recoil energies above the displacement energy and the multiplicity is thus very low. Caution should be used in applying the (n,γ) values in Tables II and III. A correction should be made for electronic energy

Table III. Displacement Energies and (n,γ) Recoil Energies

The effective recoil energy is the true average PKA energy above E_d times the gamma multiplicity per neutron capture.

Element	Displacement Energy	(n,γ) Recoil Energy, eV	
	E_d , eV	True	Effective
Be	31	1188	1912
C	31	683	891
Na	25	297	566
Mg	25	279	745
Al	27	506	812
Si	25	334	730
K	40	209	413
Ca	40	206	468
Ti	40	367	481
V	40	360	426
Cr	40	515	670
Mn	40	281	385
Fe	40	404	473
Co	40	264	362
Ni	40	530	592
Cu	40	383	413
Zr	40	112	162
Nb	40	102	125
Mo	60	132	119
Ag	60	117	136
Ta	90	102	3
W	90	115	14
Au	30	71	68
Pb	25	140	140

losses using the Lindhard model; for most elements this correction will reduce the damage by 10-20%. It is also important to note that neutron self-shielding effects can substantially reduce the average damage for a finite-sized sample, especially when the thermal cross section is high (e.g. -Au and Co). Finally, if there is a significant epithermal flux, then resonance capture may change the cross section dramatically. Beta decay may also lead to displacements, although this reaction is not presently included in our calculations. Work is now in progress to treat (n, γ) events more exactly and to include beta-decay events.¹² Preliminary calculations for the beta-decay of ²⁸Al (2.8 MeV) indicate that the net effect will be to increase the total displacements from the (n, γ) reaction by about 50%.

Conclusions

The relative importance of neutron spectral information and displacement damage production is given in Table IV. The most important problem

Table IV. Spectral Dependence of Displacement Damage in Ni

Percent of displacements are shown in each energy range.

Fission Reactors			Accelerator Source ^c	
Energy, MeV	ORR ^a	EBR II ^b	Energy, MeV	Be(d,n), 40 MeV
<0.1	3.7	4.3	<1	2
0.1-0.4	9.2	25.8	1-5	4
0.4-1	13.8	27.3	5-10	11
1-2	24.8	20.0	10-15	23
2-4	32.2	15.9	15-20	28
4-6	11.2	4.8	20-25	19
>6	5.2	2.0	25-30	8
			>30	5

^aOak Ridge Research Reactor; mixed-spectrum, flux half-thermal and half-fast at core center.

^bExperimental Breeder Reactor II at Argonne National Laboratory; fast spectrum in blanket region (row 7).

^cZero degree spectrum measured at Oak Ridge Isochronous Cyclotron.

for reactor dosimetry is that 20-40% of the displacements are typically generated by neutrons between 0.01-1 MeV, an energy region that is very difficult to measure. It should also be pointed out that simple dosimetry using the ^{54}Fe or $^{58}\text{Ni}(n,p)$ reactions will completely ignore this energy region since the thresholds are above 1 MeV. Hence, one must use reactions with lower thresholds as well as neutronics calculations to completely define this important source of damage. Above 1 MeV there are many well-known threshold reactions and spectral unfolding can be done rather accurately up to about 30 MeV. Above this energy, there are no available cross section measurements and nuclear data is required. Fortunately, only about 5% of the damage is caused by neutrons above 30 MeV in FMIT-like spectra. However, it is not known if new damage processes arise at such high neutron energies and some reactions, such as spallation, are not included in our models.

In most cases, integral fluences and damage parameters can be determined to within relative accuracies of 10-15% using our dosimetry techniques. Absolute errors are not so well-known since no errors have been assigned to displacement cross sections. Below 10 MeV such errors probably range from 5-15% due to uncertainties in the nuclear data, although model-dependent errors are harder to assess. In any case, the present techniques can be used with some confidence to correlate data taken in a variety of facilities. Nuclear data measurements, new dosimetry techniques, and sensitivity studies which are now in progress will further refine our ability to characterize neutron sources for materials studies.

References

1. F. G. Perey, "Least Squares Dosimetry Unfolding: The Program STAYSL", ORNL-TM-6062 (1977); modified by L. Greenwood (1979).
2. Evaluated Nuclear Data File, Version V, National Neutron Cross Section Center, Brookhaven National Laboratory (1979).
3. B. P. Bayhurst, J. S. Gilmore, R. J. Prestwood, J. B. Wilhelmy, N. Jarmie, B. H. Erkkila, and R. A. Hardekopf, Phys. Rev. C12, 451(1975).
4. L. R. Greenwood, "Extrapolated Neutron Activation Cross Sections for Dosimetry to 44 MeV", ANL-FPP-TM-115 (1979).
5. L. R. Greenwood, R. R. Heinrich, M. J. Saltmarsh, and C. B. Fulmer, Nucl. Sci. Eng. 72, 175 (1979).
6. L. R. Greenwood, R. R. Heinrich, R. J. Kennerley, and R. Medrzychowski, Nucl. Technol. 41, 109(1978).
7. M. F. Vlasov, A. Fabry, and W. N. McElroy, "Status of Neutron Cross Sections for Reactor Dosimetry", IAEA, INDC (NDS)-84-L+M(1977).
8. J. T. Routti and J. V. Sandberg, Computer Physics Communications 21, 119(1980).
9. J. Lindhard, M. Scharff, and H. Schiott, Mat. Phys. Medd. Dan Vid Selsk., 33, 14(1963).
10. G. R. Odette and D. R. Dorian, Nucl. Technol. 29, 346(1976).
11. V. J. Orphan, "Line and Continuum Gamma-Ray Yields from Thermal-Neutron Capture in 75 Elements", GA-10248 (1970).
12. R. K. Smither and L. R. Greenwood, "Description of Neutron Capture and Beta-Decay Processes Leading to Displacement Production", DOE-ER-0046-5, p. 29 (May 1981).