RECENT DEVELOPMENTS IN NEUTRON DOSIMETRY AND DAMAGE
CALCULATIONS FOR FUSION MATERIALS IRRADIATIONS

L. R. Greenwood and R. K. Smither

Argonne National Laboratory

Argonne, Illinois USA

ABSTRACT

Fusion materials experiments require accurate neutron dosimetry measurements and damage calculations. Neutron cross section measurements are described for application at fission reactors, accelerator neutron sources, fusion reactors, and spallation neutron sources. For fission reactors work is in progress to resolve integral/differential discrepancies and to develop new reactions for dosimetry. Reactions for fusion reactors are designed for plasma diagnostics and waste generation. Spallation reactions are being measured to extend our techniques to 800 MeV for spallation neutron sources. The SPECTER computer code has been developed to routinely calculate displacement damage, gas production, recoil spectra, and total dose for 38 elements with a user specified neutron spectrum. New helium measurements and calculations are described. A new procedure is discussed for the thermal production of helium by nickel including previously neglected damage from the energetic ^{56}Fe recoils.

INTRODUCTION

Fusion materials experiments span a diverse variety of facilities, from fission reactors to high energy particle accelerators. In order to compare property changes between such different environments, it is first necessary to measure each radiation

MASTER

2440

environment as accurately as possible and then to calculate damage parameters which can be used to correlate materials effects. Although each type of facility presents new challenges, all have the same basic needs, namely, the measurement of nuclear data, the development of new techniques, the improvement of computer codes and procedures, and the need for interlaboratory comparisons and standardization. The following sections will describe some of the recent developments in the U.S. fusion materials program. Neutron cross section measurements and integral testing is in progress for fission reactors, fusion reactors, 14 MeV and broad-spectrum accelerator sources, and spallation neutron sources. New measurements and calculations of helium production are reported, especially concerning a new procedure for calculating the thermal production of helium from nickel and the previously neglected displacement damage from this process. The SPECTER computer code has been developed to routinely calculate displacement damage and gas production for a user specified neutron spectrum.

CROSS SECTIONS FOR FISSION REACTORS AND ACCELERATOR SOURCES

Most fusion materials experiments are being conducted in either mixed-spectrum or fast fission reactors due to the availability of large volumes and high damage rates in these facilities. Superconductors and insulators have been intensively studied at 14 MeV sources and some limited measurements have been conducted at Be(d,n) broad-spectrum sources in anticipation of the Fusion Materials Test Facility now under construction. We have been engaged in a program to measure and integrally test neutron activation dosimetry cross sections in a variety of neutron spectra. Both monoenergetic and integral measurements are being made with the eventual goals of providing new reactions for dosimetry and of correcting discrepancies between integral and differential measurements. Of particular interest for fission reactors is the $^{93}\text{Nb}(n,n')$ (14.6 y) reaction due to the low threshold and long-lived activity. Samples have been irradiated in a number of different spectra and we are currently engaged in developing counting facilities and procedures. Several reactions which have notable discrepancies between differential and integral measurements include $^{47}\text{Ti}(n,p)^{47}\text{Sc}$, $^{54}\text{Fe}(n,\alpha)^{51}\text{Cr}$, $^{60}\text{Ni}(n,p)^{60}\text{Co}$, $^{59}\text{Co}(n,p)^{59}\text{Fe}$, $^{58}\text{Ni}(n,2n)^{57}\text{Ni}$, $^{63}\text{Cu}(n,\alpha)^{60}\text{Co}$, and $^{65}\text{Cu}(n,p)^{65}\text{Ni}$. These reactions, as well as many others, have recently been measured in a well-known neutron field at Argonne in collaboration with Don Smith. These data, in conjunction with previous measurements at 14 MeV [1] and various Be(d,n) sources [2,3], as well as new monoenergetic measurements now being planned, will be used to adjust these cross sections. As an example, a preliminary test has been conducted for the $\text{Ti}(n,x)^{47}\text{Sc}$ reaction. The STAY'SL [4] computer code was used to readjust the cross section as shown in Figure 1. The ENDF/B-V

Figure 1. Cross section adjustment for the Ti(n,x)⁴⁷Sc reaction using the STAY'SL computer code. The results agree well with new measurements [5]. The adjustment started with ENDF/B-V cross sections and considered new integral activities and spectra.

values were used as the starting function and integral values and spectra from seven different reactors and accelerators were used in the readjustment. The adjusted cross sections are in good agreement with new differential measurements now in progress [5] demonstrating the utility of this technique. Similar adjustments are now in progress for many other reactions and this technique promises to significantly improve the accuracy and availability of dosimetry cross sections over a wide range of energies. Coupled with calculations and selected differential measurements, this technique could be used to provide the numerous cross sections needed for dosimetry, especially at higher neutron energies.

FUSION REACTOR CROSS SECTION MEASUREMENTS

Cross section measurements are being made for fusion reactor diagnostics, in collaboration with Princeton University. In particular, the $^{27}\text{Al}(n,2n)^{26}\text{mAl}$ (6 s) [6] and $^{54}\text{Fe}(n,2n)^{53}\text{Fe}$ (10 m) [7] cross sections have been measured near the 14 MeV thresholds. The steep rise in yield with neutron energy and the short half-lives make these reactions quite sensitive to the plasma ion temperature as shown in Figure 2. These reactions could thus be used as a very sensitive plasma diagnostic for a single shot of current reactors such as the Tokamak Fusion Test Facility at Princeton.

Measurements are also being made of very long-lived activities which are of interest in fusion activation, dosimetry, and waste management. In particular, the $^{27}\text{Al}(n,2n)^{26}\text{Al}$ (7.3 E+5 y) has been studied by intense irradiations at the Rotating Target Neutron Source II at Lawrence Livermore National Laboratory followed both by direct gamma counting and by a relatively new technique, accelerator mass spectrometry [6]. The $^{54}\text{Fe}(n,2n)$ reaction mentioned above is also of interest since ^{53}Fe decays to the long-lived ^{53}Mn (3.7 E+6 y). Cross sections to both reactions have been found to be significantly lower (by about a factor of two) than predicted, thereby reducing their concern for fusion activation and waste generation. Other long-lived isotopes now being studied include ^{92}Nb (3.2 E+7 y), ^{59}Ni (7.5 E+4 y), ^{93}Zr (1.5 E+6 y), and ^{93}Mo (3000 y).

SPALLATION CROSS SECTION MEASUREMENTS

Low temperature (4 K) radiation damage experiments are being conducted at the Intense Pulsed Neutron Source (IPNS) at Argonne, a new facility is being prepared at the Los Alamos Meson Physics Facility (LAMPF) at Los Alamos National Laboratory, and even more powerful spallation sources are being proposed both in the U.S.

Figure 2. Cross section measurements are shown for the $^{54}\text{Fe}(n,2n)^{53}\text{Fe}$ reaction. Superimposed on the cross section data are two neutron energy distributions for d-t plasmas with ion temperatures of 1 keV and 9 keV (shown in arbitrary flux units). ^{53}Fe can be used for plasma diagnostics. Since ^{53}Fe decays to ^{53}Mn , the reaction is also of interest to fusion waste management.

and Europe. These facilities produce high energy neutrons (500-800 MeV) requiring new nuclear data for dosimetry measurements and radiation damage calculations. Spallation cross sections themselves are ideal for dosimetry since they have very high thresholds and reasonably large magnitudes. We have thus measured spallation cross sections from Al to ^7Be , ^{22}Na , and ^{24}Na and from Cu to 20 different spallation products [8,9]. Foil packages were placed directly in the proton beam of the IPNS and activation measurements were made at eight energies between 30 and 450 MeV. Selected cross sections are shown in Figure 3 along with other data for iron [10]. Figure 4 shows some adjusted spectra at these facilities. Radiation damage calculations are also being studied at these higher neutron energies [12]. The cross sections have also been compared to semi-empirical models of Rudstam [13] and of Silberberg and Tsao [14]. These models only appear to be reliable within about a factor of two but are useful for predicting weak reactions and general systematic trends in the data. Further differential and integral measurements are now being planned to develop these cross sections for routine dosimetry. Other measurements have also been made to determine the proton flux directly in the beam envelope and the secondary proton flux at the neutron irradiation positions [11,15].

HELIUM PRODUCTION CROSS SECTIONS

Gas production and other transmutation is of particular interest in radiation damage studies since these products may strongly influence the evolution of materials property changes, especially swelling. Helium production is of particular interest and is being routinely measured in fusion materials experiments by Rockwell International. Samples have been jointly irradiated in all types of facilities permitting the direct comparison of radiometric and helium cross sections, especially at 14 MeV [16] and Be(d,n) [17] neutron sources. Recent measurements also indicate serious disagreements with ENDF/B-V [18] for several elements irradiated in fission reactors. In particular, the ratio of calculated-to-measured helium production in the Oak Ridge Research Reactor and in the High Flux Isotopes Reactor has been determined to be about 2.3 for Ti and about 0.60 for Cu [19]. Many other materials are now being studied with the eventual goals of improving helium production cross sections and of developing these data for dosimetry applications.

One of the most important helium producing reactions is for nickel since this thermal neutron effect allows us to obtain the high helium-to-displacement damage characteristic of fusion reactors during irradiations in mixed spectrum fission reactors. A new evaluation of the capture cross sections on ^{59}Ni has recently been published [20] allowing us to directly calculate helium production

Figure 3. Spallation cross sections used for neutron spectral adjustments at high energy neutron sources. Reactions plotted include Fe to ^{54}Mn and ^{51}Cr , Ti to ^{46}Sc , and Al to ^{22}Na and ^7Be .

Figure 4. Calculated and adjusted neutron spectra are compared for IPNS and LAMPF. Note that the adjustment can now be done up to 800 MeV using spallation cross sections for dosimetry.

in any neutron spectrum. Joint radiometric and helium measurements in a number of fusion materials experiments have been used to test these new cross sections. Forty-five samples of nickel have been irradiated in three different reactors to fluences between $1.0 \text{ E}+21$ and $1.0 \text{ E}+23 \text{ n/cm}^2$. Excellent agreement is found between the calculated and measured helium content with an average ratio of 0.982 and a standard deviation of 6% [20].

An interesting development concerning the thermal helium production reaction in nickel is the realization that the ^{56}Fe recoils (at about 340 keV) from the $^{58}\text{Ni}(n,\gamma)^{59}\text{Ni}(n,\alpha)^{56}\text{Fe}$ two-step reaction produce significant displacement damage. A detailed analysis of this process has recently been published [21] which shows that this effect will produce one displacement per atom for every 567 appm helium generated. In a mixed spectrum reactor such as HFIR, this effect will double the number of displacements produced in nickel and produce a 13% increase in 316 stainless steel at a neutron fluence of $1.0 \text{ E}+23 \text{ n/cm}^2$. This simple ratio (567 appm He/dpa) can be used to correct any previous data since the calculation has only a minor dependence on the neutron energy involved. It should also be pointed out that the relatively high energy of the ^{56}Fe recoil leads to damage processes characteristic of irradiations at 14 MeV and higher energy neutron sources.

THE SPECTER COMPUTER CODE FOR RADIATION DAMAGE

The SPECTER computer code has been developed to routinely calculate displacement damage, recoil spectra, gas production, and other damage parameters for materials irradiations. All of the damage cross sections are derived from the DISCS [22] computer program using ENDF/B-V cross sections. A user need only specify a neutron spectrum of interest. The code will then provide spectral-averaged damage rates and compute net damage for a specified length of irradiation. The SPECTER damage library contains data for 38 elements spanning the periodic table. For each element the code lists displacements for elastic, inelastic, (n,xn) , (n,px) , and $(n,\alpha x)$ reactions. Recently we have added damage calculations for (n,γ) capture and any subsequent beta decay [23]. Gas production is included for each element; however, the new ENDF/B-V gas production file is now included separately [17]. Total dose values (Kerma) are also calculated for each irradiation using the MACKLIB program [24]. Recoil atom distributions for each element and each reaction are also presented in both differential and integral form. Provision is made for specifying a covariance uncertainty matrix for the input spectrum; this is routinely done for fusion experiments using the output of our spectral adjustment program STAY'SL [4]. SPECTER will then determine the uncertainty in each damage calculation due to the uncertainty in the flux

spectrum. Of course, this does not consider the uncertainty in the damage cross sections themselves, a deficiency of all present damage calculations which must eventually be addressed.

At present, only six elements have been extended beyond the 20 MeV cutoff in ENDF for use at accelerator neutron sources. These files were extended using the available data and calculations in the literature, although some reactions had to be extrapolated due to the general lack of adequate data beyond 14 MeV. For FMIT-type sources as well as lower energy spallation sources such as IPNS this deficiency is not so serious since only a small fraction of the damage (5-10%) is caused by enutrons with energies above 20 MeV. However, recent measurements at LAMPF (800 MeV protons on copper target) indicated that about 26% of the dpa and nearly 95% of the helium in copper is generated by neutrons above 20 MeV. In this case the VNMTC code was used to compute the higher energy damage [12]. However, the SPECTER and VNMTC calculations do not match very well indicating a serious need to address the problem of damage calculations at intermediate energies in the range from 20 to 800 MeV.

CONCLUSIONS AND FUTURE DIRECTIONS

Recent measurements of neutron cross sections are being used to improve and to expand our capabilities for dosimetry measurements and damage calculations. Some outstanding discrepancies between differential and integral experiments will be resolved and new reactions will be developed for routine use. However, there still remains a paucity of reliable data above 14 MeV and further experiments and calculations are badly needed, especially for spallation sources. Several new techniques are very promising, especially the use of spallation reactions at energies above 40 MeV and accelerator mass spectrometry for the measurement of very long-lived or stable isotopes. This latter technique offers the promise of measuring transmutation to stable products directly, a feature which may be of particular importance in fusion reactors.

Dosimetry at fusion reactors will soon become practical as new facilities such as TFTR and JET approach breakeven conditions. Some of the reactions which we are developing could be very useful both for dosimetry and as a plasma diagnostic. However, testing of these and other reactions must await the high power (d,t) operation.

Nuclear data is of course needed for many elements over a wide range of energies to improve the quality of the damage calculations. A carefully planned mix of calculations and selected measurements is needed. The theory and codes must also be improved

to consider alloys and insulators; at present only pure elements are included in an assumed metallic form. Finally, uncertainties should be assigned to the damage calculations. This is not easily done at present since uncertainty data is only included for a few reactions in ENDF. Assuming the inclusion of such data in future versions of ENDF and using sensitivity studies of model uncertainties, we plan to calculate uncertainties in damage rates and to interface these uncertainties with flux and spectral uncertainties to obtain more realistic estimates of damage parameters.

REFERENCES

- [1] L. R. Greenwood, J. Nucl. Mater. 108, 21-27 (1982).
- [2] L. R. Greenwood, R. R. Heinrich, R. J. Kennerley, and R. Medrzychowshi, Nucl. Technol. 41, 109-128 (1978).
- [3] L. R. Greenwood, R. R. Heinrich, M. J. Saltmarch, and C. B. Fulmer, Nucl. Sci. Eng. 72, 175-190 (1979).
- [4] F. G. Perey, ORNL/TM-6062 (1977); modified by L. R. Greenwood (1979).
- [5] D. L. Smith, Argonne National Laboratory, private communication (1984).
- [6] R. K. Smither and L. R. Greenwood, J. Nucl. Mater. 122, 1071-1077 (1984).
- [7] R. K. Smither and L. R. Greenwood, Damage Analysis and Fundamental Studies Quarterly Progress Report, DOE/ER-0046/17, May 11-13, 1984.
- [8] L. R. Greenwood and R. K. Smither, Damage Analysis and Fundamental Studies Quarterly Progress Report, DOE/ER-0046/18, August 1984.
- [9] L. R. Greenwood and R. K. Smither, Damage Analysis and Fundamental Studies Quarterly Progress Report, DOE/ER-0046/18, August 1984.
- [10] R. Michel and R. Stuck, J. Geophysical Res. 89, Supl. B673-684 (1984).
- [11] D. R. Davidson, L. R. Greenwood, R. C. Reedy, and W. F. Sommer, Measured Radiation Environment at the LAMPF Irradiation Facility, Proc. Twelfth International Symposium on the Effects of Radiation on Materials, Williamsburg, VA, June 18-20, 1984.

- [12] D. R. Davidson, L. R. Greenwood, W. F. Sommer, and M. S. Wechsler, Calculation of Displacement and Helium Production at the LAMPF Irradiation Facility, *ibid.*
- [13] G. Rudstam, Z. Naturforsch. 21A, 1027 (1966).
- [14] R. Silberberg and C. H. Tsao, Astroph. J. Suppl. 25, 315-367 (1973); parameters updated in succeeding International Cosmic Ray Conferences.
- [15] L. R. Greenwood and R. J. Popek, Proc. of the Sixth Meeting on the Int. Coll. on Adv. Neutron Sources, ICANS VI, ANL-82-80, 605-618, 1982.
- [16] D. W. Kneff, H. Farrar IV, and L. R. Greenwood, J. Nucl. Mater. 122, (1984).
- [17] D. W. Kneff, H. Farrar IV, L. R. Greenwood, and M. W. Guinan, Symp. on Neutron Cross Sections from 10-50 MeV, BNL-NCS-51245, 113-132, (1980).
- [18] Evaluated Neutron Data File Part B, Version V, National Neutron Cross Section Center, Brookhaven National Laboratory, 1979.
- [19] B. M. Oliver, D. W. Kneff, and R. P. Skowronski, Damage Analysis and Fundamental Studies Quarterly Progress Report, DOE/ER-0046-18, August 1984.
- [20] L. R. Greenwood, D. W. Kneff, R. P. Skowronski, and F. M. Mann, J. Nucl. Mater. 122, 1002-1010 (1984).
- [21] L. R. Greenwood, J. Nucl. Mater. 115, 137-142 (1983).
- [22] G. R. Odette and D. R. Dorion, Nucl. Technol. 29, 346 (1976).
- [23] R. K. Smither and L. R. Greenwood, Proc. Fourth ASTM-EURATOM Symp. on Reactor Dosimetry, NUREG/CP-0029, 793-805 (1982).
- [24] Y. Gohar and M. A. Abdou, ANL-FPP/TM-106 (1978).

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.