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for Fusion Materials Irradiations**

L. R. Greenwood

MASTER

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

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New Ideas in Dosimetry and Damage Calculations for Fusion Materials Irradiations

L. R. Greenwood
Chemical Technology Division
Argonne National Laboratory
Argonne, Illinois 60439 USA

Abstract

Nuclear data and techniques are being developed to improve our ability to characterize fusion materials irradiations in terms of neutron fluence and spectra as well as damage parameters such as atomic displacements, gas production, and other transmutation. The production of long-lived isotopes are being measured near 14 MeV, including reactions from Al, Fe, and Mo to ^{26}Al , ^{53}Mn , and ^{94}Nb , and measurements for other reactions are in progress. These data can be used for fusion reactor dosimetry, plasma diagnostics, and the estimation of radioactivity in fusion reactor materials. Dosimetry cross sections are being measured for fission reactors using Be(d,n) sources ($E_d = 7\text{--}40$ MeV) and monoenergetic neutron sources from 1 to 14 MeV. These data can be used to adjust activation cross sections. Spallation cross sections are being measured for higher-energy accelerator neutron sources. Helium production cross sections are being tested at all fusion irradiation facilities. We have recently discovered a new thermal helium production in copper, similar to the well-known effect in nickel. Recommended procedures have been published for the calculation of helium from both copper and nickel in mixed-spectrum reactors. A new program SPECOMP has recently been developed to calculate displacement damage for compounds. This program uses the SPECTER recoil atom distributions and integrates over secondary displacement functions for each combination of incident ion and matrix material to determine displacement cross sections for alloys, insulators, and breeder materials.

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Introduction

Fusion materials irradiations are being conducted in a variety of facilities including the 14 MeV T(d,n) neutron source, the Rotating Target Neutron Source II (RTNS II) at Lawrence Livermore National Laboratory; fast reactors, such as the Experimental Breeder Reactor II (EBR II) at Argonne National Laboratory; mixed-spectrum reactors, such as the High Flux Isotopes Reactor (HFIR) at Oak Ridge National Laboratory; and higher-energy accelerator neutron sources. Since none of these sources really simulates a fusion reactor environment, it is our goal to understand the irradiation characteristics of each facility so that materials performance can be correlated between them and eventually extrapolated to fusion conditions. Neutron flux and energy spectra are measured in each of these facilities using a neutron activation technique to adjust calculated neutron spectra. The induced radiation damage is then calculated using our SPECTER [1] computer code which provides the total damage, atomic displacements (dpa), gas production (H,He), and recoil atom energy spectra. These methods depend on the availability of accurate nuclear cross sections and their uncertainties. Hence, much of our recent research has focussed on the development and testing of nuclear data, as summarized below.

Integral Cross Section Measurements

Most of the nuclear cross sections which we require can be tested by performing integral experiments. If we can determine the neutron spectrum accurately by time-of-flight spectroscopy or other non-activation methods, then we can test our nuclear data and spectral unfolding techniques. Such experiments have been

performed using the Be(d,n) reaction with deuteron energies between 7 and 40 MeV [2-4]. Differential cross sections have also been measured near 14.7 MeV at the RTNS II [5]. Some of these reference neutron spectra are shown in figure 1. Combining all of these data along with previous measurements allows us to adjust the neutron cross sections. These adjustments were performed with the STAY'SL [6] computer code which performs a generalized least-squares calculation considering uncertainties in the cross sections, activation measurements, and reference spectra. This technique is especially valuable in cases where the cross sections are not well-known or difficult to measure more directly. Figure 2 shows the cross section adjustments for the relatively unknown $^{54}\text{Fe(n},\alpha)^{51}\text{Cr}$ and $^{59}\text{Co(n,p)}^{59}\text{Fe}$ reactions. In the case of $^{47}\text{Ti(n,p)}^{47}\text{Sc}$ our adjusted cross sections agree very well with new measurements [7]. At higher energies, especially above 14 MeV, this method readily provides data in an energy region where direct measurements are difficult. It should be emphasized that this technique is not a replacement for the usual differential measurements and in fact uses both types of measurements to test and adjust all known data.

At higher energies, spallation cross sections are being developed for dosimetry up to 800 MeV. These reactions produce many radioisotopes from the same target, each having a different threshold between 30 and 800 MeV, and hence are ideal for dosimetry applications. Measurements have been completed on Al and Cu [8] and the technique has been tested at the Intense Pulsed Neutron Source at Argonne and the Los Alamos Meson Physics Facility [9]. A paper describing our recent work at LAMPF is also included in these proceedings [10].

Cross Sections for Fusion Reactor Applications

Nuclear data is being developed for several fusion reactor applications. Neutron activation cross sections for 22 dosimetry reactions have been measured at several energies near 14 MeV at the RTNS II [5]. The reactions $^{27}\text{Al}(n,2n)^{26\text{m}}\text{Al}$ (6.3 s) [11] and $^{54}\text{Fe}(n,2n)^{53}\text{Fe}$ (8.5 m) [12] have been shown to be especially useful for fusion plasma diagnostics due to their extreme sensitivity to the energy and width of the neutron energy spectra near 14 MeV. These reactions are also of interest in activation calculations for maintenance or waste disposal considerations due to the very long-lived $^{26\text{g}}\text{Al}$ (7.2×10^5 y) and the decay of ^{53}Fe to ^{53}Mn (3.7×10^6 y).

The 14 MeV production cross sections to several other long-lived isotopes in fusion reactor materials are also being measured. Experiments have been completed for the production of ^{94}Nb (2.03×10^4 y) from ^{94}Mo and $Nat\text{Mo}$ [13]. The results are shown in Table I. The measurements were made at the RTNS II for an exposure of 81 days over a period of 7 months. This long exposure then permitted us to directly measure the gamma emissions from ^{94}Nb using Ge gamma spectroscopy. Measurements are also reported in Table I for the reactions $^{92}\text{Mo}(n,2n)^{91}\text{Mo}$ to $^{91\text{m}}\text{Nb}$, $^{92}\text{Mo}(n,\alpha)^{89}\text{Zr}$, and $^{95}\text{Mo}(n,p)^{95}\text{Nb}$. The reaction to $^{91\text{m}}\text{Nb}$ is also of interest since this isotope decays to the ground state $^{91\text{g}}\text{Nb}$ which has a half-life of about 700 y. Our data provides a good estimate of the production cross section to this neglected isotope, as shown in Table I. These new measurements allow us to calculate the production of ^{94}Nb and $^{91\text{g}}\text{Nb}$ in fusion reactor materials. A first-wall irradiation of Mo to 10 MW/m^2 will produce $19 \mu\text{Ci/g}$ of ^{94}Nb and 0.35 Ci/g of $^{91\text{g}}\text{Nb}$. Measurements on other long-lived iso-

topes including ^{56}Fe (2.7 y), ^{63}Ni (100 y), ^{59}Ni (7.6×10^4 y), and ^{93}Mo (3500 y) are now in progress using a variety of techniques including radiochemistry, liquid scintillation counting, and accelerator mass spectrometry.

Thermal Helium Production In Copper

Helium cross sections are being measured at all fusion irradiation facilities in collaboration with Rockwell International. A paper describing this work in HFIR is also included in these proceedings [14] and measurements at 14 MeV have recently been submitted for publication [15].

There is a well-known thermal helium production process in nickel which is being widely used to simulate fusion-like helium to displacement damage ratios in HFIR and other mixed spectrum reactors. We have recently published a comparison of measurements and calculations for this process which are in excellent agreement [16] and have also described an increase in displacements caused by the energetic ^{56}Fe recoils [17].

We have recently discovered a similar mechanism in copper whereby ^{63}Cu undergoes successive neutron captures and decays to form ^{65}Zn which has been shown to have a large thermal neutron cross section for the production of helium [18]. A combination of radiometric and mass spectrometric data from Argonne and helium measurements from Rockwell International uniquely determine all of the cross sections in this reaction chain, as shown in Table II. As shown in figure 3, we can accurately predict the level of helium in copper irradiated in HFIR to fluences of about 10^{23} n/cm². This reaction is of interest to fusion materials irradiations involving either copper or zinc since the process will produce more helium than

anticipated. Similar to the thermal effect in nickel, this effect on copper can be used to enhance the helium-to-displacement damage ratio thereby improving the simulation of fusion reactor damage for irradiations in mixed-spectrum reactors, such as HFIR. However, the effect in copper is not as pronounced as in nickel since three steps are required rather than two. Nevertheless, this process will equal the fast helium production in only a month in HFIR and will produce ten times more helium after a year. A crude equation describing this effect in HFIR is, as follows:

$$\text{Enhanced } ^4\text{He (appm)} = 0.669 \times \phi^{2.58}$$

where ϕ is the thermal neutron fluence in units of 10^{22}n/cm^2 . It must also be noted that our data shows that the fast neutron helium production is 31% higher than in ENDF[19]. The thermal reaction on Zn will also produce some extra displacement damage in the ratio of 1 dpa per 492 appm extra helium; however, this is insignificant relative to the fast neutron damage unlike the substantial damage effect in nickel.

New SPECOMP Computer Code for Damage in Compounds

The SPECTER[1] computer code is routinely used to calculate displacement damage and gas production in 38 elements of interest to the fusion materials program. However, the code does not calculate damage in alloys, compounds, or insulator materials since one has to consider all possible interactions of recoil atoms with atoms in the matrix [20]. A new addition to the SPECTER computer code package has recently been developed which performs damage calculations for compound materials. The code SPECOMP is based on the SPECTER primary recoil libraries. Given the composition of any mixture of elements in SPECTER,

SPECOMP integrates over the primary recoil distributions times the secondary recoil functions using a Lindhard model for each combination of recoil atom and matrix atom according to the prescription of Parkin, et. al.[20]. A sample calculation for LiAlO_2 is shown in figure 4 and compared to the simple sum of the elemental calculations for Li, Al, and O. As can be seen, the difference between simply adding the elemental damage and performing exact calculations for the compound are significant (20-40%) above 0.1 MeV. At lower energies the damage is dominated by the ${}^6\text{Li}(n,\alpha)t$ reaction. For the sake of comparison, the threshold displacement energies were the same in both cases. However, this is not generally the case and more work needs to be done to determine suitable threshold energies for each element in each compound of interest. Calculations for other alloys, compounds, and insulator materials are in progress. We plan to publish a more complete description of the code with damage functions for various materials and to make the code available for general use. One of the advantages of this approach with SPECTER and SPECOMP is that it is not necessary to recalculate the recoil atom energy distributions. Consequently, the codes are relatively small and can be run cheaply and quickly on small computers and they require no access to the ENDF data files.

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Figure Captions

1. Neutron energy spectra measured with time-of-flight spectroscopy are shown for thick-target Be(d,n) experiments at deuteron energies of 7, 16, 30, and 40 MeV. These spectra are used to test and adjust neutron activation cross sections.
2. Cross section adjustments are shown for the $^{54}\text{Fe}(n,\alpha)^{51}\text{Cr}$, and $^{59}\text{Co}(n,p)^{59}\text{Fe}$ reactions. The starting data is taken from BNL-325 [21] (solid lines) and the adjustments are shown as dotted lines.
3. Copper helium production measurements in HFIR (squares), shown as a function of the thermal neutron fluence, are compared to the usual ENDF/B-V [19] fast helium calculations (dashed line). The large enhancements at high fluences are well-fit by our calculations (dotted line).
4. The calculated displacement damage cross section for LiAlO_2 from SPECOMP (solid line) is compared to the elemental sums Li, Al, and O from SPECTER (dotted line). Large differences (20-40 %) are seen above 100 keV. The $^6\text{Li}(n,\alpha)t$ reaction dominates at low energies.

Table I
Measured Cross Sections (mb) for Mo

Reaction	14.55	14.60	14.78	14.80	$\pm\%$ ^a
⁹⁴ Mo(n,p) ⁹⁴ Nb	57.2	-	53.1	-	11
<i>Nat</i> Mo(n,x) ⁹⁴ Nb ^b	-	7.9	-	7.8	12
⁹⁵ Mo(n,x) ⁹⁴ Nb ^c	-	16.3	-	18.3	16
⁹⁵ Mo(n,p) ⁹⁵ Nb	40.4	-	37.1	-	9
⁹² Mo(n,x) ^{91m} Nb ^d	157.	153.	145.	145.	10
⁹⁸ Mo(n, α) ⁹⁵ Zr	6.56	6.56	6.24	6.21	8
⁹² Mo(n,x) ^{91g} Nb	≈ 300				
<i>Nat</i> Mo(n,x) ^{91g} Nb	≈ 45				

^aMajor sources of uncertainty include neutron fluence (7%)

⁹⁴Nb half-life (8%), efficiency (1.5%), statistics (1%)
deconvolution of ⁹⁵Mo (12%) for ⁹⁴Nb, (2%) for ⁹⁵Nb

^bSum of reactions from ^{94,95,96}Mo.

^cSum of (n,d+np+pn) reactions.

^dSum of (n,2n+d+np+pn) reactions.

Table II

Thermal Cross Sections(b) for He Production in Cu

Reaction	Data ^a	ENDF/B-V
$^{64}\text{Cu}(n,\gamma)^{65}\text{Cu}$	270 ± 176	< 6000
$^{65}\text{Zn}(n,\text{abs})^b$	66 ± 8	-
$^{65}\text{Zn}(n,\alpha)^{62}\text{Ni}$	4.7 ± 0.5	250 ± 150

^aData measured in HFIR with 7% epithermal flux

^bTotal absorption includes (n, γ), (n,p), and (n, α)