SAND2011-1542C

???? 225

Neutron Flux Characterization Techniques for Radiation Effects Studies

J. Graham[1]
joseph.graham@mail.utexas.edu
Phone: (719) 244-0042
Fax: (512) 471-4589

S. Landsberger[1]
s.landsberger@mail.utexas.edu

P. Ferreira[2]
ferreira@mail.utexas.edu

J. Ihlefeld[3]
jilhefe@sandia.gov

G. Brennecka[3]
glbrenn@sandia.gov

ADDRESSES:
[1]University of Texas at Austin, Nuclear Engineering Teaching Lab, R-9000 Austin, Texas USA 78712
[2]University of Texas at Austin, Deparment of Material Science and Engineering, Austin, Texas USA 78712
[3]Sandia National Laboratories, Albuquerque, New Mexico USA 87185

Neutron Flux Characterization Techniques for Radiation Effects Studies

J. Graham[1], S. Landsberger[1], P. Ferreira[2], J. Ihlefeld[3], G. Brennecka[3]

[1]University of Texas at Austin, Nuclear Engineering Teaching Lab, R-9000 Austin, Texas USA 78712
[2]University of Texas at Austin, Department of Material Science and Engineering, Austin, Texas USA 78712
[3]Sandia National Laboratories, Albuquerque, New Mexico USA 87123

In the field of radiation effects in materials, a detailed and precise description of the radiation environment used to damage samples is often required to make sense of subsequent materials analysis. The types of reactions and extent of damage that occur during irradiation strongly depend on the flux spectrum of the particular facility. Different neutron activation techniques for characterizing neutron flux spectra were performed on the University of Texas at Austin TRIGA research reactor's in-core facilities. The results were compared in terms of spectral detail and precision. Activation of Au foils with multiple correction factors, and multiple foil activation employing different deconvolution techniques comprise the methods tested. A sensitivity analysis for each technique was performed and the relative benefits of the different techniques are presented.

This work is supported, in part, by the National Institute of Nano Engineering and the Laboratory Directed Research and Development program at Sandia National Laboratories. Sandia is a multi-program laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the U.S. Department of Energy under contract DE-AC04-94AL85000.

Keywords: neutron flux, radiation effects, activation, foils

Introduction

When characterizing radiation damage to materials it is important to know what types of particle-atom interactions are taking place and the energetics of such interactions. The mechanism and degree of effect often vary dramatically with the energy of the incident particle. One category of damage effects are those due to atomic displacements as these types of crystallographic defects can permanently alter the mechanical, chemical and electrical properties of a material. Neutron collisions with matter are an important source of displacement effects since they are neutral and heavy enough to transfer large amounts of energy to atomic centers. At energies greater than tens of electron volts, elastic and inelastic collisions can transfer enough kinetic energy to a stationary target nucleus to remove it from its bound state in the crystal lattice. This type of interaction is called a knock-on event. Knock-on effects typically exhibit a fine energy structure which reflects the underlying resonance regions in the nuclear scattering cross sections as well as material resonances which originate from crystallographic focusing and channeling. Thus, in the context of neutron irradiation, knock-on effects are very sensitive to the energy dependent neutron flux. Below thermal neutron energies, knock-on interactions aren't energetic enough to result in atomic displacements. Radiative capture cross sections, however, are large making activation damage possible. If, during

decay, sufficient recoil is imparted to the daughter nucleus the daughter atom may also be displaced. Such events are called recoil events. For thermal reactors the vast majority of neutrons are at energies below 10 eV thus recoil events are the dominant mechanism for displacement damage. In contrast to knock-on cross sections, recoil displacement cross sections exhibit little fine structure. Mostly they vary as the radiative capture cross section, i.e. an inverse velocity dependence with few if any resonances. Thus for characterizing a thermal neutron reactor for materials testing, a low energy resolution is acceptable in the thermal portion of the spectrum. At epithermal and greater neutron energies a high degree of resolution is desirable but the small contribution to the total neutron flux at those energies is small enough that the gains from higher resolution are small.

A widely used method for determining neutron flux spectra for thermal reactors is by NAA of activation foils and wires. Two common techniques are explored in this paper. The first uses a pair of dilute Au foils and a Cd cover to separate thermal and epithermal flux contributions. The second technique uses activation of multiple foils without Cd shielding. The 2nd technique has the advantage that spectral resolution may be increased by increasing the number of foil/wire monitors. A novel spectral unfolding technique will be presented as well.

Theory

The principle behind the Au foil activation technique is based on spectral filtering. An ideal flux spectrum for a thermal reactor has a Maxwellian distribution at energies less than a thermal cutoff, $E_T \approx 0.2$ eV, and a slowing down behavior above E_{TC} which goes as $1/E$ [1]. Natural Cd has a high absorption cross section at thermal energies but quickly drops off around 0.4 eV. This energy is referred to as the *cadmium cutoff energy,* E_{cc}. The proximity of E_{cc} and E_{TC} allows one to construct a Cd shield as a high pass energy filter which absorbs nearly all thermal neutrons and passes through most epithermal neutrons. Armed with analytical expressions for the thermal and epithermal parts of the flux, it is possible to reconstruct the magnitudes of both distributions by comparing the activation rates of a Au foil irradiated with a Cd cover to a Au foil irradiated without one.

Assuming a flux spectrum of the form

$$\phi(E) = \begin{cases} \phi_t \dfrac{E}{(kT)^2} e^{-E/kT} & E < E_{TC} \\ \phi_0 \dfrac{1}{E} & E > E_{TC} \end{cases} \quad (1)$$

it is possible to determine the parameters from activation rates measured for the bare Au foil, R_b, and for the Cd covered foil, R_{Cd}. They are given by

$$\phi_t = \frac{1}{NG_{th}g\overline{\sigma_c}}\left[R_b - R_{Cd}\left(1+\frac{g\overline{\sigma_c}}{G_{res}I_0}f_1+\frac{\overline{\sigma_c}w}{G_{res}I_0}\right)\right] \approx \frac{R_b - R_{Cd}}{N\overline{\sigma_c}} \tag{2}$$

$$\phi_0 = \frac{R_{Cd}}{NI_0} \tag{3}$$

where N is the number of Au atoms in the foils, $\overline{\sigma_c}$ is the Maxwellian averaged radiative capture cross section and I_0 is the resonance integral of the capture cross section. The terms G_{th}, G_{res}, g, w and f_1 are correction factors which account for the effects of foil self-shielding and deviations from ideal *1/v* absorption at thermal energies [2].

The Au foil activation method may be described as a spectral filtering technique. Another foil activation technique, which utilizes multiple foils, attempts to determine the flux that solves the Fredholm equation

$$R_i = \int_0^\infty \sigma_i(E)\phi(E)dE \tag{4}$$

where R_i is a reaction rate for a given reaction for a given nuclide, i. The flux may be expanded as a linear combination of orthogonal functions, ψ_j, and, we assume, well approximated by a truncation of N terms of the linear combination. Then the Fredholm equation can be expressed as

$$R_i = \sum_{j=1}^{N} a_j \Psi_{ij} \; ; \; \Psi_{ij} = \int_0^\infty \sigma_i(E)\psi_j(E)dE \tag{5}$$

If one obtains reaction rates for $M > N$ foils and knows the cross sections for the measured interaction, it is possible to use the method of least squares to determine the coefficients a_j and hence the approximate flux

$$a_j = (\Psi_{jk}^T \Psi_{kl})^{-1} \Psi_{lm}^T R_m \tag{6}$$

$$\phi(E) \approx \sum_{i=1}^{N} a_i \psi_i(E) \tag{7}$$

Various options exist for the choice of the orthogonal functions such as Chebyshev and Laguerre polynomials [3, 4], orthogonal combinations of the monitor cross sections [5], and energy group flux.

The technique used in this characterization first uses least squares to make a two parameter fit to the ideal flux spectrum (as in equation 1) to the measured reaction rates for a set of activation foils. Then the part of the reaction

rates due to the ideal flux is removed from the measured reaction rates and a remnant flux is fit to minimize the remnant reaction rates. Using conventional matrix notation the *N* measured

$$\begin{bmatrix} R_1 \\ R_2 \\ \vdots \\ R_N \end{bmatrix} = \underbrace{\begin{bmatrix} \vec{\sigma}_1 \\ \vec{\sigma}_2 \\ \vdots \\ \vec{\sigma}_N \end{bmatrix} \begin{bmatrix} M(E,T)dE & dE/E \end{bmatrix}}_{X} \begin{bmatrix} \phi_t \\ \phi_0 \end{bmatrix} \tag{8}$$

where *M(E,T)* is the normalized Maxwellian, and the vector arrows above the cross sections indicate row vectors which get integrated upon matrix multiplication to form matrix *X*. Then using the method of least squares

$$\begin{bmatrix} \phi_t \\ \phi_0 \end{bmatrix} = \left(X^T X\right)^{-1} X^T \vec{R} \tag{9}$$

The difference between the ideal flux and the true flux can then be approximated by

$$\phi_{true} = \phi_{ideal} + \Delta\phi \tag{10}$$

Where the remnant flux, $\Delta\phi$, can be approximated by expressing it in *N* energy groups, $\Delta\phi_g$ and solving

$$\vec{R} - X \begin{bmatrix} \phi_t \\ \phi_0 \end{bmatrix} = \sigma_{ig} \Delta\phi_g \tag{11}$$

Experimental

10 foils were selected for measuring activation rates. Two 0.135 atomic percent AuAl foils were used in the Au foil technique. The Al acts as a diluting medium to allow for reasonably long irradiation times at moderate reactor power. One of the Au foils was encased in a set of Cd covers, the other left bare. For the multiple foil technique natural Fe, Mo, Zr, CuMn, Cu, NaCl, Sc and W foils were used.

The foils were irradiated in an in-core facility called a rotary specimen rack (RSR) at the University of Texas at Austin's TRIGA reactor. During irradiation the RSR orbits the fuel assembly insuring the monitors receive consistent and isotropic fluence. Irradiation times and powers were chosen to achieve measurable and safe activity levels. The AuAl foils were irradiated for 5 min at 10kW power, the Fe, Mo, Zr foils were irradiated for 2 hrs at 950 kW (full power) and the MnCu, Cu, NaCl, Sc and W foils were irradiated for 10 min at 100 kW. A linear relationship between power and flux was assumed for reaction rate reconstruction.

Upon irradiation the foils were allowed to decay for periods of hours to days, depending on the activity of the shortest lived nuclides. A high purity germanium detector was used to accumulate gamma spectra for the various films. The counting geometry was determined by selecting the closest sample to detector distance which registered a dead time less that 5% and a ^{152}Eu source was used to calibrate the efficiency at each counting geometry.

The gamma spectra were analyzed using Maestro MCA application. Radiative capture cross sections were obtained from ENDF/B-VI neutron libraries and prepared using NJOY99 data processing system. 9 group cross sections were constructed at 293 °K using a thermal + 1/E + fission weight function. The energy groups spanned 9 energy decades from 1×10^{-5} to 1×10^{4} eV.

Results

To calculate the activation rates of the two AuAl foils, the 411.8 keV gamma peak was accumulated for a total of 3 hrs for each foil. Values for the Maxwellian and *1/E* fit parameters were determined at 950 kW power to be $\phi_t = (3.5\pm0.18)\times10^{12}$ cm^{-2} s^{-1} and $\phi_0 = (1.80\pm0.05)\times10^{11}$ cm^{-2} s^{-1}.

Figure 1 shows the ideal flux with the fit parameters given above. As it is a two parameter fit based on two reaction rates, no degrees of freedom remain to estimate the systematic uncertainty in the model. The only uncertainty in the flux is due to the propagated uncertainty from the reaction rates, which are too small to display on the log-scale.

Figure 1

Table 1 displays the activation rates for the different foils and the gamma peaks used to calculate the rates. Only the rates from ^{58}Fe, ^{98}Mo, ^{94}Zr and ^{96}Zr were used to perform the fit as it was found that the residual errors in the remaining reactions were large compared to the rates themselves. Also, including the other foils resulted in an unphysically low estimate of the epithermal parameter.

Table 1

The values for the ideal flux parameters determined by the multiple foil technique are $\phi_t = (3.23 \pm 0.17) \times 10^{12}$ cm^{-2} s^{-1} and $\phi_0 = (1.22 \pm 0.02) \times 10^{11}$ cm^{-2} s^{-1}. The corresponding ideal group flux is shown figure 2.

Figure 2

Conclusions

This experiment demonstrates the ability of the multiple foil activation technique to determine the approximate neutron flux spectrum in a thermal reactor. Fit parameters for the Maxwellian and *1/E* fit agree reasonably well between the multi-foil method and the Au foil activation method. Furthermore the multi-foil method is equipped with the ability to readjust energy group fluxes to better approximate the true flux. In theory, spectral resolution is proportional to the number of flux monitors used. The multi-foil technique can easily resolve greater spectral detail with the addition of more flux monitors.

References

1. Stoughton RW, Halperin J (1959) Nucl Sci Eng 6:100
2. Standard E262-03 (2003) ASTM Intl. Doi:10.1520/E0262-03
3. Gold R (1964) Nucl Sci Eng 20:493
4. Di Cola G, Rota A (1965) Nucl Sci Eng 23:344
5. Ryufuku H (1966) J J Appl Phys 5:903

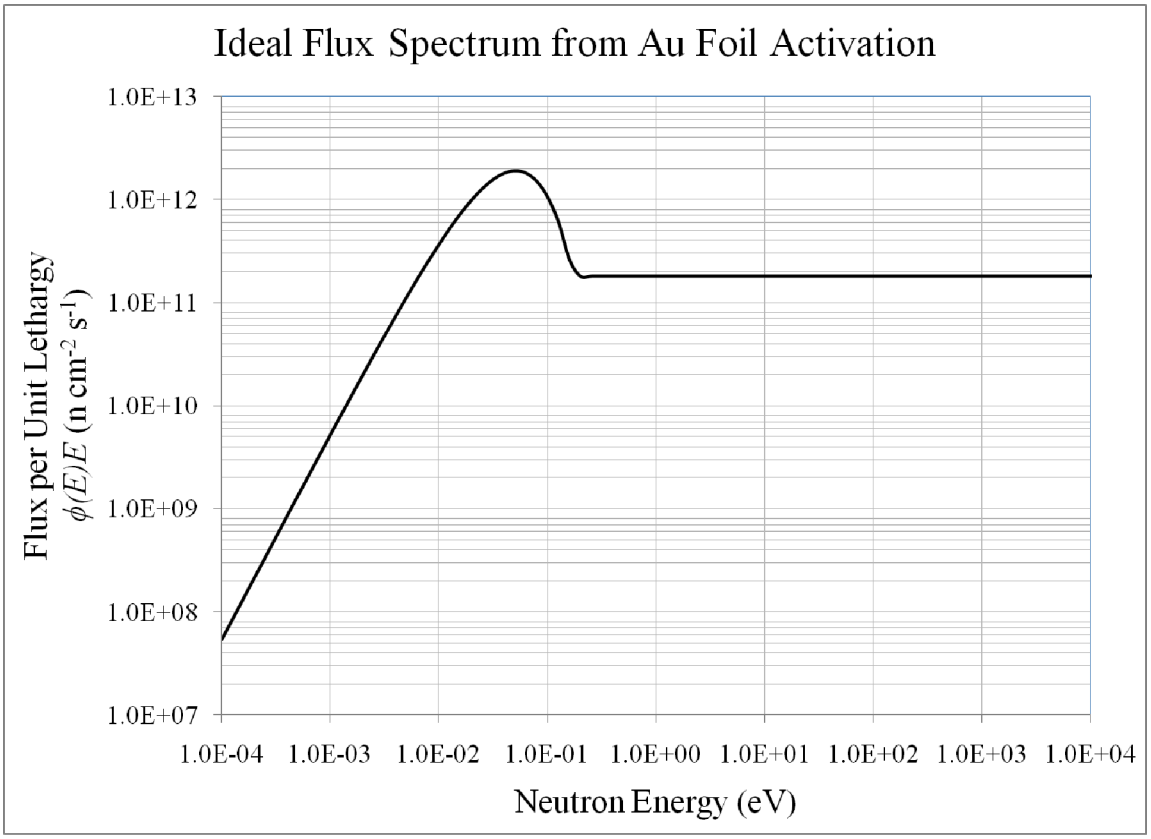

Figure 1. Ideal neutron flux spectrum for RSR facility at 950 kW power determined by activation of Au foils.

Table 1. Reaction rates of 8 activation foils at 950 kW power

Foil	Reaction	Gamma Energy *keV*	Reaction Rate s^{-1}
Fe	$^{58}Fe(n,\gamma)^{59}Fe$	192.3, 1099.2, 1291.6	$(3.5\pm0.17)\times10^{-12}$
Mo	$^{98}Mo(n,\gamma)^{99}Mo$	140.5, 181.1, 366.4, 739.5, 777.9	$(1.18\pm0.4)\times10^{-12}$
Zr	$^{94}Zr(n,\gamma)^{95}Zr$	724.2, 756.7	$(1.68\pm0.3)\times10^{-13}$
	$^{96}Zr(n,\gamma)^{97}Zr$	743.4	$(7.35\pm0.7)\times10^{-13}$
Cu-Mn	$^{55}Mn(n,\gamma)^{56}Mn$	846.8, 1810.7	$(2.9\pm0.10)\times10^{-11}$
Cu	$^{63}Cu(n,\gamma)^{64}Cu$	1345.8	$(6.1\pm0.5)\times10^{-12}$
NaCl	$^{23}Na(n,\gamma)^{24}Na$	1368.6	$(1.302\pm0.5)\times10^{-12}$
Sc	$^{45}Sc(n,\gamma)^{46}Sc$	889.3, 1120.5	$(6.98\pm0.3)\times10^{-11}$
W	$^{186}W(n,\gamma)^{187}W$	478.6, 551.5, 618.3, 625.5, 685.7, 772.9	$(1.203\pm0.8)\times10^{-10}$

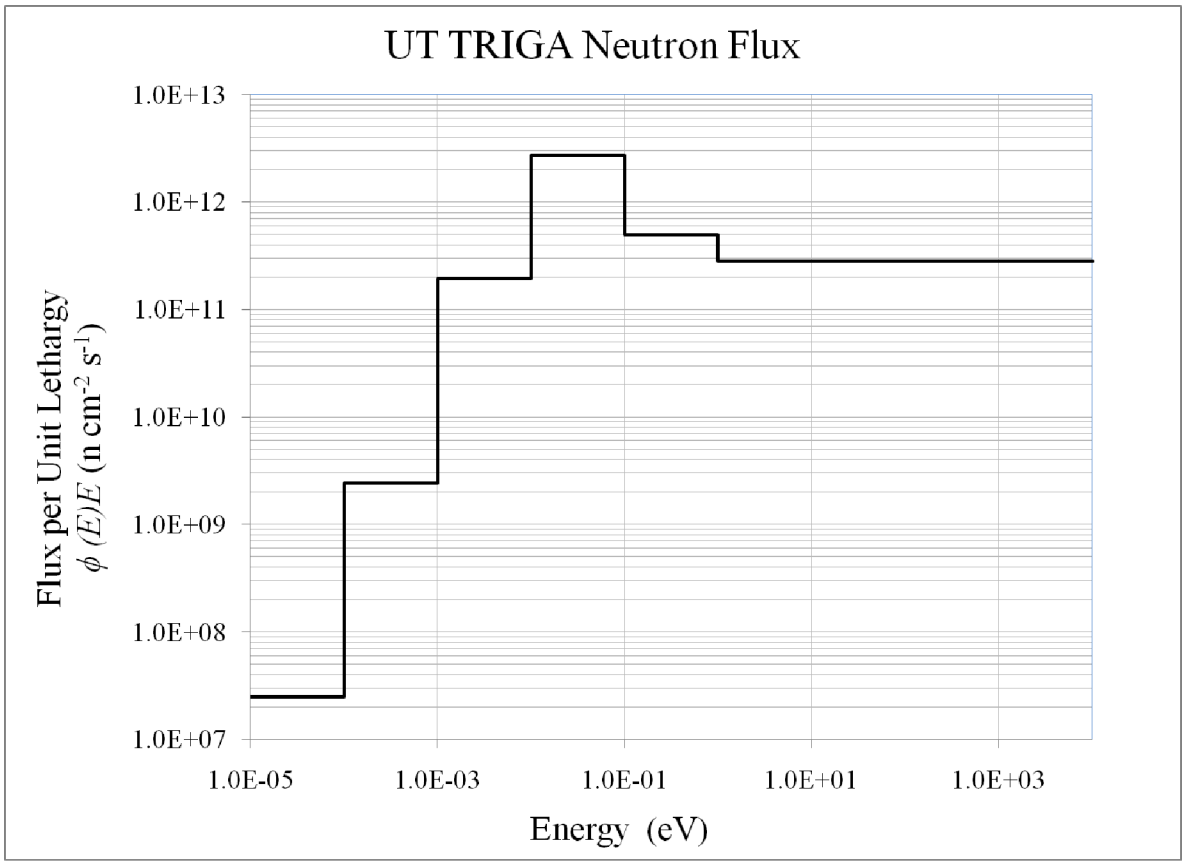

Figure 2. 9 group flux spectrum for RSR facility at 950 kW power determined by method of least squares using multiple activation foils.