ENERGY DEPENDENCE OF THE $^{238}\text{U}$ THERMAL CAPTURE CROSS SECTION

By

N. P. Baumann and M. Owais

Savannah River Laboratory
E. I. du Pont de Nemours & Co.
Aiken, South Carolina 29808

This paper is proposed for presentation at the
American Nuclear Society Annual Meeting
Las Vegas, Nevada
June 8-13, 1980
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.
DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.
INTRODUCTION

The dependence of the $^{238}\text{U}$ cross section on neutron energy in the thermal range significantly affects measurements and computations of reaction rates in nuclear reactors. The effects are particularly important for lattices of massive assemblies of natural or slightly enriched uranium in D$_2$O or graphite. For these lattices, the uncertainty enters, not in the Maxwellian energy region, but rather in the near-thermal $1/E$ energy region. Accurate measurement of these cross sections by conventional differential cross section techniques is difficult because of the dominance of neutron scattering at these energies. Assignment of this energy dependence has generally fallen into one of three categories:

- An energy dependence determined by the Breit-Wigner tail of the near-lying positive resonances with the assumption that

The information contained in this article was developed during the course of work under Contract No. DE-AC09-76SR00501 with the U.S. Department of Energy.
the residual cross section is $1/v$. The $g$ factors of Westcott\textsuperscript{1} and the ENDF/B evaluations of Leonard\textsuperscript{2} fall into this category.

- A $1/v$ energy dependence. This assumption is tacitly made by experimentalists in most measurements of reaction rates by the so-called "$1/v$ Subtraction Method." For many lattices, possible errors by this assumption are acceptable.

- An unknown energy dependence, hence one to be somewhat arbitrarily modified in order to make reactor computations agree with experiments. Such modifications have been normally made by introduction of near-lying negative-energy resonances.

The present tests were undertaken to provide integral data for making a choice of these alternatives, examples of which are shown in Figure 1.

SUMMARY

Integral activation measurements supported the thermal neutron energy dependence of $^{238}$U assumed in the ENDF/B-IV evaluation.\textsuperscript{2} The activation measurements were conducted in a thermally insulated graphite block at the side of the SP Reactor. The block was thermally heated to temperatures up to 450°C. In addition to heating, gadolinium filters were used to tailor the neutron spectra incident on the foils. The metallic foils consisted of copper and depleted uranium. Copper served as the $1/v$ reference. Activation ratios of $^{238}$U to $^{63}$Cu in the tailored
spectrum were compared to corresponding ratios in a well thermalized flux at room temperature. The difference in this ratio is strongly dependent on the energy dependence of the $^{238}\text{U}$ cross section.

DISCUSSION

Description of Method

The method involved the following steps:

- Selection of a standard foil containing an activating isotope with a known energy dependence,
- Preparation of sets of foils, each containing a number of $^{238}\text{U}$ foils and a standard foil
- Irradiation of one set of foils in a well-thermalized neutron flux.
- Simultaneous irradiation of a duplicate set of foils in a tailored neutron flux. This flux is selected to emphasize the energy region where the cross sections of $^{238}\text{U}$ and the standard are expected to differ in shape.
- Comparison of reaction rates as determined by nuclear counters.
- Comparison of measured reaction rates to computations using different assumed $^{238}\text{U}$ energy dependences.

The standard isotope chosen was $^{63}\text{Cu}$ (contained in natural copper metal foils). The advantages of copper include:
- Convenient physical form.
- Reasonable activation cross section (4.4 barns)
- Convenient half life of the activation product (12.8 hours).
- Known 1/v energy dependence with widely spread resonances at high energies.

The well-thermalized flux selected was that in the thermal column of the Standard Pile (SP) at the Savannah River Laboratory. The 1/v cadmium ratios were greater than 1000 in the regions in which the irradiations were made. This effectively eliminated errors for the small epicadmium correction in the thermal reference foils.

The tailored flux was obtained by a combination of a small (51 x 51 x 45 cm) heated graphite block and thin gadolinium filters. Neutrons were provided by the same SP reactor that fed the thermal column. Neutron temperatures incident on the foil packets were determined by the conventional paired Cu/Lu foil technique. The lutetium and all copper foils were counted on a NaI scintillation counter. The $^{238}$U foils were counted on a GeLi spectrometer.

The analysis of the experiment involved a comparison of the measured-to-calculated ratio, $R$, where

$$ R = \frac{[(^{238}\text{U} \text{ activity})/(^{63}\text{Cu} \text{ activity})]_{\text{Hot, Filtered}}}{[(^{238}\text{U} \text{ activity})/(^{63}\text{Cu} \text{ activity})]_{\text{Thermal}}} $$

(1)
On the experimental side, this ratio has the advantage that many possible sources of error cancel out. Such cancellation occurs because $R$ can be recast as a ratio of $^{238}\text{U}$ activations and a ratio of $^{63}\text{Cu}$ activations. This means that if all foils of a given type are counted under identical conditions, no efficiencies, absolute or relative, need to be determined. The simultaneous irradiation of the different foil sets also greatly reduces possible errors in time corrections. For the experiment, equation 1 assumes that the usual counting corrections have been made, e.g. epicaadmium neutron activations and differences in foil weight.

The calculation of the ratio $R$ was performed for the actual experimental conditions. The computation assumes infinite slab geometry with an isotropic neutron flux incident. The numerical integration over energy utilized the 30-energy group structure and the cross section library of GLASS. A special code was written to obtain the energy integral of the reaction rate.

$$R = \int_0^{0.625 \text{ eV}} \left[ (E_3(X+Y) - E_3(X+Y+Z)) \phi(E) dE \right]$$

(2)

where $E_3$ is the standard exponential integral and $\phi(E)$ is the temperature-dependent neutron flux spectrum given by Westcott using his $\Delta_\alpha$ function for joining the Maxwellian and $1/E$ spectra. The detector thickness $Z$, the intermediate absorber thickness $Y$, and the gadolinium filter thickness $X$, are all energy dependent and are given in units of absorption mean free paths. Application
of Equation 2 to each individual foil with the neutron flux incident on both faces of the packet and summing over all foils of the same type gives the relative total reaction rate for each foil type. An edit of the reaction rate as a function of energy was also obtained; typical reaction rate profiles are shown in Figure 2. Neutron temperatures for the paired Lu/Cu foils were obtained using the same code to give a calibration curve of activation ratios as a function of neutron temperature.

Survey computations of R were made in order to obtain near optimum conditions for the experiment. These were done in the thin foil approximation, but do not differ significantly from foil thicknesses actually used. The results are shown in Figure 3 where R is shown as a function of neutron temperature with the gadolinium filter thickness as a parameter. The results show that the expected value of R increases significantly with temperature and filter thickness. This suggests that maximum sensitivity could be obtained by maximizing both. Unfortunately, practical considerations preclude this. The difficulties of performing the experiments increase with temperature, and thick gadolinium filters reduce the activation intensity of the foils. This latter effect may reduce the count rate below that required to obtain the desired accuracy, but, more important, it may reduce the count to where the epicadmium component dominates and cannot be subtracted with sufficient accuracy. The relative count rate as a function of temperature and filter thickness is shown in Figure 4. In this figure, the $l/v$ activation rate is shown,
relative to that for an identical bare 1/ν foil in a completely thermalized spectrum at 25°C. Figures 3 and 4 served as guides at each temperature for selection of the optimum filter thickness. This selection was designed to minimize the percent error in the quantity (R-1).

**Experimental**

A schematic of the heated block assembly is shown in Figure 5. Heater plates with a maximum capacity of ≈10 kW were placed on the bottom surface. A thermocouple for controlling the temperature was imbedded near the center of the assembly. Additional thermocouples to monitor the uniformity of the temperature were scattered throughout the assembly. The insulation consisted of 2 inch thick sheets of calcium silicate. A water-cooled foil holder was positioned along the axis and close to the center of the heated block. Calcium silicate also served to thermally insulate the aluminum foil holder from the heated block. A schematic of the foil holder is shown in Figure 6. The volume of water inside the block was held to a minimum and kept distant from the foils so as not to disturb the neutron spectrum at the foils. Foil packets, shown schematically in Figure 7, were assembled in closely matched pairs. The only difference was in the filter material; 0.081 cm thick cadmium for one and thinner gadolinium metal for the other. Aluminum filler discs were added to the gadolinium to maintain the same overall dimensions. Thin aluminum catcher foils separated all copper foils from the
uranium foils. Additional filters of natural uranium were added outside the cadmium or gadolinium to suppress the resonance activation within the packet.

A schematic of the thermal column is shown in Figure 8. The configuration shown is that for the room temperature (25°C) runs which were irradiated totally within the thermal column and did not use the heated block. For the high temperature measurements, the gadolinium-covered foils were eliminated from the thermal column.

Following irradiation, the foil packets were disassembled and each foil type was counted separately. Counting of the copper and lutetium foils was straightforward. Counts were sufficiently low that dead time corrections were negligible. Verification of the half life for both foil types assured that impurity activations and fission product contamination were negligible. The $^{238}\text{U}$ was counted on a GeLi spectrometer in order to discriminate against activity from spontaneous decay and from fission products in the depleted uranium foils ($^{238}\text{U}$ foils containing 0.019 wt % $^{235}\text{U}$). Count rates were sufficiently low that pulse pileup losses were negligible. This fact was verified for the most active foils by tests with a calibrated pulser. The verification of the 2.35 day half life of the $^{239}\text{Np}$ decay served as a final test of the validity of the method. The results were consistent for both the 228 KeV gamma rays and were nearly independent of different methods of base-line subtraction.
Results

A comparison of the measured and calculated ratios (using ENDF/B-IV cross sections) is given in Table I. Indicated uncertainties are one sigma values and include counting statistics and estimates of all other uncertainties. This table includes repeat runs of all but the 300°C case. The comparison shows that the $^{238}\text{U}$ capture cross section is much closer to that of ENDF/B-IV than to $1/v$ law ($R=1.000$), but does not preclude the possibility of a small contribution from a close-lying negative energy resonance.

REFERENCES


<table>
<thead>
<tr>
<th>Graphite Temp., °C</th>
<th>Neutron Temp., °C [176\text{Lu}/63\text{Cu data}]</th>
<th>Gd Thickness ((\text{Atom/barn}) \times 10^4)</th>
<th>(R-1) Meas.</th>
<th>(R-1) Calc. (ENDFB-IV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>25</td>
<td>25</td>
<td>1.5</td>
<td>0.012 ± 0.004</td>
<td>0.015</td>
</tr>
<tr>
<td>25</td>
<td>25</td>
<td>1.9</td>
<td>0.012 ± 0.004</td>
<td>0.017</td>
</tr>
<tr>
<td>25</td>
<td>25</td>
<td>1.9</td>
<td>0.010 ± 0.006</td>
<td>0.017</td>
</tr>
<tr>
<td>25</td>
<td>25</td>
<td>1.9</td>
<td>0.020 ± 0.006</td>
<td>0.017</td>
</tr>
<tr>
<td>150</td>
<td>126</td>
<td>1.9</td>
<td>0.033 ± 0.008</td>
<td>0.022</td>
</tr>
<tr>
<td>150</td>
<td>126</td>
<td>1.9</td>
<td>0.015 ± 0.008</td>
<td>0.022</td>
</tr>
<tr>
<td>300</td>
<td>229</td>
<td>3.8</td>
<td>0.023 ± 0.008</td>
<td>0.029</td>
</tr>
<tr>
<td>450</td>
<td>335</td>
<td>3.8</td>
<td>0.027 ± 0.008</td>
<td>0.040</td>
</tr>
<tr>
<td>450</td>
<td>308</td>
<td>3.8</td>
<td>0.037 ± 0.008</td>
<td>0.039</td>
</tr>
</tbody>
</table>
FIGURE 1. Examples of Different Assumed $^{238}$U Capture Cross Sections in the Thermal Energy Region
FIGURE 2. Relative Reaction Rates for 1/v Foils Inside Gadolinium Filters (Gd Thickness = 0.00021 A/B)
FIGURE 4. Computed Activation Intensities for Different Temperatures and Filter Thicknesses
FIGURE 5. Schematic of Heated Graphite Side Block
FIGURE 6. Schematic of Water-Cooled Foil Holder
FIGURE 7. Schematic of Foil Packets
FIGURE 8. Schematic of SP Thermal Column Loading