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THE EFFECT OF CROSS SECTION VARIATION ON INTEGRALS MEASURED IN THE CFRMF[†]

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ABSTRACT

Relative capture cross section integrals for a variety of materials have been measured in the Coupled Fast Reactivity Measurement Facility at Idaho. A knowledge of the flux spectrum (peaking at about 350 keV) allows the calculation of these relative integrals from differential data where it is available. The present work describes activation integral measurements of Au, $^{238}\text{U}(n,\gamma)$, and $^{235}\text{U}(n,f)$. The experimental results are compared to calculations employing ENDF/B-II data and reveal inconsistencies beyond that expected from experimental error. These inconsistencies cannot be resolved by arbitrary adjustment of the cross section of any one of the materials. Various schemes of cross section variations and their effect on the integrals will be presented. Energy dependent reaction rates have been plotted for each of the three materials. These will be discussed in terms of specific energy regions and their importance to the total integral. This work, although not defining a unique set of cross section data, does shed important light on the questions of data testing by integral methods, and the sensitivity of the integrals to arbitrary changes in differential data over specific energy regions.

INTRODUCTION

The Coupled Fast Reactivity Measurement Facility, CFRMF, was designed and built for the purpose of measuring integral cross sections in a spectrum pertinent to fast reactor design studies. Spectrum measurements and calculations as well as relative integral cross sections have been reported elsewhere^{1,2,3} and will only be mentioned briefly in this paper. Of prime consideration will be the sensitivity of these integral measurements to differential cross section data in particular energy regions, and therefore, their utility in cross section evaluation. Integral cross section measurements of $^{238}\text{U}(n,\gamma)$, $^{238}\text{U}(n,f)$, $^{235}\text{U}(n,f)$, and $^{197}\text{Au}(n,\gamma)$ were made in an effort to find a reaction that could be used as a suitable standard for other measurements. Ratios of these integrals have been compared to calculated ratios employing ENDF/B-II data, and their sensitivity to differential cross section data is discussed.

[†]Work supported by the U. S. Atomic Energy Commission.

MEASUREMENTS

Samples of highly depleted $^{238}\text{UO}_2$ (17ppm ^{235}U), enriched $^{235}\text{UO}_2$, and ^{197}Au , were irradiated together in the measurement position of the CFRMF. Samples were of small enough size that neutron self-shielding effects were negligible. The ^{238}U capture cross section integral was determined by counting the gamma rays from the ^{239}Np , 2.35-day decay and relating this to the same activity produced in a sample irradiated in a pure thermal flux. Thus, the integral cross section is based on a knowledge of the thermal cross section rather than the decay scheme parameters appearing in the literature. The fission reactions for both ^{238}U and ^{235}U were determined by counting gamma rays from the 40.2 hour decay of ^{140}La produced from fission. The fission integral uncertainty thus depends on a knowledge of the fission yields for the mass-140 isotopes. An attempt was made to interpolate between yield data for fast fission and thermal fission (for ^{235}U) in order to arrive at a number appropriate for the CFRMF spectrum, however, the uncertainties quoted for these two numbers overlap, hence negating this refinement. The value used for the chain yield was $6.2 \pm \%$ for both ^{235}U and ^{238}U . The gold capture integral was determined by the usual ^{198}Au gamma ray counting techniques. Ratios of measured reaction integrals are given in the first column of Table 1. The quoted numbers include small corrections for sample gamma ray self-absorption, and counter efficiency variation with energy. The quoted uncertainties have been obtained by a standard propagation of uncertainties on decay scheme parameters, half-lives, yields, sample weighing, and counting statistics. In addition, the experimentally measured numbers appearing in the table are the average of two independent set of measurements the results of which fall within the uncertainty of each other.

DISCUSSION

The second column of Table 1 lists calculated reaction integral ratios. These are obtained by a numerical integration of the ENDF/B-II differential data for the particular reactions over the assumed spectrum in the CFRMF. Before a meaningful comparison between measurement and calculation can be made some discussion of the sensitivity of these integrals to variation in either the flux or cross section in particular energy ranges must be given.

Figures 1 and 2 are plots of the energy dependent reaction rate distribution for the four reactions being considered. The ratios of the integrals under these curves are the calculated values listed in Table 1. Table 2 lists the percent of the total integral occurring in the energy region indicated. It should be noted that in terms of these distributions, gold is most sensitive to the lower energy region, then $^{238}\text{U}(n,\gamma)$, and $^{235}\text{U}(n,f)$ and finally, the fast fission material has reactions essentially only within the last energy decade.

Table 1 Integral Reaction Ratios

<u>Ratio</u>	<u>Measured</u>	<u>Calculated</u>
$\frac{^{238}\text{U}(n,\gamma)}{\text{Au}(n,\gamma)}$	0.643 ± 0.07	0.574
$\frac{^{235}\text{U}(n,f)}{\text{Au}(n,\gamma)}$	5.863	3.791
$\frac{^{238}\text{U}(n,\gamma)}{^{235}\text{U}(n,f)}$	0.110	0.152
$\frac{^{235}\text{U}(n,f)}{^{238}\text{U}(n,f)}$	25.29	27.47

Table 2 Energy Distributions of Reactions in CFRMF

<u>Energy Range</u>	<u>Au(n,γ)</u>	<u>$^{238}\text{U}(n,\gamma)$</u>	<u>$^{235}\text{U}(n,f)$</u>	<u>$^{238}\text{U}(n,f)$</u>
0-1	8.16%	3.60%	3.23%	0%
1 keV - 10 keV	31.60	21.93	11.79	0
10 keV - 100 keV	26.95	33.62	25.48	0
100 keV - 1 MeV	30.5	37.09	48.66	1.10
1 MeV - 10 MeV	2.46	3.70	11.82	98.90

Table 3 Alternate Correction Factors for Measured Integrals

<u>$^{238}\text{U}(n,\gamma)$</u>	<u>Au(n,γ)</u>	<u>$^{235}\text{U}(n,f)$</u>	<u>$^{238}\text{U}(n,f)$</u>
1	0.89	1.38	1.50
1.12	1	1.55	1.68
0.72	0.65	1	1.09
0.665	0.595	0.92	1

The apparent inconsistencies noted in Table 1, must be examined both as to possible cross section errors and spectrum uncertainties. In order to define these discrepancies more completely, Table 3 has been prepared by assuming a particular integral from Table 1 to be exact, and calculating the factor by which the other integrals need to be multiplied in order to agree exactly with the measured ratios. This procedure ignores the uncertainties on the measured values; the factor derived can be assigned either to flux or cross section or more likely, may be a combination of both. The spectrum used in the numerical integration to determine integral cross section is a composite of two experimental studies and detailed transport theory calculations[†]. One can then draw an envelope around all these predictions and thereby define a conservative uncertainty for each decade of energy. The low energy (below 10 keV) region of the spectrum is the most troublesome since this region is not predicted well by foil activation techniques, because of the absence of either threshold reactions or isolated resonances, and the proton-recoil technique experiences difficulties in discriminating against gamma reactions in this region. The average estimated uncertainty to be assigned to spectrum, by decade, is shown in Table 4.

Table 4 CFRMF Spectrum Uncertainties

<u>Energy Range</u>	<u>Spectrum Uncertainty</u>
Below 10 keV	± 60%
10 - 100 keV	± 20%
100 - 1000 keV	± 10%
1 MeV - 10 MeV	± 20%

Table 2 should be studied with regard to the uncertainties given in Table 4, because these two tables reflect the sensitivity of the integral measurement to possible errors in cross sections for specific energy ranges, and also yield insight into the effect of uncertainties in spectrum which must be taken into account. It is then possible to consider the "correction factors" given in

[†]See Figure 1, Y. D. Harker, "Fast Neutron Capture Integral Measurement of ⁹⁹Tc, ¹⁰⁹Ag, and other Fission Products", Third Conference on Neutron Cross Sections and Technology, p.

Table 3 and determine the most reasonable of these choices. (One should recognize that it is improbable that any one of the four alternatives is exactly correct since this would require that one of the cross sections be known exactly). In considering that either of the capture reactions are correct, one sees that inappropriately large corrections to the two fission reactions would be required, and since these reactions are most influenced by a region of the spectrum where relatively small uncertainties exist, the large correction would have to be applied to the differential data. Therefore, the first two alternatives listed in Table 3 are considered unreasonable. The last two alternatives are, within experimental error, the same. They have the additional characteristic that the changes required for consistency take place in capture cross sections rather than fission cross sections which are generally assumed to be better known. Also, some changes in the integrals can be accounted for by the low energy spectrum where large uncertainties exist. For example, if one assumes that the $^{238}\text{U}(n,f)$ cross section is exactly known, (the last alternative in Table 3) and that the low energy spectrum may be in error by the full 60% uncertainty below 10 keV, and re-calculates the correction factors they become, 0.85 for ^{238}U , 0.76 for Au, 0.91 for $^{235}\text{U}(n,f)$, and 1.0 for $^{238}\text{U}(n,f)$. If one attempts to adjust the flux in a similar fashion at higher energies, the $^{235}\text{U}(n,f)$ begins to be adversely effected. Though they are only an example, it must be recognized that the numbers chosen above nevertheless suggest qualitatively, that both the ^{238}U and Au capture cross sections in the ENDF/B-II library are too high.

CONCLUSIONS

The study reported in this paper set out to determine a standard for the measurement of relative integral cross sections in a spectrum of the breeder reactor type. The conclusions to be drawn are as follows:

1. The fission reactions investigated are the only reasonably consistent integral ratio.
2. If the fission cross sections are correct, then the Au and ^{238}U capture data in ENDF/B-II are too high, even considering possible spectrum uncertainties.
3. More fission and threshold reactions need to be studied to gain confidence in the knowledge of the spectrum and confidence in $^{235}\text{U}(n,f)$ fission cross section, if it is to become the standard for this work.

It should also be pointed out that integral measurements, even in the absence of a well known standard, are nevertheless important relative numbers, particularly where, as is often the case, the reaction rate distribution is similar in shape from one material to another.

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Capture Intensity

Energy, KeV

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Fission Distributions

Energy, KeV