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by

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MEASUREMENT OF THE FAST NEUTRON CAPTURE CROSS SECTION OF ^{238}U RELATIVE TO $^{235}U(n,f)$

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The capture cross section of ^{238}U was measured using the activation technique and $^{235}U(n,f)$ as a reference cross section. Capture events were measured by detection of two prominent γ-transitions in the decay of the ^{239}U daughter nuclide, ^{239}Np, employing a high resolution Ge(Li) detector. The system was calibrated with samples activated in a thermal neutron flux relative to the capture cross section of gold, and with an absolutely calibrated α-emitter, ^{243}Am, which decays to ^{239}Np. Cross section measurements were carried out in the neutron energy range from 30 keV to 3 MeV. Emphasis was on absolute values between 150 keV and 1 MeV where the $^{238}U(n,\gamma)$ cross section and its ratio to $^{235}U(n,f)$ are not very sensitive to energy scale uncertainties and the $^{238}U(n,f)$ cross section is small. Background from fission products was found to restrict the accuracy of the measured data at energies > 1.5 MeV.

Introduction

The radiative capture cross section of ^{238}U is of interest in applied areas due to its important role as a fertile material in various breeding cycles. Calculations of breeding gains and criticality are sensitive to this cross section which is, unfortunately, still rather poorly known. Calculations of capture-to-fission central core reaction ratios for benchmark test facilities using current evaluated data files result in higher values than experimentally determined. This situation has prevailed for more than 10 years and has resulted in continuous requests to lower evaluated differential data. However, most experimental differential cross section data are higher than the evaluated cross section data; thus little support can be seen for further lowering of the ^{238}U capture cross section which is presently on the ENDF/B-file. Many of the problems observed in the data base seem to be associated with unfavorable factors related to measuring ^{238}U capture events. The neutron binding energy is only 4.8 MeV, thus the efficiency for counting capture events with a large liquid scintillator is low and must be expected to be rather uncertain and sensitive to changes in details of the γ-cascades. Low thresholds for inelastic scattering, and the open fission channel at higher energies, hamper all prompt detection techniques. The natural radioactivity of ^{238}U and ^{235}U and their decay products produces additional background which further impedes prompt detection and also complicates the use of the activation technique. Activation nevertheless appears to be the most promising technique because a well-defined calibration method exists for this specific approach.

The present measurements were carried out between 30 keV and 3 MeV. This energy range contributes significantly to the total capture rate in fast reactors. Emphasis was on absolute values in the range from 150 keV to 1 MeV where the cross section varies less with energy than at lower or higher energies. The activation technique was applied and the $^{235}U(n,f)$ cross section was used as a reference.

Irradiations

The $^{7}Li(p,n)^{7}Be$ reaction was used as a neutron source and protons were accelerated by an 8 MV Tandem-Dynamitron accelerator. Proton beam currents were ~15 μ amperes. Several targets consisting of metallic lithium evaporated on a 0.025 cm thick tantalum backing were used. The target thicknesses used provided neutron energy resolutions in the range of 10 to 100 keV. Irradiations were made at zero degrees to the incident proton beam. The diameter of the metallic ^{238}U capture samples was 1.27 cm and their thickness was ~0.025 cm. The ^{238}U material was specifically selected to contain a low amount of ^{235}U (< 400 ppm). The ^{238}U capture samples were mounted on the outside of a low mass ionization fission chamber as shown in Fig. 1. The diameter of the ^{235}U sample was 2.54 cm and its thickness was 210 μg/cm^2. The isotopic composition of this sample was 98.405% ^{235}U, 1.028% ^{234}U, 0.447% ^{236}U, and 0.122% ^{238}U and its mass was recently established relative to several independent mass scales[1] with an uncertainty of ~0.6%. The spacing between the capture samples and the ^{235}U sample was 0.05 cm, half of this was due to the wall of the fission counter and the other half due to the backing of the fissile deposit.

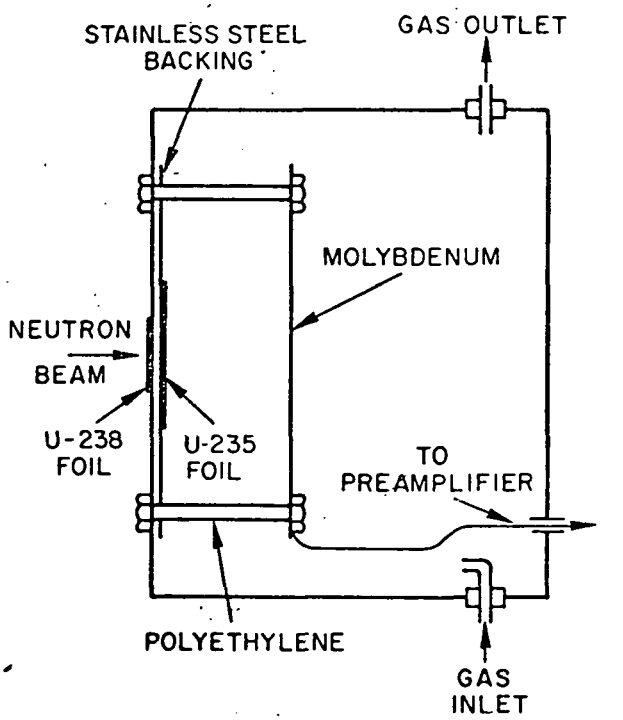

Fig. 1. Schematic of Fission Chamber and Uranium Samples

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Irradiations were carried out at 13 energies between 0.03 and 3.0 MeV with irradiation times varying from 45 minutes to 9 hours. The ^{238}U samples and the ionization chamber were usually situated ~2.54 cm from the target; however, at 0.5 MeV samples were irradiated at various distances up to 25 cm. These measurements were used to determine the background produced by neutrons which were scattered from the floor and other structures in the building. The background was found to be on the order of 1% for both the ^{238}U capture and ^{235}U fission processes at the standard position. The spatial variation of the background was known to be small over the region of the experiment from prior time-of-flight measurements with a fission counter. The absolute number of fission events was determined by extrapolating the measured fission fragment energy spectrum to zero pulse-height. A correction for the total fission fragment absorption was calculated using a fission fragment range of 4.1 mg U/cm^2. The average cross section at the average neutron energy was determined from the total number of capture and fission events. The angular distribution of the source neutrons, the path through the samples, isotopic compositions and attenuation in the various materials were taken into account.[2]

Corrections were applied for scattering of neutrons in the samples, the fission chamber and the target structural material using Monte Carlo techniques and data from current ENDF/B - files. A correction was applied to obtain the average cross section at the average energy from the measured average cross section.[3]

Determination of the Capture Events

The capture samples were irradiated in the fast neutron flux, ϕ, for a period of time, T. After the end of the irradiation, a waiting period of t > 4h permitted at least 99.9% of the ^{239}U nuclei produced with the capture events to decay to ^{239}Np. The samples were then counted with a Ge(Li) detector for a period of time, θ. The total number of counts observed in this time is given by $C_\gamma = \varepsilon_\gamma \cdot \sigma \cdot n \cdot \phi \cdot F$, where ε_γ is the effective counting efficiency, σ is the cross section to be determined, n is the number of nuclei in the sample, and ϕ is the neutron flux per cm^2 and sec. The time factor, F, is

$$F = \frac{1}{\lambda}\,(1 - \ell^{-\lambda T})\,\ell^{-\lambda t}\,(1 - \ell^{-\lambda \theta}) \quad .$$

The half-life of ^{239}Np was taken to be 2.355 ± 0.004d.[4] A correction was required for the variation of the flux, θ, during the irradiation. The correction was determined by monitoring the neutron flux with a Long Counter for a series (n) of time intervals (τ). τ was set at 15 minutes. The correction is given by

$$k = n \cdot \frac{1 - \ell^{-\lambda\tau}}{1 - \ell^{-n\lambda\tau}} \cdot \frac{\sum a_i\, \ell^{-\lambda(n-i)\tau}}{\sum a_i} \quad ,$$

with a_i being the relative monitor counts. This correction was found not to exceed 0.2%.

Major transitions in the decay of ^{239}Np result in γ-rays of 106 keV, 228 keV, and 278 keV which occur with 27.2%, 11.5% and 14.5%[5] frequency per decay, respectively. The 106-keV γ-transition cannot be distinguished from several x-rays of ^{239}Pu, therefore, the 228 keV and the 278 keV γ-rays were used in the present measurements. The stability of the counting system was checked for the period of the present experiment and found to be better than 0.4%. This represents a reproducibility limit for the present measurements and is probably caused by the limitations of the reproducibility of mounting the uranium samples in proximity to the Ge(Li) detector. The resolution of the counting system was determined using the 1.33 MeV γ-transition in the decay of ^{60}Co and was found to be 2.6 keV FWHM at 1.33 MeV.

The uranium samples were counted at a distance of ~1.4 cm from the Ge diode. At this close distance the γ-ray absorption in the sample must be determined by integrating over the space angles involved, while considering the effect of the Ge(Li) detector efficiency which is itself dependent on the direction and the position of the entering γ-ray. The 228 keV and the 278 keV γ-rays are frequently in coincidence with the 106 keV transition as well as with some other γ-rays and x-rays.[6] Thus, sum-coincidences must be expected for close-geometry counting. The sum-coincidence probabilities are expected to depend on the sample thickness because the absorption is different for the observed γ-rays and those with which they are in coincidence. Due to these complications the effective sample absorption was not calculated but experimentally determined and made part of the calibration procedure.

Calibration with ^{243}Am-Samples

The ^{239}Np - decay observed in the present experiment also occurs in the decay chain of the α-emitter ^{243}Am. Therefore, very thin ^{243}Am samples (calibrated absolutely by low-geometry α-counting) which are in equilibrium with ^{239}Np can be used for the calibration of the γ-detection efficiency.[7-9]. Two ^{243}Am samples were used in the present experiment to determine the absorption-free effective efficiency for counting ^{239}Np decays by detecting the 228 keV and the 278 keV γ-rays. These samples contained some ^{241}Am, ^{242}Cm and ^{244}Cm which could be separated in an alpha-energy spectrum analysis. The ^{243}Am-decay fraction was 90.05%. The total α-decay rates of the samples were determined by low-geometry α-counting. The values obtained for the number of gammas detected with the Ge(Li) detector to the number of α-decays for the two samples agreed within 0.4% for the 228.keV transition and within 0.3% for the 278 keV transition. The two samples differed in intensity by about a factor of three. Therefore, the result indicated that count rate effects were negligible. The diameter of the ^{243}Am samples was somewhat smaller than the capture samples. The radial dependence of the efficiency was measured with a "point" source and a correction of 0.5% was calculated by fitting the measured values with a 2nd-order polynomial and subsequently integrating over the samples of different size.

The effects of the γ-absorption in the sample and the effective detection efficiency were analyzed by a combination of measurements and Monte Carlo simulation calculations. First, the transmission, T, through the sample was measured in the same geometry as used in the actual sample counts. The measurements were performed as shown in B and C of Fig. 2 using a virgin uranium sample and an ^{243}Am sample. From this measurement, an "effective" absorption coefficient, μ, was deduced, in the parallel-beam approximation, from the formula $T = \exp(-\mu\sigma)$. This value of μ was used to calculate the effective absorption, n, for a uniformly activated uranium sample using the formula

$$n = \frac{1}{\mu\delta} \cdot (1 - e^{-\mu\delta}) \quad .$$

The validity of this experimental method was tested by means of the Monte Carlo "experiments". Calculations for geometries B and C of Fig. 2 yielded an effective calculated T and hence a μ value. Simulation of configuration A yielded the absorption, n. Substitution of the calculated μ into the formula calculating n

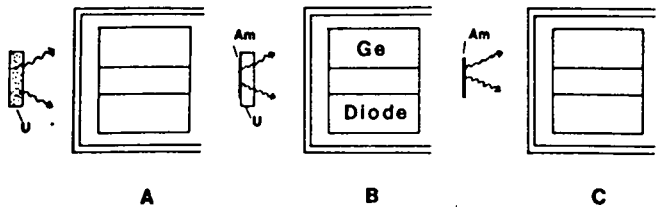

Fig. 2. Schematic Representation of the Three Cases Used in the Present Experiment and Monte Carlo Calculations

yielded a value which agreed with the Monte Carlo result to within 0.05%. This good agreement justifies the use of an experimentally-determined effective absorption coefficient in a parallel-beam approximation to calculate the absorption and efficiency correction for the geometry of the present experiment.

The above considerations do not yet include the effect of the sum-coincidences with all details. Sum-coincidences between the measured 228 keV and 278 keV γ-rays and other transitions reduce the effective absorption of the samples and are thus included in the effective thickness obtained from the transmission experiment. However, the difference between the sum-coincidences obtained for the transmission experiment and for the homogeneously-activated sample must be taken into account. A lower effective absorption is measured in the transmission experiment because the transitions which are in coincidence with the 228 keV and the 278 keV lines have lower energies and are more strongly absorbed in the transmission experiment than in the homogeneously activated sample. The effect was calculated with a simplified decay scheme and the total detection efficiencies for the emitted γ-rays which were calculated by Monte Carlo techniques. It was found that the effect due to coincidences slightly over compensates the increase in absorption caused by the effective opening angle for the detection of γ-rays.

Calibration with Samples Activated in a Thermal Neutron Flux

The thermal neutron capture cross section of ^{238}U is well known. This suggests an alternate method to calibrate the detection efficiency of the Ge(Li) detector and to obtain the effective absorption of the sample. Four capsules were placed on a wheel and rotated in the thermal column of a 10 KW reactor in order to assure equal exposure for each of the capsules. Three of the capsules were made from aluminum and one was made from cadmium. The cadmium capsule and one of the aluminum capusles contained a uranium sample sandwiched by two 0.0025 cm thick gold foils. One of the remaining aluminum capsules contained a single uranium sample and the other a single gold foil. The absolute activation of the gold samples was determined with the $4\pi\beta - \gamma$ - coincidence method employing corrections which have been discussed in detail previously.[10] The uranium samples were counted with the Ge(Li) detector using the procedures described above. The efficiency follows from

$$\epsilon = \frac{R_{U8}}{R_{Au}} \cdot \frac{\sigma_{Au}}{\sigma_{U8}} \cdot \frac{S_{Au}}{S_{U8}} \quad , R = \frac{C \cdot A}{F \cdot G}$$

where C is the number of counts, A is the atomic weight, F is the time factor defined above, and G is the mass of the sample in gms. The factor S accounts for self shielding of the samples and the activation due to neutrons scattered in the sample.[10,11] S is an average over a Maxwellian spectrum and becomes identical to the Westcott g-factor[12] for very thin samples. The neutron field was well moderated and a minor correction (0.5%) was calculated for the activation by epithermal neutrons using the Westcott formalism and the measured Cd-ratio. A value of 98.65 ± 0.09b was used for the thermal capture cross section of gold.[13] A weighted average of 2.712 ± .006b was obtained from an updated listing of all experimental values[14] of the thermal capture cross section of ^{238}U.

One of the fission products, ^{132}Te, has a half-life of 78.2h and decays with a high frequency (96.9%) by emitting a 228 keV γ-ray.[15] This gamma cannot be separated from the 228 keV γ-ray in the decay of ^{239}Np which is used in the present measurements. Fission events of ^{235}U occur in the thermal neutron activation of the ^{238}U samples and are not negligible. Corrections were calculated and found to be in the range 3.2 - 4.5% for the 228 keV photopeak. The 228 keV γ-ray of ^{132}Te is in coincidence with a 49.7 keV transition (100%),[15] thus sumcoincidences are possible which fall into the 278 keV photopeak of the ^{239}Np - decay. A correction was calculated and found to be 0.3%.

Results

The agreement between the values obtained with the two different techniques is within their respective uncertainties. The efficiency appears to be a linear function of the sample weight and a fit was used to determine the efficiency in the range of the weights of the samples used in the cross section experiment.

Table I contains the different contributions to the uncertainties of the present values. With the exception of the values above 1 MeV, the limiting factor appears to be the uncertainty of the reference cross section which was assumed to be 2%.[16]

Table I. Uncertainties of the Present Cross Sections.

Source	Range, %	Comment
Gamma Detection Efficiency	1.0	Common to all values
^{235}U Sample Mass	0.6	
Total Fission Fragment Absorption	0.5	
Half-life of ^{239}Np	0.2	
^{238}U Sample Mass	0.2	
$^{235}U(n,f)$ Cross Section	2.0	
Statistics and Reproducibility	0.9-23.1	Values are 0.9-3.0% below 1 MeV
Neutron Background	0.3-4.0	0.3% for most values except at higher energies and larger distances from the target
Second Neutron Group	0.1-1.2	Does not apply below 800 keV
Scattering of Neutrons in the Counter, Samples and Neutron Source Structural Material	0.8-1.7	
Fission Product Decay	0.2-4.0	Does not apply below 1 MeV

The cross sections obtained in the present experiment are compared in Fig. 3 with an evaluated cross section.[17] The new experimental data do not support the recent trend to lower values obtained in other measurements[19,20] which influenced the evaluated cross section.

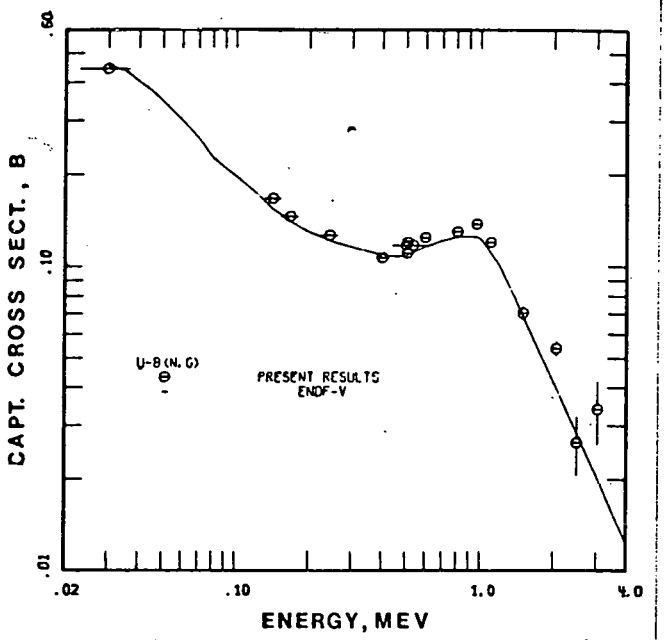

Fig. 3. Comparison of the Present Results with ENDF/B-V

The present data are compared in Figs. 4, 5, and 6 with other experimental data. Only the more recent data were selected for comparison. A detailed discussion of all available data through 1977 is given elsewhere.[17] The present result is in good agreement with that obtained with the prompt detection technique by Poenitz[21] relative to the gold standard capture cross section. Agreement with data by Lindner et al.[18] is reasonable, though the present data are somewhat higher between

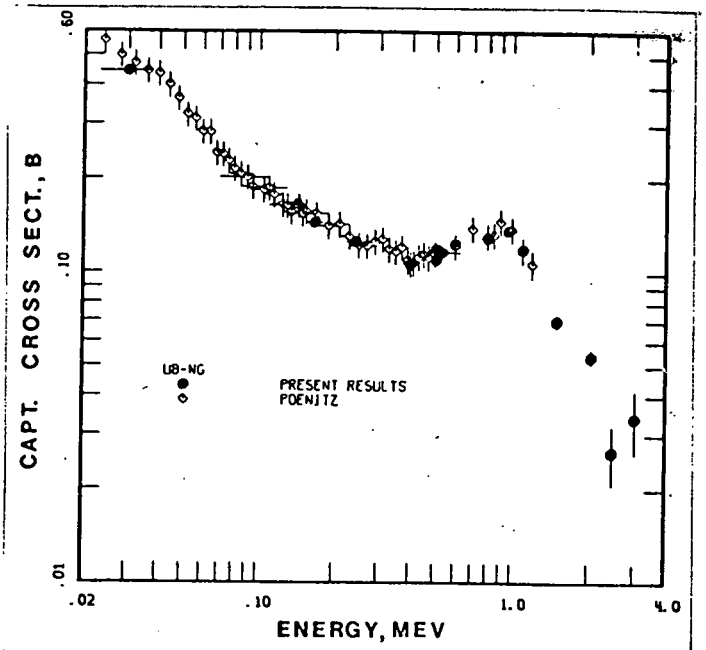

Fig. 5. Comparison of the Present Results with Other Recent Experimental Data

100 and 250 keV and around 1 MeV. The present data agree well with those by Ryves et al.[22] in the former and with the data by Davletshin et al.[23] in the latter energy region. The data by Quan et al.[20] and LeRigoleur et al.[19] agree better with those by Lindner et al. between 100 and 250 keV.

Acknowledgments

Useful discussions with J. Meadows and G. DiIorio were appreciated. This work has been supported by the U. S. Department of Energy.

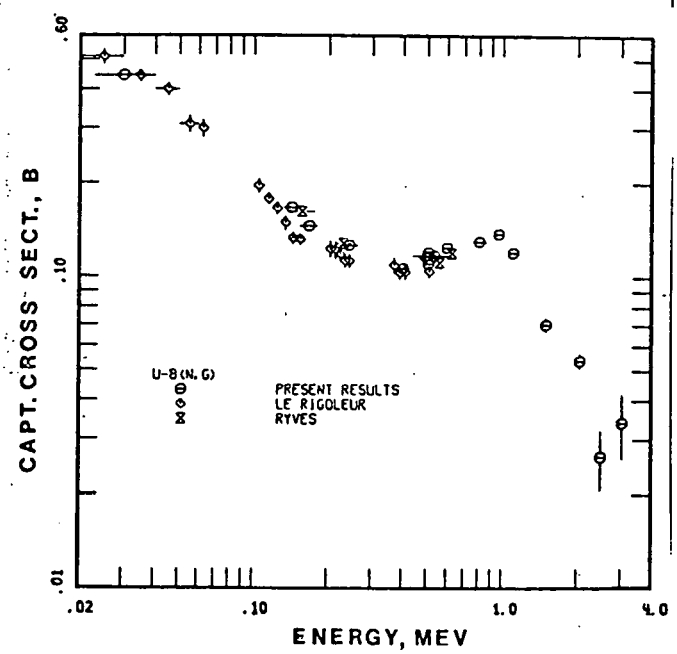

Fig. 4. Comparison of the Present Results with Other Recent Experimental Data

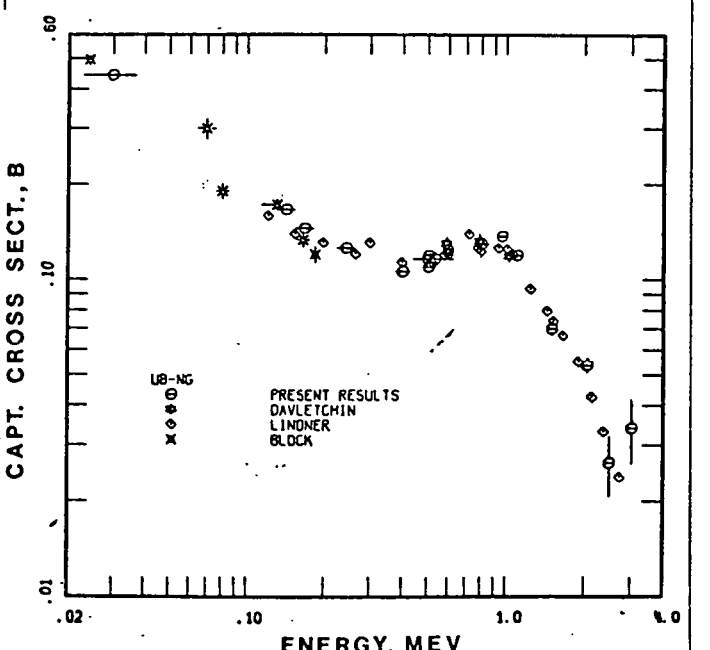

Fig. 6. Comparison of the Present Results with Other Recent Experimental Data

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