Neutron Absorption Cross Sections of U^{235} and Pu^{239} in the 5- to 50-kev Energy Range

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The absorption cross sections of U^{235} and Pu^{239} have been measured at six energies in the 5- to 50-kev energy range. Neutrons from the reaction $V^{51}(p,n)Cr^{51}$ were used in spherical-shell transmission measurements. The results show, in general, a decrease in absorption cross section with increasing energy up to about 30 kev and then a nearly constant value (up to 50 kev) of about 2.8 barns for U^{235} and 2.2 barns for Pu^{239}.

I. INTRODUCTION

THE measurement of neutron absorption cross sections of fissionable isotopes has largely been confined to energies less than one kev. A knowledge of absorption cross section (fission plus capture) in the kev region has assumed both theoretical and practical interest in the design and development of nuclear reactors.

The shell method, used in recent years in the determination of inelastic collision cross sections[1–3] in the Mev region, is used here in the determination of absorption cross sections in the kev region. The method has been discussed in detail.[3–6] A discussion of the problems peculiar to the present experiment is given in Sec. II and results are given in Sec. III.

II. EXPERIMENTAL METHOD

The spherical-shell transmission method, if combined with a uniform-response detector, offers a means of measuring neutron absorption in the presence of scattering. In the present experiment, one is faced with the added problem of secondary neutron production (fission). Thus the detector response for fission neutrons should be low compared to that for kev neutrons. Iodine was chosen as the neutron detector since it fulfills this condition and, further, has a relatively large (several barns) activation cross section in the kev region. Cylindrical NaI(Tl) crystals, 1.5 in.×1.5 in., were used.

The neutron detection sensitivity was measured as a function of crystal orientation, i.e., as a function of θ, the angle between the cylinder axis and the line from source to crystal center. For this test, an Sb-Be neutron source was used; measurements were made with each of six identical crystals oriented at each of three angles, 0°, 45°, and 90°. At any one angle, the saturated activities of the six crystals were equal to within ±1%. The asymmetry was found to be <1.5%.

The iodine activation cross section[7–9] increases with decreasing energy, so that inelastically scattered neutrons would be detected with slightly greater efficiency than unscattered or elastically scattered neutrons. Since the first excited state of Pu^{239} occurs at 49 kev,[10] and since the present measurements are made at lower energies, there is no consideration given to inelastic scattering in Pu^{239}. In the case of U^{235}, the first excited state has been reported at 13 kev,[10] although there is some question whether the lowest level given is actually the ground state. It can be shown that even if the first excited state is at 13 kev, a pessimistic value of 200 millibarns assumed for inelastic scattering cross section above 30 kev and included in the analysis changes the calculated absorption cross section less than one percent. For this reason, no consideration is given in analysis of the data to inelastic scattering in U^{235}.

The geometry of the experiment combined with background considerations indicated that the most feasible laboratory angle to use in the measurements was 0° with respect to the proton beam. This precluded such neutron sources as $Li^7(p,n)Be^7$ since center-of-mass motion produces neutrons in two energy groups below 120 kev. The reaction $V^{51}(p,n)Cr^{51}$ was used as the neutron source.[11] The low yield of neutrons from the vanadium target made necessary close spacing of components and large detector size. Target thickness corresponded to about four-kev energy loss for 1.6-Mev protons.

Shells of U^{235} and U^{238} of known purity and of weight about 5 kg were fabricated for this experiment. These shells were identical in size, with outside diameter 9.0 cm, and inside diameter 6.0 cm. A 3.6-kg Pu^{239} shell[12] was also used; the dimensions were 9.2 cm o.d. and 6.8 cm i.d. The experimental arrangement is shown in Fig. 1.

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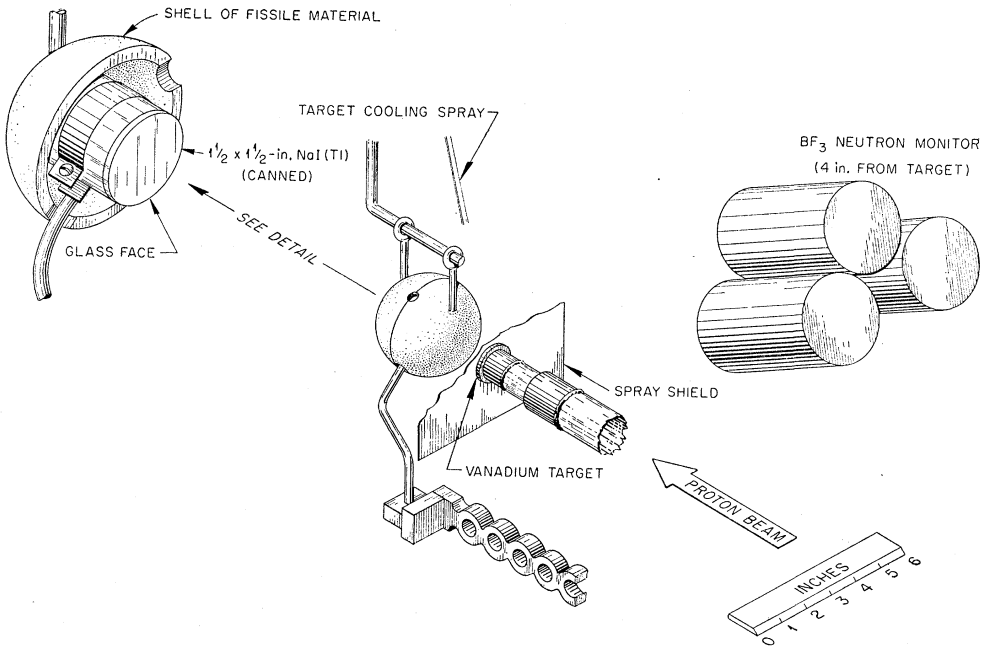

FIG. 1. Experimental arrangement. Average proton current was about 35 μa. NaI(Tl) was used as an activation detector.

In a typical experiment, a canned NaI(Tl) crystal was set in position at a standard distance (2.77±0.005 in. center-to-center) from the neutron source and in line with the proton beam. The surrounding shell was put in place and the proton beam turned on for 35.0 minutes. Cumulative counts from the BF_3 neutron monitor were recorded at 5-minute intervals to provide a basis for correcting the induced iodine activity for time variation of the neutron flux.

Proton current to the target was monitored by a current integrator, using appropriate collimation slit biasing for secondary electron collection. The neutron yield, an extremely sensitive function of proton energy,[11] was monitored with a set of three BF_3 counters in a paraffin matrix, shielded with cadmium. The monitor counting rate with proton beam stop in, was less than one percent of the rate with beam on target.

After activation the crystal was removed to a low-background counting room and mounted on an RCA 5819 photomultiplier tube. The gain was standardized with Cs^{137} 661-kev gamma radiation and the iodine beta activity ($T_{\frac{1}{2}}=25$ min) counted for a time comparable with the activation period. The amount of 15-hr Na^{24} activity induced was negligible. Decay periods of four hours or more between activations of the same crystal reduced the residual iodine activity to negligible proportions. Background counts amounted to three to six percent of the total counts.

Neutron scattering by the U^{238} shell contributed from 24% (at 4.4 kev) to 15% (at 49 kev) of the monitor count as determined by shell removal tests. For this reason, transmissions of the U^{235} and Pu^{239} shells were measured relative to that of the U^{238} shell, largely canceling this effect. A test was made at 4.4 kev to determine the monitor counting rate from fission neutrons produced by $V(p,n)$ neutron absorption in the shells. Monitor rates per proton were measured with U^{235} and U^{238} shells of identical size alternately in place. The ratios of these rates were found to be 1.00±0.01. This result is attributed to a cancellation of two small effects, fission neutron counting rate and a difference in back-scattering of $V(p,n)$ neutrons arising from the larger scattering cross section of U^{238} relative to that of U^{235}.

Normalization to either proton current integrator or BF_3 monitor gave the same value for U^{235} shell transmission relative to U^{238}, but differed by about 2% for the Pu^{239} shell relative to U^{238}. This was attributed to differences in shell thickness and total cross section. A crude calculation of the difference in scattering by the U^{238} and Pu^{239} shells predicted a 5% effect, independent of energy, to be compared with the measured 2%. The BF_3 monitor rates for Pu^{239} have therefore been decreased by 3% at each energy to allow for the relative scattering effect in computing the Pu^{239} shell transmission relative to that of the U^{238} shell. An additional uncertainty of 1.5 percent has been included in the plutonium results because of the uncertainty of this correction.

Activation by neutrons produced at locations other than the vanadium target was $\lesssim$0.7% of direct-beam activations. A single exception to this was the result of neutrons from spontaneous fission of a small amount of Pu^{240} in the plutonium shell. A shadow cone test indicated that crystal activation from $V(p,n)$ neutrons scattered by floor, walls, and air was about 1.3±0.7

TABLE I. Experimental results. Transmission of each of the fissionable shells relative to the transmission of the U^{238} shell (with all corrections applied).

E_n(kev)	$T(U^{235})/T(U^{238})$	$T(Pu^{239})/T(U^{238})$
4.4±0.4	0.641±0.015	0.865±0.025
11.8±0.5	0.700±0.015	0.892±0.025
33.1±0.4	0.802±0.015	0.958±0.025
39.6±0.5	0.786±0.015	0.930±0.025
44.0±0.5	0.806±0.015	0.921±0.025
48.4±0.5	0.793±0.015	0.915±0.025

percent of the direct-beam activation. Activation by neutrons scattered into the NaI detector by the paraffin- and cadmium-covered BF_3 monitor was shown to be $\lesssim 1.0\%$ of direct-beam activation.

The transmission of each of the fissionable shells relative to the transmission of the U^{238} shell is given in Table I. All corrections have been applied and the total uncertainty given.

III. CALCULATIONS AND RESULTS

The analysis of the data was carried out using a procedure similar to that developed by Bethe, Beyster, and Carter.[5] The ideal transmission T_∞ of the shell[4] can be calculated in terms of the absorption and total cross sections (when there is no inelastic scattering), and corrections can be applied for multiple scattering, finite source-to-detector distance, finite detector size, and detector sensitivity to fission neutrons. There are many other effects which can distort the results; the most important of these in the present measurements have been considered and shown to be small, i.e., to contribute less than 0.005 in transmission. Some of these effects are: energy loss of neutrons in elastic scattering in the shell and detector, neutron attenuation in the crystal inside the shell, variation of neutron intensity with angle, and variation of neutron energy with angle.

The observed transmission T_{obs} of a shell refers to a shell-on: shell-off measurement. This quantity can be calculated (T_{calc}) from the ideal transmission T_∞ and expressions for the corrections given above. In the analysis, T_{calc} is expressed in terms of the total and absorption cross sections, σ_t and σ_a. Since the experimentally measured quantities are values of $T_{obs}/T_{obs}(U^{238})$, a computation of $T_{calc}(U^{238})$ was made using

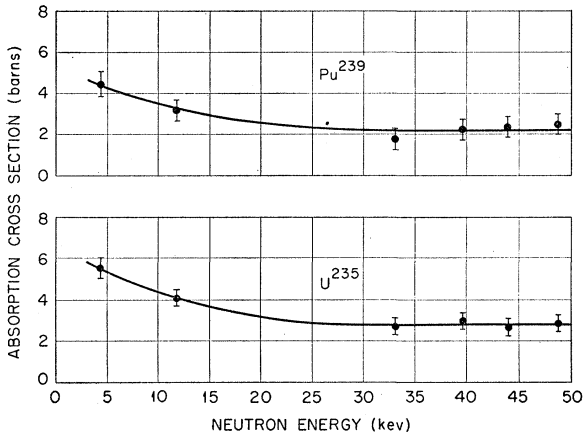

FIG. 2. Absorption cross sections of U^{235} and Pu^{239} *vs* energy.

the known total[13] and absorption[8] cross sections of U^{238}. The uncertainty introduced in these results by uncertainties in the U^{238} total and absorption cross sections is negligible.

A knowledge of the total cross sections of the fissile materials is important in determining the absorption cross sections by this method. The approximate 5% uncertainty in total cross section contributes about the same uncertainty in absorption cross section as the experimental uncertainty in T_{obs}. Another uncertainty arises from the not-well-known iodine capture cross section[7–9] at the incident neutron energies relative to that for fission neutron energies. The values of σ_t and σ_f used in this analysis were obtained by Phillips *et al.*,[1] Hibdon and Langsdorf,[13] Yeater *et al.*,[14] and Barschall and Henkel.[15]

The calculation of results, together with evaluation of the corrections and uncertainties, has been given more fully elsewhere.[16] The absorption cross sections of U^{235} and Pu^{239} as functions of energy are shown in Fig. 2.

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