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MEASUREMENTS OF (n, 3n) CROSS SECTIONS FOR 235U AND 238U

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ABSTRACT

We have measured cross sections for the ${}^{235}U(n,3n)$ reaction from 14.8 to 21 MeV, for ${}^{238}U(n,2n)$ from 14.7 to 19 MeV, and for ${}^{238}U(n,3n)$ from 14.7 to 22 MeV with ${}^{3}H(d,n)$ neutrons and a large liquid scintillator tank, and we have calculated (n,xn) and fission cross sections from 4 to 22 MeV for both nuclei using a statistical, preequilibrium model. We have also calculated the individual spectra of the two (n,2n) and three (n,3n) neutrons.

INTRODUCTION

In this paper we report (n,3n) measurements as well as calculations of the fission and (n,xn) cross sections for ²³⁵U and ²³⁸U samples at incident eutron energies up to 22 MeV. We also calculated the spectra of the individual eutrons in the (n,xn) reactions. Except for the need to correct for fission vents emitting three neutrons, the experimental method is similar to that esc_ibed previously for measurements of other (n,2n) cross sections [1], and the calculations are also similar to those of [1].

2. MEASUREMENTS

Two series of runs, one with a large sample (31 g of ²³⁸U or 18 g of ²³⁵U) and the other with a fission chamber, were necessary for each measurement. We placed the sample or fission chamber in the center of a 75-cm-diam, gadoliniumloaded liquid scintillator tank whe s it was struck by a pulsed beam of monoenergetic neutrons made by deuterons striking a tritium gas target, and we recorded the number of neutrons detected in the tank following each pulse.

A 5 x 5 cm liquid scintillator (in the neutron beam behind the tank) monitored the neutron flux. We measured the recoil protons in the upper 30% of the plateau and normalized the data using the absolute differential efficiency measurements of Verbinski, et al. [2]. The accuracy of the flux measurement is estimated to be \pm 5% or better. As a check on the flux monitor we deduced fission cross sections for the ²³⁸U runs and compared them with the known ²³⁵U(n,f) measured cross section [3]; we found agreement to within \pm 5% or better for all points except 22 MeV where the measurements were about 10% too high, presumably because the 5.5-MeV deuteron beam from the accelerator made some neutrons which were too low in energy to be monitored but high enough to cause fissions. For the ²³⁵U runs, the measured cross sections were 5-10% too high at all energies, suggesting the presence of a small lower-energy component in the neutron beam.

We measured the detector efficiency with a 252 Cf spontaneous fission source and corrected the efficiency for the difference in energy spectra between the (n,5n) neutrons and those from 252 Cf, based on our calculations of (n,3n) spectra. The efficiency was ~ 0.70 and the energy correction was < 0.035, relative to a value of $\overline{v} = 3.733$ prompt neutrons per fission for 252 Cf [4].

We measured the multiplicity of neutrons per beam pulse for a 6-cm-diam disk of the material of interest in the center of the tank. To measure the backgrounds we placed the sample on the bottom of the hole in the tank, out of the neutron beam, and repeated the run, being careful not to change the beam focal conditions and intensity. Then we replaced the sample with a fission chamber in the beam, this time recording multiplicities only for beam pulses accompanied by a signal from the fission chamber, and we determined the fission chamber backgrounds from a run in which the multiplicities were recorded for all beam pulses.

To find the (n,3n) cross section we made a small correction to all of the runs to account for dead times and then subtracted the appropriate backgrounds. For the sample run we also made corrections to the individual multiplicity probabilities of up to 0.5% for events where a fission neutron induced another fission in the sample. Next, we normalized the fission chamber run to the sample run by summing the numbers of events of multiplicity four or more [\geq 5 at energies where the (n,4n) cross section was not zero], and then we subtracted the number of three neutron fission events (normalized by the ratio of four to have neutral events for the two pass) from the three mention sample tan events to find the number of (n,3n) events. Finally, we detained cross section values by correcting for the tank efficiency, the target mass, and the incident neutron flux. At the highest energies, we used the same procedure to deduce (n,4n) cross sections. In principle it should be possible to obtain (n,2n) cross sections from our data, but unfortunately the backgrounds from scattered neutrons were high. We were able to obtain accurate (n,2n) results for ²³⁸U only where the deuteron energies were low, whereas for ²³⁵U we were completely unsuccessful in measuring (n,2n). The results of the measurements are given in Table I. The energy uncertainties arise mainly from the spread in deuteron energies in the tritium cell, while the uncertainties in the measurements include the statistical uncertainty (standard deviation), the uncertainties in the corrections, and an uncertainty of ± 0.01 in the efficiency. Not included is the estimated $\pm 5^\circ$ uncertainty in the neutron flux.

| Cross Sections for the (n,xn) Reactions on ²³⁵ U and ²³⁸ U | | | | | |
|--|------------------------|------------------------|------------------------|------------------------|--------------------------------|
| E _n (Mev) | 235 U(n,3n) (mb) | 235 U(n,4n) (mb) | 238 U(1,2n) (mb) | 238 U(n,3n) (mb) | 235 _{U(n,4n)} (mb) |
| 14.7 <u>+</u> 0.15 | | | 673 <u>+</u> 41 | 468 <u>+</u> 40 | |
| 14.8 <u>+</u> 0.20 | 87 <u>+</u> 24 | | | | |
| 16.0 <u>+</u> 0.20 | | | 451 <u>+</u> 82 | 597 <u>+</u> 62 | |
| 17.0 + 0.20 | 206 + 29 | | 366 <u>+</u> 75 | 824 <u>+</u> 62 | |
| 18.0 <u>+</u> 0.15 | | | 234 <u>+</u> 85 | 835 + 62 | |
| 19.0 <u>+</u> 0.15 | 258 <u>+</u> 27 | | 273 + 88 | 806 <u>+</u> 61 | |
| 20.0 <u>+</u> 0.12 | 265 <u>+</u> 40 | 26 <u>+</u> 28 | | 625 + 39 | |
| 21.0 + 0.12 | 277 <u>+</u> 46 | 91 <u>+</u> 32 | | 607 <u>+</u> 62 | 30 <u>+</u> 39 |
| 22.0 + 0.12 | | | | 363 <u>+</u> 80 | 135 <u>+</u> 55 |

TABLE I



Figure 1. Cross sections for $\frac{258}{9}$ U(0,xn) reactions. The open symbols are our results; circles show (n,2n), triangles show (n,3n), and squares show (n,4n) measurements. The small closed circles show (n,2n) and the dashes show (n,3n) measurements by Frehaut and Mosinski [10]. The large closed circles show (n,2n) and the closed triangles show (n,5n) measurements by Mather et al. [11]. The curves are our calculated values.

3. CALCULATIONS

We calculated (n,xn) cross sections using multistep Hauser-Feshbach statistical model techniques [5] with preequilibrium effects included using the exciton model formalism of Kalbach [6]. The equations employed in the statistical portion of the calculations are given in our earlier paper on (n,2n) and (n,3n) measurements [1]. To generate neutron penetrabilities, we used spherical optical parameters derived by Madland [7] from a fit to a large quantity of data (total cross sections, elastic scattering angular distributions, and s- and p-wave strength functions) for the uranium isotopes. Direct effects were determined through coupled channel calculations made with the JUPITOR code [8]. To represent the fission process we used discrete and continuum fission channels described by a single harmonic oscillator barrier, and we adjusted the barrier parameters of Back et al. [9], generally within their quoted errors, to obtain agreement with experimental data for the total fission cross section.

Figures 1 and 2 show our measurements (open circles) along with measurements by Frehaut and Mosinski [10] and by Mather et al. [11] at low energies, and the curves give the results of our calculations. Calculated fission cross sections are compared in Figure 3 with data of Leugers et al. [12]. For both nuclei the calculated fission cross sections generally agree with the experimental data, and such agreement, along with the use of realistic neutron optical parameter sets, provides important constraints to values calculated for (n,xn) cross sections. In Figure 1 the calculated curves are in generally good agreement with 238 U (n,xn) data. Above 15 MeV the agreement between calculated and experimental cross section results in large part from the inclusion of preequilibrium effects in the calculations; otherwise the (n,2n) curve would be much too low in this energy region. Above 20 MeV the calculated (n,3n) cross sections are higher than the measurements, perhaps in part from experimental difficulties. For 235 U (Figure 2) the agreement is not as good, perhaps because the acquisition of reliable data is not as easy, although at the highest energies the calculated (n,3n) cross section agrees reasonably well with our measurements. The dip in the calculated (n,2n) cross section near 8 MeV occurs because of competition from the (n,n'f) reaction.

The calculations of the spectra of the first, second, and third (n,3n) neutrons are of interest in model studies as well as being useful in correcting the efficiency measurements. The results of the uranium calculations are qualitatively very similar to those found for ¹⁹⁷Au(n,3n) and other heavy samples [1].

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Figure 3. Calculated cross sections for ²³⁵U(n,f) and ²³⁸U(n,f) compared with the measured points of Leugers et al. [12]. (For clarity some of the lower energy measurements are not shown.)

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