DEVELOPMENT OF A TECHNIQUE FOR MEASURING CROSS SECTIONS OF INTEREST TO ACCELERATOR TRANSMUTATION OF WASTE (ATW)

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ABSTRACT

It has been suggested that transmutation of actinide waste into fission products could be enhanced by using resonance fission of odd-odd target materials; those of interest are ^{232}Pa, ^{238}Np, and ^{242}Am. Our technique for carrying out the measurement of the resonance fission cross section of short-lived materials has three steps: 1) We produce the sample by the (d,2n) reaction at the Los Alamos Ion Beam Facility. 2) We carry out fast radiochemistry to separate the odd-odd target of interest. 3) We measure the fission rate in a high intensity pulsed neutron beam produced by 800 MeV proton spallation. Using this technique, we have successfully measured the neutron-induced fission cross sections of 1.3 day ^{232}Pa and 2.1 day ^{238}Np, from 0.01 eV to 50 keV.

I. INTRODUCTION

Following the suggestion by Bowman et. al.[1] that the transmutation of actinide waste into fission products may be enhanced by using resonance fission of odd-odd nuclei, it became clear that there was an urgent need to know the resonance properties of the nuclides of interest. The measurement of the fission cross section of isotopes with half-lives of a few days requires a neutron beam of very high intensity, but of modest resolution. The facility that is probably the best in the world for such measurements is the Los Alamos Neutron Scattering Center (LANSCE), which uses moderated spallation-neutron pulses from the Proton Storage Ring (PSR), driven by 800 MeV protons from the Clinton P. Anderson Meson Physics Facility (LAMPF).

The method of choice for producing sample material of mass number A for these measurements is by neutron irradiation of isotopes with mass number A-1 in a high flux thermal reactor. We intended, at the time that we began planning these measurements, that this method would be used. If we assume a modest 10μg of ^{231}Pa, ^{237}Np, or ^{241}Am as the starting material, and irradiate it in a thermal neutron flux of 6 x 10^{13} n/cm^2-sec for one half life of the isotope of interest, we obtain a quite adequate amount of sample--20 to 30 ng. (As it turns out, the solution of the Bateman equation for all three of the targets of interest gives roughly the same answer, and the amount of material produced is very nearly proportional to the reactor flux at these relatively low levels.) At the time we planned the measurement, the Omega West Reactor (OWR) at the Los Alamos National Laboratory was fully operational, and the 6 x 10^{13} flux level is readily obtainable in the pneumatic rabbit irradiation ports. By the time the 1993 LAMPF/PSR cycle began, however, the OWR had been shut down for extended repair. We considered two possible alternatives: the Advanced Test Reactor (ATR), at the Idaho National Engineering Laboratory, with a thermal neutron flux of 6 x 10^{14} n/cm^2-sec, or the Annular Core Research Reactor (ACRR) at the Sandia National Laboratory, which is currently configured to give about 4 x 10^{12} neutrons/cm^2-sec below 1 eV. The ACRR is clearly less than optimal, but the main reason we chose a much more exotic method of producing the sample was because of logistics. Moving the irradiated material on public highways, or, in the case of the ATR, by aircraft to Los Alamos in a reasonable time period after the irradiation was not easy. We chose to avoid the transportation problem by producing the odd-odd fissile isotopes on site, i.e., at the Laboratory, by charged-particle irradiation at the Ion Beam Facility (IBF).

II. SAMPLE PRODUCTION

The first step in producing the required samples was to measure the cross sections for (p,xn) and (d,xn) reactions as a function of the energy of the charged particle for ^{232}Th and ^{238}U targets. The cross sections for (p,n) and (p,3n) reactions with ^{232}Th are shown in Fig. 1. We found that the (p,n) and (p,3n) cross sections of ^{238}U show qualitatively the same energy dependence, but are about fifteen percent lower. Clearly, however, protons are not suitable for producing the isotopes of interest to the ATW program; the odd-odd isotopes ^{230}Pa and ^{236}Np are produced with up to ten times more efficiency than the isotopes of primary interest, ^{232}Pa and ^{238}Np, and are expected to have even larger fission cross

sections for low energy neutrons. The next step was to measure the cross sections of ^{232}Th and ^{238}U for deuterons. The results, shown in Fig. 2 for ^{232}Th, suggest that this will give an adequate sample. Even though the (d,n) cross section is about four times larger than the (d,2n) cross section, neither ^{233}Pa nor ^{239}Np is expected to have an appreciable neutron-induced fission cross section, and the fission cross sections of their decay products, ^{233}U and ^{239}Pu, are well known. In fact, their presence is an advantage in that it provides a means of normalizing the measurement.

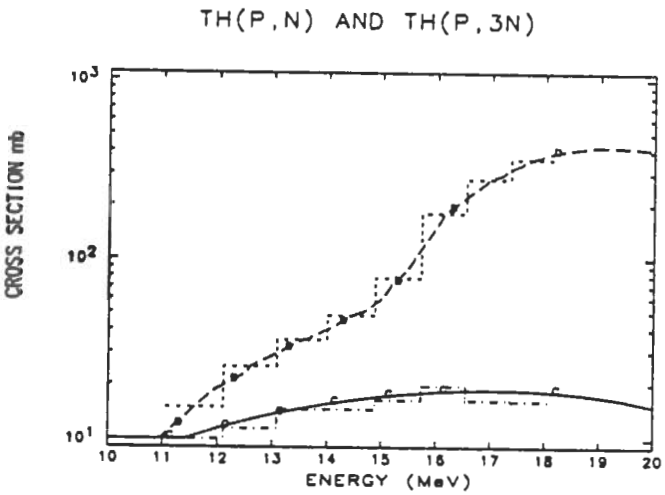

Fig. 1. Cross section, in mb, for ^{232}Th(p,n), shown as histogram C, and for ^{232}Th(p,3n), shown as histogram D. The measured cross sections for proton irradiation of ^{238}U have the same shape, but are fifteen percent lower.

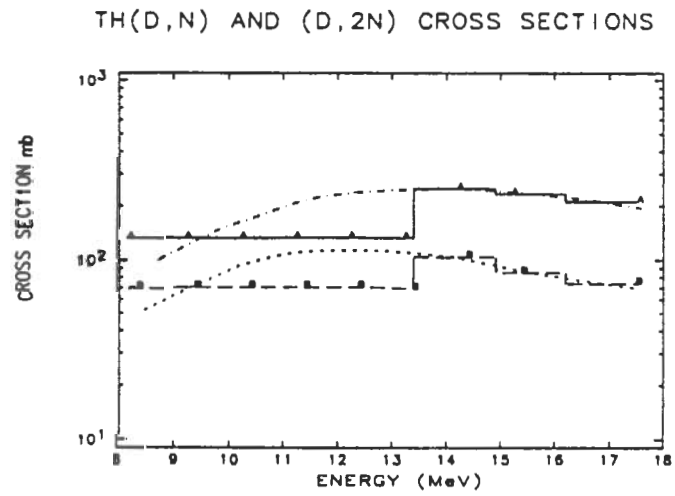

Fig. 2. Cross sections for ^{232}Th(d,n), in mb, shown as histogram A, and for ^{232}Th(d,2n), shown as histogram B. The measured deuteron cross sections for ^{238}U have roughly the same shape, but are only two-thirds as large.

We carried out one successful measurement of ^{232}Pa and two of ^{238}Np. Each of these consisted of irradiation of Th or ^{238}U at the IBF, followed by fast chemistry to separate the protactinium or neptunium, which was then deposited on a backing, assembled into a fission chamber, transported to the LANSCE facility, and inserted into the neutron beam. The procedure was not without problems. In particular, the ^{238}U foils occasionally burned through if the deuteron currents were too high or if the beam spots were too sharply focused, which led to catastrophic vacuum failure and contamination of the IBF beam line. As a result, we did not attempt to field a sample of 16-hour ^{242g}Am, which would have been produced by deuteron irradiation of ^{242}Pu.

III. FISSION CROSS SECTION MEASUREMENTS

A compensated ionization chamber with five parallel plates was used to measure the time-dependent fission rates. In this chamber, the plate spacing was 0.5 cm. The first and last plates were the anodes, at +400V. The second and fourth plates were the cathodes, at ground potential, from which the fission fragment pulses were taken. On the fourth plate, there was a deposit of ~400ng of ^{235}U as a flux monitor. On the second plate was the short-lived odd-odd isotope to be measured, facing the first anode. The middle plate served as a compensation cathode, operated at -400V, to balance the beta-gamma background from the short-lived sample.

The data were collected as event addresses in a two-dimensional array of pulse height and neutron time of flight. The data were read out every hour to permit optimization of the counting-loss corrections and statistical weighting of partial runs. The ^{235}U fission rate served only as a relative intensity and flux-shape monitor. The cross section of ^{232}Pa was determined relative to that of ^{233}U through the protactinium isotopic ratios after the sample had partially decayed; that of ^{238}Np was determined relative to ^{239}Pu.

The successful run to produce ^{232}Pa began at the IBF at 3:30 p.m. on October 6, and ended at 6 a.m. on October 8, 1993. We estimated, from the measured cross sections and the deuteron beam intensity, that we had produced about 90 ng of ^{232}Pa at the end of the irradiation. The irradiated thorium was removed from the IBF beam line, packaged, and shipped to a hot cell in the Chemistry-Metallurgy Research building, where the radiochemical separation was begun at 9:30 a.m. The final elution to separate protactinium and uranium was completed at 8:30 p.m.; the protactinium was then deposited onto the fission foil backing, assembled into the fission chamber, packaged, and shipped to the LANSCE facility. There it was unpacked, inserted into the beam line, connected to the electronics, and data collection began at 1:20 a.m. on October 9. At that time, we had about 30 ng of ^{232}Pa in the chamber, and about 130 ng of ^{233}Pa. Data collection continued until 4 p.m. on October 13; by this time 90% of the ^{232}Pa in the chamber had decayed.

A preliminary data reduction was carried out by subtracting a pointwise ^{233}U fission cross section, normalized to the area

of the 1.8-2.3 eV doublet.[2] The correction for the ^{232}U fission resonances and 1/v component was based on the BNL-325 resonance parameters.[3] Fig. 4 shows the fission cross section of ^{232}Pa below 5 eV. The solid line is a single-level Breit-Wigner fit corresponding to the parameters of Table I.

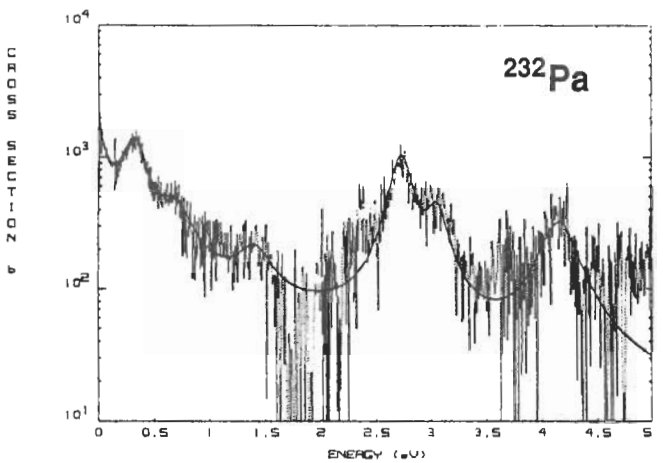

Fig. 4. The fission cross section of ^{232}Pa as a function of neutron energy below 5eV. The solid line is a single-level fit to the data, the parameters of which are listed in Table I.

Fielding of the ^{238}Np samples followed a very similar pattern, with one extra complication. In the separation of neptunium from uranium, we found that extreme care was needed to avoid the presence of ^{235}U in the fissile deposit; even a trace amount could compromise the quality of the ^{238}Np data. The two samples of ^{238}Np consisted of about 6 ng each. The fission cross section of ^{238}Np below 2 eV is shown in Fig. 4; the solid line is a single-level Breit-Wigner fit corresponding to the parameters in Table II.

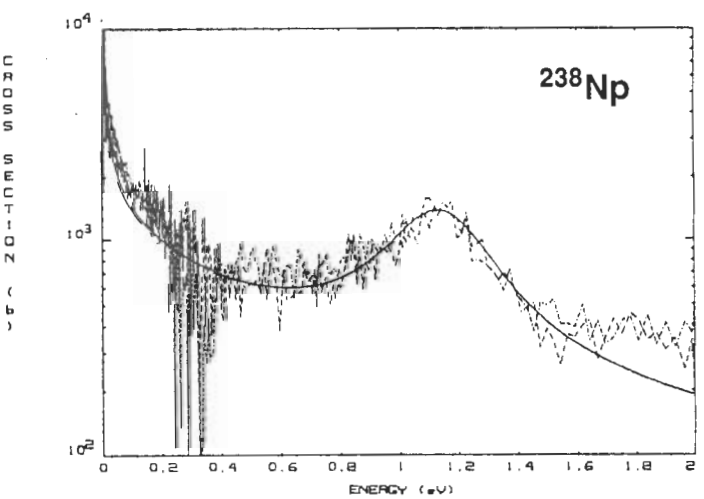

Fig. 4. The fission cross section of ^{238}Np below 2 eV. The solid line is a preliminary fit to these data corresponding to the parameters listed in Table II.

Table I. Resonance parameters of ^{232}Pa.

E_o (eV)	$2g\Gamma_n^o$ (meV)	Γ_f (meV)	Γ_γ (meV)
-5.0	14.0	400.	40.
0.33	0.067	191.	40.
0.69	0.030	300.	40.
1.40	0.019	300.	40.
2.73	0.16	177.	40.
3.05	0.054	170.	40.
4.15	0.094	335.	40.

Table II. Resonance parameters of ^{238}Np.

E_o (eV)	$2g\Gamma_n^o$ (meV)	Γ_f (meV)	Γ_γ (meV)
-5.0	37.0	400.	40.
1.14	0.175	335.	40.

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