BNL--32166

DE83 003176

Conf-821123--15

BNL-32166

COMPARISON OF THE (p,xn) CROSS SECTIONS FROM ^{238}U, ^{235}U AND ^{232}Th TARGETS IRRADIATED WITH 200-MeV PROTONS **

Y. Y. Chu and M. L. Zhou*
Department of Chemistry
Brookhaven National Laboratory
Upton, NY 11973

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

MASTER

Summary

We have measured absolute cross sections for (p,xn) reactions (x ranges from 0 to 8) from ^{238}U, ^{235}U, and ^{232}Th targets irradiated with 200 MeV protons at the Brookhaven AGS Linac injector. Chemical yields were determined by using ^{239}Np and ^{233}Pa as tracers. Yield patterns obtained in this work can be compared to the experimental results and theoretical calculations from earlier work, and they are consistent within the framework of intranuclear cascade followed by neutron evaporation and fission competition.

Introduction

Ever since the installation[1] of the Chemistry Linac Irradiation Facility (CLIF) and the Medium Energy Intense Neutron (MEIN) source at the 200 MeV Linac injector of the Brookhaven Alternating Gradient Synchrotron (AGS), irradiations of thin targets for short intervals followed by pneumatic transfer to a laboratory for chemical separations and radioactivity measurements have been carried out routinely. In addition to a number of investigations[2-5] on neutron-rich nuclides, there have been systematic studies[1,6] of production cross sections of medium-energy neutron induced reactions. However, only a few cross sections have been measured for reactions induced by 200 MeV protons.

There was considerable interest in studying competition between fission and nucleon (especially neutron) emission in excited heavy nuclei more than two decades ago. Measurements were made on products of simple spallation reactions, among them (p,xn) reaction products from ^{232}Th-targets[7-10] and two neptunium nuclides from ^{238}U-targets.[7,10] Calculations[11] were performed combining results from intranuclear cascade calculations,[12] with certain treatments of neutron evaporation and fission competition. Reasonable agreement was obtained with experimental results. In the intervening years, there have only been a few measurements[13,14] in the heavy-element region but none for uranium targets. There have been calculations[15,16] containing improvements and refinements for comparisons with early experimental data, the general conclusions remain unchanged.

In view of the important role fission plays in the spallation yield pattern among the heavy-element nuclides and the paucity of (p,xn) reaction cross sections from ^{238}U-targets (none from ^{235}U-targets) and the fact that none of the previous measurements were made using high-resolution γ-spectroscopy, we have recently measured (p,xn) cross sections for ^{238}U, ^{235}U and ^{232}Th targets irradiated with 200 MeV protons at CLIF by employing radiochemical methods and high-resolution γ-spectrosocpy.

Experimental

Irradiations

All irradiations were carried out at the CLIF facility. Target packages consisted of ^{238}U, or enriched ^{235}U (93.2%), or ^{232}Th target foils of thickness from 0.0015-in. to 0.003-in. sandwiched between aluminum guard foils, monitor foils and in an envelope. The irradiations were typically 10 minutes in length and the proton beam current was reduced to about 5 μA to avoid melting the target. In one of the experiments of the thorium target, the irradiation was carried out at the Brookhaven Linac Isotope Producer (BLIP) facility under comparable irradiation conditions.

Preparation of tracers

In order to measure absolute cross sections for the reactions studied in this work, ^{239}Np and ^{233}Pa tracers were used to determine the chemical yield in the purification procedures for each sample. They were prepared in the following manners:

A small amount (10-20 mg) of natural uranium metal was sealed in quartz tube and irradiated at the Brookhaven High Flux Beam Reactor (HFBR) for 30 seconds with a flux of ~1 x 10^{14} n/cm^2/sec. After waiting for several hours to allow ^{239}U to decay into ^{239}Np, the target was dissolved in 8 M HNO_3. Neptunium was retained on an MP1 anion exchange column after it was reduced with ferrous sulfamate and sodium nitrite; following washings with conc. HNO_3 and 8M HCl it was eluted with 0.3 M HNO_3. Aliquots from the eluent containing ^{239}Np were used for chemical yield determinations for neptunium.

A small amount (~10 mg) of thorium nitrate sealed in quartz tube was irradiated at the HFBR for several minutes. After ^{233}Th had completely decayed into ^{233}Pa, the sample was dissolved and protactinum was eluted from a Dowex 50 cation exchange column in 1.8 M HCl solution while thorium was retained on the column. Aliquots of the eluent containing ^{233}Pa were used for chemical yield determinations for protactinium.

Chemical purification for neptunium

Exhaustive chemical purifications were needed to separate the neptunium samples from the large amount of fission products present. In addition to the procedure used for the preparation of ^{239}Np tracer described above, the following steps were also included. Neptunium was extracted into 0.5 M TTA in xylene from a 1 M HCl-1 M $NH_2OH{\cdot}HCl$-0.25 M $FeCl_2$ solution. Back extraction into 10 M HNO_3 followed the washing of organic phase several times with 1 M HCl. Then the back-extracted aqueous phase was washed with TTA-xylene solution twice before neptunium was finally coprecipitated with $Fe(OH)_3$ by the addition of ferric carrier, hydrogen peroxide and conc. NH_4OH to the aqueous solution.

Chemical purification for protactinium

Thorough chemical purification was also needed for preparing the protactinium samples. The irradiated thorium target was dissolved in conc. HNO_3 and conc. HCl with a few drops of conc. $HClO_4$ and $(NH_4)_2SiF_6$. Protactinium was first retained on an MP1 anion exchange column from 8M HNO_3 and conc. HCl solutions while some fission products and thorium targets were removed. Protactinium was eluted with 1.8 M HCl and further purified when the same solution was allowed to pass through a Dowex 50 cation exchange column. Protactinium was then extracted from 9 M HCl solution into diisobutyl carbinol (DIBC) loaded on a

*Visitor from Institute of Nuclear Science and Technology, Sichuan University, Chengdu, Sichuan, China.

**Research carried out under the auspices of the U.S.Dept. of Energy under contract # DE-AC02-76CH00016.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Teflon-6 column; after washing, it was back-extracted by eluting with 12 M HCl-1 M HF solution. It was finally coprecipitated with $Fe(OH)_3$ by the addition of ferric carrier and conc. NH_4OH to the aqueous solution.

Counting

The γ-rays of the samples were counted on a calibrated Ge(Li) detector, 50 cm^3 in size with FWHM of 2.0 keV at 1332 keV. Measurements were made at the lowest possible geometry to reduce coincident summing and the decays of the γ-rays of interest were followed to ascertain the correct half-lives. The γ-ray spectra were recorded on magnetic tapes using a 4096 channel analyzer system and were analyzed later with the INTRAL code.[17] The monitor foil was assayed on a calibrated gas-flow proportional counter to determine the ^{24}Na activity produced by $^{27}Al(p,3pn)^{24}Na$ reaction. The cross section used for this monitor reaction was 9.3 mb.[18] The least squares code CLSQ[19] was used for decay curve analyses.

Results and Discussion

Table 1 lists the relevant properties of the nuclides of interest. With the exception of the half-life of ^{228}Pa, which was re-determined in this work, the rest were taken from Ref. 20. The calculated cross sections based on the counting data obtained in this work and the properties given in Table 1 are listed in Table 2. We have measured all the cross sections for (p,xn) reactions from x=0 to x=8 for ^{238}U target (except x=2 where the half-life of the reaction product is too long). Because the product nuclide for (p,γ) reaction is the same as the tracer nuclide used for the chemical yield determination, the cross section for this reaction was determined indirectly as follows: The relative yields of all neptunium nuclides were measured in two other experiments where no ^{239}Np tracer was added and the yield for ^{239}Np was converted to cross section when the yield for ^{238}Np was matched to the average value of the cross section for ^{238}Np determined in those experiments where ^{239}Np tracer was added. Since we did not measure, in the same experiments, the contribution of ^{239}Np produced by (n,γ) reaction followed by β decay, the (p,γ) cross section must be considered as upper limit. The cross section for (p,3n) reaction is partial because we only measured one of the two isomers of ^{236}Np. We were not able to measure the cross sections for (p,6n), (p,7n), and (p,8n) reactions for ^{235}U target because of the short half-lives and the lack of known and measurable γ-rays of these products. We only measured four reaction products from ^{232}Th target because of half-life and measurable γ-ray constraints. The listed uncertainties of the cross sections represent the greater of either the combination of counting statistics, detector efficiency and chemical yield uncertainties or the standard deviation of the different measurements from the mean.

Since there have been several studies[7-10,13] on (p,xn) reaction cross sections for thorium target with proton energies ranging from 82 MeV to 680 MeV, we list these results with those obtained in this work in Table 3 for comparison. Also included are the earlier measurements on the two neptunium nuclides from ^{238}U target. There is a trend of decrease in cross sections and a gradual shift of maximum to higher value of x with increase in proton energy. Our results fit in the overall trend though some of our cross sections are lower than those from 340 MeV protons. Since ours is the only measurement made with high-resolution γ-ray spectroscopy, such differences should not be taken too seriously.

Table 1. Relevant properties of the nuclides measured in this investigation

Nuclide	Half-Life	E_γ (keV)	Abundance[a]
^{227}Pa	38.3m	65.0	5.0
^{228}Pa	20.0h[b]	911.2	16.0
^{230}Pa	17.7d	952.0	27.8
^{232}Pa	1.31d	150.1	12.0
		472.8	4.0
^{231}Np	48.8m	348.4	2.7
		370.9	7.3
^{232}Np	14.7m	327.3	52.0
		819.5	33.3
^{233}Np	36.2m	299.1	0.59
		312.1	0.70
^{234}Np	4.40d	1194.1	5.5
		1435.7	6.3
		1527.5	11.8
		1558.7	19.0
		1602.2	9.7
^{235}Np	396.1d	111.0 (U $K\beta_1$ x-rays)	0.263
$^{236}Np^m$	22.5h	642.4	1.0
^{238}Np	2.12d	984.5	27.8
		1028.5	20.4
^{239}Np	2.35d	227.8	11.4
		277.5	14.5

[a] Number of photons per 100 disintegrations.
[b] Determined in this work.

Table 2. Cross sections for the different reaction products for ^{238}U, ^{235}U, and ^{232}Th targets

Reaction	σ(mb) ^{238}U-target	^{235}U-target	^{232}Th-target
(p,γ)	0.27 ± 0.07[a]		
(p,n)	1.15 ± 0.15	0.48 ± 0.05	0.56 ± 0.06
(p,2n)		0.68 ± 0.07	
(p,3n)	1.46 ± 0.15[b]	0.47 ± 0.05	1.9 ± 0.2
(p,4n)	1.02 ± 0.14	0.14 ± 0.015	
(p,5n)	0.38 ± 0.04	0.047± 0.006	1.75 ± 0.25
(p,6n)	0.15 ± 0.03		0.37 ± 0.04
(p,7n)	0.046± 0.010		
(p,8n)	0.014± 0.004		

[a] Upper limit, see text.
[b] Partial cross section, see text.

Figure 1 shows the distribution of cross sections for (p,xn) reactions versus x for these three targets. In the two recent calculations[15,16] the significant differences for the final product distributions for ^{238}U target without and with fission competition were clearly demonstrated. The former showed a relatively flat distribution over the entire range of products and the latter showed rapid decline of yields with decreasing mass number for (p,pxn) reactions with 340 MeV protons. The calculation from Ref. 15 also reproduced the observed increase in yield with decreasing mass from ^{238}Np to ^{236}Np. Though this type of calculation was not repeated in this investigation, we believe the general shape of the distributions obtained in this work could be reproduced, at least qualitatively, by the framework of intranuclear cascade followed by neutron evaporation and fission competition.

The relative magnitudes of the cross sections between ^{238}U target and ^{235}U target seem to suggest that even a slight increase of fission probability from ^{238}U to ^{235}U could lead to a substantial

Table 3. Comparison of $^{232}Th(p,xn)^{233-x}Pa$ and $^{238}U(p,xn)^{239-x}Np$ cross sections at different proton energies

Target	Reaction	Product	E_p = 82 MeV[a]	100 MeV[b]	150 MeV[a]	σ(mb) 200 MeV[c]	340MeV[d]	680 MeV[e]
^{232}Th	(p,n)	^{232}Pa	13±2		8±1	0.56±0.06	2.6±1.2	
	(p,3n)	^{230}Pa	12±2		7.5±1.0	1.9±0.2	4.2±0.3	
	(p,4n)	^{229}Pa			5.0±1.5			2.3±0.3
	(p,5n)	^{228}Pa	9 ± 1		3.25±0.50	1.75±0.2	1.7±0.2	0.8±0.2
	(p,6n)	^{227}Pa	3.2±0.5	4.8	1.5±0.2	0.37±0.04	1.0±0.2	0.26±0.04
	(p,7n)	^{226}Pa		2.1	1.0±0.3			
^{238}U	(p,n)	^{238}Np				1.15±0.15	0.46±0.05	0.25±0.04
	(p,3n)	$^{236}Np^m$				1.46±0.15	1.7±0.1	0.26±0.03

[a] Ref. 9, except (p,7n) cross section at 150 MeV is from Ref. 13. [b] Ref. 14. Cross sections not given in text, read off from graphs. [c] This work. [d] Ref. 7. [e] Ref. 10.

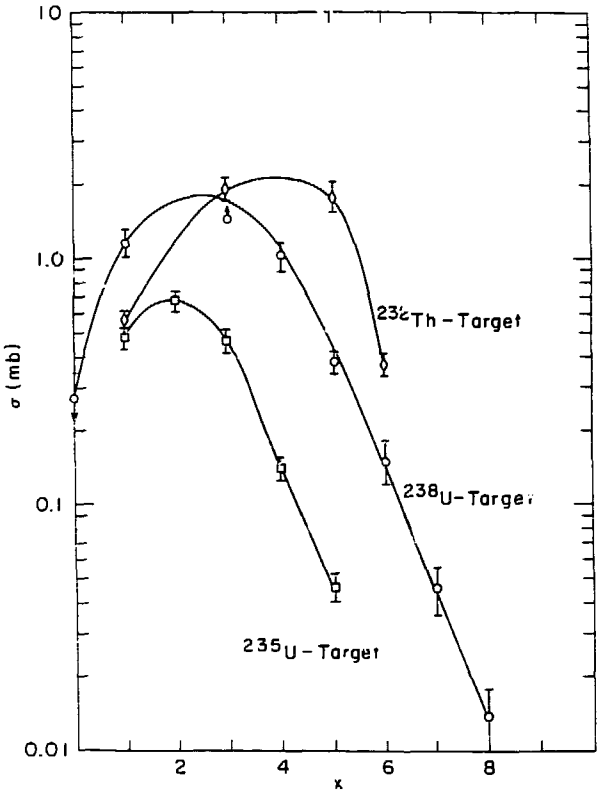

Fig. 1. Cross sections of (p,xn) reactions for ^{238}U, ^{235}U, and ^{232}Th targets irradiated with 200 MeV protons.

reduction in Γ_n/Γ_f, thus resulting in the much lower (p,xn) cross sections for ^{235}U. The fission probability for ^{232}Th and the Z^2/A values for different product nuclides are probably responsible for the observed distribution for ^{232}Th target.

References

1. S. Katcoff, J. B. Cumming, J. Godel, V. J. Buchanan, H. Susskind, and C. J. Hsu, Nucl. Instr. and Methods, 129, 473 (1975).

2. E. M. Franz, S. Katcoff, H. A. Smith, Jr. and T. E. Ward, Phys. Rev. C12, 616 (1975).

3. P. E. Haustein, H. C. Hseuh, R. L. Klobuchar, E. M. Franz, S. Katcoff and L. K. Peker, Phys. Rev. C19, 22,32 (1979).

4. J. Gilat, S. Katcoff and L. K. Peker, Phys. Rev. C21, 2041 (1980).

5. P. P. Parekh, E. M. Franz, S. Katcoff and L. K. Peker, Phys. Rev. C24, 2240 (1981).

6. S. Mirzadeh, P. P. Parekh, S. Katcoff and Y. Y. Chu, to be published (private communication).

7. M. Lindner and R. N. Osborne, Phys. Rev. 103, 378 (1956).

8. W. W. Meinke, G. C. Wick and G. T. Seaborg, J. Inorg. and Nucl. Chem. 3, 69 (1956).

9. M. Lefort, G. N. Simonoff and X. Tarrago, Nucl. Phys. 25, 216 (1961).

10. B. D. Pate and A. M. Poskanzer, Phys. Rev. 123, 647 (1961).

11. M. Lindner and A. Turkevich, Phys. Rev. 119, 1632 (1960).

12. N. Metropolis, R. Bivins, M. Storm, A. Turkevich, J. M. Miller, and G. Friedlander, Phys. Rev. 110, 185 (1958).

13. M. Hussonnois, J. de Physique, 25, 747 (1964).

14. E. C. Suk, J. E. Crawford and R. B. Moore, Nucl. Phys. A218, 418 (1974).

15. R. L. Hahn and H. W. Bertini, Phys. Rev. C6, 660 (1972).

16. V. S. Barashenkov, F. G. Gereghi, A. S. Iljinov and V. D. Toneev, Nucl. Phys. A222, 204 (1974).

17. J. B. Cumming, based on modifications of the program by R. Gunnick, H. B. Levy, and J. B. Niday, University of California, Radiation Laboratory, Livermore, Report No. UCID-15140 (1967) (unpublished).

18. J. B. Cumming, Annu. Rev. Nucl. Sci. 13, 261 (1963).

19. J. B. Cumming, (NAS-NS-3107), Application of Computers in Nuclear and Radiochemistry, G. D. O'Kelly, Ed., U. S. Atomic Energy Commission (1962), p. 25.

20. Table of Isotopes, 7th ed., edited by C. M. Lederer and V. S. Shirley, 1978, John Wiley and Sons, Inc.