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THE PRODUCTION OF NEUTRON-RICH NUCLIDES IN THE HEAVY-ELEMENT REGION VIA ^{3}He-INDUCED REACTIONS**

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DISCLAIMER

Summary

We have measured the production cross sections for ^{233}Th and ^{231}Th from the bombardment of ^{238}U with ^{3}He ions at 46-, 53-, and 60-MeV at the Brookhaven 60-in. isochronous cyclotron. We have also attempted to observe the decay of ^{233}Ac produced via ^{238}U(^{3}He,^{8}B) or equivalent reactions using 61 MeV ^{3}He ions by first separating thorium from actinium and then performing chemical purifications on the second thorium sample into which the actinium has decayed. In the four experiments we performed, three gave results consistent with the β half-life of ^{233}Ac somewhat longer than 120 s and the production cross section from this target-projectile combination in the order of 1-2 μb.

Introduction

Among the commonly used methods to produce neutron-rich nuclides outside the fission product regions are photonuclear reactions[1], medium- or high-energy proton induced reactions[2-5] and medium-energy neutron induced reactions.[6-7] There are distinct advantages and drawbacks for each. Photonuclear reactions yield relatively simple product mixtures but only a limited number of products can be expected. The medium- and high-energy proton induced reactions result in complex product mixture in which the more-favored neutron-deficient nuclides tend to mask the less-favored neutron-rich nuclides. High-energy proton induced reactions, however, have been used successfully in the study of neutron-rich nuclides among the light elements[4,5] where the valley of stability is narrow and the on-line particle identification technique could be employed. Medium-energy neutron induced reactions have been shown[8] to be effective for producing neutron-rich nuclides if there is an adequate neutron flux.

Heavy-ion induced reaction have generally been used to produce neutron-deficient nuclides mainly via (HI,xn) reactions because of the neutron-to-proton ratio of the compound nucleus and the Coulomb barrier for charged particle emissions. There are, however, special examples to produce neutron-rich nuclides in the light-element region by multinucleon transfer[9] or by fragmentation of the heavy ion beam.[10] Although ^{3}He is not usually considered as a typical heavy ion, being the most neutron-deficient complex nuclide (by virtue of its neutron-to- proton ratio of 0.5), it has been used mainly to produce neutron-deficient nuclides. However, exotic reactions may lead to products on the neutron-rich side of stability. Kashy et al.[11] have used a 74 MeV ^{3}He beam to produce the highly neutron-rich ^{43}Cl and ^{59}Mn via a five-nucleon pickup reaction. They measured the masses of ^{43}Cl and ^{59}Mn by recording the energy spectra of ^{8}B ions and estimated the cross section for the five-nucleon pickup (^{3}He,^{8}B) reaction to be in the order of a few nb/sr.

Since these measurements were made by using very thin targets for this particular reaction channel, we thought it might be feasible to observe other neutron-rich nuclides produced via such exotic nuclear reactions by using a radiochemical method.

As a continuation of recent investigations at Brookhaven, our interest has been centered on the heavy-element region. A quick survey of this region reveals that ^{233}Ac is a prime candidate for such study. It has never been observed in the laboratory, it has an estimated partial β half-life of 80 s[12] and it can be produced by ^{238}U(^{3}He,^{8}B) or equivalent nuclear reactions.

Since the highest available energy of the ^{3}He ions at the Brookhaven cyclotron is only 62 MeV, one should ask whether this energy is high enough to surmount the Coulomb barrier for the ^{238}U(^{3}He,^{8}B)^{233}Ac reaction. Depending on the value for r_0 used, the calculated Coulomb barrier varies from 52.4 MeV for r_0 = 1.5 f to 65.5 MeV for r_0 = 1.2 f. However, unlike the observation made in Ref. 11, the sample prepared by the radiochemical method should also include ^{233}Ac produced in all other equivalent reaction channels where the barrier is less. This will enhance the probability of observing ^{233}Ac using this target-projectile combination.

The next question then is even if ^{233}Ac were to be produced, what is the chance of observing and identifying it. Since the bulk of the reaction products is expected to consist of fission products with cross sections many orders of magnitude higher than that of ^{233}Ac, exhaustive chemical purification would be required and the short-lived ^{233}Ac might decay away before the purification is completed. A method often used to identify a short-lived nuclide is through the observation of its known descendent(s). In the case of ^{233}Ac, its daughter nuclide ^{233}Th has a half-life of 22.2 m, sufficiently long to permit thorough chemical purification procedures. In fact, the exhaustive purification of thorium from the mixture of large amount of fission products has recently been worked out.[13]

There is also the need to separate thorium from actinium initially before ^{233}Ac is allowed to decay into ^{233}Th, because in addition to the β decay of ^{233}Ac, ^{233}Th could also be produced directly by ^{238}U(^{3}He,2α) or equivalent reactions. The cross sections for these latter reactions may very well be much higher than those of the ^{238}U(^{3}He,^{8}B) or equivalent reactions. Therefore, during the early part of this investigation we measured the production cross sections for ^{233}Th and ^{231}Th from ^{238}U via (^{3}He,2α) and (^{3}He,2α2n) reactions at three different ^{3}He energies between 46- and 60-MeV. Later, we attempted to observe the decay of ^{233}Ac produced in ^{238}U(^{3}He,^{8}B) or equivalent reactions from the thorium samples into which actinium has decayed. For the rapid thorium and actinium separation we have chosen the very selective solvent extraction with HDEHP/toluene from 8.0 M HCl solution.[14]

Experimental

Irradiations

All the irradiations were carried out at the Brookhaven 60-in. isochronous cyclotron, with a ^{3}He energy of ~ 62 MeV. After the beam passed through a collimator window, it was allowed to impinge on the

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target which was mounted on a water-cooled aluminum block. For the irradiations at the highest energy, the collimator window (0.0015-in. thick Dural foil) was removed and the target block became an integral part of the cyclotron vacuum system. The beam current (1.0-3.5 μA) was measured with a Faraday cup and it was later used to calculate the production cross sections. Irradition time varied from 5 to 20 minutes.

Both uranium oxide and uranium foil (0.0015-in. thick) were used as targets. To prepare the oxide sample, uranium metal was dissolved in 10 M HNO_3; after passing through two MP1 anion exchange columns, the peroxide was precipitated at pH 2-3 with NH_4OH; H_2O_2 was added in excess and the sample chilled. After filtration and drying, the peroxide was converted into oxide at 850°C and then pressed into a pallet.

Chemical Procedures

Immediately after the irradiation, the target was removed for chemical processing. It was dissolved in con. HCl with a few drops of conc. HNO_3 and conc. $HClO_4$ and 2-3 mg of thorium carrier. The initial steps varied depending on the purpose of that particular experiment. If it was to measure the cross sections of the thorium nuclides produced directly in the irradiation, then the purification of thorium began. If it was an attempt to observe ^{233}Ac, then a separation of thorium from actinium was performed first. This was accomplished by extracting thorium from a 8 M HCl solution into a 50% HDEHP in toluene. According to the data given in Ref. 14, thorium should be extracted into the organic phase quantitatively ($K_d = 10^5$) while actinium should remain in the aqueous phase ($K_d = 10^{-6}$). (The actual decontamination factor of thorium from actinium under our experimental conditions — that is, to perform the separation within the shortest possible time, was found to be at least several orders of magnitude worse than the literature value. This point will be discussed more in detail later.) The desired thorium sample was extracted into HDEHP again several minutes after the initial separation to allow ^{233}Ac to decay into ^{233}Th and then back-extracted with 6 M HCl and 0.5 M HF solution.

The purification of thorium began with the retention of thorium on an MP1 anion exchange column from 10 M HNO_3 solution, conditions under which the bulk of uranium and some fission products would be eluted. Then thorium was eluted with conc. HCl; extracted into 0.2 M TTA in benzene from 0.01 M HCl solution; washed four times with 0.01 M HCl containing ytterium carrier and $NH_2OH{\cdot}HCl$ and back-extracted with 1 M HCl. After another pass through an MP1 anion exchange column in conc. HCl solution, thorium was finally precipitated as oxalate from strong HCl solution for radioactivity assay. The entire purification procedure usually took about 40 minutes.

Counting

The samples were counted either on a calibrated Ge(Li) detector (50 cm^3 in size with FWHM of 2.0 keV at 1332 keV) or on a standard gas-flow proportional counter for γ- or β-measurements, respectively. The γ-measurements were made only on those samples having a sufficiently high level of γ-activity. This included mainly the samples prepared for measuring cross sections of ^{233}Th and ^{231}Th directly produced in the irradiation. The thorium samples prepared from the growth of actinium were usually too weak to be counted on the Ge(Li) detector and were therefore only counted on the gas-flow proportional counter. The γ-ray spectra were recorded on magnetic tapes using a 4096 channel analyzer system and were analyzed later with the INTRAL code.[15] The least squares code CLSQ[16] was used for decay curve analyses.

Results and Discussion

Production Cross Sections for ^{231}Th and ^{233}Th

Production cross sections for ^{231}Th and ^{233}Th at three different ^3He energies were measured in this investigation. Uranium oxide of two different thicknesses and 0.0015-in. thick uranium foil were used in these measurements. Range data for heavy ions[17] were used to estimate the energy degradation through the target and the mean energy of ^3He through the target was calculated and used as the bombarding energy. Table 1 lists the relevant nuclear properties of ^{231}Th and ^{233}Th; these were used together with the beam current and the target thickness to calculate the production cross sections.

Table 1. Some Relevant Properties of ^{231}Th and ^{233}Th

Nuclide	Half-Life	E_γ (keV)	Abundance*
^{231}Th	25.5 h	84.5	6.5
		163.0	0.16
^{233}Th	22.2m	162.0	0.31
		169.0	0.34
		459.4	1.35
		670.1	0.65

* Number of photons per 100 disintegrations.

The results are listed in Table 2 with the mean energy for helium-3 through the target given in the first column. The errors of the cross sections

Table 2. Production Cross Sections of ^{231}Th and ^{233}Th

E_{He-3}	σ_n(μb)	
(MeV)	$^{238}U(^3He,2\alpha)^{233}Th$	$^{238}U(^3He,2\alpha 2n)^{231}Th$
46.3	1.3 ± 0.2	2.8 ± 0.3
53.0	3.8 ± 0.5	10.6 ± 1.1
59.6	6.3 ± 0.6	16.2 ± 2.0

reflect the larger quantity of either the combination of counting statistics, detector efficiency and chemical yield uncertainties or the standard deviation of the individual measurements from the mean. The excitation functions for ^{231}Th and ^{233}Th are shown in Fig. 1.

The Attempt to observe ^{233}Ac by assaying the Thorium Samples into which the Actinium has decayed

We have performed four separate experiments to search for the the decay of ^{233}Ac into ^{233}Th. All these experiments followed the same procedure: first thorium was separated from actinium by HDEHP solvent extraction; after allowing actinium to decay into thorium, a second thorium sample was extracted; following chemical purification, a sample was prepared for β counting. In one of the experiments, two extractions were performed as the initial separation. The time elapsed from the end of bombardment to the separation of phases in the solvent extraction was between 3.6 min and 4.2 min for one extraction and 5.5 min for two extractions.

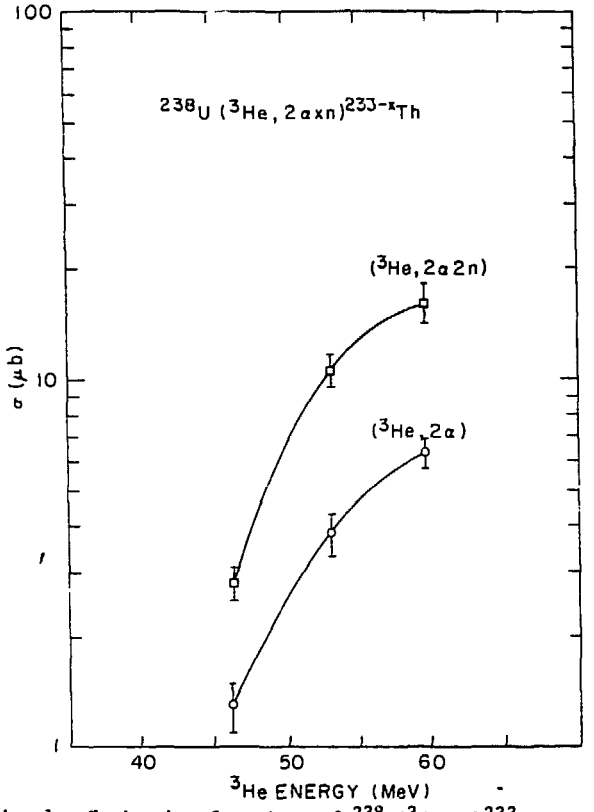

Fig. 1. Excitation functions of $^{238}U(^{3}He,2\alpha)^{233}Th$ and $^{238}U(^{3}He,2\alpha 2n)^{231}Th$ reactions measured in this work.

Because of the low activity of the samples, they were assayed on the gas-flow proportional counter. As it turned out, there was the additional problem of the background from thorium carrier in β counting. Since in the thorium samples, ^{228}Th always exists together with ^{232}Th, when a thorium sample is purified, the descendents of ^{228}Th begin to grow in, thus contributing to a gradually increasing background observed in the β counting. We have performed carrier-free chemical separations for thorium using ^{234}Th tracer and separations with small amounts of thorium carrier (< 0.5 mg), and have always found very low chemical yields. Therefore for the purification steps in these four experiments, we have used 2-3 mg of thorium carrier each time. In order to simulate the background in the β counting due to the decay of ^{228}Th and descendents, we have prepared a blank thorium sample following exactly the same chemical procedure as the real samples. The sample was followed on a proportional counter over a long period of time to establish a standard shape for background subtraction.

The counting results from each sample were analyzed first by subtracting the normalized background contribution from ^{228}Th and descendants. In all samples there is unmistakably a 22.2 min. component in the remaining counting data.

In order to compare the results from these four experiments, one must assume certain values for the half-life of ^{233}Ac. We have listed in Table 3 the calculated apparent cross sections for ^{233}Ac based on the observed ^{233}Th activity and the different values assumed for the half-life of ^{233}Ac. Note that Run 311 was the only experiment that had two solvent extractions for the initial thorium-actinium separation. Column 2 lists the time of separation of phases of the initial solvent extraction after the end-of-bombardment. The next seven columns list the calculated apparent cross sections in μb for ^{233}Ac based on counting rates, counter efficiency, time of initial separation, chemical yields and that particular assumed half-life for ^{233}Ac at the head of that column. We have used values between 60 s and 180s with 20 s increments. The last column lists the cross sections calculated using the value of the half-life of ^{233}Th, 22.2 min.

In addition to the expected decrease in calculated cross section with increasing value of the assumed half-life for ^{233}Ac within each experiment, another trend emerges from the data listed in Table 3. That is, the results of the last three experiments converge to each other as the assumed half-life for ^{233}Ac increases. The result from the first experiment remains very different from the other three regardless what is the assumed half-life for ^{233}Ac. This inconsistency may suggest serious contamination occurred in the first experiment. Since two solvent extractions were performed as the initial separation in the second experiment and only one was used in the last two experiments, the convergence of the data for these three experiments at longer assumed half-lives for ^{233}Ac seems to be significant.

However, the magnitude of 1-2 μb as the production cross section for ^{233}Ac seems to be surprisingly high in view of the estimate given in Ref. 11. Even taking the generous assumption that the same estimate applies here (using the excitation

Table 3. Calculated apparent cross sections for ^{233}Ac based on the observed ^{233}Th activities and different values assumed for the half-life of ^{233}Ac

		σ (μb)							
Run	T_s [a]	$t_{1/2}$=60s	80s	100s	120s	140s	160s	180s	22.2m
310	4.2 min.	93.4	35.0	18.9	12.4	9.1	7.2	6.0	1.5
311	5.5 min.	28.4	8.6	4.1	2.5	1.7	1.3	1.1	0.23
313	3.6 min.	16.8	6.8	3.8	2.5	1.8	1.4	1.2	0.26
314	3.9 min.	11.6	4.6	2.6	1.8	1.3	1.1	0.9	0.26

[a] T_s is the time of separation of phases of the initial solvent extraction for the thorium-actinium separation from the end of bombardment. Only one solvent extraction for initial separation was performed in all runs except 311, where two successive solvent extractions were performed as initial separation, and T_s is the time for the second extraction.

function results for (^{3}He,2α) reaction listed in Table 2, a five-fold decrease in cross section is expected when the projectile energy decreases by 13 MeV; furthermore, the estimate given in Ref. 11 was for medium elements and not for heavy elements), the cross section for ^{238}U(^{3}He,^{8}B)^{233}Ac is only expected to be less than 100 nb.

Therefore it is important to address the question of contamination more in detail. Although the data given in Ref. 14 suggest a decontamination factor of 10^{11} between thorium and actinium, under actual experimental conditions, trying to separate the phases immediately after the solvent extraction may lead to a decontamination factor many orders of magnitude less than this factor. In a separate experiment, we have used about 100 mg of thorium and performed the initial thorium-actinium separation under similar conditions and found about 0.01% of thorium left behind in the aqueous phase after one solvent extraction. Therefore one can conclude based on these observations that in the first experiment the unusually high cross section probably resulted from the incomplete extraction of thorium. Furthermore, the general agreement of the other three experiments under rather different experimental conditions (the length of bombardment, the beam current, the number of initial separations, and the time of the initial separation) seems to suggest that these data are consistent with the half-life of ^{233}Ac somewhat longer than 120 s and with the production cross section from this target-projectile combination to be of the order of 1-2 μb. This means the contributions from other equivalent reaction channels must be much higher than that of the (^{3}He,^{8}B) channel.

It does not seem feasible for us to improve these measurements at the Brookhaven cyclotron with the available energy and beam current of ^{3}He ions. However, we are planning to continue this investigation at the AGS (Alternating Gradient Synchrotron), using 28-GeV protons instead of 62-MeV ^{3}He ions. Based on the results of Pate and Poskanzer[18] and some recent exploratory measurements[19], we are hopeful that we can confirm the tentative conclusion arrived at in this investigation.

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