Nuclear Properties of 93^{237} *

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The isotope Np^{237}, produced as the decay product of the previously known 6.8-day U^{237}, has been isolated with carrier material and some of its nuclear properties measured. The U^{237} was formed by means of the reaction $U^{238}(n, 2n)U^{237}$. Np^{237} decays with the emission of alpha-particles with a half-life of about 3×10^6 years. It has been shown that the upper limit for the cross section for the fission of Np^{237} with slow neutrons is less than 1 percent of that of U^{235}.

THE bombardment of uranium with fast neutrons produces[1,2] the beta-emitting 6.8-day U^{237}, formed in the reaction $U^{238}(n,2n)U^{237}$. We have investigated 93^{237}, the daughter of U^{237}, the primary purpose of our investigation being to measure the fission properties of this isotope. Our measurements, performed upon a sample weighing about 0.6 microgram, show that 93^{237} does not undergo appreciable fission with slow neutrons. The experiments also show that 93^{237} is an alpha-emitting isotope with a half-life of about 3×10^6 years.

For the preparation of the sample of 93^{237} about 1200 grams of uranyl nitrate hexahydrate, $UO_2(NO_3)_2\cdot6H_2O$, were placed directly behind the beryllium target of the 60-inch Berkeley cyclotron and exposed to the fast neutrons produced in a 15,000-microampere hour bombardment of the beryllium with deuterons. After the lapse of several months, so that substantially all of the U^{237} had decayed to 93^{237}, a chemical separation was performed on this bombarded uranyl nitrate in order to isolate the 93^{237} into a thin rare earth fluoride precipitate (approximately 0.3 mg/cm^2), special care being exercised to remove all of the 94, uranium, protactinium, and thorium. This procedure has been described in principle in another report.[3]

The chemical procedure, in outline, is as follows. The uranyl nitrate hexahydrate is subjected to an ether extraction process in order to remove the bulk of the uranium, and rare earth fluoride is precipitated from the water phase. This rare earth fluoride contains the 94, 93, and thorium, the uranium and the protactinium (when zirconium carrier is present) remaining in solution. This rare earth fluoride is dissolved in sulfuric acid, and from a cold potassium bromate ($KBrO_3$) solution the rare earth fluoride is again precipitated. In this step the 94, in its reduced form, and the thorium and the rare earth fission products are precipitated as insoluble fluorides, and the 93 remains in solution in its oxidized form. After reduction with sulfur dioxide, the 93 in the solution is precipitated with a small amount of rare earth fluoride. Of course, it is necessary that a number of cycles be performed in order to remove completely from the final sample the 94, uranium, protactinium, and thorium, and in our procedure there were three separations from 94, six separations from uranium, five separations from protactinium, and five separations from thorium. The last 94-containing rare earth precipitate was mounted for alpha-counting and was found to contain practically no 94 (<30 counts per minute) while the final sample containing the 93^{237} had a counting rate of about 300 alphas per minute. In order to keep track of the yield of 93 through this rather lengthy chemical procedure there was added at the beginning some 2.3-day 93^{239} as tracer for the 93, and it was found that the yield of 93 in the final sample amounted to about 50 percent of that originally added to the bombarded uranium nitrate.

In order to prove that in this chemical pro-

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[1] E. M. McMillan, Phys. Rev. **58**, 178 (1940).

[2] Y. Nishina, T. Yasaki, H. Ezoe, K. Kimura, and M. Ikawa, Phys. Rev. **57**, 1182 (1940).

[3] G. T. Seaborg and A. C. Wahl, J. Am. Chem. Soc. (March, 1948).

cedure no uranium, protactinium, or thorium remained in the final sample, a chemical separation, of a similar type, was performed on non-irradiated uranyl nitrate using U^{233} as tracer for uranium, Pa^{233} as tracer for protactinium, and UX_1 as tracer for thorium. This "blank experiment" proved that in our chemical procedure there remains in the final sample much less than a microgram of each of these elements.

In order to know how much 93^{237} was formed in the bombardment described above, it was necessary, of course, to determine the yield of U^{237} in the bombardment of uranyl nitrate hexahydrate under similar conditions. A sample of 700 grams of $UO_2(NO_3)_2 \cdot 6H_2O$ was placed directly behind the beryllium target and subjected to a 500-microampere hour bombardment. This sample was carefully purified from all of the fission products and from element 93 by the ether extraction method; the ether phase, containing the bulk of the uranyl nitrate and the U^{237}, was washed 8 or 10 times with water in order to insure a very thorough purification. Aliquot amounts, determined by weighing the uranyl nitrate containing the 6.8-day U^{237}, were taken from the ether phase and the intensity of the activities was measured with a Lauritsen quartz fiber electroscope calibrated in an absolute manner for the radiation from the 6.8-day U^{237}. This calibration was accomplished by preparing thin electrolytically deposited aliquots of the uranium and counting the beta-particles from the 6.8-day U^{237} with a thin-window Geiger-Müller counter of calibrated efficiency; the weight of uranium in these small aliquots was determined by counting its natural alpha-particles with an ionization chamber of calibrated efficiency. (Correction for the self-absorption in the samples and absorption in the Geiger-Müller counter window of the 6.8-day U^{237} beta-particles was made by using the value for the half thickness of the absorption of these beta-particles, which we have determined as 6 mg Al/cm^2.) From the measurements on the calibrated Lauritsen electroscope of the chemically purified aliquots from the 500-microampere hour bombardment, we found that the yield of 6.8-day U^{237} was 0.0054 microcurie per gram of

$$UO_2(NO_3)_2 \cdot 6H_2O$$

per microampere hour of deuterons on beryllium.

From this yield it follows that there was formed in the 15,000-microampere hour bombardment of 1200 grams of $UO_2(NO_3)_2 \cdot 6H_2O$ about 97 millicuries of U^{237}, which corresponds to about 1.2 microgram. Since substantially all of this decayed to 93^{237} before the chemical separation was performed and since the yield in the chemical procedure for the isolation of 93 was about 50 percent, as mentioned above, this means that there was present in our final sample about 0.6 microgram of 93^{237}. We consider this estimate of the amount to be accurate to within a factor of about 2. The alpha-counting rate of this sample, as mentioned above, was about 300 counts per minute, and, from this value, together with the calibrated efficiency (45 percent) of the ionization chamber, it is calculated that the half-life of alpha-emitting 93^{237} is about 3×10^6 years.

This sample was placed on one electrode of an ionization chamber connected to a linear amplifier and recording system which was adjusted so as to record the pulses due to fissions. As a neutron source we used 300 milligrams of radium mixed with beryllium powder. The ionization chamber and the 300-mg radium-beryllium neutron source were completely surrounded by paraffin. Under these conditions the 0.6 microgram 93^{237} sample gave four fission counts in about 56 hours of counting time. When the ionization chamber was surrounded by cadmium, there were recorded two counts in 56 hours of counting time. Thus, the counting rate due to slow neutrons was two counts in 56 hours or 0.035 fission counts per hour. For a calibration experiment the 0.6-microgram sample of 93^{237} was replaced by 200 micrograms of electrolytically deposited uranium, whose weight was determined by counting its alpha-particles with an ionization chamber of calibrated efficiency. The easily measurable fission rate of this uranium sample, containing 1.4 micrograms of U^{235}, when compared to that for 93^{237} showed that the slow neutron fission cross section of 93^{237} is less than one percent[4] of that for U^{235}.

[4] Note added at time of publication. More recent work by A. Ghiorso, D. W. Osborne, and L. B. Magnusson has demonstrated that Np^{237} undergoes slow neutron fission with a cross section a few thousandths of a percent of that of U^{235}. (Science **104**, 386 (1946).)