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THERMAL NEUTRON FISSION IN Pa^{231}, Th^{232}, U^{234}, and Np^{237}

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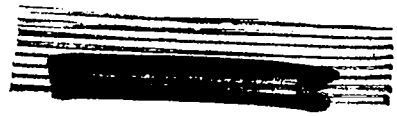
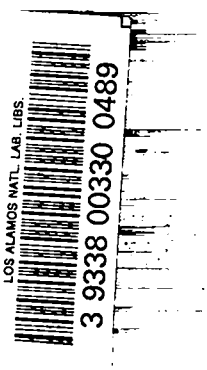


ABSTRACT

Samples of Pa^{231}, Th^{232}, U^{234}, and Np^{237} show apparent slow-neutron fission cross sections of 0.09×10^{-24}, 0.0007×10^{-24}, 1.9×10^{-24}, and 0.10×10^{-24} cm^2, respectively. These cross sections are regarded as upper limits, as the observed slow-neutron fission effects could be due to impurities.



THERMAL NEUTRON FISSION IN Pa^{231}, Th^{232}, U^{234}, and Np^{237}.

In order to check samples of Pa^{231}, Th^{232}, U^{234} and Np^{237} for possible plutonium or normal uranium contamination which might have an effect on the spontaneous fission rate observed in the samples, it was decided to look for fissions in the samples when they were placed in a very strong thermal-neutron flux. The observed thermal-neutron fission rates made it possible to place upper limits on the thermal-neutron fission cross sections of the substances studied.

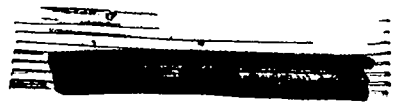
Each sample was prepared as a thin film on platinum and compared with a thin foil of normal uranium containing 3.4 micrograms of U^{235}. The comparison chamber was an argon-filled flat cylindrical steel chamber in which the samples were mounted back to back on a central electrode at a potential of about -500 volts. Collecting electrodes opposite each sample and about 1 cm distant from the central electrode were connected to twin linear amplifiers and counting circuits. The chamber was placed in the graphite column of the water boiler at Omega at a point at which the cadmium ratio for the normal uranium sample was several thousand. With the boiler operating at about 1 KW, fission counting rates of 15,000 to 20,000 counts per minute were observed for the normal uranium sample. In every comparison, cadmium shielding was found to eliminate the fissions observed in the substance being compared with uranium.

Pa^{231}:

A sample (1) of 490 micrograms was masked so that about 25 micrograms were exposed. This was necessary in order to reduce the alpha activity background in the chamber. Observed counting rates were 15,500 c/m for 3.4 micrograms of 25 against about 19 c/m for 25 micrograms of Pa^{231}.

Assuming a thermal-neutron cross section of 542 x 10^{-24} cm^2 for 25, the apparent thermal-neutron cross section of Pa^{231} is from these data 0.092 x 10^{-24} cm^2. This is considered as an upper limit, since about 0.6 micrograms of normal uranium contamination could produce the observed thermal neutron fission effect.

(1) The material used was obtained from Dr. Agruss.



Th^{232}:

A sample of 2770 micrograms of Th^{232} showed an average counting rate (for two runs) of about 15 c/m against an average counting rate of 15,000 c/m for 3.4 micrograms of 25, giving an apparent thermal neutron cross section of 0.00068 x 10^{-24} cm^2 for Th^{232}. Again, this is an upper limit, since 0.5 micrograms of normal uranium contamination could produce the observed effect.

U^{234}:

The U^{234} sample of about 10 micrograms effective weight was obtained on loan from Dr. Latimer in Berkeley. The material was prepared by extraction of UX_1 from uranium and subsequent β - decay of this substance.

The U^{234} sample showed a counting rate of 170 ± 15 c/m against 16,800 c/m for 3.4 micrograms of 25, corresponding to a slow neutron fission cross section of 1.9 x 10^{-24} cm^2 for U^{234}. It is considered unlikely that 5 micrograms of normal uranium contamination, which would be necessary to produce the observed fission rate, are present in the sample.

Linenberger[2] reports an apparent slow-neutron cross section of 2.1 x 10^{-24} cm^2 for U^{234}, based on slow neutron irradiation of the same sample, but assuming a weight of 13 micrograms. A figure of 10 micrograms for the effective weight of the sample is probably more accurate; this would bring his reported cross section to 2.7 x 10^{-24} cm^2. Considering the rather large probable error due to counting statistic present in this measurement, and the appreciable but somewhat smaller similar uncertainty in the present measurement, the agreement is reasonable.

Np^{237}:

The first fission cross section run was made with a sample[3] of 1040 micrograms of Np^{237}. This sample showed a fission counting rate of 950 ± 50 c/m against 15,100

(2) LA 140 A

[illegible]stigated were supplied by the Metallurgical Laboratory in Chicago.

c/m for 3.4 micrograms of 25. Alpha range analysis of the sample, using a variable pressure differential range chamber, showed that 0.2 c/c of the alpha activity was due to 49, so that 0.002 c/c of the sample (by weight) was 49. The 49 contaminant accounted for about 125 c/m of the observed 950 c/m. The resulting slow neutron fission cross section for 37 is 0.096×10^{-24} cm^2.

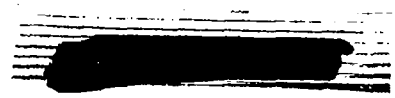
A second sample of about the same weight showed an apparent cross section of about 0.4×10^{-24} cm^2. No range analysis was made on this sample.

It was decided to purify the first sample for uranium and plutonium in order to establish if possible whether the thermal neutron fission effect was real. A description of the purification procedure follows:

While it is very probable that the chemical procedures[4] usually employed in the separation of the elements uranium, neptunium, and plutonium would afford a good purification of neptunium with respect to uranium and plutonium, it was considered very desirable to have a positive proof of the extent to which the uranium and plutonium had been removed from the neptunium. For this purpose Pu^{238} and U^{232} were used as tracers for plutonium and uranium, respectively, using alpha range apparatus to follow the individual activities.

A solution 1 ml in volume and 1 N in HNO_3, containing approximately 0.6 mg of Np^{237} in the 4^+ state, 184,000 alpha disintegrations per minute of U^{232} in the 6^+ state, 735,000 alpha disintegrations per minute of Pu^{238} in the 4^+ state, and approximately 0.1 mg of La^{+++} was prepared. The La^{+++} and Np^{4+} were precipitated with H F, carrying with them the plutonium. The precipitate was washed with a 1N H F - HNO_3 mixture and transferred to a platinum dish, where it was fumed with H Cl O_4. The residue was taken up in a 1 N H_2SO_4 solution containing SO_2. The SO_2 was boiled off and the Np^{4+} was oxidized to Np^{6+} by adding enough K Br O_3 to make the

(4) See Wahl and Seaborg, A 135



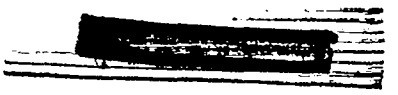
final solution 0.1 M in K Br O_3. The oxidation(2) was carried out over a period of ten minutes at room temperature. The LaF_3 was then precipitated with H F, carrying with it the plutonium. The supernatant containing the Np^{237} and K Br O_3 was evaporated to dryness and reduced with an H_2 SO_3 solution. The residue was taken up in 1 N HNO_3 and 3 mg of Fe^{+++} were added. The Fe^{+++} was precipitated with $N H_4$ O H, carrying with it practically all of the Np. The Np was then separated from the Fe by an ether extraction of an approximately 6 N HCl solution of the Fe and Np^{4+}.

The final Np plate was prepared by D. Hufford of Group CMR-4 by the zapon technique. It was found to contain only about 25 micrograms of Np^{237}. Comparison with the uranium sample showed an apparent slow neutron fission cross section of about 0.27×10^{-24} cm^2 for the Np^{237}, which is higher than the apparent cross section as observed before purification.

Alpha range analysis of the final sample showed that more than 1/3 of the initial percentage of Pu^{238} and hence of the initial plutonium percentage remained after the purification. The U^{232} alpha activity, if any, was masked by the Pu^{238} activity; it is only possible to say that the uranium purification factor was at least 4 and probably many times better.

The higher apparent cross section for the purified sample is probably due to fissionable contamination introduced at some stage in the purification or final plating. The U^{232} was checked for thermal fission before being added to the Np^{237}, and the data obtained show that its contribution to the observed fission in the Np^{237} sample cannot be greater than about 10 o/o, and is probably much less. Data on therm neutron irradiation of the Pu^{238} (5) indicate that it should be responsible for about 10 o/o of the observed effect, and a correction for this was made. Of course, the possibility of 49 contamination of the Pu^{238} subsequent to this irradiation exist

It would be of considerable interest to perform a more thorough purification on a larger amount of Np^{237} and to make a better determination of its thermal-neutron



fission cross section. As suggested by Segre[6], this datum, together with data on the photo-fission threshold and the thermal-neutron capture cross section, would make possible an approximate calculation of the transparency (and ultimately of the width) of the fission barrier at a known distance of a few hundred Kev below the top of the barrier. A similar calculation is possible for Pa^{231}.

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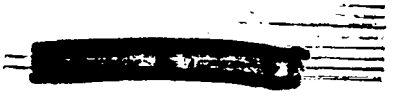
(5) Anderson and Sugarman, LA 356.

(6) Segre, Chapter V, Los Alamos Technical Series.



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