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SEARCH FOR SLOW-NEUTRON-INDUCED FISSION OF N_p^{239}

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ABSTRACT

A sample of about 5×10^{-7} gm of Np^{239} was prepared by slow-neutron exposure of uranium depleted of U^{235}. An apparent cross section of Np^{239} for slow-neutron fission of $(14 \pm 4) \times 10^{-24}$ cm^2 was observed. Because of the possibility that normal (undepleted) uranium may have been introduced as an impurity during manipulation of the sample this value is interpreted as indicating only an upper limit of $\sigma_f(Np^{239}) < 20 \times 10^{-24}$ cm^2.

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SEARCH FOR SLOW-NEUTRON-INDUCED FISSION OF Np^{239}

The question whether fission of Np^{239} can be induced by slow neutrons seemed worth answering not only to round out our knowledge of transuranic nuclei but also because a very large cross section for this process might conceivably be important in future very-high-intensity chain-reacting installations. In analogy with Pa^{231}, Np^{239} should not show slow-neutron fission. Also, since Np^{237} has an (n,f) threshold of about 400 Kev, the heavier Np^{239} should certainly require fairly-high-energy neutrons.

Np^{239} was prepared by slow-neutron irradiation of U^{238} and the subsequent beta decay of U^{239}. Uranium impoverished in the light isotopes was used. This has the advantage that the sample is less radioactive after bombardment, making manipulation less cumbersome. Furthermore the danger of U^{235} contamination of the neptunium is correspondingly reduced. The ratio of the isotopes 238:235 in our sample was 3500. About 200 gm of UO_3 prepared of this material was exposed in the BeO tamper of the Los Alamos "waterboiler" for three days of intermittent operation.

The chemical operation procedure was carried out with a view to obtain neptunium free of uranium and plutonium. The UO_3 was dissolved in concentrated nitric acid, a 3 percent sulphurous acid was added to reduce the neptunium, until the solution was about 2M in HNO_3 and contained about 2 gm/liter of uranium. 50 mg of lanthanum was added. The solution was made 2M in HF, precipitating LaF_3 and the neptunium in the +3 state. The fluoride precipitate was dissolved by

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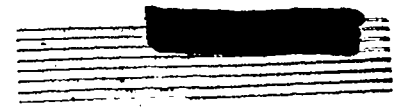
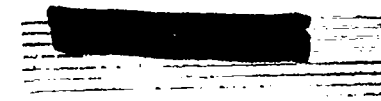


fuming with sulphuric acid. The neptunium was then oxidized quantitatively by treating with 0.3M $KBrO_3$ in 2M H_2SO_4 for half hour. This does not oxidize the plutonium which was then precipitated with the lanthanum carrier by adding HF. The supernatant, containing the neptunium was taken to dryness and fumed with sulphuric acid. This cycle of fluoride precipitation with alternate oxidation and reduction was repeated three times. The last precipitation of the (reduced) neptunium fluoride was carried out with only 0.25 mg of lanthanum carrier. Before this last precipitation a small aliquot (about 6 percent of the total) was taken out. The fluoride precipitate was slurried and deposited on a platinum foil. This was mounted in a small parallel-plate ionization chamber for fission counting. A fraction of the aliquot, 0.5 percent of the total neptunium, was deposited on another platinum foil which was mounted in a second parallel-plate ionization chamber for alpha counting.

Both chambers were filled with a mixture of argon and 5 percent CO_2. The gas in the chamber used for alpha-ray counting was continuously purified by circulating over hot calcium metal. Identical "fast" feedback linear amplifiers were used with the two chambers. The rise time (high-frequency cutoff) of the amplifiers was about 0.1 microseconds and the decay time of the pulses was about 0.3 microseconds. This short resolving time was required to avoid spurious pulses due to accidental "piling up" of a large number of neptunium beta rays. The number of beta particles in the alpha chamber was about 2×10 per second and in the fission chamber about 4×10^9 per second. The absence of pile-up pulses in the alpha counting chamber was proved by placing a beta ray source of similar strength into it. The fission chamber was placed in the graphite column of the



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waterboiler and exposed to a neutron flux of about 10^9 neutrons/cm^2 sec. Counting of both samples started less than four hours after the last separation from plutonium. No pulses were observed. In the case of the fission counting chamber no pulses were observed in the absence of slow neutrons, as was demonstrated by lowering the cadmium curtain in the graphite column.

Fig. 1 shows the counting rates of the alpha ray and fission counters as functions of the time after the last separation from plutonium. The counting rate of the alpha-ray sample is represented very well by the equation

$$n_\alpha = (2.4 \pm 0.7) + (230 \pm 3)\ (1 - e^{-\lambda t})\ \text{counts/min}$$

and that of the fission counter

$$n_f = (510 \pm 50) + (16600 \pm 200)\ (1 - e^{-\lambda t})\ \text{counts/min}$$

where λ is the decay constant of Np^{239}, taken to be 0.0258 hr^{-1} and t is in hours. The background counts of 2.5 ± 0.2 min^{-1} in the alpha counter and 170 ± 10 in the fission counter were subtracted in Fig. 1 and in the above equations.

It may be safely assumed that all of the observed alpha activity was due to plutonium either growing from neptunium or present initially because of incomplete separation. The activity of all reasonable contaminants is probably negligible. Thus we find that initially the ratio of the number of plutonium atoms N_{49} to that of neptunium atoms N_{39} was

$$N_{49}/N_{39} = 2.4/230 = (1.04 \pm 0.3) \times 10^{-2}$$

In the case of the fission count the situation is more complicated. Besides the plutonium, possible fission of neptunium and contamination by U^{235} may contribute to the observed activity. We denote the number of U^{235} atoms by N_{25} and the several cross sections for thermal neutrons by σ with appropriate indices. Then

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we have, neglecting square terms in small ratios:

$$(N_{49}\sigma_{49} + N_{39}\sigma_{39} + N_{25}\sigma_{25})/N_{39}\sigma_{49} = 510/16600 = (3.07 \pm 0.3) \times 10^{-2}$$

The first term on the left of this equation was determined by the alpha count as shown above. In order to determine uranium contamination, the main sample was removed from the platinum foil and a fluorimetric determination of uranium was made by the analytical group of the chemistry division. Complete removal of the sample was checked both by alpha count and fission count. The result indicated that there was $(4 \pm 1) \times 10^{-6}$ gm of uranium present. If all of this was due to incomplete separation in our chemical procedure, i.e., if it was depleted material, then it corresponded to $(1.15 \pm 0.25) \times 10^{-9}$ gm of U^{235}. On the other hand the final alpha activity of the sample indicated that there was initially about 5.8×10^{-7} gm of Np^{239} present. Taking into account the different fission cross sections of uranium and plutonium, we have

$$N_{25}\sigma_{25}/N_{39}\sigma_{49} = (542/765)(1.15/5.8) \times 10^{-2} = (1.4 \pm 0.5) \times 10^{-3}$$

Combining this with the fission activity due to the plutonium initially present, we find

$$\sigma_{39}/\sigma_{49} = (3.07 - 1.04 - 0.14) \times 10^{-2} = (1.9 \pm 0.5) \times 10^{-2}$$

or, using the fission cross section of Pu^{239}, 765×10^{24} cm^2

$$\sigma_{39} = (14.5 \pm 4) \times 10^{-24}\ cm^2$$

However it is quite possible that some or all of the observed uranium contamination was introduced into the sample during manipulation and was therefore not depleted of U^{235}. In this case the uranium contamination alone could easily account for all of the observed fission activity. Thus we are inclined to interpret our result-

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as an upper limit, indicating for Np^{239} a fission cross section

$$\sigma_f(Np^{239}) < 20 \times 10^{-24} \text{ cm}^2$$

The departure of both authors from the project has prevented a repetition of the experiment at this time.

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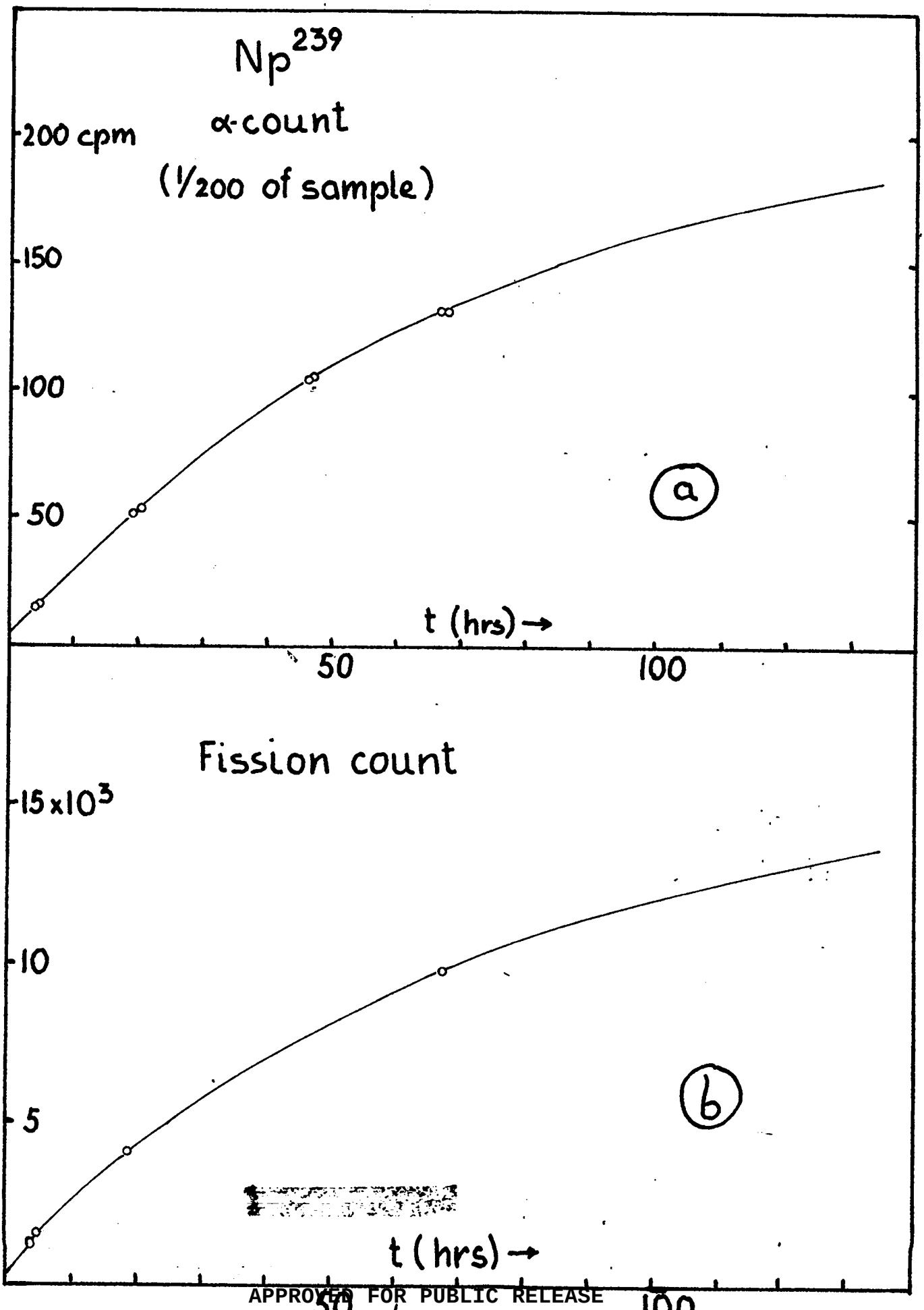

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