229. Neutron Irradiation of $^{237}\text{Np}$

$^{237}\text{Np}$ の中性子照射

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Irradiation of $^{237}\text{Np}$ target in JRR-1 and subsequent activity measurement clarified that $^{238}\text{Np}$ was formed as a consequence of neutron capture. The identification was carried out by tracing the decay and by $\beta$- and $\gamma$-spectrometry. Gamma-rays due to fission products did not interfere with the above measurements. $^{238}\text{Np}$ decays into $^{238}\text{Pu}$, and therefore, $^{238}\text{Pu}$ was isolated chemically from the target material after suitable cooling time, and its presence was confirmed by $\alpha$-spectrometry.

INTRODUCTION

$^{237}\text{Np}$, an $\alpha$ emitter, is important for studying neptunium in macro amounts, possessing as it does the longest half life among the twelve known isotopes of neptunium. Neutron capture of $^{237}\text{Np}$ gives $^{238}\text{Np}$, which disintegrates into $^{238}\text{Pu}$. So these two nuclides can be expected to be existing in the irradiated sample.

EXPERIMENTAL METHOD AND RESULTS

$^{237}\text{Np}$ was imported from AERE, Harwell, UK, in the form of 1N nitric acid solution presenting the green color of quinquivalent neptunium. Platinum plate, on which an aliquot of this solution containing 0.05 mg of $^{237}\text{Np}$ was plated, was used as the target material.

Irradiation was carried out in JRR-1 for 15 hr with a neutron flux of $10^{13}$n/cm$^2$/sec. The irradiated sample was taken out into 2 ml of 1N nitric acid from the platinum plate. 0.05 ml of the obtained solution was submitted to measurement of decay curve, and to $\beta$- and $\gamma$-spectrometry.

Beta decay was followed for 36 days by gas flow counter, as is represented in Fig. 1. The small activity remaining after 30 days may be attributed to minute amounts of fission products or impurities. Subtraction of this activity from the measured decay curve gave a straight line representing a half life of 2.10 days. This value agreed very well with those for $^{238}\text{Np}$ published previously by other investigators\(^{(1)(2)}\). The amount of $^{238}\text{Np}$ was determined from extrapolation of the obtained decay curve to correspond to $3.6 \times 10^5$ cpm at the moment of irradiation completion, and the $\alpha$ activity of the sample corresponded to $5.0 \times 10^{-4}$ mg of $^{237}\text{Np}$. It can therefore be concluded that 160 $\mu$g of $^{238}\text{Np}$ was produced from 0.05 mg of $^{237}\text{Np}$.

Beta-spectrometry was carried out by the use of a scintillation spectrometer. A Kurie plot of the result obtained is shown in Fig. 2, where the maximum $\beta$ energy originating from the sample was determined to be 1.20 MeV. A value of 1.24 MeV has been previously reported\(^{(1)(2)}\), which coincides well with the present value. This serves to confirm the existence of $^{238}\text{Np}$ in the irradiated $^{237}\text{Np}$.

Gamma spectrum measurement was performed by means of a TMC 256 channel pulse height analyzer, as is given in Fig. 3. Observed $\gamma$ peaks at 45 keV and 1.00 MeV were assigned to the intrinsic $\gamma$ rays from $^{238}\text{Np}$ and that at 75 keV to X-rays\(^{(1)(2)}\). The $\gamma$ spectrum obtained did not give any $\gamma$ peak.

of nuclides releasing γ rays over 1.00 MeV, or in fact, of any fission product at all. Upon the elapse of 30 days after irradiation, plutonium was separated by the isolating method illustrated in Fig. 4, upon considering that $^{239}\text{Pu}$ was expected to be contained in the sample as the β decay product of $^{238}\text{Np}$.

The separation method was the same in principle as that used in the previous study\(^6\). After converting to chlorides, neptunium and plutonium were reduced to quadri- and tri-valent states respectively with 0.1 M hydroquinone and 0.1 M potassium iodide in 12 N hydrochloric acid solution. Only Np($IV$) was extracted into 50 % tri-n-butyl phosphate toluene solution in this condition.

A small portion of the organic solution obtained was examined by α-spectrometry, which proved that $^{237}\text{Np}$ was pure, since only the α peak of 4.78 MeV was observed, as can be seen in Fig. 5. All of the aqueous
solution obtained gave an $\alpha$ activity of 156 cpm with the use of a 2 pi gas flow counter. Alpha peaks of 5.52, 5.15 and 4.87 MeV were detected in the $\alpha$ spectrogram for the aqueous fraction, as is also seen in Fig. 5. Peaks at 5.52 and 5.15 MeV were presumed to arise from $^{238}\text{Pu}$ and $^{239}\text{Pu}$ respectively. The latter was supposed to come from the original solution of $^{237}\text{Np}$. A nuclide having an $\alpha$ energy of 4.87 MeV could not be assigned. This fact showed that $^{238}\text{Pu}$ was contained in the neutron irradiated $^{237}\text{Np}$.

It is concluded that $^{238}\text{Np}$ was produced upon neutron irradiation of $^{237}\text{Np}$ in JRR-1, and $^{238}\text{Pu}$ decaying from $^{238}\text{Np}$ was detected.

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