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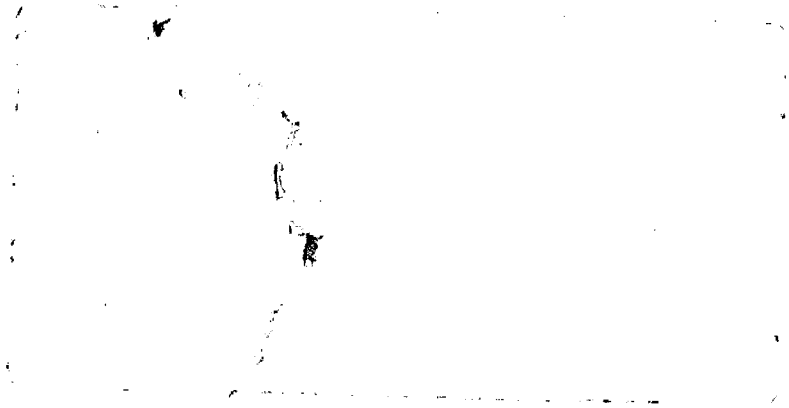

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ABSTRACT

The 1-hour neptunium activity which had previously been assigned to Np^{241} has been identified as the lower isomer of Np^{240}. The decay energy of the 1-hour Np^{240} has been found to be 2.05 Mev compared with 2.156 Mev for that of the 7.3-min Np^{240}. Gamma rays of energies 1160, 1000, 915, 595, 435, 245, 160, and 85 kev have been found to be associated with the decay of Np^{240}. The best value for the half-life of Np^{240} is 63 ± 2 min. The isotope Np^{241} has been found to have a 16-min half-life with strong evidence for an isomer with a 3.4-hour half-life.

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INTRODUCTION

The 7.3-min Np^{240} isomer was first identified by Hyde and Studier.[1] Later Orth and Street bombarded a U^{238} target with 35-Mev helium ions and found a 1-hour neptunium activity which they assigned to Np^{241}.[2] Further investigation by the present authors, using the time-of-flight isotope separator, has shown that this 1-hour activity is to be assigned the mass number 240.[3,4] In addition, Np^{241} has been found to have a half-life of 16 min and a probable isomer with a half-life of 3.4 hour.[3,4,5]

IDENTIFICATION OF 1-hr Np^{240}

The following experiments were performed to identify the mass number of the 1-hour neptunium activity. Two separate bombardments of 0.005-in. foil composed of natural uranium were made on the Crocker Laboratory 60-inch cyclotron, with 35-Mev helium ions. The neptunium was chemically separated from the target, primarily by use of a thenoyltrifluoroacetone (TTA) extraction from a 1.5 N hydrochloric acid solution and then a back-extraction into 8 N nitric acid. A time-of-flight mass separation of the neptunium fraction was made in each case. In the first bombardment only a mass 240 sample was collected and in the second bombardment, in which the time for chemical separation was reduced from 45 to 25 minutes, only a mass 241 sample was collected.

The first bombardment yielded decay periods of 1 hour and 8.7 hour at mass 240. The second bombardment gave periods of 16 min and 3.3 hour at mass 241. The time from end of bombardment until the plate was counted was 2.8 hour for the first bombardment and 1.8 hour for the second. Since the 1-hour

activity, which is known to be neptunium, was found only at mass 240, this half-life has been definitely assigned to Np^{240}. Thus the possibility previously proposed by Orth and Street that Np^{241} has a 1-hour half-life now seems quite remote.[2]

DECAY PROPERTIES OF Np^{240}

Measurements on the 1-hour neptunium activity by Orth and Street with a beta-ray spectrometer had shown a beta spectrum of upper energy limit 0.89 ± 0.03 Mev and conversion electrons corresponding to gamma rays of energies 150, 200, 260, and 580 kev.[2] Gamma-ray measurements performed in the course of this work on a NaI scintillation spectrometer showed gamma rays of energies 1160, 1000, 915, 580, and 435 kev, but additional gamma rays of less than 350 kev were obscured by the background due to the radiations from Np^{239}. By the use of gamma-gamma coincidence techniques, in collaboration with Dr. Frank S. Stephens, additional gamma rays of 245, 160, and 85 kev energy were found. The "580-kev gamma ray" was found to be complex, with components of about 595 and 565 kev in coincidence with each other. The most energetic beta-particle group had an end-point energy of about 900 kev and is in coincidence with 1160-kev gamma radiations. No other beta-ray was observed in coincidence with the gamma rays. Thus the decay energy is measured as 2.05 Mev (890 β + 1160 γ), which is consistent with the mass assignment, since from closed decay-energy cycles[6] one would estimate only 1.32 Mev for the decay energy of Np^{241}, whereas 2.05 Mev is available for the decay of Np^{240}. The 7.3-min Np^{240} has a decay energy of 2.156 Mev.[7] Therefore the 1-hour neptunium is the lower 240 isomer. The gamma-ray spectrum is consistent with this mass assignment, since gamma rays of 920 and 1020 kev are seen in the decay of Am^{240} to Pu^{240}, gamma rays of 557 and 600 kev are seen in the decay of the 7.3-min Np^{240}, and a gamma ray of 150 kev is seen in the alpha decay of Cm^{244} to Pu^{240}.[8] Within the limits of experimental error these gamma rays correspond respectively to the 915, 1000, 565, 595, and 150 kev gamma rays measured in the decay of 1-hour Np^{240}. Lefevre, Kinderman, and Van Tuyl[9] have since produced both Np^{240} isomers by bombarding Np^{239} with pile neutrons, and set an

upper limit of < 5% for the genetic lineage between the two Np^{240} isomers. In addition Vandenbosch et al.[10] have now determined the excitation function for the production of 1-hour neptunium from U^{238} bombarded with helium ions.

A close search of decay curves previously determined with chemically separated neptunium shows no evidence of any half-life between those of the 1-hour and 2-day neptunium isotopes. Careful resolution of these curves gave 63 ± 2 min as the best value for the half-life of Np^{240}.

IDENTIFICATION OF Np^{241}

The assignment to neptunium of the 16-min activity seen in the mass 241 fraction is considered fairly well established by this work. Both TTA extraction and anion-column separations had been used in previous bombardments and in each case the 16-min activity followed the neptunium fraction.[3] Vandenbosch has verified these experiments on the chemical identification of the 16-min activity as neptunium.[11] In addition he found the maximum beta-particle energy to be 1.36 Mev which is in good agreement with the 1.32-Mev value predicted by the closed decay-energy cycles.[6] This gives rise to a log ft value of 5.9, an allowed transition, if all the decay is to the ground state.

Another target composed of natural uranium was bombarded in the 60-inch cyclotron with 48-Mev helium ions, and a more careful chemical separation of the resulting neptunium was carried out in a period of about 1 hour. This included the separate precipitation of zirconium phosphate, lanthanum fluoride, and lanthanum hydroxide which co-precipitated the neptunium. Then the neptunium was extracted from 1 M hydrochloric solution into 0.4 M TTA in benzene, washed with 1 M hydrochloric acid and back-extracted into 8 M hydrochloric acid. The resulting neptunium fraction was mass-separated in the time-of-flight isotope separator with exclusive collection at the mass number 241 position. The radiations from this mass-separated sample were first counted 3.7 hour after the end of the bombardment, and showed a single decay period of about 3.5 hour. It was not possible to make observations on the radiations from the 16-min activity again because of the longer separation time. Since a clean chemical

separation was obtained in this experiment, it appears that the 3.3 to 3.5-hour activity is due to Np^{241}. One must be certain of the elemental assignment of any isotope whose mass number is to be determined in this way because of the possibility that an isotope of another element of considerably different mass but much higher ionization efficiency could be collected in low yield by the isotope separator.[12] Thus it is possible, but not probable, that the 3.4-hour activity collected at mass 241 could be a more easily ionized fission product, unseparated from neptunium by the chemistry. However, a search of the table of isotopes failed to reveal any fission products that could account for this 3.4-hour activity. The fact that this period is not found in the gross decay of the neptunium fraction is not difficult to explain, as this activity is masked by the more abundant radiations from the 1-hour Np^{240} and ~ 2-day Np^{238} and Np^{239}. This is also not inconsistent with the upper limit of 0.2 mb set by Vandenbosch[11] for the production of the 3.4-hour activity with 43-Mev helium ions. Another mass separation using additional steps in the chemical separation procedure is planned in order to be absolutely certain of this assignment.

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