SPONTANEOUS FISSION PERIODS OF Np^{237}, Pu^{238}, AND Pu^{242}

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An attempt has been made to determine the true spontaneous-fission half-period of Np^{237} by employing nuclear photographic emulsions. Prior to development, the photographic plates were treated with potassium ferrocyanide to remove the background α-particle tracks. The reliability of this method was checked by measuring the spontaneous-fission half-periods of Pu^{238} and Pu^{242}, which were independently determined using a proportional counter. The results obtained for plutonium by the various methods are identical and agree with other available data. Only three fragment tracks were detected in the Np^{237} photographic measurements. Thus, only a lower limit of 10^{18} years can be established for the half-period, as compared with the usually accepted value of 4×10^{16} years.

1. In the experiments of Segrè and his collaborators, the half-periods for spontaneous fission were already determined in 1945 — 1946 for a large number of both even and odd nuclei.[1] However, not all the data were obtained with the same accuracy. A check of the decay half-periods of Th^{232} (reference 2) and Am^{241} (reference 3) showed that their true values are higher by one to three orders of magnitude. The purpose of the present experiment was to achieve a higher accuracy in the measurement of the half-periods for the spontaneous fission of the odd-even isotope Np^{237} and the even-even isotopes Pu^{238} and Pu^{242}.

2. The investigated elements were coated electrolytically on a platinum support. The amount of the substance and the layer thickness were determined by counting the rate of α particles using a special arrangement with known geometry.[4] The uniformity of the samples was checked by counting α rays over separate parts of the layer. In addition, radiography of certain samples was carried out.

Seven samples were prepared, three of Np^{237} of ~ 20 mg each (density ~ 1 mg/cm^2), two of pure Pu^{238} (0.27 and 0.35 μg), and two consisting of a mixture of the isotopes Pu^{238} (14%) and Pu^{242} (86%), and containing 0.37 and 0.30 μg of Pu^{238} respectively. The isotope content of the mixture in the latter case was determined using a large magnetic α spectrometer.[5] In all calculations, the value of the partial half-periods of α decay for Pu^{238} was assumed to equal 86.4 years.[6]

3. The measurement of the spontaneous-fission half-period of the plutonium isotopes was carried out by two methods. In the first series of experiments, a cylindrical proportional counter 100 mm in diameter, 250 mm long, and filled with methane at 80 mm Hg was used for counting the fragments. The integral pulse spectrum was analyzed. The counting characteristics of the array had a sufficiently good plateau. The mean counting rate of the fragments was equal to 0.92 particles per hour for pure Pu^{238}, and to 7 particles per hour for the mixture of Pu^{242} + Pu^{238}. The statistical accuracy was not less than 5%, and the total error was not greater than 10%. As a result of the measurements, the following values for the half periods were obtained: 5.2×10^{10} years for Pu^{238}, and 6.7×10^{10} years for Pu^{242}.

In a second series of experiments, the method of oxidizing the latent image in nuclear emulsions was used in order to discriminate fission fragments against the background of a large number of α particles.[7] The work was carried out using an NIKFI (Research Institute of the Motion Picture Industry) P-8 emulsion 100μ thick. $K_3Fe(CN)_6$ was used for the oxidation. By means of special tests, it was established that the maximum acceptable $K_3Fe(CN)_6$ concentration amounts to $1:8 \times 10^3$. In our experiment, the concentration was not higher than $1:2 \times 10^4$. Such a concentration ensures good detection of fission fragments, and is sufficient to remove the background up to 10^{11} α particles per cm^2.

The plates were irradiated by samples of Pu^{238} and Pu^{242} for ten to one hundred hours. After the exposure, the emulsions were subjected to the action of the oxidizer (at 15° C for 30 min), were then washed with distilled water (15° C for 30 min),

and then developed in diluted D-19 developer (1:3) (at 19° C for 30 min).

As has been shown by experiments, the effect studied could be imitated by tracks of α particles which appeared due to the decay of radioactive nuclei during the process of washing out the oxidizer and of the development. Special measures were therefore taken to prevent the introduction of impurities into the emulsion during the exposure. On the scanned plates, the density of tracks of spontaneous fission amounted to 800 per cm^2 for Pu^{242} and 200 per cm^2 for Pu^{238}. It has been established that the fragment tracks are detected in the angle interval $20 - 70°$, i.e., the detection efficiency amounts to $(66 \pm 5)\%$.

The measured half-periods for the spontaneous fission of Pu^{238} and Pu^{242} were found to be $(5.0 \pm 0.6) \times 10^{10}$ years and $(6.5 \pm 0.7) \times 10^{10}$ years respectively.

The fact that the results obtained by us using various methods are identical, and are also in good agreement with data already known, permits us to conclude that the method of using nuclear emulsions may be successfully applied for measuring the half-period for the spontaneous fission of Np^{237}. The difficulty consists of the fact that long exposures (several days) are necessary to obtain a marked effect. In such a case, there is a danger of the fading of the latent image when the emulsion is kept for a long time at normal room temperature and humidity. However, control experiments on the fission of U^{235} induced by thermal neutrons showed that the fading could not lead to a considerable decrease in the detection efficiency of spontaneous-fission fragments of Np^{237}.

45 cm^2 of the emulsion were scanned altogether, and two-thirds of the area was scanned consecutively by two observers to avoid bias. Instead of an expected 100 events, only three tracks were found. The observed effect is too small to enable us to speak about an exact value of the decay constant of Np^{237}. It only determines the lower limit of the spontaneous-fission half-period of Np^{237}, for which a value of 10^{18} years is indicated. The difference between our results and those of Segre shows that the fission probability of Np^{237} is evidently considerably less than has been believed.

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