Nuclear Charge Distribution in Symmetric Fission—Sn^{121} Independent Yield*

ARTHUR C. WAHL
Department of Chemistry, Washington University, St. Louis, Missouri

AND

DAVID R. NETHAWAY
Lawrence Radiation Laboratory, University of California, Livermore, California

(Received 8 March 1963)

Less than 12% of 27.5-h Sn^{121} is formed independently in thermal-neutron fission of U^{235}; 24±3% of it is formed by beta decay of 3.1-min In^{121} and 74±10% by beta decay of 30-sec In^{121}. The small independent yield of 27.5-h Sn^{121} and the very small total fission yield of ~25-yr Sn^{121m} show that "the most probable charge" Z_p for fission products with $A=121$ must be less than 50. This result indicates that the large decrease in the total kinetic energy of fission fragments that has been observed as the fragment mass ratio approaches unity cannot be explained entirely by a distribution of nuclear charge in which heavy fragments retain, on the average, 50 or more protons.

It has been suggested that the yield of fission fragments having a closed shell of fifty protons may be favored in low-energy fission.[1] Such a preferred distribution of nuclear charge in which the "most probable charge" Z_P remains near 50 over a large range of mass numbers (~120 to 130) would explain the observed large decrease in the total kinetic energy of fission fragments as their mass ratio approaches unity.[2] We wish to present evidence against this hypothesis and, therefore, in support of the alternative hypothesis[2] that many prompt neutrons are evaporated from fragments of nearly equal mass, an hypothesis for which there is some direct experimental evidence.[3]

The hypothesis that Z_P values remain close to 50 for mass numbers in the approximate range of 120 to 130 was tenable because no fission products with $A>120$ and $Z<50$ had been reported.[4]

Indium isotopes ($Z=49$) with mass numbers 121 and 123 have been reported by Yuta and Morinaga,[5] who observed that irradiation of $Sn^{122}O_2$ with 25-MeV bremsstrahlung produced activities with periods of 3.1 min and 30 sec, and with periods of 36 sec and 10 sec in irradiation of $Sn^{124}O_2$. From studies of the radiations emitted and from considerations of nuclear systematics, the periods were assigned to isomeric states of In^{121} and In^{123}, which were believed to decay, respectively, to 27.5-h Sn^{121} and 41-min Sn^{123}. No chemical identification was made.

We have obtained evidence that the In^{121} isomers are formed in thermal-neutron fission of U^{235}. We quickly separated tin from indium in a freshly irradiated solution of U^{235} and determined the 27.5-h Sn^{121} that grew into the indium fraction.

The separation of tin from indium was accomplished by precipitating a mixture of tin(II) and tin(IV) sulfides from an aqueous HCl solution in which most of the indium remained. The mixture of tin(II) and tin(IV) sulfides was chosen for precipitation because the oxidation states of tin fission products are not known, and by selection of conditions such that 99% of tin(II) and 99% of tin(IV) were retained on the filter, the distribution of fission-product tin between the two oxidation states did not affect the outcome of the experiment. About 60% of the indium came through the filter; the exact amount was determined in each experiment.

The fraction of 27.5-h Sn^{121} growing from In^{121} after separation of tin and indium was determined for various time intervals τ between the occurrence of fission and the separation. These data allowed the calculation of the decay constants λ_1 and λ_2 of the In^{121} isomers and the fractions y_1 and y_2 of 27.5-h Sn^{121} formed from the isomers. The above mentioned quantities are related by the laws of radioactive decay; a good approximation of the relationship is

$$f=y_1e^{-\lambda_1\tau}+y_2e^{-\lambda_2\tau}. \quad (1)$$

Analysis of the data from 15 experiments by the method of least squares via a program for the IBM 7072 computer gives the following results:

Half-life (1): 3.3 ±1.0 min
Half-life (2): 29 ±8 sec
$y_1=$ 0.23±0.07
$y_2=$ 0.78±0.17.

The uncertainties are standard deviations. The half-life values are in excellent agreement with the values of 3.1±0.3 min and 30±3 sec determined by Yuta and Morinaga[5] and confirm their assignment of these

* Work supported by the U. S. Atomic Energy Commission and by the Washington University Computer Center through NSF Grant G-22296. Experimental work was done at the Lawrence Radiation Laboratory, Livermore, California while one of us (A. C. W.) was a summer visitor there.

[1] A. C. Wahl, R. L. Ferguson, D. R. Nethaway, D. E. Troutner, and K. Wolfsberg, Phys. Rev. **126**, 1112 (1962).
[2] J. C. D. Milton and J. S. Fraser, Can. J. Phys. **40**, 1626 (1962).
[3] V. F. Apalin, Yu. M. Gritsyuk, I. E. Kutikov, V. I. Lebedev, and L. A. Mikaelyan, Zh. Eksperim. i Teor. Fiz. **43**, 327 (1962) [translation: Soviet Phys.—JETP **16**, 235 (1963)].
[4] S. Katcoff, Nucleonics **18**, 201 (1960).
[5] H. Yuta and H. Morinaga, Nucl. Phys. **16**, 119 (1960).

periods to isomeric states of In^{121} that decay to the 27.5-h Sn^{121} isomer.

Using Yuta and Morinaga's values for the half-lives and our data only for establishing the values of y_1 and y_2 we obtain the following values:

$$y_1 = 0.24 \pm 0.03$$

$$y_2 = 0.74 \pm 0.10.$$

The fractional independent yield of 27.5-h Sn^{121} is given by

$$z = 1 - y_1 - y_2 = 0.02 \pm 0.10 \quad \text{or} \quad \leqslant 0.12.$$

The good agreement shown in Fig. 1 between the curve derived from these values and the experimental data indicates that the possible fission-product precursors of the In^{121} isomers, Cd^{121} and Ag^{121}, must have appreciably shorter half-lives and/or lower yields than do the In^{121} isomers.

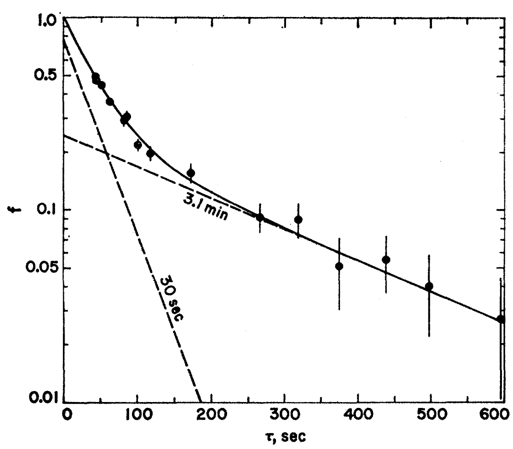

FIG. 1. Plot of the fraction f of 27.5-h Sn^{121} growing from the In^{121} isomers after time interval τ since fission. Points are experimental. The curve represents Eq. (1); values of the parameters are given in the text. The broken lines represent the two terms of Eq. (1) corresponding to decay of the individual components.

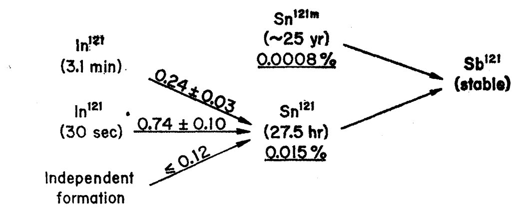

FIG. 2. Genetic relationships between fission products with $A = 121$ from thermal-neutron fission of U^{235}.

Less than 2% of 9.6-day Sn^{125} was found in the filtrate even for the shortest separation times (~40 sec). Therefore, fission-product precursors of this nuclide must have half-lives much shorter than ~40 sec and/or must have fission yields much less than that of 9.6-day Sn^{125} (0.013%[4]).

The present information available concerning the genetic relationships between radionuclides of mass 121 formed in thermal-neutron fission of U^{235} is summarized in Fig. 2. The underlined quantities are fission yields. The half-life and yield of Sn^{121m} were determined by Orth.[6]

The fission yield of Sn^{121m} is a small fraction, (0.0008%)/(0.015%+0.0008%) = 0.05, of the total Sn^{121} yield, so even if all fission-product Sn^{121m} were independently formed, the total fractional independent yield of the Sn^{121} isomers is less than 0.17. Thus, for $A = 121$ the value of Z_P is definitely less than 50, and, if the distribution of nuclear charge is Gaussian and the width parameter σ is 0.62 ± 0.06, as has been determined[1] for some other mass numbers, the value of Z_P is less than 48.9.

[6] C. J. Orth (private communication from the Los Alamos Scientific Laboratory, 1962).