Decay Constant and Mass-Yield Curve for the Spontaneous Fission of Uranium-238*

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The equilibrium ratio of Te^{132}/U^{238} in nonirradiated natural and depleted uranium was measured to be $(2.2\pm0.2)\times10^{-4}$ disintegrations per second per gram of uranium. This value was used to normalize the mass-spectrometric data for the relative yields of krypton and xenon isotopes from the U^{238} spontaneous fission. Combining the mass-spectrometric and radiochemical data obtained by previous investigators, a "mass-yield" curve for the spontaneous fission of U^{238} was constructed in which the equilibrium ratios of the fission products in nonirradiated uranium were plotted against the mass numbers. By summing the area under the curve, a value of $(7.8\pm0.9)\times10^{-17}$ yr^{-1} was obtained for the U^{238} spontaneous fission decay constant. This value is intermediate between the values reported by Fleischer and Price and by Segrè.

I. INTRODUCTION

RECENTLY, Fleischer and Price[1] have reported the spontaneous fission decay constant of uranium-238 to be $(6.9\pm0.2)\times10^{-17}$ yr^{-1}. This value is intermediate between the values of $(8.7\pm0.3)\times10^{-17}$ yr^{-1} reported in 1952 by Segrè[2] and $(5.3\pm0.9)\times10^{-17}$ yr^{-1} reported in 1947 by Perfilov.[3]

Kenna and Kuroda,[4] in 1960, attempted to calculate the spontaneous fission decay constant of uranium-238 from the equilibrium activities of individual fission products in nonirradiated uranium. The result of their preliminary calculation of the decay constant seemed to agree with Segrè's value. The radiochemical method requires low-level counting techniques, as well as the tedious radiochemical purification processes for each mass chain, and hence it may not be quite as accurate as the direct counting methods. Nevertheless, the radiochemical method offers an attractive feature; it simultaneously yields the shape of the mass-yield curve for the spontaneous fission.

Relative yields for the mass chains 83, 84, 86, 131, 132, 134, and 136 from the spontaneous fission of uranium-238 were measured mass spectrometrically by Wetherill[5] and by Young and Thode.[6] The fission decay constant cannot be calculated, however, from the mass-spectrometric data alone. In order to be able to utilize the mass-spectrometric data, we have measured in the present work the Te^{132}/U^{238} ratio in nonirradiated uranium as accurately as possible. Taking advantage of the fact that Te^{132} and Xe^{132} belong to the same fission mass chain, Wetherill's krypton and xenon data, measured for euxenite which had no thermal neutron-induced fission contribution of U^{235}, were converted to the equilibrium ratios of the fission products to uranium in each mass chain (expressed in terms of disintegrations/sec per g U^{238}). By plotting these values and reported values for other mass chains against the mass numbers, and by summing up the area under the curve, the spontaneous fission decay constant of uranium-238 was calculated.

II. EXPERIMENTAL

A. Depleted Uranium

The sample of depleted uranium in the form of $U^{238}O_3$ (99.989% U^{238} and 0.011% U^{235}) was supplied to us by Dr. H. M. Roth of the U. S. Atomic Energy Commission, Oak Ridge, Tennessee. The uranium sample was first exhaustively purified and was converted to uranyl chloride.

The sample was stored both in a limestone cave of Zero Mountains (40 ft deep underground) near Fayetteville, Arkansas and in the Chemistry Building of the University of Arkansas. The sample stored in the cave was brought back to the laboratory and the chemical processing was started within about 30 min.

B. Natural Uranium

A commercial uranyl nitrate (Mallinkrodt chemical works) was also converted to uranyl chloride.

C. Counting Equipment

A pancake-type G. M. counter (Anton 1007-TA; halogen quenched, 2 mg/cm² mica window) was used for the radioactivity measurements. When the counter was operated in anticoincidence with a mantle of surrounding cosmic-ray detectors, it had a background of about 0.9 counts/min. The counter was calibrated with Sr^{90}, K^{40}, and Y^{90} standards for various sample thicknesses.

D. Chemical Procedures

Te^{132} was isolated from kilogram quantities of nonirradiated uranyl chloride and was exhaustively purified. The radiochemical purification procedure consisted of initial precipitation of Te metal by SO_2 gas from the uranium solution containing 40 mg of Te

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[1] R. L. Fleischer and P. B. Price, Phys. Rev. 133, B63 (1964).
[2] E. Segrè, Phys. Rev. 86, 21 (1952).
[3] N. A. Perfilov, Zh. Eksperim. i Teor. Fiz. 17, 746 (1947).
[4] B. T. Kenna and P. K. Kuroda, J. Inorg. Nucl. Chem. 16, 1 (1960).
[5] G. W. Wetherill, Phys. Rev. 92, 907 (1953).
[6] B. G. Young and H. G. Thode, Can. J. Phys. 38, 1 (1960).

carrier, repeated cycles of oxidation of the metal to H_2TeO_3 by concentrated HNO_3, and ferric hydroxide scavenging process. After the radiochemical purification procedure, which usually required 6 to 7 h, more than 50% of the added Te carrier was recovered and the radiochemical and chemical purity of the final Te metal sample was estimated to be greater than 99%.

III. RESULTS AND DISCUSSION

The counting data are plotted in Fig. 1 and the equilibrium Te^{132}/U^{238} ratios are shown in Table I. The most probable charge Z_p for the mass-132 chain lies around 50, and hence the cumulative yields observed at $Z=52$ and 53 should be practically the same. The data obtained in this work are lower than the values obtained by previous investigators.[7,8] The data also indicate that the contribution from the cosmic-ray neutron-induced fission was not appreciable even when the natural uranium stored in the laboratory was used.

TABLE I. Te^{132} and I^{132} in nonirradiated uranium.

Isotope	Equilibrium ratio (10^{-4} disintegrations/sec per g U)	Uranium sample	Reference
Te^{132}	2.2±0.2	Natural	This work
Te^{132}	2.2±0.2	Depleted	This work
Te^{132}	2.2±0.2	Depleted U stored in cave	This work
Te^{132}	3.2±0.3[a]	Natural	Russel[b]
I^{132}	2.5±0.3	Natural	Ashizawa and Kuroda[c]
I^{132}	2.5±0.3	Depleted	Ashizawa and Kuroda[c]

[a] This value is calculated from the Russel's (Ref. 7) value of 4.5±0.5% for the Te^{132} yield in U^{238} spontaneous fission, using the Segrè's (Ref. 2) value of $(8.7\pm0.3)\times10^{-17}$ yr^{-1} for U^{238} spontaneous fission decay constant.
[b] See Ref. 7.
[c] See Ref. 8.

According to Wetherill,[5] the Xe^{132} yield is 3.57 ±0.06%, if the Xe^{136} yield is assumed to be 6.00%. We now adopt the value of $(2.2\pm0.2)\times10^{-4}$ disintegrations/sec per g U^{238} obtained in this work as the cumulative yield for the mass-132 chain, and normalize Wetherill's mass-spectrometric data to this Te^{132} value.

The mass-spectrometric data, which are now expressed in terms of the equilibrium fission product activities (10^{-4} disintegrations/sec per g U^{238}), are plotted against the mass numbers in Fig. 2, together

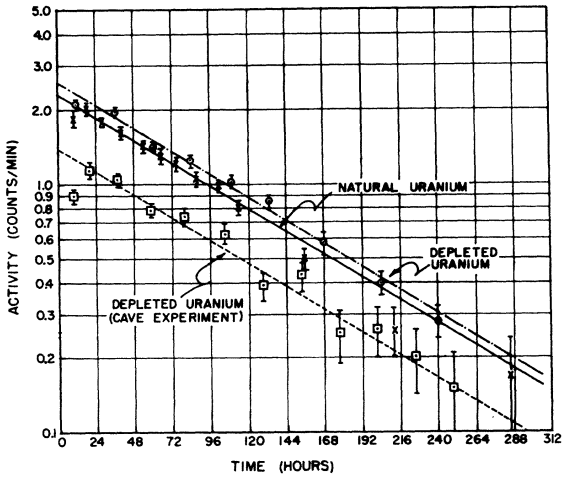

FIG. 1. Tellurium activity isolated from nonirradiated uranium salts—Natural uranium: 1300 g of natural uranium chloride (=900 g U^{238}) were used and the chemical yield was 61%. Depleted uranium: 1740 g of depleted uranyl chloride (=1215 g U^{238}) were used and the chemical yield was 49%. Depleted uranium (Cave experiment): 1740 g of depleted uranyl chloride (=1215 g U^{238}) were stored in Zero Mountain Cave, Fayetteville, Arkansas for 15 days and brought back to the laboratory in 30 min and processed. The chemical yield was 28%.

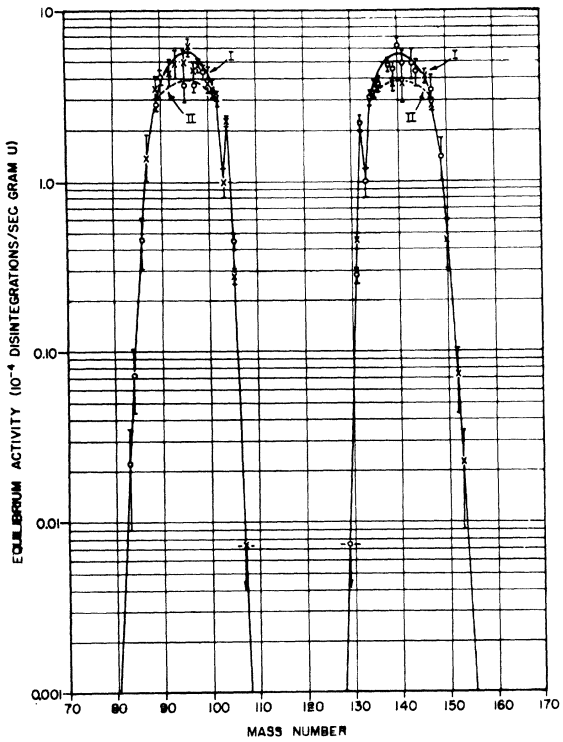

FIG. 2. Equilibrium ratios of the fission products/U^{238} in nonirradiated uranium. O–Experimental points. ×–Mirror points. The mirror points are calculated, using the values of 2 for the number of prompt neutrons emitted per fission. Curve I corresponds to Segrè's value of 8.7×10^{-17} yr^{-1} and curve II corresponds to the value of 6.9×10^{-17} yr^{-1}, reported by Fleisher and Price for the U^{238}-spontaneous fission decay constant.

[7] I. J. Russel, Ph.D. dissertation, University of Chicago, 1956 (unpublished).
[8] F. T. Ashizawa and P. K. Kuroda, J. Inorg. Nucl. Chem. 5, 12 (1957).

with the radiochemical data obtained by other investigators.[7–17]

The equilibrium ratio of relatively long-lived nuclides such as Sr^{90} and Tc^{99} in uranium minerals tends to be higher than the ratio in depleted uranium because of the contribution from the neutron-induced fission, and hence the radiochemical data obtained with uranium minerals[18–20] are not plotted in Fig. 2.

A few additional radiochemical data reported by previous investigators are not plotted in Fig. 2. For example, the Sr^{91} and Sr^{92} values reported by Heydegger and Kuroda[9] are not reliable, as stated by the authors themselves, and hence are omitted. When more than one value is available for one mass chain, the value with minimum uncertainty was selected in Fig. 2.

The mirror points are plotted in Fig. 2, using the value of 2 for the number of prompt neutrons emitted per fission. A rather well-defined "mass-yield curve" is thus obtained. The shape of the U^{238} spontaneous fission mass-yield curve is, of course, markedly different from that of thermal neutron-induced fission of U^{235}, and is characterized by a narrow mass distribution with humps and an extremely low yield in the equal-mass region.

In Fig. 2, curve I corresponds to Segrè's decay constant value[2] of 8.7×10^{-17} yr^{-1} and curve II represents the value of 6.9×10^{-17} yr^{-1} reported by Fleischer and Price[1] in the following expression:

$$N^{238}\lambda_{238,f}=\tfrac{1}{2}\sum_{i=80}^{155}N_i\lambda_i, \qquad (1)$$

where N^{238} is the number of U^{238} atoms and $N_i\lambda_i$ is the cumulative activities of the fission products at mass number i in equilibrium with N^{238}.

In Fig. 2, the experimental points fall mostly within the area between curves I and II, indicating that the decay constant corresponds to a value of $(7.8\pm0.9)\times10^{-17}$ yr^{-1}, which is the average of the values reported by Fleischer and Price[1] and Segrè.[2] It is probably worthy of note that the experimental values for the light peak region tend to agree with curve II, while the values for the heavy peak agree with curve I. It is perhaps significant that the data for the mass chains 138, 139, 141, 143, and 144 were all obtained with natural uranium only, and the measurements for these mass chains should be repeated in future.

ACKNOWLEDGMENT

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[9] H. R. Heydegger and P. K. Kuroda, J. Inorg. Nucl. Chem. **12**, 12 (1959).
[10] H. Arino, AEC Contract At-(40-1)-3235, Annual report 1964, p. 88, 1964 (unpublished).
[11] M. N. Rao, Ph.D. dissertation, University of Cologne, 1962 (unpublished).
[12] M. N. Rao and C. J. Sahani, J. Inorg. Nucl. Chem. **27**, 2679 (1965).
[13] P. L. Parker and P. K. Kuroda, J. Inorg. Nucl. Chem. **5**, 153 (1958).
[14] Tin Mo and P. K. Kuroda, J. Inorg. Nucl. Chem. **27**, 503 (1965).
[15] M. P. Menon and P. K. Kuroda, J. Inorg. Nucl. Chem. **26**, 401 (1964).
[16] M. Attrep. Ph.D. dissertation, University of Arkansas, 1965 (unpublished).
[17] P. K. Kuroda and R. R. Edwards, J. Inorg. Nucl. Chem. **3**, 345 (1957).
[18] B. T. Kenna and P. K. Kuroda, J. Inorg. Nucl. Chem. **26**, 493 (1964).
[19] P. K. Kuroda and R. R. Edwards, J. Chem. Phys. **22**, 1940 (1954).
[20] B. C. Purukayastha and G. W. Martin, Can. J. Chem. **34**, 293 (1956).

Erratum

Coupling Constants in Muon Capture, L. L. FOLDY AND J. D. WALECKA [Phys. Rev. **140,** B1339 (1965)]. The sign of one of our small nucleon recoil corrections is incorrect. Equation (13) and therefore Eq. (35) and the last term in Eq. (43) should have opposite signs. This increases our value of $F_A{}^\mu/F_A{}^\beta$ by 2% and that of $\mu(\nu^2)$ by 8% (which is well within our quoted error). We are grateful to J. Friar for calling this mistake to our attention.