

Yields and Half-Lives in the Mass-99 Fission-Product Chain

DAVID E. TROUTNER,* ROBERT L. FERGUSON, AND G. DAVIS O'KELLEY
Oak Ridge National Laboratory,[†] Oak Ridge, Tennessee

(Received 7 January 1963)

Half-lives, fission yields, and genetic relationships of Zr, Nb, and Mo members of the mass-99 fission-product chain were studied by radiochemical methods. Evidence was obtained for a Nb⁹⁹ isomer of half-life 10 ± 2 sec, which decays directly to Mo⁹⁹ in at least 52% of its disintegrations. Attempts to observe gamma radiation from this isomer in rapidly separated niobium fission products were not successful. A half-life of 2.3 ± 0.3 min was obtained for a known isomer of Nb⁹⁹, in good agreement with reported values. The fraction of the mass-99 fission product chain which passes through the longer-lived isomer was found to be ≤ 0.36 . The cumulative fractional chain yield of both isomers was determined to be 0.94 ± 0.11 , consistent with the predicted value of ~ 0.99 . Although this result is not sufficiently precise to be a stringent test of the proposed preference in low-energy uranium fission for a 50-42 proton split, it does indicate that the maximum effect would be sufficient to distort the "normal" charge-distribution curve by an amount equivalent to approximately one charge unit. No evidence was found for 35-sec Zr⁹⁹, previously reported to be a fission product. If it occurs in fission, its cumulative fractional chain yield is ≤ 0.03 , in contrast to the expected value of ~ 0.80 . An upper limit of 1.6 sec was estimated for the half-life of Zr⁹⁹.

INTRODUCTION

IT has been proposed that the independent fission yields of Mo isotopes from thermal-neutron fission of ${}_{92}\text{U}^{235}$ may be high due to a favored 50-42 proton split during the fission process.¹ The mass-99 fission-product chain appeared to be one for which this hypothesis could be tested by measuring the activity of Mo⁹⁹ separated soon after fission from its reported precursors (35-sec Zr⁹⁹ and 2.4-min Nb⁹⁹).² Preliminary experiments, in which intervals between irradiation and separation were ≥ 45 sec, indicated an apparent fractional independent yield of Mo⁹⁹ of ~ 0.55 , in agreement with the previous unpublished result (0.51 ± 0.03) of Vallis.³ The so-called normal yield, as predicted by the method of Wahl *et al.*,⁴ is 0.01.

This apparent high yield of Mo⁹⁹ seemed to offer evidence for the favored 50-42 proton split; it might, however, have been due to a previously undetected short-lived isomer of Nb⁹⁹ decaying directly to Mo⁹⁹. Therefore, an investigation was undertaken to search for this suspected isomer and, if possible, to determine its half-life, as well as to measure fission yields in the mass-99 chain. The results of this investigation are presented below.

EXPERIMENTAL

Irradiations

Irradiations were made at the Oak Ridge Research Reactor in the pneumatic tube facility where the thermal-neutron flux density was $\sim 6 \times 10^{13}$ neutrons cm^{-2} sec^{-1} . A cadmium ratio for U²³⁵ fission of ~ 60 was meas-

ured by comparing the number of fissions produced in unshielded uranium samples with the number produced in samples shielded by a 0.040-in.-thick cadmium absorber. Both Zr⁹⁷ and Mo⁹⁹ were used as fission monitors. Solutions of uranium (93% U²³⁵) were irradiated in high-density polyethylene capsules for periods of either 2 or 10 sec. All uranium solutions were $\sim 0.1M$ in HNO₃ and $\sim 0.05M$ in HF or H₂C₂O₄, and some also contained Fe(III), Zr(IV), and Mo(VI) carriers.

Procedure

Zirconium, niobium, and molybdenum were separated from one another at various times after irradiation. This gave a means of measuring the fraction of mass-99 fission products present as Zr⁹⁹, as Nb⁹⁹, and as Mo⁹⁹ at the time of separation. In one series of experiments, zirconium and niobium were coprecipitated with Fe(OH)₃ from an NH₄OH solution, leaving molybdenum in solution. In another series of experiments, zirconium was coprecipitated with Fe(OH)₃ from an NH₄OH-H₂O₂ solution, leaving niobium and molybdenum in solution.

For some experiments of the first series, an irradiated sample was divided into two portions (Procedure A, Appendix). In one portion, molybdenum was separated from zirconium and niobium relatively soon after irradiation. The molybdenum from this separation retained less than 0.5% of the zirconium and niobium present and was further purified by a solvent extraction procedure.⁵ In the second portion, molybdenum was purified after essentially complete decay of Zr⁹⁹ and Nb⁹⁹. Therefore, the ratio of Mo⁹⁹ from the first portion to that from the second gave directly the fraction of the mass-99 fission products present as Mo⁹⁹ at the early separation time. This ratio is the quantity Q used below (see Calculations).

In Procedure B (Appendix), which allowed earlier separation times, zirconium and niobium were separated

* Permanent address: University of Missouri, Columbia.

[†] Operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.

¹ A. C. Wahl, *J. Inorg. Nucl. Chem.* **6**, 263 (1958).

² C. J. Orth and R. K. Smith, *J. Inorg. Nucl. Chem.* **15**, 4 (1960), and references therein.

³ D. G. Vallis, quoted by I. F. Croall, Harwell (private communication, 1960).

⁴ A. C. Wahl, R. L. Ferguson, D. R. Nethaway, D. E. Troutner, and K. Wolfsberg, *Phys. Rev.* **126**, 1112 (1962).

⁵ D. R. Wiles and C. D. Coryell, *Phys. Rev.* **96**, 696 (1954).

from molybdenum using the entire irradiated sample. Approximately 0.5% of the molybdenum present was retained in the zirconium-niobium sample. Then, after Zr^{99} and Nb^{99} in the zirconium-plus-niobium fraction had decayed, the resulting Mo^{99} was purified. This Mo^{99} radioactivity was compared to that of Zr^{97} , which was also present in the zirconium-plus-niobium fraction and was purified by a solvent-extraction procedure.⁶ The Zr^{97} was an internal standard and served to correct for incomplete recovery of fission products from the irradiation capsule and for variations in the number of fissions due to differences in irradiation time, reactor flux, and amount of uranium irradiated. The standard was calibrated by the use of the activity ratio of Mo^{99} to Zr^{97} as determined from irradiations in which the recovery of fission products was complete. These two Mo^{99}/Zr^{97} activity ratios were used to calculate the fraction of mass-99 fission products present as Zr^{99} plus Nb^{99} at the early separation time. This is the ratio $1-Q$ used below (see Calculations).

Procedure *C* (Appendix), used in the second series of experiments, was similar to Procedure *B* except that zirconium was separated from both niobium and molybdenum. The fraction of mass-99 fission products calculated in this case was that present as Zr^{99} at the early separation time. However, in this separation approximately 2% of the niobium and 0.5% of the molybdenum present followed the zirconium.

In every experiment, the specific radioactivity of a Mo^{99} sample was compared to that of another Mo^{99} sample from the same irradiation or from the standard-calibration irradiation. Similarly when a Zr^{97} sample was counted, its radioactivity was compared to that of another Zr^{97} sample from the standard-calibration irradiation. Since all Mo^{99} and Zr^{97} samples were mounted and counted in a standard way, the only counting corrections necessary were for variation in sample thickness; these were less than 10% and introduced an uncertainty in the measurement of less than 0.5%.

The initial separations and counting methods are described in further detail in the Appendix.

CALCULATIONS

Consider the fission-product decay chain, shown in Fig. 1, where *A* and *B* represent isomers of Nb^{99} , and *C* represents Mo^{99} . The effects of Zr^{99} precursors of Nb^{99} have been neglected (see Results). The cumulative yield of Mo^{99} has been assumed to be the total chain yield, since the expected independent yields of Tc^{99} , Tc^{99m} , and Ru^{99} are less than 10^{-4} of the chain yield. Define:

x = fractional cumulative yield of *A*, based on the cumulative yield of *C*.

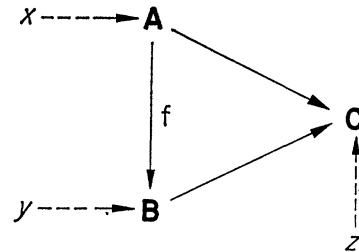


FIG. 1. Generalized decay chain used in defining terms of Eq. (1).

y = fractional cumulative yield of *B* less its fractional yield from *A*, both based on the cumulative yield of *C*.

z = fractional independent yield of *C*, based on the cumulative yield of *C*.

f = fraction of *A* which decays to *B*.

T = duration of irradiation producing fissions at a constant rate.

t, t' = time intervals between the end of an irradiation and the separation of *C* from *A* and *B*.

$\tau = T/2 + t; \tau' = T/2 + t'$.

Q = ratio of activity of *C* separated after t to activity of *C* separated after a substantially longer time t' , corrected for chemical yield, to the same number of fissions, and for decay to the same time after irradiation.

$1-Q$ = ratio of activity of *C* produced from decay of *A* and *B* separated after t to activity of *C* separated after a substantially longer time t' , corrected for chemical yield, to the same number of fissions, and for decay to the same time after irradiation.

For $\lambda_A \gg \lambda_C, \lambda_B \gg \lambda_C, t$ very short compared to the period of *C*, and t' very long compared to the periods of *A* and *B*, and with the approximation (in error by $\sim 2\%$ for the longer irradiation time) that $[1 - \exp(-\lambda T)] = \lambda T [\exp(-\lambda T/2)]$, the following equation has been derived from the equations of radioactive transformation.

$$1-Q = c_1 \exp(-\lambda_A \tau) + c_2 \exp(-\lambda_B \tau), \quad (1)$$

where

$$c_1 = x + \frac{f\lambda_A x}{\lambda_B - \lambda_A}, \quad c_2 = y - \frac{f\lambda_A x}{\lambda_B - \lambda_A}, \quad \text{and} \quad 1 - c_1 - c_2 = z.$$

Equation (1) was programmed for the IBM 7090 computer and the parameters c_1, c_2, λ_A , and λ_B were evaluated by a least-squares analysis in which the data are weighted in proportion to the reciprocal of the square of the estimated uncertainty in $1-Q$.

RESULTS

Zr^{99}

Small amounts of Mo^{99} activity were observed in the zirconium fraction separated by Procedure *C*. These amounts, expressed as fractions of the total chain, are

⁶ K. F. Flynn, L. E. Glendenin, and E. P. Steinberg, quoted in E. P. Steinberg, *The Radiochemistry of Zirconium and Hafnium*, Radiochemistry Monograph No. NAS-NS 3011 (National Academy of Sciences—National Research Council, Washington, D. C., 1960).

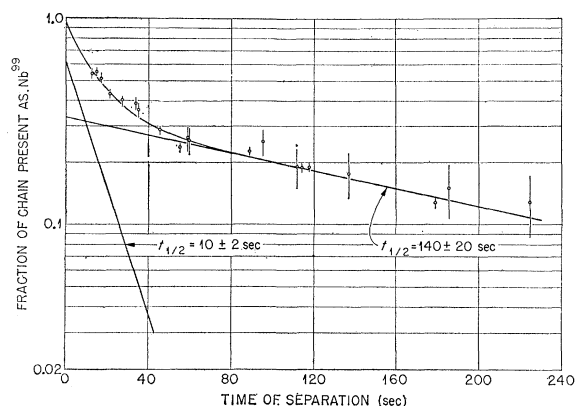


FIG. 2. Mo^{99} from Zr-Nb Fraction. $(1-Q)$ vs τ .

shown in Table I with the corresponding times of separation.

A least-squares fit of a single-component exponential decay curve to these data gave a half-life of 75 ± 13 sec, in very poor agreement with the reported 35 ± 5 -sec half-life² of Zr^{99} , and an apparent fractional cumulative yield of 0.019 ± 0.001 for a 75-sec Zr^{99} , in disagreement with the predicted⁴ yield, ~ 0.80 . In addition, the Mo^{99} observed was never more than 5% of the amount which could have grown from Nb^{99} isomers present at the time of separation. Since control experiments with Nb^{95} tracer showed that at least 2% of the Nb^{95} was carried by $\text{Fe}(\text{OH})_3$ precipitated from $\text{NH}_4\text{OH}-\text{H}_2\text{O}_2$ solution, it is a reasonable assumption that the Mo^{99} observed in zirconium fraction was due to Nb^{99} contamination. The data from these experiments, therefore, provide no evidence for a 35-sec Zr^{99} precursor of Mo^{99} .

If it is assumed that the fractional cumulative yield of Zr^{99} is ~ 0.80 , the upper limit of the half-life of Zr^{99} , as calculated from the minimum amount of Mo^{99} observed for $\tau=9$ sec, is 1.6 sec. However, the Mo^{99} activities observed for $\tau=13$ sec, $\tau=24$ sec, and $\tau=27$ sec were equal within experimental error to that observed at $\tau=9$ sec, indicating that only a small part of the observed Mo^{99} could be due to decay of a short-lived Zr^{99} . Therefore, the half-life of Zr^{99} is probably much less than 1.6 sec and has been neglected in further calculations.

TABLE I. Mo^{99} from Zr fraction.

Irradiation time, T (sec)	Separation time, τ (sec)	Mo^{99} (fraction of chain) ^a
2	9	0.023
2	9	0.015
2	12	0.018
2	13	0.015
10	24	0.014
10	27	0.015
10	53	0.012
10	73	0.008
10	105	0.008

^a Uncertainty due to radiochemical errors was estimated to be 0.001; however, uncertainty due to varying niobium contamination was not assessed.

TABLE II. Mo^{99} from separated Zr-Nb.

Separation time, ^a τ (sec)	Procedure ^b	Fraction present as Nb $(1-Q)$
12.4	B	0.540 ± 0.027
14.9	B	0.549 ± 0.028
17.3	A	0.508 ± 0.025
21.1	B	0.437 ± 0.022
27.2	A	0.400 ± 0.030
33.8	A	0.384 ± 0.031
35.0	A	0.357 ± 0.032
45.5	B	0.285 ± 0.015
55.3	B	0.235 ± 0.012
58.8	A	0.262 ± 0.037
59.5	A	0.254 ± 0.038
89.1	B	0.226 ± 0.012
95.7	A	0.251 ± 0.038
111.9	A	0.190 ± 0.041
114.1	B	0.188 ± 0.010
117.8	B	0.189 ± 0.010
136.9	A	0.177 ± 0.042
179.1	B	0.127 ± 0.007
185.4	A	0.150 ± 0.043
224.5	A	0.128 ± 0.044

^a All irradiations were 10.0 sec.

^b See text for description of procedures used.

Nb^{99}

The results from Procedures A and B, in which Zr and Nb were separated from Mo, are given in Table II. The errors shown represent an estimated $\pm 5\%$ error in the radiochemical measurement of Q by Procedure A and of $1-Q$ by Procedure B.

A plot of $1-Q$ as a function of τ is shown in Fig. 2. This curve was resolved into two components by a least-squares analysis. The two half-lives obtained are 10 ± 2 sec and 140 ± 20 sec. The half-life of the longer lived component is in excellent agreement with the 2.4 ± 0.3 -min half-life² previously reported for Nb^{99} . Since it has been shown that the shorter-lived component is not Zr^{99} (see preceding section), it must be an isomer of 2.4-min Nb^{99} .

The intercepts of the shorter-lived and longer-lived components are 0.61 ± 0.11 and 0.33 ± 0.03 , respectively. Since $c_1 + c_2 = x + y$, the cumulative yield of both isomers ($x + y$) is 0.94 ± 0.11 , in agreement with the predicted⁴ yield, ~ 0.99 .

It is impossible, of course, to determine the genetic relation of the two isomers from our data. However, by assuming that A was the shorter lived Nb^{99} , limits were determined for $x (\geq 0.50)$, $y (\leq 0.36)$, and $f (\leq 0.48)$. This was done by calculating their maximum and minimum values consistent with the experimental limits found for c_1 , c_2 , λ_A , and λ_B . From these limits, it can be seen that at least 0.50 of the chain passes through the shorter lived isomer and not more than 0.36 of the chain passes through the longer lived isomer.

Attempts were made to observe directly any gamma rays which might result from an isomeric transition of the 10-sec Nb^{99} by taking gamma scintillation spectra of quickly purified Nb fission products. However, because of the relatively long time (62 sec) between irradiation and counting and because of the exceedingly com-

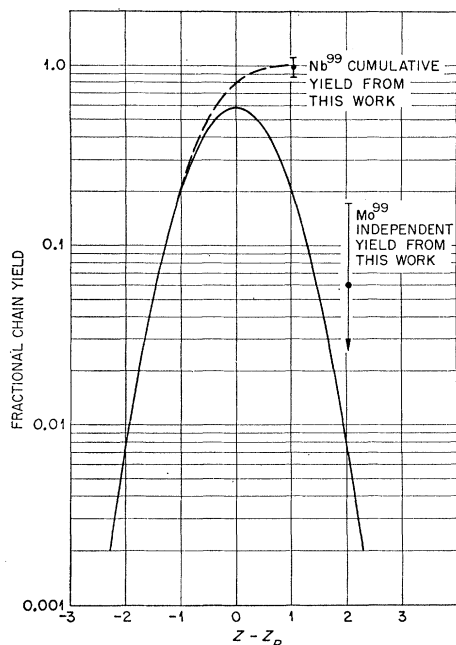


FIG. 3. Distribution of yield with Z in the mass-99 chain. Curve from Wahl *et al.* (see reference 4); for $A=99$, $Z_P=39.95$.

plex spectra of short lived Nb fission products, the results were inconclusive.

Mo^{99}

The data obtained do not permit an accurate calculation of the fractional independent yield of Mo^{99} . However, the value determined from the relation $z=1-c_1-c_2$ is 0.06 ± 0.11 , which is consistent with the predicted⁴ normal yield, ~ 0.01 . These results are shown in Fig. 3 with the normal charge distribution curve of Wahl *et al.*⁴

DISCUSSION

As can be seen from Fig. 3, the maximum distortion of the normal charge distribution curve is confined by the Mo^{99} fractional independent yield to about one charge unit. The error limits shown are the standard deviations of the measured yields. Unfortunately, this result is not sufficiently accurate to test the 50-42 proton-split hypothesis, because it was proposed¹ to explain low yields of Nb^{96} and Tc^{98} which are displaced from the normal curve by ≤ 0.5 charge unit.⁴

All of the well-characterized niobium isotopes with odd mass numbers exhibit nuclear isomerism.⁷ The ground-state properties of these nuclei are largely determined by the odd proton, and the nuclear shell model leads to the conclusion that the $p_{1/2}$ and $g_{9/2}$ levels are in close competition. In the niobium region, experimental evidence indicates a strong preference for a $g_{9/2}$ ground state, and the spin of the ground state in ${}_{41}Nb_{52}^{93}$

⁷ *Nuclear Data Sheets*, compiled by K. Way *et al.*, (Printing and Publishing Office, National Academy of Sciences-National Research Council, Washington, 25, D. C.).

has been measured as $9/2+$. Studies of the decay schemes of Nb^{91} , Nb^{93} , Nb^{95} , and Nb^{97} indicate two isomeric levels in each of these nuclei with spins of $1/2-$ for the upper and $9/2+$ for the lower level.

The existence of the 10-sec Nb^{99} isomer reported here is consistent with the properties of the other odd- A niobium isotopes. A separation between the $p_{1/2}$ and $g_{9/2}$ levels of about 0.9 MeV is expected for Nb^{99} , using as a guide the spacings observed for its neighbors, Nb^{95} and Nb^{97} . Systematics of the comparative lifetimes for $M4$ transitions in odd- A nuclei predict that the partial half-life of a 0.9-MeV, $M4$ transition in Nb^{99} should be about 7 sec.^{8,9} The effect of internal conversion can be neglected at this energy, but the beta branching from 10-sec Nb^{99} will make the total half-life computed for the Nb^{99} isomer somewhat different from the 7 sec estimated for the gamma-ray transition alone. The close agreement between the measured half-life and the half-life estimated from systematics suggests very strongly that the isomer of Nb^{99} reported above has spin and parity ($1/2-$), spaced about 0.9 MeV above the ($9/2+$) ground state.

Although the data obtained here provide no affirmative evidence for the occurrence of 35-sec Zr^{99} in fission, it is not impossible that part of the activity observed in the zirconium fraction was due to Zr^{99} . If this is true, however, the upper limit for the Zr^{99} fractional cumulative yield (~ 0.03) would indicate a distortion of the normal charge distribution curve equivalent to ≥ 1.6 charge units—an amount much greater than any observed deviation. The 1.6-sec upper limit estimated here for the half-life of Zr^{99} is based on a normal yield (~ 0.80). Isomerism is not expected in ${}_{40}Zr_{59}^{99}$ because, according to the shell model,⁹ the levels $d_{5/2}$ and $g_{7/2}$ compete for the odd neutron. Since competition between these levels cannot give rise to long-lived isomerism, it is not likely that the reported 35-sec activity is due to a low-yield isomer of Zr^{99} .

ACKNOWLEDGMENTS

The authors are grateful to M. H. Lietzke for his assistance in the least-squares calculations, to J. F. Emery for help with some of the experiments, and to J. Halperin for his careful criticism of the manuscript.

APPENDIX

Initial Separations

Procedure A

In these experiments, 1 ml of a 0.01M HNO_3 solution of 93% enriched U^{235} (0.1 mg/ml) and a complexing agent for zirconium and niobium (0.05M $H_2C_2O_4$

⁸ D. E. Alburger, in *Handbuch der Physik*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 42; M. Goldhaber and A. W. Sunyar in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (North-Holland Publishing Company, Amsterdam, 1955), pp. 461-7.

⁹ M. G. Mayer and J. Hans D. Jensen, *Elementary Theory of Nuclear Shell Structure* (John Wiley & Sons, Inc., New York 1955), p. 211ff.

or HF) was irradiated. To provide additional opportunity for isotopic exchange, in some runs Mo(VI) [0.05 mg/ml], Nb(V) [5 mg/ml], Zr(IV) [5 mg/ml], and Fe(III) [20 mg/ml] were included in the rabbit. (Similar results were obtained whether or not the carriers were included in the rabbit.) Within five seconds after the irradiation ended, the rabbit was pierced with a stainless steel hypodermic needle, and the irradiated solution was drawn into a glass mixing vessel. This vessel contained ~ 7 ml of a solution of zirconium (0.5 mg/ml), niobium (0.5 mg/ml), and iron (3 mg/ml) carriers (unless these were present in the rabbit), as well as Mo(VI) [6 mg/ml] and NaBrO₃ (14 mg/ml) to ensure that the molybdenum was in the +6 oxidation state. The irradiated solution was stirred with this carrier solution for ~ 5 sec, or longer for the later separation times. Approximately one-half of this solution was then drained into a second vessel which contained 5 ml of 15*M* NH₄OH. A precipitate, which consisted of ferric hydroxide, zirconium hydroxide, and hydrous niobium oxide, formed immediately. The time at which this precipitate formed was taken to be the time at which separation of zirconium and niobium from molybdenum occurred. All times were measured by electrical timers and were accurate to ± 0.05 sec. The mixture was quickly drained into a fritted glass funnel and filtered by suction. (Iron carrier was used in these separations so that the hydroxide precipitate could be filtered quickly). Subsequently, molybdenum in the filtrate, as well as that in the remaining half of the solution in the first vessel, was purified⁵ and the two samples mounted for counting. Control experiments using Zr⁹⁵ - Nb⁹⁵ tracer indicated that $\leq 0.1\%$ of the zirconium and niobium separated as described above contaminated the molybdenum in the filtrate. Furthermore, when the Zr⁹⁷ content of the filtrate from a typical experiment was determined and compared to that in the precipitate, $\leq 0.5\%$ of the total Zr⁹⁷ was found in the filtrate.

Procedure B

In this procedure, 1 ml of a 0.05*M* HF-0.01*M* HNO₃ solution containing 0.1 mg of 93% enriched U²³⁵ and 0.5 mg Zr(IV), 1 mg Nb(V), and 1.5 mg Fe(III) carriers was irradiated. After irradiation (and an appropriate delay when later separation times were desired) this solution was drawn into the mixing vessel which contained 7 ml of a solution of Mo(VI) [4 mg/ml], NaBrO₃ (14 mg/ml), and NH₄OH (4.3*M*). The hydroxide precipitate formed immediately and was stirred for ~ 5 sec. The mixture was then filtered and the precipitate washed quickly with two 10 ml portions of 6*M* NH₄OH. Some time later the precipitate was dissolved and washed through the funnel by 1 ml of a 0.3*M* HF-1*M* HNO₃ solution containing 10 mg Mo(VI) and 5 mg Zr(IV) carriers. This washing was followed by another with 14 ml of 1*M* HNO₃. Zirconium was extracted from this solution into a xylene-thenoyltrifluoroacetone mixture and purified by the procedure of Flynn.⁶ The aqueous

phase from this extraction contained the Mo⁹⁹, and, after evaporation to dryness with 0.5 ml of 18*M* H₂SO₄, molybdenum was taken up in 6*M* HCL and purified by the procedure of Wiles and Coryell.⁵ Control experiments using Mo⁹⁹ tracer indicated that $\sim 0.5\%$ of the Mo⁹⁹ present at separation time was retained in the hydroxide precipitate.

The Zr⁹⁷ internal standard was calibrated in runs similar to those described above except that the irradiated solution was removed from the rabbit only after essentially complete decay of Zr⁹⁹ and Nb⁹⁹ and that the hydroxides were not precipitated. The carrier solution and 1*M* HNO₃ were flushed through the rabbit to obtain complete recovery of the fission products. Samples of Zr⁹⁷ and Mo⁹⁹ were then processed in the same way as described above. The standard deviation of the mean Mo⁹⁹/Zr⁹⁷ activity ratio determined in this way for five irradiations was 0.7% of the ratio.

Procedure C

In experiments in which this procedure was used, the solutions irradiated contained 93% enriched U²³⁵ (0.1 mg/ml for the 10-sec irradiations; 1 mg/ml for the 2-sec irradiations), HNO₃ (0.01*M*), and HF (0.05*M*). The volume was again 1 ml. After irradiation (and a delay for later separation times) the solution was transferred to a mixing vessel containing 5 ml of a solution of Zr(IV) [0.1 mg/ml], Nb(V) [0.2 mg/ml], Mo(VI) [6 mg/ml], and Fe(III) [0.3 mg/ml] carriers, NaBrO₃ (50 mg/ml), and H₂O₂ (1.2%). The resulting solution was stirred 3-5 sec and drained into a second vessel which contained ~ 7 ml of an 11*M* NH₄OH-0.9% H₂O₂ solution. A ferric hydroxide-zirconium hydroxide precipitate formed immediately, and after the mixture was stirred 2-5 sec, it was filtered. The precipitate was quickly washed with two 10-ml portions of a solution containing NH₄OH (5*M*), H₂O₂ (1%), and Nb(V) [0.1 mg/ml] and Mo(VI) [0.1 mg/ml] carriers. In this separation, the H₂O₂ complexed the niobium and kept it in solution. Control experiments with Nb⁹⁵ tracer indicated that $\sim 2\%$ of the niobium was carried by the precipitate in the separation. The precipitate was removed from the filter funnel, and Zr⁹⁷ and Mo⁹⁹ were determined as described under procedure B. The calibration of the Zr⁹⁷ internal standard, which was also described under Procedure B, was applicable to these experiments as well.

Counting

All samples were filtered, dried, and weighed on filter paper disks 25 mm in diameter. The disks were mounted on aluminum counting plates and held in place by strips of polyester film adhesive tape (6.65 mg/cm²). Decay of the beta activity was followed on a flow-type proportional counter for at least two weeks for all samples. As an additional check of radiochemical purity, gamma spectra of some samples were determined with a scintillation spectrometer; in all cases only the expected radiations were found.