

to contamination of the foil. In any case it seems quite clear at this point that the radiation emitted by rhenium, which in total intensity is in agreement with the earlier result, 0.14 count per minute per cm<sup>2</sup> of area of the metal under  $2\pi$  geometry counting conditions is much less energetic than a 43-keV beta spectrum.

When the screen wall counter was used,<sup>2,3</sup> with a distance of 1.26 cm between the sample cylinder on which the metallic rhenium was mounted and the screen gauze defining the wall of the counter, it was observed that with the wall of the counter negative by any voltage larger than 10 volts with respect to the sample cylinder the rhenium radiation was undetectable with the lowest pressures of gas obtainable. For example, 3 millimeters of Hg pressure of ethylene mixed with 1.1 cm Hg pressure of argon gave less than 3 percent transmission of the rhenium radiation. This would correspond, for an exponential absorption law, to a mean free path of 10 micrograms per cm<sup>2</sup>, or using a ratio of 10 empirically deduced between the range and the mean free path of soft beta radiation,<sup>4</sup> we calculate that the range of the beta radiation from rhenium must be less than 100 micrograms per cm<sup>2</sup>, which according to the relation of Katz and Penfold<sup>5</sup> gives an upper energy limit to the beta radiation from rhenium of 8 keV instead of the 43 keV given earlier.<sup>1</sup>

It is also clear that having reduced the energy of the radiation, the half-life must be recalculated. Whereas the earlier half-life was thought to be about  $10^{12}$  yr, it now is clear that it must be reduced by at least the ratio of the range of 43-keV radiation—about 3 milligrams per cm<sup>2</sup>—to the new upper limit to the range—0.1 milligram per cm<sup>2</sup>—or to  $10^{11}$  years or less. It cannot be clear how much less than  $10^{11}$  years the half-life of rhenium 187 is until a successful attempt to measure the radioactivity in a gaseous state has been made. The systematics of the penetrating power of radiations as soft as rhenium is relatively unknown, since there is no other known beta spectrum as soft as this natural radioactivity seems to be.

Several attempts have been made in our laboratory to prepare gaseous compounds of rhenium which could be used in proportional counters to determine not only the half-life, but also the energy of the radiation. The compounds tried were: rhenium trimethyl, which we failed to prepare, and rhenium oxy-chloride and rhenium hexafluoride. The latter two were prepared successfully, but proved to be rather recalcitrant counter gases, and no important data have yet been obtained.

It would seem reasonable to attempt to test for the possibility that the natural radioactivity of rhenium may correspond to a half-life of as short as a few billion years, in which case the accumulation of osmium 187, the daughter isotope, in old rocks should be observable, and use of this observation might produce the first reliable value of the half-life of this most interesting naturally radioactive isotope. The shell

model and beta decay systematics<sup>6</sup> say that rhenium 187 should have an  $ft$  value of about  $10^{10}$  and be only first-forbidden with a spin change of two. This indicates that a one-keV transition should correspond to a half-life of about  $10^{11}$  years, which is in permissive agreement with the facts as stated previously.

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<sup>1</sup> S. N. Naldrett and W. F. Libby, *Phys. Rev.* **73**, 487 (1948); N. Sugarman and H. Richter, *Phys. Rev.* **73**, 1411 (1948); B. Gauthé and J. M. Blum, *Compt. rend.* **236**, 1255 (1953).

<sup>2</sup> W. F. Libby, *Phys. Rev.* **46**, 496 (1934).

<sup>3</sup> D. D. Lee and W. F. Libby, **55**, 245 (1939).

<sup>4</sup> A. D. Suttle, Jr., thesis, University of Chicago, 1952 (unpublished).

<sup>5</sup> L. Katz and A. S. Penfold, *Revs. Modern Phys.* **24**, 1 (1952).

<sup>6</sup> M. G. Mayer (private communication).

### Fine Structure in U<sup>233</sup> Fission

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**F**INE structure in the light group of the mass distribution for slow-neutron-induced fission of U<sup>235</sup> was reported in a previous publication.<sup>1</sup> An abnormally high yield was observed at mass 100 suggesting a preference for this mass in the fission act, perhaps as the complement of a preferred 82-neutron

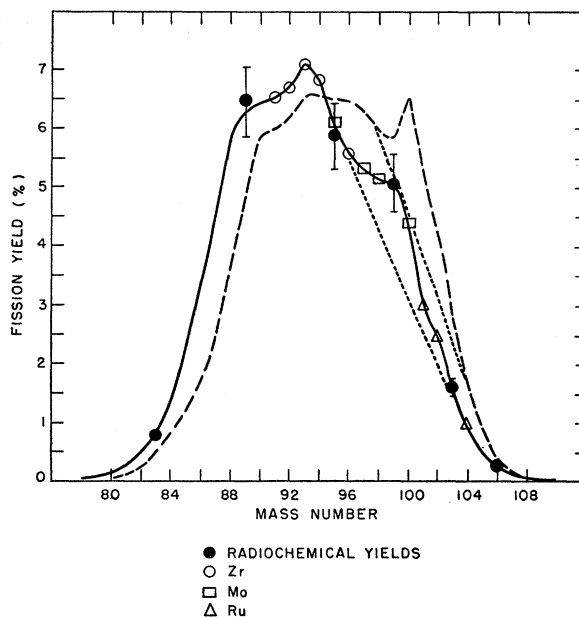


FIG. 1. Yield-mass curves for the light groups in slow neutron-induced fission of U<sup>233</sup> (solid line) and U<sup>235</sup> (dashed line).

shell in the heavy fragment at about mass 134. The possibility of such a preference in fission was first suggested by Wiles.<sup>2</sup> Preliminary mass spectrometric determinations of the relative isotopic abundances of fission-produced zirconium, molybdenum, and ruthenium show that this phenomenon also occurs in slow-neutron-induced fission of  $U^{233}$ . The relative abundance data for three elements were fitted together with the aid of the radiochemically determined yields<sup>3</sup> of  $Sr^{89}$  (6.5 percent),  $Zr^{95}$  (5.9 percent),  $Mo^{99}$  (5.1 percent), and  $Ru^{103}$  (1.6 percent), to give a yield-mass curve for the light group (solid line in Fig. 1). It is seen that, regardless of normalization, fine structure is indicated in the region around mass 99. The corresponding curve for  $U^{235}$  fission<sup>1</sup> (dashed line in Fig. 1) is shown for comparison. Hypothetical smooth curves (dotted lines) in the fine structure region serve to show the effect more clearly. A dip in the  $U^{233}$  yield-mass curve at masses 91 and 92 is also apparent as in the case of  $U^{235}$  fission.

The difference in appearance of the fine structure in  $U^{233}$  fission is probably the result of a narrowing of the light group peak and the shift toward lower mass. The maximum of the effect occurs at about mass 99 which is complementary to the heavy fission products containing 82 neutrons (around mass 133), and is

consistent with the interpretation in terms of preferential in fission. It is interesting to note that fine structure in the light group for spontaneous fission of  $Cm^{242}$  occurs at mass 105,<sup>4</sup> which is again complementary to the 82-neutron shell region, thus providing additional evidence for this interpretation.

A detailed investigation of the mass distribution in neutron-induced fission of  $U^{235}$  covering both light and heavy groups is nearing completion and will be reported soon. Mass spectrometry and the isotope dilution technique have been used to obtain absolute fission yields, thus avoiding the difficulties of normalizing relative isotopic abundances of one element to those of another. Similar investigations of  $U^{233}$  and  $Pu^{239}$  fission will be undertaken.

<sup>1</sup> Glendenin, Steinberg, Inghram, and Hess, *Phys. Rev.* **84**, 860 (1951).

<sup>2</sup> D. R. Wiles, M.Sc. thesis, Department of Chemistry, McMaster University, Hamilton, Ontario, August, 1950 (unpublished); Wiles, Smith, Horsley, and Thode, *Can. J. Phys.* **31**, 419 (1953).

<sup>3</sup> Steinberg, Seiler, Goldstein, and Dudley, U. S. Atomic Energy Commission Document MDDC-1632, January, 1948 (unpublished); *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Energy Series, Plutonium Project Record, Vol. 9, Div. IV, Appendix B. Revised values to be published.

<sup>4</sup> E. P. Steinberg and L. E. Glendenin, *Phys. Rev.* **95**, 431 (1954).