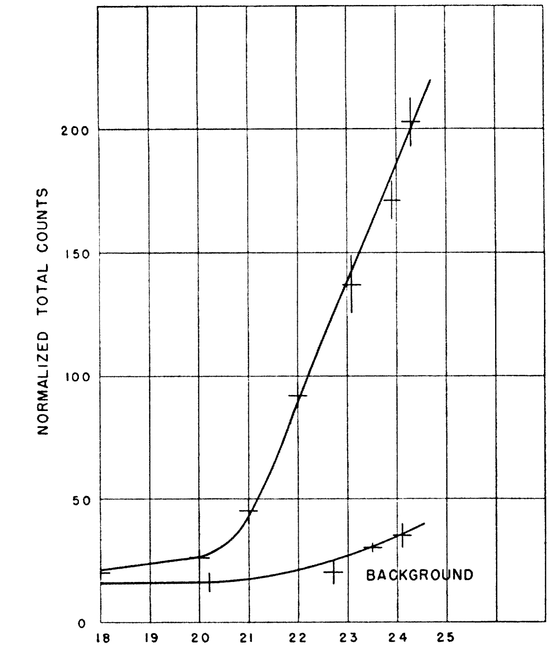

FIG. 2. Yield curve $C^{12}(\gamma,p)B^{11}$. The total counts at each energy are normalized to the same number of roentgens as measured by an ionization chamber.

for any given maximum energy is known.[1] Using these results and our yield curve, the excitation function shown in Fig. 3 is obtained.

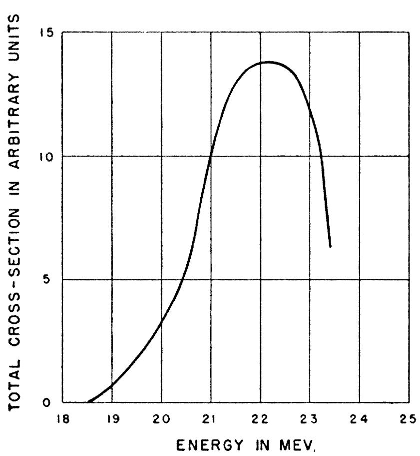

FIG. 3. Excitation function $C^{12}(\gamma,p)B^{11}$.

Since the yield curve is that of a thick (4.8-Mev) target, and since the protons must possess an energy of at least 2 Mev in order to reach the detector, the threshold of this excitation function is probably too high and its shape near the threshold is distorted. However, the threshold calculated from the mass values is 16 Mev. If the proton barrier and air absorption are taken into account, then the value of the threshold in Fig. 3 is within one Mev of the expected value. From the known geometry of the experimental arrangement and the effective target thickness, it is possible to assign an absolute value of 1×10^{-26} cm^2 to the cross section at the peak of the excitation function. It should be emphasized that this value has not been corrected for counter efficiency and target absorption, and therefore represents a lower limit of the actual value. We estimate that the upper limit does not exceed this value by more than a factor of five.

We are at present applying the techniques described here to a survey of the relative yields and angular distributions of photoprotons from various elements and are continuing the investigation of $\gamma-p$ excitation functions.

Each of us wishes to acknowledge a grant from the Committee on the Advancement of Research of the University of Pennsylvania.

* This work was supported in part by the joint program of the ONR and AEC.

[1] H. W. Koch and R. E. Carter, Phys. Rev. **77**, 165 (1950); R. N. H. Haslam *et al.*, Phys. Rev. **80**, 318 (1950). We are indebted to Professor Katz for a private communication.

The Isotopes of Xenon and Krypton in Pitchblende and the Spontaneous Fission of U^{238}

J. MACNAMARA AND H. G. THODE
Department of Chemistry, McMaster University, Hamilton, Ontario, Canada
August 31, 1950

It has been known for some time that the uranium nucleus undergoes spontaneous fission.[1,2] Recent values for the spontaneous fission half-life of uranium range from $(1.3\pm0.2)10^{16}$ to $(1.9\pm0.1)10^{15}$ years.[3–5] Since the spontaneous fission rate for U^{235} has been found to be approximately the same as that for natural uranium,[6] the spontaneous fission rate of natural uranium will be essentially that of the much more abundant isotope, U^{238}. The small concentration of plutonium in pitchblende[7] would seem to preclude the possibility of appreciable neutron fission of U^{235}.

As one would expect a similar distribution of fission fragments originating in the spontaneous fission of U^{238} as in the neutron fission of U^{235}, especially in the neighborhood of the 82-neutron shell, appreciable quantities of fission product krypton and xenon should be present in pitchblende. Since very little is known of the fission products from the spontaneous fission of uranium, experiments were designed to extract these rare gases with the hope that sufficient quantities could be obtained for mass spectrometer investigations. During the course of this work Khlopin, Gerling, and Baranovskaya[8] reported a much higher ratio of Xe to Kr in pitchblende than is normally found in the atmosphere, indicating the presence of appreciable quantities of fission product xenon.

The rare gases in a sample of pitchblende from Great Bear Lake, Canada, have now been extracted, purified, and analyzed with a mass spectrometer. The purified gas samples analyzed contained about 5×10^{-4} c.c. at N.T.P. of total xenon and krypton. Five fission product isotopes of xenon (Xe^{129}, Xe^{131}, Xe^{132}, Xe^{134}, Xe^{136}), and three of krypton (Kr^{83}, Kr^{84}, Kr^{86}) have been identified. The samples also contained some normal xenon and krypton as indicated by the presence of non-fission product isotopes in abundances proportional to their concentration in normal atmospheric gases.

Thus it was possible to determine with considerable accuracy the ratio of fission product to normal material. The ratios of fission product xenon to normal xenon and fission product krypton to normal krypton were 4:1 and 1:24, respectively.

The abundance data for the fission product isotopes of xenon and krypton are given in Table I.

TABLE I. The abundance data for the fission product isotopes of xenon and krypton from the spontaneous fission of U^{238}.

Xenon Mass	Xenon Yield[a] (percent)	Krypton Mass	Krypton Yield[b] (percent)
129	0.088 ±0.013	83	0.12 ±0.01
131	0.74 ±0.02	84	0.45 ±0.05
132	3.46 ±0.025	86	1.64 ±0.15
134	5.10 ±0.014		
136	6.00		

[a] Xe^{136} taken as 6.00 percent.
[b] Xe/Kr ratio taken as 7.0.

It is interesting to note that Xe^{129} is a product in the spontaneous fission of U^{238}. This is not surprising, since the half-life of I^{129} is estimated[9] to be 10^8 yr., or about 1/14 the age of the pitchblende sample.[10] By extracting the xenon from pitchblende of different ages and determining the abundance of Xe^{129}, relative to another stable isotope of xenon, it is hoped to obtain a more accurate half-life for I^{129}.

Figure 1 is a mass fission yield curve obtained by plotting the data of Table I. The curve will vary slightly depending on the yield value assigned to Xe^{136} and to the ratio of xenon to krypton formed in the fission process. The ratio of xenon to krypton of 7.0 used in the calculations was determined by direct mass spectrometer measurements and is a preliminary value. The results indicate an asymmetrical mass fission yield curve similar to that obtained in the fission of U^{234} and U^{236} with maxima at approximately 95 and 140 in agreement with the results of Whitehouse and Galbraith.[11] The higher ratio of Xe to Kr found, however. would indicate a shifting of the whole mass fission yield curve toward the heavier masses which would be expected for the spontaneous fission of U^{238}. The relatively high yield of Xe^{132} is of particular interest. It can be seen from Figs. 1 and 2 that the fission yield of this isotope is about 65 percent above the smooth curve drawn through yield values for the other xenon isotopes. Since the relative precision of these yield values is better than one percent (except for Xe^{129}), it is clear that Xe^{132} has an abnormally high yield.

The fine structure previously reported in the mass fission yield curve for U^{236} fission[12] appears also in the neighborhood of nuclides with 82 neutrons, but the high yields occur at Xe^{133} and Xe^{134}, not Xe^{132}. This shifting of the fine structure to the lower masses would be expected, if we accepted the hypothesis of Glendenin[13] to account for these abnormal yields.

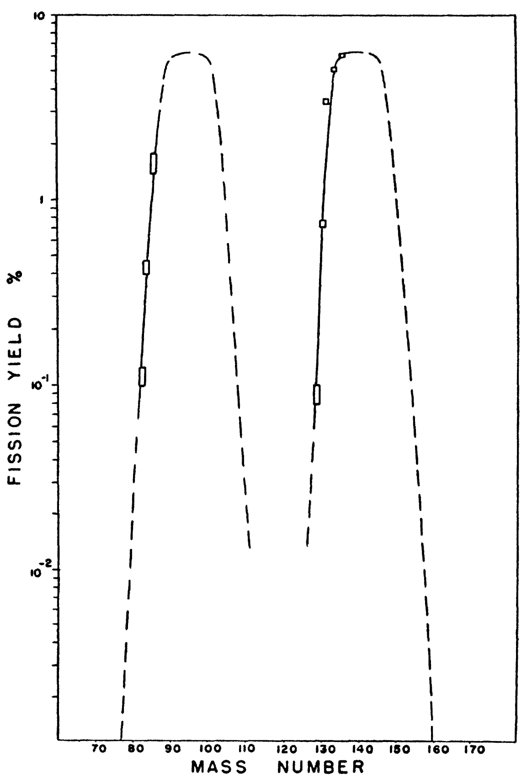

FIG. 1. Mass fission yield curve for spontaneous fission of U^{238}.

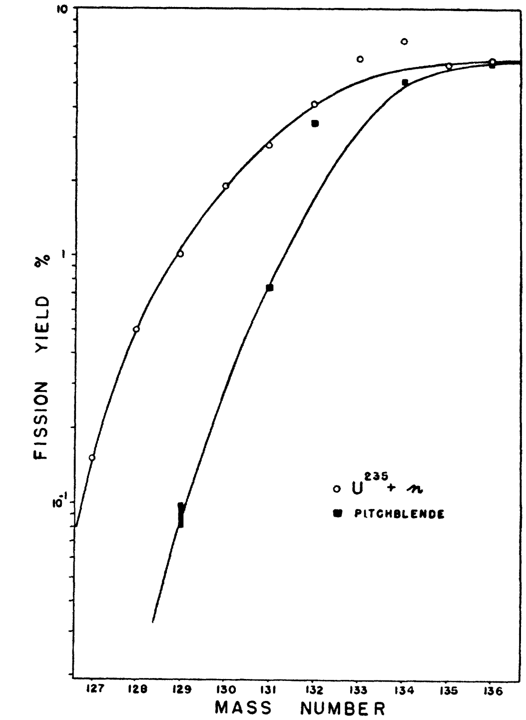

FIG. 2. Mass fission yield curve for the fission of U^{236} and U^{238} showing fine structure.

A careful investigation of the relative abundances of the argon isotopes occluded in the pitchblende sample showed less than a three percent variation in the argon 40 to argon 36 ratio when compared with atmospheric argon. This would seem to indicate that there is little fractionation of the xenon and krypton isotopes due to different rates of diffusion out of the mineral.

Experiments are in progress to determine actual volumes of fission product xenon and krypton in pitchblende samples of different ages. These determinations will yield an accurate ratio of Xe to Kr and will make possible estimates of the total amount of fission per gram of uranium for geophysical age studies. Also, similar studies are under way with thorium minerals to determine the extent of spontaneous fission of thorium.

The authors wish to thank M. Dubeck for his assistance in preparing the samples and R. F. Errington and the Eldorado Mining and Refining Company, Limited for the samples of pitchblende. We also wish to acknowledge the financial assistance from the National Research Council of Canada.

[1] K. A. Petrzhak and G. N. Flerov, J. Phys. U.S.S.R. **3**, 275 (1940); Chem. Abs. **35**, 4677[1] (1941).
[2] G. N. Flerov and K. A. Petrzhak, Phys. Rev. **58**, 89 (1940).
[3] N. A. Perfilov, J. Phys. U.S.S.R. **11**, No. 3 (1947), Chem. Abs. **42**, 2864 (b) (1948).
[4] H. Yagoda and N. Kaplan, Phys. Rev. **76**, 702 (1949).
[5] W. Maurer and H. Pose, Zeits. f. Physik **121**, 285 (1943).
[6] R. R. Williams, *Principles of Nuclear Chemistry* (D. Van Nostrand and Company, Inc., New York, 1950), p. 27.
[7] Peppard, Studier, Gergel, Mason, Sullivan, and Mech, paper presented at 118th Meeting of American Chemical Society, Chicago, September 1950.
[8] Khlopin, Gerling, and Baranovskaya, Bull. Acad. Sci. U.R.S.S. Classe. Sci. Chim. 599-604, 1947; Chem. Abs. **42**, 3664[a], (1948).
[9] S. Katcoff, Phys. Rev. **71**, 826 (1947).
[10] A. O. Nier, Phys. Rev. **55**, 153 (1939).
[11] W. G. Whitehouse and W. Galbraith, Phil. Mag. **41** (May, 1950).
[12] Macnamara, Collins, and Thode, Phys. Rev. **78**, 129 (1950).
[13] L. E. Glendenin, Phys. Rev. **75**, 337 (1949).