that Pb has relatively few levels so that the statistical theory might not be applicable, just as in the case of Bi.

Two of the elements investigated, Zr and Pb, show deviations from a smooth variation of the cross section with energy. These anomalies were quite reproducible and are shown in two typical runs in Figs. 1 and 7. It is somewhat surprising to find structure in heavy elements, considering the poor resolution used. Evidence for such a structure in Pb had previously been found by Bretscher and Murrell.[7] In view of the fact that the anomalies have the shape of dips rather than peaks, one might suspect that they could be caused by inelastic scattering. As the energy of the primary neutrons becomes just sufficient to produce the final nucleus in an excited state, low energy neutrons will be emitted from the scatterer, and the detector used will detect these neutrons with high efficiency. This effect would result in a decrease of the observed cross section. It does not appear likely, however, that this explanation could account for the observations. It is known[8] that at these energies the cross section for inelastic scattering, at least in the case of Pb, is very small (not more than 0.3 barn), and only three percent of the scattered neutrons will reach the detector in the geometry used. Furthermore, because of the wide spread in energy of the primary neutrons, only a very small fraction of the scattered neutrons will have energies in the range in which the sensitivity of the counter is very high.

TABLE I. Samples used in transmission experiments.

Element	Atomic weight	Thickness (cm)	Number of atoms/cm^2 $\times 10^{-24}$
Zr	91	1.27	0.0551
Ag	108	1.27	0.0746
In	115	1.25	0.0471
In	115	2.42	0.0923
Sb	122	3.18	0.0657
I	127	3.18	0.0723
Ta	181	1.27	0.0705
Pb	207	1.90	0.0630
Pb	207	3.18	0.1081

It is interesting to note that the most abundant isotopes of both Zr and Pb have closed neutron shells[9] (50 and 126 neutrons, respectively), and might, therefore, be expected to have broad and widely spaced levels. It is planned to investigate, with better resolving power, the energy regions in which the anomalies were observed.

The authors wish to thank Mr. R. L. Henkel for assistance in the experiments. This work was supported by the Wisconsin Alumni Research Foundation and by the AEC.

[7] Bretscher and Murrell, quoted Goldsmith, Ibser, and Feld, Rev. Mod. Phys. **19**, 259 (1947).

[8] Barschall, Battat, Bright, Graves, Jorgensen, and Manley, Phys. Rev. **72**, 881 (1947).

[9] M. G. Mayer, Phys. Rev. **74**, 235 (1948).

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Measurements on Radioactive Krypton Isotopes from Fission after Mass-Spectrographic Separation

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Radioactive isotopes of krypton resulting from fission of uranium have been separated in a mass-spectrograph. The half-lives and the relative fission yields of the isotopes Kr^{85}, Kr^{87}, and Kr^{88} have been measured. The maximum energies of the β-particles from Kr^{85} and Kr^{87} have been checked by absorption. It has been shown that Kr^{88} emits soft β-particles and an intense γ-radiation. This result combined with other measurements indicates that the β-spectrum of Kr^{88} is complex.

INTRODUCTION

AMONG the radioactive nuclei formed in neutron-induced fission of uranium there are many krypton isotopes.[1,2] By using mixtures of these isotopes several radioactive periods have been isolated, either from the analysis of complex decay curves[3] or by the application of less direct methods.[4] The mass-numbers of the radioactive isotopes could be determined with considerable certainty from such experiments.

The present note describes experiments which were carried out with some long-lived krypton isotopes after they had been separated by means of the mass-spectrograph of this Institute.[5] This procedure is a crucial test of the assignment of activities to mass-numbers and,

[1] Plutonium Project, Rev. Mod. Phys. **18**, 513 (1946).

[2] G. T. Seaborg and J. Perlman, Rev. Mod. Phys. **20**, 585 (1948).

[3] A. H. Snell, Phys. Rev. **52**, 1007 (1937). W. Seelmann-Eggebert and H. J. Born, Naturwiss. **31**, 59 (1943). E. P. Clancy, Phys. Rev. **60**, 87 (1941).

[4] G. N. Glasoe and J. Steigman, Phys. Rev. **58**, 1 (1940).

[5] J. Koch and B. Bendt-Nielsen, Kgl. Danske Vid. Sels. Mathfys. Medd. **21**, No. 8 (1944). J. Koch, Phys. Rev. **69**, 238 (1946).

TABLE I. Half-lives of krypton isotopes. (a) Measurements with pure isotopes. (b) Measurements according to reference 2.

Isotope	Half-life (a)	Half-life (b)
Kr^{88}	2.77 hr.	2.8 hr.
Kr^{87}	1.30 hr.	1.25 hr.
Kr^{85}	4.36 hr.	(4–4.6) hr.

moreover, it offers the possibility of determining the half-lives and relative fission yields with some precision. Due to the strong activity of each isotopic sample this technique, in principle, also permits a closer study of the β-spectrum of the different krypton isotopes, but in lack of a β-spectrograph only the maximum energy of the β-particles could be checked by absorption.

Our interest has been especially devoted to the question whether γ-rays are emitted in the decay of Kr^{88}. The closer knowledge of the decay scheme of this isotope is in itself of course of some interest. It is, however, of primary importance to the interpretation of the recoil experiments with Kr^{88}, carried out by Jacobsen and Kofoed-Hansen.[6] The consequences which may be drawn from such experiments regarding the type of the coupling in β-decay have been discussed in more detail elsewhere.[7]

EXPERIMENTAL PROCEDURE

Ten kilograms of powdered uranium oxide, mixed with a small portion of ammonium carbonate, were placed in a glass container which was surrounded by a few centimetres of paraffin. After having evacuated the container, the tube leading to the pumps was closed by turning a stopcock. The uranium container was inserted between the coils of the cyclotron magnet, as close to the $(D+Be)$ neutron source as possible, and then heavily irradiated for about three hours. The inert gases emanating from the uranium compound were pumped off by means of a Toepler pump, using the gases given off by the ammonium carbonate (NH_3, CO_2, and H_2O) as a carrier of the activity, and concentrated in a volume of about 100 cm^3. After having added a great amount of normal krypton, the gas mixture was introduced to the capillary ion source of the mass-spectrograph. These operations were carried out quickly, so that the separation of the isotopes could be started about half an hour after stopping the cyclotron. The technique of collecting gaseous isotopes by bombarding a metal with energetic ions has already been described elsewhere.[8] In the present case, the collector was built of a stack of brass plates, which were mounted as shown in Fig. 1, and which divided the mass range in question into fifty steps. By means of this arrangement it was possible after mass-spectrographic separation to investigate the radiation from the individual parts of the collector without being disturbed by any radiation originating from active atoms deposited in the neighborhood. The use of a fluorescent screen of crown glass, mounted on top of the collector, permitted the adjustment of the intensity and resolving power of the mass-spectrum of the stable krypton isotopes before operation and the observation of the beams during the time of collection. While the isotopes were being collected, either Kr^{82}, Kr^{83}, or Kr^{84} was focused on a wire. In front of the collector a shutter was inserted, which was opened magnetically as soon as good operating conditions were established. After running the mass-spectrograph for about one hour, the collector was quickly removed from the apparatus in order to study the active deposits.

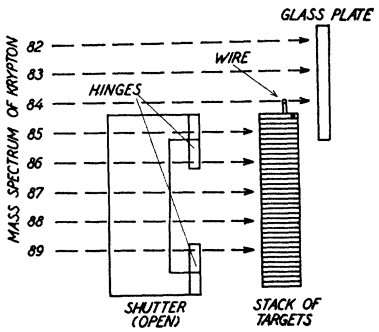

FIG. 1. Detail of mass-spectrograph. Arrangement for the collection of radioactive krypton isotopes. Thickness of each target 1.5 mm.

RESULTS

Mass Determination and Relative Fission Yields

The distribution of radioactivity along the stack of collector plates is plotted in Fig. 2, showing three clearly resolved peaks. From visible marks on the collector plates, due to the sputtering action of the beams of stable krypton isotopes, these peaks can be identified to belong to Kr^{85}, Kr^{87}, and Kr^{88}. The figures of the diagram have been corrected for radioactive decay

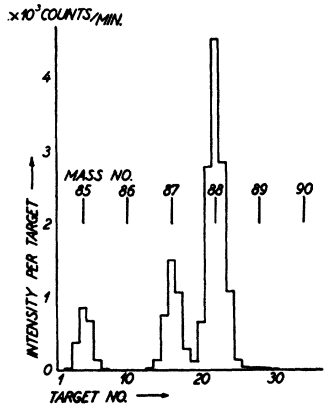

FIG. 2. Mass-spectrum of some radioactive krypton isotopes.

[6] J. C. Jacobsen and O. Kofoed-Hansen, Phys. Rev. **73**, 675 (1948).

[7] O. Kofoed-Hansen, Phys. Rev. **74**, 1785 (1948).

[8] J. Koch, Nature **161**, 566 (1948).

during the time of measurement, and they refer to the situation 2.5 hr. after stopping the neutron bombardment from the cyclotron. The dispersion per mass unit is found to be about 9 mm and the breadth of the peaks about 3 mm.[9] This breadth is somewhat smaller for Kr^{85} than for Kr^{88} due to a slight change in the focusing properties of the mass-spectrograph along the mass scale.

Because of the relatively long time interval between the termination of the neutron irradiation and the radioactive measurements, there was no possibility of finding the short-lived Kr^{89} (2.5 m) or its successor Rb^{89} (15.4 m). The faint activity ($\sim$1 percent of Kr^{88}) at the mass number 89 probably originates from $(Kr^{88}H)^+$ ions, which may be formed in the ion source due to the presence of hydrogen in the gas mixture. A search for the 2-hr. isomer of mass number 83 also gave a negative result. This, however, is in agreement with the facts that the number of atoms of this isomer, formed in fission, is relatively very small, and that the radiation mainly consists of γ-rays which have only a low efficiency for being detected by the counter. Also the 10-yr. Kr^{85} isomer could not be observed here.

The half-lives of the isotopes Kr^{85}, Kr^{87}, and Kr^{88} were determined by following the decrease in activity of the strongest isotopic samples for about 24 hr. These decay curves are shown in Fig. 3 and the resulting half-lives are listed in Table I together with the hitherto accepted values. The accuracy here obtained is estimated to be about 2 percent.

The calculation of the total number of atoms of the different krypton isotopes decaying per unit time (see Fig. 2) makes it possible to determine the course of the curve of fission yields in the mass region in question. It is well known that the constant production of a radioactive substance during the time T, followed by a decay during the time t, leads to a number of atoms of the substance N which is given by

$$N=(y/\lambda)[1-\exp(-\lambda T)]\exp(-\lambda t), \qquad (1)$$

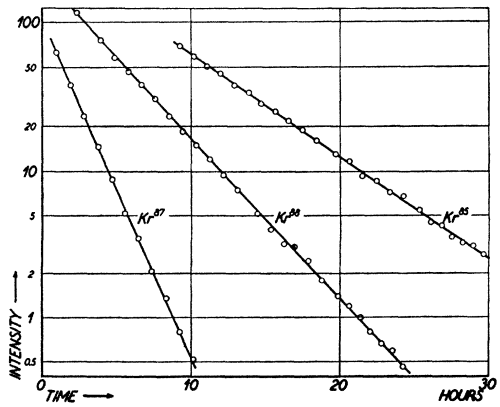

FIG. 3. Decay curves of Kr^{88}, Kr^{87}, and Kr^{85}.

[9] The breadth is defined as twice the standard deviation when approximating the measured curve by a Gaussian distribution.

TABLE II. Fission yields in relation to the production of Kr^{88}.

Isotope	y_{rel} (measured)	y_{rel} (by interpolation)
Kr^{88}	100	100
Kr^{87}	70±15	78
Kr^{85}	33±10	44

where y is the yield of production per unit time, and λ is the decay constant. The application of this formula for the determination of the relative fission yields (y_{rel}) of the isotopes in question is permitted, since all the predecessors are short-lived ($\lesssim$3 m).[10] In the case of Kr^{88} it must, however, be remembered that this isotope is in equilibrium with its successor Rb^{88}, having a half-life of 17.8 m. For Kr^{88} the expression (1) must therefore be multiplied by a factor

$$1+\lambda_{Rb}/(\lambda_{Rb}-\lambda_{Kr}). \qquad (2)$$

The average values of y_{rel} for two series of measurements are given in Table II and compared with values taken from the course of the known fission yield curve (see reference 1). The agreement is satisfactory, considering the presence of irregularities during the time of irradiation with neutrons, from which the large error mainly arises. The rather low value listed for Kr^{85} is partly due to the fact that the contribution of the long-lived Kr^{85} isomer has been neglected.

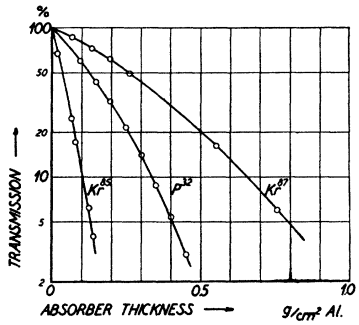

FIG. 4. Absorption curves of Kr^{87}, Kr^{85}, and P^{32}. The curves have been corrected for the thickness of the counter window.

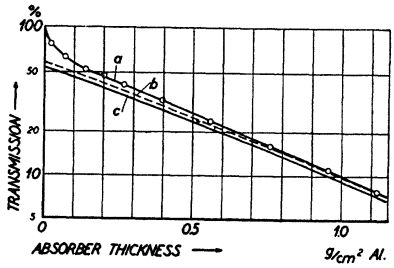

FIG. 5. *a*, Absorption curve for Kr^{88}+Rb^{88} in equilibrium. *b*, Absorption curve for Rb^{88}, normalized so as to fit curve *a* for thick absorbers. *c*, Absorption curve for Rb^{88}, representing the contribution from this isotope to the course of curve *a*.

[10] N. Sugarman, J. Chem. Phys. 17, 11 (1949).

TABLE III. Maximum energies of β-rays from Kr^{87} and Kr^{85}.

Isotope	Max. energy in Mev (measured)	Max. energy in Mev (reference 2)
Kr^{87}	3.2 ±0.3	4
Kr^{85}	0.75±0.1	0.85−0.94

Beta-Ray Energies

The maximum energies for the β-rays from Kr^{85} and Kr^{87} were simply estimated by measuring the absorption in aluminum, neglecting the existence of γ-rays from these isotopes, which give rise to only a small error. These absorption curves are shown in Fig. 4, together with an absorption curve for P^{32}, which was taken for comparison. The unknown ranges of the β-particles from the two krypton isotopes were determined relative to the range of the β-particles from P^{32}. The corresponding β-energies, which are listed in Table III together with former values, were derived from Feather's formula.

The absorption curve for the radiations from the mixture of Kr^{88} and Rb^{88} in equilibrium has also been measured and has been compared with the absorption curve for the radiations from pure Rb^{88} (Fig. 5, curves *a* and *b*). For this purpose, samples of Rb^{88} were collected on a set of collector plates, which were mounted in a bulb containing radioactive krypton, and which was connected to a negative potential.[4,6] In carrying out the absorption experiments, special precaution was taken to ensure the same geometrical arrangement. An analysis of the absorption curve *a* clearly shows that the main number of Kr^{88} atoms decay with a maximum energy of about 0.5 Mev. The maximum energy of the only hitherto known group of β-particles from Kr^{88} is reported to be 2.43 Mev.[6,11] The small difference in the slope of the curves *a* and *b* for absorbers of thicknesses larger than the range of the low energy group is probably due to the presence of the group of 2.43 Mev. From the present experiments, however, it cannot definitely be concluded that this group exists; but as far as one may rely on the conclusion from the above mentioned measurements on the existence of the 2.43-Mev group, it is seen that the decay scheme of Kr^{88} is complex.

Gamma-Rays from Kr^{88}

The demonstrated group of short range β-particles from the decay of Kr^{88} indicates the existence of a γ-ray group having an energy of about 2 Mev. A special experiment was designed with the purpose of finding this radiation. Due to the disturbing effect of the γ-radiation from Rb^{88}, which is in equilibrium with Kr^{88}, it was necessary to liberate the krypton from the samples collected in the mass-spectrograph. This was carried out in vacuum by means of induction heating, with the result that not only the krypton was given off, but also a great amount of other gases being occluded

[11] G. L. Weil, Phys. Rev. **62**, 229 (1942).

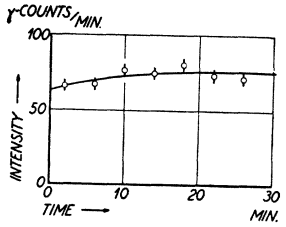

FIG. 6. Time dependence of gamma-ray intensity from a sample which at time zero consists of pure Kr^{88}.

in the brass collectors. Using these gases as carrier of the activity, the gas mixture was first compressed by means of a Toepler pump, and then transferred to a small glass vessel filled with mercury and having the bottom turned upwards. A counter was mounted above the vessel and lead absorbers were interposed in order to eliminate β-rays.

Under these conditions, the curve of Fig. 6 was measured, showing the change of γ-rays intensity with time. The extrapolation of this curve to time zero, which corresponds to the time of inlet of active gas into the vessel, clearly shows the presence of γ-rays which must originate from pure Kr^{88}. The further course of the curve is governed by the decay of Kr^{88}, combined with the production and decay of the γ-active isotope Rb^{88}. The full-drawn curve in the figure corresponds to the expression

$$a\cdot\exp(-\lambda_{Kr}\cdot t)-b\cdot\exp(-\lambda_{Rb}\cdot t), \qquad (3)$$

where $a=109$ and $b=47$. The measurements of Fig. 6 could be repeated several times by replacing the glass vessel by another one. The active gas mixture was sucked over into the Toepler pump during this procedure.

The presence of an intense γ-radiation from Kr^{88} implies that the absorption curve for the mixture of Kr^{88} and Rb^{88} (see Fig. 5*a*) must be somewhat above the absorption curve for pure Rb^{88} in the case of very thick absorbers. The more exact position of this Rb^{88} curve, in relation to the curve *a*, can be determined by applying considerations leading to the expression (2). Curve *c* in Fig. 5 is constructed on this basis. Any further conclusions which may be drawn from the relative adjustment of the curves *a* and *c* must, however, be taken with considerable reservation, since we have no evidence that the efficiency of the counter is the same for low as for high energy β-particles.

This fact and the above-mentioned possibility of a complex β-decay of Kr^{88} show that any closer interpretation of the recoil experiments with Kr^{88}, which were mentioned in the Introduction, must be postponed at the present time. The further study of the decay scheme of Kr^{88} is, therefore, highly desirable.

We wish to thank Professor Niels Bohr for his never-failing interest and encouragement. We also wish to express our gratitude to Professor J. C. Jacobsen for valuable discussions. Furthermore, our thanks are due Mr. B. Madsen for putting a counter equipment at our disposal, Mr. K. O. Nielsen and Mr. S. Møller-Holst for their help in the work with the cyclotron and the mass-spectrograph, respectively.