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RADIATIONS FROM SHORT-LIVED RARE GAS FISSION PRODUCTS*†

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Abstract—The energies of the primary γ-rays emitted in the decay of 3·2 min ^{89}Kr, 33 sec ^{90}Kr, 1·2 min ^{91m}Rb, 41 sec ^{139}Xe, and 66 sec ^{140}Cs were determined and relative γ-ray intensities measured. Photon per disintegration values were estimated for ^{89}Kr, ^{90}Kr, ^{139}Xe and for 9·5 min ^{139}Cs. β-ray energy endpoints were measured for ^{89}Kr, ^{90}Kr, ^{139}Xe and ^{139}Cs, and lower limits established for the decay energies of these nuclides. A simple and rapid gas chromatographic separation which was developed for these studies is described.

A SYSTEMATIC study of the half-lives of the short-lived ($T_{\frac{1}{2}} < 5$ min) gaseous fission products was made by DILLARD *et al.*,[1] utilizing the collection of the daughter activities on a charged wire in a gas flow tube, with subsequent radiochemical analysis to determine the distribution along the length of the wire. They obtained half-lives for the krypton isotopes of mass numbers 89, 91, 92, 93, 94, 95 and 97, and for the xenon isotopes of mass numbers 139, 140, 141, 143 and 144.

Half-life measurements and β-ray endpoint determinations for the krypton and rubidium isotopes of mass numbers 89, 90 and 91 were made by KOFOED-HANSEN and NIELSEN[2] by means of an isotope separator. They also reported the detection of gamma radiations from ^{90}Rb, ^{91}Rb and ^{91m}Rb.

The nuclear characteristics reported in the literature for the nuclides of interest in the present study are given in Table 1.

In the present work, the radiations of several gaseous fission products and daughters with half-lives in the range of 30 sec to several minutes have been studied by γ- and β-ray spectrometry. These data are of considerable interest in the study of the fission process, in nuclear level systematics, and in reactor shielding calculations.

EXPERIMENTAL

Irradiation and sampling

The gas samples were obtained by irradiation of ~50 mg samples of uranyl nitrate for 10–20 sec in the pneumatic tube facility[8,9] of the 1 MW Ford Nuclear Reactor at the University of Michigan.

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† Based in part on a dissertation by M. A. WAHLGREN, submitted to the University of Michigan in partial fulfillment for the Ph.D. degree in Chemistry.

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[1] C. R. DILLARD, R. M. ADAMS, H. FINSTON and A. TURKEVICH, in Coryell and Sugarman, *Radiochemical Studies: The Fission Products* (Edited by C. D. CORYELL and N. SUGARMAN) NNES Plutonium Project Record p. 624. McGraw-Hill, New York (1951).

[2] O. KOFOED-HANSEN and K. O. NIELSEN, *Phys. Rev.* **82,** 96 (1951).

[8] W. W. MEINKE, *Nucleonics* **17,** No. 9, 86 (1959).

[9] The instrumentation, techniques, and experimental results are described in considerably more detail in the Ph.D. thesis of M. WAHLGREN, University of Michigan, 1961 (unpublished); and U.S. Atomic Energy Commission Report TID-11807 (1961).

TABLE 1.—SUMMARY OF PERTINENT NUCLEAR DATA FROM THE LITERATURE[3,4]

Nuclide	Half-life	E_γ (MeV)	E_β −(MeV)
^{89}Kr	3·2, 2·6 or 2·5 min	—	4·0, 3·9
^{90}Kr	33 sec	—	3·2
^{91}Kr	9·8, 10 or 6 sec	—	3·6
^{89}Rb	15·4, 14·9 or 15·5 min	Several[6]	3·9, 4·5, 3·8
^{90}Rb	2·7 min	Several[6]	5·7
^{91m}Rb	1·67 or 1·2 min[9]	Detected	4·6
^{139}Xe	41 sec	—	—
^{140}Xe	16 or 9·8 sec	—	—
^{139}Cs	9·5, 10 or 7 min	0·630, 1·270[7]	—
^{140}Cs	66 sec	—	—
^{139}Ba	84 or 86 min	0·163, 1·43	2·4, 2·2, 0·8

[3] D. STROMINGER, J. M. HOLLANDER and G. T. SEABORG, *Rev. Mod. Phys.* **30**, 585–904 (1958).
[4] K. WAY (Editor) Nuclear Data Group, *Nuclear Data Sheets*, National Academy of Sciences, National Research Council, Washington.
[5] A. C. WAHL, R. L. FERGUSON, D. R. NETHAWAY, D. E. TROUTNER and K. WOLFSBERG, U.S.A.E.C. Report TID-13127 (1961).
[6] G. D. O'KELLEY, E. EICHLER and N. R. JOHNSON, Paper P/672, Vol. 15, *Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva*, 1958. Paper P/672 Vol. 15 United Nations (1959).
[7] J. WASSON, MIT Laboratory for Nuclear Science, Progress Report, May 31, 1958 (unpublished).

Sample transfer from the irradiation position to a hood in the chemistry laboratory required three seconds.

The inert fission gases which separate from the gross fission products by emanation[10] were drawn from the polyethylene irradiation capsule with the sampling device shown in Fig. 1. Although the

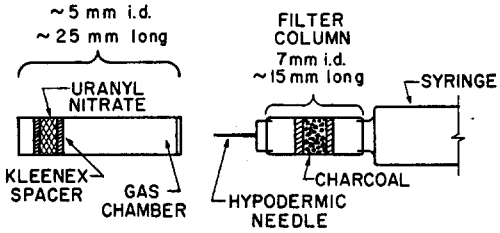

FIG. 1.—System for sampling fission gas.

emanation from organic uranium salts has been shown to be more quantitative than from the nitrate, the small sample size possible with the nitrate is important in this case because of experimental conditions.

Preliminary experiments established that the differences in adsorptivity and in effusion rates through the pores of the charcoal were sufficient to accomplish the separation of the submicro amounts of gas into kyrpton and xenon fractions.[9] The krypton was drawn through the column into the syringe while the xenon was retained in the charcoal column. With this system the gas sample could be drawn and separated within 15 sec after the end of irradiation.

The xenon release from a typical column (3–4 mm thick and 7 mm in diameter) left unsealed was studied with 9·1 hr ^{135}Xe tracer. At 25°C and atmospheric pressure the xenon activity decreased by a factor of two each 5 min. Abrupt reduction of pressure by as much as a factor of two across the column in the sampling process was found to lead to carry-through of xenon with the krypton fraction. Pressure reduction sufficient to give a flow rate of carrier gas (air) of 0·1 cm^3 sec^{-1} yielded quantitative separation under the conditions of these experiments, but limited the minimum sampling time to ~5 sec.

By variation of irradiation time, decay time, and by passing the residual gas through paper filters, it was possible to obtain spectra of the various daughter activities as well as those of the longer-lived

[10] A. C. WAHL and N. A. BONNER, *Radioactivity Applied to Chemistry*, Chap. 9. J. Wiley, New York (1951).

gaseous nuclides. These spectra are necessary for the analysis of the complex spectra obtained with the short-lived gases.

Instrumentation and counting techniques

The dual memory 100-channel pulse-height analyser with associated rapid transfer equipment and detectors used in these studies has been described previously.[8,9] The handling and transfer time from hood to counter (from the midpoint of the sampling period to the start of counting) was 12 sec for γ-ray counting and 24 sec for β-ray counting samples.

All γ-ray spectra were taken at a standard source distance of 10 cm from the 3 in $\times$ 3 in. NaI(Tl) crystal using a 2·5 g cm^{-2} Be absorber, with the exception of ^{91m}Rb which was run at 6 cm due to the low counting rate. Sample size was chosen to give at least 75 per cent analyser "live time" to minimize spectral distortion. The resolution of the 3 in. $\times$ 3 in. crystal was 7·3 per cent at 0·662 MeV.

The analyser was recycled at 0·5, 5 and 50 min intervals during the decay of the short-lived parent, the daughter, and the longer-lived components respectively. Semi-logarithmic plots of these data readily show the parent-daughter relationships.

For γ-ray spectra of the short-lived xenon fraction the sample was drawn, the charcoal column sealed into a polyethylene "rabbit", and sent by a secondary pneumatic loop to the counting position at the γ-detecting crystal. For the krypton fraction a special polyethylene syringe was used which could be sealed and utilized as a rabbit for transfer to the counter.[9]

A 1 in $\times$ 1½ in. right-cylindrical plastic phosphor was used to obtain β-ray spectra. Although subject to distortion from the ampoules, Compton recoil electrons and the general multi-nuclide background, the spectra allow an approximate measurement of the maximum or predominant β-ray endpoint observed in the decay of the short-lived fractions. Energy calibration was based on the known endpoints of ^{89}Rb (3·9 MeV) and ^{138}Cs (3·4 MeV) daughter activities present in the samples. The contribution of Compton electrons was estimated by comparison with a set of similar samples counted through a 1 g cm^{-2} Be absorber. The calibration was checked with ^{90}Sr – ^{90}Y and ^{38}Cl sources. Due to the difficulty of reproducing the exact geometry, scattering and absorption of the experimental samples, it was felt that the most reliable calibration was that obtained from known daughter activities in the same samples. The reproducibility of the endpoints measured by this technique is $\pm$0·2 MeV.

Analysis of data

Because of the complexity and possible ambiguity of the β-ray spectra, the quantitative results quoted have been derived from analysis of the gamma-ray spectra in the manner of O'KELLEY *et al.*[6] The pulse height distribution parameters of resolution, Compton continuum, Compton edge, and escape peaks were established from spectra of thirteen nuclides giving essentially monoenergetic γ-ray spectra, for the energy range from 0·191 to 4·1 MeV. From these parameters additional pulse height distributions were synthesized where necessary to provide a reference spectrum at each 0·3 MeV interval in the range from 1 to 4·5 MeV.

Corrections were made where appropriate for the analyser "dead time" and for the time of count, if long compared to the half-life. Subtraction of the contribution of the other nuclides observed in the decay of the sample was then performed if the contribution to the first spectra was significant. The subtraction of each successive gamma ray followed fitting of the top and leading edge from the appropriate reference pulse height distribution. In several cases it was not possible to begin with the highest energy peak observed because of poor statistics. In these cases the first clearly defined peak in order of decreasing energy was used as a starting point.

For the calculation of relative intensities, peak efficiencies based on the data of HEATH[11] and of WOLICKI *et al.*[12] were used. Correction was made for attenuation by the β-absorber based on experimental absorption measurements.

The photon per disintegration rates were derived from analysis of the spectra of the appropriate daughter activity. For this determination neither absolute counting nor intercalibration of counters is required. The geometry of the source is the same for both parent and daughter, and the relative peak efficiencies are well known. The accuracy of the calculation is determined by the accuracy of the half-lives assumed for the calculation, the error in γ-ray spectral analysis, and the validity of the literature values for the decay schemes of the respective daughter activities.

Each of the experiments described in the following section was repeated at least three times. The relative abundances were reproducible to within 5 per cent, and the photon per disintegration values to within 10 per cent, for the primary γ-rays. For precise analysis the coincidence summing should be

[11] R. L. HEATH, U.S.A.E.C. Report IDO-16408 (1957).
[12] E. A. WOLICKI, R. JASTROW and F. BROOKS, Naval Research Laboratory Report NRL-4833 (1956).

TABLE 2.—SUMMARY OF EXPERIMENTAL DATA

Nuclide	Half-life (observed)	Irradiation time (sec)	Cooling time (sec)	γ Energy (MeV)	Relative abundance	Photon/disintegration	β Energy (MeV)	Decay Energy (MeV) (Estimated)	Fig. No.
^{89}Kr	$3{\cdot}2 \pm 0{\cdot}3$ (min)	15	240	0·215	80	0·31*	4·0	4·6	2,3
				0·375	35	0·13			
				0·430	33	0·13			
				0·510	49	0·19			
				0·595	100	0·39			
				0·730	39	0·15			
				0·880	58	0·23			
				1·12	50	0·19			
				1·32	34	0·13			
				1·52	70	0·27			
				1·67	31	0·12			
				1·90	16	0·061			
				2·05	26	0·10			
				2·23	14	0·054			
				2·35	25	0·099			
				2·62	8·0	0·031			
				2·70	5·7	0·022			
				2·85	18	0·071			
				3·00	8·9	0·035			
				3·18	11	0·043			
				3·38	15	0·060			
				3·56	11	0·043			
				3·72	11	0·043			
				3·90	9·6	0·037			
				4·05	3·4	0·014			
				4·16	2·7	0·010			
				4·31	2·7	0·010			
				4·58	1·1	0·005			

^{90}Kr	33 ± 2 sec	10	12–30	0·125	100	0·67†	1·8	3·6	4
				0·240	10	0·07			
				0·540	100	0·67			
				1·10	100	0·67			
				1·54	42	0·28			
				1·78	19	0·12			
^{91m}Rb	1·2 ± 0·1 min	20	30	0·095	100	—	—	—	5
				0·350	47				
^{139}Xe	41 ± 2 sec	15	30	0·170	40	0·20‡	4·6	4·6	6
				0·220	100	0·49			
				0·295	53	0·26			
				0·395	21	0·10			
^{139}Cs	9·5 min§	20	100	1·270	100	0·12‡	3·8	3·8	7
				0·630	18	0·02			
^{140}Cs	66 ± 2 sec	15	40	0·610	—	1·0 (estimated)	—	—	8

* Assuming 1250 keV photons in 54 per cent of the disintegrations[6] of ^{89}Rb, and half-lives of 3·2 min and 14·9 min for ^{89}Kr and ^{89}Rb, respectively.

† Assuming 840 keV photons in 61 per cent of the disintegrations[6] of ^{90}Rb, and half-lives of 33 sec and 2·7 min for ^{90}Kr and ^{90}Rb, respectively.

‡ Assuming 165 keV photons in 23 per cent of the disintegrations[13] of ^{139}Ba, and half-lives of 41 sec, 9·5 min, and 85 min for ^{139}Xe, ^{139}Cs and ^{139}Ba, respectively.

§ Accuracy is less than for other nuclides because of an interfering peak at about the same energy.

[13] R. L. MACKLIN. N. H. LAZAR and W. S. LYON, *Phys. Rev.* **107**, 504 (1957).

taken into account. This effect was checked experimentally by comparison of spectra taken at 1 and at 10 cm. The differences, however, were within the experimental error and have been ignored in the data summation which follows.

EXPERIMENTAL RESULTS

The experimental data are summarized in Table 2. A brief discussion of the experiments and the γ-ray spectra obtained are given in the following section. Due to the

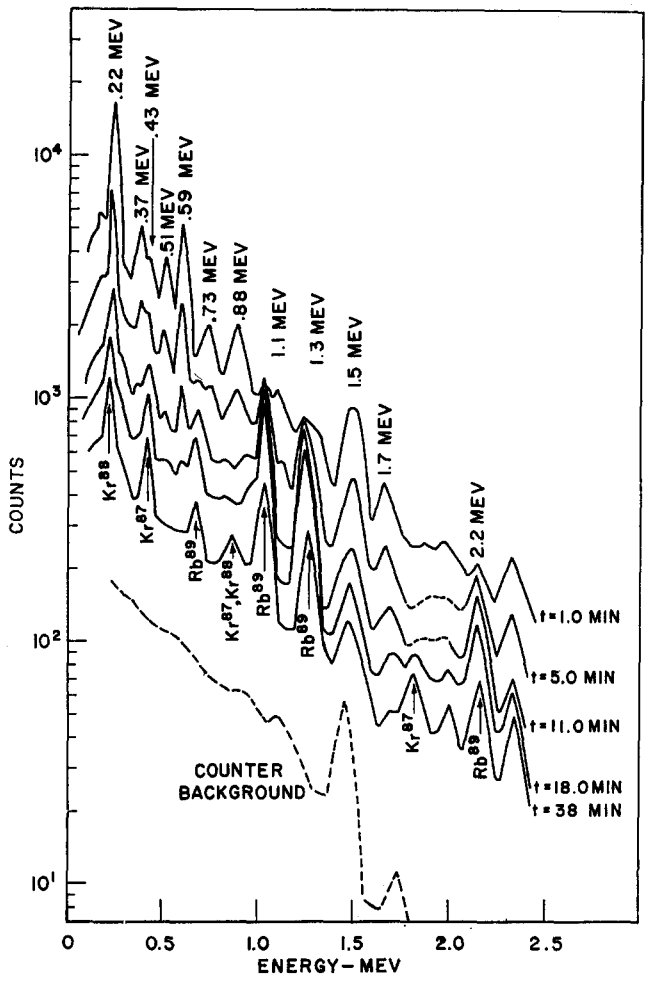

FIG. 2.—Series of γ-spectra of ^{89}Kr separated 4 min after end of 15 sec irradiation. Count length: 2 min.

multi-component nature of some of the spectra and to the poor statistics in counting short-lived samples, low intensity γ rays present may not have been detected.

Mass assignments have been made on the basis of the agreement with the previously reported half-lives, as well as the growth and decay of known daughter activities.

1. *3·2 min* ^{89}Kr

The krypton fraction was allowed to decay for 4 min after the separation to reduce the contribution of 33 sec ^{90}Kr to an insignificant level. It was then filtered to remove

the ^{90}Rb and ^{89}Rb formed during the decay period and counted immediately. The 0–2·5 MeV γ-ray spectrum as a function of decay time is shown in Fig. 2.

A limited number of coincidence measurements were made on ^{89}Kr, and γ-γ coincidences were observed between the γ-ray at 215 kev and those at 370, 510, 880 and 1500 keV. No γ-rays were observed in coincidence with the 595 keV γ-ray. This γ-ray was, however, found to be in coincidence with the 4·0 MeV β-ray component. This γ-β coincidence indicates a value of ~4·6 MeV for the decay energy of ^{89}Kr. The detection of γ-rays of energy up to 4·5 MeV established that there is no appreciable β-ray transition to the ground state of ^{89}Rb.

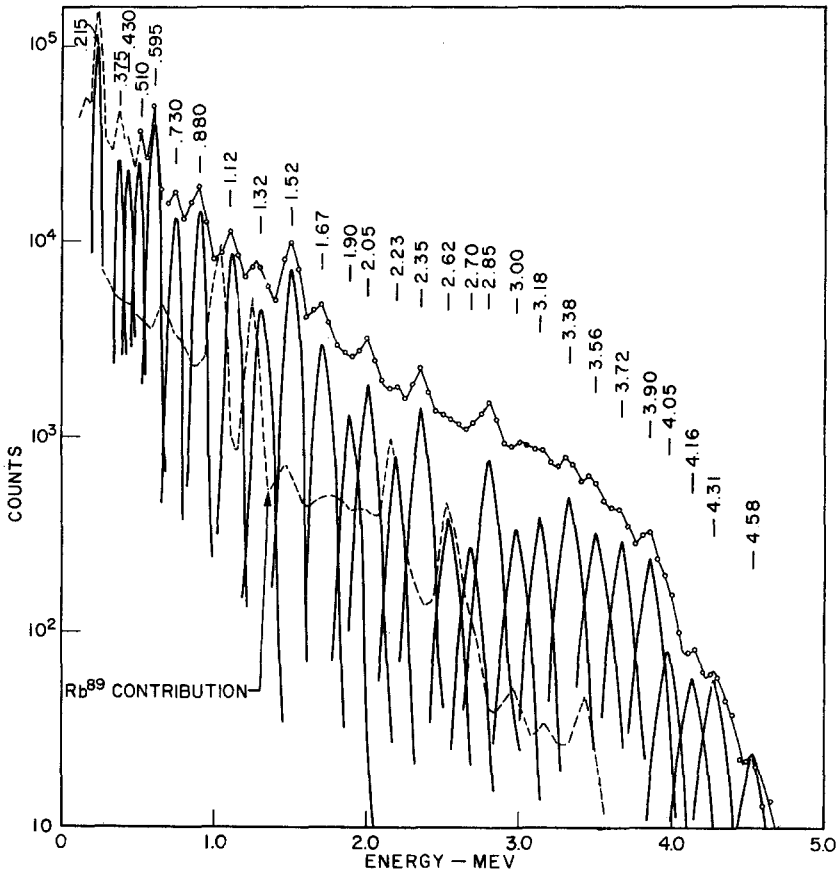

FIG. 3.—Spectrum of ^{89}Kr after subtraction of ^{89}Rb, ^{87}Kr and ^{88}Kr.

The analysis of the complex spectrum into component γ-ray lines is shown in Fig. 3. Due to the large number of γ-rays involved, the analysis is necessary but not sufficient to establish uniquely the energies and intensities of the γ-ray lines of ^{89}Kr. The background correction for 78 min ^{87}Kr and 2·8 hr ^{88}Kr was minimized by utilization of short irradiation and cooling of the sample before the separation. The radiochemical purity of the samples was confirmed by separation of the rubidium daughter after an initial decay period and establishing the absence of detectable amounts of 32 min ^{138}Cs.

2. *33 ses* ^{90}Kr

The series of spectra obtained from the short-lived krypton fraction is shown in Fig. 4. The uncertainty in the half-life determination on the peaks at 1·54 and 1·78 MeV is somewhat large because of the lower count rate and the ^{89}Kr background. These γ-rays were not observed in separated daughter fractions nor in the xenon fraction and are therefore assigned to ^{90}Kr along with those at 125, 540 and 1100 keV.

In addition to the 1·8 MeV β-ray listed in Table 2, a more energetic component of lower abundance was observed which was masked by the growth of ^{90}Rb, and is assumed to be the 3·2 MeV β-ray previously reported.[2]

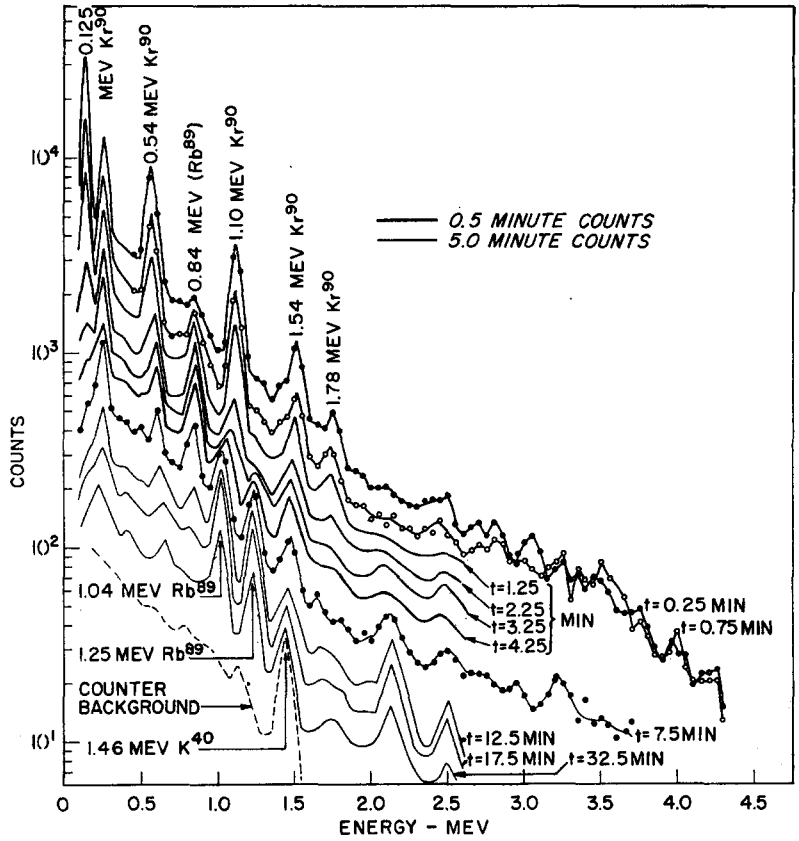

FIG. 4.—Series of γ-spectra of ^{90}Kr separated 15 sec after end of 10 sec irradiation.

Because of the equivalence of the relative intensities of the three major γ-rays, and of the photon per disintegration values (>0·6), a cascade originating at 1·78 MeV is postulated, competitive with the 1·78 MeV crossover transition. This level must then be fed by the abundant 1·8 MeV β-ray transition and also by the 1·54 MeV γ-ray. From the photon per disintegration values a β-ray transition to the ground state of ^{90}Rb in ~20 per cent of the decays is indicated, consistent with the low-abundance high-energy β-ray transition observed.

3. *1·2 min* ^{91m}Rb

The decay of the short-lived rubidium activity is shown in Fig. 5. To obtain this activity, the krypton fraction was allowed to decay for 30 sec, the residual gas purged, and the rubidium fraction sent to the counter. The photopeaks were not observed in similar spectra when the fission product sample was allowed to decay for 45 sec before sampling, and are therefore not associated with the decay of ^{89}Rb or ^{90}Rb. The short half-life of ^{92}Kr (~3 sec) makes it improbable that 80 sec ^{92}Rb would be detected in

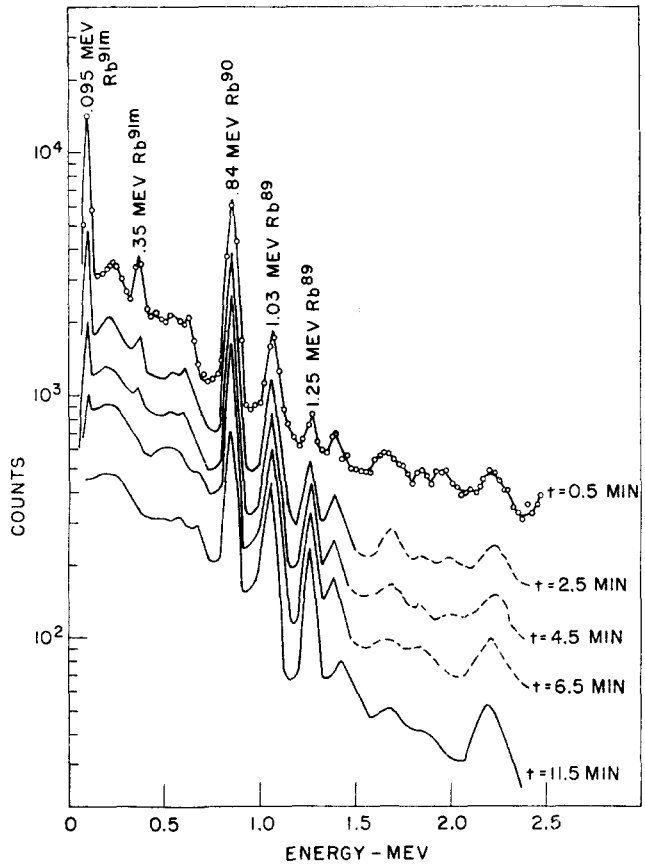

FIG. 5.—Series of γ-spectra of ^{91m}Rb filtered from krypton fraction separated 15 sec after end of 20 sec irradiation and allowed to decay 30 sec. Count length: 1 min.

this experiment. A 4·6 MeV β-ray was observed by previous workers[2] in the decay of the nuclide. The rubidium X-ray region was not observed in this experiment; such information would, however, be useful in establishing whether the γ-rays are competitive with or preceded by the enegetic β-ray transition.

4. *41 sec* ^{139}Xe

The only resolvable γ-rays of 41 sec half-life are of low energy, as can be seen from Fig. 6. The β-ray spectra showed an ~4·6 MeV endpoint with one or more components at ~3·5 MeV. The primary transitions are energetic β-ray transitions to

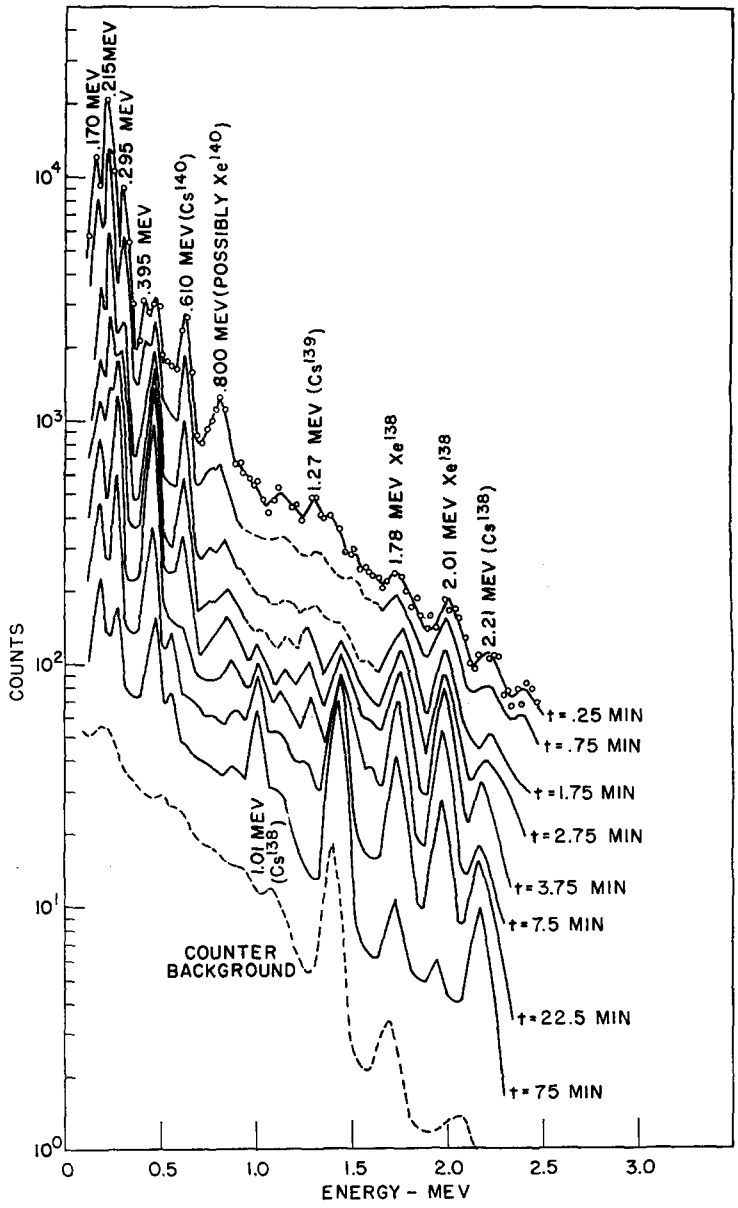

FIG. 6.—Series of γ-spectra of Xe separated 30 sec after end of 15 sec irradiation. Count length: 0·5, 5 and 50 min.

low-lying levels and probably to the ground state of ^{139}Cs. The relative contribution of ^{140}Xe and ^{140}Cs which may have been present should have been negligible under the conditions of the experiment.

5. *9·5 min* ^{139}Cs

The xenon fraction was allowed to decay for 100 sec, and the residual xenon then purged from the charcoal filter with hot air and vacuum. The only γ-rays of ~9·5

min half-life detected were those at 630 and 1270 keV previously reported by WASSON.[7] The relative intensities were resolved from the spectrum taken at 7·5 min, to allow decay of the 66 sec ^{140}Cs peak at 610 keV as shown in Fig. 7. The photon per disintegration values were calculated using the same assumptions as in the case of ^{139}Xe (see Table 2). The Rb activity observed in these spectra results from decay of Kr during the separation step.

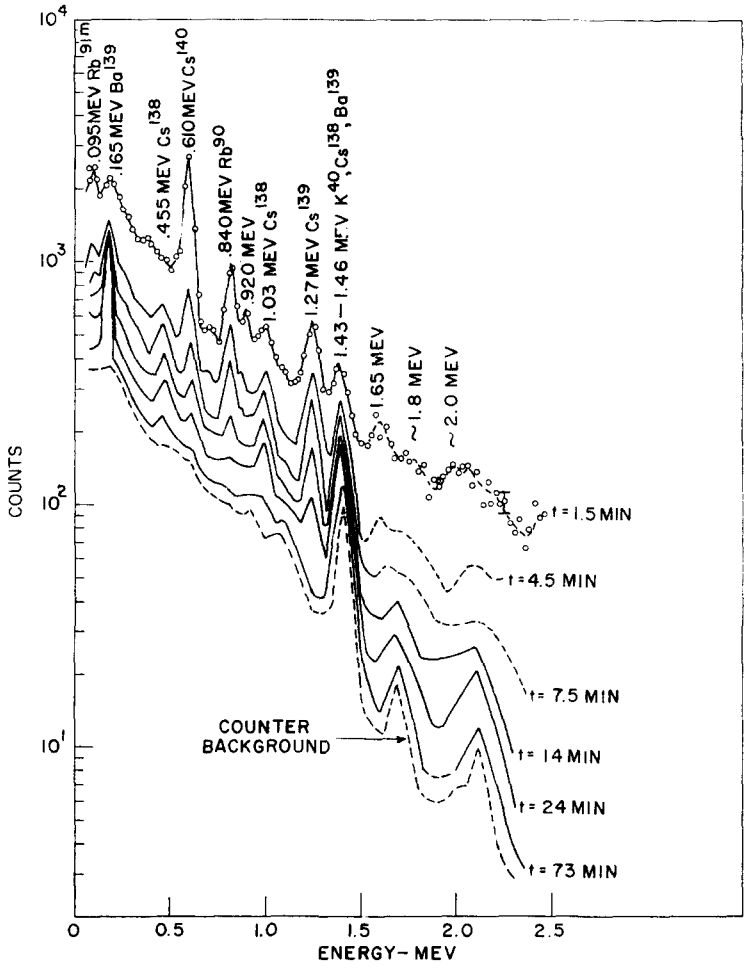

FIG. 7.—Series of γ-spectra of Cs filtered from Xe fraction separated 15 sec after end of 20 sec irradiation and allowed to decay 100 sec. Count length: 3 min.

The maximum β-ray endpoint observed under conditions optimized for the detection of ^{139}Cs was ~3·8 MeV. Because of the low intensity of the observed γ-radiation, this β-ray endpoint is taken as the decay energy.

6. *66 sec* ^{140}Cs

In a similar xenon sample purged after 40 sec cooling, an intense γ-ray of 66 $\pm$ 2 sec half-life was observed at 610 keV and is shown in Fig. 8. The presence of higher

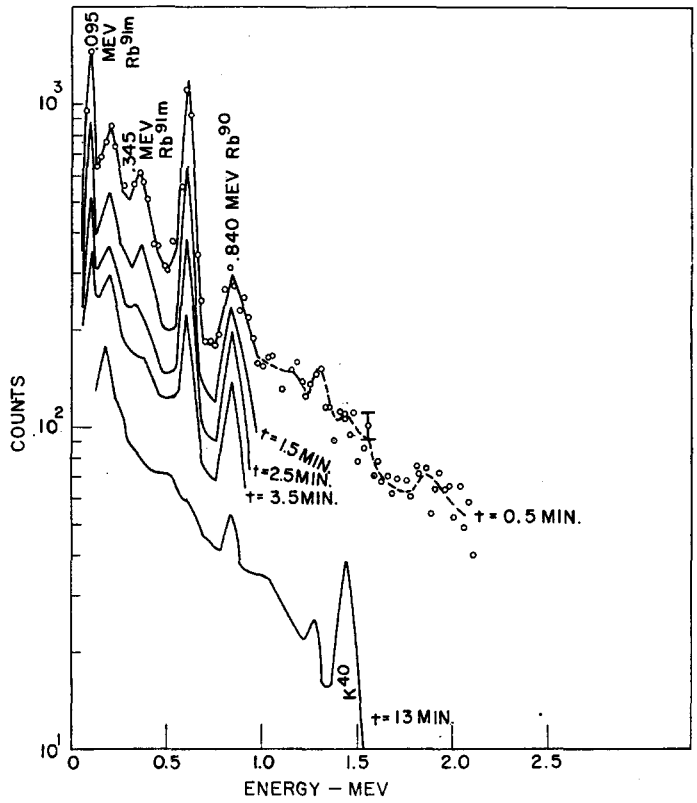

FIG. 8.—Series of γ-spectra of Cs filtered from Xe fraction separated 15 secs after end of 15 sec irradiation and allowed to cool 40 secs. Count length: 1 min.

energy γ-ray radiation of 66 sec half-life was indicated by the gross decay of the higher channels of the spectra. No β-ray spectra of this nuclide were obtained.

CONCLUSIONS

The feasibility of the study of the short-lived fission gases by a simple and rapid gas chromatographic separation has been established. The correlation of the β- and γ-radiations observed in this work into definitive decay schemes will require a considerable amount of further investigation. Extensive coincidence measurements are not practical with the system as set up for these experiments because of the large number of samples necessary to obtain useful statistics. The decay energies given in Table 2 are in fair agreement with those computed by RIDDELL,[14] but somewhat lower than the revised values quoted by CAMERON.[15]

Acknowledgements—The authors wish to thank Professor H. J. GOMBERG, C. W. RICKER, W. H. DUNBAR, and the staff of the Ford Nuclear Reactor for their cooperation in making available the laboratory and irradiation facilities. The assistance of R. SHIDELER and H. NASS in many phases of the work is gratefully acknowledged.

[14] J. RIDDELL, Atomic Energy of Canada Limited, Report CRP-654.
[15] A. G. W. CAMERON, Atomic Energy of Canada Limited, Report CRP-690 (1957).