STUDY OF THE γ-RADIATION FROM Xe^{137} *AND* Xe^{138}

V. V. OVECHKIN and N. N. DEMIDOVICH

Submitted to JETP editor May 23, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) **47**, 1671-1676 (November, 1964)

The isotopes Xe^{137} and Xe^{138} were obtained by bombarding a water solution of U^{235} with Po–Be neutrons moderated in paraffin. The gaseous fission products were removed from the solution by a current of helium gas, and the xenon isotopes were absorbed by cooled activated charcoal. γ-ray measurements were made in a scintillation γ spectrometer. New data have been obtained on the relative intensities and total γ-ray yields per β decay for Xe^{137} and Xe^{138}, which make possible considerable refinement of the decay scheme of these isotopes. The total γ-ray yields are: for Xe^{137}, 0.15 MeV—5%, 0.44 MeV—33%; and for Xe^{138}, 0.16 MeV—5%, 0.25 MeV—37%, 0.42 MeV—23%, 1.73 MeV—16%, and 1.99 MeV—11%.

INTRODUCTION

THE first indication of the complex nature of the Xe^{137} and Xe^{138} β spectra appeared in the work of Nassif and Seelmann-Eggebert[1] and Thulin[2] in 1955. A detailed study of the γ radiation from Xe^{137} was made in 1960 by Prakash.[3] More recently Ockenden and Tomlinson[4] obtained new data on Xe^{137} and Xe^{138}. According to the data of Prakash the 0.15, 0.24, 0.35, 0.44, and 1.60 MeV γ lines are assigned to the nucleus Xe^{137}. On the contrary, Ockenden and Tomlinson find only one γ line at 0.44 MeV for Xe^{137} and therefore they consider the Xe^{137} decay scheme presented by Prakash to be incorrect.

In view of the contradiction of the above results on the γ radiation from Xe^{137} and the absence of data on the absolute γ-ray yield per β decay, we undertook a study of the γ radiation of this isotope. This also presented the possibility of verifying the data of Thulin[2] and Ockenden and Tomlinson[4] on the γ radiation of Xe^{138}, whose decay gives rise to the γ lines at 0.16, 0.26, 0.42, 0.51, 1.78, and 2.02 MeV.

Prakash[3] and Ockenden and Tomlinson[4] used a reactor as a souree of thermal neutrons; calculations carried out by us showed that similar work could be performed with a Po–Be neutron source (with an intensity $\sim 2 \times 10^7$ sec^{-1}), using enriched U^{235} (~ 6.0 g) and a special process for separating the xenon isotopes in activated charcoal.

EXPERIMENTAL DETAILS

Figure 1 shows the essential features of the apparatus for obtaining and separating the gaseous fission products Xe and Kr.

In the center of the paraffin block 15 is a quartz ampoule 6 containing an aqueous solution of uranyl nitrate (~ 6.0 g U^{235}). Helium from the container 1

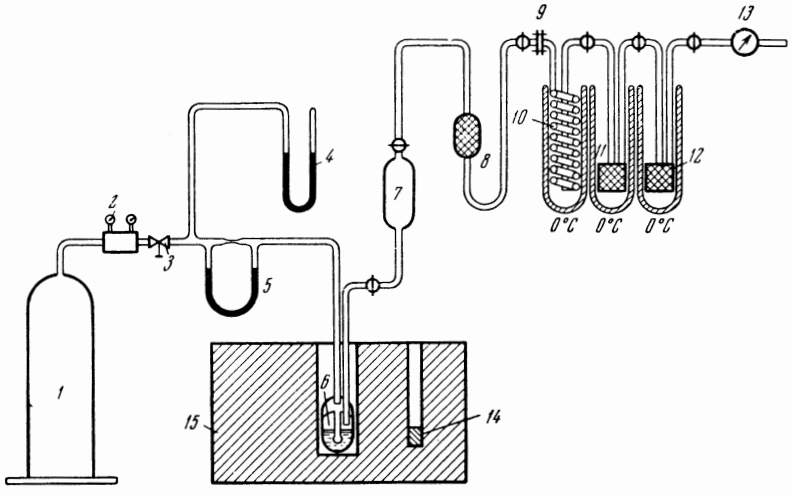

FIG. 1. Diagram of equipment for obtaining and separating the gaseous fission products Xe and Kr: 1 – Helium container, 2 – pressure reducer, 3 – valve, 4 – oil manometer, 5 – flow meter, 6 – quartz ampoule with uranium solution, 7 – 1.5-liter reservoir, 8 – silica gel column, 9 – aerosol filter type AFA-RMP, 10 – cooling coil, 11 and 12 – activated charcoal columns, 13 – gas flow meter type GSB-400, 14 – Po-Be neutron source, 15 – paraffin block.

passes through the flow meter 5, enters the ampoule 6, and carries away the gaseous fission products; the fission fragments with a half-life of a few seconds decay in the vessel 7 which has a volume of 1.5 liters. The gas then enters a silica gel column 8 which serves for removal of water and iodine vapor, passes through the aerosol filter 9, the cold trap 10, and enters the activated charcoal adsorption columns 11 and 12. After passing through the flow meter 13 the gas is released to the atmosphere.

The activated charcoal adsorption columns 11 and 12 are intended for trapping Xe from the Xe + Kr gas mixture in the helium flow. The size of the charcoal adsorption layer was chosen from the condition for most complete separation of xenon and krypton at a given adsorbant temperature, a given velocity, and a definite period of helium pumping. It was found that about six times more xenon was retained in the first column (11) than in the second column (12).

Before each irradiation the entire system is evacuated, with baking of columns 11 and 12 at ~350°C. After outgassing, columns 11 and 12 are immersed in a vessel with a coolant mixture at a temperature of ≈0°C and the helium flow is turned on. The necessary helium flow rate is established and the ampoule 6 with the uranium is lowered into the paraffin block 15, the distance from the ampoule to the neutron source 14 being 5.5 cm. The thermal neutron flux at the ampoule is $\sim 4.0 \times 10^4\,cm^{-2}\,sec^{-1}$.

The helium flow is cut off one minute after the end of the irradiation. Then the adsorption columns 11 and 12 are removed and transferred to a scintillation γ spectrometer consisting of a type USD-1 universal scintillation probe with NaI(Tl) crystals 30 mm in diameter and 14 mm thick, and 40 mm in diameter and 50 mm thick, with a 100-channel pulse height analyzer, type AI-100. The γ-spectrometer energy resolution for the Cs^{137} line (E_γ = 0.66 MeV) was about 10% for the 40 × 50 mm crystal and 8.5% for the 30 × 14 mm crystal. In the course of the measurements an energy calibration was made with the following γ sources: Co^{60}, Kr^{85}, Cs^{137}, Ra^{226}, and U^{235}.

In the present work we made sixteen tests with different rates of helium flow in the range 0.4—1.8 liters per minute and different times of irradiation of the uranium (4—25 min). We measured and analyzed about 300 γ spectra in the region 10—2500 keV. As an illustration Fig. 2 shows typical γ spectra of adsorption column 11 obtained with the 40 × 50 mm crystal in the energy interval 30—850 keV.

After recording the γ-spectra we calculated the areas of the peaks and plotted the decay curves for each of the observed peaks. From decomposition of the decay curves we have reliably identified by their half-lives the following isotopes: Xe^{137}, Xe^{138}, Cs^{138}, and Ba^{139}. Figure 2,b shows a decomposition curve for the decay of the peak at 150 keV.

In order to obtain relative γ-ray intensities from the peak areas, it is necessary to have a calibration curve of the efficiency for recording γ rays as a function of energy for the crystals used. In the present work this curve was obtained using the method described by Ovechkin et al,[5] with corrections for the finite dimensions of column 11 and for the thickness of the protective covering on the crystal.

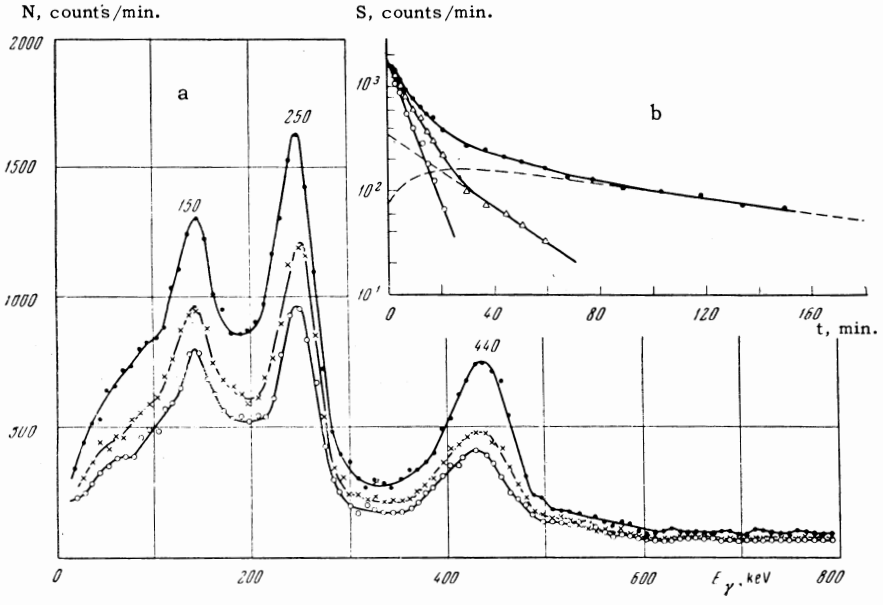

FIG. 2. a) Typical γ spectra of column 11 (Fig. 1): • – after 3 min 25 sec, × – after 8 min 20 sec, ○ – after 13 min 5 sec (time measured from end of irradiation; duration of measurement 2 min). b) Decomposition of decay curve into the three component isotopes (150 keV peak): • – S_{150} ($T_{1/2}$ = 82 min) for Ba^{139}, Δ – S_{150} ($T_{1/2}$ = 17 min) for Xe^{138}, and ○ – S_{150} ($T_{1/2}$ = 4.2 min) for Xe^{137}; the dashed line is the calculated accumulation curve for Ba^{139}.

Table I

γ-ray energy, MeV	Relative Intensities			
	Data of [6]	Data of [2]	Data of [7]	Present work
0.46	33	20	32	37
1.0	43	25	34	33
1.4	100	100	100	100
2.2		20	25	20

The adequate accuracy of this calibration is illustrated by the data in Table I, where our final results for the relative intensities of Cs^{138} γ rays are compared with the results of other authors.[6,7,2]

In addition to the relative intensities, we also determined the total γ-ray yields per β decay for Xe^{137} and Xe^{138}. For the total yield of 0.46-MeV γ rays from Cs^{138}, we used the value 23 ± 3% determined by Bunker et al,[7] which allowed the total Cs^{138} β activity to be estimated from the γ spectra. Then, using the known experimental conditions and the fission yield data, we can determine the Xe^{138} and Xe^{137} β activity. The total γ-ray yields are obtained by taking the corresponding ratios of the intensities of γ rays from the measured γ spectra to the total β activities of these isotopes.

EXPERIMENTAL RESULTS AND DISCUSSION

Xe^{137}. Table II lists the relative intensities of the Xe^{137} γ rays, measured by Prakash,[3] Ockenden and Tomlinson,[4] and the present work.

As can be seen from Table II, the results of the present work are closer to the data of Ockenden and Tomlinson[4] and confirm their conclusion as to the incorrectness of Prakash's results. In addition it should be noted that both the additional Xe^{137} lines at 0.15 MeV ($T_{1/2}$ = 3.9 ± 0.3 min) and 0.03 MeV ($T_{1/2}$ = 4.0 ± 0.3 min) appear rather reliably in our measurements when the decay curves are decomposed. According to our measurements the total yields of 30-, 150-, and 440-keV γ rays per Xe^{137} β decay are respectively 7, 5, and 33% (maximum error ±23%).

Table II

γ-ray energy, MeV	Relative Intensities		
	Data of [3]	Data of [4]	Present work
0.03			0,21
0.15	0.29		0,16
0,24	1.0		
0,35	0.14		
0.44		1.0	1,0
0.46	0.88		
0.60	0.40		

In Cs^{137}, according to the shell model picture,[8] the occurrence of isomeric transitions is improbable since the 82-neutron shell is completely filled.

In this connection and in considering the results of the γ-ray intensity and energy measurements, it is necessary to assume that the 30-keV γ rays correspond to K x-ray lines. If we make a further comparison of the experimental results with the theoretical conversion coefficients α_K for Cs as given by Sliv and Band,[9] we can draw the following conclusions concerning the β decay of Xe^{137} (see diagram in Fig. 3).

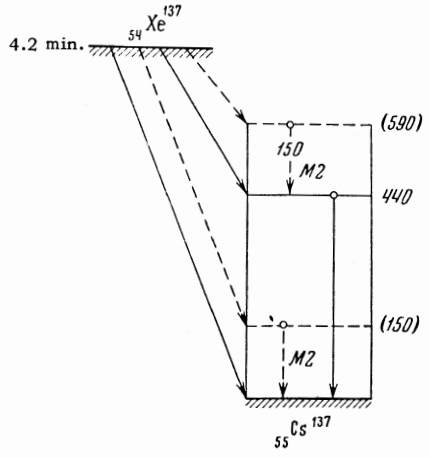

FIG. 3.

1. There are two types of γ rays: 0.15 and 0.44 MeV, and also K x-rays. The main contribution to the x-ray peak is due to the 0.15-MeV γ rays, to which we must assign type M2 multipolarity.

2. Cs^{137} has an excited level at 0.44 MeV which decays to the ground state via an extremely intense γ ray (33% per β decay). There is an additional level either at 0.15 MeV or 0.59 MeV.

3. In the β spectrum of Xe^{137} at least three groups of β particles should be observed; the intensity of the group associated with the transition to the ground state of Cs^{137} presumably is 55–67%.

Xe^{138}. According to our data the following lines can be associated with Xe^{138}: 30 keV ($T_{1/2}$ = 17.3 ± 0.3 min); 0.16 MeV ($T_{1/2}$ = 17.0 ± 0.4 min); 0.25 MeV ($T_{1/2}$ = 16.0 ± 0.7 min); 0.44 MeV ($T_{1/2}$ = 16.5 ± 0.3 min); 1.73 MeV ($T_{1/2}$ = 16.9 ± 0.1 min); 1.99 MeV ($T_{1/2}$ = 16.6 ± 0.1 min). The assignment of the three γ lines (0.15, 0.25, and 0.44 MeV) to Xe^{138} was established in an additional experiment with rapid purging of adsorption column 11 by heated air. This experiment was started 30 min after the end of irradiation, so that the Xe^{137} had already decayed. After the purging it turned out that the 250 keV peak disappeared completely, the intensity of the 160 and 440 keV peaks decreased

Table III

γ-ray energy, MeV	Relative Intensities		
	Data of [2]	Data of [4]	Present work
0.03			0.05
0.16		0.33	0.14
0.26		1.0	1.0
0.42	1.0	0.40	0.63
0.51	0.20	0.08	
1.78	Not determined	0.66	0.44
2.02	Not determined	0.58	0.30

sharply, and their half-lives began to correspond to Ba^{139} ($T_{1/2}$ = 81 min) and Cs^{138} ($T_{1/2}$ = 32 min).

Table III compares our measurements on the relative intensities of Xe^{138} γ rays with those of other authors.[2,4] It is evident from this table that our data on Xe^{138} γ-ray energies agree with those of Ockenden and Tomlinson,[4] but that the values of relative intensities are considerably different (by a factor of 1.5–2). The good agreement of our similar data on Cs^{138} with those of Bunker et al[7] allows us to conclude that the calibration of our spectrometer was carried out correctly and that our determinations of the Xe^{138} relative intensities are trustworthy.

Our measurements of the Xe^{138} total γ-ray yields per β decay are listed below (the maximum possible error is ±22%):

γ-ray energy, MeV:	0.03	0.16	0.25	0.42	1.73	1.99
γ-ray yield, %:	3	5	37	23	16	11

Analysis of the experimental data obtained and comparison with the conversion coefficients α_K permit us to construct a decay scheme for Xe^{138}, into which the two observed γ lines have been fitted (Fig. 4). The decay scheme which we propose has the following features.

1. The 2.4-MeV end-point β decay[1] occurs to the ground state of Cs^{138} with an intensity of 28%. In addition to the main group, three other groups of β particles should be observed.

2. Instead of the 2.40-MeV excited level, we introduce a 2.00-MeV level which is depopulated by a direct γ transition to the ground state and a cascade transition through a new level at 0.26 MeV.

3. The 0.16- and 0.26-MeV γ rays make roughly equal contributions to the 30-keV x ray peak observed by us; the multipolarity of these γ lines must be M1 and E2, respectively.

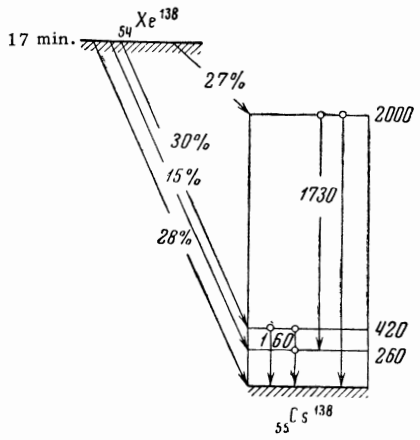

FIG. 4.

4. The authors owe an expression of gratitude to I. E. Nakhutin for the interest which he has shown in this work, and to G. A. Loshakov and Yu. V. Linda for their assistance.

[1] S. Nassiff and W. Seelmann-Eggebert, Zs. Naturf. **10a**, 83 (1955).

[2] S. Thulin, Arkiv Fys. **9**, 137 (1955).

[3] S. Prakash, Zs. Elektrochem. **64**, 1037 (1960).

[4] D. Ockenden and R. Tomlinson, Canad. J. Chem. **40**, 1594 (1962).

[5] Ovechkin, Pirkin, Polyakov, and Ochkin, PTÉ, **5**, 126 (1960), translation: Instruments and Experimental Techniques, No. 5, 817 (1960).

[6] Langer, Duffield, and Stanley, Phys. Rev. **89**, 907A (1953).

[7] Bunker, Duffield, Mize, and Starner, Phys. Rev. **103**, 1417 (1956).

[8] M. Goeppert Mayer and J. Jensen, Elementary Theory of Nuclear Shell Structure, New York, Wiley, 1955. Russian translation IIL, 1958.

[9] L. Sliv and I. Band, Tablitsy koeffitsientov vnutrenneĭ konversii (Tables of Internal Conversion Coefficients) part 1, AN SSSR, 1956.

Translated by C. S. Robinson
237