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Characteristics of the Fission Product Cs^{135}

NATHAN SUGARMAN
*Los Alamos Scientific Laboratory, University of California, Los Alamos, New Mexico**

and

Institute for Nuclear Studies, University of Chicago, Chicago, Illinois
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Two samples of Xe^{135} of high intensity were prepared at the Los Alamos homogeneous pile. The decay products of the gas contained a Cs activity presumed to be Cs^{135}. The half-life of Cs^{135} was found to be $(2.1\pm0.7)\times10^6$ yr. and the maximum energy of its β^--radiations 0.21 Mev. No γ-radiations were detected. The capture cross section of Cs^{135} for pile neutrons was determined to be 14.5 ± 4 barns.

INTRODUCTION

IN earlier fission product studies the attempts to detect radiation from Se^{79}, Pd^{107}, I^{129}, and Cs^{135} were unsuccessful. On the assumption that the half-lives of all four species were long, minimum half-lives were estimated from the failure to detect the radiations and the factors limiting observation.[1] Chief among the latter have been the detection apparatus for extremely soft β^--radiation and the strength of the sources prepared.

The three earlier experiments designed to measure the half-life of Cs^{135} have given minimum half-life estimates ranging from 1200 years to 3.5×10^6 years. In the first experiment Finkle and Sugarman[2] isolated an iodine sample from irradiated uranium and after decay of the 6.7-hr. I^{135} and 9.2-hr. Xe^{135} analyzed for Cs. No radiations were detected by a mica end-window counter. On the assumption that the radiations of Cs^{135} would have been detected in this apparatus, a minimum half-life for Cs^{135} was calculated to be 1200 years. In a similar experiment Glendenin and Metcalf[3] estimated the half-life as >2000 years. In a third experiment Engelkemeir[4] looked for radiations from Cs^{135} in the decay products of 30 millicuries of 9.2-hr. Xe^{135}. The absence of radiations in a mica end-window counter led to a half-life estimate of $>3.5\times10^6$ years for β^--particles sufficiently energetic to penetrate the 3.5-mg/cm^2 mica window and the $\sim$12-mg/cm^2 Cs carrier source without appreciable loss in intensity. The Cs sample was also examined with a windowless counter and if a β^--energy of 30 kev was assumed, the minimum half-life estimate fell to 2.5×10^4 years.

In the present experiments, two samples of Xe^{135}, containing 36 curies and 30 curies of activity, were prepared at the Los Alamos homogeneous pile.[5] After decay of the Xe activity, the decay products were examined for Cs activity. A new Cs activity was found, the maximum energy of the β^--radiations being 0.21 ± 0.01 Mev as determined by Feather analysis of the Al absorption curve. The timing of the experiments was such as to indicate that the new activity is Cs^{135}, the daughter of 9.2-hr. Xe^{135}. The half-life for Cs^{135} calculated from the observed activity and the total activity of the Xe parent is $(2.1\pm0.7)\times10^6$ yrs. This value is not inconsistent with the limiting value set by Engelkemeir[4] if one considers the loss in intensity arising

* The Xe^{135} samples were prepared during the summer of 1948 when the author was a consultant to the Los Alamos Scientific Laboratory.

[1] "Nuclei formed in fission," issued by The Plutonium Project, J. Am. Chem. Soc. **68**, 2411 (1946); Rev. Mod. Phys. **18**, 513 (1946).

[2] B. Finkle and N. Sugarman, Plutonium Project Record, **9B**, 7.40.1 (1946).

[3] L. E. Glendenin and R. P. Metcalf, Plutonium Project Report, CC-2219 (1945).

[4] D. W. Engelkemeir, Plutonium Project Record, **9B**, 7.40.2 (1946).

[5] R. F. Christy, AEC declassified document, MDDC No 72 (June 18, 1946).

TABLE I. Activity of 9.2-hr. Xe^{135} prepared at water boiler.

Experiment	Gas cell	Activity of 9.2-hr. Xe^{135} at time of bulb filling (c/min.)	Counting geometry*	Fraction of gas in cell	Calculated activity of 9.2-hr. Xe^{135} at bulb filling time (dis/sec.)
1	1	14,000 (1st filling)	0.033	5.73×10^{-9}	1.23×10^{12}
	2	12,870 (1st filling)	0.033	5.14×10^{-9}	1.26×10^{12}
		14,200 (2nd filling)	0.033	5.14×10^{-9}	1.39×10^{12}
2	1	11,800 (1st filling)	0.033	5.88×10^{-9}	1.01×10^{12}
		13,800 (2nd filling)	0.033	5.88×10^{-9}	1.18×10^{12}
	2	11,700 (1st filling)	0.033	5.26×10^{-9}	1.12×10^{12}

* Average counting geometry of cell determined from measurements with a U_3O_8 β^--standard.

from the absorption of the β^--rays in the source and counter window.

PREPARATION OF Xe^{135}

In the operation of the Los Alamos homogeneous pile (water boiler) the active Kr and Xe isotopes formed in the fission process are swept out of solution and discarded. This pile is, therefore, a convenient source of gaseous fission products freed from the host of non-volatile active products formed in fission. The separation of 9.2-hr. Xe^{135} from the gases emanating from the water boiler during its operation presents a formidable shielding problem because of the many short-lived Kr and Xe activities. The radiation hazard can be substantially reduced if advantage is taken of the fact that only two of the gaseous fission products have parents of appreciable half-life, namely, 5.3-day Xe^{133} growing from a 22-hr. I^{133} and 9.2-hr. Xe^{135} from 6.7-hr. I^{135}. The active gases produced during the operation of the boiler are removed quite efficiently by the internal bubbling from the radiation decomposition of the water. These gases are discarded. If one gas-sweeps the boiler after its operation and examines the sweeping gas, the only fission products which should be present are those whose parents are sufficiently long-lived to continue generating these species, namely, Xe^{133} and Xe^{135}.

In the isolation of the Xe samples the boiler was operated for some twelve hours. The gases during this period were discarded. The boiler was shut down and the solution swept with He for about 30 minutes at a rate of 50 l/hr. to remove any residual gases. This gas was also discarded. The solution was then swept with He for five hours and the gas was passed through a vacuum line for collection of the Xe^{133} and Xe^{135} activities growing from their I parents during this interval.

The vacuum line for collecting of Xe was shielded by two inches of lead and consisted of a liquid NaOH trap for removal of acid vapor and spray, two glass wool traps for removal of spray, a Drierite trap for removal of the bulk of the water vapor, a dry ice cooled trap for removal of the residual water vapor, an activated charcoal trap cooled by dry ice for adsorption of Xe, and a Toepler pump and 1-l bulb for collection of the Xe after desorption from the charcoal.

After five hours of sweeping at the rate of ~30 l/hr. the He flow was stopped. At this time the activity on the charcoal trap was ~35 r/hr. at ~2 ft. Little activity was present elsewhere in the line except for the NaOH trap which had an observed level of ~2 r/hr. in contact. The dry ice was removed from the charcoal trap and the charcoal allowed to warm up to room temperature. Most of the air swept out of the boiler and adsorbed on the trap is removed but very little Xe is desorbed. To desorb the Xe the trap was heated by passing current through the resistance wire wound around it. The active Xe and residual air thus desorbed from the charcoal were transferred to a 1-l bulb on the line by means of the Toepler pump. The activity level of the Xe at this time corresponded roughly to 10 curies of Ra as measured with a radiation survey meter.

The bulb containing the Xe was then attached to another lead-shielded vacuum line for the transfer of a small aliquot of the gas to a counting cell. This vacuum line consisted of three 1-l bulbs, including the one containing the Xe, joined by sections of small volume. The volume of the bulbs and of each section of the line was measured. A small aliquot of the original gas (~2 ml) was diluted with air and expanded into a 1-l bulb. A small aliquot of this gas was again diluted with air and expanded into the third 1-l bulb. Finally, a small aliquot of the twice-expanded gas was transferred to a counting cell for measurement of the radioactivity.

The two counting cells were of glass, 2 cm across and 1 cm deep, with a wide flange for sealing of a 3-mg/cm^2 mica window. A micro stopcock was sealed at the bottom of each cell. The volumes of the cells were 3.09 ml and 2.65 ml. The average fractions of the original gas transferred to the cells after the dilution process were 5.8×10^{-9} and 5.2×10^{-9} in the two experiments. The radioactivity in the cells was measured with a 3-mg/cm^2 mica end-window counter at ~3.3 percent geometry.

Absorption and decay curves taken on the Xe samples showed that the major activity (94 percent) at the time of isolation was the 9.2-hr. Xe^{135}. The other activity present was the 5.3-day Xe^{133}. Two experiments were performed, in each of which samples of Xe^{135} in high intensity were prepared.

In each experiment, both gas cells were filled from the same diluted gas mixture and the activity in each was measured for some time. One of the cells was then emptied and a new diluted gas mixture was prepared by a different dilution procedure and the cell was refilled and the activity followed. The purpose in filling the cell from a new gas dilution was to test the dilution technique and the homogeneity of the gas. The data from the two experiments appear in Table I. The data are not corrected for the absorption or scattering of the β^--radiations because of the difficulty in estimating these corrections. Much of the error will, however, cancel since the two effects contribute oppositely to the observed counting rate.

The discrepancy in the activity of a cell from two different fillings arises from the difference in the dilution technique for each filling, the technique so designed as to give the maximum discrepancy from the inhomogeneity in the diluted gas mixture. Averaging the results from each experiment, weighing the two fillings equally, one finds that the total 9.2-hr. Xe^{135} activity present at the time the bulb was filled was 36 curies in experiment 1 and 30 curies in experiment 2. The estimated error in these values is ~20 percent. Much of this error comes from the uncertainty in the counting geometry and scattering and absorption corrections for the β^--radiations.

RADIOACTIVITY OF Cs^{135}

About a week after the preparation of the Xe^{135} samples, the bulbs containing the active Xe were washed with dilute acetic acid solution. The acid wash solution was introduced into the bulbs and shaken vigorously in order to wash the decay products out of the gas phase and from the walls. This process was repeated five times. The combined wash solutions from each bulb were examined for radioactivity some two months later. Samples were prepared for radioactivity detection by evaporation and by chemical analysis for radio-Cs. The analysis consisted of addition of Cs carrier to an aliquot of the wash solution, $CsClO_4$ precipitations, $Fe(OH)_3$ scavengings, precipitations of a mixed $CsI\text{-}BiI_3$ compound, and finally precipitation of cesium cobaltinitrite.[6] Measurements with a 2-mg/cm^2 mica end-window tube showed the presence of activity in the evaporated samples and the Cs precipitates. The absorption characteristics of the radiations of the evaporated samples were the same as those from the chemically separated Cs samples, and the level of activity in the evaporated samples from an aliquot of solution agreed with that of the Cs samples after self-absorption corrections were made. Analysis by the Feather method[7] of the Al absorption curve of the radiations of this activity led to an estimate of the maximum β^--ray energy of 0.21 ± 0.01 Mev (Fig. 1). The γ-counting rate was less than 1 c/min. for a Cs sample of 600 β^- c/min. This result indicates the absence of γ-rays of moderate energy in high yield.

Assignment of the new Cs activity to Cs^{135} is very probable from the way in which it was prepared. This assignment is consistent with the timing of the experiments in which the He gas-sweeping of the pile was retained for Xe adsorption after discarding the first half-hour sweeping to eliminate active gases from short-lived halogens. The only Cs isotopes which should be present in the decay products of the gas are Cs^{133} arising from the decay of Xe^{133} and Cs^{135} from Xe^{135}. Since Cs^{133} is a naturally occurring stable isotope, assignment of the activity to Cs^{133} would mean that the expected mode of decay would be isomeric transition. The shape of the Al absorption curve of the Cs activity is characteristic of a β^- spectrum rather than that of a conversion electron spectrum. The mass assignment of 135 appears most probable.

The half-life of Cs^{135} was determined from the measured activity of an aliquot of the wash solution and the previously determined Xe^{135} activity of the samples. Small aliquots of each wash solution were evaporated on 2-mil platinum foils and counted. No decay in the activity of the Cs samples was observed over a period of three months. Corrections for geometry, backscattering,[8] and absorp-

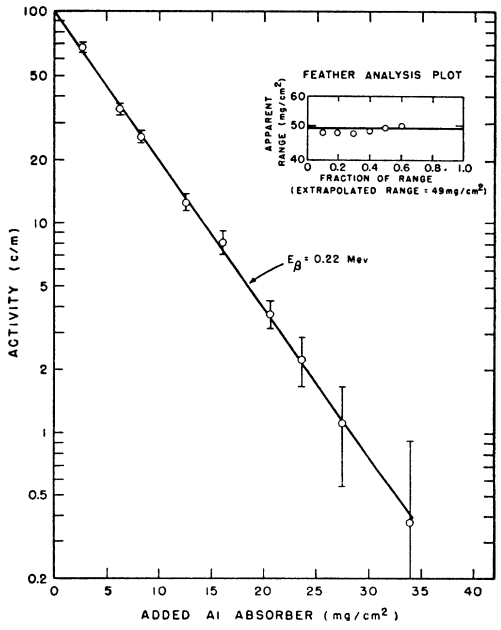

FIG. 1. Aluminum absorption curve of Cs^{135}. At zero added Al absorber the absorber thickness is about 3 mg/cm^2 from the counter window and air gap.

[6] Nathan Sugarman, J. Chem. Phys. **17**, 11 (1949).
[7] N. Feather, Proc. Camb. Phil. Soc. **34**, 599 (1938).
[8] Engelkemeir, Seiler, Steinberg, Winsberg, and Novey, Plutonium Project Record, **9B**, 2.4 (1946).

TABLE II. Half-life of Cs^{135}.

Experiment	Average activity of 3 samples (c/min.)	Aliquot	Counting geometry	Absorption correction (1-mg/cm² air, 2-mg/cm² mica)	Back-scattering correction	Total activity Cs^{135} in wash solution (dis/sec.)	Total activity 9.2-hr. Xe^{135} originally present (dis/sec.)	Half-life Cs^{135} (years)
1	111.0	0.02	0.175	1.71	0.67	600	1.32×10^{12}	2.30×10^{6}
2	116.5	0.02	0.175	1.71	0.67	635	1.12×10^{12}	1.85×10^{6}

tion of the β^--radiation in the 1-mg/cm² air and 2-mg/cm² mica window of the counter were made. The average half-life of Cs^{135} determined from these measurements is 2.1×10^6 years. The error estimated in the half-life of Cs^{135} is ~35 percent which includes the error in the Xe^{135} activity. The data are given in Table II.

NEUTRON CAPTURE CROSS SECTION OF Cs^{135}

Aliquots of the Cs^{135} wash solutions were irradiated in the thimble of the Argonne heavy water pile for about four days for the production of Cs^{136}. Radiochemical analyses for Cs were performed on the irradiated solutions three days after the end of the irradiation. Decay and absorption measurements on the Cs samples showed the presence of the 13.7-day Cs^{136} activity.[9] The cross section of Cs^{135} for the capture of pile neutrons was determined to be 13.7 barns from samples of the Cs solution from experiment 1, and 15.3 barns for the Cs solution from experiment 2. The average cross section is 14.5 barns with an estimated error of ~30 percent.

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[9] L. E. Glendenin, private communication on nuclear characteristics of Cs^{136}.