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U-235 Gamma-Radiation

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Evidence for the existence of a gamma-ray accompanying the alpha-decay of U-235 is presented. The energy of the gamma-ray, as determined by several independent absorption measurements, is 162 kev.

INTRODUCTION

THE three naturally occurring uranium isotopes, U-234, U-235, and U-238, are spontaneous alpha-emitters. In a previous report[1] it was shown that the U-234 alpha-decay is accompanied by x-rays, and the present paper reports the discovery and measurement of gamma-rays accompanying the alpha-decay of U-235. The 162 kev gamma-ray found associated with the U-235 decay was discovered and clearly identified at this laboratory in April 1946.[2] A similar radiation was observed somewhat earlier at the University of Chicago, but was attributed to U-234.

Discovery of the Gamma-Radiation

Sample Preparation

The significant steps in the purification of the samples of U-235 used were as follows. The bulk of the radioactive and other impurities were removed by ether extraction of uranyl nitrate from a nitric acid solution of the original ore. Reduction and precipitation of the uranium as UF_4 probably removed few impurities other than the very volatile fluorides such as silicon tetrafluoride. The precipitated, dried UF_4 was then converted with gaseous fluorine to the volatile UF_6, which was purified by condensation and re-evaporation. The UF_6 gas was then passed through a great many fine pored filters which increased by isotopic separation the proportion of U-235 to about one-third, and the UF_6 was then condensed directly into a clean, uncontaminated nickel cylinder with quarter-inch walls. It is felt that no radioactive materials except U-234, U-235, U-238, and insignificant traces of radon were present at the time of condensation into the sample cylinder.

Radiation

A large sample of solid U-235 enriched UF_6 in a quarter-inch wall nickel cylinder was placed at a standard distance (about six inches) from a screen wall Geiger counter. An initial activity of about 1000 c/m was observed. Superimposed on this was a growing activity of 24 day half-life (UX_1, UX_2, and UZ), extrapolating to 300–400 c/m at equilibrium. To prove that a residue of the non-volatile fluoride of UX_1, possibly swept through the filters and condensed with the sample, was not the cause of the initial 1000 c/m activity, a sample of natural uranium was examined. This was prepared in the same manner as the preceding sample but was passed through only a single filter with somewhat larger pores. The initial gamma-activity of this sample was only slightly above background (net rate 24 c/m) and increased with a 24-day half-life toward 500 c/m. Since this sample contained half again as much U-238, parent of the UX_1, UX_2, and UZ, as the enriched sample, the equilibrium activity of the latter sample should have been about 330 c/m. The initial activity found then (1000 c/m) was clearly not attributable to UX_1 inclusion in the sample of enriched uranium to the extent of more than a few percent. Indeed, the initial activity (24 c/m) of the natural uranium sample with much poorer filtration led to the expectation that UX_1 dust carry over in the enriched UF_6 sample was entirely negligible.

[1] R. L. Macklin and G. B. Knight, Phys. Rev. 72, 435 (1947).
[2] R. L. Macklin and W. S. Miller, Uranium Project Report A-3640.

Energy of the Radiation

A gram sample of nearly pure U-235F_4 was used for preliminary absorption measurements. These showed a prominent gamma-ray component at about 162 kev.

For more accurate measurements, UF_6 samples such as those described above were hydrolyzed and heated to give U_3O_8 powder. This was spread on sample holders in a thin layer of measured mass. Samples of enriched uranium (one-third U-235, two-thirds U-238) were prepared at the same time and made up with equal quantities of U-238. Thus, the gamma- and beta-rays of UX_1, UX_2, and UZ at any time were equal for a pair of samples. Hence, the difference in counting rate of a pair corresponded to a known quantity of U-235 (or U-234). Absorption measurements with both lead and aluminum were made at these laboratories and later (July, 1946) at Clinton National Laboratories. The 162 kev gamma-radiation was confirmed (see Fig. 1) and two weaker radiations corresponding to *L* and *M* x-rays

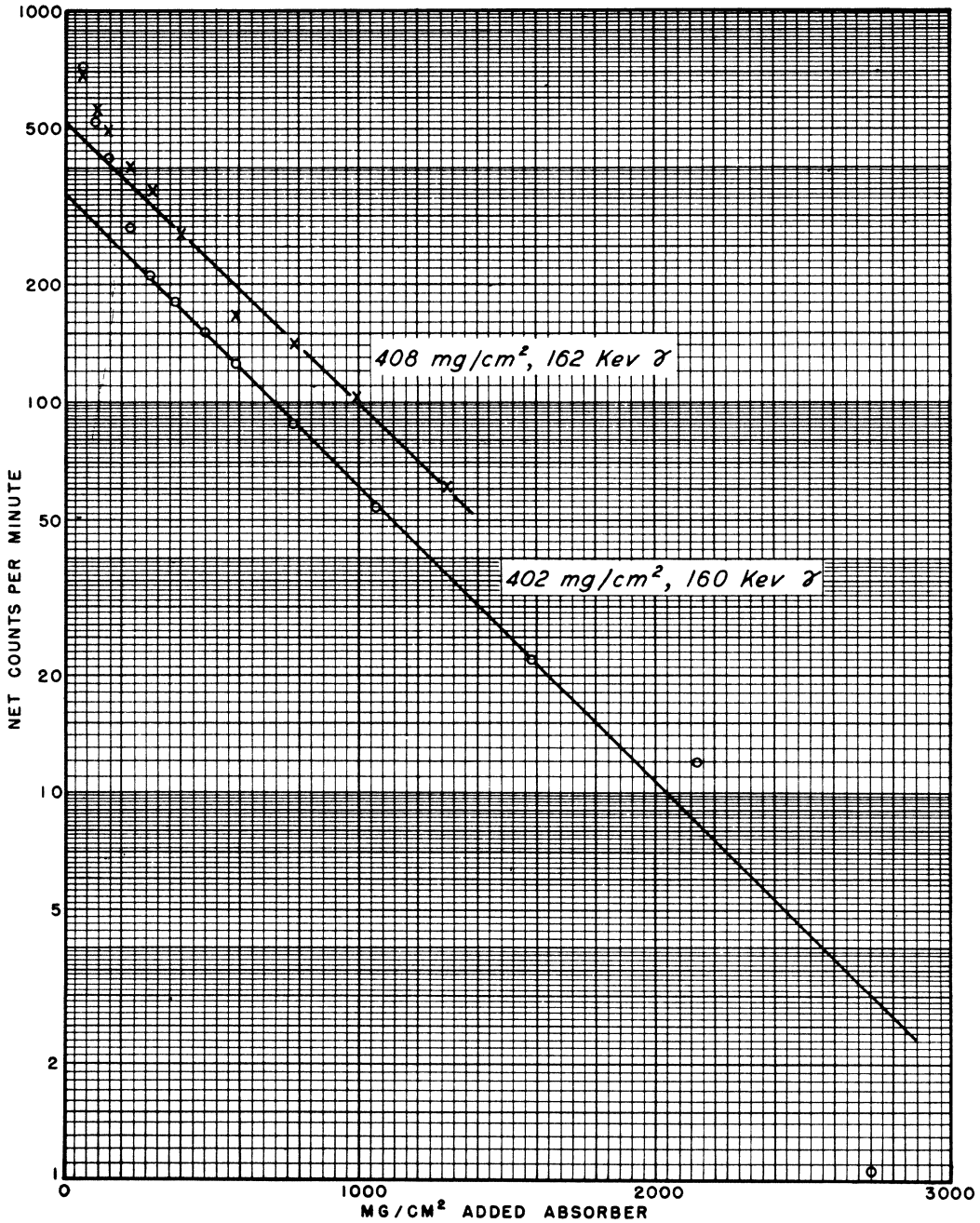

FIG. 1. Two Pb absorption curves of U-235 by difference (see text).

were found. These latter were later shown to be associated with U-234 (see reference 1).

The yield of gamma-rays per alpha-disintegration can be only roughly estimated. Values near 100 percent of the U-235 alpha-disintegration rate seem most reasonable although the gamma-counter efficiencies are so poorly known as to make this little more than a guess. One type counter yielded estimates of 10–100 percent and another gave 125–250 percent.

Origin of the Radiation

To determine which uranium isotope gave rise to the gamma-activity observed, samples of varying composition were used. The experiments with natural and enriched uranium already described showed, of course, that the 162 kev gamma-activity was not proportional to the U-238 content. Two samples were obtained which were nearly equal in U-234 content but differed by a factor of three in U-235 content as shown by mass spectrometer. These samples were several months old and hence contained virtually equilibrium quantities of UX_1, UX_2, and UZ proportional to the U-238 present.

Using the previous results for the gamma-radiation relative intensities measured through a quarter-inch of nickel, the ratio of gamma-activities of these samples (through $\frac{1}{4}''$ of nickel) was predicted on the basis of two hypotheses. If the 162 kev gamma-activity were due to the U-234, the activity ratio should be 0.63 because of the difference in U-238, UX_1, UX_2, and UZ content of the two samples. However, if the 162 kev gamma-activity were due to the U-235, the ratio would be 2.32. The ratio observed was 2.31 ± 0.04. Hence, the gamma-ray appears definitely to be associated with U-235.

DISCUSSION

The high U-234 alpha-activity of available U-235 samples has made it impossible to observe alpha-gamma-coincidences or to discover U-235 alpha-rays unaccompanied by gamma-rays. Hence, experimental confirmation of the decay scheme proposed below is not at present available.

The alpha-decay of U-235 is supposed to lead in most, perhaps all, cases to an excited state of Th-231 (UY) which rapidly omits a 162 kev gamma-ray to reach the ground state. A half-life for de-excitation longer than ten minutes would have been readily observed. The conversion of the gamma-ray has not been detected and is probably slight.

The decay of UY(Th-231) from its ground state has been investigated and reported separately.[3]

This document is based on work performed for the AEC by Carbide and Carbon Chemicals Corporation, at Oak Ridge, Tennessee.

[3] G. B. Knight and R. L. Macklin, Phys. Rev. **75**, 34 (1949).