Search for the decay of ⁹⁶Zr

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We used the Oak Ridge National Laboratory low-background γ -ray spectrometer in an attempt to measure the decay of ⁹⁶Zr. Less than 0.13 disintegration min⁻¹ was observed from a sample enriched in ⁹⁶Zr; this corresponds to a $t_{1/2} \ge 3.56 \times 10^{17}$ yr and a log *t* value of ≥ 21.6 .

RADIOACTIVITY ⁹⁶Zr (from nature); measured I_{γ} from ⁹⁶Nb; set limit on specific activity and $t_{1/2}$.

There are 19 nuclei that are found in nature even though they are thermodynamically capable of radioactive decay. Eight of these are α emitters including ²³²Th, ²³⁵U, and ²³⁸U. The remaining 11 are β unstable but possess half-lives long compared to the age of the universe, since their only decay mode is a highly forbidden one. Details of many of these "fossil nuclei" have been summarized by Raman and Gove.¹

The half-lives of only two of the four nuclei which can decay by a fourth-forbidden mode have been measured. This note describes an attempt to observe the decay of 96 Zr by means of the Oak Ridge National Laboratory (ORNL) low-background γ -ray spectrometer designed for lunar sample studies.²

Since the Q value for β decay of ⁹⁶Zr is 183 ± 6 keV,³ the only levels of ⁹⁶Nb which can be populated are the ground state (6⁺) and the excited states⁴ at 45 keV (5⁺) and 152 keV (4⁺). The expected β transition is the fourth-forbidden non-unique transition to the 4⁺ 152-keV excited state. The energy of this transition is calculated to be only 31 keV.

We attempted to detect the β decay of ⁹⁶Zr by the presence of its daughter nuclide 23.4-h ⁹⁶Nb, which has a large decay energy (~3.1 MeV) and decays via a series of triple γ -ray cascades.⁵ In the ORNL low-background γ spectrometer,² this pattern of coincident γ -ray transitions can lead to both a high detection sensitivity and an identification of the radionuclide.

The sample consisted of 9.64 g of zirconium enriched to 57.4% 96 Zr. The zirconium as the chemical compound ZrO₂ was compressed into a cylinder and loaded into a plastic holder. The sample was measured in a low-background γ -ray spectrometer for 9573 min, and the background for 6637 min. A preliminary analysis of the data showed that the ZrO₂ sample as received from the ORNL Isotopes Division contained small but detectable amounts of thorium and uranium. Spectrum libraries for calibration of the instrument were prepared from ZrO_2 samples of natural isotopic composition, each containing a known amount of either ⁹⁶Nb, Th, or U.

The data analysis followed the general approach used for lunar sample studies.² The experimental singles and coincidence spectra were fitted by use of a least-squares computer program developed by Schonfeld.⁶ The computer analysis showed that our ability to detect ⁹⁶Nb decays in the enriched zirconium sample was seriously impaired by the presence of γ rays from 3.8 ppm Th and 0.8 ppm U. No ⁹⁶Nb was detected in this experiment. An upper limit of ≤ 0.13 dis min⁻¹ of ⁹⁶Nb was established by least-squares analysis, from which we compute a half-life of $\geq 3.56 \times 10^{17}$ yr, possibly the longest half-life known for a β unstable nucleus.

The energy of 31 keV for the $0^+ \rightarrow 4^+ \beta$ transition and the lower limit for the half-life lead to a value of $\log ft \ge 21.6$. This result is consistent with values¹ of $\log ft$ for the two measured fourth-forbidden nonunique β transitions: 22.6 for the transition ¹¹⁵In($\frac{9}{2}^+$) \rightarrow ¹¹⁵Sn($\frac{1}{2}^+$), and 23.2 for the transition ¹¹³Cd($\frac{1}{2}^+$) \rightarrow ¹¹³In($\frac{9}{2}^+$).

In a search for the double- β decay of 96 Zr. Awschalom⁷ utilized two NaI(Tl) scintillation crystals with an over-all efficiency of about 60% for electrons in the 2.5- to 4.5-MeV region-well above the β -ray end point energy for single β decay of either ⁹⁶Zr or ⁹⁶Nb. He reported the partial half-life of ⁹⁶Zr for double- β decay to be >3.6×10¹⁷ vr. Awschalom might have detected the single- β decay of ⁹⁶Zr via the summing of radiations following ⁹⁶Nb decay. For example, the 749-keV β group with an average energy of ~380 keV might sum with the 812-850-778 keV γ -ray cascade to exceed the 2500-keV discriminator setting. However, such a process would imply a miniscule detection efficiency and thus Awschalom's result has no bearing on the single- β decay $t_{1/2}$ of 96 Zr.

- *Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.
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