Improved Limit on the Electron Capture Decay Branch of ^{176}Lu

E. B. Norman* and E. Browne
Nuclear Science Division, Lawrence Berkeley National Laboratory,
Berkeley, CA 94720 U.S.A.

I. D. Goldman
Instituto de Física, Universidade de Sao Paulo, 05508-900 Sao Paulo, SP, Brazil

P. R. Renne
Berkeley Geochronology Center, 2455 Ridge Road, Berkeley, CA 94709 U.S.A. and Department of Earth and Planetary Science, University of California, Berkeley, CA 94720 U.S.A.

Abstract

We have performed searches for the electron-capture decay branches of ^{176}Lu to the ground state and first excited state of ^{176}Yb. No evidence of either decay mode was observed. From these measurements we have established upper limits on both of these possible branches that are each >20 times more stringent than the single previously published limit.

Keywords: Natural radioactivity, geochronology, electron-capture, Ge detector

***Corresponding author. Tel.: 1-510-486-7846; fax: 1-510-486-6738**

E-mail address: ebnorman@lbl.gov (Eric B. Norman)

1. Introduction

The long-lived naturally occurring nuclide ^{176}Lu ($J^{\pi} = 7^{-}$) β^{-} decays to levels in ^{176}Hf. This decay is the basis for a dating method increasingly applied to rocks and meteorites. The utility of this dating method has been hampered by uncertainty in the half-life, as recently reviewed by Begemann et al. (2001). To some extent the half-life estimates vary systematically according to the approach used for its determination, with the lowest values, (3.50 to 3.57)x10^{10} years, implied from mass spectrometric measurements of Lu- correlated ^{176}Hf excesses in meteorites whose ages were independently known (Patchett and Tatsumoto, 1980; Blichert-Toft et al., 2002; Bizzarro et al., 2003). A half-life value of (3.73 $\pm$ 0.02)x10^{10} years obtained from the direct counting experiment of Nir-El and Lavi (1998) agrees with the (3.72 $\pm$ 0.03)x10^{10} years) value determined by Scherer et al. (2001) through age comparisons for terrestrial rocks. However, Browne and Junde (1998) evaluated the results of a large number of direct counting measurements and recommended a higher value of (4.00 $\pm$ 0.22)x10^{10} years. Moreover, the recent and precise γ-γ coincidence measurement of the ^{176}Lu half-life of (4.08 $\pm$ 0.03)x10^{10} years by Grinyer et al. (2003), which agrees very well with the earlier direct γ-ray counting results of (4.08 $\pm$ 0.24)x10^{10} years by Norman (1980) and (4.05 $\pm$ 0.09)x10^{10} years by Gerke et al. (1990), reaffirms a systematic difference of order 15% in the half-life deduced from these two approaches, which must be resolved before the ^{176}Lu-^{176}Hf method can be established as a reliable geo- and cosmo-chronometer.

It has been suggested that the discrepancies involving age comparisons could be reconciled if ^{176}Lu also underwent significant electron capture decay. ^{176}Lu is unstable with respect to electron-capture decay to ^{176}Yb. The Q_{EC} for decay to the ^{176}Yb ground state is 106.2 keV (Firestone, 1996). Thus, EC decays to both the $J^{\pi} = 0^{+}$ ground state and $J^{\pi} = 2^{+}$ 82-keV first excited state of ^{176}Yb are possible. These EC decay branches would be 7th and 5th forbidden transitions, respectively, and thus are expected to be negligibly small. In fact, there are no observed cases of such highly-forbidden transitions (Singh et al., 1998). The decay scheme of ^{176}Lu is illustrated in Figure 1. The published limit on the EC decay branch of ^{176}Lu of < 10% was reported by Arnold (1954) in his study of the decay of ^{176}Lu. This upper limit was deduced from a search for Yb K x-rays that would be produced by the EC decay of ^{176}Lu. Because of the recent inconsistencies encountered in using the ^{176}Lu/^{176}Hf chronometer, it was felt that a new search for the EC decay of ^{176}Lu was warranted.

2. Experimental

Two plastic bottles, each containing 5 grams of 99.9% pure $LuCl_3 \cdot 6H_2O$ purchased from Alfa-Aesar were placed against the plastic endcap of a 110 cm^3 high-purity germanium detector. X- and γ-ray data from approximately 20 – 800 keV were acquired in 4096 channels for a period of 65 hours using an ORTEC PC-based acquisition system. The energy resolution of this system was measured to be 1.4 keV full width at half maximum at 88 keV. Portions of the spectrum obtained from the Lu sample are shown in figures 2 and 3. The Hf K x rays and the 88-, 202-, 307-, and 401-keV γ rays produced by the β^- decay of ^{176}Lu were clearly observed. However, no evidence of Yb K x rays or 82-keV γ rays was seen. In order to have reference spectra of Yb and Hf K x rays, we also fluoresced samples of Hf metal powder and Yb_2O_3 with 122-kev γ rays from a ^{57}Co source.

3. Results

As can be seen from the decay scheme shown in Figure 1, all of the beta decays of ^{176}Lu eventually produce γ-ray transitions through the 88-keV level in ^{176}Hf. Thus, in order to establish a limit on the EC decay branch to the 82-keV level in ^{176}Yb, we determined both the net area of the 88-keV peak and the gross area of an equal width energy interval centered on 82 keV in the spectrum obtained from the Lu_2O_3 sample. The net area of the 88-keV peak was determined to be N_{88} = 79111 $\pm$ 534 counts, whereas the gross area of the 82-keV region was found to be 46623 $\pm$ 216 counts. Since no peak was observed at 82 keV, we multiplied the gross area of this interval by 2 and then took the square root to obtain a 1σ upper limit, (N_{82} < 305) on the net area of 82-keV peak. Both the 88-keV level in ^{176}Hf and the 82-keV level in ^{176}Yb decay mainly by internal conversion electrons rather than by photon emission. The total internal conversion coefficient, α_T for the 88-keV transition is 5.914, and that for the 82-keV transition is 7.125 (Hager and Seltzer, 1968). Because of the small energy difference between these two transitions, we assumed the same photopeak detection efficiencies for them. Thus, the upper limit we can establish on the ^{176}Lu EC branch to the 82-keV level in ^{176}Yb is

$$B_{82} < \frac{(1 + \alpha_T(82))\, N_{82}}{(1 + \alpha_T(88))\, N_{88}} = \frac{(8.125)(305)}{(6.914)(79111)} = 0.0045 \text{ (or 0.45\%).}$$

The beta decay of ^{176}Lu produces Hf x rays through the atomic vacancies created by internal conversion electrons emitted from the decays of the 998-, 597-, 290-, and 88-keV levels in ^{176}Hf. Hf K_α x rays are emitted in 25.4% of all ^{176}Lu beta decays (Browne and Firestone, 1986). Yb K_α x rays would be produced in 38.4% of all ground state to ground state EC decays of ^{176}Lu (Browne and Firestone, 1986; Schonfeld, 1998). Yb K_α x rays are 3.3 keV lower in energy than those of Hf (Browne and Firestone, 1986). Thus in order to establish an upper limit on the EC decay of ^{176}Lu to the ground state of ^{176}Yb, we determined the net area of the Hf K_α x-ray doublet, N_{Hf} = 331382 $\pm$ 576, and established an upper limit on the net area (N_{Yb}) of a possible Yb K_α x-ray peak in the spectrum obtained from the Lu_2O_3 sample. To establish this upper limit, we added a scaled version of the spectrum obtained from the fluorescence of the Yb_2O_3 sample to the spectrum obtained from counting the Lu_2O_3 sample. We then compared the resulting sum to the original Lu spectrum to see if we could observe "shoulders" on the low-energy sides of the Hf x-ray peaks that would be caused by the presence of the Yb x rays. In the Yb_2O_3 fluorescence spectrum N_{Yb} = 18065 $\pm$ 247. Our upper limit on the scale factor at which we could still detect the Yb x rays in the summed spectrum was 0.10. Again we assumed that because of the small energy difference between these two transitions, the photopeak detection efficiencies for the Yb and Hf K_α x rays were equal. Thus, our 1σ upper limit on the EC decay of ^{176}Lu to the ^{176}Yb ground state is:

$$B_{gs} < [0.10 \times 18065 \times 0.254] / [331382 \times 0.384] = 0.0036 \text{ (or 0.36\%).}$$

4. Conclusion

As expected, no evidence for the EC decay of ^{176}Lu to either the ground state or first excited state of ^{176}Yb was observed. The upper limits we have been able to establish on these possible EC transitions are each >20 more stringent than the single previously published limit of <10% for the EC decay of ^{176}Lu. Our results are also about a factor of two more stringent than that recently obtained from a mass spectrometric search for excess ^{176}Yb in $(1.0 – 2.7)x10^9$ year-old Lu-bearing samples (Amelin and Davis, 2003). Taken together, these results indicate that an explanation for discrepancies between ^{176}Lu half-life estimates based on age comparisons and those based on direct counting methods must be sought elsewhere.

References

Amelin, Y. and Davis, W. J., 2003. Geochemical test for branching decay of ^{176}Lu. *Goldschmidt Conf. Abstracts*, p. A15, Kurashiki, Japan.

Arnold, J. R., 1954. Energy levels in ^{176}Lu and ^{176}Hf. *Phys. Rev.* **93**, 743-745.

Begemann, F., Ludwig, K.R., Lugmair, G.W., Min, K., Nyquist, L.E., Patchett, P.J., Renne, P.R., Shih, C.-Y., Villa, I.M., Walker, R.J.,2001. Call for an improved set of decay constants for geochronological use. *Geochimica et Cosmochimica Acta* **65**: 111-121.

Bizzarro, M., Baker, J.A., Haack, H., Ulfbeck, D., Rosing, M., 2003. Early history of Earth's crust-mantle system inferred from hafnium isotopes in chondrites, *Nature* **421** (6926): 931-933.

Blichert-Toft, J., Boyet, M., Telouk, P., and Albarede, F., ^{147}Sm-^{143}Nd and ^{176}Lu-^{176}Hf in eucrites and the differentiation of the HED parent body, 2002. *Earth and Planetary Science Letters* **204** (1-2): 167-181.

Browne, E. and Firestone, R. B., 1986. *Table of Radioactive Isotopes*. Wiley, New York.

Browne, E. and Junde H., 1998. Nuclear Data Sheets for A=176. *Nucl. Data Sheets* **84**, 337-486 .

Firestone, R.B., 1996. *Table of Isotopes*, 8th ed. Wiley, New York, p. 2092.

Gehrke, R.J., Casey, C., and Murray, R. K., 1990. Half-life of ^{176}Lu, *Phys. Rev.* C **41**, 2878-2882.

Grinyer, G.F., Waddington, J.C., Svensson, C.E., Austin, R.A.E., Ball, G.C., Hackman, G., O'Meara, J.M., Osborne, C., Sarazin, F., Scraggs, H.C., Stöver, H.D.H., 2003. Half-life of ^{176}Lu. *Phys. Rev.* C **67**, 014302/1-4.

Hager, R. S. and Seltzer, E. C., 1968. Internal Conversion Tables. Part I: K-, L-, M-Shell Conversion Coefficients for Z = 30 to Z = 103. *Nucl. Data Tables* **A4**, 1-235.

Nir-El, Y., and Lavi, N., 1998. Measurement of the half-life of ^{176}Lu. *Appl. Radiat. Isot.* **49**,1653.

Norman, E. B., 1980. Half-life of ^{176}Lu, *Phys. Rev.* C **21**, 1109-1110.

Patchett, P.J., and Tatsumoto, 1980. M., Lu-Hf total-rock isochron for the eucrite meteorites, *Nature* **288** (5791): 571-574.

Scherer, E., Munker, C., and Mezger, K., 2001. Calibration of the lutetium-hafnium clock, *Science* **293** (5530): 683-687.

Schonfeld, E., 1998. Calculation of Fractional Electron Capture Probabilities. *Appl. Rad. & Isotopes* **49**, 1353- 1357.

Singh, B., Rodriguez, J. L., Wong, S. S. M., and Tuli, J. K., 1998. Review of Log*ft* Values in β Decay. *Nucl. Data Sheets* **84**, 487-563.

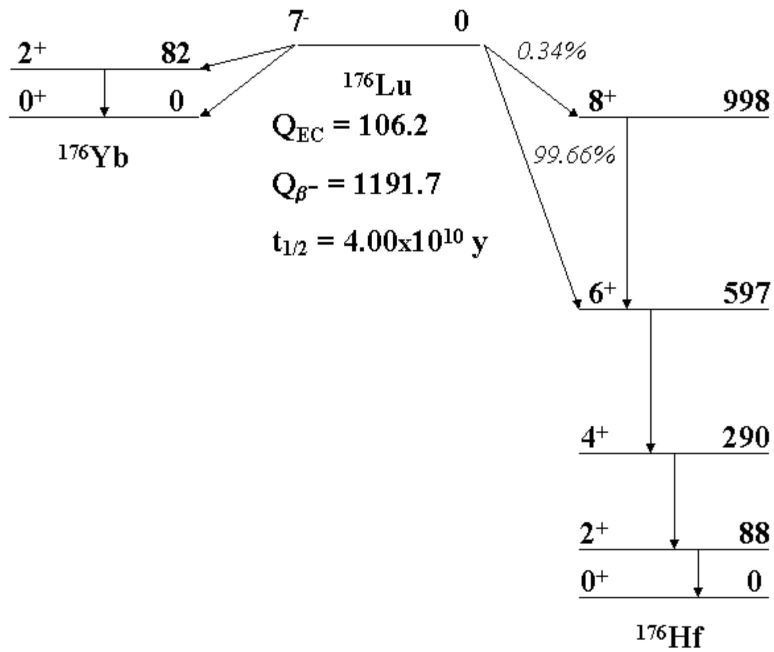

Figure 1. Decay scheme of ^{176}Lu. The possible electron-capture decay branches to the ground and first-excited states of ^{176}Yb are illustrated. All energies are given in keV.

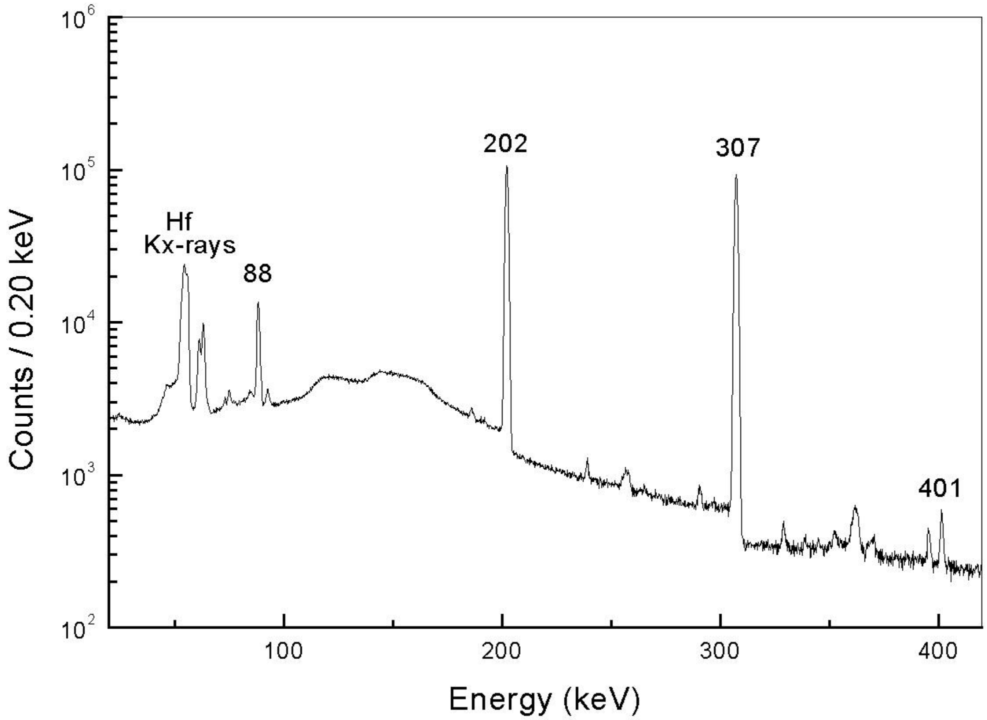

Figure 2. Gamma–ray spectrum observed from the 10-g sample of Lu_2O_3. Peaks attributed to the β^- decay of ^{176}Lu are indicated.

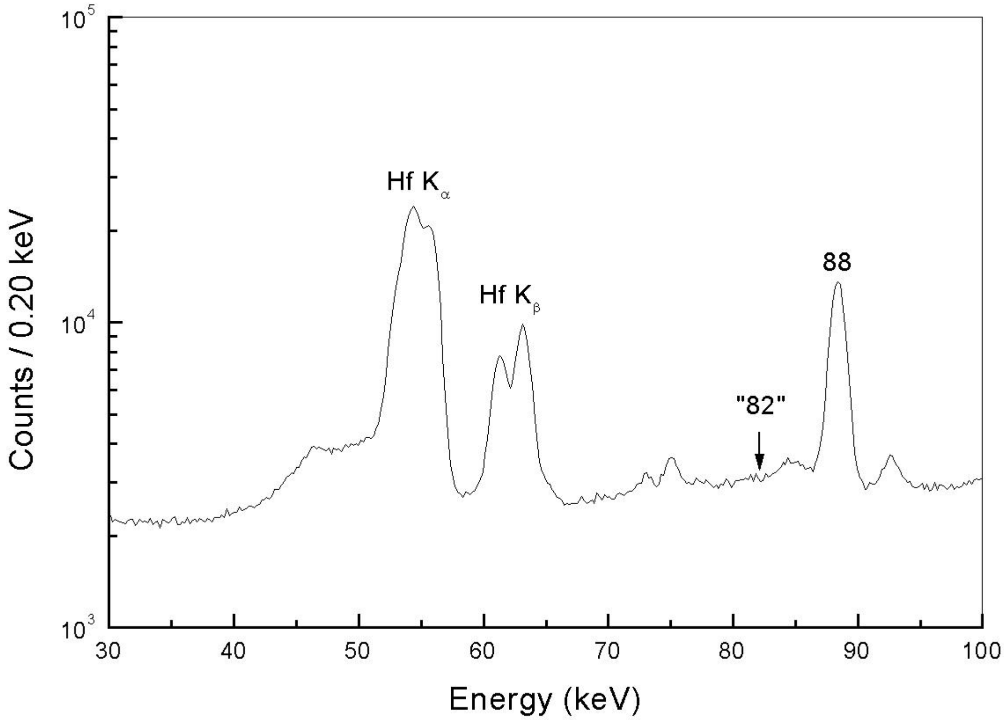

Figure 3. Expanded region of the spectrum observed from the 10-g sample of Lu_2O3. The expected position of the 82-keV γ ray from the possible EC decay of ^{176}Lu is indicated.