ISOTOPIC COMPOSITION OF THORIUM IN LUNAR SAMPLES

J. N. Rosholt, U. S. Geological Survey, Denver, Colorado 80225

Alpha spectrometer measurements of natural thorium in lunar samples are expressed as a comparison of the measured Th^{232}/Th^{230} activity ratio to the Th^{232}/Th^{230} activity ratio expected from the Th^{232}/U^{238} concentration ratio. This expected ratio is calculated by $[Th^{232}/U^{238}]_{atom} \times [\lambda_{232}/\lambda_{238}]$ as previously described (1). Concentrations of uranium, thorium, expected Th^{232}/Th^{230} ratio, measured Th^{232}/Th^{230} ratio, and their quotient for samples that have not been reported previously are listed in Table 1. Uranium and thorium concentrations were taken from Tatsumoto and others (2, 3).

Table 1. Concentrations of uranium, thorium, and radioactivity ratios of thorium isotopes.

Sample	Rock type	U (ppm)	Th (ppm)	Th^{232}/U^{238} (atom ratio)	Th^{232}/Th^{230} (expected activity ratio)	Th^{232}/Th^{230} (measured activity ratio)	Expected ratio / Measured ratio
				Apollo 14			
14063,37*	Breccia (matrix)	3.40	12.64	3.84	1.21	1.19	1.02
14307,26*	Breccia (matrix)	3.36	11.85	3.64	1.16	1.18	.98
14307,26*	Breccia (clast)	4.99	17.29	3.58	1.14	1.18	.96
14318,26	Breccia	3.56	12.46	3.62	1.15	1.18	.97
				Apollo 15			
15071,36*	Fines	.680	2.456	3.73	1.18	1.15	1.03
15080,01*	Fines	.785	2.924	3.85	1.22	1.13	1.08
15515,13	Fines, clod	.974	3.619	3.84	1.22	1.19	1.02
15600,3	Fines	.522	1.889	3.74	1.19	1.16	1.02
15505,25	Breccia	1.01	3.563	3.74	1.19	1.17	1.02
15065,45*	Crystalline	.137	.522	3.96	1.26	1.03	1.22
15076,20*	Crystalline	.153	.590	3.98	1.26	.85	1.48
15085,20*	Crystalline	.118	.459	4.01	1.27	.93	1.37
15476,12*	Crystalline	.192	.733	3.95	1.25	1.11	1.13
15555,10*	Crystalline	.126	.460	3.76	1.19	.98	1.21
				Apollo 16			
66041,22*	Fines	.638	2.546	4.12	1.31	1.08	1.21
66081,19*	Fines	.668	2.764	4.28	1.36	1.15	1.18
68501,45*	Fines	.641	2.533	4.11	1.31	1.24	1.06
67015,11*	Breccia (black clast)	1.21	4.449	3.77	1.20	1.15	1.04
67015,12*	Breccia (matrix)	.200	.732	3.76	1.19	1.08	1.10
64435,57	Breccia	.023	.092	4.19	1.33	.95	1.40
				Apollo 17			
74220,16	Fines	.161	.555	3.57	1.13	1.00	1.13

*Solutions of samples containing U and Th obtained from M. Tatsumoto.

The results of the comparison of measured thorium isotopic ratios to the expected ratios for all samples analyzed from the six Apollo missions are shown in Figure 1. Analyzed samples from Apollo 11, 12, and 14 missions not included in Table 1 were reported (1, 4, 5). Values that exceed unity on the abscissa of this diagram indicate an excess of radioactivity over that expected at the α-particle energy of Th^{230} (4.66 MeV peak) or a deficiency in the alpha-particle intensity of Th^{232} (4.00 MeV peak). I do not attribute the variations in the Th^{232}/Th^{230} ratios to disequilibrium in the U^{238}-U^{234}-Th^{230} decay sequence, because of the equilibrium conditions demonstrated between U^{234} and U^{238} and equilibrium between Pa^{231} and U^{235} in all samples in which these ratios have been measured.

ISOTOPIC COMPOSITION OF THORIUM

Rosholt, J. N.

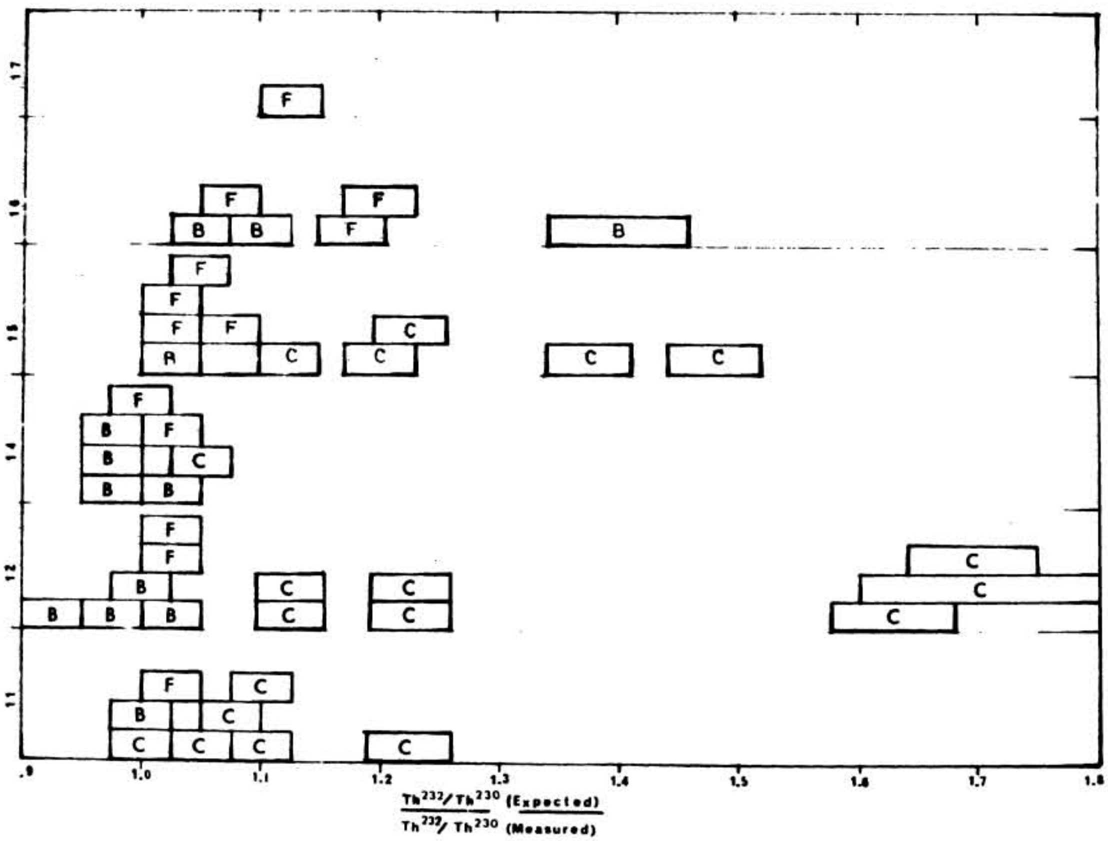

Figure 1. Variations of Th^{232}/Th^{230} ratios in lunar samples with Apollo mission number shown on ordinate. B, breccia; C, crystalline rock, F, fines.

Examination of the fine structure at 4.66 MeV alpha energy was made possible by improved resolution in the alpha spectrometer. Recent measurements on lunar samples that have anomalous Th^{232}/Th^{230} activity ratios indicate that only Th^{230} activity contributes to the 4.66 MeV energy peak. The previous suggestion (1) that an isomer of Th^{232} (Th^{232m}) decayed with 4.7 MeV alpha emission was not correct. Anomalous Th^{232}/Th^{230} activity ratios caused by a deficiency in the alpha-particle intensity of Th^{232} could mean that Th^{232m} decays by gamma transition to Th^{232} at the ground-energy state.

If such an isomer exists, Figure 1 illustrates that the isomeric ratio (Th^{232m}/Th^{232}) appears to be greatest in mare basalts and the least or nil in samples with a large KREEP component such as the breccia samples from Apollo 12 and 14. Samples of fines appear to be intermediate between breccias and igneous rocks; breccias and fines analyzed from Apollo 16 and fines (orange soil) analyzed from Apollo 17 appear to have larger Th^{232m}/Th^{232} ratios than breccias and fines from any of the previous missions.

ISOTOPIC COMPOSITION OF THORIUM

Rosholt, J. N.

Previous reports (1, 6) have discussed clues, regarding the existence of an isomer of Th^{232}, from neutron irradiation of lunar and terrestrial thorium. A significant amount of 5.3 MeV alpha activity was produced. More recent measurements indicate that this activity results from disintegration of Po^{210} (138-day half-life). Gamma spectrometry measurements on samples irradiated with a smaller neutron flux showed that a significant amount of a 412 KeV gamma emitter was produced in thorium separated from lunar samples. A higher activity of the 412 KeV gamma emitter was produced in thorium from lunar samples 15085 and 74220 than in thorium from terrestrial samples. Subsequent irradiations and half-life measurements indicate that this activity actually results from the decay of Au^{198} (65-hour half-life). I have not been able to reproduce a decay of this energy with a 30-hour half-life reported as a preliminary value (6). Unfortunately, both Po^{210} and Au^{198} can be produced by interfering nuclear reactions Bi^{209} (n,γ) $Bi^{210} \xrightarrow{\beta} Po^{210}$ and Au^{197} (n,γ) Au^{198}. These interfering reactions would occur if small amounts of bismuth and gold were present in the purified thorium or in the material upon which the thorium was mounted for irradiation. The amounts of Au^{198} and Po^{210} produced in thorium from selected lunar samples and the lack of this activity produced in thorium from terrestrial samples suggest a correlation with anomalous isotopic composition of thorium. It would appear difficult to produce such correlations from interfering nuclear reactions alone.

REFERENCES

(1) Rosholt, J. N., and Tatsumoto, M. (1971) Proc. Second Lunar Sci. Conf., Geochim. Cosmochim. Acta, Suppl. 2, v. 2, p. 1577-1584.

(2) Tatsumoto, M., Hedge, C. E., Doe, B. R., and Unruh, D. M. (1972) Proc. Third Lunar Sci. Conf., Geochim. Cosmochim. Acta, Suppl. 3, v. 2, p. 1531-1555.

(3) Tatsumoto, M., Hedge, C. E., Knight, R. J., Unruh, D. M., and Doe, B. R. (1972) The Apollo 15 Lunar Samples (editors, J. W. Chamberlain and C. Watkins), Lunar Science Institute, p. 391-393.

(4) Rosholt, J. N., and Tatsumoto, M. (1970) Proc. Apollo 11 Lunar Sci. Conf., Geochim. Cosmochim. Acta, Suppl. 1, v. 2, p. 1499-1502.

(5) Rosholt, J. N. (1972) Lunar Science-III (editor, C. Watkins), Lunar Science Institute Contribution No. 88, p. 663-665.

(6) Rosholt, J. N., and Tatsumoto, M. (1972) Lunar Science-IV (editors, J. W. Chamberlain and C. Watkins), p. 634-636.

(7) I thank Philip Aruscavage and Charles Bush for their help in obtaining numerous gamma-ray spectra of neutron-irradiated samples.