ISOTOPIC COMPOSITION OF URANIUM, THORIUM, AND PROTACTIONIUM


U-Pa-Th. As found for 14163, (1) breccia and fines samples 14318, 15505, 15515, and 15600 have Pa\(^{231}\) in radioactive equilibrium with U\(^{235}\). The U\(^{234}\) daughter is in equilibrium with the U\(^{238}\) parent in these samples as it is in breccia samples 14063 and 14307. (2) The Th\(^{232}/\text{Th}\(^{230}\) activity ratios in these Apollo 14 breccia samples indicate that Th\(^{230}\) is in equilibrium with parent U\(^{238}\), and no anomalous thorium isotopic composition was found.

The situation is different for Apollo 15 crystalline rocks that were analyzed because none have Th\(^{232}/\text{Th}\(^{230}\) activity ratios that agree with the expected ratios calculated from Th\(^{232}/\text{U}\(^{238}\). Concentrations of uranium and thorium in the Apollo 15 samples have been published. (3) Samples 15065, 15076, 15085, 15476, and 15555 have Th\(^{232}/\text{Th}\(^{230}\) activity ratios of 1.0 or less in comparison to expected ratios of about 1.26 as calculated from the observed values of Th\(^{232}/\text{U}\(^{238}\). This set of thorium isotopic results for Apollo 15 crystalline rocks is similar to the thorium isotopic results reported for Apollo 12 crystalline rocks. (4) Apollo 15 fines and breccias 15071, 15080, 15085, 15515, and 15600, similar to fines 12070, (4) have Th\(^{232}/\text{Th}\(^{230}\) activity ratios slightly less than or equal to the expected Th\(^{232}/\text{Th}\(^{230}\) ratios calculated from the Th\(^{232}/\text{U}\(^{238}\) ratios. Thus, the same interpretations, regarding results of thorium isotopic composition, appear valid for Apollo 15 samples as those described for Apollo 12 samples; (3) namely, the differences in thorium isotopic composition between different sample types suggest the existence of a Th\(^{232}\) isomer of variable isomeric abundance in lunar samples. If such an isomer exists, the isomeric ratio (Th\(^{232m}/\text{Th}\(^{232}\)) appears to be greatest in mare basalts and the least or nil in samples with a large KREEP component such as sample 12033. (5)

Clues Regarding Existence of an Isomer of Th\(^{232}\). Of special interest are observations resulting from neutron irradiation of purified thorium separated from lunar rocks. Previous to irradiation with a total flux of approximately \(5 \times 10^{18}\) neutrons, purified thorium had been evaporated from TTA (Thenoyl trifluoroacetone--complexed thorium) on pure aluminum discs. The purpose of this investigation is to determine if a suspected isomer of Th\(^{232}\) (Th\(^{232m}\)) can be distinguished from Th\(^{232}\) by a difference in the reaction products produced from nuclear irradiation of thorium.
The $^{232}\text{Th}(n,\gamma)^{233}\text{Th} \stackrel{\beta^-}{\rightarrow}^{233}\text{Pa} \stackrel{\beta^-}{\rightarrow}U^{233}$ reaction will produce a measurable quantity of alpha-particle emitting $U^{233}$ on the counting disc. In addition to the expected $U^{233}$ produced, a relatively large and unexpected activity of 5.3 MeV alpha emitter was also produced in some lunar thorium. Extended half-life measurements of 5.3 MeV activity have been made and are being continued on aliquots of thorium extracted from rock 14310. Preliminary measurements on aliquots irradiated at different times indicate that the half-life of anomalous 5.3 MeV activity is approximately 180 days, and the extrapolated 5.3 MeV/$U^{233}$ activity ratio is about 0.5 for this sample. Irradiation of thorium extracted from sample 12033 produced a 5.3 MeV/$U^{233}$ activity ratio of less than 0.08. Very lengthy half-life measurements are required to distinguish the anomalous 5.3 MeV activity from interfering activity of $^{210}\text{Po}$ (5.30 MeV, 138.4-day half-life). Neutron capture by bismuth impurities in the metal discs can produce interfering $^{210}\text{Po}$ activity by the reaction $^{209}\text{Bi}(n,\gamma)^{210}\text{Bi} \stackrel{\beta^-}{\rightarrow}^{210}\text{Po}$. Most of the efforts to find the proper irradiation conditions and backing materials to be used in the investigation of the properties of irradiated thorium have been on terrestrial thorium standards. The aluminum discs now being used (99.999% Al) apparently contain only very small amounts of bismuth impurities, if any, but previously used discs of titanium and molybdenum contain a sufficient amount of bismuth impurities to produce more $^{210}\text{Po}$ interference than could be tolerated. Previously reported activity at the 5.3 MeV alpha energy in irradiated terrestrial thorium plated on titanium has been found to be virtually all $^{210}\text{Po}$.

Additional clues regarding the existence of an isomer of $^{232}\text{Th}$ ($^{232}\text{Th}^{m}$) in lunar rocks have been found in purified lunar thorium that was irradiated with epithermal neutrons; the aluminum discs, on which thorium had been evaporated, were shielded from thermal neutrons by their encapsulation in a cadmium cup. A relatively high activity of 412 KeV gamma emitter was found by gamma-ray spectroscopy using a Ge(Li) detector. Preliminary measurements on 14310 and 15085 indicate that the 412 KeV gamma emitter is an isomer of $^{233}\text{Pa}$ ($^{233}\text{Pa}^{m}$) that decays by isomeric transition (I. T.) to ground-state $^{233}\text{Pa}$ with approximately 30-hour half-life. Interference at this energy could be contributed by $^{198}\text{Au}$ produced by neutron capture in gold; however, $^{198}\text{Au}$ has a half-life of 64.8 hours and the difference between the 30-hour and 64.8-hour decay rates can be used to distinguish the two isotopes of similar gamma energy.

Interpretation of the data indicates the following tentative neutron-capture and decay scheme of an isomeric series collateral to that of ground-state $^{232}\text{Th}$. 

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ISOTOPIC COMPOSITION OF U, Th, AND Pa

Rosholt, John N.

\[
\begin{align*}
\text{Th}^{232}_{\eta, \gamma} &\rightarrow \text{Th}^{233}_{\text{m}} \rightarrow \text{Pa}^{233}_{\text{m}} \\
\text{Th}^{232}_{\eta, \gamma} &\rightarrow \text{Th}^{233} \rightarrow \text{Po}^{233} \rightarrow \text{U}^{233} \\
&\rightarrow \text{Th}^{229}
\end{align*}
\]

REFERENCES AND NOTES


(6) We thank Philip Aruscavage and Charles Bush for their help in obtaining numerous gamma-ray spectra of neutron-irradiated samples. Publication authorized by the Director, U. S. Geological Survey.