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The isotopic composition of Lu in meteorites and lunar rocks: implications for the decay constant of ^{176}Lu

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Recent determinations of the ^{176}Lu decay constant λ^{176}Lu made by age comparison against the U-Pb chronometer display a dichotomy between values derived from $<$4 billion year old terrestrial samples on the one hand (1.867 x 10^{-11}yr^{-1}, [1-3]), and those derived from $>$4.5 billion year old meteorites on the other ($\sim$ 1.96 x 10^{-11}yr^{-1}, [2, 4-7]). For a given set of Lu-Hf parameters (^{176}Hf/^{177}Hf and ^{176}Lu/^{177}Hf) for the bulk silicate Earth, this $\sim$5% difference results in vastly different early-Earth differentiation scenarios as inferred from the initial εHf values of Archean and Hadean rocks and zircons [e.g., 1,7]. Settling the discrepancy between "terrestrial" and "meteorite" λ^{176}Lu values has therefore become a key issue in understanding the geochemical evolution of the early Earth.

One hypothesis for explaining the difference between λ^{176}Lu values is that the decay rate of ^{176}Lu to ^{176}Hf was briefly enhanced through the excitation of ^{176}Lu to its short-lived isomer by gamma irradiation of early solar system materials before they coalesced into larger bodies [8]. Such a mechanism could have generated significant initial heterogeneity in Hf isotopic composition and perhaps even the end-members of a positively-sloped mixing line upon which meteorite isochrons would later be superimposed. These hypothetical "isochrons" would be too steep, yielding erroneously high λ^{176}Lu values. (We note, however, that meteorite suites from parent bodies that underwent early, large-scale melting and isotopic re-equilibration should not show this effect.) Given the difference between the observed and expected (i.e., at 4.56 Ga; λ^{176}Lu = 1.867 x 10^{-11}yr^{-1}) slopes of published meteorite isochrons, the

burnout of ^{176}Lu, if it occurred, might manifest itself as a slight (∼0.1-0.4%) variability among the ^{176}Lu/^{175}Lu of Earth, Mars, the moon, chondrites, and eucrites. Relative to these planetary bodies, the ^{176}Lu/^{175}Lu of non-irradiated material should be 0.3-0.7% higher). Previous TIMS-based studies [4,9] have not resolved such Lu isotope anomalies in meteorites.

To search further for anomalies, we used an MC-ICPMS technique to measure the Lu isotopic compositions of several terrestrial and extraterrestrial materials, including a Martian meteorite, 6 lunar samples (low- and high-Ti basalts, a KREEP basalt, and a soil), 2 eucrites, 2 ordinary- and 2 carbonaceous chondrites, an Allende CAI, and BIR-1. Lutetium was separated from the digested samples using a 50W-x12 cation exchange column, yielding a heavy rare-earth element fraction containing mostly Yb and Lu. Although such naturally occurring Yb is now commonly used to apply a mass bias correction to Lu during isotope dilution measurements by MC-ICPMS [e.g., 6], corrections for the isobaric interference of ^{176}Yb on ^{176}Lu are large for non-spiked Lu samples because of the low natural abundance (∼2.6%) of ^{176}Lu. To overcome this difficulty, and to eliminate the need to assume invariable Yb isotope composition among all samples, we processed the HREE through an additional column (αHIBA, [10]) to separate Yb from Lu. Admixed Re and standard-sample bracketing were used to correct for mass bias during MC-ICPMS analysis. The mean ^{176}Lu/^{175}Lu values for all the individual parent bodies represented by our samples lie within a narrow range of +0.05% to -0.03% relative to our terrestrial AMES Lu standard solution and are indistinguishable from each other given the ∼0.1% (2 s.d.) external reproducibility for multiple digestions of terrestrial rock standards. Though our preliminary data set contains no anomalies of a magnitude that would clearly support the gamma irradiation hypothesis of [8], it does not rule out such a possibility either. Internal Lu-Hf isochron data for individual meteorites will hopefully provide further constraints on the causes of the λ^{176}Lu discrepancy.

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