Extinct radionuclide decay signatures and mantle evolution

C.L. Harper Jr.
S.B. Jacobsen

Department of Earth and Planetary Sciences, Harvard University, 20 Oxford Street, Cambridge, MA 02138, USA.

Introduction

We are developing three promising extinct radionuclide systems for studies of early Earth differentiation and geodynamics: ^{146}Sm-^{142}Nd ($t_{1/2}$ = 103 Ma), ^{182}Hf-^{182}W ($t_{1/2}$ = 9 Ma), and ^{244}Pu-Ru,Pd ($t_{1/2}$ = 80 Ma). Each of these systems has potential for advancing understanding of different aspects of Earth history. The utility of an extinct isotope system is determined by four factors. First, the cosmo/geochemical properties of the parent/daughter element pair determine the potential advantages or disadvantages of the system for investigating different modes of planetary differentiation and geodynamic transport. Second, the half-life of the parent defines the timescale for development of isotopic differences between differentiated reservoirs at early times. Relevant 'boundary conditions' of Earth history (accretion rate, giant impacts, late additions, geodynamic mixing rates, etc.) are a third generic factor and set the stage for the geochemical behavior associated with the events and processes one seeks to understand. The final factor is the range of radiogenic isotope effects in rocks which determines the practicality of signal resolution by high precision isotopic measurements. We have focused recently on developing mass spectrometry for Nd and are now able to reproduce some isotope ratios to better than ± 4 ppm (± 0.04 ε-units) at the 2σ level for single measurements. This experience has encouraged us to think optimistically about applications requiring precise measurement capabilities of radiogenic isotope effects at the 5 to 10 ppm level.

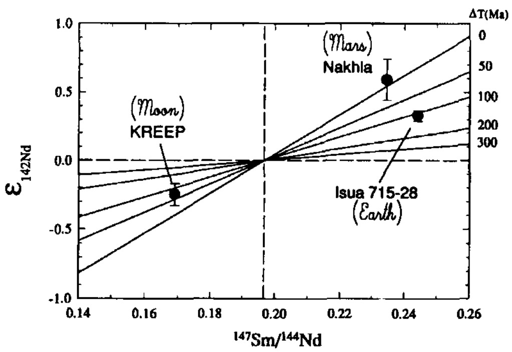

FIG. 1. ^{146}Sm-^{142}Nd fossil isochron diagram.

^{146}Sm-^{142}Nd

The presence of a small but significant abundance of ^{146}Sm in the early solar system provides a chronometry particularly well suited for dating very early episodes of differentiation in planetary bodies (Harper and Jacobsen, 1992). Our approach is based on the preservation of isotopic signatures in large-scale subcrustal reservoirs and is advantageous in cases where early-formed crustal samples are either unavailable, as in the case of Mars (for sampling reasons), and the Earth (for tectonic reasons), or severely disturbed, as in the case of the Moon. In principle, this system is ideal for dating differentiation episodes in the silicate portions of planetary bodies because other large scale cosmochemical processes such as volatile depletion or core formation do not fractionate Sm/Nd. Large-scale merger events are expected to rehomogenize the silicate portion of the merged object and reset the isotopic clock. Consequently the age of the oldest differentiated mantle reservoir should provide a lower limit on cessation of hierarchical accretion series. These properties and linkage to the long-lived ^{147}Sm-^{143}Nd system make the ^{146}Sm-^{142}Nd system attractive.

Our first order question is to determine a lower limit to the age of the last big merger event in the accretion history of the Earth for comparison with parallel efforts to determine the age of the Moon. A summary of the most significant ^{142}Nd data for the Earth, Moon (and Mars) is shown in figure 1. The terrestrial datum is from the 3.81 Ga old Isua sample 715-28, which appears to have been derived from a mantle reservoir depleted very early in Earth history (Harper and Jacobsen, 1992; Sharma, *et al.*, 1994). The lunar KREEP datum is from Harvard measurements of two 3.85 Ga old KREEP basalts (14078 and 15386). The ^{142}Nd shift in KREEP probably dates the late-stage freeze-up of the lunar magma ocean. The Mars datum is for Nakhla (Harper *et al.*, in prep.). Mars appears to have accreted rapidly (by 4.54 Ga). The inferred accretion interval may be a reasonable approximation to the timescale for formation of Mars-sized objects in the Earth's accretion series. Model ages for the Isua and lunar 'urKREEP" sources agree at 4.47 Ga, delimiting the interval for formation of the Earth-Moon system to be < ~100 Ma.

A second objective is to know why so few early Archean samples, (many of which apparently came from much older depleted mantle sources), have normal ^{142}Nd. Chondritic ^{142}Nd in 3.7 to 4.0 Ga old rocks may be explained in two ways. Either the actual initial ε_{143Nd} for the source(s) is not high, (*ergo* no problem to begin with), or very strongly-fractionated depleted source reservoirs formed at ~4.2 Ga. If we assume the former explanation, then we again have two different scenarios. The chondritic ^{142}Nd sources were either parts of the mantle that remained undifferentiated through the early Hadean, or products of geodynamic remixing. In the latter case, the non-chondritic 715-28 source was isolated (mantle lithosphere?). We also wish to address the question of the initial mode of origin of the high-^{142}Nd Isua 715-28 mantle source. Did it form by partial melting to form 'primordial crust', or by magma ocean differentiation (crystal fractionation)?

^{244}Pu-Pd,Ru

^{244}Pu-Pd,Ru chronometry is based on the spontaneous fission of ^{244}Pu. Pu is a refractory lithophile element expected to follow Nd in most differentiation processes, and Pd and Ru are both refractory and highly siderophile. Equilibrium partitioning during segregation of metal and sulfide into the core is expected to have depleted Pd and Ru by factors $> 10^6$. Accretion of a 'late' (post-core formation) 'veneer' has been suggested to explain the relatively small and similar mantle depletion factors (~300) observed for all the highly siderophile refractory elements. The radiogenic component from ^{244}Pu in Ru and Pu isotopes ranges from 3 to 10 ppm for $f^{Pu/Ru,Pd}$ = 300. Notable features of the veneer scenario are: (i) initial deposition of highly siderophile element mantle budgets at the early Earth's surface; and (ii) geodynamic mixing into the mantle over geological time. There is tentative evidence that the mixing remains incomplete today. The interesting prospect is that early source reservoirs may have had much larger $f^{Pu/Ru,Pd}$ and proportionally greater radiogenic effects. Pd and Ru isotopes might therefore provide a tracer of the tectonic/geodynamic cycle over Earth history and may be particularly useful in addressing the question of whether or not contemporary-style plate tectonics was operative in the Hadean. They may also provide a test between some of the scenarios mentioned in the previous section. For example, if the 715-28 source is a residue of partial melting to form the protocrust, and if the veneer component was partly sequestered within the protocrust, then ^{142}Nd should correlate with Pd and Ru isotopes.

^{182}Hf-^{182}W

Hf is a refractory lithophile element, whereas W is refractory, moderately siderophile and depleted relative to Hf by a factor of 15 to 30 due to partitioning into the core. A reasonable assumption about most of the history of core growth is that it was rate-determined by accretion. Consequently the ^{182}Hf-^{182}W system should provide a chronometer of both terrestrial accretion and core formation. In principle one would want to determine the initial abundance of ^{182}Hf in the early solar system first from internal isochron meteorite studies. However, for technical reasons we are initially using the Hf/W fractionation in the silicate Earth to look for evidence of live initial ^{182}Hf against W from large samples of the Toluca iron meteorite used as a proxy for the solar system initial composition. A W shelf standard represents the silicate Earth. We measure $^{182}W/^{183}W$ as WO^{3-}, (normalized to $^{184}W/^{183}W$), by static multicollection with simultaneous $^{18}O/^{16}O$ determination. The standard exhibits an apparent radiogenic mass-182 shift of ~3 ± 1 ε-units relative to Toluca, confirming previous results (Harper *et al.*, 1991). Neutron transport modeling (Masarik and Reedy, pers. comm.) suggests this effect is too large to be due to neutron capture burnout during space exposure. If our measurements are accurate (and we have no evidence of interferences), then the isotopic measurements indicate that ^{182}Hf was live in the early solar system, and that the early stages of terrestrial accretion proceeded rapidly (*viz.*, within a few ^{182}Hf half-lives).

References

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