

^{176}Lu - ^{176}Hf ZIRCON EVIDENCE FOR RAPID LUNAR DIFFERENTIATION. Dianne J. Taylor¹, Kevin D. McKeegan¹ and T. Mark Harrison^{1,2} ¹Dept. of Earth and Space Sciences & IGPP, UCLA, Los Angeles, CA, 90095, dtaylor@ess.ucla.edu. ²Research School of Earth Sciences, Australian National University, Canberra, A.C.T. 0200

Introduction: The most accessible record of the earliest phases of planetary formation and evolution is preserved in rocks returned from the surface of the Moon by the Apollo missions. Examination of their mineralogy and chemical compositions suggests that a large portion of the Moon was initially completely molten due to the high temperatures associated with its formation [1]. A timescale for the cooling and solidification (and accompanying chemical differentiation) of this planetary-sized magma body has not been firmly established, with estimates based on various isotope systems (both short-lived and long-lived) ranging from ~30 to 300 million years after Solar System formation (e.g. [2, 3, 4, 5, 6]). Here we report a coupled U-Pb and ^{176}Lu - ^{176}Hf isotope study of individual lunar zircons from the KREEP-rich Apollo 14 landing site. As the final layer of the lunar magma ocean (LMO) to solidify, a closure age for the so-called KREEP reservoir would establish an upper bound for the duration of LMO crystallization. The zircon crystallization ages and $\epsilon_{\text{Hf}(T)}$ values from our study yield a separation age of the KREEP source region of 4505^{+36}_{-24} Ma (1σ , Fig. 1b), implying that primary differentiation of the Moon was complete within 60 million years of the formation of CAIs [7].

Samples and technique: The lunar zircons analyzed in this study were isolated from the clast-rich polymict breccias 14304, 14305 and 14321. We utilized ion microprobes to obtain crystallization ages (^{207}Pb - ^{206}Pb and U-Pb ages) in $\sim 20 \mu\text{m}$ spots of each zircon as well as rare earth element (REE) and Ti concentrations on select samples (ages, REE and Ti crystallization temperatures were previously reported in [8]). Lu and Hf isotope analyses were undertaken by using laser ablation inductively-coupled plasma mass spectrometry (LA-ICPMS) according to the techniques described by Harrison et al. [9].

The Apollo 14 zircons show a distribution of crystallization ages ranging from 3.9 Ga to 4.4 Ga. With the exception of 4 zircons, all U-Pb ages are concordant at the 4% level or better, which we take as evidence that the ^{207}Pb - ^{206}Pb ages correspond to original crystallization ages. The lack of zoning and core-rim relationships in the zircons give us confidence in relating the Hf isotope composition to the ^{207}Pb - ^{206}Pb age used to calculate $\epsilon_{\text{Hf}(T)}$.

Results and discussion: Our ^{176}Hf / ^{177}Hf data for 29 zircons (Fig. 1) show $\epsilon_{\text{Hf}(T)}$ values ranging from -7

to +0.3. The lunar zircon results extend the age range over which Hf isotope data are available for the Moon back to 4.35 Ga and represent the least radiogenic Hf results thus far in lunar materials. Although there is some scatter toward less negative $\epsilon_{\text{Hf}(T)}$, which is not unexpected given the likelihood of some reservoir mixing, the data show a clearly defined trend in negative $\epsilon_{\text{Hf}(T)}$ vs. age with a distinct lower bound (which, however, does not transgress into the ‘forbidden region’ corresponding to $\text{Lu/Hf} < 0$). Such a correlation testifies to the existence of a coherent enriched source region which was isotopically isolated from mantle sources and continued to evolve independently over time. The well-resolved negative $\epsilon_{\text{Hf}(T)}$ values, in conjunction with the REE concentrations in these zircons [10], clearly point toward a KREEP source for the magmas from which the zircons crystallized.

We can derive a two-stage model age for the isotopic closure of the KREEP source region from the intersection of the linear trend with the chondritic evolution line (CHUR). We assume that the Moon formed with a chondritic Lu/Hf ratio, which steadily decreased as the LMO cooled and crystallized until a minimum Lu/Hf ratio is attained in the ITE-enriched KREEP reservoir. If the KREEP reservoir remains chemically isolated after formation, then later-crystallizing minerals containing a KREEP component will reflect a time-integrated subchondritic ^{176}Lu / ^{177}Hf signature. An estimate for the formation of the KREEP reservoir is provided by a fit to the 23 least radiogenic data points, yielding a model age of 4505^{+36}_{-24} Ma (1σ) and corresponding $^{176}\text{Lu}/^{177}\text{Hf} = 0.014 \pm 0.002$ (given by the slope of the fit line). The scatter of the data about this evolution line is roughly commensurate with analytical uncertainties (MSWD = 1.29), and the error on the intersection with CHUR is determined from the 1σ error envelope propagated around the regression line. Notably, the $^{176}\text{Lu}/^{177}\text{Hf}$ inferred from this fit is within uncertainty of the lower bound for $^{176}\text{Lu}/^{177}\text{Hf}$ of KREEP estimated by Warren [11] (= 0.01420) thus lending confidence that these zircons represent the KREEP source region. An absolute lower limit on the closure age of KREEP can be obtained from the intersection of the $\text{Lu/Hf} = 0$ evolution line with the least radiogenic $\epsilon_{\text{Hf}(T)}$ zircon data; our data require a closure age no younger than 4.45 Ga.

The zircon $\epsilon_{\text{Hf}(T)}$ data are not consistent with the younger magma ocean crystallization age determined by using the coupled ^{147}Sm - ^{143}Nd / ^{146}Sm - ^{142}Nd system (4.352 ± 0.023 [5]) but are consistent with ^{182}Hf - ^{182}W data suggesting an LMO crystallization age of 4.527 ± 0.010 Ga [4] (although that result has recently been called into question [12]) and with the requirement that the lunar mantle be shielded from accretion of highly siderophile elements prior to 4.4 Ga [13]. The younger ^{142}Nd age may indicate resetting of the Sm-Nd system and/or component mixing in bulk analyses. Thermal models of magma ocean evolution which suggest that it may have taken up to 200 million years for a magma layer insulated by a plagioclase crust to cool [14] also do not appear to be viable, according to our estimate. More recent thermal models [15, 16] suggest that LMO cooling can proceed quite rapidly if the crust is fractured by repeated impacts during this early period in the Moon's history.

Our results do not strictly require that the entire magma ocean had completely crystallized by 4.50 Ga. However, in order for the enriched signature seen in the Apollo 14 zircons to have evolved, the LMO must have proceeded to greater than 99% crystallization by this time. Once formed, this KREEP reservoir must have remained isolated from any depleted Hf-isotope signature originating from the lower part of the lunar mantle. At 4.50 Ga, the KREEP layer could still have been a mush of liquid and crystals which did not communicate with the lower mantle or the over-riding plagioclase crust, at least not in the Procellarum KREEP terrain region of the Moon sampled by the Apollo 14 mission. Other regions of the Moon's crust,

during this time, could still have experienced some remelting through interaction with the upper mantle, leading to the formation of new crustal rocks, either Mg-suite rocks or ferroan anorthosites. Thus, the <4.50 Ga ages found for some ferroan anorthosites [6], are not inconsistent with an early crystallization age of the magma ocean. Our data indicate that geophysical models of magma ocean crystallization should be revised to account for a short timescale of differentiation of no more than a few tens of millions of years following the Moon-forming impact.

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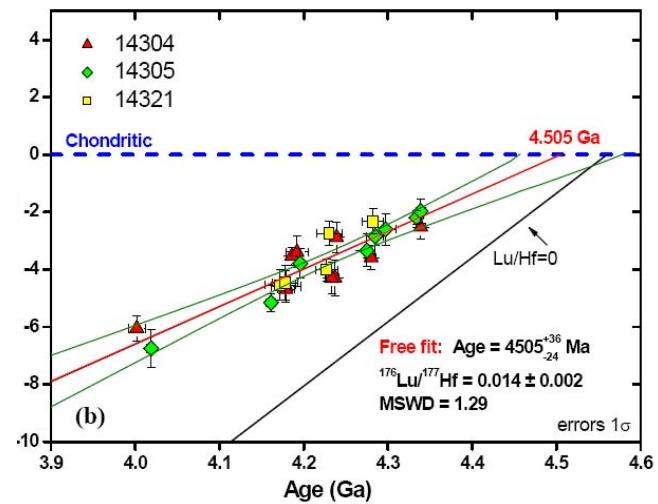
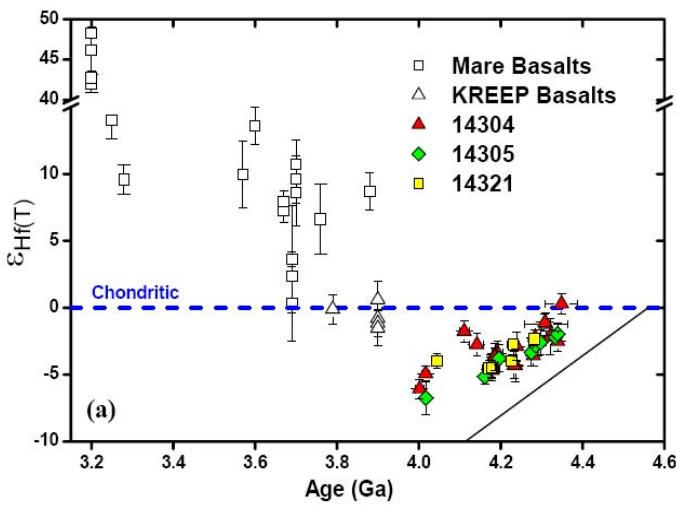


Figure 1. $\epsilon_{\text{Hf}(T)}$ vs. T (a) LA-MC-ICPMS *in situ* data for lunar zircons from this study along with bulk TIMS data for mare and KREEP basalts from the literature [17-20] (grey data points). (b) Subset of the 23 least radiogenic data points. A weighted least-squares fit yields a closure age of KREEP of 4505^{+36}_{-24} Ma (1σ), $^{176}\text{Lu}/^{177}\text{Hf} = 0.014 \pm 0.002$. Also plotted is the $^{176}\text{Lu}/^{177}\text{Hf} = 0$ evolution line which marks the boundary of the “forbidden zone.” Datapoint errors are 1 sigma; the confidence interval on the fit is 95%.