



Tungsten isotopes and the origin of the Moon



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ABSTRACT

The giant impact model of lunar origin predicts that the Moon mainly consists of impactor material. As a result, the Moon is expected to be isotopically distinct from the Earth, but it is not. To account for this unexpected isotopic similarity of the Earth and Moon, several solutions have been proposed, including (i) post-giant impact Earth–Moon equilibration, (ii) alternative models that make the Moon predominantly out of proto-Earth mantle, and (iii) formation of the Earth and Moon from an isotopically homogeneous disk reservoir. Here we use W isotope systematics of lunar samples to distinguish between these scenarios. We report high-precision ^{182}W data for several low-Ti and high-Ti mare basalts, as well as for Mg-suite sample 77215, and lunar meteorite Kalahari 009, which complement data previously obtained for KREEP-rich samples. In addition, we utilize high-precision Hf isotope and Ta/W ratio measurements to empirically quantify the superimposed effects of secondary neutron capture on measured ^{182}W compositions. Our results demonstrate that there are no resolvable radiogenic ^{182}W variations within the Moon, implying that the Moon differentiated later than 70 Ma after Solar System formation. In addition, we find that samples derived from different lunar sources have indistinguishable ^{182}W excesses, confirming that the Moon is characterized by a small, uniform $\sim +26$ parts-per-million excess in ^{182}W over the present-day bulk silicate Earth. This ^{182}W excess is most likely caused by disproportional late accretion to the Earth and Moon, and after considering this effect, the pre-late veneer bulk silicate Earth and the Moon have indistinguishable ^{182}W compositions. Mixing calculations demonstrate that this Earth–Moon ^{182}W similarity is an unlikely outcome of the giant impact, which regardless of the amount of impactor material incorporated into the Moon should have generated a significant ^{182}W excess in the Moon. Consequently, our results imply that post-giant impact processes might have modified ^{182}W , leading to the similar ^{182}W compositions of the pre-late veneer Earth's mantle and the Moon.

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1. Introduction

The Moon is generally thought to have formed from debris produced during a giant impact between the proto-Earth and a roughly Mars-sized body (Cameron and Benz, 1991; Canup and Asphaug, 2001; Hartmann and Davis, 1975), but important details about this model remain incompletely understood. Recent work has focused on reconciling observed isotopic similarities between the Earth and the Moon (Armitage et al., 2012; Wiechert et al., 2001; Young et al., 2016; Zhang et al., 2012) with the prediction that in the canonical giant impact model the Moon predominantly consists of impactor material (Canup and Asphaug, 2001). Consequently, in this model the Moon is expected to show an isotopic difference relative to the Earth (Pahlevan and Stevenson, 2007), in

marked contrast with the observed isotopic similarity. To explain this paradox, a variety of solutions have been proposed, including (i) giant impact models in which the Moon predominantly derives from the proto-Earth (Canup, 2012; Ćuk and Stewart, 2012; Reufer et al., 2012), (ii) post-giant impact isotopic equilibration (Pahlevan and Stevenson, 2007), and (iii) derivation of proto-Earth and impactor from the same isotopically homogeneous reservoir in the inner solar system (Dauphas et al., 2014; Mastrobuono-Battisti et al., 2015; Wiechert et al., 2001).

The short-lived ^{182}Hf – ^{182}W system ($t_{1/2} = 8.9$ Ma) is particularly useful to examine these issues and constrain models of lunar origin. This is because the Moon-forming impact involved mixing between impactor and proto-Earth components with presumably different ^{182}W compositions. These radiogenic ^{182}W variations result from Hf/W fractionation during core formation, which for the impactor and proto-Earth likely occurred at different times and under different conditions, ultimately leading to distinct ^{182}W com-

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positions of proto-Earth's mantle and the impactor's mantle and core (e.g., Kleine et al., 2009). Mixing of these three components during the giant impact, therefore, likely induced a ^{182}W difference between the Moon and the immediate post-giant impact mantle of the Earth. This also holds true if the Moon predominantly consists of proto-Earth mantle material, because the pre- and post-giant impact ^{182}W compositions of Earth's mantle were likely different (Kruijjer et al., 2015).

Determining the ^{182}W composition of the Moon has proven difficult, because most lunar samples show cosmogenic ^{182}W excesses from neutron capture on Ta during prolonged exposure of the lunar samples to galactic cosmic rays (Leya et al., 2000). One way to overcome this problem is to analyze lunar metals, which are devoid of Ta-derived cosmogenic ^{182}W and hence should define the pre-exposure ^{182}W compositions directly (Kleine et al., 2005; Touboul et al., 2015, 2007). Using this approach, Touboul et al. (2007) neither found statistically meaningful ^{182}W differences among different lunar rock types nor between these samples and the present-day bulk silicate Earth (BSE). Nevertheless, two more recent studies, using higher precision measurement techniques, demonstrated that the Moon has a ^{182}W excess of ~ 25 ppm over the present-day BSE (Kruijjer et al., 2015; Touboul et al., 2015). These two studies exclusively investigated KREEP-rich samples, raising the question of whether other lunar reservoirs—such as the mare basalt sources—exhibit similar or larger ^{182}W excesses. Addressing this question is important not only for precisely determining the ^{182}W composition of the bulk Moon, but also for obtaining insights into the timescale of lunar magma ocean crystallization. This is because magma ocean crystallization is thought to have produced compositionally distinct lunar mantle sources, including the high-Ti mare basalt sources, which are characterized by the highest Hf/W known on the Moon (e.g., Righter and Shearer, 2003). Thus, if magma ocean crystallization occurred during the lifetime of ^{182}Hf , then high-Ti mare basalts should have a radiogenic ^{182}W excess relative to other lunar rocks.

Determining the ^{182}W signatures of mare basalts is challenging because due to their high Ta/W, neutron capture effects may be significant even for weakly irradiated samples. Prior studies analyzed metal samples (Kleine et al., 2005; Touboul et al., 2007), but given the low abundance of metals in mare basalts, large sample masses must be processed to obtain sufficient W for precise isotope analyses. In this study, we therefore use a different approach and combine ^{182}W measurements on lunar whole-rock samples with high-precision Hf isotope and Ta/W ratio measurements to empirically quantify the effects of secondary neutron capture on measured ^{182}W compositions. Using this approach, we derive pre-exposure ^{182}W for different lunar source lithologies, which makes it possible (i) to assess the magnitude of any potential radiogenic ^{182}W variations within the Moon, and (ii) to precisely define the ^{182}W composition of the bulk silicate Moon. We then use these results to assess the timescale of lunar mantle differentiation and use the ^{182}W composition of the Moon to test current giant impact models.

2. Samples and analytical methods

A total of 10 lunar samples were selected for combined W and Hf isotopic, as well as Ta/W analyses, including 2 low-Ti mare basalts (12004, 15495), 6 high-Ti mare basalts (10057, 70017, 70035, 70215, 74255, 75035), Mg-suite norite 77215, and lunar meteorite Kalahari 009. The latter sample was selected because of its very low cosmic ray exposure age of only ~ 230 yr (Nishiizumi et al., 2005), and accordingly, the expectation that neutron capture effects are minimal for this sample. Moreover, to permit a direct comparison to the results previously obtained for KREEP, we also

measured the Ta/W and Hf isotope compositions of the KREEP-rich samples investigated by Kruijjer et al. (2015).

All samples were received as rock fragments and were ultrasonically cleaned and rinsed with ethanol, and then crushed and ground to a fine powder in an agate mortar. The analytical techniques for sample digestion, chemical separation of W, and W isotope ratio measurements by MC-ICPMS are largely based on previously developed procedures (Kruijjer et al., 2015, 2014). In brief, the lunar samples (~ 0.5 – 1 g) were digested in ~ 20 – 40 ml HF–HNO₃ (2:1) at 130 – 150 °C on a hotplate for 2–3 days. Then ~ 2 – 5% aliquots (equivalent to ~ 2 ng W and 10 – 20 ng Ta) were taken for the determination of Ta and W concentrations by isotope dilution. For the isotope composition measurements, W was separated from the sample matrix using a two-stage anion exchange chromatography (Kruijjer et al., 2015, 2014). While the first column separates W from most matrix elements, the second anion exchange chromatography step quantitatively separates W from other high field strength elements (HFSE; Ti, Zr, Hf, Ta). The Hf in these HFSE cuts was subsequently separated using Ln-spec resin as described in Bast et al. (2015). Note that this chromatography step was repeated twice to ensure that the final Hf cuts had Ti/Hf < 0.01 and Zr/Hf < 2 . The total yields of the chemical separation were ~ 80 – 95% for W and 50 – 90% for Hf. Total procedural blanks for the isotope composition analyses were ~ 50 – 100 pg W and ~ 20 – 40 pg Hf, and insignificant given the amounts analyzed (~ 30 ng W, ~ 100 – 1000 ng Hf).

The W and Hf isotope measurements were performed using a ThermoScientific® Neptune Plus MC-ICPMS in the Institut für Planetologie at the University of Münster. The procedures for high-precision W isotope analyses are described in detail elsewhere (Kruijjer et al., 2015, 2014). Samples and standards for W and Hf isotope analyses were introduced using self-aspirating Cetac® C-flow or ESI® PFA nebulizers (50 – 60 $\mu\text{L}/\text{min}$) connected to a Cetac® Aridus II desolvator. The W isotope measurements were performed using Jet sampler and X-skimmer cones which resulted in total ion beams of ~ 1.5 – 2.5×10^{-10} A obtained for a ~ 30 ppb W standard solution at uptake rates of ~ 50 – 60 $\mu\text{L}/\text{min}$. Electronic baselines were obtained prior to each sample measurement by deflecting the beam using the electrostatic analyzer for 60 s and then subtracted from sample signal intensities. A single W isotope measurement comprised 200 cycles of 4.2 s integration time each, and each sample was measured once or twice depending on the amount of W available for analysis. Small isobaric interferences from ^{184}Os and ^{186}Os on W isotope ratios were corrected by monitoring interference-free ^{188}Os , and were smaller than 10 parts-per-million (ppm) on $^{182}\text{W}/^{184}\text{W}$ and hence insignificant. Instrumental mass bias was corrected by internal normalization to $^{186}\text{W}/^{184}\text{W} = 0.92767$ (denoted '6/4') using the exponential law. Note that we only used $^{186}\text{W}/^{184}\text{W}$ -normalized data to avoid normalizations involving ^{183}W ; the latter can be biased by a small analytical artefact on ^{183}W introduced during sample preparation, as observed in this and several earlier studies (e.g., Cook and Schönbachler, 2016; Kruijjer et al., 2012; Willbold et al., 2011). The Hf isotope analyses were performed using standard sampler and 'H' skimmer cones and all major 'non-radiogenic' Hf isotopes (^{177}Hf , ^{178}Hf , ^{179}Hf , ^{180}Hf) were measured simultaneously. Total ion beams of ~ 3.5 – 4.5×10^{-10} were obtained for a ~ 150 ppb Hf standard solution at uptake rates of ~ 50 $\mu\text{L}/\text{min}$. Electronic baselines were obtained prior to each sample measurement by deflecting the beam using the electrostatic analyzer for 60 s and then subtracted from sample signal intensities. A single Hf isotope measurement comprised 200 cycles of 4.2 s integration time each but sample solutions were measured 4–6 times to improve the precision. Potential isobaric interferences from ^{180}W and ^{180}Ta on ^{180}Hf were corrected by monitoring interference-free ^{183}W and ^{181}Ta , and were negligible (< 10 ppm on $^{180}\text{Hf}/^{177}\text{Hf}$). Instrumental mass bias was corrected

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