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## Late delivery of chondritic hydrogen into the lunar mantle: Insights from mare basalts

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### ABSTRACT

Recent analytical advances have enabled first successful in-situ detection of water (measured as OH) in lunar volcanic glasses, and, melt inclusions and minerals from mare basalts. These in-situ measurements in lunar materials, coupled with observations made by orbiting spacecraft missions have challenged the traditional view of the Moon as an anhydrous body. By synthesizing and modeling of previously published data on OH contents and H isotope compositions of apatite from mare basalts, we demonstrate that a model of hydrogen delivery into the lunar interior by late accretion of chondritic materials adequately accounts for the measured “water” content and its hydrogen isotopic composition in mare basalts. In our proposed model, “water” in the lunar interior was mostly constituted by hydrogen, delivered by the late accretion of chondrite-type materials. Our model is also consistent with previously proposed models to account for other geochemical characteristics of the lunar samples.

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### 1. Introduction

For several decades since the successful Apollo and Luna missions brought back more than 380 kg of lunar rock and soil samples, the Moon was considered to be an anhydrous body (Papike et al., 1991). Although, small quantities of water and hydrogen had been reported from bulk lunar samples from measurements made in early 1970s, the preferred interpretation was that hydrogen in lunar soils and regolith breccias was derived from solar wind implantation and that the measured water was not indigenous lunar water but a result of terrestrial contamination (see for example Epstein and Taylor (1973), references therein, and Supplementary Material). Interestingly, three mare basalts were also analyzed in these early studies (Friedman et al., 1971; Merlivat et al., 1974, 1976) and were found to contain few part per million (ppm) H with an elevated hydrogen isotopic composition ( $\delta D \sim 200\text{--}300\text{‰}$ ); an enriched deuterium signature compared to any known terrestrial reservoir. The favored interpretation of this dataset was in terms of cosmic-ray spallation reactions (Epstein and Taylor, 1973) (see Supplementary Material for further details). Since these early measurements, a few decades passed until recent analytical advances allowed first successful in-situ detection of water (measured as OH) in lunar

volcanic glasses formed during fire-fountain eruptions (Saal et al., 2008). Subsequently, water has been measured in melt inclusions trapped in olivine in these volcanic glasses (Hauri et al., 2011) and in mineral apatite (Boyce et al., 2010; Greenwood et al., 2011a, 2012; McCubbin et al., 2010a, 2010b, 2011; Robinson et al., 2012), which is an accessory phase found ubiquitously in basaltic lunar rocks. In apatite, water is structurally bound in its hydroxyl form (OH<sup>-</sup>). These in-situ measurements in lunar samples and minerals, coupled with observations made by orbiting spacecraft missions have challenged the traditional view of the Moon as an anhydrous body (see review by Anand (2010)). However, the presence of hydroxyl in apatite does not necessarily imply a wet lunar interior. Indeed, hydrogen is likely to be an important H species at the very low oxygen fugacity prevailing in the lunar mantle (e.g., Elkins-Tanton and Grove, 2011; Hirschmann et al., 2012).

Mare basalts contain appreciable amounts of hydroxyl located into the structure of mineral apatite (Boyce et al., 2010; Greenwood et al., 2011a, 2012; McCubbin et al., 2010a, 2010b, 2011). In contrast, apatites in lunar crustal rocks and in KREEPy rocks are almost devoid of OH (Greenwood et al., 2011a, 2012; McCubbin et al., 2010b, 2011; Robinson et al., 2012). Based on the OH content in mare-basalt magmas from which apatites crystallized, estimated through the partitioning of OH between apatite and basaltic melts, and applying appropriate fractional crystallization and partial melting corrections, it has been suggested that the mantle source rocks of these basalts may have contained

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between a few ppm to hundreds of ppm water (Boyce et al., 2010; Hauri et al., 2011; McCubbin et al., 2010b). However, exploiting similar latest advances in analytical instrumentation, analysis of lunar samples for their chlorine isotopic composition seems to be more compatible with an anhydrous lunar interior (Sharp et al., 2010). In lunar samples, the measured  $\delta^{37}\text{Cl}$  values range from  $-1$  to  $+24\%$ ,  $\sim 25$  times the range observed in terrestrial samples, pointing towards volatilization of metal halides instead of HCl during the eruption of mare magmas, and therefore, implying very low H concentration during Cl degassing (Sharp et al., 2010). Modeling of the lunar mantle petrogenesis through the crystallization of a lunar magma ocean (LMO) also requires a relatively dry lunar interior (Elkins-Tanton and Grove, 2011). Indeed, Elkins-Tanton and Grove (2011) have shown that a lunar mantle with 10–200 ppm water requires a bulk LMO with 100–1000 ppm water. The late-stage crystallization products of this LMO, known as urKREEP (K, REE and P standing for Potassium, Rare Earth Elements and Phosphorus, respectively), should thus be even more enriched in water considering the incompatible behavior of water during the LMO crystallization. However, this is contrary to the measurements made so far in KREEPy rocks (McCubbin et al., 2011; Robinson et al., 2012). Elkins-Tanton and Grove (2011), therefore, concluded that lunar interior must have contained  $< 10$  ppm water although their model cannot rule out addition of hydrogen and/or water in the lunar mantle by impacts after the main phase of crystallization of the LMO. Finally, retaining large amounts of primordial hydrogen, if it was ever accreted (Albarède, 2009), on a low-gravity body such as the Moon seems unlikely. It would have required a shielding atmosphere on the early Moon, for which currently there is no evidence (Pahlevan and Stevenson, 2007).

By synthesizing and modeling of previously published data on OH contents and H isotope compositions of apatite from mare basalts, we demonstrate that a model of hydrogen delivery into the lunar interior by late accretion of chondritic materials

adequately accounts for the “water” content and its hydrogen isotopic composition in mare basalts. In our proposed model, “water” in the lunar interior was thus mostly constituted by hydrogen delivered by late accretion of chondrite-type materials. It is possible that the Moon may have also retained some primordial hydrogen. However, on the basis of the existing geochemical dataset on lunar samples, we argue that the late-accretion of chondritic materials to the Moon dominates the inventory of hydrogen in the bulk-silicate-Moon. Our model is also consistent with previously proposed models to account for other geochemical characteristics of the lunar samples (Day et al., 2007; Taylor and Robinson, 2012).

## 2. Hydrogen content of differentiated lunar basaltic melts

In order to fully understand the evolution of the lunar interior, mare basalts provide ideal samples as they are directly derived from the lunar mantle through partial melting. Availability of additional non-mare samples such as Mg-suite and KREEP basalts provide further constraints on lunar magmatic processes. For the present purposes, we have focused on mare basalts in which the mineral apatite occurs ubiquitously. At the very low oxygen fugacity that prevails in the lunar interior,  $\text{H}_2$  is likely to be an important H-bearing species in lunar magmas (Elkins-Tanton and Grove, 2011; Hirschmann et al., 2012). Given the complexity involved in the petrogenesis of mare basalts, it is very difficult to obtain any reliable estimates of the water content of primitive lunar magmas before they underwent secondary processes such as fractional crystallization or degassing, for example. The best that can be done is to estimate the OH content of the differentiated magma from which apatites have crystallized. Hydroxyl groups could indeed form very late during the magma evolution through coupled Fe reduction and H oxidation (Hirschmann et al., 2012; Wetzel et al., 2012). Overall, the average OH content of

**Table 1**

Measured OH contents in apatites from mare basalts and estimated OH and equivalent H contents of evolved magmas from which apatites have crystallized.

Ref. <sup>a</sup>	Sample #	Type	Measured OH content (ppm) <sup>b</sup>				OH in evolved melt (ppm) <sup>c</sup>				Eq. H in evolved melt (ppm) <sup>d</sup>				Age (Gy) <sup>e</sup>	TiO <sub>2</sub> (wt%) <sup>f</sup>
			Min	Max	Avg	SD	Min	Max	Avg	SD	Min	Max	Avg	SD		
a	14053	High-Al mare basalt	2935	4543	3873	602	11741	18171	15491	2407	691	1069	911	142	3.87 ± 0.03	2.6
b	10044	High-Ti mare basalt	1606	2361	1931	319	6422	9444	7726	1275	378	556	454	75	3.63 ± 0.07	9.0
b	12039	Low-Ti mare basalt	1813	11428	7288	3067	7253	45711	29153	12269	427	2689	1715	722	3.10 ± 0.07	3.5
b	75055	High-Ti mare basalt	–	–	2021	–	–	–	8084	–	–	–	476	–	3.76 ± 0.05	10.8
b	14053	High-Al mare basalt	1077	1757	1429	280	4307	7027	5717	1118	253	413	336	66	3.87 ± 0.03	2.6
c	10047	High-Ti mare basalt	1094	1538	1316	314	4376	6152	5264	1256	257	362	310	74	3.71 ± 0.004	9.4
c	12064	Low-Ti mare basalt	–	–	330	–	–	–	1320	–	–	–	78	–	3.17 ± 0.07	4.0
d	15058	Low-Ti mare basalt	0	4420	1797	1639	0	17680	7189	6556	0	1040	423	386	3.37 ± 0.02	1.7
e	14053	High-Al mare basalt	1300	2700	1775	640	5200	10800	7100	2559	306	635	418	151	3.87 ± 0.03	2.6
e	NWA2977	VLT basalt	4000	7000	5500	2121	16000	28000	22000	8485	941	1647	1294	499	3.12 ± 0.01	0.2
f	12021	Low-Ti mare basalt	0	4837	1430	1752	0	19349	5722	7009	0	1138	337	412	3.25 ± 0.03	3.5
f	LAP02205	Low-Ti basalt	766	13834	7374	5358	3066	55337	29498	21431	180	3255	1735	1261	2.99 ± 0.01	3.2
f	NWA2977	VLT basalt	0	2260	565	1130	0	9042	2260	4521	0	532	133	266	3.12 ± 0.01	0.2
f	12023	Mare basalt <sup>g</sup>	0	8447	3405	4455	0	33788	13618	17821	0	1988	801	1048	3.30 ± 0.10	2.5
f	12037	Mare basalt <sup>g</sup>	0	15843	5321	4635	0	63371	21285	18540	0	3728	1252	1091	$\sim 3.20^h$	3.0
f	79195	Mare basalt <sup>g</sup>	0	3882	857	1275	0	15530	3428	5102	0	914	202	300	$\sim 3.75^h$	12.0

<sup>a</sup> References: (a) Boyce et al. (2010); (b) Greenwood et al. (2011a); (c) Greenwood et al. (2012); (d) McCubbin et al. (2010a); (e) McCubbin et al. (2010b); (f) McCubbin et al. (2011).

<sup>b</sup> Equivalent OH when reported as H<sub>2</sub>O (see Supplementary Material for details).

<sup>c</sup> Calculated using a partition coefficient  $D_{\text{OH}}^{\text{Ap/melt}}$  of 0.25 (see McCubbin et al. (2011)).

<sup>d</sup> Equivalent mass of H (see Supplementary Material for details).

<sup>e</sup> Best age estimate from published data (see Supplementary Material for details).

<sup>f</sup> TiO<sub>2</sub> contents are from the online supporting Table A3.11. Mare basalt group compositions of Wieczorek et al. (2006) and from the lunar sample compendium (<http://curator.jsc.nasa.gov/lunar/compendium.cfm>).

<sup>g</sup> Mare basalt fragments recovered from soil samples.

<sup>h</sup> Approximate ages corresponding to the typical age range reported for Apollo 12 and 17 samples (see Supplementary Material for details).

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