



Speciation and dissolution of hydrogen in the proto-lunar disk



Kaveh Pahlevan^{a,b,*}, Shun-ichiro Karato^b, Bruce Fegley^c

^a Laboratoire Lagrange, Université Côte d'Azur, Observatoire de la Côte d'Azur, CNRS, Nice, France

^b Department of Geology and Geophysics, Yale University, New Haven, CT 06520, USA

^c Planetary Chemistry Laboratory, Department of Earth and Planetary Sciences and The McDonnell Center for the Space Sciences, Washington University, St. Louis, MO 63130, USA

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ABSTRACT

Despite very high temperatures accompanying lunar origin, indigenous water in the form of OH has been unambiguously observed in Apollo samples in recent years. Such observations have prompted questions about the abundance and distribution of lunar hydrogen. Here, we investigate the related question of the origin of lunar H: is the hydrogen observed a remnant of a much larger initial inventory that was inherited from a “wet” Earth but partly depleted during the process of origin, or was primordial hydrogen quantitatively lost from the lunar material, with water being delivered to lunar reservoirs via subsequent impacts after the origins sequence? Motivated by recent results pointing to a limited extent of hydrogen escape from the gravity field of the Earth during lunar origin, we apply a newly developed thermodynamic model of liquid–vapor silicates to the proto-lunar disk to interrogate the behavior of H as a trace element in the energetic aftermath of the giant impact. We find that: (1) pre-existing H-bearing molecules are rapidly dissociated at the temperatures considered (3100–4200 K) and vaporized hydrogen predominantly exists as OH(v), H(v) and MgOH(v) for nearly the full range of thermal states encountered in the proto-lunar disk, (2) despite such a diversity in the vapor speciation – which reduces the water fugacity and favors hydrogen exsolution from co-existing liquids – the equilibration of the vapor atmosphere with the disk liquid results in significant dissolution of H into proto-lunar magmas, and (3) equilibrium H isotopic fractionation in this setting is limited to <10 per mil and the “terrestrial” character of lunar D/H recently inferred should extend to such a precision if liquid–vapor equilibration in the proto-lunar disk is the process that gave rise to lunar hydrogen. Taken together, these results implicate dissolution as the process responsible for establishing lunar H abundances.

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1. Introduction

After decades of scientific thought maintaining that the Moon is essentially dry (<1 ppb H₂O), lunar samples have, in recent years, become recognized as carriers of measurable indigenous water (Hauri et al., 2015). In this work, we use “indigenous” to refer to non-solar-wind, non-cosmogenic sources such as inheritance from birth or implantation via impactors (Bottke et al., 2010) and “water” to refer to various forms of hydrogen in silicate glasses, minerals and melts (mostly as OH) with concentrations representing the weight percent of H₂O that would be released upon degassing. In a pioneering study, Saal et al. (2008) measured tens of ppm wt H₂O in lunar volcanic glasses and inferred much higher H₂O contents in the pre-degassed magma. In a follow-up study,

Hauri et al. (2011) analyzed olivine-hosted melt inclusions from lunar pyroclastic glass deposit 74220 (“orange glass”) sampling a pre-eruptive magma and found upwards of hundreds of ppm wt H₂O, confirming the earlier inference of high water contents for the source magma. These discoveries have motivated a number of subsequent investigations intended to reevaluate the abundance and distribution of lunar water.

Karato (2013) reanalyzed the lunar geophysical observables, exploiting the high sensitivity of electrical conductivity to the presence of protons to infer bulk Moon H₂O abundances of 10–100 ppm wt. Sharp et al. (2010) observed a wide variation in the chlorine isotopic composition of lunar samples and argued that such variation was due to a dearth of H₂O in the host lava and that the water content of the lunar mantle was constrained to <0.1 ppm wt. In a subsequent publication, Sharp et al. (2013) showed that rapid degassing of H₂ could dehydrate lunar magmas and reconcile much more hydrous magma compositions with the anhydrous conditions inferred by the chlorine isotopic data. Hui et al. (2013) measured H₂O contents in plagioclase grains in

* Corresponding author at: Laboratoire Lagrange, Observatoire de la Côte d'Azur, Boulevard de l'Observatoire, C.S. 34229, 06304, Nice, Cedex 4, France.

E-mail address: pahlevan@oca.eu (K. Pahlevan).

anorthosite and troctolite from the lunar crust, inferring an initial water content of ~ 320 ppm wt for the lunar magma ocean from which these grains presumably crystallized. Albarède et al. (2015) measured Zn, Rb and K abundances in a range of lunar samples, constructed a volatility scale, extrapolated the behavior of moderately volatile elements to H and concluded that the water content of the bulk Moon is ≤ 1 ppm wt. Finally, Chen et al. (2015) analyzed melt inclusions from a range of mare basalts and found H contents spanning 5–100 times less than that measured by Hauri et al. (2011), interpreted this range as a signature of degassing and inferred minimum H₂O concentrations for the primitive lunar mantle of 110 ppm wt.

The range in the inferred lunar water abundances attests to the uncertainties in interpreting the lunar volatile record. For sample studies, this uncertainty can arise from: (i) the question of whether the samples in question are characteristic or anomalous in the lunar interior, (ii) an incomplete knowledge of mineral–melt partition coefficients necessary to infer magma compositions from crystalline samples, and (iii) the process of degassing, which, for extrusive volcanism, can eliminate the vast bulk of the inventory of volatiles in magmas, but leave a record from which the history is difficult to reconstruct. Geophysical studies have their own uncertainties; for example, interpretation of the observed electrical conductivity and tidal dissipation in the lunar interior is subject to uncertainty partly due to an incomplete understanding of the sensitivity of the observables to water contents and partly due to an incomplete knowledge of the values of the geophysical observables themselves (Karato, 2013). Such uncertainty allows a variety of inferences to be made from the observable lunar volatile record, with recent inferences ranging from 1 ppm wt H₂O (Albarède et al., 2015) to 110 ppm wt (Chen et al., 2015). For the purposes of this study, we assume that the bulk lunar H abundances are within this range.

Here, we investigate the question of the origin of lunar water: is the measured H a remnant of primordial hydrogen inherited at birth or is it implanted via late-accretionary impacts by, perhaps, water-rich impactors? The discovery of indigenous lunar water has come amidst modern isotopic observations that provide new evidence for a “terrestrial” origin of the lunar material (Kruijer et al., 2015; Pahlevan, 2014; Touboul et al., 2015; Young et al., 2016) motivating new developments to the giant impact hypothesis (Canup, 2012; Cuk and Stewart, 2012; Pahlevan and Stevenson, 2007; Pahlevan et al., 2011) and rendering possible the inheritance of lunar water from the proto-Earth. Motivated by the nearly closed-system behavior of hydrogen in the post-impact Earth-disk system (Nakajima and Stevenson, 2014), we investigate whether the processes accompanying lunar origin can reproduce the bulk elemental abundances recently inferred for the lunar interior. We address this question by constructing a model capable of calculating snapshots of the evolution of the proto-lunar disk and the equilibrium partitioning of hydrogen and its isotopes between the proto-lunar liquid and co-existing vapor to gain insight into the behavior of H in the aftermath of the Moon-forming impact. If the “water” being measured is indeed primordial (i.e. inherited at birth), then its isotopic composition can be used to constrain the process of volatile depletion with the advantage that hydrogen isotopes may exhibit significant equilibrium fractionation even at the high temperatures encountered after the giant impact (see Section 4.2). While a detailed understanding of the processes that established the lunar volatile abundances is – at present – non-existent, the process of liquid–vapor partitioning illustrated here must play a role in any satisfactory scenario of volatile depletion. In Section 2, we describe the adopted disk model. Section 3 covers the thermodynamic model we use to characterize high-T major element silicates – and the chemical conditions they establish – in the proto-lunar disk, and the modeled behavior of hydrogen as a

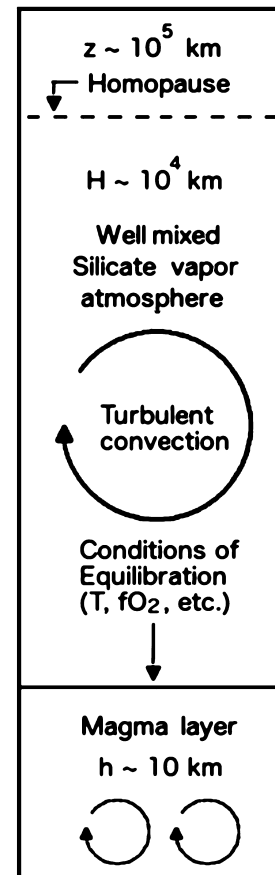


Fig. 1. A schematic diagram of a proto-lunar disk column, with a midplane liquid layer, a liquid–vapor interface, and an overlying atmosphere. The physical and chemical conditions at the interface (e.g. T , fO_2) determine the partitioning and therefore the composition of the liquid layer and complementary atmosphere. The proto-lunar liquid is assumed to become the lunar magma ocean liquid without further fractionation. The parameter that characterizes the thermal state is the mass fraction of the atmosphere (f_v) and can vary between 0–1. Vigorous (convective and other) fluid motions ensure that the vapor atmosphere is largely well-mixed and the homopause – above which vapor species can gravitationally separate according to distinct scale heights – appears only at very high altitudes.

passive tracer. In Section 4, results of elemental and isotopic partitioning between the silicate vapor atmosphere and an underlying disk liquid layer are presented. In Sections 5 and 6, the implications of these results for the origin of lunar water are discussed and the conclusions of the study are summarized.

2. Disk model

At present, only one evolutionary model of the proto-lunar disk has been developed that attempts to make a connection between a disk process and a chemical or isotopic signature in the lunar material (Canup et al., 2015). Such an evolutionary model is beyond the scope of the present work. Instead, here we describe a physical-chemical model that permits calculation of snapshots of the disk evolution, including the physical and chemical conditions (e.g. T , fO_2) that determine trace element and isotopic partitioning between the liquid layer and the atmosphere as a function of the thermal state (Fig. 1). With this model, we can explore the behavior of tracers for the full range of conditions encountered in the proto-lunar disk during its lifetime.

The physical model is of a 1-dimensional stratified column of disk material with a convective liquid layer near the mid-plane, a liquid–vapor interface, and an overlying convective atmosphere. The partitioning calculations here pertain to the conditions at the

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