



Water in evolved lunar rocks: Evidence for multiple reservoirs

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Abstract

We have measured the abundance and isotopic composition of water in apatites from several lunar rocks representing Potassium (K), Rare Earth Elements (REE), and Phosphorus (P) – KREEP – rich lithologies, including felsites, quartz monzodiorites (QMDs), a troctolite, and an alkali anorthosite. The H-isotope data from apatite provide evidence for multiple reservoirs in the lunar interior. Apatite measurements from some KREEP-rich intrusive rocks display moderately elevated δD signatures, while other samples show δD signatures similar to the range known for the terrestrial upper mantle. Apatite grains in Apollo 15 quartz monzodiorites have the lowest δD values measured from the Moon so far (as low as -749‰), and could potentially represent a D-depleted reservoir in the lunar interior that had not been identified until now. Apatite in all of these intrusive rocks contains <267 ppm H_2O , which is relatively low compared to apatites from the majority of studied mare basalts (200 to >6500 ppm H_2O). Complexities in partitioning of volatiles into apatite make this comparison uncertain, but measurements of residual glass in KREEP basalt fragments in breccia 15358 independently show that the KREEP basaltic magmas were low in water. The source of 15358 contained ~ 10 ppm H_2O , about an order of magnitude lower than the source of the Apollo 17 pyroclastic glass beads, suggesting potential variations in the distribution of water in the lunar interior.

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1. INTRODUCTION

The detection of water in lunar volcanic glasses, apatites, and melt inclusions has implications for planetary accretion, the source(s) of water in the Earth–Moon system, and the role of water in lunar evolution (Saal et al., 2008; McCubbin et al., 2010; Boyce et al., 2010; Greenwood

et al., 2011; Hauri et al., 2011; Tartèse et al., 2013, 2014; Barnes et al., 2013, 2014a). (In this paper we use “water” as a shorthand way of referring to all hydrogen species, H_2O , OH, and H_2 . More than one may be present, depending on oxygen fugacity and pressure (e.g., Hirschmann et al., 2012). Recent work has shown that apatites in mare basalts contain appreciable amounts of water, and are generally enriched in deuterium (^2H , or D) with respect to Earth, possibly due to the addition of D-rich material early in the Moon’s history (Greenwood et al., 2011) or to the loss of H preferentially over D during magma degassing

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(e.g. Saal et al., 2008, 2013; Tartèse and Anand, 2013; Tartèse et al., 2013). The final fraction of the global lunar magma ocean (LMO) model is considered to be urKREEP, so named for its enrichment in incompatible elements compared to other lunar materials (e.g., Warren and Wasson, 1979). Because water behaves as an incompatible element in major silicate phases that formed during the LMO crystallization (Koga et al., 2003; Aubaud et al., 2004; Grant et al., 2007), KREEP-rich rocks are expected to be enriched in water relative to other lunar rocks. Many KREEP-rich lithologies consist of evolved rocks that formed as intrusions, which means that they would have avoided or experienced minimal water loss (unlike the mare basalts), and potential hydrogen isotope fractionation due to magmatic degassing.

The mineral apatite [$\text{Ca}_5(\text{PO}_4)_3(\text{F}, \text{Cl}, \text{OH})$] incorporates OH into its crystal structure, making it a potential recorder of the concentration of OH in magma at the time of apatite crystallization. The OH in apatite is resistant to exchanging O or H with adsorbed terrestrial water on thin section surfaces (Greenwood et al., 2011) hence it is useful for H isotopic measurements by secondary ion mass spectrometry (SIMS). The D/H ratio, expressed as $\delta\text{D} (\text{‰}) = ([\text{D}/\text{H}]_{\text{sample}}/[\text{D}/\text{H}]_{\text{standard}} - 1) \times 1000$, relative to Vienna Standard Mean Ocean Water (V-SMOW), is important for identifying the source of the Moon's water and the extent of water loss during magmatic processing (e.g., Greenwood et al., 2011; Elkins-Tanton and Grove, 2011; Tartèse et al., 2013). Previous studies have demonstrated that the water content of lunar apatite varies among different rock types. Apatites in Apollo mare basalts record the highest H_2O contents (up to ~ 7500 ppm) and δD (+390 to +1100‰) (McCubbin et al., 2010; Greenwood et al., 2011; Barnes et al., 2013; Tartèse et al., 2013), while apatites in more evolved, KREEP-related rocks generally have lower H_2O contents (<3000 ppm) and δD values (−384 to +791‰, Greenwood et al., 2011; Barnes et al., 2014a; Robinson et al., 2013; Tartèse et al., 2014; Robinson and Taylor, 2014).

Though it was initially thought that apatite water contents could be used to infer initial magmatic water abundances (McCubbin et al., 2010; Boyce et al., 2010; Tartèse et al., 2013, 2014; Barnes et al., 2014a), Boyce et al. (2014) demonstrated that the F–Cl–OH partitioning into apatite is not well-described by a simple apatite–melt partition coefficient. Apatite water content thus cannot be used to quantitatively determine the amount of water in the co-existing melt, unless the F or Cl content are also known for both melt and apatite, and that it can be confirmed that apatite crystallized in equilibrium with the melt. However, the measured D/H ratios are still useful for gaining insights into processes that might have affected the parental magmas.

Rocks formed in an intrusive environment could have experienced minimal D–H fractionation and water loss prior to apatite crystallization because they formed at pressures where water is far more soluble in silicate melt than at low pressures near or on the surface (e.g., Dixon et al., 1995). Water solubility in magmas decreases with decreasing pressure, so when a hydrous magma approaches a planetary surface, the melt degasses and water is lost assuming water is present as OH and H_2O . Diffusion modeling of

water loss from volcanic lunar pyroclastic glasses indicates that they lost up to 98% of their initial water content upon eruption (Saal et al., 2008). Mare basaltic magmas would also have degassed during eruption onto the lunar surface, but probably less than the 98% loss experienced by pyroclastic glasses. Based on water contents of melt inclusions in olivine from subaerial and submarine Hawaiian lavas (which have little water loss due to the pressure at which they erupted; Hauri, 2002), we estimate that lava flows lose up to 90% of their pre-eruptive H_2O . Low-Ti mare magmas could have lost 85 to 99% of their pre-eruptive water contents (Tartèse et al., 2013), but such estimates assume an initial D/H ratio. The degassing of hydrogen from a magma or lava also fractionates lighter H from heavier D, especially if H_2 is lost rather than H_2O (e.g. Richet et al., 1977; Tartèse and Anand, 2013; Tartèse et al., 2013). Degassing experiments with apatite suggest that volatile loss occurs rapidly in extrusive magmas, and that apatite, which forms late, will not preserve pre-eruption volatile contents and will instead reflect the post-degassing volatile composition of the magma (Ustunisik et al., 2015). The erupted mare basalts have the highest δD values (up to $\sim +1100$ ‰) found in lunar rocks so far (Greenwood et al., 2011; Barnes et al., 2013; Tartèse et al., 2013).

Apatite in intrusive rocks may represent a more pristine sampling of the lunar interior water than the water bound in apatites in the mare basalts, due to their formation at depth at higher pressures (Robinson et al., 2013; Robinson and Taylor, 2014; Barnes et al., 2014a). Reaction relations in symplectitic intergrowths in troctolite 76535 indicate a depth of origin of ~ 40 km (Gooley et al., 1974; McCallum et al., 2006), corresponding to a pressure of over 1.5 kb. On the other hand, gabbonorite sample 76255 may have formed as shallow as a few kilometers (McCallum et al., 2006), and quartz-monzodiorite 14161,7373 as shallow as 1 km (Jolliff et al., 1999), at a pressure of 50 b. The liquidus temperatures and presence of quartz in highly fractionated lunar felsites (Hess et al., 1975, 1978, 1989; Robinson and Taylor, 2011; Robinson et al., 2015) indicate crystallization at a pressure of at least ~ 1 kb (Tuttle and Bowen, 1958), corresponding to a depth of over 20 km, assuming a crustal density of 2550 kg/m^3 (Wieczorek et al., 2013). Up to 3 wt.% H_2O is soluble in rhyolitic melts at ~ 1 kb, and even 0.5 wt.% would have been soluble at the 50 b pressure experienced by the parental melt for QMD 14161, 7373 (VolatileCalc, Newman and Lowenstern, 2002; Jolliff et al., 1999). Common mineral phases associated with hydrous melts such as amphibole and micas have never been reported in lunar rocks, and no lunar magma has been shown to have contained weight percent levels of water (e.g. Robinson and Taylor, 2014 and references therein). While the concentration of water in the melt would have increased during crystallization, it would have had to reach weight percent levels to degas *before* apatite crystallization began and removed F, Cl, and OH from the melt (Boyce et al., 2014). Assuming that water is completely incompatible in the crystallizing silicates and that a magma had an initial H_2O content of 0.1 wt% (similar to A17 melt inclusions, Saal et al., 2013), the H_2O concentration would have only increased to 1 wt.% after 90% crystallization.

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