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Experimental constraints on the solidification of a nominally dry lunar magma ocean

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ABSTRACT

The lunar magma ocean (LMO) concept has been used extensively for lunar evolution models for decades, but to date the full cooling and crystallization path of the LMO has not been studied experimentally. Here we present results of a high-pressure, high-temperature experimental study of the mineralogical and geochemical evolution accompanying the full solidification of a nominally dry LMO. Experiments used a bulk composition based on geophysical data, and assumed an initial LMO depth of 700 km. The effect of pressure within a deep magma ocean on solidification at different levels in the ocean was explicitly taken into account, by performing experiments at multiple pressures and constant temperature during each solidification step. Results show formation of a deep harzburgite (olivine + low-Ca pyroxene) layer in the first \sim 50% of equilibrium crystallization. The crystallising mineral assemblage does not change until plagioclase and clinopyroxene appear at 68 PCS (per cent solid by volume), while low-Ca pyroxene stops forming. Olivine disappears at 83 PCS, and ilmenite and β -quartz start crystallizing at 91 and 96 PCS, respectively. At 99 PCS, we observe an extremely iron-rich (26.5 wt.% FeO) residual LMO liquid. Our results differ substantially from the oft-cited LMO solidification study of Snyder et al. (1992), which was based on a limited number of experiments at a single pressure. Differences include the mineralogy of the deepest sections of the solidified LMO (harzburgitic instead of dunitic), the formation of SiO₂ in the lunar interior, and the development of extreme iron enrichment in the last remaining dregs of the LMO. Our findings shed new light on several aspects of lunar petrology, including the formation of felsic and iron-rich magmas in the Moon. Finally, based on our experiments the lunar crust, consisting of the light minerals plagioclase and quartz, would reach a thickness of \sim 67.5 km. This is far greater than crustal thickness estimates from recent GRAIL mission gravitational data (34-43 km, Wieczorek et al., 2013). Although the initial depth of the LMO has an effect on the thickness of crust produced, this effect is not large enough to explain this discrepancy. Inefficient plagioclase segregation, trapping of magma in cumulate reservoirs, and Al sequestration in spinel cannot explain the discrepancy either. As plagioclase crystallization can be suppressed by the presence of H₂O, this implies that the lunar magma ocean was water-bearing.

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1. Introduction

The Moon is thought to have formed by accretion of materials ejected during a giant impact of a large impactor with the Earth (e.g., Canup and Asphaug, 2001; Canup, 2012; Ćuk and Stewart, 2012). This collision would have resulted in the generation of significant heat, leading to hot lunar formation conditions. As a result, the young Moon is thought to have been covered by a global magma ocean, referred to as the lunar magma ocean (e.g.,

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http://dx.doi.org/10.1016/j.epsl.2017.04.045 0012-821X/© 2017 Elsevier B.V. All rights reserved. Smith et al., 1970; Wood et al., 1970; Warren, 1985; Rai and van Westrenen, 2014; Steenstra et al., 2016). Crystallization of the LMO is thought to have created a series of concentric cumulate layers with different chemical compositions and mineralogical assemblages, and a ferroan anorthosite primary crust via flotation of less-dense plagioclase-rich cumulates (e.g., Warren, 1985; Snyder et al., 1992; Shearer et al., 2006; Elkins-Tanton et al., 2011).

Because the crystallization sequence and composition of these cumulate layers are of primary importance for understanding subsequent key events in lunar evolution, including the formation of a plagioclase-rich crust and an overturn in the mantle leading to mare basalt volcanism, numerous authors have attempted to model the crystallization of the lunar magma ocean (e.g., Longhi, 1980, 2003, 2006; Taylor and Jakeš, 1974; Tonks and Melosh, 1990; 2

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Snyder et al., 1992; Longhi et al., 2010; Elardo et al., 2011; Elkins-Tanton et al., 2011). Most previous models produce broadly similar cumulate assemblages despite differences in assumed bulk lunar composition and model parameters, with Mg-rich olivine crystallizing first, followed by orthopyroxene, combining to form a harzburgite layer in the lower lunar mantle (Snyder et al., 1992; Elardo et al., 2011: Elkins-Tanton et al., 2011). Anorthitic plagioclase begins to crystallize late (after >70 per cent solid by volume, PCS) floating to the surface to form the primary lunar highland crust. The corresponding mafic cumulates become more Fe-rich as crystallization proceeds and eventually (after >95 PCS) a very dense ilmenite-rich layer is thought to form (Snyder et al., 1992). The final residual LMO dregs become enriched in potassium (K), rare earth elements (REE), and phosphorus (P), forming the source for the later-formed KREEP-rich magmatic rocks (Snyder et al., 1992; Warren and Wasson, 1979; Shearer et al., 2006).

Surprisingly, these models were based on a limited set of high-pressure, high-temperature experiments, requiring theoretical models of phase stability to extrapolate to full magma ocean solidification conditions. Most models ignored the possible effect of variable pressures within the molten ocean on crystallization.

To better quantify the evolutionary history of the LMO, including its crystallization sequence, the chemical compositions of cumulates and corresponding residual LMO during progressive solidification, we performed a fully experimental model of lunar magma ocean solidification, based on anhydrous experiments at pressuretemperature (P-T) conditions that are directly relevant for the evolution of the lunar interior. Parts of our results were previously reported in Lin et al. (2017).

During each step of LMO crystallization, the effects of pressure in the molten ocean on crystallization of the magma were taken into account explicitly. Our results differ significantly from previous studies, in terms of the primary mineralogical composition of the Moon after LMO solidification, and in terms of the compositional evolution of the magma as solidification proceeds. We find that primary SiO₂ formed in the Moon towards the end of LMO crystallization, predict the presence of extreme Fe-rich melts accompanying the formation of the KREEP reservoir. Finally, we show that a nominally dry Moon should have produced an anorthositic crust of \sim 68 km, far thicker than observed, and discuss possible reasons for this discrepancy.

2. Methods

2.1. Experimental

2.1.1. Starting material composition

High-pressure, high-temperature experiments were performed in the chemical system Ca-Fe-Mg-Al-Ti-Si-O (CFMATS), under nominally anhydrous conditions. The starting composition, based on geophysical data for the Moon (Khan et al., 2007), is slightly lower in aluminium abundance than the Lunar Primitive Upper Mantle (LPUM) composition used for the oft-cited LMO numerical model (Snyder et al., 1992), and has a composition that is similar to that of the Bulk Silicate Earth (BSE, McDonough and Sun, 1995). Choosing a bulk composition that is close to the composition of the BSE is consistent with recent high-precision measurements of the chemical and isotopic composition of the Moon that show a very high degree of similarity between BSE and the Moon (e.g., Savage et al., 2010; Armytage et al., 2011; Zhang et al., 2012; Kruijer et al., 2015; Touboul et al., 2015). Table 1 shows a comparison between the bulk composition of the LMO chosen for this study, other LMO compositional models, and the Bulk Silicate Earth (Lin et al., 2017).



Fig. 1. Cartoons (after Tonks and Melosh, 1990) illustrating the experimentally derived solidification evolution of a nominally dry lunar magma ocean. (a) Initial LMO. (b) Turbulently convecting, isothermal LMO at <50 PCS (Per Cent Solidification by volume). Crystals nucleate at all depths and do not dissolve when carried to shallower/deeper depths in the LMO. (c) Compositions of the residual LMO and cumulate pile at 50 PCS, after suspended crystals have settled. (d) During fractional crystals (Pl and Qz) float. (e) The location and composition of the residual 1% LMO and cumulates at 99 PCS. OI = olivine; Opx = orthopyroxene; Cpx = clinopyroxene; Pl = plagioclase; Ilm = ilmenite; $Qz = \beta$ -quartz.

Starting materials were prepared by mixing appropriate amounts of high purity (99.5-99.99%, Alfa Aesar) powdered oxides (MgO, Fe₂O₃, Al₂O₃, TiO₂, SiO₂) and CaCO₃ (99.95-100.05%, Alfa Aesar). The oxides MgO, Al₂O₃, TiO₂ and SiO₂ were fired overnight at 1000 °C and then stored at 110 °C. The other oxides and calcium carbonate were dried at 110 °C overnight prior to use. After mixing the starting materials under ethanol in an agate mortar for 1 h, they were dried in air and decarbonated in a Pt crucible in a box furnace by gradually raising the temperature from 650 to 1000 °C in about 7 h. The Pt crucible had previously been iron-saturated to minimise iron loss. The resulting mixtures were melted in air for 20 min at 1550 °C to promote homogeneity, and to reduce most of the iron in the starting material to 2^+ . They were quenched by immersing the bottom of the Pt crucible in water. Small fragments of resulting glasses were embedded in epoxy, polished, carbon-coated and analyzed for homogeneity by electron microprobe to precisely constrain starting material compositions. The glasses were subsequently crushed, dried, and reground under ethanol in an agate mortar for 1 h and then kept at 110°C until use.

2.1.2. Experimental approach

Our experiments assume an initial LMO depth of 700 km (Fig. 1a), in the range of current estimates between 400 and 1400 km (Elkins-Tanton et al., 2011; Elardo et al., 2011; Rai and van Westrenen, 2014; Steenstra et al., 2016). Similar to Snyder et al. (1992) and Elardo et al. (2011), the process of LMO crystallization was investigated by adopting a 'two-stage' model, with equilibrium crystallization persisting until 50 PCS based on calculations

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