



Evidence for a sulfur-undersaturated lunar interior from the solubility of sulfur in lunar melts and sulfide-silicate partitioning of siderophile elements

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Abstract

Sulfur concentrations at sulfide saturation (SCSS) were determined for a range of low- to high-Ti lunar melt compositions (synthetic equivalents of Apollo 14 black and yellow glass, Apollo 15 green glass, Apollo 17 orange glass and a late-stage lunar magma ocean melt, containing between 0.2 and 25 wt.% TiO₂) as a function of pressure (1–2.5 GPa) and temperature (1683–1883 K). For the same experiments, sulfide-silicate partition coefficients were derived for elements V, Cr, Mn, Co, Cu, Zn, Ga, Ge, As, Se, Mo, Sn, Sb, Te, W and Pb. The SCSS is a strong function of silicate melt composition, most notably FeO content. An increase in temperature increases the SCSS and an increase in pressure decreases the SCSS, both in agreement with previous work on terrestrial, lunar and martian compositions. Previously reported SCSS values for high-FeO melts were combined with the experimental data reported here to obtain a new predictive equation to calculate the SCSS for high-FeO lunar melt compositions. Calculated SCSS values, combined with previously estimated S contents of lunar low-Ti basalts and primitive pyroclastic glasses, suggest their source regions were not sulfide saturated. Even when correcting for the currently inferred maximum extent of S degassing during or after eruption, sample S abundances are still > 700 ppm lower than the calculated SCSS values for these compositions. To achieve sulfide saturation in the source regions of low-Ti basalts and lunar pyroclastic glasses, the extent of degassing of S in lunar magma would have to be orders of magnitude higher than currently thought, inconsistent with S isotopic and core-to-rim S diffusion profile data. The only lunar samples that could have experienced sulfide saturation are some of the more evolved A17 high-Ti basalts, if sulfides are Ni- and/or Cu rich.

Sulfide saturation in the source regions of lunar melts is also inconsistent with the sulfide-silicate partitioning systematics of Ni, Co and Cu. Segregation of significant quantities of (non)-stoichiometric sulfides during fractional crystallization would result in far larger depletions of Ni, Co and Cu than observed, whereas trends in their abundances are more likely explained by olivine fractionation. The sulfide exhaustion of the lunar magma source regions agrees with previously proposed low S abundances in the lunar core and mantle, and by extension with relatively minor degassing of S during the Moon-forming event. Our results support the hypothesis that refractory chalcophile and highly siderophile element systematics of low-Ti basalts and pyroclastic glasses reflect the geochemical characteristics of their source regions, instead of indicating the presence of residual sulfides in the lunar interior.

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1. INTRODUCTION

Sulfur (S) is a volatile element and understanding its origin, abundance, and distribution in planetary interiors is important due to the effects of the sulfur cycle on properties of planetary crusts, atmospheres and, in the case of the Earth, the biosphere (Farquhar et al., 2000). Constraining S abundances in lunar reservoirs is important to constrain the early volatile budget of, and volatile fluxes in, the Earth-Moon system (e.g., Wing and Farquhar, 2015; Righter et al., 2017a,b; Steenstra et al., 2017a,b). One key aspect of the lunar sulfur cycle relates to the question whether the lunar interior was ever saturated in sulfide minerals. Sulfides are sinks for chalcophile elements that will preferentially partition into these phases (Kiseeva and Wood, 2013; Mungall and Brenan, 2014; Wood et al., 2014; Steenstra et al., 2017a). Many of the chalcophile elements are also (highly) volatile and their depletion in samples derived from the lunar interior may provide important constraints on models of the early evolution of the Earth-Moon system (Paniello et al., 2012; Wang and Becker, 2013; Steenstra et al., 2016; Wang et al., 2016; Kato and Moynier, 2017; Righter et al., 2017a,b). Quantifying chalcophile element depletions however requires knowledge about the presence or absence of sulfides in the lunar interior. The interpretations of the systematics of highly siderophile element (HSE's) abundances in lunar samples are also highly dependent on the assumption of sulfide undersaturation or saturation in the lunar mantle, given the fact that the HSE behave significantly less siderophile in the presence of FeS (Mungall and Brenan, 2014; Laurenz et al., 2016).

Several previous studies focused on determination of the SCSS of terrestrial compositions (Wendlandt, 1982; Mavrogenes and O'Neill, 1999; Liu et al., 2007; Jugo et al., 2005; Wykes et al., 2015; Fortin et al., 2015; Smythe et al., 2017). Lunar melts are distinctly different due to their higher TiO₂ and FeO contents, where low-Ti basalts contain < 6 wt.% TiO₂ and high-Ti basalts > 6 wt.% (Neal and Taylor, 1992). Some studies determined the SCSS values for high-FeO martian basalts, but these compositions have low TiO₂ contents (Righter et al., 2009; Ding et al., 2014). Only three studies experimentally determined the solubility of S for a limited range of high-Ti lunar melt compositions (Danckwerth et al., 1979; O'Neil and Mavrogenes, 2002; Ding et al., 2017).

Gibson et al. (1975, 1976) and Brett (1976) argued for sulfide saturation in the Apollo 17 source regions, based on SCSS data of terrestrial basalts. The latter studies did not explore the effects of pressure (*P*), temperature (*T*) or a wider range of (lunar) melt compositions. Danckwerth et al. (1979) studied the solubility of S in the A17 high-Ti basalt 74275 as a function of its FeO content and found that up to 3400 ppm S could be dissolved in this composition at a temperature of 1523 K and a pressure of 1 bar. Given the S contents measured in this basalt, they concluded that A17 high TiO₂ basalts were not sulfide saturated near their liquidus temperatures. O'Neil and Mavrogenes (2002) studied the overall effects of TiO₂ and reported a subtle increase of SCSS with increasing silicate melt TiO₂ content. The most recent study (Ding et al., 2017) experimentally determined the SCSS for an average Luna 16 mare basalt composition (TiO₂ ~5 wt.%) and the Apollo 11 B3 mare basalt (TiO₂ ~11 wt.%). Using a revised model for predicting the SCSS, Ding et al. (2017) found that the SCSS at the conditions of last equilibration of intermediate and high-Ti lunar basalts and glasses are higher than the measured S contents in these samples. This suggests that no sulfide retention occurred in the lunar mantle during these partial melting events.

Determination of the SCSS for low- and high-Ti melts over a wide *P-T* and compositional range is required to assess if the source regions of lunar magmas sampled during the Apollo missions were sulfide saturated. Here, we quantify the SCSS for a suite of low- to high-Ti lunar melt compositions (synthetic equivalents of Apollo 14 (A14) black and yellow glass, Apollo 15 (A15) green glass, Apollo 17 (A17) orange glass and a composition representative of late-stage lunar magma ocean (LMO) residual melt), together covering a range in TiO₂ content of 0.2–25 wt.%, as a function of *P-T* (Tables 1–4). The new SCSS dataset is combined with previously determined SCSS data for high-FeO silicate melts to obtain predictive SCSS model values specifically suitable for high-FeO lunar melts. This model is then used to calculate the SCSS values for the various lunar low- and high-Ti melts and resulting values are compared with the measured S contents in these samples to assess the likelihood of sulfide saturation in their source regions.

Sulfide saturation in the lunar mantle can also be addressed by studying siderophile element systematics in lunar samples in conjunction with the experimentally

Table 1
Measured starting compositions of silicate melt (in wt.%) obtained with EPMA.

	<i>N</i>	SiO ₂	FeO	MgO	CaO	Al ₂ O ₃	TiO ₂	Cr ₂ O ₃	MnO	K ₂ O	Total
A14BG ^a	17	32.1(1)	23.3(1)	12.2(0)	7.0(0)	4.8(0)	18.9(1)	0.96(1)	0.33(1)	0.1(0)	99.8(1)
A14YG ^{a,b}	–	–	–	–	–	–	–	–	–	–	–
A15GG ^a	17	48.3(1)	15.6(6)	17.2(4)	8.9(0)	7.3(0)	0.25(1)	0.53(1)	0.19(1)	–	98.3(1)
A17OG ^a	25	37.4(3)	21.0(2)	11.1(2)	8.7(0)	8.1(2)	10.1(5)	0.72(1)	0.32(2)	0.013(4)	97.7(1)
LBS10 ^c	25	48.2(3)	16.8(1)	4.37(2)	9.91(2)	10.9(1)	6.7(1)	–	–	0.015(3)	97.1(1)

^a Composition after Delano (1986a).

^b The Apollo 14 yellow glass (A14YG) starting composition was not measured.

^c Composition after Lin et al. (2017).

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