



New bulk sulfur measurements of Martian meteorites and modeling the fate of sulfur during melting and crystallization – Implications for sulfur transfer from Martian mantle to crust–atmosphere system



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ABSTRACT

Sulfur storage and transport between different reservoirs such as core, mantle, crust and atmosphere of Mars are tied to igneous processes. Martian meteorites carry a record of mantle melting and subsequent differentiation history of Martian magmas. Investigation of S geochemistry of Martian meteorites can thus provide an understanding of how S is transferred from the Martian interior to the exosphere. In this study we measured bulk S concentration of 7 Martian meteorites and modeled the behavior of S during both isobaric crystallization of primary Martian magmas and isentropic partial melting of Martian mantle. Comparisons between measured data and modeled results suggest that (1) sulfides may become exhausted at the source during decompression melting of the mantle and mantle-derived basalts may only become sulfide-saturated after cooling and crystallization at shallow depths and (2) in addition to degassing induced S loss, mixing between these differentiated sulfide-saturated basaltic melts and cumulus minerals with/without cumulate sulfides could also be responsible for the bulk sulfur contents in some Martian meteorites. In this case, a significant quantity of S could remain in Martian crust as cumulate sulfides or in trapped interstitial liquid varying from 2 to 95 percent by weight. Our modeling also suggests that generation of sulfide-undersaturated parental magmas requires that the mantle source of Martian meteorites contain <700–1000 ppm S if melting degree estimation of 2–17 wt.% based on compositions of shergottites is relevant.

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1. Introduction

Sulfur fluxes and storage are important in understanding processes of terrestrial planets from the core to exosphere. Sulfur is thought to be a dominant volatile element on Mars (Gaillard et al., 2012; Gibson et al., 1985; King and McLennan, 2010; Stewart et al., 2007). Understanding S distribution among Martian mantle, crust and the atmosphere as well as the fate of sulfur in various magmatic processes is important for constraining the differentiation history of Mars.

The primary source of sulfur to the Martian surface–atmosphere system is volcanism (Gaillard and Scaillet, 2009; Halevy et al., 2007; Johnson et al., 2008). Further the distribution of S between Martian crust and atmosphere, which can potentially affect the ancient climate on Mars and the formation of sulfate deposits on the surface, is also intimately related to the behavior of sulfur dur-

ing magmatic differentiation including cooling, crystallization, and degassing. The amount of S released to the Martian atmosphere may be estimated as the difference between (1) the S content of the mantle-derived magma and (2) the budget of magmatic S trapped in the basaltic crust. The former could be estimated by S contents of basaltic melts at sulfide saturation (SCSS) (Ding et al., 2014; Richter et al., 2009), assuming mantle derived magma is sulfide saturated. The latter is usually approximated by S contents in Martian meteorites, although bulk S concentration data of the latter remained limited until recently (Franz et al., 2014; Lodders, 1998). Based on the assumptions that erupted Martian basalts are sulfide saturated and the difference between the expected solubility limit and the measured concentration in olivine-phyric shergottites reflects S loss owing to degassing, Richter et al. (2009) estimated that mantle derived basalts on Mars degassed 2400 ppm S. Similar approaches are also applied to constrain sulfur degassing flux for terrestrial settings (e.g., Self et al., 2008; Zhang et al., 2013). However, both above assumptions can be problematic. First, it is not considered whether sulfide, if present in the Martian mantle, can remain in the residue during partial melting;

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Second, most Martian meteorites contain cumulus minerals and some have experienced secondary alteration (weathering/impact effects), which could either deplete or enrich S in meteorites. Another approach to estimate magmatic degassing of S is by simulating equilibrium degassing of primary magmas, which has been quantitatively investigated by Gaillard et al. (2012). A maximum of 2100 ppm S degassing from an initial concentration of 3500 ppm S in primitive Martian magmas is obtained in Gaillard et al. (2012) for hydrated-oxidized melts (0.4 wt.% H₂O, FMQ-0.5). The calculations by Gaillard et al. (2012) consider several parameters, such as atmospheric pressure, oxygen fugacity of emplaced magmas and other volatiles (H₂O and CO₂) contents in the melts, for the degassing process. However, these authors do not consider the possible SCSS and S concentration changes in the evolving melt before degassing, though by analogy to the case on Earth, both SCSS and S concentration in the melt must change during fractional crystallization because SCSS is a function of pressure, temperature, oxygen fugacity, and melt composition (Baker and Moretti, 2011; Holzheid and Grove, 2002; Jégo and Dasgupta, 2013, 2014; Jugo, 2009; Jugo et al., 2010; Lee et al., 2012; Li and Ripley, 2009; Liu et al., 2007) which in turn would control the onset of sulfide precipitation. How sulfur behaves during emplacement and crystallization of basalt into the crust is a missing link in sulfur cycle. Magmatic S budget of Martian crust is key information in this regard, which needs to be evaluated in the context of differentiation of primitive Martian basalts. Therefore here we focus on S budgets of Martian meteorites.

Martian meteorites range from evolved basaltic shergottites with small fractions of phenocrysts to cumulates with small or negligible amount of trapped liquids. Sulfur contents of different types of Martian meteorites thus can provide important insights into igneous processes that operated on Mars. Righter et al. (2009) calculated sulfur carrying capacity change along differentiation path of a model Martian magma taken from Symes et al. (2008) and compared the result to the S contents of some olivine-phyric and basaltic shergottites. Similarly, Ding et al. (2014) applied their new SCSS model to investigate the change in SCSS along the liquid line of descent and sulfide precipitation in corresponding cumulates derived from Yamato 98049. However, no study has combined petrology of different Martian meteorites and their bulk S data and interpreted them in the context of magma generation via melting of a sulfide-bearing mantle and differentiation of various putative parental melts via shallow fractional crystallization.

In this study we measured bulk S contents of 7 Martian meteorites using solution ICP-MS (Erdman et al., 2014) and discuss the bulk sulfur data of meteorites from this study and those from literature by modeling the fate of S during fractional crystallization of Martian magmas and the fate of sulfide during mantle partial melting. By comparing bulk S data of Martian meteorites to the modeling results, we estimate the plausible S budgets of the Martian mantle, crust, and atmosphere.

2. Methods

2.1. Analytical technique

Bulk S contents of 7 Martian meteorites (the shergottites Los Angeles, Zagami, NWA 856, NWA 1068, and Tissint, and the nakhlites NWA 998 and Nakhla) were measured using high mass-resolution solution ICP-MS following the method in Erdman et al. (2014). Interior chips of each of these Martian meteorites were obtained from the meteorite collection in the Center for Meteorite Studies at Arizona State University. Fractions, ranging from ~13 to ~60 mg, were taken from each of these interior chips. For three of the samples (NWA 856, NWA 1068 and NWA 998), two separate fractions were taken to assess sample heterogeneity. Each of

these fractions was then dissolved and processed separately. One of the fractions of NWA 998 was leached with 1 mol/part HCl first. After being sonicated and centrifuged, the supernatant and solid residues were carefully separated. The supernatant was directly diluted by 1 mol/part HCl while the solid residues went through same steps as other bulk rock fractions. Sulfur in the bulk rock fractions was dissolved and converted to the S⁶⁺ species in the form of SO₄²⁻. Both bulk rock solutions and the leachate were analyzed with the Thermo Finnigan Element 2 magnetic sector ICP-MS at Rice University. To check the accuracy of the sulfur analyses, several terrestrial rock standards including one basalt (United States Geological Survey, BHVO-2), one gabbro (Japanese Geological Survey, JGb-1) and one marine mud (United States Geological Survey, MAG-1) were run as external standards in the same sequence. The limit of detection on sulfur ranges from 40 to 90 ppm, determined by the instrumental detection limit in solution and the dilution factor (Erdman et al., 2014).

2.2. Geochemical data compilation

We have compiled the bulk S data of Martian meteorites from this study and previous studies. 32 Martian meteorites with available bulk rock mineralogy and composition are included in Table 1. Most of the 32 Martian meteorites, including 7 measured in this study, belong to 3 groups: basaltic shergottite, ol-phyric shergottite and nakhlite. In addition, two lherzolitic shergottites (LEW88516 and ALH77005), one dunite (Chassigny) and one orthopyroxenite (ALH84001) are also included in the data compilation.

Basaltic shergottites chiefly comprise pyroxene and plagioclase without early crystallizing olivine or chromite (Papike et al., 2009). Stolper and McSween (1979) demonstrated by optical analysis and melting experiments that both Shergotty and Zagami contain cumulus pyroxene, probably without cumulus plagioclase. Cumulus pyroxenes are commonly observed in basaltic shergottites with a few exceptions such as Los Angeles (Rubin et al., 2000) and QUE94201 (McSween et al., 1996), which have been shown as liquid composition. Ol-phyric basalts share the petrographic features of olivine megacrysts, presence of chromite and Fe-Ti oxides, and low augite proportions (Goodrich, 2002; Papike et al., 2009). Textural studies and experimental results suggest that at least some olivine-phyric shergottites contain excess olivine (Filiberto and Dasgupta, 2011; Goodrich, 2003, 2002; Sarbadhikari et al., 2009; Usui et al., 2008).

Nakhlites are clinopyroxenites composed mostly of cumulus grains of subcalcic augite as the most abundant mineral and less abundant olivine and mesostasis (Treiman, 2005). Lherzolitic shergottites are also cumulates containing mainly poikilitic olivine often enclosed by pyroxene along with minor modal plagioclase (Papike et al., 2009).

Chassigny has similar texture and mineralogy to terrestrial dunites (Floran et al., 1978). Chassigny contains 91.6% olivine, 5% pyroxene, 1.7% feldspar, 1.4% chromite, 0.3% melt inclusions and trace amount of other minerals (Floran et al., 1978). Orthopyroxenite consists of only one sample, ALH84001, which contains 97% coarse grained orthopyroxene, 2% chromite, 1% plagioclase and other accessory minerals (Mittlefehldt, 1994; Papike et al., 2009).

Throughout the paper, we follow the definition of cumulus phase from Stolper and McSween (1979) as crystals accumulated or concentrated in a liquid; intercumulus liquid as the liquid from which the cumulus crystals formed and which once surrounded these crystals. Cumulate fraction refers to the mass fraction of cumulate minerals. Melt fraction refers to the mass fraction of intercumulus liquid, which is assumed to be the difference between 1 and cumulate fraction. In order to estimate the plausible fraction of cumulate minerals in Martian meteorites, we first approximate mass fraction of each mineral using the mineral modes (pyroxene,

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