



Chondritic late accretion to Mars and the nature of shergottite reservoirs

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ABSTRACT

Mars is considered to have formed as a planetary embryo that experienced extensive differentiation early in its history. Shergottite meteorites preserve evidence for this history, and for late accretion events that affected their mantle sources within Mars. Here we report the first coupled ^{187}Re – ^{187}Os , $^{87}\text{Sr}/^{86}\text{Sr}$, highly siderophile element (HSE: Os, Ir, Ru, Pt, Pd, Re) and major element abundance dataset for martian shergottites that span a range of MgO contents, from 6.4 to 30.3 wt.%. The shergottites range from micro-basalt to basaltic-andesite compositions, have enriched to depleted incompatible trace-element compositions, and define fractional crystallization trends, enabling the determination of HSE compatibility for martian magmatism in the order: $\text{Os} > \text{Ir} \geq \text{Ru} \gg \text{Pt} \geq \text{Pd} \geq \text{Re}$. This order of compatibility is like that defined previously for Earth and the Moon, but the fractionation of strongly compatible Os, Ir and Ru appears to take place at higher MgO contents in martian magmas, due to early onset of sulfide fractionation. In general, enriched shergottites have lower MgO contents than intermediate or depleted shergottites and have fractionated HSE patterns ($\text{Re} + \text{Pd} + \text{Pt} > \text{Ru} + \text{Ir} + \text{Os}$) and more radiogenic measured $^{87}\text{Sr}/^{86}\text{Sr}$ (0.7127–0.7235) and $^{187}\text{Os}/^{188}\text{Os}$ (0.140–0.247) than intermediate or depleted shergottite meteorites ($^{87}\text{Sr}/^{86}\text{Sr} = 0.7010$ – 0.7132 ; $^{187}\text{Os}/^{188}\text{Os} = 0.127$ – 0.141). Osmium isotope compositions, corrected for crystallization age, define compositions that are implausibly unradiogenic in some enriched shergottites, implying recent mobilization of Re in some samples. Filtering for the effects of alteration and high Re/Os through crystal-liquid fractionation leads to a positive correlation between age-corrected Sr and Os isotope compositions. Mixing between hypothetical martian crustal and mantle reservoirs are unable to generate the observed Sr–Os isotope compositions of shergottites, which require either distinct and discrete long-term incompatible-element depleted and enriched mantle sources, or originate from hybridized melting of deep melts with metasomatized martian lithosphere. Using MgO-regression methods, we obtain a modified estimate of the bulk silicate Mars HSE composition of (in ng g^{-1}) 0.4 [Re], 7.4 [Pd], 9.6 [Pt], 6.2 [Ru], 3.7 [Ir], 4 [Os], and a long-term chondritic $^{187}\text{Os}/^{188}\text{Os}$ ratio (~ 0.1312). This result does not permit existing models invoking high-pressure and temperature partitioning of the HSE. Instead, our estimate implies 0.6–0.7% by mass of late accretion of broadly chondritic material to Mars. Our results indicate that Mars could have accreted earlier than Earth, but that disproportional accretion of large bodies and a relative constant flux of accretion of available materials in the first 50–100 Ma of Solar System led to the broad similarity in HSE abundances between Earth and Mars.

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1. Introduction

Planets within the Solar System formed from common accretion processes that ultimately led to different outcomes in their final mass and composition. To zeroth order, differences in planetary mass and composition can be explained by accretion from a nebu-

lar disk that was heterogeneous with respect to mass distribution and to a lesser or greater degree in chemical components. However, planetary formation processes are difficult to elucidate, most especially as the later stages of planet accretion remain enigmatic. For example, Mars is considered to have accreted by collisions between 1000–5000 km diameter planetary embryos (Chambers and Wetherill, 1998), and reached half of its present-day size in only 1.8 million years or less (Dauphas and Pourmand, 2011), which is far more rapid than the presumed accretion time-scale for Earth of >50 Ma (Kleine et al., 2009). Among the distinct geochemical

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and structural features of Mars are the estimated elevated abundances of the highly siderophile elements (HSE: Au, Re, Pd, Pt, Rh, Ru, Ir, Os) in its mantle, which appear to be like that in Earth (Day et al., 2016). This geochemical feature has been explained through late accretion of broadly chondritic materials to Mars (Brandon et al., 2000, 2012), perhaps during an early and large impact event (Bottke et al., 2010; Brasser and Mojzsis, 2017). Alternatively, the elevated HSE abundances in Mars have been explained through high-pressure and temperature metal-silicate partitioning within a magma ocean (Righter et al., 2015). To distinguish between these models, properly determining the relative and absolute abundances of the HSE within bulk silicate Mars, the relative timing of their delivery, and explaining the similar distribution of these elements between Mars and Earth remain fundamental science questions.

Currently, determination of the composition of Mars requires the use of meteorites that are delivered to Earth through impact events on the martian surface. Of the variety of martian meteorites, the shergottites are the most geochemically diverse, and can be described as broadly basaltic and magmatically-derived rocks. Geochemical groupings, based on incompatible trace element abundances and lithophile isotope systematics have led to classification of shergottites as depleted (low La/Yb, $^{87}\text{Sr}/^{86}\text{Sr} < 0.712$) intermediate, and enriched (high La/Yb, $^{87}\text{Sr}/^{86}\text{Sr} > 0.712$) variants. Shergottite geochemical diversity implies long-lived incompatible trace element depleted- and enriched-sources in Mars, either representing incompatible trace-element enriched crust and depleted mantle mixing (Jones, 1989), or highly-differentiated mantle sources (e.g., Symes et al., 2008; Basu Sarbadhikari et al., 2009). To date, no martian mantle samples have been recognized in the meteorite collection. Consequently, the martian mantle or bulk silicate Mars composition must be estimated from the properties of martian meteorites, including the shergottites, nakhlites (cumulate clinopyroxenites), chassignites (cumulate dunites), and ancient martian meteorites, including an orthopyroxenite (Allan Hills 84001), and a basaltic regolith breccia (Northwest Africa 7033/7533 and their pairs).

We have investigated a suite of shergottite meteorites to better understand fractional crystallization and mixing processes that their parental magmas experienced, and to investigate and more precisely define the distribution of the highly siderophile elements (HSE: Os, Ir, Ru, Pt, Pd, Re,) within bulk silicate Mars. Our new data, for 21 shergottite meteorites, significantly expands and complements existing Re–Os isotope and HSE data (Birck and Allègre, 1994; Brandon et al., 2000, 2012; Jones et al., 2003; Baumgartner et al., 2017; Goderis et al., 2016) and provides the first coupled analysis of ^{187}Re – ^{187}Os , $^{87}\text{Sr}/^{86}\text{Sr}$, HSE concentration and major-element data for the same martian meteorite sample aliquots, obviating issues related to inter-laboratory bias or sample heterogeneity. We utilize these data to examine the lower apparent Pt and Pd abundances estimate for bulk silicate Mars, relative to the terrestrial mantle (Day et al., 2016), and to examine models for the distribution of the HSE within Mars, including models of high-pressure temperature partitioning, or late accretion.

2. Samples and methods

Seven Antarctic martian shergottite meteorites were obtained from the Meteorite Working Group (Larkman Nunatak [LAR] 12095, LAR 06319, LAR 12240,7 and LAR 12011,12), Roberts Massif [RBT] 04262; Elephant Moraine [EETA] 79001 and Allan Hills [ALHA] 77005. The remainder of the shergottite meteorites came from the Royal Ontario Museum's collection (Northwest Africa [NWA] 3171, NWA 5298, NWA 6342, NWA 6963, NWA 7042, NWA 7257, NWA 7397, NWA 7721, NWA 8679, NWA 10593, Dhofar [Dho] 019, Dar Al Gani [DaG] 476 and Tissint). For the desert finds, a Wells low-loss diamond wire saw was used to access material away from the

fusion crust surface. For all materials, weathering or fusion crust surfaces were removed prior to generation of fine-ground sample powders that were prepared using a clean alumina mortar and pestle.

Analytical procedures were undertaken at the *Scripps Isotope Geochemistry Laboratory (SIGL)*. For major-element abundances and Sr isotopes, ~50 mg of sample powder was digested in Teflon-distilled concentrated HF (4 mL) and HNO₃ (1 mL) for >72 hrs on a hotplate at 150 °C, along with total procedural blanks and terrestrial basalt and andesite standards (BHVO-2, BCR-2, BIR-1a, AGV-2). Samples were sequentially dried and taken up in concentrated HNO₃ to destroy fluorides, followed by doping with indium to monitor instrumental drift during analysis, and then diluted to a factor of 50,000 for major-element determination. Major-element abundances were obtained using a *ThermoScientific* iCAP Qc quadrupole inductively coupled plasma mass spectrometer (ICP-MS) in low resolution mode. For major-elements, Si was derived by difference, with reproducibility of other elements measured on the BHVO-2 reference material being better than 3%, except Na₂O (7.1%). To assess the veracity of our technique, we measured 15 ocean island basalt and mid-ocean ridge basalt samples using both solution ICP-MS and X-ray fluorescence and find good agreement between the two methods (*Supplementary Information; Table S1*).

Strontium isotopic compositions and Rb and Sr abundances were obtained on residual solutions used for major-element abundances. Samples were up-taken in 4M HNO₃ and loaded onto Sr-specification resin, where Sr was separated from other elements using the method described in Moynier et al. (2012). Separated Sr-cuts were measured on a *ThermoScientific* Triton thermal ionization mass spectrometer, using TaF₅ as the activator. Blanks were between 21 and 59 pg of Sr and were negligible relative to total Sr in loaded masses (<1%). 300 ng loads of the NBS 987 Sr standard gave an average $^{87}\text{Sr}/^{86}\text{Sr}$ value of 0.710233 ± 5 ($n = 7$). Three separate digestions of the BHVO-2 standard gave 0.703458 ± 2 , which is within uncertainty of the suggested value from GEOREM of 0.703469 ± 17 (Jochum et al., 2005).

Osmium isotope and HSE abundance analyses were performed using methods described in Day et al. (2016b). Homogenized powder aliquots and total procedural blanks were digested in sealed borosilicate Carius tubes, with isotopically enriched multi-element spikes (^{99}Ru , ^{106}Pd , ^{185}Re , ^{190}Os , ^{191}Ir , ^{194}Pt – Rh and Au are monoisotopic, and were not measured), and 8 mL of a 1:2 mixture of multiply Teflon distilled HCl, and multiply Teflon distilled HNO₃ that was treated with H₂O₂ to purge Os. Samples were digested to a maximum temperature of 270 °C in an oven for 72 hrs. Osmium was triply extracted from the acid using CCl₄ and then back-extracted into HBr, prior to purification by micro-distillation. Rhenium and the other HSE were recovered and purified from the residual solutions using standard anion exchange separation techniques. Isotopic compositions of Os were measured in negative-ion mode on a *ThermoScientific* Triton thermal ionization mass spectrometer. Rhenium, Pd, Pt, Ru and Ir were measured using a *Cetac* Aridus II desolvating nebuliser coupled to a *ThermoScientific* iCAP q ICP-MS. Offline corrections for Os involved an oxide correction, an iterative fractionation correction using $^{192}\text{Os}/^{188}\text{Os} = 3.08271$, a ^{190}Os spike subtraction, and finally, an Os blank subtraction. Reported precision for $^{187}\text{Os}/^{188}\text{Os}$, determined by repeated measurement of the UMCP Johnson–Matthey standard was better than $\pm 0.2\%$ (2 St. Dev.; 0.11385 ± 15 ; $n = 10$). Measured Re, Ir, Pt, Pd and Ru isotopic ratios for sample solutions were corrected for mass fractionation using the deviation of the standard average run on the day over the natural ratio for the element. Reproducibility of HSE analyses using the iCAP q ICP-MS was better than 0.4% (2 St. Dev.) for 0.5 ppb solutions, and all reported values are blank corrected. The total procedural blanks ($n = 4$) run with the samples had $^{187}\text{Os}/^{188}\text{Os} = 0.168 \pm 0.043$, with quantities (in picograms)

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