



On the iron isotope composition of Mars and volatile depletion in the terrestrial planets



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ABSTRACT

Iron is the most abundant multivalent element in planetary reservoirs, meaning its isotope composition (expressed as $\delta^{57}\text{Fe}$) may record signatures of processes that occurred during the formation and subsequent differentiation of the terrestrial planets. Chondritic meteorites, putative constituents of the planets and remnants of undifferentiated inner solar system bodies, have $\delta^{57}\text{Fe} \approx 0\%$; an isotopic signature shared with the Martian Shergottite–Nakhilite–Chassignite (SNC) suite of meteorites. The silicate Earth and Moon, as represented by basaltic rocks, are distinctly heavier, $\delta^{57}\text{Fe} \approx +0.1\%$. However, some authors have recently argued, on the basis of iron isotope measurements of abyssal peridotites, that the composition of the Earth's mantle is $\delta^{57}\text{Fe} = +0.04 \pm 0.04\%$, indistinguishable from the mean Martian value. To provide a more robust estimate for Mars, we present new high-precision iron isotope data on 17 SNC meteorites and 5 mineral separates. We find that the iron isotope compositions of Martian meteorites reflect igneous processes, with nakhilites and evolved shergottites displaying heavier $\delta^{57}\text{Fe} (+0.05 \pm 0.03\%)$, whereas MgO-rich rocks are lighter ($\delta^{57}\text{Fe} \approx -0.01 \pm 0.02\%$). These systematics are controlled by the fractionation of olivine and pyroxene, attested to by the lighter isotope composition of pyroxene compared to whole rock nakhilites. Extrapolation of the $\delta^{57}\text{Fe}$ SNC liquid line of descent to a putative Martian mantle yields a $\delta^{57}\text{Fe}$ value lighter than its terrestrial counterpart, but indistinguishable from chondrites. Iron isotopes in planetary basalts of the inner solar system correlate positively with Fe/Mn and silicon isotopes. While Mars and IV-Vesta are undepleted in iron and accordingly have chondritic $\delta^{57}\text{Fe}$, the Earth experienced volatile depletion at low (1300 K) temperatures, likely at an early stage in the solar nebula, whereas additional post-nebular Fe loss is possible for the Moon and angrites.

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1. Introduction

How planets in the inner solar system accreted and differentiated is a question fundamental to understanding the chemical differences that exist between them today. Iron offers a unique opportunity to address this question. Not only is it abundant in each of the major planetary reservoirs, the crust, mantle and core, it exists in three different oxidation states – Fe^{3+} , Fe^{2+} and Fe^0 – the relative proportions of which are constrained by local redox conditions. These redox states confer different properties to the element, making it capable of tracing a variety of processes.

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Despite the range of iron oxidation states, its isotopes are remarkably constant in chondritic meteorites, identical to the IRMM-014 reference material (Schoenberg and von Blanckenburg, 2006; Craddock and Dauphas, 2011; Needham et al., 2009; Theis et al., 2008; Wang et al., 2013); i.e., $((^{57}\text{Fe}/^{54}\text{Fe})_{\text{chondrite}} / (^{57}\text{Fe}/^{54}\text{Fe})_{\text{IRMM-014}} - 1) * 1000 = \delta^{57}\text{Fe} = 0\%$. This constancy is independent of chondrite class, thus, if planets accreted from chondritic material, deviations from 0‰ should reflect post-nebular processes. Mass-dependent fractionation laws dictate that, in general, Fe^{3+} -bearing phases are heavier than Fe^{2+} -bearing phases, i.e., $\delta\text{Fe}^{3+} - \delta\text{Fe}^{2+} = \Delta\text{Fe}^{3+-2+} > 0\%$ (Polyakov and Mineev, 2000; Shahar et al., 2008), although no apparent isotopic fractionation exists between Fe^{2+} and Fe^0 under terrestrial core-forming conditions (Hin et al., 2012; Poitrasson et al., 2009; Roskosz et al., 2006).

Using this behaviour as a guide, causes for isotopic differences in basaltic rocks from different planetary bodies can be evaluated. Initial studies considered that $\delta^{57}\text{Fe}$ in the BSE $\approx 0.1\%$, owing to the overlap of ultramafic to intermediate magmatic rocks, (Beard and Johnson, 2004; Poitrasson et al., 2004). This signature is also observed in the lunar low-Ti basalts (Liu et al., 2010; Poitrasson et al., 2004; Weyer et al., 2005), whereas the lunar high-Ti basalts are systematically enriched in $\delta^{57}\text{Fe}$ ($\approx 0.2\%$). Iron isotope signatures for the Earth–Moon system, based on basaltic rocks, are resolvably heavier than estimates for SNC meteorites (Anand et al., 2006; Poitrasson et al., 2004; Wang et al., 2012; Weyer et al., 2005) and the Howardite–Eucrite–Diogenite rocks from asteroid IV-Vesta (Poitrasson et al., 2004; Schoenberg and von Blanckenburg, 2006; Wang et al., 2012; Weyer et al., 2005), both of which give chondritic values. Further, the recognition of basaltic achondrites extending to even heavier values than those of the Earth–Moon system (i.e., angrites, $\approx 0.2\%$) suggests heavy isotope enrichment is a consequence of planetary differentiation. Possible explanations include vaporisation and loss of isotopically-light Fe during planetary collisions (Poitrasson et al., 2004), isotope fractionation during mantle partial melting (Dauphas et al., 2009; Weyer and Ionov, 2007) and fractionation by high-pressure phases in the lower mantle during core formation (Polyakov, 2009; Williams et al., 2012).

However, and while not widely agreed upon (see, e.g., Zhao et al., 2012; Poitrasson et al., 2013) some recent studies on the composition of mantle peridotites (Craddock et al., 2013; Weyer and Ionov, 2007) suggest a BSE value that is lighter ($\delta^{57}\text{Fe} \approx 0.04 \pm 0.04\%$) than mean values for terrestrial and lunar basalts, as represented by Mid-Ocean Ridge Basalt, MORB ($\delta^{57}\text{Fe} \approx 0.1\%$; Teng et al., 2013) and the low-Ti suite, respectively. Importantly, this value is similar to chondrites, Mars and IV-Vesta, removing the need to appeal to fractionation mechanisms between Earth and these bodies. In order to shed light on the isotopic characteristics of Mars, we have analysed 17 whole-rock SNC meteorites and 5 mineral separates using high-precision techniques (Poitrasson and Freydisier, 2005; Sossi et al., 2015). Through identification of the primary and parental magma compositions of SNCs, and applying corrections for the effects of magmatic differentiation, the iron isotopic composition of Martian primary magmas is calculated and shown to be lighter than that of primary MORB, meaning the Martian and terrestrial mantles are isotopically distinct. A comparison of planetary basalts shows a systematic dependence on Fe/Mn ratios, implying a common process leading to iron isotope fractionation. Mass balance constrains the transfer of material to be significant on the planetary-scale, namely by volatile depletion or core-formation. We interpret these results in the context of the accretion and differentiation histories of the terrestrial planets.

2. Samples and methods

2.1. Samples and dissolution

Samples Shergotty, Zagami, Los Angeles, SaU 005, NWA 1183, Nakhla, Governador Valadares, Lafayette and Chassigny were provided by the Natural History Museum, London. Samples Y 980459 and Y 000953 were obtained from the National Institute for Polar Research, Japan. The Australian National University samples DaG 476 and Zagami were purchased from a meteorite dealer, whereas samples ALHA 77005, EETA 79001A, LAR 06319, RBT 4262 and MIL 03346 were provided by the Meteorite Working Group (MWG), NASA, in the form of 1 g rock chips, free of fusion crusts. Exactly 500 mg of these chips was hand-crushed in a boron carbide mortar and pestle to create a fine powder. Of this batch, 100 mg was dissolved in duplicate for the stable isotope analyses, first by a mixture of concentrated HCl–HF–HNO₃ in the ratio 1:0.5:0.2 in a 3 mL Savillex Teflon beaker on a table-top hotplate set to $\approx 130^\circ\text{C}$.

After drying down in nitrate form, 15 M HNO₃ with a few drops of HF were added to the samples and then placed, still in their 3 mL Teflon beaker, inside 20 mL FEP Teflon vessels with 2–3 mL of MQ H₂O to create a vapour pressure, and inserted into steel bombs. These were left to dissolve in an oven set to $\approx 210^\circ\text{C}$ for 7 days. Subsequent evaporation with addition of concentrated HNO₃ confirmed that the samples had been dissolved in their entirety.

2.2. Column chromatography and mass spectrometry

Samples were analysed in two batches – those marked ‘Poitrasson’ were processed at Geosciences Environnement Toulouse (GET) CNRS laboratory, following the procedure detailed in Poitrasson and Freydisier (2005). Owing to the reproducibility of data acquired during multiple sessions over the period, standard-sample bracketing (SSB) results were used rather than the combined SSB and Ni-doped regression method of Poitrasson and Freydisier (2005). Samples labelled ‘Sossi’ were analysed at the Australian National University, following the procedure described in Sossi et al. (2015). Three well-characterised standard materials, Allende, BHVO-2 and Milhas Hematite were run throughout to ensure total procedural accuracy and precision were maintained (Table 1) in agreement with published values (Sossi et al., 2015).

3. Results

Iron isotope measurements for the SNC meteorites are summarised in Table 1. Whole-rock major- and trace-element data and literature iron isotope data are listed in Appendix A.

The iron isotope compositions of whole rocks show a restricted range, $-0.07 \pm 0.01 < \delta^{57}\text{Fe}(\%) < +0.10 \pm 0.03$, overlapping with those of both chondrites and mantle peridotites. Measured $\delta^{57}\text{Fe}$ values are inversely correlated with whole-rock MgO contents (Fig. 1a), mimicking trends observed in terrestrial mafic rocks (Sossi et al., 2012; Teng et al., 2008). Within the shergottites, a similar correlation is found with iron isotopes and CaO (Fig. 1b), with the nakhlites displaced towards higher CaO values as a result of clinopyroxene accumulation. Similar systematics are observed when compared with other indices of magmatic differentiation, such as Ni, Sc and modal olivine content (Fig. 1c). Individual analyses compare favourably with other recent measurements (shown in grey, Fig. 1), and most notably with recent high precision measurements (Wang et al., 2012). For meteorites analysed in both studies ($n = 7$), a correlation coefficient of 0.6 is observed, respectable considering the limited isotopic range (0.15%) compared to analytical uncertainty ($0.03\text{--}0.05\%$). This argues against any significant sample heterogeneity or bias in analytical methods, such that values reported herein can be interpreted with confidence.

Although based on a smaller dataset ($n = 5$), pyroxene (from nakhlites) and olivine (from Chassigny) separates define an even narrower spread than the whole rocks, extending from $\delta^{57}\text{Fe} = -0.04 \pm 0.05\%$ to $+0.05 \pm 0.06\%$. Calculated pyroxene–whole rock fractionation factors are resolvable in the Y 000593 and Lafayette nakhlites, where $\Delta^{57}\text{Fe}_{\text{px-nak}} = -0.08 \pm 0.01\%$ and $-0.07 \pm 0.05\%$, respectively (Fig. 1d). Pyroxene–nakhlite pairs from Nakhla and MIL 03346 show no resolvable difference, a result mirrored by the olivine–chassigny pair (Fig. 1d).

4. Discussion

4.1. Petrogenesis of Martian SNCs

In attempting to link the geochemistry and iron isotope compositions of SNC meteorites to that of the Martian mantle and terrestrial rocks, estimates of parental magma compositions are required. On the basis of geochemical criteria, Martian basalts are derived

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