



A complex history of silicate differentiation of Mars from Nd and Hf isotopes in crustal breccia NWA 7034

Rosalind M.G. Armytage^{a,*}, Vinciane Debaille^a, Alan D. Brandon^b, Carl B. Agee^c

^a Laboratoire G-Time, CP 160/02, Université Libre de Bruxelles, Av. F. Roosevelt 50, 1050 Bruxelles, Belgium

^b Department of Earth and Atmospheric Sciences, University of Houston, Houston, TX, 77204, USA

^c Institute of Meteoritics, University of New Mexico, Albuquerque, NM, 87131, USA

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ABSTRACT

Resolving the possible mantle and crustal sources for shergottite meteorites is crucial for understanding the formation and early differentiation of Mars. Orbiter and rover characterization of the martian surface reveal that the major element composition of most of its surface does not match the shergottites (McSween et al., 2009) leaving the relationship between them poorly understood. The identification of the meteorite NWA 7034 and its pairs as a Mars surface rock (Cartwright et al., 2014) provides access to a representative sample of Mars' crust (Agee et al., 2013; Humayun et al., 2013). Utilizing the short-lived ^{146}Sm – ^{142}Nd , and long-lived ^{147}Sm – ^{143}Nd and ^{176}Lu – ^{176}Hf chronometers, which are sensitive to silicate differentiation, we analyzed three fragments of NWA 7034. The very negative mean isotopic compositions for this breccia, $\mu^{142}\text{Nd}_{\text{Jndi-1}} = -45 \pm 5$ (2SD), $\varepsilon^{143}\text{Nd}_{\text{CHUR}} = -16.7 \pm 0.4$ (2SD) and $\varepsilon^{176}\text{Hf}_{\text{CHUR}} = -61 \pm 9$ (2SD) point to an ancient origin for this martian crust. However, modeling of the data shows that the crust sampled by NWA 7034 possesses a Hf/Nd ratio and coupled $\varepsilon^{143}\text{Nd}$ – $\mu^{142}\text{Nd}$ model age that are incompatible with this crustal reservoir being an end-member that generated the shergottite source mixing array. In addition, this crust is not juvenile, despite its rare earth element profile, but has had a multistage formation history. Therefore, early crustal extraction alone was not responsible for the creation of the reservoirs that produced the shergottites. Instead mantle reservoirs formed via other early differentiation processes such as in a Mars magma ocean must be responsible for the trace element and isotopic signatures present in shergottites.

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1. Introduction

The martian shergottite meteorites have a range of igneous crystallization ages from ~ 170 Myr to ~ 574 Myr (Nyquist et al., 2001; Brennecka et al., 2014). They exhibit significant variation in their incompatible trace element (ITE) contents and radiogenic isotopic compositions, indicative of multiple distinct sources (e.g. Borg et al., 2002). Based on these bulk rock characteristics, shergottites fall into three subgroups (depleted, intermediate, and enriched), forming compositional arrays that can be generated by mixing between a martian reservoir depleted in ITE, and one that is more ITE-enriched. These reservoirs have also been shown to differ in redox state, with a range in oxygen fugacity -2.5 to -1 log units relative to the quartz–fayalite–magnetite buffer (e.g. Herd et al., 2002). The depleted end-member is thought to repre-

sent the depleted martian mantle (Borg and Draper, 2003; Debaille et al., 2007, 2008). The enriched end-member reservoir remains enigmatic with both the martian crust (Humayun et al., 2013) or ITE-enriched late cumulates resulting from the crystallization of a martian magma ocean (Blichert-Toft et al., 1999; Borg and Draper, 2003; Debaille et al., 2007) being proposed. From coupling the short-lived ^{146}Sm – ^{142}Nd and long-lived ^{147}Sm – ^{143}Nd chronometers in shergottites, the resultant mixing line between their sources has been interpreted as a planetary isochron, representing either a single major differentiation event on Mars ~ 50 Myr after solar system formation (SSF) (Borg et al., 2003; Caro et al., 2008), or the termination of protracted crystallization of a martian magma ocean (MMO) ~ 100 Myr after SSF (Debaille et al., 2007, 2009). As such, identifying the enriched end-member for the shergottite mixing array is crucial for understanding the nature of differentiation on early Mars and its timing.

The martian crust is an obvious candidate for the enriched end-member reservoir that is added to shergottite magmas through assimilation-fractional crystallization (AFC) processes, as ITEs will be concentrated in the extracted melts that cool to form crust

* Corresponding author. Now at: Jacobs/JETS, NASA Johnson Space Center, 2101 NASA Parkway, Mailcode X13, Houston, TX, 77058, USA.

E-mail address: rosalind.m.armytage@nasa.gov (R.M.G. Armytage).

during silicate differentiation. Explaining the shergottite ITE and isotopic compositional arrays in terms of crustal AFC is problematic when considered in conjunction with the textural diversity and major element variations in these meteorites (Borg and Draper, 2003; Ferdous et al., 2017). In addition, a crustal contamination origin does not easily replicate the correlation between the initial $\epsilon^{143}\text{Nd}$ and $\gamma^{187}\text{Os}$ in shergottites (Brandon et al., 2012). An alternative scenario where the enriched mantle source formed as a late-stage residual melt during crystallization of a MMO (Borg et al., 2016; Brandon et al., 2012; Debaille et al., 2007) can account for the decoupling. The similarity of many of the compositional outcomes for shergottites in both scenarios, requires additional independent constraints to examine their relative likelihood, and hence our understanding of the early evolution of Mars.

The discovery of a unique martian polymict breccia meteorite (NWA 7034 meteorite, and its pairs, Agee et al., 2013; Humayun et al., 2013), and the confirmation of its martian origin (Cartwright et al., 2014) allows for such additional testing of the two scenarios. The major element composition of NWA 7034 matches the orbital and rover data bulk compositions of the martian crust (Agee et al., 2013; Humayun et al., 2013), unlike shergottites which do not match the composition of majority of the martian surface (McSween et al., 2009). The meteorite contains clasts with 4.428 Ga igneous zircons (Humayun et al., 2013) consistent with ancient exposed crust on Mars, and older than the 4.1 Ga orthopyroxenite ALH 84001 (Lapen et al., 2010). The clast types in NWA 7034 include igneous lithologies (basalt, basaltic andesite, trachyandesite, and a Fe-Ti-P rich lithology) as well as sedimentary, and those of impact origin (Santos et al., 2015). Despite the textural diversity and major element composition variation between the clasts, the trace element profiles of the component clasts as well as the matrix are remarkably similar, with the exception of the monzonite (similar to the trachyandesite from Santos et al., 2015) clasts (Humayun et al., 2013). The ITE concentrations in bulk analyses of NWA 7034 are considerably higher than in enriched shergottites, and their rare earth element (REE) patterns emulate a partial melt of a primitive mantle composition (Humayun et al., 2013), a signature of “juvenile” crust that conflicts with the petrologic diversity in the clasts (Hewins et al., 2016; Santos et al., 2015). As such this meteorite represents ancient crustal material that could conceivably be the enriched end-member reservoir for the shergottite source mixing arrays, providing a unique opportunity to test this hypothesis that has implications for the earliest differentiation history of Mars.

Radiogenic chronometers sensitive to silicate differentiation such as $^{146,147}\text{Sm}$ – $^{142,143}\text{Nd}$ and ^{176}Lu – ^{176}Hf can address whether crust represented by NWA 7034 is the ITE enriched end-member for shergottites, and also provide constraints on the formation of currently exposed ancient martian crust. Here we report $^{146,147}\text{Sm}$ – $^{142,143}\text{Nd}$ and ^{176}Lu – ^{176}Hf systematics, and REE abundances for three bulk breccia fragments of NWA 7034. Modeling of the chemical and isotopic data was carried out to determine the origin of the NWA 7034 crust and its relationship to the shergottites, and explore the concomitant implications for the early silicate differentiation history of Mars.

2. Methods

2.1. Sample preparation and chemistry

As NWA 7034 is a polymict breccia (e.g. Santos et al., 2015), three separate bulk fragments A, B, and C (fragments 1, 2, and 3 from Goderis et al., 2016) were measured to assess any possible isotopic heterogeneity. The three chips of NWA 7034 were individually crushed using an agate pestle and mortar, specific to achondrite samples. The respective dissolved sample powder

masses were 123.4 mg, 118.2 mg, and 106.4 mg. Dissolution was initially by HF-HNO₃ in Savillex vials on the hotplate at 110 °C. An additional dissolution step was carried out in Parr bombs at 120 °C to ensure the destruction of refractory phases such as zircon. The resulting fluorides were attacked with multiple steps of HCl evaporation to ensure clear solutions. Subsequent to dissolution, a ~5% aliquot was removed and spiked with a ^{176}Lu – ^{179}Hf and a ^{150}Sm – ^{148}Nd spike. These aliquots were passed over a four-column procedure to isolate Hf, Nd, Sm and Lu, beginning with a cation exchange resin to isolate a Hf cut and a REE (rare earth element) cut from the matrix. The REEs were subsequently separated from each other using HDEHP (di(2-ethylhexyl)orthophosphoric acid) resin, created in-house from Teflon beads and HDEHP. The Hf cut was purified using first an anion column to remove Fe, followed by an Ln-spec column (Eichrom). Two other ~5% aliquots were removed, one for trace element analysis on the 7700 Agilent ICP-MS (inductively-coupled-plasma mass spectrometer) at Université Libre de Bruxelles (Table 1), and an additional 5% aliquot was removed for future possible isotope analysis. The remainder was passed over a seven column procedure to isolate Nd, Hf and Sm for isotopic ratio analysis. The main difference from the chemistry for the spiked aliquots (besides using a separate set of columns and smaller mesh sizes) is the purification of the REE cut. After the first pass on the HDEHP column, the Nd cut is oxidized with a NaBrO₃–10M HNO₃ mixture and passed over another, smaller HDEHP column to remove the oxidized Ce (modified after Li et al., 2015). The cut is then passed over a 0.5 ml cation resin column to remove the Na prior to analysis. The Sm cut is passed a second time over the HDEHP column, but with a slight change in acid molarity to ensure the complete removal of possible Nd interferences. Total procedural blanks are on the order of Hf < 160 pg, Lu < 26 pg, Nd < 74 pg and Sm < 6 pg, where the mass of each element that was processed was on the order Hf ~ 500 ng, Lu ~ 40 ng, Nd ~ 1500 ng, and Sm ~ 400 ng.

2.2. Isotope mass spectrometry

All the elemental and isotope analyses were carried out at Université Libre de Bruxelles (ULB). The spiked aliquots and the unspiked Hf were measured on the Nu Instruments Nu Plasma HR multi-collector inductively-coupled-plasma mass spectrometer (MC-ICP-MS) using an Aridus II desolvator. The unspiked Hf was introduced into the plasma using a 75 $\mu\text{l}/\text{min}$ nebulizer and an Aridus II desolvator. The samples were bracketed every two samples using the JMC 475 standard to correct for mass fractionation. Potential isobaric interferences were monitored using ^{175}Lu , ^{172}Yb and ^{182}W and found to be below the level of detection. The mean value for the standard was $^{176}\text{Hf}/^{177}\text{Hf} = 0.282174 \pm 0.00019$ ($n = 20$) and the sample data were normalized to the literature value of $^{176}\text{Hf}/^{177}\text{Hf} = 0.282163 \pm 0.00009$ (Blichert-Toft et al., 1997) (Table 2). The spiked aliquots were run at concentrations between 10–25 ppb, and corrected for mass fractionation using an exponential law followed by spike stripping using an iterative method (Debaille et al., 2007). The error on the $^{147}\text{Sm}/^{144}\text{Nd}$ and $^{176}\text{Lu}/^{177}\text{Hf}$ ratios is taken to be 5%, based on previous analysis of total procedural replicates, though the errors on the individual concentrations are much smaller as they are based purely on that of the spike, which is better than 0.5%. The measured $^{147}\text{Sm}/^{144}\text{Nd}$ and $^{176}\text{Lu}/^{177}\text{Hf}$ ratios of an aliquot of the USGS (US Geological Survey) standard BHVO-2, processed the same way, were 0.155, and 0.0093 respectively. These values correspond to the reference values of $^{147}\text{Sm}/^{144}\text{Nd} = 0.150$ and $^{176}\text{Lu}/^{177}\text{Hf} = 0.0097$ (Wilson, 1997), which are within the assigned 5% uncertainty. The unspiked high-precision Sm and Nd isotopic analyses were carried out on the Triton Plus TIMS (thermal ionization mass spectrometer) instrument also at ULB. Both were run on double Re filaments loaded

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