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## Silicon isotopic variation in enstatite meteorites: Clues to their origin and Earth-forming material

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### ABSTRACT

Of the primitive meteorite groups, the enstatite chondrites are among the most chemically dissimilar to terrestrial, which should preclude them as major components of the proto-Earth. However, for many isotope systems (most notably oxygen), enstatite chondrites show very little variation away from terrestrial, hinting at a common origin. One isotope system which appears to differ from this trend is silicon, but no satisfactory explanation has been proposed as to why these meteorites should be significantly lighter than both the silicate Earth and other primitive meteorite groups.

This study presents a comprehensive investigation into the Si isotope composition of enstatite chondrites and their differentiated counterparts, the aubrites, and confirms that these meteorites are, with respect to Si isotopes, the lightest macroscale solar system objects so far analysed. Crucially, the results show that EH chondrites are significantly lighter ( $\delta^{30}\text{Si} = -0.77 \pm 0.08\%$ ) than EL chondrites ( $\delta^{30}\text{Si} = -0.59 \pm 0.09\%$ ) and aubrites ( $\delta^{30}\text{Si} = -0.60 \pm 0.11\%$ ). Silicon isotope analyses of the metal-free components of EH and EL reveal that these are identical, within error, to each other and with carbonaceous/ordinary chondrite bulk measurements (viz.,  $\delta^{30}\text{Si} \sim -0.47$ ), which is taken as evidence that Si isotope variation in the nebular gas is not the cause of the light Si isotope enrichment in enstatite chondrites. From this, one can infer that silicates condensing from the solar nebula over a wide range of compositions and, presumably, heliocentric distances have very similar Si isotope compositions. A statistically significant negative correlation between bulk  $\delta^{30}\text{Si}$  and Si content in enstatite chondrite kamacite indicates that the presence of isotopically light Si in the metal phase (as the result of formation under reducing conditions) is the principal cause of the bulk enrichment in lighter Si isotopes.

Based on the Si isotope offset between enstatite meteorites and the silicate Earth, the extant enstatite meteorites cannot represent a significant proportion of the material that accreted to form the proto-Earth, as unfeasibly large amounts of Si would have to enter the core to satisfy mass balance equations. However, we posit that material enriched in refractory lithophile elements which condensed in the same region as E-chondrites, could still be an important component.

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### 1. Introduction

The enstatite chondrites (E-chondrites) are unique amongst the primitive (undifferentiated) meteorites, in that they have formed under extremely reducing conditions (e.g., Mason, 1966; Keil, 1989). As a result, these meteorites are composed predominantly of Fe-poor enstatite ( $\text{MgSiO}_3$ , typically with Fe contents < 1%), Si-bearing Fe–Ni metal (kamacite) as well as a variety of rare mineral phases (e.g., Mg-, Ca- and Mn-sulphides, TiN and  $\text{Si}_2\text{N}_2\text{O}$ ) not typically stable on Earth (Keil, 1968). Another feature of E-chondrites is that they appear to have undergone significant refractory lithophile element (RLE) fractionation, as evinced by

low Mg/Si, Al/Si ratios, compared to other primitive meteorites (Larimer and Anders, 1970; Baedeker and Wasson, 1975). Such compositions might preclude their involvement as precursor materials of the proto-Earth; however, of all of the primitive meteorites groups, they are isotopically the most similar materials to Earth.

In particular, E-chondrites exhibit no isotopic variations in O (Newton et al., 2000), N (Javoy et al., 1986), Mo (Dauphas et al., 2002), Ni (Regelous et al., 2008), Cr and Ti (Trinquier et al., 2007, 2009) and Sr (Moynier et al., 2012) relative to terrestrial composition, which, in most cases, is not true for the ordinary (O-) and carbonaceous (C-) chondrite groups. This has led some authors to suggest that E-chondrites are the primary building blocks of Earth (Javoy, 1995; Javoy et al., 2010). However, the Mg/Si ratio of bulk E-chondrites (0.65–0.75) is the lowest of all primitive meteorites, much lower than that of Bulk Silicate Earth (BSE = 1.05; Jagoutz et al., 1979); such a

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substantial fractionation of these elements is difficult to explain by any known process that might occur during Earth's accretion and differentiation (e.g., Drake and Righter, 2002). If this dichotomy is to be resolved, more information into the processes of formation and composition of E-chondrites is required.

Silicon is one element where there are apparent resolvable isotopic differences between E-chondrites and BSE yet, of the three major chondrite groups, E-chondrites are the least well characterised in terms of Si isotopes. Only 8 bulk analyses of 6 separate meteorites are present in the literature (EH: Abee, Indarch, Sahara 87168, St. Marks; EL: Khairpur, Pillistfer; Georg et al., 2007; Chakrabarti and Jacobsen, 2010; Armytage et al., 2011; Fitoussi and Bourdon, 2012). Intriguingly, E-chondrites are the lightest meteorite samples analysed in all of the above studies, often exhibiting Si isotope compositions that are resolvably lighter than O- and C-chondrite samples. The Si isotope system therefore has the potential to reveal new insights into E-chondrites.

In the last decade interest in the Si isotope system has increased, largely due to the discovery that BSE is non-chondritic with respect to Si isotopes (Georg et al., 2007). This implies that, if Earth accreted from chondritic material, some process or variety of processes have fractionated the Si isotopic composition of BSE during or post-accretion. The most common explanation for this offset is that Si is one of the "light elements" present in the Earth's core (e.g., Birch, 1964; Poirier, 1994) and that partitioning of Si between metal and silicate has preferentially enriched the silicate phase in the heavier isotopes (Georg et al., 2007; Fitoussi et al., 2009; Shahar et al., 2009, 2011; Ziegler et al., 2010; Armytage et al., 2011). This isotopic offset can be used to calculate, via mass-balance, an independent estimate of the amount of Si present in the core. For instance, using an isotopic offset of  $\Delta^{30}\text{Si}_{\text{BSE-chond}} = 0.15\text{‰}$  (where  $\Delta^{30}\text{Si}_{\text{BSE-chond}} = \delta^{30}\text{Si}_{\text{BSE}} - \delta^{30}\text{Si}_{\text{chondrite}}$ ; see Section 2 for definition of  $\delta$  value) Armytage et al. (2011) calculated an estimate of  $\sim 9$  wt% Si in the core. Although this is based on a number of non-trivial assumptions (Section 4.5), this value is consistent with estimates derived from other criteria (McDonough, 2003).

The very light Si isotopic composition of E-chondrites has led workers to preclude these meteorites as representative of major components of the Earth; Armytage et al. (2011) omitted E-chondrites from their "meteorite\*" average (\* denoting the absence of E-chondrite data in this mean) when calculating  $\Delta^{30}\text{Si}_{\text{BSE-chond}}$ , and Fitoussi and Bourdon (2012) specifically used the significant Si isotopic offset between BSE and E-chondrites to suggest that only  $< 15\%$  of Earth's precursor material could be derived from E-chondrite-like material. This is because the large  $\Delta^{30}\text{Si}_{\text{BSE-E-chondrite}}$  value (in some cases up to  $\sim 0.30\text{‰}$ ) implies that unrealistically large amounts (up to  $\sim 23$  wt%) of Si should be present in the Earth's core, which is implausible based on physical and chemical estimates (Badro et al., 2007).

However, as mentioned above, these conclusions are based on scant data and, furthermore, in none of the studies has there been a specific attempt to explain why E-chondrites are isotopically lighter than other meteorites. Therefore, here we investigate the Si isotope composition of E-chondrites by analysing a representative suite of such meteorites as well as their differentiated counterparts, the aubrites.

## 2. Samples and analytical methods

The enstatite chondrites are classified as either Fe-rich EH chondrites (Fe  $\sim 30$  wt%) or Fe-poor EL chondrites (Fe  $\sim 25$  wt%; Sears et al., 1982). Associated with E-chondrites are the differentiated enstatite achondrites, the aubrites. Aubrites are composed predominantly of Fe-free enstatite and have mineralogies consistent with formation under very reducing conditions

(Keil, 1989, 2010). Although most aubrites are fragmental or regolith breccias, their chemistry indicates formation as a result of partial (or total) melting and fractional crystallisation of material similar to that of the E-chondrite parent body(ies) (e.g., Wolf et al., 1983; Keil, 2010). Petrological and geochemical studies (Keil, 1989; Lin and El Goresy, 2002), cosmic ray exposure ages (e.g., Eugster, 2003) and extinct radionuclide systematics (Shukolyukov and Lugmair, 2004) all suggest that EH, EL and aubrites derive from separate bodies that formed in close proximity in the solar nebula. Based on the reduced mineralogy of all three groups, it is often suggested that these materials formed closer to the Sun than both O- and C-chondrites, at around 1 AU (Baedecker and Wasson, 1975; Kallemeyn and Wasson, 1986).

The samples analysed in this research comprise a representative selection of both the enstatite chondrites and achondrites (see Table 1): six EH samples (four falls and two finds); seven EL samples (five falls and two finds); three aubrites (all falls) and the anomalous achondrite Shallowater (thought to have formed on a separate parent body; Keil, 1989). The E-chondrite samples span a range of petrological grades (EH3, 4, 5 and EL3 and 6), which refer to that sample's degree of thermal alteration, 3 being the least altered and 6 being the most equilibrated. For comparison we also analysed three primitive terrestrial samples (two ocean-island basalts and a peridotite), four carbonaceous chondrites (Allende, Cold Bokkeveld, Murray and PCA91467) and an ordinary chondrite (Parnallee). Previous Si isotope studies have better characterised these three sample groups (Savage et al., 2010; Fitoussi et al., 2009; Chakrabarti and Jacobsen, 2010; Armytage et al., 2011); hence we sometimes use their more robust compositional averages in the discussion.

Meteorite samples were received as  $\sim 2$ – $5$  g pieces and ground to a fine powder using an agate pestle and mortar. The use of agate as a powdering medium does not contaminate the Si isotope composition of a sample (Savage et al., 2011; Zambardi and Poitrasson, 2011). To further test this, a separate piece of Khairpur (EL6) was ground in an alumina pestle and mortar; the Si isotope compositions of both powders are indistinguishable (Table 1). In addition, E-chondrite silicate (metal-free) fractions were separated for analysis with the use of a hand magnet. These fractions were processed as for the bulk samples. For the aubrites, any magnetic phases were removed before analysis; this is because, although nominally metal-free, some aubrites retain large masses of metal that have not been removed during differentiation, which could lead to significant sample inhomogeneity.

Samples were processed using a HF-free alkali fusion technique, as described in detail by Georg et al. (2006) and in the Supporting Online Material (SOM) (S1.1 and S1.2). Sample Si concentrations were measured on a photospectrometer; yields typically exceeded 95%. Before isotope analysis, the samples were purified for Si using a single-step cation exchange chromatography procedure, as described in Georg et al. (2006) and in Section S1.3. This method quantitatively removes all cationic species from the matrix and results in  $> 97\%$  recovery of Si. All samples and standards were processed through this chemistry before analysis.

Silicon isotope measurements were made on a Thermo Scientific Neptune Plus Multi-Collector Inductively-Coupled-Plasma Mass-Spectrometer (MC-ICP-MS) at Washington University in St. Louis, running at medium resolution. Analytical details, procedures and instrumental running conditions are described in Section S1.5 and Table S4. Isotope measurements were calculated using the standard-sample bracketing protocol relative to the silica sand standard NBS28. Silicon isotope variations are defined using the delta notation ( $\delta^{30}\text{Si}$  or  $\delta^{29}\text{Si}$ ) as the per mil (‰) deviation from the standard, as such:

$$\delta^x\text{Si} = [({}^x\text{Si}/{}^{28}\text{Si})_{\text{sample}}/({}^x\text{Si}/{}^{28}\text{Si})_{\text{NBS28}} - 1] \times 1000$$

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