Si AND Fe ISOTOPE HETEROGENEITY IN ENSTATITE CHONDRITES: IMPLICATIONS FOR NEBULAR FRACTIONATION AND TERRESTRIAL CORE FORMATION. J. Sikdar[1], H. Becker[1] and J. A. Schuessler[2]. [1]Institut für Geologische Wissenschaften, Freie Universität Berlin (jinia@zedat.fu-berlin.de), [2]GFZ German Research Centre for Geosciences, Potsdam, Earth Surface Geochemistry, Telegrafenberg, Potsdam.

Introduction: Geochemical studies along with metal-silicate partitioning experiments carried out at high temperatures and pressures have established that the core of Earth likely contains a minor fraction of Earth's total silicon budget (~3-8 wt.% Si, [1-3]). Accordingly, the elevated δ^{56}Fe and δ^{30}Si of bulk silicate Earth (BSE) relative to chondrites by at least ~0.1‰ was interpreted as a fingerprint of Earth's core-mantle segregation event [3-4]. Based on the assumption that the isotopic offset between BSE and chondrites reflects equilibrium isotope fractionation during core formation, a number of subsequent studies have been carried out to deduce the physico-chemical conditions prevailing at the core-mantle boundary of Earth, estimate the nature of light elements in Earth's core and trace the composition of the potential building blocks of Earth and other terrestrial planets [3-6].

However, the measured mean force constants of iron bonds in basaltic glasses and iron-rich alloys suggest negligible equilibrium Fe isotope fractionation between silicate and metal at the proposed conditions of metal-silicate equilibration (3000-4000 K, 40-60 GPa [7]). Also, the small metal-silicate Si isotope fractionation factor at high temperatures question the role of equilibrium Si isotope fractionation during metal-silicate segregation as a main cause for the significant isotopic offset between BSE and bulk chondrites [8]. Therefore, it has been hypothesized that the isotopic offset between BSE and chondrites could be associated with nebular fractionation rather than equilibrium isotope fractionation during planetary core formation [8, 9]. Hence, the effect of nebular chemistry in controlling the Si and Fe isotopic signature of undifferentiated solar system materials and terrestrial rock samples should be better explored.

To distinguish the extent to which Earth might have inherited its Si and Fe isotopic signature from solar nebula processing or evolved as a result of core formation (or both), we have carried out high precision Si and Fe isotope analyses in different components of EH3 and EH4 type enstatite chondrites (EC). Formed under highly reduced conditions, EC are the only class of undifferentiated meteorites where Si is found to alloy with metallic Fe, with Si concentration reaching up to 5 wt. % in metals. Since materials similar to enstatite chondrites may have contributed to the growth of Earth and other terrestrial planets [10], enstatite chondrites and their components might yield useful information on nebular isotopic fractionation processes that affected the building materials of terrestrial planets.

Materials and Methods: In the present work, we have carried out Si and Fe isotopic analyses of bulk meteorites, metal-sulfide spherules, matrix metals and magnetic/slightly-magnetic/non-magnetic fractions from following enstatite chondrites - Sahara 97072, Kota Kota, MAC 88184, Abee and Indarch. The metal-troilite spherules (MTS) are round to sub-round ~50 to 800 µm sized concentrically layered opaque assemblages found in least equilibrated EH3 chondrites [11]. Matrix metal grains, on the other hand, are anhedral to subhedral shaped metal grains (100-500 µm) found in close association with the metal spherules. Because of their prominent size, both matrix metals and MTS were sampled after visual inspection under binocular microscope. The magnetic, slightly magnetic and non-magnetic fractions of EC were separated using a hand-magnet based on their decreasing magnetic susceptibility. For Si isotope analyses, samples were digested using NaOH flux in Teflon vials at 190°C and column chromatography was carried out using 1.8 ml AG 50W-X12 (200–400 mesh) resin to purify Si [12]. In the case of metallic fractions, a larger amount of resin (3 ml) was used and column chromatography was typically repeated twice to efficiently separate Si from Fe-Ni matrices. For Fe isotope analyses, aliquots of EC components were digested in a HF-HNO_3-HCl mixture and purification of Fe was carried out using 1 ml AG 1-X8 (200–400 mesh) anion exchange resin. After purification, high precision Si and Fe isotope analyses were carried out using the Neptune Plus MC-ICP-MS at GFZ Potsdam. For Si isotope analyses, sample-standard bracketing using NBS-28 and Mg doping was applied, whereas Fe isotope analyses were carried out using sample-standard bracketing with IRMM-014.

Results: The data reveal that Si and Fe isotopes are distributed heterogeneously among different components of enstatite chondrite and δ^{30}Si-δ^{56}Fe values are negatively correlated, *i.e.,* δ^{30}Si becomes progressively heavier from metallic to silicate fractions of EC whereas δ^{56}Fe becomes progressively lighter. The MTS display the lightest δ^{30}Si (-5.4±0.29‰) and the heaviest δ^{56}Fe (0.20±0.04‰). On the other hand, the non-magnetic fractions of EC display much heavier δ^{30}Si (-0.30±0.11‰, which is remarkably similar to BSE) and much lighter δ^{56}Fe (-0.15±0.06‰, compared to 0.01±0.07‰ of terrestrial peridotites). The δ^{30}Si and δ^{56}Fe of matrix metals, magnetic and slightly-magnetic fractions were found to vary between MTS and non-magnetic fractions of EC (Fig. 1).

Discussion: According to the condensation sequence of minerals in highly reduced nebular environments (*i.e.* at C/O >0.8), metals should condense earlier than silicates with a temperature gap of ~400°C [13]. In such environments, early-condensed Fe-Ni metal should become enriched in heavier Fe isotopes, which is consistent with the heavy δ^{56}Fe composition of MTS and matrix metals of EC. Unlike Fe, Si associated with metallic fractions of EC did not form a separate condensed phase but instead alloys with Fe-Ni. Alloy formation is a chemical reaction where the lighter isotopes react faster and thus are concentrated in early-formed Fe-Ni metal. Partitioning of lighter Si isotopes into early-condensed metals in reduced domains of the nebula leads to a reservoir effect, with enrichment of heavy Si isotopes in the residual gas and consequently heavier δ^{30}Si in later-condensed enstatite. Therefore, nebular fractionation associated with condensation from reduced nebular gas is expected to create complementary isotopic signatures of Si and Fe in metallic and silicate reservoirs, which is indicated by the combined Si and Fe isotope data in EC components. Thus the extremely light δ^{30}Si and heavy δ^{56}Fe in metallic fractions of EC and heavier δ^{30}Si/light δ^{56}Fe in its non-magnetic fractions can be explained by the early condensation of Si bearing metals followed by sulfides and silicates in reduced environments.

Enstatite chondrites display close similarity to BSE with respect to numerous isotopic tracers [O, Ca, S, N etc]. Although EC were ruled out as a major building material of Earth [6], this study and other recent work [14] showing similarities in δ^{30}Si between BSE and non-magnetic/silicate fractions of EC suggest that they may have genetic significance. We note that in contrast to Si, δ^{56}Fe of the non-magnetic fractions of EC are considerably lighter than BSE. However, unlike Si, Fe in non-magnetic fractions of EC is predominantly hosted by sulfides and Fe isotopic fractionation related to sulfide formation may have caused the light δ^{56}Fe in non-magnetic fractions of EC.

Therefore, based on the isotopic similarities of Si isotopes in silicate-rich fractions of EC and BSE, it can be said that Earth and EC might have formed from a uniform isotopic reservoir of the inner protoplanetary disk. Thus, Earth may have inherited its Si and Fe isotopic signature predominantly from a mixture of materials derived from reduced and more oxidized nebular reservoirs where isotopic characteristics were set by the partitioning of Si and Fe isotopes between early condensed metals followed by sulfides and silicates at nebular conditions. This would imply that the Si and Fe isotope composition of BSE has not been (or only little) affected by high temperature equilibrium isotope fractionation at core-mantle boundary of Earth.

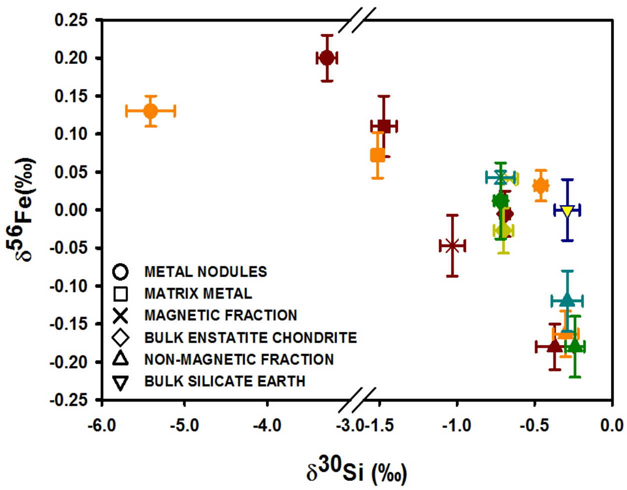

Fig 1: Heterogeneous distribution of Si and Fe isotopes in different components of Enstatite chondrites. Color codes of meteorites: Sahara 97072 (red), Kota Kota (orange), Abee (dark green), Indarch (blue), MAC 88184 (green). Error bars refer to 2SD of the mean of multiple measurements. $\delta^{30}Si_{BSE}$ is taken from [15] and $\delta^{56}Fe_{BSE}$ (mantle peridotites) is from [16].

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