Constraints on chondrule-matrix complementarity from silicon isotope compositions of components of the Allende CV3 meteorite

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Introduction: Carbonaceous chondrites (CC) are mainly composed of different proportions of various types of chondrules, refractory inclusions and fine-grained matrix material [1]. Hezel and Palme [2] found complementarity Mg/Si ratios in chondrules and matrix of some CC. However, the complementarity and formation of chondrules and matrix from a common reservoir was challenged by the study of Olsen et al. [3]. Recently, Dauphas et al. [4] showed that bulk chondrites and other planetary materials display a positive correlation of δ^{30}Si the Mg/Si ratio. This correlation may have been caused by equilibrium isotope fractionation during the condensation of forsterite, which is leaving behind isotopically lighter silicon in gaseous species such as SiO. Silicon isotopic compositions and correlations with major element variations in chondritic components may serve as tools to understand the processing of chondrules and chondrule-matrix complementarity.

In order to evaluate the effects of chondrule formation on Si isotopic fractionation in chondrite components and the complementarity issue, we have determined silicon isotope compositions and corresponding major element abundances of minerals in chondrules and matrix from the carbonaceous chondrite Allende (CV3).

Methods: The major element compositions of olivines and pyroxenes in chondrules and matrix were determined using the 'JEOL JXA 8200 Superprobe' electron microprobe at FU Berlin. Based on their mineralogy and textures, chondrules are classified as type I and type II porphyritic olivine-pyroxene chondrules and barred olivine chondrules. In situ Si isotope composition and major and trace element compositions of olivines and pyroxenes were determined by femtosecond LA-MC-ICPMS and a quadrupole ICP-MS, respectively, using a laser ablation split stream method [5]. Silicon isotopic compositions were determined in high resolution clipped ion beam mode, which resolves interferences from molecular interferences (e.g. C and N oxides like $^{14}N^{16}O^+$ or $^{12}C^{16}O$). δ^{30}Si of samples were determined against NBS-28, a quartz sand reference standard which is reproducible within 0.23 ‰ δ^{30}Si (2SD). The accuracy of δ^{30}Si values was tested by in situ analysis of δ^{30}Si in reference materials with a range of compositions (e.g. NIST SRM 610, BHVO-2, IRMM17, NBS-28) and comparison with δ^{30}Si data of these materials obtained by bulk analysis of δ^{30}Si using fusion, chemical separation of Si and conventional MC-ICPMS. The abundances of major elements were calibrated against SRM 610 and normalized to 100 wt-% element oxides with uncertainties less than 10 % for most elements.

Results and discussion: Chondrules show values ranging from -1.33 ± 0.15 to 0.55 ± 0.20 ‰ (2SE) with overall run precisions varying between 0.08 to 0.21 ‰. The δ^{30}Si of matrix ranges from -0.88 ± 0.11 to -0.26 ± 0.11‰ (2SE) and tends to be lighter and less variable, although isotopic composition of matrix overlap with those in the rims of chondrules. In most chondrules the isotopic composition of Si becomes lighter from chondrule core to the rim. The matrix mainly contains Fe-rich olivine with an iron-content of 30 ± 7wt% (2σ) and Mg# (Mg# = molar Mg/(Mg+Fe²⁺). varying between 0.42 to 0.53. Porphyritic type I and barred olivine chondrules show both Mg# of 0.88 on average, whereas porphyritic type II chondrules display lower values with 0.74 on average. Positive linear correlations between Mg/Si and δ^{30}Si are detectable for all porphyritic type I chondrules and in a few porphyritic type II and barred olivine chondrules, whereas olivines from the matrix do not show any correlation between Mg/Si and δ^{30}Si.

Assuming that chondrules were molten partially or completely, the Mg-rich and isotopically heavier center of the chondrules can be explained as evaporation residues of molten chondrules before recondensation of evaporated chondrule material. The latter process introduced isotopically light Si in the rim area of many chondrules by diffusional processes during cooling of the chondrules. Lighter δ^{30}Si of the Fe-rich matrix olivine suggests later condensation of these grains, also from isotopically light nebular gas. The Si isotope data represents strong evidence for chondrule-matrix complementarity and suggests that it originated from evaporation and condensation processes during chondrule formation.

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