



0.1 Carbon and silicon isotope fractionation in synthetic and natural Silicon Carbide

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Though potentially an important fractionation process, structure-related isotopic fractionation (SRIF) remains very poorly studied. According to this concept an isotopic fractionation between co-crystallised phases and/or domains having different defect densities is induced by different affinities for the light vs. heavy isotopes between crystalline phases with variable crystallochemistry. Estimation of SRIF's magnitude at geologically-relevant temperatures is of considerable interest. In our work we have investigated isotopic composition of synthetic and natural silicon carbide (SiC) using two principal experimental approaches: a) Study of SRIF between polytypes in SiC; and b) Investigation of simultaneously grown C-rich Si_xC_y phases with different crystal chemistries. Since the polytypes have slightly different crystal chemistries and chemical compositions (i.e., ratio Si/C) differences in $\delta^{13}\text{C}$ between them are to be expected. The principal question is the magnitude of this effect.

C and Si isotopic composition of natural SiC grains (heavy fraction from kimberlites, grain size ≤ 0.5 mm) and of SiC wafers grown by the sublimation method at 2700 K and containing several simultaneously grown polytypes were studied by conducting SIMS profiles across clearly visible polytype boundaries after which Raman microspectroscopy was used to verify the position of SIMS spots vs. polytype. Two main polytypes: 6H and 15R were identified. SIMS analyses employed a 500 pA, 17.5 kV

$^{133}\text{Cs}^+$ primary beam. Ion counting based precision values were typically ± 0.5 ‰ for $(^{13}\text{C}/^{12}\text{C})_{meas}$, whereas the observed internal precisions were typically ± 0.6 ‰. Values of ± 0.20 ‰ (counting based) and ± 0.25 and ± 0.45 ‰ (internal) were typical for both the $^{29}\text{Si}/^{28}\text{Si}$ and $^{30}\text{Si}/^{28}\text{Si}$ measured ratios. Carbon in the studied samples was isotopically light with $\delta^{13}\text{C}_{PDB}$ values around -21 - -23 ‰ as determined by gas source mass spectrometry.

The differences in isotopic composition between the polytypes are generally small and no clear differences between the polytypes were found. However, we have observed regions with significant scatter (≥ 1 ‰) in $^{13}\text{C}/^{12}\text{C}$ on the scale ≤ 100 microns in both synthetic and natural SiC. Such scatter was also observed for Si isotopes, in many cases exceeding measurement uncertainty. Keeping in mind the very high growth temperatures of the synthetic samples (~ 2700 K) such an isotopic effect is considerable. The temperature gradient over the growing wafer was very small (~ 10 - 20 K over 5 cm) and the lateral growth was negligible (only the wafer thickness is increasing).

The analysed natural SiC grains were mostly 6H polytype. 4 out of 5 grains have similar $^{13}\text{C}/^{12}\text{C}$, whereas one of the grains was on average 3.5 ‰ lighter. Remarkably, two of the grains appear to be isotopically heterogeneous with variations about 1.5-2 ‰ within a grain.

At present we are investigating different possible causes for the observed isotopic fractionation. One possible explanation for the observed isotopic heterogeneity would be fluctuations of growth rate and/or variable densities of extended defects (such as micropipes) which would lead to preferential trapping of one of the isotopes. Less likely possibility is that the polytype stability and defect density depend on the relative vapour pressures of Si, SiC_2 and Si_2C species, suggesting that significant isotopic fractionation between these gas species may have played a role. Detailed consideration of these possibilities will be given in the presentation. In any case, our study shows that under certain conditions isotopic fractionation may be significant even at extremely high temperatures and relevant to the Geoscience problems.

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