

Dual Si and O Isotope Measurements Using IRMS-BrF₅ Fluorination

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Silicon and oxygen are the most abundant non volatile elements on Earth in the form of silicates. Both elements have three stable isotopes producing ½-slope, mass-dependent fractionation lines on a triple isotope plot of terrestrial samples caused by chemical and biological processes. Non-mass-dependent variations in these isotope systems occur during stellar nucleosynthesis processes. Different methods have been used to analyze these isotopes from the same or separate aliquots of a sample including fluorination-IRMS [1,2], MC-ICP-MS [3] and SIMS [4].

We have made advances in precise determinations of Si- and O-isotopes from a single aliquot of 1 mg silica using a fluorination-IRMS method. Bromine pentafluoride (BrF₅) fluorination is used to extract oxygen and silicon tetrafluoride (SiF₄) from the same sample after reaction in heated Ni-tubes. Triple O-isotope analyses are conducted on O₂ gas collected using 5A molecular sieve. Triple Si-isotope analyses are conducted on SiF₄ gas from the same sample after the collection of O₂. Both Si- and O-isotopes are measured using a custom-built MAT 253 isotope ratio mass spectrometer.

Replicate analyses of NBS-28 have a precision of 0.065‰ and 0.056‰ for δ²⁹Si and δ³⁰Si values, respectively. All Si-isotope data are reported on an NBS-28 corrected scale. Using our method, the Diatomite SRM has a δ²⁹Si_{NBS-28} and δ³⁰Si_{NBS-28} of 0.623 ± 0.060‰ and 1.269 ± 0.060‰, respectively, in good agreement with [5]. Silicified volcanic rocks possess δ³⁰Si values close to 0‰, while chert bands in BIF have heterogeneous δ³⁰Si compositions, which indicates multiple chert precipitation processes. The δ¹⁸O and δ³⁰Si values of biogenic silica phytoliths varied little as a result of partial (up to 30%) dissolution, except under extreme pH conditions, indicating that phytoliths preserved in typical soils may be used as robust paleoclimate indicators. The δ³⁰Si values among BSE, lunar, and enstatite chondrite meteorites vary from 0.3-0.9‰ [6]. This variation requires better resolution to understand planetary processes and we are currently working to resolve these differences.

[1] Leng & Sloane, 2008, *JQS* 23, 313-319. [2] Ding *et al* 2004, *GCA* 68, 205-216. [3] Reynolds *et al* 2007, *JAAS* 22, 561-568. [4] Basile-Doelsch *et al* 2005, *Nature* 433, 399-402. [5] Wille *et al* 2010, *EPSL* 292, 281-289. [6] Fitoussi & Bourden, 2012, *Science* 335, 1477-1480.