

Nickel isotope anomalies: Neutron-rich or neutron-poor?

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The solar system is a mixture of many different nucleosynthetic sources. Numerous studies have investigated the nucleosynthetic origins of the solar system by examining isotopic anomalies in meteorites [e.g. 1]. Isotopic anomalies in the most neutron-rich isotopes of Ca, Ti, Cr in primitive meteorites have been interpreted as incomplete-mixing of a highly neutron enriched nucleosynthetic source into the early solar system [2-4]. This source has been variously hypothesised to be a type Ia supernova [5], type II supernova [6] or an AGB star [7]. However, nearly all of the reported measurements of these anomalies have employed normalisation to a pair of lighter-isotopes, assumed not to be anomalous. Hence, in these cases the inference of a neutron-rich anomaly is a point of interpretation since the anomaly could equally well reside on one of the normalising isotopes.

More recently, Ni has been added to the list of elements exhibiting apparent anomalies in its most neutron-rich isotopes, ⁶²Ni and ⁶⁴Ni [8, 9, 10]. We have made further measurements of Ni isotopes in bulk meteorites, both mass-independently (internally-normalised) and mass-dependently (double-spike), to yield absolute Ni isotope ratios that will better constrain the nucleosynthetic environment from which this incompletely mixed component arose. Preliminary results suggest that, in fact, the anomalies reside on ⁵⁸Ni and not on ⁶²Ni or ⁶⁴Ni. If these findings of neutron-poor anomalies are confirmed, they have far-reaching implications for the nucleosynthetic sources of the Ni isotopic anomalies and the identification of the nucleosynthetic sources of the solar system in general.

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Silica speciation in aqueous sodium silicate solutions

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Aqueous alkali silicate fluids can significantly contribute to the mobility of high field-strength elements in the lower crust and upper mantle [1]. The speciation of silica in these fluids is important for understanding the transport of such elements as alkali-silica complexes. Speciation of aqueous fluids and melts in the Na₂O-SiO₂-H₂O system has been previously studied *in situ* for one composition on the H₂O-NS4 join [2]; here, we investigate silica speciation of fluids over a range of compositions in the Na₂O-SiO₂-H₂O system.

The speciation of dissolved silica in aqueous sodium silicate solutions has been studied *in situ* up to 600°C and 2 GPa using Raman spectroscopy and a Bassett-type hydrothermal diamond-anvil cell [3]. Starting materials were either aqueous NaOH solution plus quartz, or water plus sodium silicate glass (NS2 and NS3), yielding bulk compositions from 10 to 70 wt% SiO₂ and various Na/Si ratios. Composition was determined from the dimensions of the sample chamber and the quartz or glass chip, and the densities of the individual phases. The cell was heated until all phases except zircon were dissolved to yield a single fluid phase, which permitted analysis at known fluid composition. Pressure in the cell was determined from the shift in wavenumber of the ν₃-SiO₄ (~1008 cm⁻¹) band of zircon [4].

The assignment of Raman bands to particular dissolved species remains somewhat elusive, so the interpretation is partly qualitative. The main trends observed in Raman spectra of fluids with increasing SiO₂/H₂O ratio include 1) decrease in the intensity of the ~770 cm⁻¹ band (monomer); 2) increase in the intensity of the ~1050 cm⁻¹ band (assigned to bridging oxygen Si-O stretch [2]); and 3) increase in spectral contributions at ~550-600 cm⁻¹ and ~830-1000 cm⁻¹, which have been assigned to Qⁿ species [e.g. 2]. These trends are interpreted to represent increasing polymerization. Opposite trends, interpreted to indicate decreasing polymerization, are observed with increasing Na/Si ratio.

[1] Manning *et al.* (2008) *EPSL* **272**, 730–737. [2] Mysen (2009) *Geochim. Cosmochim. Acta* **73** 5748–5763. [3] Bassett *et al.* (1993) *Rev. Sci. Instr.* **64**, 2340–2345. [4] Schmidt *et al.* (2011) *MinMag*, this volume.