

## Measurement of mass-independent $\epsilon^{64}\text{Ni}$ isotopic anomalies in meteorites

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Mass-independent isotopic variations in meteorites can yield information about mixing of material with different nucleosynthetic histories in the early solar system [e.g. 1, 2]. As a moderately siderophile, moderately refractory major element with five stable isotopes Ni is an attractive target for study. However, Ni isotopic measurements present a significant analytical challenge, because of the small magnitude of anomalies ( $\sim 50$  ppm [3]). Different results have been reported for Ni isotopic variability between the same range of meteorite groups. Notably, some workers have argued for a range in  $\epsilon^{62}\text{Ni}$  between ordinary and carbonaceous chondrites [3], in keeping with other neutron-rich iron peak nuclides [e.g. 4, 5]. However, others have suggested there is no resolvable mass-independent variability in  $\epsilon^{62}\text{Ni}$ , but this may result from the lower level of precision achieved by some studies [6, 2, 7].

This debate can be further explored using  $^{64}\text{Ni}$ , the most neutron rich, stable Ni isotope. From theory and measurements on CAIs, larger  $\epsilon^{64}\text{Ni}$  variability is expected than  $\epsilon^{62}\text{Ni}$  [8]. Even with these larger potential variations, resolving  $\epsilon^{64}\text{Ni}$  anomalies remains problematic owing to difficulties in obtaining accurate measurements given the presence of an isobaric  $^{64}\text{Zn}$  interference.

Here we present advances, building on the work of Regelous *et al* [3], in both the chemical separation, with a dramatically reduced Zn blank, and the MC-ICP-MS procedures. Preliminary data from this study show larger range of  $\epsilon^{64}\text{Ni}$  anomalies with respect to  $\epsilon^{62}\text{Ni}$  and increased precision of  $\epsilon^{64}\text{Ni}$  data ( $\sim \pm 20$  ppm 2s.d.). These advances will provide a new tool with which to draw inferences about early solar system processes, the provenance of meteorites and the protolith of Earth.

[1] Cameron and Truran (1977) *Icarus*, **30** 447–461 [2] Chen, *et al.* (2009) *Geochim et Cosmochim Acta*, **73** 1461–1471 [3] Regelous *et al.* (2008) *EPSL*, **212** 330–338 [4] Trinquier *et al.* (2007) *APJ* **655** 1179–1185 [5] Niemeyer *et al.* (1985) *GRL* **12** 733–736 [7] Dauphas, *et al.* (2008) *APJ*, **686** 560–569. [6] Cook, *et al.* *Analytical Chemistry* (2006) **78** 8477–8484 [8] Birck and Lugmair (1988) *EPSL* **90** 131–143

## Thermodynamics of natural geothermal systems

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During the last two decades extensive data on fluid chemistry of geothermal fluids in Iceland and in other locations have been collected. Samples include non-volcanic and volcanic waters and vapour, hot springs, mud pools, and single and two phase well discharges. The fluid composition range from ambient to over 300°C, salinity from <10 to >30,000 ppm Cl and pH between <1.5 and >10. This data set provide a unique opportunity to study gas-water-rock interaction and equilibrium state, fluxes and composition of source fluids and effects of boiling (phase separation) on the chemistry and evolution of natural geothermal fluids.

Quantitative interpretation of geothermal fluid chemistry has involved aqueous thermodynamic speciation and *in situ* speciation measurements, the study of mineral and gas-water saturation, liquid-vapour separation models and reaction path modelling both including thermodynamic and kinetic data. The results indicate that equilibrium has been attained between most major components and the associated alteration minerals of the deep aquifer fluids, the concentrations depending on temperature as well as source fluid and primary rock composition. Exceptions are observed in the case of high magmatic gas flux into the system. Upon ascent to the surface and subsequent cooling and boiling the equilibrium state changes. Some components respond to these changes, like observed for calcite precipitation upon boiling and phase segregation. Others do not, like silica concentrations in hot springs that are commonly close to quartz saturation at the aquifer temperatures. Moreover, redox sensitive components like sulphur, hydrogen and carbon, may not show equilibrium species distribution between various oxidation states at temperatures as high as <300°C. This is based on *in situ* determination of various species and oxidation states. Based on these findings it is concluded that the redox chemistry of natural open aqueous systems are controlled by steady state between supply of the elements to the system and the kinetics of reactions involving these elements.