

How did chondrules/CAIs acquire their O anomalies?

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The O isotopes of type B CAIs and chondrules, except in enstatite chondrites, lie on mixing lines with slopes of ~ 1 . These O isotopic compositions were likely established by gas-melt exchange. The initial isotopic compositions of the gas and the dust/melt remain unresolved. The Earth-Moon, Mars, CI- and E-chondrites, fall on very similar mass fractionation lines, consistent with the bulk composition of the nebula being close to terrestrial values. The pattern of O isotopic compositions in CC-chondrite chondrules and CAIs points to dust/melt ($\delta^{17,18}\text{O} \approx -50\%$) exchanging with a gas composition lying near $\delta^{17,18}\text{O} \approx 0\%$ (e.g. Clayton, 1993). The lack of mass independent fractionation in EC chondrules hints at a fractionation mechanism that is redox sensitive.

The main O-bearing species in the nebular gas were H_2O and CO. Modelling indicates that under relatively oxidizing conditions H_2O would have interacted with a melt more rapidly than CO, via evaporation/condensation [e.g. $\text{SiO}(\text{g}) + \text{H}_2\text{O}(\text{g}) = \text{SiO}_2(\text{l}) + \text{H}_2$] and gas-melt exchange. Also, CO- H_2O isotopic exchange would have been rapid compared to gas-melt exchange.

If the initial dust/melt composition was $\delta^{17,18}\text{O} \approx -50\%$, the initial gas composition must have been above the terrestrial mass fractionation line. It requires that gas-dust equilibration in the Earth-Moon, Mars, CI-, and E-chondrite formation regions involved remarkably similar dust/gas ratios despite their very different locations and bulk chemistries. It also leaves the lack of mass independent fractionation in EC chondrules unexplained.

Alternatively, the gas and dust both had compositions near $\delta^{17,18}\text{O} \approx 0\%$, and the mass independent fractionation occurred in the gas during chondrule/CAI formation. To explain the generally lighter O isotopic compositions of the more refractory chondrules/CAIs, isotopically light H_2O ($\delta^{17,18}\text{O} < -50\%$) must be generated near peak temperatures, but H_2O become heavier with time/cooling. Of the 4 reactions not involving ozone known to produce mass independent fractionation (Thiemens, 1999), only $\text{OH} + \text{CO} = \text{CO}_2 + \text{H}$ has been modelled to date. This reaction preferentially consumes C^{16}O . To produce $\delta^{17,18}\text{O} \approx -50\%$ H_2O , requires that C^{16}O react $\sim 25\%$ faster than the other isotopomers. The reaction would become less important under reducing conditions as the OH and H_2O would be converted to CO. However, the overall reaction rate does not exhibit the required temperature sensitivity. It remains to be seen whether any of the other reactions have the appropriate properties to make this a viable explanation.

References

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Study of diatoms/aqueous solution interface: I. Testing a surface complexation approach

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This work reports on a concerted study of diatom trace metals-water interfaces with four different species: two marine planktonic species (*Thalassiosira weissflogii*, *Skeletonema costatum*) and two freshwater periphytic species (*Achnantes minutissima*, *Navicula minima*). Cell surface charge was measured by potentiometric titration using a limited residence time reactor. The zeta potential of living cells was determined in a wide range of solution composition (pH, ionic strength, dissolved organic matter and metals concentrations). Information on the chemical composition and molecular structure of diatoms surfaces was obtained using FT-IR (Diffuse Reflectance and Attenuated Total Reflectance) and X-ray Photoelectron Spectroscopy (XPS). The surface area of living cells in aqueous solutions was quantified using Small Angle X-ray Scattering Spectroscopy (SAXS).

These observations allowed the identification of the major specific surface functional groups. Taking into account the high specific surface area of viable cells as measured by SAXS (on the order of $10^{-7} \text{ m}^2/\text{cell}$) and relative proportion of carboxylic, amine and silanol surface sites of 1:1:0.02 as inferred from XPS and FT-IR quantification of *Achnantes Minutissima* cells wall composition, a surface complexation model (SCM) was constructed that allows a rigorous thermodynamic description of diatom-solution interfaces. A good agreement between the experimental data and surface complexation modeling demonstrates the validity of this thermodynamic approach.

Preliminary experiments on metal adsorption (Cd, Pb, U, and Al) on two marine and two freshwater diatom species evidenced very fast and reversible processes which can be modeled within the framework of SCMs recently developed for complex multioxide minerals and carbonates (Pokrovsky & Schott, 2002) and bacterial surfaces (Fein et al., 1997).