



## **Non-mass dependent oxygen isotopic composition of refractory oxide dust produced by a gas phase chemical process: Implications for the evolution of the solar nebula**

S. Chakraborty (1), **M. H. Thiemens** (1), Y. Kimura (2) and J. A. Nuth III (2)

(1) Department of Chemistry and Biochemistry, University of California San Diego, La Jolla, CA 92093-0356, USA, (2) Code 691, Astrochemistry Laboratory, NASA's Goddard Space Flight Center, Greenbelt, MD 20771, USA (subrata@chem.ucsd.edu / Fax: +1 858 534 7042 / Phone: +1 858 534 6053)

Meteorites (early solar system objects) possess a wide range of oxygen isotopic compositions, reflecting the existence of heterogeneity in the solar nebula. Along with the spread in composition, a striking feature is the departure from the normal mass-fractionation line (represented by a slope  $1/2$  line in a 3-isotope plot of oxygen).

Recently, through a detailed kinetic model, photochemical self-shielding of nebular CO (the most abundant oxygen carrying molecule in a nebular environment), was proposed to be the origin of anomalous oxygen in the early solar nebula. Though the self-shielding model calculates the water-ice isotopic composition, our recent experimental observations demonstrate that photochemical processes involving electronically excited CO are likely to be an important contributor to early solar system processes as well.

Apart from photochemistry, the actual process of gas to solid conversion, required for production of planetary materials, is a process will also mediate final isotopic composition. It was demonstrated in previous laboratory experiments and quantum mechanically based model calculations that the symmetry governed gas-phase recombination reactions (such as,  $O + O_2 + M \rightarrow O_3 + M$  and  $O + CO$ ) are the predominant source of mass-independent compositions. Marcus (2004) proposed a nebular symmetry dependent process, including a surface phase and an entropic enrichment factor on grain

surfaces to produce a  $^{17}\text{O}$  and  $^{18}\text{O}$  depleted oxygen reservoir followed by reaction of the adsorbed metallic oxides. The effect arises due to a symmetry dependent stabilization of the metal dioxide and thus is a rather general process leading to production of calcium-aluminum solids as suggested previously by Thiemens. The present work is the first report of laboratory experiments leading to solid production.

We report here the first replicable production of non-mass dependently fractionated iron and silicate condensates. We produced these materials in the Smoke Generator at GSFC from a stream of silane and pentacarbonyl iron in a molecular hydrogen flow mixed with molecular oxygen or nitrous oxide. The smokes were formed at total pressures less than about 100 Torr in an electrical discharge powered by a Tesla coil. The smokes themselves were collected from the surfaces of the electrodes (though samples from other locations were occasionally analyzed) after each experiment and analyzed for  $^{17,18}\text{O}$  by laser fluorination.

It is interesting to see that none of the products (with respect to the initial composition) define a trend line not passing through the initial oxygen reservoir composition. The product SiO data defines a slope and intercept of  $0.68 \pm 0.03$  and  $2.52 \pm 0.29$  respectively and is interpreted as involving two fractionation steps. First, the thermolysis, breakdown and oxidation of the silicon to form SiO in a *mass dependent* manner and, secondly, a symmetry dependent reaction (e.g.  $\text{O} + \text{SiO}$ ) resulting in a *non-mass dependent* fractionation. More details will be presented during the conference.