Goldschmidt 2021 Abstract
https://doi.org/10.7185/gold2021.5364

A POSSIBLE ORIGIN FOR THE MASS-INDEPENDENT ISOTOPIC FRACTIONATION: IMPLICATION FOR COSMOCHEMISTRY

FRANÇOIS ROBERT

CNRS/Museum National Histoire Naturelle

Presenting Author: francois.robert@mnhn.fr

Using the experimental results obtained for oxygen isotopes during the synthesis of ozone[1], we have proposed a theoretical interpretation of the mass independent isotopic fractionation effect[2] (MIF). I will review its consequences for laboratory experiments.

Theoretical. A and B, 2 isotopes of the same element and M a chemical bond. The atom A resulting from photodissociation of MA reacts with its parent molecule (A+MA or A+MB). An isotopic exchange takes place during this reaction as A+MB®[AMB]*®B+MA. If a fraction of the the activated complex [AMB]* formed by this exchange reaction is involved in a chemical reaction, the chemical products should exhibit the MIF effect. This effect is caused by the fact that it is not possible to mathematically separate the exchange and the non-exchange processes when the complex is formed by the same isotopes (so called indistinguishable as [AMA]*).

Experimental[3]: The vapor pressure of liquids ($TiCl_4$ or $MgCl_2$ in hydrocarbons) carried by N_2 or N_2O or CO_2 gas flow at ≈1 mbar, are submitted to a microwave discharge. Grains condensed in the so-formed plasma are deposited on silicon wafers (Æ » 1 mm) on top of organic films (thickness 10-100 nm).

Analytical: Isotopic compositions were measured using NanoSIMS. In grains, the variations in Ti, Mg or O exceed 1000‰ (expressed in the usual d units per mil). As suggested by Marcus[4], the surface of a growing grain acts as a concentrator and enhances the rate of isotopic exchange compared to gas phase reactions. A similar effect may have occurred in the protosolar nebula during high temperature oxidation reactions of atoms.

References: [1] Thiemens M.H. & Heidenreich J.E., (1983) *Science*, *219*, 1073-1075. [2] Reinhardt P. and Robert F. (2018) *J. Chem. Phys.* **513**, 287–294. [3]. Robert et al., *Nat. Astron*. (2020), 1-7 DOI: org/10.1038/s41550-020-1043-1.[4] Marcus R.A., (2004) *J. Chem. Phys. 121*, 8201-8211.