In search of the mass-independent isotope fractionation effect: the role of plasmas.

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The first doubts on the general character of the mass dependent isotopic fractionation theory (MDF) appeared in 1983 with the synthesis of ozone(1). Oxygen isotopes are often used to illustrate the MDF: a relative variation of 1% in the 17O/16O isotopic ratio must be accompanied by 2% in the 18O/16O because the mass difference between 18 and 16 is twice that between 17 and 16. The origin of the MDF is rooted in the most fundamental principles of quantum mechanics. It is ubiquitous, observed for isotope exchange reactions and for 2-body chemical reactions. It was theorized in 1947 by H. Urey(2).

On the contrary, ozone results from a 3 body reaction (O+O2+ M → O3 + M where M is the third body stabilizing the complex O3\*) and shows equal relative variations in 17O/16O and 18O/16O (referred to as the *MIF effect*). This result reproduces the variations observed in meteorites(3) and more generally in the whole solar system. Just as the physical origin of the MIF effect is still an open question in quantum mechanics, its possible application to the formation of the solar system is difficult to judge.

The seminar contains 3 parts: (1) theoretical, (2) experimental and (3) analytical. In part (1), I will show that the results on ozone can be reproduced if the fundamental quantum mechanical requirement that, for indistinguishable isotopes, the two possible reaction pathways (elastic scattering and particle exchange i.e. isotopic exchange and non-exchange) are *superimposed*(4,5). In part (2) I will discuss how the equations that govern the effect in ozone led us to consider condensation experiments in plasma. The part (3) is dedicated to the results obtained for Ti, Mg, O and N isotopes using mixtures of TiCl4+Pentane, MgCl3+Pentanol and CO2 or N2O+Pentane(6,7). The spatial resolution of the NanoSims revealed considerable isotopic variations for all these elements in micrometric size carbonaceous particles condensed in plasma (in the range 1000-10,000 ‰). These results are reasonably well accounted by the model built for ozone. As proposed by(1), they imply that the MIF effects becomes serious candidates to explain the origin of solids in the solar system. They nevertheless need to be documented experimentally under physical and chemical conditions similar to those modeled in cosmochemistry.

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