MASS-INDEPENDENT ISOTOPE FRACTIONATION IN PLASMA:
THEORY AND EXPERIMENTS.

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Introduction: Although the mass dependent isotopic fractionation theory (MDF) is ubiquitously observed in 2-body chemical reactions, the first doubts on its general character appeared in 1983 with the synthesis of ozone [1]. Ozone results from a 3 body reaction (O+O₂+ X → O₃ + X where X is the third body stabilizing the complex O₃*). During its synthesis, equal relative variations in \(^{17}\)O/\(^{16}\)O and \(^{18}\)O/\(^{16}\)O are observed, in clear contradiction with MDF predictions but mimic those observed in meteorites and in solar system planets [2]. 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 seems difficult to judge.

Theoretical proposal: According to a recent proposal [3], the lifetime ratio of two complexes is different from unity if their formations involve identical or non-identical isotopes. The MIF effect would result from the quantum mechanical requirement according to which, for identical isotopes, the two possible reaction channels (elastic scattering and particle exchange) have to be superposed (such as, for example: \(^{16}\)O + \(^{16}\)O \rightarrow \(^{16}\)O\(^{16}\)O*). Numerical calculations are in reasonable agreement with laboratory isotopic determinations. Beside ozone the theory could be extended [3] to all chemical elements involved in reactions provided the activated complex resulting from isotope recombination is retrieved by a chemical reaction faster than its spontaneous dissociation.

Experimental: In microwave plasma, the molecules are dissociated by electron impact producing highly reactive –CH radicals ultimately forming solid grains. The isotopic composition at the surface of these 1-3 \(\mu\)m grains was analyzed with the NanoSims. We have used methane, pentane or pentanol to generate these carbonaceous plasmas while Ti, Mg (or O) atoms are carried by chlorides or CO₂ (for oxygen).

Results: Large (up to 1000 ‰) MIF effects are observed. Examples are reported in the Figures 1 and 2. These results are reasonably well accounted for by the model developed for ozone.