Mass-independent isotope fractionation: a general behavior of elements in plasma chemical reaction ?

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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 $(O+O_2+ X \otimes O_3 + X \text{ where } X \text{ is the third body stabilizing the complex O_3*})$. During its synthesis, equal relative variations in ¹⁷O/¹⁶O and ¹⁸O/¹⁶O are observed, in clear contradiction with MDF predictions but mimic those observed in meteorites [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.

We have attempted an approach that is at the same time theoretical, experimental and analytical [3,4,5,6]. The MIF effect can be accounted for if the quantum mechanical requirement for indistinguishable isotopes are introduced in the theory [4]. If correct, this mechanism should not be limited to ozone nor to oxygen isotopes but to reactions where the intermediate activated complex is involved in two reactional channels. To illustrate this proposal, we have performed chemical reactions in non thermal plasma. We will show results obtained for several isotopic systems (O, Mg, Ti, etc.) using mixture of TiCl₄+Pentane, MgCl₃+Pentanol and CO₂ or N₂O+Pentane [5,6]. Large (in the 1000 ‰ range) MIF effects are observed in micrometric size carbonaceous particles deposited in the plasma during the polymerization of hydrocarbon radicals. These results are reasonably well accounted by the model built for ozone. As proposed by [1], they imply that the MIF effect becomes a serious candidate to explain several isotopic anomalies observed in meteorites.

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