

The role of plasma in the mass independent isotopic fractionation : theory and experiments

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In contrast to mass dependent isotopic fractionation (MDF) characterizing 2-body chemical reactions, 3-body reactions yield mass independent fractionation (referred to as Chem-MIF) [1]. This conclusion is based on the theoretical treatment of the Chem-MIF effect observed for oxygen isotopes in ozone [2] ($O+O_2+X \rightleftharpoons O_3+X$ where X is the third body stabilizing the complex O_3^*). This type of reactions is promoted in plasma where atoms react with the non-dissociated fraction of their parent molecule [3].

In the Chem-MIF effect, the lifetime ratio h 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: $^{16}O + ^{16}O^{16}O \rightleftharpoons ^{16}O^{16}O^{16}O^*$). Several parameters dictate the h value via the complex stabilization mechanism by the third body X , namely: (i) the interaction potential, (ii) the complex lifetime and (iii) its steric definition. The numerical sensitivity of h with respect to these three parameters will be presented.

The theory predicts the isotopic variations patterns for the different chemical elements. Experiments performed with Mg and Ti chlorides in carbonaceous plasma were in agreement with these theoretical expectations [3]. These tests have been extended to the 7 isotopes of Ruthenium. However, contrary to the possible Chem-MIF occurrences in meteorite data, the mass dependent fractionation was negligible relative to h in these experiments. Therefore, experimental simulations in protosolar conditions seem unavoidable to compare the data on natural samples with the Chem-MIF model predictions [4].

References: [1] M.H. Thiemens, J.E. Heidenreich III, *Science* **219** 1073–1075, (1983). [2] F. Robert, P. Reinhardt. *Chem. Phys. Impact* **4**, 100073, [2022]. [3] F. Robert, M. Chaussidon, A. Gonzalez-Cano, S. Mostefaoui, PNAS **118** 52, (2021). [4] N. Asset et al., PNAS (2025, in press) ; cf. oral presentation at this conference.