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Nuclear field shift effect in chemical exchange reactions: a new interpretation of mass-independent isotope fractionation in the early Solar System

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Most of isotopic variability in the Solar System is accounted for by four types of processes: (i) mass-dependent thermodynamic fractionation, (ii) radioactive decay, (iii) spallation by cosmic rays, and (iv) incomplete mixing in the solar nebula. Here, we propose the adoption of "nuclear field shift effect" into the cosmochemistry, which shows quite good agreement with isotopic anomalies found in the primitive meteorites.

In 1996, the classic theory of stable isotope fractionation of the Bigeleisen-Mayer equation [1] has been expanded by the original author to include the mass-independent term named the nuclear field shift effect [2]. The nuclear charge distribution is attributable to the nuclear shell structure, and hence, it possesses mass-independent property. This causes an isotope shift in the atomic and molecular energy levels via the interaction between the nucleus and orbital electrons. The nuclear field shift is account for the origin of mass-independent isotope fractionation in chemical exchange reactions [2].

We demonstrated on several elements (Cr, Mo, Ru, Cd, and Te) the existence of the mass-independent isotope fractionation in laboratory experiments with a ligand exchange system or a redox system. The isotope ratios were analyzed by multiple-collector inductively coupled plasma mass spectrometry with a typical precision of $<$100 ppm. For each element, obtained isotope fractionation factors showed mass-independent property, which was explicable with the nuclear field shift theory [2]. These results suggest that the isotope fractionation chemically created in the nature would have mass-independent property due to their nuclear radii. We subsequently set out to compare the prediction of Bigeleisen's (1996) theory with the isotopic anomalies found in meteorites ("FUN inclusions" of Allende, carbonaceous chondrites, or ordinary chondrites). Some of these anomalies are clearly inconsistent with nucleosynthetic effects (either s- or r-processes), and for these, the nuclear field shift theory fitted extremely well. In meteorites, the nuclear field shift effect may originate both during condensation/evaporation processes in the nebular gas and during the metamorphism of the meteorite parent bodies. The isotope exchange reactions between chemical species which have been generated in the condensation/evaporation processes and metamorphism may have been affected by the nuclear field shift.

[1] Bigeleisen J. and Mayer M. G. 1947, *J. Chem. Phys*. 15, 261-267. [2] Bigeleisen J. 1996, *J. Am. Chem. Soc*. 118, 3676-3680.