NUCLEAR FIELD vs NUCLEOSYNTHETIC EFFECTS AS CAUSE OF ISOTOPIC ANOMALIES IN FUN INCLUSIONS

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Introduction: The anomalous abundances observed, notably for Mg, Si, Ca, Ti, Cr, Sr, Ba, Nd, and Sm in the so-called FUN inclusions (see [1] for a review), epitomize the occurrence of isotopic anomalies in early nebular condensates. Some isotopic anomalies cannot, however, be accounted for by nucleosynthetic processes [1,2] and we therefore explore the possibility that they may instead be due to mass-independent isotope fractionation induced by nuclear field shift. We will discuss the extreme conditions of the solar nebula in which these fractionation effects are the most likely to be operative.

Results and discussion: The theory of first-order mass-dependent fractionation has recently been revised to include the so-called nuclear field shift effect [3]. The spatial distribution of protons in the nucleus, which impacts the charge distribution interacting with electronic shells, obeys symmetry requirements that in turn cause the nuclear charge radii to vary unevenly with the number of neutrons characterizing the different isotopes of a same element [4]. The ensuing shift of the nuclear field imparts a mass-independent character to the electric field around the nucleus of the different isotopomers [3] and therefore to isotopic fractionation among coexisting species. A number of experiments, mostly involving solvent extraction and liquid chromatography, confirmed the existence of mass-independent fractionation as predicted by the nuclear field shift effect [5].

Striking isotopic anomalies are present in the alkaline-earth elements of FUN inclusions. The case for Ba in FUN inclusions, notably in the EK-1-4-1 and C-1 inclusions is particularly illuminating. While EK-1-4-1 is unique in its isotopic properties, a number of inclusions are closely related to C-1 in their isotopic compositions [6]. The mass-independent isotope effects are calculated in ε units using the equation 1 (see [7] for derivation) :

$$\varepsilon_{m_i} = \left(\delta\langle r^2\rangle_{m_1,m_i} - \frac{m_2(m_i - m_1)}{m_i(m_2 - m_1)}\delta\langle r^2\rangle_{m_1,m_2} \right)\times a \qquad \mathbf{(1)}$$

in which m_i stands for the atomic mass of a nuclide indexed with the variable i and a is an adjustable parameter representing the overall extent of mass-independent fractionation. This equation will be used to test for a possible effect of the nuclear field shift in FUN inclusions.

As in the original literature, the measured Ba isotope compositions are first normalized to a reference value for ^{134}Ba/^{138}Ba. The ε values plotted as a function of mass number show that the abundances of the other isotopes corrected for mass-fractionation in EK-1-4-1 and C-1 clearly are anomalous (Figure 1a and 1b). In contrast, the isotopic variation patterns closely follow the variation of nuclear charge radius. Both enrichment (EK-1-4-1) and depletion (C-1) of odd atomic-mass isotopes with respect to the even atomic-mass isotopes are observed. Furthermore, this isotopic pattern reproduces the mass-independent fractionation effects consistently observed in chemical fractionation experiments on Ba [8]. Overall, the calculations reproduce the anomalous isotopic variations well, indicating that the nuclear field shift effect accounts adequately for the anomalous isotopic variations of Ba observed in FUN inclusions.

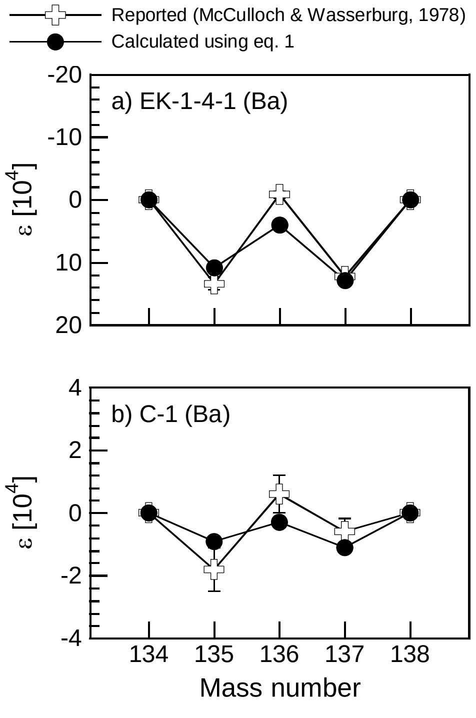

Figure 1 *Isotopic anomalies of Ba. Open symbols are for literature data [8] (errors are 2σ uncertainties) and closed symbols for the data calculated using Eq. (1). The isotope pairs used for normalization are 134 and 138.*

Again, the pattern of isotopic variations of Ca, especially for C-1, is similar to that of <r^2>. A large excess (EK-1-4-1) and a large deficit (C-1) are seen for ^{48}Ca. In spite of a small discrepancy for ^{42}Ca, which possibly represents a true nucleosynthetic anomaly, the calculation reproduces, as for Ba, the isotopic anomalies well. Most experimental work on Ca isotope fractionation during chemical exchange unfortunately only report results on ^{40}Ca, ^{44}Ca, and ^{48}Ca. In some systems, however, isotope separation factors of the ^{40}Ca-^{44}Ca and ^{40}Ca-^{48}Ca pairs show a breakdown of the mass-dependant law ($\propto \delta m/mm'$), which may also be assigned to the nuclear field shift effect. These experimental observations suggest that the anomalous isotopic variations of ^{48}Ca found in FUN inclusions may result from mass-independent chemical isotope fractionation.

A similar dependence on the nuclear field shift effect of Sr isotope anomalies found in FUN inclusions is expected. Unfortunately, because of ^{87}Sr variations resulting from the radioactive decay of ^{87}Rb, the anomaly is only incontrovertible for ^{84}Sr. The predicted anomaly is present in both the C-1 and EK-1-4-1 inclusions, but also in the refractory inclusion USNM 1623-5 from the Vigarano carbonaceous chondrite, which is thought to be isotopically similar to C-1 [6]. Surprisingly, however, once the ^{87}Sr abundance in USNM 1623-5 has been corrected for radiogenic ingrowth from ^{87}Rb over 4.56 Ga, the anomaly (ε_{87}=5.8) is consistent with the effect predicted by the nuclear field shift theory. It nevertheless remains small with respect to the magnitude of radioactive effects.

Even Mg seems isotopically anomalous in EK-1-4-1 and C-1. The lack of correlation between the excesses of ^{26}Mg and the Al/Mg ratios in the different phases of the inclusions of C-1 and EK-1-4-1 demonstrates that the observed isotopic anomalies cannot be attributed to the decay of ^{26}Al, and thus the possibility of some unidentified nucleosynthetic effect on one or both of the other two isotopes of Mg (^{24}Mg and ^{25}Mg) has been considered. Since Mg has only three isotopes, evidence of the nuclear field shift effect can only be based on one nuclide Nonetheless, the mean-squared charge radii decrease in the order ^{24}Mg >^{26}Mg >^{25}Mg, Remarkably, a mass-independent isotope fractionation effect has also been observed in chemical extraction experiments [10]. The isotopic anomalies of Mg may therefore also be attributable to the nuclear field shift effect.

Mass-independent fractionation effects also have been observed in chemical exchange experiments on Ti and Cr that are successfully explained by nuclear field shift effects [5]. This work prompted us to review the isotopic evidence on FUN inclusions. The overall isotopic pattern of Ti is reproduced by the nuclear field shift theory. For Cr, the observed excesses of ^{53}Cr and ^{54}Cr are also well predicted by the $\delta\langle r^2\rangle$ dependence. In contrast, the isotopic pattern of Ti and Cr observed in C-1 are different from those of EK-1-4-1 and these patterns are poorly reproduced by the nuclear field shift effect. This suggests that the isotopic anomalies observed in C-1 are of either radioactive (^{53}Cr) or nucleosynthetic origin.

Likewise, the isotopic anomalies observed for Nd and Sm in FUN inclusions cannot be even approximately reproduced by the nuclear field shift theory. For these elements, $\langle r^2\rangle$ changes smoothly with the neutron number and the changes are minor. The mass-independent isotopic variations of Nd and Sm in FUN inclusions appear to be genuine nucleosynthetic anomalies related to a mixture of *s*- and *r*-processes, which probably reflects the very large neutron cross-sections of lanthanides.

Conclusion: As to where in the solar nebula and how these isotopic effects arose remain speculative at best, but their apparent restriction to refractory inclusions seems to at least point to very high-temperature processes, such as evaporation and condensation of silicate material in the inner nebular disk. These effects should be most visible in the relative stable isotope abundances of some elements. The recently described excesses of ^{54}Cr characteristic of different planetary objects [11] may be a prime example of mass-independent fractionation and may signal isotopic heterogeneities of the Solar Nebula related to variable proportions of volatile and refractory components.

The potential overlap of mass-independent effects with radiogenic ingrowth suggests that a re-evaluation of some chronological results may also be in order, notably for ^{87}Sr, ^{26}Mg, and ^{53}Cr. For instance, the time scales of planetary accretion inferred from ^{53}Cr and ^{26}Al are not fully consistent [12,13]. The small and outward increasing excess of ^{53}Cr in the Solar System (<0.5 ε) may also reflect mass-independent effect on the scale of the nebular disk.

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