A Mg SURVEY FOR MORE "FUN" ISOTOPIC ANOMALIES


The most spectacular example of isotopic anomaly is of the "FUN" type which is characterized by large mass fractionation coupled with correlated nuclear effects in many elements. These correlated nuclear effects may be compared with the characteristic signatures of nucleosynthetic sources and hopefully may be used to identify the origin of solar system matter. At present there exist only three FUN samples with a combined mass <1g [cf. review (1)]. A survey for more FUN samples is urgently needed. More FUN samples will represent more mixtures of distinct nucleosynthetic components and thus would facilitate the determination of the composition of these end components. The discovery of new more massive FUN samples will permit the analysis of more elements, especially those of low abundances. This will in turn yield more correlations, hence constraints, for the nucleosynthesis theory. Furthermore, with more FUN samples of the appropriate mineralogical composition, it may be possible to determine whether \(^{26}\)Al correlates with any of the nucleosynthetic components. If such a correlation exists then the nucleosynthetic anomalies probably were synthesized immediately before the solar system formation and the \(^{26}\)Al was most likely made in extra-solar sources. In such a case supernova trigger models would receive strong support. Finally, the discovery of more FUN samples may clarify the relationship between the fractionation and nucleosynthetic effects which is apparently a key to the origin of the isotopic anomalies.

Since only \(\sim 3\%\) of Allende inclusions are of the "FUN" type a FUN-seeking survey is a high risk, low yield, and long term project. Such a survey should also be economical in the amount of sample consumed and time required per analysis. We have started a survey combining optical examination and Mg isotopic analysis. This combination is capable of detecting all the existing FUN samples. The Mg analysis is rapid because crystals can be selected, characterized, and directly loaded for mass spectrometry without chemical treatment (2). It consumes \(\sim 1\) mg material and looks for rather large \((>1%/amu)\) fractionation which is a distinct signature of FUN samples.

With gracious help from the staff of the National Museum of Natural History, we have examined part of its collection of the Allende meteorite and selected those fragments containing large \((\sim 100\) mg) exposed inclusions. We then carefully examined them individually in our clean room with a binocular microscope and removed from each of them several \(\mu g\)-sized crystals or clumps. These were analyzed following the method described in (2). Initially we could not mass-produce V-shaped filaments and used flat filaments instead. These early analyses seemed to yield variable mass fractionation of up to \(\sim 1%/amu\). Thirteen inclusions were analyzed in this way and no fractionation \(\sim 2%/amu\) (as those in other FUN samples) was found (3). More recently, we have finished a V-filament jig, based on the Caltech design. The result is summarized in Fig. 1 which is a histogram for the mean...
Fig. 1. Histogram of $\Delta^{25}\text{Mg}/^{24}\text{Mg}$ for terrestrial spinel and Allende inclusions. Permil deviation relative to 0.12475 is plotted. In the upper panel, the $\Delta^{25}\text{Mg}$ of all normal runs (except the one at $-5.5^\circ/oo$) lie between $-4$ and $+10^\circ/oo$ with a mean of $-1.5^\circ/oo$ defining the likely range of instrumental fractionation. Ten Allende inclusions define a distribution overlapping with this range and thus have no resolvable intrinsic fractionation. NMNH3675A is the suspected FUN sample with a clearly distinct Mg fractionation $-9^\circ/oo$ favoring the heavy isotopes. A pure spinel sample of this inclusion has similar $\Delta^{25}\text{Mg}$ to that of the Ti-fassaites and melilites covered with alteration products and possibly terrestrial dirt. This result indicates that neither late alteration nor terrestrial contamination have introduced substantial amount of normal Mg.

$\Delta^{25}\text{Mg}/^{24}\text{Mg}$ for sets of 10 ratios. The only selection criterion for these sets was that the standard deviation of the set mean must be $\leq 1^\circ/oo$. The per mil deviation of $\Delta^{25}\text{Mg}/^{24}\text{Mg}$ ($\Delta^{25}\text{Mg}$) relative to the reference value 0.12475 (2) is plotted. Eight terrestrial Mg spinel runs were used to establish the distribution for the normal standard. All normal runs except one can be described by a distribution centered on $-1.5^\circ/oo$. The extremes are $\pm 2.5^\circ/oo$ which we consider to be the likely range for instrumental fractionation. The "fluky" run has a $\Delta^{25}\text{Mg}$ of $-5.5^\circ/oo$. Data for ten Allende inclusions range from $-5$ to $+4^\circ/oo$ and thus have no resolvable intrinsic fractionation. Five analyses of inclusion NMNH 3675A yielded a distribution ranging from 6 to $12^\circ/oo$, well displaced from that of the normal and remaining samples. Therefore it has an intrinsic mass fractionation of $-9^\circ/oo$ favoring the heavy isotopes.

NMNH 3675A is a type B coarse-grained inclusion exposed on
the surface of an Allende fragment. Only a small portion (≈50 mg) of the original inclusion remained on this fragment. Terrestrial contamination is evident as dirt particles and yellow stains are clearly visible. Some dirt was blown away with a compressed freon duster. The major minerals are Ti-fassaite and melilite, both with tiny (≤3 μm) spinel inclusions. This inclusion has been severely altered after its formation. White fine-grained powder rich in alkali and halogen elements, presumably secondary phases due to alteration, permeate the entire inclusion while clean crystals are extremely rare. The data are mostly from four 100 μm-sized samples: three pyroxene crystals and one melilite crystal with attached alteration products. To check if terrestrial contamination and/or late alteration has introduced substantial amounts of normal Mg, we removed some stained material (≈0.1 mg) and dissolved it in HF and HNO₃. The acid residue was examined with an SEM equipped with an X-ray energy dispersive analyzer and found to consist of pure spinel crystals. No other oxide grains were present. This spinel sample is presumably free from both terrestrial contamination and secondary alteration products. Its $\Delta^{26}$Mg lies between 7 and 11‰ indicating that neither processes have introduced large amounts of normal Mg. The deviation in $^{26}$Mg/$^{24}$Mg is nearly twice that of $^{25}$Mg/$^{24}$Mg confirming that it is a fractionation effect. The fractionation corrected $^{26}$Mg/$^{24}$Mg ratio ($\delta^{26}$Mg) is within $\pm$1‰ of the normal. Since dissolved Mg normal solutions have not been extensively studied in our mass spectrometer we cannot report the precise $\delta^{26}$Mg at present.

Judging from its Mg fractionation we suspect that NMNH 3675A is a FUN sample. Its fractionation is intermediate between the FUN sample EKL-4-1 ($\Delta^{25}$Mg ≈ 20‰) and normal. It does not have a large nuclear effect in $^{26}$Mg like that found in EKL-4-1 ($\delta^{26}$Mg ≈ -3.7‰). These two characteristics are similar to those of inclusions EGG-3 (4) and B29 (5), the two other suspects. Therefore, there may be a class of Allende inclusions of substantial Mg fractionation but without associated nucleosynthetic effects. Alternatively, these samples may be intermediate between the usual inclusions and FUN inclusions and have accordingly diminishing nuclear effects bordering on the detection limit. Hopefully, the continuing search for fractionated Mg samples will help resolving the relation between the fractionation in Mg and other elements as well as nucleosynthetic effects.

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