MORE ANOMALIES FROM THE ALLENDE METEORITE: SAMARIUM

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Abstract. The isotopic composition of Sm has been measured in the Allende inclusions C1 and EK1-4-1. These inclusions were previously shown to have isotopic anomalies for the elements O, Mg, Ca, Ba and Nd. Sample C1 has a normal Sm isotopic composition except for the "p-process" isotope ^{144}Sm, which has an excess of 15±5 parts in 10^4. Sample EK1-4-1 was found to have Sm with a highly aberrant composition, indicating excess at all unshielded isotopes and an excess at ^{144}Sm. These data confirm the previous evidence for an addition of "r-process" isotopes to this material and also indicate an addition to the "p-process" isotope ^{144}Sm. From the observations on C1 it is inferred that the "p-process" addition is decoupled from the "r-process" addition.

Introduction

Sm isotopic anomalies are reported in the two known FUN (Fractionation and Unknown Nuclear effects) inclusions EK1-4-1 and C1 from the Allende meteorite. Extensive data have been reported on these two Ca-Al-rich inclusions (type B) as they are the only known materials with definitive isotopic anomalies for several elements. Anomalies in Sr from the same inclusions are reported in a companion letter by *Papanastassiou and Wasserburg* [1978]. Previous work has shown inclusion C1 to contain anomalous O, Mg, Ca, Ba and Nd [*Clayton and Mayeda,* 1977; *Wasserburg, Lee and Papanastassiou,* 1977; *Lee, Papanastassiou and Wasserburg,* 1978; *McCulloch and Wasserburg,* 1978]. The results obtained on Ba for EK1-4-1 showed that the isotopic effects were compatible with the addition of matter only to the unshielded nuclei ^{135}Ba and ^{137}Ba. The results on Nd were also compatible with the addition of matter to the unshielded nuclei. However, as Nd has only one shielded isotope (^{142}Nd), a definitive interpretation was not possible. For this reason we pointed out [*McCulloch and Wasserburg,* 1978] that to demonstrate general excesses compatible with r-process addition, it will be necessary to obtain data on Sm and other refractory elements in this mass region with two shielded isotopes. In addition, it was pointed out that any reliable astrophysical model must await the experimental determination of the yields of exotic nuclei in the region of the rare earths where chemical fractionation will not obscure the relative contributions. Reports of Sm isotopic effects in EK1-4-1 were subsequently presented by *Lugmair, Marti and Scheinin* [1978] and by ourselves at the Ninth Lunar and Planetary Science Conference. Isotopic anomalies for Sr in EK1-4-1 and C1 were also reported by *Papanastassiou, Huneke, Esat and Wasserburg* [1978].

Results

The experimental procedures used for the separation and measurement of Sm are based on the procedures developed in this laboratory by *Russ, Burnett, Lingenfelter and Wasserburg* [1971]. All samples were analyzed using our standard Sm chemistry apart from EK1-4-1 PYX and C1 PYX(2), which were purified using an additional miniature column to remove Ba. The results are given in Table 1 as deviations from the normal ratios [*Russ et al.,* 1971] in parts in 10^4 after correction for isotopic fractionation by normalizing to ^{147}Sm/^{154}Sm, and using ^{154}Sm as the index isotope. The results of analyses of a terrestrial sample, a nephelinite from the Oslo Rift, Norway, are shown in Table 1 and are in excellent agreement with the high precision abundances reported by *Russ et al.,* [1971]. For the C1 and EK1-4-1 samples the mass fractionation correction applied to Sm was less than ~3‰ per mass unit and within the same range as applied to the terrestrial sample.

Three analyses of different samples from the inclusion EK1-4-1 are given in Table 1. These samples are a scoop of the crushed inclusion, a pyroxene and melilite mineral separate. All EK1-4-1 samples show large deviations from normal which exceed the uncertainties by over a factor of 10. The deviations for the different samples are identical within error with the exception of ^{144}Sm/^{154}Sm deviations for EK1-4-1 SC and EK1-4-1 PYX which are at the limit of 2σ uncertainty. For EK1-4-1 SC peaks at masses 151 and 153 of 10^{-13} amps were present. The relative intensity of these peaks was consistent with Eu which does not interfere with Sm. The larger uncertainties for the melilite analysis are a consequence of a factor of 10 smaller sample size (4 ng Sm). These variations are in agreement with a recent report by *Lugmair et al.* [1978].

From inclusion C1 two different pyroxene separates and a split of the crushed inclusion were analyzed. These analyses all show an enrichment in only ^{144}Sm/^{154}Sm of 15±5 parts in 10^4. All other isotopic ratios are identical within error to the terrestrial normal. The only exception is for C1 PYX(1), which shows possible deviation from normal equal to the 4σ uncertainty for ^{148}Sm/^{154}Sm and 3σ uncertainty for ^{150}Sm/^{154}Sm. This was not present in the higher precision analyses of the other pyroxene separate C1 PYX(2a) or in the analysis of the total inclusion C1S1 and is therefore probably an artifact. A second analysis was carried out on another aliquot of C1 PYX(2a) after repeating the Sm separation on the final column. The values of ^{144}Sm/^{154}Sm for repeat analyses of this sample (C1 PYX(2b)) are in good agreement within experimental uncertainty.

As ^{144}Sm is the only anomalous Sm isotope in C1, we investigated the possibility of an isobaric interference. For all runs the mass spectrum was scanned at high sensitivity between masses 130 and 180, and no interfering species were identified. In particular, no ^{142}Nd$^+$ or ^{146}Nd$^+$ ions were detected and for the analysis of sample C1 PYX(2a), we can place an upper limit to any possible interference to ^{144}Sm from ^{144}Nd of less than 3 parts in 10^4. However, for the other samples of C1 and EK1-4-1, the smaller Sm ion beam intensities only allowed an upper limit of ^{144}Nd interference to ^{144}Sm of less than approximately 10 parts in 10^4. To test whether ^{144}Nd interference could produce an effect of this magnitude, a sample consisting of approximately equal proportions of normal Sm and Nd was loaded on a filament and analyzed. At the Sm running temperature, an apparent excess of ^{144}Sm of 10 parts in 10^4 was measured in this sample with the ratios Nd/NdO = 10^{-4} and ^{144}Sm/^{144}NdO = 0.1. In the samples C1 and EK1-4-1 the Sm fraction analyzed is essentially free of NdO with the measured ratio ^{144}Sm/^{144}NdO$\geq$100. Thus, assuming the same Nd/NdO ratio in C1 and EK1-4-1, the maximum ^{144}Nd interference to ^{144}Sm in C1 and EK1-4-1 is 1 part in 10^6. This is a factor of 10^3 smaller than the observed excesses and we conclude that interference from ^{144}Nd cannot account for the observed ^{144}Sm excesses.

Discussion

To elucidate the effects in Sm it is more appropriate to use the two shielded isotopes ^{150}Sm and ^{148}Sm to correct for fractionation with ^{148}Sm as an index isotope. The weighted average of data renormalized from the raw measured ratios are shown for EK1-4-1, C1 and the terrestrial sample in the bottom of Table 1 and is consistent with data transformed using equation 2 of *McCulloch and Wasserburg* [1978].

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Table 1. Samarium Isotopic Variations[a]

Sample[b]	Sm(ppm)	ε144	ε148	ε149	ε150	ε152
		Normalized to $^{147}Sm/^{154}Sm$, Index Isotope ^{154}Sm				
EK1-4-1 SC	4.1	−17.0±5.2	−36.2±2.2	−0.7±1.5	−35.1±2.2	−9.1±1.4
EK1-4-1 PYX	5.3	−5.2±5.9	−38.2±2.6	+0.2±2.1	−33.9±3.1	−9.4±1.7
EK1-4-1 MEL	1.4	−7.4±13.0	−37.0±4.9	+0.2±4.1	−31.4±7.7	−6.5±3.7
C1 S1		+15.5±5.9	−0.4±1.8	−0.2±1.5	−1.2±1.5	−1.7±1.2
C1 PYX (1)	4.9	+11.8±4.0	−4.2±1.8	−0.8±1.8	−3.1±2.2	+0.2±0.6
C1 PYX (2a)	4.4	+17.0±3.0	−0.6±1.0	−0.5±0.7	−1.2±1.2	−0.6±0.5
C1 PYX (2b)		+17.8±3.0	+0.2±1.2	−1.0±1.3	−1.2±1.5	−0.3±0.8
Terr. Sample	20.0	0.0±3.7	−1.0±1.8	−0.3±1.3	−0.3±2.1	+0.6±0.8
		+1.5±3.0	−0.4±0.6	+0.8±1.0	−1.8±2.2	+0.3±0.5
		+0.7±1.5	−0.6±0.8	+0.3±1.3	−0.3±1.5	+0.7±0.7
		Normalized to $^{150}Sm/^{148}Sm$, Index Isotope ^{148}Sm				
		ε144	ε147	ε149	ε152	ε154
EK1-4-1		+34.4±8.4	+38.5±3.3	+36.5±2.1	+24.1±4.0	+34.3±4.2
C1		+15.4±5.0	+0.4±1.6	+1.3±1.0	+2.0±2.1	+2.0±3.9
Terr. Sample		+1.6±4.0	+0.4±1.6	+0.1±1.6	−0.8±3.3	−1.4±4.7

[a]Errors are 2σ mean. Deviations in parts per 10^4 relative to the grand mean of terrestrial normals [*Russ et al.*, 1971]; $^{144}Sm/^{154}Sm = 0.13516\pm1$; $^{148}Sm/^{154}Sm = 0.49419\pm2$; $^{149}Sm/^{154}Sm = 0.60750\pm2$; $^{150}Sm/^{154}Sm = 0.32440\pm2$; $^{152}Sm/^{154}Sm = 1.17537\pm4$; $^{147}Sm/^{154}Sm = 0.65918$.
[b]PYX = pyroxene; MEL = melilite; S1, SC = splits of crushed inclusion; Terr. = Terrestrial.

For inclusion EK1-4-1, the renormalized data show positive anomalies at the unshielded isotopes ^{154}Sm, ^{152}Sm, ^{149}Sm, ^{147}Sm and at the low abundance shielded isotope ^{144}Sm. The addition of material to the unshielded Sm isotopes is fully consistent with the additions found in the unshielded Ba and Nd isotopes [*McCulloch and Wasserburg*, 1978].

The melilite and pyroxene mineral phases from EK1-4-1 have an identical enrichment although these phases contain different proportions of Sr, Ba, Nd, and Sm. This is not consistent with *in situ* fission subsequent to crystallization but implies that these minerals formed from an isotopically homogeneous liquid or gas phase or were subsequently isotopically reequilibrated. Xe data from EK1-4-1 [*Papanastassiou et al.*, 1978] are also not consistent with an origin of these anomalies by *in situ* fission. The Xe data are consistent with mixtures of ^{244}Pu fission Xe, air Xe, Xe from cosmic-ray-induced reactions on Ba, REE and I, and Xe from ^{129}I decay. There is no evidence for isotopic enrichments from any other sources (e.g., Xe as found in bulk acid-insoluble residues; *Lewis, Srinivasan and Anders*, 1975).

The number of excess atoms in EK1-4-1 can be calculated by assuming that the isotopic effects are due to addition of nuclides to average solar system material with cosmic abundances. This is given by $\epsilon(A)N(A) \times 10^{-4}$ where $\epsilon(A)$ are the measured deviations given in Table 1 and N(A) is the cosmic abundance [*Cameron*, 1973]. These excess atoms are tabulated in Table 2 and plotted in Figure 1. For Ba and Nd the normalizations are as described by *McCulloch and Wasserburg* [1978] with $^{134}Ba/^{138}Ba$ and $^{144}Nd/^{142}Nd$ used for normalization and ^{138}Ba and ^{142}Nd as index isotopes. For Nd the arbitrary assignment of an excess to ^{144}Nd of +20 has been adjusted to +18 to produce a smooth curve between excess atoms of ^{150}Nd and ^{152}Sm. An alternative normalization for Nd using $^{148}Nd/^{150}Nd$ has been proposed by *Clayton* [1978] but this has the disadvantage of assuming a normal cosmic ratio for the $^{148}Nd/^{150}Nd$ excesses. The main feature of the curve in Figure 1 is the pronounced peak between Ba and Nd. Chemical fractionation between the exotic rare earth and Ba nuclei or the addition of unfractionated exotic nuclei to fractionated "solar" material prior to the formation of the inclusion would modify this peak. A small odd-even effect is also apparent in Nd and Sm, with the excess atoms being greater in the even atomic number nuclei. This effect is not present in fission yield curves [*Meek and Rider*, 1974].

The additions to the unshielded isotopes can be attributed to beta decay from unstable neutron-rich species. Isotopes

Table 2. Nucleosynthetic Components in Ba, Nd and Sm for EK1-4-1

Nuclide	N(A)[a]	σ(A)[b]	$N_s(A)$[c]	$N_r(A)$[d]	ε(A)[e]	ε(A)x[f] N(A)10^{-4}	ε(A)N(A)[g] / ε(154)N_r(A)
^{130}Ba	0.005[m]	2000[k]	0.0	0.0	0.0	0.0	—
^{132}Ba	0.005[m]	650[k]	0.0	0.0	0.0	0.0	—
^{134}Ba	0.116	225[h]	0.116	0.0	0.0	0.0	—
^{135}Ba	0.316	470[i]	0.055	0.261	13.4	4.23×10^{-4}	0.47
^{136}Ba	0.375	70[h]	0.375	0.0	0.0	0.0	—
^{137}Ba	0.543	72.6[i]	0.363	0.180	12.3	6.68×10^{-4}	1.08
^{138}Ba	3.44	5.7[h]	3.30	0.14	0.0	0.0	—
^{142}Nd	0.211	45[j]	0.211	0.0	0.0	0.0	—
^{143}Nd	0.095	332[i]	0.025	0.07	27.8[l]	2.64×10^{-4}	1.1
^{144}Nd	0.186	67[i]	0.116	0.07	18.0	3.35×10^{-4}	1.39
^{145}Nd	0.065	485[i]	0.016	0.049	24.9	1.61×10^{-4}	0.96
^{146}Nd	0.134	105[i]	0.069	0.065	12.0	1.61×10^{-4}	0.72
^{148}Nd	0.045	210[k]	0.0	0.045	33.6	1.50×10^{-4}	0.98
^{150}Nd	0.044	240[k]	0.0	0.044	33.6	1.47×10^{-4}	0.98
^{144}Sm	0.007[m]	120[k]	0.0	0.0	34.4	0.24×10^{-4}	—
^{147}Sm	0.035	1150[k]	0.006	0.029	38.5	1.34×10^{-4}	1.35
^{148}Sm	0.025	260[k]	0.025	0.0	0.0	0.0	—
^{149}Sm	0.031	1620[k]	0.004	0.027	36.5	1.14×10^{-4}	1.23
^{150}Sm	0.017	370[k]	0.017	0.0	0.0	0.0	—
^{152}Sm	0.060	450[k]	0.014	0.046	24.1	1.46×10^{-4}	0.92
^{154}Sm	0.051	380[k]	0.0	0.051	34.3	1.76×10^{-4}	1.0

[a]Cosmic abundance (per Si = 10^6), *Cameron* [1973]. [b]30 KeV cross sections (millibarns). [c]s-process abundance. [d]r-process abundance given by $N_r(A) = N(A) - N_s(A)$. [e]Deviations from normal in parts in 10^4 from Table 1 and *McCulloch and Wasserburg* [1978]. [f]Excess atoms (see fig. 1). [g]Ratio of excess atoms to cosmic r-process atoms, normalized to ^{154}Sm (see fig. 2). [h]*Musgrove, Boldman and Macklin* [1976]. [i]*Holmes, Woosley, Fowler and Zimmerman* [1976]. [j]*Conrad*, [1976]. [k]*Allen, Gibbons and Macklin* [1971]. [l]Corrected for ^{147}Sm decay for an age of 4.56×10^9 years. [m]p-process only nuclide.

produced by this general mechanism are often referred to as "r-process." To test whether the excess atoms shown in Figure 1 are produced by an average "cosmic" r-process, we have calculated the ratios of excess atoms in EK1-4-1 to cosmic r-process atoms, normalized to the r-process only isotope ^{154}Sm. This ratio is given by $\epsilon(A)N(A)/\epsilon(154)N_r(A)$ where $N_r(A)$ is the calculated cosmic r-process abundance. To obtain $N_r(A)$, the s-process component $N_s(A)$ is subtracted from the total isotopic abundance N(A). Thus $N_r(A) = N(A) - N_s(A)$. The isotopes ^{154}Sm, ^{150}Nd and ^{148}Nd are produced only by the r-process. However, several other isotopes also contain a significant s-process component. For these isotopes, $N_s(A)$ was calculated using the local equilibrium approximation, $N_s(A-1)\sigma(A-1) = N_s(A)\sigma(A)$ as modified by the general trend of the $N_s(A)\sigma(A)$ curve and closed neutron shell at ^{138}Ba (see for example *Seeger, Fowler and Clayton*, 1965). Improved cross section and abundance data were used (see Table 2) together with allowance for branching in the s-process chain at ^{134}Cs and ^{151}Sm [*Ward, Newman and Clayton*, 1976]. The uncertainty in calculating the r-process contribution depends on the accuracy of the calculated s-process and the relative proportions of s and r components. In figure 2 the number of excess atoms in EK1-4-1 relative to cosmic r-process are shown as a function of atomic number and are in general *similar* to the cosmic r-process distribution. However, significant deviations at ^{149}Sm, ^{147}Sm and ^{135}Ba show that the exotic material was not *identical* to the average solar system r-process, or that the additions were made to material which did not have the present average solar system isotopic composition.

Sample EK1-4-1 also shows an excess in the proton-rich or "p-process" isotope ^{144}Sm. No corresponding enrichment was found in the "p-process" isotopes ^{132}Ba and ^{130}Ba. *Woosley and Howard* [1978] have indicated that the production of these nuclei by photodisintegration reactions operating on r- and s-process seeds is temperature dependent. Using this model, it may be possible to overproduce ^{144}Sm relative to its cosmic abundance without corresponding additions to ^{132}Ba and ^{130}Ba. Sr data from EK1-4-1 [*Papanastassiou et al.*, 1978] show that the p-process isotope ^{84}Sr has a deficiency of -32 ± 2 parts in 10^4 when normalized to $^{86}Sr/^{88}Sr = 0.1194$. This is clearly not compatible with the excess in ^{144}Sm. However, as ^{87}Sr has a contribution from ^{87}Rb($\tau_{1/2} = 5.0 \times 10^{10}$ yr) decay the data for the three remaining Sr isotopes can also be interpreted as an excess in ^{86}Sr (s-process) or a deficiency in ^{88}Sr (s- and r-process). None of these possibilities are in consonance with the Ba, Nd and Sm results and data on p-process nuclides in EK1-4-1 between Sr and Ba will be required to resolve this apparent inconsistency.

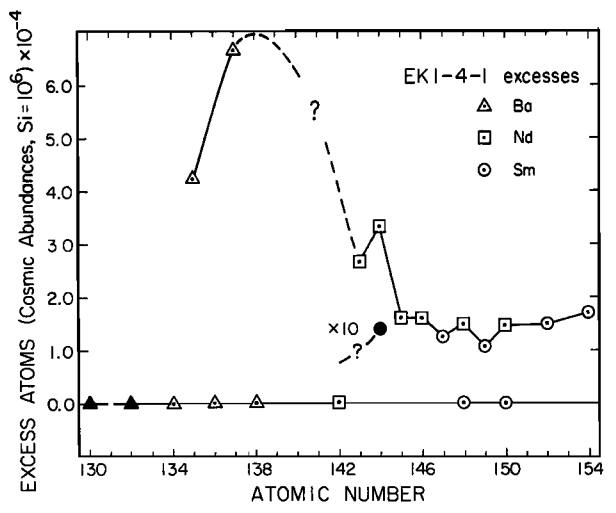

Fig. 1. Excess atoms of Ba, Nd and Sm in EK1-4-1 assuming addition of exotic nuclides to material with cosmic abundances. No excesses are assumed for the shielded nuclides ^{150}Sm, ^{148}Sm, ^{142}Nd and ^{134}Ba and for ^{138}Ba. Solid symbols show "p-process" only isotopes ^{130}Ba, ^{132}Ba and ^{144}Sm.

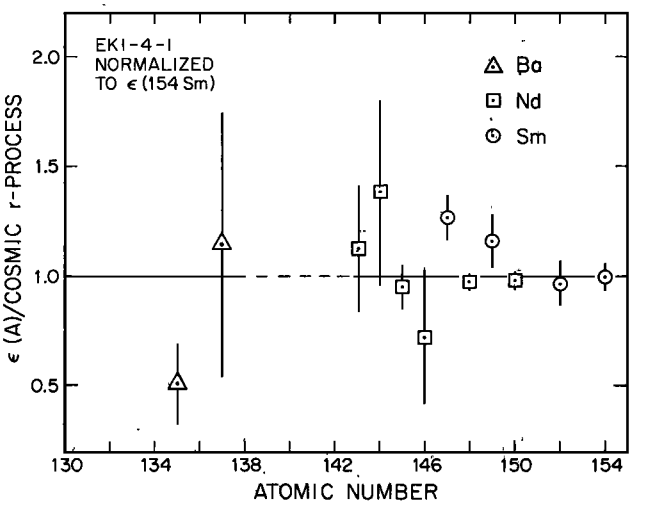

Fig. 2. Ratios of EK1-4-1 excess atoms to an average solar system r-process abundance as a function of atomic number (normalized to ^{154}Sm = 1.0).

In the other FUN sample, C1, an excess in only the "p-process" isotope ^{144}Sm of 1.0×10^{-5} atoms (per cosmic Si = 10^6 atoms) is present. All other isotopic ratios are normal. The presence of only a "p-process" addition to C1 is important as it shows that the "p-process" is not necessarily coupled to the r-process. This is in contrast to EK1-4-1, where both p- and r-process additions are present. The ^{144}Sm "p-process" excess in C1 is approximately a factor of 2 smaller than that in EK1-4-1. No corresponding addition has been detected in the "p-process" isotopes ^{132}Ba and ^{130}Ba in C1 [*McCulloch and Wasserburg*, 1978], although this may be close to the limit of resolution. The normal Nd isotopic composition in C1 [*McCulloch and Wasserburg*, 1978] is in consonance with these results as it has no "p-process" only isotopes. An addition to ^{142}Nd of the same number of atoms as excess ^{144}Sm* in C1 would only produce an effect in $^{142}Nd/^{144}Nd$ of 5 parts in 10^5 which is at the limit of our resolution. ^{142}Nd may also possibly have an indirect p-process contribution from decay by the extinct nuclide ^{146}Sm ($\tau_{1/2} = 1.0 \times 10^8$ yr). *Scheinin, Lugmair and Marti* [1976] and *Lugmair and Marti* [1977] have reported measurements of $^{142}Nd/^{144}Nd$ and from marginal data have inferred small excesses in ^{142}Nd attributed to decay of ^{146}Sm. Our Nd measurements in C1 show no evidence for the presence of ^{146}Sm, although the Sm/Nd ratio in this sample is not substantially fractionated from chondritic. A deficiency in the p-process isotope ^{84}Sr of -9 ± 2 parts in 10^4 is also present in C1, in contrast with the observed excess in ^{144}Sm. This indicates that simple addition of "p-process" material to normal solar system material is not an adequate explanation of the isotopic effects.

From the data reported here, as well as the wide range of Xe isotopic anomalies, it is reasonable to consider that the r-process isotope ^{129}I ($\tau_{1/2} = 1.6 \times 10^7$ yr) may also have been added in the same process. It was shown by *McCulloch and Wasserburg* [1978] that if the negative ^{135}Ba anomaly in C1 was representative of the initial solar system, then the additional ^{135}Ba* required to fill in the negative anomaly would give $^{135}Ba^*/^{127}I \sim 0.5 \times 10^{-4}$. Thus, if the same number of ^{129}I and ^{135}Ba* atoms were added, this would give $^{129}I/^{127}I \sim 10^{-4}$, which is similar to the observed ratio. It is also of interest to note that the addition of approximately the same number of ^{129}I atoms and exotic nuclei tabulated in table 2, would also give $^{129}I/^{127}I \sim 10^{-4}$. This model of small last-"minute" injection of r-process material is consistent with

the time scale of several million years as indicated by the presence of ^{26}Al with $\tau_{1/2} = 0.7 \times 10^6$ years [*Lee, Papanastassiou and Wasserburg*, 1977].

The discussion of Ba, Nd and Sm data for EK1-4-1 and the Sm data for C1 has been based on the premise of addition of exotic materials to material with a normal solar system isotopic composition. However, the presence of negative isotopic anomalies in other elements, ^{135}Ba and ^{48}Ca in C1 and ^{84}Sr and ^{26}Mg in EK1-4-1 and C1, suggests that this may not have been the case and materials which received these additions may have been depleted in some isotopes. This is particularly apparent in Sr [*Papanastassiou et al.*, 1978] which shows negative anomalies in ^{84}Sr/^{88}Sr for both EK1-4-1 and C1, when normalized to ^{86}Sr/^{88}Sr = 0.1194. As suggested by *McCulloch and Wasserburg* [1978], the negative ^{135}Ba anomaly may also imply that C1 represents precursor solar system material before r-process addition. However, the depletions of ^{135}Ba in C1 and the smaller ^{135}Ba excess relative to cosmic r-process in EK1-4-1 could also be due to ^{135}Cs, which is the progenitor for ^{135}Ba. If C1 and EK1-4-1 condensed before the more volatile ^{135}Cs ($\tau_{1/2} = 2.3 \times 10^6$ yr) decayed to ^{135}Ba, this could account for these discrepancies.

The extent and significance of the correlated isotopic anomalies which have at present only been found in the two Allende FUN inclusions, C1 and EK1-4-1, are still not apparent. An extensive survey of the Ba isotopic composition in a variety of Allende inclusions [*McCulloch and Wasserburg*, 1978] showed that 90% of these inclusions had a normal isotopic composition. Inclusions with a normal Ba isotopic composition included samples which had large ^{26}Mg excesses correlated with Al/Mg ratio, samples with a distinctive oxygen isotopic composition but lying on the mixing line of ^{16}O and normal O [*Clayton, Grossman and Mayeda*, 1973] and a sample with a suggested Hg isotopic anomaly [*Reed and Jovanovic*, 1969]. The Ba isotopic composition in meteorites [*Eugster, Tera and Wasserburg*, 1969; 1969; *de Laeter and Date*, 1973] and lunar samples [*McCulloch and Wasserburg*, 1978] has also been shown to be identical to terrestrial Ba. Thus, in contrast to the widespread O anomalies, only a small proportion of the materials sampled show correlated anomalies for other elements.

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