

CALCIUM ISOTOPIC ANOMALIES IN THE ALLENDE METEORITE

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ABSTRACT

We report isotopic anomalies in Ca which were found in two Ca-Al-rich inclusions of the Allende meteorite. These inclusions previously had been shown to contain special anomalies for Mg and O which were attributed to fractionation and unknown nuclear effects. The Ca data, when corrected for mass fractionation by using $^{40}\text{Ca}/^{44}\text{Ca}$ as a standard, show nonlinear isotopic effects in ^{48}Ca of +13.5 per mil and in ^{42}Ca of +1.7 per mil for one sample. The second sample shows a ^{48}Ca depletion of -2.9 per mil, but all other isotopes are normal. Samples with large excesses in ^{26}Mg show no Ca anomalies. The effects demonstrate that isotopic anomalies exist for higher-atomic-number refractory elements in solar-system materials and do not appear to be readily explainable by a simple model. The correlation of O, Mg, and Ca results on the same inclusions requires the addition and preservation in the solar system of components from diverse nucleosynthetic sources. Observed anomalous Mg and Ca compositions for coexisting mineral phases are uniform within each inclusion, and require initial isotopic homogeneity within an inclusion but the preservation of wide variations between inclusions. Assuming formation of these inclusions by condensation from a gaseous part of the solar nebula, this implies isotopic heterogeneity on a scale of $10\text{--}10^2$ km within the nebula.

Subject headings: meteors and meteorites — nucleosynthesis — stars: supernovae

I. INTRODUCTION

This *Letter* reports the discovery of isotopic anomalies in the element calcium in two peculiar inclusions of the Allende meteorite. Isotopic anomalies for Ba and Nd on the same samples are reported in a companion *Letter* by McCulloch and Wasserburg (1978). The two inclusions were previously found to contain distinct Mg nonlinear isotopic anomalies which could not be attributed to ^{26}Al decay (Wasserburg, Lee, and Papanastassiou 1977, hereafter WLP). This study is part of a continuing attempt to establish the range in atomic number over which isotopic shifts exist in solar-system materials in order to aid in identifying the nuclear and astrophysical processes which must be their ultimate cause. In addition to the well-recognized problems of Mg and of ^{26}Al , it was anticipated that, besides C and Si, the element Ca might be a good choice in the search for isotopic anomalies because its many stable isotopes (^{40}Ca , ^{42}Ca , ^{43}Ca , ^{44}Ca , ^{46}Ca , ^{48}Ca) are produced by distinct processes, although it is rather distant from oxygen and magnesium.

The samples investigated are Ca-Al-rich inclusions with the typical morphology and mineral phases which have been associated with early condensates from the solar nebula (Grossman 1972). Most inclusions so far studied show oxygen isotopic anomalies which may be explained as a mixture of extraordinary oxygen (O_E) consisting of essentially pure ^{16}O and "normal" solar-system oxygen (O_N) (Clayton, Grossman, and Mayeda 1973; Clayton *et al.* 1977), and exhibit "normal" magnesium with an excess of ^{26}Mg correlated with ^{27}Al (Lee, Papanastassiou, and Wasserburg 1976, hereafter LPW). The excess of ^{26}Mg ($^{26}\text{Mg}^*$) appears to

be due to the decay of ^{26}Al ($\tau_{1/2} = 7 \times 10^5$ years) in the solar system (Lee, Papanastassiou, and Wasserburg 1977*a*, hereafter LPW 1977*a*). The two inclusions which are found to show Ca anomalies were both previously found to contain isotopically anomalous oxygen and magnesium of a distinctive type (Clayton and Mayeda 1977; WLP). In contrast to the usual inclusions, the Mg in the two isotopically distinctive inclusions C1 and EK1-4-1 appears to be highly fractionated (F) and the $^{26}\text{Mg}/^{24}\text{Mg}$, when normalized for fractionation, exhibits nonlinear anomalies which correspond to deficiencies of ^{26}Mg or ^{24}Mg or excesses of ^{25}Mg due to unknown nuclear (UN) effects (Lee and Papanastassiou 1974). This type of anomaly has been designated FUN (WLP). The Mg fractionation and the negative $\delta^{26}\text{Mg}$ values in each of the two inclusions were found to be the same for different phases in the same inclusion but different between inclusions. Clayton and Mayeda (1977) found that spinel and pyroxene in the two inclusions with FUN anomalies lie off of the $O_E\text{--}O_N$ mixing line and appeared to suggest fractionation in a manner corresponding to the Mg isotopes. An indication of other isotopic anomalies was found by McCulloch and Wasserburg (1978), who showed a small depletion in ^{135}Ba in sample C1. We have investigated Ca (1) in inclusions which showed large $^{26}\text{Mg}^*$ correlated with ^{27}Al but normal $^{25}\text{Mg}/^{24}\text{Mg}$, and (2) in the two samples with FUN anomalies. Different mineral phases were analyzed to establish whether Ca is isotopically homogeneous within an inclusion.

II. RESULTS

The experimental procedures used for determining the isotopic ratios follow the high-precision mass spectro-

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metric procedures described by Russell *et al.* (1977) and Russell, Papanastassiou, and Tombrello (1977, hereafter RPT). A description of the techniques and procedures can be found in RPT. Data were obtained with a ^{40}Ca ion beam of typically $1.5\text{--}4 \times 10^{-10}$ A. Data were taken over a range of a factor of 3 in ion beam intensity, and the amplifier scales and resistors were varied for some of the ratios. The isotopic ratios for each sample were, to within experimental precision, constant throughout the experiment and independent of the above factors. The isotopic composition of Ca has been determined by RPT for many terrestrial materials, some lunar samples, and several bulk meteorite samples, and found to be the same and with no large mass fractionation (at most 0.7‰ μu^{-1}). We have used the isotopic abundances of normal Ca by RPT for reference values. Two terrestrial normals were analyzed during these experiments, and one enriched standard was prepared gravimetrically from normal Ca, to which ^{48}Ca was added to shift the abundance of only ^{48}Ca by 2.1‰ . Our data are reported as shifts in the isotopic abundance of each isotope relative to the RPT values in parts per thousand:

$$\delta i \equiv [(R_{i/44}^{\text{C}}/R_{i/44}^{\text{RPT}}) - 1] \times 10^3.$$

These shifts are calculated after correction (C) for instrumental and natural mass fractionation as determined from the ratio of $^{40}\text{Ca}/^{44}\text{Ca}$ by using the reference value of $^{40}\text{Ca}/^{44}\text{Ca} = 47.153$. Mass fractionation effects are corrected by using a power law; the total range in the raw measured $^{40}\text{Ca}/^{44}\text{Ca}$ was from 48 to 45. Calcium-44 was used as the index isotope. The values of $R_{i/44}^{\text{RPT}}$ are given in Table 1. Inspection of the data for the normals in Table 1 shows our results to be in good agreement with those of RPT for all isotopes and of comparable precision for individual runs. There are some shifts away from $\delta i = 0$ outside of the limits of error ($2 \sigma_{\text{mean}}$) but all well within $4 \sigma_{\text{mean}}$. Some of

these shifts may be due to the problems associated with normalization over a wide mass range as found by RPT. We consider as reliable those shifts which are substantially larger than the quoted uncertainties. The measured $\delta 48$ value of the 2.1‰ (gravimetric) enriched standard is $2.1\text{‰} \pm 0.2\text{‰}$ and demonstrates our ability to quantitatively determine shifts of this magnitude on an isotope with an abundance of 1.8×10^{-3} .

Two inclusions which contain the "usual" O and Mg anomalies were investigated. Analyses were done on inclusion WA, for which we had previously obtained a ^{26}Al - ^{26}Mg isochron on both macroscopic and microscopic samples (LPW; Lee, Papanastassiou, and Wasserburg 1977b, hereafter LPW 1977b). The anorthite showed an excess of ^{26}Mg of 100‰ and a $^{25}\text{Mg}/^{24}\text{Mg}$ close to normal. The raw $^{25}\text{Mg}/^{24}\text{Mg}$ in the pyroxene was also close to normal in composition. The normalized Ca data on the same coexisting phases are shown in Table 1 and are not distinguishable from the terrestrial standards for all isotopes. Sample BG2-6 has an excess of ^{26}Mg of 13‰ and no suggestion of other Mg anomalies for either spinel or grossular. This sample also gives no evidence of Ca isotopic effects, except possibly for a hint at ^{48}Ca .

Sample EK1-4-1 is one of the two FUN inclusions. Two analyses were carried out on separate dissolutions of the pyroxene sample previously analyzed for Mg. The results show a clear positive anomaly of about $+1.7\text{‰}$ for $\delta 42$ and a very large positive anomaly of $+13.5\text{‰}$ for $\delta 48$. To confirm this result, a new sample of EK1-4-1 was obtained through the cooperation and generosity of H. Nagasawa. This sample, consisting predominantly of pyroxene and some melilite and altered phases, was analyzed. The sample was first confirmed to show the same distinctive Mg isotopic composition as the previously studied mineral separate (T. Esat, personal communication). The results on this sample (SC) are in excellent agreement with the other

TABLE 1
CALCIUM ISOTOPIC EFFECTS*

Sample [†]	$\delta 42^{\#}$	$\delta 42^{**}$	$\delta 43$	$\delta 46$	$\delta 48$
EK1-4-1 PYX-A	$+1.5 \pm 0.3$	$+1.8 \pm 0.3$	$+0.9 \pm 0.5$	-13 ± 13	$+13.6 \pm 0.5$
PYX-B [‡]	$+2.0 \pm 0.2$	$+1.4 \pm 1.2$	$+0.5 \pm 1.7$	-13 ± 40	$+13.0 \pm 1.0$
SC-A	$+1.7 \pm 0.2$	$+1.6 \pm 0.6$	$+1.1 \pm 0.6$	-26 ± 13	$+13.3 \pm 0.6$
SC-B [§]	$+1.7 \pm 0.4$	$+1.5 \pm 0.4$	$+0.8 \pm 0.6$	—	$+13.5 \pm 0.4$
MEL	$+1.8 \pm 0.1$	$+1.5 \pm 0.1$	$+0.6 \pm 0.6$	-2 ± 12	$+13.4 \pm 0.6$
C1 S1	$+0.2 \pm 0.2$	-0.2 ± 0.3	-0.2 ± 0.5	-13 ± 7	-2.7 ± 0.2
PYX	-0.1 ± 0.2	-0.1 ± 0.3	0.0 ± 0.8	-26 ± 26	-2.9 ± 0.5
MEL	0.0 ± 0.2	$+0.1 \pm 0.4$	-0.5 ± 0.8	-7 ± 7	-3.4 ± 0.3
WA PYX	-0.2 ± 0.4	$+0.1 \pm 0.6$	$+0.5 \pm 0.6$	—	$+0.8 \pm 0.5$
AN	-0.1 ± 0.2	-0.2 ± 0.5	$+0.5 \pm 0.3$	0 ± 13	$+0.5 \pm 0.5$
BG2-6	0.0 ± 0.1	-0.2 ± 0.2	$+0.3 \pm 0.3$	-7 ± 20	$+1.0 \pm 0.5$
Normals CaF ₂	0.0 ± 0.1	-0.2 ± 0.3	-0.3 ± 0.3	-7 ± 20	-0.5 ± 0.2
CaCO ₃	-0.2 ± 0.2	0.0 ± 0.4	-0.5 ± 0.9	-7 ± 13	$+0.1 \pm 0.5$
$\delta 48 = 2.1\text{‰}$ ^{††}	-0.1 ± 0.1	-0.4 ± 0.4	$+0.3 \pm 0.5$	-20 ± 27	$+2.1 \pm 0.2$

*Deviations in parts per thousand in the Ca isotopic ratios after normalization of $^{40}\text{Ca}/^{44}\text{Ca}$ to remove natural and instrumental mass fractionation. Errors are $2\sigma_{\text{mean}}$. The normalization and deviations are given relative to the grand mean of RPT (1977): $^{40}\text{Ca}/^{44}\text{Ca} = 47.153 \pm 0.003$; $^{42}\text{Ca}/^{44}\text{Ca} = 0.31221 \pm 0.00002$; $^{43}\text{Ca}/^{44}\text{Ca} = 0.06485 \pm 0.00001$; $^{46}\text{Ca}/^{44}\text{Ca} = 0.00152 \pm 0.00001$; $^{48}\text{Ca}/^{44}\text{Ca} = 0.08870 \pm 0.00002$ (Note $\delta 40 = 0$). [†]PYX = pyroxene; MEL = melilite; AN = anorthite; SC, S1 = splits of crushed inclusion. [‡]Second dissolution of same sample. [§]Samples processed using distinct elution schemes. ^{||}Mg from same sample shows ^{26}Mg of 100‰ . [#]Measured on $10^{11}\Omega$. ^{**}Measured on $10^1\Omega$. ^{††}Enriched Standard in ^{48}Ca .

analyses. In addition, a sample highly enriched in melilite was also analyzed and yields results in agreement with all other EK1-4-1 analyses. The data on the melilite were obtained over a range of beam intensity from 0.5×10^{-10} to 7.5×10^{-10} A and yield consistent results independent of beam intensity. This indicates the absence of interfering species. The interference effects would be expected to vary significantly rather than to scale with the Ca beam intensity over a factor of 15 change in the Ca beam. The Ca data on EK1-4-1 clearly indicate uniform isotopic composition in coexisting mineral phases within this inclusion, analogous to the homogeneity observed for Mg (WLP).

The second FUN sample studied is C1. A solution of the bulk inclusion (S1) shows a deficiency of -2.7% for $\delta 48$ and no evidence of effects on the other isotopes. To establish whether this effect is real and whether it is uniform in the coexisting phases, we analyzed pyroxene and melilite, in which Ca is a major constituent and which differ in Ca content by a factor of 2.5. The results clearly confirm the existence of a deficiency at ^{48}Ca of -2.9% and show that, to within experimental errors, this deficiency is uniform between these phases. Data on both C1 and EK1-4-1 indicate anomalous but uniform Ca isotopic composition for coexisting phases in the individual inclusions.

In carrying out the experiments, we made a thorough search of the mass spectrum between mass 35 and mass 100. No possible interfering species were identified. K was absent during data acquisition. The analytical results were independent of the phases analyzed, some of which contain abundant Ti and in some of which Ti is absent, indicating that $^{49}\text{Ti}^+$ could not be the cause of the positive $\delta 48$ values.

To determine whether there is evidence of mass fractionation, a double spike highly enriched in ^{42}Ca and ^{48}Ca was added to aliquots of the Ca solutions of EK1-4-1, C1, and WA, which permits a reliable determination of $^{40}\text{Ca}/^{44}\text{Ca}$ in the sample relative to the standard normal (RPT). The results on EK1-4-1 show a fractionation of 1.8% per mass unit, favoring the lighter isotopes (Table 2). This is about a factor of 10 smaller than that observed for Mg in the same sample and opposite in sign. The fractionation determined for

EK1-4-1 is relatively large, being a factor of 2.6 greater than found by RPT in a wide variety of terrestrial and extraterrestrial samples. Sample C1 shows a much smaller Ca mass fractionation of 0.3% μu^{-1} . Sample WA shows a substantial fractionation of 0.7% μu^{-1} , favoring the lighter isotopes.

III. DISCUSSION

Isotopic analyses of Ca in Allende show that, for typical Ca-Al-rich inclusions, $\delta i \approx 0$ for all isotopes independent of the magnitude of $^{26}\text{Mg}^*$. For the unusual inclusions with FUN-type Mg and O anomalies, there exist well-defined nonlinear anomalies in Ca. The anomalies are the same for different phases within one inclusion. There does not appear to be a correlation between the magnitude of the nonlinear anomalies with the degree of fractionation in the FUN samples, but there is a qualitative correlation with $\delta^{26}\text{Mg}$. The observed values of $\delta 43$ and $\delta 46$ are zero within errors. It is surprising that major anomalies do not appear in the rare isotope ^{46}Ca , which is only 1/58 as abundant as ^{48}Ca .

These observations provide a plausible basis for assigning the nonlinear anomalies determined for $\delta 42$ and $\delta 48$ in the FUN samples to variations in the number of ^{42}Ca and ^{48}Ca nuclei and not to other isotopes. In this case, we conclude that there must exist both excesses and deficiencies in ^{48}Ca and excesses in ^{42}Ca . As yet, no reliable theoretical model for the formation of all the Ca isotopes is available to aid in decoding the observations. According to current models, ^{40}Ca and ^{42}Ca are made in explosive oxygen burning and ^{44}Ca in explosive Si burning (Woosley, Arnett, and Clayton 1973), while ^{43}Ca , ^{46}Ca , and ^{48}Ca can be produced by capture reactions on seed nuclei during explosive carbon burning (Howard *et al.* 1972). The Ca anomalies cannot be easily explained by these theoretical models. For example, both ^{42}Ca and ^{48}Ca show isotopic effects, while they are not thought to be made in the same process. The observations appear to require the mixing of distinctive nucleosynthetic sources. However, it is interesting to note that ^{48}Ca can be produced in the same process which may be responsible for both the $^{16}\text{O}_E$ and ^{26}Al .

TABLE 2
ISOTOPE EFFECTS (in per mil)

Sample	Fractionation* (% per amu)			$\text{O}_E\text{-O}_N$ Line	$^{26}\text{Mg}^*\ \S$	$\delta^{26}\text{Mg}^{\parallel}$	$\delta^{42}\text{Ca}^{\#}$
	Ca \dagger	Mg	O \ddagger				
C1	+0.3	+30	+15	off	?	-1.7	-2.9
EK1-4-1	-1.8	+20	+10	off	?	-3.4	+13.5
WA	-0.7	~ 0	0	on**	Yes	+100	(+0.5)
BG2-6	--	~ 0	--	--	Yes	+13	(+1.0)

*Fractionation factor per atomic mass unit difference. Positive sign corresponds to relative enrichment of the heavier isotopes. \dagger From doubly spiked Ca analyses. \ddagger Based on an end point of $\text{O}_E/\text{O}_N \sim 5 \times 10^{-2}$ and assuming that deviations off $\text{O}_E\text{-O}_N$ line are due to mass fractionation. \S Excess ^{26}Mg (in per mil) which correlates with ^{27}Al ; value is to be found in next column. \parallel Non-linear anomaly after normalization to $^{25}\text{Mg}/^{24}\text{Mg} = 0.12663$. $\#$ Non-linear anomaly after normalization to $^{40}\text{Ca}/^{44}\text{Ca} = 47.153$; values in parentheses are not resolvable effects. **S. Epstein, personal communication.

It should be noted that the choice of Ca normalization is arbitrary. If the two isotopes used for normalization are changed, then the isotopes which manifest nonlinear anomalies will change, as will the magnitude of the anomalies. Which of the isotopes have anomalies is not unequivocally established. Further, if the large "fractionation" determined on EK1-4-1 relative to "normal" Ca actually represents the addition of exotic nuclei to "normal" unfractionated Ca, then the composition of the exotic component will be distinct from that calculated from the nonlinear anomalies. The apparent fractionation determined from $^{40}\text{Ca}/^{44}\text{Ca}$ in EK1-4-1 is opposite in sign to that observed for Mg. This is surprising, as Ca and Mg have similar elemental properties. This observation may cast doubt on the validity of assigning the fractionation (F) type of behavior in Mg, O, and Ca to actual fractionation processes. This behavior may instead reflect major shifts in the isotopic abundance of these elements due to a special nucleosynthetic mixture (cf. WLP). It is possible that the apparent large (up to 60%) fractionation in Mg in FUN samples is due to incomplete mixing of ^{24}Mg , ^{25}Mg , and ^{26}Mg from the basic sources which provided solar-system Mg, and that the shift in the oxygen isotopic composition off of the O_E - O_N line found by Clayton and Mayeda (1977) may result from the existence of a third component with exotic $^{17}\text{O}/^{18}\text{O}$.

If we assume the normalization of $^{40}\text{Ca}/^{44}\text{Ca}$ used in Table 1 to be correct and consider the excesses of ^{48}Ca and ^{42}Ca in EK1-4-1 as due to their addition to "normal" Ca, this implies the existence of a nucleosynthetic component with composition of $^{48}\text{Ca}/^{42}\text{Ca} = 2.3$. It is plausible to consider "normal" Ca to be a mixture of Ca nuclei produced in different stars or within different zones in a supernova. If normal solar-system Ca is made up of only two components, this requires that one component have excesses in ^{42}Ca and ^{48}Ca and that the second component have a deficiency in ^{48}Ca at least as large as found in sample C1. A simple two-component model for both Ca and Mg with ^{26}Al as a contributor to ^{26}Mg cannot explain both the Mg and Ca isotopic variations unless there exists chemical fractionation between Ca and Mg between the two sources contributing to the formation of the chondrules. It is not known whether ^{26}Al or $^{26}\text{Mg}^*$ was present in samples with FUN anomalies. The Ca data demonstrate the presence of significant general isotopic anomalies. The data are not consistent with a special anomaly at ^{44}Ca due to ^{44}Ti decay ($\tau_{1/2} = 47$ yr) as predicted by Clayton (1975). The isotopic patterns of EK1-4-1 and C1 are not consistent with addition or depletion of only ^{44}Ca ; furthermore, the isotopic effects are uniform among distinct phases, within each inclusion, which have distinct Ca and Ti abundances and Ca/Ti ratios varying from ~ 5 to $\sim 10^3$ (for fassaïtic pyroxene and melilite).

It has been proposed that negative $\delta^{26}\text{Mg}$ is a result of proton bombardment in the early solar system (Heymann and Dziczkaniec 1976). However, we have not been able to conceive of a bombardment mechanism which could alter the isotopic composition of solar-system Ca in such a manner as to cause excesses of

^{48}Ca . It is possible that a different Ca normalization may yield a pattern of isotopic anomalies compatible with an irradiation model. However, at present, we do not believe that the modification of "normal" solar-system material by local proton irradiation is compatible with the observations (Heymann and Dziczkaniec 1976; Clayton, Dwek, and Woosley 1977), or that the addition of a single homogeneous exotic material from a nearby supernova to "normal" solar-system material is an adequate explanation for the observations. It appears that the average solar value must be accounted for by several separate constituent components, which include end members, responsible for the observed variations. Substantial fluctuations about the average exist on the small scale and may exist on the grand scale within the solar system owing to incomplete mixing between the diverse components. From the observed ^{26}Al abundance in Allende inclusions and using an estimate of the production ratio of $^{26}\text{Al}/^{27}\text{Al} \sim 10^{-3}$ (Truran and Cameron 1978), we have inferred (LPW 1976, LPW 1977a) substantial late contributions of ^{27}Al to the solar system. The observed O_E contents in different planetary bodies indicate a widespread contribution of exotic oxygen to the solid bodies of the solar system. From these observations there is a distinct implication that there was an addition of relatively large amounts of exotic materials of low atomic number immediately before formation of the solar system. This late and large contribution must now be extended to a wide range of elements (see McCulloch and Wasserburg 1978).

In addition to the results previously reported by us on Mg, the new data on Ca and those in the companion paper on Ba and Nd show that in some instances there are substantial isotopic anomalies in refractory elements with a wide range in atomic number. The data on refractory elements may be explained by assuming that the samples are mixtures of material made up of dust grains from different sources which were melted or vaporized and coalesced, thereby preserving the variations represented by the sources but destroying the original grains and initial isotopic heterogeneities within the inclusions now sampled. The data on ^{26}Al - ^{26}Mg still indicate the production or injection of freshly synthesized materials into the early solar system. If the model of condensation from a gaseous part of the solar nebula for the formation of the Ca-Al-rich Allende inclusions is correct, it will require steep gradients in the relative abundances of originally dusty material from distinct nucleosynthetic sources over a scale of 10 to 100 km in the solar nebula at the time of condensation. Such steep gradients appear to be more compatible with a solar irradiation model, but the observed isotopic anomalies do not seem to support that view.

In 1973 October the authors met in conference with W. A. Fowler, T. A. Tombrello, J. C. Huneke, and S. E. Woosley in the Lauritsen Library of the Kellogg Radiation Laboratory to discuss candidates for identifying exotic nuclear components in early solar-system "condensates" which were then known to have an excess of ^{16}O . That discussion plus the interest expressed

by S. Woosley gave support to the endeavors reported here and aided the junior author in persuading the senior author to again pursue this work. We wish to acknowledge our debt to W. A. Russell for freely making available his knowledge of Ca and its isotopes. We have benefited from the interchange of both samples and ideas with R. N. Clayton. We thank H. Nagasawa for

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