

SEARCH FOR ^{26}Al EFFECTS IN THE ALLENDE FUN INCLUSION C1

Tezer M. Esat,* Typhoon Lee,† D. A. Papanastassiou† and G. J. Wasserburg†

*W. K. Kellogg Radiation Laboratory, Division of Physics, Mathematics and Astronomy, Pasadena, California 91125

†The Lunatic Asylum, Division of Geological and Planetary Sciences, California Institute of Technology, Pasadena, California 91125

Abstract. The Mg isotopic composition in plagioclase from the Allende inclusion C1 has been measured to correlate excess $^{26}\text{Mg}^*$ with isotopic anomalies for elements of higher atomic number. No large $^{26}\text{Mg}^*$ excesses were observed in samples with $^{27}\text{Al}/^{24}\text{Mg}$ up to 165. The inferred initial $^{26}\text{Al}/^{27}\text{Al}$ is 5×10^6 which is a factor of ten less than previously observed in other Allende inclusions. The low ^{26}Al abundance may be the result of late formation of C1 (by 2.5 m.y.) or due to the non-uniform addition of ^{26}Al to the solar system. The major mineral phases in C1 which are Mg rich and the bulk alteration products all have a uniform Mg isotopic composition with 30‰ per mass unit fractionation and $\delta^{26}\text{Mg} \approx -1.6\text{‰}$. The plagioclase data show variable Mg isotopic fractionation which ranges from 0 to 30‰ per mass unit. Isotopic homogeneity was demonstrated for the major mineral phases rich in Mg both in macroscopic samples and in microscopic individual crystals. The detailed correlation of ^{26}Al with the more general isotopic anomalies and its use as a chronometer marking the production and introduction of isotopically anomalous material into the solar system remains an open question.

Isotopic anomalies have been discovered in two Allende inclusions, C1 and EK1-4-1. The anomalies range from low-Z elements O [Clayton, Onuma, Grossman and Mayeda, 1977; Clayton and Mayeda (CM), 1977], Mg [Wasserburg, Lee and Papanastassiou (WLP), 1977a] and Si [Clayton, Mayeda and Epstein, 1978; Yeh and Epstein, 1978] to the heavier elements Ca [Lee, Papanastassiou and Wasserburg (LPW), 1978], Sr [Papanastassiou and Wasserburg, 1978], Ba and Nd [McCulloch and Wasserburg, 1978a], and Sm [Lugmair, Marti and Scheinin, 1978; McCulloch and Wasserburg, 1978b]. The Mg and O isotopic patterns found in C1 and EK1-4-1 were the first indicators of the more general anomalies in these inclusions designated by WLP [1977a] as FUN anomalies (for Fractionation and Unknown Nuclear effects). Both C1 and EK1-4-1 were found to contain highly fractionated Mg of 30‰ and 20‰ per mass unit (mu) and negative $\delta^{26}\text{Mg}$ of -1.8‰ and -3.4‰ , respectively, after normalizing for mass fractionation. The negative $\delta^{26}\text{Mg}$ values are due to deficiencies in ^{26}Mg or ^{24}Mg or excesses in ^{25}Mg . Oxygen isotopic data in C1 and EK1-4-1 do not lie along the mixing line observed for Allende inclusions with end members of "normal" oxygen (O_N) and "extraordinary" oxygen (O_E) consisting of essentially pure ^{16}O [Clayton, Grossman and Mayeda, 1973]. The oxygen isotopic pattern in these two inclusions is consistent with mixtures of O_N and O_E which were subsequently fractionated [CM, 1977]. Recently a fractionated pattern was found in the two FUN samples in Si [Clayton et al., 1978; Yeh and Epstein, 1978]. These results show that the large isotopic shifts in Mg and Si in the FUN inclusions must be due to mass fractionation and not to nuclear effects. It is unlikely that a nucleosynthetic event could produce an abundance pattern closely mimicking fractionation for both these elements.

In this letter we report the Mg isotopic composition of major and minor mineral phases in inclusion C1. The purpose was to establish (a) whether the Mg isotopic composition is the same in different phases, and (b) whether ^{26}Al was present in a sample exhibiting fractionation and non-linear nuclear effects in other elements. Mineral phases rich in ^{27}Al were sought out as they could contain detectable excesses of ^{26}Mg ($^{26}\text{Mg}^*$) due to the *in situ* decay of ^{26}Al . The first ^{26}Al - ^{26}Mg internal isochrons were obtained on inclusions BG2-6 and WA [LPW, 1976; WLP, 1976; LPW, 1977a]. These data demonstrated the correlation of ^{26}Mg excesses with ^{27}Al within each inclusion for a variety of mineral phases in which Mg ranges from being a trace element to a major mineral constituent. The data for $^{26}\text{Mg}^*$, up to this point, strongly favor the

presence of "live" ^{26}Al in the solar nebula within a time interval of $\sim 3 \times 10^6$ years prior to condensation. Establishing the presence of $^{26}\text{Mg}^*$ correlated with the $^{27}\text{Al}/^{24}\text{Mg}$ ratio in the Al-rich mineral phases of C1 would lend support to the association of the more general nuclear anomalies with a nearby supernova and provide a time scale for the event.

Results

The minerals in C1 consist of fassaite pyroxene, melilite, spinel, plagioclase and fine grained alteration products that permeate the inclusion, and are most often associated with melilite and plagioclase but are also seen as rims on pyroxene. Plagioclase is present in small amounts ($\sim 0.1\%$). All minerals in C1 appear to be corroded to various degrees as manifested by the presence of alteration products consisting of sodalite, Fe-Mg-rich pyroxene, wollastonite and other phases not yet identified. The alteration products which have a typical grain size of a few micrometers were called fuzz. Bulk samples and individual crystals of each major phase were studied. The comparison of macroscopic and microscopic samples is important for establishing the extent of isotopic homogeneity within minerals of this inclusion. The fractionated Mg composition of C1 provides a rare opportunity to address this matter. The macroscopic samples consisted of density separates or of higher quality, hand-picked mineral grains. The samples were dissolved, Mg chemically separated and analyzed. Single crystals were characterized with an electron probe micro-analyzer (EPMA) and a scanning electron microscope (SEM) and the Mg was analyzed by direct loading. Procedures as developed in this laboratory have been described in detail [WLP, 1977b; LPW, 1977b]. A summary of the samples analyzed in the present work follows.

Pyroxene: (A) bulk separate of pyroxene with abundant spinel inclusions; (B) from sample A, ~ 200 water-clear, inclusion-free grains; (C) one clear pyroxene crystal.

Melilite: (A) bulk separate with some spinel inclusions; (B) 28 hand-picked grains individually studied with the petrographic microscope (under oil immersion) and the EPMA; (C) a similarly characterized melilite crystal for a direct loading experiment.

Spinel: (A) the insoluble residue from pyroxene-A was verified to be pure spinel. A portion of this sample was fused and Mg chemically separated and analyzed; (B) the insoluble spinel residue from fuzz-A was found to include some grains which contained Fe, Cr, Ti and Mn. A single Fe-rich crystal ($\sim 20 \mu\text{m}$) was selected for direct loading.

Fuzz: (A) the lower density separate ($\rho < 2.9 \text{ g/cm}^3$) was ground to less than $60 \mu\text{m}$ and separated by heavy liquids. The portion $\rho < 2.5 \text{ g/cm}^3$ was composed mainly of clumps of micron sized grains rich in Na, Cl, Fe, Mg and Al (Table 1, footnote f) interspersed with fragments ($\leq 30 \mu\text{m}$) of the major phases. This sample was analyzed as the representative of the "sodalite" type material; (B) a clump of fuzz ($\sim 200 \mu\text{m}$) was selected for analysis by direct loading.

Among the pyroxene grains in sample pyroxene-A, a few water-clear, brown-colored ($\sim 100 \mu\text{m}$) crystals were found consisting of Fe-rich pyroxene (Table 1, footnote f). It is not clear whether the Fe-rich pyroxenes were located within the inclusion or whether they were introduced from the outside rim or from the surrounding matrix during sampling. A search of the rim and matrix material and of a thin section of the inclusion did not reveal their origin. Given the presence of abundant iron-rich pyroxenes in the alteration products, it is plausible that these Fe-rich pyroxenes are from the C1 inclusion.

The Mg isotopic compositions of the pyroxene, melilite, spinel and fuzz samples are listed in Table 1. All samples, except for the brown-pyroxene which will be discussed separately, show large fractionation ($\sim 30\text{‰}$ per mu). After correction for mass fractionation [LPW,

1976], a uniform offset of $\delta^{26}\text{Mg} \approx -1.6 \pm 0.2\%$ is found. The uniformity in fractionation and in $\delta^{26}\text{Mg}$ is evident both in the macroscopic and microscopic samples. This confirms the previous observation of isotopic homogeneity for "whole-rock" and spinel samples from inclusion C1 [WLP, 1977a] and extends it to all the major, "primary", mineral phases.

Plagioclase samples were hand-picked from the low density separates ($\rho \leq 2.9 \text{ g/cm}^3$). Examination of the plagioclase crystals with the EPMA showed that the contaminant fuzz was unevenly distributed over the surfaces and in fractures. The Mg content of the fuzz was variable but ranged up to 8%. Since plagioclase grains with very high $^{27}\text{Al}/^{24}\text{Mg}$ ratios are required in order to best demonstrate ^{26}Mg effects correlated with ^{26}Al decay, microsurgical techniques were employed to remove contaminant material from individual crystals. These efforts did not always produce crystals with high $^{27}\text{Al}/^{24}\text{Mg}$ ratios (Table 2) due to the low abundance of plagioclase in C1 and the smaller crystal size as compared to inclusion WA in which similar procedures were applied [LPW, 1977a]. In addition, the cleaning procedures were not pursued with the necessary extreme measures for fear of losing the rare plagioclase crystals. The surfaces of each crystal were scanned with the EPMA to determine the level of Mg contamination and major element composition of each crystal. Following the EPMA and optical microscope verification that over 90% of the volume of a crystal was plagioclase, it was dissolved in 0.01 ml HF and HNO_3 on a teflon (FEP) planchet under the binocular microscope. An aliquot of the solution (~20%) was used to determine the Ca/Mg ratio by isotope dilution. The $^{27}\text{Al}/^{24}\text{Mg}$ ratio was obtained by combining the EPMA result for Ca/Al with the Ca/Mg value as determined by isotope dilution [LPW, 1977b]. The total blank for the directly loaded plagioclase analysis was ~0.06 ng. In addition, two "macroscopic" samples each consisting roughly of 30 crystals which were individually screened were dissolved; the Mg was chemically separated and analyzed. As an additional check of the Mg contamination introduced through the total analytical procedures, high purity plagioclase crystals from the Moore County eucrite were analyzed (Table 2). High $^{27}\text{Al}/^{24}\text{Mg}$ ratios (300-350) were obtained. The Mg contents in Moore County plagioclase correspond to Mg concentrations of 900-400 ppm. Neither Mg amounts as found in Moore County plagioclase nor the measured blanks seriously affect the conclusions in this paper based on the C1 plagioclase data.

Table 1. Mg Isotopic Composition of C1 Samples Except for Plagioclase

Sample	Weight ^a (g)	Fractionation ^b		$\delta^{26}\text{Mg}^c$ (‰)	$^{27}\text{Al}/^{24}\text{Mg}^d$
		min.	max.		
Pyroxene—A	1.5×10^{-2}	29.6	30.8	-1.6 ± 0.2	1.5-2.5
—B	3×10^{-4}	29.3	30.6	-2.0 ± 0.2	1.5-2.5
—C ^e	2×10^{-6}	27.8	30.4	-1.3 ± 0.6	2.4
Melilite —A	5×10^{-4}	29.4	30.8	-1.5 ± 0.2	1-2
—B	1×10^{-5}	31.2	32.6	-1.6 ± 0.2	1-2
—C ^e	1.5×10^{-6}	27.8	30.0	-1.2 ± 0.6	1.4
Fuzz —A ^f	1×10^{-4}	29.6	31.0	-1.9 ± 0.3	2
—B ^{e,f}	4×10^{-6}	27.6	28.8	-1.8 ± 0.5	2
Spinel —A	2×10^{-4}	30.0	31.8	-2.0 ± 0.2	2.5
—B ^e	5×10^{-8}	28.6	30.9	-1.4 ± 0.6	2.5
Brown Pyroxene ^{e,f}	1×10^{-6}	-1.8	+0.2	-0.3 ± 0.6	1

^aFor μg -sized samples the weight was estimated from crystal volume.

^bFractionation (in per mil per m.u.) relative to the measured raw value obtained for normal Mg: $^{25}\text{Mg}/^{24}\text{Mg} = 0.12475$. Five normals run concurrently with the present samples yield raw ratios in good agreement with the previously established raw $^{25}\text{Mg}/^{24}\text{Mg}$ and $^{26}\text{Mg}/^{24}\text{Mg}$ ratios; furthermore the normalized data yield $\delta^{26}\text{Mg} = -0.1 \pm 0.2\%$. ^cThe quoted errors are $2\sigma(\text{mean})$. ^dRange of $^{27}\text{Al}/^{24}\text{Mg}$ obtained from electron microprobe analyses of thin sections and individual crystals. ^eDirectly loaded crystal. ^fEPMA analyses yield: SiO_2 43.5, $\text{TiO}_2 < 0.1$, Al_2O_3 33.4, $\text{Cr}_2\text{O}_3 < 0.1$, FeO 2.5, $\text{MnO} < 0.1$, MgO 7.7, CaO 1.5, Na_2O 3.4, K_2O 0.3, Cl 5.2 for fuzz; and SiO_2 46.0, TiO_2 2.3, Al_2O_3 10.0, Cr_2O_3 0.2, FeO 7.1, MnO 0.1, MgO 10.7, CaO 24.8, $\text{Na}_2\text{O} < 0.1$ for brown pyroxene (amounts in weight per cent).

The data on plagioclase crystals are shown in Table 2 and Fig. 1. The range in $^{27}\text{Al}/^{24}\text{Mg}$ is from 20 to 165. The low values of Al/Mg are most probably due to attached fuzz since measurements of Mg concentration in plagioclase by EPMA give values of 0.2% Mg. It is evident that samples highly enriched in ^{27}Al do not show excesses of $^{26}\text{Mg}^*$ comparable to those observed in inclusions WA and BG2-6. For example, for the initial $(^{26}\text{Al}/^{27}\text{Al})_0 = 5.1 \times 10^{-5}$ as determined for inclusion WA (line A in Fig. 1) the calculated $\delta^{26}\text{Mg}$ for plagioclase-A is 60‰ in contrast to the measured value of +3.0‰. The relatively large errors of directly loaded samples are due to low Mg contents and to the uncertainty of the bias in $\delta^{26}\text{Mg}$ for directly loaded samples (Table 2, footnote e). In order to avoid this bias in data obtained by the direct loading technique [LPW, 1977b], Mg was chemically separated from the larger "bulk" samples F and H. Data on plagioclase-H indicate that $(^{26}\text{Al}/^{27}\text{Al})_0$ in C1 must be lower than in WA by a factor of at least 11.

The plagioclase data show variable mass fractionation which is occasionally much lower than the 30‰ per mu found for the major Mg-rich phases. No regular relationship was observed between the fractionation observed for the plagioclase analyses and the amount of Mg. We believe that the variability is not due to contamination or to mass-spectrometry but is intrinsic to the minerals themselves. The variable fractionation indicates that Mg in the analyzed plagioclase crystals (and the attached fuzz) is a mixture of at least three components: a) fractionated Mg as observed in major phases in C1; b) unfractionated Mg; and c) excess $^{26}\text{Mg}^*$ from ^{26}Al decay. A complexity arises from the possible different initial $^{26}\text{Mg}/^{24}\text{Mg}$ values for components a and b. In particular, if, in the plagioclase, the Mg which is not produced by *in situ* ^{26}Al decay does not have a uniform isotopic composition, the data on plagioclase need not define a ^{26}Al - ^{26}Mg isochron in Fig. 1. It is evident that although the data define a good linear array with very shallow slope, not all data lie on a single straight line. The lack of a well defined isochron is consistent with the observation of variable fractionation in plagioclase samples which implies a lack of isotopic homogeneity in plagioclase for non-radiogenic Mg.

While a strong case for the presence of $^{26}\text{Mg}^*$ cannot be made for C1 from the directly loaded samples due to the uncertainty in the bias correction, the analyses of Mg chemically separated from bulk plagioclase samples H and F show that definite small excesses of ^{26}Mg exist in C1 plagioclase. Sample H shows a clear excess of ^{26}Mg with respect to both normal (N) Mg ($\delta^{26}\text{Mg} = 0\%$, fractionation, $F = 0\%$) and FUN Mg ($\delta^{26}\text{Mg} = -1.6\%$, $F = 30\%$). Since sample H shows a low fractionation it is most plausible to assume that the actual $^{26}\text{Mg}^*$ should be calculated relative to normal Mg. Sample F has $\delta^{26}\text{Mg}$ in the interval 0 to -1.6% . This could be due either to $^{26}\text{Mg}^*$ or to the admixture of normal Mg to FUN Mg. Such admixture would, however, affect both $\delta^{26}\text{Mg}$ and F in the mixture (M). If $X_{\text{FUN}} = \text{Mg}_{\text{FUN}} / (\text{Mg}_{\text{FUN}} + \text{Mg}_{\text{N}})$ represents the fraction of FUN Mg in the mixture we obtain: $F_M = 30\% \cdot X_{\text{FUN}}$ and $\delta^{26}\text{Mg}_M = -1.6\% \cdot X_{\text{FUN}}$. Since sample F has the full complement of 30‰ fractionation, we calculate for this sample $X_{\text{FUN}} \geq 0.9$ and $\delta^{26}\text{Mg}_M = -1.4\%$. The distinct shift of the measured $\delta^{26}\text{Mg}$ to -0.6% for sample F must therefore be due to the presence of $^{26}\text{Mg}^*$ and not to the admixture of normal Mg.

These relationships can also be used to exclude the possibility for some samples that the low fractionation values are due to contamination with laboratory normal Mg. For sample H we calculate $X_{\text{FUN}} = 0.2$ corresponding to 24 ng of Mg_{N} . The measured procedural blank of 1.2 ng and the concurrently run Moore County plagioclase experiments make it highly unlikely that a 24 ng Mg blank could have been introduced. The possibility that the variable fractionation effects were caused by nonstandard mass spectrometer operating conditions for direct loading analyses can be excluded by the Moore County experiments which show no fractionation and were done under the same conditions as C1 plagioclase. The only remaining viable alternative is that unfractionated Mg of normal isotopic composition exists in the plagioclase or on the surfaces and in inclusions in the analyzed plagioclase crystals. We believe that only a small fraction of the Mg can be in the plagioclase crystal structure. As further evidence that our analytical techniques could have easily detected excesses of $^{26}\text{Mg}^*$ in C1 plagioclase, we show in Table 2 analyses of plagioclase and pyroxene from another type B inclusion labelled Egg-1. The data show that the directly loaded plagioclase for this sample has $^{27}\text{Al}/^{24}\text{Mg} = 93$ and $\delta^{26}\text{Mg} = 33.5\%$.

Table 2. Mg Isotopic Composition of Plagioclase Samples

Sample	Weight ^a (μg)	Fractionation ^b		$\delta^{26}\text{Mg}^c$ (‰)	$^{27}\text{Al}/^{24}\text{Mg}$	Mg^d (ng)
		min.	max.			
Allende C1 Plagioclase						
A	2.1	18.5	24.6	3.0±1.8	165	3
B	2.4	26.0	29.1	0.1±1.0	21	27
C	—	24.0	29.2	2.4±1.4	—	—
D	2.4	18.0	25.3	2.1±1.5	78	8
E	—	13.2	19.0	3.0±1.2	—	—
F ^e	8.5	29.4	31.5	-0.6±0.3	20	90
G	1.9	5.1	10.6	0.9±1.1	63	6
H ^e	10.3	2.8	8.7	1.4±0.4	55	30
I	3.0	7.5	14.5	2.6±1.6	94	8
Moore County Plagioclase						
A	2.3	-0.5	-4.3	0.0±1.0	300	2
B	9.5	-6.3	+1.0	-0.1±2.4	320	4
C ^f	2.5	—	—	—	350	1.5
Allende Egg-1 (type B inclusion)						
Pyroxene ^e	—	-0.3	+2.7	0.0±0.4	1.5-2.5	400
Plagioclase	1.8	-2.9	+1.4	33.5±1.0	93	4.4

^aWeight of anorthite crystals determined from the Ca content. ^bSee Table 1, footnote b. ^cErrors are 2σ mean. ^dMg amount in the sample. ^eSample processed through chemistry; other samples analyzed by direct loading. A bias of +1.5±0.5‰ has been added to $\delta^{26}\text{Mg}$ for directly loaded samples, and the errors were combined quadratically. ^fOnly the Ca and Mg concentrations were measured on this sample.

while the pyroxene yields $\delta^{26}\text{Mg} \approx 0$ and $^{27}\text{Al}/^{24}\text{Mg} \approx 2$. The $(^{26}\text{Al}/^{27}\text{Al})_0$ calculated for Egg-1 from these data is identical to the value in inclusion WA [LPW, 1977a].

Discussion

A schematic representation of the Mg isotopic properties of the various mineral phases in inclusion C1 with regard to fractionation (F), unknown nuclear anomalies (UN), and ^{26}Al effects (^{26}Mg excesses) is given in Table 3. The data on C1 show that: 1) microscopic and macroscopic samples of spinel, pyroxene (Fe-free), melilite and the alteration products all have a homogeneous Mg isotopic composition, showing a fractionation of 30‰ per mass unit and $\delta^{26}\text{Mg} \approx -1.6$ ‰; 2) plagioclase samples with high $^{27}\text{Al}/^{24}\text{Mg}$ ratios exhibit only small but distinct excesses of ^{26}Mg *; 3) Mg isotope fractionation in plagioclase is variable; 4) Mg in a rare Fe-rich pyroxene is unfractionated and normal in composition.

The isotopic homogeneity of Mg in Mg-rich major phases in C1 is evident both in the bulk samples and in single microscopic crystals. These observations are in agreement with the homogeneous Mg isotopic composition found in EK1-4-1 [WLP, 1977a]. In contrast, in EK1-4-1, spinel and pyroxene have distinctly different O isotopic composition compared to melilite [CM, 1977]. The spinel and pyroxene lie off the O_E-O_N mixing line, while melilite is on the line close to "normal" oxygen. The difference between the O and Mg isotopic patterns together with the presence of low temperature phases (including alkalis, halogens and Fe) penetrating throughout the inclusion which consists predominantly of high temperature condensates, was accounted for by invoking a late-stage back-reaction mechanism [WLP, 1977a; CM, 1977]. In this scheme, the constituent mineral phases within the inclusion were initially assumed to be isotopically uniform. The inclusion was then immersed in a relatively cool region of the solar nebula containing oxygen of normal composition. Substantial oxygen isotope exchange took place, particularly for melilite and plagioclase which are assumed to be the more reactive mineral phases. The Mg isotopic composition between the various Mg-rich phases remained unaltered as there was too little Mg left in the gas phase. This is supported by the observation of Mg with a fractionated composition for fuzz in C1. This fuzz contains an abundance of residual unreacted melilite. The bulk Mg in the fuzz must represent material obtained directly from the melilite in C1. This view is compatible with the fact that the isotopic compositions of the refractory elements Ca, Sr, Ba, Nd and Sm are anomalous but uniform in pyroxene and melilite.

The variable fractionation observed in plagioclase grains presents a new constraint for the Allende inclusions and requires that, in addition to the FUN Mg which comprises the bulk of the Mg in C1, there exist small amounts of "normal" Mg in C1. This material is in C1 and cannot plausibly be explained as an artifact of laboratory Mg contamination. The back-reaction mechanism introduced previously to account for the O isotopic heterogeneity between melilite and the other phases can be extended to explain the observed effects in Mg. The following is a plausible sequence of events:

a) At the time of solidification of C1, the intergrown mineral phases had uniform but fractionated Mg isotopic composition with a deficit of $\delta^{26}\text{Mg} \approx -1.6$ ‰. Spinel, pyroxene, melilite and plagioclase were formed as primary minerals crystallizing from a supercooled melt [Blander and Fuchs, 1975; WLP, 1977a]. Furthermore, some ^{26}Al was present in this chondrule.

b) The C1 chondrule was later immersed in a cool, volatile-rich region of the solar nebula with which it reacted. Among other elements, alkalis, halogens, Fe, O_N and a small amount of refractory elements, including Mg with normal isotopic composition diffused into the inclusion. The corrosive gas reacted with the various mineral phases exchanging the O in some phases and producing a variety of reaction products (mostly from melilite and plagioclase) with the addition of O, alkalis, halogens and Fe. Primary phases rich in Mg and with characteristic C1 FUN Mg reacted to form fuzz with essentially all Mg in the fuzz provided by the pre-existing Mg-rich primary phases.

c) Plagioclase presumably exchanged its O and was extensively altered by the reaction. As the intrinsic Mg content (originally of the C1, FUN-type) was very low in the plagioclase, the addition of a very small amount of normal Mg would significantly alter the isotopic composition of Mg in the plagioclase. The relative extent to which FUN Mg would be present in the plagioclase would depend on the degree of transport of Mg from nearby crystals during the chemical attack, the geometric isolation of individual plagioclase crystals, and the amount of normal Mg introduced by the corrosive medium in the neighborhood of Mg poor plagioclase crystals. The plagioclase grains and the intimately associated reaction products are, therefore, considered to contain four types of Mg: 1) the original FUN Mg, 2) $^{26}\text{Mg}^*$ from *in situ* ^{26}Al decay, 3) normal Mg introduced with normal O in the back-reaction exchange with the corrosive medium either before or after ^{26}Al decay, 4) FUN Mg remobilized from adjacent Mg-rich primary phases and contributing to the reaction products.

Evidence for the presence of isotopically normal Mg in C1 is found

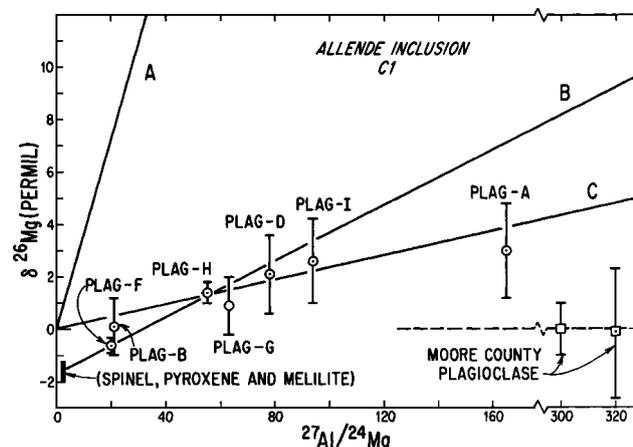


Fig. 1. Plagioclase data from Allende inclusion C1 and Moore County. Line A represents the isochrons obtained from Allende inclusions BG2-6, BG3-13 and WA with slope $(^{26}\text{Al}/^{27}\text{Al})_0 \approx 5 \times 10^{-5}$. Lines B and C represent fits to two groups of data: a) spinel, pyroxene and melilite with $\delta^{26}\text{Mg} = -1.6$ ‰ and 30‰ per mu fractionation and the chemically separated Mg from sample F with fractionation of 30‰ per mu; b) normal Mg ($\delta^{26}\text{Mg} = 0$) and the chemically separated Mg from sample H which shows nearly zero fractionation. Note that $\delta^{26}\text{Mg}$ for plagioclase F and H have small errors.

also in the few Fe-rich pyroxene grains, although their association with C1 has not been conclusively established. The composition of this pyroxene is similar to the mineralogy of the late-stage rims discovered by Wark and Lovering [1978]. In the framework of the above scenario, the iron-rich pyroxene represents rare "coarse-grained" crystals of the late-stage condensation and reaction products which grew on pre-existing material. The presence of microscopic crystals of wollastonite, Fe-rich pyroxene, grossular and Na-Cl-Fe-rich fuzz in vugs, cracks, and reaction surfaces in C1 and other type B inclusions makes this scenario plausible. Insofar as the typical alteration products in C1 contain fractionated Mg, it is not possible to grow these products *in situ* in the meteorite by migration of Mg from the matrix. Instead, this alteration must precede the aggregation of the meteorite. The implied early introduction of alkalis and halogens is consistent with the observation of excess ¹²⁹Xe in sodalite rich material in EK1-4-1 [Papanastassiou, Huneke, Esat and Wasserburg, 1978].

The most striking feature of the results on plagioclase in C1 is the presence of only small excesses of ²⁶Mg* in phases highly enriched in ²⁷Al/²⁴Mg. The present data indicate that ²⁶Al existed in C1 and that (²⁶Al/²⁷Al)₀ is about a factor of 10 lower in inclusion C1 than in WA. Evidence for ²⁶Mg* effects exists in only a limited number of Allende inclusions. The existing data and the present results for C1 suggest that the ²⁶Al abundance in the early solar nebula was non-uniform.

Inclusion C1 appears to be a "poor relative" of EK1-4-1 in that nuclear anomalies in EK1-4-1 are much more extensive, both in the number of anomalous isotopes and magnitude of effects. In particular ¹³⁶Ba and ⁴⁸Ca appear to be depleted in C1 suggesting that this sample represents solar system material which did not receive its full complement of exotic material to bring it up even to the solar system average. Following this pattern, it is possible that C1 did not receive its full complement of ²⁶Al. If the added material (of high Z) is produced in the same event and stellar site as ²⁶Al, then we would expect EK1-4-1 which has substantial excesses of many isotopes to contain an overabundance of ²⁶Al relative to ²⁶Al/²⁷Al ~ 5 x 10⁻⁵. This argument implies that ²⁶Al/²⁷Al mixtures in the solar nebula were heterogeneous and that differences in ²⁶Al/²⁷Al may not be used as a chronometer. Alternative postulates are: a) C1 condensed or remelted at a somewhat later time than WA (ΔT ≈ 2.5 x 10⁶ years); b) arbitrary amounts of ²⁶Al or ²⁶Mg* were introduced into the plagioclase crystals during the late-stage back reaction or the plagioclase crystals are a secondary and not primary phase in the type B inclusions.

We conclude that the present data show that a small ²⁶Mg excess exists in C1 and that it is correlated with the ²⁷Al/²⁴Mg ratio. This indicates that ²⁶Al was present in C1 together with the more general nuclear anomalies. It is possible that study of EK1-4-1 will better clarify the relationship between ²⁶Al and the FUN anomalies. The absence of nuclear anomalies in Ca, Sr and Ba in inclusion WA, which shows a high initial (²⁶Al/²⁷Al)₀ ratio indicates that the correlation of ²⁶Al with the FUN anomalies is not a simple matter. The observation of both positive and negative non-linear anomalies [LPW, 1978; McCulloch and Wasserburg, 1978a] has necessitated the presence of at least two components with distinct isotopic compositions. The association of ²⁶Al with either or both of these components remains an open question. We note that chemical alteration has significantly affected the C1 inclusion and modified the isotopic characteristics of mineral phases both for oxygen and magnesium. This requires that we consider, as part of the general problem, ongoing chemistry in the early solar system over an extended time, possibly up to the present. We are not simply dealing with mixtures of various distinct nucleosynthetic components but instead with products of complex physical-chemical processes which have significantly modified the isotopic characteristics of the original nucleosynthetic components.

Table 3. Truth Table for Mg Isotopic Properties of Minerals in C1

	Spinel	Pyroxene	Melilite	Fuzz	Plagioclase	Brown Pyroxene
F	+	+	+	+	—	—
UN	+	+	+	+	I	—
²⁶ Al	I	I	I	I	+	I

Symbols: F: Fractionation; UN: Unknown Nuclear anomalies; +: present; —: absent; I: indeterminate.

Acknowledgements. We thank A. L. Albee for ready access to the electron microprobe facility and R. L. Lewis for generous assistance in the use of the scanning electron microscope. The paper benefited greatly from constructive detailed reviews by J. H. Reynolds and L. E. Nyquist. T. L. thanks the Enrico Fermi Institute of the University of Chicago and the Robert R. McCormick Trust for allowing him the freedom to complete the final phase of this work at Caltech. This work was supported by NASA NGL 05-002-188 and NSF 76-83685. This is contribution 3106 (277) of the Division of Geological and Planetary Sciences.

References

- Blander, M., and L. H. Fuchs, Ca-Al-rich inclusions in Allende: evidence for liquid origin, *Geochim. Cosmochim. Acta*, 39, 1605-1619, 1975.
- Clayton, R. N., L. Grossman, and T. K. Mayeda, A component of primitive nuclear composition in carbonaceous meteorites, *Science*, 182, 485-488, 1973.
- Clayton, R. N., N. Onuma, L. Grossman, and T. K. Mayeda, Distribution of the presolar component in Allende and other carbonaceous chondrites, *Earth Planet. Sci. Lett.*, 34, 209-224, 1977.
- Clayton, R. N., and T. K. Mayeda, Correlated O and Mg isotope anomalies in Allende inclusions: I. Oxygen, *Geophys. Res. Letters*, 4, 295-298, 1977.
- Clayton, R. N., T. K. Mayeda, and S. Epstein, Isotopic fractionation of Si in Allende inclusions, in *Lunar and Planetary Science IX*, Lunar and Planetary Institute, Houston, 186-188, 1978.
- Lee, T., D. A. Papanastassiou, and G. J. Wasserburg, Demonstration of ²⁶Mg excess in Allende and evidence for ²⁶Al, *Geophys. Res. Letters*, 3, 109-112, 1976.
- Lee, T., D. A. Papanastassiou, and G. J. Wasserburg, ²⁶Al in the early solar system: fossil or fuel?, *Ap. J. (Letters)*, 211, 107-110, 1977a.
- Lee, T., D. A. Papanastassiou, and G. J. Wasserburg, Mg and Ca isotopic study of individual microscopic crystals from the Allende meteorite by the direct loading technique, *Geochim. Cosmochim. Acta*, 41, 1473-1485, 1977b.
- Lee, T., D. A. Papanastassiou, and G. J. Wasserburg, Ca isotopic anomalies in the Allende meteorite, *Ap. J. (Letters)*, 220, 21-25, 1978.
- Lugmair, G. W., K. Marti, and N. B. Scheinin, Incomplete mixing of products from r-, p-, and s-process nucleosynthesis: Sm-Nd systematics in Allende inclusion EK1-4-1, in *Lunar and Planetary Science IX*, Lunar and Planetary Institute, Houston, 672-674, 1978.
- McCulloch, M. T., and G. J. Wasserburg, Ba and Nd isotopic anomalies in the Allende meteorite, *Ap. J. (Letters)*, 220, 15-19, 1978a.
- McCulloch, M. T., and G. J. Wasserburg, More anomalies from the Allende meteorite: Sm, *Geophys. Res. Letters*, 5, 599-602, 1978b.
- Papanastassiou, D. A., J. C. Huneke, T. M. Esat, and G. J. Wasserburg, Pandora's box of the nuclides, in *Lunar and Planetary Science IX*, Lunar and Planetary Institute, Houston, 859-861, 1978.
- Papanastassiou, D. A., and G. J. Wasserburg, Strontium isotopic anomalies in the Allende meteorite, *Geophys. Res. Letters*, 5, 595-598, 1978.
- Wark, D. A., and J. F. Lovering, Marker events in the early evolution of the solar system: evidence from rims on Ca-Al-rich inclusions in carbonaceous chondrites, *Proc. Lunar Sci. Conf. 8th*, 95-112, 1978.
- Wasserburg, G. J., T. Lee, and D. A. Papanastassiou, Large ²⁶Mg excesses in an Allende inclusion and a ²⁶Al-²⁶Mg internal isochron, *Meteoritics*, 11, 384-385, 1976.
- Wasserburg, G. J., T. Lee, and D. A. Papanastassiou, Correlated O and Mg isotopic anomalies in Allende inclusions: II. Magnesium, *Geophys. Res. Letters*, 4, 299-302, 1977a.
- Wasserburg, G. J., T. Lee, and D. A. Papanastassiou, Mg and Ca isotopic study of individual microscopic crystals from the Allende meteorite by the direct loading technique, in *Lunar Science VIII*, Lunar and Planetary Institute, Houston, 991-993, 1977b.
- Yeh, H.-W., and S. Epstein, ²⁹Si/²⁸Si and ³⁰Si/²⁸Si of meteorites and Allende inclusions, in *Lunar and Planetary Science IX*, Lunar and Planetary Institute, Houston, 1289-1291, 1978.

(Received June 13, 1978;
accepted July 17, 1978.)