

## CORRECTION\*

DEMONSTRATION of  $^{26}\text{Mg}$  EXCESS in ALLENDE and EVIDENCE for  $^{26}\text{Al}$ 

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**Abstract.** We report the discovery of a large anomaly in the isotopic composition of Mg in a Ca-Al rich chondrule from the Allende meteorite. This anomaly is manifest independently of instrumental fractionation and is due to an enrichment of about 1.3 percent in  $^{26}\text{Mg}$  while the abundances of  $^{25}\text{Mg}$  and  $^{24}\text{Mg}$  are terrestrial in value. There is a strong correlation in this chondrule between the  $^{26}\text{Mg}$  excess and the Al/Mg ratio so that the most plausible cause of the anomaly is the *in situ* decay of now extinct  $^{26}\text{Al}$  ( $\tau_{1/2} = 0.72 \times 10^6$  yr). Mineral phases extracted from a Ca-Al-rich aggregate have distinct Al/Mg but show identical, small Mg anomalies which are apparent after correction for fractionation ( $\delta^{26}\text{Mg} = 0.3\%$ ). These data indicate that this aggregate was isotopically homogenized in a high Al/Mg environment after the decay of  $^{26}\text{Al}$  had occurred or that some of the Mg anomalies are due to effects other than *in situ* decay of  $^{26}\text{Al}$ .

Measurements of the isotopic composition of many elements have demonstrated that the earth, meteorites and the moon appear to be nearly homogeneous in isotopic composition, although highly heterogeneous in chemical composition. This suggests that materials from different nucleosynthetic sources were well mixed prior to the formation of planetary bodies. It is possible to search for and identify variations in isotopic abundances relative to a standard or "normal" state, e.g., the terrestrial abundances. Such variations must reflect:

- 1) the preservation of material from distinct nucleosynthetic processes prior to the formation of the solar system; or
- 2) the modification of "normal" isotopic abundances by nuclear or physico-chemical processes in the solar system.

Most of the isotopic variations which have been detected are attributed to the second type and result from radioactive decay, nuclear reactions in the solar system, or isotopic fractionation within the solar system. However, variations in Ne reported by Black [1972] and in O by Clayton, Grossman and Mayeda [1973] have been interpreted as due to the first type and resulting from incorporation of interstellar dust grains into solar system materials in a manner which preserves the isotopic character of distinct nucleosynthetic sources. Many of the variations in isotopic abundances which occur in nature result from fractionation of normal materials by physico-chemical and biologic processes. The smooth isotopic abundance patterns resulting from small fractionation processes are readily recognizable and are found in nature and as a result of laboratory procedures and instrumental effects. The relationship for small fractionation for a chemical element is given approximately by  $R_{ij}/R_{ij}^0 = 1 + \alpha(i-j)$ , where  $R_{ij}$  is the abundance ratio of the isotopes of mass numbers  $i$  and  $j$ ,  $R_{ij}^0$  is the normal ratio and  $\alpha$  is the fractionation factor per unit mass difference. For variations much larger than instrumental effects, it is possible to identify which isotopes are enriched or depleted. However, in many cases the magnitude of the effects is comparable to, or smaller than, instrumental fractionation. For these cases, identification of an isotopic abundance shift is based on the recognition of differences in isotopic abundances relative to the pattern generated by small fractionation effects. Isotopic abundance shifts which are not the product of a small degree of isotopic fractionation, e.g., due to diffusion or chemical phenomena, have been called "nuclear anomalies". It would perhaps be better to call such variations *non-linear* isotopic effects. For small effects, for an element with only three isotopes, it is not possible to determine uniquely the isotope responsible for the

non-linear effects. For four or more isotopes the assignment of the shift in abundance is well defined if it appears only at one mass number.

It is possible to obtain non-linear isotopic effects by extreme fractionation processes which might be attributed to nuclear effects. For example, if a reservoir of oxygen enriched by a factor of 10-100 in  $^{16}\text{O}$  were produced by isotopic fractionation of normal solar system material, and were mixed later with another reservoir of normal oxygen, then the mixture would not be recognizable as the result of fractionation processes and might be interpreted as a nuclear effect.

A search for non-linear Mg variations was carried out by Schramm, Tera and Wasserburg [1970]. These workers developed the high sensitivity and high precision procedures currently in use for Mg and showed that a wide variety of materials had no non-linear variations to within 0.03 percent. The discovery in Allende of "high temperature" material [Marvin, Wood and Dickey, 1970; Grossman, 1972] and of very primitive Sr [Gray, Papanastassiou and Wasserburg, 1973], and the discovery of the  $^{16}\text{O}$  anomaly by Clayton, Grossman, and Mayeda [1973] led to a new Mg search. Two groups recently reported effects in Mg from inclusions in Allende using a normalization relative to  $^{25}\text{Mg}/^{24}\text{Mg}$ . Lee and Papanastassiou [1974] found anomalies in a number of inclusions with  $\delta^{26}\text{Mg}$  ranging from  $-0.2\%$  to  $+0.3\%$  and Gray and Compston [1974] found an anomaly of  $\delta^{26}\text{Mg} = +0.4\%$ . Since these effects are comparable to, or smaller than, instrumental fractionation effects it was impossible to determine whether the positive  $\delta^{26}\text{Mg}$  shifts were due to an excess of  $^{24}\text{Mg}$  or  $^{26}\text{Mg}$  or a depletion of  $^{25}\text{Mg}$ . The negative effect could not be attributed to an excess of  $^{26}\text{Mg}$ .

As we will be considering evidence that some of the isotopic effects are due to *in situ* decay of  $^{26}\text{Al}$ , let us consider the basic relationships. Suppose that a short lived nuclide (e.g.,  $^{26}\text{Al}$ ) were present at some stage in the solar nebula. If a sample which condensed from the solar nebula was isotopically homogeneous in the constituent mineral phases and if these phases were preserved until the present, then the isotopic abundance of Mg in the different phases (p) would be given by

$$(^{26}\text{Mg}/^{24}\text{Mg})_p = (^{26}\text{Mg}/^{24}\text{Mg})_0 + (^{26}\text{Al}/^{27}\text{Al})_0 (^{27}\text{Al}/^{24}\text{Mg})_p \quad (1)$$

This is the basic isochron equation for an extinct nuclide.  $(^{26}\text{Mg}/^{24}\text{Mg})_0$  and  $(^{26}\text{Al}/^{27}\text{Al})_0$  are the isotopic ratios in the inclusion at the last time (0) of homogenization. Phases (p') of another inclusion which formed at a later time  $\tau$  from the same parcel of the local solar nebula (LSN) would give

$$(^{26}\text{Mg}/^{24}\text{Mg})_{p'} = [(^{26}\text{Mg}/^{24}\text{Mg})_0 + (^{26}\text{Al}/^{27}\text{Al})_0 (^{27}\text{Al}/^{24}\text{Mg})_{\text{LSN}}] (1 - e^{-\lambda\tau}) + (^{26}\text{Al}/^{27}\text{Al})_0 e^{-\lambda\tau} (^{27}\text{Al}/^{24}\text{Mg})_{p'}$$

This isochron corresponds to both a different slope and initial  $^{26}\text{Mg}/^{24}\text{Mg}$  than equation (1).

**Sample Description.** We have continued our search for isotopic variations in a wider variety of inclusions and mineral phases. A large fragment (BG2) was provided by A. Burlingame. This yielded a distinctive buff-white spherical inclusion (BG2-6) about 2mm diameter with a grain size typically  $\sim 2\mu\text{m}$  and larger crystals up to  $20\mu\text{m}$ . This chondrule was strikingly zoned with the grain size increasing toward the center. Samples taken from near the exterior were identified as grossular by electron microprobe and x-ray diffraction (Table 1). Coarser grained samples were identified by electron microprobe as close to hedenbergite ( $\text{CaFeSi}_2\text{O}_6$ ). X-ray diffraction data are consistent with this identification. Sample AAI was kindly provided by

\*The first two paragraphs of this paper were omitted from the January 1976 issue due to an oversight by the AGU Office.

Table 1. Quantitative Electron Microprobe Analyses<sup>a</sup>

	Grossular Ca <sub>3</sub> Al <sub>2</sub> Si <sub>3</sub> O <sub>12</sub>	Hedenbergite CaFeSi <sub>2</sub> O <sub>6</sub>			Hibonite <sup>b</sup> CaAl <sub>12</sub> O <sub>19</sub>	Spinel <sup>b</sup> MgAl <sub>2</sub> O <sub>4</sub>
SiO <sub>2</sub>	39.12	48.27	48.57	45.59	0.23	<0.01
TiO <sub>2</sub>	0.11	0.03	0.02	0.10	3.62	0.1
Al <sub>2</sub> O <sub>3</sub>	23.88	<0.01	<0.01	8.88	87.19	66.0
Cr <sub>2</sub> O <sub>3</sub>	0.14	0.16	0.16	<0.01	n.d. <sup>c</sup>	n.d.
FeO	0.53	28.11	25.01	21.11	0.05	3.0
MnO	<0.01	0.35	<0.01	<0.01	n.d.	n.d.
MgO	0.59	0.05	0.02	0.09	2.24	23.6
CaO	37.25	23.22	27.96	24.75	8.08	0.1
Na <sub>2</sub> O	0.02	<0.01	<0.01	<0.01	n.d.	n.d.
Sum	101.7	100.2	101.7	100.4	101.4	92.8

<sup>a</sup>Weight percent Analysis of grossular and hedenbergite from BG2-6, hibonite and spinel analyses are from AAl residue. <sup>b</sup>Unpolished small grains on graphite disc. <sup>c</sup>n.d. = not determined.

C. Gray and W. Compston. We pursued the study of C1, a melilite-fassaite chondrule containing spinel [see Gray et al., 1973].

**Procedures.** All procedures were as reported in Lee and Papanastassiou [1974] referred to as L & P. We dissolved two separate pieces of BG2-6A and B. Some material from BG2-6 (~1 μg) was directly loaded on a filament. A split of AAl was dissolved. After dissolution of BG2-6A and AAl small residues remained. Seventy-four grains from the BG2-6A residue were analyzed semi-quantitatively using an energy dispersive x-ray detector on the electron microprobe; 60% of the grains were rich in Mg and Al with small amounts of Fe while the rest were rich in Al, Ca and Ti. Of 100 grains from the residue of AAl, 20% were rich in Mg and Al and 80% were rich in Al, Ca and Ti. Quantitative microprobe analysis of the AAl and BG2-6A residues revealed that the Mg and Al rich phase is spinel (MgAl<sub>2</sub>O<sub>4</sub>) while the Ca, Ti and Al phase is hibonite, CaAl<sub>12</sub>O<sub>19</sub> (Table 1). From grain counts we estimate Al/Mg of the BG2-6A residue as 3.5 ± 1 and that of AAl as 9.5 ± 2. For other samples Al and Mg were determined by atomic absorption and isotope dilution.

Scattered Al ions contribute to the background of <sup>24</sup>Mg [L & P, 1974]. Extensive tests were performed for directly loaded samples which typically show a high intensity <sup>27</sup>Al beam. After correction for fractionation, using the procedures of L & P (1974), the <sup>26</sup>Mg/<sup>24</sup>Mg of directly loaded samples was too low by 0.15%. We have increased <sup>26</sup>Mg/<sup>24</sup>Mg for directly loaded samples by 0.15%. The conclusions of this paper are not sensitive to this correction.

**Isotopic Results Without Normalization.** Fig. 1 shows histograms for all runs of BG2-6 and AAl without fractionation corrections. Results for C1 are also shown [L & P, 1974, and present work]. Each entry in the histograms is the mean of every set of ten sequential ratios taken throughout the run. We show all data on normal samples over the past two years. These consist of 25 runs yielding 396 sets of ten ratios on reagent MgO, terrestrial peridotite and spinel, and a total sample of the chondrite Guarena. The variation in normal samples is due to instrumental fractionation. The fractionation during a single run varies much less than from run to run so that we shall use the

Table 2. Un-normalized Mg Ratios

Sample	Sets	<sup>25</sup> Mg/ <sup>24</sup> Mg			<sup>26</sup> Mg/ <sup>24</sup> Mg		
		Mean±σ <sup>a</sup>	min	max	Mean±σ <sup>a</sup>	min	max
Normals	396	0.12475±13	0.12440	0.12540	0.13569±27	0.13529	0.13734
BG2-6A	16	0.12486±3	0.12483	0.12491	0.13770±7	0.13759	0.13784
	14 <sup>b</sup>	0.12464±5	0.12458	0.12472	0.13719±10	0.13699	0.13745
BG2-6B	15	0.12481±5	0.12474	0.12493	0.13779±16	0.13766	0.13830
AAl	16	0.12491±8	0.12478	0.12504	0.13672±19	0.13653	0.13728
	18	0.12479±9	0.12471	0.12513	0.13644±25	0.13629	0.13741
C1	60	0.12850±10	0.12832	0.12881	0.14370±22	0.14347	0.14442
C1 <sup>b</sup> Res	15	0.12840±7	0.12856	0.12832	0.14328±17	0.14310	0.14373
C1 <sup>d</sup> Res	10	0.12878±12	0.12858	0.12897	0.14436±31	0.14407	0.14486
Normals		Number of Sets <sup>c</sup>					
		<-3σ <sub>D</sub>	<-2σ <sub>D</sub>	<-σ <sub>D</sub>	[-σ <sub>D</sub> , +σ <sub>D</sub> ]	>σ <sub>D</sub>	>2σ <sub>D</sub>
<sup>25</sup> Mg/ <sup>24</sup> Mg	396	0	2	57	276	63	14
<sup>26</sup> Mg/ <sup>24</sup> Mg	396	0	1	61	230	105	32

<sup>a</sup>Standard deviation of the distribution. <sup>b</sup>High Al signal during mass spectrometer run. <sup>c</sup>Residue directly loaded on filament <sup>d</sup>Residue fused and processed <sup>d</sup>Distribution of sets in different intervals; e.g. entry under "<-2σ<sub>D</sub>" is the total number of sets whose value is lower than the grand mean minus 2σ<sub>D</sub>.

extensive data on normals to establish the range of our instrumental fractionation. It is well known that the observance of very limited variations in raw isotopic ratios during a given run offers no assurance that the run is not subject to fractionation.

The standard deviations σ<sub>D5</sub>, σ<sub>D6</sub> of the distribution for all runs on normals and the minimum and maximum values obtained are given in Table 2. The distribution for <sup>25</sup>Mg/<sup>24</sup>Mg appears to be Gaussian. The distribution for <sup>26</sup>Mg/<sup>24</sup>Mg shows a marked positive skewness. From this variety of experiments we conclude that the instrumental fractionation is well described by the parameters for normals. Ratios from an individual run which differ from normal values by more than the 3σ<sub>D</sub> values determined for the normal samples, are resolvable from normal Mg. We conclude that if the <sup>25</sup>Mg/<sup>24</sup>Mg ratios agree with the mean normal value to well within the observed standard deviation σ<sub>D5</sub> in the normal runs and if the <sup>26</sup>Mg/<sup>24</sup>Mg ratios differ from normal by significantly more than 3σ<sub>D6</sub>, then the isotopic anomaly is reasonably assignable to <sup>26</sup>Mg.

The data in Table 2 and Fig. 1 show that: 1) both BG2-6 samples have <sup>25</sup>Mg/<sup>24</sup>Mg which are well within 1σ<sub>D5</sub> from the mean of the normals; 2) both samples have <sup>26</sup>Mg/<sup>24</sup>Mg which exceed the mean of the normals by 8σ<sub>D6</sub>; and 3) the minimum value of <sup>26</sup>Mg/<sup>24</sup>Mg observed for any set on these samples is slightly greater than the maximum value for all normal sets. Therefore, we conclude that sample BG2-6 shows essentially normal <sup>25</sup>Mg/<sup>24</sup>Mg and has a distinct excess in <sup>26</sup>Mg.

For sample AAl the mean for <sup>25</sup>Mg/<sup>24</sup>Mg lies within 1σ<sub>D5</sub> of the mean of normal runs; the mean for <sup>26</sup>Mg/<sup>24</sup>Mg is 4σ<sub>D6</sub> greater than for normal runs. However, the minimum and maximum <sup>26</sup>Mg/<sup>24</sup>Mg for AAl are within the bound of the maximum <sup>26</sup>Mg/<sup>24</sup>Mg for normal runs. These data are strongly suggestive of an excess in <sup>26</sup>Mg but the arguments are not as cogent as for BG2-6 Gray and Compston [1974] used their data on AAl to argue for an excess in <sup>26</sup>Mg of 0.4% despite extensive instrumental isotopic fractionation of ±1.5% in <sup>26</sup>Mg/<sup>24</sup>Mg. Their range of fractionation is a factor of 2.5 times greater than our resolution in <sup>26</sup>Mg/<sup>24</sup>Mg (3σ<sub>D6</sub> = 0.6%, Table 2). Our analyses confirm their normalized data for this sample. However, it is clear that their results do not permit the assignment of the isotopic shift in AAl to an excess in <sup>26</sup>Mg rather than an excess in <sup>24</sup>Mg or a deficiency in <sup>25</sup>Mg. This discussion is not intended to obfuscate their independent discovery of a Mg isotopic effect.

Unnormalized Mg ratios of C1 were previously found to be significantly fractionated in four experiments. At that time we were not certain that large instrumental fractionation could be excluded [L & P 1974]. We have further analyzed the insoluble residue of C1 by direct loading and after fusion followed by chemistry. These analyses show the same large fractionation as the soluble part of C1 (Table 2). These observations coupled

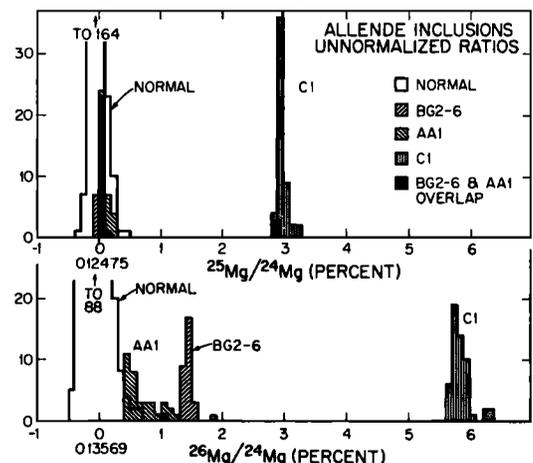


Fig. 1. Histograms of un-normalized Mg ratios. Each entry corresponds to the mean of a set of ten ratios. Note that Mg which is fractionated from the normal values would show a <sup>25</sup>Mg/<sup>24</sup>Mg enrichment half as large as the <sup>26</sup>Mg/<sup>24</sup>Mg enrichment (similar to C1).

Table 3. Normalized Analytical Results<sup>a</sup>

Sample	<sup>26</sup> Mg/ <sup>24</sup> Mg 2σ errors	<sup>26</sup> Mg Excess <sup>b</sup> percent	<sup>27</sup> Al/ <sup>24</sup> Mg	( <sup>26</sup> Al/ <sup>27</sup> Al) <sub>0</sub> x10 <sup>4</sup>
BG2-6A	0.14164± 3	1.31±0.02	32	0.58
	0.14159± 3	1.28±0.02		
Residue <sup>c</sup>	0.14007± 6 <sup>d</sup>	0.19±0.04 <sup>d</sup>	(4.4) <sup>e</sup>	(0.60)
BG2-6B	0.14186± 6	1.47±0.04		
BG2-6C <sup>f</sup>	0.14160±20 <sup>d</sup>	1.3 ±0.1 <sup>d</sup>		
AAI	0.14049± 4	0.49±0.03	13	0.53
	0.14052± 6	0.51±0.04		
Residue <sup>c</sup>	0.14042± 6 <sup>d</sup>	0.41±0.01 <sup>g</sup>	15 <sup>g</sup>	0.38 <sup>g</sup>
		0.44±0.04 <sup>d</sup>	(12) <sup>e</sup>	(0.51)
B30	0.14019± 3	0.27±0.02	10.4	{0.37} <sup>i</sup>
Residue <sup>h</sup>	0.14022± 6	0.30±0.04	5.2	{0.81} <sup>i</sup>

<sup>a</sup>Normalized to <sup>25</sup>Mg/<sup>24</sup>Mg = 0.12663. <sup>b</sup>Using <sup>26</sup>Mg/<sup>24</sup>Mg = 0.139805 for normals as corrected for fractionation. <sup>c</sup>Residue directly loaded on filament and corrected (see text). <sup>d</sup>High Al signal during mass spectrometer run. <sup>e</sup>Estimate based on grain counts using electron microprobe. <sup>f</sup>Mini-chip directly loaded on filament. <sup>g</sup>Datum from Gray and Compston (1974). <sup>h</sup>Residue was fused and then processed. <sup>i</sup>Calculated as if due to *in situ* <sup>26</sup>Al decay.

with the better defined distribution of the unnormalized ratios for normal Mg permit the clear identification of significantly fractionated Mg in C1.

**Results after Normalization.** We conclude that for BG2-6 we may assign the non-linear isotopic effect to an excess of <sup>26</sup>Mg and plausibly assume that <sup>25</sup>Mg/<sup>24</sup>Mg is normal. The normalized results are shown in Table 3. Two samples of BG2-6A are in agreement and sample B shows a similar but distinctly higher excess, which is attributed to sample heterogeneity.

To compare Mg isotopic effects on different phases from the same inclusion, acid insoluble residues were analyzed (Table 3). The spinel-rich residue of BG2-6 has a small <sup>26</sup>Mg excess which is apparent after normalization. These results show that different phases of this inclusion are isotopically heterogeneous and that this heterogeneity is correlated with the Al/Mg ratio. The data are displayed on a Mg-Al evolution diagram (Fig. 2). The acid insoluble residue and bulk AAI inclusion show identical Mg anomalies and the two fractions have similar Al/Mg. These data are compatible with the presence in the two inclusions of normal <sup>25</sup>Mg/<sup>24</sup>Mg and of <sup>26</sup>Al with an initially uniform <sup>26</sup>Al/<sup>27</sup>Al. The unnormalized data for BG2-6 indicate an *initial* <sup>26</sup>Mg/<sup>24</sup>Mg value close to modern terrestrial. If we interpret the two data points of BG2-6 as an internal isochron due to the *in situ* decay of <sup>26</sup>Al we obtain (<sup>26</sup>Al/<sup>27</sup>Al)<sub>0</sub> = 0.6x10<sup>-4</sup>. The data for AAI lie in this isochron and are consistent with the same (<sup>26</sup>Al/<sup>27</sup>Al)<sub>0</sub>.

From the value of (<sup>26</sup>Al/<sup>27</sup>Al)<sub>0</sub> we may calculate the change in <sup>26</sup>Mg/<sup>24</sup>Mg in the reservoir from which the samples came, assuming that Al/Mg is near the solar value 0.08. The total shift of <sup>26</sup>Mg/<sup>24</sup>Mg due to the subsequent decay of <sup>26</sup>Al after the reference time defined by BG2-6 is 0.005%. If the <sup>26</sup>Al abundance were higher at an earlier time by a factor of ten, then the effect would be 0.05%. This shows that recrystallization effects which equilibrate <sup>26</sup>Mg produced from <sup>26</sup>Al with materials with solar Al/Mg would mask any enrichment.

Let us now assume that all of the non-linear Mg effects so far reported are due to an excess of <sup>26</sup>Mg. If all of the isotopic effects are due to the presence of <sup>26</sup>Al in normal material, then the deviations of these data points from the isochron of BG2-6 would correspond to an interval in time of 1.4x10<sup>6</sup> years in the early solar system using <sup>26</sup>Al.

B30 is an aggregate that showed a δ<sup>26</sup>Mg = 0.3% [L & P, 1974]. We have analyzed the acid insoluble residue of B30 after fusion and chemical separation. The residue shows δ<sup>26</sup>Mg = 0.3% although its Al/Mg is one-half that of the bulk sample. This result differs markedly from the strong correlation of Al/Mg and isotopic effects observed in BG2-6.

Sample C1 is unique in that the unnormalized data show marked enrichment in <sup>25</sup>Mg and <sup>26</sup>Mg relative to <sup>24</sup>Mg (Fig. 1). The general pattern appears to be one of large isotopic

fractionation. We conclude that significant isotopic fractionation processes were active during the formation of some Allende inclusions. Following L & P [1974], C1 has a negative <sup>26</sup>Mg anomaly of 0.2% after normalization of <sup>25</sup>Mg/<sup>24</sup>Mg. As this negative anomaly cannot be due to <sup>26</sup>Al, it will be important to determine if this is truly a non-linear effect due to nuclear effects or if it is the result of incorrect normalization for large fractionation effects.

The question of normalization requires attention particularly for large fractionation effects. As pointed out by Schramm *et al.* [1970], their normalized <sup>26</sup>Mg/<sup>24</sup>Mg did not agree with the value of Catanzaro, Murphy, Garner and Shields [1966]. Our normalized <sup>26</sup>Mg/<sup>24</sup>Mg for normals agrees with that of Schramm *et al.* [1970]. A redetermination of the absolute isotopic composition of Mg appears to be necessary.

**Discussion.** The observed large non-linear isotopic shifts in Mg can be assigned to a definite excess of <sup>26</sup>Mg whereas the <sup>24</sup>Mg and <sup>25</sup>Mg abundances are approximately normal. Correlation of the excess <sup>26</sup>Mg with Al/Mg in different phases of the same inclusion provides strong evidence that the <sup>26</sup>Mg is produced by the *in situ* decay of <sup>26</sup>Al added to solar system material with normal Mg composition and that the correlation on an Al-Mg evolution diagram represents an internal isochron for BG2-6. In contradistinction, the identical δ<sup>26</sup>Mg for the two samples of B30 with different Al/Mg could be an indication that some of the non-linear Mg effects are not due to an excess of <sup>26</sup>Mg. However, the B30 data can be explained by the decay of <sup>26</sup>Al if we assume that B30 remained a closed system but that the phases reached isotopic equilibrium, possibly by melting, after the <sup>26</sup>Al had decayed.

It is possible that the correlation line for BG2-6 is a mixing line resulting from the presence of an Al rich phase containing highly enriched <sup>26</sup>Mg and a phase low in Al and containing normal solar system Mg. In this case <sup>26</sup>Al would be a fossil nuclide at the time of incorporation in the solar system. The Al rich host phases could represent aggregates of circumstellar dust grains condensed from ejecta of the central star which formed rapidly after the nucleosynthesis of <sup>26</sup>Al in the star. This model is constructed to permit the preservation of fossil anomalies and may be tested by a search for other isotopic anomalies which would be associated with <sup>26</sup>Al nucleosynthesis and would be trapped in Al rich condensates.

If BG2-6 does not contain preserved extra-solar material then <sup>26</sup>Al must have been made in the solar system or freshly synthesized <sup>26</sup>Al was added to the solar system immediately before condensation started. The time interval between the <sup>26</sup>Al producing event and the formation of BG2-6 cannot be more than a few million years based on the calculated (<sup>26</sup>Al/<sup>27</sup>Al)<sub>0</sub> and any plausible production mechanism for <sup>26</sup>Al [c.f. Schramm, 1974]. The contribution of the last addition of material to the total production for a stable nuclide made in the same process as <sup>26</sup>Al could be significant. For example, if <sup>26</sup>Al is synthesized along with <sup>27</sup>Al with a

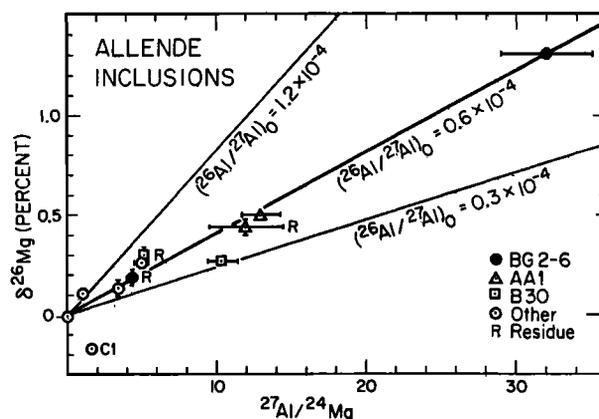


Fig. 2. Al-Mg evolution diagram for Allende samples. The correlation line for BG2-6 yields (<sup>26</sup>Al/<sup>27</sup>Al)<sub>0</sub> = 0.6 x 10<sup>-4</sup> and contrasts sharply with the correlation line for B30 which has essentially zero slope and much higher initial <sup>26</sup>Mg/<sup>24</sup>Mg.

production ratio  $^{26}\text{Al}/^{27}\text{Al} \approx 10^{-3}$ , and if the  $(^{26}\text{Al}/^{27}\text{Al})_0$  of BG2-6 is representative of the solar system, then the last minute spike which provided the  $^{26}\text{Al}$  must have contributed at least 5% of the  $^{27}\text{Al}$  in the solar system.

Xe anomalies due to  $^{129}\text{I}$  and  $^{244}\text{Pu}$  decays [Reynolds, 1960; Rowe and Kuroda, 1965] require addition of r-process material to the solar system about  $2 \times 10^8$  years before Xe retention in meteorites [c.f., Schramm and Wasserburg, 1971]. The observed Xe anomalies in Allende [Podosek and Lewis, 1972] and the very short half life of  $^{26}\text{Al}$  require that either the  $^{26}\text{Al}$  event and the  $^{129}\text{I}$  and  $^{244}\text{Pu}$  events were independent or that Xe retention started  $\sim 2 \times 10^8$  years after condensation of Al-rich materials.

If  $^{26}\text{Al}$  is made in the solar system, the bombardment of Al, Si and Mg by protons with energy greater than 10 Mev seems to be the most plausible mechanism as first discussed by Fowler, Greenstein and Hoyle [1962]. Some workers have interpreted the Mg effects as due to proton irradiation in the solar system (D. Heymann, private communication). The minimum proton fluence needed to produce the observed  $^{26}\text{Al}$  abundance is  $\sim 10^{20}/\text{cm}^2$  [see Schramm, 1971, and Reedy and Arnold, 1972]. If proton irradiation occurred in the nebula prior to condensation, then only a small fraction of the nebula could be irradiated by any single source of protons since the range of protons is limited due to shielding by gas. In this case the  $(^{26}\text{Al}/^{27}\text{Al})_0$  value reported here would not be an average value for the solar system but a lower limit for the irradiated material. A "uniform irradiation" model implies short range accelerating mechanisms operating throughout the whole gas cloud which could generate high energy protons over large regions or continuous mixing of the whole nebula during the irradiation. If irradiation of condensed debris occurred (a la FGH) the shielding problem is avoided. In this case other isotopic effects due to spallation should be present in Allende unless remixing occurred. The effects of secondary neutrons generated in proton reactions depend on the specific configuration of the target material. It is interesting to note that a pure  $^{16}\text{O}$  component can also be generated with intense proton irradiation. The appropriate proton energy range for a complete destruction of  $^{17}\text{O}$  and  $^{18}\text{O}$  without significant depletion of  $^{16}\text{O}$  is between 4 and 9 Mev. In principle the O anomaly could be caused by mixing this  $^{17}\text{O}$  and  $^{18}\text{O}$  poor material with normal material.

If the  $^{26}\text{Mg}$  effects are due to *in situ*  $^{26}\text{Al}$  decay, then effects due to other short-lived nuclides are possible [Lee et al., 1975].  $^{53}\text{Mn}$  decays into  $^{53}\text{Cr}$  with  $\tau_{1/2} = 3.7 \times 10^6$  years. It is produced abundantly in the spallation of  $^{56}\text{Fe}$  by protons of energy similar to that required for  $^{26}\text{Al}$  production [Reedy and Arnold, 1972]. Further, if  $^{26}\text{Al}$  were a late addition of newly synthesized stellar material and if that material also contained Fe group nuclei then  $^{53}\text{Cr}$  should show pronounced isotopic effects because all  $^{53}\text{Cr}$  is thought to be produced as  $^{53}\text{Mn}$  in nucleosynthesis of the Fe group [see Bodansky, Clayton and Fowler, 1968].

Urey [1955] first pointed out that  $^{26}\text{Al}$  might be a possible heat source in the early solar system. Using  $^{26}\text{Al}/^{27}\text{Al} = 0.6 \times 10^{-4}$  and assuming a solid body of chondritic composition, we calculate [Carlsaw and Jaeger, 1959] the minimum size of a planetary body which will melt. For a thermal diffusivity of  $\kappa = 0.007 \text{ cm}^2/\text{sec}$ , the center of a body of 6km radius will reach 1800°K (zero initial temperature). For a body nearer a "fluffy" dust aggregate and with thermal diffusivity similar to the lunar regolith,  $\kappa = 0.00013 \text{ cm}^2/\text{sec}$ , the minimum radius of the body becomes 0.6km. A high  $^{26}\text{Al}$  abundance as used here would provide a heat source for effectively all early formed bodies a few kilometers in size and could explain the metamorphism in meteorites as well as allow the early production of differentiated bodies (achondrites, iron meteorites) on small parent bodies.

The present work demonstrates the existence of a  $^{26}\text{Mg}$  excess and shows the correlation of this excess with the Al/Mg ratio in the same inclusion. These observations can be explained by the *in situ* decay of  $^{26}\text{Al}$ . Another inclusion shows the same small  $\delta^{26}\text{Mg}$  for phases with different Al/Mg which can be explained by remelting after the decay of  $^{26}\text{Al}$ . However, the general pattern of Mg isotopic anomalies is not yet shown to be fully consistent with the  $^{26}\text{Al}$  hypothesis and an apparently negative  $^{26}\text{Mg}$  anomaly was obtained which cannot be explained by  $^{26}\text{Al}$ . A large natural fractionation effect has been found for Mg isotopes. The resolution of the O anomaly and its relation with Mg effects are still eluding us.

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#### References

- Black, D.C., On the origin of trapped He, Ne and Ar isotopic variations in meteorites - II. Carbonaceous meteorites. *Geochim. Cosmochim. Acta*, **36**, 377-394, 1972.
- Bodansky, D., D.D. Clayton, and W.A. Fowler, Nuclear quasi-equilibrium during silicon burning. *Ap. J. Suppl.*, **16**, 299-371.
- Carlsaw, H.S., and J.C. Jaeger, Conduction of heat in solids, 2nd ed., Oxford Univ. Press, London, 1959.
- Catanzaro, E.J., T.J. Murphy, E.L. Garner, and W.R. Shields, Absolute isotopic abundance ratios and atomic weight of magnesium, *J. Res. N.B.S.* **70A**, 453, 1966.
- Clayton, R.N., L. Grossman, and T.K. Mayeda, A component of primitive nuclear composition in carbonaceous meteorites, *Science*, **182**, 485-488, 1973.
- Fowler, W.A., J.L. Greenstein, and F. Hoyle, Nucleosynthesis during the early history of the solar system, *Geophys. J. Roy. Astron. Soc.*, **6**, 148-220, 1962.
- Gray, C.M., and W. Compston, Excess  $^{26}\text{Mg}$  in the Allende meteorite, *Nature*, **251**, 495-497, 1974.
- Grossman, L., Condensation in the primitive solar nebula, *Geochim. Cosmochim. Acta*, **36**, 597-618, 1972.
- Lee, T., and D.A. Papanastassiou, Mg isotopic anomalies in the Allende meteorite and correlation with O and Sr effects, *Geophys. Res. Letters*, **1**, 225-228, 1974.
- Lee, T., D.A. Papanastassiou, and G.J. Wasserburg, Discovery of a large  $^{26}\text{Mg}$  isotope anomaly and evidence for extinct  $^{26}\text{Al}$ , *Bull. Amer. Phys. Soc.*, **20**, 1486, 1975.
- Marvin, U.B., J.A. Wood, and J.S. Dickey, Jr., Ca-Al rich phases in the Allende meteorite, *Earth Planet. Sci. Letters*, **7**, 346-350, 1970.
- Podosek, F.A., and R.S. Lewis,  $^{129}\text{I}$  and  $^{244}\text{Pu}$  abundances in white inclusions of the Allende meteorite, *Earth Planet. Sci. Letters*, **15**, 101-109, 1972.
- Reynolds, J.H., Determination of the age of the elements, *Phys. Rev. Letters*, **4**, 8-10, 1960.
- Rowe, M.W., and P.K. Kuroda, Fissionogenic xenon from the Pasamonte meteorite, *J. Geophys. Res.*, **70**, 709-714, 1965.
- Reedy, R.C., and J.R. Arnold, Interaction of solar and galactic cosmic ray particles with the moon, *J. of Geophys. Res.*, **77**, 537-555, 1972.
- Schramm, D.N., Nucleosynthesis of  $^{26}\text{Al}$  in the early solar system and in cosmic rays, *Astrophys. Space Sci.*, **13**, 249-266, 1971.
- Schramm, D.N., F. Tera, and G.J. Wasserburg, The isotopic abundance of  $^{26}\text{Mg}$  and limits on  $^{26}\text{Al}$  in the early solar system, *Earth Planet. Sci. Letters*, **10**, 44-59, 1970.
- Schramm, D.N., and G.J. Wasserburg, Nucleochronologies and the mean age of the elements, *Ap. J.*, **162**, 57-69, 1970.
- Urey, H.C., The cosmic abundances of potassium, uranium, and the heat balance of the earth, the moon, and mars, *Proc. Natl. Acad. Sci. U.S.A.*, **41**, 127-144, 1955.

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