

ALUMINUM-26 IN THE EARLY SOLAR SYSTEM: FOSSIL OR FUEL?

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ABSTRACT

The isotopic composition of Mg was measured in different phases of a Ca-Al rich inclusion in the Allende meteorite. Large excesses of ^{26}Mg of up to 10% were found. These excesses correlate strictly with the $^{27}\text{Al}/^{24}\text{Mg}$ for four coexisting phases with distinctive chemical compositions. Models of in situ decay of ^{26}Al within the solar system and of mixing of interstellar dust grains containing fossil ^{26}Al with normal solar system material are presented. The observed correlation provides definitive evidence for the presence of ^{26}Al in the early solar system. This requires either injection of freshly synthesized nucleosynthetic material into the solar system immediately before condensation and planet formation, or local production within the solar system by intense activity of the early Sun. Planets promptly produced from material with the inferred $^{26}\text{Al}/^{27}\text{Al}$ would melt within $\sim 3 \times 10^6$ yr.

Subject headings: abundances — meteors and meteorites — nucleosynthesis — solar system: general

I. INTRODUCTION

Because of the considerable significance of ^{26}Al as a short-time astrophysical chronometer and indicator of nuclear processes as well as a heat source for the planets, we have continued our studies of Mg isotopic anomalies. We here present new observations on the isotopic composition of Mg in the Allende meteorite and show data that demonstrate excesses of ^{26}Mg of up to 10% which are strongly correlated with the element Al and which appear to be due to the decay of ^{26}Al with a mean life of 10^6 years. We will attempt to deduce whether these effects are the result of in situ decay of ^{26}Al within the solar system or if they could be due to the entrapment and preservation of Al-rich interstellar grains which originally contained ^{26}Al but which contained only the fossil $^{26}\text{Mg}^*$ when incorporated into the solar system.

Techniques for high-precision Mg isotopic analyses were first developed by Schramm, Tera, and Wasserburg (1970), who showed that terrestrial, lunar, and ordinary meteoritic materials showed no detectable Mg effects. Variations in the isotopic composition of Mg which are not attributable to isotope fractionation were discovered in the Allende meteorite by Lee and Papanastassiou (1974) and Gray and Compston (1974). These first discoveries showed clear but small effects which could not be unequivocally attributed to a particular Mg isotope. In a more extended search, Lee, Papanastassiou, and Wasserburg (LPW) (1976a) established in one chondrule (BG-2-6) a large excess of $\sim 1.3\%$ in ^{26}Mg , with ^{24}Mg and ^{25}Mg of approximately "normal" abundance. The effects in this chondrule correlated with Al/Mg in different phases and provided strong evidence that the $^{26}\text{Mg}^*$ was due to the in situ decay of ^{26}Al and that the correlation of the effect with Al/Mg represented an internal isochron. Another

sample was found with small but distinct Mg anomalies which, if attributed to ^{26}Mg , did not correlate with Al/Mg. A third chondrule (BG-3-13) showed a ^{26}Mg excess and a correlation with Al/Mg (LPW 1976b).

One of the key tests of the ^{26}Al hypothesis is that a crystal which originally contained Al but no Mg would now contain pure ^{26}Mg with no ^{24}Mg or ^{25}Mg . While such a pure end member may not exist, it was deemed important to find crystals of Al-rich phases which intrinsically contain low Mg and which have no Mg-rich crystals of other phases attached or included. Prior to this report there were only two samples which were found to exhibit ^{26}Mg anomalies lying well outside of possible instrumental fractionation effects. Because of the small sample size of these inclusions, and the intimate intergrowth of phases of high and low Al/Mg, it was not possible to obtain analyses of several pure phases in the same inclusion.

II. RESULTS

Recently Chen and Tilton (1976) in a Pb investigation of Allende, reported on a coarse-grained chondrule (WA) containing anorthite, pyroxene, spinel, and melilite. These workers generously provided us samples of WA. A single anorthite crystal ($\sim 250 \mu\text{m}$, Anorthite A) was first selected and found to be almost free of inclusions. The crystal was loaded on a filament for direct measurement. The Mg was determined to be essentially normal for $^{25}\text{Mg}/^{24}\text{Mg}$ and enriched by 3% for $^{26}\text{Mg}/^{24}\text{Mg}$. A spinel crystal (A) taken from within 1 mm of the anorthite crystal was also analyzed by direct loading and yielded Mg of normal composition. While the precise Al/Mg ratio of the anorthite crystal is not known, the data on these two crystals with distinctive chemical compositions demonstrate a correlation between ^{26}Mg excess and the Al/Mg ratio (Table 1).

* Contribution No. 2811.

TABLE 1
SUMMARY OF RESULTS

Phase		$^{27}\text{Al}/^{24}\text{Mg}^*$	$\delta^{26}\text{Mg}_N^\dagger$ (%)	$\delta^{26}\text{Mg}_C^\ddagger$ (%)	$(^{26}\text{Al}/^{27}\text{Al})_0^\S$ $\times 10^5$
Anorthite	G	—	95 ± 2	97	—
CaAl ₂ Si ₂ O ₈	G1	245**	81 ± 3 #	85	4.8
	G2	(235)	77.4 ± 0.5	86	
	B1	128	47 ± 2 #	50	5.4
	B2	(235)	46.7 ± 0.5 #	49	
	B3		34.1 ± 0.3	49	
A		—	29 ± 2	31	—
Melilite		9.1	3.9 ± 0.3	3.9	6.0
Ca ₂ (Mg,Al,Si) ₃ O ₇	(12.0)				
Spinel	A	—	-1.0 ± 0.3	0.5	—
	B	(2.5)	0.3 ± 0.2	0.3	—
MgAl ₂ O ₄					
Fassaite		2.0	0.1 ± 0.2	0.1	—
Ca(Mg,Al,Ti)(Si,Al) ₂ O ₆	(2.5)				

* "Bulk" value of dissolved sample separate, uncertainty $\pm 6\%$; parenthesized values are from electron microprobe analyses with a 20 μm spot size.

† Corrected for mass fractionation by normalizing $^{25}\text{Mg}/^{24}\text{Mg}$ to 0.12663.

‡ Normalized data further corrected for blank and for offsets in runs with high Al signal when applicable (see LPW, 1976a).

§ Calculated as $[\delta^{26}\text{Mg}_C / (^{27}\text{Al}/^{24}\text{Mg})] \times 0.139805$.

|| Direct load of sample grains.

Direct load of dissolved sample without chemistry.

** Al calculated from Ca concentration by assuming the sample is pure anorthite (uncertainty $\pm 3\%$).

To establish a quantitative relationship with Al/Mg, 30 grains ($\sim 10^{-4}$ g) of anorthite (B) essentially free of foreign phases were prepared. Similar separates of melilite, fassaite, and spinel were also prepared. The dissolution, chemical separation, and mass spectrometric techniques follow our standard procedure (LPW 1976a). Al concentrations were determined on aliquots (2–5 μg Al) by a neutron activation technique adapted and miniaturized from the macroprocedures used by R. Schmitt. Two aliquots of the dissolved anorthite B (B1 and B2 containing 10 ng and 40 ng of Mg, respectively) were measured by direct loading prior to chemical separation. Both aliquots yielded the same ^{26}Mg excess of $\sim 5\%$ (Table 1), confirming a low and constant Mg loading blank (~ 0.2 ng). Because of small sample size, the Mg blank contamination for the chemically separated aliquot (B3) was 40%; but the data for B3, when corrected for the measured blank, agree with the directly loaded aliquots. An additional larger anorthite separate of much superior purity was prepared from which two grains (sample G) were directly loaded and found to have an excess $^{26}\text{Mg}^*$ of 9.7%. The remaining 100 grains were dissolved, and an aliquot (G1) showed $^{26}\text{Mg}^*$ of 8.5%. The same result was obtained from the chemically separated larger aliquot (G2) after correction for a 10% measured blank for Mg.

The results are shown in Table 1 and Figure 1. All raw $^{25}\text{Mg}/^{24}\text{Mg}$ ratios (uncorrected for instrumental effects and independent of sample size and beam stability) are within 0.7% of the normal value, indicating no significant variations in $^{25}\text{Mg}/^{24}\text{Mg}$. The raw $^{26}\text{Mg}/^{24}\text{Mg}$ for all the anorthite samples are greatly

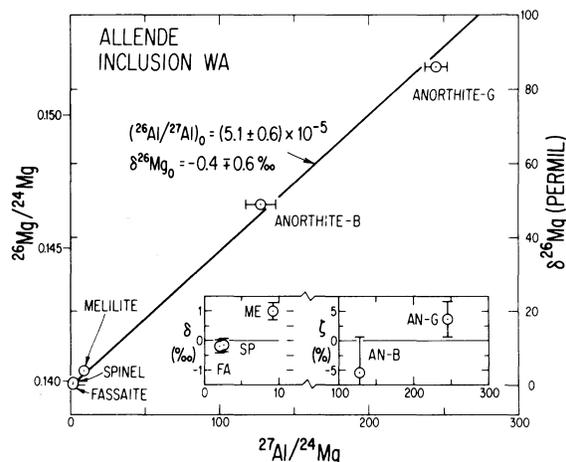


FIG. 1.—Internal isochron defined by four coexisting phases. Note that the melilite point appears near the origin only because of the scale. The insert shows fractional deviations of the data from the best-fit line in the ordinate (δ ; permil) or in the abscissa (ξ ; percent) depending on which data uncertainty is dominant. The slope and initial Mg are indicated (errors are 2σ).

enriched. These data again show that the anomaly is due to an excess of ^{26}Mg far beyond possible instrumental effects. Assuming that $^{25}\text{Mg}/^{24}\text{Mg}$ is precisely normal (N) and correcting for mass discrimination, we then calculate the fractional ^{26}Mg isotopic shift as $\delta^{26}\text{Mg}$, given in per mil (‰) relative to $(^{26}\text{Mg}/^{24}\text{Mg})_N = 0.139805$. The data define a good linear array and give a best fit slope corresponding to $^{26}\text{Mg}^*/^{27}\text{Al} = (5.1 \pm 0.6) \times 10^{-5}$ with an intercept at $^{27}\text{Al}/^{24}\text{Mg} = 0$ of $(\delta^{26}\text{Mg})_0 = -0.4 \pm 0.6$ per mil (2σ errors). The $^{26}\text{Mg}^*/^{27}\text{Al}$ is close to that of BG-2-6. Note that the $^{27}\text{Al}/^{24}\text{Mg}$ values of the "bulk" dissolved sample separates of the phases approach those of the pure phases determined by electron microprobe (values in parentheses in Table 1). This indicates that the melilite separate is not a mixture of low Al/Mg melilite and of anorthite. Therefore, the observed linear array is defined not by arbitrary mixtures, but by four, well defined, coexisting phases with Al/Mg governed by the chemical composition of each phase.

III. FOSSIL OR FUEL?

In situ decay.—Let us assume that ^{26}Al was present in the solar nebula at the time that small (~ 1 cm) solid objects formed and that some of these objects are preserved until today without subsequent alteration. If the isotopic composition of each of the elements were homogeneous within a small object, either because of melting and isotopic mixing of the object itself or because of local homogeneity of the volume of the solar nebula from which the object formed, then the different phases p within an object with different Al/Mg would at the present time follow the basic "isochron" equation (LPW 1976a):

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

where $(^{27}\text{Al}/^{24}\text{Mg})_p$ is the ratio in phase p . On a plot of $^{26}\text{Mg}/^{24}\text{Mg}$ and $^{27}\text{Al}/^{24}\text{Mg}$ (Fig. 2a) the data will form a straight line with intercept $(^{26}\text{Mg}/^{24}\text{Mg})_0$ (initial Mg) and slope $(^{26}\text{Al}/^{27}\text{Al})_0$. Another object formed τ years later from the same reservoir will have an isochron of less steep (but positive) slope and a higher intercept reflecting the growth of ^{26}Mg in the reservoir due to ^{26}Al decay during the time τ . Objects formed from a reservoir with Al/Mg of average solar system abundance will have essentially the same initial $(^{26}\text{Mg}/^{24}\text{Mg})_0$ because $(^{27}\text{Al}/^{24}\text{Mg})_0 \approx 0.1$ (Cameron 1973). Objects formed from reservoirs highly enriched in Al relative to Mg will have initial $^{26}\text{Mg}/^{24}\text{Mg}$ distinctly higher than normal solar system material. The systematics of in situ decay of ^{26}Al , for systems which were initially isotopically homogenized, require that coexisting phases of undisturbed samples form a

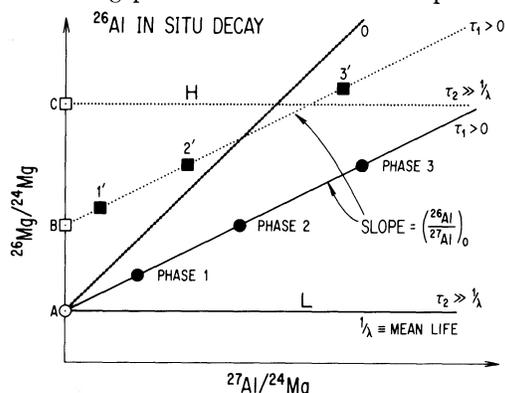


FIG. 2a.—Al-Mg evolution diagram for two synchronous systems formed from reservoirs with very low (L) and high (H) $^{27}\text{Al}/^{24}\text{Mg}$ and which were initially isotopically homogeneous at time 0. Different objects of age 0 and the phases in these objects all lie on A0. Phases (1, 2, 3) from objects formed from L at τ_1 will lie on an isochron passing through A, while phases (1', 2', 3') in objects formed from H at τ_1 will lie on a parallel isochron through B.

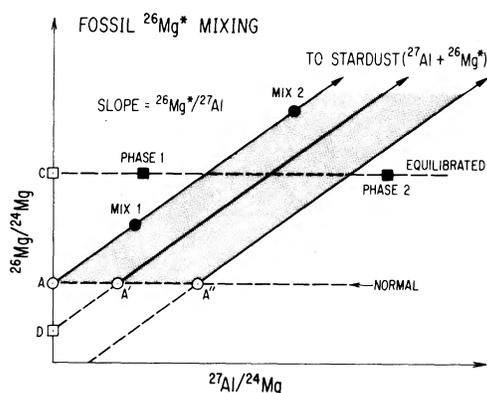


FIG. 2b.—Systematics of mixing of normal material with stardust containing ^{27}Al and fossil $^{26}\text{Mg}^*$ produced by ^{26}Al decay prior to incorporation in the solar system. Mixtures (1 and 2) of stardust with normal material A containing no Al fall on a mixing line with slope equal to the $^{26}\text{Mg}^*/^{27}\text{Al}$ in the stardust. Mixtures with solar system ^{27}Al (A' and A'') populate the shaded zone and extrapolate to erroneous intercepts. Phases formed from recrystallization of mixtures after isotopic homogenization fall on the horizontal line through C, which corresponds to $^{26}\text{Mg}/^{24}\text{Mg}$ of the total mixture.

straight line on a ^{26}Al evolution diagram and that high $(^{26}\text{Mg}/^{24}\text{Mg})_0$ be observed if high $(^{27}\text{Al}/^{24}\text{Mg})$ reservoirs existed. If the ratio $^{26}\text{Al}/^{27}\text{Al}$ were uniform in the solar system, the isochrons would provide a chronometer for systems with Al-rich phases.

Mixing of fossil $^{26}\text{Mg}^$ from ^{26}Al decay.*—Consider an alternate model in which the $^{26}\text{Mg}^*$ is the result of ^{26}Al decay in interstellar dust grains (which we will call “stardust”) and that no ^{26}Al was present when these grains were mixed with “normal” solar system material. Similar models have been proposed by Clayton (1975) to explain the ^{129}Xe anomalies produced by the decay of ^{129}I (Reynolds 1960). We assume that the grains are pure Al_2O_3 except for the $^{26}\text{Mg}^*$. Then arbitrary mixtures of such material with normal solar system material which is free from Al will define a straight line which passes through a point representing normal Mg (point A in Fig. 2b). The slope of this line is $^{26}\text{Mg}^*/^{27}\text{Al}$ in the stardust. Samples of all objects representing different mixtures would lie on a single line independent of time. If an object composed of this mechanical mixture were melted and isotopically homogenized, then new phases formed within the object would have the same enriched $^{26}\text{Mg}/^{24}\text{Mg}$ ratios but different $^{27}\text{Al}/^{24}\text{Mg}$. However, any chemical reaction between the grains in the original mechanical mixture which disproportionates Al, Mg, and $^{26}\text{Mg}^*$ to produce crystals of different composition would produce arbitrary Mg anomalies independent of the Al/Mg ratio (e.g., consider a $^{26}\text{Mg}^*\text{O}$ crystal grown from the stardust). Further, mixtures of stardust with solar system material A' containing Al and Mg will yield a parallel line which extrapolates to an erroneous initial value below A. Thus samples of different mixtures in which variable amounts of solar system Al are present will not generate a straight line. The only model of this type compatible with the data would require that the preponderance of Al in the mixtures come from the stardust. This model would be consistent with horizontal lines such as through C, but it would not yield linear arrays with positive slope if the phases in an “aggregate” were produced with significant chemical disproportionation.

Comparison.—From consideration of the two types of systematics it is clear that the data cannot be explained by mixing fossil $^{26}\text{Mg}^*$ with solar system material unless each crystal analyzed within a single inclusion represents mixtures of stardust and solar system material nearly free of Al. The fact that we analyzed well-defined phases makes this an implausible alternative. Furthermore, the textural evidence in this inclusion points overwhelmingly toward chemical reaction and recrystallization, most plausibly from a molten stage. It follows from these arguments that ^{26}Al must have been present in the inclusions at the time of formation of the crystals now observed and thus in the solar system itself. This ^{26}Al may have been produced in the solar system or introduced by presolar interstellar grains or gas. In any case, the data require isotopic homogenization of Al and Mg in the inclusions at a time when ^{26}Al was still present. No plausible case can be made that the Allende chondrules are extra-

solar-system interstellar "marbles" with otherwise normal isotopic characteristics. This is evident from the uniformity of $^{25}\text{Mg}/^{24}\text{Mg}$ and the negative results of searches for isotopic effects in Rb and Sr (Gray, Papanastassiou, and Wasserburg 1973), Ba (McCulloch, Papanastassiou, and Wasserburg 1976), and Sm and Gd (Curtis and Wasserburg 1976). We have analyzed Ca in three inclusions which show O and Mg effects and Ca in the anorthite G1 in this work. After normalizing $^{44}\text{Ca}/^{40}\text{Ca}$, we find that ^{42}Ca , ^{43}Ca , ^{46}Ca , and ^{48}Ca abundances in these samples are indistinguishable from those of normal Ca to within 0.05%, 0.1%, 2%, and 0.3%, respectively.

IV. DISCUSSION

Urey (1955) first suggested ^{26}Al as a possible heat source for early planetary differentiation (see also Fish, Goles, and Anders 1960). For material with chondritic composition and an initial $^{26}\text{Al}/^{27}\text{Al} = 5 \times 10^{-5}$ the available heat is 2 kcal g^{-1} . This is enough to melt the center of planets and solid objects which formed early with radii larger than a few kilometers (LPW 1976a).

The existence of ^{26}Al at the observed abundance requires a nucleosynthetic event immediately before or during solar system formation. The production of ^{26}Al was last reviewed by Schramm (1971). This matter now requires reassessment. The time interval between the last r -process event and Xe retention in solar system objects calculated from ^{129}I and ^{244}Pu is $\Delta \sim 10^8$ years (Schramm and Wasserburg 1970). If Xe retention started soon after condensation, then the ^{26}Al event and the last r -process event cannot be from the same source. Furthermore, Δ cannot represent a period of isolation of the protosolar cloud if ^{26}Al represents an addition to the solar system. If all the ^{129}I were produced in the ^{26}Al event instead of in an r -process, then the calculation of Δ breaks down. An estimate of Δ can then be obtained which is an order of magnitude greater, as may be seen from the upper limit $\Delta_{\text{max}} \sim 10^9$ years estimated by assuming ^{244}Pu and ^{232}Th are from a single r -process event.

Isotopic effects discovered by Clayton, Grossman, and Mayeda (1973) suggest that oxygen in the solar system is the result of incomplete mixing of two reservoirs. We have constructed phenomenological models in which the contaminating reservoir is assumed to contain all the ^{17}O , ^{18}O , and ^{26}Al in the solar system, but without much ^{16}O or Mg, and the other reservoir represents the bulk of solar system material with most of the ^{16}O (Wasserburg, Papanastassiou, and Lee 1976). This model permits fewer nuclei to be added to the solar system as compared to a model that adds ^{16}O nuclei. The model also explains the lack of correlation between $\delta^{26}\text{Mg}$ and $\delta^{18}\text{O}$ and the apparent contradiction between the observed O isotopic heterogeneity and the approximate homogeneity of $^{26}\text{Al}/^{27}\text{Al}$. It also implies that $^{26}\text{Al}/^{18}\text{O} \sim 10^{-4}$ in the contaminating reservoir and that the observed O effects are not gross heterogeneities but residual fluctuations of a mixing process which is 90% complete. If the oxygen effects are due to a contaminating reservoir containing ^{16}O and ^{26}Al , then the approximate homogeneity of $^{26}\text{Al}/^{27}\text{Al}$ indicates that the added component provided more than 30% of the ^{16}O in at least local volumes of the solar nebula. Consequences of importing ^{17}O , ^{18}O , and ^{26}Al and possibly other rare isotopes of the low- Z elements such as ^{13}C , ^{15}N , ^{19}F , ^{21}Ne , and ^{22}Ne in one single component must be examined under the light of nuclear astrophysics.

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REFERENCES

- Cameron, A. G. W. 1973, *Space Science Rev.*, **15**, 121.
 Chen, J. H., and Tilton, G. R. 1976, *Geochim. Cosmochim. Acta*, **40**, 635.
 Clayton, D. D. 1975, *Ap. J.*, **199**, 765.
 Clayton, R. N., Grossman, L., and Mayeda, T. K. 1973, *Science*, **182**, 484.
 Curtis, D. B., and Wasserburg, G. J. 1976, personal communication.
 Fish, R. A., Goles, G. G., and Anders, E. 1960, *Ap. J.*, **132**, 243.
 Gray, C. M., and Compston, W. 1974, *Nature*, **251**, 495.
 Gray, C. M., Papanastassiou, D. A., and Wasserburg, G. J. 1973, *Icarus*, **20**, 213.
 Lee, T., and Papanastassiou, D. A. 1974, *Geophys. Res. Letters*, **1**, 225.
 Lee, T., Papanastassiou, D. A., and Wasserburg, G. J. 1976a, *Geophys. Res. Letters*, **3**, 109 (LPW).
 ———. 1976b, *Trans. Am. Geophys. Union*, **57**, 278 (LPW).
 McCulloch, M. T., Papanastassiou, D. A., and Wasserburg, G. J. 1976, Abstract, 39th Annual Meeting of the Meteoritical Society.
 Reynolds, J. H. 1960, *Phys. Rev. Letters*, **4**, 8.
 Schramm, D. N. 1971, *Ap. Space Sci.*, **13**, 249.
 Schramm, D. N., Tera, F., and Wasserburg, G. J. 1970, *Earth Planet. Sci. Letters*, **10**, 44.
 Schramm, D. N., and Wasserburg, G. J. 1970, *Ap. J.*, **162**, 57.
 Urey, H. C. 1955, *Proc. Nat. Acad. Sci. U.S.*, **41**, 127.
 Wasserburg, G. J., Papanastassiou, D. A., and Lee, T. 1976, submitted to *Proc. Nat. Acad. Sci. U.S.*

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Note added in proof.—The production of ^{26}Al in the solar system would require the existence of isotopic anomalies in many other elements.