Is so-called Al-Mg isochron for a meteorite CAI a two component mixing line?

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An Al-Mg pseudo-isochron is expected for the type B inclusion (CAI) of carbonaceous chondrites on the basis of stoichiometry of anorthite (or hibonite). The linear correlation of excess ^{26}Mg with $^{27}\text{Al}/^{24}\text{Mg}$ ratio found for the Allende and Leoville CAI's is well-interpreted as a result of mixing in various proportions (<2%) of two end-members: one with no substantial excess of ^{26}Mg such as spinel, fassaite and melilite, and the other nearly pure anorthite with excess ^{26}Mg (possibly fossil ^{26}Mg) or its precursor. I would like to call attention to the difficulties in attaining to a "true" Al-Mg internal isochron for meteorite CAI's.

INTRODUCTION

Discovery of the excess ^{26}Mg by Gray and Compston (1974) and Lee and Papanastassiou (1974) and the demonstration of Al-Mg internal isochron by Lee *et al.* (1977) for the Allende CAI's were crucial contributions to the understanding of existence of extinct ^{26}Al in the early solar system materials. The presence of ^{27}Al -correlated ^{26}Mg excesses in meteorite CAI's was reconfirmed by the later works of other groups (Hutcheon *et al.* 1978; Stegmann and Begemann 1981; Lorin *et al.* 1977). Based on the live ^{26}Al model, these workers obtained almost the same initial $^{26}\text{Al}/^{27}\text{Al}$ ratio of $\sim 5 \times 10^{-5}$ for the type B CAI's of the Allende and Leoville (C3) meteorites. The live ^{26}Al abundance in the earliest solar system materials estimated from the ratio is considered to be enough to melt a small planetary body as well as larger planets (Lee *et al.* 1977), which substantiates the previous idea (Urey 1955; Fish *et al.* 1960).

Validity of the Al-Mg isochron is of basic importance for evaluation of live ^{26}Al in the early solar system. As discussed by Lee *et al.* (1977), if an Al-Mg isotopic equilibration was established at the time of CAI formation then

the linear correlation is eventually ascribed to *in situ* decay of ^{26}Al within the solar system. However, chemical and petrological works so far reached are not enough to clarify this problem, since the chemical and isotopic nature of CAI is so complex and has been still a controversial question.

In this article, I would like to point out a possibility that the linear Al-Mg correlation of the meteorite CAI which has been normally interpreted as a "true" isochron may represent more or less a two-component mixing line, and to emphasize importance of further, basic searches for the Al-Mg system of meteorite CAI's to establish its chronological meaning.

DISCUSSION

Isochron vs. two-component mixing line

Among many isotopic analyses of Al-Mg systems in the meteorite CAI's only three or four CAI's are known to have well-documented Al-Mg internal isochrons (Lee *et al.*, 1977; Hutcheon *et al.*, 1978; Stegmann and Begemann, 1981; Hutcheon and Steele, 1980; Huneke *et al.*, 1983). In any case the slope of isochron is defined solely by the data points of anorthites with different amounts of excess ^{26}Mg ($\delta^{26}\text{Mg}$).

The anorthites analyzed are reported to be "quite pure". Hence, in some cases, the Al concentrations in the mineral were calculated stoichiometrically via Ca determined by isotope dilution (Lee *et al.*, 1977). The nearly stoichiometric concentration of Al in anorthite grains of CAI is rather clear for three CAI's under consideration. For example, deviations of the total chemical compositions within a large anorthite grain of the well-known Allende CAI WA from the stoichiometry ($\text{CaAl}_2\text{Si}_2\text{O}_8$) are only 1% or less for four areas examined by electron probe (and ion probe) micro-analyses, although large variations are observed for Mg content (240-570ppm) (Bradley *et al.*, 1978). Similar features are also reported for other two CAI's (Hutcheon *et al.*, 1978; Stegmann and Begemann, 1981). Such high anorthitic nature is considered to be general for the plagioclase of Allende CAI (Grossman, 1975). A highly pure mineral chemistry has been also found for a hibonite grain from the Dhajala ordinary chondrite for which a well defined Al-Mg isochron was obtained (Horton and Bischoff, 1984).

Therefore, the ^{27}Al concentrations in anorthite or hibonite grains analyzed for Al-Mg isochron plot are considered to be almost stoichiometric and thus approximated to be constant (k) normally within 3% (possibly 5-10% in some cases (Stegmann and Begeman., 1981)); $(^{27}\text{Al}) = 2/(\text{CaAl}_2\text{Si}_2\text{O}_8) = 7.2 \times 10^{-3} \text{ mol/g}$ for plagioclase and $(^{27}\text{Al}) = 12/(\text{CaAl}_{12}\text{O}_{19}) = 1.8 \times 10^{-2} \text{ mol/g}$ for hibonite. If so, the so-called isochron relationship can be converted to a two-component mixing equation as follows, which is in commonest use for geochronologists (Faure, 1977) to test a validity of isochron:

$$^{26}\text{Mg}/^{24}\text{Mg} = a(^{27}\text{Al}/^{24}\text{Mg}) + b$$

Isochron relationship
(a = slope, b = intercept)

$$= a k / (^{24}\text{Mg}) + b$$

Two-component mixing relationship

Thus it turned out that regardless of validity of

isochron the linear Al-Mg correlation obtained for one Al-rich (stoichiometric) mineral and other Mg-rich phases does potentially represent a two-component mixing line.

Therefore, the data points of anorthites or hibonite in an isochron are explicable by relatively small additions of Mg-rich phases into the nearly pure anorthite or hibonite with excess ^{26}Mg component. For example, let us consider spinel (Mg=17wt%) to be Mg-rich phase. In Fig. 1, a minor addition of spinel (e.g., 0.5% mixing of spinel (Sp) in Fig. 1) yields extremely large variations in $^{27}\text{Al}/^{24}\text{Mg}$ ratio which substantially cover the whole excess ^{26}Mg data so

Fig. 1. Two-component mixing diagram for the Al-Mg system of meteorite CAI. The solid line is calculated on the assumption of mixing of varying proportions of spinel with no excess ^{26}Mg with anorthite with excess ^{26}Mg component: $ak = 5.1 \times 10^{-5} (^{27}\text{Al}) = 3.7 \times 10^{-7} \text{ mol g}^{-1}$. Note that variations of excess ^{26}Mg observed for many meteorite CAI's, typically the Allende CAI WA (Lee *et al.*, 1977), are explained by minor addition (less than 2%) of spinel to the anorthite. Symbols Sp and An represent spinel and anorthite, respectively.

far observed for the meteorite CAI's: data points of anorthites in WA isochron obtained by Lee *et al.* (Fig. 1) (Lee *et al.*, 1977) are explained by addition of 0.5-1% spinel to the anorthite. In the same way the other CAI isochrons (Hutcheon *et al.*, 1977; Stegmann and Begemann, 1981; Hutcheon and Steele, 1980; Huneke *et al.*, 1983) are also explained by addition of mostly less than 0.5% (up to 1.8%) spinel.

The linear Al-Mg correlations for meteorite CAI's have been interpreted normally by isochron model (Lee *et al.*, 1977; Hutcheon *et al.*, 1977; Stegmann and Begemann, 1981) or the *ad hoc* mixing model (Clayton, 1977a). These workers, however, seem to have dismissed the stoichiometric view as expressed above.

Was Al-Mg system really in equilibrium at the time of CAI formation?

Because anorthite and hibonite have no crystal site for Mg, Mg in the minerals may exist as micro or submicron Mg-rich inclusions dispersed within anorthite grains like "Fremdlinge" (Goresy *et al.*, 1978), or possibly in lattice defects or deformed crystal sites. In this respect, detailed searches for excess ^{26}Mg as well as normal Mg by Bradley *et al.* (1978), Hutcheon *et al.* (1978) and Huneke *et al.* (1983) are valuable. They found heterogeneous and large variations of normal Mg (300 - 1,000 ppm) and homogeneous distributions of excess ^{26}Mg components within anorthite grains. They detected no particular excess ^{26}Mg carriers larger than several μm size in anorthite such as Al_2O_3 -grains as suggested by Clayton (1977a), and thus the results appeared to be consistent with the ^{26}Al - ^{26}Mg isochron model. However, it is pointed out that this situation is quite similar to that of oxygen isotopic anomaly of meteorite CAI's for which two-components mixing relationship has been well-established by Clayton group; Clayton *et al.* (1977) had also failed to identify any ^{16}O -carried phase in CAI's.

It is thus possible that the large variations of the $^{27}\text{Al}/^{24}\text{Mg}$ ratios of anorthite are results

of the heterogeneous distributions of Mg-inclusions within the mineral grains. The question then arises whether the Mg-inclusions were co-genetic with anorthite from a same reservoir and the normal Mg isotopes were in equilibrium with Al? In this connection, it is interesting that some type B CAI's (B1) have the clear Al-Mg correlation but the B2's exhibit larger dispersions from the isochron with somewhat lower slope (Hutcheon and Steele 1980). Similar trend of the Al-Mg system has been found even for a single B1 CAI, the well-known Allende CAI WA (Huneke *et al.*, 1983).

From these Al-Mg isotopic data, as well as many other chemical and isotopic characteristics of CAI's so far reported, it is suggested that the majority of Al-Mg systems of meteorite CAI's were more or less lacking in complete isotopic equilibration at the time of formation or subjected to post-formation disturbances (Meeker *et al.*, 1983). As argued by Wood (1981), discovery of heterogeneous distributions of isotopes and elements in CAI's and their unusual petrologic features have led cosmochemists to believe that precursor materials of CAI's were formed in different stellar environments and then mixed into the presolar system interstellar medium and the independently-formed dust grains had been subjected to reheating, melting and/or distillation, back-reaction with the distilled-off vapor or recondensates and so on, which might have led to metamorphic to igneous origin of CAI's. It is thus possible that some type B CAI's were formed from partial to total melts (Kurat *et al.*, 1975; Blander and Fuchs 1975; Wark *et al.*, 1982) or by sub-solidus sintering of pre-existed dust grains, and therefore some CAI's (Nagasawa *et al.*, 1977) might have attained chemical equilibrium but some others were in incomplete equilibration (Palme and Wlotzka 1979; Nagasawa *et al.*, 1982), depending on their formation conditions.

It should be noted that as far as we know there is neither clear evidence that the Al-Mg isotopic equilibration was established at the time of CAI formation nor other experimental base such as diffusion data applicable to the CAI

which might constrain the basic criteria of the Al-Mg isochron.

Therefore, the meaning of the age or the initial $^{26}\text{Al}/^{27}\text{Al}$ ratio deduced from these isochrons is solely model dependent; for example, the age represents the stellar time scale (Clayton, 1977b) or the early solar system time scale (Lee *et al.*, 1977).

CONCLUSION

Because of stoichiometry of Al-rich minerals the reported Al-Mg internal isochrons for meteorite CAI's (Lee *et al.*, 1977; Hutcheon *et al.*, 1978; Stegmann and Begemann, 1981; Hutcheon and Steele, 1980; Hunkeke *et al.*, 1983; Hinton and Bischoff, 1984) are well-interpreted as two-component mixing lines between Mg-rich phase such as spinel, fassaite and melilite, and nearly pure anorthite or hibonite with fossil ^{26}Al . Therefore, a live ^{26}Al model for the time scale and heat source of planetary bodies in the early solar system are still uncertain. In order to clarify the validity of so-called Al-Mg isochron for a meteorite CAI, more detailed searches for distributions of Mg and Al isotopes (possibly submicron order examinations) would be required.

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REFERENCES

- Blander, M. and Fuchs, L. H. (1975) Calcium-aluminum-rich inclusions in the Allende meteorite: evidence for a liquid origin. *Geochim. Cosmochim. Acta* **39**, 1605–1619.
- Bradley, J. G., Huneke, J. C. and Wasserburg, G. J. (1978) Ion microprobe evidence for the presence of excess ^{26}Mg in an Allende anorthite crystal. *J. Geophys. Res.* **83**, 244–254.
- Clayton, D. D. (1977a) Cosmoradiogenic ghost and the origin of Ca-Al-rich inclusions. *Earth Planet. Sci. Lett.* **35**, 433–454.
- Clayton, D. D. (1977b) Solar system isotopic anomalies: Supernova neighbor or presolar carriers? *Icarus* **32**, 244–269.
- Clayton, R. N., Onuma, N., Grossman, L. and Mayeda, T. K. (1977) Distribution of the presolar component in Allende and other carbonaceous chondrites. *Earth Planet. Sci. Lett.* **34**, 209–224.
- Faure, G. (1977) *Principles of isotope geology*, 97–106. John Wiley & Sons: New York.
- Fish, R. A., Goles, G. G. and Anders, E. (1960) The record in the meteorites. III. On the development of meteorites in asteroidal bodies. *Ap. J.* **132**, 243–258.
- Goresy, A. El., Nagel, K. and Ramdohr, P. (1978) Fremdlinge and their noble relatives. *Proc. Lunar Sci. Conf.* **9th**, 1279–1303.
- Gray, C. M. and Compston, W. (1974) Excess ^{26}Mg in the Allende meteorite. *Nature* **251**, 495–497.
- Grossman, L. (1975) Petrology and mineral chemistry of Ca-rich inclusions in the Allende meteorite. *Geochim. Cosmochim. Acta* **39**, 433–454.
- Hinton, R. W. and Bischoff, A. (1984) Ion microprobe magnesium isotope analysis of plagioclase and hibonite from ordinary chondrites. *Nature* **308**, 169–172.
- Huneke, J. C., Armstrong, J. T. and Wasserburg, G. J. (1983) FUN with PANURGE: High mass resolution ion microprobe measurements of Mg in Allende inclusions. *Geochim. Cosmochim. Acta* **47**, 1635–1650.
- Hutcheon, I. D., Steele, I. M., Smith, J. V. and Clayton, R. N. (1978) Ion microprobe, electron microprobe and cathodoluminescence data for Allende inclusions with emphasis on plagioclase chemistry. *Proc. Lunar Planet. Sci. Conf.* **9th**, 1345–1368.
- Hutcheon, I. D. and Steele, I. M. (1980) Mineralogy and Mg isotopic composition of type B2 inclusions from Leoville and Allende. *Lunar Planet. Sci.* **XI**, 496–498.
- Kurat, G., Hoinkes, G. and Fredriksson, K. (1975) Zoned Ca-Al-rich chondrule in Bali: new evidence against the primordial condensation model. *Earth Planet. Sci. Lett.* **26**, 140–144.
- Lee, T. and Papanastassiou, D. A. (1974) Mg isotopic anomalies in the Allende meteorite and correlation with O and Sr effects. *Geophys. Res. Lett.* **1**, 225–228.
- Lee, T., Papanastassiou, D. A. and Wasserburg, G. J. (1977) ^{26}Al with early solar system: Fossil or fuel? *Astrophys. J.* **211**, L107–L110.
- Lorin, J. -C., Shimizu, N., Christophe Michel-Levy, M. and Allègre, C. (1977) The Mg isotope anomaly

- in carbonaceous chondrites: An ion-probe study. *Meteoritics* **12**, 299–300.
- Meeker, G. P., Wasserburg, G. J. and Armstrong, J. T. (1983) Replacement textures in CAI and implications regarding planetary metamorphism. *Geochim. Cosmochim. Acta* **47**, 707–721.
- Nagasawa, H., Blanchard, D. P., Jacobs, J. W., Brannon, J. W., Philpotts, J. A. and Onuma, N. (1977) Trace element distribution in mineral separates of the Allende inclusions and their genetic implications. *Geochim. Cosmochim. Acta* **41**, 1587–1600.
- Nagasawa, H., Blanchard, D. P., Shimizu, H. and Masuda, A. (1982) Trace element concentrations in the isotopically unique Allende inclusion, EK1-4-1. *Geochim. Cosmochim. Acta* **46**, 1669–1673.
- Palme, H. and Wlotzka, F. (1979) A Ca, Al-rich inclusion from the Leoville (C3V) meteorite. *Meteoritics* **14**, 508–511.
- Stegmann, W. and Begemann, F. (1981) Al-correlated ^{26}Mg excess in a large Ca-Al-rich inclusion of the Leoville meteorite. *Earth Planet. Sci. Lett.* **55**, 266–272.
- Urey, H. C. (1955) The cosmic abundances of potassium, uranium, and the heat balance of the earth, the moon, and mars. *Proc. Nat. Acad. Sci. U.S.* **41**, 127–144.
- Wark, D. A. and Lovering, J. F. (1982) The nature and origin of type B1 and B2 Ca-Al-rich inclusions in the Allende meteorite. *Geochim. Cosmochim. Acta* **46**, 2581–2594.
- Wood, J. A. (1981) The interstellar dust as a precursor of Ca, Al-rich inclusions in carbonaceous chondrites. *Earth Planet. Sci. Lett.* **56**, 32–44.