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Aluminum-26 in H4 chondrites: Implications for its production and its usefulness as a fine-scale chronometer for early solar system events

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Abstract–In order to investigate whether or not ^{26}Al can be used as a fine-scale chronometer for early solar system events we measured, with an ion microprobe, Mg isotopes and Al/Mg ratios in separated plagioclase, olivine, and pyroxene crystals from the H4 chondrites Ste Marguerite (SM), Forest Vale (FV), Beaver Creek and Quenggouk and compared the results with the canonical ^{26}Al/^{27}Al ratio for calcium-aluminum-rich inclusions (CAIs). For SM and FV, Pb/Pb and Mn-Cr ages have previously been determined (Göpel *et al.*, 1994; Polnau *et al.*, 2000; Polnau and Lugmair, 2001). Plagioclase grains from these two meteorites show clear excesses of ^{26}Mg. The ^{26}Al/^{27}Al ratios inferred from these excesses and from isotopically normal Mg in pyroxene and olivine are $(2.87 \pm 0.64) \times 10^{-7}$ for SM and $(1.52 \pm 0.52) \times 10^{-7}$ for FV. The differences between these ratios and the ratio of 5×10^{-5} in CAIs indicate time differences of 5.4 ± 0.1 Ma and 6.1 ± 0.2 Ma for SM and FV, respectively. These differences are in agreement with the absolute Pb/Pb ages for CAIs and SM and FV phosphates but there are large discrepancies between the U-Pb and Mn-Cr system for the relative ages for CAIs, SM and FV. For example, Mn-Cr ages of carbonates from Kaidun are older than the Pb/Pb age of CAIs. However, even if we require that CAIs are older than these carbonates, the time difference between this "adjusted" CAI age and the Mn-Cr ages of SM and FV require that ^{26}Al was widely distributed in the early solar system at the time of CAI formation and was not mostly present in CAIs, a feature of the X-wind model proposed by Shu and collaborators (Gounelle *et al.*, 2001; Shu *et al.*, 2001). From this we conclude that there was enough ^{26}Al to melt small planetary bodies as long as they formed within 2 Ma of CAIs, and that ^{26}Al can serve as a fine-scale chronometer for early solar system events.

INTRODUCTION

Evidence for the presence of the short-lived ($t_{1/2} = 7.3 \times 10^5$ years) nuclide ^{26}Al in early solar system solids was established in 1974 by measurements of excesses of its daughter isotope ^{26}Mg in refractory calcium-aluminum-rich inclusions (CAIs) (Gray and Compston, 1974; Lee and Papanastassiou, 1974; see also Lee *et al.*, 1976, 1977). Since then a wealth of data, mostly from ion microprobe measurements, have been obtained (MacPherson *et al.*, 1995). The vast majority of these are from CAIs but, mostly recently, measurements have also been made on chondrules and other materials (Hutcheon and Hutchison, 1989; Hutcheon and Jones, 1995; Hutcheon *et al.*, 1994, 2000; Russell *et al.*, 1996; Srinivasan *et al.*, 1999, 2000a,b; Kita *et al.*, 2000; Marhas *et al.*, 2000; Huss *et al.*, 2001; Mostefaoui *et al.*, 2002).

The presence of ^{26}Al in the early solar system gives rise to two important questions: (1) Can ^{26}Al be used as a fine-scale chronometer for early system events? (2) Can it serve as a heat source for the melting of asteroidal bodies? (Urey, 1955; Fish *et al.*, 1960). A positive answer to the first question requires a uniform distribution of ^{26}Al in the solar system. If this condition is met then the ^{26}Al/^{27}Al ratio of 5×10^{-5} measured in CAIs implies that enough ^{26}Al was present for melting as long as (1) planetesimals formed relatively early (within 2 Ma of the formation of CAIs), (2) had on average approximately chondritic composition, and (3) had a certain minimum size (Schramm *et al.*, 1970).

The majority consensus of scientists concerned with these problems seems to have been in favor of a positive answer to the two questions posed above (see, for example, Podosek *et al.*, 1991; Caillet *et al.*, 1993; MacPherson *et al.*, 1995; Hsu *et al.*, 2000), with the assumption that ^{26}Al was produced by a stellar source and was injected into the solar system at its birth (Cameron and Truran, 1977; Cameron, 1984; Wasserburg *et al.*, 1994).

However, lately this consensus has been challenged by the X-wind model put forward by Shu and collaborators (Shu *et* *al.*, 1996, 1997, 2001). According to this model not only is ^{26}Al (and other short-lived isotopes) produced by local irradiation in the X-wind region of the early Sun but CAIs and chondrules themselves are produced in this region (Lee *et al.*, 1998; Gounelle *et al.*, 2001; Shu *et al.*, 2001). Furthermore, only CAIs are produced with the high ^{26}Al/^{27}Al ratio of 5×10^{-5}, whereas chondrules have much smaller ratios. The consequences of this model are that there is not enough ^{26}Al for the melting of small asteroids and, because ^{26}Al is not distributed uniformly, it cannot serve as a fine-scale chronometer.

Although various objections have been raised against the irradiation production of the short-lived isotopes for which evidence has been found in early solar system objects (Goswami and Vanhala, 2000; Goswami *et al.*, 2001), the recent discovery of initial ^{10}Be, a radionuclide that can only be produced by energetic particle irradiation and not by stellar nucleosynthesis, in CAIs (McKeegan *et al.*, 2000, 2001; MacPherson and Huss, 2001; Sugiura *et al.*, 2001) appears to strengthen the Shu model. However, the fact that ^{10}Be apparently can be produced in supernova jets (Cameron, 2002) should add a little caution.

A way to test whether ^{26}Al can be used as a chronometer is to measure the Al-Mg system in objects for which age information can be obtained from other, independent, chronometers. CAIs and samples from H4 chondrites provide a good opportunity for such a comparison because Pb/Pb ages for both are available (Chen and Wasserburg, 1981; Manhès *et al.*, 1988; Göpel *et al.*, 1994; Amelin *et al.*, 2002). The U-Pb system is the only absolute chronometer with a time resolution (<1 Ma) that is useful for comparison with the results of age dating based on ^{26}Al and other short-lived isotopes such as ^{53}Mn. There is evidence that the H4 chondrites studied here experienced fast cooling (Pellas and Storzer, 1981; Lipschutz *et al.*, 1989; Göpel *et al.*, 1994). Recently, chronometric information on two H4 chondrites, Ste Marguerite and Forest Vale, has been obtained from the Mn-Cr system (Polnau *et al.*, 2000; Polnau and Lugmair, 2001). What makes such an investigation possible is the fact that H4 chondrites contain plagioclase grains with extremely high Al/Mg ratios. We thus undertook an ion microprobe study of the Al-Mg system in plagioclase crystals from H4 chondrites. Preliminary results have been reported by Zinner and Göpel (1992).

In order to avoid confusion, we note that there are many different aspects of the X-wind model. In this paper we do not attempt to address all of them but concentrate on the conclusion that only CAIs received an amount of ^{26}Al corresponding to a ^{26}Al/^{27}Al ratio of 5×10^{-5} and that all other solar system materials received much less ^{26}Al, a conclusion reached by the proponents of the X-wind model (Gounelle *et al.*, 2001; Shu *et al.*, 2001).

EXPERIMENTAL

We measured plagioclase, olivine and pyroxene grains from the H4 chondrites Ste Marguerite (SM), Forest Vale (FV), Beaver Creek and Quenggouk. Feldspar in H4 chondrites is quite rare and the grains we analyzed were obtained from mineral separates. The feldspar separates of SM, FV and Quenggouk were obtained as a byproduct of the mineral separation of phosphates. Twenty to 50 g of chondrite starting material was ground in a boron carbide mortar until the grain size of the main fraction was between 37 and 74 μm. After eliminating the metal phase by a hand magnet the sieved meteorite powder >37 μm was passed several times through a Frantz magnetic separator where phosphates, feldspar and impurities (small chips from the boron carbide mortar) pass through undeflected and are found in the >1.6 A fraction. Phosphates were eliminated with bromoform and feldspars were then handpicked under a binocular microscope.

Two mineral fractions could be distinguished: (a) clear transparent crystals and (b) white milky crystals. Later analysis revealed that the transparent clear grains were plagioclase crystals while the milky grains were feldspathic glass or feldspars with tiny Mg-rich inclusions. Additional separates from FV and Beaver Creek were provided by Paul Pellas. Individual grains of feldspar, but also of pyroxene and olivine, were mounted in epoxy and polished for ion probe analysis. Some of the feldspar grains were as large as 150 μm but most of the measured grains were ~50 μm in size. One of them is shown in Fig. 1.

Energy dispersive x-ray (EDX) data were obtained on selected plagioclase grains after ion probe analysis. The clear crystals have fairly albitic compositions. Na_2O contents range from 2.3 to 9.5 wt% (most ~8), CaO from 4.6 to 16.1 wt% and K_2O from 0.1 to 0.6 wt%. Thus the range of the analyzed plagioclase grains is An_{15-80}. However, the An_{80} represents only one grain, the anorthite content of all the others was 55% or below. Mg contents are extremely low, which posed quite a challenge for the ion probe analyses (see below). The milky grains of glass have higher Mg concentrations but were not analyzed in the ion probe.

Ion probe measurements, employing techniques described before (McKeegan *et al.*, 1985; Fahey *et al.*, 1987), were made at a mass resolving power of ~3000, sufficient to eliminate all isobaric interferences ($^{48}Ca^{++}$, Mg hydride). They were made in a peak-jumping mode, with the Mg isotopes being measured with an electron multiplier and the Al^+ signal with a Faraday cup. With primary beam currents of 1.7–4.8 nA $^{27}Al^+$ count rates ranged up to 10^7 counts (cts)/s. One run typically consisted of 50 blocks of six cycles each (1, 5, 5 s/cycle for the Mg isotopes) and a run lasted more than 2 h. Several such runs were made on each spot so that a single measurement lasted as long as 10 h. Magnesium isotopic ratios were corrected for mass fractionation determined from the ^{25}Mg/^{24}Mg ratio. Al/Mg ratios were obtained from the ion ratios and the sensitivity factor measured on an anorthite standard (Miakejima plagioclase, provided by Ian Hutcheon). The detection efficiency of the electron multiplier for Mg ions was calibrated against the Faraday cup at count rates of ~10^6 cts/s.

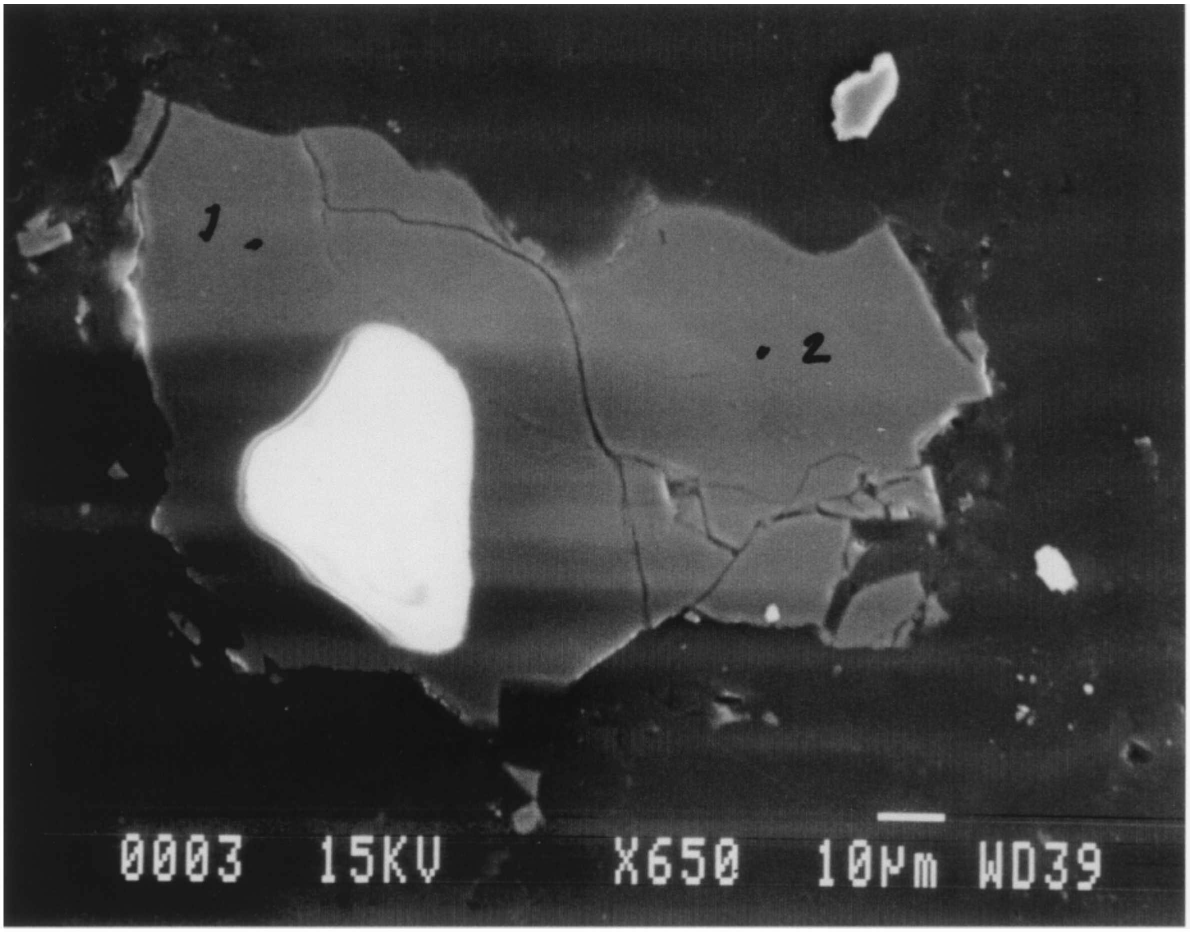

FIG. 1. Scanning electron micrograph of a polished plagioclase grain from Ste Marguerite. The large white spot is the area where Mg isotopes and Al were analyzed in the ion microprobe. The ^{27}Al/^{24}Mg ratio in this spot is 4600. The two numbered dots indicate spots where EDX analyses were made.

RESULTS

We measured twelve individual plagioclase grains from SM, three from FV, four from Beaver Creek and two from Quenggouk. Along with these plagioclase grains we analyzed two olivine and three pyroxene grains from SM, three olivines and one pyroxene from FV, and four olivines from Quenggouk. The results are given in Table 1 and plotted in Fig. 2. In SM and FV we observe clear ^{26}Mg excesses in the plagioclase crystals, in Beaver Creek this excess is marginal, and we cannot claim a definite ^{26}Mg excess for Quenggouk plagioclase.

Figure 2 shows also the least-square lines fitted to the plagioclase, olivine and pyroxene data points. For Beaver Creek, where we did not measure any olivines and pyroxenes, we used the combined olivine + pyroxene data for SM and FV. For Quenggouk no line fit has been attempted. The slopes of these lines, which are given with 2σ errors in the plots, represent the inferred initial ^{26}Al/^{27}Al ratio at the time of closure of the Al-Mg system. For the three meteorites SM, FV, and Beaver Creek, within the analytical errors, the data points are consistent with a single evolution line in each case; the reduced χ^2 of the fits is 1.0, 1.3, and 0.9, respectively, for these three meteorites. The inferred ^{26}Al/^{27}Al ratios are $(2.87 \pm 0.64) \times 10^{-7}$, $(1.52 \pm 0.52) \times 10^{-7}$, and $(1.09 \pm 1.26) \times 10^{-7}$. However, for Beaver Creek the slope of the fitted line agrees with zero within 2σ errors. For FV the ^{26}Mg/^{24}Mg ratios measured in three plagioclase crystals are not correlated with the ^{27}Al/^{24}Mg ratios. However, this could be just the consequence of the measurement errors since, within these errors, the data points are consistent with a single evolution line. Recent analysis of many spots within a large single plagioclase crystal from FV by Zinner *et al.* (2002) yielded a good correlation and a slope of 1.56×10^{-7}, within the errors identical to the slope obtained here. Since the data points in the plots of Fig. 2 were obtained from different individual crystals, this means that the time of closure was the same for different plagioclase grains from a given meteorite.

DISCUSSION

If we interpret the differences between the "canonical" initial ^{26}Al/^{27}Al ratio of 5×10^{-5} for CAIs and the ratios

TABLE 1. Al/Mg ratios and Mg isotopic ratios measured in individual grains from four H4 chondrites.

Meteorite/ mineral	^{27}Al/^{24}Mg ($\pm 2\sigma$)	^{26}Mg/^{24}Mg ($\pm 2\sigma$)
Ste. Marguerite		
Plagioclase	4009 ± 554	0.14122 ± 0.00148
	3618 ± 370	0.14034 ± 0.00070
	3992 ± 790	0.14031 ± 0.00036
	4576 ± 1178	0.14031 ± 0.00068
	5279 ± 448	0.14089 ± 0.00080
	1286 ± 58	0.13963 ± 0.00051
	3019 ± 375	0.14092 ± 0.00068
	3815 ± 1128	0.14111 ± 0.00074
	3914 ± 165	0.14078 ± 0.00088
	2455 ± 184	0.14022 ± 0.00038
	3244 ± 293	0.14041 ± 0.00046
	3169 ± 339	0.14033 ± 0.00074
Pyroxene + Olivine*	0.17 ± 0.42	0.13942 ± 0.00014
Forest Vale		
Plagioclase	10 199 ± 282	0.14075 ± 0.00076
	8438 ± 844	0.14064 ± 0.00062
	6060 ± 678	0.14081 ± 0.00060
Pyroxene + Olivine*	0.29 ± 0.10	0.13945 ± 0.00020
Beaver Creek		
Plagioclase	2769 ± 13	0.13930 ± 0.00073
	1977 ± 135	0.13983 ± 0.00067
	2461 ± 246	0.13971 ± 0.00032
	2515 ± 74	0.13994 ± 0.00050
Quenggouk		
Plagioclase	5771 ± 1674	0.13905 ± 0.00054
	6711 ± 1129	0.13989 ± 0.00043
Olivine*	5.07 ± 1.24	0.13933 ± 0.00018

*Averages of measurements on several grains.

determined for the different H4 chondrites of the present study as being due to a time difference in formation/closure we obtain for the Al-Mg ages of these meteorites relative to CAIs the following values: SM, 5.43 ± 0.12 Ma; FV, 6.10 ± 0.18 Ma; Beaver Creek, 6.45$^{+0.91}_{-0.48}$Ma. For the first two meteorites we can compare these relative ages with the absolute Pb/Pb ages of 4.566 ± 0.002 Ga (Chen and Wasserburg, 1981; Allègre *et al.*, 1995; Manhès *et al.*, 1988; Amelin *et al.*, 2002) measured in CAIs, and 4.5627 ± 0.0006 Ga and 4.5609 ± 0.0007 Ga measured in phosphates of SM and FV (Göpel *et al.*, 1994). These values translate into time differences relative to CAIs of 3.3 ± 2.1 and 5.1 ± 2.1 Ma for SM and FV, respectively.

This relationship is shown in Fig. 3 where we plot the initial ^{26}Al/^{27}Al ratios against the Pb/Pb ages of CAIs and the H4 meteorites SM and FV (lower scale of the *x*-axis). The bold line with the arrow depicts the evolution of the ^{26}Al/^{27}Al ratio with time if it is assumed that this ratio was 5×10^{-5} at 4.566 Ga B.P., the Pb/Pb age of CAIs. From this line and the ^{26}Al/^{27}Al ratios measured in SM and FV, relative Al-Mg ages are obtained, which are indicated on the upper scale of the *x*-axis. From this plot it is clear that, within the analytical errors, the age differences between CAIs and the H4 chondrites obtained from the Pb/Pb and the Al-Mg measurements agree with one another. As can be seen, the largest uncertainty stems from the Pb/Pb age of CAIs and the two broken lines in the figure indicate the effect of this uncertainty on the evolution of the ^{26}Al/^{27}Al ratio. Thus on the surface it appears that we have obtained a positive answer to our original question whether ^{26}Al can be used as a fine-scale chronometer. However, there are several other questions that have to be discussed before we can be certain of this answer.

Are the Plagioclase Measured of Metamorphic Origin?

There is little doubt that phosphate in H4 chondrites is of metamorphic origin (Göpel *et al.*, 1994). Our chronological interpretation of the Al-Mg data obtained from the H4 plagioclase grains assumes that these grains are also of metamorphic origin (*i.e.*, they are a secondary mineral), formed as modification of some other, preexisting, phases during the metamorphic cooling of the H4 parent body with complete Mg homogenization. An alternative, considered by MacPherson *et al.* (1995), is that the grains have a primary (igneous) origin and are relics from Al-rich chondrules. Unfortunately, the grains of this study were obtained as isolated grains from mineral separates and we do not have any information about their petrographic context in the meteorite.

Therefore, in order to better constrain the origin of these isolated crystals, we petrographically studied thin sections that were made from small bulk fragments of SM selected during the separation procedure. Our purpose was to correlate the petrographic observations (grain size, crystal shape) and chemical composition of the feldspars observed *in situ* with the information on separated grains that were studied with the ion probe.

Feldspar is a relatively rare mineral phase in grade 4 chondrites. Three principal types of occurrences can be distinguished:

(1) Metamorphic Feldspar–Mesostasis and glass crystallize during metamorphism and are transformed into plagioclase. The correlation of grain size and abundance of feldspar with the metamorphic grade of its host meteorite support this interpretation. The major element composition of these newly formed feldspars, which are best documented in type 6 chondrites, is albitic and ranges from Ab$_{82}$An$_{12}$Or$_{6}$ to Ab$_{84}$An$_{10}$Or$_{6}$ (Van Schmus and Ribbe, 1968).

(2) Feldpars in Aluminum-Rich Objects–Al-rich objects are widespread constituents of ordinary chondrites. Their petrography is described in more detail by Bischoff and Keil (1984). These authors define different subtypes and report

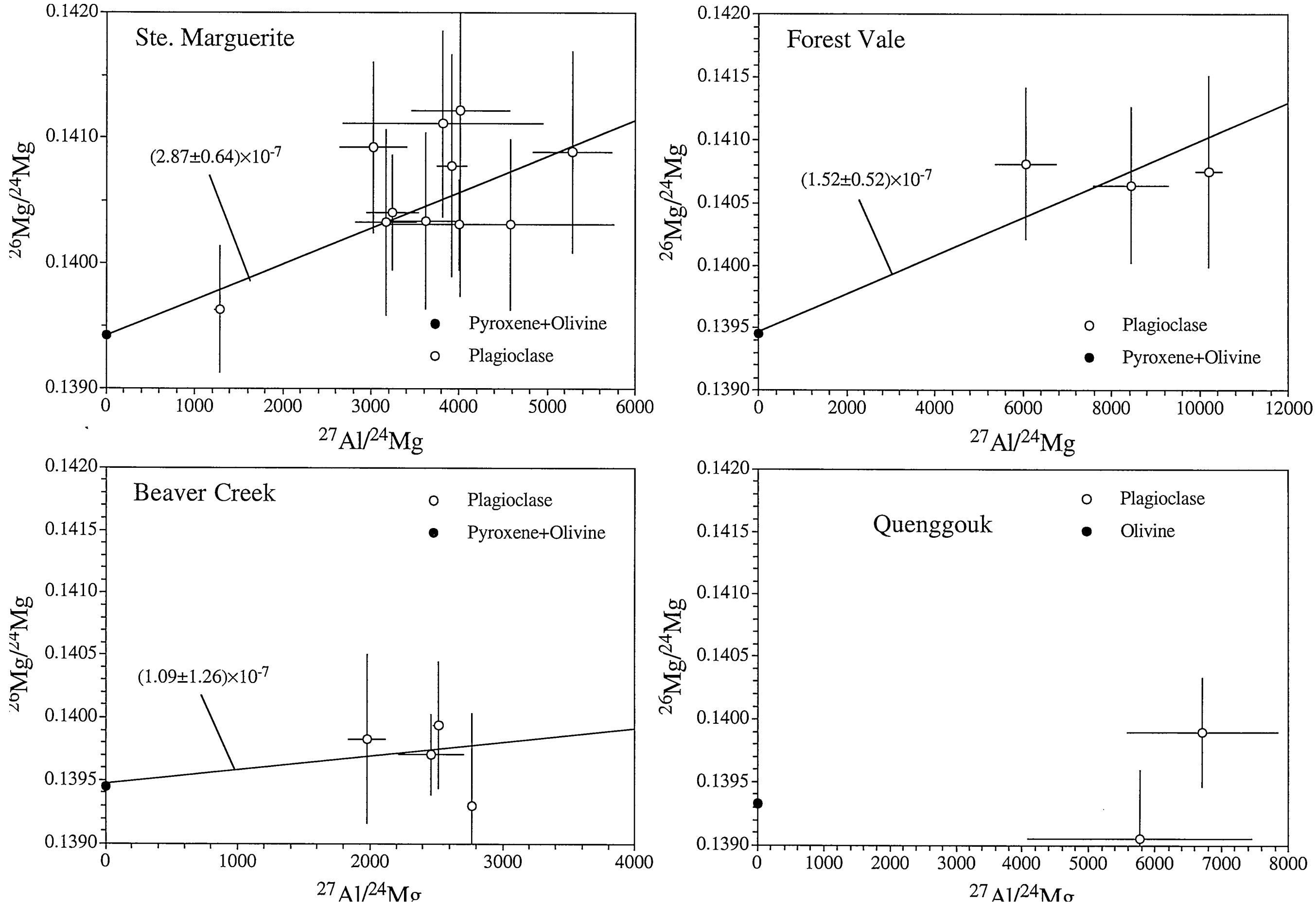

FIG. 2. Al-Mg plots of plagioclase, olivine and pyroxene grains from four H4 chondrites. The lines for three of them are least-square fits to the data points. The slopes of these lines represent inferred initial ^{26}Al/^{27}Al ratios. Errors of the data points and of the ^{26}Al/^{27}Al ratios are 2σ as they are in all subsequent figures.

that the major element composition is variable, ranging from albitic compositions in Na-rich inclusions to fairly anorthitic compositions in refractory CAIs.

(3) Feldspar in Chromium-Rich Chondrules and Inclusions–Ramdohr (1967) and more recently Krot *et al.* (1993) and Christophe Michel-Levy *et al.* (1995) studied Al-Cr-rich objects, chondrules and inclusions in more detail. The feldspars associated with these chromitic inclusions have an albitic composition (Ab_{43-88}).

The separation procedure yielding only a few crystals extracted from several tens of grams of material as well as the scarcity of the petrographic observations (seven feldspar occurrences in four thin sections) confirm that feldspar is a rare mineral and heterogeneously distributed in type H4 chondrites. However, even without petrographic information on the isolated crystals of this study we can make some conclusions about their origin. First, with the exception of one An_{80} grain, the anorthite content of the plagioclase crystals of this study is much lower than that of any plagioclase in chondrules from unequilibrated ordinary chondrites analyzed for ^{26}Mg excesses (see Table 2 of Huss *et al.*, 2001). The same observation holds for the Mg concentrations. ^{27}Al/^{24}Mg ratios in anorthite in CAIs range up to 1000; in plagioclase from chondrules they are usually much smaller (Russell *et al.*, 1996; Hutcheon *et al.*, 2000; Kita *et al.*, 2000; Marhas *et al.*, 2000; Srinivasan *et al.*, 2000a; Huss *et al.*, 2001; Mostefaoui *et al.*, 2002). In contrast, the ^{27}Al/^{24}Al ratios in the plagioclase grains of this study range up to 5300 for SM, 10 200 for FV and 6700 for Quenggouk (Table 1 and Fig. 2). These are ratios comparable to those seen in plagioclase from the eucrite Piplia Kalan (Srinivasan *et al.*, 1999). Both, the high Na_2O content and the extremely low Mg content of the plagioclase from H4 chondrites indicate a metamorphic origin. Lacking petrographic information, we do not know the nature of the precursor phase(s) of the plagioclase, but it could have been Na- and Al-rich glass as found in chondrules from various meteorites. The Al/Mg ratios of this glass are usually fairly low (see, for example, Table 2 of Huss *et al.*, 2001) and it apparently required long exposure to elevated temperatures during the metamorphic process to remove most of the Mg. Plagioclase does not readily accommodate Mg in its structure but it would take high temperature to mobilize it enough to

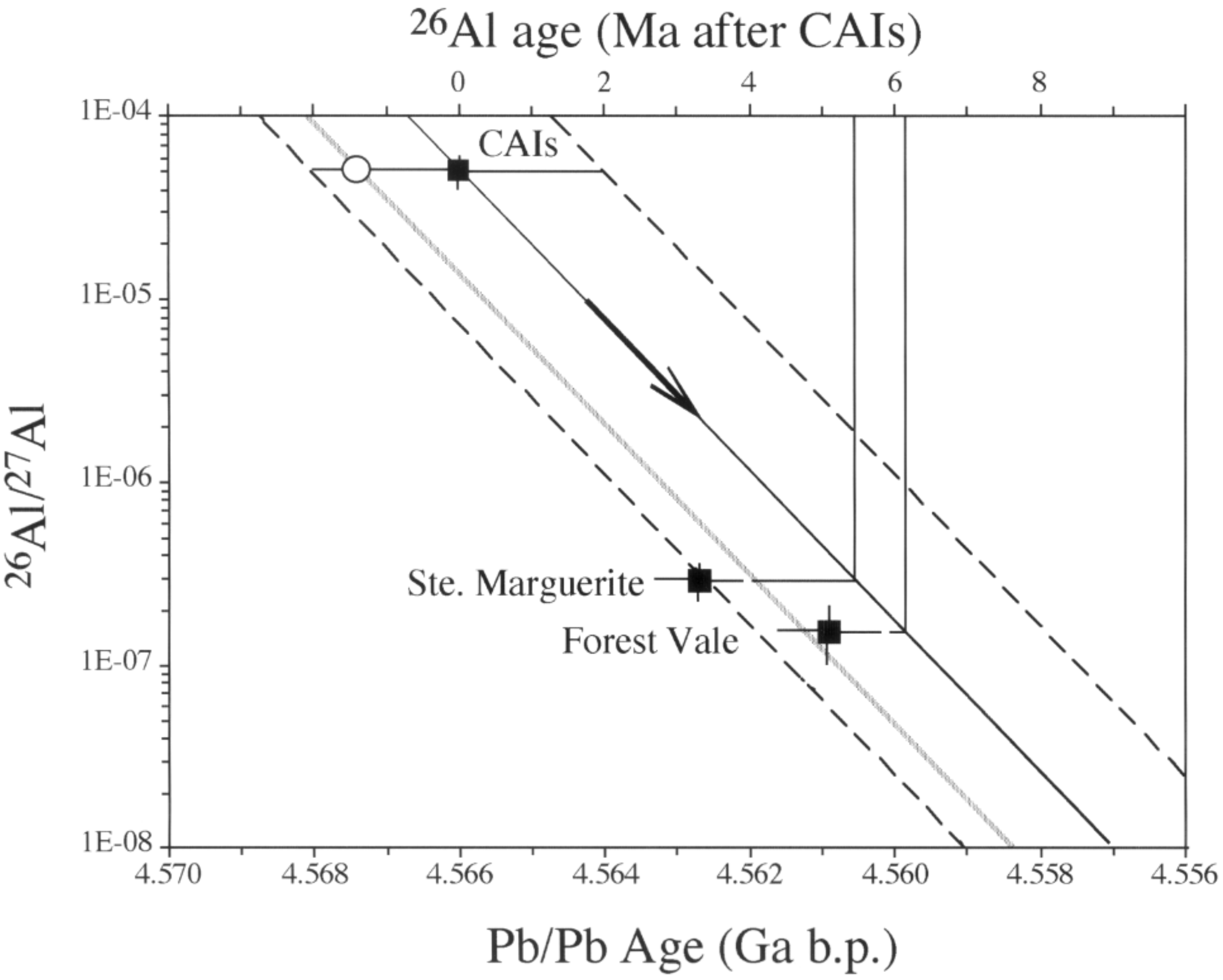

FIG. 3. Plot of inferred initial ^{26}Al/^{27}Al ratios of CAIs, Ste Marguerite and Forest Vale (solid squares) *vs.* the Pb/Pb ages of these objects (lower scale). Also plotted is the temporal evolution of the ^{26}Al/^{27}Al ratio from the canonical value of 5×10^{-5} in CAIs at their Pb/Pb formation time (solid line). The broken lines indicate the uncertainties in this evolution due to the error in the CAI age determination. The upper scale represents Al-Mg ages relative to CAIs. According to their ^{26}Al/^{27}Al ratios, Ste Marguerite and Forest Vale are 5.4 and 6.1 Ma younger than CAIs. The open circle represents the recent U-Pb measurements of CAIs from Allende and Efremovka by Amelin *et al.* (2002). Their average age gives an even better fit to the Ste Marguerite and Forest Vale ages. However, throughout the paper we use the average value of 4.566 Ga from the older determinations (Chen and Wasserburg, 1981; Manhès *et al.*, 1988; Göpel *et al.*, 1994).

achieve the extremely low Mg concentrations observed in the H4 plagioclase.

However, the more detailed petrographic observations in the thin sections do not allow us to strictly exclude the possibility that at least some of the feldspar from SM come from Cr-Al-rich inclusions or chondrules: all but two plagioclase occur in the close vicinity of such Cr-rich inclusions, which, as stated by Christophe Michel-Levy *et al.* (1995), are especially abundant in SM and FV. In these aggregates the rather dense chromite is loosely intergrown with feldspatic mesostasis and partially crystallized plagioclase. The anhedral plagioclase crystals reach a size of up to 200 μm. The second type of plagioclase occurrences consists of subhedral olivine or orthopyroxene crystals that include anhedral plagioclase crystals.

Do the Aluminum–Magnesium and the Uranium–Lead Chronometers Measure the Same Events?

In general, a given chronometer measures the closure time of the corresponding isotopic system and it cannot be expected that different isotopic systems have the same closure times, especially if their solid carriers undergo cooling which is slow with respect to the half lives of the radioactive species under consideration. This is a general problem with all radioactive clocks. Closure is achieved when the temperature drops below a value where the isotopic system used as a chronometer does not equilibrate any longer. However, as has been discussed in detail for the U-Pb system by Göpel *et al.* (1994), the meaning of a radiometric age depends on the precise diffusive behavior of both the parent and the daughter elements, which in turn depends on the metamorphic grade and peak temperature reached in the parent rock. The H4 chondrites were subjected to temperatures between 650 and 950 K (Göpel *et al.*, 1994). This may be lower than the U/Pb closure temperature of the phosphates, which means that the U/Pb closure in the phosphates may not be related to the cooling period for these objects and thus the interpretation of the Pb/Pb age as thermal closure of the phosphates is not guaranteed.

The general situation might be similar for the Al-Mg system but the diffusion constants for Al and Mg in feldspar are probably quite different from those of U and Pb in phosphate. It is thus clear that we cannot simply compare radiometric ages based on different chronometers. In addition, we lack the relevant parameters such as diffusion rates and the exact temperature-time profiles experienced by the different H4 chondrites. It is not even necessary that the feldspar crystals of this study have a metamorphic origin, the essential point is that the Mg isotopes were equilibrated during the heating of the H4 parent rocks. Although from deviations from a simple isochron relationship in CAIs it can be concluded that Mg in plagioclase (and melilite) equilibrates more readily than in spinel and pyroxene (Podosek *et al.*, 1991), the exact diffusive behavior of Mg in feldspar is not known. If we take the diffusion parameters determined by Sheng *et al.* (1992) for spinel and assume a temperature of 800 K for a duration of 1 Ma (Manhès and Göpel, 1998) we obtain a diffusive length of only 0.20 μm. The diffusion rate in feldspar is certainly higher than in spinel but we do not know whether it is high enough for Mg equilibration in 50–100 μm large grains.

The ^{244}Pu fission track technique did not allow the detection of a time difference between the track retention in whitlockite and pyroxene (Pellas and Storzer, 1981) and also the metallographic technique yields only a lower limit of 1000 K/Ma for SM and FV (see Table IV in Lipschutz *et al.*, 1989). If complete closure would have been achieved for both the U-Pb and Al-Mg systems during metamorphic heating of the H4 parent rocks, the ages determined from these two systems would signify cooling below the respective closure temperatures. Then such a large cooling rate would imply a time difference of only a fraction of 1 Ma, less than the uncertainties of the Pb/Pb ages for the phosphates of these two meteorites. However, since closure and complete equilibration in these two systems is not guaranteed, there could be a significant time difference (a couple of million years?) for the ages determined by the two chronometers.

Other Chronometers

Other short-lived isotopes used for determining chronologies of early solar system events include ^{53}Mn and ^{129}I. Since only relative ages can be measured by such chronometers, in each case absolute ages were obtained by anchoring the relevant systems to samples for which precise Pb/Pb ages have been obtained. Lugmair and Shukolyukov (1998, 2001) have done so for the Mn-Cr system by using the Pb/Pb age of 4.5578 ± 0.0005 Ga measured for the angrites Lewis Cliff 86010 and Angra dos Reis (Lugmair and Galer, 1992). Ages obtained in this way are shown in Fig. 4 where the evolution of the ^{53}Mn/^{55}Mn ratios is plotted as a function of time when the time calibration is obtained from the angrites. They include Mn-Cr ages of 4.5650 ± 0.0007 for SM (Polnau *et al.*, 2000) and 4.5613 ± 0.0008 for FV (Polnau and Lugmair, 2001). As can be seen from the figure, the Mn-Cr and Pb/Pb ages of FV agree within the errors, while SM's Mn-Cr age is 2.3 Ma larger than the Pb/Pb age of its phosphate. Göpel *et al.* (1994) also obtained a whole-rock Pb-Pb age of 4.5667 ± 0.0016 Ga for SM that is only 1.7 Ma older than the Mn-Cr age (Polnau and Lugmair, 2001; see also Gilmour and Saxton, 2001). However, we prefer the Pb/Pb age of the phosphates because the Pb/Pb ages of bulk meteorites are *model* ages. All fragments display an excess of radiogenic Pb relative to that produced by *in situ* decay of U, assuming a primordial Pb isotopic composition for the measured ^{204}Pb. This discordant U/Pb feature indicates a recent (about 0–0.5 Ga) perturbation of the U-Pb system which is similar to that observed in all bulk samples of equilibrated chondrites. Consequently, the Pb/Pb ages of all bulk meteorites do not have a precise chronological meaning.

Of course, the same caveat as stated for the Al-Mg and U-Pb systems applies for the Mn-Cr and U-Pb chronometers: they do not necessarily have the same closure temperatures and do not necessarily yield the same time information. In case of Mn-Cr, isochrons are obtained by determining the ^{53}Cr/^{52}Cr ratios in chromite, which has a low ^{55}Mn/^{52}Cr ratio, and silicates, which have a higher ratio. In Fig. 4 we also plotted Mn-Cr age data for the eucrite Chervony Kut, which lies between SM and FV, averages for CI and CM chondrites and CAIs (Birck and Allègre, 1985; Rotaru *et al.*, 1992; Birck *et al.*, 1999), as well as ion probe data from carbonates from the meteorite Kaidun (Hutcheon *et al.*, 1999). There are many more Mn-Cr ages, which are not plotted here. For a more detailed discussion of other samples, see Nyquist *et al.* (2001).

The comparison of the Pb/Pb ages of CAIs and the inferred Mn-Cr age of Kaidun carbonates leads to an immediate problem. Carbonates are believed to be secondary phases, formed by aqueous activity on meteorite parent bodies, and cannot be older than CAIs, which are believed to be the oldest solids formed in the solar system. Gilmour (2000) and Lugmair and Shukolyokov (2001) have suggested that the Pb/Pb age of CAIs is not the primary formation age of these objects but reflects alteration processes or late equilibration. However, the age of 4.5768 ± 0.0012 Ga obtained for CAIs from the inferred ^{53}Mn/^{55}Mn ratio of 4.4×10^{-5} (Birck *et al.*, 1999) is not necessarily the correct age for the formation of the inclusions. As has been discussed by Lugmair and Shokulyokov (1998, 2001), CAIs contain nucleosynthetic anomalies in several elements, including the Cr isotopes (see also Bogdanovski *et al.*, 2002). Thus, there exists an intrinsic

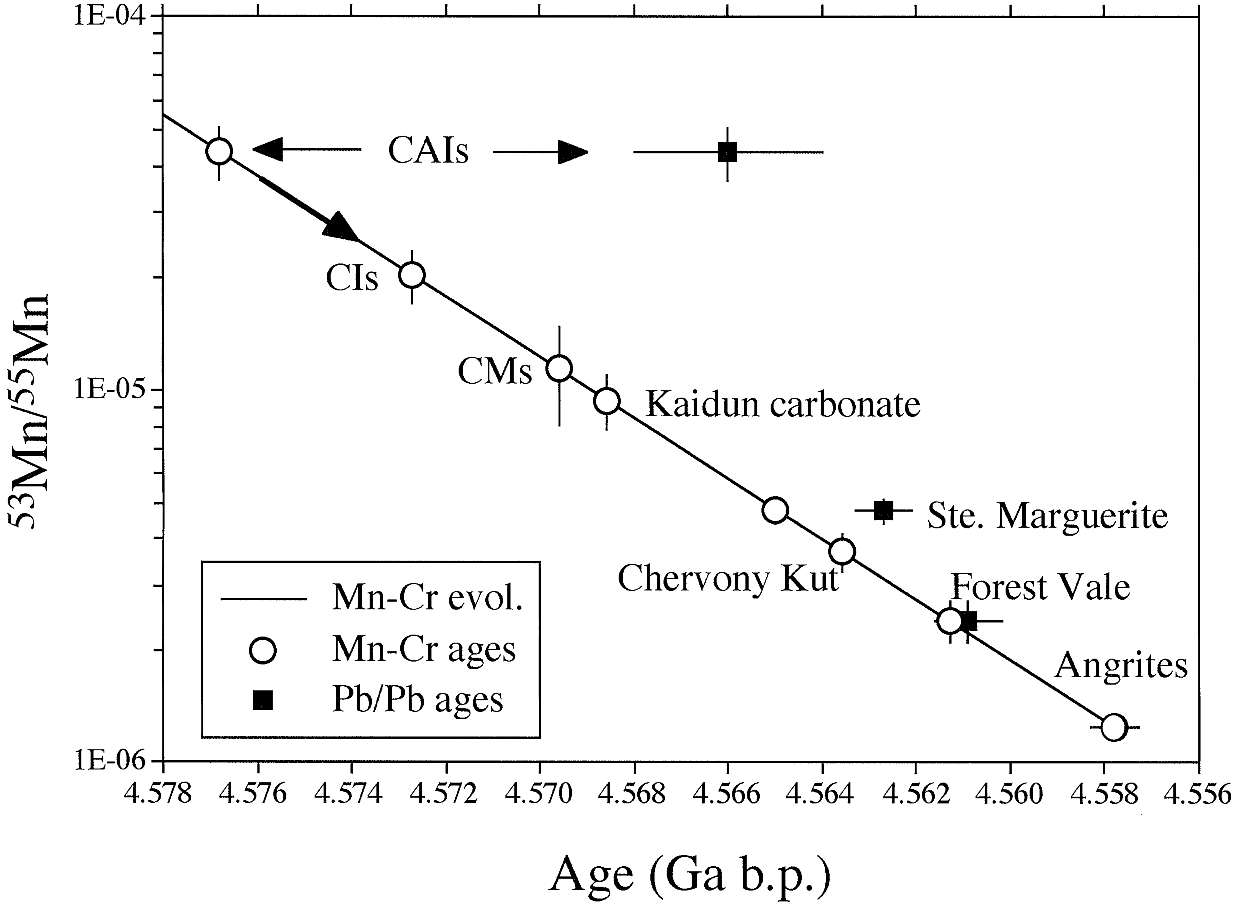

FIG. 4. Ages of different early solar system objects based on their inferred ^{53}Mn/^{55}Mn ratios. The Mn-Cr evolution line is tied to the ^{53}Mn/^{55}Mn ratio in angrites, for which a precise Pb/Pb age has been determined. Also plotted are the Pb/Pb ages of CAIs, Ste Marguerite and Forest Vale phosphates. While the Mn-Cr and Pb/Pb ages are within the errors for Forest Vale, there exists a noticeable difference for Ste Marguerite. Chervony Kut is a eucrite. There are large discrepancies between the Mn-Cr and Pb/Pb ages, especially for CAIs, but also for Ste Marguerite.

ambiguity concerning the Mn-Cr ages of CAIs. Furthermore, there is an incompatibility of the Mn-Cr and U-Pb systems for the age difference between the CAIs and the ordinary chondrites.

The ^{129}I chronometer might also add some information concerning the relative formation ages of H4 plagioclase. In Fig. 5 we plot the evolution of the ^{129}I/^{127}I ratio if it is calibrated against the Pb/Pb of 4.557 ± 0.002 Ga of Acapulco phosphates (Göpel *et al.*, 1992; Brazzle *et al.*, 1999). According to the I-Xe time scale, plagioclase from SM formed 9.5 ± 0.4 Ma earlier than Acapulco phosphates. Unfortunately, the I-Xe age of 4.565 ± 0.006 for SM phosphate has a large error of 6 Ma (Brazzle *et al.*, 1999), so that the I-Xe system does not yield a precise time difference in the formation of SM phosphate and plagioclase. Based on the Acapulco phosphate calibration, the absolute age of SM plagioclase is 4.567 ± 0.002 Ga as compared to the Pb/Pb age of 4.5627 ± 0.0006 for SM phosphate, a time difference of 3.7 ± 2.1 Ma. On the other hand, the I-Xe age of plagioclase agrees with the Mn-Cr age of SM within the errors. Also plotted in Fig. 5 are the ages of sodalite in Allende CAIs (Hohenberg *et al.*, 1998). Since sodalite is an alteration phase, the relative young ages are no surprise.

Figure 6 shows a comparison of ages based on the U-Pb, Mn-Cr, and I-Xe systems. Only the first provides absolute ages, the others have been anchored to the Pb/Pb ages of the angrites and of Acapulco, respectively, as discussed above. The errors plotted for the Mn-Cr ages and I-Xe indicate only the errors for the time differences relative to the calibration ages but do not include the uncertainties of the latter. Here we concentrated only on objects relevant for this discussion, more detailed discussions of relative ages can be found in the papers by Brazzle *et al.* (1999), Gilmour (2000), Gilmour and Saxton (2001), and Lugmair and Shukolyukov (2001). There are several discrepancies. One is the time difference between SM and FV as determined by the U-Pb and the Mn-Cr systems. The largest discrepancy exists between the Pb/Pb and Mn-Cr ages of CAIs and, quite independent of any absolute age calibration, there is a huge difference in the U-Pb and Mn-Cr ages between CAIs and the H4 chondrites. There cannot be much

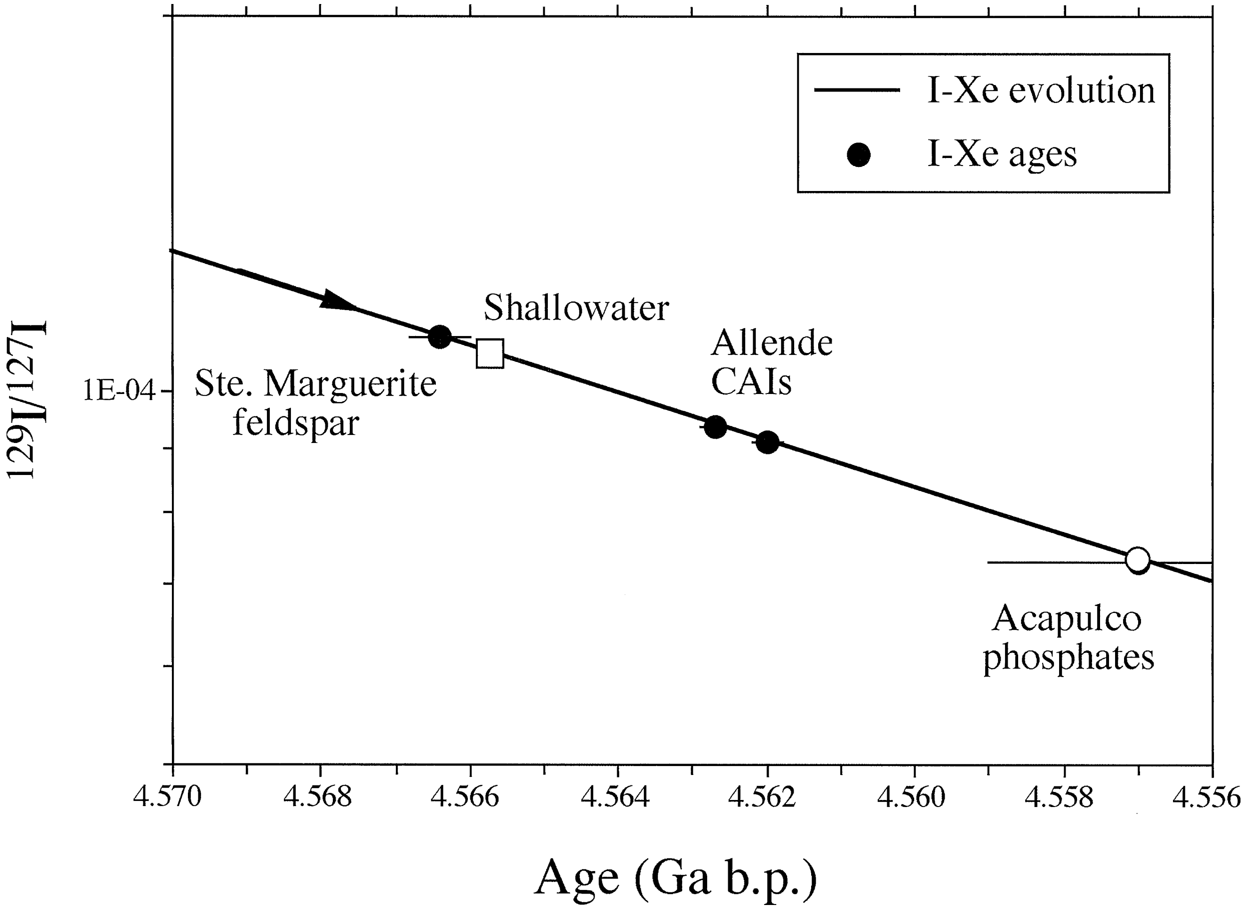

FIG. 5. Ages of Allende CAIs and Ste Marguerite feldspar based on their inferred $^{129}I/^{127}I$ ratios. The I-Xe evolution lied is anchored to the $^{129}I/^{127}I$ ratio and the Pb/Pb age of Acapulco phosphate. Shallowater is used as a reference standard for I-Xe measurements; however, its absolute age is determined from that of Acapulco. According to the ^{129}I clock, Ste Marguerite feldspar is older than its phosphate (Fig. 3).

doubt that CAIs must be older than carbonates. Incidentally, chondrules from Bishunpur and Chainpur give the same Mn-Cr age as these carbonates (Nyquist *et al.*, 2001). In Fig. 6 we assigned Al-Mg ages to CAIs and the H4 plagioclase samples by fixing the CAI age at 4.570 Ga. This was done under the assumption that the Mn-Cr age of Kaidun carbonates is correct and that the time interval between CAI formation and the formation of meteorite parent bodies, on which aqueous activity could take place, was at least 1 Ma. With this assignment, the SM Al-Mg age agrees within errors with the I-Xe age of its plagioclase and with its Mn-Cr age. There is a discrepancy for FV but that would exist whatever Al-Mg age we assign: the difference in inferred $^{26}Al/^{27}Al$ ratios between SM and FV is too small to be in agreement with the difference in Mn-Cr ages between these two meteorites. Alternatively, if we tie the Al-Mg age of FV to its Mn-Cr age we would have to conclude that CAIs are younger than carbonates in Kaidun, a thought that makes us quite uncomfortable. This, of course, is all said under the assumption that the Al-Mg system does provide ages and that the Al-Mg clock for SM and FV started approximately at the same time as the clocks of the other chronometers started for these H4 chondrites. On the other extreme, the Mn-Cr age for CAIs seems to be excluded by the simple observation of ^{26}Mg excesses in SM and FV plagioclase *if* the Mg isotopes in this phase had been reset by the heating of the H4 chondrites. If the inferred $^{26}Al/^{27}Al$ ratios for these two meteorites are representative for their Mn-Cr age, then the $^{26}Al/^{27}Al$ ratio in CAIs would have to be at least 3.0×10^{-3} and 3.3×10^{-2}, respectively, which is clearly not the case.

Can Aluminum-26 Serve as a Fine-Scale Chronometer?

Let us return to our original question whether or not ^{26}Al can be used as a chronometer for early solar system events. In the previous discussion of Fig. 6 we have already implicitly made this assumption. It is clear from this figure that there are discrepancies between ages based on different chronometers. These differences exist not only in terms of absolute ages which, for the short-lived chronometers, can only be achieved by anchoring them to U-Pb ages, but also for relative ages. One obvious example is provided by the differences in relative U-Pb and Mn-Cr ages for CAIs and the angrites. As has already

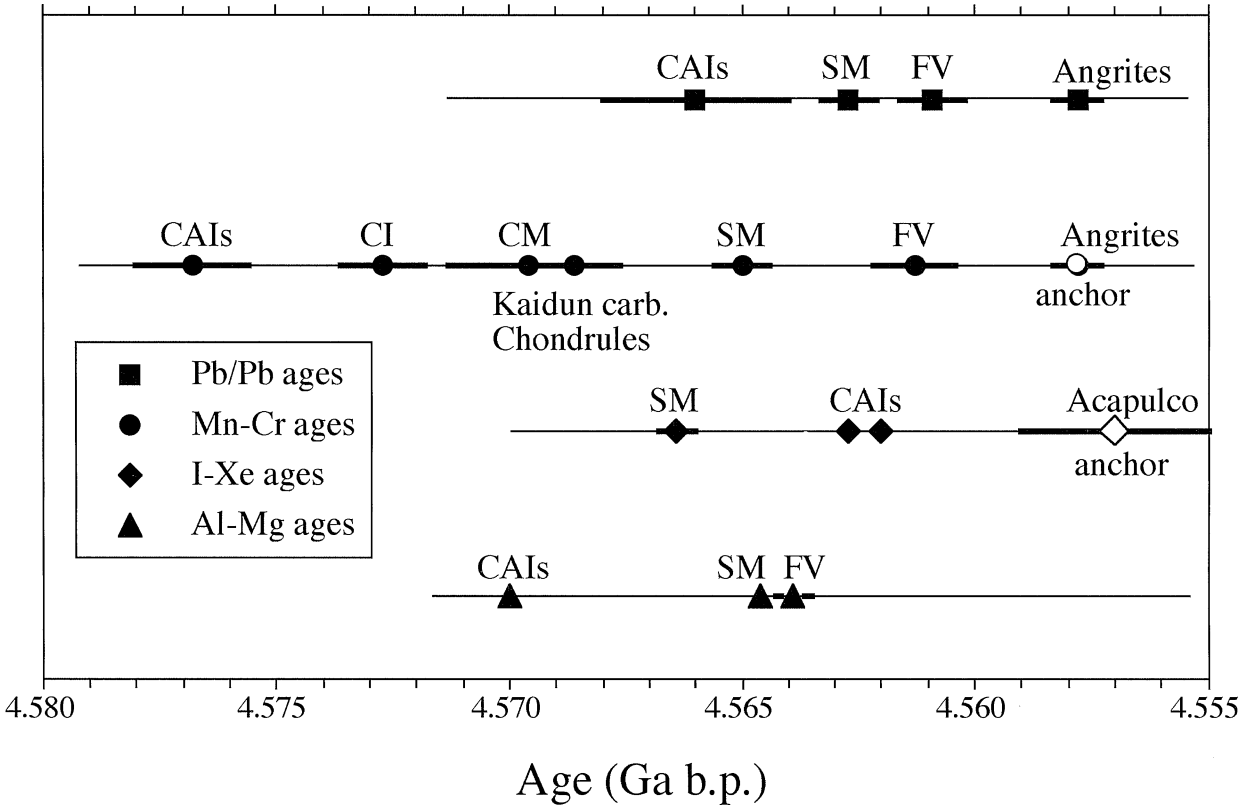

FIG. 6. Comparison of ages based on different chronometers. Only the Pb/Pb ratio gives absolute ages, the Mn-Cr and I-Xe ages are tied to the Pb/Pb ages of angrites and Acapulco, respectively. Al-Mg ages are plotted under the assumption that CAIs are at least 1 Ma older than carbonates from Kaidun. According to this assumption, CAIs are still considerably younger than the age derived from their ^{53}Mn/^{55}Mn ratio. However, such an old age is ruled out by the presence of radiogenic ^{26}Mg in plagioclase from Ste. Marguerite and Forest Vale. Given the ages of these objects determined by other chronometers, ^{26}Al must have been widely distributed in the early solar system.

been pointed out at the beginning of the discussion section, the Pb/Pb ages of CAIs and SM and FV phosphate are compatible with the different inferred ^{26}Al/^{27}Al ratios for these objects. However, we immediately run into problems with the Pb/Pb age of CAIs as soon as we compare it with Mn-Cr ages, not only with that for CAIs (which might not be correct) but also with that of Kaidun carbonates.

Still, with all necessary caution, we believe we can answer the question about the chronometer ^{26}Al in a positive sense. If, as we have done in Fig. 6, we assume that CAIs are ~1 Ma older than carbonates from chondrite parent bodies, then the age differences relative to SM and FV agree with the ^{26}Al/^{27}Al ratios at least for SM. Furthermore, if the plagioclase in SM and FV is of metamorphic origin and obtained its Al not just from CAIs or if the Mg in feldspar has been isotopically equilibrated (*i.e.*, the Al-Mg clock has been reset) during metamorphic heating of the H4 parent body, then the inferred ^{26}Al/^{27}Al ratios can be as high as measured only if the ^{26}Al/^{27}Al ratio at the time of CAI formation was 5×10^{-5} not just in CAIs but in most of the material present in the early solar system.

However, we cannot strictly rule out that the inferred ^{26}Al/^{27}Al ratios in H4 chondrites have nothing to do with the metamorphic ages of these rocks and that the plagioclase crystals formed much earlier, received much lower initial ^{26}Al/^{27}Al ratios and that Mg in these crystals was not equilibrated during metamorphic heating of the H4 parent body. Still, if these feldspar crystals were from chondrules and if these chondrules formed more or less contemporaneously with CAIs in the X-wind of an early Sun (Shu *et al.*, 2001), it has to be explained why the inferred ^{26}Al/^{27}Al ratios in H4 chondrites are distinctly lower than those found in carbonaceous and ordinary chondrites (Hutcheon *et al.*, 1994, 2000; Hutcheon and Jones, 1995; Russell *et al.*, 1996; Kita *et al.*, 2000; Marhas *et al.*, 2000; Srinivasan *et al.*, 2000a,b; Huss *et al.*, 2001; Mostefaoui *et al.*, 2002), why there is a difference in these ratios between SM and FV, and why there seems to be a dependence of the ^{26}Al/^{27}Al ratios in chondrules from a variety of ordinary chondrites on metamorphic grade (Huss *et al.*, 2001).

Remaining Questions

Even though we believe that the existing evidence is in favor of widespread distribution of ^{26}Al in the early solar system, there remain many discrepancies, uncertainties and questions. Some have already become apparent in the previous discussion such as the discrepancies arising from comparison of the U-Pb with the Mn-Cr system. One of the most important is posed by the initial presence of ^{10}Be in CAIs. This radioisotope ($t_{1/2}$ = 1.5 Ma) is produced in spallation reactions by energetic particles. According to the X-wind model, this irradiation happened during an active early Sun and produced also the ^{26}Al in CAIs and chondrules (Gounelle *et al.*, 2001; Shu *et al.*, 2001). Most measurements in CAIs made so far indicate a fairly constant ratio between ^{26}Al/^{27}Al and ^{10}Be/^{9}Be (McKeegan *et al.*, 2000; Sugiura *et al.*, 2001), although some variations have been observed (MacPherson and Huss, 2001). Because ^{10}Be and ^{26}Al are produced from different target elements, no strict correlation is expected, even if both isotopes *had* a spallation origin unless some precursor material was irradiated and subsequently thoroughly mixed before the formation of CAIs from this homogeneous mix. Although Gounelle *et al.* (2001) make the (very ad hoc) assumption that solid proto-CAIs were irradiated, the very uniform ^{26}Al/^{27}Al ratios in CAIs can only be achieved by thorough mixing of irradiated precursor material. Still, most CAIs apparently were produced during a short time span. It would therefore be extremely important to look for the presence of radiogenic ^{10}B in samples that are not CAIs and that are younger than CAIs. If ^{10}Be was widely distributed in the early solar system (as we argued ^{26}Al was), we can make estimates of the inferred ^{10}Be/^{9}Be ratios expected for SM and FV. With an assumed initial ^{10}Be/^{9}Be ratio of 8×10^{-4} in CAIs and time differences of 5.4 and 6.1 Ma derived from the ^{26}Al/^{27}Al ratios we obtain ^{10}Be/^{9}Be ratios of 6.5×10^{-5} for SM and 4.8×10^{-5} FV. The actual measurements would pose a formidable challenge. The ratios themselves do not look prohibitive and reasonably high Be/B ratios have been found in CAIs. However, it should be remembered that even then Be remains a trace element. A further complication is that B has only two stable isotopes so that no mass fractionation correction can be applied when measuring radiogenic ^{10}B excesses. Thus, a search for ^{10}Be in non-CAI materials probably will depend on whether any phases with high Be/B *and* high Be content can be found.

Even if such measurements prove to be impossible or unsuccessful, there are other improvements one would hope for to clarify the question of whether ^{26}Al is a chronometer. Al-Mg measurements in H4 plagioclase should be made on samples whose petrological origin is better known (Zinner *et al.*, 2002). Furthermore, the closure behavior of different isotopic systems should be better known. This, together with better knowledge of the temperature profile experienced by the H4 chondrites and more precise ages from different clocks would make it possible to clarify some chronological discrepancies. For example, the difference between the Pb/Pb age of SM phosphate and the I-Xe age of SM feldspar is 3.7 ± 2.1 Ma. If this time difference is real and if both phosphate and plagioclase are of metamorphic origin, it appears to be incompatible with a metallographic cooling rate of >1000 K/Ma. This is just one example, but there are many more unresolved questions.

CONCLUSIONS

Among plagioclase grains from the H4 chondrites SM, FV, Beaver Creek and Quenggouk, those from the first two meteorites show clear excesses of ^{26}Mg. The ^{26}Al/^{27}Al ratios inferred from theses excesses and from isotopically normal Mg in pyroxene and olivine are $(2.87 \pm 0.64) \times 10^{-7}$ for SM and $(1.52 \pm 0.52) \times 10^{-7}$ for FV. If interpreted chronologically, the difference between these ratios and the ratio of 5×10^{-5} in CAIs indicate time differences of 5.4 ± 0.1 and 6.1 ± 0.2 Ma between CAIs and plagioclase from SM and FV, respectively. These differences are, within errors, in agreement with the absolute Pb/Pb ages for CAIs and SM and FV phosphates. However, chronology based on the ^{53}Mn chronometer has consistency problems with respect to the other chronometers and at present does not provide a unique interpretation in terms of timescale.

If we require that CAIs are older than carbonates from Kaidun, the time difference between this "adjusted" CAI age and the Mn-Cr ages of SM and FV requires that ^{26}Al was widely distributed in the early solar system at the time of CAI formation and was not mostly present in CAIs, a feature of the X-wind model for the production of ^{26}Al and the formation of CAIs. This in turn means that ^{26}Al can serve as a fine-scale chronometer for early solar system events. In addition, although there are still inconsistency problems between the different chronometers which require further study, the major results indicate a short timescale for the formation, metamorphism and igneous activity of the first planetary bodies and is in agreement with ^{26}Al having been widespread and having acted as a heat source.

Dedication–This paper is dedicated to the memory of Paul Pellas. His passion for meteoritics and his boundless enthusiasm for some of the problems treated in this paper will never be forgotten. His stimulation and guidance were essential for this study. We also thank him for providing grain separates from Forest Vale and Beaver Creek and for many heated discussions.

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