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2 ***Title: Samarium-neodymium chronology and Rubidium-Strontium***
3 **systematics of an Allende calcium-aluminum-rich inclusion with implications**
4 **for ^{146}Sm half-life**

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ABSTRACT

Calcium-aluminum-rich inclusions (CAIs) are primitive objects that formed within the protoplanetary disk surrounding the young Sun. Recent Pb–Pb chronologic studies have demonstrated that CAIs are the oldest solar system solids, crystallizing 4567 Ma ago (Amelin et al., 2002; Connelly et al., 2012). The isotope systematics of CAIs therefore provide critical insight into the earliest history of the Solar System. Although Sm–Nd and Rb–Sr geochronometers are highly effective tools for investigating cosmochemical evolution in the early Solar System, previous studies of CAIs have revealed evidence for isotopically disturbed systems. Here we report new age data for Allende CAI Al3S4 derived from both the long-lived (^{147}Sm – ^{143}Nd) and short-lived (^{146}Sm – ^{142}Nd) isotopic systems. The ^{147}Sm – ^{143}Nd chronometer yields an age of 4560 ± 34 Ma that is concordant with ^{207}Pb – ^{206}Pb ages for CAIs and indicates that the Sm–Nd system was not significantly disturbed by secondary alteration or nucleosynthetic processes. The slope of the ^{146}Sm – ^{142}Nd isochron defines the Solar System initial $^{146}\text{Sm}/^{144}\text{Sm}$ of 0.00828 ± 0.00044 . This value is significantly different from the value of 0.0094 determined by Kinoshita et al (2012). Ages recalculated from all published ^{146}Sm – ^{142}Nd isochron data using the traditional 103 Ma half-life and the initial $^{146}\text{Sm}/^{144}\text{Sm}$ value determined here closely match Pb–Pb and ^{147}Sm – ^{143}Nd ages determined on the same samples. In contrast, ages recalculated using the 68 Ma half-life determined by Kinoshita et al. (2012) and either of the initial $^{146}\text{Sm}/^{144}\text{Sm}$ values are often anomalously old. This is particularly true for the youngest samples with ^{146}Sm – ^{142}Nd isochron ages that are most sensitive to the choice of ^{146}Sm half-life used in the age calculation. In contrast to the Sm–Nd isotope system, the Rb–Sr system is affected by alteration but yields an apparent isochron with a slope corresponding to a much younger age of 4247 ± 110 Ma. Although the Rb–Sr system in CAIs appears to be disturbed, the

initial $^{87}\text{Sr}/^{86}\text{Sr}$ value determined from the isochron is 0.698942 ± 0.000008 , and closely approximates estimates of the initial Solar System value. Although this isochron may be a mixing line, it might also record alteration on the Allende parent body in which Rb was added to the Al₃S₄ CAI that was initially largely devoid of Rb.

1. Introduction

Calcium-aluminum-rich inclusions formed in the protoplanetary disk surrounding the young Sun as a result of multiple transient heating events that thermally processed presolar dust and solar system condensates (Hood et al., 2009; Scott and Krot, 2007). In order to better understand the earliest history of the Solar System and the formation of CAIs, we investigated the chronometry of Allende CAI Al₃S₄ with improved methods for determining both absolute and relative ages. Isotopic analyses of the Sm–Nd and Rb–Sr systems were performed on CAI Al₃S₄ (also known as TS₃₄), previously described by (Clayton et al., 1977), from the Allende CV3 carbonaceous chondrite. The Al₃S₄ CAI is an igneous type B inclusion in which the major minerals - melilite, clinopyroxene (Al, Ti-rich diopside), and anorthite exhibit chemical equilibrium (e.g. Simon et al. 1991).

The ^{147}Sm – ^{143}Nd system has had relatively limited application in dating the earliest solar system materials, such as chondritic meteorites and CAIs. This is in part due to the limited spread in the $^{147}\text{Sm}/^{144}\text{Nd}$ ratios present in these bulk samples that makes age determinations difficult. Analyses of minerals separated from several Allende and Efremovka CAIs have generally resulted in discrepant ages. For example, a four point isochron for Allende CAI Egg 2 yielded an age of 4.80 ± 0.07 Ga, whereas two point melilite-pyroxene tie-lines for Egg 6 and

Big Al yielded ages of 4.80 ± 0.18 Ga and 4.53 ± 0.09 Ga, respectively (Papanastassiou et al., 1987). Other Sm–Nd data reported for CAIs demonstrate clear evidence for isotopic disturbance, although some individual data points fall near a 4.55 Ga reference line (Bogdanovski and Jagoutz, 1999, Bogdanovski and Jagoutz, 1997). Thus, with the exception of the two-point tie-line for Allende CAI Big Al by (Papanastassiou et al., 1987), all published Sm–Nd data for CAIs demonstrate evidence for isotopic disturbance. The relative paucity of Sm–Nd age investigations of CAIs reflects the difficulties associated with: 1) obtaining large, petrographically well characterized samples, 2) producing high purity mineral separates that exhibit substantial fractionation of Sm and Nd, and 3) obtaining sufficiently accurate isotopic ratios on very small amounts of Nd and Sm. This study is based on large mineral fractions from Allende CAI Al3S4 produced at the University of Chicago circa 1975 that allowed long duration, high intensity, multi-dynamic runs of Nd^+ (rather than NdO^+ as in previous studies) on the newest generation of thermal ionization mass spectrometer. This approach minimizes measurement errors, facilitates high precision $^{142}\text{Nd}/^{144}\text{Nd}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratio determinations, and ultimately allowed us to obtain undisturbed Sm–Nd isotopic systematics for a CAI.

The alpha decay of short-lived radionuclide ^{146}Sm to ^{142}Nd has been used as a geochronometer for determining the chronology of Solar System formation and early planetary differentiation, and provides insights into *p*-process nucleosynthesis of solar ^{146}Sm (e.g. Caro, 2011; Boyet et al., 2010; Wassurburg et al., 2006; Audouze and Schramm, 1972). The half-life of ^{146}Sm has been measured five times (Meissner et al. 1987; Friedman et al. 1966; Nurmia et al., 1964; Dunlavey and Seaborg, 1953), and includes a recent measurement by Kinoshita et al., (2012). The currently adopted half-life, derived from the work of Friedman et al. (1966) and Meissner et al., (1987) is 103 ± 5 Ma. Kinoshita et al., (2012) determined a value for the ^{146}Sm

half-life of 68 ± 14 Ma based on alpha activity counting experiments. This value is outside analytical uncertainty of the traditional 103 Ma half-life changing the time scales derived from the isotopic system. The coherent ^{146}Sm - ^{142}Nd isochron derived from mineral separates of CAI Al3S4 in this study, provides an independent evaluation of the ^{146}Sm half-life.

There is abundant evidence for isotopic disturbance of the Rb–Sr system in Allende CAIs (Gray et al., 1973; Tatsumoto et al., 1976; Podosek et al., 1991; Moynier et al., 2010). Young model ages of CAIs have been interpreted to reflect disturbance of the Rb–Sr system after the formation of the CAIs but before ~ 3.6 Ga, implying that alteration occurred on the Allende parent body at least 1 Ga after CAI formation. In addition to the Sm–Nd data, we also present Rb–Sr isotopic data that strongly implies a disturbance to the system during alteration on the Allende parent body.

2. Materials and Methods

Allende CAI Al3S4 is also known as TS-34 and has been studied extensively (Clayton et al., 1977; Beckett, 1986; Simon et al., 1991; Davis et al., 1992; Beckett et al., 2000). A photomicrograph produced by Clayton et al. (1977) is presented in Figure 1. Al3S4 is a relatively large CAI, measuring 1.2 cm at its maximum dimension. The petrography of Al3S4 was described in detail by Clayton et al. (1977). The CAI has a well-defined mantle of melilite with ~ 38 volume percent (vol %) clinopyroxene (Al, Ti-rich diopside) and ~ 4 vol % spinel. The core contains approximately 14 vol % spinel, 32 vol % melilite, 46 vol % clinopyroxene, < 2 vol% anorthite, and ~ 6 vol% secondary phases (Beckett, 1986; Connolly et al., 2003). Melilite and pyroxene (Al, Ti-rich diopside) crystals in the interior range up to ~ 1 mm in size and are surrounded by a 1.5 mm thick mantle composed primarily of melilite. Spinel crystals 40–150

108 μm in size are included in all major phases throughout the CAI. Cracks and veins of alteration,
109 typical of most Allende inclusions, are present throughout TS-34. Secondary minerals are
110 primarily grossular, monticellite, and wollastonite (Beckett, 1986).

111 Allende CAI Al3S4 was separated into constituent phases by a combination of magnetic
112 and heavy liquid separations, as well as hand-picking, at the University of Chicago (Clayton et
113 al., 1977). Mineral fractions were prepared using two density separation steps. In the first,
114 melilite and pyroxene-dominated fractions were prepared from a 100–300 μm size fraction using
115 a solution of density 3.26 g/cm^3 . The melilite and pyroxene-dominated fractions were further
116 crushed to $<50 \mu\text{m}$ and separated into melilite and pyroxene fractions by hand-picking. Two
117 density separates of the bulk CAI above and below 3.26 g/cm^3 were also analyzed. The
118 composition of the mineral fractions was determined by x-ray diffraction at the University of
119 Chicago (Clayton et al., 1977) and confirmed by SEM/EDS at Lawrence Livermore National
120 Laboratory. A brief description of the composition of the mineral fractions follows: 1) the
121 $d<3.26 \text{ g}/\text{cm}^3$ fraction consists primarily of melilite with minor anorthite and secondary minerals;
122 2) the $d>3.26 \text{ g}/\text{cm}^3$ fraction consists of pyroxene and spinel; 3) the pyroxene fraction consists of
123 pyroxene (Al, Ti-rich diopside), spinel, and perovskite, 4) the melilite + pyroxene fraction
124 consists primarily of melilite with minor pyroxene, 5) the melilite fraction consists of melilite
125 with minor anorthite, and 6) the fines fraction consists of a split of the fine-grained, <325 mesh
126 powder, produced during sieving of the sample in preparation for mineral separations. Although
127 the mineralogy of this fraction was not determined it is likely to contain a disproportionate
128 amount of fine-grained alteration minerals, making it somewhat more vulnerable to isotopic
129 disturbance.

The five mineral and fines fractions were washed and sonicated in deionized water at LLNL for 10 minutes, and then digested in concentrated HF+HNO₃ using Parr bombs. Samples were heated to 150°C for 24 hours in the bombs. Samples were transferred from the bombs and underwent repeated dry downs in aqua regia until completely digested with no residual material remaining. Following dissolution, 5% aliquots were spiked with ¹⁴⁹Sm and ¹⁵⁰Nd tracers (97.7% and 97.8% purity, respectively). In order to minimize uncertainty on the ⁸⁷Rb/⁸⁶Sr ratio associated with the Rb laboratory blank, the 95% fractions were spiked with ⁸⁷Rb and ⁸⁴Sr tracers (98.0% and 99.2% purity, respectively). Rubidium, strontium, and REE were purified using BioRad AG-50W-X8 cation exchange resin and 2N and 6N HCL. Samarium and neodymium were separated using 0.2M α-HIBA acid in pressurized quartz columns. Samples analyzed for Nd isotope composition were passed through these columns twice to reduce ¹⁴²Ce interferences on ¹⁴²Nd. Total procedural blanks were 35 pg Sm, 117 pg Nd, 9 pg Rb, and 35 pg Sr. Although the need to digest the samples using bombs resulted in blanks that were significantly higher than usual, the high abundances of Sr and REE in the samples makes the blank contributions to the total elemental budget unimportant (0.0034–0.014% for Sm, 0.0040–0.017% for Nd, 0.0001–0.0063% for Sr, and 0.054–0.010% for Rb) in all but one fraction. The pyroxene fraction had a Rb contribution from the blank of 1.3% and required a significantly larger correction.

Mass spectrometry was performed on a ThermoScientific Triton thermal ionization mass spectrometer at LLNL. Neodymium isotopic compositions and ¹⁴⁷Sm/¹⁴⁴Nd ratios of individual mineral fractions were determined on separate aliquots of the mineral separates, alleviating the need for correcting Nd isotopic compositions for the contribution of the 97.8% ¹⁵⁰Nd tracer (Table 1). Neodymium and samarium were loaded on double zone refined Re filaments in 2N HCL. Samarium was corrected for internal mass fraction using ¹⁴⁷Sm/¹⁵²Sm = 0.56083.

Neodymium was run in dynamic mode at high intensity (2.7–4.8V ^{144}Nd ; 10^{11} ohm amplifiers) for 510 to 1080 ratios (8 second integrations) for all but one fraction to obtain very high precision $^{142}\text{Nd}/^{144}\text{Nd}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios. The $d < 3.26 \text{ g/cm}^3$ fraction was run at 1.3V ^{144}Nd (10^{11} ohm amplifier) for 130 ratios. Ratios were corrected for mass fractionation to $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$ using an exponential law. Long term $^{142}\text{Nd}/^{144}\text{Nd}$ and $^{143}\text{Nd}/^{144}\text{Nd}$ ratios measured for the JNdi-1 neodymium isotopic reference (Tanaka et al., 2000) was 1.141837 ± 0.000007 and 0.512101 ± 0.0000007 (2 stdev; $N = 37$), respectively. This is in good agreement with the average values obtained on 7 JNdi-1 standards run during the course of this investigation which were 1.141837 ± 0.000005 and 0.512102 ± 0.0000003 (2 stdev). The cerium interference on ^{142}Nd is 12-24 ppm for all mineral fractions except for Mel-Pyx (75 ppm) and Pyx (1400 ppm). Rubidium and strontium data are presented in Table 2. Strontium was run on single zone refined Re filaments with a Ta_2O_5 emitter suspended in H_3PO_4 . Data were collected in static mode using 10^{12} ohm amplifiers on ^{84}Sr , ^{86}Sr and ^{87}Sr . Individual mass spectrometer runs were performed at 2–5V ^{88}Sr intensities (10^{11} ohm amplifier) and consisted of 200 cycles with 16.8 second integration times. Internal mass fractionation was corrected using $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$. Replicate analyses of the NBS-987 Sr standard during the course of this investigation yielded an external reproducibility corresponding to $^{87}\text{Sr}/^{86}\text{Sr} = 0.710243 \pm 0.000009$ (2 stdev; $N=12$). Note that all of the published Sr isotopic data used for comparison were renormalized to our long-term NBS-987 value of $^{87}\text{Sr}/^{86}\text{Sr} = 0.710249 \pm 0.000009$ (2 stdev; $N=49$). Rubidium was run on single zone refined Re filaments. Instrumental mass fraction was corrected using the NBS-984 external standard assuming an $^{85}\text{Rb}/^{86}\text{Sr}$ of 2.593; the uncertainty of Rb fractionation was 0.5%, and was propagated through the total uncertainty of 1%.

3. Results

3.1 Sm-Nd systematics

The Sm-Nd isotopic data for the mineral separates from CAI Al3S4 are listed in Table 1 and are plotted Figures 1 and 2. The $^{147}\text{Sm}/^{144}\text{Nd}$ ratios for the mineral fractions demonstrate significant variation ranging from 0.17076 for the melilite fraction to 0.23204 for the pyroxene fraction. The mineral fractions fall on or very close to a single correlation line on both ^{147}Sm – ^{143}Nd and ^{146}Sm – ^{142}Nd isochron plots. The isochrons were plotted with *Isoplot 4.1* (Ludwig et al., 2009) and include all data, although the ^{147}Sm – ^{142}Nd isochron does exclude data for the fines fraction.

3.2 Rb-Sr systematics

The Rb-Sr isotopic data for the same mineral separates analyzed for Sm-Nd are listed in Table 2 and plotted in Figure 3. The $^{87}\text{Rb}/^{86}\text{Sr}$ ratios range from 0.0076 for the melilite to 0.071 for the density >3.26 separate. The mineral separates plot on a single correlation line with an MSWD of 1.4. The $^{84}\text{Sr}/^{86}\text{Sr}$ ratios ($\epsilon^{84}\text{Sr}$) of the mineral separates are listed in Table 2. The $^{84}\text{Sr}/^{86}\text{Sr}$ ratios of the mineral separates have $\epsilon^{84}\text{Sr}$ values ranging from 0.51 for the density separate >3.26 to 1.34 for the pyroxene separate, with a mean $\epsilon^{84}\text{Sr} = 1.08 \pm 0.30$. The $\epsilon^{84}\text{Sr}$ values are within error of the values measured in recent studies of nucleosynthetic anomalies in bulk CAIs (e.g. Brennecka et al., 2013; Hans et al., 2013; Moynier et al., 2010).

4. Discussion

4.1 Sm–Nd ages of CAI crystallization

The Sm–Nd isotopic data from Al3S4 form a well-defined linear array with a slope that reflects the crystallization age of the CAI (Table 1; Figs. 2 and 3). The ^{147}Sm – ^{143}Nd chronometer yields an age of 4560 ± 34 Ma with a relatively small mean squared weighted deviation (MSWD) of 2.4. The agreement between the ^{147}Sm – ^{143}Nd age and the accepted Pb–Pb age of 4567.2 ± 0.6 Ma for CAIs (Amelin et al., 2002; Connelly et al., 2012; Amelin et al., 2010; Jacobsen et al., 2008; Bouvier and Wadhwa, 2010) provides strong evidence that the Sm–Nd isotopic system in Al3S4 has been minimally disturbed since the CAI cooled below the closure temperature for Sm and Nd diffusion. This age is concordant with the two-point melilite and pyroxene tie line age of 4.53 ± 0.09 Ga reported for Allende CAI Big Al by Papanastassiou et al. (1987), but is more precise. Some evidence for disturbance of the Sm–Nd isotopic system in CAI Al3S4 is revealed by the fines fraction, however. This separate lies off the ^{147}Sm – ^{143}Nd isochron, but on the ^{146}Sm – ^{142}Nd isochron (see below). This fraction was produced during initial crushing of the CAI and was separated from the other fractions during the sieving process. As a result, this fraction contains the finest-grained, most easily friable, component of the CAI and is expected to have a larger proportion of secondary minerals than the other mineral fractions. The initial $^{143}\text{Nd}/^{144}\text{Nd}$ ratio derived from the isochron is 0.506657 ± 0.000043 ($\epsilon^{143}\text{Nd} = -0.34 \pm 0.85$), and agrees with $^{143}\text{Nd}/^{144}\text{Nd}$ ratios previously determined on primitive chondritic meteorites of 0.506686 ± 0.000070 ($\epsilon^{143}\text{Nd} = 0.24 \pm 1.4$; Bouvier et al., 2008) and 0.50665 ± 0.00014 ($\epsilon^{143}\text{Nd} = -0.47 \pm 2.8$; Amelin and Rotenberg, 2004), but is more precise. The initial $^{143}\text{Nd}/^{144}\text{Nd}$ determined from CAI Al3S4 isochron is within uncertainty of the value of 0.506674 ($\epsilon^{143}\text{Nd} \equiv 0$) proposed by Jacobsen and Wasserburg (1980, 1984) for the CHondritic Uniform Reservoir (CHUR). This agreement confirms the current CHUR parameters, and provides

additional evidence that the Sm–Nd isotopic systematics of Al3S4 record the crystallization age of the sample.

The availability of relatively large mineral fractions allowed the first internal ^{146}Sm – ^{142}Nd isochron for a CAI to be produced. The initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratio defined by the slope of the isochron is 0.00828 ± 0.00044 (Fig. 3). This value is likely to be one of the most accurate representations of the initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratio of the Solar System, not only because CAIs are the earliest solids formed in the protoplanetary disk, but also because the linearity of the isochron provides assurance that the ^{146}Sm – ^{142}Nd isotopic systematics are not disturbed. The initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratio defined by the CAI isochron is in excellent agreement with the value of 0.0085 ± 0.0007 determined from basaltic eucrite meteorites by (Boyet et al., 2010), but differs significantly from the $^{146}\text{Sm}/^{144}\text{Sm}$ ratio of 0.0094 ± 0.0005 determined by (Kinoshita et al., 2012). The $^{146}\text{Sm}/^{144}\text{Sm}$ ratios calculated by (Boyet et al., 2010) and (Kinoshita et al., 2012) are based on Sm–Nd isotopic measurements of the same set of eucrite samples but determined using the 103 Ma and 68 Ma ^{146}Sm half-lives, respectively.

The ^{146}Sm – ^{142}Nd isochron yields an initial $^{142}\text{Nd}/^{144}\text{Nd}$ ratio of 1.141496 ± 0.000018 . Relative to our terrestrial standard this corresponds to an $\epsilon^{142}\text{Nd}$ value of -3.01 ± 0.16 . Initial $\epsilon^{142}\text{Nd}$ values for bulk Earth and chondritic meteorites are calculated using estimated bulk $^{147}\text{Sm}/^{144}\text{Nd}$ ratios that range from 0.1960 (Bouvier et al., 2008) to 0.1967 (Jacobsen and Wasserburg 1980, 1984; Amelin and Rotenberg, 2004) and $^{142}\text{Nd}/^{144}\text{Nd}$ ratios determined on terrestrial standard and chondritic meteorites (e.g., Boyet et al., 2005), respectively. The estimated initial terrestrial $\epsilon^{142}\text{Nd}$ value is -2.92 ± 0.07 , whereas the initial value inferred from chondritic meteorites is -3.13 ± 0.07 . Uncertainties include measurement uncertainties on $^{142}\text{Nd}/^{144}\text{Nd}$ ratios of ± 0.000007 . This analysis demonstrates that the initial $^{142}\text{Nd}/^{144}\text{Nd}$ inferred

from the CAI isochron is intermediate between the value inferred for the Earth and chondritic meteorites, but is within analytical uncertainty of both.

3.2 Role of nucleosynthetic anomalies in Sm–Nd chronometry

Small nucleosynthetic anomalies in Sm and Nd have recently been reported in Allende CAIs by Brennecka et al. (2013). These anomalies have the potential to affect both the inferred Sm and Nd concentrations and the $^{147}\text{Sm}/^{144}\text{Nd}$ ratios for the CAI mineral fractions because the $^{150}\text{Nd}/^{144}\text{Nd}$ and $^{149}\text{Sm}/^{152}\text{Sm}$ ratios in CAIs may not have (the assumed) terrestrial values. We addressed these effects using the average Sm and Nd isotopic compositions determined by Brennecka et al. (2013) on eleven un-spiked Allende CAIs to deconvolve our isotope dilution measurements. The magnitude of these corrections on the $^{147}\text{Sm}/^{144}\text{Nd}$ ratios was approximately 0.015%. Any isotopic anomalies, therefore, have no effect on the slope and only a extremely minor shift in the initial $^{142}\text{Nd}/^{144}\text{Nd}$ ratio determined from the ^{146}Sm – ^{142}Nd isochron.

Isotopic anomalies produced by *r*-, *s*-, and *p*-process nucleosynthesis could, in principle, cause the Sm–Nd isotopic systematics of CAIs to differ from the Sm–Nd isotopic systematics of other Solar System materials. Specifically, negative anomalies in the *p*-process isotope ^{144}Sm suggest that similar deficits could exist in other *p*-process isotopes such as ^{146}Sm . However, it is difficult to quantitatively evaluate the magnitude of potential *p*-process anomalies in a radionuclide such as ^{146}Sm because the proportion of ^{146}Sm present in the molecular cloud is dependent on the time since it was produced in a supernova. Nevertheless, *p*-process anomalies in ^{144}Sm are extremely small, ~2 parts in 10,000 (0.02%). A shift in the abundance of ^{146}Sm by 0.02% changes the initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratio by an amount encompassed by the analytical

uncertainty on the $^{146}\text{Sm}/^{144}\text{Sm}$ ratio inferred from the slope of the ^{146}Sm – ^{142}Nd isochron. Furthermore, the good agreement between the initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratio for the Solar System calculated from the Sm-Nd isotopic systematics of angrite (Lugmair and Galer, 1992; Nyquist et al. 1994) and eucrite meteorites (Boyet et al., 2010) suggest that all of these early Solar System materials are derived from a source with a common $^{146}\text{Sm}/^{144}\text{Sm}$ ratio, and that differences in $^{146}\text{Sm}/^{144}\text{Sm}$ due to nucleosynthesis cannot be distinguished with analytical uncertainty.

Although the initial $^{142}\text{Nd}/^{144}\text{Nd}$ ratio determined from the Al3S4 isochron is similar to the values derived from both chondritic meteorites and bulk Earth, it is important to note that the abundances of the other stable Nd isotopes in CAIs are measurably different (Brennecka et al., 2013). The stable Nd isotopic compositions of the Al3S4 mineral fractions are presented in Table 1 and permit nucleosynthetic effects to be evaluated for this sample. The $^{145}\text{Nd}/^{144}\text{Nd}$ and $^{148}\text{Nd}/^{144}\text{Nd}$ ratios are lower than the corresponding terrestrial values by ~0.2 epsilon units, whereas the $^{150}\text{Nd}/^{144}\text{Nd}$ ratio is lower by ~1.0 epsilon unit (Table 1), consistent with the magnitude of anomalies measured in eleven bulk Allende CAIs by Brennecka et al. (2013). A 0.14 ± 0.26 epsilon unit correction for nucleosynthetic effects on $^{142}\text{Nd}/^{144}\text{Nd}$ can be calculated using the $^{148}\text{Nd}/^{144}\text{Nd}$ data measured on the mineral fractions and the correlation of ^{148}Nd – ^{142}Nd determined from primitive chondrites (Qin et al., 2011 and Boyet and Gannoun 2013). A correction of 0.14 ± 0.14 epsilon units is calculated if the more precise $^{148}\text{Nd}/^{144}\text{Nd}$ measurements determined from bulk Allende CAIs by Brennecka et al (2013) are used. This implies that the initial $^{142}\text{Nd}/^{144}\text{Nd}$ determined from the isochron reflects a minimum estimate for the bulk Nd isotopic composition of the Solar System. However, because the errors associated with the corrections are the same size as the corrections themselves, a more definitive constraint on the initial Nd isotopic composition of the solar System is not possible. It should be noted,

however, the small deviations in the initial $^{142}\text{Nd}/^{144}\text{Nd}$ ratio of Al3S4 from the bulk Nd isotopic composition of primitive chondritic meteorites or bulk Earth will have no effect on the slope of the $^{146}\text{Sm}-^{142}\text{Nd}$ isochron. Thus, differences between the $^{146}\text{Sm}/^{144}\text{Sm}$ initial ratio calculated by Kinoshita et al. (2012) and the value determined from the Al3S4 isochron are not the result of nucleosynthetic processes that occurred prior to the formation of the Solar System.

3.3 Evaluation of the ^{146}Sm half-life and initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratio

The accuracy of the new ^{146}Sm half-life presented by (Kinoshita et al., 2012) cannot be directly evaluated using the Al3S4 $^{146}\text{Sm}-^{142}\text{Nd}$ isochron presented here because this isochron likely defines the initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratio of the Solar System. The disagreement between the initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratio determined by (Kinoshita et al., 2012) and the value determined by both (Boyet et al., 2010) and inferred from the CAI data presented here, however, suggests the 68 Ma half-life reported by Kinoshita et al. may be incorrect.

We evaluated the accuracy of the ^{146}Sm half-life determined by Kinoshita et al. (2012) by comparing the Pb–Pb and $^{147}\text{Sm}-^{143}\text{Nd}$ ages determined on meteorite and lunar samples with $^{146}\text{Sm}-^{142}\text{Nd}$ mineral isochron ages calculated for the same samples using the different ^{146}Sm half-lives. Table 3 presents $^{146}\text{Sm}-^{142}\text{Nd}$ isochron ages calculated from available Sm–Nd data in the literature using both potential ^{146}Sm half-lives and initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratios. Although the uncertainty on the 68 Ma half life is ± 14 Ma, and the uncertainty on the 103 Ma half life of ± 5 Ma, these are not taken into account in this comparison because they are not typically included in the uncertainties of most published $^{146}\text{Sm}-^{142}\text{Nd}$ ages. Figure 4 compares ages derived from the $^{146}\text{Sm}-^{142}\text{Nd}$ system with either Pb–Pb ages or $^{147}\text{Sm}-^{143}\text{Nd}$ ages determined on the same

samples. Also plotted on Figure 4 is a 1:1 line illustrating concordance between the various
 chronometers. Lines are regressed through the age data to assess which ^{146}Sm half-life and
 initial $^{146}\text{Sm}/^{144}\text{Sm}$ most closely approximates the 1:1 line. Although there is significant scatter,
 this analysis permits the two half-lives to be evaluated within the context of the published Sm–
 Nd data. From Figure 4 it is apparent that a line regressed through all of the ages determined
 using the 103 Ma half-life and initial $^{146}\text{Sm}/^{144}\text{Sm}$ of 0.00828 lies very close to the 1:1 line,
 whereas ages calculated using the 68 Ma half-life and either of the initial $^{146}\text{Sm}/^{144}\text{Sm}$ values
 does not. The ^{146}Sm – ^{142}Nd ages calculated using the 68 Ma half-life and initial $^{146}\text{Sm}/^{144}\text{Sm}$ of
 0.0094 proposed by (Kinoshita et al., 2012) are very similar to those calculated using the initial
 $^{146}\text{Sm}/^{144}\text{Sm}$ ratio of 0.00828 measured in this investigation. Thus, the observation that the 103
 Ma half-life provides a better fit to the published age data is largely independent of the initial
 $^{146}\text{Sm}/^{144}\text{Sm}$ ratio assumed in the age calculation. The concordance between the ^{146}Sm – ^{142}Nd
 isochron ages calculated using $^{146}\text{Sm}/^{144}\text{Sm} = 0.00828$ and ^{146}Sm $t_{1/2} = 103$ Ma with Pb–Pb and
 ^{147}Sm – ^{143}Nd ages determined on the same samples implies that the 103 Ma half-life provides the
 most accurate representation of the ^{146}Sm decay rate.

Note that the slopes of the regressions in Figure 4 are strongly dependent on four
 relatively young lunar sample ages. This observation stems from the inability of ancient samples
 to constrain the ^{146}Sm half-life because the uncertainty on ^{146}Sm – ^{142}Nd isochron slopes is large in
 comparison to the small difference between the isochron slopes derived from ancient samples
 and the initial Solar System $^{146}\text{Sm}/^{144}\text{Sm}$ value. For example, the ^{146}Sm – ^{142}Nd age for angrite
 LEW86010 recalculated from data presented by Nyquist et al. (1994) using the 68 Ma half-life
 and initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratio of 0.0094 is 4546^{+11}_{-12} Ma, in general agreement with the $4557.8 \pm$
 0.2 Ma Pb–Pb age for this meteorite (Galer and Lugmair, 1996; Amelin, 2008). However, an

essentially identical age of 4554^{+17}_{-19} Ma is derived using the 103 Ma half-life. It is therefore necessary to examine the ^{146}Sm – ^{142}Nd ages of relatively young samples for which measurably different amounts of ^{142}Nd ingrowth has occurred since the beginning of the Solar System in order to evaluate the accuracy of the two ^{146}Sm half-lives. Ages calculated for relatively young lunar ferroan anorthosites 60025 and 60016, as well as lunar troctolite 76535 and lunar norite 77215 (Borg et al., 2011; Marks et al., 2014; Borg et al., 2013; Carlson et al. 2013) using the 68 Ma ^{146}Sm half-life and initial Solar System $^{146}\text{Sm}/^{144}\text{Sm}$ of 0.0094 are 4406^{+19}_{-22} Ma, 4377^{+38}_{-62} Ma, 4392^{+19}_{-24} Ma, and 4412^{+38}_{-64} respectively. These ages are significantly older than the corresponding Pb–Pb and ^{147}Sm – ^{143}Nd ages of these samples: 4359 ± 3 Ma (60025), 4300 ± 32 Ma (60016), 4306 ± 11 Ma (76535), and 4283 ± 23 Ma (77215). We find far better agreement between the absolute ages of these lunar samples and the ^{146}Sm – ^{142}Nd ages calculated using the 103 Ma ^{146}Sm half-life and initial Solar System $^{146}\text{Sm}/^{144}\text{Sm}$ of 0.00828. These calculations yield ages of 4322^{+27}_{-33} Ma for 60025, 4298^{+57}_{-94} Ma for 60016, 4301^{+29}_{-36} Ma for 76535, and 4351^{+57}_{-96} Ma for 77215. This analysis supports the accuracy of the 103 Ma half-life of ^{146}Sm .

3.4 Rb–Sr isotopic systematics

Rubidium–strontium isotopic compositions were determined for the same separates of CAI Al3S4 analyzed for Sm–Nd (Table 3). The Rb–Sr data form a highly linear array (MSWD of 1.3) on an isochron plot, yielding an age of 4247 ± 110 Ma and an initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.698942 ± 0.000008 using an ^{87}Rb decay constant of 1.402×10^{-11} (Begemann et al., 2001; Fig. 5). The fines fraction does not fall along the isochron, suggesting the presence of isotopically

352 disturbed minerals in this fraction. The relatively large uncertainty in age reflects the limited
 353 spread in $^{87}\text{Rb}/^{86}\text{Sr}$ ratios relative to the precision of $^{87}\text{Sr}/^{86}\text{Sr}$ ratio measurements ($\sim 0.0007\%$;
 354 2σ). The initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio determined from the isochron is slightly lower than the Solar
 355 System initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of 0.69887 ± 0.00002 determined by (Gray et al., 1973) and the
 356 value of 0.69891 ± 0.00002 determined by (Podosek et al., 1991) from analyses of Allende CAIs
 357 (both values are renormalized to the $^{87}\text{Sr}/^{86}\text{Sr}$ for the NBS standard value used here of 0.710243
 358 ± 0.000009). Like previous Sr isotopic data from CAIs, the initial Sr isotopic composition
 359 determined from the Al3S4 data is slightly lower than the initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratios determined for
 360 the angrite meteorites Angra Dos Reis (0.698978 ± 0.000004) and LEW86010 ($0.698971 \pm$
 361 0.000008) by (Hans et al., 2013; Nyquist et al., 1994). The value inferred from Al3S4 is also
 362 substantially lower than the Basaltic Achondrite Best Initial (BABI) $^{87}\text{Sr}/^{86}\text{Sr}$ ratio of $0.699083 \pm$
 363 0.000055 (Papanastassiou and Wasserburg, 1969) or the recently revised BABI value of
 364 0.698970 ± 0.000028 (Hans et al., 2013). The fact that the measured initial $^{87}\text{Sr}/^{86}\text{Sr}$ ratio lies
 365 very near, or slightly below, previous estimates for the initial Solar System value demonstrates
 366 that Al3S4 formed very early in Solar System history in an environment that was essentially Rb-
 367 free. Note that the data for Al3S4 show an apparent ^{84}Sr excess, similar to the excesses observed
 368 in several recent studies of Allende CAIs (Table 2; Brennecka et al., 2013; Hans et al., 2013;
 369 Moynier et al., 2010). Furthermore, it has been argued that the ^{84}Sr anomaly is an artifact of an
 370 unobserved anomaly on ^{86}Sr that stems from the fractionation correction procedure that assumes
 371 $^{86}\text{Sr}/^{88}\text{Sr}$ is equal to the terrestrial value of 0.1194 . Applying a correction of 0.62 ± 0.35 epsilon
 372 units (half of the ^{84}Sr anomaly value from Hans et al., 2013) to the initial $^{87}\text{Sr}/^{86}\text{Sr}$ of CAI Al3S4
 373 shifts the initial $^{87}\text{Sr}/^{86}\text{Sr}$ from 0.698942 to 0.699004 . The adjusted value is in good, but not

perfect, agreement with the Solar System initial value estimated from CAIs and primitive meteorites by Hans et al. (2013).

Previous work has provided strong evidence for isotopic disturbance of the Rb–Sr system in Allende CAIs (Gray et al., 1973; Tatsumoto et al., 1976; Podosek et al., 1991; Moynier et al., 2010). Gray et al., (1973) determined Rb–Sr model ages (T_{BABI}) on a number of Rb-rich aggregates ($^{87}\text{Rb}/^{86}\text{Sr} > 0.3$) and found them to be as young as 3.6 Ga. They interpreted these ages to reflect disturbance of the Rb–Sr system later than 3.6 Ga, implying that alteration occurred on the Allende parent body at least 1000 Ma following CAI formation. Late formation of some secondary phases found in CAIs is consistent with the absence of radiogenic $^{26}\text{Mg}^*$ produced by in-growth of short-lived ^{26}Al (Jacobsen et al., 2011). It is possible that the radiogenic ^{87}Sr in the Al₃Si₄ mineral fractions is derived almost exclusively from secondary phases (i.e. sodalite and nepheline). Variations in $^{87}\text{Rb}/^{86}\text{Sr}$ ratios could reflect both the abundances of these phases in the various separates as well as the concentration of Sr in the primary CAI minerals in the separates.

The Rb–Sr isochron has several possible interpretations. On one hand, it might reflect mixing of a small amount of a Rb-rich contaminant into mineral fractions initially devoid of Rb and consequently having a non-radiogenic Sr isotopic composition at 4567 Ma. Analysis of Allende bulk matrix material by Tatsumoto et al. (1976), however, demonstrate that this material falls significantly above the 4.25 Ga slope line regressed through the Al₃Si₄ mineral fractions. Allende bulk matrix, therefore, cannot serve as the contaminant. Likewise, Wetherill et al. (1973) analyzed Rb–Sr systematics from a fine-grained, sodium-rich Allende CAI with a high Rb/Sr ratio ($^{87}\text{Rb}/^{86}\text{Sr} = 0.521$); the data plot well above the 4.25 Ga line. In fact, a line regressed through the Al₃Si₄ mineral fractions and the fine-grained Allende inclusion analyzed

by Wetherill et al. (1973) has a slope corresponding to 4642 ± 34 Ma (MSWD = 12). It therefore seems likely that the behavior of the Rb–Sr system in fine-grained inclusions primarily reflects fractionation of Rb/Sr occurring soon after (or during) the formation of fine-grained inclusions. Thus, simple mixing of matrix materials that have Rb–Sr isotopic systematics similar to other components in the Allende meteorite cannot produce the 4.25 Ga isochron. Instead, material with higher Rb/Sr or lower $^{87}\text{Sr}/^{86}\text{Sr}$ is required. Another interpretation involves addition of a laboratory contaminant. This scenario seems unlikely, however, because several Allende CAIs, analyzed in different laboratories, all demonstrate similar Rb–Sr isotopic systematics (Gray et al., 1973; Tatsumoto et al., 1976; Podosek et al., 1991). Perhaps the most feasible contaminant would be derived from Allende matrix but having a Rb/Sr ratio modified during metamorphism.

A final possibility is that the disturbance of the Rb–Sr isotopic system involves the addition of a small amount of very high Rb/Sr material into Al3S4 at ~4.25 Ga. In this case Rb would have been introduced in a single event or over a relatively short time interval to an object previously devoid of Rb. Due to the absence of ^{87}Rb , the Sr isotopic composition of minerals in Al3S4 would not have evolved significantly from the initial Solar System $^{87}\text{Sr}/^{86}\text{Sr}$ value. If this scenario is correct, an extremely small amount (i.e., less than 100 ng) of Rb was added to the CAI as a result of metamorphism on the Allende parent body at ~4.25 Ga. Secondary minerals, such as grossular, monticellite, sodalite, nepheline, and wollastonite, forming during metamorphism would have incorporated this Rb over a time period of hours to days. Radiogenic growth of ^{87}Sr in these minerals would then yield a Rb–Sr age reflecting the time of parent body metamorphism. However, if the alteration process added significant amounts of Sr, as well as Rb, the linear array on the Rb–Sr plot defined by the mineral fractions would reflect mixing, rather than ingrowth of ^{87}Sr . -In any case, the Rb–Sr age of 4.25 Ga suggests that there was a late

metamorphic event on the Allende parent asteroid, although the timing of this event is not uniquely defined.

3.5 Chronology and metamorphism in CAIs

The observation that the event that introduced Rb into the CAI did not significantly affect other chronometers provides insight into metamorphic processes on the Allende parent asteroid. The extremely low concentration of Rb in primary CAI minerals ($<0.06\text{--}0.23\text{ }\mu\text{g/g}$; Table 2) makes the Rb–Sr system extremely sensitive to late addition of small amounts of Rb. In contrast, Sm and Nd have significantly higher abundances in CAIs and are refractory elements, difficult to redistribute by metamorphic processes that effectively mobilize Rb and other more soluble elements. As a consequence, the Sm–Nd system has preserved a record of crystallization in CAI Al₃S₄. The Pb–Pb chronometer, like the Rb–Sr chronometer, is susceptible to contamination during metamorphism. However, sequential leaching processes are commonly applied for Pb–Pb chronometry, specifically to remove secondary Pb added during metamorphism. As a result, Pb–Pb ages of CAIs are derived from mineral fractions that do not contain significant amounts of extraneous Pb and are consequently less likely to reflect the effects of post-crystallization metamorphic processes. In summary, crystallization ages of moderately altered CAIs may be obtained using the Pb–Pb, Al–Mg, and Sm–Nd systems, but are unlikely to be obtained from the Rb–Sr isotopic system.

4. Conclusions

440 A ^{147}Sm – ^{143}Nd age of 4560 ± 34 Ma and an initial $^{143}\text{Nd}/^{144}\text{Nd}$ of 0.506657 ± 43 was determined
 441 for mineral fractions from the coarse-grained type B Allende inclusion Al3S4. This is the first
 442 age determined on a CAI that is not based on an assumed initial isotopic composition of the
 443 Solar System and is in good agreement with previously determined Al–Mg and Pb–Pb ages of
 444 CAIs. This concordance, combined with an initial $^{143}\text{Nd}/^{144}\text{Nd}$ value similar to initial Solar
 445 System values from bulk meteorites (e.g., Bouvier et al., 2008; Amelin and Rottenberg, 2004;
 446 Jacobsen and Wasserburg, 1980), suggests that the Sm–Nd isotopic systematics of Al3S4 are
 447 minimally disturbed. A ^{146}Sm – ^{142}Nd isochron with a slope corresponding to an initial
 448 $^{146}\text{Sm}/^{144}\text{Sm} = 0.00828 \pm 0.00044$ was obtained from the same mineral fractions. This initial
 449 ratio is in excellent agreement with the initial $^{146}\text{Sm}/^{144}\text{Sm}$ value of the Solar System determined
 450 by Boyet et al. 2010, but measurably different from the value of 0.0094 calculated by Kinoshita
 451 et al. (2012) using the newly determined 68 Ma ^{146}Sm half-life. All published ^{146}Sm – ^{142}Nd
 452 isochron ages were recalculated using both the 68 Ma and 103 Ma half-lives in conjunction with
 453 the initial $^{146}\text{Sm}/^{144}\text{Sm}$ of 0.00828 determined here and the value of 0.0094 proposed by
 454 Kinoshita et al. (2012). This evaluation demonstrates that ^{146}Sm – ^{142}Nd ages calculated using the
 455 103 Ma half-life and initial $^{146}\text{Sm}/^{144}\text{Sm}$ of 0.00828 reproduce Pb–Pb and ^{147}Sm – ^{143}Nd ages
 456 determined on the same samples significantly better than ages calculated using the 68 Ma half-
 457 life and either of the initial $^{146}\text{Sm}/^{144}\text{Sm}$ values. This analysis is strongly based on several young
 458 lunar samples because these samples yield initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratios lying significantly outside
 459 analytical uncertainty of the Solar System initial value. Revision of the ^{146}Sm half-life to the
 460 previous value of 103 Ma has significant implications for the timing of terrestrial, lunar, and
 461 Martian planetary silicate mantle differentiation, requiring planetary differentiation to have
 462 occurred somewhat later than would be predicted from calculations based on the 68 Ma value.

The Rb–Sr systematics of CAI Al₃S₄ are clearly disturbed, but form a well defined linear array with a slope corresponding to 4.25 Ga. The significance of this age is not clear, although it could represent the age of alteration on the Allende parent asteroid if metamorphism involved the addition of a very small amount of Rb, and essentially no Sr, into Al₃S₄.

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Table 1. A13S4 CAI Sm-Nd isotopic data

Fraction	Wt (mg)	Sm (ppm)	Sm (ng) ^e	Nd (ppm)	Nd (ng) ^e	$\frac{147\text{Sm}}{144\text{Nd}}$ a,d	$\frac{142\text{Nd}}{144\text{Nd}}$ a,b	$\frac{143\text{Nd}}{144\text{Nd}}$ a,b	$\frac{145\text{Nd}}{144\text{Nd}}$ a,b	$\frac{148\text{Nd}}{144\text{Nd}}$ a,b	$\frac{150\text{Nd}}{144\text{Nd}}$ a,b
Mel-Pyx	70.95	4.202	298	13.54	961	0.18751 ± 19	1.141814 ± 4	0.512332 ± 1	0.348395 ± 1	0.241573 ± 1	0.236430 ± 2
Melilite	142.51	1.936	276	6.852	976	0.17076 ± 17	1.141788 ± 4	0.511825 ± 1	0.348395 ± 1	0.241573 ± 1	0.236442 ± 2
Pyx	11.93	7.26	87	18.91	226	0.23204 ± 52	1.141894 ± 5	0.513665 ± 3	0.348394 ± 1	0.241577 ± 1	0.236433 ± 2
d>3.26	54.23	6.262	340	18.13	983	0.20878 ± 21	1.141849 ± 3	0.512986 ± 1	0.348397 ± 1	0.241568 ± 1	0.236421 ± 2
d<3.26	49.12	2.802	138	9.863	484	0.17172 ± 19	1.141789 ± 11	0.511855 ± 4	0.348395 ± 1	0.241571 ± 3	0.236433 ± 7
Fines	152.82	2.874	44	9.141	1397	0.19006 ± 19	1.141819 ± 4	0.512479 ± 1	0.348397 ± 1	0.241568 ± 1	0.236438 ± 2
JNdi Nd Std (N=37)					500		1.141837 ± 7 ^c	0.512101 ± 7 ^c	0.348402 ± 4	0.241578 ± 5	0.236456 ± 6

Abbreviations for mineral fractions are as follows: Mel. = melilite, Pyx = pyroxene, Mel-Pyx = melilite separated from the pyroxene fraction. All samples and standards run as Nd⁺.

^a. Error limits apply to last digits and include a minimum uncertainty of 0.5% plus 50% of the blank correction for Sm and Nd added quadratically.

^b. Normalized to $^{146}\text{Nd}/^{144}\text{Nd} = 0.7219$. Uncertainties refer to last digits and are $2\sigma_m$ ($2 \times$ standard error of measured isotopic ratios).

^c. Uncertainties refer to last digits and are $2\sigma_p$ ($2 \times$ standard deviation of population of mass spectrometry runs on isotopic standard). Isochron are calculated using either using either $2\sigma_p$ (from standard runs) or $2\sigma_m$ (from measured isotopic ratios).

^d. $^{147}\text{Sm}/^{144}\text{Nd}$ corrected for nucleosynthetic effects using Sm and Nd isotopic compositions measured on 11 bulk Allende CAIs by Brennecke et al. (2011, 2012, 2013). Total effect is < 0.015%.

^e. Abundances of elements calculated for entire mineral fractions. 95% splits were analyzed for Nd isotopic composition determinations and 5% splits were analyzed for Nd and Sm concentrations.

Table 2. Al3S4 CAI Rb-Sr isotopic data

Fraction	Wt (mg)	Rb (ppm) ^a	Rb (ng) ^b	Sr (ppm) ^a	Sr (ng) ^b	⁸⁷ Rb/ ⁸⁶ Sr ^{a,c}	⁸⁷ Sr/ ⁸⁶ Sr ^d	$\epsilon^{84}\text{Sr} (\pm 2\sigma)^f$
Melilitite	142.51	0.064	9.1	242.8	34601	0.00076 ± 1	0.698987 ± 8	0.98 ± 0.19
d<3.26	49.12	0.231	11.3	224.4	11023	0.00298 ± 3	0.699129 ± 8	1.24 ± 0.22
Fines	152.82	0.123	18.8	116.5	17804	0.00305 ± 3	0.699110 ± 8	
Pyx	11.93	0.056	0.7	46.3	553	0.00351 ± 12	0.699152 ± 8	1.34 ± 0.67
Mel-Pyx	70.95	0.234	16.6	115.4	8188	0.00587 ± 6	0.699306 ± 8	1.30 ± 0.21
d>3.26	54.23	0.211	11.5	86.2	4675	0.00709 ± 7	0.699373 ± 8	0.51 ± 0.20
NBS-987 Std (N=49)							0.710249 ± 9 ^e	

Abbreviations for mineral fractions are as follows: Mel. = melilitite, Pyx = pyroxene, Mel-Pyx = melilitite separated from the pyroxene fraction. The long term reproducibility of ⁸⁷Rb/⁸⁷Rb measured on 30 runs of NBS-984 Rb standard = 2.603 ± 17 and are 2σ_p (2×standard deviation of population of mass spectrometry runs on isotopic standard) and were used to correct for instrument mass fractionation.

- ^a. Error limits apply to last digits and include a minimum uncertainty of 1% plus 50% of the blank correction for Rb and Sr added quadratically.
^b. Abundances of elements calculated for entire mineral fractions. 95% splits were analyzed for Rb and Sr concentration determinations and 5% splits were analyzed for Sr isotopic composition.
^c. Uncertainties refer to last digits and are 2σ_p. (2 x standard deviation of population of mass spectrometry runs on isotopic standard).
^d. Isochron are calculated using either 2σ_p (from standard runs) or 2σ_m (from measured isotopic ratios), whichever is larger.
^e. Normalized to ⁸⁶Sr/⁸⁸Sr = 0.1194. Uncertainties refer to last digits and are 2σ_m (2×standard error of measured isotopic ratios).

$$\epsilon_{\text{Sr}}^{84} = \left(\frac{\left(\frac{\text{Sr}^{84}}{\text{Sr}^{86}} \right)_{\text{sample}}}{\left(\frac{\text{Sr}^{84}}{\text{Sr}^{86}} \right)_{\text{standard}}} - 1 \right) \times 10^4.$$

Table 3. ^{146}Sm - ^{142}Nd ages of primitive meteorites and lunar samples calculated using different parameters (Sm-Nd).

Meteorite/ sample type	Sample Name	Absolute Age (Ma)	Absolute Age Reference ^a	Initial $^{146}\text{Sm}/^{144}\text{Sm}$	Sm-Nd age ^b (Ma)	Sm-Nd age ^c (Ma)	Sm-Nd age ^d (Ma)	^{146}Sm - ^{142}Nd Ref
CV3	CAI	4567.1 ± 0.9			(+) (-)	(+) (-)	(+) (-)	
Angrites	LEW 86010	4558 ± 0.5	Pb-Pb ¹	0.0076 ± 0.0009	4554 19 17	4559 12 11	4546 12 11	Nyquist et al., 1994
	LEW 86010	4558 ± 0.5	Pb-Pb ¹	0.0071 ± 0.0017	4544 41 32	4552 27 21	4539 27 21	Nyquist et al., 1994
	Angra dos Reis	4512 ± 85	Sm-Nd ²	0.0063 ± 0.0017	4527 47 35	4540 31 23	4528 31 23	Lugmair & Marti (1977)
Euclrites	Caldera	4544 ± 19	Sm-Nd ⁵	0.0074 ± 0.0013	4549 29 25	4555 19 16	4543 19 16	Wadhwa & Lugmair (1996)
	EET87520	4547 ± 9	Sm-Nd ⁶	0.0069 ± 0.0045	4540 157 75	4549 104 49	4537 104 49	Lugmair et al. (1991)
	EET90020	4482 ± 30	Sm-Nd ^{7,8}	0.0048 ± 0.0020	4486 80 52	4514 53 34	4501 53 34	Nyquist et al. (1997a, 1997b)
	Ibitira	4557.4 ± 0.55	Pb-Pb ⁹	0.0091 ± 0.0028	4580 55 40	4576 36 26	4563 36 26	Prinzhofer et al. (1992)
	Ibitira	4557.4 ± 0.55	Pb-Pb ⁹	0.0082 ± 0.0008	4566 15 14	4566 10 9	4554 10 9	Nyquist et al. (1999)
	Moama	4466 ± 42	Sm-Nd ³	0.0041 ± 0.0013	4463 57 41	4498 37 27	4486 37 27	Jacobsen and Wasserburg
	Moore county	4484 ± 19	Pb-Pb ¹⁰	0.0066 ± 0.0012	4533 30 25	4545 20 16	4532 20 16	Boyett et al. (2010)
	Y980318	4567 ± 24	Sm-Nd ¹¹	0.0060 ± 0.0009	4519 24 21	4535 16 14	4523 16 14	Nyquist et al. (2004); Nyquist et al. (1994)
	Y980433	4542 ± 42	Sm-Nd ¹²	0.0057 ± 0.0005	4512 14 12	4530 9 8	4518 9 8	Nyquist et al. (1994)
Mesosiderite	Mt. Padbury	4494 ± 45	Sm-Nd ¹⁴	0.0056 ± 0.0008	4509 22 19	4529 15 13	4516 15 13	Stewart et al. (1994)
	Acapulco	4556.4 ± 3	Pb-Pb ¹⁶	0.0070 ± 0.0018	4542 43 34	4551 29 22	4538 29 22	Prinzhofer et al. (1992)
Lunar	Lunar 60025	4359 ± 3	Pb-Pb ¹⁷	0.0016 ± 0.0003	4321 33 27	4405 22 18	4392 22 18	Borg et al. (2011)
	Lunar 60016	4300 ± 32	Sm-Nd ¹⁸	0.0003 ± 0.0001	4298 60 102	4389 40 67	4377 40 67	Marks et al. (2014)
	Lunar 76535	4307 ± 11	Sm-Nd ¹⁹	0.0014 ± 0.0003	4300 36 29	4391 24 19	4378 24 19	Borg et al. (2013)
	Lunar 77215	4283 ± 23	Sm-Nd ²⁰	0.0019 ± 0.0009	4351 97 58	4424 64 38	4412 64 38	Carlson et al. (2013)

All ^{146}Sm - ^{142}Nd ages calculated from reference age of CAI of 4567 Ma. Absolute ages are either Pb-Pb or ^{147}Sm - ^{143}Nd . (+) and (-) values represent two standard deviations. Samples with ages older than 4567 Ma are not plotted in Figure 4.

- 1 Lugmair and Galer, (1992); 2 Lugmair & Marti (1977); 3 Jacobsen & Wasserburg (1984); 4 Boyett et al. (2010) 5 Wadhwa & Lugmair (1996); 6 Lugmair et al. (1991); 7,8 Nyquist et al. (1997a, 1997b); 9 Amelin et al. (2006); 10 Tera et al. (1997); 11 Nyquist et al. (2004); Nyquist et al. (1994); 13 Prinzhofer et al. (1992); 14 Stewart et al. (1994); 15 Sharma et al. (1995); 16 Amelin et al. (2005); 17 Borg et al. (2011); 18 Marks et al. (2014); 19 Borg et al. (2013); 20 Carlson et al. (2013)
- ^{146}Sm $t_{1/2}$ = 103 Ma, $^{146}\text{Sm}/^{144}\text{Sm}$ = 0.00828.
- ^{146}Sm $t_{1/2}$ = 68 Ma, $^{146}\text{Sm}/^{144}\text{Sm}$ = 0.00828.
- ^{146}Sm $t_{1/2}$ = 68 Ma, $^{146}\text{Sm}/^{144}\text{Sm}$ = 0.0094.

Figure Captions:

Figure 1. Photomicrograph of Allende CAI Al3S4 from Clayton et al. (1977). The outer 1.5 mm border consists of radially oriented melilite crystals, with spinel content increasing inwards. The interior of the inclusion is primarily crystalline clinopyroxene, melilite, and anorthite with included euhedral spinel. Field of view is 22mm x 17mm.

Figure 2. A ^{147}Sm - ^{143}Nd isochron plot of mineral fractions from Allende CAI Al3S4 yielding an age of 4560 ± 34 million years and an initial $^{143}\text{Nd}/^{144}\text{Nd}$ of 0.506657 ± 43 . Isochron has a mean squared weighted derivative of 2.4. The Fines fraction is plotted as an open symbol and is excluded from the regression. Inset represents deviation of individual points from the isochron in epsilon units:

$$\epsilon_{\text{Nd}}^{143} = \left(\frac{(\text{Nd}^{143}/\text{Nd}^{144})^{\text{sample}}}{(\text{Nd}^{143}/\text{Nd}^{144})^{\text{standard}}} - 1 \right) \times 10^4.$$

Figure 3. A ^{146}Sm - ^{142}Nd isochron plot of mineral fractions from Allende CAI Al3S4. The slope of this isochron yields an initial $^{146}\text{Sm}/^{144}\text{Sm}$ ratio of the Solar System of 0.00828 ± 0.00044 . The y-intercept defines an initial $^{142}\text{Nd}/^{144}\text{Nd}$ ratio of 1.141496 ± 0.000018 ($\epsilon^{142}\text{Nd} = -3.01 \pm 0.16$), which is in good agreement with the initial $^{142}\text{Nd}/^{144}\text{Nd}$ ratios calculated for both chondritic meteorites ($\epsilon^{142}\text{Nd} = -3.13 \pm 0.07$; Boyet et al., 2005) and Earth from our Nd standard ($\epsilon^{142}\text{Nd} = -2.92 \pm 0.07$) using $^{147}\text{Sm}/^{144}\text{Nd} = 0.1960$ to 0.1967 (Bouvier et al., 2008; Jacobsen and Wasserburg, 1980, 1984) and assuming a half-life of $^{146}\text{Sm} = 103$ Ma. Inset represents deviation of individual points from the isochron in epsilon units:

$$\epsilon_{\text{Nd}}^{142} = \left(\frac{(\text{Nd}^{142}/\text{Nd}^{144})^{\text{sample}}}{(\text{Nd}^{142}/\text{Nd}^{144})^{\text{standard}}} - 1 \right) \times 10^4.$$

Figure 4. Comparison of ^{146}Sm - ^{142}Nd ages and Pb-Pb and ^{147}Sm - ^{143}Nd ages. The ^{146}Sm - ^{142}Nd ages are calculated assuming A) ^{146}Sm $t_{1/2} = 103$ Ma and initial solar system $^{146}\text{Sm}/^{144}\text{Sm} = 0.00828$ (filled circles), B) ^{146}Sm $t_{1/2} = 68$ Ma and initial solar system $^{146}\text{Sm}/^{144}\text{Sm} = 0.0094$ (gray triangles), and C) ^{146}Sm $t_{1/2} = 68$ Ma and initial solar system $^{146}\text{Sm}/^{144}\text{Sm} = 0.00828$ (open squares). The lines represent best fits to the data. The regression for the 103 Ma half-life lies closest to the 1:1 line representing perfect agreement between the different chronometers.

Figure 5. A ^{87}Rb - ^{86}Sr isochron plot of Allende CAI Al3S4 mineral fractions yielding an age of 4247 ± 110 million years. Age calculated using a decay constant of $\lambda = 0.01402 \text{ Ga}^{-1}$ (Begemann et al., 2001). The Fines fraction is plotted as an open symbol and is excluded from the regression. Inset represents deviation of individual points from the isochron in epsilon units

$$\epsilon_{\text{Sr}}^{87} = \left(\frac{(\text{Sr}^{87}/\text{Sr}^{86})^{\text{sample}}}{(\text{Sr}^{87}/\text{Sr}^{86})^{\text{standard}}} - 1 \right) \times 10^4.$$

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5