Primordial formation of major silicates in a protoplanetary disc with homogeneous $^{26}$Al/$^{27}$Al

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Understanding the spatial variability of initial $^{26}$Al/$^{27}$Al in the solar system, i.e., ($^{26}$Al/$^{27}$Al)$_{0}$, is of prime importance to meteorite chronology, planetary heat production, and protoplanetary disc mixing dynamics. The ($^{26}$Al/$^{27}$Al)$_{0}$ of calcium-aluminum–rich inclusions (CAIs) in primitive meteorites ($\approx 5 \times 10^{-5}$) is frequently assumed to reflect the ($^{26}$Al/$^{27}$Al)$_{0}$ of the entire protoplanetary disc, and predicts its initial $^{26}$Mg/$^{24}$Mg to be $\approx 35$ parts per million (ppm) less radiogenic than modern Earth (i.e., $\Delta^{26}$Mg$_{0} = -35$ ppm). Others argue for spatially heterogeneous ($^{26}$Al/$^{27}$Al)$_{0}$, where the source reservoirs of most primitive meteorite components have lower ($^{26}$Al/$^{27}$Al)$_{0}$ at $\approx 2.7 \times 10^{-5}$ and $\Delta^{26}$Mg$_{0}$ of $\approx 16$ ppm. We measured the magnesium isotope compositions of primitive meteoritic olivine, which originated outside of the CAI-forming reservoirs, and report five grains whose $\Delta^{26}$Mg$_{0}$ are within uncertainty of $\approx 35$ ppm. Our data thus affirm a model of a largely homogeneous protoplanetary disc with ($^{26}$Al/$^{27}$Al)$_{0}$ of $\approx 5 \times 10^{-5}$, supporting the accuracy of the $^{26}$Al/$^{26}$Mg chronometer.

INTRODUCTION

The discovery of correlated $^{26}$Mg/$^{24}$Mg with Al/Mg in refractory inclusions in primitive meteorites (1)—chondrites—bore witness to the previous presence of live $^{26}$Al ($^{26}$Al$\rightarrow^{26}$Mg; $\lambda_{1/2} = -0.730$ million years (Ma); see the Supplementary Materials) in the nascent solar system, in abundances sufficient to drive melting and metamorphism in planetesimals (2), and provide a valuable high-resolution chronometer of early solar system processes (2, 3). Moreover, the inferred ($^{26}$Al/$^{27}$Al)$_{0}$ was sufficiently high to place important constraints on the birth environment of the solar system and the processes that mixed together in the solar nebula and protoplanetary disc [see (4)].

Solar system ($^{26}$Al/$^{27}$Al)$_{0}$ has largely been derived from analyses of “normal” calcium-aluminum–rich inclusions (CAIs): ultrarefractory condensates found in unequilibrated chondrites that are the oldest dated objects in the solar system (5, 6), and whose age, with a weighted mean of 4567.30 ± 0.16 Ma, is commonly taken to represent “time zero” of solar system history. Their antiquity and high elemental Al/Mg ratios enable precise determination of ($^{26}$Al/$^{27}$Al)$_{0}$. These works (7–9) have yielded a canonical ($^{26}$Al/$^{27}$Al)$_{0}$ of $\approx 5.3 \times 10^{-5}$ that is frequently assumed to reflect the ($^{26}$Al/$^{27}$Al)$_{0}$ of the solar system as a whole.

Canonical ($^{26}$Al/$^{27}$Al)$_{0}$ is one order of magnitude higher than the galactic background, as measured by γ-ray spectroscopy (10), indicating that $^{26}$Al was injected into the nascent solar system from its stellar source (11) shortly before or just after the formation of the protoplanetary disc. This may not have allowed sufficient time for $^{26}$Al to be spatially homogenized before the CAIs formed. Heterogeneity in solar system ($^{26}$Al/$^{27}$Al)$_{0}$ is evident in some rare refractory objects (12)—namely, some FUN (fractionation and unidentified nuclear isotope effects) CAIs (13), BAGs (blue aggregates), and PLACs (platy crystal fragments) (14)—which contain no evidence for live $^{26}$Al. This observation is commonly interpreted to indicate that these inclusions formed before $^{26}$Al was injected into the protoplanetary disc (15). These unusual objects preserve an early window into the solar system mixing, but we believe are not representative of the bulk of material in the protostellar disc. In this study, we focus only on the solar system’s evolution after the condensation of normal CAIs.

Nonetheless, even normal CAIs (hereafter referred to as CAIs) are demonstrably anomalous in their isotopic compositions of many elements relative to the material that comprises bulk meteorites and the terrestrial planets (16, 17). It is therefore reasonable to question whether ($^{26}$Al/$^{27}$Al)$_{0}$ determined from CAIs is representative of the solar system as a whole. Spatially heterogeneous ($^{26}$Al/$^{27}$Al)$_{0}$ within the protoplanetary disc would compromise the utility of the $^{26}$Al/$^{26}$Mg decay system for dating early solar system processes, as Al-Mg chronometry traditionally assumes the same ($^{26}$Al/$^{27}$Al)$_{0}$ in CAIs and the object being dated. Much of the understanding of early solar system chronology was developed from the Al-Mg chronometer, so assessing the robustness of its underlying assumptions is of paramount importance. Previous attempts to assess spatial ($^{26}$Al/$^{27}$Al)$_{0}$ homogeneity using concordance between Al-Mg and other radioisotope chronometers have yielded conflicting conclusions (18–21).

Consequently, there has been much interest in trying to establish independent constraints on whether or not ($^{26}$Al/$^{27}$Al)$_{0}$ was spatially homogeneous in the protoplanetary disc. An important perspective is provided by the evolution of the radiogenic daughter isotope ratio, $^{26}$Mg/$^{24}$Mg, with time. For ($^{26}$Al/$^{27}$Al)$_{0}$ = 5.32 × 10$^{-5}$ (8), the initial solar system $^{26}$Mg/$^{24}$Mg, expressed in linearized delta notation as $\Delta^{26}$Mg$_{0}$ [see (22) and the Supplementary Materials], should be $\approx 34.7 \pm 1.4$ ppm in order for chondritic meteorite reservoirs to evolve to their modern compositions (Fig. 1). We refer to this as the “canonical model.”

The most recent, highest-precision analyses of bulk refractory inclusions in chondrites define an isochron slope in keeping with previous studies, ($^{26}$Al/$^{27}$Al)$_{0}$ of (5.26 ± 0.01) × 10$^{-5}$, but $\Delta^{26}$Mg$_{0}$ of $\approx 15.8 \pm 1.2$ ppm (9); this initial $\Delta^{26}$Mg$_{0}$ implies that bulk CI chondrites (Ivuna-like carbonaceous chondrites, which are the chondrite group thought to best represent the bulk solar system composition) had a reduced ($^{26}$Al/$^{27}$Al)$_{0}$ of (2.71 ± 0.21) × 10$^{-5}$ to evolve to their modern $\Delta^{26}$Mg$_{0}$ (Fig. 1). Calculations of ($^{26}$Al/$^{27}$Al)$_{0}$ for bulk ordinary and bulk enstatite chondrites based on their modern $\Delta^{26}$Mg and $^{26}$Al/$^{24}$Mg, assuming that they each had $\Delta^{12}$Mg$_{0}$ of $\approx 15.8$ ppm (fig. S1), also yield
similarly subcanonical (26Al/27Al)0. These observations seemingly provide evidence for differences in (26Al/27Al)0 between the portion of the protoplanetary disc that condensed CAIs and that which contributed to the bulk chondrites and that which contributed to bulk chondrules. Uncertainty bars/areas are ±2 SE.

RESULTS

The refractory nature of RFs is evident in their highly forsteritic compositions (Fo98.5) and elevated refractory element contents compared to most chondrule olivine and also AOAs (Fig. 3A). With Δ17O (mass-independent oxygen isotope composition; see the Supplementary Materials) of ~−5.6‰, they are 16O poor compared to CAIs and AOAs (Fig. 3B) but are similar to bulk chondrules from carbonaceous chondrites (32).

RFs have Δ26Mg0 ranging from 8.1 ± 2.7 to −40.2 ± 16.9 ppm (Fig. 3C). Critically, 4 (of 13) of our RFs have Δ26Mg0 values that are significantly lower than the lowest possible Δ26Mg0 of −15.8 ± 1.2 ppm of the AOA-CAI model (9), while none are lower than the lowest possible Δ26Mg0 of −34.7 ± 1.4 ppm of the canonical model (Fig. 3C). Because of the low Al/Mg of these objects, this holds true even if the minor amount of 26Mg ingrowth is corrected for. The Δ26Mg0 model ages of RFs, calculated relative to the Δ26Mg evolution curve (Fig. 1), range from −0.14 ± 0.40 to >4 Ma after CAIs (Fig. 4A). The oldest RFs (i.e., lowest Δ26Mg0) all have high refractory element concentrations (Fig. 3, C and D), whereas, in the younger samples, refractory element abundances decrease to those of more typical chondrule olivines.

At the same time, we would anticipate no values less than −34.7 ± 1.4 ppm if the canonical model is correct.

A challenge for this crucial test is to identify for analysis sufficiently old olivine that formed outside of the CAI-forming reservoir(s). Given that Δ26Mg can routinely be measured at the University of Bristol to a precision of ±5.0 ppm (2 SE) for the small amounts of magnesium available in individual olivine grains (typically <5 μg for an olivine grain of ~200 μm), we can only differentiate olivines that have formed before the two modeled curves converge to within ~5.0 ppm of one another. This corresponds to a time of formation no later than ~1.4 Ma after CAIs.

Previously, the magnesium isotope evolution of the early solar system has been investigated using in situ measurements of olivine dated in chondrules (25)—quenched melt droplets that formed in the protoplanetary disc that are the dominant component of primitive meteorites (26)—but these grains were too young, given the precision of analysis, to resolve the two scenarios illustrated in Fig. 1 (see also fig. S1). Rather than analyze typical chondrule olivine, here, we target refractory forsterite grains (RFs) in unequilibrated carbonaceous chondrites. RFs are volumetrically minor (27) but ubiquitous in unequilibrated chondrites occurring in three petrographic settings: as (i) isolated grains in chondrite matrix that formed via fragmentation of preexisting chondrules (28), (ii) in situ phenocrysts in magnesium-rich (“type I”) chondrules (27) (Fig. 2), and (iii) so-called relict grains in the cores of olivine phenocrysts in iron-rich (“type II”) chondrules, which represent unmolten chondrule precursors (29).

The eponymous feature of these grains is their high-Mg/(Mg + Fe) and relatively high, but still trace, concentrations of refractory elements Al, Ti, and Ca in their structure compared to more common meteoritic olivine (30). These characteristics are compatible with their formation at an early stage of disc evolution in high-temperature, low-fO2 conditions (31). Moreover, their petrographic relationships with later-formed silicates (29), namely, their presence as “relict” grains in some type II chondrules, show that they predate at least some chondrules. So, although they are not absolutely dated, RFs are demonstrably older than at least some chondrules and therefore preserve isotopic information from the solar system’s earliest history.

Fig. 1. Illustration of two Δ26Mg evolution models for chondrite parent bodies. The canonical model (purple curve), consistent with widespread (26Al/27Al)0 homogeneity, uses the modern composition of CI chondrites (9, 37, 47) and (26Al/27Al)0 of (5.32 ± 0.11) × 10−5 (8, 9) to yield Δ26Mg0 = −34.7 ± 1.4 ppm. Ordinary chondrites (OC) and enstatite chondrites (EC), two major classes of chondrites, yield statistically identical Δ26Mg0 based on their modern compositions (9, 37, 47). (ii) The alternative “AOA-CAI” model (orange curve) assumes Δ26Mg0 of −15.8 ± 1.2 ppm (9), consequently requiring (26Al/27Al)0 of a factor of ~2 lower than the canonical model to evolve to modern CI composition, reflecting (26Al/27Al)0 heterogeneity between the portion of the protoplanetary disc that condensed CAIs and that which contributed to bulk chondrites. Uncertainty bars/areas are ±2 SE.
DISCUSSION

While there is oxygen isotope heterogeneity among CAIs, the majority from the least equilibrated (i.e., most petrologically pristine) chondrites have isotopically uniform $\Delta^{17}$O at $\approx -24\%$, likely reflecting the composition of their source reservoir(s) (33). Chondrules have a range in $\Delta^{17}$O, clustering between $\Delta^{17}$O of $\approx -8\%$ and $+2\%$. It is therefore reasonable to use the $\Delta^{17}$O of RFs to genetically link them with the chondrule-forming region(s) and distinguish them from the region(s) of the solar system that condensed CAIs. Although $\Delta^{17}$O variability is commonly argued to result from photochemical reactions within the solar system (34), meteorites show covariations of $\Delta^{17}$O with a range of mass-independent isotope anomalies that reflect variable inputs from different stellar sources (35). Why isotopic anomalies with such different origins covary is not well understood, but empirically, $\Delta^{17}$O is a good proxy for heterogeneous distribution of pre-solar material in the nebula. The $\Delta^{17}$O measurements of our RFs link them to the reservoir of material that formed the major silicate component of chondrites, including chondrules (Fig. 3B).

The idea that four RFs have $\Delta^{26}$Mg$_0$ lower than the lowest possible value predicted by the “CAI-AOA model” argues against this model’s general applicability and strengthens concerns that inclusion of AOAs and CAIs on the same isochron is ill advised. Rather, these four most unradiogenic RFs have $\Delta^{26}$Mg$_0$ within uncertainty of $-34.7 \pm 1.4$ ppm, the value for the solar system at the onset of CAI formation, as calculated using canonical ($^{26}$Al/$^{27}$Al)$_0$ for CI chondrites (Fig. 1). Because no RF has $\Delta^{26}$Mg$_0$ significantly lower than this “canonical” value, it seems unlikely that their distinctive magnesium isotopic compositions are of a nucleosynthetic origin (i.e., isotope anomalies inherited from heterogeneously distributed pre-solar carriers). While possible in principle, it would seem implausibly serendipitous for these nucleosynthetic compositions to fit exactly in the small window predicted by independently constrained radiogenic decay.

Previously, a positive array of correlating $^{26}$Mg and $^{54}$Cr anomalies in bulk meteorites and CAIs (9, 36) was argued to track coupled heterogeneous distribution of ($^{26}$Al/$^{27}$Al)$_0$ and stable nucleosynthetic anomalies in the protoplanetary disc. The purported correlation was strongly pinned by a model $\Delta^{26}$Mg$_0$ for the “CAI-AOA reservoir,” derived from the intercept of the CAI-AOA isochron (9). As discussed above, our measurements argue against the validity of this value. Moreover, subsequent work on bulk chondrites has illustrated that their variable $\Delta^{26}$Mg can be explained by their variable Al/Mg from a common canonical initial $\Delta^{26}$Mg and $^{26}$Al/$^{27}$Al (37, 38). Thus, the arguments made in (9) appear no longer relevant. It has become
Fig. 3. The chemical and isotopic compositions of RFs compared to CAIs, AOAIs, chondrules, and both $\Delta^{26}\text{Mg}$ evolution models. (A) RFs ($\text{Fo}_{90-98}$) are Ca-rich relative to AOAIs and CAIs. (B) Oxygen isotope compositions similar to bulk carbonaceous chondrite (CC) chondrules distinguish RFs from AOAIs and CAIs, linking them to the major silicates in chondrites. We show the primitive chondrule mineral (PCM) line (48), the terrestrial fractionation line, and a fractionation line at $\Delta^{17}\text{O} = -5.6\%$ around which our RF data cluster. Measured $\Delta^{26}\text{Mg}$ of RFs relative to the end-member $\Delta^{26}\text{Mg}_0$ models (vertical bars) plotted against (C) calcium and (D) aluminum concentrations. Four RFs are well resolved from the AOA-CAI model. All uncertainties are ±2 SE (omitted on literature data and smaller than symbols for our oxygen data). Literature references are given in the Supplementary Materials.

The similarity of RFs to chondrules in terms of their oxygen isotope compositions, and their presence as large phenocrysts in type I chondrules, suggests that RFs are the products of crystallization from parental melts—i.e., they are the products of crystallization of chondrule-like objects—rather than direct gas-solid condensates. This is consistent with the view that RFs crystallized from condensed silicate melts at high-temperature and low-$f_O_2$ conditions (27). Therefore, one interpretation of the model ages of RFs is that they represent the crystallization of refractory element–rich condensed melts (i.e., refractory element–rich chondrule-forming events).

The range in model ages of RFs indicates either a protracted period of formation over ~4 Myr or early formation followed by variable reequilibration with an evolving nebula. This latter notion is in keeping with ideas of continued chondrule reprocessing and interaction with nebula gas [e.g., (41)]. The continuum of RF $\Delta^{26}\text{Mg}$ model ages from values as old as CAIs to several Ma younger is consistent with single-chondrule Pb-Pb ages (6, 42) but contrasts with the marked peak in relatively young ages for internal Al-Mg isochrons for single chondrules (Fig. 4, B and C). We attribute the ~2 Myr offset between Al-Mg ages of CAIs and chondrules, evident in literature data, to the effects of transient thermal events (43) in the protoplanetary disc that reset Al-Mg internal isochrons but incompletely reset the Pb-Pb chronometer. We suggest that these thermal events largely ceased ~2 to 3 Ma after CAIs, resulting in most chondrules recording this age in their internal Al-Mg ages. Most internal Al-Mg isochrons of chondrules are pinned by high-Al/Mg phases [e.g., small plagioclase (<20 µm) or microcrystalline mesostasis], which are both more fusible
and have faster solid-state magnesium diffusion than the larger RFs (~100 μm). Chondrule ages are thus more readily reset than model \( \Delta^{26}\text{Mg}_{\text{ODM-3}} \) isotope ages in RFs. While the internal Al-Mg isochrons in chondrules may constrain the timing of thermal events in the protoplanetary disc, we suggest that they likely do not represent formation ages.

Although RFs formed within at least ~300,000 years of CAIs, they have very different \( \Delta^{17}\text{O} \), illustrating that large-scale oxygen isotope heterogeneities were established early in the solar system. This suggests that the process(es) that produced these differences [e.g., photodissociation of CO (34, 44)] was highly efficient or that there was preexisting \( \Delta^{17}\text{O} \) heterogeneity in the protosolar molecular cloud that was not homogenized by the time CAI formation began.

Our inference of common \( ^{26}\text{Al}/^{27}\text{Al} \)_0 (at ~5.3 × 10\(^{-5}\)) between CAIs and the major silicate phases from the terrestrial planets—and asteroid-forming reservoirs—supports the underlying assumption of the \( ^{26}\text{Al} \rightarrow ^{26}\text{Mg} \) dating system and therefore reaffirms its validity as a widely applicable, high–temporal resolution, early solar system chronometer. Moreover, the remarkable antiquity of RFs, calculated from their \( ^{26}\text{Mg} \), demonstrates an important before-unseen consistency with chondrule formation ages determined by the extant \( ^{207}\text{Pb} \rightarrow ^{206}\text{Pb} \) system (6), another cornerstone of early solar system chronology.

**MATERIALS AND METHODS**

We targeted polished sections of two unequilibrated chondrites (primitive meteorites that did not experience high degrees of thermal metamorphism or aqueous alteration on their parent asteroids) in this study: Northwest Africa 4502, a type 3 (45) oxidized Vigarano-like carbonaceous chondrite (CV3 ox), and Felix, a type 3.3 (46) Ornans-like carbonaceous chondrite (CO3.3) borrowed from the Natural History Museum, London (identification number: P13341). Candidate grains were identified and imaged using scanning electron microscopy (backscattered electrons and x-ray energy-dispersive spectroscopy) at the University of Bristol (UK), and their in situ chemical composition was measured using electron probe microanalysis (EPMA) at the University of Bristol. Oxygen isotopes were measured in situ via secondary ionization mass spectrometry at CRPG (Nancy, France). Before ex situ magnesium isotope measurements, each RF was excavated from its host section using a newly developed technique that combines laser excavation and microsampling. Magnesium isotope compositions were measured ex situ via multicollector inductively coupled plasma source mass spectrometry (MC-ICP-MS) at the University of Bristol. These measurements were conducted using a modified protocol that allows for small masses of magnesium (<5 μg) to be measured to high precision (typically better than ±3 ppm on \( ^{26}\text{Mg}_{\text{ODM-3}} \), ±2 SE). The reader is referred to the Supplementary Materials for the detailed analytical and microsampling protocols.
Table S1. A summary of the chemical composition and 27 Al/24 Mg of RFs measured in situ by secondary ion mass spectrometry.

Fig. S16. Detailed scanning electron microscope image of RF C21 (NWA 4502).

Fig. S13. Detailed scanning electron microscope image of RF C4 (NWA 4502).

Fig. S9. Detailed scanning electron microscope image of RF C9 (Felix).

Fig. S8. A summary of false-color K content/full/6/11/eaay9626/DC1

Table S2. A summary of the oxygen isotope composition of RFs measured in situ by secondary ion mass spectrometry.

Fig. S4. Microexcavation of an RF.

6. The heating of solids in the variable 238 U/235 U.

7. The distribution of U-Pb, Rb-Sr and Sm-Nd.

8. The distribution of the initial solar system 26 Al/27 Al.

9. The distribution of 26 Al cross section: Implication for the heterogeneous distribution of 26 Al in the solar protoplanetary disk.

10. The distribution of the Mg isotopic composition of RFs measured in situ by secondary ion mass spectrometry.

11. The distribution of Mg isotopic anomalies in forsterites from the Murchison (C2) and Allende (C3) carbonaceous chondrites.

12. The distribution of magnesium isotope compositions—Implications for the formation of the early solar system.

13. The distribution of Mg and Al isotopic homogenization in the early inner Solar System.

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