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Episodic formation of refractory inclusions in the Solar System and their presolar heritage

K.K. Larsen a,*, D. Wielandt a, M. Schiller a, A.N. Krot a, M. Bizzarro a

a Centre for Star and Planet Formation, Natural History Museum of Denmark, University of Copenhagen, Copenhagen DK-1350, Denmark
b Hawai’i Institute of Geophysics and Planetology, University of Hawai’i at Manoa, HI 96822, USA

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ABSTRACT

Refractory inclusions [Ca-Al-rich inclusions (CAIs) and Amoeboid Olivine Aggregates (AOAs)] in primitive meteorites are the oldest Solar System solids. They formed in the hot inner protoplanetary disk and, as such, provide insights into the earliest disk dynamics and physicochemical processing of the dust and gas that accreted to form the Sun and its planetary system. Using the short-lived 26Al to 26Mg decay system, we show that bulk refractory inclusions in CV (Vigarano-type) and CR (Renazzo-type) carbonaceous chondrites captured at least two distinct 26Al-rich (26Al/27Al ratios of ~5 × 10^-5) populations of refractory inclusions characterized by different initial 26Al/26Mg isotope compositions (μ26Mg0). Another 26Al-poor CAI records an even larger μ26Mg0 deficit. This suggests that formation of refractory inclusions was punctuated and recurrent, possibly associated with episodic outbursts from the accreting proto-Sun lasting as short as ∼8000 yr. Our results support a model in which refractory inclusions formed close to the hot proto-Sun and were subsequently redistributed to the outer disk, beyond the orbit of Jupiter, plausibly via stellar outflows with progressively decreasing transport efficiency. We show that the magnesium isotope signatures in refractory inclusions mirrors the presolar grain record, demonstrating a mutual exclusivity between 26Al enrichments and large nucleosynthetic Mg isotope effects. This suggests that refractory inclusions formed by incomplete thermal processing of presolar dust, thereby inheriting a diluted signature of their isotope systematics. As such, they record snapshots in the progressive sublimation of isotopically anomalous presolar carriers through selective thermal processing of young dust components from the proto-Solar molecular cloud. We infer that 26Al-rich refractory inclusions incorporated 26Al-rich dust which formed <5 Myr prior to our Sun, whereas 26Al-poor inclusions (such as FUN- and PLAC-type CAIs) incorporated >10 Myr old dust.

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1. Introduction

Our Solar System formed through the collapse of a well-mixed molecular cloud core made up of dust and gas originating from a multitude of nucleosynthetic sources (Vasileiadis et al., 2013; Kuffmeier et al., 2016). This nucleosynthetic diversity is preserved in presolar grains from primitive meteorites. Through gravitational core collapse, the earliest energetic phase of protostellar evolution was characterized by high mass accretion rates onto the forming proto-Sun (∼10^-5 M⊙ yr^-1) (D’Alessio et al., 2005), punctuated by protostellar episodic outbursts (Frimann et al., 2016; Hsieh et al., 2018). This resulted in a very hot inner protoplanetary disk. Based on their high-temperature mineralogy and irradiation signatures (Wielandt et al., 2012; Sossi et al., 2017), refractory inclusions [Ca-Al-rich inclusions (CAIs) and Amoeboid Olivine Aggregates (AOAs)] are thought to have condensed at this stage from a gas in the innermost region of the protoplanetary disk (Scott and Krot, 2014). As such, CAIs from CV (Vigarano-type) carbonaceous chondrites record an absolute age of 4,567.30 ± 0.16 Myr (Connelly et al., 2012), defining the onset of our Solar System. Hence, refractory inclusions captured the earliest physicochemical processing of the presolar dust that accreted to form the Sun and its planetary system.

Our understanding of the formation of refractory inclusions primarily comes from petrographic, chemical, and isotopic analysis of the large, mm- to cm-sized CAIs in CV chondrites (MacPherson, 2014). Except for a few rare refractory inclusions, CV CAIs and AOAs generally exhibit excesses in 26Mg that correlate with their 27Al/24Mg ratio, signifying the former presence of short-lived radioactive 26Al with an initial 26Al/27Al0 ratio of (5.25 ± 0.02) × 10^-5 (Larsen et al., 2011). This has been interpreted to reflect formation of these inclusions within less than ∼8000 yr from a
CAI-forming gas enriched in $^{26}$Al via chemical processing of $^{26}$Al-rich presolar carriers (Larsen et al., 2011). The idea of enrichment of $^{26}$Al in the CAI-forming reservoir is supported by correlated minor enrichments in neutron-rich isotopes of the iron peak elements ($^{48}$Ca, $^{50}$Ti, $^{54}$Cr) in $^{26}$Al-rich CV CAIs; interpreted to be the result of selective thermal processing of isotopically anomalous presolar carriers (Trinquier et al., 2009; Larsen et al., 2011; Schiller et al., 2015, 2018). Much larger nucleosynthetic effects are known to exist in $^{26}$Al-poor FUN/UN-type (Fractionation and Unidentified Nuclear effects) CAIs from CV chondrites, as well as $^{26}$Al-poor PLAC-type CAIs (PLATy hibonite Crystals) from CM chondrites, indicating residual nucleosynthetic variability in some refractory inclusions (Ireland, 1990; Liu et al., 2009; Köpp et al., 2016, 2018). It has been proposed to result from incomplete chemical homogenization of isotopically diverse presolar dust components and that refractory inclusions, contrary to current belief, did not form exclusively from a single, completely homogenized gas of solar composition (Ireland, 1990). These observations are not compatible with complete sublimation of all presolar material into a gaseous state prior to CAI-formation. As such, it is not clear if the $^{26}$Al-$^{26}$Mg isotope systematics of $^{26}$Al-rich CV-type CAIs and AOAs are representative for refractory inclusions in other types of carbonaceous chondrites or if they merely represent a snapshot of isotopically evolving protoplanetary gas and dust reservoirs.

It is well known that features such as size, type, mineralogy and abundance of refractory inclusions differ between different groups of carbonaceous chondrites (Scott and Krot, 2014), which are believed to have accreted at various heliocentric distances from the Sun in the outer Solar System (Warren, 2011). For example, in CR chondrites (Renazzo-type carbonaceous chondrites) refractory inclusions are typically very small (10–300 μm), about an order of magnitude less abundant (<1 vol%) than in CV chondrites and on average mineralogically distinct (Krot et al., 2002). Thus, it is conceivable that their isotope systematics also record differences in their formation conditions and reveal whether refractory inclusions in different chondrite groups represent distinct populations with unique isotope signatures. Importantly, CR chondrites are some of the most pristine early Solar System materials that escaped secondary asteroidal thermal metamorphism and experienced only minor aqueous alteration (Krot et al., 2002; Makide et al., 2009). As such, CR CAIs represent ideal targets to search for primordial isotopic differences to other CAIs that is associated with their formation process. Here, we report on the first high-precision Multiple-Collector Inductively-Coupled-Plasma source Mass-Spectrometer (MC-ICP-MS) located at the Centre for Star and Planet Formation (Natural History Museum of Denmark, University of Copenhagen), following procedure outlined in Bizzarro et al., 2011 and Larsen et al., 2018. In brief, purified Mg was introduced into the plasma source by means of an Apex IR desolvating nebulizer in a 2% HNO$_3$ run solution at 50 ml min$^{-1}$ using a Thermo Fisher Jet sample cone and skimmer X-cone interface in high-resolution mode. Corrections for instrumental mass bias were obtained by sample-standard bracketing with sample/standard peak intensities matching to better than 5%. Samples and standards were analyzed 10 times with each analysis consisting of 100 cycles of 16.78 second integrations separated by a total of 922 seconds of on-peak zero blank measurements in clean 2% HNO$_3$ solution.

Stable magnesium isotope ratios are reported in parts per million (ppm) deviation relative to the composition of the Mg reference standard DTS-2B (Olsen et al., 2013) according to: $^{25}$Mg$^*$ = $\left(\frac{[^{25}\text{Mg}]/[^{24}\text{Mg}]_{\text{sample}}}{[^{25}\text{Mg}]/[^{24}\text{Mg}]_{\text{DTS-2B}}} - 1\right) \times 10^6$. The mass-independent $^{25}$Mg$^*$ (non-radiogenic $^{26}$Mg + radiogenic $^{26}$Mg ingrowth from $^{26}$Al decay) is reported in the same fashion after mass bias correction using the exponential fractionation law and internal normalization to $^{25}$Mg$^{24}$Mg = 0.126896 (Bizzarro et al., 2011). Data reduction was conducted offline using the software package lollite (Paton et al., 2011) and is reported in Table 1.

$^{27}$Al/$^{24}$Mg ratios were determined using the Thermo Fisher Scientific quadrupole ICP-MS located at the Centre for Star and Planet Formation (Natural History Museum of Denmark, University of Copenhagen), by standard-sample bracketing on sample-matched gravimetric standards with $^{27}$Al/$^{24}$Mg ratios of 0.1, 1.3, 3.4 and 5.7, thereby covering the range of $^{27}$Al/$^{24}$Mg ratios for refractory inclusions reported in this study. Repeat measurements of two basaltic USGS standards (BHVO-2 and BCR-2) and two CAIs (CP1 and CP2) showed that accurate $^{27}$Al/$^{24}$Mg ratios, as compared to spiked $^{27}$Al/$^{24}$Mg ratios reported in Paton et al., 2012 and nominal USGS values, can be obtained to a precision of ~2% (Fig. S1). Data reduction was conducted using the software package lollite (Paton et al., 2011).
3. 26Al-26Mg isotope systematics of refractory inclusions from CR and CV chondrites.

In the 26Al-26Mg isochron diagram, two FG CV CAIs (EK12 and EK14) plot on the bulk CV CAI-AOA isochron ([26Al/27Al]0 = (5.252 ± 0.019) × 10−5, μ26Mg0 = −15.9 ± 1.4 ppm) previously determined for refractory inclusions in the reduced CR chondrite Efremovka (Larsen et al., 2011), while all of the CR CAIs and one FG CV CAI (EK3) plot consistently below it (Fig. 1a). Three of the CR AOA plots on the CV chondrite, while another three CR AOA plots slightly above it. Refining the CV isochron using the old data including the two new FG CV CAI results in an isochron with a (26Al/27Al)0 ratio of (5.252 ± 0.018) × 10−5 and μ26Mg0 of −15.9 ± 1.4 ppm (MSWD = 0.98), i.e. identical to that reported in Larsen et al., 2011. A regression solely using bulk CV CAIs (i.e. excluding AOA) also defines an isochron with an indistinguishable (26Al/27Al)0 ratio of (5.26 ± 0.05) × 10−5 and intercept of μ26Mg0 = −17.8 ± 9.6 ppm. This is distinct from the isochron intercept of −34 ppm predicted for a homogeneous distribution of 26Al in the Solar protoplanetary disk (Kita et al., 2013). Mineral fractions from the KT-1 FUN CV CAI record significant deficits in μ26Mg* corresponding to a μ26Mg0* value of −545 ± 300 ppm with no evidence for the former presence of 26Al, i.e. (26Al/27Al)0 of (0.7 ± 1.0) × 10−5 (Fig. 1a).

Fine-grained spinel-rich CV and CR refractory inclusions have textural and mineralogical characteristics consistent with their primary condensation origin, which is also corroborated by the pristine internal inclusions for FG CV CAIs [average (26Al/27Al)0 ratio of (5.1 ± 0.2) × 10−5 (MacPherson et al., 2012)], i.e. consistent with the (26Al/27Al)0 ratio defined by bulk CV CAIs. Other, typically coarse-grained igneous CAI types record (26Al/27Al)0 ratios as low as −4.3 × 10−5, suggesting their later resetting up to ~200,000 yr after formation of FG CAIs (MacPherson et al., 2012). CR CAIs are typically fragmented in nature such that individual fragments may not reflect their true bulk 26Al-26Mg isotope systematics (Simon and Grossman, 2006), especially for coarse-grained igneous CAIs. Therefore, we restrict our analysis to the five fine-grained CR CAIs to constrain the primary condensation history for CR CAIs.

An isochron using these data results in an (26Al/27Al)0 ratio of (5.1 ± 0.2) × 10−5, consistent with the bulk CV isochron and internal inclusions for CR CAIs recording (26Al/27Al)0 of (4.4–5.4) × 10−5 (Macike et al., 2009), but with lower initial μ26Mg0 of −95 ± 26 ppm, i.e. 79 ± 26 ppm lower than the CV isochron. Four of the re-molten CR CAIs analyzed here are not well-constrained by the CR isochron, a feature that we attribute to their fragmental nature and, hence, unrepresentative sampling of their bulk composition as mentioned above.

While the initial μ26Mg0* values determined from internal 26Al-26Mg CAI isochrons measured by secondary ion mass-spectrometry are generally poorly constrained, one CR CAI [with the inferred (26Al/27Al)0 of (5.4 ± 0.6) × 10−5] shows a resolvable μ26Mg0* deficit of −400 ± 300 ppm (Macike et al., 2009), i.e. within error of the bulk CR CAI isochron. Moreover, a high-precision internal isochron for a type B CV CAI (Egg-3; Wasserburg et al., 2012) has identical (26Al/27Al)0 and μ26Mg0 to the CR isochron (Fig. 1a). Similarly, a few Fo8 (Forsterite-rich type B) CAIs (MacPherson et al., 2016) record identical μ26Mg0* values within error to that defined here for CR CAIs. We further note that another bulk CV CAI (EK3) measured in this study plots on the CR isochron. When comparing the isochron intercepts, μ26Mg0* versus their slope, 26Al/27Al for these inclusions, (Fig. 1b) it is evident that the 26Al-26Mg isotope composition of such CV and CR CAIs cannot be explained by their formation from an evolving reservoir constrained by the CV isochron. Instead they must have formed from a reservoir characterized by non-Solar Mg-isotope composition with a lower μ26Mg0 similar to that defined here for CR CAIs (unrelated to radiogenic decay of 26Al). Thus, μ26Mg0 in such inclusions record both a radiogenic 26Mg component related to 26Al decay and a non-radiogenic 26Mg component.

4. Episodic formation of refractory inclusions and their transport to the accretion region of the giant planets.

The distinct μ26Mg0* values for bulk refractory inclusions in CV and CR chondrites indicate at least two separate CAI-forming reservoirs/episodes with (26Al/27Al)0 of ~5 × 10−5. The three CR
Fig. 1. a) $^{26}$Al-$^{26}$Mg isochron diagram for bulk refractory inclusions from CR and CV chondrites showing the presence of at least two distinct populations of refractory inclusions with significantly different $\mu^{26}$Mg*0 (where $\mu^{26}$Mg*0 is the intercept of the isochron defined as the parts per million deviation of the initial $^{26}$Mg/$^{24}$Mg ratio relative to Earth after mass-dependent fractionation correction using the exponential law and normalization to the solar $^{25}$Mg/$^{24}$Mg ratio of 0.126896; Bizzarro et al., 2011). Also shown is the internal mineral isochron for the Egg-3 CV3 CAI from Wasserburg et al., 2012, which coincides with the CR isochron. b) Magnesium isotope evolution diagram for refractory inclusions, in which each data point represents the slope ($^{26}$Al/$^{27}$Al) and intercept ($\mu^{26}$Mg*0) in the $^{26}$Al-$^{26}$Mg isochron diagram. The red shaded area represents the range of Mg isotope compositions that can be explained by in-situ $^{26}$Al decay from a “canonical” CV CAI reservoir ($^{26}$Al/$^{27}$Al = 5.25 × 10^{-5} and $\mu^{26}$Mg*0 = −15.9 ppm, reservoir A), along trajectories determined by their bulk $^{27}$Al/$^{24}$Mg ratios. The green shaded area represents the evolution from a CR CAI reservoir for $^{27}$Al/$^{24}$Mg ratios up to 2 typical for forsterite-rich type B (FoB) CAIs. Most CV CAIs (MacPherson et al., 2010; 2012; Kita et al., 2012) can be explained by evolution from the CV reservoir, whereas most FoB CAIs (MacPherson et al., 2016), the type B CAI Egg-3 (Wasserburg et al., 2012) and an igneous CR CAI (Makide et al., 2009) require evolution from a distinct isotope reservoir with deficient $\mu^{26}$Mg* similar to CR CAIs (reservoir B, this study). Hence, these inclusions belong to a separate population characterized by isotopically anomalous $\mu^{26}$ Mg* relative to “normal” CV CAIs. To facilitate comparison, literature data was recalculated using $\beta = 0.511$. (For interpretation of the colors in the figure(s), the reader is referred to the web version of this article.)
AOAs that plot above the CV isochron cannot be regressed with any other AOAs from this study to define a statistically meaningful regression line. Thus, we speculate that these objects hint at the possibility of yet another population of refractory inclusions with a different initial $^{26}\text{Mg}^\text{+}$ although additional data is required to test this hypothesis. Thus, the existence of different CAI populations with contrasting $\mu^{28}\text{Mg}_0^\text{+}$ suggests that formation of $^{26}\text{Al}$-rich refractory inclusions was a punctuated and recurrent process, plausibly associated with protostellar episodic outbursts from the active proto-Sun (Frimann et al., 2016; Hsieh et al., 2018). Such episodic outbursts are observed for young stellar objects in the process of accreting mass via their disks from the surrounding envelope with episodes lasting for less than a few thousand years (Hsieh et al., 2018), which is consistent with the timeframe deduced from the error envelopes of CV and CR isochrons of $<8,000$ and $<82,000$ yr, respectively. Therefore, episodic accretion and CAI-formation may be intrinsically linked, reflecting a generic process inherent to the evolution of protostars.

The presence of CAI material in the comet 81P/Wild 2 has been taken as evidence for CAI transport and recycling into distant regions of the Solar protoplanetary disk (Brownlee et al., 2006). The near absence of such refractory materials in the inner accretion regions of the terrestrial planets (Trinquart et al., 2009; Olsen et al., 2016; Bizzarro et al., 2017) can be explained if refractory inclusions were episodically lofted above the disk midplane and recycled back into the outer disk. Carbonaceous chondrites, such as CV and CR, accreted in the outer volatile-rich part of the disk beyond the orbits of Jupiter, with the CR chondrites most likely accreting at larger heliocentric distances compared to the close proximity of the accretion region of CV chondrites to the orbit of Jupiter (Van Kooten et al., 2016, 2017; Kruijser et al., 2017). If correct, the fact that our new data supports evidence for formation of two distinct populations of refractory inclusions, which now resides in two different types of chondrites (CV and CR), suggests that refractory inclusions belonging to the CV isochron (population A) were predominantly transported to disk regions just beyond Jupiter. On the other hand, population B inclusions (all CR CAIs and some CV CAIs) were present in both the CV and CR accretion regions (Fig. 2). In other words, CR chondrites only captured the inclusions belonging to population B, whereas CV chondrites captured inclusions of both populations A and B. This can be explained if population B inclusions were transported to larger orbital distances than those from population A. Successive inward drift could have introduced inclusions from population B to the CV accretion region, but not vice versa (Fig. 2). Following this reasoning, the high abundance ($\sim$85%) and small size of $^{26}\text{Al}$-poor CAIs in CH chondrites, inferred to have accreted even beyond CR chondrites (Krot et al., 2012) implies that such $^{26}\text{Al}$-poor inclusions were transported further out in the disk, plausibly at much earlier times. Variable travel distance of refractory inclusions can plausibly be explained by secular changes in the outward transport efficiency of the disk winds associated with episodic outbursts (Audard et al., 2014) and/or size-sorting effects during transport above the disk midplane and subsequently within the outer disk (Hansen, 2014). The latter possibility seems consistent with the size distribution of refractory inclusions in CV and CR chondrites, for which the characteristically large inclusions present in CV chondrites are expected to re-enter the disk midplane at smaller orbital radii, as well as drift faster inwards compared to the much smaller inclusions typical of CR chondrites.

This distribution of refractory inclusions is also consistent with recent numerical simulations showing that an early formation ($<1$ Myr) of Jupiter can trap CAIs in a pressure maximum beyond it (Desch et al., 2018). In this model, the high abundance of CAIs in CV chondrites is explained by trapping and locally enhancing their modal abundance at the far-Sun side of Jupiter. CR parent bodies accreting at larger orbital distances do not experience this enhancement and consequently contain a lower abundance of CAIs. Apart from the size and abundance differences of refractory inclusions in CV and CR chondrites, the presence of both population A and B in CV chondrites contrasting the apparent exclusive presence of population B in CR chondrites (Figs. 1 and 2) supports this idea. Further, it is generally believed that the isotopically anomalous composition of $^{26}\text{Al}$-poor inclusions, such as FUN CAIs, requires their formation prior to sufficient isotopic homogenization and admixing of $^{26}\text{Al}$ to the CAI-forming reservoir (Holst et al., 2013). Hence, the predominance of $^{26}\text{Al}$-poor refractory inclusions at larger heliocentric distances compared to $^{26}\text{Al}$-rich inclusions suggests that the transport efficiency was progressively decreasing. As such, refractory inclusions record the dynamical evolution of the Solar protoplanetary disk, where populations of inner disk refractory inclusions are periodically lofted above the disk midplane and redistributed to the outer Solar System through large-scale mass transport processes with progressively decreasing efficiency.

5. The presolar heritage of isotopic signatures in refractory inclusions

Through continuous Galactic Chemical Evolution (GCE), stellar nucleosynthesis progressively enriches $^{25}\text{Mg}$ and $^{26}\text{Mg}$ relative to $^{24}\text{Mg}$ in the interstellar medium (Fenner et al., 2003). We note that
this results in an anticorrelation between 25Mg/24Mg and μ26Mg*,
trending towards μ26Mg* deficits in young dust generations com-
pared to the average dust – a trend which is also corroborated
by type-II supernova (SNIll) models (Fig. 3). Thus, it appears un-
avoidable that presolar dust generations of different age on average
record divergent Mg-isotope compositions (Clayton, 1988).
Presolar grains from pristine chondritic meteorites are sam-
ple of the molecular cloud dust that made up our Solar Sys-
tem. We find that the presolar grain data for graphites, silicates
and oxides agree with the theoretical predictions from GCE and
SNIll models for an anticorrelation between 25Mg/24Mg and μ26Mg*;
thereby reflecting the galactically inherited non-radiogenic 26Mg*
background (Fig. 4a). Nucleosynthetic addition of 26Al would
produce a radiogenic spike in its decay product 26Mg* in young
dust through radioactive decay, thereby overprint this galactic
background. Indeed, a clear 26Mg* spike is observed for presolar
grains with about solar 25Mg/24Mg. This is most noticeably in low-
density presolar graphite grains believed to originate from SNIll
(Lodders and Amari, 2005) and group 3 and 4 oxide grains orig-
inating from either SNIll or low-mass and low-metalllicity Asymp-
totic Giant Branch (AGB) stars (Lodders and Amari, 2005; Nittler
et al., 2008) (Fig. 4a). It is also consistent with the Mg-isotope sys-
tematics in SNIll presolar SiC grains of the X type (Lodders and
Amari, 2005). Thus, the presolar grain record demonstrates a mu-
tual exclusivity between large 25Mg/24Mg excesses (coupled with
μ26Mg* deficits) and evidence for 26Al, and reveal three main dust
components/generations; 1) an ancient galactically inherited
component with excess μ26Mg* (coupled with deficit 25Mg/24Mg), 2)
a newer molecular cloud component with deficit μ26Mg* (cou-
pled with excess 25Mg/24Mg), and 3) a young 26Al-rich component
with about solar non-radiogenic μ26Mg* (coupled with about solar
25Mg/24Mg) (Fig. 4a inset). Hence, if refractory inclusions in-
deed formed by incomplete vaporization of such dust components,
they are predicted to capture diluted signatures of its isotopically
anomalous compositions. The distinct Mg-isotopic signatures of re-
fractory inclusions reported here indicate progressive admixing of
exotic presolar dust to the hot inner disk CAI-forming region.

For example, a characteristic feature of FUN-type CAIs is the
presence of large mass-independent nucleosynthetic isotope effects
in 48Ca, 50Ti and 54Cr, as well as 26Mg* deficits coupled with the
general lack of evidence for the former presence of 26Al (Schiller
et al., 2015; Köpp et al., 2018; Lee et al., 1979; Holst et al., 2013).
The KT-1 FUN CV CAI reported here (µ25Mg* = −545 ± 300
ppm, [26Al/27Al]⊙ < 1.7 × 10−5) is an example of this. Although
the extreme 25Mg/24Mg enrichments typical for FUN-type refrac-
tory inclusions could in principle be explained by kinetic mass-
dependent fractionation effects (Davis et al., 2015), these processes
cannot account for their large μ26Mg* deficits, down to ∼−4000
ppm (Makide et al., 2009; Köpp et al., 2016), and not either those
observed in 26Al-poor PLAC-type CAIs (Liu et al., 2009; Köpp
et al., 2016). This is because the range of beta values (0.511–0.513)
xperimentally determined for Mg isotope fractionation through
evaporation of such CAI compositions cannot account for such
large deficits (e.g. Davis et al., 2015). Instead, we propose that
these features can be explained if FUN- and PLAC-type refractory
inclusions formed from a gas dominated by vaporized new, but
26Al-poor (i.e. > 10 Myr prior to Solar System formation), presolar
dust characterized by µ25Mg* deficits (component 2 in Fig. 4a).
As such, these types of CAI’s may represent a diluted signature of
the more extreme presolar component 2, for which FUN-type inclusions
potentially record a secondary mass-fractionation overprint
(Fig. 4b).

On the other hand, 26Al-rich refractory inclusions [with (26Al/27Al]⊙
of ∼5 × 10−5], such as ‘normal’ CV CAIs and SHIB-type (Spinel-HiBonite
inclusions) CAIs in CM chondrites lack the extreme nucleosynthetic effects seen
in FUN/UN CAIs and PLACs (Fig. 3b), thereby mirroring the mutual exclusivity between
26Al and μ26Mg* deficits recorded by presolar grains (Fig. 4). The formation of 26Al-rich refractory inclusions can be explained by their formation from a gas dominated by vaporized young dust
(component 3 in Fig. 4a inset) rich in 26Al and of roughly solar non-radiogenic μ26Mg* and solar 25Mg/24Mg. Thus, these 26Al-
rich CAI types seem to represent diluted signatures of the more
extreme presolar component 3. If this dust originated from SNIll
in the protosolar molecular cloud, as indicated by the preso-
lar grain record (Fig. 4a), we find that the average free-decay age of
this dust, resulting from the difference between the [26Al/27Al]⊙
ratio of ∼5 × 10−5 in ‘normal’ CAIs and the average [26Al/27Al]⊙
yields of ∼5 × 10−5 from SNIll nucleosynthesis models of sol-
lar metallicity massive stars (Ms = 30–35) with near solar
25Mg/24Mg (Fig. S11 in the supplementary material), is less than 5
Myr depending on the relative proportion of SNIll-derived 26Al in
CAIs. Hence, this dust was less than 5 Myr old on average when it became incorporated into 26Al-rich CAIs.

Finally, we note that all of the fine-grained CR CAI condens-
ates measured in this study record solar to super-solar 25Mg/24Mg
(up to ∼800 ppm; Table 1). This is unexpected from theore-
tical predictions for condensation from a solar composition gas,
in which the lighter isotopes are preferentially partitioned into
the condensate (Davis and Richter, 2014). This suggests that
the CR CAI-forming gas was characterized by a slight enrichment in
25Mg/24Mg (>800 ppm) and deficit in μ26Mg* (<−5 ppm) rela-
tive to the average solar composition. This further mirrors the
anticorrelated 25Mg/24Mg and μ26Mg* observed for the presolar
grains record and suggest that the CR CAI-forming gas was of
slightly non-solar composition generated via preferential selec-
tive admixing of isotopically anomalous young dust components
(2 & 3).

6. Thermal processing and the distribution of Mg isotopes and
26Al in the early Solar System

The CAI-forming gas was generated by thermal processing of
presolar dust accreting to the proto-Sun. However, the presence of
widespread nucleosynthetic isotope anomalies in a range of ele-
ments (e.g., Mg, Al, Cr, Ca, Ti) in refractory inclusions is not com-
patible with a homogeneous solar composition gas (Ireland, 1990).
As such, dust sublimation was incomplete prior to formation of these solids. We infer that this gas composition was governed by the thermal properties of isotopically anomalous presolar carriers. Our analysis suggests that populations of refractory inclusions formed through episodic transient heating events, thereby capturing distinct snapshots of an isotopically evolving CAI-forming reservoir. Some CAIs (PLACs and FUN/UN CAIs) clearly preserved nucleosynthetic ‘nugget’ effects (% range for $^{48}$Ca and $^{50}$Ti [Köpp et al., 2018]) from refractory oxide residues rich in Ca and Ti (e.g., presolar hibonite and corundum), whereas nucleosynthetic effects in more volatile elements, such as Mg (enriched in presolar silicates such as pyroxene and olivine), were significantly reduced ($^{25}$Mg/$^{26}$Mg$_0$ < 0.5%; Fig. 4b). These observations corroborate a volatility-controlled sublimation of mineralogically diverse presolar carriers, implying a closer to solar composition in the gas for volatile elements, such as Mg, than those of refractory elements such as Ca and Ti.

Extraction of new dust components (2 & 3), carrying $^{26}$Mg$^*$ deficits and $^{26}$Al/$^{27}$Al enrichments, into the CAI-forming gas suggests a relatively volatile mineralogy. This generates a complementarity in the residual thermally processed midplane dust in the inner protoplanetary disk, resulting in a net non-radiogenic $^{26}$Mg$^*$ excess coupled with a $^{25}$Mg/$^{24}$Mg and $^{26}$Al/$^{27}$Al deficit relative to the solar average (Fig. 5). This dust accretes to form the terrestrial planets, thereby providing a mechanism for explaining the reduced initial ($^{26}$Al/$^{27}$Al)$_0$ in the inner solar protoplanetary disk relative to CI chondrites (Larsen et al., 2011; 2016; Schiller et al., 2015). The $^{26}$Mg$^*$ deficit recorded by inner Solar System bodies (such as ordinary chondrites and angrites [Larsen et al., 2011]) can therefore only be explained by a reduced radiogenic $^{26}$Mg$^*$ ingrowth relative to the solar average represented by CI chondrites (Fig. 5 and Fig. S12 in the supplementary material).

We conclude that the Mg-isotope systematics of the Solar System can be justified by chemical unwinding by thermal processing of the three molecular cloud components; 1) an ancient galactically inherited and thermally resilient $^{26}$Al-poor presolar dust component enriched in non-radiogenic $^{26}$Mg$^*$, which became enriched in the inner terrestrial planet-forming region by virtue of loss of some of the other components, 2) a newer $^{26}$Al-poor thermally
labile dust component deficient in non-radiogenic $^{26}$Mg, plausibly inherited from the proto-solar molecular cloud >10 Myr prior to Solar System formation, which is recorded in FUN/UN/PLAC-type CAIs, and 3) a freshly synthesized (on average <5 Myr old) $^{26}$Al-rich molecular cloud component, which was enriched in the CAI-forming gas and subsequently incorporated into $^{26}$Al-rich refractory inclusions, such as ‘normal’ CV, CR and SHIB-type CAIs.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary material

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References