



Correlated nanoscale characterization of a unique complex oxygen-rich stardust grain: Implications for circumstellar dust formation

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Received 4 October 2016; accepted in revised form 4 May 2017; available online 11 May 2017

Abstract

We report the light to intermediate-mass element abundances as well as the oxygen, magnesium, silicon, and titanium isotope compositions of a unique and unusually large ($0.8 \mu\text{m} \times 3.75 \mu\text{m}$) presolar O-rich grain from the Krymka LL3.2 chondrite. The O-, Al-, and Ti-isotopic compositions are largely compatible with an origin from an asymptotic giant branch (AGB) star of 1.5 solar masses with a metallicity that is 15% higher than the solar metallicity. The grain has an elevated $^{17}\text{O}/^{16}\text{O}$ ratio ($8.40 \pm 0.16 \times 10^{-4}$) compared to solar, and slightly sub-solar $^{18}\text{O}/^{16}\text{O}$ ratio ($1.83 \pm 0.03 \times 10^{-3}$). It shows evidence for the presence of initial ^{26}Al , suggesting formation after the first dredge-up, during one of the early third dredge-up (TDU) episodes. Titanium isotopic data indicate condensation of the grain before significant amounts of material from the He-burning shell were admixed to the stellar surface with progressive TDUs. We observed a small excess in ^{30}Si ($\delta^{30}\text{Si} = 41 \pm 5\%$), which most likely is inherited from the parent star's initial Si-isotopic composition. For such stars stellar models predict a C/O-ratio < 1 even after the onset of TDU, thus allowing the condensation of O-rich dust.

The grain is an unusual complex presolar grain, consisting of an Al-Ca-Ti-oxide core, surrounded by an Mg-Ca-silicate mantle, and resembles the condensation sequence for a cooling gas of solar composition at pressures and dust/gas ratios typically observed for circumstellar envelopes around evolved stars. We also report the first observation of phosphorus in a presolar grain, although the origin of the P-bearing phase remains ambiguous.

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Keywords: Astrochemistry; Circumstellar matter; Nuclear reactions, nucleosynthesis, abundances; Stars: late-type; Stars: winds, outflows

1. INTRODUCTION

Refractory dust grains with highly anomalous isotopic compositions, so-called “presolar” or “stardust” grains, are found in small quantities in primitive Solar System materials such as unequilibrated meteorites, interplanetary dust particles (IDPs), and cometary dust (Zinner, 2014).

The isotopic anomalies cannot be explained by chemical or physical Solar System processes, but require nucleosynthetic reactions in stellar environments. These stardust grains condensed in the winds of evolved stars and in the ejecta of stellar explosions and carry the isotopic fingerprints of their parent stars. During their passage through the interstellar medium (ISM), they were exposed to shockwaves from nearby supernova explosions, grain-grain-collisions, and high-energetic irradiation (e.g., Woitke et al., 1993; Jones et al., 1996; Dwek, 1998).

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Subsequently, stardust grains were incorporated into the gas and dust cloud from which our Solar System formed, and a fraction of them survived equilibration and alteration processes during the various Solar System formation stages (e.g., Cameron, 1962, 1973; Lodders and Amari, 2005; Zinner, 2014). The study of the isotopic compositions of these grains, as well as their elemental compositions and mineralogy, provides valuable information on stellar nucleosynthesis and evolution, grain growth in circumstellar environments, the types of stars that contributed material to the molecular cloud from which our Solar System formed, and chemical and physical processes in the ISM and in the Solar System.

With the exception of nanodiamonds, whose origins are still a matter of debate, silicates are the most abundant type of presolar grain (e.g., Zinner, 2014). Primitive IDPs contain ~400 ppm (ppm) on average (Floss et al., 2006), with abundances up to the percent level for individual particles (Messenger et al., 2003; Busemann et al., 2009; Davidson et al., 2012). In the most pristine unequilibrated meteorites, matrix-normalized abundances of ~200 ppm are observed (e.g., Nguyen et al., 2007, 2010; Floss and Stadermann, 2009; Vollmer et al., 2009; Nittler et al., 2013).

Oxide stardust grains are less abundant, with abundances of up to tens of ppm in meteorites (e.g., Vollmer et al., 2009; Leitner et al., 2012) and, on average, on the order of 20 ppm for IDPs, although concentrations for individual IDPs tend to be much higher, but contain large uncertainties, since they are based, in each case, on only one grain (Stadermann et al., 2006; Floss and Stadermann, 2009; Nguyen et al., 2014). The latter number is based on the identification of one single presolar Al_2O_3 grain and thus bears large uncertainties.

For material from comet 81P/Wild 2, statistics are limited. A distinction between presolar silicates and oxides is not possible for these samples, since all O-rich presolar signatures were found in high-velocity impact craters in aluminum foil, and structural information was largely erased in the impact process. To date, only 5 presolar O-rich grains have been found (Stadermann et al., 2008; Leitner et al., 2010; Floss et al., 2013).

Based on their O-isotopic compositions, presolar silicate and oxide grains are divided into four distinct groups, with a few “unusual” grains outside of this classification (Nittler et al., 1997, 2008).

Group 1 grains, which make up the majority of the presolar O-anomalous grains, have higher than solar $^{17}\text{O}/^{16}\text{O}$ and $^{18}\text{O}/^{16}\text{O}$ ratios ranging from ~solar (2.01×10^{-3}) down to $\sim 1 \times 10^{-3}$. They formed in the outflows of 1.2–2.2 M_{\odot} red giant and asymptotic giant branch (AGB) stars of approximately solar metallicity (the metallicity, Z , denotes the mass fraction of elements heavier than helium) (Nittler, 2009; Palmerini et al., 2011). Their O-isotopic compositions are well-explained by the so-called “first dredge-up” (FDU) process, which occurs when a star becomes a red giant star (e.g., Lattanzio and Boothroyd, 1997; Boothroyd and Sackmann, 1999). The products of partial H-burning are mixed into the convective envelope and modify the isotopic composition of the stellar surface. The $^{17}\text{O}/^{16}\text{O}$ ratio at the stellar surface is heavily

influenced by this event and depends strongly on the stellar mass (e.g., El Eid, 1994; Boothroyd and Sackmann, 1999). The depth to which the convective envelope extends depends on the maximum temperature, and thus on the stellar mass. As can be seen from El Eid (1994) and Boothroyd and Sackmann (1999), this leads to a gradually enhancement of $^{17}\text{O}/^{16}\text{O}$ on the stellar surface until $M = 2$ to 2.5 M_{\odot} . For stars of higher masses, $^{17}\text{O}/^{16}\text{O}$ decreases again, because the convective envelope extends into region below the “ ^{17}O -peak” (El Eid, 1994), where ^{17}O is depleted. The $^{18}\text{O}/^{16}\text{O}$ ratio, on the other hand, is only slightly affected by the FDU, and mainly represents the initial composition of the stellar envelope.

Group 2 grains are also characterized by enhanced $^{17}\text{O}/^{16}\text{O}$ ratios, but are significantly depleted in ^{18}O ($^{18}\text{O}/^{16}\text{O} < 1 \times 10^{-3}$). These depletions cannot be explained by the first dredge-up in the parent stars, but require additional mixing processes, such as cool bottom processing (CBP) occurring in red giant and AGB stars with $M < 1.5 M_{\odot}$ and sub-solar metallicity (Nollett et al., 2003; Palmerini et al., 2011), or hot bottom burning (HBB) in intermediate mass stars of 4–8 M_{\odot} (Lugaro et al., 2017).

The origin of the grains of Group 3 is still not well-established. The majority of these grains show depletions in both ^{17}O and ^{18}O , pointing to a formation around red giant and AGB stars with low masses ($< 1.4 M_{\odot}$) and low metallicities ($Z \leq Z_{\odot}$) (Nittler et al., 1997). According to Galactic chemical evolution (GCE) models, an increase of the $^{17,18}\text{O}/^{16}\text{O}$ ratios is expected with increasing metallicity. Thus, for low-metallicity stars one expects sub-solar $^{17,18}\text{O}/^{16}\text{O}$ ratios at the time of their birth (Timmes et al., 1995; Kobayashi et al., 2011). For stars with $M < 1.2 M_{\odot}$, the first dredge-up should only have minor effects on the O-isotopic compositions (even at low metallicities) (Boothroyd and Sackmann, 1999). Combined, these effects can account for the majority of the Group 3 grains’ isotopic compositions (Nittler, 2009). However, several Group 3 grains with sub-solar $^{17}\text{O}/^{18}\text{O}$ ratios are possibly of supernova origin (Nittler et al., 2008). Grains of Group 4 are characterized by significantly enhanced $^{18}\text{O}/^{16}\text{O}$ ratios, while the $^{17}\text{O}/^{16}\text{O}$ shows some variation. SNeII have been identified as their most probable stellar sources, based on a comparison of multi-element isotope data with model predictions (Nittler et al., 2008; Vollmer et al., 2008; Nguyen and Messenger, 2014).

Formation of O-rich and C-rich dust around AGB stars occurs during different evolutionary stages. Dust around low- and intermediate-mass AGB stars condenses almost exclusively during the thermal pulsing (TP-) AGB phase, while dust production on the First Giant Branch and in the early AGB-phase is very inefficient (Gail et al., 2009). Initially, the stellar envelope is O-rich (i.e., $\text{C}/\text{O} < 1$), which favors the formation of silicate and oxide stardust in the stellar outflow. After a series of thermal pulses, the star experiences a so-called “third dredge-up” (TDU), which mixes material from the He-intershell, including ^{12}C , to the stellar surface, resulting in a change of the envelope’s composition. The carbon-to-oxygen ratio increases to $\text{C}/\text{O} > 1$, and the star becomes a carbon star, producing carbon and SiC dust (e.g., Gail et al., 2009, and references

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