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Nucleosynthetic Signatures in Presolar SiC and Graphite Grains

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Presolar SiC and graphite grains are the grain types whose isotopic signatures have been extensively studied. Isotopic compositions of light and intermediate elements in single grains have been analyzed mostly using secondary ion mass spectrometry. Detailed information about nucleosynthetic conditions can be obtained from isotopic compositions of heavy elements. Isotopic compositions of heavy elements in SiC and graphite grains have been analyzed using resonant ionization mass spectrometry. Analyses of heavy elements and noble gases are likely to produce new insights into presolar grains using newly-developed instruments.

KEYWORDS: presolar grains, meteorites, isotopic anomalies, nucleosynthesis, supernovae, asymptotic giant branch (AGB) stars

1. Introduction

Presolar grains are grains that formed in stellar outflows or stellar ejecta, were incorporated into meteorites, and remained almost intact since their formation. They retain information of their parent stars. The first mineral type of presolar grains, diamond, was identified and isolated in 1987 [1]. The mineral types of presolar grains identified to date include diamond [1], SiC [2, 3], graphite [4], oxides [5-7], Si_3N_4 [8], silicates [9-12], and refractory carbides inside graphite or SiC host grains [13]. Their abundances range from a few hundred ppm to down to a few ppb [14, 15]. The study of presolar grains, which began in the late 80's, opened up a new field of astronomy, yielding a wealth of information about nucleosynthesis in stars, grain formation in stellar outflows or stellar ejecta, and the Galactic chemical evolution.

Of the presolar grain mineral types, SiC and graphite grains are most extensively studied. SiC grains are present in various chemical types of meteorites [14, 16]. Their abundance in the Murchison meteorite (CM2) is ~6 ppm relative to a bulk meteorite [17]. Although the majority of SiC grains are sub-μm in size, there are large grains (≥1 μm). Trace element concentrations are high compared with the other presolar grain mineral types, thus isotopic ratios of several elements can be analyzed in single grains.

Presolar graphite grains are present only in highly primitive meteorites. The Murchison and Orgueil (CI) meteorites are the only two from which graphite grains were extracted [17, 18] and systematically studied [19, 20]. The abundance of graphite grains in Murchison is 0.88 ppm [19]. There are many grains larger than 1 μm in size. Grains exhibit a range of density (1.6 – 2.2 g/cm^3). One of the most intriguing features of graphite grains is that isotopic, elemental and morphological characteristics depend on density. Low-density graphite grains show higher trace element abundances than high-density graphite grains. Large grain size (1≥ μm) and high trace elements are two main factors that make it possible to analyze isotopic ratios of multiple elements.

In this paper, we will discuss isotopic analyses of light, intermediate and heavy elements in single SiC and graphite grains and future prospects of the study of these presolar grains.

2. Isotopic Ratios of Light and Intermediate Elements

2.1 Silicon carbide

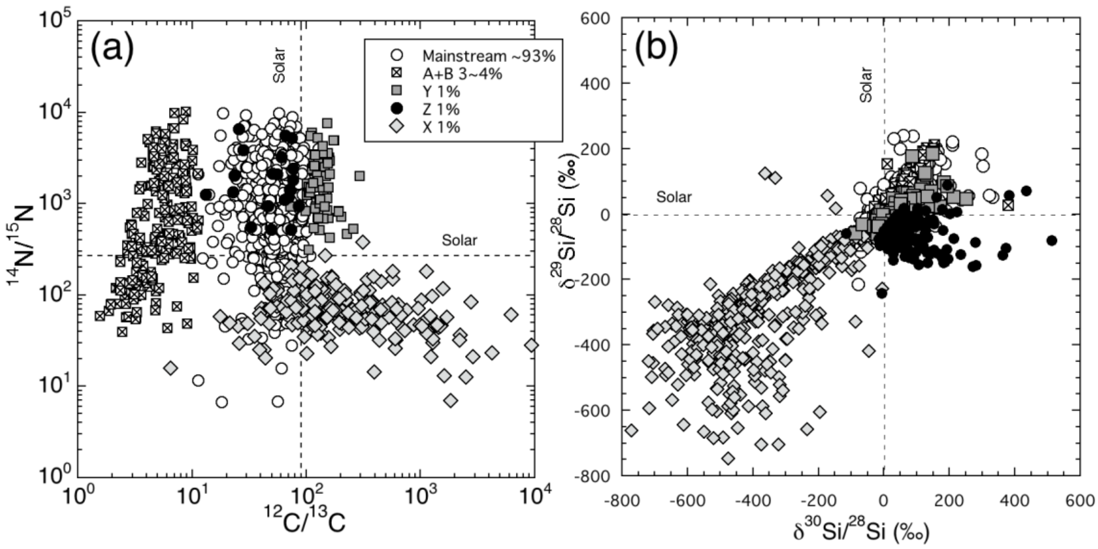

Fig. 1. Isotopic ratios of SiC grains from different populations are shown. (a) C and N isotopic ratios. (b) Si isotopic ratios expressed with δ values $\{\delta^{i}Si/^{28}Si$ (‰) = $[(^{i}Si/^{28}Si)_{grain}/(^{i}Si/^{28}Si)_{standard} - 1] \times 1000\}$. The dotted lines indicate the solar ratios (or the ratio of air for $^{14}N/^{15}N$). Note that the minor populations of grains have been located for detailed studies, thus grains in these populations are overrepresented in the plots. Real abundances are shown in the legend. The data are from the Presolar Grain Database (http://presolar.wustl.edu/Laboratory_for_Space_Sciences/Presolar_Grain_Database.html) [21].

Silicon carbide grains are classified into several populations based on their C, N and Si isotopic ratios [22, 23] (Fig. 1). Mainstream grains, defined as having $^{12}C/^{13}C$ ratios between 10 and 100, $^{14}N/^{15}N$ ratios higher than that of air (272), and Si enriched in the neutron-rich isotopes, comprise 90 – 95 % of the total SiC grains in meteorites. Grains of type Y and Z exhibit $^{14}N/^{15}N$ ratios similar to mainstream grains. Y grains are defined as having $^{12}C/^{13}C$>100 [24], while Z grains show $^{12}C/^{13}C$ ratios similar to those of mainstream grains [25]. Silicon isotopic ratios in mainstream, Y and Z grains show a systematic trend. First, average $^{29}Si/^{28}Si$ ratios systematically decrease from mainstream to Y to Z grains. Second, the spread of $\delta^{30}Si/^{28}Si$ values (see the caption of Fig. 1 for the definition of δ values) systematically becomes larger.

Bulk analyses of SiC-rich fractions show that they are enriched in *s*-process Kr [26-28], Xe [27, 28], Sr [29], Ba [30-32], Nd and Sm [31]. Since the majority of SiC grains are mainstream grains, the *s*-process enrichments were most likely carried by mainstream grains. The *s*-process signatures agree with the predicted ratios from low-mass (1.5 – 3 M_{sun}) asymptotic giant branch (AGB) stars. Isotopic signatures of C, N and Si isotopic ratios of mainstream grains also support that they formed in low-mass AGB stars [33]. The isotopic signatures of C, N and Si in Y and Z grains can be also explained by AGB stars but lower metallicity than close-to-solar. Y grains of around half solar metallicity [24], and Z grains of around one-third of solar metallicity [25].

A+B grains are defined as having $^{12}C/^{13}C$ ratios lower than 10 with a range of $^{14}N/^{15}N$ ratios. Low $^{12}C/^{13}C$ ratios of A+B grains favor J stars, which show low $^{12}C/^{13}C$ ratios. However, the spread of the $^{14}N/^{15}N$ ratios of A+B grains suggest that multiple stellar sources seem to exist for this population [34].

X grains are characterized by ^{28}Si excesses, higher-than-solar $^{12}C/^{13}C$ and lower-than-solar $^{14}N/^{15}N$ ratios, and high $^{26}Al/^{27}Al$ ratios (~ 0.1) [35-37]. A few grains show ^{44}Ca excesses due to the decay of ^{44}Ti ($T_{1/2}$ = 60 a) [38]. The initial presence of ^{44}Ti is the strongest proof that these grains formed in supernovae becaues it is produced only duirng explosive nucleosynthesis. Other isotopic signatures also agree that grains fromed in supernovae.

2.2 Graphite

$^{12}C/^{13}C$ ratios of graphite grains span more than 3 orders of magnitude [19, 20]. Carbon is the only major element in graphite and C isotopic ratios alone are not so diagnostic to distinguish grains' stellar sources. Figure 2 shows distributions of $^{12}C/^{13}C$ ratios of 4 density fractions from Murchison. Lower density fractions, KE3 (1.65 – 1.72 g/cm^3) and KFA1 (2.05 – 2.10 g/cm^3), show a broader peak, where higher density fractions, KFB1 (2.10 – 2.15 g/cm^3), and KFC1 (2.15 – 2.20 g/cm^3), show bimodal distributions with many more isotopically light grains than heavy grains.

Nitrogen is relatively abundant across the grains. $^{14}N/^{15}N$ ratios of many graphite grains are close to that of air. It has been proposed that indigenous N has been partially equilibrated with normal N either in the solar system or in the laboratory [39]. This means that close-to-normal N isotopic ratios need to be cautiously interpreted when we consider grains' stellar sources.

Oxygen and Si isotopic ratios are indicative of some of the stellar sources of graphite grains. Low-density graphite grains show ^{18}O enrichment in many of the grains that are taken as proof that they formed in core-collapse supernovae: ^{18}O is produced via $^{14}N(\alpha,\gamma)^{18}F(e^{+}\nu)^{18}O$ in the partial He burning zone, resulting $^{18}O/^{16}O$ ratio of 1.68 in a 15M_{sun} model by Rauscher et al. [40]. Many low-density graphite grains show ^{28}Si excesses, similar to SiC X grains from supernovae, while a few show ^{29}Si and ^{30}Si excesses. ^{28}Si is produced during O burning and excesses in neutron-rich isotopes are indicative of neutron captures in various nucleosynthetic zones.

The majority of high-density graphite grains have $^{12}C/^{13}C$ ratios higher than 100 (Fig. 2), up to a few thousand. These high $^{12}C/^{13}C$ ratios and high $\delta^{30}Si/^{28}Si$ values observed in a few of the grains, high enrichment of *s*-process elements [41] indicate that they formed in AGB stars of low-metallicity.

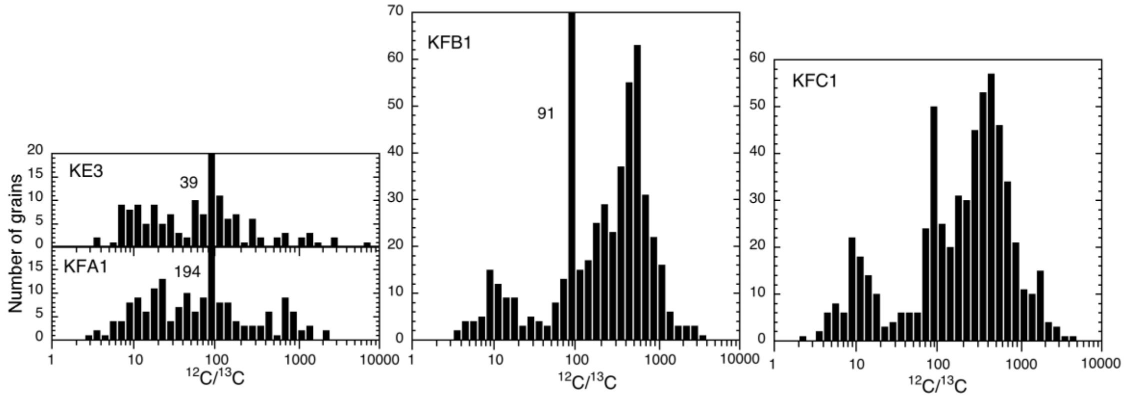

Fig. 2. $^{12}C/^{13}C$ histograms of graphite grains from the four Murchison graphite fractions, KE3, KFA1, KFB1 and KFC1. The numbers in the diagrams are the numbers of grains in the bin that includes the solar ratio (89). Data from [19].

3. Isotopic Ratios of Heavy Elements

Single grain analyses of light and intermediate elements have provided information about grains' stellar sources, while analyses of heavy elements can provide detailed information about nucleosynthetic conditions.

The difficulties are: the abundances of heavy elements are much lower than those of light and intermediate elements. Analyses of isotopic ratios of heavy elements using secondary ion mass spectrometry (SIMS) require tremendously high mass resolving power and that reduces ion counts. For example, a mass resolving power (M/ΔM) of 22,120 is required to separate ^{96}Mo and ^{96}Zr. Thus, SIMS is, in general, not an ideal instrument to analyze isotopic ratios of heavy elements.

Resonant Ion Mass Spectrometry (RIMS) has been used for isotopic studies of heavy elements in presolar grains. Unlike SIMS, which uses ions that is produced less than 1 percent of sputtered material, RIMS ionizes neutral material that comprises a major part of sputtered material. A desorbed laser produces a cloud of neutral atoms. Then the second lasers selectively ionize elements of interest. Since it is free of interferences, isotopes are analyzed with a time-of-flight mass spectrometer.

3.1 Silicon carbide

Isotopic ratios of heavy elements in SiC grains have been analyzed using the CHARISMA (Chicago-Argonne Resonant Ionization Spectrometer for Mass Analysis) at Argonne National Laboratory, USA. Analyses of mainstream grains indicate that their parent stars are close-to-solar metallicity [42] and gave constraints for the range of ^{13}C pocket efficiencies [43].

Analyses of X grains show very intriguing isotopic patterns [44]. Four out of 6 X grains showed excesses in ^{95}Mo and ^{97}Mo. That was not what was expected: from the *s*-process, we expect excesses in ^{96}Mo and ^{98}Mo, and from the *r*-process the largest excess in ^{100}Mo. Meyer et al. [45] proposed neutron-burst: it takes place in the shocked and heated ($T = 10^9$ K) He-rich region in an exploding star. The neutrons are produced by (α,n) reactions on ^{13}C and ^{22}Ne, and subsequently captured by heavy elements. For the Mo isotopes, the large abundances of ^{95}Y ($T_{1/2}$ = 10 m), ^{95}Zr ($T_{1/2}$ = 64 d) and ^{97}Zr ($T_{1/2}$ = 17 h) produce excesses in ^{95}Mo and ^{97}Mo.

3.2 Graphite

A few graphite grains were analyzed with the CHARISMA. Thirty-two KFC1 grains were analyzed for their Zr and Mo isotopic ratios [46]. Of eight grains that could be analyzed for both Mo and Zr isotopic compositions, three grains show *s*-process isotopic signatures for both elements. Four grains show almost normal Mo but anomalies in Zr, large depletions or excesses in ^{96}Zr. High ^{96}Zr can be explained by the *r*-process, or the *s*-process when neutron density is unusually high. Zr and Ba isotopic ratios of high-density grains from Orgueil show *s*-process signature from AGB stars [47].

3. Future Work

Analyses of elements that are difficult to analyze with SIMS are likely to bring new information in presolar grains. These elements include heavy elements and noble gases: the former is due to the need of extremely high-mass resolving power that would reduce ion counts, and the latter is because noble gases are not ionized with SIMS. Recent instrumental developments are encouraging steps toward a better understanding for the origins and evolutions of presolar grains.

A more advanced type of RIMS, CHILI (Chicago Instrument for Laser Ionization), has been developed at The University of Chicago, USA [48] and has been applied to analyze heavy elements in SiC grains [49]. CHILI has high spatial resolution of ~10 nm due to a liquid metal ion source. Six tunable Ti: Sapphire lasers are used for ionization and simultaneous ionization of three elements is possible. The goal for the useful yield, defined by the ratio of detected ions to sputtered atoms, is set higher (40%) than those of previous-generation RIMS.

The other instrument, LIMAS (Laser Ionization Mass Nanoscope), has been installed at Hokkaido University, Japan [50]. This is a non-resonant, post-ionization mass spectrometer. By femtosecond laser, all atoms are ionized and high mass resolving power (up to 150,000) can be achieved using multi-turn time-of-flight mass spectrometer, MULTUM II. LIMAS also uses a liquid metal ion source, thus has high spatial resolution. Helium in a GENESIS sample has been analyzed using LIMAS [51].

Acknowledgments

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