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Isotopic analysis of presolar graphite grains from Orgueil

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Abstract

We report the successful isolation and isotopic analysis of presolar graphite grains from the Orgueil CI chondrite. Isotopic measurements were made on seven density fractions, with grain sizes >1 μm: ORG1b (1.59–1.67 g cm^{-3}), 1c (1.67–1.75 g cm^{-3}), 1d (1.75–1.92 g cm^{-3}), 1f (2.02–2.04 g cm^{-3}), 1g (2.04–2.12 g cm^{-3}), 1h (2.12–2.16 g cm^{-3}) and 1i (2.16–2.30 g cm^{-3}). We measured C, N, O, and Si isotopes with the NanoSIMS. All the fractions, except ORG1b and ORG1h, contain presolar graphite as demonstrated by the large range of $^{12}C/^{13}C$ ratios (4–1746) measured in individual grains. The abundance of grains with isotopically light carbon increases with increasing density. Some of the low-density grains are enriched in ^{18}O, ^{15}N and ^{28}Si. As the density increases, the grains mostly exhibit solar oxygen and nitrogen isotopic ratios. However, the high-density grains are enriched in ^{29}Si and ^{30}Si. The ^{18}O and ^{28}Si excesses indicate that the low-density grains originated from supernovae, while the high-density grains (with ^{30}Si and ^{12}C excesses) probably originated from AGB stars with low metallicities.

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Keywords: Interstellar dust; Isotopic anomalies; Graphite; Supernovae; AGB stars; Nucleosynthesis

Contents

1. Introduction

Presolar graphite was first isolated from the Murchison CM2 meteorite as the carrier of Ne-E(L) (almost pure ^{22}Ne) by Amari et al. (1990). Since then these grains from Murchison have been extensively studied, and almost everything we know about presolar graphite is based on

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these studies (Amari et al., 1990, 1995; Hoppe et al., 1995; Zinner et al., 1995; Travaglio et al., 1999). The lack of studies on graphite grains from other meteorites rests on the fact that, although graphite can be found in the most primitive meteorites (Huss and Lewis, 1995), it has a low abundance compared to SiC (Huss and Lewis, 1995), and the separation procedure for graphite is far more complicated than that for SiC (Amari et al., 1994). Complications during the separation arise because graphite is chemically not as resistant as SiC or oxide minerals, and because it needs to be isolated from other carbonaceous material in the meteorite that has similar chemical properties. Carbonaceous phases, in particular presolar graphite grains, are believed to condense from gases with $C/O > 1$. The envelopes of carbon stars, Wolf–Rayet (WR) stars, novae, and supernova (SN) zones all satisfy this condition at some time in their lifecycles. In order to gain a proper understanding of condensation of carbon phases from stellar atmospheres and their survival in the interstellar medium (ISM) as well as to obtain information on stellar nucleosynthesis, presolar graphite populations from other meteorites need to be identified and studied for their isotopic compositions.

Based on noble gas analyses, Huss and Lewis (1995) estimated the abundance of presolar graphite in the carbonaceous chondrite (CI) Orgueil to be an order of magnitude higher than that in Murchison. Thus, an effort was made by Pravdivtseva et al. (2004) to isolate presolar graphite in Orgueil, using the separation procedure previously applied to Murchison. They obtained a fraction with a density of $\sim$1.8 g cm^{-3} and grain size >1 μm. Although these grains closely resembled the onion-type grains of Murchison (Hoppe et al., 1995), NanoSIMS isotopic analyses of C and N of 162 grains yielded only normal ratios, indicating that the grains had a solar system origin (Pravdivtseva et al., 2004). Additional Ne isotopic analysis of 14 individual grains did not detect any excesses in ^{22}Ne above the blank.

We undertook a new separation of carbonaceous and refractory presolar grains from Orgueil (Jadhav et al., 2005). The main objective was to isolate presolar graphite from Orgueil, which, despite its inferred high abundance in this meteorite, has been elusive until now. We present C, N, O and Si isotopic analyses of graphite grains from seven density fractions of Orgueil. We also discuss the possible stellar sources for the anomalous graphite grains found.

2. Experimental procedure and methods

Our sample of Orgueil was obtained from the National Museum of Natural History in Paris. We subjected 24.16 g to essentially the same separation procedure carried out for Murchison graphite by Amari et al. (1994) to obtain ten final density fractions. The lightest density fraction, ORG1a (<1.59 g cm^{-3}) contains organic matter and the heaviest fraction, ORG1j (>2.3 g cm^{-3}) is expected to contain SiC and oxides. At least a few of the remaining eight density fractions were expected to contain presolar graphite grains. These fractions were then separated according to size with a cutoff of 1 μm.

Carbonaceous grains were located in a JEOL-840A SEM and identified by energy dispersive X-ray (EDX) analysis. Candidate grains were selected on the basis of high C content and morphological features characteristic of Murchison graphite (spherules with a platy ‘onion’ or knobby ‘cauliflower’ appearance (Hoppe et al., 1995)).

This initial characterization in the SEM was followed by isotopic analyses of the candidate grains in the NanoSIMS. $^{12}C^-$, $^{13}C^-$, $^{16}O^-$ and $^{18}O^-$ (analysis phase 1) and, $^{12}C^{14}N^-$, $^{12}C^{15}N^-$, $^{28}Si^-$, $^{29}Si^-$ and $^{30}Si^-$ (phase 2) secondary ions produced by bombarding the sample with a Cs^+ primary beam were counted in multidetection mode.

3. Results

Table 1 lists the number of carbonaceous grains identified in the different fractions, the kind of isotopic data obtained on them and the number of grains that were found to contain isotopic anomalies.

Some of our density fractions of Orgueil contain large concentrations (much higher than in graphite separates from Murchison) of macromolecular carbonaceous material, in which graphite grains are often found embedded. This material made identification of graphite grains in Orgueil difficult. Most of the grains from the density separates are spherules and look very similar to Murchison presolar graphite grains; they have smooth surfaces and a platy, onion-like morphology (Hoppe et al., 1995). Some potato-shaped presolar graphite grains were also found. We did not find any grains with cauliflower-type morphology, which had been seen in the low-density fractions of Murchison (Hoppe et al., 1995). The sizes of the grains range from 2 to 30 μm. Unlike for Murchison, in Orgueil the average grain size increases with density.

Five of the seven density fractions studied have isotopically anomalous graphite (Table 1). The $^{12}C/^{13}C$ ratios of the grains range from 4 to 1746 (Figs. 1 and 2). The C ratios of the ORG1b and 1h grains are close to solar and do not show the large variations that are expected in presolar graphite (Hoppe et al., 1995). However, 29% of all grains in ORG1c, 59% in ORG1d, 90% in ORG1f, 73% in ORG1g, and 67% of the grains in ORG1i have C isotopic anomalies. The grains from ORG1h are morphologically different from the grains of the other fractions, in that they are smooth spherules with very little surface morphology. Grains of such morphology have been found to be isotopically normal in Murchison studies as well (Zinner et al., 1995). In general, grains with isotopically light carbon are more abundant in the higher density fractions (ORG1f, 1g and 1i; Figs. 1 and 2). A minor population of grains with $^{12}C/^{13}C \sim 10$ is observed in all fractions heavier than ORG1c (Figs. 1 and 2). Sim-

Table 1
Number of carbonaceous grains identified in each density fraction, the type of isotopic ratios measured for them and the number of presolar grains identified in each fraction

Fraction name	Density (g cm^{-3})	# of carbonaceous grains identified and type of isotopic data obtained				# of presolar grains found
		C	N	O	Si	
ORG1b	1.59–1.67	22	–	22	–	0
ORG1c	1.67–1.75	7	7	7	7	2
ORG1d	1.75–1.92	32	32	32	32	19
ORG1e	1.93–2.02	–	–	–	–	–
ORG1f	2.02–2.04	72	72	72	72	65
ORG1g	2.04–2.12	52	51	52	51	38
ORG1h	2.12–2.16	18	–	18	–	0
ORG1i	2.16–2.30	9	9	9	9	6

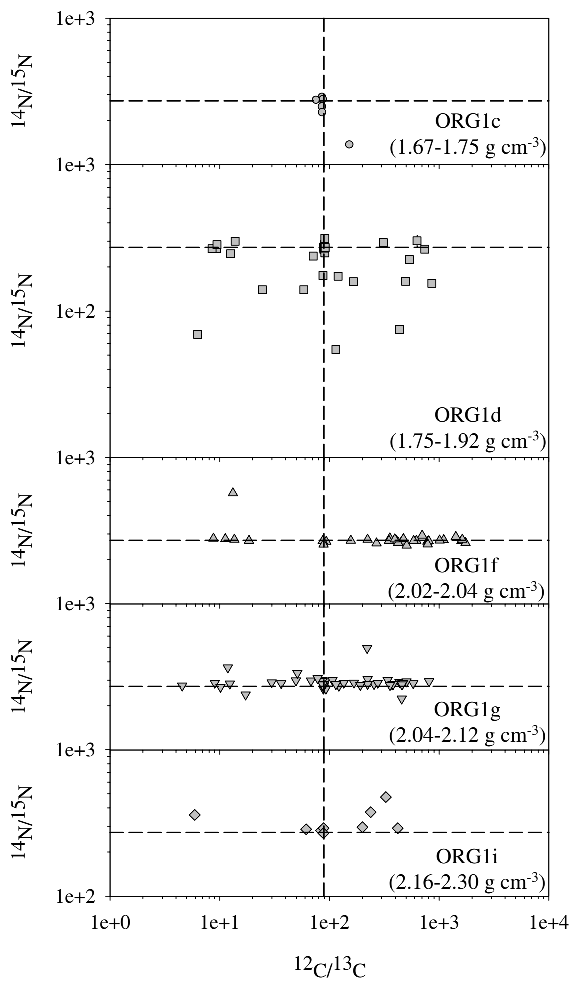

Fig. 1. $^{14}N/^{15}N$ and $^{12}C/^{13}C$ measured in individual graphite grains from different density fractions of the Orgueil meteorite. Solar values of $^{14}N/^{15}N = 272$ and $^{12}C/^{13}C = 89$ are indicated by the dashed lines in the plots.

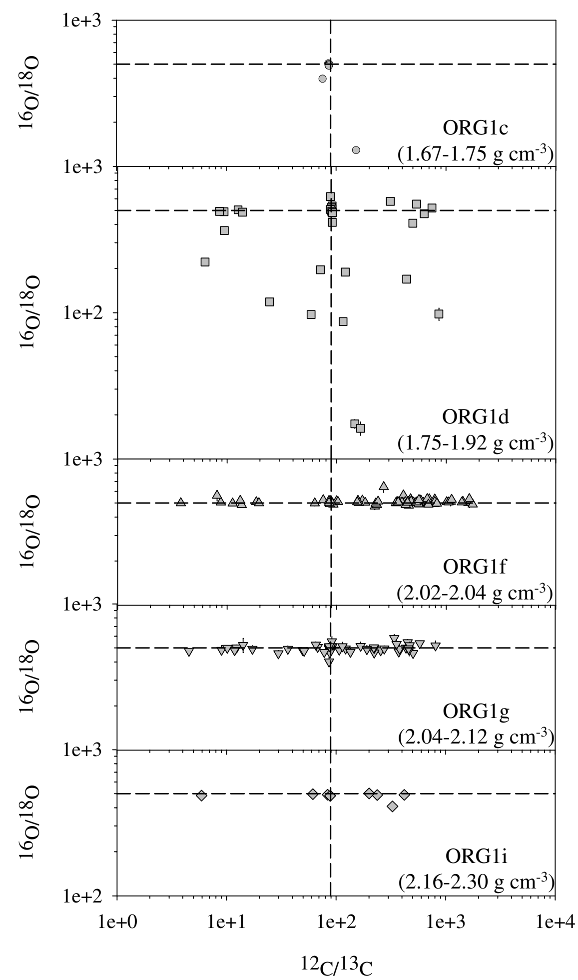

Fig. 2. Oxygen and carbon isotopic ratios of graphite grains from different density fractions. The dashed lines in each plot indicate the solar ratios of $^{16}O/^{18}O = 499$ and $^{12}C/^{13}C = 89$.

ilar C isotopic distributions, a higher abundance of grains with $^{12}C/^{13}C >$ solar in grains with higher density, and the presence of a population around $^{12}C/^{13}C \sim 10$, have also been observed in Murchison graphite (Hoppe et al., 1995).

Figs. 1 and 2 show the C, N and O isotopic compositions measured in the grains of the various density fractions. The low-density fractions, ORG1c and 1d, contain some grains that have ^{15}N and ^{18}O excesses. Most of the grains that contain isotopically heavy nitrogen also have ^{18}O excesses. Since ^{18}O excesses are a signature of a SN origin (Travaglio et al., 1999), they indicate that ORG1c and 1d contain SN graphite. At higher densities (ORG1f, 1g and 1i), the $^{14}N/^{15}N$ and $^{16}O/^{18}O$ ratios become surprisingly normal. This is especially puzzling in view of the large range of

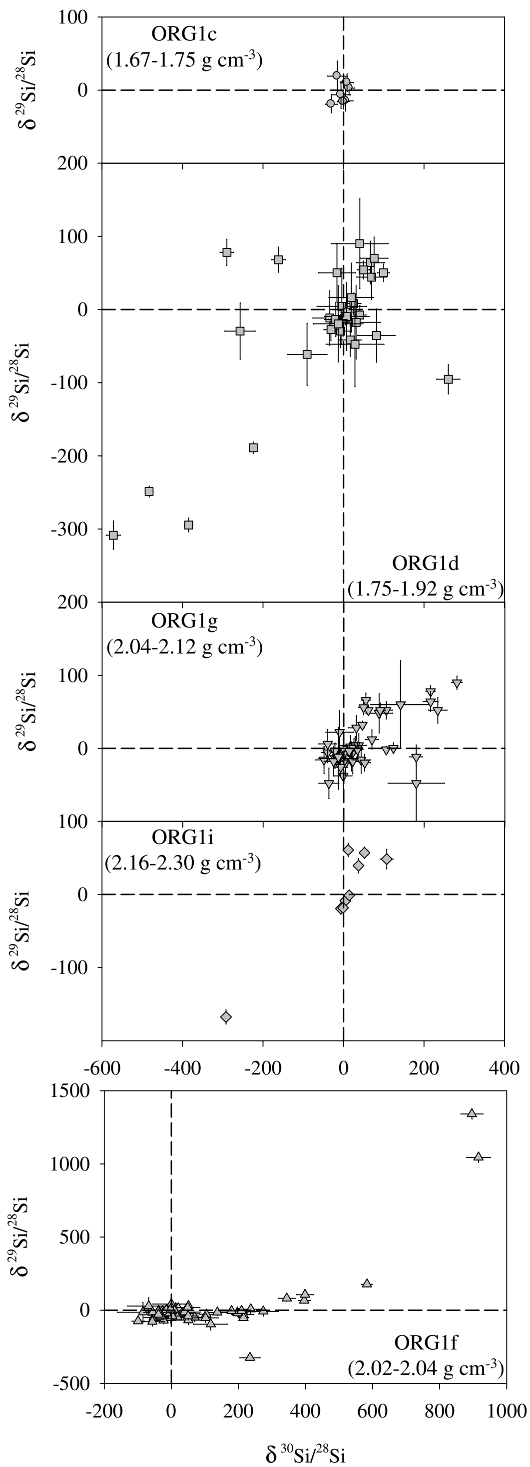

Fig. 3. Delta-value three-isotope plots of the Si ratios measured in graphite grains from different density fractions. The ratios are plotted as δ-values, deviations from the terrestrial ratios $^{29}Si/^{28}Si = 0.050633$ and $^{30}Si/^{28}Si = 0.033621$, in permil (‰). Error bars are 1σ. Dashed lines indicate the solar ratios ($\delta^{29}Si/^{28}Si = \delta^{30}Si/^{28}Si = 0$).

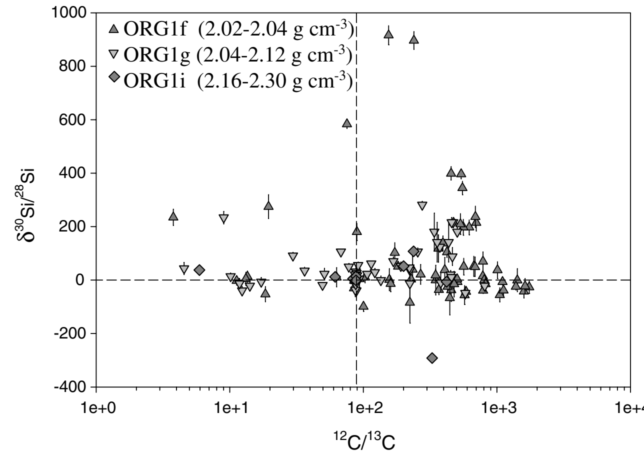

Fig. 4. The $\delta^{30}Si/^{28}Si$ values of graphite grains from the high-density fractions, ORG1f, 1g and 1i, are plotted against their $^{12}C/^{13}C$ ratios. The dashed lines indicate solar values and error bars are 1σ.

$^{12}C/^{13}C$ ratios seen in these grains. A possible explanation for this fact is that initially anomalous N and O isotopically equilibrated by exchange with normal N and O, either on the parent body or in the laboratory (Zinner et al., 1995), and the normal N and O ratios of high-density grains cannot be considered to represent the isotopic signatures of their stellar sources.

The silicon isotopic compositions of the grains also depend on density (Fig. 3). The low-density fractions ORG1c and 1d contain grains with ^{28}Si excesses. Some of these excesses correlate with ^{18}O and ^{15}N excesses. Excesses in ^{28}Si and ^{18}O are indicative of a SN origin. The high-density grains from ORG1f, 1g and 1i, however, are enriched in ^{29}Si and ^{30}Si. Most ^{30}Si-rich grains contain isotopically light carbon (Fig. 4). These signatures are expected for low-metallicity AGB stars where during the thermally pulsing phase ^{12}C and $^{29,30}Si$ are mixed by the third dredge-up (TDU) from the He shell into their envelopes (Hoppe et al., 1997; Busso et al., 1999).

In general, Orgueil graphite grains appear to be rather similar to Murchison graphite, both morphologically and in their isotopic properties, an exception being the lack of cauliflower-type grains in Orgueil.

4. Discussion

While there still remains a multitude of unanswered questions about presolar graphite grains and their stellar sources, one issue seems to have cleared up. There exists a large population of SiC grains from AGB stars. However, in spite of the fact that Kr isotopic measurements of bulk samples indicated the presence of AGB graphite grains in the highest density fraction from Murchison (Amari et al., 1995), before this study and a recent study by Amari et al. (2005) only few such grains have been identified from single-grain isotopic analysis. This was a puzzle because both graphite and SiC grains are expected to condense around similar C-rich stars. High-density graphite grains from the Murchison fractions KFB1 and KFC1 have been analyzed with the IMS-3f in the past (Hoppe et al., 1995), but due to the low sensitivity of the instrument it was difficult to obtain Si isotopic data for these grains with sufficiently high precision. The NanoSIMS, however, with its high sensitivity, allows us to measure the Si isotopes of high-density graphite grains with much higher precision and hence, finally, allows us to investigate graphite grains from low-metallicity AGB stars. A recent NanoSIMS study by Amari et al. (2005) of the Murchison KFB1 and KFC1 fractions, has also found graphite grains that originated from low-metallicity AGB stars.

While high-density graphite grains have high $^{12}C/^{13}C$ ratios and large ^{30}Si excesses, no SiC grains with similar C and Si isotopic signatures have been found (Hoppe et al., 1997; Nittler and Alexander, 2003). The relatively low $^{12}C/^{13}C$ ratios in type Z SiC grains, which have large ^{30}Si excesses, have been explained by invoking extra mixing, cool bottom processing (CBP) (Hoppe et al., 1997; Nollett et al., 2003). This process leads to proton capture on ^{12}C mixed into the envelope by the TDU, lowering the $^{12}C/^{13}C$ ratio. A possible explanation for the high $^{12}C/^{13}C$ ratios in high-density graphite grains could be that these grains formed around low-metallicity AGB stars that did not experience any CBP. Such stars are predicted to have not only high $^{12}C/^{13}C$ ratios but also very high C/O ratios. Because of the large relative excess of C over Si in the envelope of such stars, graphite, but not SiC grains are expected to condense (Bernatowicz et al., 2006). Thus it appears that the presence or absence of CBP in low-metallicity stars is a criterion whether SiC or graphite grains form in the envelope of such stars, both with large ^{30}Si excesses, but SiC with low and graphite with high $^{12}C/^{13}C$ ratios.

5. Conclusions

Presolar graphite has been successfully isolated from Orgueil. Five of the seven density fractions analyzed contain presolar graphite as indicated by isotopically anomalous carbon. The morphologic and isotopic characteristics of Orgueil graphite are similar to those of Murchison. We have found evidence of SN graphite in the low-density fractions and grains from low-metallicity AGB stars in the high-density fractions.

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