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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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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-