

## On the stellar sources of presolar graphite

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Primitive meteorites contain graphite spherules whose anomalous isotopic compositions indicate a stellar origin. The isolation of presolar graphite grains is difficult and they have been less well studied than presolar silicon carbide grains. While previous isotopic measurements have been made on graphite from the Murchison meteorite and mostly on low-density grains, a new separation of graphite from the Orgueil meteorite and the application of a new ion microprobe, the NanoSIMS, have provided a wealth of C, N, O, Al, and Si isotopic data on grains with a range of densities. These data confirm that low-density grains come from supernovae. These grains are characterized by  $^{15}\text{N}$ ,  $^{18}\text{O}$ ,  $^{28}\text{Si}$  and  $^{49}\text{Ti}$  excesses, high inferred  $^{26}\text{Al}/^{27}\text{Al}$  ratios, as well as evidence for the initial presence of the short-lived radionuclides  $^{22}\text{Na}$ ,  $^{41}\text{Ca}$ , and  $^{44}\text{Ti}$ . All these signatures point to an origin in Type II supernovae. The Ne-E(L) component, almost pure  $^{22}\text{Ne}$ , is characteristic of presolar graphite and led to its discovery. In SN grains it is due to the decay of short-lived ( $T_{1/2} = 2.6$  yr)  $^{22}\text{Na}$ , which apparently was implanted into the grains. Many high-density grains have high  $^{12}\text{C}/^{13}\text{C}$  ratios ( $>300$ ) and large excesses in  $^{30}\text{Si}$  and smaller excesses in  $^{29}\text{Si}$ . These signatures are best explained by an origin in low-metallicity AGB stars. In these stars the enrichments of the envelope in the heavy Si isotopes and in  $^{12}\text{C}$ , products of nucleosynthesis in the He shell, are much larger than those expected for solar-metallicity parent stars. The high  $^{12}\text{C}/^{13}\text{C}$  ratios imply also high C/O ratios, which probably caused the preferential condensation of graphite grains over SiC grains. An AGB origin of high-density grains is compatible with the presence of internal sub-grains with high concentrations of the s-process elements Zr, Mo, and Ru, reflecting the high abundances of these elements in the envelope of such stars. The stellar source of high-density grains with  $^{12}\text{C}/^{13}\text{C}$  ratios around 10 remains elusive.

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## 1. Introduction

Primitive meteorites contain presolar grains that condensed in the outflows of evolved stars and SN ejecta [e.g., 1]. They are characterized by large isotopic anomalies and their study in the laboratory provides information on stellar nucleosynthesis and evolution, Galactic chemical evolution, physical and chemical conditions of stellar atmospheres, and conditions in the early Solar nebula and on meteorite parent bodies. Among the different types of presolar grains, silicon carbide has been studied in most detail. SiC is chemically very refractory and by chemical separation can be isolated in almost pure form [2]. In addition, because SiC condenses at  $C/O > 1$ , it is not produced in the Solar nebula and all grains are of stellar origin. This is in marked contrast to oxygen-rich minerals, which are abundant in Solar System materials. As a consequence, hundreds to ten thousands of grains have to be analyzed for their isotopic compositions in order to find the rare O-rich grains of stellar origin [e.g., 3, 4]. Presolar graphite grains can be isolated from meteorites by chemical and physical separation methods, but the separation procedure is much more difficult than that for SiC [2, 5] and presolar graphite has been less well studied than SiC. For years, analysis of graphite has been restricted to grains from the Murchison carbonaceous meteorite [e.g., 6, 7]. Recently, graphite separates have been obtained from a second meteorite, Orgueil [8]. In addition, a new type of ion microprobe, the NanoSIMS, allows isotopic analysis of small grains with high sensitivity [9]. The new wealth of isotopic data for individual grains prompted us to reassess the stellar origin of presolar graphite.

## 2. Isotopic ratios of individual presolar graphite grains

### 2.1 Classification of presolar graphite

It has been known for some time that the isotopic compositions of graphite grains depend on their density. Figure 1 shows histograms of the  $^{12}\text{C}/^{13}\text{C}$  ratios measured in single grains from four density fractions obtained from Murchison [5]. The isotopic ratios of the noble gases depend on density as well [5]. Detailed isotopic measurements on single grains have been made only from the low-density fraction KE3 [7] and the results have shown that most of these grains must have originated from Type II supernovae. For the purpose of the present work we divide the graphite grains into two classes, low- and high-density grains. The first class comprises the Murchison fractions KE3 and KFA1, and the second class the fractions KFB1 and KFC1 (Fig. 1). There exists no exact agreement on the density scales for Murchison and Orgueil grains. Based on their isotopic characteristics, we classify Orgueil graphite grains with  $\rho < 2.02 \text{ g cm}^{-3}$  as low-density and those with  $\rho > 2.02 \text{ g cm}^{-3}$  as high-density [8].

### 2.2 Results

Figures 2-4 show C, N, O, Al, and Si isotopic ratios measured with the Cameca IMS 3f and NanoSIMS ion microprobes in single low- and high-density graphite grains from Murchison and Orgueil. The Al and Si isotopic ratios in Figs. 3 and 4 are compared with those observed in different types of presolar SiC grains [1]. Although there is undoubtedly some overlap of grains from different stellar sources in the two graphite classes, the isotopic ratio

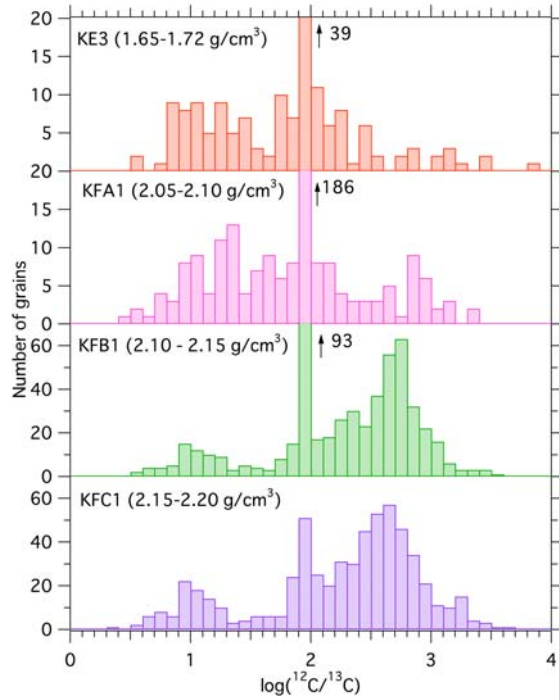


Figure 1. Carbon isotopic ratios of different density fractions of graphite grains from the Murchison Meteorite.

distributions in these two classes are clearly distinct. A much larger fraction of low-density grains has  $^{15}\text{N}$  and  $^{18}\text{O}$  excesses, both a signature of the He/C shell of Type II supernovae [10]. The essentially normal N and O isotopic ratios of many grains with a huge range in  $^{12}\text{C}/^{13}\text{C}$  ratios cannot be explained by stellar models and are most likely the result of isotopic equilibration or contamination. Low-density grains resemble SiC X grains, which have a SNII origin, in their high inferred (from  $^{26}\text{Mg}$  excesses)  $^{26}\text{Al}/^{27}\text{Al}$  ratios (Fig. 3). There exist only few analyses of high-density grains and grains with high  $^{26}\text{Al}/^{27}\text{Al}$  ratios have been selected. Si isotopic ratios of some indicate also a SNII origin. The  $^{28}\text{Si}$  excesses of low-density grains, similar to those of SiC X grains indicate a SNII origin, but also the few grains with large  $^{29}\text{Si}$  and  $^{30}\text{Si}$  excesses could have such an origin (Fig. 4). In summary, the new data confirm a SN origin for most low-density graphite grains. Evidence for short-lived  $^{41}\text{Ca}$  and  $^{44}\text{Ti}$  as well as large  $^{49}\text{Ti}$  excesses

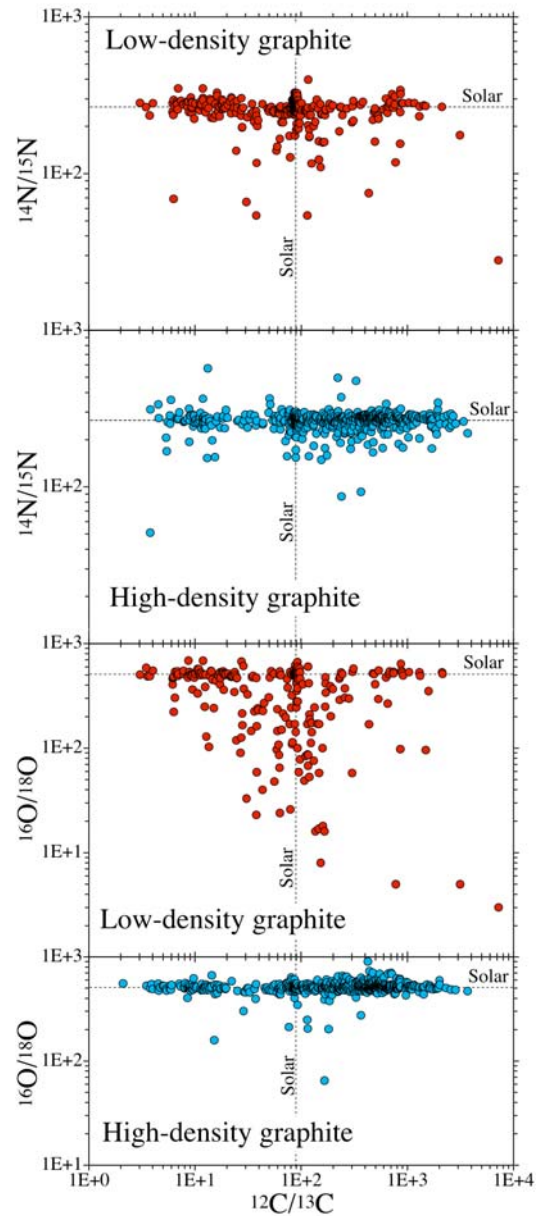


Figure 2. Carbon, N, and O isotopic ratios of individual low- and high-density graphite grains.

(possibly from the decay of  $^{49}\text{V}$ ) also indicate an origin in Type II supernovae [7, 11].

The most diagnostic isotopic signatures in high-density graphite grains are the C and Si isotopic ratios. The  $^{30}\text{Si}$  excesses are similar to such excesses in SiC Z grains, which are believed to originate in low-metallicity AGB stars [12], but are even larger (Fig. 4). Models of AGB nucleosynthesis predict that such  $^{30}\text{Si}$  excesses, resulting from neutron capture during thermal pulses in the He shell, should

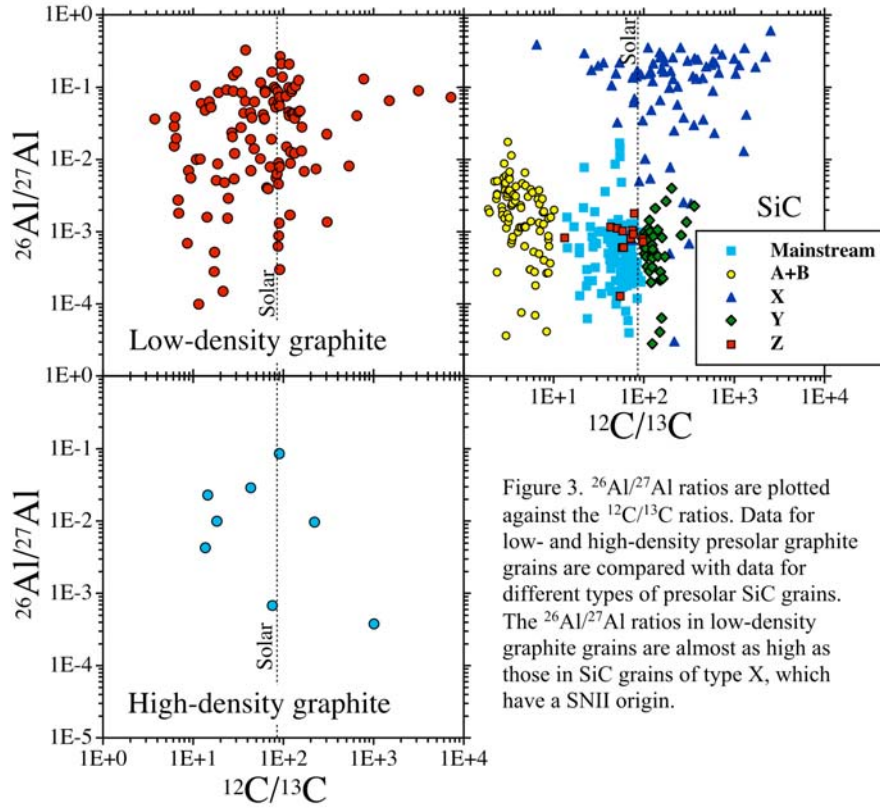


Figure 3.  $^{26}\text{Al}/^{27}\text{Al}$  ratios are plotted against the  $^{12}\text{C}/^{13}\text{C}$  ratios. Data for low- and high-density presolar graphite grains are compared with data for different types of presolar SiC grains. The  $^{26}\text{Al}/^{27}\text{Al}$  ratios in low-density graphite grains are almost as high as those in SiC grains of type X, which have a SNII origin.

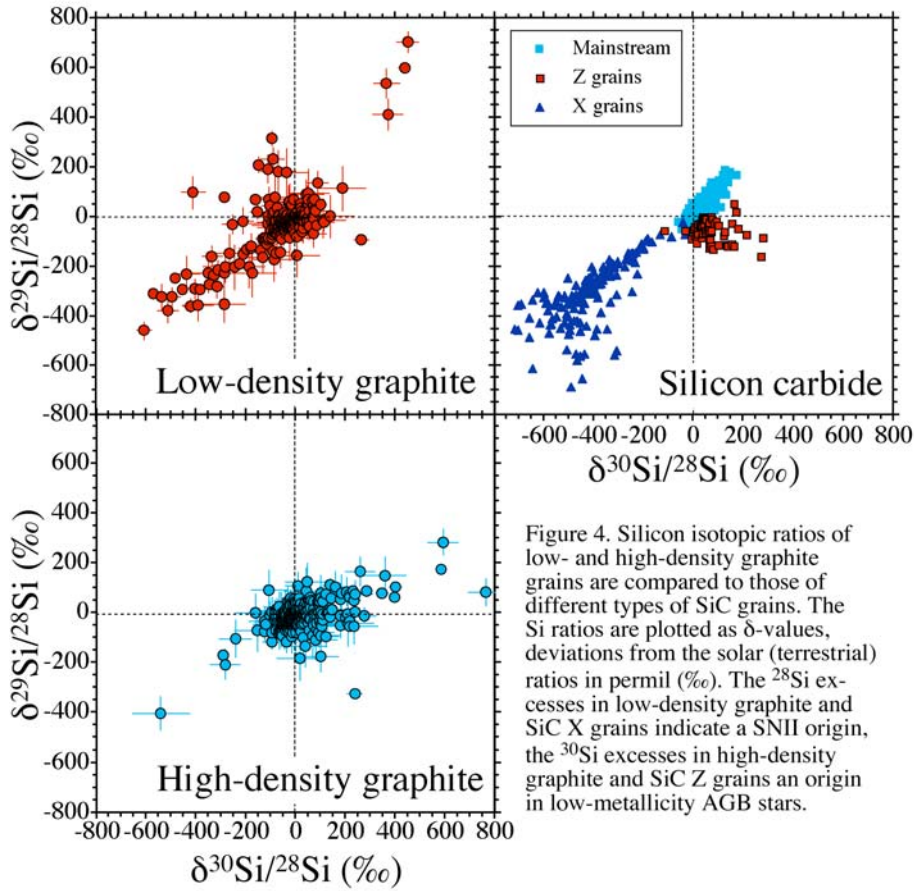


Figure 4. Silicon isotopic ratios of low- and high-density graphite grains are compared to those of different types of SiC grains. The Si ratios are plotted as  $\delta$ -values, deviations from the solar (terrestrial) ratios in permil (‰). The  $^{28}\text{Si}$  excesses in low-density graphite and SiC X grains indicate a SNII origin, the  $^{30}\text{Si}$  excesses in high-density graphite and SiC Z grains an origin in low-metallicity AGB stars.

be accompanied by high  $^{12}\text{C}/^{13}\text{C}$  ratios, resulting from He burning. The  $^{30}\text{Si}$  and  $^{12}\text{C}$  are mixed into the star's envelope by the third dredge-up. Both enrichments are predicted to be much larger in low-metallicity stars than in stars of solar metallicity [13]. Many high-density graphite grains indeed have high  $^{12}\text{C}/^{13}\text{C}$  ratios (Fig. 1). High  $^{12}\text{C}/^{13}\text{C}$  ratios imply high C/O ratios, which might have led to the preferential condensation of graphite over SiC and could explain why we do not find SiC grains with high  $^{12}\text{C}/^{13}\text{C}$  ratios and  $^{30}\text{Si}$  excesses. TEM studies of high-density graphite grains are consistent with an AGB origin. Both low- and high-density grains contain internal sub-grains of carbides, mostly TiC. Whereas such internal grains from low-density graphite have negligible amounts of heavy elements, those from high-density grains have high concentrations of the *s*-process elements Zr, Mo, and Ru [14]. These elements are predicted to be highly enriched in the envelope of AGB stars [15]. Thus several pieces of evidence point to an origin of high-density graphite grains with high  $^{12}\text{C}/^{13}\text{C}$  ratios in low-metallicity AGB stars. However, the stellar origin of the population of high-density grains with  $^{12}\text{C}/^{13}\text{C}$  ratios around 10 (Fig. 1), several of which have substantial  $^{30}\text{Si}$  excesses, remains elusive.

### 3. Neon isotopes

Presolar graphite was originally isolated because it is the carrier of Ne-E(L), almost pure  $^{22}\text{Ne}$ , which is released at low temperature [16]. While Ne-E(H) in presolar SiC grains seems to come from AGB stars, Ne-E(L) is much closer to pure  $^{22}\text{Ne}$  than Ne in the He shell of such stars and in the He/C zone of Type II supernovae. It has therefore been concluded that it originates from the decay of  $^{22}\text{Na}$  ( $T_{1/2} = 2.6$  yr), possibly in novae. However, analysis of individual graphite grains [17] shows that  $^{22}\text{Ne}$ -rich grains have  $^{18}\text{O}$  and/or  $^{28}\text{Si}$  excesses and evidence for the initial presence of  $^{44}\text{Ti}$ , all being signatures of a SN origin. Sodium-22 was apparently implanted into SN graphite grains in the SN ejecta while Ne, much more abundant in the ejecta, was not ionized because of its much higher ionization potential and was not implanted [18].

### 4. Conclusions

Based on the isotopic compositions of presolar graphite grains from primitive meteorites we can identify two stellar sources. Low-density grains are characterized by  $^{15}\text{N}$ ,  $^{18}\text{O}$ ,  $^{28}\text{Si}$  and  $^{49}\text{Ti}$  excesses, high inferred  $^{26}\text{Al}/^{27}\text{Al}$  ratios, as well as evidence for the initial presence of the short-lived radionuclides  $^{22}\text{Na}$ ,  $^{41}\text{Ca}$ , and  $^{44}\text{Ti}$ . All these signatures point to an origin in Type II supernovae. Many high-density graphite grains have high  $^{12}\text{C}/^{13}\text{C}$  ratios, large  $^{30}\text{Si}$  excesses and contain internal grains with high concentrations of the *s*-process elements Zr, Mo, and Ru. These features indicate an origin in low-metallicity AGB stars. The stellar source of high-density grains with  $^{12}\text{C}/^{13}\text{C}$  ratios around 10 remains elusive.

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