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NEW STELLAR SOURCES FOR HIGH-DENSITY, PRESOLAR GRAPHITE GRAINS

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ABSTRACT

We present C, N, O, Si, Al-Mg, K, Ca, and Ti isotopic analyses of seven high-density (ORG1f, $\rho \sim 2.02-2.04$ g cm^{-3}) graphite grains from Orgueil with $^{12}C/^{13}C$ ratios smaller than 20. The presence of ^{44}Ti in three of these grains indicates an origin in Type II supernovae (SNe). The ^{13}C excesses in these SNe grains, however, remain enigmatic. The remaining grains have extremely large Ca and Ti isotopic anomalies, some of which are much larger than those predicted for envelopes of asymptotic giant branch (AGB) stars. These anomalies in conjunction with low $^{12}C/^{13}C$ ratios can only be explained by pure nucleosynthetic He-shell components of AGB stars. Born-again AGB stars that experience a late He flash are able to explain the low $^{12}C/^{13}C$ ratios of some of the grains along with the presence of extreme enrichments in the Ca and Ti isotopes. This study indicates that high-density graphite grains have multiple stellar sources: SNe and born-again AGB stars, in addition to the previously established low-metallicity AGB stars.

Subject headings: dust, extinction — meteors, meteoroids — nuclear reactions, nucleosynthesis, abundances — stars: abundances — stars: AGB and post-AGB — supernovae: general

1. INTRODUCTION

Presolar graphite grains were 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 presolar graphites have also been isolated from the CI chondrite Orgueil (Jadhav et al. 2006). Extensive isotopic studies (Amari et al. 1990, 1995; Hoppe et al. 1995; Zinner et al. 1995; Travaglio et al. 1999; Jadhav et al. 2006) of graphite grains from these meteorites have shown that the isotopic properties of presolar graphites are density-dependent. Many low-density (LD) graphite grains have large excesses in ^{15}N, ^{18}O, and ^{28}Si (Travaglio et al. 1999; Jadhav et al. 2006). A few of these LD grains show evidence for ^{44}Ti (Nittler et al. 1996), have large excesses in ^{41}K (due to the decay of ^{41}Ca) (Amari et al. 1996) and high inferred $^{26}Al/^{27}Al$ ratios. These isotopic compositions, in addition to high $^{12}C/^{13}C$ ratios, are similar to those of SiC-X grains, which are known to have a supernova (SN) origin. Travaglio et al. (1999) were able to match these major isotopic signatures of LD grains by performing mixing calculations of different layers of Type II SNe. On the other hand, a large fraction of high-density (HD) graphite grains appears to have an origin in low-metallicity, asymptotic giant branch (AGB) stars (Zinner et al. 2006a). In transmission electron microscopy (TEM) studies, Bernatowicz et al. (1996) and Croat et al. (2005) found HD grains from Murchison that contain subgrains rich in the *s*-process elements Zr, Mo, and Ru, indicating an AGB origin. Many HD graphites have large ^{30}Si excesses that are correlated with high $^{12}C/^{13}C$ ratios (Amari et al. 2005; Jadhav et al. 2006). Similar signatures are expected for low-metallicity AGB stars where, during the thermally pulsing phase, ^{12}C and $^{29,30}Si$ from the He shell are mixed into the envelope by third dredge-up (TDU) (Busso et al. 1999; Zinner et al. 2006a). These models not only predict high $^{12}C/^{13}C$ ratios but also high C/O ratios, which produce an ideal environment for graphite condensation (Lodders & Fegley 1997). The large, relative excess of C over Si in the envelopes of such stars causes graphite and not SiC grains to condense (Bernatowicz et al. 2006), which could be an explanation for the absence of any SiC grains that have similar high $^{12}C/^{13}C$ ratios and large ^{30}Si excesses (Hoppe et al. 1997; Nittler & Alexander 2003).

Both LD and HD graphite grains from Murchison and Orgueil contain a minor population of grains that have $^{12}C/^{13}C$ ratios $\sim$10. The nucleosynthetic source(s) of grains with such low $^{12}C/^{13}C$ ratios is still unknown. Amari et al. (2001) have suggested J stars and born-again giants like Sakurai's object as the sources of SiC grains of type A and B, which have $^{12}C/^{13}C < 10$. We present here the results of isotopic analyses of seven HD graphite grains with $^{12}C/^{13}C < 20$ from the Orgueil, ORG1f (2.02–2.04 g cm^{-3}) density fraction.

2. EXPERIMENTAL METHODS

Large grains (>2 μm) from the high-density fraction of Orgueil, ORG1f (2.02–2.04 g cm^{-3}), were selected for these measurements. This size selection was made to ensure that multielement isotopic analyses can be made on the grains. Spherical, carbonaceous grains were first identified by X-ray analysis in the scanning electron microscope (SEM) and then transferred with a micromanipulator onto a gold-foil mount. This procedure was essential

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to isolate the grains from the large amounts of macromolecular carbonaceous material, in which graphite grains from Orgueil are often found embedded. Analysis of isolated grains reduces contamination from the surrounding material. The Cameca NanoSIMS 50 at Washington University was used to measure C, N, O, Si, Al-Mg, K, Ca, and Ti isotopes on 44 grains from ORG1f. We used a Cs^+ primary beam to generate negative secondary ions of ^{12}C, ^{13}C, ^{16}O, and ^{18}O in phase 1 of the analyses, and of $^{12}C^{14}N$, $^{12}C^{15}N$, ^{28}Si, ^{29}Si, and ^{30}Si in phase 2. These ions were counted in multidetection mode. In phase 3 of the analyses, positive secondary ions of ^{12}C, ^{24}Mg, ^{25}Mg, ^{26}Mg, and ^{27}Al, produced with an O^- beam, were detected. The K, Ca, and Ti measurements were carried out with the O^- beam in a combination of peak-jumping and multidetection modes. Positive secondary ions of ^{39}K, ^{41}K and ^{43}Ca (B field 1) and ^{12}C, ^{40}Ca, ^{42}Ca, ^{44}Ca and ^{48}Ti (B field 2) were measured to obtain K and Ca ratios. Ti isotopes were measured using three magnetic fields: at B_1 we detected ^{46}Ti, ^{48}Ti, and ^{50}Ti; B_2–^{47}Ti, ^{49}Ti, and ^{51}V: and B_3–^{12}C, ^{40}Ca, ^{48}Ti, ^{50}Ti, and ^{52}Cr. ^{51}V and ^{52}Cr were used to correct the ^{50}Ti signal for isobaric interferences from V and Cr, and, ^{40}Ca was measured to correct for Ca interferences at masses 46 and 48.

The isotopic measurements were carried out at sufficiently high mass resolutions in order to avoid molecular interferences (e.g., SiO at mass 44). The unresolvable isobaric peaks were as follows: (1) ^{46}Ca and ^{46}Ti at mass 46, (2) ^{48}Ca and ^{48}Ti at mass 48, and (3) ^{50}Ti and ^{50}Cr at mass 50. The abundances of ^{46}Ca and ^{48}Ca are so low that we do not expect these interferences to produce significant anomalies. We applied corrections for these interferences by assuming normal or terrestrial $^{46}Ca/^{40}Ca$ and $^{48}Ca/^{40}Ca$ ratios. For the one grain that was found to have a large ^{46}Ti anomaly, we are able to rule out the possibility of a large contribution from the ^{46}Ca signal (see discussion on grain g-9 in § 4). Further, a large ^{48}Ca signal will cause deficits in the Ti isotopic ratios that are normalized to ^{48}Ti. The observed excesses in such grains will, as a result, be lower limits. We also assume that all the contribution from ^{50}Cr to the ^{50}Ti signal is terrestrial. This is a reasonable assumption because the grains acquire large amounts of terrestrial Cr in the laboratory from $Na_2Cr_2O_7$, which is used as an oxidizing agent during the chemical separation procedure for these grains.

Similar to previous measurements (Jadhav et al. 2006), a majority of the HD graphite grains in this study have large $^{12}C/^{13}C$ ratios. We present here the results of our isotopic analyses on a subset of seven grains that have $^{12}C/^{13}C < 20$.

3. RESULTS

C, N, O, Si, *and* Al-Mg *isotopes*.—The $^{12}C/^{13}C$ ratios of the seven grains range from 4 to 18 (Table 1). The N, O, Si, Al-Mg and K isotopic ratios of these grains are listed in Table 1. We were able to obtain $^{14}N/^{15}N$ ratios for only four of these grains. Grain g-29 exhibits an excess in ^{14}N ($^{14}N/^{15}N = 412 \pm 6$), while grain g-o67 is enriched in ^{15}N ($^{14}N/^{15}N = 51 \pm 1$). The other two grains have solar $^{14}N/^{15}N$ ratio ($\sim$272) within errors. Only one grain, g-38, has an anomalous $^{16}O/^{18}O$ ratio of 602 ± 14 (solar $^{16}O/^{18}O \sim 499$), the rest have terrestrial O isotopic ratios. Solar N and O ratios have also been previously obtained on Murchison and Orgueil HD grains (Hoppe et al. 1995; Jadhav et al. 2006). This puzzling result in view of the large range of C isotopic ratios seen in these grains has been attributed to equilibration with normal N and O, either on the parent body or in the laboratory (Hoppe et al. 1995). No stellar source is known to produce anomalous C and normal N simultaneously. The Si isotopes of these seven grains show a similar behavior. Six grains have close to normal $^{29,30}Si/^{2828}Si$ ratios. The largest anomaly was seen in grain g-o67, which is depleted in ^{29}Si ($\delta^{29}Si = -325‰ \pm 18‰$) and enriched in ^{30}Si ($\delta^{30}Si = 235‰ \pm 30‰$). This grain is highly enriched in ^{13}C ($^{12}C/^{13}C \sim 4$; solar 89) and ^{15}N ($^{14}N/^{15}N = 51 \pm 1$). All the grains were found to be normal in $^{25}Mg/^{24}Mg$ within errors. Grain g-o67 has the largest ^{26}Mg excess ($\delta^{26}Mg = 1593‰ \pm 140‰$) and a large inferred $^{26}Al/^{27}Al$ ratio of 0.010 ± 0.001. Another grain, g-38, with a $\delta^{26}Mg$ value of $336‰ \pm 49‰$ has an inferred $^{26}Al/^{27}Al$ ratio of 0.00050 ± 0.00007. The Al/Mg ratios for the remaining grains were too low for detection of radiogenic ^{26}Mg. It is possible that the equilibration processes affecting the N and O isotopes also dilute the Si and Mg isotopes in these HD graphite grains. Thus, apart from the large ^{13}C excesses, these grains are mostly normal in the N, O, Si, and Al-Mg isotopic ratios (Table 1).

K, Ca, *and* Ti *isotopes*.—Six of the grains have a normal $^{41}K/^{39}K$ ratio (0.072) within errors. Grain g-9 is the only one with an elevated $^{41}K/^{39}K$ ratio of 0.080 ± 0.002 (Table 1). Since the intrinsic concentration of K in graphite grains is expected to be very low, we attribute the ^{41}K excess in this grain to the decay of ^{41}Ca ($T_{1/2} = 1.03 \times 10^5 a$) (Amari et al. 1996) and obtain an inferred $^{41}Ca/^{40}Ca$ ratio of 0.002 (Table 1). The Ca and Ti isotopic compositions of the seven grains are given in Table 1 and plotted as isotopic patterns in Figures 1–4. Three of the grains (g-1, g-38, and g-o67) have solar (within errors) or subsolar $^{42}Ca/^{40}Ca$ and $^{43}Ca/^{40}Ca$ ratios but display enormous ^{44}Ca excesses. Their $^{46}Ti/^{48}Ti$ and $^{47}Ti/^{48}Ti$ ratios are normal (within errors) or subsolar, whereas the $^{49}Ti/^{48}Ti$ and $^{50}Ti/^{48}Ti$ ratios are larger than the terrestrial values. The other four grains exhibit large excesses in ^{42}Ca, ^{43}Ca, and ^{44}Ca, and these excesses are especially large in grain g-9. This grain also has large excesses in ^{46}Ti and ^{50}Ti and relatively smaller excesses in ^{47}Ti and ^{49}Ti. The Ti concentrations in grains g-29, g-40, and g-34 were too low to be able to obtain Ti isotopic data.

In the following section we discuss in detail the C, Ca and Ti isotopic ratios and the clues they provide in determining the possible stellar sources for these grains.

4. DISCUSSION

Figure 1*a* shows the Ca isotopic patterns for the three grains, g-1, g-38, and g-o67, which contain large ^{44}Ca excesses. Neutron capture either in AGB stars or in interior zones of Type II SNe can produce excesses in ^{44}Ca relative to ^{40}Ca. However, in all these cases the excesses in ^{42}Ca and ^{43}Ca are expected to be larger than those in ^{44}Ca. This is clearly not the case for these three grains. Furthermore, the ^{44}Ca excesses are much greater than those predicted for the He/C and O/C zones of a type II SNe (Woosley & Weaver 1995). Thus the ^{44}Ca excesses must come from the decay of the short-lived radionuclide, ^{44}Ti ($T_{1/2} = 60a$). During SIMS analysis we observed the ^{44}Ca ion signals for these three grains to be perfectly correlated with the ^{48}Ti ion signals but not with those of the other Ca isotopes, providing additional evidence that the ^{44}Ca excesses must be from the decay of ^{44}Ti. Titanium-44 is produced only in SN explosions (Timmes et al. 1996; Woosley & Weaver 1995; Iyudin et al. 1994; Vink et al. 2001) and hence the initial presence of this isotope in grains g-1, g-38, and g-o67 indicates a SN origin. The derived $^{44}Ti/^{48}Ti$ ratios for these grains are given in Table 1. Titanium-44 is produced in the Ni- and Si-rich zones of type II SNe by α-rich freeze-out from quasi-statistical equilibrium (Woosley et al. 1973; Woosley & Weaver 1995; Timmes et al. 1996) and can be found in graphite grains provided some ^{44}Ti from the inner zones is mixed with material from the carbon-rich, outer zones. The Ti isotopic patterns for these grains (Fig. 1*b*) match those seen in the C/O and He/C zones of a 15 $M_\odot$ SN (Woosley & Weaver 1995). Grain g-o67

TABLE 1

C, N, O, Si, Al-Mg, K, Ca, Ti Isotopic Ratios of High-Density Graphite Grains from ORG1f with $^{12}C/^{12}C < 20$

Grain	$^{14}N/^{15}N$	$^{16}O/^{18}O$	$\delta^{29}Si/^{28}Si$ (‰)	$\delta^{30}Si/^{28}Si$ (‰)	$\delta^{25}Mg/^{24}Mg$ (‰)	$\delta^{26}Mg/^{24}Mg$ (‰)	$^{26}Al/^{27}Al$ ($\times 10^{-3}$)	$^{41}K/^{39}K$ ($\times 10^{-2}$)
g-1	267 ± 18	520 ± 7	−20 ± 13	−35 ± 18	22 ± 25	−15 ± 23	[b]	7.310 ± 0.238
g-38	[a]	602 ± 14	−68 ± 21	−20 ± 28	−6 ± 42	336 ± 49	0.472 ± 0.069	7.938 ± 0.771
g-o67	51 ± 1	500 ± 11	−325 ± 18	235 ± 30	1 ± 78	1593 ± 140	9.732 ± 0.857	7.302 ± 0.139
g-9	294 ± 7	518 ± 10	−27 ± 24	−74 ± 30	1 ± 23	−23 ± 22	[b]	8.031 ± 0.184
g-29	412 ± 6	525 ± 9	−34 ± 19	−65 ± 25	−26 ± 15	−1 ± 15	[b]	7.281 ± 0.091
g-40	[a]	519 ± 9	−7 ± 19	−38 ± 24	−22 ± 11	−13 ± 11	[b]	7.247 ± 0.058
g-34	[a]	515 ± 11	−16 ± 20	−47 ± 25	5 ± 26	6 ± 25	[b]	7.249 ± 0.073

Grain	$^{12}C/^{13}C$	$^{41}Ca/^{40}Ca$ ($\times 10^{-3}$)	$\delta^{42}Ca/^{40}Ca$ (‰)	$\delta^{43}Ca/^{40}Ca$ (‰)	$\delta^{44}Ca/^{40}Ca$ (‰)	$^{44}Ti/^{48}Ti$ ($\times 10^{-3}$)	$\delta^{46}Ti/^{48}Ti$ (‰)	$\delta^{47}Ti/^{48}Ti$ (‰)	$\delta^{49}Ti/^{48}Ti$ (‰)	$\delta^{50}Ti/^{48}Ti$ (‰)
g-1	14	0.3 ± 0.9	−152 ± 96	156 ± 239	6018 ± 203	0.179 ± 0.008	−15 ± 8	−28 ± 9	161 ± 11	44 ± 15
g-38	4	1.2 ± 1.2	−118 ± 177	−602 ± 274	51886 ± 1499	0.765 ± 0.028	−77 ± 12	−98 ± 15	125 ± 6	242 ± 6
g-o67	4	0.3 ± 0.3	63 ± 52	15 ± 107	3767 ± 77	0.595 ± 0.025	2 ± 4	14 ± 4	169 ± 5	236 ± 6
g-9	18	2.3 ± 0.4	16028 ± 316	27641 ± 805	9396 ± 151	[c]	35032 ± 4432	1376 ± 371	2278 ± 298	32827 ± 4594
g-29	10	0.2 ± 0.2	1451 ± 50	1628 ± 100	540 ± 24	[c]	[d]	[d]	[d]	[d]
g-40	11	0.2 ± 0.3	3375 ± 65	3662 ± 138	554 ± 22	[c]	[d]	[d]	[d]	[d]
g-34	8	0.1 ± 0.1	5064 ± 60	7410 ± 146	2179 ± 25	[c]	[d]	[d]	[d]	[d]

Note.—Errors are 1 σ; δ values are deviations from solar ratios per mil.

[a] Not measured because a large signal from $^{13}C_2$ gives rise to an unresolvable interference at mass 26 while measuring $^{12}C^{14}N$.

[b] No $^{26}Al/^{27}Al$ ratios were derived for these grains since they do not exhibit any ^{26}Mg excesses.

[c] Did not derive $^{44}Ti/^{48}Ti$ ratio for these grains because the $\delta^{44}Ca/^{40}Ca$ values are comparable to the $\delta^{42}Ca/^{40}Ca$ and $\delta^{43}Ca/^{40}Ca$ values.

[d] Not measured because of very low Ti signal.

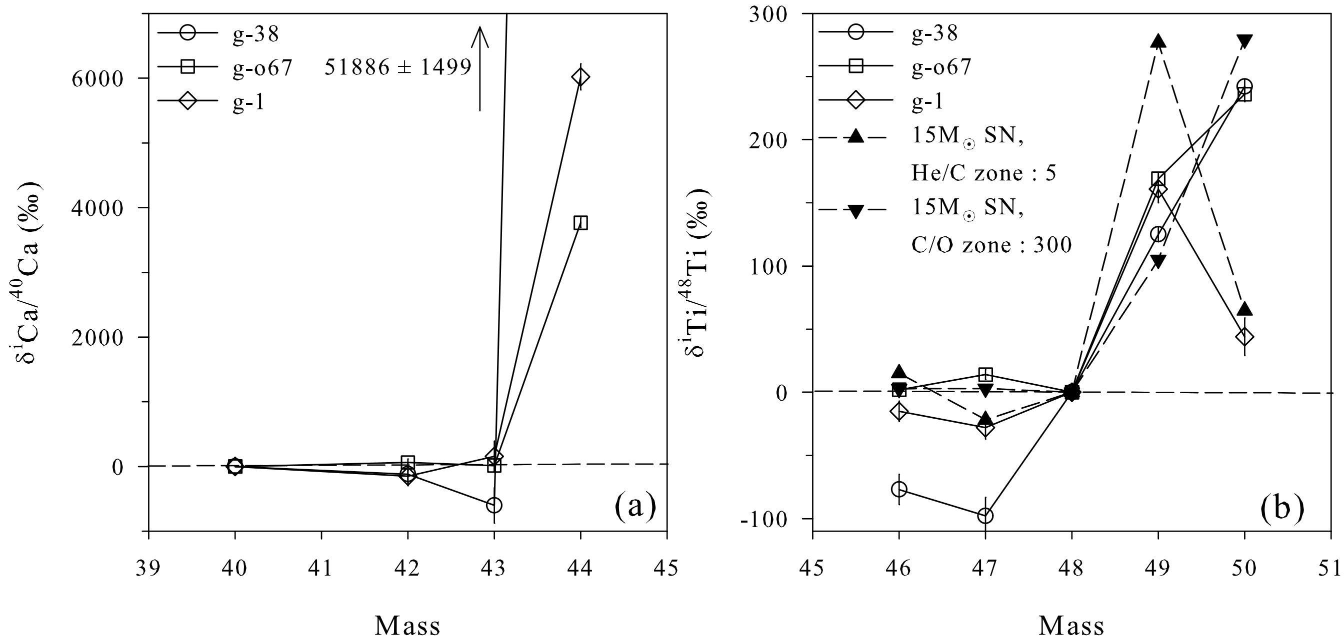

FIG. 1.—(*a*) Ca and (*b*) Ti isotopic patterns observed in three graphite grains are plotted vs. mass number and compared with theoretical models of stellar sources. The ratios are plotted as delta values, deviations from solar ratios in per mil (‰). Error bars in this and subsequent plots are 1 σ. (*a*) The grains, g-38, g-o67, g-1, have ^{44}Ca excesses that vastly exceed any anomalies in ^{42,43}Ca, indicating the initial presence of the short-lived radionuclide ^{44}Ti. (*b*) Grain g-1 has a Ti isotopic pattern similar to that seen in the He/C zone of a 15 $M_\odot$ SN, while the patterns for grains g-38 and g-o67 agree well with those predicted for the C/O zone of a 15 $M_\odot$ SN. The theoretical isotopic ratios seen in these zones are normalized to match the size of the anomalies observed in the grains.

exhibits an excess in ^{15}N and has a high ^{26}Al/^{27}Al ratio—signatures consistent with a SN origin. Nitrogen-15 is abundant at the bottom of the He/C zone and high ^{26}Al/^{27}Al ratios are found in the He/N zone that had undergone H burning (Forestini et al. 1991; Mowlavi & Meynet 2000; Karakas & Lattanzio 2003). The only highly unusual signature in this grain is its low ^{12}C/^{13}C ratio ($\sim$4). Most of the SiC X grains that show evidence for ^{44}Ti have higher ^{12}C/^{13}C ratios (Nittler et al. 1996). The He/N zone has a low ^{12}C/^{13}C ratio, however, in order to achieve the low ^{14}N/^{15}N ratio and the Ti isotopic signatures of this grain, material from the underlying He/C and C/O zones has to be mixed to material from the He/N zone. These two zones have not only extremely high ^{12}C/^{13}C ratios but also C contents that are higher than that in the He/N zone by large factors. Thus, it is unclear how grain g-o67 can have such a low ^{12}C/^{13}C ratio after admixture of material from lower zones to match the Ti isotopic ratios. A similar problem is posed by the other two SN grains. Grain g-38 has a moderately high ^{26}Al/^{27}Al ratio (0.0005 $\pm$ 0.00007), lower than what is found in almost all SN zones with the exception of the lower layers of the He/C zone. Grain g-1 is normal (within errors) in N, O, Si, Al-Mg, and K isotopes. Both grains have low ^{12}C/^{13}C ratios ($\sim$4 and 14) but have ^{49}Ti and ^{50}Ti excesses that require contributions from zones with large abundances of almost pure ^{12}C.

Grain g-9 has extreme ^{42,43,44}Ca and ^{46,47,49,50,}Ti excesses (Figs. 2*a* and 2*b*). These excesses and the ^{41}Ca/^{40}Ca ratio (0.0023 $\pm$ 0.0004) are incompatible with an AGB origin. The ^{41}Ca/^{40}Ca ratios expected in AGB stars are on the order of 10^{-4} to 10^{-5} (Amari et al. 1996; Gallino et al. 1998; Wasserburg et al. 1995). High ratios can be obtained in the He/C, C/O, and O-rich zones of Type II SNe where the ^{22}Ne(α, n)^{25}Mg reaction provides ample neutrons for the production of ^{41}Ca (Woosley & Weaver 1995). That the observed ^{41}Ca/^{40}Ca ratio in grain g-9 is smaller can be explained by mixing with material from the He/N zone or isotopic equilibration. On the other hand, as seen in Figure 2*a*, the Ca isotopic excesses observed in grain g-9 are too high to permit any mixing with the He/N zones. They are higher than those found in the He/C or C/O zones of a 15 $M_\odot$ SN, and one has to go to the O/C and O/Ne zones to match the excesses seen in the grain (Fig. 2*a*). Figure 2*b* shows that the C/O zone could explain the large ^{49,50}Ti excesses but not the ^{46}Ti excess. The O/C and O/Ne zones are able to account for the large ^{47,49,50}Ti anomalies (but not ^{46}Ti) seen in this grain. If grain g-9 originated in a SN then it requires pure O/C or O/Ne zone materials to explain its Ca and Ti isotopic compositions. However, the C in all these zones is essentially pure ^{12}C and the carbon abundances are much higher than that in the He/N zone, the only place with a low ^{12}C/^{13}C ratio of $\sim$3.6 (Woosley & Weaver 1995). This makes it impossible to achieve the low ^{12}C/^{13}C ratio of 18 in grain g-9 while maintaining the large Ca and Ti excesses by mixing of material from the He/N zone with material from the C/O, O/C, or O/Ne zones. What makes the situation even more difficult is that the extremely high Ca isotopic ratios in the O/C and O/Ne zones are in part the result of a substantial drop in the abundance of ^{40}Ca (the reference isotope in these ratios) as one goes below the He/C zone.

The origin of the huge ^{46}Ti excess in g-9 is unknown. Two possible explanations for this large ^{46}Ti excess can be ruled out. A large contribution could have been from ^{46}Ca because in our SIMS measurement we could not resolve the ^{46}Ca and ^{46}Ti signals from one another (this requires a mass resolving power of 43,233, which is beyond the capability of the NanoSIMS). We used the measured Ca/Ti ratio of this grain ($\sim$35) to calculate the ^{46}Ca signal expected from the abundance pattern in a AGB He-shell (see below) or a SN zone that matches the other Ca isotopic ratios of the grain. We found that the expected contribution from ^{46}Ca is much too small to explain the large ^{46}Ti excess observed in this grain. Alternatively, an $\sim$1% contribution from the Si/O zone of a 15 $M_\odot$ SN, which has a high abundance of

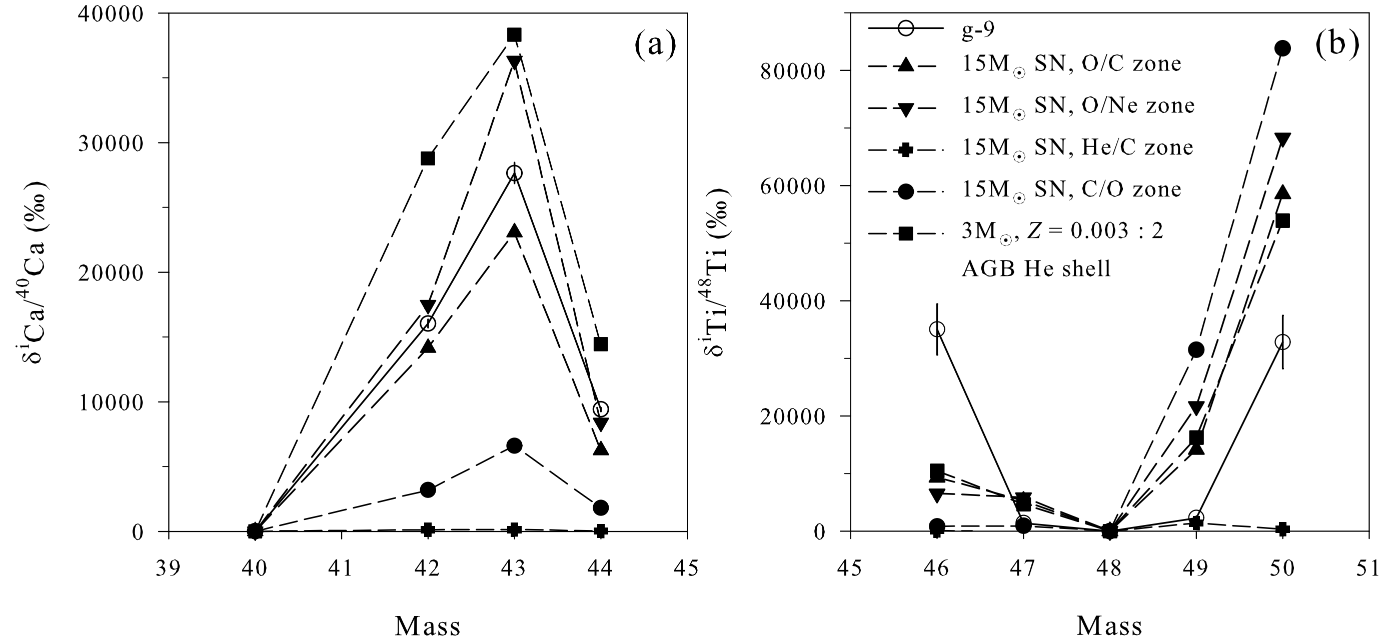

FIG. 2.—(*a*) Ca and (*b*) Ti isotopic patterns observed in grain g-9 are plotted vs. mass number and compared with theoretical predictions. The ratios are plotted as delta values. This grain has Ca and Ti isotopic patterns that are similar to those predicted for O-rich (O/C and O/Ne) zones of a 15 $M_{\odot}$ SN and for the He shell of a 3 $M_{\odot}$ AGB star with metallicity $Z = 0.003$. The predicted pattern of the He shell was diluted by a factor of 2 to fit the Ca isotopic pattern seen in the grain. However, the large ^{46}Ti excess cannot be explained. This grain has a $^{12}C/^{13}C$ ratio $\sim$18.

^{46}Ti (Woosley & Weaver 1995), could explain the ^{46}Ti excess. However, such a contribution will result in a large ^{40}Ca excess and in turn, deficits in ^{43,44}Ca, which are not observed. It is essential to point out at this stage that it is not necessary for both the Ca and Ti anomalies in a single graphite to originate from the same stellar source. This is because all the Ti is obtained from subgrains within the graphite grain while Ca is found to be uniformly distributed in the host grain. The parent grain and the subgrain could have condensed in completely different stellar environments and the Ca and Ti anomalies could represent these distinct stellar sources.

In the remainder of this discussion we will attempt to explain the Ca and Ti excesses in grains g-9, g-34, g-29, and g-40 by comparing them with predictions for the He shell of AGB nucleosynthesis models. These models have been discussed in detail by Gallino et al. (1998) and more recently, by Zinner et al. (2006b). The Gallino et al. (1998) models calculate nuclear abundances at the surface of AGB stars by mixing He shell material into the envelope by TDU. However, we use pure He shell predictions (without dilution with envelope material) from these models to explain the excesses seen in the aforementioned grains.

The size of the Ca and Ti excesses seen in grain g-9 is incompatible with an AGB star origin since no AGB nucleosynthetic model can account for such high $\delta^{42,43,44}$Ca and $\delta^{47,49,50}$Ti values in its envelope. These signatures can be matched by slight dilution of pure He shell material of a 3 $M_{\odot}$, low-metallicity ($Z = 0.003$) AGB star (Figs. 2*a* and 2*b*). However, the He shell of an AGB star consists of almost pure ^{12}C, making it difficult to explain the ^{13}C excess in this grain. We shall discuss a possible stellar source that can explain the signatures seen in grain g-9 in detail further below, since the remaining grains also require a similar explanation.

Grain g-34 has a low $^{12}C/^{13}C$ ratio of $\sim$8 and has a Ca isotopic pattern that matches closely that expected in the C/O zone of a 15 $M_{\odot}$ SN (Fig. 3). However, this and underlying zones contain essentially pure ^{12}C. Similar to grain g-9, mixing between the He/N zone and the O/C and O/Ne zones cannot match both the Ca isotopic ratios and the low $^{12}C/^{13}C$ ratio of this grain. Alternatively, the Ca excesses of this grain can be explained by the He shell of a 2 $M_{\odot}$, $Z = 0.003$ AGB star (Fig. 3).

The situation is the same for the last two grains, g-29 and g-40. Although excesses in ^{42,43,44}Ca are smaller than those in the previously discussed grains, any admixture from the underlying SN zones to material from the He/N zones required to match the Ca isotopic ratios increases the $^{12}C/^{13}C$ ratio far beyond those (10 and 11) measured in these grains. On the other hand, the Ca

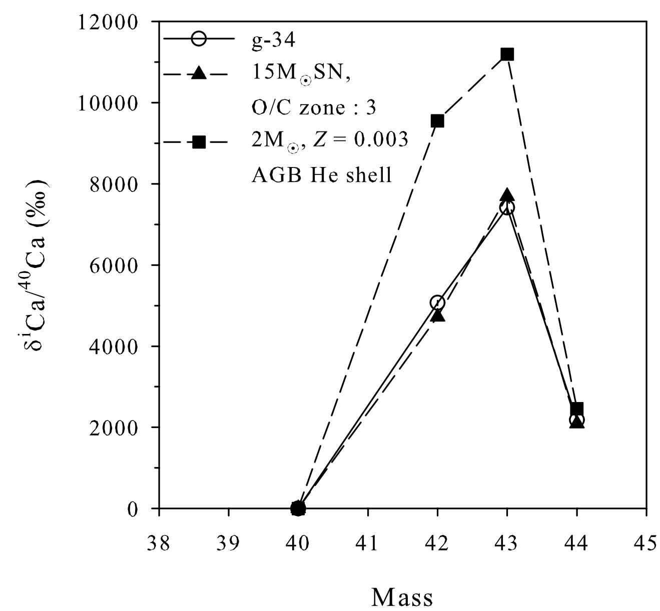

FIG. 3.—Ca isotopic pattern observed in grain g-34, is plotted vs. mass number and compared with model predictions. The ratios are plotted as delta values. This grain has a $^{12}C/^{13}C$ ratio of $\sim$8 and a Ca isotopic pattern that matches the pattern predicted for the C/O zone of a 15 $M_{\odot}$ SN and that of the He shell of a 2 $M_{\odot}$ AGB star with metallicity, $Z = 0.003$ (divided by 3).

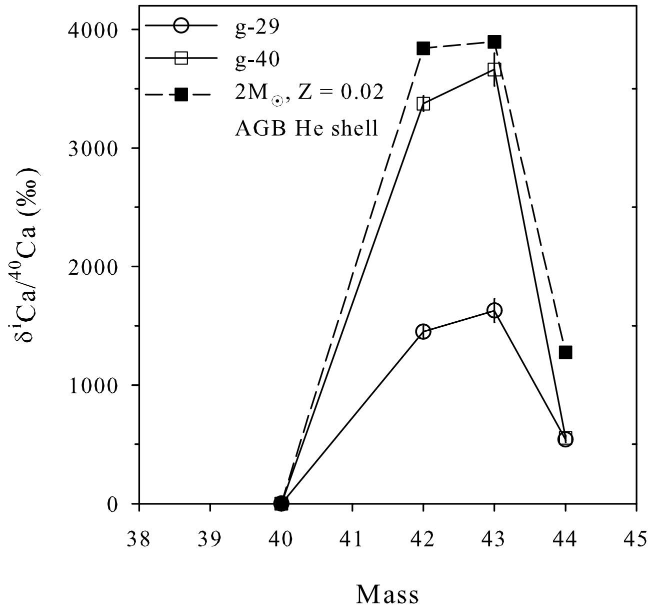

FIG. 4.—Ca isotopic patterns obtained for grains g-29 and g-40, are plotted vs. mass number and compared with that predicted for the He shell of a 2 $M_\odot$ AGB star of solar metallicity. The ratios are plotted as delta values. Both grains are highly enriched in ^{13}C ($^{12}C/^{13}C$ ratio of g-29 is 10 and of g-40 is 11). These grains could have originated from born-again AGB stars, which have envelopes with very low $^{12}C/^{13}C$ ratios and enrichments in s-process elemental abundances.

anomalies are much larger than those predicted for the envelope of AGB stars. Apart from the high C and Ca isotopic anomalies in both these grains, grain g-29 also exhibits an excess in ^{14}N ($^{14}N/^{15}N \sim 412$).

The Ca patterns of g-29 and g-40 match those of pure nucleosynthetic components theoretically predicted for the He shell of a 2 $M_\odot$ AGB star of solar metallicity (Fig. 4). However, both grains have $^{12}C/^{13}C$ ratios of $\sim$10 that completely disagree with the type of nucleosynthesis taking place in a He-burning shell. The basic problem is that the Ca (and Ti) isotopic compositions of our graphite grains show the signature of neutron capture, requiring He burning, which results in the production of ^{12}C. This is in disagreement with the low $^{12}C/^{13}C$ ratios found in the grains, which are the result of H burning in the CNO cycle. One possible stellar source that combines an *s*-process signature with that of H burning are born-again AGB or post-AGB stars that undergo a very late He flash. These stars, typified by Sakurai's object (V4334Sgr), undergo a very late thermal pulse (VLTP) during their descent along the white dwarf cooling track, long after they have left the AGB phase (Asplund et al. 1999). The spectroscopic observations of Sakurai's object by Duerbeck & Benetti (1996), Asplund et al. (1997, 1999), and Kipper & Klochkova (1997) have shown that it is rich in He, C, N, O (C > O) and the *s*-process elements, along with being deficient in H. The object also has a $^{12}C/^{13}C$ ratio of 1.5–5.0. Complete H-burning is evidenced by H deficiency and high He abundances. The simultaneous occurrence of a low $^{12}C/^{13}C$ ratio along with *s*-process elemental enrichments indicates that H-burning occurred after the star left the AGB track. At this stage in its evolution the star has lost most of its envelope as a planetary nebula and only a thin residual H-rich layer is left. Herwig et al. (1999) and Herwig (2001) studied evolutionary models of 2 $M_\odot$ stars that undergo a VLTP. They showed that in such objects the He-flash-powered convection zone, which is rich in freshly produced ^{12}C, can mix with the residual H-rich envelope and cause convective H burning via the CN cycle. The $^{12}C/^{13}C$ ratio is thus drastically reduced as ^{12}C is converted to ^{13}C. The surface of the star is H-rich before the convection zone extends into the envelope but becomes rapidly depleted in H as convective H-burning continues. The envelope continues to remain C-rich because it is thin and the residual envelope had only a limited number of protons, which prevents the CN cycle from reaching equilibrium (^{14}N is less abundant than ^{12}C) before all the H is exhausted. Iben (1984) and Renzini (1982) estimated that 10%–25% of stars that leave the AGB phase and are on the white dwarf track undergo a late He flash.

Thus, we propose that grains g-29 and g-40, and possibly g-34 and g-9, condensed in the atmosphere of such an object. The low $^{12}C/^{13}C$ ratios in the grains are explained by limited H burning of the residual envelope. The large neutron-capture effects in Ca and Ti are explained by the fact that the heavy-element compositions on the surface of a born-again AGB star are very similar to those of the He shell because there is not much envelope material to dilute them. The currently cooling Sakurai's object is known to be producing large quantities of C-rich dust (Asplund et al. 1997, 1999). Infrared observations of the atmosphere of this star indicate the presence of graphite dust in the atmosphere (Eyres et al. 1998). Born-again stars, like Sakurai's object, have also been considered to be viable candidates for the sources of SiC grains of type A and B (Amari et al. 2001). These grains are characterized by their low $^{12}C/^{13}C$ ratios (<10) and a large range of $^{14}N/^{15}N$ ratios (40–10^4). Some of them were also found to be enriched in *s*-process elements.

In summary, we find that seven graphite grains with $^{12}C/^{13}C <$ 20 have puzzling origins. The three grains g-1, g-38, and g-o67 certainly come from type II SNe as indicated by the presence of ^{44}Ti. There exists no explanation for their low $^{12}C/^{13}C$ ratios in conjunction with their Ti isotopic compositions. Similar signatures have been seen by Nittler et al. (1996) in one LD graphite grain from Murchison (KE3c-242). This grain shows evidence for ^{44}Ti, has a $^{12}C/^{13}C$ ratio of 6.91 $\pm$ 0.02, an $^{26}Al/^{27}Al$ ratio of 0.0018 $\pm$ 0.0003 and has normal N and O. Nittler & Hoppe (2005) found one SiC grain that has ^{28}Si, ^{49}Ti, and ^{44}Ca excesses and a high inferred $^{26}Al/^{27}Al$ ratio but a very low $^{12}C/^{13}C$ ratio. All the isotopes in this SiC grain indicate a SN origin except the C isotopic compositions.

One remaining puzzle is why grains g-9, g-29, g-34, and g-40 seem to retain almost pure nucleosynthetic components in Ca and Ti, but are essentially normal in their Mg and Si isotopic compositions. The neutron-capture reactions that affected the Ca and Ti isotopes of the grains should also have changed their Mg and Si isotopic compositions significantly. Isotopic equilibrations, as invoked for N, are a possibility but we have to better understand siting and bonding of various trace elements in graphite grains before we can arrive at a conclusion.

Material for the seven grains that have been discussed above still remains after the SIMS analyses. We intend to measure heavy element (Mo, Zr, Sr, Ba) isotopes in these grains with resonant ionization mass spectrometry (RIMS) at the Argonne National Laboratory. Such coordinated analyses might help to address some of the unanswered questions regarding these grains.

5. CONCLUSIONS

The measurements of Ca and Ti isotopic compositions in high-density graphite grains with $^{12}C/^{12}C < 20$ lead us to consider Type II SNe and born-again AGB stars as stellar sources of high-density graphite grains, in addition to low-metallicity AGB stars. Thus, high-density graphite grains seem to have multiple stellar sources. More isotopic systems need to be measured on the grains discussed in this paper and additional grains with similar ^{13}C excesses need to be isolated and studied extensively in order to better constrain the stellar sources of this particular population of presolar graphite grains. We hope that this study will trigger theoretical efforts to create additional nucleosynthetic models that will result in a better understanding of the isotopic signatures seen in such grains. The detection and study of more objects like Sakurai's object could strengthen the hypothesis that these objects are valid stellar sources of some presolar grains.

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