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Ne ISOTOPES IN INDIVIDUAL PRESOLAR GRAPHITE GRAINS FROM THE MURCHISON METEORITE TOGETHER WITH He, C, O, Mg-Al ISOTOPIC ANALYSES AS TRACERS OF THEIR ORIGINS

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ABSTRACT

Ne isotopes measured in individual presolar graphite grains, solid samples of extinct stars preserved in primitive meteorites, provide information on the type of stellar sources of the grains and on nucleosynthetic mixing and ion-trapping processes which were operating. We present Ne and He isotope analyses of single presolar graphite grains from the KFB1 density fraction extracted from the carbonaceous chondrite Murchison. In addition, we measured isotopes of C, O, and Mg-Al with the NanoSIMS ion microprobe to better constrain the origin of the grains. Eleven out of 51 presolar graphite grains contain nucleosynthetic ^{22}Ne above our detection limit. This fraction of ^{22}Ne-rich grains is similar to the one reported by Nichols et al. although we have a lower ^{22}Ne detection limit. We detected rare He-shell ^{20}Ne in one ^{22}Ne-rich grain and obtained the ^{20}Ne/^{22}Ne ratio (0.03 ± 0.02) of the He-shell of an Asymptotic Giant Branch (AGB) star with 1.5–2 $M_\odot$ and subsolar metallicity. We also detected ^{4}He in this grain, while in the other grains, which originally acquired He, He-loss seems to be significant. We found unequivocal evidence for radiogenic ^{22}Ne (Ne-R) in another graphite grain, which likely condensed in a core-collapse supernova and which incorporated live radioactive ^{22}Na ($t_{1/2}$ = 2.6 yr). For the other grains, a clear assignment to a stellar source is more difficult to make. Putative stellar sources are supernovae, AGB stars, born-again AGB stars, J-type carbon stars, and CO novae.

Key words: circumstellar matter – dust, extinction – methods: analytical – methods: laboratory – nuclear reactions, nucleosynthesis, abundances

1. INTRODUCTION

Isotopically highly anomalous neon was discovered by Black & Pepin (1969) in carbonaceous chondrites. This neon, highly enriched in its heaviest isotope ^{22}Ne and named Ne-E, was assigned to a circumstellar source. This anomalous Ne component later led to the discovery and isolation of its carrier phase presolar graphite, primarily round grains >1 μm in diameter (Amari et al. 1990). The primary motivations for laboratory investigations of presolar graphite are to elucidate their stellar sources, to investigate nucleosynthetic processes, and to study dust condensation around stars.

Noble gas studies of different density fractions of bulk samples of millions of presolar graphite grains isolated from the carbonaceous chondrite Murchison (Amari et al. 1995) revealed that the samples actually contained two different ^{22}Ne-rich components. One component, released from the bulk samples during analysis at low temperature heating steps, was named Ne-E(L). The second one, released at high-temperature steps, was named Ne-E(H) (Eberhardt et al. 1981) and is slightly less ^{22}Ne-rich than the former. Subsequent work (Amari et al. 1995) found that Ne-E(L) released from the millions of grains actually consists itself of two distinct components: mainly radiogenic Ne-R (^{22}Ne from the decay of radioactive ^{22}Na, $t_{1/2}$ = 2.6 yr; Clayton 1975) as well as a minor contribution of nucleosynthetic Ne-G, produced in the He-shell in Asymptotic Giant Branch (AGB) stars (Gallino et al. 1990, [^{20}Ne/^{22}Ne]-G ≈ 3×10^{-2} to 1×10^{-1}, Heck et al. 2007) carried by a different population of grains. In contrast, the component Ne-E(H) exclusively consists of Ne-G. In this study, we only use the pure endmember components Ne-G and Ne-R. Na-22 is produced in supernovae and novae (Clayton 1975; Clayton & Hoyle 1974). Graphite grains from density fractions where Ne-R has been detected have ^{18}O excesses and Si isotopic anomalies (mainly in the form of ^{28}Si excesses). This leads to the assumption that many of the graphite grains originated in supernova explosions. Subsequent studies pointed out that novae only play a minor role as sources for graphite (e.g., Amari 2006). Gehrz et al. (1998) estimated that novae contribute only 3‰ to the interstellar dust inventory. The work on bulk samples showed that the original Ne isotopic composition acquired in a presolar, stellar environment has been largely preserved in presolar graphite.

To constrain the graphite's origin further, it is important to know the isotopic composition of noble gases as well as other diagnostic nuclides from individual grains. Helium and Ne analyses of single presolar graphite grains from Murchison were pioneered by Nichols et al. (1992). They discovered that in the density fraction KFB1 from Murchison (2.10–2.15 g cm^{-3}), ∼29% of the presolar graphites (14 out of 49 grains) contained measurable amounts of ^{22}Ne. This is the same density fraction studied again in this work. In the low-density fraction KE3 (1.65–1.72 g cm^{-3}), Nichols et al. (1994) found a similar proportion (33%; 7 out of 21 grains) of ^{22}Ne-rich grains, while in the higher density fraction KFC1 (2.15–2.20 g cm^{-3}), only ∼7% of the grains (3 out of 46 grains) are ^{22}Ne-rich (Kehm et al. 1996). None of the grains studied by these authors revealed the presence of any ^{20}Ne, ^{21}Ne, or ^{4}He above their detection limits. The most gas-rich grains account for most of the total gas amount. It remains an open question whether the Ne-R

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was implanted into the grains after they formed or whether the Ne-R precursor nuclide ^{22}Na was incorporated into the grains during their condensation (e.g., Amari 2006; Amari 2009). It also remains elusive whether the grains with no noble gases detected never acquired any gas or the gas amounts are simply below detection limits.

A large database of C, N, O, Mg-Al, Si, and Ca-Ti isotopes of single presolar graphite grains was obtained by Hoppe et al. (1995) and Travaglio et al. (1999). It has been observed that each graphite density fraction has its own isotopic and trace element characteristics and hence represents a distinct mixture of products from different stellar sources. For example, in low-density graphite grains, nucleosynthetic signatures are preserved in the isotopes of C, O, Si, and N, while in high-density graphite grains, O and N show normal composition which is most likely due to equilibrium exchange with solar/terrestrial O and N. Radiogenic ^{26}Mg, ^{41}K, and ^{44}Ca (from the short-lived radionuclides ^{26}Al, ^{41}Ca, and ^{44}Ti, respectively) were detected in several graphite grains (Hoppe et al. 1995; Nittler et al. 1996; Amari et al. 1996; Travaglio et al. 1999). These analyses suggest that core-collapse supernovae (Type II) play an important role as sources particularly of low-density presolar graphite. In contrast, more high-density grains (KFC1 density fraction) bear signatures, e.g., the chemical composition of internal refractory carbides and the isotopic compositions of heavy elements, from AGB stars (e.g., Croat et al. 2005; 2008). The isotopic compositions of a few grains are qualitatively consistent with a nova origin (Amari et al. 2001a; José & Hernanz 2007).

Presolar ^{22}Ne without any detectable amounts of ^{20}Ne has also been observed in single presolar SiC grains from the carbonaceous chondrites Murchison (Nichols et al. 1992; Heck et al. 2007) and Murray (Heck et al. 2007), where it was mostly accompanied by ^{4}He. This information, together with C, N, and Si isotopic data of these grains, indicated that in contrast to presolar graphite, most of the SiC grains condensed in the outflows of low-mass AGB stars and the noble gases were implanted later by a fast stellar wind in the post-AGB phase (Heck et al. 2007). Only 1% of presolar SiC formed in supernovae (e.g., Amari et al. 1992; Hoppe et al. 2000). It is still an open question why the fraction of presolar graphite grains with a likely supernova origin is distinctly higher than that of presolar SiC grains (Zinner 2004).

In this study, we aim to determine the fraction of ^{22}Ne-rich graphite grains with the advantage of a higher Ne-sensitivity compared with the first study of noble gases in single graphite grains (Nichols et al. 1992). Rare gas analyses of single micron-sized presolar gains are very challenging due to the extremely small gas amounts. Furthermore, we investigate the grain-size dependency of the Ne concentration to shed light on the trapping mechanism of Ne into presolar graphite. One of our main goals is also to search for the rare, previously not detected ^{20}Ne and ^{4}He in individual presolar graphite grains. The Ne isotopic composition (i.e., the ^{20}Ne/^{22}Ne ratio), in conjunction with other isotope systems, is useful to to discern between supernova, nova, and AGB star origins, and in particular to distinguish between different nova types. We compare the light noble gas composition, together with isotopic data of C, O, and Mg-Al, with stellar nucleosynthetic model predictions to better constrain the stellar sources of gas-rich presolar graphite grains. Such an approach is only possible with single grain analyses.

Table 1

He and Ne detection limits (10^{-15} cm^3 STP) of the three noble gas analysis sessions. Data from session 1 have been rejected due to the high ^{4}He, ^{20}Ne, and ^{22}Ne detection limits

	^{4}He	^{20}Ne	^{21}Ne	^{22}Ne
Session 1	1300	60	0.51	6.2
Session 2	230	31	0.27	3.6
Session 3	230	0.33	37	1.7

2. SAMPLES AND EXPERIMENTS

Presolar graphite was extracted from Murchison at the University of Chicago using an acid dissolution and density separation technique described by Amari et al. (1994). Graphite grains from the acid residue of the KFB1 density fraction (2.10–2.15 g cm^{-3}) were mounted on ultraclean Au-foil and imaged in the secondary electron microscope (SEM). Thereafter, secondary ions ^{12}C^{-}, ^{13}C^{-}, ^{16}O^{-}, ^{18}O^{-}, and ^{28}Si^{-} from 134 selected grains were analyzed in multicollection mode using a Cs^{+} primary ion beam with the NanoSIMS ion-microprobe at Washington University in St. Louis. Special care was taken to consume only small sample amounts in order to provide enough material for subsequent rare gas analyses. The primary beam was rastered over the exposed grain surface (2×2 μm^2 and 3×3 μm^2 raster fields). The measurement was stopped when uncertainties of C isotope ratios based on counting statistics were better than 1%. A previous study suggests that short NanoSIMS analyses of major element isotopes do not have a detectable effect on the noble gas concentrations (Heck et al. 2007). Sixty presolar graphite grains (ø 1.7–6.2 μm), with a spatial separation to their neighbors considerably larger than the laser beam spot diameter (~50 μm), were analyzed for He and Ne isotopic composition at ETH Zurich. Gases were extracted by melting single grains with an Nd-YAG IR laser. The glowing of the grains was monitored with a video camera and reached its maximum intensity usually in less than one minute of continuous energy increase. After gas cleaning with getters and cold traps the sample gas was pumped almost quantitatively into the ion-source by a special-purpose molecular drag pump to achieve high sensitivity (Baur 1999). The He-4^{+}, ^{20}Ne^{+}, ^{21}Ne^{+}, and ^{22}Ne^{+} as well as interfering ions were detected with an electron multiplier in ion-counting mode using peak jumping. For each measurement the instrument's memory and background signals were recorded and subtracted from the sample data. Our detection limits—defined by the 2σ scatter of blank measurements—for the three analyses sessions are given in Table 1. This procedure has been developed for the analyses of individual presolar SiC grains. A more detailed description is given by Heck et al. (2007). Data from the first analysis session were discarded due to the high scatter of the blank data. We subsequently focus on the data of 51 grains from sessions 2 and 3 (Figure 1, Tables 2 and 3).

After noble gas analyses, the melt residues of the grains were imaged in the SEM at the MPI for Chemistry in Mainz to verify whether grains had been completely melted, and neighboring grains were checked whether they had remained unaffected by the laser beam.

Thirteen of 51 melt residues of graphite grains which completely melted but still contained large enough amounts of material for ion microprobe study (Figure 2) were selected for analysis of the diagnostic isotope system Mg-Al. The secondary ions ^{24}Mg^{+}, ^{25}Mg^{+}, ^{26}Mg^{+}, and ^{27}Al^{+} were measured in

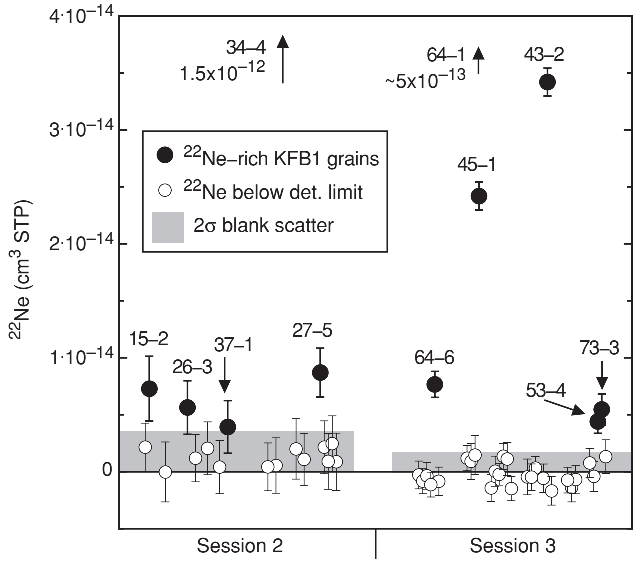

Figure 1. ^{22}Ne gas amounts of presolar graphite grains from the KFB1 density fraction from Murchison during two analysis sessions. The average blank value has been subtracted from the data. Detection limits are defined by the 2σ scatter of blank data of one session. Data from the first session have been rejected due to a high detection limit. Error bars on data points in all figures are 1σ and include analytical errors. 1 cm^3 STP = 2.6868×10^{19} atoms.

multicollection mode with the NanoSIMS in Mainz using a primary O^- beam. Due to the limited amount of material left in the melt residue, we have chosen to measure the Mg-Al isotopes instead of Ca-Ti isotopes since Al has a relatively high abundance in presolar graphite. Previous work has shown that presolar spinel grains have on average $\delta^{25}\text{Mg} \approx 0$‰ (Zinner et al. 2005). We have assumed an average $\delta^{25}\text{Mg} = 0$‰ for the graphite grains here as well. The observed difference in the measured average $^{25}\text{Mg}/^{24}\text{Mg}$ ratios between our graphite grains and the Burma spinel standard of –34‰ to –37‰ amu^{-1} (depending on the measurement session) has been interpreted to be due to instrumental mass fractionation (matrix effects, sample topography, etc.) and Mg isotope ratios were corrected accordingly. Extinct ^{26}Al was inferred from excess ^{26}Mg, which is considered to be entirely radiogenic, using the following expression (Hoppe et al. 1995):

$$\left(\frac{^{26}\text{Al}}{^{27}\text{Al}}\right)_{\text{inferred}} = \frac{\delta^{26}\text{Mg}}{1000} \times \left(\frac{^{26}\text{Mg}}{^{24}\text{Mg}}\right)_{\text{terrestrial}} \times \left(\frac{^{24}\text{Mg}}{^{27}\text{Al}}\right)_{\text{measured}} \times \varepsilon, \quad (1)$$

where δ^{26}Mg is the sample's permill-deviation[6] from the terrestrial values $(^{26}\text{Mg}/^{24}\text{Mg})_{\text{terrestrial}} = 1.3932 \times 10^{-1}$ and the sensitivity factor $\varepsilon = (\text{Al}^+/\text{Mg}^+)/(\text{Al}/\text{Mg}) = 1.17$ was derived by comparing the measured ratio of the standard with its true ratio.

3. RESULTS

3.1. C and O Isotopes

The $^{12}\text{C}/^{13}\text{C}$ ratios range from 4 to 1300 ($^{12}\text{C}/^{13}\text{C}_\odot = 89$). Except for two grains, which differ from solar composition by slightly more than 2σ, the ratios of all other grains are distinct from solar by $>4\sigma$. From the carbon isotopic compositions alone we can conclude that the grains are of extrasolar origin. Ten graphite grains for which noble gases were measured have relatively low $^{12}\text{C}/^{13}\text{C}$ ratios (<16). One of these grains is KFB1g 45-1 with both ^{22}Ne and ^{20}Ne above detection limit (see below). Six of the other grains have ratios $^{12}\text{C}/^{13}\text{C} < 10$, with three of them containing ^{22}Ne above detection limit. The $^{16}\text{O}/^{18}\text{O}$ ratios range from 300 to 550 ($^{16}\text{O}/^{18}\text{O}_\odot = 499$), but are normal within error for most of the samples. Hoppe et al. (1995) and Croat et al. (2008) concluded that the O-isotopic composition reflects dilution with normal O at some time after grain formation. The dilution process probably occurred in the solar nebula, in the parent asteroid, and in the laboratory. The $^{12}\text{C}/^{13}\text{C}$ and $^{16}\text{O}/^{18}\text{O}$ ratios for grains where noble gases were measured are given in Table 3.

3.2. Ne Isotopes

Eleven graphite grains out of the 51 grains analyzed in two sessions contain ^{22}Ne above our detection limits (see Figure 1; Table 3). This fraction (22%), while smaller, is statistically indistinguishable from Nichols et al.'s (1992) fraction of ^{22}Ne-rich grains (30%; 14 out of 49 grains) for the same density fraction. We detected for the first time ^{20}Ne in an individual graphite grain and could determine the $^{20}\text{Ne}/^{22}\text{Ne}$ ratio (KFB1g 34-4: $^{20}\text{Ne}/^{22}\text{Ne} = [3.2 \pm 1.8] \times 10^{-2}$). If not mentioned otherwise, all errors are given as 1σ and include analytical uncertainties based on counting statistics. For grains containing only ^{22}Ne above detection limit, upper limits of the $^{20}\text{Ne}/^{22}$Ne

6 $\delta^A\text{Mg} = \left(\frac{(^A\text{Mg}/^{24}\text{Mg})_{\text{sample}}}{(^A\text{Mg}/^{24}\text{Mg})_{\text{standard}}} - 1\right) \times 10^3$.

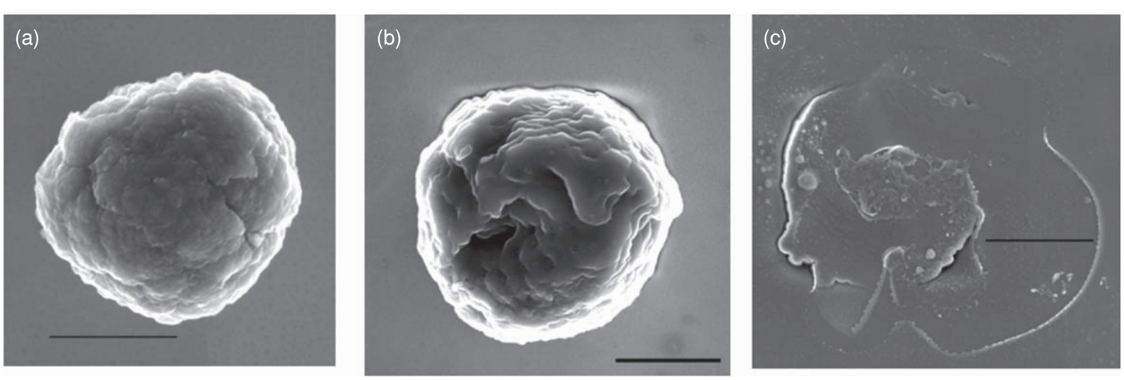

Figure 2. SEM images of three different KFB1 graphite grains showing their typical appearance (a) after physiochemical separation from the meteorite matrix, (b) after surface sputtering due to NanoSIMS analyses, and (c) after IR laser melting for extraction of noble gases. The melt residue has about the same size as the unmelted grain and still allowed detection of significant amounts of Mg-Al isotopes using the NanoSIMS. The black scale bar is 1 μm in length.

Table 3
Gas Amount and Concentrations of ^{22}Ne-rich KFB1g Graphite Grains and Upper Limits of ^{20}Ne/^{22}Ne Ratios of the Other Grains

Grain KFB1g-	Diameter (μm)	^{22}Ne (10^{-14} cm^3 STP)	[^{22}Ne] (10^{-3} cm^3 STP g^{-1})	^{20}Ne/^{22}Ne
07-1[a]	4.37			
10-1[a]	1.94			
12/22-1/1[a]	1.75			
15-2[a]	6.17	0.73 ± 0.28	0.028 ± 0.011	⩽8.6
24-2[a]	1.97			
25-3[a]	2.58			
26-2[a]	3.65			
26-3[a]	2.21	0.56 ± 0.23	0.47 ± 0.20	⩽8.6
27-1[a]	2.75			
27-5[a]	1.78	0.87 ± 0.21	1.39 ± 0.34	⩽7.0
30-1[a]	2.58			
30-2[a]	1.96			
31-1[a]	3.16			
31-2[a]	2.24			
32-1[a]	3.05			
32-2/3[a]	1.89			
34-4[a]	2.17	145.00 ± 0.71	128.0 ± 1.6	0.032 ± 0.018
37-1[a]	3.82	0.39 ± 0.23	0.064 ± 0.037	⩽8.6
40-1[b]	2.65			
41-1[b]	2.30			
42-1[b]	2.03			
43-1[b]	2.41			
43-2[b]	2.36	3.42 ± 0.12	2.340 ± 0.088	⩽0.01
43-3[b]	2.00			
45-1[b]	3.73	2.42 ± 0.12	0.419 ± 0.022	1.7 ± 1.0
45-2[b]	2.99			
46-1[b]	3.76			
51-1[b]	2.53			
51-2[b]	2.20			
53–1[b]	2.28			
53-2[b]	3.31			
53-4[b]	2.24	0.44 ± 0.10	0.354 ± 0.084	⩽0.1
56-1[b]	3.62			
57-1[b]	2.74			
57-3[b]	2.21			
58-1[b]	4.65			
61-1[b]	3.90			
62-1[b]	4.17			
63-1[b]	3.09			
63-4[b]	2.70			
64-1[b]	5.18	48.40 ± 0.25	3.140 ± 0.040	⩽0.0007
64-4[b]	1.68			
64-6[b]	3.18	0.77 ± 0.11	0.215 ± 0.032	⩽0.06
65-1[b]	1.84			
71-1[b]	4.08			
71-2[b]	1.87			
73-1[b]	3.29			
73-2[b]	2.69			
73-3[b]	1.88	0.55 ± 0.14	0.74 ± 0.18	⩽0.1

Notes. Uncertainties are 1σ analytical errors.
[a] Noble gas analysis session 2. [b] Noble gas analysis session 3.

ratio were determined (Table 3) as follows:

$$\left(\frac{^{20}\mathrm{Ne}}{^{22}\mathrm{Ne}}\right)_{\mathrm{upper\,limit}} = \frac{^{20}\mathrm{Ne}_{\mathrm{det.limit}}}{^{22}\mathrm{Ne}_{\mathrm{measured}} - 2\sigma_{\mathrm{error}}} . \quad (2)$$

This gives the highest possible ^{20}Ne/^{22}Ne ratio within 2σ uncertainty based on ion-counting statistics, a conservative estimate if only ^{22}Ne is known. We obtained a particularly low upper ^{20}Ne/^{22}Ne limit for grain 64-1 (^{20}Ne/^{22}Ne $\leqslant 7 \times 10^{-4}$) and low upper limits for grain 64-6 (^{20}Ne/^{22}Ne $\leqslant 6 \times 10^{-2}$) and for grains 43-2, 53-4, 73-3, and 53-4 (^{20}Ne/^{22}Ne $\leqslant 0.1$).

The ^{22}Ne amounts range from 3.9×10^{-15} to 1.5×10^{-12} cm^3 STP (standard temperature and pressure; 1 cm^3 STP = 2.6868 $\times 10^{19}$ atoms). Approximate ^{22}Ne concentrations range from 2.8×10^{-5} to 1.3×10^{-1} cm^3 STP g^{-1}. Concentrations were calculated by assuming spherical grains with a diameter being the average of the measured major and minor axes in SEM images of the grains. The concentration range partially overlaps