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# **Presolar Graphite: Noble Gases and their Origins**

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**Abstract:** Presolar graphite contains a <sup>22</sup>Ne-rich component called Ne-E(L). Noble gas studies on graphite aggregates and single grains have shown that although a dominant source of the <sup>22</sup>Ne is <sup>22</sup>Na, <sup>22</sup>Ne in the He-shell of asymptotic giant branch stars have also contributed to the Ne-E(L). In addition to novae that have been considered to be a possible source of <sup>22</sup>Na, supernovae are a likely source as well. Krypton isotopic ratios of the separates indicate that part of graphite formed in low-mass ( $\leq 3 M_{\odot}$ ) asymptotic giant branch stars of low metallicity ( $Z \leq 0.006$ ).

Keywords: meteorites — presolar grains — Ne-E(L) — isotopic anomalies — nucleosynthesis

#### **1** Introduction

Our solar system formed from a collapsing molecular cloud 4.6 billion years ago. Until the late 1960s, it was believed that solid material in the molecular cloud completely evaporated during this event and thus the solar system was isotopically homogenised. A hint that the solar system was not entirely homogenised came from noble gas analyses of meteorites. Three main components, Ne-A, Ne-B and cosmogenic Ne were identified in meteorites. Ne-A is believed to a component that was present in the solar nebula and trapped in primitive carbonaceous meteorites, whereas Ne-B, or solar Ne, is a component which originated from the sun. If Ne in meteorites is a mixture of these components, in a <sup>20</sup>Ne/<sup>22</sup>Ne-<sup>21</sup>Ne/<sup>22</sup>Ne diagram it should plot inside the triangle bounded by the three components. When Black & Pepin (1969) heated a fragment of the Orgueil meteorite at incremental temperature steps (stepwise heating) and measured Ne released at each step, they found that Ne isotopic ratios in the high  $(>800^{\circ}C)$  temperature releases fell below the triangle, indicating the presence of a new, <sup>22</sup>Ne-rich component.

They named the component Ne-E, as C and D were already taken for minor components. Subsequently Jungck (1982) found that there were two kinds of Ne-E. One kind, Ne-E(H), was released at high temperatures (1100–1500°C) and the carrier phase was concentrated in high-density fractions (2.5–3.1 g cm<sup>-3</sup>). The other kind, Ne-E(L), was released at low temperatures (500–800°C) and the carrier was concentrated in low-density fractions (2.2–2.5 g cm<sup>-3</sup>).

Isotopically anomalous noble gas components were also found in Kr and Xe from meteorites. The quest for the carrier phases of the anomalous components ultimately led to the discovery of stardust, or presolar grains, in meteorites. Presolar grains identified to date include diamond (Lewis et al. 1987), SiC (Bernatowicz et al. 1987; Tang & Anders 1988), graphite (Amari et al. 1990), oxide (Huss et al. 1994; Hutcheon et al. 1994; Nittler et al. 1997),  $Si_3N_4$  (Nittler et al. 1995), and refractory carbide such as TiC inside graphite (Bernatowicz et al. 1991, 1996).

Silicon carbide is the carrier phase of Ne-E(H),  $^{22}$ Ne from the He shell in asymptotic giant branch (AGB) stars, while graphite is the carrier of Ne-E(L). Silicon carbide from different kinds of meteorites has been extensively studied. In contrast, essentially all the data on graphite have been obtained from the study of graphite from the Murchison meteorite. This is because graphite is present in only very primitive meteorites and the extraction of graphite from meteorites is not as straightforward as that of SiC.

In this paper, noble gas results on graphite from four different densities, KE1  $(1.6-2.05 \text{ g cm}^{-3})$  and its daughter fraction KE3  $(1.65-1.72 \text{ g cm}^{-3})$ , KFA1  $(2.05-2.10 \text{ g cm}^{-3})$ , KFB1  $(2.10-2.15 \text{ g cm}^{-3})$  and KFC1  $(2.15-2.20 \text{ g cm}^{-3})$  from the Murchison meteorites will be discussed. One of the most interesting characteristics of graphite is that its isotopic properties depend on density. The purpose of this paper is to focus mainly on the noble gas data available to date on graphite and discuss the origins of presolar graphite and the <sup>22</sup>Ne.

#### 2 Noble Gases in Graphite and their Origin

#### 2.1 Krypton

Amari, Lewis & Anders (1995) analysed Ne, Ar, Kr and Xe in grain aggregates of KE1, KFA1, KFB1 and KFC1 by stepwise heating, the method that is commonly used to differentiate components of different origins in noble gas studies.

Krypton-80 and <sup>86</sup>Kr abundances are strongly indicative of s-process conditions such as neutron density and temperature because of the s-process branches at <sup>79</sup>Se and <sup>85</sup>Kr. In Figure 1 where <sup>86</sup>Kr and <sup>83</sup>Kr abundances are normalised to <sup>82</sup>Kr, two-component mixtures lie on a straight line because both axes have a common denominator. There are two lines: one defined by KFC1 and the other by all



**Figure 1** Krypton isotopic ratios of the graphite fractions KE1, KFA1, KFB1 and KFC1 from the Murchison meteorite (Amari et al. 1995). There are two line trends, one defined by the KFC1 data, the other by the data of the other fractions (see text). The data obtained on size-separated SiC from the Murchison meteorite (Lewis, Amari & Anders, 1994) are also plotted. Kr–N indicates a close-to-normal component derived from the SiC data, while Kenna indicates a typical planetary Kr from the Kenna ureilite.

the other fractions. The lines can be interpreted as mixing between s-process Kr and close-to-normal Kr. The latter is possibly Kr in the envelope of AGB stars, and/or so-called solar or planetary Kr typically found in meteorites. The two lines indicate that two kinds of s-process Kr are present in graphite and that KFC1, the highest density fraction, contains s-process Kr that is totally different from that of the lower density fractions.

The s-process <sup>86</sup>Kr/<sup>82</sup>Kr ratios  $[({}^{86}Kr/{}^{82}Kr)_s]$  can be obtained by extrapolating the lines to the pure s-process <sup>83</sup>Kr/<sup>82</sup>Kr ratio of 0.375 because this ratio is nearly independent of the mean neutron exposure ( $\tau_0$ ). (The <sup>83</sup>Kr/<sup>82</sup>Kr ratio is revised from 0.3 used by Amari et al. 1995.) s-Process <sup>80</sup>Kr/<sup>82</sup>Kr ratios  $[({}^{80}Kr/{}^{82}Kr)_s]$  were obtained in the same manner by extrapolating regression lines to <sup>83</sup>Kr/<sup>82</sup>Kr = 0.375 in a <sup>80</sup>Kr/<sup>82</sup>Kr-<sup>83</sup>Kr/<sup>82</sup>Kr diagram. ( ${}^{86}Kr/{}^{82}Kr)_s$  for KFC1 is 4.43 ± 0.46 and for the other fractions is 0.10 ± 0.25, whereas ( ${}^{80}Kr/{}^{82}Kr)_s$ for KFC1 is 0.030 ± 0.047 and for the other fractions is 0.08 ± 0.042.

The <sup>80</sup>Kr/<sup>82</sup>Kr ratios of both kinds of s-process Kr are indistinguishable within errors. However, the <sup>86</sup>Kr/<sup>82</sup>Kr ratios are quite different. The high (<sup>86</sup>Kr/<sup>82</sup>Kr)<sub>s</sub> for KFC1 can be best explained by low-metallicity AGB stars ( $Z \le 0.006$ ) (Gallino, personal communication). The (<sup>86</sup>Kr/<sup>82</sup>Kr)<sub>s</sub> for the other fractions can be attributed to close-to-solar metallicity AGB stars (Gallino, personal communication; also see Gallino et al. 1990). An interesting possibility is that at least part of the grains in the three lower-density fractions contains s-process Kr from massive stars, because KE3 grains are believed to have formed in supernovae (Travaglio et al. 1999). In massive stars  ${}^{86}$ Kr/ ${}^{82}$ Kr ratios are predicted to be low (0.45), however  ${}^{80}$ Kr/ ${}^{82}$ Kr ratios are expected to be high (1.4–2.3) (Raiteri et al. 1991a, 1991b), which does not agree with the s-process Kr observed in the lower-density fractions. It remains to been seen whether massive stars with different parameters (such as mass and metallicity) can reproduce the s-process Kr isotopic ratios observed in the lower-density fractions.

### 2.2 Neon

A Ne 3-isotope plot of Ne isotopic data obtained by stepwise heating of the graphite fractions is shown in Figure 2. Predicted <sup>20</sup>Ne/<sup>22</sup>Ne ratios in the He-shell of AGB stars with different masses (1.5, 2, and  $3 M_{\odot}$ ) and metallicity (Z = 0.003, 0.006 and 0.02) range from 0.0204 to 0.0746, with an average ratio of 0.055 (Gallino, personal communication). Many <sup>20</sup>Ne/<sup>22</sup>Ne ratios of graphite are lower than the average and a few are even lower than the lowest predicted He-shell ratio. This indicates that another <sup>22</sup>Nerich component enriched in <sup>22</sup>Ne, most likely <sup>22</sup>Ne from the decay of <sup>22</sup>Na, is necessary to explain the graphite data. Amari et al. (1995) assumed that the Ne in the samples was a three-component mixture of Ne from the He-shell, <sup>22</sup>Ne from <sup>22</sup>Na, and close-to-normal Ne from the envelope of AGB stars and/or meteorites, and resolved the Ne into the above three components after correcting for Ne produced by cosmic-rays. (Carbon is too light to produce Ne



**Figure 2** Neon isotopic ratios of the graphite fractions KE1, KFA1, KFB1, and KFC1 from the Murchison meteorite (Amari et al. 1995). The open circles in the figures are isotopic ratios of the total Ne. Predicted Ne ratios in the He-shell of AGB stars are also plotted (Gallino, personal communication). The lines indicate mixing between pure  $^{22}$ Ne (presumably from the decay of  $^{22}$ Na) and solar wind Ne.

by spallation, although retaining Ne recoiled from adjacent silicate grains in meteorites is a possibility.) Neon in the samples is predominantly <sup>22</sup>Ne from <sup>22</sup>Na (80 to 95% in KE1, KFA1 and KFC1, 50 to 75% in KFB1) as first suggested by Clayton (1975).

## 2.3 Origin of <sup>22</sup>Na

Since the discovery of Ne-E, novae have been a favourite stellar source of <sup>22</sup>Na. Hydrodynamic model calculations of nova outbursts have shown that ONe novae produce a significant amount of <sup>22</sup>Na (José & Hernanz 1998; Starrfield et al. 1998).

Nichols et al. (2003) measured He and Ne in single graphite grains from KE3, KFB1 and KFC1 that had been analysed for their isotopic ratios of elements such as C, N and Si prior to noble gas analysis. They found that only a portion of the grains (7 to 43% of the analysed grains) have  $^{22}$ Ne above the detection limit of the noble gas mass spectrometer. Of special interest are the two  $^{22}$ Nerich grains, KE3-573 and KFB1-161, for which the upper limit of  $^{20}$ Ne/ $^{22}$ Ne is determined to be 0.01. It is lower than  $^{20}$ Ne/ $^{22}$ Ne ratios predicted from the He-shell, indicating that the two grains contain  $^{22}$ Ne from the decay of

<sup>22</sup>Na. The <sup>12</sup>C/<sup>13</sup>C ratio of KE3-573 is  $27 \pm 1$  and that of KFB1-161 is  $3.8 \pm 0.1$ . Their <sup>14</sup>N/<sup>15</sup>N ratios are solar within errors. There is evidence that N in graphite grains is at least partially equilibrated with solar N. Thus, N isotopic ratios of the grains are not diagnostic for distinguishing stellar sources.

The low  ${}^{12}C/{}^{13}C$  ratio of KFB1-161 is indicative of a nova origin (Nichols et al. 2003). Nova ejecta are characterised by low  ${}^{12}C/{}^{13}C$  (0.3–1.8), a large range of  ${}^{14}N/{}^{15}N$  (3–130 for CO novae and 0.3–4 for ONe novae) and high  ${}^{26}Al/{}^{27}Al$  (0.01–0.6) (José & Hernanz, 1998; Starrfield, Gehrz & Truran, 1997; Starrfield et al. 1998; José et al. 2003). In fact, a few presolar grains of a putative nova origin show low  ${}^{12}C/{}^{13}C$  ratios (4–9) (Amari et al. 2001). The  ${}^{22}Na/C$  ratio of KFB1-161 is estimated to be 9 × 10<sup>-6</sup>. Since Na is expected to be highly depleted relative to C due to its volatility in grain condensation, Nichols et al. (2003) have argued that an ONe nova is a possible source of  ${}^{22}Na$  and the grain.

The  ${}^{12}C/{}^{13}C$  ratio of KE3-573 is too high to be accounted for by a nova. This grain has most likely formed in a supernova because its  ${}^{29}Si/{}^{28}Si$  and  ${}^{30}Si/{}^{28}Si$  ratios are 0.765 × solar and 0.673 × solar, respectively.

<sup>28</sup>Si excesses are characteristic of presolar grains of a supernova origin (Nittler et al. 1995; Travaglio et al. 1999; Hoppe et al. 2000) and many low-density graphite grains in the fraction KE3 are believed to have formed in supernovae (Travaglio et al. 1999). <sup>22</sup>Na is produced in the O/Ne zone where carbon burning takes place (Woosley & Weaver, 1995) and where C is thus depleted. It is commonly assumed that a C-rich environment (C > O) is required to condense carbonaceous grains. However, Clayton, Deneault & Meyer (2001) proposed that graphite grains could form in O-rich zones in supernovae because CO molecules that normally lock up C were dissociated by the radiation from radioactive isotopes. It is of interest to further investigate whether supernovae can be one of the sources of Ne-E(L) and its carriers.

#### **3** Conclusions and Future Work

Of the two kinds of Ne-E found in primitive meteorites, Ne-E(H) is carried by SiC that contains <sup>22</sup>Ne from the He-shell of AGB stars. The other kind, Ne-E(L) is carried by graphite. While its <sup>22</sup>Ne is predominantly due to <sup>22</sup>Na (50 to 95% of the <sup>22</sup>Ne), a portion of it is <sup>22</sup>Ne from the He-shell of AGB stars. The inferred s-process <sup>86</sup>Kr/<sup>82</sup>Kr ratio (4.43 ± 0.46) of KFC1 (2.15–2.2 g cm<sup>-3</sup>), which is markedly different from that of the other density fractions (0.10 ± 0.25), indicates that KFC1 graphite formed in lowmetallicity ( $Z \le 0.006$ ) AGB stars. There are two possible sources of <sup>22</sup>Na, novae and supernovae.

To better understand the origin of graphite and its anomalous noble gases, coordinated studies of graphite by various techniques are necessary.

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