Presolar Graphite from the Murchison Meteorite: Imprint of Nucleosynthesis and Grain Formation

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Abstract. Presolar graphite is the carrier of Ne-E(L) and most ^{22}Ne in Ne-E(L) had long been attributed to radiogenic decay of ^{22}Na from novae. Of presolar graphite grains with a range of density (1.6-2.2g/cm^3), low-density graphite grains extracted from the Murchison meteorite are characterized by ^{18}O excesses and Si isotopic anomalies and are believed to have formed in supernova ejecta. From noble gas analyses of low-density graphite grains, we conclude that ^{22}Ne in the grains is from the *in situ* decay of ^{22}Na ($T_{1/2}$=2.6a) produced in the C-burning zone in presupernova stars. The grains also contain Kr that was produced by neutron capture, either in the He-burning zone or the C-burning zone during hydrostatic burning. The ^{22}Ne of a ^{22}Na origin indicates that the grains formed shortly after the explosion. The presence of ^{22}Ne of a ^{22}Na origin and Kr, and the absence of ^{22}Ne of a non-radiogenic origin might give us a further clue for graphite formation in supernova ejecta.

Keywords: supernovae; nucleosynthesis; slow neutron capture process; dust; meteorite; Ne-E(L)
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INTRODUCTION

Noble gases in meteorites have played an important role in cosmochemistry. They are literally rare, thus a small addition of isotopically anomalous noble gas components could be easily detected. The anomalous components include Ne-E (enriched in ^{22}Ne), Kr-S (*s*-process Kr), Xe-S (*s*-process Xe) and Xe-HL (enriched in both light and heavy Xe isotopes) [see 1]. Ne-E was first discovered when Black and Pepin [2] analyzed the Ne isotopic composition in a fragment of the Orgueil meteorite by stepwise heating. Stepwise heating is a method commonly applied for noble gas studies. Temperature is incrementally increased and the released noble gases are analyzed in each step. The concept behind this technique is that different noble gas components are trapped in different sites in the minerals (e.g., surface or interior) or different minerals and thermal properties of the minerals/trapping sites are different, thus different noble gas components are released at different temperatures. The Ne components commonly observed in meteorites are called Ne-A, Ne-B and Ne-S [e.g., see Fig. 13.1.1 of reference 1]. If the Ne in the Orgueil meteorite was a mixture of these components, Ne isotopic ratios of all the temperature steps should have fallen in the triangle bounded by the components. However, in high (900 – 1000°C) temperature steps, the ratios fell below the triangle, indicating the presence of a ^{22}Ne-rich component. Black [3] named the component Ne-E. Subsequently, Jungck [4] separated the Orgueil meteorite into several fractions and found that there are two kinds of Ne-E. One was released at *l*ow temperatures (500 – 700°C) and was concentrated in *l*ow-density fractions (2.2 – 2.5 g/cm^3), whereas the other was released at *h*igh temperatures (1200 – 1400°C) and was enriched in a *h*igh-density fraction (2.5 – 3.1 g/cm^3). They were dubbed Ne-E(L) and Ne-E(H), respectively.

The identification of carriers of the anomalous noble gas components came almost two decades after the discovery of the components in meteorites. Diamond was identified as the carrier of Xe-HL in 1987 [5]. Subsequently, silicon carbide (SiC), the carrier of Ne-E(H), Kr-S and Xe-S, was isolated and found [6, 7]. The ^{22}Ne of Ne-E(H) is from the He-shell in asymptotic giant branch (AGB) stars [8-10]. Finally, graphite was identified as the carrier of Ne-E(L) in 1990 [11]. Its ^{22}Ne is predominantly from ^{22}Na with an addition of ^{22}Ne from He-shell in AGB stars [12].

The carrier grains exhibit huge isotopic anomalies not only in noble gases but also other elements, indicating that they formed in the ejecta of supernovae or in the circumstellar envelopes of asymptotic giant branch (AGB) stars. They are called presolar grains because they formed before the solar system. Extensive studies of presolar grains have provided new information on nucleosynthesis in stars, mixing in supernova ejecta, Galactic chemical evolution and grain formation in the outflow of stars [e., g., 13, 14, 15]. In this paper, we will reexamine noble gas data on presolar graphite grains from the Murchison meteorite and discuss the implications of the data on nucleosynthesis and grain formation.

DATA SOURCES

Most studies on presolar graphite have been performed on four graphite-rich separates extracted from the Murchison meteorite [16]. They are KE1 (1.6–2.05 g/cm^3), KFA1 (2.05–2.10 g/cm^3), KFB1 (2.10–2.15 g/cm^3) and KFC1 (2.15–2.20 g/cm^3). The separate KE1 was further purified, yielding KE3 (1.65–1.72 g/cm^3) [17]. Concerning isotopic signatures, KE1 and KE3 can be regarded as the same separate.

There exist two sets of noble gas analyses of the separates. Amari et al. [12] analyzed Ne, Ar, Kr and Xe in bulk (=aggregates of millions of grains) samples by stepwise heating. Nichols et al. [18] measured ^{4}He and ^{20,22}Ne in single grains of known C and Si isotopic ratios from KE3, KFB1 and KFC1.

In this paper, we will focus on the lowest-density separates KE1 and KE3.

DISCUSSION

Neon

Low-density Graphite

Low-density graphite grains from the separate KE3 have been most extensively studied with ion probe: their large grain size (median grain size: 4.9 μm) and high trace element concentrations made it possible to analyze isotopic ratios of multiple elements [17, 19, 20]. Many grains (~50 %) have ^{18}O excesses and Si isotopic anomalies, mainly in the form of ^{28}Si excess. Highest ^{26}Al/^{27}Al ratios inferred from ^{26}Mg excesses are ~0.1. ^{28}Si excesses and high ^{26}Al/^{27}Al ratios are also observed in SiC grains of type X (X grains), which are believed to have formed in Type II supernovae [21, 22]. Proof for their supernova origin came from the presence of ^{44}Ti ($T_{1/2}$ = 60a) in a few low-density graphite and SiC X grains [23, 24] because ^{44}Ti is produced only by explosive nucleosynthesis.

In order to quantitatively examine whether the graphite grain data could be explained with supernova models, Travaglio et al. [20] used yields of Type II supernova models by Woosley and Weaver [25] and mixed different zones. They found that general isotopic features of the low-density graphite grains are explained if a small amount of material from the inner Si-rich zone is ejected and mixed at a microscopic scale before grain formation into the outer He-rich zones.

^{22}Ne-rich low-density grains

Of 21 KE3 grains analyzed for He and Ne, nine grains contained measurable amount of ^{22}Ne. ^{4}He and ^{20}Ne were under the detection limit in all grains. One ^{22}Ne-rich grain was not analyzed for its C, O, and Si isotopic ratios. Seven ^{22}Ne-rich grains show elevated ^{18}O/^{16}O ratios (6.83×10^{-3} to 0.119) above the solar ratio (2×10^{-3}). Grain KE3a-573 has the normal ^{18}O/^{16}O ratio, but ^{28}Si excess {δ^{29}Si/^{28}Si = –235±97 ‰, δ^{30}Si/^{28}Si = –327±128 ‰, where δ^{i}Si/^{28}Si ≡ $[(^{i}\mathrm{Si}/^{28}\mathrm{Si})_{\mathrm{grain}}/(^{i}\mathrm{Si}/^{28}\mathrm{Si})_{\mathrm{solar}} - 1] \times 1000$.} This grain was so enriched in ^{22}Ne that an upper limit of its ^{20}Ne/^{22}Ne (<0.01) could be determined. Two grains with ^{18}O excesses show the presence of ^{44}Ti [^{44}Ti/^{48}Ti = $(1.06\pm0.22)\times10^{-3}$ and $(2.40\pm1.10)\times10^{-3}$]. Thus all ^{22}Ne-rich grains bear supernova signatures such as ^{18}O and ^{28}Si excesses, and the presence of ^{44}Ti, indicating that they formed in supernovae.

In Type II supernovae, the lowest ^{20}Ne/^{22}Ne ratio is found in the partial He burning zone (or He/C zone, according to the terminology by Meyer et al. [26], whose name indicates most abundant elements) where ^{14}N is converted into ^{22}Ne. The predicted ^{20}Ne/^{22}Ne ratio in the zone in a 25$M_{\odot}$ star of solar metallicity is 0.088 by Chieffi and Limongi [27] and 0.096 by Heger et al. [28]. They are much higher than the upper limit observed in grain KE3a-573. This implies that the ^{22}Ne in the grain was not implanted because if it was the case ^{20}Ne should have been also implanted, resulting in a much higher upper limit than 0.01. This leaves ^{22}Na as a sole source of the ^{22}Ne in the grain.

Although upper limits of ^{20}Ne/^{22}Ne ratios of the other grains are not available, the portion of ^{22}Ne from ^{22}Na can be determined from the Ne isotopic ratios of the separate KE1 (^{20}Ne/^{22}Ne = 0.0301±0.0018, ^{21}Ne/^{22}Ne = 0.000118±0.000017). Assuming that the Ne in KE1 is a mixture of ^{22}Ne from ^{22}Na, Ne from the He/C zone, and solar Ne, more than 99% of the ^{22}Ne in KE1 originated from ^{22}Na. Thus, the ^{22}Ne-rich grains most likely contain ^{22}Ne solely from the decay of ^{22}Na, as first suggested by Nichols et al. [18]. ^{22}Na is produced in the C convective shell (O/Ne zone) during hydrostatic C burning by ^{21}Ne(p,γ)^{22}Na, where ^{21}Ne is produced by ^{20}Ne(n,γ)^{21}Ne and protons are produced by ^{12}C(^{12}C,p)^{23}Na [27].

Krypton

^{86}Kr/^{82}Kr and ^{80}Kr/^{82}Kr ratios are very sensitive indicators for nucleosynthetic conditions in stars. Neutron capture on ^{84}Kr feeds both the ground state and the isomeric state of ^{85}Kr [29]. The unstable isotope ^{85}Kr decays to ^{85}Rb with the half-life of 11 years when it is at the ground state. At the isomeric state, its decay rate is much faster ($T_{1/2}$ = 4.48h). During convective core He burning, the ground and the isomeric states are not thermalised and need to be treated independently. During shell C burning, there is full thermalization between the ground state and the isomeric state. In any case, ^{86}Kr yields depend on neutron density. Selenium-79 is at a branching point of the *s*-process and its behavior is critical to ^{80}Kr yields. The half-life of ^{79}Se strongly depends on temperature: it is much shorter at stellar conditions (one month at ~1×10^{9}K) than in terrestrial conditions (650,000 years) [30]. As a consequence, ^{80}Kr yields depend on neutron density and temperature.

TABLE 1. Krypton isotopic ratios.

Separate/Zone	80/82	83/82	84/82	86/82
KE1+KFA1	0.070±0.045	≡0.334	1.26±0.65	0.02±0.26
KE1+KFA1	0.127±0.023	≡0.623	2.86±0.33	0.67±0.14
KFC1	0.030±0.047	≡0.375	2.58±0.41	4.43±0.46
O/C*	0.405	0.334	1.41	0.0938
O/Ne*	0.0656	0.623	1.97	0.726

* A $25M_{\odot}$ model by Chieffi and Limongi [27]

Amari et al. [12] have found that the four Murchison separates are enriched *s*-process Kr (Kr-S) and that in a ^{86}Kr/^{82}Kr–^{83}Kr/^{82}Kr plot (Fig. 1), KE1+KFA1 and KFC1 form two distinct lines, indicating that the Kr in the separates is a mixture of close-to-normal Kr (on the right in Fig. 1) and Kr-S (on the left) and that there are two Kr-S components: Kr-SH in KFC1 and Kr-SL in KE1+KFA1. To infer the isotopic composition of Kr-SH, we assumed (^{83}Kr/^{82}Kr)s ≡ 0.375 instead of 0.30 used by Amari et al. [12], reflecting the improvement of precisions of analyses in neutron capture cross sections (Table 1). Kr-SH, with a high ^{86}Kr/^{82}Kr ratio, most likely originated from low-metallicity AGB stars as concluded by Amari et al. [12]. Kr-SL was originally associated with high-metallicity AGB stars or a mixture of AGB and massive stars, thus its isotopic ratios were inferred by assuming ^{83}Kr/^{82}Kr = 0.30. The ^{83}Kr/^{82}Kr ratio was used to infer other Kr isotopic ratios because it is predicted to be constant and defined by the inverse ratio of their neutron capture cross sections at relatively low temperature (kT ~ 30 keV). However, at the higher temperature [~10^9K (~90 keV)] realized during convective shell C-burning in the O/Ne zone, deviations from the classical 1/v rule (v: thermal velocity) for the cross section of ^{83}Kr become significant. Thus, it is necessary to reevaluate the ^{83}Kr/^{82}Kr ratio to apply for low-density graphite grains, which formed in supernovae. Since the lowest ^{86}Kr/^{82}Kr and ^{83}Kr/^{82}Kr ratios observed in the grains are 1 and 0.7, respectively (Fig. 1), Kr-SL must have smaller ratios than those.

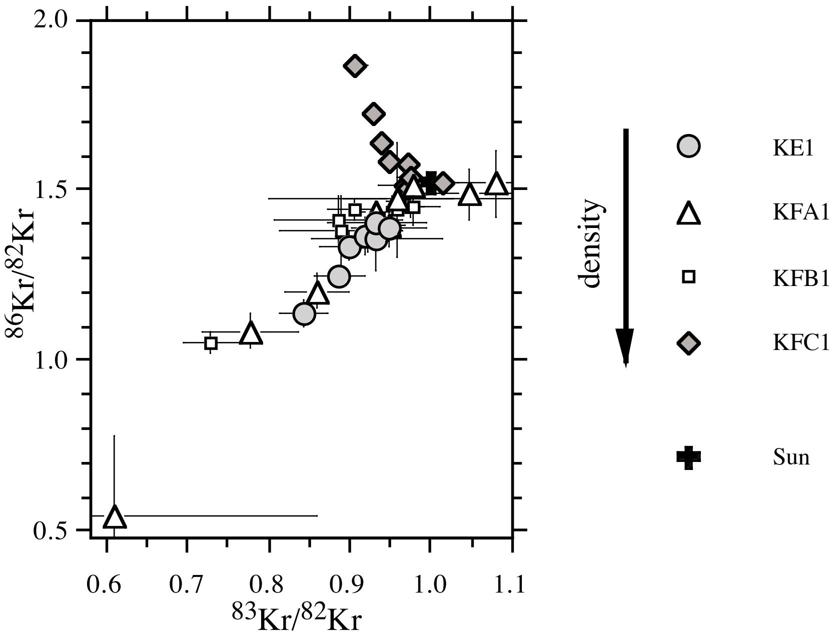

FIGURE 1. Krypton isotopic ratios of the graphite separates from the Murchison meteorite. KFC1 and KE1+KFA1 form two distinct lines. Errors are 1 σ. Data from [12].

Chieffi and Limongi [27] constructed a set of explosive yields of massive stars of solar metallicity in the mass range of 11–120M$_{\odot}$, using the latest version of the FRANEC code (5.05218) that includes a nuclear network extending to ^{98}Mo. Comparing the grain data and their predicted ratios, the 15M$_{\odot}$ model can be excluded from a source of the Kr in the low-density grains (KE1+KFA1) because the ^{86}Kr/^{82}Kr ratio in the O/Ne zone (C burning zone) is 5.05 and the ^{83}Kr/^{82}Kr ratio of the bottom of the O/C zone (the relic of the He convective core) is 2.504, which are much higher than 1 and 0.7, respectively. In their 25M$_{\odot}$ and 35M$_{\odot}$ models, the ^{83}Kr/^{82}Kr ratio is ~0.3 (0.334 in 25M$_{\odot}$ and 0.310 for 35M$_{\odot}$) in the O/C zone (6.2–6.6M$_{\odot}$ in Fig. 2), and ~0.6 (0.623 in 25M$_{\odot}$ and 0.597 in 35M$_{\odot}$) in the O/Ne zone (2.6–6.2M$_{\odot}$ in Fig. 2). The average neutron density in the O/C zone is ≤ 10^6 n/cm^3 [31], whereas that in the O/Ne zone reaches 10^{12} n/cm^3 [32], which by far exceeds the range of a classical notion of the *s*-process. The KE1+KFA1 data were extrapolated to ^{83}Kr/^{82}Kr = 0.334 and 0.623

to examine the zone where the Kr in the grains originated (Table 1). When extrapolated to 0.334 (hence assuming the Kr was produced in the O/C zone), both ^{80}Kr/^{82}Kr and ^{86}Kr/^{82}Kr ratios are close to zero, whereas the model predicts a much higher ^{80}Kr/^{82}Kr ratio. When extrapolated to 0.623, the ^{80}Kr/^{82}Kr ratio from the grains is still higher than the ratio from the model.

It is difficult to further narrow down which zone is responsible for the Kr in the grains because of huge uncertainties in ^{86}Kr and ^{80}Kr yields. The neutron capture cross section of ^{85}Kr is theoretically estimated with ~80 % uncertainty [29]. The cross section of ^{79}Se is theoretically determined with a huge uncertainty up to 50 %. Moreover, as the half-life of ^{79}Se strongly depends on temperature, a slight difference in temperature can result in a big difference in ^{80}Kr yields.

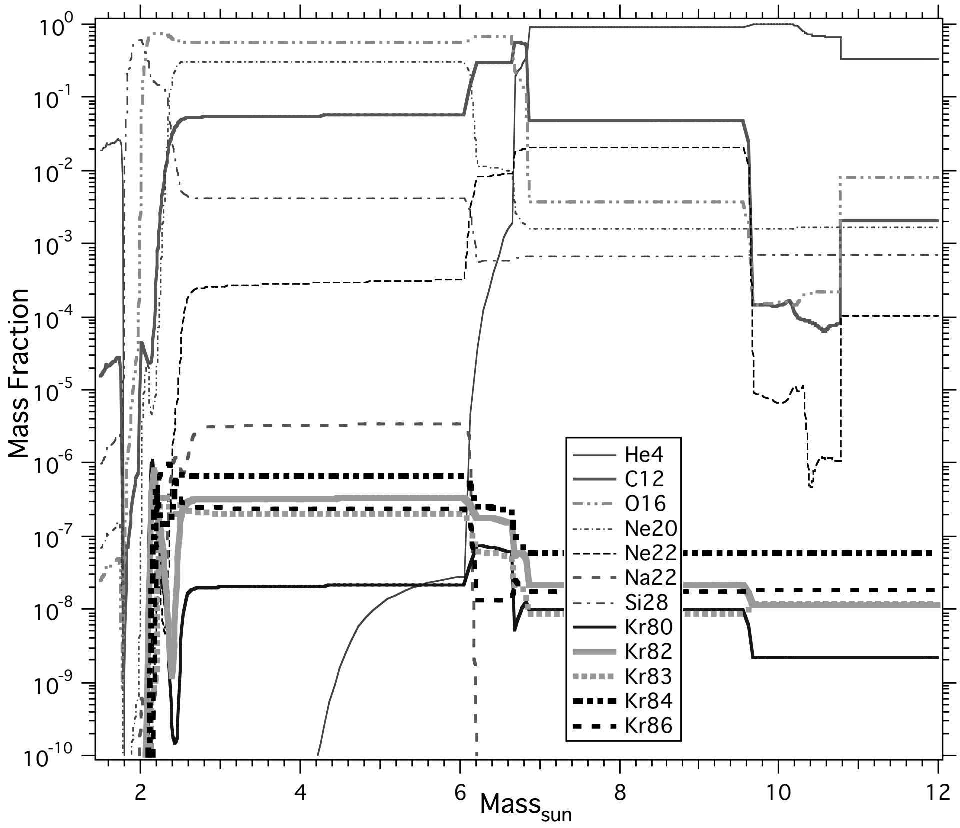

FIGURE 2. Elemental yields for a $25M_{\odot}$ model with the solar metallicity 6.8×10^{6} seconds after the explosion [27].

Implications for Grain Formation

The noble gas data of low-density grains reflect conditions of grain formation. The presence of the ^{22}Ne of a ^{22}Na origin indicates that the grains formed within a few years after the explosion before ^{22}Na completely decayed. This does not contradict to observations of supernovae: in SN 1987A formation of dust grains has been shown to occur in the ejecta about 400 days after the explosion, as implied by the sudden decrease in the visible light accompanied by a huge infrared counterpart [33].

Furthermore, the presence of the s-process Kr and absence of ^{22}Ne of a non-radiogenic origin in the grains could provide a further clue. Neon and Kr are inert and do not form compounds with other elements, thus implantation was the only way that the grains acquired these gases. In the O/C and O/Ne zones where the Kr in the grains was synthesized, ^{20}Ne yields are more than four orders of magnitude higher than ^{84}Kr yields [27].

When supernova ejecta expand, it is generally assumed that elements in the same zones have the same velocity. At the same velocity, Kr is implanted into a much deeper region than Na and Ne. For example, if their velocity relative to the grains is 3100 km/s, the penetration depth of Ne into carbonaceous grains is 0.65 μm and that of Kr is 3.4 μm, respectively. If Ne was lost by diffusion loss, Ne of a ^{22}Na origin should have been equally lost: penetration depths of Na and Ne are almost identical.

Grain formation in supernova ejecta is a complex process and we need to take various things into account to explain the formation conditions. When supernova ejecta hit the interstellar medium or a circumstellar shell that had been expelled from a progenitor star, the ejecta were heated by the reverse shock and elements in the same zones obtained the same energies thus different velocities. It is observed that there are zones with a different degree of ionization [34]. A work to disentangle the clues on grain formation is in progress.

CONCLUSIONS

Low-density graphite grains, characterized by ^{18}O excesses and Si isotopic anomalies (mainly ^{28}Si excesses), formed in supernova ejecta. They contain ^{22}Ne from the decay of ^{22}Na and s-process Kr. The former was produced in the O/Ne zone (C convective shell) during C burning, while the latter was produced either in the O/Ne zone (C convective shell) or the O/C zone during core He burning. The presence of the initial ^{22}Na and Kr, and the absence of non-radiogenic ^{22}Ne may provide a further clue on graphite grain formation in supernova ejecta.

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