

INTERSTELLAR SiC WITH UNUSUAL ISOTOPIC COMPOSITIONS:
GRAINS FROM A SUPERNOVA?SACHIKO AMARI,^{1,2} PETER HOPPE,^{1,3} ERNST ZINNER,¹ AND ROY S. LEWIS²*Received 1992 April 6; accepted 1992 May 13*

ABSTRACT

Five single SiC grains, 2–9 μm in size, from the Murchison carbonaceous meteorite have been analyzed by ion microprobe mass spectrometry for their C, N, Si, Mg-Al, Ca, and Ti isotopic compositions. While most interstellar SiC grains from primitive meteorites are characterized by ^{13}C , ^{14}N , ^{29}Si , ^{30}Si , ^{46}Ti , ^{47}Ti , ^{49}Ti , and ^{50}Ti excesses relative to solar isotopic ratios, and $^{26}\text{Al}/^{27}\text{Al}$ ratios of up to $\sim 10^{-2}$, carrying the signatures of H- and He-burning, these five grains (\equiv grains *X*) have large excesses in ^{12}C (up to 28 times solar) and ^{15}N (up to 22 times solar), depletion in ^{29}Si and ^{30}Si (up to 59%), and $^{26}\text{Al}/^{27}\text{Al}$ ratios between 0.1 and 0.6. They furthermore have ^{49}Ti excesses (up to 95%), and one grain has a large ^{44}Ca excess (300%). While the Ca and Ti anomalies point toward explosive nucleosynthesis in supernovae and the in situ decay of the radioactive precursors ^{44}Ti and ^{49}V in SiC grains formed in supernova ejecta, there is no simple formation scenario that can give a consistent explanation for the isotopic compositions of these grains.

Subject headings: dust, extinction — ISM: abundances — nuclear reactions, nucleosynthesis, abundances — stars: carbon — supernovae: general

1. INTRODUCTION

Interstellar silicon carbide isolated from the Murchison CM2 chondrite is highly anomalous in its C, Si, N, Mg, Ti, Sr, Ba, Nd, and noble gas isotopic compositions (Zinner, Tang, & Anders 1989; Ireland, Zinner, & Amari 1991a; Ott & Bege-mann 1990a, b; Prombo et al. 1992; Zinner, Amari, & Lewis 1991b; Zinner et al. 1991a; Lewis, Amari, & Anders 1990; Stone et al. 1991). Most SiC grains are characterized by heavy C, light N, excesses in ^{29}Si and ^{30}Si , and $^{26}\text{Al}/^{27}\text{Al}$ ratios of up to $\sim 10^{-2}$, carrying the signature of H- and He-burning. However, we found one SiC grain (grain 4 of Zinner et al. 1991a) whose C, N, and Si isotopic compositions are completely different from all the other grains. After this discovery we systematically searched for other such grains in the Murchison SiC separates KJG and KJH, having mass-weighted average grain sizes of 3.02 and 4.57 μm (Amari, Lewis, & Anders 1992). Ion microprobe isotopic analysis of 180 KJG grains revealed one additional grain of similar isotopic properties, and of 506 KJH grains, three more. These five grains, which exhibit extremely exotic isotopic compositions, distinct from the majority of the SiC grains, were named grains *X* (Zinner et al. 1991c). We report ion microprobe isotopic measurements of C, N, Mg, Ca, and Ti in the five grains *X*.

2. RESULTS

The techniques for SIMS isotopic measurements on single SiC grains have been described previously (Zinner et al. 1989; Ireland et al. 1991a). Before ion microprobe analysis the grains were examined for their morphology and major element chemistry in the scanning electron microscope (SEM).

Table 1 lists the sizes of grains *X* as determined in the SEM and their elemental and isotopic compositions determined by ion microprobe analysis (because of small sizes and/or low elemental concentrations we could not obtain all isotopic

ratios). Figures 1–3 show their isotopic compositions in comparison to the other KJG and KJH grains. Most SiC grains have ^{13}C and ^{14}N excesses (Fig. 1) as well as excesses in ^{29}Si and ^{30}Si (Fig. 2). Many also show large excesses in ^{26}Mg attributable to the decay of ^{26}Al (Zinner et al. 1991a), with $(^{26}\text{Al}/^{27}\text{Al})_0$ ratios ranging from 4×10^{-5} to 2×10^{-2} (Fig. 3). SiC grains furthermore contain Ti—in one case shown to be in the form of TiC (Bernatowicz, Amari, & Lewis 1992)—with V-shaped isotopic abundance patterns, that is, excesses of all Ti isotopes relative to ^{48}Ti (Ireland et al. 1991a, b) (see Fig. 4). These isotopic features have been interpreted as the nucleosynthetic signatures of H-burning (producing the observed C and N isotopic composition and ^{26}Al) and of neutron capture during He-burning (producing the Si and Ti isotopic compositions) (Ireland et al. 1991a; Stone et al. 1991; Virag et al. 1992). Possible astrophysical sources for these grains are carbon stars on the asymptotic giant branch (AGB) (Gallino et al. 1990) or massive stars during their Wolf-Rayet stage (Dearborn & Blake 1985; Prantzos & Cassé 1986; Busso & Gallino 1985; Prantzos, Arnould, & Arcoragi 1987; Prantzos, Hashimoto, & Nomoto 1990). A detailed discussion of these data will be given elsewhere; here we wish to concentrate on the extremely exotic grains *X*.

In their morphology the grains *X* are indistinguishable from the other grains: they have a platy appearance with euhedral, often hexagonal, features similar to grains from Murchison and Orgueil described previously (Stone et al. 1991; Virag et al. 1992).

In contrast, they differ completely in their isotopic properties, having ^{12}C and ^{15}N excesses relative to solar (Fig. 1), large depletions in ^{29}Si and ^{30}Si (Fig. 2), and $(^{26}\text{Al}/^{27}\text{Al})_0$ ratios between 0.1 and 0.6 (Fig. 3), substantially higher than those found in any other grains. Four grains have large excesses in ^{49}Ti (Table 1 and Fig. 4); in grain *X1* the error on $\delta^{49}\text{Ti}$ is large (Table 1) because hardly any material was left for analysis, and in grain *X5* the Ti concentration was too low for isotopic analysis. Grain *X4* also has an excess of ^{50}Ti . This could be an isobaric interference from an excess in ^{50}V or ^{50}Cr , but in this case the $^{50}\text{C}/^{50}\text{V}$ ratio would have to be 119 times solar or the

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TABLE 1
ELEMENTAL AND ISOTOPIC COMPOSITIONS OF SiC GRAINS X

Grain	X1	X2	X3	X4	X5
Size (μm)	3×4.5	4.5×5	2×2.5	9×9	3.5×5
Al (%)	2.24	4.88	6.35	0.32	0.07
Ca (ppm)	380	576	68	26	<22
Ti (ppm)	200	360	168	67	<20
V (ppm)	140	35	62	9.6	<2.4
Cr (ppm)	nd ^a	66	nd	6.5	<3.8
$^{12}\text{C}/^{13}\text{C}$	1135 ± 58	304 ± 3	191 ± 7	2525 ± 143	168 ± 4
$^{14}\text{N}/^{15}\text{N}$	181 ± 2	106 ± 1	71 ± 3	12.6 ± 0.1	159 ± 8
$\delta^{29}\text{Si}/^{28}\text{Si}$ (‰)	-376 ± 4	-459 ± 2	-318 ± 2	-363 ± 2	-249 ± 4
$\delta^{30}\text{Si}/^{28}\text{Si}$ (‰)	-585 ± 4	-454 ± 3	-517 ± 10	-529 ± 2	-388 ± 8
$^{26}\text{Al}/^{27}\text{Al}$	0.20 ± 0.01	0.23 ± 0.03	0.10 ± 0.01	0.61 ± 0.04	0.23 ± 0.01
$\delta^{42}\text{Ca}/^{40}\text{Ca}$ (‰)	57 ± 208	-167 ± 80	nd	14 ± 200	nd
$\delta^{43}\text{Ca}/^{40}\text{Ca}$ (‰)	276 ± 386	390 ± 170	nd	79 ± 356	nd
$\delta^{44}\text{Ca}/^{40}\text{Ca}$ (‰)	48 ± 118	3040 ± 237	nd	-55 ± 78	174 ± 362
$\delta^{46}\text{Ti}/^{48}\text{Ti}$ (‰)	nd	82 ± 48^b	nd	33 ± 66	nd
$\delta^{47}\text{Ti}/^{48}\text{Ti}$ (‰)	nd	23 ± 50^b	nd	-79 ± 62	nd
$\delta^{49}\text{Ti}/^{48}\text{Ti}$ (‰)	600 ± 300	239 ± 58^b	473 ± 104	949 ± 88	nd
$\delta^{50}\text{Ti}/^{48}\text{Ti}$ (‰)	nd	-52 ± 38^b	nd	306 ± 59	nd

NOTES.—All errors are 1σ . δ -values denote the deviation of an isotopic ratio from the terrestrial ratio in permil, e.g., $\delta^{29}\text{Si}/^{28}\text{Si} = 1000 \times [({}^{29}\text{Si}/^{28}\text{Si})_{\text{sample}}/({}^{29}\text{Si}/^{28}\text{Si})_{\text{terr}} - 1]$.

^a Here "nd" equals not determined.

^b Combined measurements at Washington University and by Ireland et al. 1991b at the Australian National University.

$^{50}\text{Cr}/^{52}\text{Cr}$ ratio 11 times solar. The last possibility is unlikely since the $^{53}\text{Cr}/^{52}\text{Cr}$ ratio is normal. Grain X2 exhibits a large excess of ^{44}Ca and a much smaller excess in ^{43}Ca , but the other grains in which Ca isotopes were measured have normal Ca, notably grains X1 and X4 for which the errors on the $^{44}\text{Ca}/^{40}\text{Ca}$ ratio are much smaller than in the other two grains.

3. DISCUSSION

The case for a circumstellar origin of SiC grains from primitive meteorites has been made before, including a discussion of possible stellar sources (Zinner et al. 1989; Stone et al. 1991; Virag et al. 1992). The problem is that none of the stellar sites whose chemical environment enable the condensation of SiC can provide a simple explanation for the isotopic compositions of the grains X. Carbon-rich red giants, massive mass-losing

stars during the WC stage, novae, and C-rich shells from supernovae all satisfy the condition $C > O$ for SiC condensation (Virag et al. 1992).

3.1. AGB Stars

Both AGB stars and novae can account for the high $^{26}\text{Al}/^{27}\text{Al}$ ratios observed in grains X. Nucleosynthetic processes in AGB stars take place in a H-burning shell and underlying He-burning shell (see, e.g., Iben 1991). Cameron (1992) proposed an AGB star origin for grain X2 by invoking high-temperature H-burning in a hot-bottomed convective envelope in the late stage of such a star for increased ^{26}Al production.

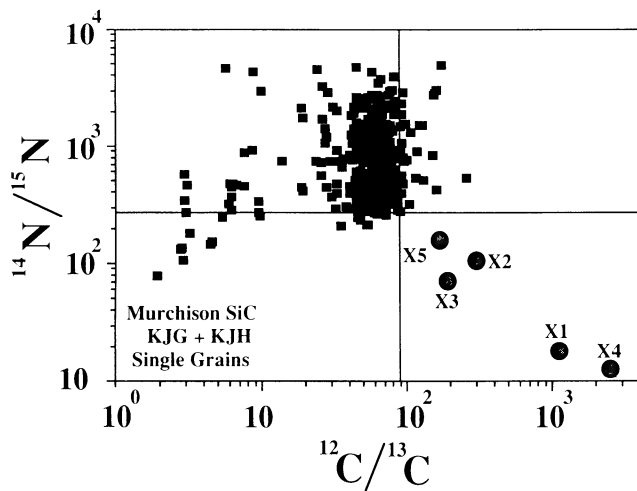


FIG. 1.—Carbon and nitrogen isotopic compositions of individual SiC grains from Murchison with diameters between 1.5 and 11.0 μm . Solid lines depict solar $^{14}\text{N}/^{15}\text{N}$ and $^{12}\text{C}/^{13}\text{C}$ ratios. While most data points fall in the upper left quadrant, the five grains X (circles) plot in the lower right quadrant.

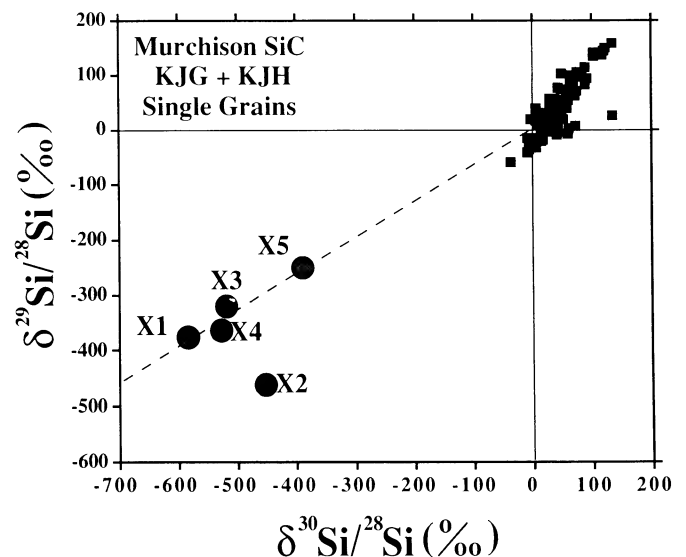


FIG. 2.—Three-isotope plot of Si compositions measured in single SiC grains from Murchison separates KJG and KJH. Plotted are δ -values, the deviations from terrestrial ratios in permil. Solid lines depict solar isotopic ratios ($\delta^{29}\text{Si}/^{28}\text{Si} = 0$). Four of the grains X (circles) lie close to a line (dashed) through isotopically normal Si; X2 deviates substantially from this line.

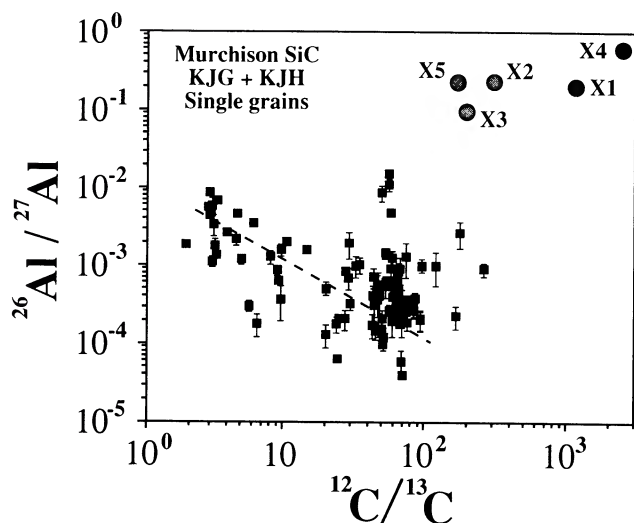


FIG. 3.—Plot of $(^{26}\text{Al}/^{27}\text{Al})_0$ inferred from ^{26}Mg excesses in single SiC grains from Murchison vs. their C isotopic ratios. The broken line indicates the main trend for typical SiC grains. Grains X (circles) are characterized by extremely high $^{26}\text{Al}/^{27}\text{Al}$ and $^{12}\text{C}/^{13}\text{C}$ ratios.

This process would explain higher $^{15}\text{N}/^{14}\text{N}$ ratios, but it would also yield high ^{13}C . This might actually be observed in the correlation of $^{26}\text{Al}/^{27}\text{Al}$ with $^{13}\text{C}/^{12}\text{C}$ in the “nonexotic” (i.e., non-X) grains (Fig. 3) and the fact that grains with very low $^{12}\text{C}/^{13}\text{C}$ (high ^{13}C) tend to have low $^{14}\text{N}/^{15}\text{N}$ (^{15}N excesses) (Fig. 1). Virag et al. (1992) discussed the possibility that SiC grains with very low $^{12}\text{C}/^{13}\text{C}$ ratios carry the products of hot-bottom burning. In order to account for the isotopically light C in grains X Cameron (1992) assumed mixing of essentially pure ^{12}C from the underlying He-burning shell; the required mixing ratio would be from 100:1 to 1000:1 for $C_{\text{He-shell}}:C_{\text{H-shell}}$. Such mixing, however, would also dilute ^{26}Al relative to ^{27}Al , and it is not clear whether a final $^{26}\text{Al}/^{27}\text{Al}$ ratio of up to 0.6 could be achieved.

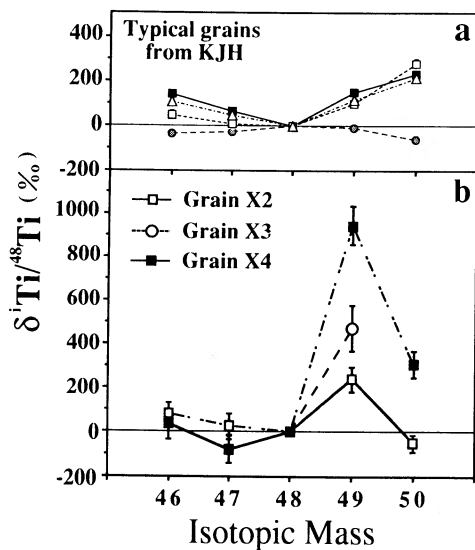


FIG. 4.—Most SiC grains from Murchison have a V-shaped pattern in their Ti isotopic composition (i.e., excesses in all Ti isotopes relative to ^{48}Ti when compared to solar isotopic ratios). In one grain this pattern is inverted (a). Grains X have large excesses in ^{49}Ti and much lesser anomalies in the other isotopes (b). Data for (a) are from Ireland et al. (1991b).

The problem with Si is even more serious. Cameron (1992) explained the ^{29}Si and ^{30}Si depletions in grains X by more rapid proton capture on the heavy Si isotopes than on ^{28}Si at elevated temperatures. A more detailed model calculation of hot-bottom burning (Brown & Clayton 1992) shows that indeed ^{28}Si first becomes enriched relative to ^{29}Si and ^{30}Si by p -capture on ^{27}Al , but this process leads to $\delta^{29,30}\text{Si}$ values of only $\sim -40\%$. At higher temperatures $^{29}\text{Si}(p, \gamma)^{30}\text{P} \rightarrow ^{30}\text{Si}$ dominates and results in ^{29}Si depletions and large ^{30}Si excesses, that is, an isotopic composition in the lower right corner of Figure 2. Such compositions are also obtained by model calculations of hot H-burning in novae (Woosley 1986; Wiescher et al. 1986). Furthermore, the Si-composition in the H-shell would be significantly changed by the admixture of (isotopically heavy) Si from the He-shell accompanying ^{12}C whose dredge-up is necessary to explain the isotopically light C. An additional dilution affecting both the $^{26}\text{Al}/^{27}\text{Al}$ ratio and the Si isotopic composition would take place in the envelope of the AGB star (Forestini, Paulus, & Arnould 1991). This effect, however, would be minimized if most of the envelope had already been expelled during the planetary nebula phase of the star (Cameron 1992; Virag et al. 1992).

At the temperatures characteristic for hot-bottom burning Ca and Ti are not affected by proton reactions. Cameron (1992) interpreted the large ^{44}Ca excess in grain X2 to be the result of neutron capture in the He-shell. This interpretation requires the n -capture cross section of ^{44}Ca to be seven times lower than the accepted value (Beer, Voss, & Winters 1992) and a dilution by a factor 62 with isotopically normal Ca. However, even with this reduced cross section a neutron dose sufficient to produce the observed ^{44}Ca excess would result in a V-shaped isotopic pattern for Ti, such as exhibited by the typical SiC grains (Fig. 4), and not in the ^{49}Ti excess seen in X2. Isotopically anomalies in Ca are found only in grain X2 (which also differs from the other grains X in its Si isotopes; see Fig. 2), but all grains seem to have excesses in ^{49}Ti . This isotopic pattern is the signature of small n -exposures rather than the much larger n -exposures expected for the He-shell of AGB stars (Gallino et al. 1990; Hollowell & Iben 1989). A neutron exposure $\tau = 2.4 \times 10^{-3} \text{ mb}^{-1}$ of isotopically normal Ti would yield $\delta^{47}\text{Ti} = -18\%$, $\delta^{49}\text{Ti} = +1000\%$ and $\delta^{50}\text{Ti} = +140\%$, but such a small exposure would not result in any measurable isotopic effects in Ca. Thus AGB stars cannot provide a consistent explanation of the isotopic compositions of grains X.

3.2. Wolf-Rayet Stars and Novae

Wolf-Rayet stars have also H- and He-burning zones, but the H-burning would not reach high enough temperatures to result in ^{15}N excesses relative to solar or high $^{26}\text{Al}/^{27}\text{Al}$ ratios (Dearborn & Blake 1985; Prantzos & Cassé 1986). As in AGB stars, nucleosynthesis in WR stars cannot explain the Si isotopic composition, and n -exposures in the He-shell are much too high (Prantzos et al. 1990) to account for the Ti-isotopic compositions of the grains X.

Explosive H-burning in novae can produce ^{26}Al (Clayton 1984) with $^{26}\text{Al}/^{27}\text{Al}$ ratios of up to 30 (Woosley 1986) and large ^{15}N excesses (Starrfield, Sparks, & Truran 1985; Wiescher et al. 1986). However, this process produces also large amounts of ^{13}C , and nova ejecta are predicted to have low $^{12}\text{C}/^{13}\text{C}$ ratios (Truran 1986). While most model calculations of nucleosynthesis in novae yield ^{29}Si deficits and ^{30}Si excesses (Woosley 1986; Wiescher et al. 1986), one of the hydrodynamic

nova models of Starrfield, Truran, & Sparks (1978) does result in depletions in both ^{29}Si and ^{30}Si (Wiescher et al. 1986). Finally, neither the ^{44}Ca excess in X2 nor the ^{49}Ti excesses in X2 and the other grains X can be explained by nuclear reactions taking place in novae.

3.3. Supernovae

In contrast to the previously discussed stellar models, supernovae (SNs) can, in principle, account for almost all the isotopic compositions observed in grains X if one considers different zones in these stellar objects. Here the problem is that contributions from different zones have to be mixed together selectively. While it has now become clear that supernova ejecta are extremely turbulent, so that mixing of different zones is not unlikely, it is not clear whether such mixing can yield the correct isotopic compositions in the final product. The He-burning shell of a pre-SN star would provide a C-rich environment for the condensation of SiC, and its C would be essentially pure ^{12}C . ^{15}N enrichment could either be found in this zone or originate from explosive H-burning. The Si isotopic signature of grains X is in general agreement with that predicted from hydrostatic and explosive O-burning (Woosley, Arnett, & Clayton 1973; Woosley 1986), which produce essentially pure ^{28}Si .

One of the most attractive features of a SN source for grains X is that such a source can account for the ^{44}Ca and ^{49}Ti excesses in X2 by the in situ decay of radioactive precursors. Large ^{44}Ca anomalies have been predicted from the decay of ^{44}Ti ($\tau_{1/2} = 47$ yr) by Clayton (1975), and it has been pointed out that ^{44}Ti is the precursor of ^{44}Ca in explosive He-, O-, and Si-burning (Woosley et al. 1973; Woosley 1986). Likewise, ^{49}Ti results from the decay of ^{49}V ($\tau_{1/2} = 331$ days) and is also produced by explosive nucleosynthesis (He-, Ne-, and O-burning) (Woosley 1986). It is intriguing that of all radioactive nuclei in the nuclear mass region of Ca and Ti, ^{44}Ti and ^{49}V are the only ones that have long enough half-lives for in situ decay after grain formation in SN ejecta. Such a scenario is strengthened as there now appears to be evidence for SN grains from SN 1987A (Lucy et al. 1991). On the other hand, if there was live ^{44}Ti in SiC condensed in SN ejecta, the other Ti isotopes in X2 are expected to be much more anomalous than observed (Woosley 1986). Likewise, for grain X4 one would

have to invoke chemical separation between Ti and V in order to explain its ^{49}Ti excess by the in situ decay of ^{49}V .

A major problem for a SN origin of grains X is their high $^{26}\text{Al}/^{27}\text{Al}$ ratio, far exceeding the production ratio of up to 6×10^{-3} expected in Type II SNs (Woosley 1986). Of the total ^{26}Al from such stars, a smaller fraction comes from H-burning in the H-zone. The larger fraction is produced by explosive Ne-burning, but even there the maximum expected $^{26}\text{Al}/^{27}\text{Al}$ ratio is only 2×10^{-2} (Woosley 1986). A possible solution is that we are dealing with a Type I SN (Thielemann, Nomoto, & Yokoi 1986), and the ^{26}Al was produced during the AGB phase of the precursor star and had been present at the surface of the exploding white dwarf.

Even more complicated scenarios can be invoked by considering that in a binary star system *both* stars contributed to the isotopic compositions of grains X. For example, the binary contributing mass to an accreting white dwarf (that becomes a Type I SN) could be an AGB star and provide sufficient ^{26}Al . However, this is just an extension of ad hoc complicated scenarios, and it should be obvious that there is no consistent explanation for the isotopic compositions of grains X. We can only hope that continued theoretical work on nucleosynthesis in stellar sources as well as determinations of the isotopic compositions of additional elements in such grains will one day provide more insight regarding the origin of these stellar messengers.

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