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Presolar Stardust Grains

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Abstract

Meteorites and interplanetary dust particles contain presolar stardust grains: solid samples of stars that can be studied in the laboratory. We review the role of these grains for the sciences of nucleosynthesis, stellar evolution, grain condensation, and chemical and dynamic evolution of the Galaxy. We explain what stardust is, prediscovery ideas about it, how it is collected and recognized, and how evidence of thermal condensation and measured isotopic abundance ratios places individual grains into related families. Unique scientific information derives primarily from the high precision (in some cases $<1\%$) of the measured isotopic ratios of large numbers of elements in single stardust grains. To clarify the new scientific scope, we focus on three of the stardust families: mainstream SiC grains from asymptotic giant branch carbon stars, oxide grains from oxygen-rich red giants, and carbide grains condensed within interiors of young expanding supernovae. Stardust science is just now reaching maturity and will remain an increasingly significant aspect of nucleosynthesis applications.

1.1 Introduction

Presolar stardust grains are solid samples of stars that can be studied in the laboratory. They condensed during cooling of gases in stellar outflows and explosions and became a part of the interstellar medium (ISM) from which our Solar System ultimately formed. The word "stardust" distinguishes such interstellar dust from the larger mass of interstellar dust that formed in other ways. The word "presolar" indicates that these grains formed in stars that evolved prior to the birth of the Sun. Presolar grains are overwhelmingly extracted from the most primitive meteorites, small asteroid fragments whose orbits have intersected the Earth, and from interplanetary dust particles, cometary and asteroidal particles collected at high altitudes in the atmosphere. Asteroids and comets both formed during the earliest stages of solar system formation. The stardust grains they contain must necessarily have formed from pre-existing stars, and each stardust grain is hence older than the Sun. Stardust grains that condensed in stars younger than the Sun certainly exist in the ISM today, but samples of them have not yet been collected. The current NASA STARDUST mission may return the first samples of younger stardust.

Presolar grains are recognized as such by their highly unusual isotopic compositions, in essentially every element that they contain, relative to all other materials available for laboratory study. Their isotopic variations are too large (>4 orders of magnitude in some cases)

Table 1.1. *Types of presolar grains in meteorites and interplanetary dust particles (IDPs).*

Phase	Abundance (ppm)	Size	References
Diamond	1400	2 nm	[1]
SiC	14	0.1–20 μm	[1]
Graphite	10	1–20 μm	[1]
TiC, ZrC, MoC, RuC, FeC, Fe-Ni	(sub-grains in graphite)	5–220 nm	[2,3]
Silicon Nitride (Si_3N_4)	>0.002	$\sim$1 μm	[4]
Spinel ($MgAl_2O_4$)	1	0.1–3 μm	[4]
Corundum (Al_2O_3)	<0.1	0.1–3 μm	[5]
Hibonite ($CaAl_{12}O_{19}$)	0.002	$\sim$2 μm	[6]
Silicates	500 (in IDPs)	0.3–1 μm	[7]

Key to references: [1] Huss, Hutcheon, & Wasserburg 1997, [2] Bernatowicz et al. 1996, [3] Croat et al. 2004, [4] Nittler et al. 1995, [5] Zinner et al. 2004, [6] Choi, Wasserburg, & Huss 1999, [7] Messenger et al. 2003.

to be explained by chemical or physical fractionations. Rather, many of these variations clearly point to nuclear reactions occurring in individual stars. Quite a large literature exists documenting the discovery of these grains and their astrophysical consequences. We cite several key papers as we proceed, but our goal is more to indicate the types of arguments and conclusions that they have uniquely brought to astrophysics. The state-of-the-art in the field has been reviewed recently by Zinner (1998) and Nittler (2003). Many details of the astrophysical implications of stardust may be found in the volume edited by Bernatowicz & Zinner (1997).

The known types of presolar stardust are listed along with their sizes and abundances in Table 1.1. Figure 1.1 shows scanning electron microscope images of two presolar stardust grains. Both examples are single crystals of silicon carbide (SiC), but their isotopic compositions (indicated in the caption) reveal that they formed in quite different stars. Figure 1.1*a* shows a so-called mainstream grain, and Figure 1.1*b* shows a so-called X grain. Both grains are a few μm in diameter, larger than is typical for both interstellar dust and meteoritic stardust, but amenable to isotopic analysis. New technologies are now allowing isotopic analysis of presolar stardust grains with more typical sizes (<500 nm). A 1μm SiC grain contains $> 10^{11}$ atoms, sufficient for statistically precise measurements of isotopic ratios not only for C and Si but even for minor and trace elements within the SiC (such as N, Mg, Ca, Ti, and others in some cases). Such grains are also large enough for experimental determination of their mineralogy and microstructure. For example, most SiC stardust grains have been found to be single crystals with the cubic structure (β-SiC), rather than one of the hexagonal polymorphs (α-SiC) common in industrially produced SiC (Daulton et al. 2002). Crystalline SiC is expected to condense in a carbon-rich gas as it cools from high temperatures due to expansion, and β-SiC has, in fact, been identified in circumstellar envelopes of carbon stars (Speck, Hofmeister, & Barlow 1999). The extremely nonsolar isotopic compositions of the grains indicate that they completely condensed and cooled prior to any mixing with interstellar matter.

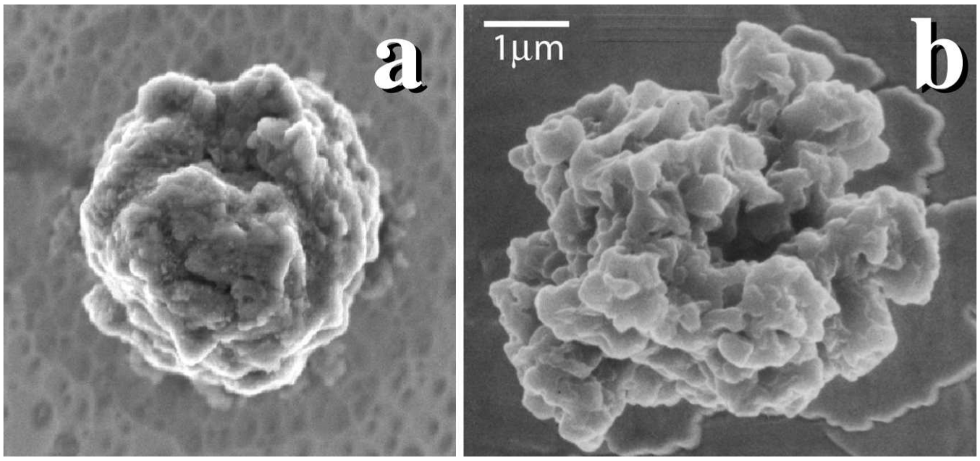

Fig. 1.1. Scanning electron microscope images of two presolar stardust grains of silicon carbide (SiC). (*a*) "Mainstream" grain with: $^{12}C/^{13}C = 60$ (solar ratio is 89), $^{14}N/^{15}N$ = 1,000 (solar ratio is 272), $^{29,30}Si/^{28}Si = 1.05 \times$ solar, inferred $^{26}Al/^{27}Al \approx 10^{-3}$. This grain formed in a presolar C-rich AGB star. (*b*) "X" grain with: $^{12}C/^{13}C = 300$, $^{14}N/^{15}N = 60$, $^{29}Si/^{28}Si = 0.7 \times$ solar,$^{30}Si/^{28}Si = 0.5 \times$ solar, inferred $^{26}Al/^{27}Al \approx 0.3$. This grain formed in the ejecta of a presolar supernova.

Note that the grains discussed here are not typical of the main mass of dust in the ISM. Most dust mass is believed to consist of atoms and molecules accreted onto refractory grain cores when the diffuse ISM joins clouds (Draine 2003). Because the atoms and molecules that make up the grain mantles originated in diverse nucleosynthetic sites, most of the interstellar dust mass is probably isotopically homogeneous (∼solar in the Sun's parent molecular cloud). It is evident that the stardust grains could not have formed by a process of heating of interstellar mantles accreted in molecular clouds, both because they are minerals (not amorphous solids) and because their isotopic ratios are too extreme and indicative of individual stars. For example, the highly nonsolar isotopic ratios of the SiC grains shown in Figure 1.1 are not expected in interstellar-grain mantles, but, based on both observations and theoretical models, these ratios speak strongly in favor of thermal condensation within asymptotic giant branch (AGB) C-star winds (mainstream grain) and supernova interiors (X grain).

1.1.1 Isolation and Analysis of Presolar Stardust

Presolar stardust comprises a small fraction of interstellar dust and of meteorites (Table 1.1). Meteoritic evidence is that stardust accounts for much less than 1% of refractory atoms in solar system solids, though how much preexisting dust was destroyed during solar system formation is unknown. Identifying this small fraction of stardust within a meteorite rock is not an easy matter. Most presolar grains have been identified in acid-resistant residues left over after most of the mass of the meteorite has been dissolved in strong acids. The chemical dissolution procedures were developed experimentally as a means of concentrating the unknown carriers of isotopically anomalous noble gases (Lewis & Anders 1983). Once these carrier concentrations were achieved, the residues were shown to contain many of the mineral phases listed in Table 1.1 (Amari, Lewis, & Anders 1994). These then were analyzed grain by grain for mineral structure, primarily by electron diffraction, and for isotopic ratios by secondary-ion mass spectrometry (SIMS). Such studies, which required breathtaking techniques for handling and analysis of microparticles, confirmed that individual stardust grains were the carriers of the noble gas anomalies and had isotopic variations in their major elements as well (Bernatowicz et al. 1987; Zinner, Tang, & Anders 1987). Identification of even rarer presolar phases (e.g., oxide minerals, Si_3N_4, and TiC) required development of automated particle analysis techniques (Nittler et al. 1995, 1997; Choi et al. 1998) and ultramicrotomy and transmission electron microscopy techniques (Bernatowicz et al. 1996). Recently, the development of new SIMS instrumentation has resulted in the discovery of presolar silicate stardust in interplanetary dust particles (Messenger et al. 2003). Such grains would have been destroyed in the prior acid treatments.

1.1.2 Astronomical Context

To glimpse the astrophysical potential of presolar stardust one can imagine a telescope capable of measuring isotope ratios to better than 1% precision in stars. Imagine what science one could do with such a telescope: nucleosynthesis in stars, stellar structure, and chemical evolution of the Galaxy. These are exactly the topics that the measured presolar grains clarify. But because the isotopic ratios are measured with considerably greater precision than is possible for astronomers, the questions that can be pursued are more precise and novel. The problem with this rosy picture is that each grain bears no label identifying its parent star! The parent star has long been gone, so at best one can identify the type of star and its evolutionary status. Knowledge of stars and their nucleosynthesis is required to identify the donor stars. Knowledge of condensation chemistry is also required. However, once the type of donor star is identified, the grain data can provide new knowledge about it. New science always follows from precise new information, and stardust is no exception to that rule.

1.1.3 Theoretical Precursors

The 1987 discovery of presolar grains from stars did not occur in a vacuum. In addition to a huge literature on interstellar dust there existed the following specific idea: dust condensed within stars and stellar outflows will record the isotopic signatures of that star and, as a natural component of the ISM, may be found in meteorites. Much of the observable expectation at the time lay in possible cosmic chemical memory (Clayton 1982) of anomalous ISM dust within solids that were grown from that dust in the solar system. Early suggestions were that the ^{16}O-richness of millimeter-sized solar system rocks called calcium-aluminum-rich inclusions resulted from their containing in their precursors dust that condensed in some undefined manner near supernovae (Clayton, Grossman, & Mayeda 1973) and that a ^{22}Ne-rich gas component within meteorites arose from the meteorites containing presolar dust rich in ^{22}Ne (Black 1972). These works did not suggest an origin for the isotopic anomalies in thermal condensation in stars, but they contributed to the emergence of the idea that isotopes might identify presolar material or rocks grown from it. What came as a surprise was the later isolation and characterization of several ISM stardust components themselves, apparently unchanged, within the meteorites.

In that early and fragmentary setting, the following ideas were set forth.

(1) Dust should condense thermally both in the interior of expanding supernovae and in red giant winds and be evidenced by huge and predictable isotopic anomalies (Clayton 1975, 1978).
(2) Supernova dust should have excess daughter abundances from extinct radioactivities (^{22}Na, ^{26}Al, ^{40}K, ^{41}Ca, ^{44}Ti, and ^{53}Mn) that were still alive when the grains condensed and stable isotope anomalies reflecting the compositions of the distinct supernova condensation zones.
(3) Differing grain phases would reflect differing supernova zone chemical compositions, such as sulfides replacing oxides in O-depleted inner supernova shells (Clayton & Ramadurai 1977).
(4) Stardust from red giant stars should contain *s*-process isotopes (Clayton & Ward 1978; Srinivasan & Anders 1978), with carbonaceous *s*-process carriers originating in AGB carbon stars (Clayton 1982, 1983; Swart et al. 1983).
(5) Dust should also condense in novae and be evidenced by extinct ^{26}Al and ^{22}Na, ^{30}Si-rich Si, ^{13}C-rich C, and ^{15}N-rich N (Clayton & Hoyle 1976).
(6) Finally, ^{13}C-rich carbon residues correlated with *s*-process Xe were attributed to stardust from a late-type red giant having C/O > 0.9 in order to condense carbon (Swart et al. 1983).

Once presolar grains were isolated in 1987, their isotopic compositions were investigated for every element measurable at the time. Many of the isotopic signatures were found to agree with the prior predictions. Moreover, these theoretical precursors, and even the controversy that they generated, provided fertile soil for quick interpretation of the first measurements of extreme isotopic anomalies in presolar grains after they were found in meteorites. The issues then became identification of the parent stars from the isotopic signatures of the presolar grains and the ways in which the precise isotopic ratios confronted stellar models.

1.1.4 A Watershed of s/r Decomposition

Many papers at this conference evaluate implications of astronomical observations of *s*-process and *r*-process elements, requiring that each heavy element be decomposed into contributions from those nucleosynthesis components. Based on such a decomposition, presolar stardust provided the first unambiguous demonstration that the *s*-process isotopes were synthesized in a special type of star and admixed into the ISM, including our solar system 4.6 Gyr ago. Quantitative decomposition of Solar heavy-element abundances into *s*-process and *r*-process contributions was first performed by Clayton et al. (1961), following their mathematical solution and evaluation of the *s*-process abundances. Their Table 4 listed N_r/N_s for each heavy element, and Clayton & Fowler (1961) provided the same for each of their isotopes. These first estimates were numerically rough because for N_r they utilized coarse, steady-state *r*-process calculations by B^{2}FH (Burbidge et al. 1957). An improved idea was introduced by Seeger, Fowler, & Clayton (1965), who introduced the exponential distribution of neutron fluences to calculate N_s and determined N_r by subtracting N_s from the total isotopic abundance. This improved procedure has become the method of choice, and was adopted by the Karlsruhe program of *s*-process studies, which has continued to present improved numerical evaluations of that same procedure (Käppeler et al. 1982; Käppeler 1999).

Nonetheless, the decomposition of solar abundances remained an act of theoretical faith until apparently pure *s*-process xenon gas was discovered in an acid-resistant residue of the Murchison carbonaceous meteorite (Srinivasan & Anders 1978). Their Figure 2 compared the isotopic composition of the Xe gas driven from that carbonaceous residue by high heating with the *s*-process decomposition of Xe calculated earlier by Clayton & Ward (1978). The

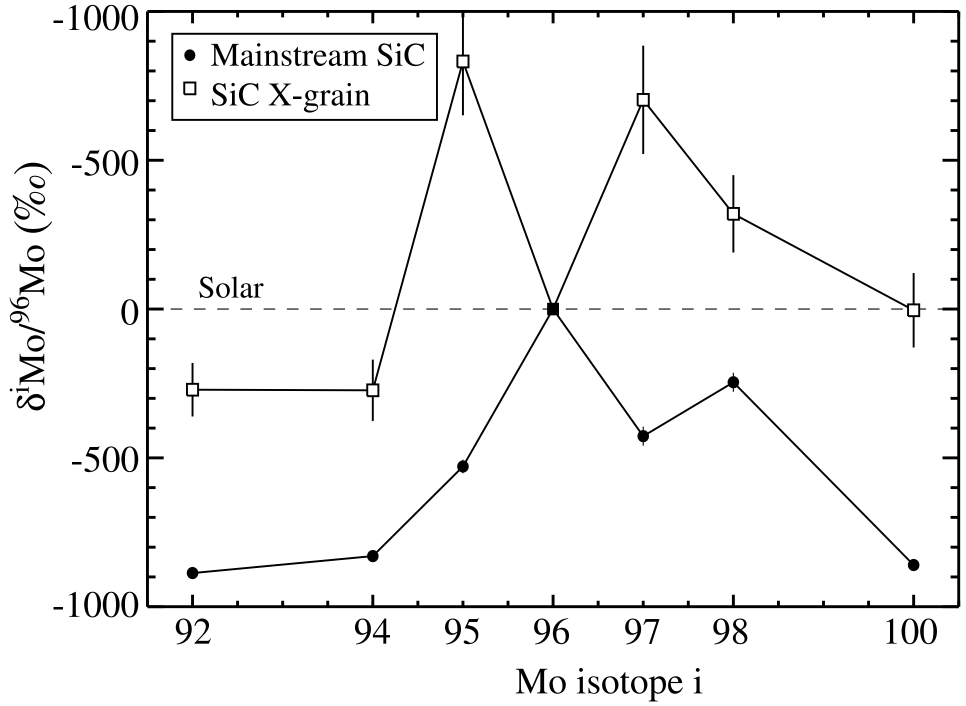

Fig. 1.2. Mo isotopic ratios measured in two presolar SiC grains. Ratios are expressed as δ-values, permil deviations from a terrestrial isotope standard: $\delta R = (R_{\mathrm{meas}}/R_{\mathrm{standard}} - 1) \times 10^3$. All isotopes in the mainstream grain are depleted relative to the *s*-process only ^{96}Mo. Thus, this grain clearly reveals an *s*-process signature, confirming an origin in a low-mass AGB star atmosphere. In contrast, the X grain (known to have formed in a supernova) is unusually enriched in ^{95}Mo and ^{97}Mo, indicating "neutron burst" nucleosynthesis (§1.5). Data from Nicolussi et al. (1998) and Pellin et al. (2000).

stunning agreement shown in that figure was the first occasion on which essentially pure *s*-process isotopes could be measured experimentally. It advanced natural philosophy by demonstrating that the *s*-process idea is correct, that it occurred in individual stars, and that the theoretical decomposition of the Solar mixture into *s* and *r* was justified. Clayton & Ward (1978) also argued that purely *s*-process isotopes could be sequestered in the ISM within red giant stardust, and they had emphasized Xe because other isotopic variations of Xe isotopes were already known; but, curiously, their prediction in 1975 was not accepted for publication until 1978 when the detection (Srinivasan & Anders 1978) supported its ideas. Building on this finding, Swart et al. (1983) showed that the *s*-process-rich residue was also very ^{13}C-rich, about twice the Solar complement, compared to ^{12}C, and suggested that dust from carbon stars carried both signatures into the meteoritic acid-resistant residue. An engaging historical account of this development can be found in Scientific American (Lewis & Anders 1983).

The demonstration that some unknown ^{13}C-rich chemical form of red giant stardust was present within the meteorites provided one of several motivations for further purification of that residue until the *s*-process carriers were identified as micron-sized grains of crystalline

SiC (Bernatowicz et al. 1987), which were then shown to collectively carry *s*-process Xe (Lewis, Amari, & Anders 1994) and ^{13}C-rich carbon (Zinner et al. 1987). In this way the *s*-process pattern, and also ^{22}Ne-rich gas, contributed to the discovery of stardust and the isotopic astronomy that has been made possible by it. Newer high-sensitivity techniques in M. Pellin's laboratory at Argonne have been able to measure *s*-process isotopic patterns in individual presolar stardust grains (rather than collections of grains) for some refractory elements (Nicolussi et al. 1997, 1998; Pellin et al. 2000). Figure1.2 shows their measurement of molybdenum isotopes in two SiC grains (Nicolussi et al. 1998). The "mainstream" grain contains essentially pure *s*-process Mo. As discussed in greater detail below, other isotopic data for the mainstream SiC stardust grains indicate an origin in AGB carbon-star atmospheres. Thus, the grains provide an unprecedented opportunity for confronting AGB star *s*-process nucleosynthesis models with high-precision observational data (Lugaro et al. 2003).

1.2 Classification of Isotopic Families of Stardust

For sake of illustration, suppose four elements can be measured isotopically (frequently true) in a single stardust grain. This provides at least one, and probably more, isotope ratio for each element. For example, in the most-studied stardust grain type, SiC (Fig. 1.1), elements routinely measured are Si, C, and N. The independent isotopic ratios that can be formed are Si (2), C (1), and N (1). Those measured ratios form the basis of a multidimensional space, 4-dimensional in this example, within which each grain assumes a unique position (a point). That space is mostly empty; but filled volumes enable identification of clustered groups and trends. Isotopically related grains form the basis of a classification system and enable science to be constructed. The significance of this classification system for stardust grains can be likened to the significance for stars of spectral types, the main sequence, and the Hertzsprung-Russell diagram.

The SiC classification is illustrated by Figure 1.3, which shows the ^{14}N/^{15}N vs. ^{12}C/^{13}C isotopic subspace. Differing symbols shown in these plots and associated labels (mainstream, A+B, X, Y, Z) have arisen historically (Hoppe et al. 1994) to label clusters or trends of individual SiC grains that are believed to be related. That the groupings are not arbitrary is indicated also by considering the Si-isotope space (Fig. 1.4). For example, the Y grains are defined on the basis of having ^{12}C/^{13}C > 100, but plot largely to the right of the slope 4/3 line formed by the mainstream grains on the Si 3-isotope plot. The Z grains, on the other hand have similar C isotopes to the mainstream, but quite different Si isotopes. Such considerations show the alphabet families to be less arbitrary than they might at first seem. In many SiC grains, it is also possible to measure more isotopic ratios than those of Si, C, and N (Hoppe et al. 1994, 1996; Hoppe & Ott 1997; Nittler & Alexander 2004). These include: ^{46,47,49,50}Ti/^{48}Ti; initial ^{26}Al/^{27}Al ratios inferred from excess radiogenic ^{26}Mg; Ca isotope ratios, enabling also the determination of initial ^{44}Ti/^{48}Ti ratios from radiogenic ^{44}Ca excesses and Ti/Ca ratios (Nittler et al., 1996); and, with special laser techniques, isotopic ratios of noble gases He and Ne (Nichols et al. 2004) and the heavy trace elements Ba, Fe, Sr, Mo, and Zr (Nicolussi et al. 1997; Pellin et al. 2000; Savina et al. 2003). Any of these help further constrain the donor star types for individual SiC grains.

Similar isotopic clustering has allowed subclassification of other stardust minerals as well, including graphite and oxides. In this paper we will discuss but a few presolar grain classes to illustrate main arenas of scientific impact, beginning with the mainstream SiC grains,

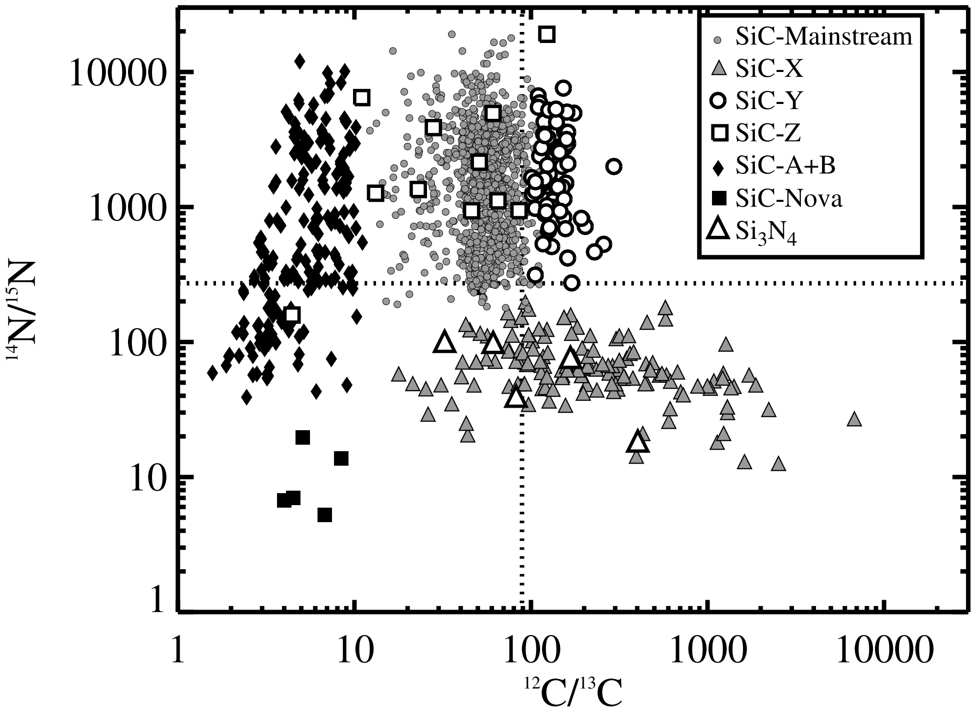

Fig. 1.3. C and N isotopic ratios measured in individual presolar SiC and Si_3N_4 grains (Data from: Hoppe et al. 1994, 2000; Nittler et al. 1995; Hoppe & Ott 1997; Huss et al. 1997). Dotted lines indicate the Solar isotope ratios here and in subsequent figures.

which serve to illustrate how distinctive grain families are recognized and attributed to stellar sources. Space will prevent detailed discussion of the SiC grains of type Y (Amari et al. 2001a), A+B (Amari et al. 2001b), and of nova grains (Amari et al. 2001c). Type X SiC will be discussed in §1.5. Also it must be said that the numbers of isotopic ratios that can be measured in stardust grains depends on the type of grain (Table 1.1), whose mineral structure and properties determine the abundances of trace elements that condensed sufficiently abundantly to yield isotopic measurements and grain size, which determines the total number of atoms available for measurement. Moreover, the SIMS technique is destructive, so that some grains can be consumed before all elements can be measured.

1.3 Mainstream SiC

The mainstream grains, which comprise $\sim$90% of the presolar SiC population (Figs. 1.3 and 1.4) were formed during mass loss from AGB carbon stars. This is attested to by: (1) their $^{12}C/^{13}C$ ratios distributed about the value 60, just as are the carbon stars (Smith & Lambert 1990); (2) large $^{14}N/^{15}N$ ratios owing to the dredge-up events that enrich the atmosphere in CN-burning products (Boothroyd & Sackmann 1999) along with the He-shell-burning products (Busso, Gallino, & Wasserburg 1999); (3) nearly pure *s*-process compositions of heavy elements (Srinivasan & Anders 1978; Prombo et al. 1993; Nicolussi et al. 1997), but including evidence of *s*-process branching unique to AGB stars (Gallino,

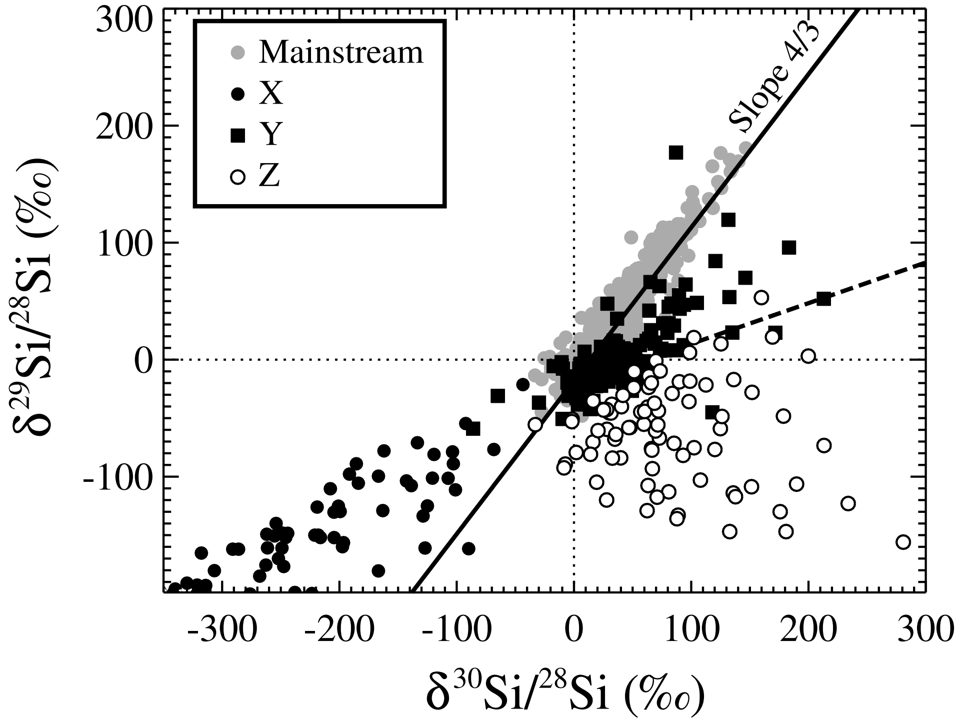

Fig. 1.4. Si isotopic ratios, expressed as δ-values, for presolar SiC grains of type mainstream, X, Y and Z (Data from: Hoppe et al. 1994, 2000; Hoppe & Ott 1997; Huss et al. 1997; Nittler & Alexander 2004). A+B grains have similar isotopic ratios to the mainstream, while the X grains extend to much more ^{28}Si-rich compositions than shown (lower left quadrant). Mainstream grains form an array with slope 1.3 (solid line). Long-dashed line indicates trajectory expected for dredge-up of He-shell material in an individual AGB star (see text).

Busso, & Lugaro 1997; Lugaro et al. 2003); (4) ^{26}Al/^{27}Al initial ratios up to $\sim 10^{-3}$ (Hoppe & Ott 1997) as expected for AGB stars (Forestini, Paulus, & Arnould 1991); and (5) noble gas isotopes reflecting a mixture between He-shell *s*-process and initial atmospheric gas (Lewis, Amari, & Anders 1990; Nichols et al. 2004), just as AGB models yield. Furthermore, an infrared feature associated with SiC has been observed around C stars (Speck, Barlow, & Skinner 1997), confirming the thermodynamic expectation that C-based solids (e.g., SiC) condense only in gas having C abundance comparable to or greater than O abundance (e.g., Lodders & Fegley 1995). Today it seems beyond doubt that AGB C stars are the birthplaces of mainstream SiC.

The good linear correlation of mainstream grain Si isotopes in Figure 1.4 surely attests to the mainstream being a physical family rather than an arbitrarily defined subgroup of stardust. However, a key observation is that the mainstream Si array has a much steeper slope on the Si 3-isotope plot and a larger range than expected from AGB star dredge-up processes (Gallino et al. 1994; Lugaro et al. 1999). Almost certainly, this array reflects a range of initial compositions in the Si isotopes of the stardust parent AGB stars. A related

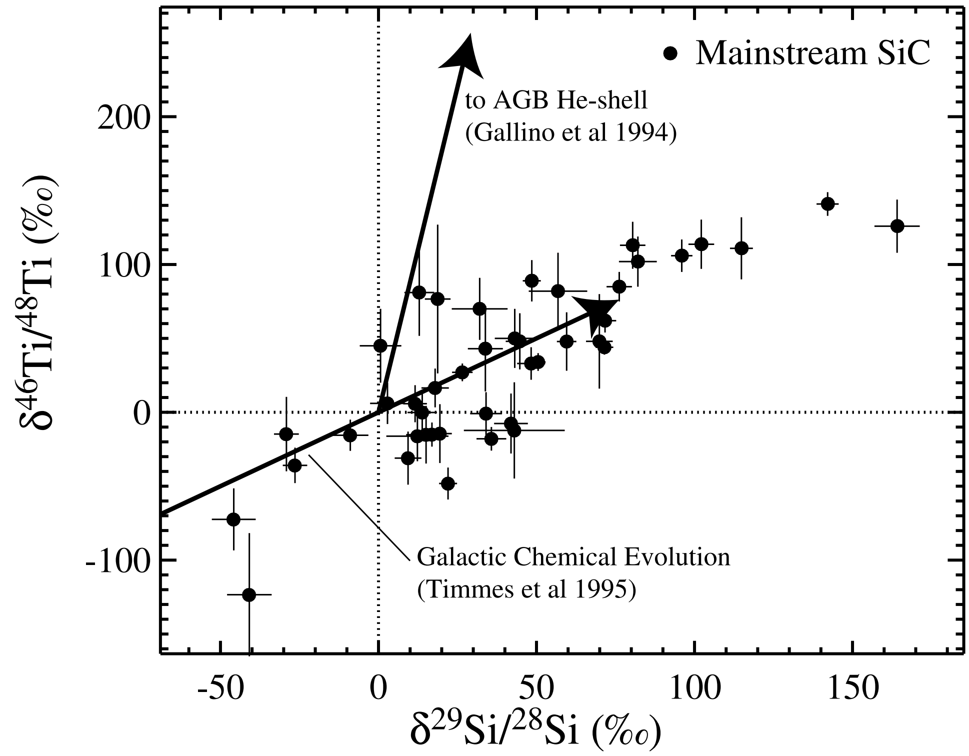

Fig. 1.5. Correlation of Si and Ti isotopic ratios in mainstream presolar SiC grains. The slope of the correlation is distinct from that expected for AGB star dredge-up, but compatible with models of Galactic chemical evolution. Data are from Hoppe et al. (1994) and Alexander & Nittler (1999).

property of the mainstream grains is a strong correlation between Si and Ti isotopic ratios (Hoppe et al. 1994; Alexander & Nittler 1999), illustrated in Figure 1.5 for $^{29}Si/^{28}Si$ and $^{46}Ti/^{48}Ti$. Again, the observed correlation is primarily one of initial compositions for the AGB stars because the slope and extent of the line differ strongly from the expected shifts of these isotopes owing to *s*-process dredge-up in individual AGB stars.

1.3.1 New Astrophysics with Mainstream Grains

Several astrophysical issues are raised by the mainstream grains. These have led to new astrophysical knowledge. We illustrate only a few major issues here.

1.3.1.1 AGB Neutron Sources

Solar abundances already offered evidence of competition at branch points in the *s*-process path between neutron capture and temperature-dependent β decay rates (Ward, Newman, & Clayton 1976). The mainstream grains intensified this greatly since they are so close to pure *s*-process in their heavy elements. Their data revealed that no single temperature and neutron flux could simultaneously match the many branchings. A new physical model of the AGB *s*-process was developed in part because of this data-induced crisis. In that model, the ^{13}C(α,n)^{16}O neutron source is the major source of the neutron fluence. The ^{13}C is produced by a pocket of mixing between envelope H and new ^{12}C after each thermal shell flash. The ^{13}C so produced burns, releasing its neutrons in hydrostatic equilibrium ($n_n = 10^7$ cm^{-3}, kT = 8 keV) during the interpulse phase of the AGB (Gallino et al. 1998; Arlandini et al. 1999). Subsequently during the He-shell flash, the ^{22}Ne source is activated to provide differing branch conditions ($n_n = 10^{10}$ cm^{-3}, kT = 23 keV) for a smaller neutron fluence, and this higher-T, higher-neutron-flux capture partially resets the branches established during the ^{13}C-burning interpulse. The resulting agreements not only brought many branch points into closer concordance but clearly established the ^{13}C neutron source as provider of the major fluence for the *s*-process. The new AGB model even provides an explanation for the bizarre observation that larger SiC grains have a greater *s*-process production ratio for ^{86}Kr/^{82}Kr than do smaller grains (Lewis et al. 1994), if higher-mass AGB stars (which have hotter ^{22}Ne flashes, larger n_n, and thus more ^{86}Kr) also produce the larger SiC grains. This two-neutron-source AGB model has not only been established by the SiC data and by models of AGB stars, it has also rejuvenated the science of accurate 10–30 keV neutron capture measurements for the *s*-process (Arlandini et al. 1999; Käppeler 1999).

1.3.1.2 Presolar Metallicity Greater than Solar with Slope = 4/3

Figures 1.4 and 1.5 show that most of the AGB stars donating mainstream grains had ^{29}Si/^{28}Si and ^{30}Si/^{28}Si ratios greater than solar (lying between about 95% and 120% of solar). This raises the question of how stars that were born and evolved prior to the birth of the Sun acquired such compositions. Clayton (1988) had suggested that the chemical evolution of the Galaxy would correlate ^{29}Si/^{28}Si and ^{30}Si/^{28}Si initial ratios within stars; but in a mixed-ISM, one-zone model that produces the Sun, presolar ratios should all be less than solar (Timmes & Clayton 1996). Furthermore, Timmes & Clayton (1996) showed that their deviations from solar should be equal when normalized to solar abundances (correlated along a line of slope 1), rather than distributed along a line of slope 4/3, as observed. Because the *s*-process in AGB stars enriches these ratios by only about 10 and 30 permil, respectively (Lugaro et al. 1999), much less than their observed spread along the mainstream line in Figure 1.4, the mainstream correlation is indeed a correlation in initial stellar abundances, as chemical evolution anticipates. But why the different slope and why are the mainstream grains isotopically heavier in Si than the Sun? This is a great and unexpected finding about the presolar Milky Way and/or the Sun. Attempts to resolve this crisis have taken four directions.

First, if the Sun (rather than the mainstream grains) is taken to be abnormal, then the heavy-isotope richness of presolar AGB stars can be regarded as a heavy-isotope deficiency of the Sun. Clayton & Timmes (1997) showed that it is hard to understand how the slope 4/3, which requires very different solar ^{29}Si/^{28}Si and ^{30}Si/^{28}Si isotope ratios than those of the evolving ISM, can, miraculously, also produce a Sun that lies almost on the mainstream line. Clayton & Timmes' Figure 4 shows that to do so requires a very large (about 30%) enrichment of ^{30}Si within each AGB star, much in excess of AGB model expectations and also apparently ruled out by C-isotopic ratios of AGB stars and mainstream grains (Alexander & Nittler 1999). Alternatively, it cannot be ruled out that the true ISM Si isotope evolution occurs along a slope 4/3 line. However, this would likely require that ^{30}Si is greatly overproduced relative to ^{29}Si in low-metallicity supernovae, and there is no hint of such behavior in current supernova calculations or reason to expect it from astrophysical considerations.

Second, Clayton (1997) proposed that new AGB stars born central to the solar orbit, where the Galactic metallicity gradient could result in stars having higher initial $^{29,30}Si/^{28}Si$ ratios, scatter outward from molecular clouds or other Galactic features to donate their SiC grains at the larger solar birth radius. Sellwood & Binney (2002) have given support to the general astronomical idea by calculating radial migration of low-mass stars as result of their "surfing" spiral density waves (rather than scattering from clouds). A semi-analytic model by Nittler & Alexander (1999) indicates that such outward orbital diffusion of stars would not necessarily result in the observed Si isotopic distribution of the SiC, but additional research is clearly needed.

Third, Lugaro et al. (1999) utilized the distinction (Timmes & Clayton 1996, Fig. 2) between low-mass and high-mass supernovae Si ejecta to construct an inhomogeneous chemical evolution model that produced stochastically a slope 4/3 in the Si three-isotope plot. Their model is certainly oversimplified and contains some arbitrary aspects that could invalidate it. Moreover, Nittler (2002) has shown that it does not reproduce the strong observed correlation between $^{46}Ti/^{48}Ti$ and $^{29}Si/^{28}Si$ ratios. The Si-Ti correlation does allow some of the spread in the data to be explained by such a model, but it seems unlikely that heterogeneous evolution can account for the entire mainstream Si distribution.

Fourth, the initial compositions of the AGB stars would have correlated isotopic compositions if they were varying mixtures of two distinct isotopic reservoirs. Clayton (2003) has advanced this as having been caused by a galactic merger about 6.5 Gyr ago between the gas of the Milky Way disk and that of a lower-metallicity satellite galaxy that therefore has lower $^{29}Si/^{28}Si$ and $^{30}Si/^{28}Si$ isotope ratios. The AGB stars appeared in a burst of star formation stimulated by the merger. The Sun, which formed about 2 Gyr later, had been enriched by supernova less massive than 25 $M_\odot$, whose ejecta do not increase and may even lower the Sun's $^{29,30}Si/^{28}Si$ ratios, allowing the Sun to remain at the lower extreme of the mainstream three-isotope Si plot (Fig. 1.4). This suggestion still needs to be rigorously investigated, but it appears promising to explain a range of observations.

1.4 Presolar Oxide Grains

Presolar oxide minerals (spinel, corundum, hibonite, TiO_2, and silicates) are more difficult to locate in meteorites than is C-rich dust due to a large background of isotopically solar dust formed in the solar system. Nonetheless, several hundred grains have now been found, mostly through automated techniques (e.g., Nittler et al. 1997; Choi et al. 1998). O isotope data are shown in Figure 1.6; the isotope ratios span several orders of magnitude and, as was seen above for SiC, tend to define distinct trends allowing for a classification system. Additional isotopic data for some grains, including inferred $^{26}Al/^{27}Al$ ratios and Ti and Mg isotopic ratios, have helped further constrain stellar origins of the grains. The majority of the grains (Groups 1 and 3, according to Nittler et al. 1997) have isotopic compositions in good agreement with observations of O-rich red giant and AGB stars (e.g., Harris & Lambert 1984) and with theoretical expectations for ^{17}O dredge-up in such stars (Boothroyd & Sackmann 1999). Comparison with the stellar models indicates that the grains formed from stars with a range of masses ($\sim 1.2-5 M_\odot$) and with initial $^{18}O/^{16}O$ ratios probably reflecting Galactic chemical evolution. A supernova origin has been suggested for two grains, and the origin of the Group 4 grains remains enigmatic (Galactic chemical evolution and supernova mixing have been suggested). The ^{18}O-depleted Group 2 grains are discussed below.

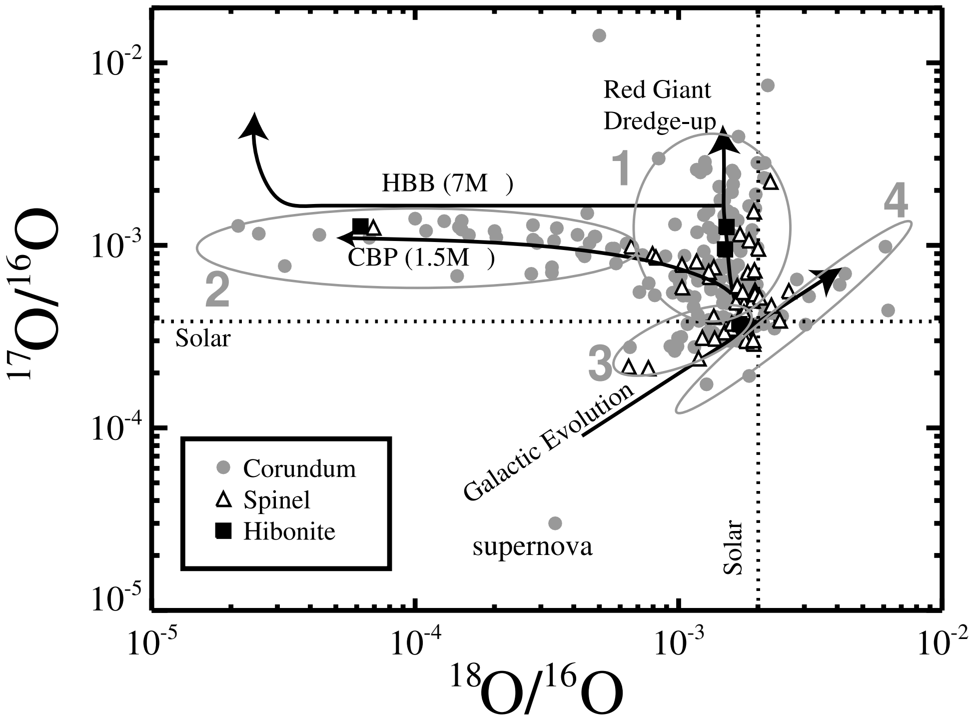

Fig. 1.6. O isotopic ratios measured in presolar oxide stardust. Grey ellipses indicate Group definitions of Nittler et al. (1997). Theoretical expectations for Galactic evolution (Timmes, Woosley, & Weaver 1995), red giant dredge-up (Boothroyd & Sackmann 1999), cool-bottom processing (CBP; Nollett, Busso, & Wasserburg 2003) and hot-bottom burning (HBB; Boothroyd, Sackmann, & Wasserburg 1995) are schematically shown. Data from Nittler (1997, and references therein), Choi et al. (1998, 1999), Nittler et al. (2003), and Zinner et al. (2004).

1.4.1 New Astrophysics with Presolar Oxide Grains

1.4.1.1 Extra Mixing in Red Giant and AGB Stars

Some 10% of presolar oxide grains (Group 2) have lower $^{18}O/^{16}O$ ratios and higher $^{26}Al/^{27}Al$ ratios than can be explained by standard stellar evolutionary models, indicating partial H-burning of the parent stars' envelopes. Two proposed mechanisms for this processing are hot-bottom burning (HBB) in intermediate-mass AGB stars (Boothroyd et al. 1995) and cool-bottom processing (CBP) in low-mass AGB stars (Wasserburg et al. 1995). CBP, also called "extra" or "deep" mixing, has previously been invoked to explain anomalously low $^{12}C/^{13}C$ ratios in low-mass red giants (Charbonnel 1994) as well as high Na and Al abundances in globular cluster giants (Shetrone 1996). Recent calculations by Nollett et al. (2003) strongly favor a CBP explanation for most of the Group 2 oxides and indicate that the grain compositions can constrain mixing rates and temperatures. Thus, the stardust grains reveal that CBP occurs in AGB stars as well as red giants, and it is hoped that the grain data will help identify the still unknown physical mechanism(s) driving the extra mixing. HBB is ruled out for most of the Group 2 oxide grains, because it can explain neither grains with intermediate $^{18}O/^{16}O$ ratios nor grains with $^{17}O/^{16}O$ ratios <0.001 (Boothroyd et al. 1995). However, Nittler et al. (2003) found an extreme Group 2 spinel grain with very high $^{25}Mg/^{24}Mg$ and $^{26}Mg/^{24}Mg$ ratios. The composition of this grain is most consistent with an origin in a $\sim 4-5\,M_{\odot}$ HBB AGB star. This result indicates that intermediate-mass stars were dust contributors to the protosolar cloud. Moreover, it emphasizes the importance of using multiple elements in individual grains to better constrain stellar origins.

1.4.1.2 Galactic Chemical Evolution and the Age of the Galaxy

The distribution of $^{18}O/^{16}O$ ratios observed in Group 1 and 3 oxide grains is in good agreement with expectations for Galactic chemical evolution, which predicts that $^{17}O/^{16}O$ and $^{18}O/^{16}O$ ratios should increase with metallicity (Clayton 1988; Nittler et al. 1997). This agreement indicates that the Galactic chemical evolution of O isotopes is reasonably well understood, and thus metallicities of parent stars can be inferred from grain O-isotope ratios and theoretical models. The grains can then be used to trace evolutionary histories of other isotope systems. For example, $^{25}Mg/^{24}Mg$ ratios measured in presolar spinel and corundum grains (Nittler et al. 2003) indicate that this ratio evolves much more slowly near solar metallicity than is predicted by numerical evolution models (e.g., Timmes et al. 1995). This result is in agreement with astronomical observations (Gay & Lambert 2000) and might indicate an important AGB star contribution to the Mg isotope budget at relatively low metallicity. Ti isotopes in a few corundum grains also seem to reflect Galactic chemical evolution (Choi et al. 1998; Hoppe et al. 2003). Finally, Nittler & Cowsik (1997) used the inferred masses and metallicities of the parent stars of Group 3 grains to put bounds on the age of the Milky Way. The age they derived (14 Gyr) has large systematic uncertainties, but is reasonably consistent with other estimates and was derived in a fundamentally new way.

1.5 Presolar Supernova Grains

Figures 1.3 and 1.4 show that SiC "X" grains form an isotopically distinct class, in many ways complementary to the mainstream. It is now well established that these grains, as well as a major fraction of presolar graphite grains and all known presolar Si_3N_4 grains, all condensed within the expanding ejecta of supernova explosions (see review by Amari & Zinner 1997). Although a special Type Ia supernova model was proposed to explain many of the observed isotopic signatures (Clayton et al. 1997), it now appears most likely that the grains formed in Type II events (SN II). Many of the observed isotope signatures (e.g., ^{28}Si and ^{15}N excesses, a wide range of $^{12}C/^{13}C$ ratios, and extremely high inferred $^{26}Al/^{27}Al$ ratios (up to 0.6) are qualitatively consistent with models of SN II nucleosynthesis. The smoking gun for a supernova origin (Clayton 1975) is the inferred presence of the extinct nuclides ^{44}Ti (Fig. 1.7) and ^{49}V, seen in the grains as ^{44}Ca and ^{49}Ti (Nittler et al. 1996; Hoppe & Besmehn 2002), since these short-lived nuclei are only synthesized within supernovae. Excesses in ^{22}Ne can in some cases also be attributed to supernova production of ^{22}Na (Nichols et al. 2004). The inferred $^{44}Ti/^{48}Ti$ ratios in ^{44}Ca-enriched grains are consistent with theoretical expectations for SN II. Moreover, they correlate with $^{29,30}Si/^{28}Si$ ratios (Fig. 1.7), consistent with the grains containing material from the innermost ^{28}Si-rich zones, where ^{44}Ti is synthesized during an α-rich freeze-out. Detailed discussions of the isotopic signatures of supernova grains can be found in Amari & Zinner (1997), Travaglio et al. (1999), and Hoppe et al. (2000). Of key importance is the observation that the grains carry the isotopic signatures of different mass zones of the parent supernovae. This indicates

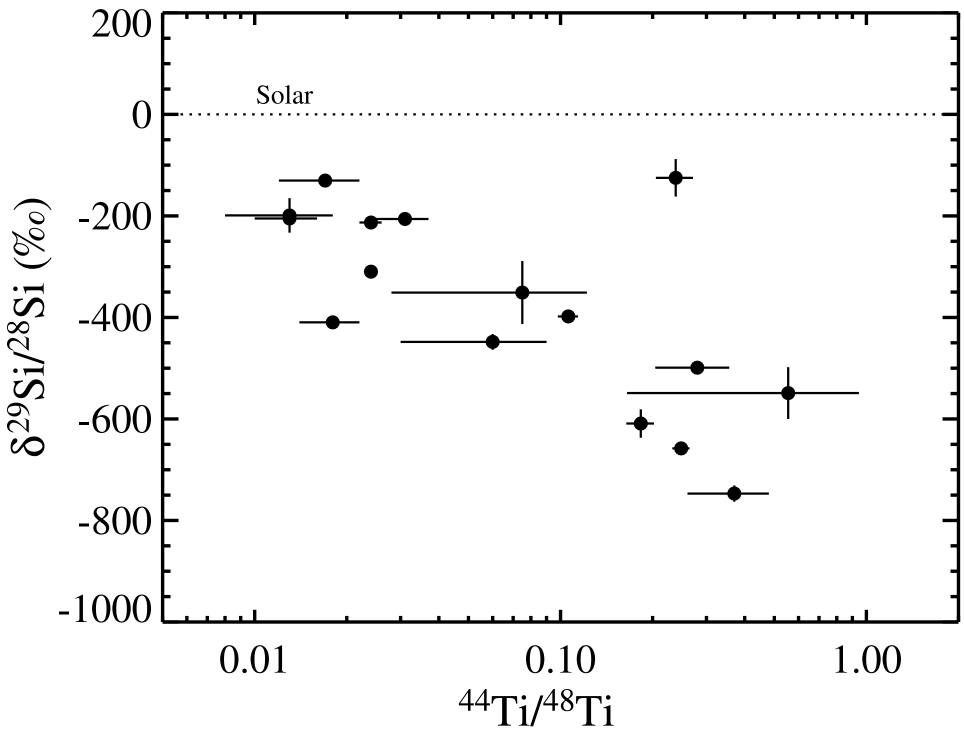

Fig. 1.7. δ-^{29}Si ratios plotted versus inferred ^{44}Ti/^{48}Ti ratios for presolar SiC X grains (Nittler et al. 1996; Hoppe et al. 2000; Besmehn & Hoppe 2004). The presence of extinct ^{44}Ti in the grains proves a supernova origin.

the need for selective mixing of material from different layers, with profound implications for processes of gas transport and dust nucleation and growth in SN II ejecta (see § 1.5.2).

1.5.1 New Nucleosynthesis Information from Supernova Grains

As we have already discussed for mainstream SiC grains and presolar oxides, the supernova grains provide new information about nucleosynthesis within their formation environments. We discuss two illustrative examples.

1.5.1.1 Nitrogen-15

Nitrogen-15 is produced in SN II models primarily by neutrino spallation from ^{16}O in inner shells (Woosley & Weaver 1995). Supernova presolar grains have large ^{15}N excesses coupled with isotopic signatures of H burning (very high ^{26}Al/^{27}Al ratios, low ^{12}C/^{13}C ratios in some grains). This has posed a significant problem for quantitatively explaining the grain data, since H-burning produces abundant ^{14}N-rich N. The measured ^{15}N/^{14}N ratios are higher than can be readily explained by SN II mixing models, if it is assumed that C $>$ O for SiC or graphite condensation (Travaglio et al. 1999; Hoppe et al. 2000). Relaxing the C $>$ O restriction might allow for SiC formation in the ^{15}N- and O-rich inner zones (see § 1.5.2), possibly removing the problem. Alternatively, recent observational evidence (Chin et al. 1999) supports much higher bulk ^{15}N production by SN II than is predicted by current models. Moreover, massive star models incorporating rotation indicate that abundant ^{15}N could be made hydrostatically when protons from the H shell get mixed into the He shell (Langer et al. 1998). These recent theoretical and observation results could help reduce the difficulty in explaining the grain data, if the ^{13}C and ^{15}N in the grains in fact originated in the He shell.

1.5.1.2 Neutron-Burst Nucleosynthesis

Pellin et al. (2000) and Davis et al. (2002) reported Sr, Zr, Mo, Ba, and Fe isotopic compositions for several SiC X grains. Figure 1.2 shows the Mo isotope pattern for one X grain. This ^{95}Mo- and ^{97}Mo-enriched pattern is clearly distinct, not only from the *s*-process signature of the plotted mainstream grain, but also from the composition expected for the *r*-process, associated with SN II. Similar results are found for the other studied elements. Meyer, Clayton, & The (2000) showed that the X grain Zr and Mo isotope signatures could be explained by a new kind of "neutron burst" nucleosynthesis, occurring when a large flux of neutrons are released as the shock wave passes through the He shell of the SN II. Although this process is but a minor contributor to bulk Mo isotopes in comparison with the *s* and *r* processes, unknown reasons apparently exist for its preferential enrichment of SiC X grains. This result clearly shows the power of presolar grains for probing conditions and processes in local regions of SN II ejecta.

1.5.2 Condensation Problems within Supernovae

The supernova interior offers a unique laboratory for condensation physics. It guarantees that chemistry must begin with gaseous atoms, with no trace of previous molecules or grains. The nucleosynthesis problems posed by isotopic ratios in supernova grains can not be decoupled from physical questions about their condensation. It is not physically clear whether the mixing apparently required by the isotope data represents molecular mixing in the young remnant or transport of a growing grain from one composition zone into another. Intimately related is an elemental composition question, namely, whether the bulk C abundance must exceed the O abundance to condense C-rich dust within supernovae. If the C/O ratio is less than unity, equilibrium condensation dictates that all carbon is locked up in the stable CO molecule, precluding condensation of SiC and graphite. Travaglio et al. (1999) performed SN II mixing models, assuming that the mixing is molecular, prior to condensation, and that only material with C $>$ O could support graphite formation. In contrast, Clayton, Liu, & Dalgarno (1999) and Clayton, Deneault, & Meyer (2001) argued that radioactive destruction of CO molecules removes the C $>$ O requirement. They advanced a kinetic theory of graphite growth and calculated its consequences in detail after advocating a specific nucleation model. They further argued that small graphite particles in a hot gas of C and O will associate with C faster than they will be oxidized by the more abundant O. Thus, even though oxidation would be the ultimate end, given adequate time, the expansion will terminate the chemistry after about two years, with large graphite grains remaining. This theory built on the finding that a large mass of CO was indeed destroyed in supernova 1987A (Liu & Dalgarno 1994, 1995). However, this theory is far from complete. For example, it is not known whether grains of TiC and Fe-Ni metal would condense with the graphite in this scenario, as required by observations of supernova graphites (Croat et al. 2004).

Similar but different questions surround the condensation of supernova SiC, the X grains of Figures 1.3 and 1.4. It seems plausible that radioactive liberation of free C atoms from CO molecules could also facilitate the condensation of SiC in O-rich gas. Although a kinetic route to SiC condensation has not been laid out, Deneault, Clayton, & Heger (2003) have formulated a physical description of the ejecta, which appears promising to explain many properties of SiC X grains. In their model, a reverse shock from increasing ρr^3 in the H envelope builds, after about a month, a dense shell in an inner layer of the supernova. Silicon-28 rich SiC condenses in this layer, allowed by the enhanced density and the radioactive dissociation of CO and SiO molecules. Mixing of a new type during condensation occurs if the reverse shock from the presupernova wind arrives at the condensation zone between six months to a year, because that shock slows the gas and forces the partially condensed SiC grains to propel forward through the decelerating gas into regions with different isotopic compositions. After $\sim 10^3$ yr, a third strong reverse shock from the ambient ISM propels SiC grains forward through overlying ejecta at high speed (typically 500 km s^{-1}) such that other atoms are implanted into the grains, perhaps accounting for some trace isotopes, as suggested for Mo and Fe (Clayton et al. 2002). Velocity-mixing instabilities prior to these reverse shocks will result in a spectrum of overlying column compositions so that grains from different supernovae (or different regions of a given supernova) could have a diversity of compositions.

Such physical modeling suggests that X grains provide new sampling techniques of the detailed physical structures of supernova ejecta, but it remains to be seen if the ideas will withstand the scrutiny of more detailed models. Especially threatening is the possibility of SiC destruction by sputtering, oxidation, or a shock that is too strong from the presupernova wind (Deneault et al. 2003). A key point becomes the spectrum of wind masses that accompany Type II supernovae, for those masses provide the mass of the shock-generating obstacle. Especially needed are 2-D and 3-D hydrodynamic calculations of the reverse shocks and of instability-induced velocity mixing by the primary outgoing shock and a detailed study of molecular mixing to ascertain the degree to which microscopic mixing can be called upon during the first year. Additionally, detailed microstructural investigations of supernova grains by analytical electron microscopy are likely to provide a great deal of information about their formation processes (e.g., Croat et al. 2004).

What now appears certain, despite these many open questions, is that supernova grains studied by isotopic analysis will provide, through details of condensation chemistry, a new sampling spectrum of young supernova interiors, just as have gamma-ray lines and hard X rays. The radioactivity that causes each type of sampling raises fundamental chemical questions as well.

1.6 Conclusions

Presolar stardust grains have been identifiable by cosmochemists because their several isotopic ratios are too unusual to be explained by any origin other than within ejected stellar matter prior to its mixing with the ISM. The high precision of this isotopic data, higher than traditionally obtainable by astronomical spectroscopy, supports detailed questioning about the stars that produced the grains. Stellar evolution theory, nucleosynthesis theory, and chemical evolution theory for the Galaxy must each be called upon to interpret the solid samples of the exploding or mass-losing stars. New insights into each have been demanded or suggested by the precise isotopic ratios of not only individual grains but of families of grains whose relatedness is established by iterative procedures. Grains grouped initially by purely isotopic criteria are examined with standard techniques of science: hypotheses about stars of origin are iterated with computed stellar models; hypotheses about trends within related grains are iterated with astrophysical models of trend-determining evolution; hypotheses about nuclear reactions in stars are iterated with the nuclear laboratories. By these processes of classification, posed hypotheses, and challenge by astrophysical theory, an increasingly refined scientific corpus has been established. This may be likened to the historic challenge of the spectral types of stars and of their positions and populations in the Hertzsprung-Russell diagram.

Stardust grains have also opened purely chemical frontiers of stellar physics. It is necessary to come to grips with details of the condensation processes and of physical aspects of stars that had not been previously demanded. How well mixed is the AGB dredge-up matter in the envelope? How is its wind initiated and does its stardust select overdense epochs of mass loss? How does the radioactive dissociation of the CO molecule alter chemical consequences of the C/O ratio? How do reverse shocks in supernovae enable supernova grains to grow and survive with the isotopes we find within them? How and when does supernova matter mix? These questions are but examples of many new cosmochemical frontiers that automatically open by the ability to study grains of stardust in detail in the laboratory.

Each presolar stardust grain is a measurement of some unknown star, a measurement undeveloped until cosmochemists isolate that grain and investigate it in the laboratory. Perhaps their laboratory techniques may justifiably be called new telescopes. They are the only telescopes capable of observing Galactic stars that died more than 5 Gyr ago. Understanding the early structure and chemical evolution of our Galaxy will increasingly rely on their high-precision message.

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