CSIRO PUBLISHING

Publications of the Astronomical Society of Australia, 2009, **26**, 278–283 www.publish.csiro.au/journals/pasa

Cosmic-Ray Exposure Ages of Large Presolar SiC Grains

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Received 2008 November 18, accepted 2009 February 16

Abstract: Among presolar SiC grains found in the Murchison carbonaceous meteorites (average size less than 0.5 μm) are very large grains, ranging in size up to 50 μm. We interpret ^{6}Li excesses measured in eight of these grains as being the result of spallation reactions by Galactic cosmic rays during the time the grains spent in the interstellar medium before their incorporation into the meteorite. Derived interstellar exposure ages range from 40 My to 1 Gy, the highest values being consistent with theoretical expectations of interstellar grain lifetimes. Although six grains have almost identical C and Si isotopic compositions, their exposure ages are very different. This observation, combined with low trace element contents, and unusual grain sizes, raises fundamental questions about their stellar sources.

Keywords: stardust — circumstellar matter — cosmic rays — isotopic ratios — ISM: kinematics and dynamics — stars: AGB stars — stars: winds, outflows

1 Introduction

Primitive meteorites, interplanetary dust particles, micrometeorites, and comets contain tiny grains of stardust (e.g. Clayton & Nittler 2004; Lodders & Amari 2005; Zinner 2007). These small grains condensed in the expanding atmospheres of red-giant branch and Asymptotic Giant Branch (AGB) stars and in the ejecta of supernovae, survived travel in the interstellar medium (ISM), and were included in the molecular cloud from which our Solar System formed. Their stellar origin is demonstrated by their isotopic compositions, which reflect those of their stellar sources and are completely different from that of the Solar System. These presolar grains include carbonaceous phases such as diamond, silicon carbide, and graphite, and oxygen-rich phases such as silicates, corundum (Al_2O_3), and spinel ($MgAl_2O_4$) (Zinner 2007).

Among presolar grains, SiC has been studied in most detail. The reason is that, because of its chemically refractory nature, SiC can be fairly easily separated from meteorites by physical and chemical processing, and although the average size of SiC grains is less than 0.5 μm (Amari, Lewis & Anders 1994), many grains larger than 1–2 μm are available for study. Another reason is that SiC has relatively high contents of minor and trace elements, making multi-element isotopic analysis of single grains possible. Most presolar SiC studied thus far has been extracted from the Murchison meteorite because large amounts of this meteorite are available. Furthermore, for still unknown reasons, the average grain size of SiC from Murchison is larger than that of SiC from other meteorites (Russell et al. 1997). Of the two separation series of Murchison, the K-series and L-series, undertaken at the University of Chicago (Amari et al. 1994), the LS and LU fractions of the latter contain unusually large grains, ranging up to 50 μm in size (Figure 1).

Previous studies of Murchison LS+LU grains have included isotopic analysis of C, N, Si, Al–Mg, and Ti, Raman spectroscopic measurements and the determination of trace element abundances (Ireland, Zinner & Amari 1991; Virag et al. 1992; Amari et al. 1995). They indicate that the large LS+LU grains are unique. Isotopic compositions show clustering for C and Si (Virag et al. 1992) and even for Ti (Ireland et al. 1991). Concentrations of N, Al, Ti and other trace elements in most LS+LU grains are much lower than those in smaller SiC grains (Virag et al. 1992; Hoppe et al. 1994; Amari et al. 1995). Another distinguishing characteristic of the largest LS+LU grains is not only their size but also their morphology. While most of the smaller SiC grains show euhedral crystal features (e.g. Hoppe et al. 1994; Bernatowicz et al. 2003), almost all large LS+LU grains lack them. Instead, they have blocky appearances with frequently smooth surfaces (see Figure 1) and often look as if they are fragments of even larger pieces. A remaining puzzle is why these large grains showed up only in the L-series separation but in no other SiC-rich separations from Murchison (Nittler & Alexander 2003; Marhas, Hoppe & Ott 2007). Astronomical observations indicate that circumstellar grains are a few hundred nanometers in size (Mathis, Rumpl & Nordsieck 1977; Mathis 1990) and most presolar SiC grains are in this size range. Astronomers consider grains of a few μm

10.1071/AS08037 1323-3580/09/03278

in size 'bricks'; the large LS+LU grains are thus true boulders.

In order to obtain more information about the large SiC grains and their stellar sources we started a series of new investigations of the Murchison LS+LU grains, including noble gas analysis (Heck et al. 2008; Ott et al. this volume) and isotopic measurements of the heavy elements Ba, Eu, Gd, and W (Avila et al. 2008a,b, 2009). Here we report Li isotopic data and the determination of exposure ages in the ISM.

2 Lithium Isotopes and Cosmic-Ray Exposure Ages

One of the important pieces of information one wishes to obtain about presolar grains is their age, i.e. the time they condensed in stellar atmospheres. Grain ages are not only important for obtaining information on the history of the parent stars of the grains but also for setting constraints on interstellar processes, such a grain collisions and grain sputtering in supernova shocks. There are several obstacles that stand in the way of radiometric dating: the grains are small, their original isotopic compositions are anomalous, and many elements used for age dating, such as K and Rb, are rather volatile and not likely to be included into SiC grains. It still has to be seen whether conventional dating with radionuclides (U-Pb?) is possible (Avila et al. 2007). An alternative is to determine residence times in the ISM from the contents of cosmogenic (i.e. cosmic-ray-produced) nuclides in the grains. Tang & Anders (1988) and Lewis, Amari & Anders (1994) used ^{21}Ne excesses, believed to be of cosmogenic origin, to obtain cosmic-ray (CR) exposure ages in size fractions of bulk SiC from Murchison. Inferred ages ranged up to 133 My, less than theoretical estimates of interstellar lifetimes between 500 My and 1.5 Gy (Jones et al. 1997). However, Ott & Begemann (2000) measured the recoil range of ^{21}Ne nuclei produced by spallation reactions in SiC and found it to be much larger than the range assumed by Lewis et al. (1994). This means that recoil losses would be much larger, and Ott & Begemann (2000) proposed that the ^{21}Ne in presolar SiC was of stellar origin. Recoil ranges of Xe are shorter, and Ott et al. (2005) set a limit of ~50 My on CR-exposure ages from cosmogenic ^{126}Xe in SiC. The large size of LS+LU grains and their low trace element concentrations offer a unique opportunity to determine CR exposure ages from cosmogenic Li: recoil Li produced by spallation on C might be retained in the grains, and low indigenous Li contents might allow the detection of cosmogenic Li.

With the NanoSIMS ion microprobe we measured the C, Si, Li and B isotopic ratios of 9 large LS+LU grains. Carbon and Si were measured as negative secondary ions produced by bombarding the grains with a Cs$^+$ beam, Li and B as positive secondary ions produced with an O$^-$ primary beam. Because Li and B have very low concentrations, it is important to avoid any contamination of these elements. Before isotopic measurements, we obtained secondary ion images of the grains to determine areas without any obvious contamination. Measurements in multidetection (the Li and B isotopes and ^{30}Si) were made on 'clean' areas by rastering the primary beam over areas a few μm wide. Lithium concentrations (Figure 2) are lower than those of B, resulting in clear cosmogenic signatures. We therefore concentrate only on the Li isotopic data.

Table 1 and Figure 2 show Li isotopic ratios measured in the LS+LU grains. The ratios are expressed as δ-values, deviations from the terrestrial ratio of 0.0829 in permil (‰). Eight of the grains have ^{6}Li excesses outside of 2σ errors. The only reasonable explanation for these ^{6}Li excesses is that they are of cosmogenic origin. Lithium-6 is a fragile nuclide and is completely destroyed

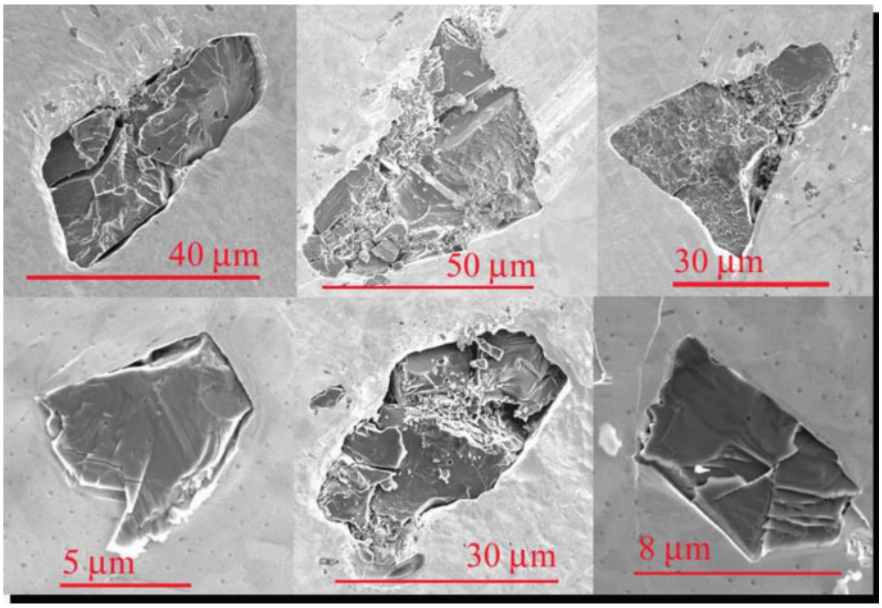

Figure 1 Scanning electron micrographs of large presolar SiC grains from the L-series fractions LS+LU. The grains were pressed into a gold foil for ion probe analysis, resulting in some of the cracks in several of the grains.

in stellar environments. Under special circumstances, ^{7}Li can be produced by the Cameron-Fowler mechanism (Cameron & Fowler 1971) via ^{3}He(α,γ)^{7}Be(e^{-},ν)^{7}Li in red giant and AGB stars. Thus, if there is any stellar Li left in the SiC grains, it must be mostly ^{7}Li. Both Li isotopes are produced in approximately equal amounts by collisions of cosmic rays with interstellar atoms. While all ^{6}Li is produced in this way, most of ^{7}Li is produced by big bang nucleosynthesis and some in AGB stars. In order to determine the amount of cosmogenic Li in the SiC grains, we assumed the measured Li to be a mixture of terrestrial and CR-produced Li and the grains to contain no stellar Li. Because Li is a fairly volatile element, it is unlikely that it condensed into SiC. The Li with terrestrial isotopic composition is most likely contamination. The possible presence of some stellar ^{7}Li means that the cosmogenic Li contents we obtain are lower limits.

Derivation of CR exposure ages requires knowledge of (1) the production rates of the Li isotopes by Galactic cosmic rays; (2) the CR flux and its energy distributions in the ISM; (3) the retention of spallation-produced Li in the grains. For the production rates we used the calculations by Reedy (1989), which are based on his earlier estimate of the Galactic cosmic ray flux (Reedy 1987). For determining the Li retention, we used the SRIM code (Ziegler 2004) to calculate ranges of spallation recoil ^{6}Li for a distribution of recoil energies derived from the momentum distribution given by Greiner et al. (1975). These authors studied the breakup of ^{12}C and ^{16}O projectiles into lighter nuclei, among them ^{6}Li. Because the analysed grains have irregular shapes (Figure 1), we approximated the grains by spheres with diameters being the geometric mean of the dimensions inferred from the secondary electron microscope images. More details about the calculation of the production rates and the retention of recoil ^{6}Li, as well as a discussion of errors on exposure ages resulting from uncertainties of the CR flux and the irregular shapes of the grains, can be found in Gyngard et al. (2009).

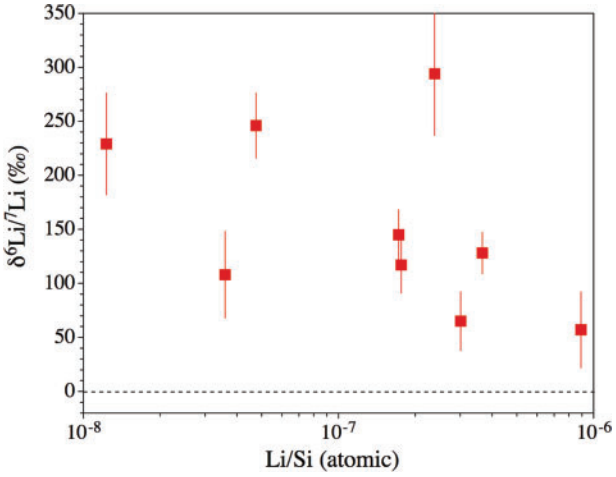

Figure 2 Lithium isotopic ratios measured in individual LS+LU grains are plotted against the atomic Li/Si ratio. Errors are 1σ.

Cosmic ray exposure ages for the LS+LU grains are shown in Figure 3. As can be seen, the ages cover a large range. Most are larger than the limit set by ^{126}Xe (Ott et al. 2005) and the highest values overlap with estimates for grain lifetimes in the ISM (Jones et al. 1997).

3 Discussion

The large range of CR exposure ages raises several important questions. The C and Si isotopic ratios of the grains are in the range displayed by the so-called mainstream SiC grains (e.g. Zinner 2007) and indicate that the grains originated in the winds from AGB stars. Different ages imply that the grains have different parent stars that reached the thermally pulsing (TP) AGB phase at different times before Solar System formation. During the TP-AGB phase, ^{12}C synthesised in the He-burning shell is mixed into the originally O-rich envelope, increasing the C/O ratio until the star becomes a carbon star. Carbon-rich conditions (C > O) are required for carbonaceous phases such as SiC to condense (Lodders & Fegley 1997). On the other hand, the very large LS+LU grains are extremely rare in Murchison. In addition, under typical conditions in carbon star atmospheres, expected grain sizes are much smaller (Bernatowicz et al. 2005), and it is still not clear

Table 1. Isotopic compositions and GCR exposure ages of LS+LU SiC grains

Diameter[a] (μm)	^{12}C/^{13}C	δ^{29}Si/^{28}Si (‰)	δ^{30}Si/^{28}Si (‰)	δ^{6}Li/^{7}Li (‰)	Li/Si (atomic)	^{6}Li retention (%)	Corrected age[b] (My)
37	47.5 ± 0.3	45 ± 4	73 ± 7	294 ± 57	2.4×10^{-7}	74	860
8	55.7 ± 0.4	70 ± 4	75 ± 7	128 ± 19	3.7×10^{-7}	44	980
23	48.0 ± 0.3	51 ± 4	74 ± 7	229 ± 47	1.2×10^{-8}	65	40
43	47.4 ± 0.3	36 ± 4	57 ± 7	108 ± 40	3.6×10^{-8}	77	50
5	47.5 ± 0.3	57 ± 4	71 ± 7	145 ± 23	1.7×10^{-7}	35	660
18	47.9 ± 0.3	55 ± 4	65 ± 7	117 ± 26	1.8×10^{-7}	61	310
20	47.9 ± 0.3	57 ± 4	66 ± 7	246 ± 30	4.8×10^{-8}	63	170
8	90.9 ± 0.3	15 ± 3	21 ± 7	65 ± 27	3.0×10^{-7}	43	420
5	47.4 ± 0.3	54 ± 4	76 ± 7	57 ± 35			

Errors are 1σ.

[a] Geometric mean of the three dimensions of the grains.

[b] Relative errors are ~50% and absolute errors are probably at least a factor of 2. For a more detailed discussion see Gyngard et al. (2009).

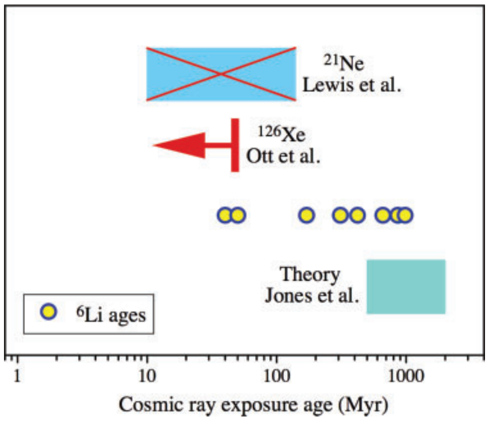

Figure 3 Cosmic ray exposure ages of LS+LU grains are compared to previous estimates and to theoretical interstellar grain lifetimes. The ages obtained by Lewis et al. (1994) were invalidated by subsequent measurements of recoil ranges (Ott & Begemann 2000).

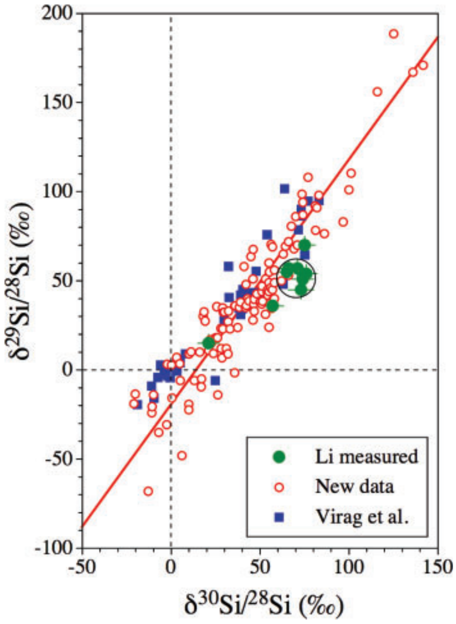

Figure 4 Silicon isotopic ratios (expressed as δ-values) of LS+LU SiC grains analysed for their Li isotopic compositions are plotted together with those of other LS+LU grains. Error bars of the Li-analysed grains are 1σ. Six grains with essentially identical Si isotopic compositions are encircled. Among these six grains we did not obtain an age for the grain with the highest δ^{30}Si value (see Table 1). The red line is the mainstream correlation line (Zinner et al. 2007).

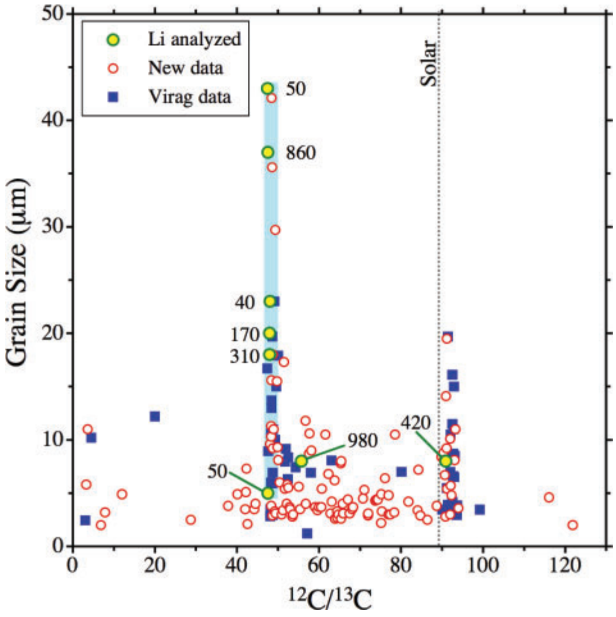

Figure 5 Grain sizes of LS+LU grains are plotted against their C isotopic ratios. The numbers next to the grains analysed for their Li isotopic ratios are the inferred CR exposure ages in My. The largest grains have ^{12}C/^{13}C ratios close to 46 (light blue area). The five grains >15 µm with known ages have the same Si isotopic ratios. Some other unusually large grains have C isotopic ratios close to the solar ratio of 89.

how such large SiC grains can form. These considerations and clustering of the isotopic compositions of LS+LU grains led to the suggestion that grains in an isotopic cluster originated from a single parent star (Virag et al. 1992). The isotopic ratios of the SiC analysed for their Li isotopic compositions are not random. Figure 4 shows their Si isotopic ratios and those of all previously (Virag et al. 1992) and newly analysed LS+LU grains. New C and Si isotopic data are reported in Heck et al. (2009) and include unpublished data obtained in connection with the analysis of heavy elements in LS+LU grains (Avila et al. 2008a,b, 2009). Six of the grains whose Li isotopes were measured have identical Si isotopic ratios within errors (δ^{29}Si/^{28}Si = 53.2‰ and δ^{30}Si/^{28}Si = 70.8‰).

These six grains have also identical C isotopic ratios within errors (^{12}C/^{13}C = 46.1). Five of them, for which we could determine CR exposure ages, are plotted in Figure 5 together with the other LS+LU grains. They are the largest grains analysed. The reason that grains analysed for Li have similar isotopic compositions is that large grains and grains with smooth surfaces were preferentially selected and such grains seem to have similar isotopic compositions (see also Virag et al. 1992). It would be highly desirable to perform Li isotopic ratio measurements also on grains with other C and Si isotopic compositions. However, most of these grains are smaller and it remains to be seen whether meaningful Li isotopic data can be obtained on such grains.

We are left with the puzzle of how SiC grains with essentially identical C and Si isotopic ratios can have different CR exposure ages. Several grains with ^{12}C/^{13}C ratios of around 46 also give different ^{3}He and ^{21}Ne exposure ages, although, on average, these ages are somewhat lower than those derived from Li (Heck et al. 2009; Ott et al. this volume). As already mentioned, the C and Si isotopic ratios of almost all LS+LU grains indicate an origin in AGB stars. However, only a small fraction of AGB stars during the TP stage are expected to have the same C and Si isotopic compositions. Furthermore, the isotopic ratios of these two elements in AGB stars are largely determined by independent processes. During the main sequence phase of a star, H burning via the CNO cycle lowers the ^{12}C/^{13}C ratio in interior layers and, after

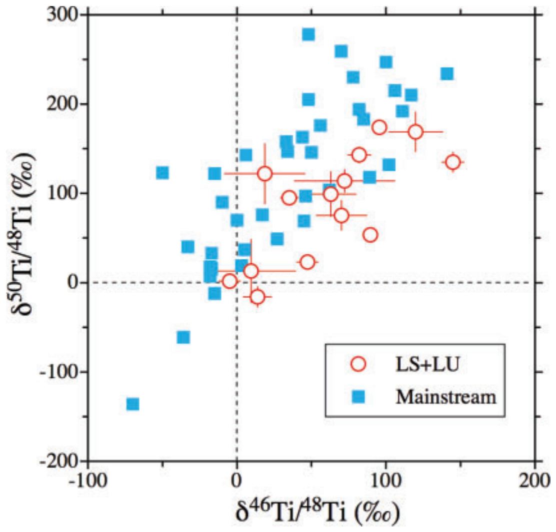

Figure 6 Titanium isotopic ratios (expressed as δ values) of LS+LU mainstream SiC grains (Ireland et al. 1991) are compared with those of smaller mainstream grains labelled 'mainstream' (Hoppe et al. 1994; Alexander & Nittler 1999). Error bars of the LS+LU grains are 1σ.

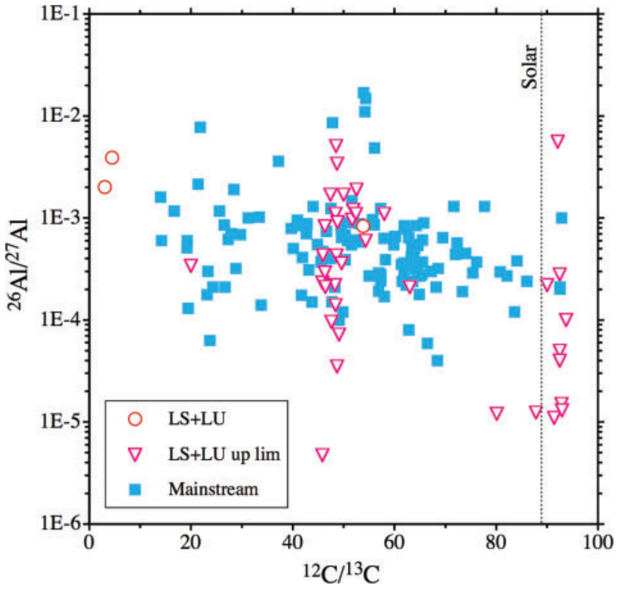

Figure 7 Inferred ^{26}Al/^{27}Al ratios of LS+LU grains (Virag et al. 1992; and unpublished data from this laboratory) and of mainstream grains are plotted against the grains' ^{12}C/^{13}C ratios. The triangles indicate upper limits.

the first (and second) dredge-up, mixing of interior zones into the envelope, also at the surface of the star (e.g. Boothroyd & Sackmann 1999). Shell He burning during the TP-AGB phase produces ^{12}C, which is mixed into the envelope by third dredge-up (TDU) episodes (Busso, Gallino & Wasserburg 1999) and results in an increase in the ^{12}C/^{13}C and C/O ratios until the star becomes a carbon star. Predicted final ^{12}C/^{13}C ratios depend on stellar mass, metallicity, and mass loss rate (Zinner et al. 2006). In addition, extra mixing or cool bottom processing (Nollett, Busso & Wasserburg 2003) has been invoked to explain lower-than-expected ^{12}C/^{13}C ratios in certain SiC grains and in RGB stars (Charbonnel 1995; Wasserburg, Boothroyd & Sackmann 1995). Different combinations of all these parameters, especially for different degrees of the TDU, could result in the same ^{12}C/^{13}C ratio of the envelope, but this is highly unlikely.

In contrast to C, the range of Si isotopic ratios in SiC grains cannot be explained by nucleosynthesis in their parent stars, but in addition reflects the original Si isotopic compositions of the parent stars (Clayton et al. 1991; Alexander 1993). Neutron capture in the He intershell during the TP-AGB phase and mixing into the envelope by the TDU leads to ^{29}Si and ^{30}Si excesses and would move the isotopic composition mostly to the right in the Si 3-isotope plot of Figure 4. The fact that the grains analysed for Li plot to the right of the mainstream correlation line indicates that their Si experienced more neutron capture than average mainstream grains (Lugaro et al. 1999; Zinner et al. 2007). Grains of type Y also plot to the right of the line but they are characterised by ^{12}C/^{13}C > 100 (Hoppe et al. 1994; Amari et al. 2001), a signature not shared by the LS+LU grains. In addition, most Y grains have lower ^{29}Si/^{28}Si ratios. Figure 6 shows ^{50}Ti/^{48}Ti ratios of LS+LU grains as well as mainstream grains plotted against their ^{46}Ti/^{48}Ti ratios. Titanium-50 is the Ti isotope most affected by neutron capture. The fact that, on average, the ^{50}Ti excesses of the LS+LU grains are smaller than those of smaller mainstream grains indicates that their Ti received less neutron exposure or that less material had been mixed into the envelope by the TDU. Unfortunately, we do not have the Ti isotopic compositions of the LS+LU grains analysed for Li.

Another indication that the parent stars of LS+LU grains experienced only limited TDU comes from ^{26}Al. While most mainstream grains show evidence for the initial presence of ^{26}Al in the form of large ^{26}Mg excesses, clear evidence for ^{26}Al was obtained in only three LS+LU grains, two of which are SiC grains of type AB (^{12}C/^{13}C <10). For the other LS+LU grains only upper limits were obtained (Figure 7). Aluminum-26 in AGB stars is produced in the H-burning shell and brought to the surface by the TDU. Cool bottom processing has been invoked for additional ^{26}Al production (Nollett et al. 2003; Zinner et al. 2007). The reason that many of the upper limits are fairly high is that Al concentrations and Al/Mg ratios in LS+LU grains are much lower than those in smaller SiC grains from the K series (Hoppe et al. 1994; Amari et al. 1995).

The low concentrations of heavy elements such as Zr and Ba in LS+LU grains (Virag et al. 1992; Amari et al. 1995) could possibly be attributed to little TDU of these s-process elements. However, the low concentrations of minor and trace elements such as Al, Ti, and V cannot be explained in this way, but must rather reflect special formation conditions. It is tempting to hypothesise that the physical and chemical conditions that led to the formation of large SiC grains also were responsible for their extremely low trace element concentrations. Formation of SiC grains of the sizes studied here not only requires high gas pressure and C/O ratios not much higher than 1.05 (Lodders & Fegley 1995) but also formation times of hundreds of years (Bernatowicz et al. 2005). Low trace element concentrations imply relatively low condensation temperatures (Lodders & Fegley 1997). These conditions are generally not met by stellar winds from AGB stars (Sedlmayr & Krüger 1997). We would expect that such conditions are extremely rare and it is therefore even more puzzling that they occurred in several stars at different times in the past, moreover in stars that in some cases had essentially identical C and Si isotopic compositions. If there is some intrinsic connection between these unique C and Si isotopic compositions and the unique conditions for the formation of jumbo grains, we do not have an explanation.

A possible solution to this conundrum is that the large grains with identical C and Si isotopic ratios actually formed some ≥860 My before the solar system (the largest CR exposure age of these grains, see Figure 5) from a single AGB star. Subsequently, different grains from this star were shielded from Galactic cosmic rays for different amounts of time so that their CR exposure ages are different. The problem with this scenario is that we simply do not know how shielding by several meters of material can be accomplished in the ISM in a way that these grains still arrived in the Solar System and were together incorporated in the Murchison meteorite. Another complication is that these large grains were found only in the piece of Murchison that underwent the L-series separation and not in other pieces.

At present, all possible explanations, either several parent stars with identical isotopic compositions or shielding of grains in the ISM, appear very unlikely. We can only hope that further studies, such as isotopic analysis of other elements, will help in solving this puzzle.

Acknowledgments

We thank Frank Stadermann and Tim Smolar for help with the NanoSIMS and are grateful to Roy Lewis for providing the LS+LU grains. Detailed comments by a reviewer have improved this paper. This work was supported by NASA grant NNX08G71G.

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