



Heavy elements in presolar grains: constraints on conditions in asymptotic giant branch stars

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Presolar SiC grains come from a variety of kinds of stars, but the most common type, the mainstream grains, are believed to have formed in the outflows of low mass, carbon-rich asymptotic giant branch (AGB) stars. Measurements of the isotopic composition of the *s*-process elements Sr, Zr, Mo, Ru and Ba in individual mainstream SiC grains allow constraints of the range of conditions used in stellar models of AGB stars.

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1. Introduction

Presolar silicon carbide (SiC) grains were discovered to be present in primitive meteorites nearly twenty years ago [1]. They formed in carbon-rich ($C/O > 1$) outflows from stars, survived potentially destructive processes in the interstellar medium, during solar system formation, during parent body metamorphism, during atmospheric entry and during laboratory separation of the grains from their host meteorites. The mainstream grains are the most common type of SiC and are believed to have come from low mass asymptotic giant branch (AGB) stars. This conclusion was initially reached on the basis of isotopic compositions of krypton and xenon in aggregates of presolar SiC and of C, N and Si in individual grains, but has been corroborated by many other isotopic measurements. Several recent reviews on presolar grains are available [2, 3, 4]. In the past few years, techniques have been developed to measure the isotopic compositions of heavy trace elements in individual presolar grains. The implications of these measurements for processes in AGB stars are reviewed here.

2. Analytical techniques for heavy elements in presolar grains

All measurements of isotopic compositions of presolar grains, whether in bulk or of individual grains, are made by mass spectrometry. The heavy elements present significant challenges, because there are many isobaric interferences. For example, consider the mass range from 90 to 100 atomic mass units (a.m.u.). There are overlaps between ^{92}Zr and ^{92}Mo ; ^{94}Zr and ^{94}Mo ; ^{96}Zr , ^{96}Mo and ^{96}Ru ; ^{98}Mo and ^{98}Mo ; and ^{100}Mo and ^{100}Ru . These isobaric interferences require mass resolutions ($M/\Delta M$) of 27,000 to 840,000, well beyond the capabilities of high sensitivity mass spectrometers. In order to get around this problem, three approaches can be used: (1) simply avoid elements or isotopes with isobaric interferences; (2) dissolve samples and partially or completely chemically separate the elements from one another; or (3) use lasers to resonantly ionize only the element of interest. The first approach was used in early measurements of Ba, Nd and Sm in aggregates of presolar SiC by secondary ion mass spectrometry [5] and in recent measurements of SiC-bearing presolar diamond aggregates by inductively coupled plasma (ICP) mass spectrometry [6]. The second approach has been used to measure the isotopic compositions of Sr [7], Ba [8,9], Nd [10], Sm [10] and Dy [11] in SiC aggregates by thermal ionization mass spectrometry. When elements are only partially separated from one another, the properties of elements can be used to reduce interferences. For example, before measuring Sr isotopes, the filament is heated to a temperature where Rb evaporates and Sr remains on the filament, reducing the isobaric interference between ^{87}Rb and ^{87}Sr [7]. The third approach has been applied to measurements of Sr [12], Zr [13], Mo [14], Ru [15] and Ba [16] in individual presolar SiC grains by laser desorption and laser resonant ionization mass spectrometry [17].

Analytical sensitivity is also an important issue. The abundances of heavy trace elements in presolar grains are low, even when overproduction due to the s-process in mainstream SiC grains is taken into account [18]. For this reason, all early measurements were made on aggregates of thousands of presolar grains [5-11]. With one exception, all measurements of

heavy element isotopic composition on individual SiC grains have been made by laser desorption laser resonant ionization mass spectrometry. The one exception is for Ba, for which there are no significant isobaric interferences and for which modern secondary ion mass spectrometers have sufficient sensitivity and precision [19].

All reported presolar grain isotopic analyses by laser resonant ionization mass spectrometry have been done on the CHARISMA instrument, built at Argonne National Laboratory several years ago [17]. In this instrument, a UV laser is introduced through a reflected light microscope, focused to less than 1 μm and used to ablate atoms from a SiC grain embedded in gold. Two or three lasers tuned to wavelengths appropriate to ionize one element are passed through the atom cloud above the sample. The photoions are then accelerated with an electrostatic field and passed through a time-of-flight mass spectrometer. The instrument has high sensitivity (a few percent of atoms removed from the sample are counted in the detector) and excellent rejection of isobaric interferences. For example, this instrument has been used to measure isotopic compositions of Zr, Mo or Ru without any nonresonant ionization of the other two elements.

3. Stellar models

Mainstream presolar SiC grains are thought to come from low mass AGB stars. At this stage of stellar evolution, hydrogen and helium burn alternatively in two shells in the narrow region between the convective H-rich envelope and the degenerate C-O core. During interpulse periods, hydrogen burns, gradually increasing the mass of the He-rich intershell. Eventually, conditions allow the triple- α reaction to occur, causing a thermal runaway or thermal pulse (TP), which convectively mixes the He intershell. The s -process occurs in the He intershell in low mass AGB stars, via two neutron sources. s -Process products and ^{12}C are mixed into the envelope after each TP when the convective envelope penetrates below the H-He discontinuity at the quenching of each thermal instability. This is known as third dredge-up (TDU). The major neutron exposure for the s -process occurs at low density over long times, via the $^{13}\text{C}(\alpha, n)^{16}\text{O}$ reaction during interpulse periods. The source of the ^{13}C is H introduced from the convective envelope into the top layers of the intershell after each TDU episode, which reacts with ^{12}C to form a ^{13}C pocket. The mechanism of H mixing is currently being debated, but observational evidence strongly supports the activation of the ^{13}C neutron source. A second neutron source, $^{22}\text{Ne}(\alpha, n)^{25}\text{Mg}$ is marginally activated during TPs, providing a high neutron density but short exposure. The latter neutron exposure is mostly felt at branch points in the s -process path. In this work, we compare isotopic compositions measured in presolar grains with those predicted by R. Gallino and colleagues. These are produced by a s -process network postprocessing calculation that uses the FRANEC stellar evolution code as an input. These calculations have been described in detail [20, 21, 22]. In the models we consider, we assume solar metallicity and allow the ^{13}C pocket efficiency to cover a range of a factor of 24, from twice the standard case, U2, to one-twelfth the standard case, D12. The standard case matches the solar system s -process distribution for a star of one-half solar metallicity [20]. Model predictions have been shown previously for Sr, Zr, Mo, and Ba for 1.5 and 3 M_{\odot} stars [21]; we show models for 2 M_{\odot} here, which are not significantly different from 1.5 M_{\odot} models.

4. Comparison of presolar SiC and AGB models

Presolar SiC grain data for Zr are compared with a $2 M_{\odot}$ model in Fig. 1. For each choice of ^{13}C pocket efficiency, there is a line with some symbols along it. The envelope has an initial isotopic composition of the solar system. With each TP with TDU, new s -process material is mixed into the envelope and the composition moves away from solar composition in the direction of lower $\delta(^{96}\text{Zr}/^{94}\text{Zr})$. The envelope initially has $\text{C}<\text{O}$ and will condense oxides and silicates. After the 12th TP with TDU in this model, enough C has been dredged up such that $\text{C}>\text{O}$ and graphite, SiC and TiC will condense [23]. With increasing ^{13}C pocket efficiency, lower and lower $\delta(^{96}\text{Zr}/^{94}\text{Zr})$ values are predicted, but at higher ^{13}C pocket efficiencies, it can be seen that $\delta(^{96}\text{Zr}/^{94}\text{Zr})$ turns around and begins to increase in later pulses. This is due to the action of the ^{22}Ne exposure during TPs, which activates the neutron capture at the s -process branch point at ^{95}Zr , which begins to produce ^{96}Zr . The models cover most of the range of data shown in Fig. 1, but do not explain the three SiC grains with the lowest $\delta(^{96}\text{Zr}/^{94}\text{Zr})$ values. Models for $3 M_{\odot}$ AGB stars tend to turn around earlier and produce $\delta(^{96}\text{Zr}/^{94}\text{Zr})$ values that match grains less well [21].

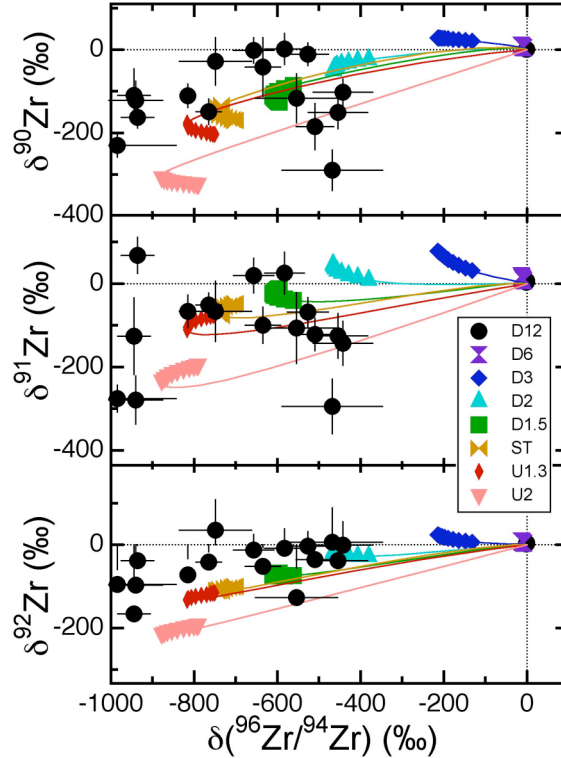


Fig. 1. Isotopic compositions of Zr in presolar SiC grains [13, 21] compared with those predicted for a $2 M_{\odot}$ AGB star with initially solar metallicity. Uncertainties are $\pm 2\sigma$.

Several conclusions can be reached by the comparison of grains with predictions for Zr isotopes: (1) mainstream SiC grains come from low-mass AGB stars, probably mostly from $1.5\text{--}2 M_{\odot}$ stars, a few from $3 M_{\odot}$ stars; (2) a variety of ^{13}C pocket efficiencies are needed to explain the range of grain data; (3) the grains with the lowest $\delta(^{96}\text{Zr}/^{94}\text{Zr})$ are difficult to explain and point out the need for improved measurement of Zr neutron capture cross sections.

Presolar SiC grains and $2 M_{\odot}$ AGB star predictions are compared for Mo in Fig. 2. The predictions mostly lie along straight lines rather than curves, because branching effects are much less strong for Mo than they are for Zr. The excellent match between grains and predictions is a measure of the quality of the neutron capture cross section measurements that are an input parameter into the predictions. The only consistent discrepancy is that the grains tend to lie below the predictions for $\delta(^{95}\text{Mo}/^{96}\text{Mo})$ vs. $\delta(^{92}\text{Mo}/^{96}\text{Mo})$, suggesting a need to remeasure the neutron capture cross section for ^{95}Mo .

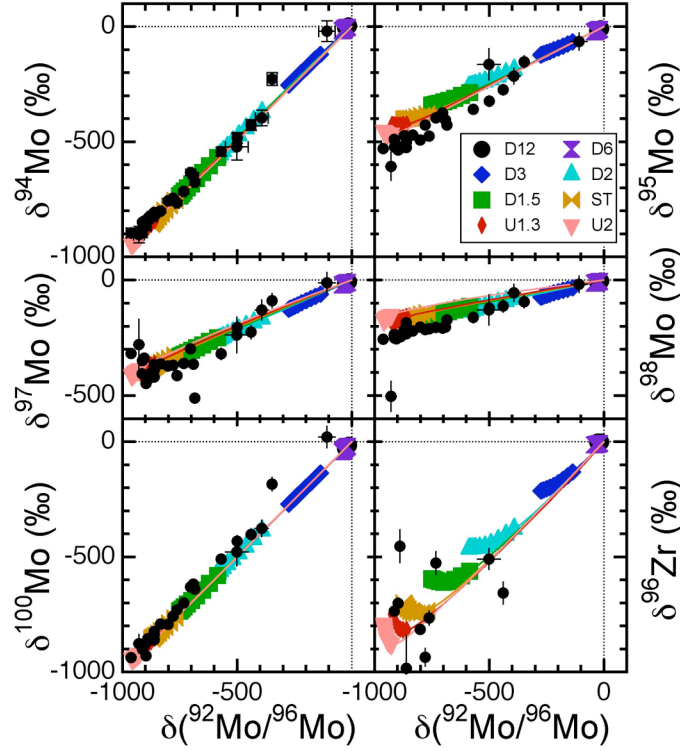


Fig. 2. Isotopic compositions of Mo in presolar SiC grains [14, 21] compared with those predicted for a $2 M_{\odot}$ AGB star with initially solar metallicity. Uncertainties are $\pm 2\sigma$.

The comparison between grains and predictions for Mo reinforces the conclusions reached from Zr and allows an additional conclusion to be reached: the alignment along straight lines confirms the basic picture of mixing of s-process material into an initially solar composition component, as expected for TDU into an initially solar composition AGB envelope. In recent work, Zr, Mo and Ba have all been measured in a number of mainstream grains along with C, N and Si [24]. Although the new data is not shown here, it shows some interesting effects. Examination of the lower right panel of Fig. 2 shows that there are three grains that lie below the predictions. Grains in this part of the diagram not only cannot be explained by these models, they cannot be explained by any neutron capture process acting on material of initially solar composition. The new data show additional grains in the “impossible” field as well as many grains that plot within the range allowed by models. The most likely explanation of the grains with “impossible” isotopic compositions is that they are contaminated with solar system Mo, but not with solar system Zr. This could have occurred in the meteorite parent body, during the extensive chemical processing needed to separate SiC from the host meteorite, or during mounting of the grains on their sample mounts. If grains that are contaminated with Mo are

eliminated from further consideration, the remaining, apparently uncontaminated, grains cover a narrower range of isotopic composition and can be explained with a narrower range of ^{13}C pocket efficiencies, from D1.5 to U1.3 [24].

Predictions for Ba for a $2 M_{\odot}$ AGB star are compared with mainstream presolar grains in Fig. 3. The grains match the predictions well, but the entire range of ^{13}C pocket efficiencies are needed to explain the full range of the data. In recent measurements comparing Zr and Ba measurements on the same grains, similar behavior to Mo vs. Zr were seen, that there are some grains with isotopic compositions that cannot be explained by neutron capture on material of initially solar isotopic composition [24]. If, as in the case of Mo, the grains of “impossible” composition are assumed to be contaminated with solar system Ba, the remaining grains are found to cover a narrower range of ^{13}C pocket efficiencies, from D1.5 to U1.3 [24].

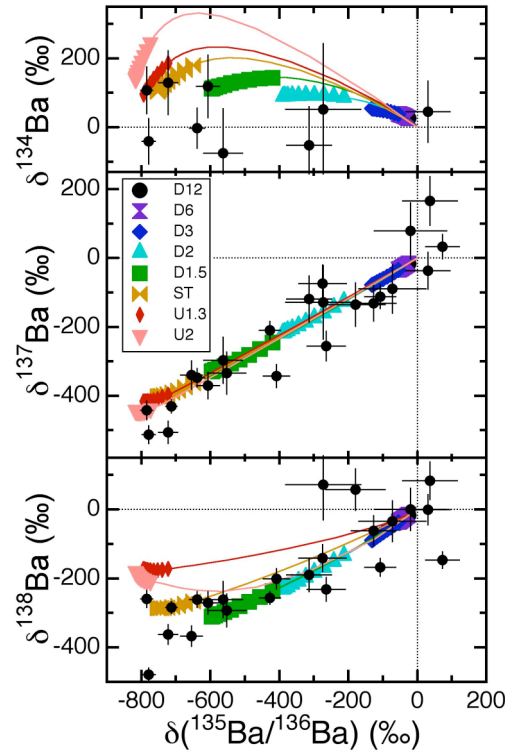


Fig. 3. Isotopic compositions of Ba in presolar SiC grains [16, 21] compared with those predicted for a $2 M_{\odot}$ AGB star with initially solar metallicity. Uncertainties are $\pm 2\sigma$.

Predictions for Sr for a $2 M_{\odot}$ AGB star are compared with mainstream presolar grains in Fig. 4. The relatively large error bars for the data allow agreement between predictions and data for $\delta(^{87}\text{Sr}/^{86}\text{Sr})$ vs. $\delta(^{84}\text{Sr}/^{86}\text{Sr})$, but on the upper plot, the predictions follow a curve that almost seems designed to avoid the data. This was noted earlier [21] and two explanations were offered: (1) some grains are contaminated with solar system Sr; or (2) the parent AGB stars had initial isotopic compositions that were enriched in $^{88}\text{Sr}/^{86}\text{Sr}$ relative to solar composition. In view of the new evidence [24] that some grains are contaminated with solar system Mo and/or Ba, the first explanation now seems likely. Contamination with solar system Sr would move data points along straight lines between their original uncontaminated compositions (which are expected to be along the curve of the $2 M_{\odot}$ predictions) and solar system composition, at (0,0) on the upper panel of Fig. 4. The recent work on Zr, Mo and Ba [24] suggests that ^{13}C pocket

efficiencies can be restricted to the range D1.5 to U1.3, but the upper panel of Fig. 4 shows three data points that seem to require an efficiency of U2. However, ^{88}Sr is more strongly produced in the late TPs of $3 M_{\odot}$ stars [21] and it may be that a few grains are from $3 M_{\odot}$ AGB stars.

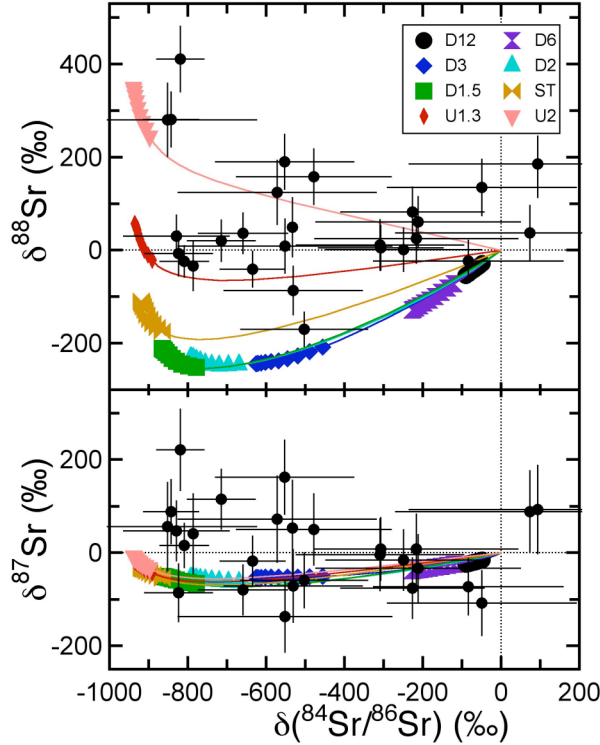


Fig. 4. Isotopic compositions of Sr in presolar SiC grains [12, 21] compared with those predicted for a $2 M_{\odot}$ AGB star with initially solar metallicity. Uncertainties are $\pm 2\sigma$.

5. Conclusions and outlook

Heavy element isotopic compositions of single presolar grains have confirmed the general picture of how the s -process isotopes are produced deep in AGB stars, mixed into the outer parts of those stars and ejected into the interstellar medium. Although a wide range of ^{13}C pocket efficiencies are allowed by current models of AGB stars, grains appear to come from a narrower range centered on the “standard case” (which explains the solar system s -process isotopic abundances with half-solar metallicity AGB stars). The timescales from models are consistent with the levels of short-lived radionuclides inferred to have been present in mainstream presolar SiC grains [25].

Measurement of the isotopic compositions of heavy elements in individual presolar grains has allowed considerable progress in understanding of AGB stars and providing “ground truth” for stellar nucleosynthesis modeling. There is much still to do. A number of elements likely occur in concentrations high enough to measure and some of these are sensitive to branch points in the s -process nucleosynthesis path. Measurement of multiple elements on the same grain has proven to be quite powerful for recognizing grains for which solar system contamination seems to be an issue. Further work is clearly needed in trying to remove this contamination from

grains. Relatively few measurements exist on elemental abundances in individual presolar grains and there are no grains for which both elemental and isotopic abundances are known. Such measurements will be very important for disentangling the effects of nucleosynthesis and high temperature chemistry in stellar photospheres. Recent measurements of isotopic and elemental abundances in aggregates of presolar grains have shown the power of this approach [6].

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References

- [1] T. Bernatowicz, G. Fraundorf, T. Ming, E. Anders, B. Wopenka, E. Zinner, and P. Fraundorf *Evidence for interstellar silicon carbide in the Murray carbonaceous meteorite*, *Nature* **330**, 728–730, 1987.
- [2] E. Zinner 2004 *Presolar grains*. In *Meteorites, Planets, and Comets* (Ed. A. M. Davis), Vol. 1 *Treatise on Geochemistry* (Eds. H. D. Holland and K. K. Turekian), Elsevier-Pergamon, Oxford, pp. 17–39, 2004.
- [3] D. D. Clayton and L. R. Nittler *Astrophysics with presolar stardust*. *Annu. Rev. Astron. Astrophys.* **42**, 39–78, 2004.
- [4] L. R. Nittler *Presolar stardust in meteorites: recent advances and scientific frontiers*. *Earth Planet. Sci. Lett.* **209**, 259–273, 2003.
- [5] E. Zinner, S. Amari, and R. S. Lewis *s-Process Ba, Nd, and Sm in presolar SiC from the Murchison meteorite*. *Astrophys. J.* **382**, L47–L50, 1991.
- [6] Q.-Z. Yin, C.-T. A. Lee, and U. Ott *Signatures of the s-process in presolar silicon carbide grains: barium through hafnium*. *Astrophys. J.* **647**, 676–684, 2006.
- [7] F. A. Podosek, C. A. Prombo, S. Amari, and R. S. Lewis *s-Process Sr isotopic compositions in presolar SiC from the Murchison meteorite*. *Astrophys. J.* **605**, 960–965, 2004.
- [8] U. Ott and F. Begemann *Discovery of s-process barium in the Murchison meteorite*. *Astrophys. J.* **353**, L57–L60, 1990.
- [9] C. A. Prombo, F. A. Podosek, S. Amari, and R. S. Lewis *s-Process Ba isotopic compositions in presolar SiC from the Murchison meteorite*. *Astrophys. J.* **410**, 393–399, 1993.
- [10] S. Richter, U. Ott, and F. Begemann *s-Process isotope abundance anomalies in meteoritic silicon carbide: new data*. In *Nuclei in the Cosmos* (Eds. F. Käppeler and K. Wisshak). Institute of Physics Publishing, Bristol and Philadelphia, pp. 127–132, 1993.
- [11] S. Richter, U. Ott, and F. Begemann *s-Process isotope abundance anomalies in meteoritic silicon carbide: data for Dy*. In *Proc. European Workshop on Heavy Element Nucleosynthesis* (Eds. E. Somorjai and Z. Fülöp). *Inst. Nucl. Res. Hungarian Acad. Sci., Debrecen*, pp. 44–46, 1994.
- [12] G. K. Nicolussi, M. J. Pellin, R. S. Lewis, A. M. Davis, R. N. Clayton, and S. Amari *Strontium isotopic composition in individual circumstellar silicon carbide grains: a record of s-process nucleosynthesis*. *Phys. Rev. Lett.* **81**, 3583–3586, 1998.

- [13] G. K. Nicolussi, A. M. Davis, M. J. Pellin, R. S. Lewis, R. N. Clayton, and S. Amari *s-Process zirconium in presolar silicon carbide grains*. *Science* **277**, 1281–1283, 1997.
- [14] G. K. Nicolussi, M. J. Pellin, R. S. Lewis, A. M. Davis, S. Amari, and R. N. Clayton *Molybdenum isotopic anomalies in individual presolar silicon carbide grains from the Murchison meteorite*. *Geochim. Cosmochim. Acta* **62**, 1093–1104, 1998.
- [15] M. R. Savina, A. M. Davis, C. E. Tripa, M. J. Pellin, R. Gallino, R. S. Lewis and S. Amari *Extinct technetium in presolar silicon carbide grains*. *Science* **303**, 649–652, 2004.
- [16] M. R. Savina, A. M. Davis, C. E. Tripa, M. J. Pellin, R. N. Clayton, R. S. Lewis, S. Amari, R. Gallino, and M. Lugaro (2003) *Barium isotopes in individual presolar silicon carbide grains from the Murchison meteorite*. *Geochim. Cosmochim. Acta* **67**, 3201–3214, 2003.
- [17] M. R. Savina, M. J. Pellin, C. E. Tripa, I. V. Veryovkin, W. F. Calaway, and A. M. Davis *Analyzing individual presolar grains with CHARISMA*. *Geochim. Cosmochim. Acta* **67**, 3215–3225, 2003.
- [18] S. Amari, P. Hoppe, E. Zinner, and R. S. Lewis *Trace-element concentrations in single circumstellar silicon carbide grains from the Murchison meteorite*. *Meteoritics* **30**, 679–693, 1995.
- [19] K. K. Marhas, P. Hoppe, and U. Ott *A NanoSIMS study of C-, Si- and Ba-isotopic compositions of presolar silicon carbide grains from the Murchison meteorite*. *Meteorit. Planet. Sci.* **38**, A58.
- [20] R. Gallino, C. Arlandini, M. Busso, M. Lugaro, C. Travaglio, O. Straniero, A. Chieffi, and M. Limongi *Evolution and nucleosynthesis in low-mass asymptotic giant branch stars. II. Neutron capture and the s-process*. *Astrophys. J.* **497**, 388–403, 1998.
- [21] M. Lugaro, A. M. Davis, R. Gallino, M. J. Pellin, O. Straniero, and F. Käppeler *Isotopic compositions of strontium, zirconium, molybdenum, and barium in single presolar SiC grains and asymptotic giant branch stars*. *Astrophys. J.* **593**, 486–508, 2003.
- [22] O. Straniero, R. Gallino, and S. Cristallo *s-Process in low-mass asymptotic giant branch stars*. *Nucl. Phys. A* **777**, 311–339, 2006.
- [23] K. Lodders and B. Fegley, Jr. *The origin of circumstellar silicon carbide grains found in meteorites*. *Meteoritics* **30**, 661–678, 1995.
- [24] J. G. Barzyk, M. R. Savina, A. M. Davis, R. Gallino, M. J. Pellin, R. S. Lewis, S. Amari, and R. N. Clayton *Multi-element isotopic analysis of single presolar SiC grains*. *New Astron. Rev.* **50**, 587–590, 2006.
- [25] A. M. Davis and R. Gallino *Short-lived radionuclides in presolar SiC grains: constraints on timescales in AGB stars*. *Mem. Soc. Astronom. Ital.* **77**, 885–890, 2006.