HEAVY ELEMENT ISOTOPIC ANALYSIS OF LOW DENSITY GRAPHITE GRAINS WITH LION.
Ishita Pal[1], Manavi Jadhav[1], Danielle Z. Shulaker[2], and Michael R. Savina[2], [1]Department of Physics, University of Louisiana Lafayette, Lafayette, LA 70503 (palishita13@gmail.com), [2]Nuclear and Chemical Sciences Division, Lawrence Livermore National Laboratory, CA.

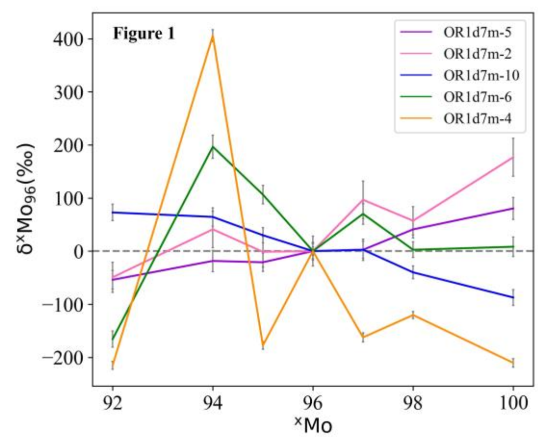
Introduction: Presolar SiC grains are the most widely studied presolar grain type because of their high abundance and ease of separation from their host meteorites [1]. Most SiC (i.e., mainstream grains) originate from low mass asymptotic giant branch (AGB) stars; only about 1.5% of SiC (SiC-X) come from supernovae (SNe) [2]. Presolar graphite grains from low density (LD) fractions exhibit isotopic signatures that indicate SN sources [e.g., 3]. In contrast to SiC SNe grains, about 26% of presolar graphite grains originate in type II SNe [4]. Thus, studying LD graphite grains increases our chances of systematically studying nucleosynthetic reactions in high mass stars. LD graphites are also expected to have higher concentrations of heavy elements than mainstream grains and therefore should yield useful data [5]. Investigations of isotopic patterns of heavy elements in such grains will reveal important insights into neutron capture processes, like the *n*- and -*r* processes, as well as help constrain the astrophysical sites of the enigmatic *p*-process nucleosynthesis.

In this study, we measured Zr, Mo, and Ru isotopes in LD graphite grains from Orgueil to constrain their stellar sources. A recent theoretical study by [6] compared Zr and Mo isotopic data in SiC-X grains to stellar yields. We aim to perform a similar comparison of our grains with data/conclusions presented in [6]. This is the first heavy element study by Resonance Ionization Mass Spectrometry (RIMS) of LD presolar graphite.

Methods: Thirteen LD graphite grains from Orgueil (OR1d: 1.75−1.92 g cm^{-3}) mounted on gold that were previously measured for C, N, Si isotopes and trace element abundances [7,8] were imaged in an SEM. Six of 13 grains had sufficient material for RIMS analyses. The grains were C-welded to the mount and marked with fiducials using a Focused Ion Beam at the Shared Instrumentation Facility at Louisiana State University. ^{90,91,92,94,96}Zr, ^{92,94,95,96,97,98,100}Mo, and ^{96,98,99,100,101,102,104}Ru were measured simultaneously on each grain using the Laser Ionization of Neutrals (LION) RIMS at the Lawrence Livermore National Laboratory using six tunable Ti: Sa lasers. Lasers tuned to wavelengths to resonantly ionize Zr, Mo, and Ru were pulsed at slightly differing times to resolve isobaric interferences. We report data on five grains for which we collected data with LION.

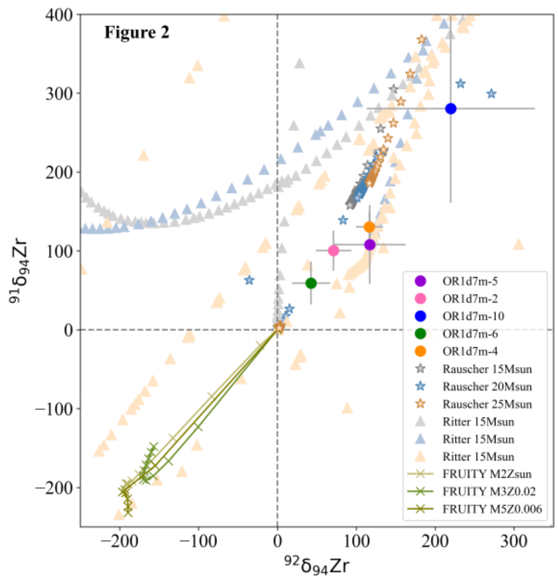
Results:

C, N, Si isotopes. Light element isotopes were measured at the University of Hawai'i Cameca ims 1280 ion microprobe. Details of the measurements can be found in [7,8]. Three OR1d7m grains (-2, -5, and -10) show significant excesses in ^{13}C ($^{12}C/^{13}C$=13−19±0.4−1.0), while OR1d7m-4 shows a minor excess ($^{12}C/^{13}C$=58.4±2.6) compared to the solar value of $^{12}C/^{13}C$=89. Only OR1d7m-6 is slightly ^{12}C enriched ($^{12}C/^{13}C$=125.3±3.8). Nitrogen isotopic ratios in three of the five grains are close to that of air ($^{14}N/^{15}N$=272). Two anomalous OR1d7m grains (-4 and -6) have $^{14}N/^{15}N$ values of 108.7±4.2 and 112.6±1.6, respectively. Along with an excess in ^{15}N, OR1d7m-4 also contains an excess in ^{28}Si, further indicating an origin in a core-collapse SN [e.g., 9]. OR1d7m-6 is depleted in ^{30}Si but has an excess of ^{29}Si, while OR1d7m grains -2, -5, and -10 are solar within 2σ.



Mo isotopes. All Mo data were normalized to the pure *s*-process isotope, ^{96}Mo. Three isotope plots of LD grains are markedly different from those for high density graphite grains from Murchison [10] which show a significant number of grains with classic *s*-process signatures in which all isotopes are depleted with respect to ^{96}Mo. In contrast, pure *s*-process signatures were not seen in the grains from this study. As seen in Figure 1, grain OR1d7m-4 exhibits an M-shaped pattern similar to the pure *s*-process signature where it is moderately depleted in ^{92,95,97,98,100}Mo, however it has $\delta^{94}Mo_{96}$ ~ +400. ^{94}Mo is predominantly produced by the *p*-process, with a minor *s*-process contribution. However, ^{92}Mo is also a pure *p*-process isotope and is depleted rather than enhanced in this grain. Four of the measured grains are depleted in ^{92}Mo. OR1d7m-10 has excesses in both the *p*-process isotopes ^{92,94}Mo. In contrast, OR1d7m grains -2 and -5 have excesses in only ^{100}Mo and have close to solar values for all other isotopes of Mo (within 2s). The Mo isotopes in OR1d7m-6 are intriguing as it is depleted in ^{92}Mo, has excesses in $^{94.95,97}$Mo and is solar in ^{98,100}Mo.

Ru isotopes: The seven stable isotopes of Ru have similar isotopic characteristics as the isotopes of Mo. The lighter isotopes, ^{96,98}Ru, are produced by *p*-process, while the heaviest isotope, ^{104}Ru, is a pure *r*-process isotope. ^{100}Ru is a pure *s*-process isotope, while the remaining have contributions from both *s*- and *r*-processes. Similar to Mo, Ru data are normalized to the pure *s*-process isotope ^{100}Ru. OR1d7m grains -2 and -4 show significant depletion in all Ru isotopes relative to ^{100}Ru, suggesting *s*-process nucleosynthesis. OR1d7m-5 and -6 show Ru isotope ratios within 2σ of the solar composition. Unfortunately, we did not obtain statistically significant ion counts on OR1d7m-10.



Zr isotopes: Zr has five stable isotopes. ^{90,91,92,94}Zr are predominantly produced by the *s*-process, while ^{96}Zr is traditionally considered to be produced by *r*-process but can also be fed by the *s*-process at neutron fluxes of > 3×10^{8} neutron cm^{-3} [11] or by neutron bursts in SNe [12]. In this study, Zr data are normalized to the *s*-process isotope, ^{94}Zr. Figure 2 shows the Zr isotopic grain data where all the grains have positive $\delta^{91,92}Zr_{94}$ values. Three grains show normal to solar values within 2σ for all Zr isotopes. OR1d7m-4 has excesses in $^{90,91,92}Zr_{94}$ while OR1d7m-2 has excesses in $^{91,92}Zr_{94}$. When compared with stellar nucleosynthesis models (Figure 2), low mass FRUITY AGB star models [13] do not explain the grains' isotopic distributions. Figure 2 also plots the isotopic yields from the He shells and shallower regions of 15, 20 and 25 solar mass SNe models with solar metallicity [14, 15]. The yields include radiogenic contributions from Sr, Y, Rb, and Kr to the Zr isotopes [6]. The SNe models can explain the ^{94}Zr depletion in the grains from this study.

Discussion: Excesses of ^{95,97}Mo and ^{96}Zr in OR1d7m-6 show evidence of *n*-process in SNe [12, 6]. It also has a ^{15}N excess reaffirming a SN origin. However, we cannot explain the ^{94}Mo excess in OR1d7m-6. OR1d7m-4 also has an excess of ^{94}Mo while all the other Mo isotopes are depleted relative to ^{96}Mo. This grain shows SNe signatures as excesses in ^{28}Si and ^{15}N [16]. Further, OR1d7m-10 does not show any SNe signatures in light element isotopes but contains enrichments in the *p*-process isotopes of Mo, expected in high energy stellar environments. OR1d7m-2 and -5 contain mild enrichments in the *r*-process-only isotope ^{100}Mo. Lastly, the ^{13}C enrichments in OR1d7m-10, -5, and -2 can be attributed to H ingestion in the He shell of core collapse SNe [6].

Conclusion: In this study, we identified grains that show an *n*-process and a *p*-process nucleosynthetic signature and two grains with *r*-process signatures. Overall, all the five LD graphite grains exhibit isotopic features consistent with origins in type II SNe.

Acknowledgments: IP acknowledges NASA FINESST award 21-PLANET21-0133. IP and MJ acknowledge NSF 20-543 award #2033380. MRS and DZS acknowledge funding from the LLNL Glen T. Seaborg Institute and LLNL-LDRD Program under Project 20-ERD-030. MRS and DZS work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. LLNL-ABS-843839.

References: [1] Amari S. et al. (1994) *GCA, 58, 459-470* [2] Zinner E. (2014) *Treatise on Geochem., Elsevier. 2nd ed, Vol 1.4, 181* [3] Hoppe P. et al. (1995) *GCA, 59, 4029-4056* [4] Jadhav M. et al. (2014) *PoS NIC XIII, 004* [5] Croat T.K. et al. (2003) *GCA, 67, 4705-4725* [6] Pignatari M. et al. (2018) *GCA, 221, 37-46* [7] Jadhav M. et al. (2014) *LPS XLV Abstract # 1015* [8] Jadhav M. et al. (2015) *LPS XLVI Abstract # 2882* [9] Jadhav M. et al. (2013) *GCA, 113, 193-224* [10] Pal I. et al. (2022) *LPS LII, Abstract #1262* [11] Lugaro M. et al. (2003) *ApJ, 593, 486-502* [12] Meyer B.S. et al. (2000) *ApJ Lett., 540, 49-52* [13] Cristallo S. et al. [2011] *ApJ Suppl., 197, 17* [14] Rauscher T. et al. (2002) *ApJ, 576, 323-348* [15] Ritter C. et al. (2018) *MNRAS, 480, 538-571* [16] Meyer B.S. and Zinner E. (2006) *MESS II, U. Arizona Press, 69-108*