

HEAVY ELEMENT ISOTOPIC MEASUREMENTS OF HIGH DENSITY PRESOLAR GRAPHITE GRAINS WITH LION. Ishita Pal¹, Manavi Jadhav¹, Michael R. Savina², Danielle Z. Shulaker², Christopher J. Dory², Frank Gyngard³, Noriko Kita⁴, Sachiko Amari⁵. ¹Department of Physics, University of Louisiana at Lafayette, Lafayette, LA 70503 (palishita13@gmail.com), ²Nuclear and Chemical Sciences Division, Lawrence Livermore National Laboratory, CA, ³Center for NanoImaging, Department of Medicine, Brigham and Women's Hospital, Cambridge, MA 02139, ⁴Department of Geoscience, University of Wisconsin-Madison, Madison, WI 53706, ⁵McDonnell Center for the Space Sciences & Physics Department, Washington University, St. Louis, MO 63130.

Introduction: Based on multi-element isotopic analyses of major, minor, and trace light elements, a majority of high density (HD) presolar graphite grains are known to come from Asymptotic Giant Branch (AGB) Stars [1,2]. Supernova (SN) signatures have been observed in a few HD graphite grains [1,2] and some grains also contain evidence for *i*-process nucleosynthesis observed in post-AGB stars [3]. Early Resonance Ionization Mass Spectrometry (RIMS) was used to collect Zr and Mo isotopes in HD grains from Murchison [4], but light-element data, which are essential for classifying presolar grains, were not collected on the same grains. A later study used a previous generation RIMS instrument (CHARISMA) to obtain Zr, Mo, and Ba isotopic data on Orgueil HD grains but the data were mostly inconclusive as grains were extensively measured for light elements, resulting in very little grain left over, severely reducing the total ion counts obtained on individual grains [5,6].

In a continued quest to better understand the nucleosynthesis sources of HD graphite grains, we measured C, N, O, Sr, Zr, and Mo isotopes of Murchison HD graphite grains.

Methods: Fifty-six graphite grains from the KFB1h density (2.10–2.15 g cm⁻³) fraction of Murchison were identified and documented with a scanning electron microscope (SEM). Carbon and N isotopes were measured with the NanoSIMS 50 at Washington University, St. Louis. Oxygen isotopes were measured at the WiseSIMS lab (CAMECA IMS-1280) of the University of Wisconsin-Madison [7].

Prior to the RIMS measurements, the graphite grains were FIB welded to the gold mount using a C weld to prevent the grains from hopping off the mount, as is common while measuring graphites by RIMS [5,6]. Fiducial markers were also milled into the mount to simplify navigation during RIMS analyses. This work was done at the Shared Instrumentation Facility at Louisiana State University and the W.M. Keck Center for Nanoscale Optofluidics at the University of California, Santa Cruz. The grains were then redocumented in an SEM at Lawrence Livermore National Laboratory (LLNL). RIMS data were obtained on 46 KFB1h graphite grains at LLNL using the new generation, Laser Ionization of Neutrals (LION) instrument. Each grain was simultaneously measured for ^{84,86,87,88}Sr, ^{90,91,92,94,96}Zr,

and ^{92,94,95,96,97,98,100}Mo. A Nd:YAG laser (1064 nm, 7 ns full width at half-maximum, 1000 Hz) focused to a 1–2 μm spot on the sample volatilized material. Six tunable Ti:sapphire lasers were aligned collinearly ~1 mm above the sample surface to resonantly ionize Sr, Mo, and Zr atoms. Two-color two-photon RIS schemes employed to ionize Sr and Zr and Mo, respectively, are described in [8,9]. To discriminate between Mo and Zr isobaric interferences, Sr and Mo resonant ionization lasers were pulsed first. At the same time, the sample potential was raised rapidly to 3 kV to immediately accelerate the Sr and Mo photoions into the TOF mass spectrometer. Then, after an additional 200 ns, Zr ionization lasers were pulsed to ionize Zr in the same cloud of neutral atoms. The time-of-birth difference between the Zr and Mo ions resolved the isobars at m/z 92, 94, and 96.

Results: *C, N, O isotopes.* The C, N, O isotopic ratios of the grains in this study are discussed in detail in [7]. The ¹²C/¹³C ratios measured in the 56 graphite grains varied over a large range, from 11 to 1777. HD graphite fractions from Murchison and Orgueil are known to mostly contain grains with isotopically light C [1,2] and the grains from this study follow the same trend. Five grains were found to be highly enriched in ¹³C. Three grains contain large ¹⁸O and/or ¹⁵N excesses indicating origins in supernovae. Similar to previously studied HD presolar graphite grains [1,2], a majority of the ¹⁴N/¹⁵N isotopic ratios had values close to that of air (272). Except the three SN grains, the remaining grains had excesses in ¹⁷O and ¹⁸O up to 15% relative to solar [7].

Mo isotopes. All the Mo isotopes were normalized to ⁹⁶Mo, a pure *s*-process isotope. ^{92,94}Mo are *p*-process isotopes; however, ⁹⁴Mo can also be produced by the *s*-process. ¹⁰⁰Mo is a pure *r*-process isotope. We obtained a consistently high ion yield for Mo isotopes in all the measured grains. Twenty one out of 46 grains show strong to moderate *s*-process signatures, while three contain diluted *s*-process signatures (Figure 1). These signatures indicate an origin in low mass AGB stars [10]. One of these three grains is highly ¹³C-enriched. The remaining grains had solar Mo isotopic compositions.

Interestingly, Mo data on 8 grains might indicate that some portion of the Mo signal originates from sub-grains. We will discuss this issue further at the meeting.

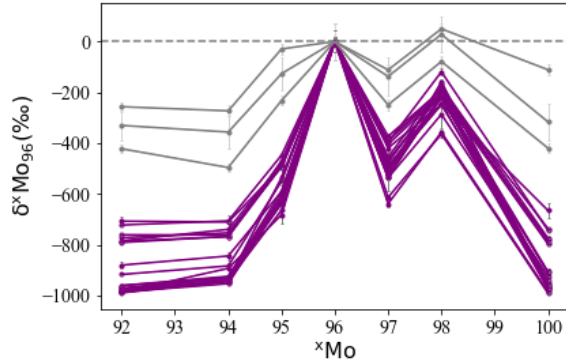


Figure 1: Mo mass plot showing grains with strong to moderate (purple) and diluted (grey) s -process signatures in KFB1h HD grains. All errors shown are 1σ .

Sr isotopes. Sr isotopes were normalized to ^{86}Sr , a pure s -process isotope. ^{84}Sr is a pure p -process isotope, $^{86,87}\text{Sr}$ are s -process isotopes while ^{88}Sr is mostly an s -process isotope but can also be made by the r -process. Seven grains in this study show large depletions in ^{84}Sr (Figure 2) which can be attributed to the large abundance of ^{86}Sr in low mass AGB star envelopes [10]. Three out of the seven ^{84}Sr depleted grains were also depleted in ^{88}Sr , while three had excesses in the neutron magic isotope. One of the seven grains had deficits in $^{84,88}\text{Sr}$, indicating an excess in ^{86}Sr . This grain (KFB1h-441) also shows SN signatures with large excesses in ^{15}N and ^{18}O and a $^{12}\text{C}/^{13}\text{C} = 40.7 \pm 0.2$. Apart from the Sr anomalies in the 7 grains discussed earlier, we found a range of ^{88}Sr excesses in 12 grains ($\delta^{88}\text{Sr} = 115 \pm 15 - 667 \pm 118$ ‰) and ^{88}Sr deficits in four grains ($\delta^{88}\text{Sr} = -85 \pm 29 - -461 \pm 32$ ‰). These signatures (apart from the 1 SN grain described above) are typical of s -process nucleosynthesis in low mass AGB stars [10].

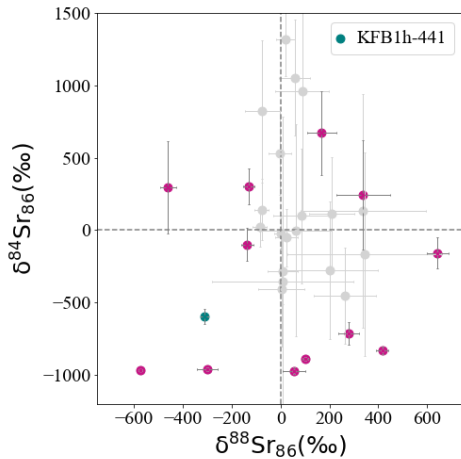


Figure 2: 3-isotope δ -plot of $^{84,86,88}\text{Sr}$. Pink data points are grains with anomalous $\delta^{84,88}\text{Sr}$ within 2σ . Grey data points are grains with solar values of $\delta^{84,88}\text{Sr}$. SN grain KFB1h-441 is shown in teal. All errors shown are 1σ .

Zr isotopes. We obtained very low Zr ion yields on grains in this study resulting in large errors on our measurements. However, 10 grains yielded good statistics. A few of these grains show ^{90}Zr depletions with respect to solar. All Zr isotopes are generated by the s - and r -processes except ^{96}Zr which is an r -process isotope. Most of the grains had negligible ^{96}Zr counts or were found to be depleted in $\delta^{96}\text{Zr}$ with respect to solar. However, we found a prominent ^{96}Zr excess ($\delta^{96}\text{Zr} = 4233 \pm 530$ ‰) in grain KFB1h-351. It has a $^{12}\text{C}/^{13}\text{C} = 14.0 \pm 0.1$ and also exhibited a diluted s -process signature in Mo isotopes. While these signatures appear to be contradictory, it must be noted that Sr, Zr, and Mo are also present in refractory carbide subgrains [11,12] and these signatures need not correlate as they might arise from different subgrains within the same parent grain. Earlier studies of HD graphites found similar ^{96}Zr excesses in three grains [4,6].

Conclusion: This study reports the first successful systematic attempt to carry out coordinated light- and heavy-element isotopic analyses on presolar graphite grains. We observed clear s -process signatures in Mo and Sr isotopes that point to low mass AGB star origins for most of the grains. We will discuss further implications of this study and the stellar sources of the grains in detail at the meeting.

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