

Ti ISOTOPIC RATIOS IN LOW-DENSITY GRAPHITE GRAINS FROM ORGUEIL. M. Jadhav¹, S. Amari², E. Zinner² and T. Maruoka^{2*}, ¹Laboratory for Space Sciences and the Department of Earth and Planetary Sciences (manavijadhav@wustl.edu), ²Laboratory for Space Sciences and the Physics Department, Washington University in St. Louis, One Brookings Dr., St. Louis, MO 63130, USA., * present address: Graduate School of Life and Environmental Sciences, University of Tsukuba, Ibaraki 305-8572, Japan.

Introduction: In previous studies [1, 2], some of the grains from the low-density graphite fractions of Orgueil, ORG1c and ORG1d ($\rho \sim 1.59$ - 1.67 g cm⁻³ and 1.75 - 1.92 g cm⁻³, respectively), were found to have isotopic signatures in N, O, Si and Al-Mg that indicate a Type II supernova (SN) origin. In order to characterize these grains further we carried out multi-element (C, N, O, Si, Al-Mg, K, Ca, and Ti) isotopic analyses of a new set of low-density graphite grains from the ORG1d fraction of Orgueil. We also expect to be able to analyze the same grains for heavy element isotopes (Sr, Zr, Mo, Ru, Ba) by resonant ionization mass spectrometry (RIMS).

Experimental Methods: Thirty carbonaceous grains from ORG1d were isolated from the large amounts of macromolecular carbonaceous material, in which these grains from Orgueil are often found embedded. This was done to reduce contamination by the carbonaceous material and, hence, facilitate the isotopic analyses. After mounting the grains on a gold foil mount, we coated the mount with a 40 nm layer of gold to prevent the grains from falling off.

The isotopic measurements of these grains were carried out with the NanoSIMS at Washington University. A Cs⁺ primary beam was used to generate negative secondary ions of ¹²C, ¹³C, ²⁸Si, ²⁹Si, and ³⁰Si in phase 1 of the analyses, and of ¹⁶O, ¹⁸O, ¹²C¹⁴N, and ¹²C¹⁵N, in phase 2. Positive secondary ions of ¹²C, ²⁴Mg, ²⁵Mg, ²⁶Mg, and ²⁷Al, were produced by an O⁻ primary beam in phase 3 of the analyses. The measurements of these three phases were made in multidetection mode. The K, Ca, and Ti measurements were carried out with the O⁻ beam in a combination of peak-jumping and multidetection modes. Positive secondary ions of ³⁹K, ⁴¹K and ⁴³Ca (B field 1) and ¹²C, ⁴⁰Ca, ⁴²Ca, ⁴⁴Ca and ⁴⁸Ti (B field 2) were measured to obtain K and Ca ratios. Ti isotopes were measured at 3 magnetic fields: at B₁ we detected ⁴⁶Ti, ⁴⁸Ti, and ⁵⁰Ti; B₂ – ⁴⁷Ti, ⁴⁹Ti, and ⁵¹V; and B₃ – ¹²C, ⁴⁰Ca, ⁴⁸Ti, ⁵⁰Ti, and ⁵²Cr. ⁵¹V and ⁵²Cr were used to correct the ⁵⁰Ti signal for isobaric interferences from V and Cr, and, ⁴⁰Ca was measured to correct for Ca interferences at masses 46 and 48.

Results: Figures 1 – 4 show the C, N, O, Si, Al-Mg isotopic ratios of the measured grains along with results from previous studies [1, 2]. The ¹²C/¹³C ratios vary from 11 to 2400. Most grains have sub-solar or solar C isotopic ratios (solar ¹²C/¹³C ~ 89). There exists a distinct population of grains with ¹²C/¹³C ratios $< \sim 20$. Eight of the grains exhibit ¹⁸O excesses, with ¹⁸O/¹⁶O ratios up to 4 times the solar value. Two of these ¹⁸O-enriched grains have the largest ¹⁵N excesses while four of them have the largest ²⁸Si excesses. One grain (g-5) is ³⁰Si enriched ($\delta^{30}\text{Si} =$

136 ± 28 ‰). The ¹⁵N, ¹⁸O and ²⁸Si excesses are indicative of a SNII origin for these grains [3]. In addition, nine grains have large ²⁶Al/²⁷Al ratios (ranging from 10^{-3} up to 0.04) that were derived from ²⁶Mg excesses. All of these grains contain excesses in ¹⁵N, ¹⁸O, or ²⁸Si, if not in all these three isotopes. These signatures in conjunction with high inferred ²⁶Al/²⁷Al ratios are consistent with a Type II SN origin of the grains.

It is clear from Figures 1 – 4 that the anomalies observed in the present study are not as large as those observed in previous studies of grains from the same density fraction. We suspect that this is due to contamination from the large quantity of gold that was deposited on the grain mount prior to the analyses. This contamination dilutes the anomalous isotopic ratios of the grains.

The mount was also found to be contaminated by large amounts of terrestrial K and Ca, making the detection of radiogenic ⁴¹Ca impossible. The ^{42,43,44}Ca data was also affected by the large Ca contamination. All the grains are normal in ⁴²Ca/⁴⁰Ca (within errors), and one grain (g-5) has an excess in ⁴³Ca ($\delta^{43}\text{Ca}/^{40}\text{Ca} = 154 \pm 43$ ‰). Grain g-5 also has a high $\delta^{44}\text{Ca}/^{40}\text{Ca}$ value of 222 ± 22 ‰. Since the ⁴³Ca and ⁴⁴Ca excesses are of approximately the same magnitude, they can be explained by neutron capture in the interior zones (He/C or O/C zones) of a type II SN and the ⁴⁴Ca excess does not constitute evidence for the presence of ⁴⁴Ti. The rest of the grains have normal ⁴⁴Ca/⁴⁰Ca ratios. Most grains have normal ⁴⁶Ti/⁴⁸Ti and ⁴⁷Ti/⁴⁸Ti ratios (within errors). Several grains have correlated excesses in these two isotopes and one has a large ⁴⁶Ti depletion, but a normal ⁴⁷Ti/⁴⁸Ti ratio (Figure 5). Five grains have large ⁴⁹Ti excesses (as high as 1597 ± 85 ‰ in grain g-2) while six have moderate ⁴⁹Ti excesses (Figure 6). Due to a very high ⁵⁰Cr signal that interfered with ⁵⁰Ti, we were unable to obtain good ⁵⁰Ti data on these grains. The grains acquire Cr during chemical separation in the laboratory from Na₂Cr₂O₇, which is used as an oxidizing agent to remove macromolecular carbon. Three grains that have less than a 60% ⁵⁰Cr contribution to their ⁵⁰Ti ion signal have elevated $\delta^{50}\text{Ti}/^{48}\text{Ti}$ values of 259 ± 97 ‰ (g-6), 185 ± 22 ‰ (g-14) and 144 ± 38 ‰ (g-18). The magnitude of the ^{49,50}Ti excesses seen in the grains in this study can be explained by admixture from the O-rich and He/C zones of a SNII to the outer zones, which have normal Ti isotopic ratios [3].

Most of the grains with Ti anomalies have ¹⁸O, ¹⁵N, ²⁸Si excesses and high ²⁶Al/²⁷Al ratios. This confirms their SN origin.

Conclusions: Our Ti data confirm that low-density graphite grains from Orgueil seem to originate from Type II supernovae. A further scrutiny of the Ti isotopic pat-

terns in these grains is necessary. For example, the three grains (g-2, g-24, g-27) with the largest ^{49}Ti excesses have close-to-solar C isotopic ratios but substantial ^{18}O excesses. It is still unclear why grains with the highest ^{18}O excesses have C ratios that do not differ too much from the solar ratio in view of the fact that the He/C zone, responsible for the ^{18}O excesses, contains almost pure ^{12}C . In addition, the population of grains that are ^{13}C -enriched ($^{12}\text{C}/^{13}\text{C}$ ratios $< \sim 20$) need to be studied in depth in order to determine their stellar source.

References: [1] Jadhav M. et al. (2006) *New Astron. Rev.* 50, 591–595. [2] Jadhav M. et al. (2006) *Meteoritics & Planet. Sci.*, 41, A87. [3] Travaglio C. et al. 1999. *ApJ*, 510, 325-354.

