

CONTINUED STUDY OF PRESOLAR GRAPHITE FROM MURCHISON SEPARATE KFA1. S. Amari¹, F. J. Stadermann¹, E. Zinner¹ and R. S. Lewis², ¹Laboratory for Space Sciences and the Physics Department, Washington University, One Brookings Dr., St. Louis, MO 63130, USA (sa@wuphys.wustl.edu, fjs@wuphys.wustl.edu, ekz@wuphys.wustl.edu), ²Enrico Fermi Institute, University of Chicago, 5630 S. Ellis Ave., Chicago, IL 60637, USA (r-lewis@uchicago.edu)

Introduction: Presolar graphite was isolated from the Murchison meteorite [1] only 3 years after the discovery of presolar SiC [2]. Yet it has not been studied as extensively as SiC. One of the reasons is that trace element concentrations in graphite grains are, in general, not high enough to perform isotopic analysis with sufficient precision. A new type of ion microprobe, the NanoSIMS, has recently been installed at Washington University in St. Louis. Its high sensitivity at high-mass resolution and its multi-detection capability [3] make it possible to measure isotopic ratios of trace elements with unprecedented precision. We have previously reported C, N, O, and Si isotopic ratios of graphite from the Murchison KFA1 separate (2.05-2.10g/cm³) [4]. We extended our studies to Mg-Al, K, Ca, and Ti in the same KFA1 graphite grains that we had previously analyzed in order to obtain correlated isotopic ratios.

Experimental Procedures: The measurements were made with the NanoSIMS at Washington University. We analyzed Mg-Al ratios in 75 grains, K-Ca isotopes in 24 grains, and Ti isotopes in 32 grains. An O⁻ primary beam was rastered over a 3×3μm² area on the grains. For Mg-Al analysis, ¹²C⁺, ²⁴Mg⁺, ²⁵Mg⁺, ²⁶Mg⁺ and ²⁷Al⁺ were simultaneously collected with five small electron multipliers. Potassium and Ca isotopic analysis was performed in the combined analysis mode, which uses magnetic peak jumping in connection with multi-detection. In the first step, ¹²C⁺, ³⁹K⁺, ⁴¹K⁺, ⁴³Ca⁺ and ⁴⁷Ti⁺ were measured, in the second ⁴⁰Ca⁺, ⁴²Ca⁺, ⁴⁴Ca⁺, and ⁴⁸Ti⁺ after changing the magnetic field and deflection voltages of the electron multipliers. Titanium isotopes were also analyzed in the combined mode: ¹²C⁺, ⁴⁶Ti⁺, ⁴⁸Ti⁺, ⁵⁰Ti⁺, and ⁵²Cr⁺ in the first step and ⁴⁷Ti⁺, ⁴⁹Ti⁺, ⁵¹V⁺, and ⁵³Cr⁺ in the second step. Murchison matrix was used as standard for Mg and K, and a mixture of perovskite and chromite was used for Ca, Ti and Cr as well as for centering ⁵¹V.

Results and Discussion: Mg-Al. ²⁶Al/²⁷Al ratios were calculated from ²⁶Mg excesses, where δ²⁵Mg/²⁴Mg values were subtracted from δ²⁶Mg/²⁴Mg values under the assumption that nuclear processes (such as neutron capture) would produce approximately equal excesses in the neutron-rich Mg isotopes [5-8]. Of 22 grains with detectable ²⁶Al, 14 grains have inferred ²⁶Al/²⁷Al ratios higher than 1×10⁻², 11 of those have ¹²C/¹³C ratios lower than solar (Fig. 1). Interestingly, also higher δ²⁵Mg/²⁴Mg values are observed in grains with ¹²C/¹³C ratios lower than solar (Fig. 2). This is puzzling because grains with high ¹²C/¹³C ratios, which are likely to be of a supernova origin, are expected to have high δ²⁵Mg/²⁴Mg values: in the He/N zone of supernovae [5, 6], ²⁵Mg is destroyed via the ²⁵Mg(p,γ)²⁶Al reaction and the ¹²C/¹³C ratio is expected to be low (8 for 15M_⊙ supernovae) due to the CNO cycle, while in the He/C zone as well as in the O/C and

O/Ne zones ²⁵Mg is produced by neutron capture. The ¹²C/¹³C ratio in the He/C zone is extremely high (10⁵) due to the triple-α reaction. Thus, to obtain lower ¹²C/¹³C ratios, material from the He/N zone had to be incorporated into the mix from which the grains formed (if they indeed formed in supernovae), bringing down δ²⁵Mg/²⁴Mg values. On the other hand, in AGB (asymptotic giant branch) stars, ²⁵Mg and ¹²C that are produced in the He-burning zone are brought into the envelope during third dredge-up episodes [7, 8]. Thus, in both types of stellar sources ²⁵Mg/²⁴Mg and ¹²C/¹³C ratios are expected to be positively correlated.

Ca. Unfortunately, for unknown reasons the analyzed mount was heavily contaminated and isotopically anomalous Ca in some of the grains was undoubtedly swamped by normal Ca. Nonetheless, 14 grains turned out to be anomalous. Seven grains have excesses in ⁴²Ca, ⁴³Ca, and ⁴⁴Ca, 2 grains in ⁴²Ca and ⁴⁴Ca. The highest δ⁴³Ca/⁴²Ca value is 691±50‰. Three grains have deficits in ⁴²Ca and one of the three in ⁴³Ca as well. Eight grains show evidence for the initial presence of short-lived ⁴¹Ca from ⁴¹K excesses; inferred ⁴¹Ca/⁴⁰Ca ratios range from 1×10⁻³ to 8×10⁻³. Because of the high level of the Ca contamination, these ratios should be regarded as lower limits.

Ti. Ti analysis was also hindered by Ca and Cr contamination. Many grains have lower than solar apparent ⁴⁷Ti/⁴⁸Ti ratios, but this is a result of large ⁴⁸Ca interferences with ⁴⁸Ti⁺. Since we did not measure ⁴⁰Ca during the Ti isotopic analysis, we tried to correct for the ⁴⁸Ca⁺ interference from ⁴⁰Ca counts obtained during the K-Ca analysis and by assuming the ⁴⁸Ca/⁴⁰Ca ratio to be solar. We discarded the data if the inferred ⁴⁸Ca/⁴⁸Ti ratio was larger than 0.1. Of the 24 grains, only 7 grains had acceptably low ⁴⁸Ca/⁴⁸Ti ratios. If no K-Ca analysis was made, grains with large negative δ⁴⁷Ti/⁴⁸Ti values (-240 to -60‰) were also discarded. Because of the large ⁵⁰Cr interference we could not determine ⁵⁰Ti/⁴⁸Ti ratios.

All 7 grains with anomalous Ti have ⁴⁹Ti excesses ranging up to 1500‰. One of them shows a V-shape isotopic pattern when normalized to ⁴⁸Ti, with about equal amounts of excesses in ⁴⁶Ti and ⁴⁹Ti (1200 and 1500‰). Another grain has a ⁴⁷Ti excess of 180‰.

Stellar Sources. Because of the contamination, we are certain that at least Si and Ca isotopic ratios of the KFA1 grains in this mount have to be regarded as upper or lower limits, depending on the anomalies. With this constraint in mind, we consider stellar sources of KFA1 graphite.

Low-density graphite grains from Murchison separate KE3 (1.65-1.72g/cm³) have been most extensively studied among the graphite fractions from this meteorite [9]. Many KE3 grains have ¹⁸O and ²⁸Si excesses

and a few grains show evidence for the initial presence of short-lived ^{44}Ti (from large ^{44}Ca excesses). Those isotopic features indicate that they formed in supernova ejecta. Some of the KFA1 grains have ^{18}O and ^{28}Si excesses, similar to KE3 grains. A supernova origin can be assigned to these grains.

Of 43 grains analyzed for O isotopic ratios, 22 grains are ^{16}O -rich, with $^{18}\text{O}/^{16}\text{O}$ ratios down to $0.75\times$ solar and $^{17}\text{O}/^{16}\text{O}$ ratios down to $0.68\times$ solar. In the early Galaxy, isotopes made from α particles such as ^{16}O (primary isotopes), were enriched relative to those produced from primary isotopes (secondary isotopes). Oxygen in the envelope in low-mass low-metallicity stars, which were born in the early stages of the Galaxy, are predicted to be ^{16}O -rich, even after the first dredge-up, and the subsequent evolution of the star does not change the O isotopic composition significantly [10]. $^{12}\text{C}/^{13}\text{C}$ ratios of the ^{16}O -rich grains range from 3.5 to 1400. If these grains were produced in low-mass low-metallicity stars and the stars experienced third dredge-up episodes, $\delta^{25}\text{Mg}/^{24}\text{Mg}$ and $\delta^{30}\text{Si}/^{28}\text{Si}$ values of the grains should increase along with $^{12}\text{C}/^{13}\text{C}$ ratios. However, a negative correlation is observed between $^{12}\text{C}/^{13}\text{C}$ ratios and $\delta^{25}\text{Mg}/^{24}\text{Mg}$ values (Fig. 2), contrary to what is expected if the grains formed in AGB stars. A better test would be to check whether there exists any correlation between Si and Ti isotopic ratios, because such a correlation has been observed in grains of an AGB star origin [11]. However, Ti data in many grains are of no use due to the overwhelming ^{48}Ca interference with ^{48}Ti .

$^{41}\text{Ca}/^{40}\text{Ca}$ ratios in the envelope of AGB stars are expected to be on the order of 10^{-4} [12] while those in the He/C, O/C and the other O-rich zones of supernovae range up to 0.02 [5, 6]. The observed ratios in the grains are thus consistent with a supernova origin.

In summary, some of the KFA1 grains most likely have formed in supernovae. However, we are not able to infer any other stellar sources, partly because of the high level of contamination of the mount. In the future, we hope to analyze clean mounts to better understand the origin of the KFA1 graphite grains.

References: [1] Amari S. et al. (1990) *Nature*, 345, 238-240. [2] Bernatowicz T. et al. (1987) *Nature*, 330, 728-730. [3] Stadermann F. J. et al. (1999) *Lunar Planet. Sci.*, XXX, Abstract #1407. [4] Amari S. et al. (2002) *Meteorit. Planet. Sci.*, 37, A11. [5] Meyer B. S. et al. (1995) *Meteoritics*, 30, 325-334. [6] Woosley S. E. and Weaver T. A. (1995) *Astrophys. J. Suppl.*, 101, 181-235. [7] Busso M. et al. (1999) *Ann. Rev. Astron. Astrophys.*, 37, 239-309. [8] Karakas A. L. and Lattanzio J. C. (2002) *Astrophys. J.*, submitted. [9] Travaglio C. et al. (1999) *Astrophys. J.*, 510, 325-354. [10] Boothroyd A. I. et al. (1994) *Astrophys. J.*, 430, L77-L80. [11] Hoppe P. et al. (1994) *Astrophys. J.*, 430, 870-890. [12] Wasserburg G. J. et al. (1995) *Astrophys. J.*, 440, L101-L104.

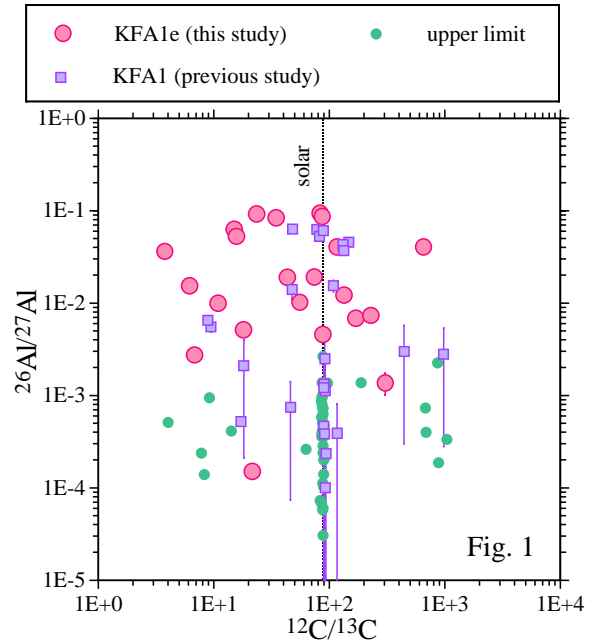


Fig. 1

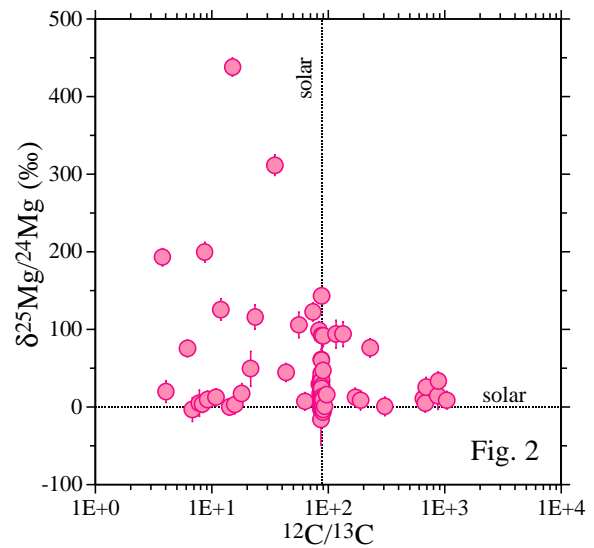


Fig. 2