OXYGEN ISOTOPIC STUDY OF HIGH-DENSITY GRAPHITE GRAINS FROM THE MURCHISON METEORITE (CM2).

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Introduction: Presolar grains are stardust that formed in stellar ejecta and were incorporated in meteorites. They have large isotopic anomalies that attest to their stellar origin. Of mineral types of presolar grains, graphite grains exhibit a range of densities (1.6 – 2.2 g/cm^3) [1]. Their $^{12}C/^{13}C$ ratios vary more than 3 orders of magnitude, ranging from 2.1 to 7223 [2]. One of the most intriguing features of graphite grains is that isotopic characteristics depend on density. There are four density fractions extracted from Murchison (CM2): KE3 (1.65 – 1.72 g/cm^3), KFA1 (2.05 – 2.10 g/cm^3), KFB1 (2.10 – 2.15 g/cm^3), and KFC1 (2.15 – 2.20 g/cm^3) [3]. Grains from KE3 and KFA1, low-density grains, show a broad distribution of $^{12}C/^{13}C$ ratios. High $^{26}Al/^{27}Al$ ratios (up to 0.15) and Si isotopic anomalies (mostly ^{28}Si excesses, but, in a few cases, $^{29,30}Si$ excesses) of many grains indicate that they formed in supernovae. Grains from KFB1 and KFC1, high-density grains, have two distinct peaks, one with $^{12}C/^{13}C$ ratios around 10, the other with ratios around 400 – 630 [2]. Many low-density grains from Murchison show ^{18}O excesses with $^{18}O/^{16}O$ ratios up to 185 times solar [2, 4], indicative of a supernova origin. In contrast, many high-density graphite grains show close-to-normal $^{18}O/^{16}O$ and $^{17}O/^{16}O$ ratios. This is attributed to isotopic exchange with close-to-normal O either in the solar system or in the laboratory, as in the case of N [2, 5]. In this study, we focused on obtaining high-precision O isotopic ratios to further investigate the affects of Galactic chemical evolution and/or other processes that might have changed the original O composition of high-density graphite grains.

Experimental: KFC1 grains, deposited onto a gold foil from suspension, were documented for their locations and grain sizes using a JEOL JSM-840A scanning electron microscope. Carbon and N isotopic ratios were analyzed using the NanoSIMS 50 at Washington University: $^{12}C^-$, $^{13}C^-$, $^{12}C^{14}N^-$, and $^{12}C^{15}N^-$ were simultaneously collected. DAG (carbon paint) was used as a standard for the C isotopes, and synthetic Si_3N_4 for the N isotopes. Oxygen isotopic ratios of 36 of the same grains were analyzed using the CAMECA IMS-1280 (WiscSIMS) at The University of Wisconsin-Madison. A focused beam of 5pA was used for analysis, and $^{16}O^-$, $^{17}O^-$ and $^{18}O^-$ were detected with electron multipliers. Terrestrial organic matter containing ~2wt.% of O (WI-STD-64, UWMA1) was used as a standard. Since this standard is usually used as a standard for C isotopic ratios [6], the O isotopic composition of this standard has not been determined yet. We estimated it from 23 terrestrial kerrogen samples [7] that are similar to our standard: we took the average of the 23 samples ($\delta^{18}O$ = 21.1 ± 10.8 ‰, 2σ) and used this average as the O isotopic composition of our standard. The errors of the data reported here are 2σ.

Results and Discussion: Carbon and N isotopic ratios of the grains agree with those of previous studies [2, 5]. Two grains have ^{18}O excesses ($\delta^{18}O$ = 1480 ± 26 ‰, and 119 ± 30 ‰), showing their supernova origin. $\delta^{18}O$ and $\delta^{17}O$ of the rest of the grains range from 40 to 80 ‰, and from 10 to 60 ‰, respectively, regardless of their $^{12}C/^{13}C$ ratios. The average values, excluding the two ^{18}O-rich grains, are $\delta^{18}O$ = 55 ± 12 ‰ and $\delta^{17}O$ = 38 ± 16 ‰.

KFC1 grains with $^{12}C/^{13}C$ ratios higher than 100 are believed to have formed in low-metallicity ($Z = 3 \times 10^{-3}$ and 6×10^{-3}, $Z_{sun} = 2 \times 10^{-2}$) AGB stars [2]. During the third dredge-up phase, $\delta^{18}O$ values in the envelope remain negative up to half a solar metallicity, while $\delta^{17}O$ values become positive at $Z = 6 \times 10^{-3}$ ($\delta^{17}O$ = 21‰) [2]. This implies that negative $\delta^{18}O$ and negative/positive $\delta^{17}O$ values are expected in these grains, which is not consistent with our observations of positive values for both $\delta^{18}O$ and $\delta^{17}O$. Fujiya [8] estimated the O isotopic composition of primordial water in CM chondrites from the O isotopic compositions of anhydrous silicates [9] and phyllosilicates [10] in CM chondrites. The $\delta^{18}O$ and $\delta^{17}O$ values of primordial water in CM chondrites were calculated to be 65 ‰ and 43‰, respectively. The average $\delta^{18}O$ and $\delta^{17}O$ values of our grains agree with this estimation, suggesting that the O isotope compositions of high-density graphite grains were affected by aqueous alteration in the Murchison parent body, and that the O in high-density graphite grains does not reflect the Galactic chemical evolution.

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