IS THERE PRESOLAR DUST IN THE ALLENDE CURIOUS MARIE CALCIUM-ALUMINUM-RICH INCLUSION?

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Introduction: Presolar grains have been identified in primitive chondrites based on their large isotopic anomalies, which were inherited from their parent stars that died prior to the solar system formation [1]. The most abundant presolar mineral phase is silicates, which can be identified only in situ by high-resolution isotopic mapping and are up to several hundred ppm in primitive fine-grained matrix [2]. In comparison, presolar silicon carbide (SiC) has been found in both meteoritic acid residues (SiC is acid resistant) and primitive fine-grained matrix. The inferred comparable presolar SiC abundances (a matrix normalized abundance of ~30 ppm) between the acid residues and the matrix suggest that primitive fine-grained matrix is the dominant host of presolar dust [3]. In contrast to this commonly held view, a recent study observed the release of *s*-process noble gas isotopic signatures at high temperatures during stepwise heating of *Curious Marie* [4], a fine-grained Ca, Al-rich inclusion (CAI) from Allende [5]. The authors suggest that presolar SiC, a known *s*-process noble gas carrier [6], was incorporated into fine-grained CAIs in the Allende carbonaceous chondrite and have survived parent-body processing [4]. It, however, remains possible that the observed *s*-process carrier was transported from the surrounding matrix to the CAI interior by a fluid because *Curious Marie* experienced extensive aqueous alteration [5]. Secondary processes aside, the *s*-process carrier may not be presolar SiC, as other refractory presolar phases such as oxides could also explain the required *s*-process isotopic signature and grain abundance. Furthermore, in fine-grained CAIs, oxides are more likely than SiC to survive from oxidation at high temperatures during CAI formation. Thus, we were motivated to conduct an in-situ search of presolar grains in *Curious Marie* by high-resolution isotopic imaging using the Cameca NanoSIMS 50 microprobe at Washington University in St. Louis. Our goal is to investigate whether fine-grained CAIs host any presolar dust and, if so, whether presolar SiC survived from high-temperature oxidation during fine-grained CAI formation.

Experimental Methods: We studied both the original *Curious Marie* section (Fig. 1a) [4,5] and a new section consisting of one edge of *Curious Marie* and its adjacent Allende matrix (Fig. 1b). We first obtained backscattered electron (BSE) maps of the two sections in a scanning electron microscope (SEM). We then carried out NanoSIMS C and O isotope ion imaging analyses in both the matrix and fine-grained CAI samples with a total analysis area of ~12,000 and 30,450 μm^2, respectively. We also obtained $^{12}C^-$, $^{13}C^-$, $^{12}C^{14}N^-$, $^{12}C^{15}N^-$ and $^{28}Si^-$ ion images of 26,775 μm^2 of the fine-grained CAI to further examine if presolar SiC is present. The measurements were made in semi-automated mode and involved rastering a ~1 pA Cs^+ ion beam over 15 × 15 μm areas within larger (18 ×18 μm) areas presputtered to remove the Au or C coat. The primary Cs^+ beam was ~100–150 nm in size for all the analyses. The NanoSIMS analysis conditions were comparable to those adopted in the literature for in situ search of presolar grains in fine-grained matrix [e.g., 7]. Isotope images were reduced using the L'Image software [7].

Results: We searched for presolar O-rich and C-rich grain candidates by manual examination of the isotope ratio images. A candidate is confirmed if at least one isotope ratio of carbon and oxygen is >4 σ away from the surrounding meteorite matrix. With this identification criterion, we identified zero C-anomalous grains in our NanoSIMS isotope measurements. Our detection of no C-anomalous grains translates to an upper limit of 2.1 ppm and 7.5 ppm (2σ error, single sided [8]) presolar SiC in *Curious Marie* by assuming a mean grain diameter of 0.20 μm and 0.38 μm, respectively. In comparison, our data give an upper limit of 36 ppm presolar SiC in Allende matrix by adopting the average presolar SiC grain diameter, 0.38 μm, reported in [3]. Also, we did not
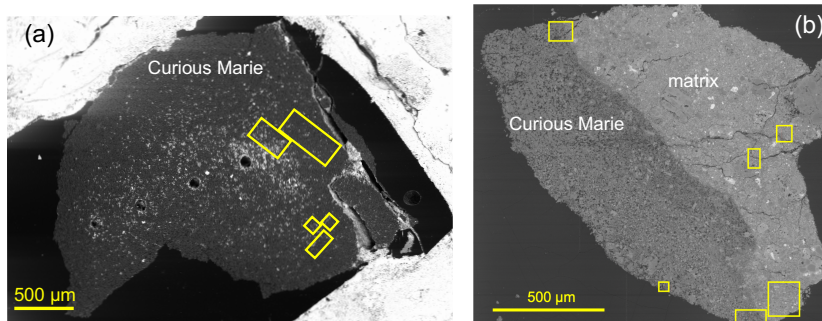

Fig. 1. BSE maps of (a) the original *Curious Marie* section and (b) a new section consisting of fine-grained CAI (dark-colored) and matrix (light-colored) in Allende. NanoSIMS analysis areas are highlighted in yellow.

identify any C-rich grains with C/Si of ~1 in the C-N-Si ion images, further excluding the potential existence of SiC in *Curious Marie*.
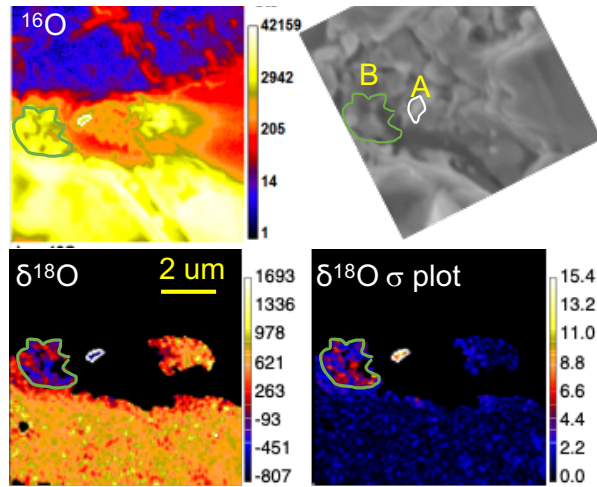

Fig. 2. Images of area A9-08 in *Curious Marie* that contains two ^{18}O-depleted grains A and B (highlighted by white and green contours, respectively). The SE image (upper right) was obtained with an Auger nanoprobe.

Out of 140 C and O isotope images (15×15 μm) for *Curious Marie*, we identified two regions in area A9-08 that had significant ^{18}O depletions (Fig. 2). In the "σ" plot of Fig. 2, each pixel represents the number of standard deviations (Poisson statistics) by which its measured isotopic ratio is away from the terrestrial value. During the automated analysis of area A9, we collected C and O ion images of 20 15×15 μm areas, and none of the δ^{18}O "σ" plots of the other 19 areas exceeds a maximum of 8σ deviation. In comparison, the maximum of the "σ" plot for A9-8 is almost twice as high, meaning that the identified ^{18}O depletions are indeed statistically significant when analytical uncertainties, *e.g.*, long-term drift, topographic and matrix effects, are taken into account (estimated based on variations in the data of the other 19 areas). The identified O-anomalous grain A (white contour) is 0.55 μm in diameter and has an O isotopic composition of δ^{17}O = −124±137‰ and δ^{18}O = −726±32‰. The identified O-anomalous grain B (green contour) is 1.9 μm in diameter and has an O isotopic composition of δ^{17}O = 75±59‰ and δ^{18}O = −398±10‰. The ^{18}O depletions of both the grains were present through all measurement cycles. According to the SE image in Fig. 2, grain B seems to be an aggregate instead of a single grain. We attempted Auger analyses of grains A and B to characterize their compositions. However, we could not obtain reliable Auger spectral data due to significant sample charging. The identification of O-anomalous grains A and B over 30,450 μm^2 areas corresponds to a presolar O-rich grain abundance of 101^{+270}_{-89} ppm (2σ error, single sided).

Discussion: Previous studies suggested that presolar SiC is destroyed in fine-grained matrix when heated to above 400 °C [3,6]. The lack of presolar SiC in Allende matrix shown by our data is in line with this suggestion and excludes the possibility that presolar SiC was transported from adjacent matrix to *Curious Marie* by aqueous alteration. The presolar SiC abundance inferred by the *s*-process noble gas data of *Curious Marie* are 7 ppm and 25 ppm for a mean grain diameter of 0.20 μm and 0.38 μm, respectively [4]. In comparison, our in situ NanoSIMS survey of *Curious Marie* points out an abundance of at least three times lower, questioning the proposal of SiC being the carrier of *s*-process noble gas by [4]. Our data suggest that refractory oxides could instead explain the *Curious Marie*'s release of *s*-process noble gas at high temperatures. The O isotopic composition of grains A and B, however, seem peculiar when compared to presolar silicates and oxides identified in fine-grained matrix. This could suggest either that *Curious Marie* incorporated a different population of presolar grains or that there was some unidentified analytical artifact in the O isotope analysis. We plan to remeasure the O isotopic compositions of grains A and B by using a smaller raster to confirm the O isotopic anomalies.
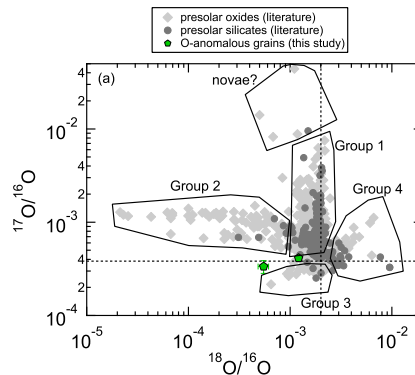

Fig. 3. Oxygen 3-isotope plot comparing the O-anomalous grains from this study with the literature presolar oxide and silicate data.

Conclusion: We did not identify any presolar SiC grains in *Curious Marie* by in situ NanoSIMS C and O isotope survey. The upper limit of our inferred presolar SiC abundance is a factor of three lower than the result of [4]. Identification of two O-anomalous candidates suggests that presolar oxides could instead be the carrier of the *s*-process noble gas identified by [4].

References: [1] Nittler L. R. & Ciesla F. (2016) *ARA&A, 54*, 53–93. [2] Floss C. & Haenecour P. (2013) *Geochem. J., 50*, 3–25. [3] Davidson J. et al. (2014) *GCA, 139*, 248–266. [4] Pravdivtseva et al. (2020) *Nat. Astron., 4*, 617–624. [5] Tissot F. L. H. et al. (2016) *Sci. Adv., 2*, e1501400. [6] Huss G. R. & Lewis R. S. (1995) *GCA, 59*, 115–160. [7] Nittler et al. (2018) *GCA, 226*, 107–131. [8] Gehrels N. (1986) *ApJ, 303*, 336–346.