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Morphologies, Isotopes, Crystal Structures, and Microstructures of Presolar Al_2O_3 Grains: a NanoSIMS, EBSD, EDS, CL, and FIB-TEM study

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Presolar grains are rare sub-micron-to-micron-sized components of primitive chondrites and interplanetary dust particles (IDPs), and are identified on the basis of their highly anomalous isotopic compositions [e.g., 1]. They formed around evolved stars prior to Solar System formation, and survived impact and radiation processing in the interstellar medium and early Solar System, before eventual incorporation into comets and asteroids. The grain morphology and crystal structure of presolar grains will reflect condensation conditions in circumstellar envelopes of asymptotic giant branch (AGB) stars, as well as processing in the interstellar medium (ISM) and protosolar disk. Transmission electron microscopy (TEM) studies of presolar oxide grains have been limited, however, due to their small sizes and low abundance relative to oxide grains with solar isotopic compositions [e.g., 2, 3]

Corundum, the thermodynamically stable phase of Al_2O_3, is observed to be the most abundant refractory dust species in envelopes around oxygen-rich AGB stars. There are few studies of the morphology of presolar Al_2O_3 grains [4, 5] because the isotopic measurements necessary to identify the presolar grains amorphize the grain surfaces and consume a substantial fraction of the grain volume. In order to reveal the relationship between the morphology, isotopic compositions, crystal structure, and microstructures of individual presolar grains, we have obtained detailed secondary electron images, energy dispersive X-ray spectra (EDS), electron backscattered diffraction (EBSD) patterns, and cathodoluminescence (CL) spectra for 280 Al_2O_3 grains using field-emission scanning electron microscopy (FE-SEM) prior to the isotopic measurements. Focused ion beam (FIB) lift-out sections were made from the identified presolar grains and the interior structures were observed with TEM.

The Al_2O_3 grains were identified from an acid residue of QUE97008 (LL3.05) by EDS and observed in detail by FE-SEM at the Carnegie Institution of Washington (CIW). Previously identified alumina grains from Semarkona (LL3.0), Roosevelt County 075 (H3.1), and Bishunpur (LL3.15) were also used in this study [5]. We obtained CL spectra of each Al_2O_3 grain using an array detector in the spot-mode with an acceleration voltage of 6 kV and an electron probe current of ~5 nA with a FE-SEM equipped with a Gatan MonoCL4 Elite system at National Institute of Standards and Technology. EBSD analysis was performed with an FEI Nova 600 FIB-SEM equipped with an HKL EBSD system at the Naval Research Laboratory (NRL).

Isotope measurements were performed with the Cameca NanoSIMS 50L ion-microprobe at CIW. Oxygen isotopes of 163 grains were measured with a ~100 nm Cs^+ beam rastered over each of the grains. Secondary ions of $^{16}O^-$, $^{17}O^-$, $^{18}O^-$, $^{27}Al^{16}O^-$, and $^{24}Mg^{16}O^-$ were simultaneously detected. An O^-

beam was subsequently used to measure the Mg-Al isotopic compositions of the presolar and some solar Al_2O_3 grains. Secondary ions of $^{24}Mg^+$, $^{25}Mg^+$, $^{26}Mg^+$, $^{27}Al^+$, and $^{48}Ti^+$ were simultaneously measured.

Eight presolar grains from QUE97008 and one from RC 075 were newly identified. Ultra-thin sections of selected grains were prepared with the FIB-SEMs at NRL (FEI Nova 600 FIB-SEM) and CIW (Zeiss Auriga 40) and TEM studies were carried out at NRL with a JEOL 2200FS FE-STEM.

We made FIB sections of seven presolar grains. Five of them were observed with TEM and electron diffraction revealed all to be corundum. Grain QUE018, for which O-isotopic compositions indicate an origin from a low-mass AGB star [1], has an irregular morphology covered with a relatively even face and other rough surfaces with cracks (Fig. 1a and 1d). EBSD patterns of QUE018 taken prior to SIMS measurements indexed to that of corundum, indicating that the surface of the grain (<30 nm in depth) is corundum (Fig. 1b). No elements other than Al and O were detected by EDS. No Mg isotopic anomalies nor a signal of ^{48}Ti were detected by the NanoSIMS, although ^{40}Ca was detected on the left side of the grain. The CL spectrum displayed several peaks at ~1.7 and 2.1-2.6 eV that were not observed on a micron-sized corundum grain prepared by crushing a single crystal (Fig. 1c); these peaks may reflect the internal defect structure of the presolar grain. Electron diffraction patterns of the QUE018 FIB section indicate multiple corundum crystallites with slight rotational variation of the lattice across the grain. Two other grains (QUE060 and QUE137) include internal voids. Grain QUE060 has a subhedral shape with facets and a cavity. The bright-field TEM images show that the voids (10-20 nm) were segregated to one side of the grain and seem to relate to the cavity. Grain QUE137 has one flat face, which seems to have formed by shear splitting from a larger grain. Many voids were observed across the grain in the bright-field image.

The morphology and microstructures of presolar Al_2O_3 grains suggest that the cracks, voids, and polycrystalline nature are secondary features after grain formation in the circumstellar envelopes. A possible process for forming these features is grain-grain collisions in a supernova shock in the ISM. Acid etching during the chemical treatments necessary to isolate these grains from the meteorites may also have changed the surface structures and cracked the grains.

References:

[1] L. R. Nittler et al. (2008) *Astrophysical Journal,* **686,** 1524.

[2] R. M. Stroud et al. (2004), *Science,* **305,** 1455.

[3] T. J. Zega et al. (2014) *Geochimica et Cosmochimica Acta,* **124,** 152.

[4] B. G. Choi et al. (1998) *Science,* **282,** 1284.

[5] A. Takigawa et al. (2014) *Geochimica et Cosmochimica Acta,* **124,** 309.

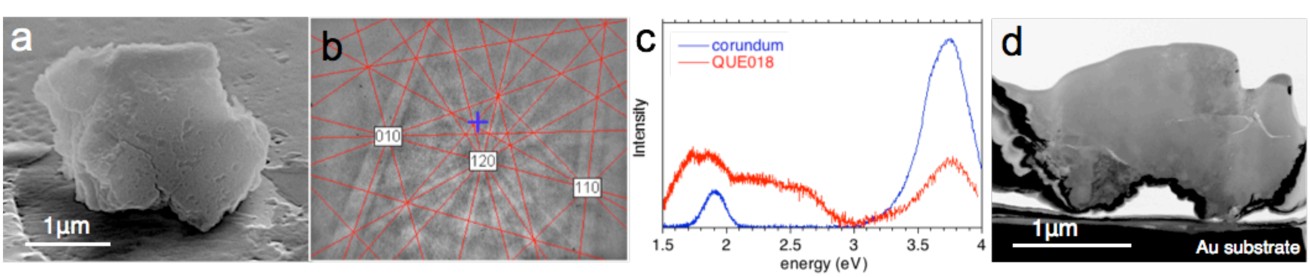

Figure 1. (a) SEM image and (b) EBSD pattern of presolar QUE018. (c) Cathodoluminescence spectra of presolar grain QUE018 and micron-sized single crystal corundum. (d) Dark-field TEM image of the FIB lift-out section of QUE018.