COORDINATED TEM AND NANOSIMS OXYGEN ISOTOPE ANALYSIS OF INTERPLANETARY DUST PARTICLES PREPARED BY FOCUSED ION BEAM. C. E. Jilly-Rehak[1], Z. Gainsforth[1], A. L. Butterworth[1], S. Hsiao[2], K. Naito[2], H. Shang[3], J. S. Rehak[4], and A. J. Westphal[1]. [1]Space Sciences Laboratory, University of California Berkeley, Berkeley CA 94720; jillyrehak@berkeley.edu. [2]Institute of Earth Science, Academia Sinica, 11529, Taipei, Taiwan. [3]ASIAA, Institute of Astronomy and Astrophysics, Academia Sinica, 11529, Taipei, Taiwan. [4]Department of Nuclear Engineering, University of California Berkeley, Berkeley CA 94720.

Introduction: Chondritic-porous interplanetary dust particles (CP-IDPs, likely originating from comets [1]) contain a complex mixture of coarse crystalline phases surrounded by an ultra-fine-grained matrix. The coarse crystalline component is typically a mixture of high-temperature silicates, such as olivine and pyroxene, and sulfides. The finer surrounding matrix is rich in silicate material, both nanocrystalline and amorphous, primitive carbonaceous material, and GEMS (Glass with Embedded Metal and Sulfide).

Though most comets are thought to have formed in the outer Solar System – either in the Kuiper belt or in the more distant Oort Cloud – oxygen isotope analyses of comet grains (including those returned by the Stardust mission) suggest incorporation of material from various reservoirs, with high temperature phases likely originating from the inner Solar System [e.g., 2-7].

The objective of this study is to develop a new method for coordinated TEM and isotopic analysis of IDP components. Most bulk and multiphase IDP analyses from literature lack petrographic context [4,5] – we aim to take oxygen isotope analyses of individual mineral grains and components (e.g., GEMS-rich fine grained material) inside of an IDP to determine their contributions to the bulk O-isotope trends. Such coordinated TEM/SIMS analyses can help to determine the origin of IDP components, thereby helping to constrain disk evolution and transport models.

Sample Preparation and Methods: Four IDPs were prepared for coordinated TEM and NanoSIMS analysis: Three from Cluster 17 (L2071,17), here named Particle 4, Humpty, and Dumpty, and one additional IDP L2076, R1, referred to here as particle R1. Humpty and Dumpty are two IDPs in one FIB section (Fig. 1).

Focused Ion Beam (FIB): Ultra-thin sections of IDPs and San Carlos olivine (standard) were prepared using the FEI Strata 235 dual beam FIB at the National Center for Electron Microscopy (NCEM) at Lawrence Berkeley National Lab (LBL). Sections were taken directly from particles on nucleopore filters. Some advantages of FIB over microtomy are that FIB preserves the petrographic context of the fine-grained material, a section can be precisely positioned to extract desired regions, and no additional handling or embedding of the particles are necessary. This comes at a slight cost compared to ultramicrotomy in that FIB does not preserve as much of the particle.

For each IDP, a conductive Pt strap was deposited over the top. A 30 keV Ga^+ ion beam was used to mill the samples to ~150-250 nm. FIB sections were attached to a Cu TEM half-grid with Pt. Final thinning and cleaning of the section at 10 keV helped remove any surficial amorphous material created during the FIB section process. After TEM analysis, the IDP sections were removed from the Cu half-grids and affixed to a 1-cm-round polished stainless steel disc using Pt deposition in the FIB. San Carlos olivine sections were prepared using the same methods, and were also affixed to the disc. The disc was coated with 20 nm of Au to ensure conductivity for NanoSIMS analysis.

Transmission Electron Microscopy (TEM): To investigate the petrography of the IDPs, we used the FEI Titan 60-300, the Philips CM200/FEG, and the Zeiss LIBRA 200MC at NCEM. The Titan Bruker and CM200 Oxford EDX detectors were used for imaging and EDX analysis and mapping. We used a Zeiss Libra 200MC TEM with an in-column Omega energy filter for imaging and diffraction. Beam voltages were typically set to 200 keV.

NanoSIMS: The NanoSIMS 50L at Academia Sinica in Taiwan was used for isotopic analysis. We measured ^{16}O, ^{17}O, ^{18}O, ^{28}Si, ^{32}S, and $^{24}Mg^{16}O$, in imaging mode. The mass resolving power was sufficient to resolve $^{16}OH^-$ from $^{17}O^-$. A 3 pA Cs^+ ion beam of ~250 nm was used to raster across 5 × 5 μm areas, each taken at an image resolution of 64 × 64 pixels at 3000 μs/px dwell time. Analyses ranged from 50-300 cycles. There were two analysis regions per IDP, since each FIB section was ~10 μm wide, and each region in particle R1 was analyzed in multiple runs of 50 cycles each. The San Carlos olivine FIB sections were used as standards.

Data Reduction Techniques: Preliminary results were processed using L'Image NanoSIMS software (by L. Nittler). However, due to the complex, fine-scale heterogeneous structure of the IDPs, we needed to define regions of interest (ROI) for phases that fade in and out throughout the analysis cycles, essentially creating 3-dimensional ROIs in the X-Y-cycle datacube. To address this, we developed a python code that visualizes, and calculates ratios for 3D ROIs. The python program reads the raw data (with the assistance of SIMS python package created by Z. Peeters to extract raw data from the .im files), corrects for a deadtime of 44 ns (method of [8]) and performs a QSA correction ($\beta = 0.75$ [9]).

The program allows the user to mask the data (here we use a threshold of ^{16}O counts ≥ 200 per pixel to eliminate background), and calculates ratios using the total counts of the ROI per run. Instrumental mass fractionation (IMF) was corrected using the weighted mean of standard compositions for a given day. Uncertainties on the standards represent the 2σ standard error, and uncertainties on the unknowns are 2σ propagated errors for the standard deviation of the unknowns and the standard error for the standard analyses.

For component analysis, the python program outputs the data to a .vtk file, which is then read using open-source program Paraview where we manually select grains and analyze data from the 3D image render using python programmable filters (Fig. 2).

Results and Discussion: Petrography of IDPs Particle 4 and Humpty and Dumpty was previously reported in [10]. Particle R1 contains three coarse sulfides ranging from 1-4 μm in size, as well as two coarse ~1 μm olivine crystals. One of these olivine grains is euhedral and Fe-rich, with composition Fo_{63}, the other has composition Fo_{73}. These grains are embedded in fine-grained and amorphous silicate matrix littered with ~50 nm sulfides and carbon-rich material.

On a standard oxygen three-isotope plot, the bulk data from particle R1 (calculated over the whole 5 μm raster) plot near chondritic silicates around $\delta^{17}O$, $\delta^{18}O = 0$, while Particle 4 and Humpty and Dumpty extend towards negative values along the TFL (Fig. 3). The large deviation in values suggests incorporation of material from various reservoirs. The data are roughly consistent with previously measured IDPs [4,5]. For the individual components, olivine crystals from the IDPs tend to plot close to chondritic silicates, while the fine-grained matrix material is less $^{17,18}O$-enriched (Fig. 3).

The trends along the TFL occur on the thinner, more fine-grained samples that are easily ablated, and on long cycles. This suggests that the mass-dependent fractionation may be partly instrumental, likely due to changing topography as analysis progressed (see Fig. 1c, analogous to crater bottom-roughness fractionation observed in [11]). We are working on improving this method by using thicker samples (up to ~300-500 nm) to increase precision, and to potentially reduce other causes of mass-dependent fractionation.

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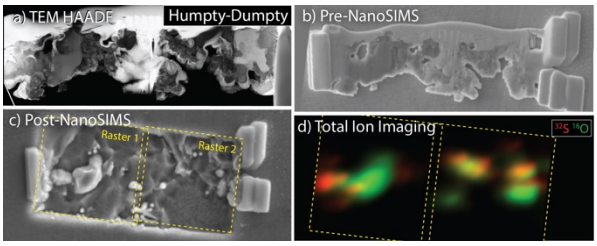

Fig. 1: Images of IDPs Humpty and Dumpty. **a)** TEM high-angle annular dark field image **b)** Secondary Electron (SE) image of the FIB section after mounting on steel disk, before NanoSIMS analysis. **c)** SE image of NanoSIMS pits after analysis. **d)** Total ion image of NanoSIMS results, $^{32}S^-$ in red, and $^{16}O^-$ in green, essentially showing silicate-rich and sulfide-rich areas.

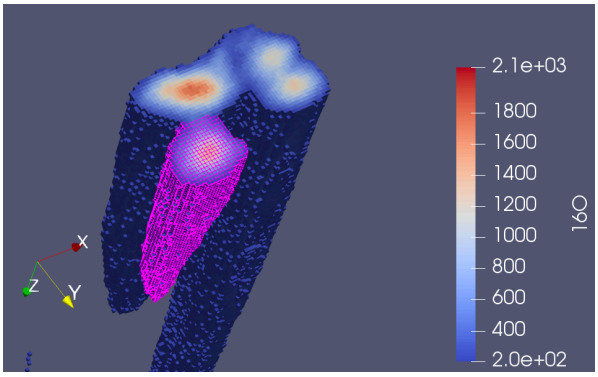

Fig. 2: Image of grain selection in Humpty using Paraview. Selection shown in pink. Color bar is ^{16}O counts, XY plane is the ion imaging raster and Z is cycles.

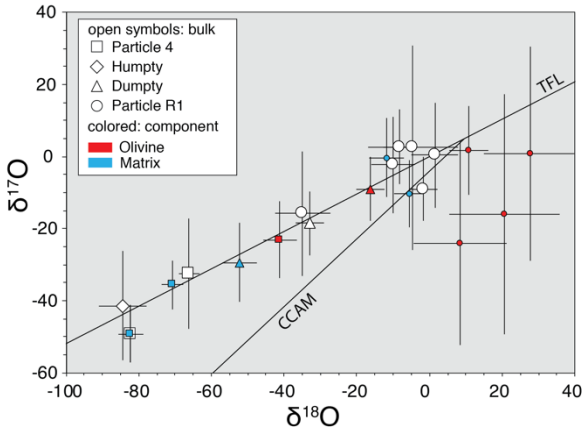

Fig. 3: IDP bulk and component O-isotope analyses in this study. Terrestrial fractionation line (TFL) and carbonaceous chondrite anhydrous mineral line (CCAM) shown for reference.