

STANDARDIZATION AND CORRECTION OF ARTIFACTS IN ATOM-PROBE TOMOGRAPHIC ANALYSIS OF ALLENDE NANODIAMONDS

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Introduction: The origins of meteoritic nanodiamonds (NDs) remain an enigma. Isotopic anomalies in trace elements, chiefly Xe, in bulk meteoritic acid dissolution residues [1], suggest a Type II supernova origin, while the bulk measured C [2] and N [3] are consistent with formation in the solar system. Poorly-crystalline to amorphous carbonaceous phases, which can comprise a significant fraction of the residue [4], are potentially of different origin than the NDs. Measurements of the $^{12}\text{C}/^{13}\text{C}$ ratios of individual (on average ~ 2.6 nm diameter) ND grains are essential to understanding where they formed. Efforts are ongoing to conduct such measurements using atom-probe tomography (APT) [5,6]. Carbon isotope abundances measured by APT yield biased isotopic ratios due to instrumental artifacts; we are investigating and quantifying these artifacts and improving our standards and data reduction methods to optimize isotopic ratio measurements.

Experimental: Needle-shaped sample “nanotips” with a radius of 20–100 nm, containing NDs or terrestrial detonation ND standards embedded between sputter-deposited Pt layers, are prepared by focused ion-beam microscope liftout. APT was performed with a Cameca LEAP 4000X Si [5,6], yielding 3D positions and time-of-flight data for $\sim 57\%$ of the atoms. Reconstructed data are corrected for background noise, interferences from other mass peaks, and detector deadtime effects [7]. To investigate remaining artifacts in isotopic measurements we conducted complementary APT/secondary ion mass spectrometry (SIMS) measurements on a ^{13}C enriched diamond superlattice [8], employing a Cameca NanoSIMS 50.

Results: The ND standards reproduce the significant instrumental artifacts noted in prior work, which lead to an underestimation of $^{12}\text{C}/^{13}\text{C}$ ratios [5,6,7]. For the few hundred Allende NDs measured to date, we do not detect significant isotopic deviations from the standards, suggesting that these NDs most likely formed in the early solar system. The ^{13}C -rich layer in the superlattice was measured by APT to have a $^{12}\text{C}/^{13}\text{C}$ ratio of ~ 0.1 , in good agreement with NanoSIMS measurements ranging from 0.1 to 0.2. These measurements will provide valuable reference data about hydride formation, charge state distribution, and molecular cluster formation.

References: [1] Lewis R. S. et al. 1987. *Nature* 326:160–162. [2] Russell S. R. et al. 1996. *MAPS* 31:343–355. [3] Marty B. et al. 2011. *Science* 332:1533–1536. [4] Stroud R. M. et al. 2011. *ApJL* 738:L27–L31. [5] Heck P. R. et al. 2014. *MAPS* 49(3):453–467. [6] Isheim D. et al. 2013. *Microsc Microanal* 19(Suppl 2):CD974–CD975. [7] Lewis J. B. et al. 2015. *Ultramicroscopy*, submitted. [8] Watanabe H. and Shikata S. 2011. *Diam Relat Mater* 20:980–982. [9] This work is supported by NASA grants NNX14AP15H (J.B.L.) and NNX13AF53G (C.F.). The atom-probe tomograph at the Northwestern University Center for Atom-Probe Tomography (NUCAPT) was acquired with support from NSF and ONR.