

SEARCH FOR ORGANIC NANOGLOBULES IN CARBONACEOUS CHONDRITES USING MICROTOMOGRAPHY. T. Matsumoto¹, A. Tsuchiyama¹, K. Nakamura-Messenger², M. E. Zolensky², T. Nakano³, and K. Uesugi⁴, ¹Department of Earth and Space Science, Osaka University, 1-1 Machikaneyama-cho, Toyonaka 560-0043, JAPAN, matsumoto@astroboy.ess.sci.osaka-u.ac.jp, ²NASA Johnson Space Center, Houston, TX, 77058, USA, ³Geological Survey of Japan / National Institute of Advanced Industrial Science and Technology, Tsukuba, 305-8567 JAPAN, ⁴SPring-8 / Japan Synchrotron Radiation Research Institute, Mikazuki, 679-5198 JAPAN.

Introduction: Primitive solar materials have various organic matters. In recent years, novel organic materials called organic nanoglobules of a few hundred micrometers in typical size were discovered in carbonaceous chondrites [1,2], IDPs [3], and comet 81P/Wild 2 [4]. The organic globules are spherical shape and in many cases with hollow structures. Composition of the globules are mainly aromatic carbon [4]. The isotopic anomalies of δD and δN^{15} observed in the globules [3,5] indicate that they were formed from photochemical reaction to ice particles at very low temperature environment, such as molecular clouds or outer protosolar disk [5]. Aqueous alteration of organic matters [6] and the gamma-ray irradiation to PAH [7] are also suggested as alternative possible formation processes. If the globules are made from organic ice particles, the hollow regions of the globules are suggested to be once filled with volatile H₂O-rich organic ices, while if they were formed by aqueous alteration, the hollow regions should be filled with a fluid which caused the aqueous alteration. However, fluids in the globules have not been detected so far in the previous studies. If fluids were originally preserved in the hollows, they might be lost during destructive processes of sample separation or preparation for TEM observation.

X-ray computed tomography (CT) is a non-destructive method which can determine 3-D internal structures of objects. SR (synchrotron radiation)-based imaging microtomography can give submicron spatial resolution [8] and was applied to micro textures in extraterrestrial materials, such as cometary grains captured by the Stardust mission [9]. If organic globules are observed non-destructively in carbonaceous chondrites by tomography, we can check the presence of fluids in the hollows. If fluids are preserved, we may analyze chemical and isotopic compositions of the fluids. The purpose of this study is to observe organic nanoglobules using imaging tomography for future analysis.

Experiments: Tagish Lake, Bells (CM), Ivuna (CI) and Orgueil (CI) meteorite are chosen for the present study. Three to six particles of few tens of micrometers in size selected from each meteorite were observed using an imaging tomographic system with a Fresnel zone plate at BL47XU in SPring-8, JAPAN [8]. The photon energy was 7.0 keV and CT images were

reconstructed by 1800 projections. Successional 800 slices were obtained for 3-D structure of each sample. The voxel size in CT images is $40.8 \times 40.8 \times 40.8$ nm, which gives effective spatial resolution of about 200 nm. After the imaging, some of the samples were embedded in epoxy, and microtomed to ultra-thin sections for observations under an transmission electron microscope (TEM). CT images which match individual TEM images were made using image analysis technique by slicing in different directions from the original CT slice direction. The CT and TEM images were compared to determine whether organic globules are observed in CT images.

Results: An example of CT images of Tagish Lake meteorite sample is shown in Figure 1a. We can see many spherical objects with hollows of several hundred nm to $\sim 1 \mu\text{m}$ in size in all samples, as candidates of organic globules. However, we cannot conclude that they are really organic globules due to CT artifact attributed to refraction by X-ray (refraction contrast); solid spherical objects might be reconstructed as hollowed objects.

The comparison between the CT image (Fig.1) and TEM image (Fig.2a) of almost the same section shows that at least some of the spherical objects with hollows are organic globules (Fig.2b). We can estimate materials from CT values, which are quantitative expression of brightness of CT images and proportional to linear attenuation coefficients (LACs) of material under a monochromatic X-ray beam in principle, in CT images. In the present study, however, we cannot examine whether or not any fluid is present in the hollows from CT values due to the refraction contrast.

Discussions: The three dimensional-shapes of the globule identified in CT images were examined. The external and hollow shapes were extracted from the CT images manually and approximated as three-axial ellipsoids with a-, b-, and c-axes. The external shape of the globule in Figure 1 has axes of $a=465$ nm, $b=573$ nm, $c=599$ nm, while the hollow of $a=203$ nm, $b=235$ nm, $c=334$ nm, respectively. On the other hands, the radii of whole globule and hollow in the TEM image (Fig.2b), 343 nm and 248 nm, respectively, are smaller than those in the CT images. Although this discrepancy results from the refraction contrast, the relative sizes of

the a-, b- and c-axes are not largely affected by the refraction contrast. Thus, the globule is not really spherical but oblate with the aspect ratio of about 0.8. The number density of globule candidates in Tagish Lake is about $1 \times 10^{-3} \mu\text{m}^{-3}$, which is smaller than that estimated by TEM ($5 \times 10^{-2} \mu\text{m}^{-3}$) [5]. This result is not consistent with the expectation that the spherical objects with hollows in the CT images is the organic globules, because only large objects can be recognized in the CT images.

As mentioned above, we cannot examine from the CT images whether or not fluid is preserved in the hollows of organic globules. However, we can recognize the positions of the globules, which are embedded in the present samples. Figure 3 shows CT slice of the sample vertical to the images of Figures 1 and 2, where an organic globule candidate is present at depth of 8.0 μm from the microtomed surface. If we can grind the sample to the position right above the globule, we may analyze a fluid in this globule using a microanalysis, such as nano SIMS, if fluid is reserved.

References: [1] Nakamura K. et al (2002) *Int. J. Astrobiol.*, 1, 179 [2] Garvie L.A.J. and Buseck P.R. (2004) *Earth. Planet.Sci.Lett.*,224, 431-439. [3] Messenger S. et al. (2008) *LPS XXXIX*, Abstract #2391. [4] De Gregorio B. T. (2009) *LPS XXXX*, Abstract #1130. [5] Nakamura-Messenger K. et al (2006) *Science*, 314 1439-1442. [6] Dworkin J.P. et al. (2001) *Proc. Nat. Acad. Sci.*, 98,815-819. [7]Saito M. and Kimura Y. (2009) *ApJ*,2,L147 [8] Uesugi K. et al (2001b) *Proc.SPIE.*,4503, 291. [9] Nakamura T. et al. (2008) *Science*.321.1664-1667.

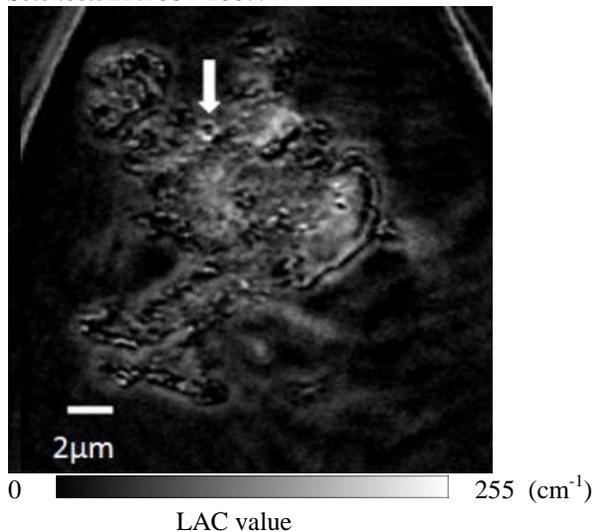


Figure 1. A CT image of a Tagish Lake meteorite sample. This slice in a direction which is different from the original slice direction was made to match the TEM image in Figure 2a. An arrow shows a candidate of an organic globule.

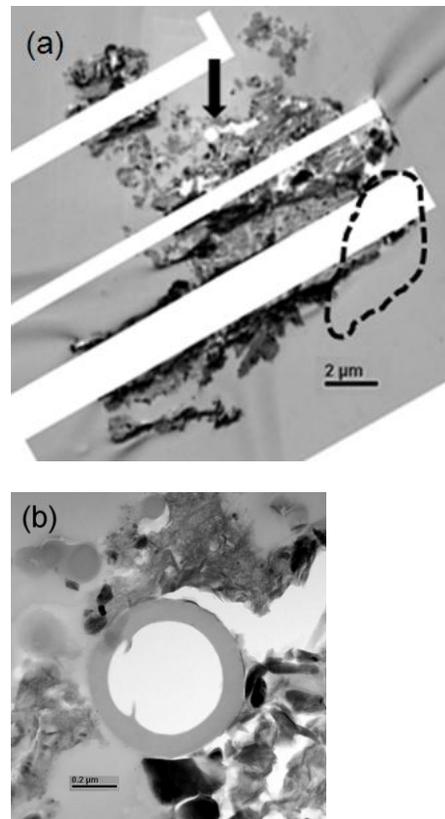


Figure 2. A transmission electron microscope images of a Tagish Lake meteorite sample. (a) An ultra-thin section corresponding to the CT image of Figure 1. A dotted area corresponds to a grain plucked during sample preparation. Diagonal white stripes roughly show folded areas of the section. (b) Close-up of the organic globule indicated by an arrow in (a).

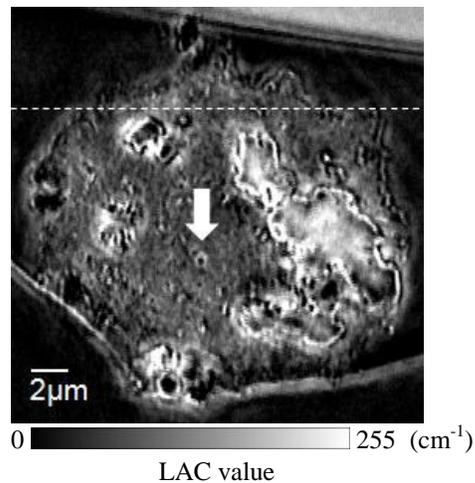


Figure 3. A CT image of slice vertical to the images of Figures 1 and 2. A dotted line shows the position of the microtomed surface. An arrow shows a candidate of organic globule, which is located at depth of 8.0 μm from the surface.