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Three-dimensional observation and morphological analysis of organic nanoglobules in a carbonaceous chondrite using X-ray micro-tomography

Toru Matsumoto[a,*], Akira Tsuchiyama[a], Keiko Nakamura-Messenger[b,c], Tsukasa Nakano[d], Kentaro Uesugi[e], Akihisa Takeuchi[e], Michael E. Zolensky[c]

[a] *Department of Earth and Space Science, Osaka University, 1-1 Machikaneyama-cho, Toyonaka 560-0043, Japan*
[b] *ESC/Jacobs Technology, Houston, TX 77058, USA*
[c] *NASA Johnson Space Center, Houston, TX 77058, USA*
[d] *Geological Survey of Japan/National Institute of Advanced Industrial Science and Technology, Tsukuba 305-8567, Japan*
[e] *SPring-8/Japan Synchrotron Radiation Research Institute, Sayo 679-5198, Japan*

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Abstract

Organic nanoglobules are submicrometer spherical, often hollow organic grains ubiquitously distributed throughout primitive solar materials, such as carbonaceous chondrites. Until now, organic nanoglobules have been examined by TEM only after sectioning by ultramicrotomy so it has not been possible to determine whether fluids or mineral grains occur in the hollow cores. H_2O-rich fluids might be present in hollows of the nanoglobules if they originate from dust particles composed of organic materials and ice prior to or in an early stage of the solar system formation or fluids incorporated into nanoglobules during aqueous alteration on the asteroidal parent body. In order to determine whether or not any fluids or mineral grains are present in the nanoglobules, a carbonaceous chondrite sample (Tagish Lake C2 meteorite) was observed non-destructively using synchrotron radiation-based X-ray CT (computed tomography), and then microtomed sections were observed using a transmission electron microscope (TEM). We observed three-dimensional shapes of thirty-eight organic nanoglobules in the meteorite sample. Their size and shape distributions are consistent with a hypothesis that nanoglobules originate from icy dust particles. Nanoglobule candidates observed in CT images were confirmed by the TEM images. However, the presence or absence of fluid could not be judged because CT images of nanoglobules are affected by X-ray refraction. Simulation of CT images by considering X-ray refraction shows that the presence or absence of water in nanoglobules cannot be distinguished with CT images alone. However the outer shapes of nanoglobules can be determined quantitatively and nanoglobules containing silicate cores can be easily identified. The thirty-eight nanoglobules we examined did not have silicate cores.

1. INTRODUCTION

Insoluble organic materials called organic nanoglobules are ubiquitously distributed throughout primitive extraterrestrial materials such as Tagish Lake ungrouped C2 carbonaceous chondrite (e.g., Nakamura et al., 2002; Garvie and Buseck, 2004; Nakamura-Messenger et al., 2006; Nittler et al., 2009; De Gregorio et al., 2010a,b), CI carbonaceous chondrites (Garvie and Buseck, 2006; Nittler et al., 2009; De Gregorio et al., 2010a), CM carbonaceous chondrites (Garvie and Buseck, 2004; Messenger et al., 2008; Nittler et al., 2009; De Gregorio et al., 2010a), CR chondrites (Nittler et al., 2009; De Gregorio et al., 2010a; Hashiguchi et al., 2011), Isheyevo CH/CB meteorite (Ishii et al., 2010), type 3.0 ordinary chondrites (Cody et al., 2011), stratospheric interplanetary dust particles (IDPs; Messenger et al.,

* Corresponding author.
E-mail address: tmatsumoto@kueps.kyoto-u.ac.jp (T. Matsumoto).

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2008; Matrajt et al., 2011), and particles from comet 81P/Wild 2 (Matrajt et al., 2008; De Gregorio et al., 2009, 2010c). They are approximately spherical from the observation of ultra-thin sections, but their 3-D shapes have not been exactly revealed. Their sizes range from several hundred to several thousand nanometers (nm), and in many cases they have single hollow spaces in their centers. It was suggested that the nanoglobules were formed by asteroidal aqueous alteration on their parent bodies (Nakamura et al., 2002; Cody et al., 2011). However, they are enriched in $^{15}N/_{14}N$ and D/H compared with terrestrial materials and the parent body materials. These isotopic anomalies indicate that the organic nanoglobules were formed from ice gains by photochemical reaction in cold molecular clouds or low temperature regions of protoplanetary disk at 10–50 K (Nakamura-Messenger et al., 2006). In this case, the ice grains that consisted of water and simple organic materials were irradiated by ultraviolet or cosmic ray and complex refractory organic mantles were formed on the surfaces of the particles. Interstellar grains in a molecular cloud are theoretically considered to have a layer structure that is composed of a silicate core, an inner organic matter mantle and an outer ice mantle (represented by Greenberg, 1998), which are very similar in size, chemical composition and texture with nanoglobules observed in astromaterials except the silicate core. In astromaterials, organic nanoglobules with silicate cores are quite rare, there have been only three reports of such material (Nakamura-Messenger et al., 2006; Hashiguchi et al., 2011; Nakamura-Messenger et al., 2012). One way to explain the lack of silicate core was that the silicate core could have been removed during the sample preparation such as physically plucked away during ultramicrotomy (e.g., Nakamura-Messenger et al., 2006 for details) or petrological thin section polishing (Hashiguchi et al., 2011) or chemically digested to isolate insoluble organic matters from rest of the meteoritic material (e.g., De Gregorio et al., 2010a).

Alternatively the core of the interstellar icy dust was not refractory silicate, but a more volatile material, which could evaporate away leaving less volatile mantle material as hollow nanoglobules, or the nanoglobules formed from organics-ice particles, the central hollow regions of the nanoglobules should have originally been filled with water or organic ice, which might be preserved as fluids in the hollow regions. Fluids may also have been incorporated into nanoglobules during aqueous alteration. However, fluids in the nanoglobules have not been detected so far because all of above mentioned destructive sample preparation prior to the observations. If fluids were originally preserved in the central hollow regions of the nanoglobules, they would have been lost during these destructive processes during sample separation for transmission electron microscope observation.

X-ray computed tomography (CT) is a non-destructive method to obtain three-dimensional (3-D) structures of materials. Absorption-contrast CT using X-ray transmittance of objects gives CT images, which show spatial distribution of X-ray linear absorption coefficient (LAC) of objects. Synchrotron radiation (SR)-based imaging tomography using X-ray microscope optics with a Fresnel zone plate (FZP) has been developed (e.g., Uesugi et al., 2006; Takeuchi et al., 2009). The system achieves a few hundred nanometer spatial resolution around 8 keV, and was applied to elucidate micro-textures of extraterrestrial materials, such as cometary particles collected by the Stardust mission (Zolensky et al., 2006; Nakamura et al., 2008a,b; Rietmeijer et al., 2008) and particles on asteroid surface collected by the Hayabusa mission (Tsuchiyama et al., 2011). We have applied this method to organic nanoglobules in a carbonaceous chondrite. The advantages of this method for observation of nanoglobules are follows: (1) 3-D internal structure of nanoglobules can be examined non-destructively *in situ*, (2) nanoglobules can be observed by its high spatial resolution, and (3) materials in nanoglobules can be estimated from a quantitative LAC values in CT images using monochromatic X-rays (Tsuchiyama et al., 2005).

The purpose of the study is to identify nanoglobules in CT images, reveal their 3-D morphologies and 3-D distributions in a carbonaceous chondrite for the first time, and determine whether or not fluids or mineral grains are present in their interior.

2. EXPERIMENTS

Tagish Lake meteorite, an ungrouped carbonaceous chondrite of C2 type (Zolensky et al., 2002), was used in this study. A particle of ~30 × 40 × 60 μm in size from the matrix area was held on a glass fiber of 5 μm in diameter with a small amount of glue (glycol phthalate). This particle was observed using the imaging tomographic system at BL47XU in SPring-8, a synchrotron facility in Japan (Uesugi et al., 2006; Takeuchi et al., 2009). The voxel size in CT images was 40.8 nm × 40.8 nm × 40.8 nm. Effective spatial resolution of the system is about 200 nm (Uesugi et al., 2006). The sample was measured under the slightly defocused condition. Defocus distance was approximately 200 μm. Monochromatized X-ray energy of 7.0 keV was used for the imaging, and CT images were reconstructed from 1800 projections using a convolution back-projection algorithm (Nakano et al., 2000). Eight hundred successive CT images were obtained for 3-D structure of the sample. A software package named SLICE (Nakano et al., 2006) was used for 3-D image processing and analysis, including creation of CT sections from the 3-D images from arbitrary directions to match a cross section of the sample, drawing external shape images (bird's eye view images) of the meteorite particle and the nanoglobules from arbitrary directions, and calculation of best-fit ellipsoids of nanoglobules.

After the 3-D imaging, the sample particle together with the attached glass fiber was embedded in low viscosity epoxy (Embed 812) and thickness and orientation controlled ultra thin sections (70–120 nm thickness) were obtained by ultramicrotomy (for the details of this dissection technique, see Nakamura-Messenger et al., 2006). These sections were transferred to amorphous C-supported Cu TEM grids. Five TEM grids with about 30 thin-sections each on top were prepared from the sample for subsequent TEM study to identify the nanoglobules and compare with the nanoglobules candidates in the 3D images. The TEM imaging was performed using a JEOL