Supporting information

# Fabrication of Metal-Organic Framework Nanosheet and Nanoroll with N-Donor Type of Bridging Ligands

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## Materials

All the reagents employed were commercially available and used as supplied without further purification. Reagent $Cu(OTf)_2$ and 4,4′-bipyridine (bpy) of 98 % purity was purchased from Tokyo Kasei Chemical Company and acetone and ethanol was purchased from Wako Chemical Company. Milli-Q filtered water with resistivity > 18 MΩ·cm was used throughout the experiments.

## Material synthesis and exfoliation

Single crystal of CuMOF, $[Cu(bpy)_2(OTf)_2]$, was prepared according to the procedure reported before.[1S] The obtained material was characterized by XRD and IR to confirm the purity of the material. Three milligrams of the obtained single crystals were dispersed in 2 ml of solvent and used for the physical exfoliation treatment. Ultrasonication was performed on an As One US cleaner at room temperature and 270W for 5 – 60 min. Shaking treatment was performed at room temperature for 1 – 12 h. All samples were left over several hours to remove large thick CuMOF crystals before TEM, SEM, and AFM observation. Only the samples treated in water were centrifuged at 1000 rpm for 30 min for further purification.

## Characterization

Powder X-ray diffraction patterns were measured on a Rigaku RINT-2100S using monochromated Cu-Kα radiation. Infrared spectra were recorded on JASCO FT/IT-4200 with using ATR attachment. TEM images were taken on a JEOL JEM1400 at an accelerating voltage of 100 kV and a JEOL JEM2010 at 120 kV. The samples were supported on carbon copper grids. SEM images were taken on a JEOL JSM-6510 electron microscope at an accelerating voltage of 10 kV. For the SEM observation, a carbon copper grid for TEM was also used. To obtain the topographical images, Veeco AFM atomic force microscope was used under operation of the contact mode. For the AFM substrate, freshly cleaved mica and Si substrates were used. The nanosheets were placed on mica or Si substrate by dropping the colloidal solution and following washing with the solvent used.

Reference

1S. A. Kondo, H. Kajiro, H. Noguchi, L. Carlucci, D. M. Proserpio, G. Ciani, K. Kato, M. Takata, H. Seki, M. Sakamoto, Y. Hattori, F. Okino, K. Maeda, T. Ohba, K. Kaneko and H. Kanoh, *J. Am. Chem. Soc*. **2011**, *133*, 10512-10522.

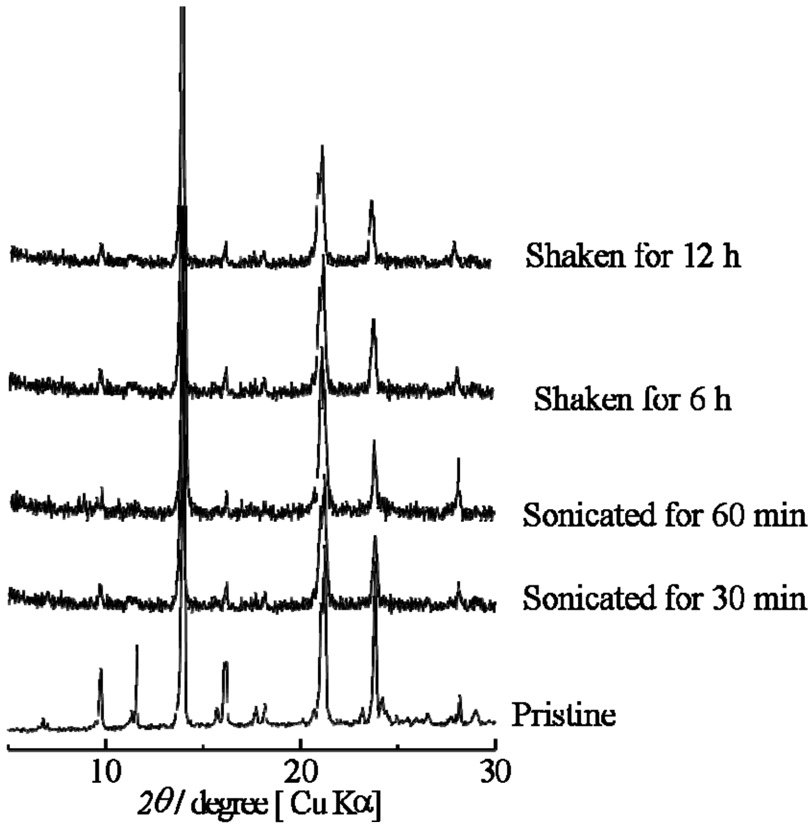

**Figure 1S**. XRD patterns of CuMOF. From bottom to top, pristine CuMOF, the sediments after ultrasonication for 30 min, for 60 min, and after shaking treatment for 6h, and for 12 h. All samples are treated in acetone.

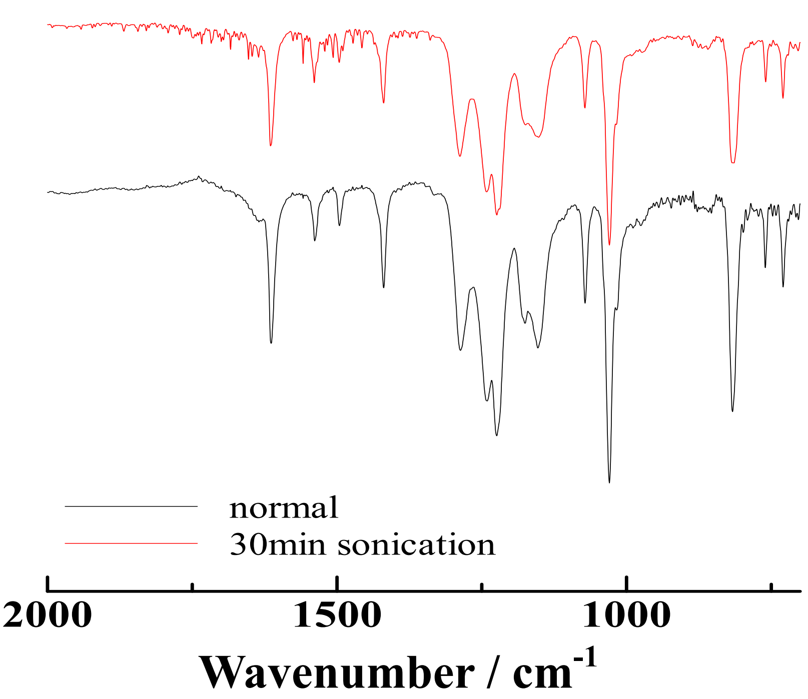

**Figure 2S**. FT-IR spectra of pristine CuMOF (black) and the sediment after ultrasonication for 30 min in acetone (red).

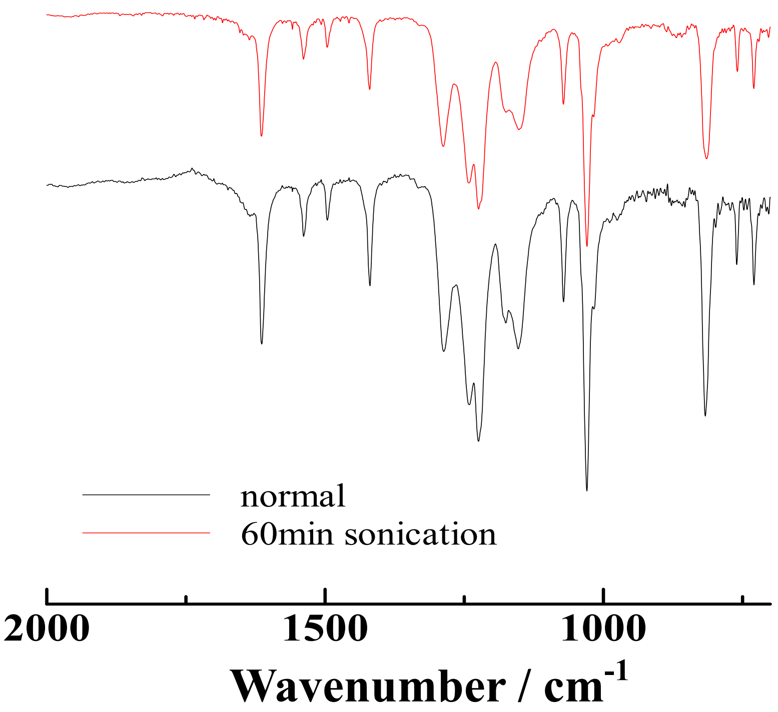

**Figure 3S**. FT-IR spectra of pristine CuMOF (black) and the sediment after ultrasonication for 60 min in acetone (red).

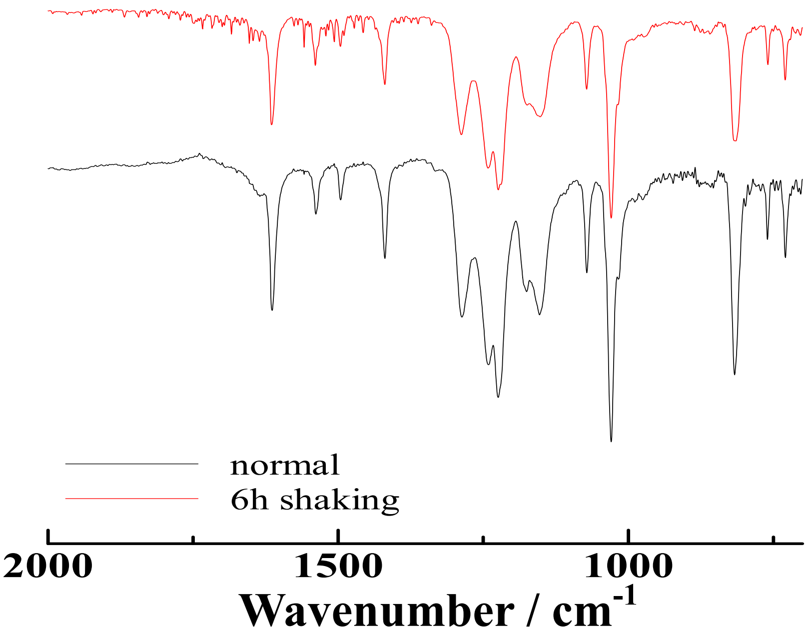

**Figure 4S**. FT-IR spectra of pristine CuMOF (black) and the sediment after shaking treatment for 6 h in acetone (red).

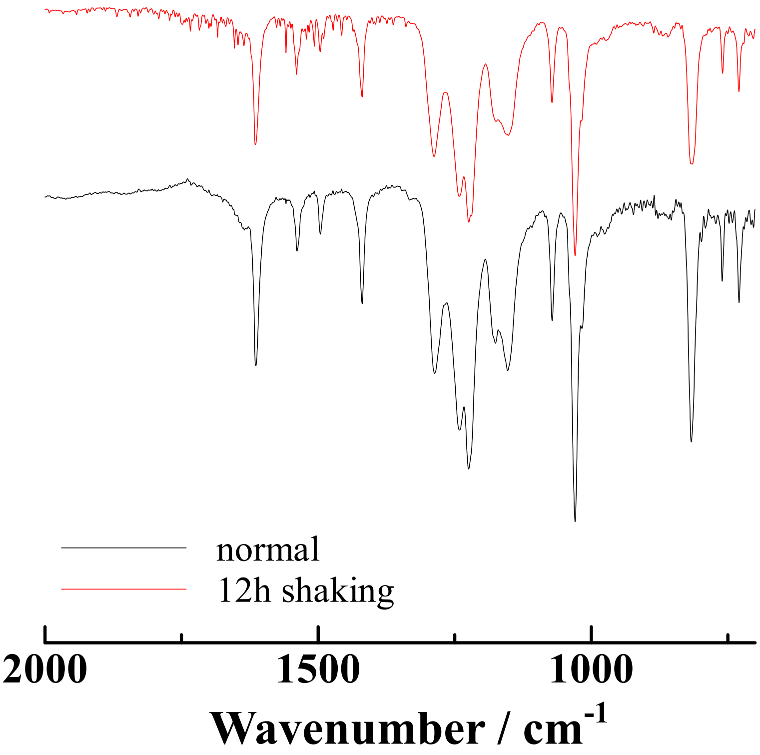

**Figure 5S**. FT-IR spectra of pristine CuMOF (black) and the sediment after shaking treatment for 12 h in acetone (red).

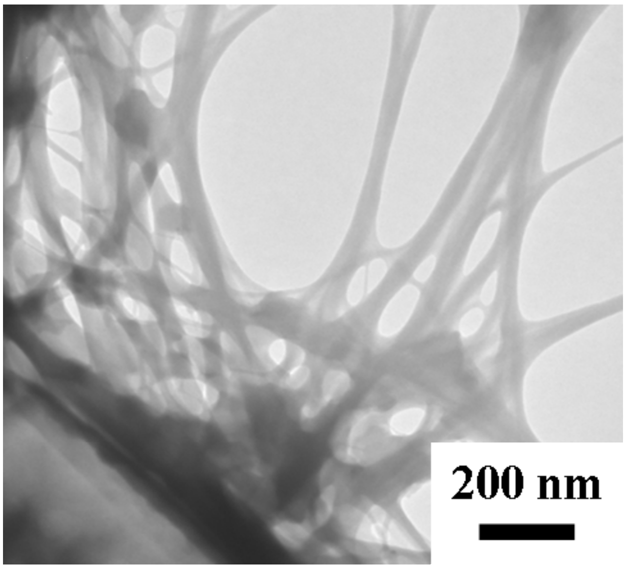

**Figure 6S**. TEM image of CuMOF sonicated for 60 min in acetone.

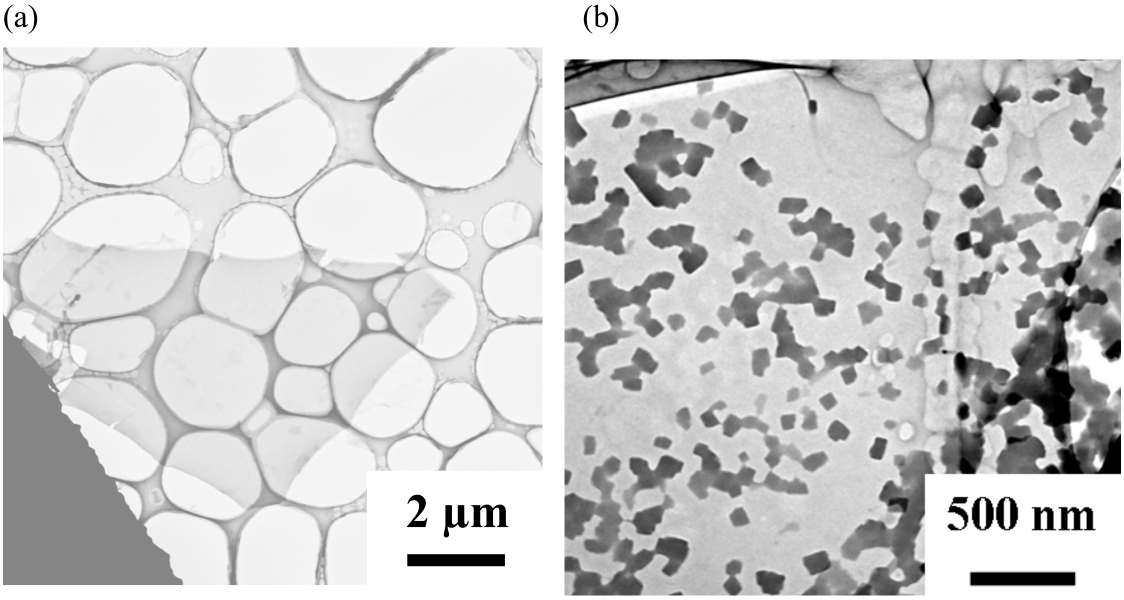

**Figure 7S**. TEM images of CuMOF shaken for (a) 6 h and for (b) 12 h in acetone.

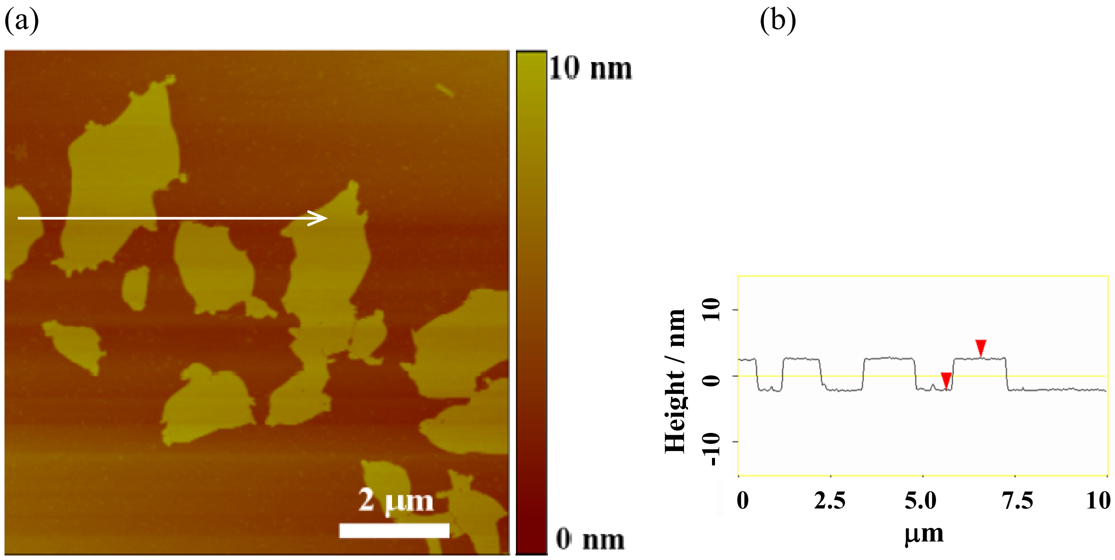

**Figure 8S**. (a) AFM image of CuMOF shaken for 6 h in acetone and (b) height profile of the area highlighted in figure 2S(a).

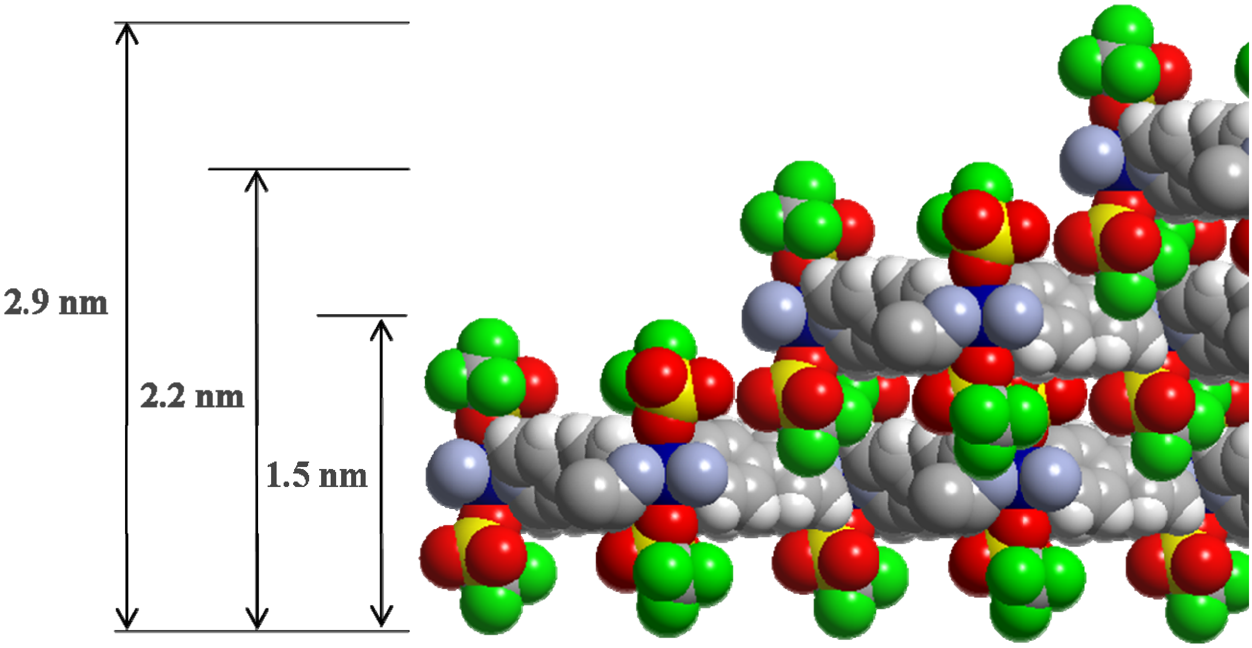

**Figure 9S**. Schematic representation of of CuMOF with the thickness of mono to triple layered parts in space filling model.