

Title:

**Moisture-Responsive Supramolecular Nanotubes**

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Main contents:

1. <sup>1</sup>H NMR spectra of the lipids; 2. Calibration curve of eugenol; 3. Time-dependent formation of nanotube; 4. STEM images of the self-assemblies of cobalt(II) ion with other lipids; 5. Morphological changes and release of nanotubes in bulk water; 6. Moisture uptake of nanotube self-assembled from 16-hydroxy-*N*-palmitoyl-glycylglycine; 7. XRD and FT-IR of the sodium salt of 15-hydroxy-*N*-pentadecanoyl-glycylglycine; 8. FT-IR spectra of sheets after dehydration treatment; 9. STEM image of Co-sheet after dehydration at 5 mg/mL.

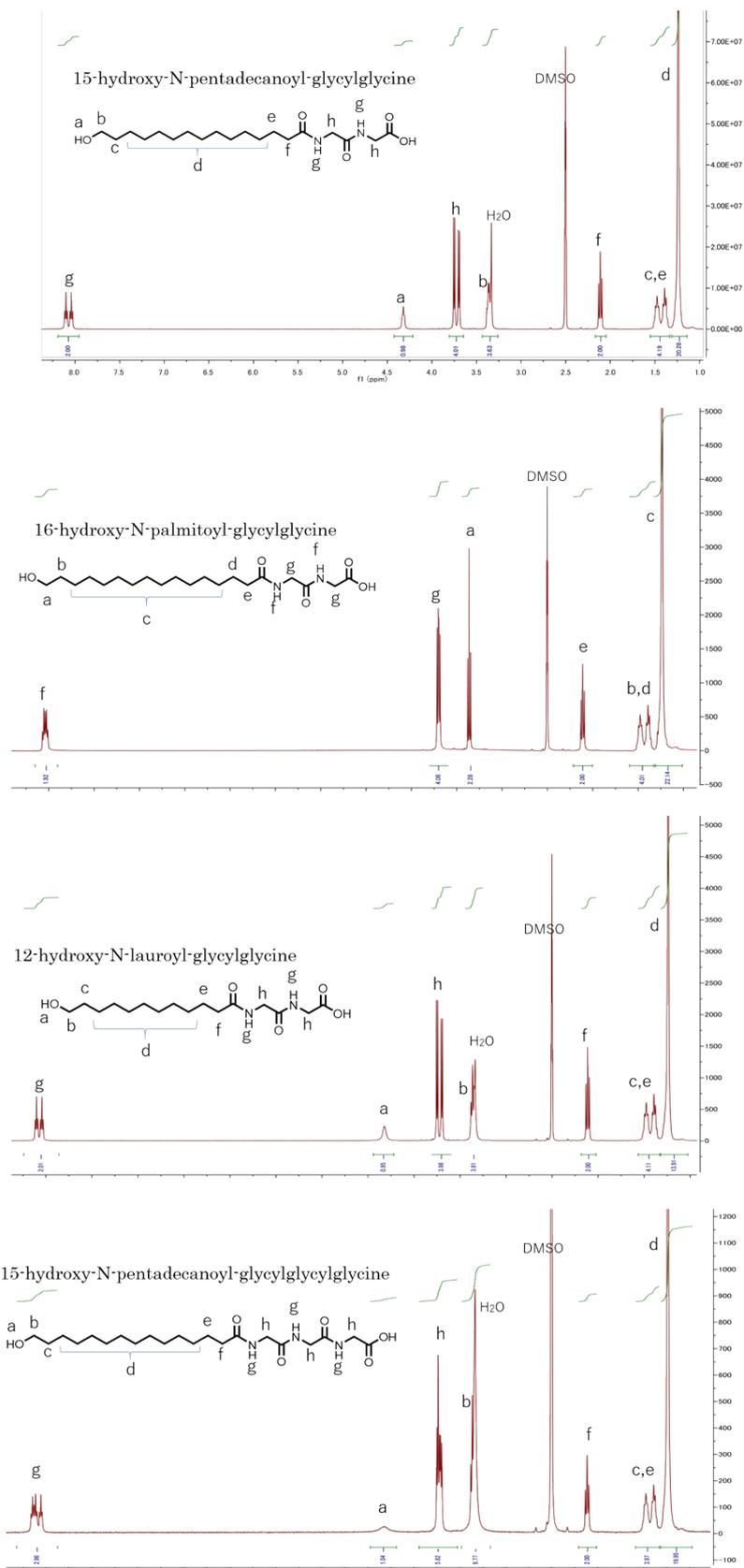


Fig. S1 <sup>1</sup>H NMR spectra of the lipids.

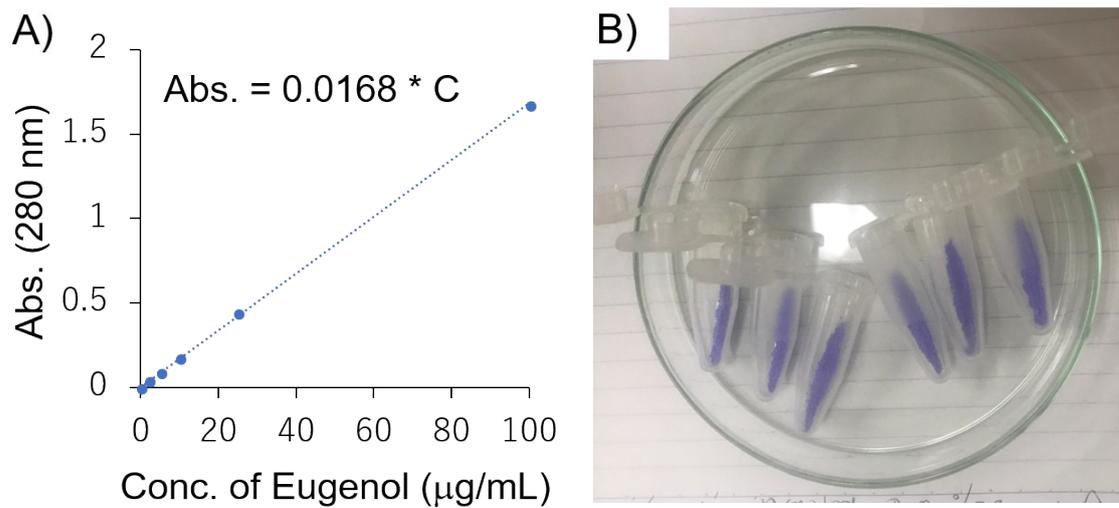


Fig. S2 (A) The calibration curve of eugenol and (B) Dry powder of Co-NT in the EP tube for the release experiment of eugenol.

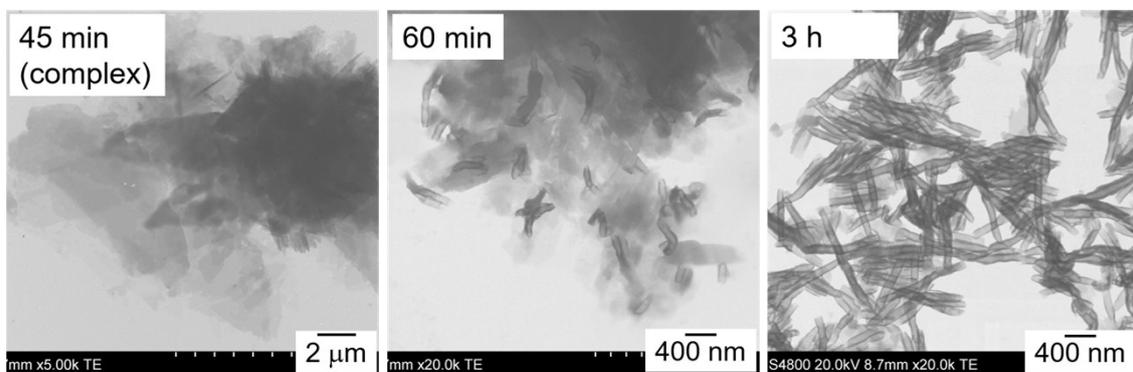


Fig. S3 Time-dependent transformation from the bis(lipid)-Zn(II) complex into Zn-NT.

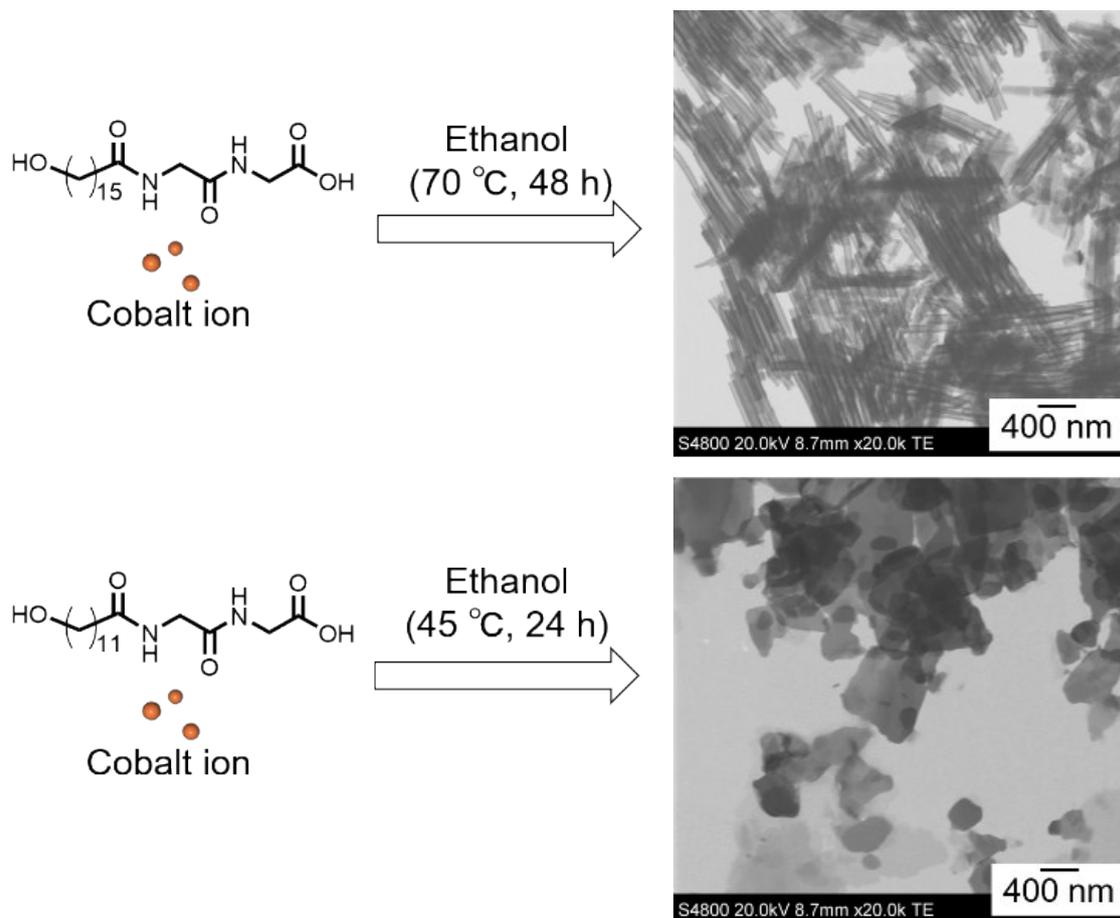


Fig. S4 STEM images of the self-assemblies of cobalt(II) ion with 16-hydroxy-N-palmitoyl-glycylglycine or 12-hydroxy-N-lauroyl-glycylglycine.

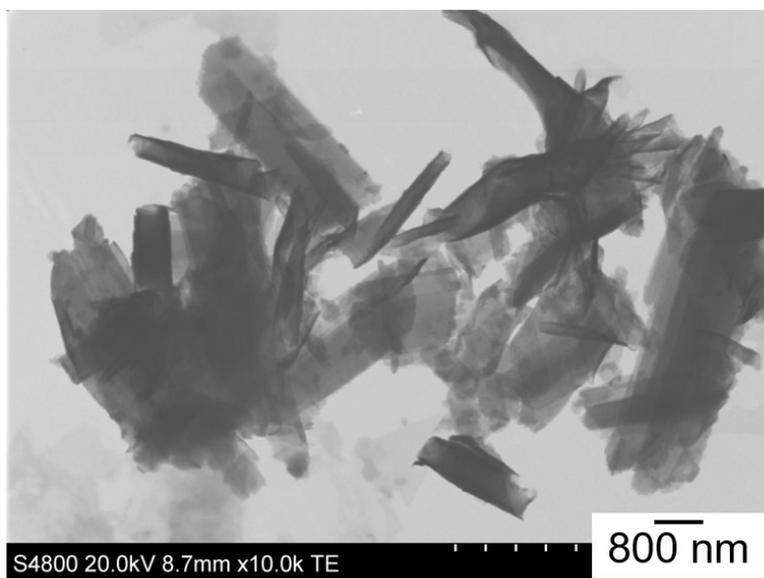
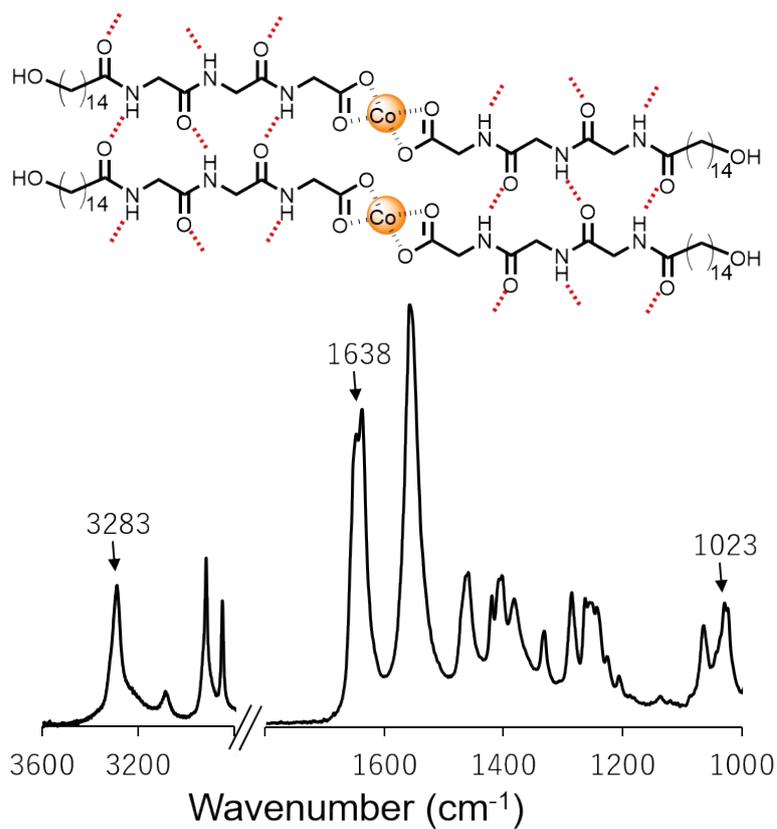


Fig. S5 H-bonding, FT-IR and STEM images of the self-assembly of cobalt(II) ion with 15-hydroxy-N-pentadecanoyl-glycylglycylglycine. In the FT-IR, Lack of 1666 cm<sup>-1</sup> peak suggests the absence of loose-type H-bond network.

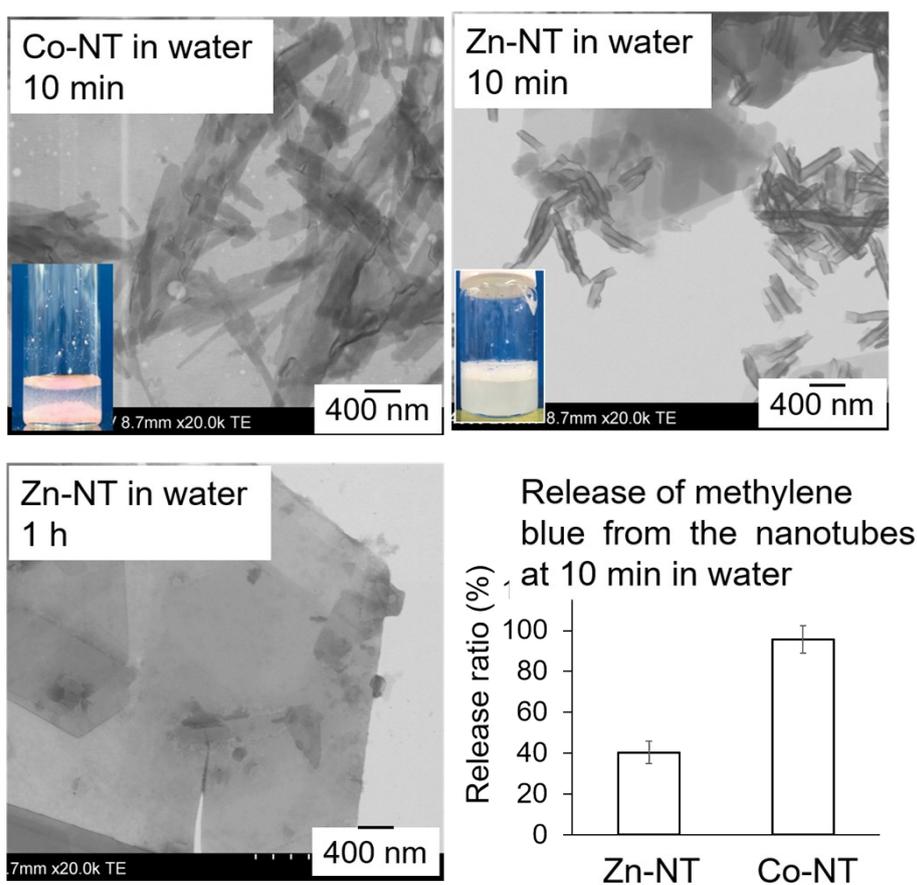


Fig. S6 STEM images of nanotubes dispersed in water and the release of methylene blue.

#### Encapsulation and release of methylene blue.

Methylene blue was encapsulated into the nanochannels of nanotubes by the mixing of methylene blue with nanotube dispersions in ethanol. Those dispersions were filtered, washed with ethanol to remove non-encapsulated methylene blue, and dried in vacuum.

100 mg of each powder were dispersed in 10 mL of distilled water and stirred under 400 rpm. At 1 min and 10 min timepoints, 2 mL of dispersions were filtered through syringe-type membrane filters (100 nm in pore size) and the filtrates were collected for measuring the absorbance at 665 nm. The concentration of methylene blue was calculated from a calibration curve. The data at 1 min were considered as baseline since non-encapsulated methylene blue will quickly release into water upon dispersion.

Total amount of methylene blue in nanotube was measured as follows: 100 mg of powders were dispersed in 10 mL of ethanol-water mixture, heating of the dispersions at 80 °C could result in the complete release of methylene blue.

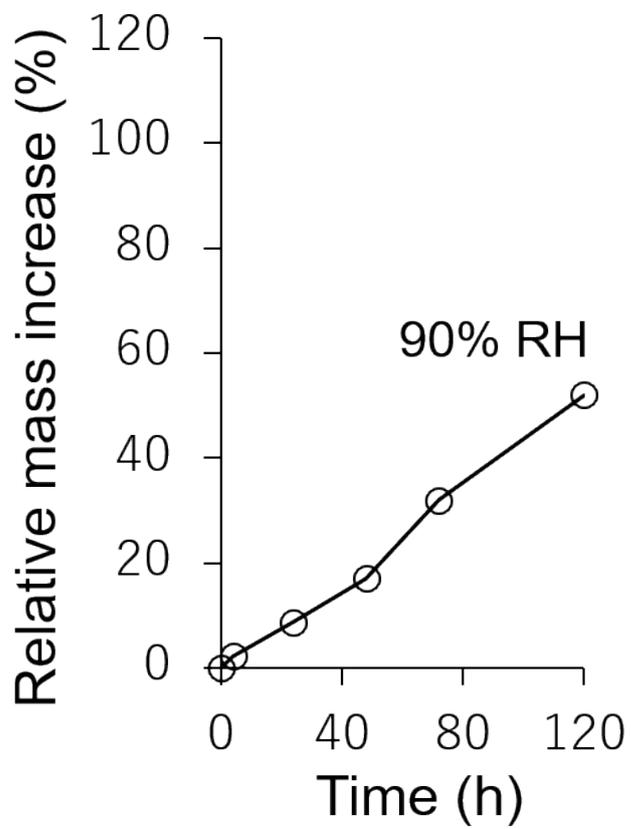


Fig. S7 Moisture uptake by the nanotube self-assembled from cobalt(II) ion with 16-hydroxy-N-palmitoyl-glycylglycine.

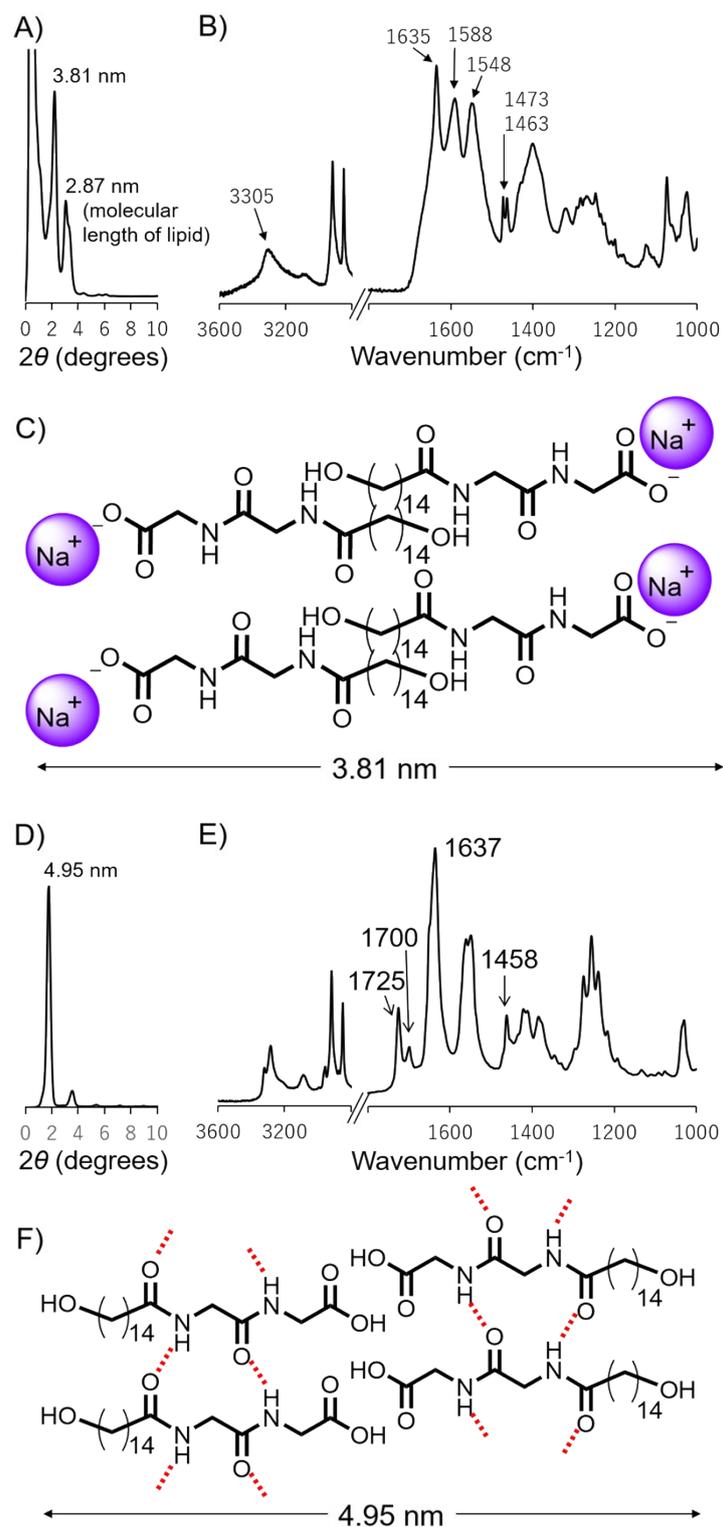
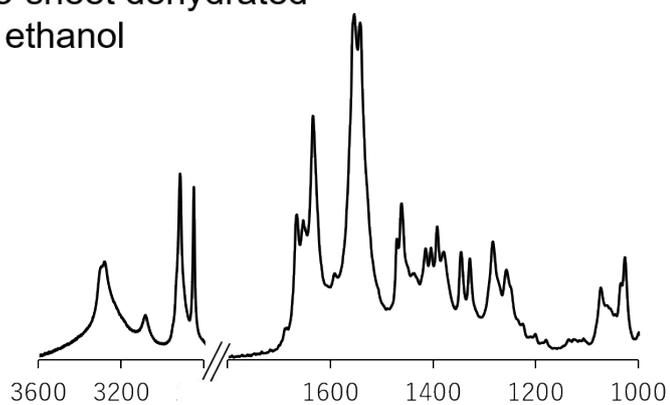


Fig. S8 The (A) XRD spectrum, (B) FT-IR spectrum, and (C) speculated molecular packing of the sodium salt of 15-hydroxy-N-pentadecanoyl-glycylglycine. Those of the acidic form of the lipid are shown in D-F.

Co-sheet dehydrated  
in ethanol



Zn-sheet dehydrated  
in ethanol

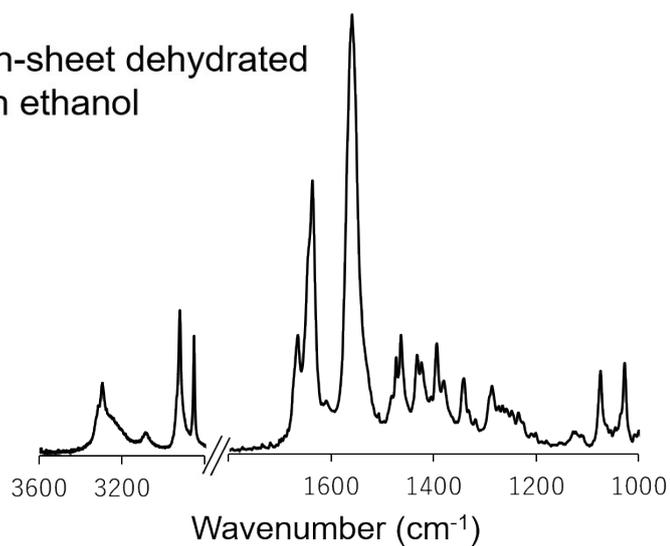


Fig. S9 FT-IR spectra of Co-sheet or Zn-sheet dehydrated in hot ethanol.

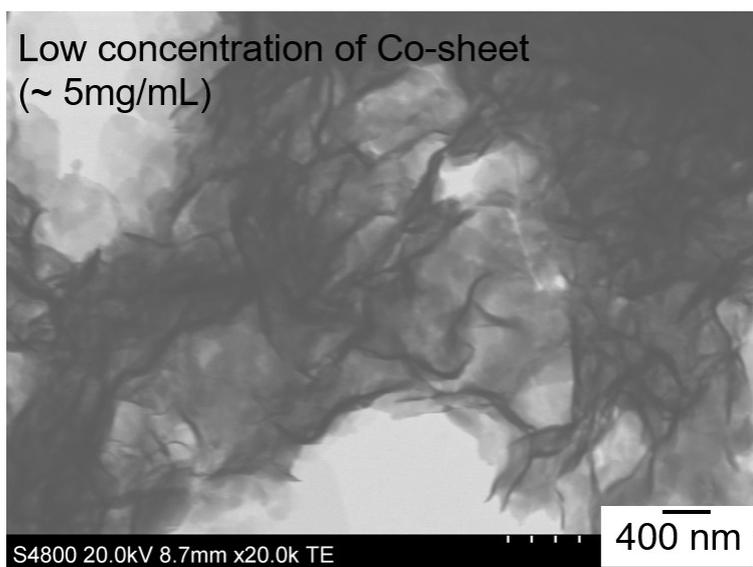


Fig. S10 Dehydration and incubation of the Co-sheet in ethanol at lipid concentration of ~ 5 mg/mL.