Supplementary Information

Environmentally friendly supramolecular nanosheet particles for surface coating

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[S1] *Capped-NSs reacted with different amounts of TMPTG*

The capped-NSs were prepared by reacting uncapped-NSs (including the $-NH_2$-terminated axis) with TMPTG. A series of capped-NS samples were prepared by changing the amount of TMPTG added. One, 10, 20, 50, and 100 mol equivalents against the axis of TMPTG were added to the uncapped-NSs (TG1NS, TG10NS, TG20NS, TG50NS, and TG100NS, respectively). They were then washed with acetone, redispersed in water, and placed on Si wafers for scanning electron microscopy (SEM) observation (Figure S1).

The samples were redispersed in water and diluted. The uncapped-NSs were dissolved in water and no particles were observed. While, TG1NS, TG10NS, TG20NS, TG50NS, and TG100NS were perfectly stable under these conditions (the several particles were lacked due to the shear forces of the attachment of the particles to the surface). We reported in our previous study that two EGDGE molecules are required to react with each –NH2 end for capping.[36] While, the addition of 1 equivalent of TMPTG against axis ends achieved this in this study, indicating that one TMPTG has sufficient bulkiness to block the dethreading of CyD. This indicates that TMPTG is more effective in stabilizing uncapped-NSs than EGDGE. The TG20NS is stable against to the dilution and was used as the capped-NSs in all experiments.

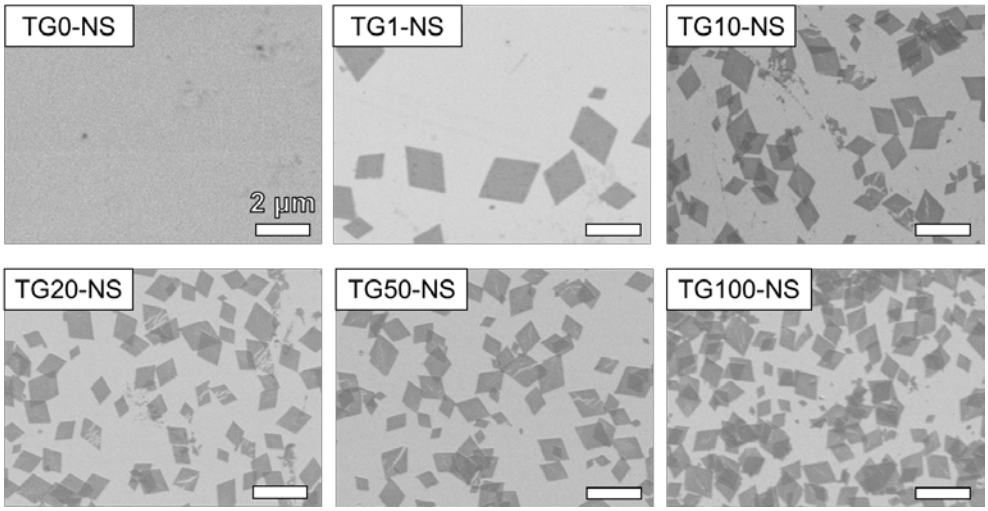

Figure S1. SEM images of the uncapped-NSs, TG1NS, TG10NS, TG20NS, TG50NS, and TG100NS.

The modification ratio of TMPTG with the axis ends in TGnNS could be calculated based on the ratio of the peak integrals of CH_3 of PPO (at 1.0 ppm) and CH_3 of TMPTG (at 0.8 ppm). Those were 74%, 158%, 184% for TG1NS, TG20NS, and TG50NS, respectively. The modification ratio less than 100 % possibly indicates that the one TMPTG reacted with one axis end. The modification ratio over 100 % indicates that two TMPTG were reacted with one axis end.

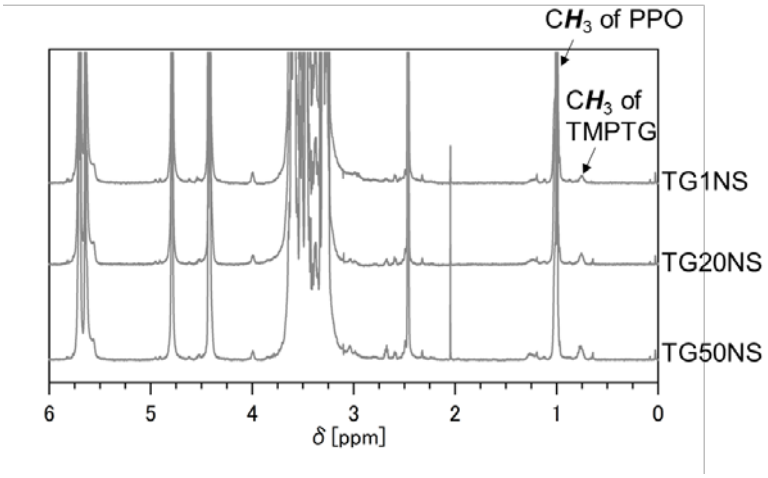

Figure S2. 1H NMR spectrum of TG1NS, TG20NS, and TG50NS in DMSO-d_6.

[S2] *Calculation of coverage of capped-NS-coated samples*

The coverage of capped-NS-coated samples were obtained by the image analysis of three SEM images for each samples. The representative of the image analysis is shown in Figure S3.

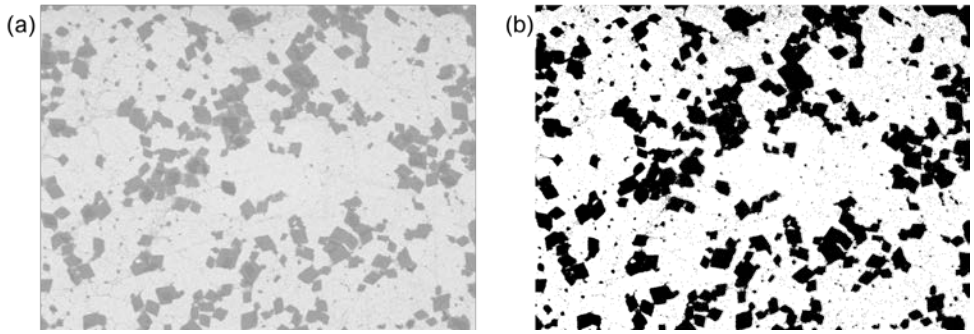

Figure S3. (a) Representative SEM image of capped-NS-coated sample and (b) its binarized images. The ratio of dark region to whole area leads the coverage of the sample.

[S3] *Oil-repellent properties of the capped-NSs on the surface of various types of materials*

Capped-NS coatings can be used on various material surfaces and render polymer (PEO) brush layers on the material surfaces. Chili oil was dropped onto the capped-NS-coated surfaces, which were then immersed in water. The chili oil droplets were strongly attached to the non-coated surfaces. The capped-NS-coated surfaces exhibited oil-repellent properties in water. The fabrication of polymer brush layers on material surfaces is difficult because synthetic methods are typically required (such as reactions and/or organic solvents). Coating with capped-NSs is a new method for constructing polymer brush layers using easy fabrication protocols (drop casting or spraying).

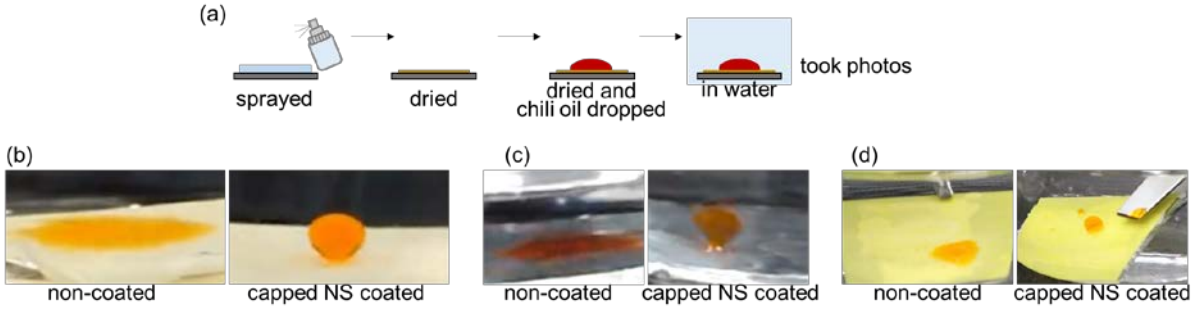

Figure S4. (a) Protocol for the construction of the capped-NSs coating and the experiment used to assess the oil-repellent properties of the coating under water. Photographs of chili oil droplets on (b) a rubber glove, (c) aluminum foil, and (d) plastic films in water. The photographs on the left-hand side of figure part b, c and d are of the non-coated samples and those on the right-hand side are of the capped-NS-coated samples.

[S4] *Abundance ratio of bacteria in the extracted sea water*

The abundance ratio of bacteria in the extracted sea water was analyzed by 16S amplicon sequencing analysis, which was outsourced to Chemical Dojin Co., Ltd.

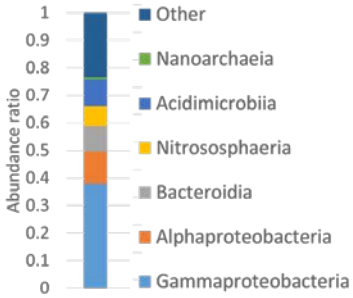

Figure S5. Abundance ratio of bacteria in the extracted sea water.