

Supplementary Material (ESI) for Chemical Communications
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Electronic Supporting Information

ESI-1. Experimental details

The chemically converted graphene (CCG) dispersions were synthesized using a similar procedure we have reported previously.¹ Graphene oxide (GO) was synthesized from graphite (SP-1, Bay Carbon) using a modified Hummers method.² 25 ml of GO (0.05 wt%) was mixed with 25 ml of water, 50 μ l of hydrazine solution (35 wt% in water, Aldrich) and 175 μ l of ammonia solution (28% in water, Crown Scientific) in a 50-ml glass vial. The mixed dispersion was then vigorously shaken and divided into five parts. Two parts of the dispersion were placed in 90 °C and 100 °C water bath, respectively; and the other three parts placed in 60 ml Teflon-lined stainless steel autoclaves and heated to 120°C, 150°C and 180°C, respectively. All the heat treatments lasted for 3 h.

The CCG membranes were prepared by vacuum filtration of 0.5 ml of CCG dispersions through polycarbonate filter membranes (47 mm in diameter, 0.2 μ m pore size, Millipore). Once the filtration was completed (no free CCG dispersion was left on the filter but the membrane still remained wet), a certain amount of water or other solvents was immediately poured on top of the as-formed CCG film, which was then subjected to continuous vacuum suction (with a pressure of around 1 bar) to allow the water to flow through the membrane. The time for the water to drain was recorded to allow calculation of the average water flux for the CCG membranes.

Au nanoparticles and Pt nanoparticles were synthesized using the classical methods reported in the literature.^{3, 4} Particle sizes of Au nanoparticles and Pt nanoparticles were examined by the transmission electron microscopy. 60 μ M Pt nanoparticle colloids and 100 μ M Au nanoparticle colloids were vacuum filtrated through the CCG membranes. The filtrated liquid was examined by UV-Vis (Cray 300, Varian) to confirm the rejection of nano-particles.

A 50 ml of direct yellow (DY) (10 μ M) solution was vacuum filtered through the graphene membranes, and the top dye solution was tested by UV-Vis spectroscopy after a certain filtration time. The filtrate was examined after performing the filtration test to confirm the rejection rate.

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The Atomic Force Microscopy (AFM) imaging in water was carried out by an AFM (Multimode IV, Veeco) equipped with a fluid cell. The cantilever (NP with the nominal spring constant of 0.32 N/m, Veeco) was treated by UV light before use.

The AFM samples for the dried samples were prepared by drop-casting of the corresponding dispersion on a silicon wafer and dried in air. The AFM images for the dried samples were obtained by a Molecular Imaging Picoscan II instrument in tapping mode; Analysis of the AFM images was performed using the WSxM software (version 3, Nanotec Electronica S.L., Spain). Imaging was conducted in tapping mode, with 512 * 512 data acquisitions at a scan speed of 1.1 Hz at room temperature in air. Oxide-sharpened, silicon nitride tips with integrated cantilevers with a nominal spring constant of 0.38 N/m were used.

All SEM images reported here were taken in an Ultraplus-SEM (Zeiss) using an In-Lens detector, and the accelerating voltage was adjusted to obtain the best-quality image.

ESI-2. Additional discussion on the difficulty in using AFM and SEM to characterize the corrugation of CCG

As discussed in the main text, in order to allow the use of the AFM technique to characterize the configuration of CCG sheets, the sample needs to be fixed on a flat substrate. However, the fixation process could dramatically alter the morphology of CCG sheets. For example, we have found that the capillary force during the drying process could flatten moderately corrugated sheets that were prepared at lower temperatures (90-150°C). In another words, CCG sheets prepared at these temperatures exhibited a similar flatness under AFM if drop cast and dried on a silicon wafer even though the difference in the water permeation rate of the membranes resulted from these sheets was prominent. Nevertheless, the corrugated morphology of the CCG sheets treated at 180°C was able to be observed under AFM (see below) because the sheets were so crumpled at this temperature that the capillary force was unable to fully flatten them. The SEM images of the dried CCG films also revealed that the 180°C-CCG film had a rougher surface than the ones obtained at lower temperatures (Fig. S2).

Performing the AFM in the solution state could prevent the effect of capillary force and we have demonstrated that CCG sheets that were absorbed on the positively charged polymer (PDMA)

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are corrugated. However, this method still poses some problems. For example, the electrostatic attraction between CCG and PDMA could alter the configuration of CCG sheets. We have also noticed that if the CCG sheets became more corrugated, less sheets were observed on the PDMA-treated substrate. The CCG sheets treated at 180°C could hardly be seen by AFM. This result seems to be understandable as more corrugated sheets have less contact areas/sites with the substrate.

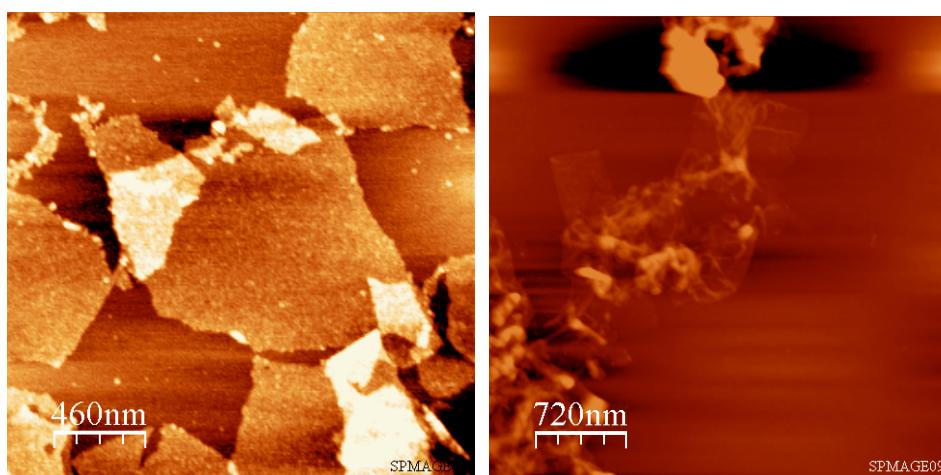


Fig. S1. AFM images of dried CCG sheets that were prepared at 90°C (left) 180°C (right), respectively.

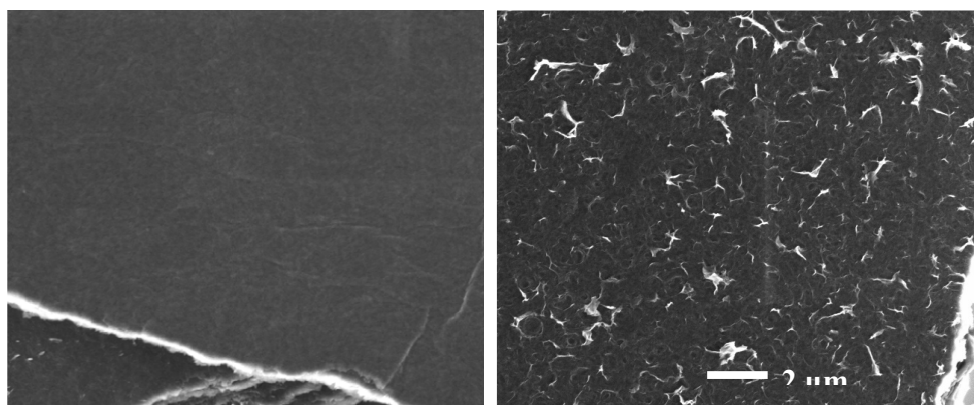


Fig. S2. SEM images of the CCG membranes. The CCG dispersions were prepared at 90°C (left) 180°C (right), respectively.

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