# **Supporting Information (SI)**

## Electrochemical delamination assisted transfer of molecular nanosheets

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#### Calculation of the effective thickness

The effective thickness d of the CNMs was calculated from the attenuation of the substrate signal using the Beer-Lambert law<sup>1</sup>

$$d_{total} = -\lambda \cos \theta \ln \frac{I}{I_0} , \qquad (1)$$

where *I* is the intensity of the substrate signal (Au 4f or Si 2p);  $I_0$  is the intensity of a bare substrate reference sample;  $d_{total}$  is the total thickness of the layer (CNM) on the substrate; the  $\lambda_{Au}$  (or  $\lambda_{Si}$ ) is the attenuation length of Au 4f (or Si 2p) electrons in the carbon layer (3.6 nm for Au or 3.0 nm for Si in aromatic carbon)<sup>2</sup> and the  $\theta$  is the angle between the normal of the sample and the analyzer (18.7°). The intensity  $I_0$  was obtained by measuring *via* Ar sputtering cleaned Au (300 nm) or SiO<sub>2</sub> (300 nm) substrate, respectively.

#### Transfer of CNMs onto gold substrates

To test functionality of the thiol groups on the bottom side of the CNMs, a BPT CNM grown on Au/mica substrate was transferred to the Au/SiO<sub>2</sub> substrate by the ECD assisted method. In order to reduce oxidation of the thiols on the bottom side of the CNM, the sample was washed only twice with degassed water. In the S 2p spectrum (Figure S3), the peak at 161.9 eV (dark yellow coloured) is assigned to the thiolates (R-S-Au) group, indicating the rebinding of the CNM to the gold substrate. The S 2p<sub>3/2</sub> peak at 163.2 eV is assigned to disulfides (R-S-S-R); the peaks at 161.0 eV and 164.5 eV are due to gold sulfides (S-Au) and sulfoxide species, respectivly.<sup>3</sup>



**Figure S1**. Electrochemical stability test of NBPT SAMs. With an increase of the negative potential on gold substrate, the oxidized nitrogen in nitro group on top of NBPT were gradually reduced to low valence state. When the potential reached a value between -2.0 V and -2.5 V, the molecules were removed from the substrate and a small amount of sulfur and sulfides was left on the surface. The parameters used for the stability test: Anode: Pt,  $0.19 \text{ cm}^2$ ; Cathode: NBPT SAM/Au/mica,  $1 \text{ cm}^2$ ; Distance between electrodes ~1 cm; Time ~2 min. Concentration of NaOH: 0.1 mol/L.



**Figure S2**. XP spectra of the Au/mica substrate after the NBPT-CNM was removed *via* ECD. Small amount of sulfur is detected on the surface.



**Figure S3**. XP spectra of a BPT CNM transferred on an Au/Si substrate *via* the ECD assisted transfer (see SI p. 2 and the main manuscript for more details)





**Figure S4**. Optical microscopy images of the CNMs successively grown on the same Au substrate and transferred onto SiO<sub>2</sub>/Si substrates using ECD. (a-c) Show the first (a), second (b) and third (c) cycle of the growth and ECD assisted transfer. The defect density of all CNMs is comparable.



**Figure S5**. AFM image of a TPT CNM synthesized on Au/mica and transferred onto SiO<sub>2</sub>/Si substrate via ECD assisted transfer. The sample was measured under N<sub>2</sub> atmosphere. The height profile at a rupture of the CNM shows the apparent thickness of ~1.7 nm. Note that the interaction of the AFM tip with the CNM surface and the SiO<sub>2</sub> substrate influences the absolute value of the measured thickness.

**Table S1:** Quantitative analysis of the high-resolution XP spectra of the transferred BPT, NBPT and TPT CNM samples on SiO<sub>2</sub> (300 nm) substrates presented in Figure 4 including the peak assignment, their binding energies, full width at half maximum (FWHM) values and areas obtained from the spectra deconvolution. The peak fitting of the sulfur doublet was performed using the fixed intensity ratios due to the spin-orbit coupling of the p-photoelectrons. For the elemental ratios mentioned in the main paper the relative sensitivity factors (RSF) of 1 for C 1s, 1.8 for N 1s, 2.9 and 1.11 for S 2p<sub>3/2</sub> were used.

Peak assignment	Binding energy, eV	FWHM, eV	Area, %		
S 2p <sub>3/2</sub>					
BPT CNM					
-SH	163.3	1.2	81		
oxidized sulfur	167.6	1.4	19		
NBPT CNM					
-SH	163.2	1.1	74		
oxidized sulfur	167.6	1.3	26		
TPT CNM					
-SH	163.3	1.1	70		
oxidized sulfur	167.5	1.6	30		
C 1s					
BPT CNM					
C-C aromatic	284.6	1.3	67		
C-S/C-N/C-C aliphatic	285.4	1.6	16		
C=O	286.7	1.7	10		
O-C=O	288.8	1.2	7		
NBPT CNM					
C-C aromatic	284.6	1.3	63		
C-S/C-N/C-C aliphatic	285.4	1.7	17.5		
C=0	286.7	1.9	12.5		
0-C=0	288.7	1.2	7		
TPT CNM					

C-C aromatic	284.6	1.2	72			
C-S/C-N/C-C aliphatic	285.6	1.6	11			
C=O	286.7	1.4	10			
O-C=O	288.8	1.7	7			
N 1s						
NBPT CNM						
-NH <sub>2</sub>	399.5	2.0	91			
-NH3+	402.0	2.4	9			

**Table S2:** Quantitative analysis of the high-resolution XP spectra of the transferred BPT CNM samples on Au/SiO<sub>2</sub> (30 nm) substrates presented in Figure S4 including the peak assignment, their binding energies, full width at half maximum (FWHM) values and areas obtained from the spectra deconvolution.

Peak assignment	Binding energy, eV	FWHM, eV	Area, %		
S 2p <sub>3/2</sub>					
S-Au	161.0	0.8	30		
-S-Au	161.9	0.9	30		
-SH or –S-S-	163.2	1.0	25		
sulfoxide	164.5	0.9	15		
C 1s					
C-C aromatic	284.6	1.5	62		
C-S/C-N/C-C aliphatic	285.8	1.2	10		
C=0	287.0	1.8	21		
O-C=O	288.9	1.2	7		

### References

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