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Electronic Supplementary Information

Stretchable Electrically Conductive and High Gas Barrier Nanocomposites

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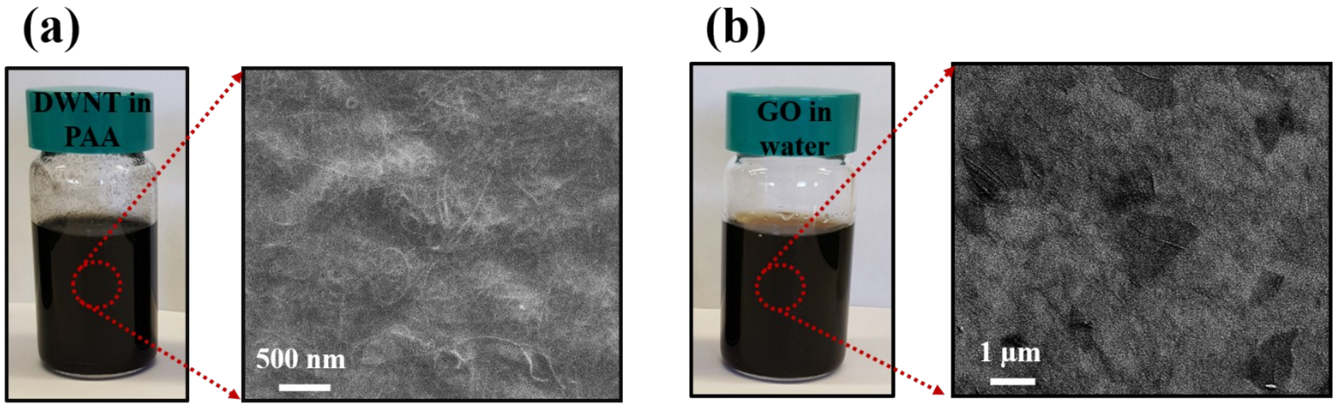

Fig. S1 Photographs of vials containing (a) DWNT stabilized by PAA in water and (b) GO in water. SEM images of the corresponding suspensions cast onto silicon wafers are shown next to each of these vials.

The composition (or concentration) of polymers and carbon nanofillers with added cycles in each system is based on quartz crystal microbalance (QCM) measurements. A linearly growing PEO/GO film has a constant composition of PEO and GO (40 and 60 wt%, respectively) during assembly. In contrast, the concentration of carbon nanofillers in the exponentially growing systems (PEO/DWNT-PAA and PEO/DWNT-PAA/GO) decreases with the number of layers deposited. This is because polymers are the main contributor to the exponential growth rather than the nanoparticles.

Table S1. Composition of multilayer nanocomposite films.

Cycles	Concentration (wt%)						
	PEO/GO		PEO/DWNT-PAA		PEO/DWNT-PAA/GO		
	PEO	GO	PEO	DWNT-PAA	PEO	DWNT-PAA	GO
10	40.7	59.3	32.2	67.8	30.1	53.4	16.5
20			36.5	63.5	32.6	54.2	13.2
30			38.9	61.1	33.6	54.5	11.9
40			40.5	59.5	33.1	56.1	10.8

Because DWNT is dispersed in PAA, thermogravimetric analysis was performed on a PEO/DWNT-PAA/GO free-standing film to obtain the composition of each individual component. The characterstic peak for each component is obtained by substracting the nitrogen data (unreactive) from the air data(reactive).[1] Figure S1 shows these graphs and corresponding characteristic peaks due to differences in the degradation rate between the two gas environments. As shown in Table S2, DWNT has a characteristic peak at 620 °C, and this value does not overlap with the characteristic peaks of other components. Commercial CNTs are generally composed of crystalline and amorphous carbons and some metallic catalyst. The metallic catalyst has the same high temperature behavior under both air and nitrogen gas. Amorphous carbon has a lower degradation temperature than the crystalline carbon (i.e., the carbon nanotube itself), suggesting that the CNT concentration can be estimated from the height of its own characteristic peak, which is only the result of nanotubes and catalyst.[2] As a result, the concentration (wt%) of DWNT in the trilayer assembly is calculated by taking the characteristic peak wt% of the assembly, multiplying by 100, and dividing by the characteristic peak wt% of the neat component. For example, the PEO/DWNT-PAA/GO films contain 18.7 wt% nanotube (plus some catalyst impurity), which was obtained by dividing 1690 by 90.5.

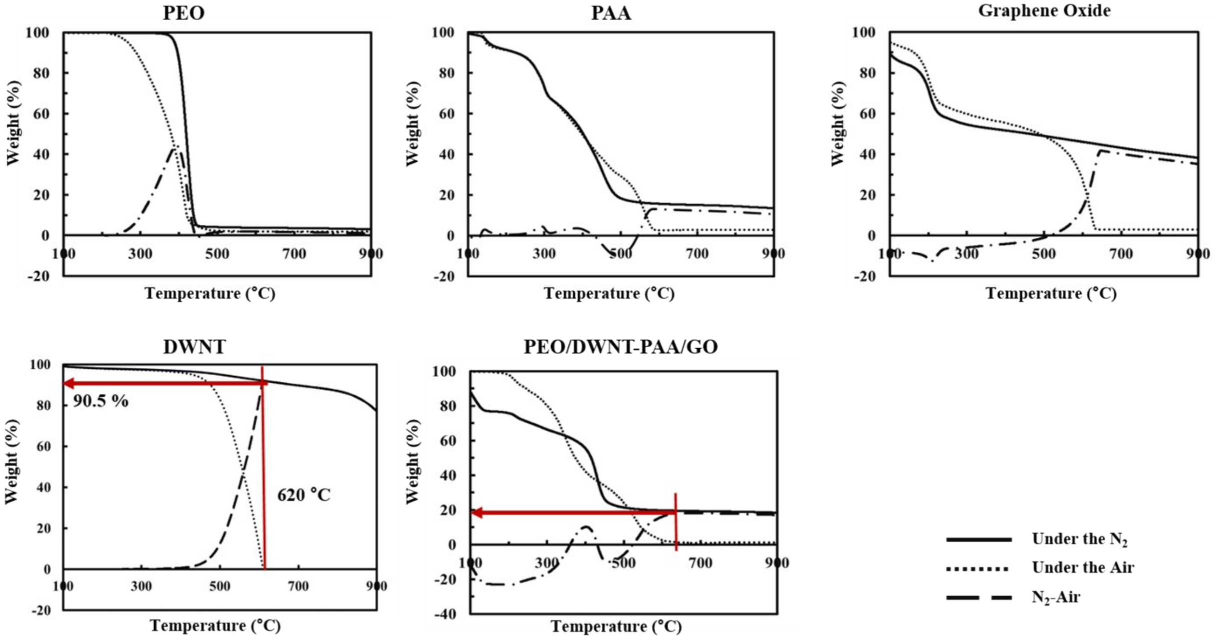

Fig. S2 Thermograms of PEO, PAA, GO, DWNT, and a 100 TL PEO/DWNT-PAA/GO film under both nitrogen gas and dry air, respectively. The difference between the air and nitrogen curves is also plotted to show the characteristic peak for a given material.

Table S2. Characteristic temperature of DWNT and composition in the trilayer assembly.

LbL System	Characteristic temp. (°C)	Conc. (wt%)	Conc. in assembly (wt%)	DWNT (wt%)
PEO/PAA-DWNT/GO	**620**	**90.5**	**16.9**	**18.7**

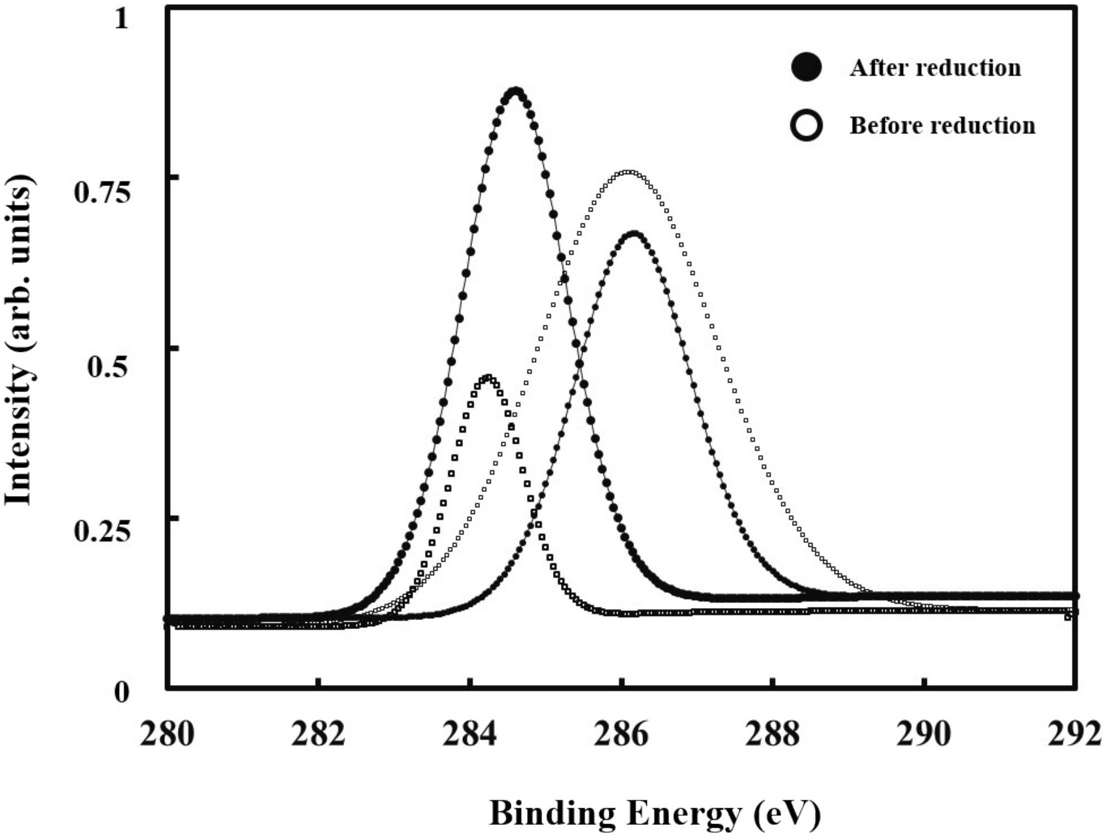

Fig. S3 C 1s XPS spectrum of graphene oxide before and after a 90 min reduction at 175 °C.

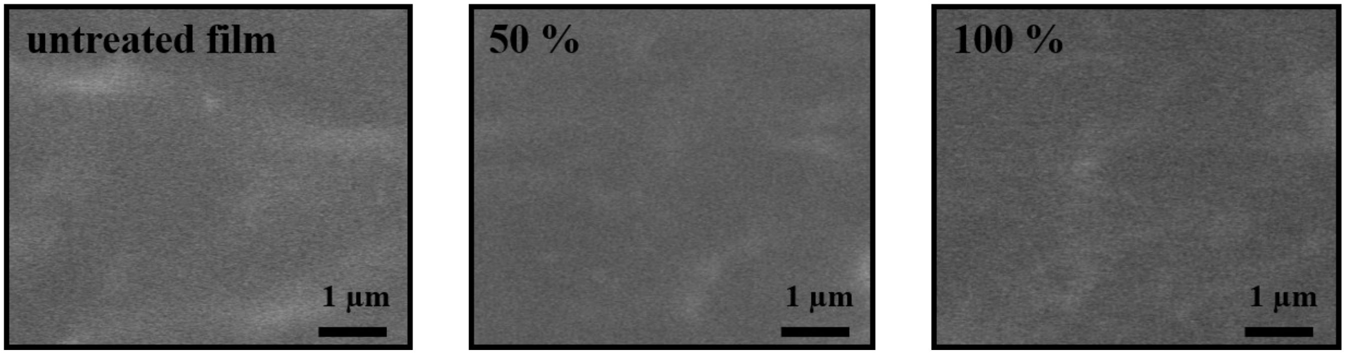

Fig. S4 SEM images of uncoated 1 mm thick polyurethane rubber before and after 50 and 100% stretching.

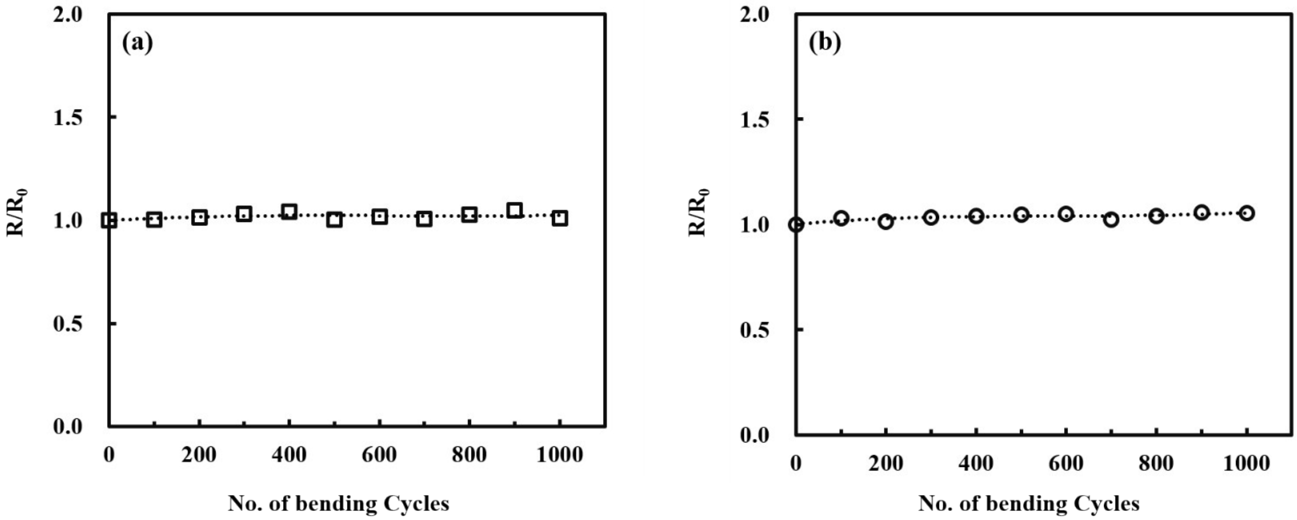

Fig. S5 Normalized resistance of 30 bilayer (940 nm thick) PEO/DWNT-PAA (open square) and 40 trilayer (900 nm thick) PEO/DWNT-PAA/GO (open circle) in cyclic bending.

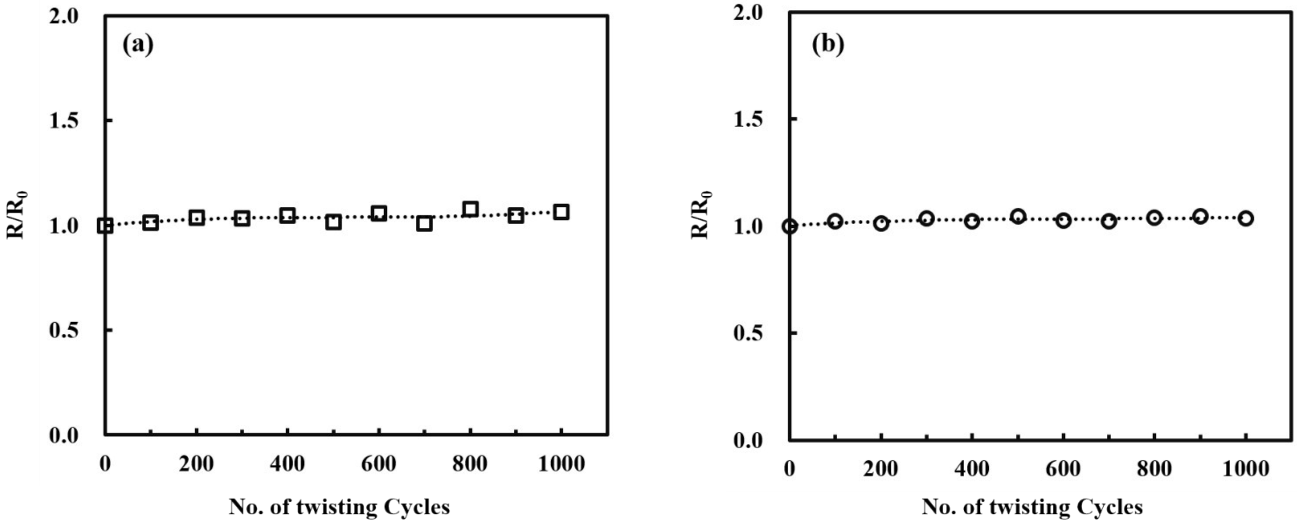

Fig. S6 Normalized resistance of 30 bilayer (940 nm thick) PEO/DWNT-PAA (open square) and 40 trilayer (900 nm thick) PEO/DWNT-PAA/GO (open circle) in cyclic twisting.

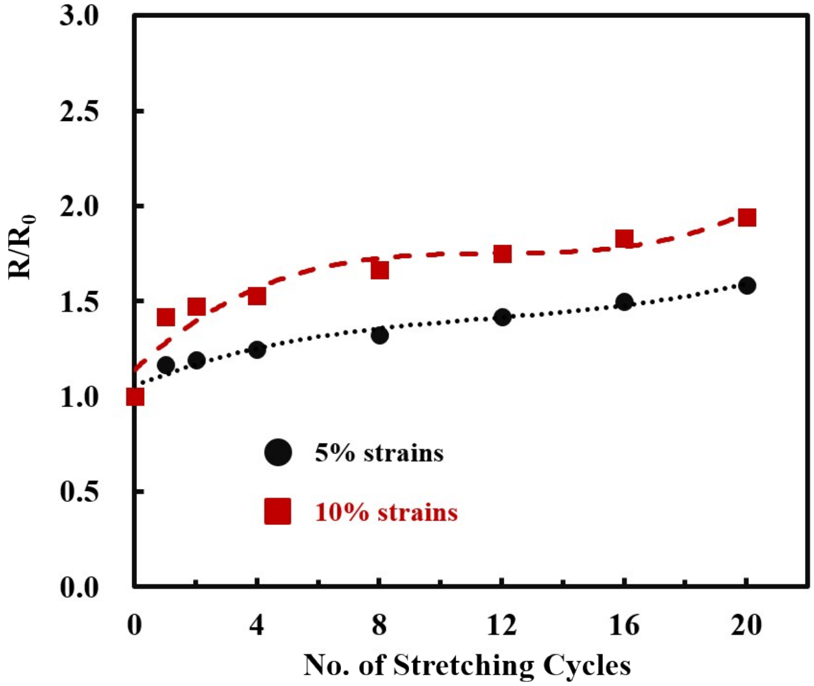

Fig. S7 Normalized resistance of 40 trilayer PEO/DWNT-PAA/rGO at a strain of 5% (circle) and 10% (square) as a function of the number of stretching cycles.

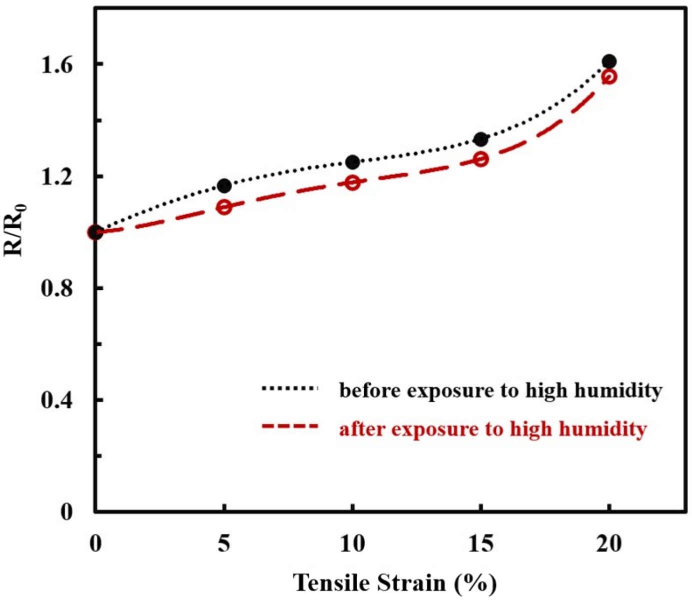

Fig. S8 Normalized resistance of 40 trilayer PEO/DWNT-PAA/rGO films as a function of tensile strain before (closed circle) and after (open circle) exposure to high humidity (~97% RH).

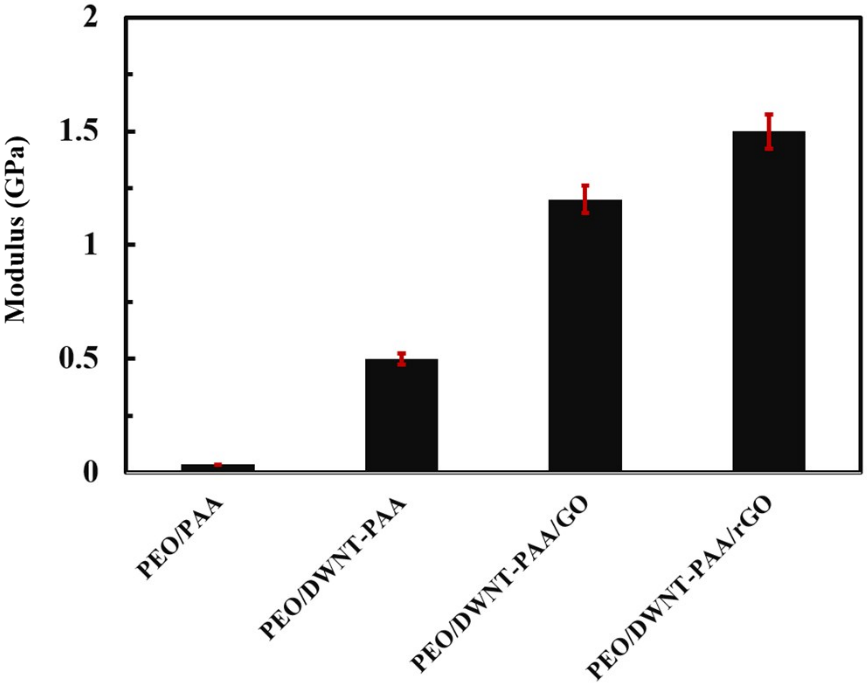

Fig. S9 Elastic modulus of 30 PEO/PAA BL, 30 PEO/DWNT-PAA BL, 40 PEO/DWNT-PAA/GO TL, and 40 PEO/DWNT-PAA/rGO TL films.

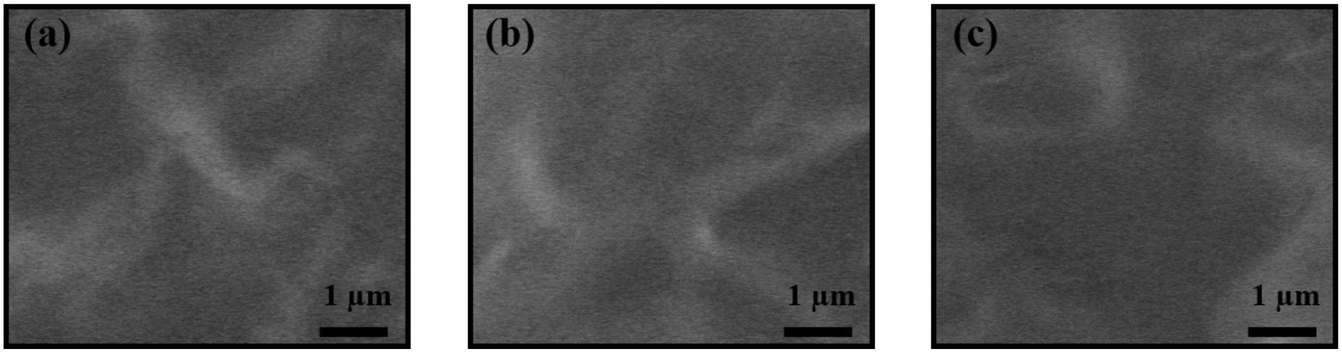

Fig. S10 SEM images of PEO/PAA films on 1 mm thick polyurethane rubber (a) before thermal treatment and after thermal treatment at 175 °C at 0% (b) and 50% strain (c).

References

(1) B. S. Shim, Z. Tang, M. P. Morabito, A. Agarwal, H. P. Hong, N. A. Kotov, *Chem. Mater.,* 2007, **9**, 5467.

(2) Y. T. Park, A. Y. Ham, J. C. Grunlan, *J. Phys. Chem. C*, 2010, **114**, 6325.