## **Electronic Supplementary Information**

## Highly Controllable Transparent and Conducting Thin Films Using Layer-by-Layer Assembly of Oppositely Charged Reduced Graphene Oxides

Dong Wook Lee, Tae-Keun Hong, Dongwoo Kang, Jisook Lee, Mihee Heo, Jin Young Kim,

Byeong-Su Kim\*, and Hyeon Suk Shin\*

Interdisciplinary School of Green Energy, UNIST (Ulsan National Institute of Science and Technology) Banyeon-ri 100, Ulsan 689-798, Republic of Korea

E-MAIL : <a href="mailto:shin@unist.ac.kr">shin@unist.ac.kr</a>; <a href="mailto:bskim19@unist.ac.kr">bskim19@unist.ac.kr</a>;

## **Supporting Information Available**



Figure S1. Zeta potentials at different pH values for negatively charged GO and rGO and positively charged GO and rGO solutions. Concentration of each solution is 0.005 wt%.



Figure S2. XPS survey spectra of negatively charged GO (top) and positively charged GO (bottom). The positively charged GO with amine groups shows N 1s peak.

	As-prepared (before annealing)			After annealing		
Bilayer (n)	Thickness (nm)	T (%)	R (KΩ/sq.)	Thickness (nm)	T (%)	R (KΩ/sq.)
2	5.0 ± 0.06	97	-	$5.2 \pm 0.02$	96	1800
4	7.7 ± 0.09	94	6900	$6.0 \pm 0.12$	93	74
6	$10.2 \pm 0.11$	92	3700	7.3 ± 0.19	91	32
8	$14.5 \pm 0.14$	91	2300	$10.5 \pm 0.31$	90	17
10	$15.2 \pm 0.12$	88	880	$12.0 \pm 0.31$	86	8.6
15	-	78	596	-	75	2.5

Figure S3. Thickness, transmittance (T), and sheet resistance (R) of rGO LbL multilayers before and after annealing.



Figure S4. AFM images and height profiles for 2, 6, and 10 bilayers of rGO LbL multilayers before and after annealing. Top three rows are for  $2 \times 2 \mu m$  regions and bottom three rows are for  $10 \times 10 \mu m$  regions.



Figure S5. SEM images of rGO multilayers with 2 bilayers (a), 6 bilayers (b), and 10 bilayers (c) on Si substrates after annealing.

	As-prepared (before annealing)			After annealing		
Bilayer (n)	Thickness (nm)	T (%)	R (KΩ/sq.)	Thickness (nm)	T (%)	R (KΩ/sq.)
2	4.6	99.5	-	2.0	97.6	-
4	7.0	98.9	-	6.4	96.0	-
6	8.5	98.0	-	7.2	94.7	524
8	14.1	97.8	-	14.4	91.0	67
10	16.4	96.5	-	20.2	87.6	21

(b)







Figure S6. (a) Thickness, transmittance (T), and sheet resistance (R) of GO LbL multilayers before and after annealing. (b) UV/Vis spectra of GO LbL multilayers before and after annealing. (c) AFM images and surface rms roughness (Rq) for 2, 6, and 10 bilayers of GO LbL multilayers before and after annealing.



Figure S7. Raman spectra of rGO (a and b) and GO (c and d) LbL films before and after annealing.