Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supporting Information

Rapid Oxygen Diffusive Lithium-Oxygen Battery using Restacking-inhibited, Free-Standing Graphene Cathode Film

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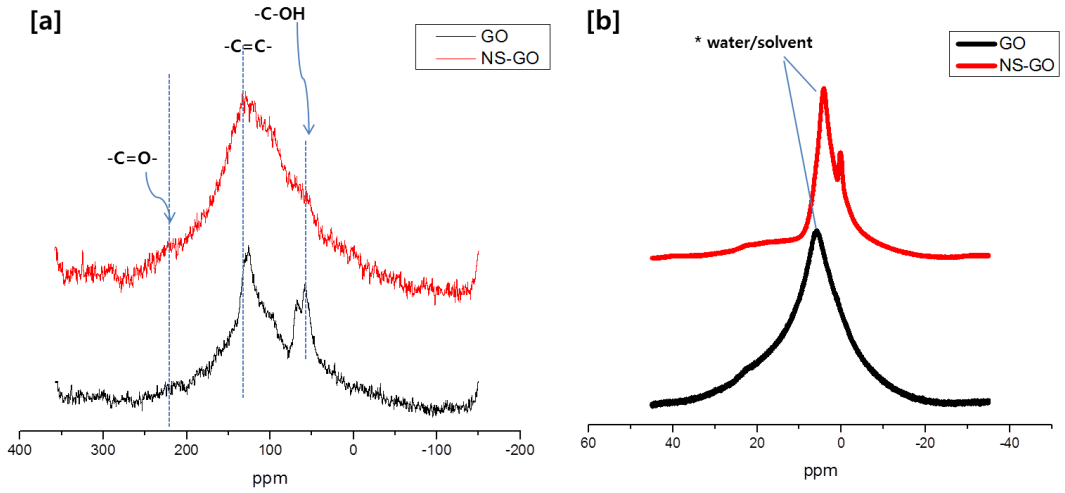
Figure S1. (a) The ^{13}C CP-MAS NMR and (b) ^{1}H MAS NMR of graphene oxide (GO, black) and non-stacked graphene oxide (NS-GO, red)



	Area	Center	Width	Height	%Area of C=C peak (SP2)	C-O peak Ratio (SP3)
GO peaks	4275500000	14.3	75.4	36078000	71	0.67
	7275200000	59.4	23.3	199130000		
	27828000000	124.0	59.5	297540000		
NS-GO peaks	736560000	1.7	21.9	21459000	77	0.29
	14445000000	70.4	85.5	107600000		
	49947000000	126.6	84.6	375680000		

Figure S2. The FT-IR spectra of graphene oxide (GO) and non-stacked graphene oxide (NS-GO)

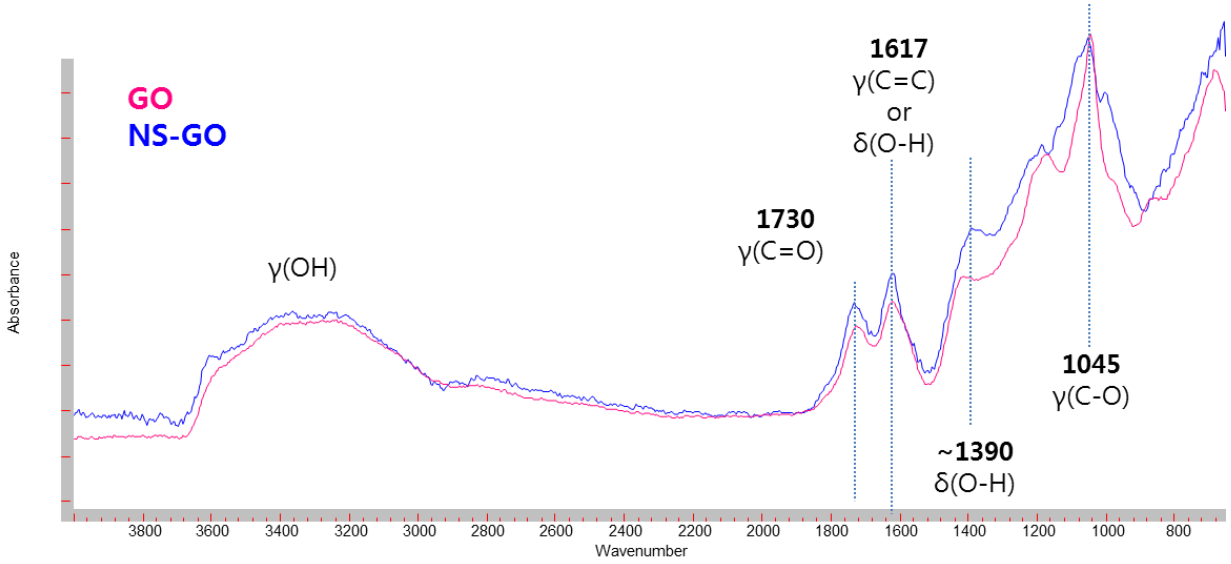


Figure S3. The ^{13}C CP-MAS NMR of (a) non-stacked reduced graphene oxide (NS-rGO) and (b) partially reduced non-stacked-GO (p*-NS-rGO)

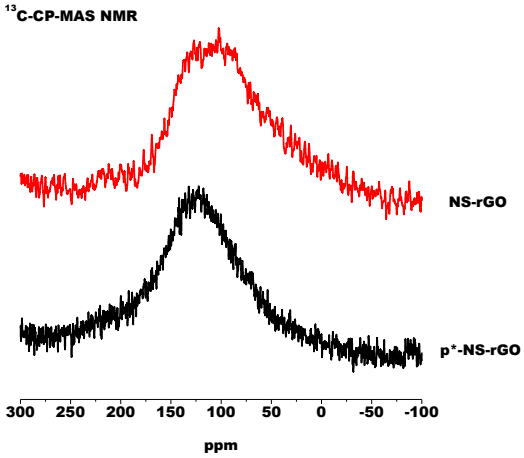
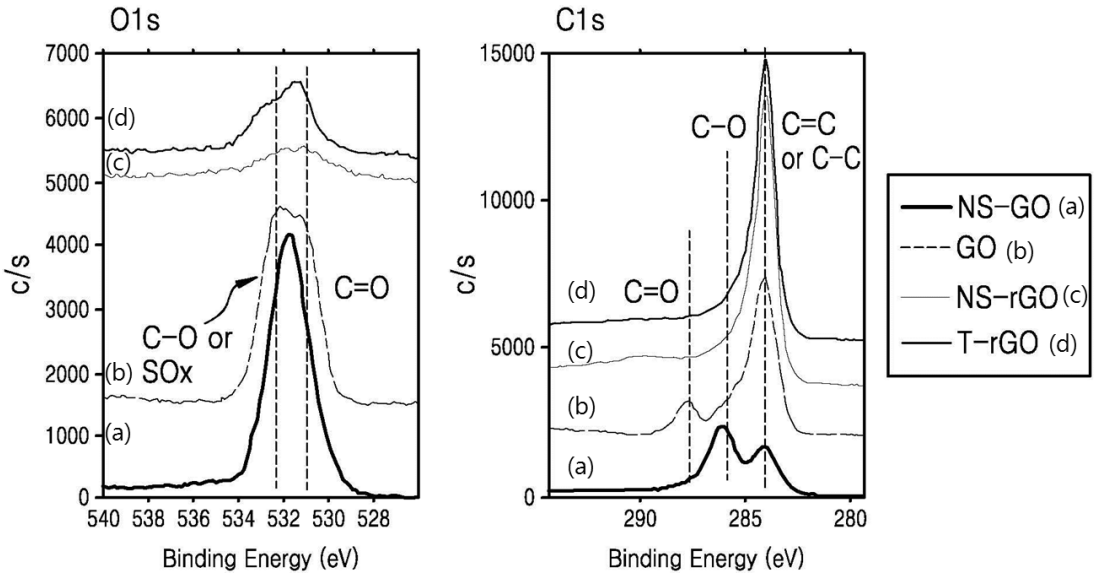


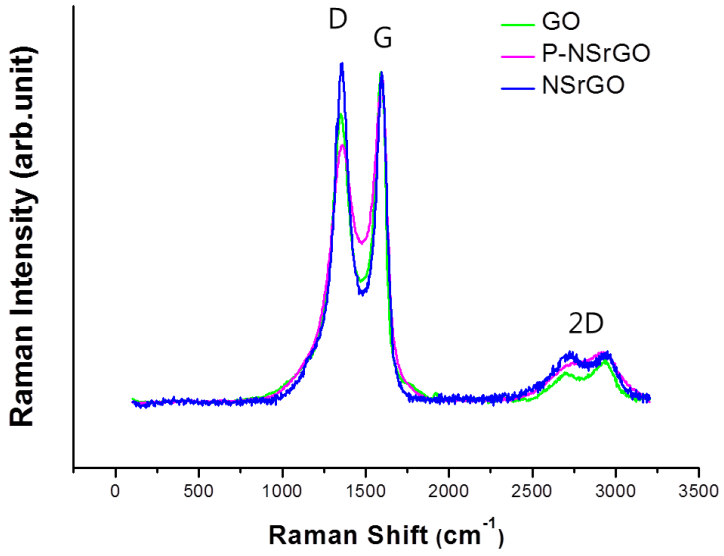
Figure S4. The XPS results of (a) graphene oxide (GO), (b) non-stacked graphene oxide (NS-GO), (c) thermally reduced GO (T-rGO), (d) non-stacked reduced graphene oxide (NS-rGO)



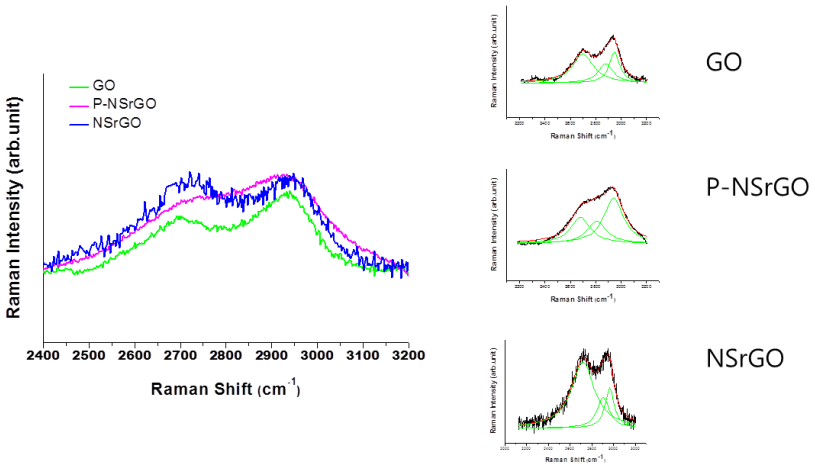
	C1s	N1s	O1s	Na1s	Si2p	S2p
(a) NS-GO	62.8	0.7	34.0	1.1	0.1	1.2
(b) GO	73.2	0.7	25.3	0.0	0.5	0.2
(c) NS-rGO	95.2	0.5	3.3	0.5	0.1	0.4
(d) T-rGO	90.7	0.0	7.0	0.1	2.0	0.2

Figure S5. The Raman spectra of graphene oxide (GO) and non-stacked graphene oxide (NS-GO)

(a) Raman spectra of Graphene oxide derivatives (I_D/I_G)



b) 2D bands of Graphene oxide derivatives



No.	D		G		ID/IG	2D	
	Xc	I_D	Xc	I_G		Xc	I_{2D}
GO	1352.6	0.82408	1595.5	0.84706	0.97	2699.4	0.077
P-NSrGO	1360.2	0.74267	1597.9	0.81399	0.91	2680.2	0.070
NSrGO	1356.1	0.96282	1598.3	0.89801	1.07	2717.4	0.128

Figure S6. The ^{7}Li MAS NMR spectra of the carbon (Printex) film (a) carbon:electrolyte=1:4, (b) carbon:electrolyte=1:8, (c) carbon:electrolyte=1:10 (d) NS-rGO:electrolyte=1:10, (e) the comparision results of peak width at the ^{7}Li MAS NMR spectra; The ^{7}Li MAS NMR spectra were obtained after soaking in electrolyte at a spinning rate of 13k, in which the graphene derivatives contain an ionic liquid electrolyte, (diethylmethyl(2-methoxyethyl)ammonium bis(trifluoromethane) sulfonamide, DEMA-TFSi) containing 1.0 M bis(trifluoromethane) sulfonimide lithium salt (LiTFSi).

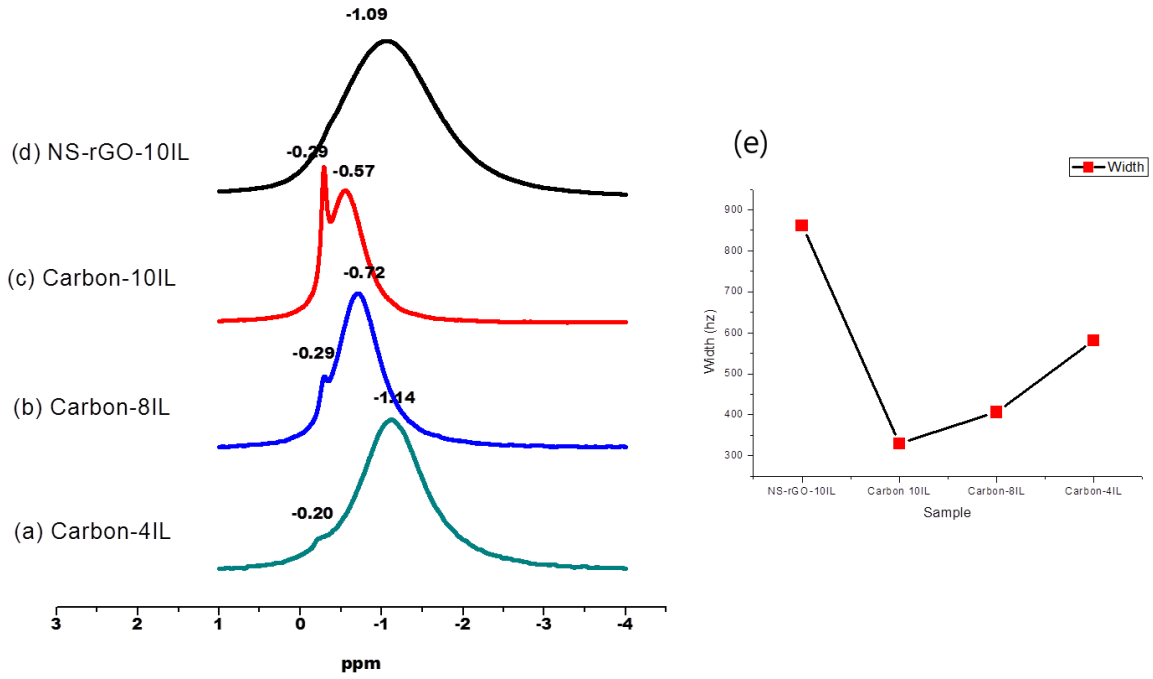


Figure S7. Cell structure (Li/interlayer/cathode/GDL/SUS) of a Li-air battery (Li-O_2 MIT cell) including the freestanding film, roll-pressed cathode; graphene derivatives/DEMA-LiTFSi-PTFE (1:5:0.2), cathode area (1.13cm^2), weight (3.9 mg/cm^2), graphene derivatives loading (0.64 mg/cm^2) at 60^oC

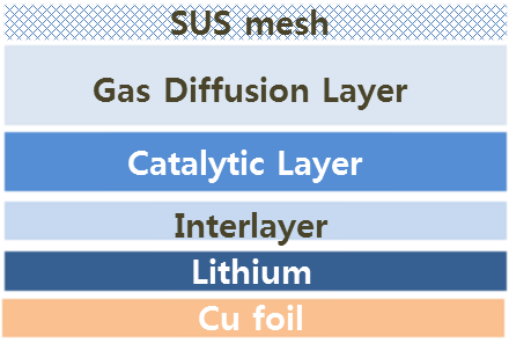


Figure S8. Discharge and charge profiles of non-stacked reduced GO (NS-rGO) for the cathode material up to 10 cycles under full discharge capacity condition. The test conditions included a cut-off voltage of ($1.8 < V < 4.5$), current density: $0.24 mA/cm^2$ under 1 atm oxygen gas and room temperature, discharge: CC mode, charge: CC-CV mode.

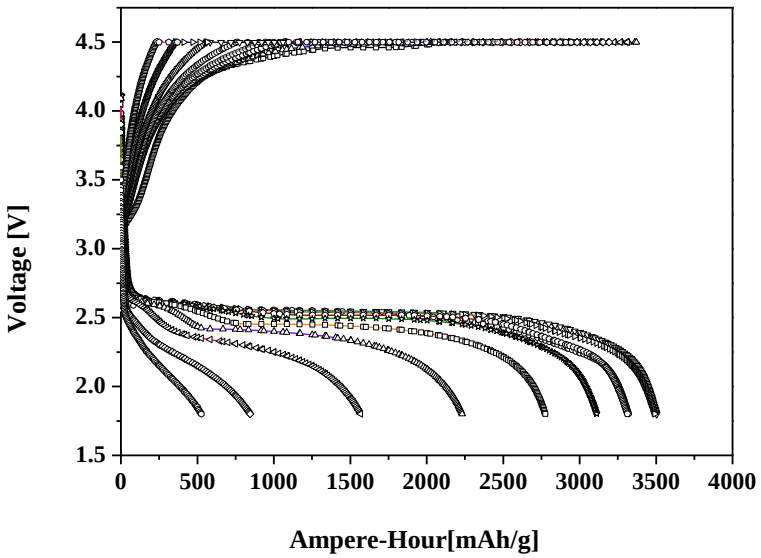
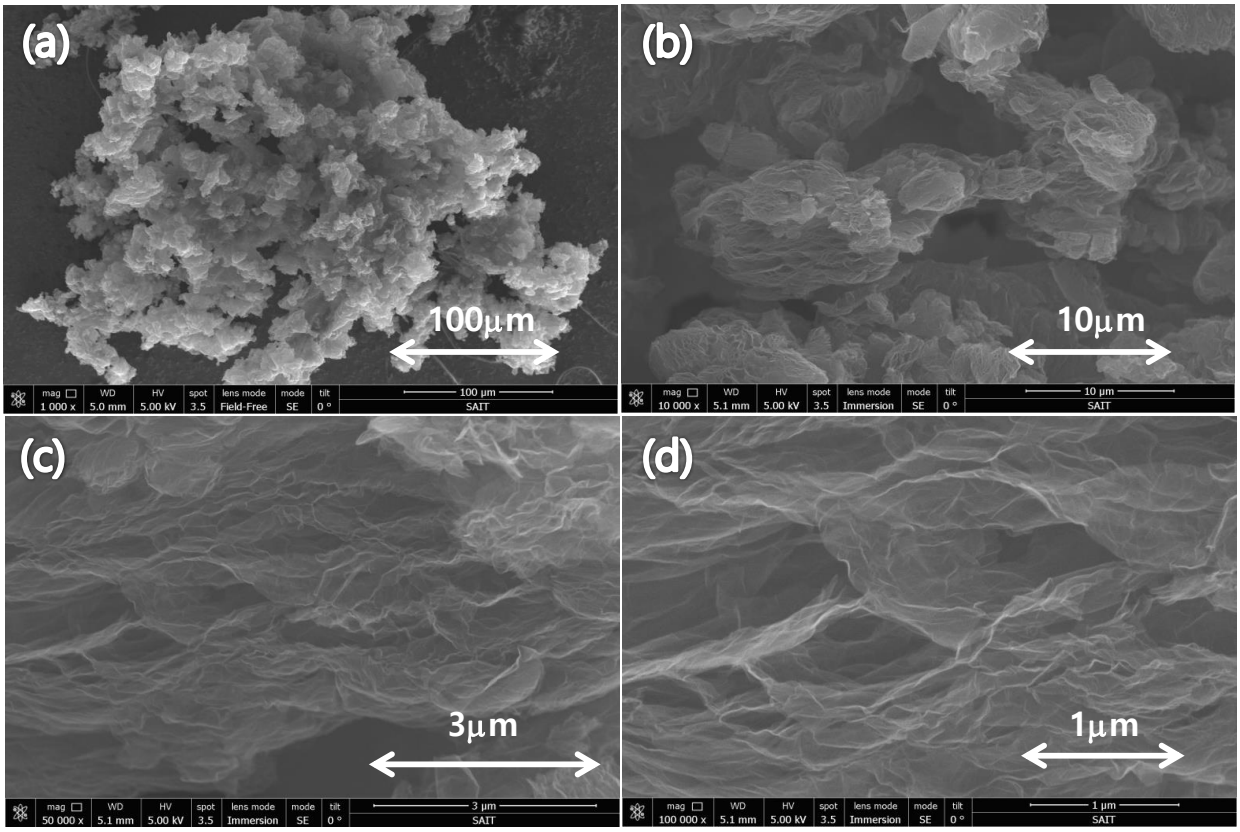


Figure S9. The cross-section SEM images of the NS-rGO powder and magnification ranging from (a)1,000X (b) 10,000X (c) 50,000X to (d) 100,000X



The synthetic process of non-stacked graphene oxide (NS-GO) and non-stacked reduced graphene oxide (NS-rGO):

Graphene oxide (GO) was prepared by the modified Hummer's method from natural graphite using sulfuric acid, potassium permanganate, and sodium nitrate. We followed our previous paper[1] to fabricate NS-GO powder and NS-rGO powder. Briefly, for NS-GO, the GO (1 g) was dispersed in ethanol (1 L) with homogenization (1 hour) and ultra-sonication (1 hour). After pouring cold hexane in dispersed GO solution, the co-solvent system GO solution was filtered by vacuum filtration with continuously stirring and adding pure hexane, and then the solvent was completely evaporated by rotary evaporator. For NS-rGO, the NS-GO (500 mg) on an alumina boat in quartz furnace was treated to form NS-rGO by thermal reduction at 1,000°C for 1 hour (1.5°C/min) under 3% hydrogen/nitrogen atmosphere or under vacuum condition. The total yield was about 40%. For comparison, p*-NS-rGO sample are performed at about 300°C for 1 hour (p*-NS-rGO).

*1. Y. Yoon, K. Lee, C. Baik , H. Yoo , M. Min , Y. Park, S. M. Lee, H. Lee, S. M. Lee , H. Lee, Adv. Mater., 2013, **25**, 4437–4444*

Table S1. Surface area and pore characterizations of the non-stacked reduced GO (powder) using BET method.

Samples	BET Surface area (m^2/g)	Pore volume (cm^3/g)	Pore diameter (nm)
Ns-rGO	3414	25.51	29.89
p*-NS-rGO	1206	6.13	20.33
NS-GO	325	0.7	8.6

Table S2. Surface area and pore characterizations of the non-stacked reduced GO (film) using BET method.

Samples	BET Surface area (m^2/g)	Pore volume (cm^3/g)	Pore diameter (nm)
Ns-rGO	164	1.04	25.3
T-rGO	107	0.24	8.96
C-rGO	103	0.23	9.06

Table S3. Discharge capacities of graphene oxides for the Li-air battery

Samples	NS-GO	p*-NS-rGO	NS-rGO	T-rGO	C-rGO
Discharge capacity [mAh/g]	150	250	3320	600	470

Table S4. Dimensions and capacities of gas-diffusion layers (a) carbon paper, (b) NS-rGO

Samples	Weight (mg/cm^2)	Thickness (um)	Capacity (mAh)	Specific capacity (mAh/g)
GDL-35BA (carbon paper)	4.3	200	1.52	174
NSrGO	0.61	7	1.62	314

* Cathode layer: CM250 (MWCNT)