Ice-templated, pH-tunable self-assembly route to hierarchically porous graphene nanoscroll networks

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Supporting Information

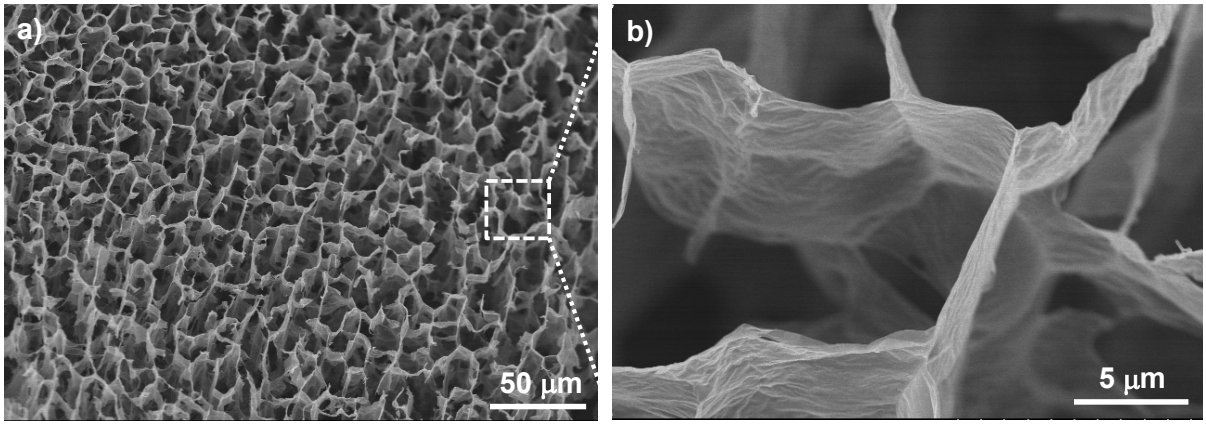

Figure S1. The ice crystal-induced formation of microscale (5-15 μm) pores for high density GO solution after a freeze-casting process. Microscale honeycomb structures of GO sheets are formed due to the formation of ice crystals during the freeze-casting process.

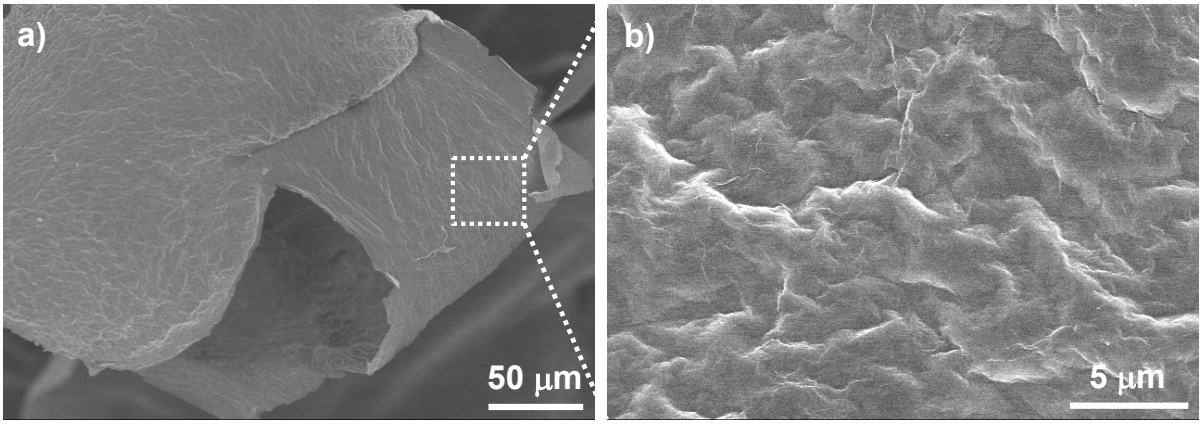

Figure S2. Stacked multilayer graphene sheets after slow-freezing of the rGO solution in the refrigerator and freeze-drying processes.

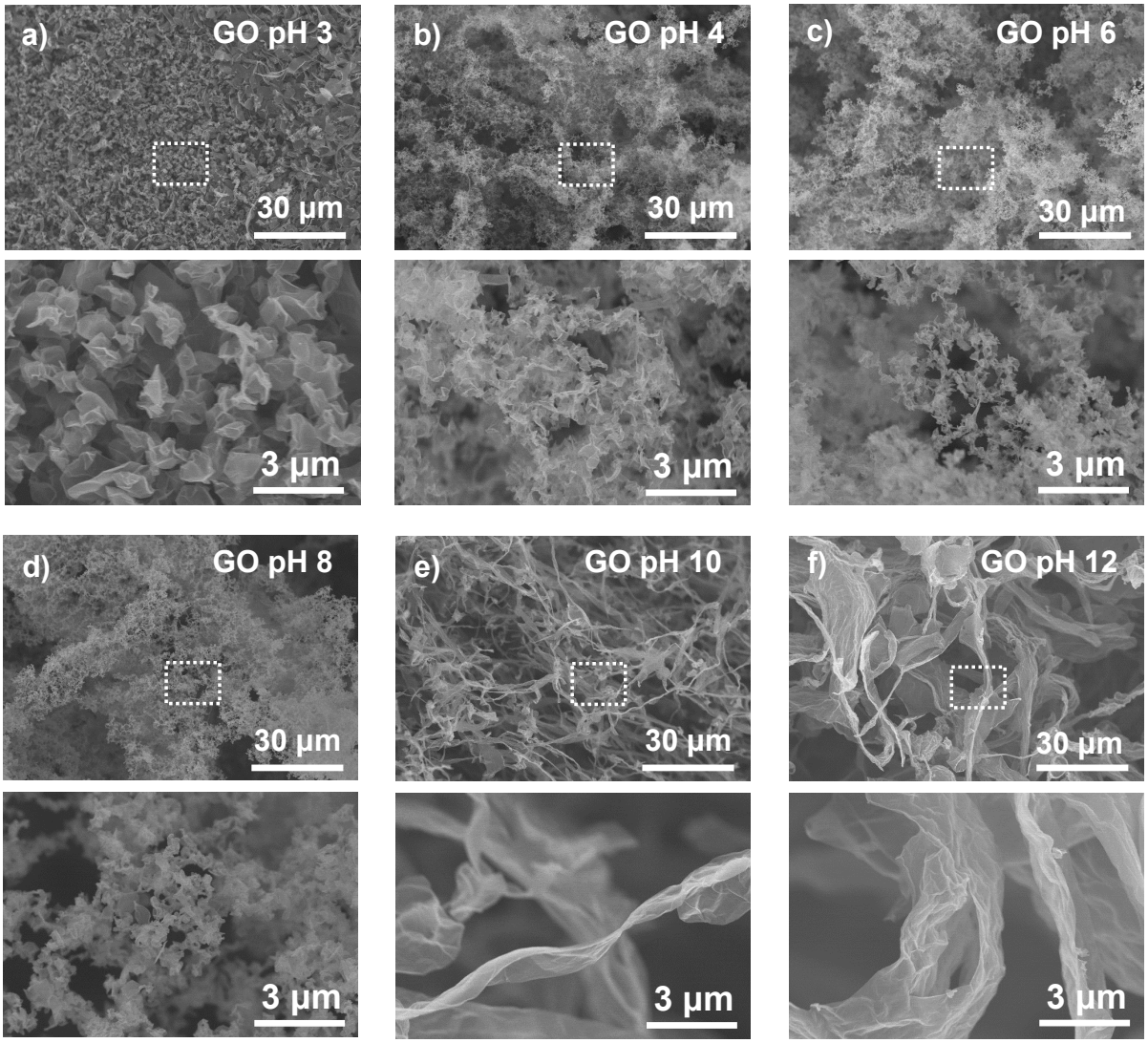

Figure S3. Various morphologies of porous GO structures after freeze-casting of GO solution at different pH conditions. At low pH conditions (a,b,c,d), GO crumpled into micrometer scale particles without stacking together. At pH 10 (e), GO tends to form mostly twisted and self-folded structures with only small portions of rolled structures.

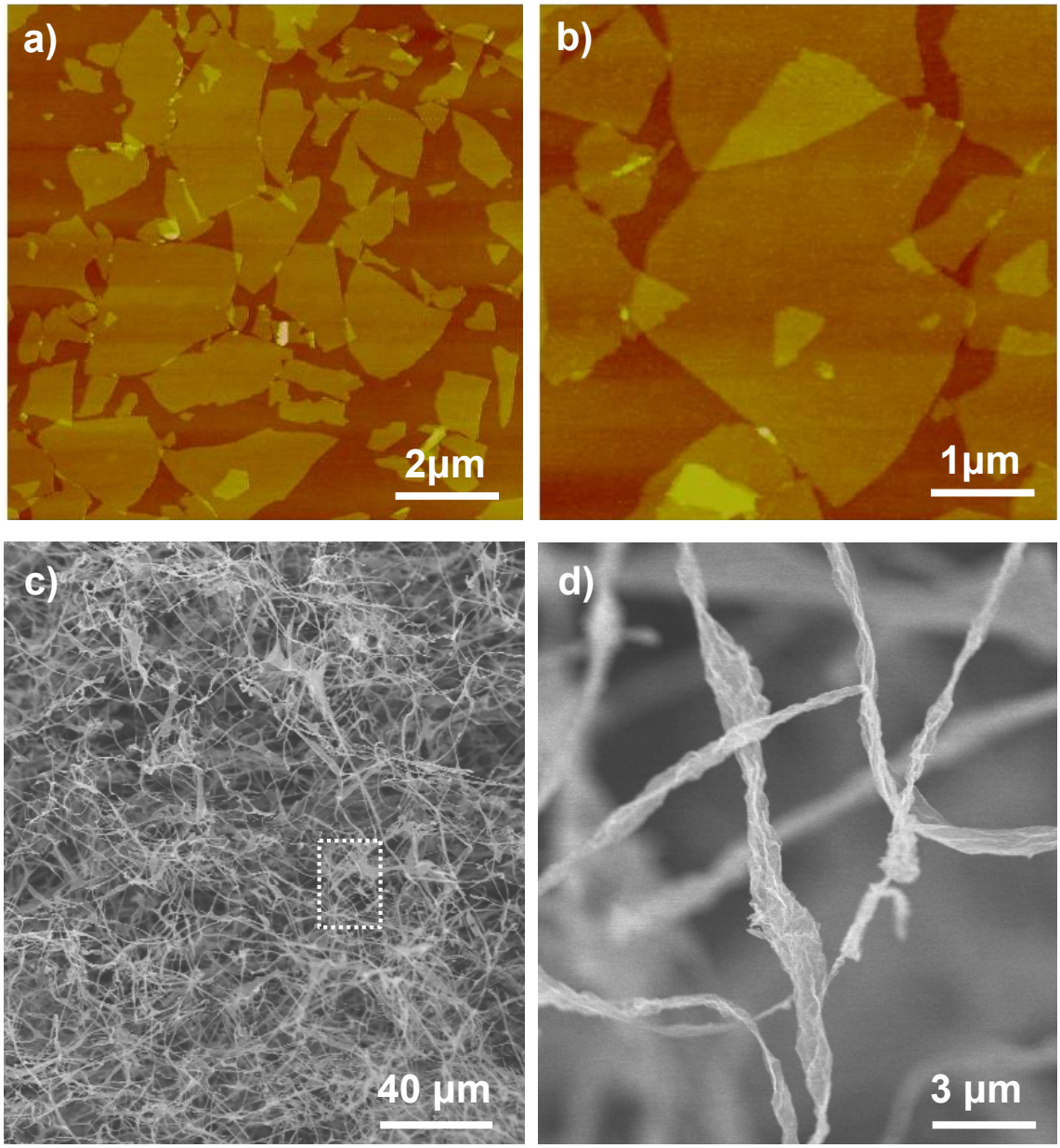

Figure S4. (a) and (b) AFM images of large size rGO (~4 μm) sheets. (c) and (d) SEM images of crumpled graphene nanoscrolls when large size rGO sheets are used in the formation of graphene nanoscrolls.

Table S1. Textural properties of graphene nanostructures assembled from GO and r-GO. BET surface area and total pore volume of the graphene nanostructures.

Sample	BET Surface Area ($m^2 g^{-1}$)[a]	Total Pore Volume ($cm^3 g^{-1}$)[b]
GO-pH6	100	0.26
GO-pH10	50	0.13
rGO-pH6	90	0.19
rGO-pH10	110	0.40

[a]BET surface area was calculated by BET equation at the relative pressure range of 0.05-0.30.

[b]Total pore volume was determined at the relative pressure of 0.98-0.99.

Table S2. Comparison of the ORR activity with previous results. Table showing catalyst l oadings, half-wave potentials, and kinetic currents (at 0.7 V (vs. RHE)) to compare the ORR activity of our rGO-pH10 catalyst with previously reported N-doped graphene nanostructure

Sample	Loading (μg cm^{-2})	Reference electrode [a]	Halfwave potential (V)	Kinetic curren t density @ 0.7 V (mA cm^{-2})	N content	Reference
rGO-pH10	60	Hg/HgO	0.65	1.5	4 % N/C = 5.3 %	This work
N-Graphene	40	Ag/AgCl	0.62	0.8	N/C = 4 at%	1
N-Graphene	n/a	Ag/AgCl	0.60	0.3	1.7 at%	2
N-Graphene hollow spher e	140	Ag/AgCl	0.54	0.2	3.8 at%	3
N-Graphene (from urea)	140	Ag/AgCl	0.45	0.2	7.9 at%	4
Pyridine-grap hene	140	Ag/AgCl	0.52	0.2	N/C = 5 at%	5
PANI-rGO	1000	Ag/AgCl	0.73	9.8	7.4 at%	6
Porous N-gra phene	255	Hg/HgO	0.70	3.6	7.8 wt%	7

[a] Potential versus RHE scale was estimated by the RHE calibration carried out in this report. The calib ration value was determined to be E(RHE) = E(Ag/AgCl) + 0.916 V = E(Hg/HgO) + 0.880 V.

References for Table S2

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