Electronic Supplementary Material (ESI) for Nanoscale.

Supplementary information

Massive Production of Graphene Nanoscrolls and Their Assist for High Rate Performance Supercapacitors

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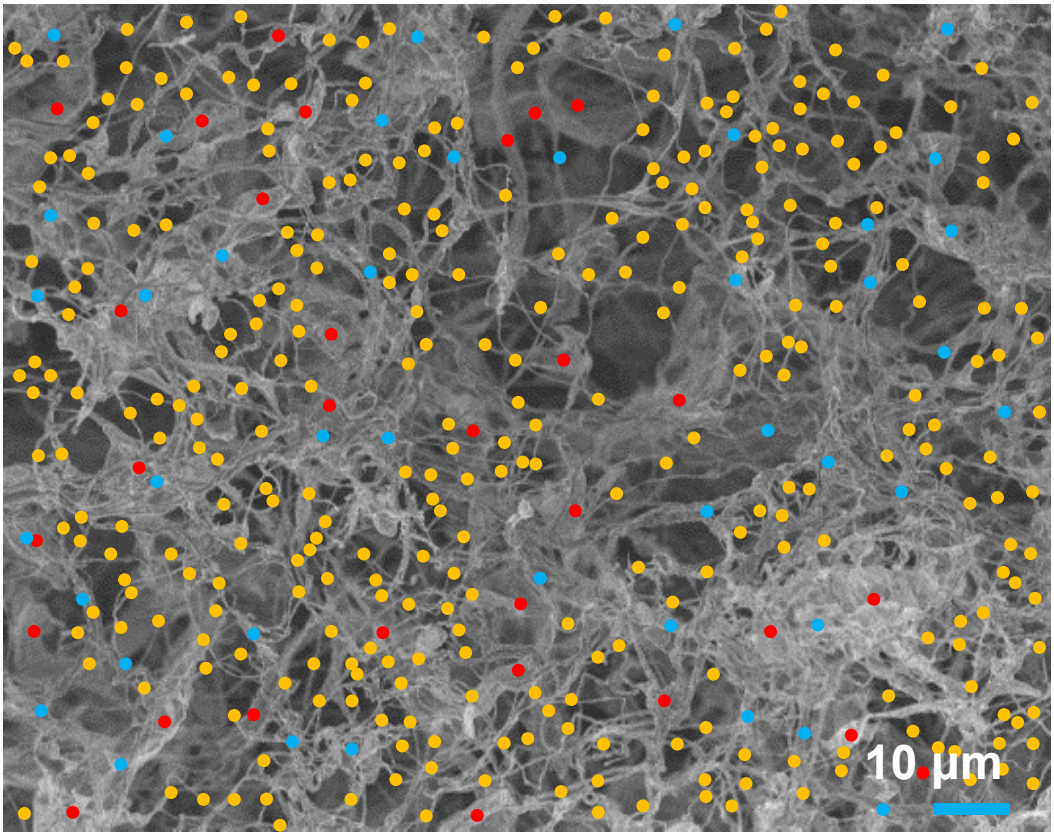


Figure S1 SEM image for the assessment of the efficiency to transform CRG to GNS at the concentration of 0.5 mg/mL. The red dots indicate CRG sheets, the yellow dots indicate GNSs and blue dots indicate GNS bundles. Through counting, there are 309 GNSs, 30 CRG sheets, and 42 GNS buddles. The minimal yield was calculated as (309+42)/(309+42+30) × 100% ≈92%.

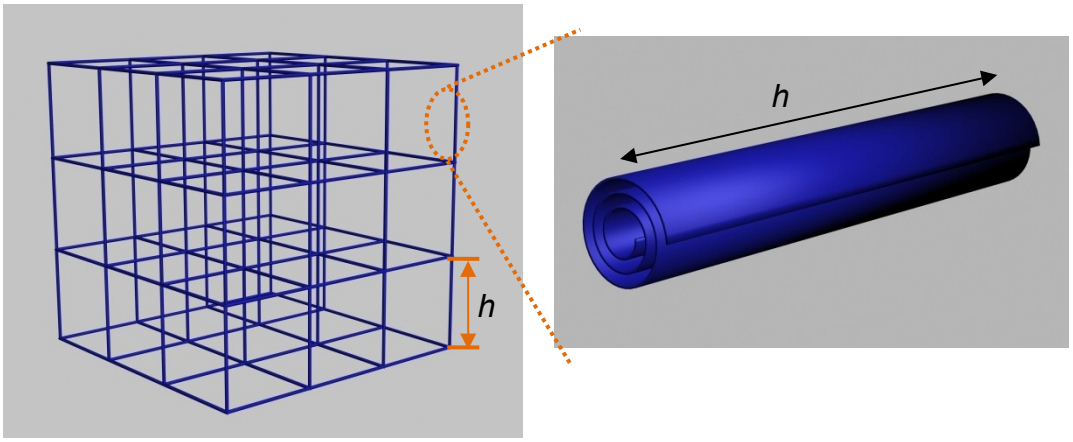


Figure S2. Density calculation model.

Here, we assume that a cubic framework is made up of 12 rigid GNSs with the same length h. Because of the open morphology of GNS, the mass of a GNS $m' = A/S$, where A is the area of one GNS and S is the specific surface area of graphene sheet. Assuming that GNS is formed by a piece of square-shaped graphene sheet and the length of GNS equals the length of the square. Therefore, the calculated density of GNS foam ρ is:

$$\rho = \frac{12m'}{4V} = \frac{12 \times (\frac{2h^2}{S})}{4h^3} = \frac{6}{Sh}$$

All the above formula is based on the assumption of rigid GNSs and easy connections between GNSs. As to the real case, GNS is flexible and the interactions between GNSs are week physical entanglements. The above reason hinders the decrease of density in experiment: GNSs tend to collapse when the intrinsic elastic force and physical entanglement force are hard to bear their self-weight.

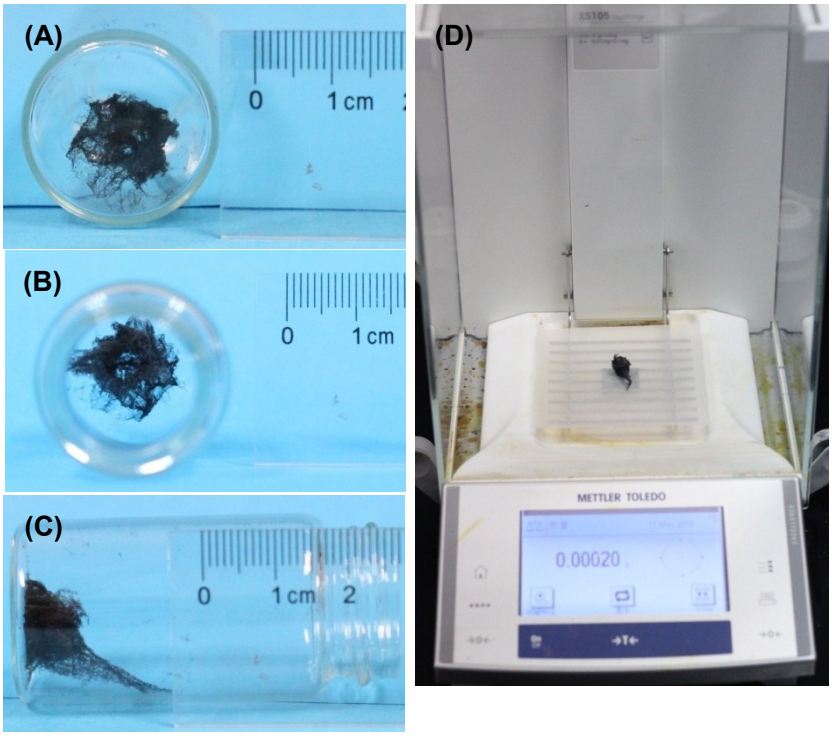


Figure S3. (A) bottom, (B) top, (C) side views of GNS1; (D) Weight of GNS1 sample. The volume of GNS1 foam is 2 ml according to the volume calculation formula of circular truncated cone: $V=1/3\ \pi\ h\ (R^2+r^2+Rr)$. (h=0.8cm, R=1.0cm, r=0.8cm)

Table S1 Specific capacitances and retention rates of GNSs and MWNTs (calculated by CV, the electrolyte is 1M H_2SO_4)

	GNS2		GNS3		GNS4		GNS5		MWNT	
Scan rate (mV/s)	***Cs* (F/g)**	**Retention (%)**	***Cs* (F/g)**	**Retention (%)**	***Cs* (F/g)**	**Retention (%)**	***Cs* (F/g)**	**Retention (%)**	***Cs* (F/g)**	**Retention (%)**
10	91.0	100	91.4	100	103.6	100	99.0	100	46.7	100
20	88.5	97.2	89.0	97.4	99.7	96.2	94.5	95.5	44.5	94.7
50	84.2	92.5	84.2	92.1	94.7	91.4	88.0	88.9	41.5	89.0
80	82.3	90.4	81.9	89.6	90.8	89.7	81.9	82.7	40.1	85.9
100	82.3	90.4	80.8	88.4	90.0	86.8	79.5	80.3	39.3	84.2
200	77.5	85.2	75.0	82.1	84.0	81.1	72.0	72.7	36.6	78.4
500	69.6	76.5	60.0	65.6	75.6	73.0	60.0	60.6	31.3	67.1
800	64.6	71.0	57.5	62.9	70.5	68.1	56.3	56.8	28.3	60.6
1000	61.8	67.9	51.0	59.1	67.9	65.5	50.0	50.5	26.4	56.6
2000	53	58.2	41.5	45.4	58.5	56.5	45.0	45.5	19.8	42.5

The specific capacitance of single electrode based on CV curve is:

$$C_m = \frac{\int_{U_1}^{U_2} IdU + \int_{U_2}^{U_1} IdU}{m \times u \times (U_2 - U_1)}$$

Where C_m (F/g) is the specific capacitance, m (g) is the mass of single electrode, u (V/s) is the scan rate, U_2 and U_1 (V) are the highest and lowest value of the potential window, I (A) is the instant current.

Table S2 Specific capacitances and retention rates of GNSs and MWNTs (calculated by GCD, the electrolyte is 1M H_2SO_4)

	GNS2		GNS3		GNS4		GNS5		MWNT	
	Cs (F/g)	Retention (%)	*Cs* (F/g)	Retention (%)	*Cs* (F/g)	Retention (%)	*Cs* (F/g)	Retention (%)	*Cs* (F/g)	Retention (%)
0.5	78.8	100	80.1	100	86.7	100	93.5	100	40.9	100
0.8	80.5	102.1	80.7	100.9	89.1	102.8	91.9	98.2	40.4	98.8
1	80.6	102.3	81.3	101.5	90.4	104.2	91.0	97.4	40.2	98.4
2	80.1	101.6	80.5	100.5	91.1	105.1	89.6	93.7	39.0	95.3
5	77.3	98.1	77.4	96.6	89.2	102.8	81.5	87.2	36.4	89.1
8	75.3	95.5	74.9	93.5	87.0	100.3	77.3	82.6	34.6	84.6
10	73.8	93.7	73.8	92.1	85.7	98.8	75.1	80.3	33.4	81.6
20	69.6	88.3	67.7	84.6	80.8	93.2	67.4	72.1	29.4	72.0
50	61.5	78.0	56.7	70.8	72.6	83.8	56.6	60.5		

The specific capacitance of single electrode based on GCD curve is:

$$C_m = \frac{I \times t}{\Delta U \times m}$$

Where C_m (F/g) is the specific capacitance, m (g) is the mass of single electrode, I (mA) is the discharge current, t (s) is the length of discharge time, ΔU (V) is the potential window.

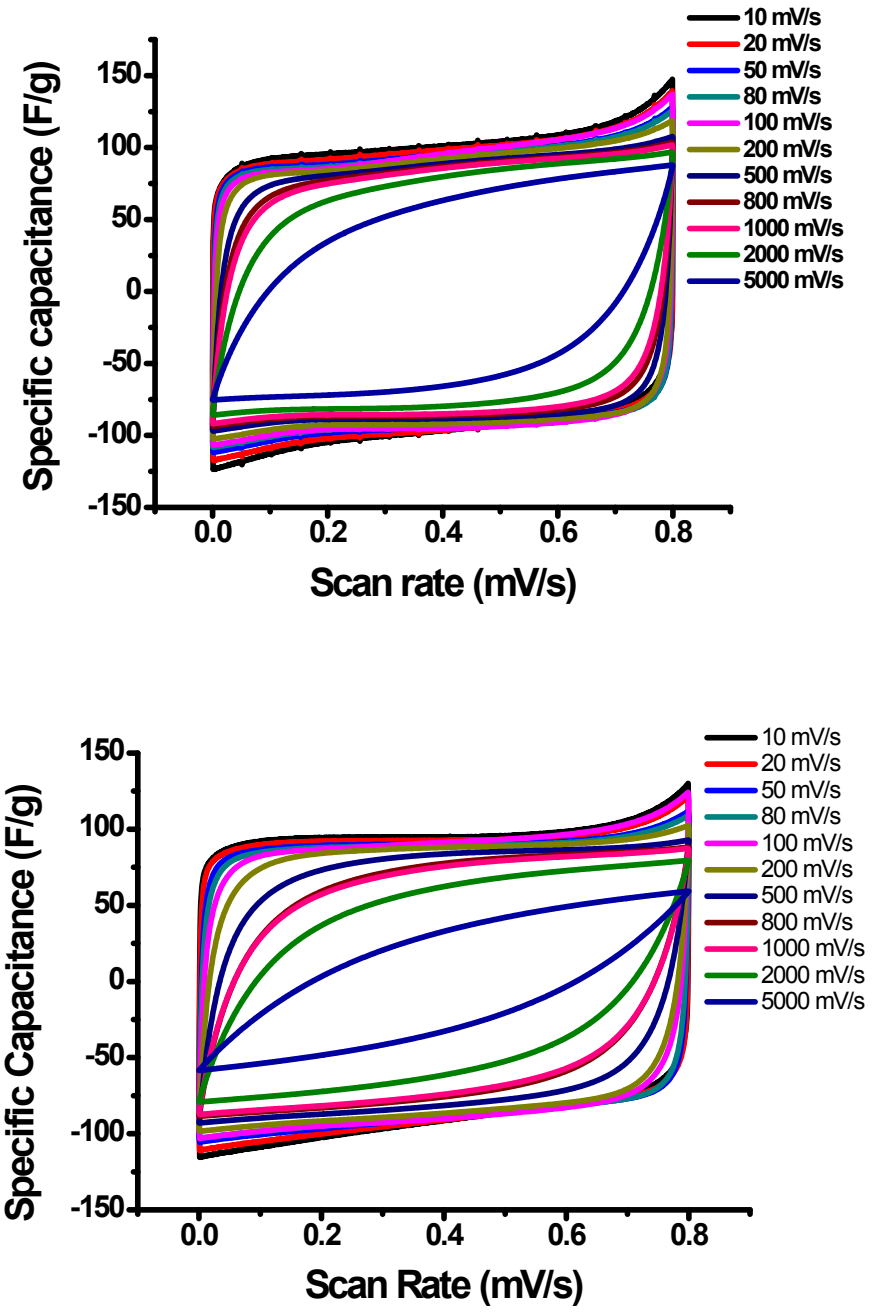


Figure S4. CV curves of AGF1 (top) and AGF2 (bottom), the electrolyte is 1M H_2SO_4.

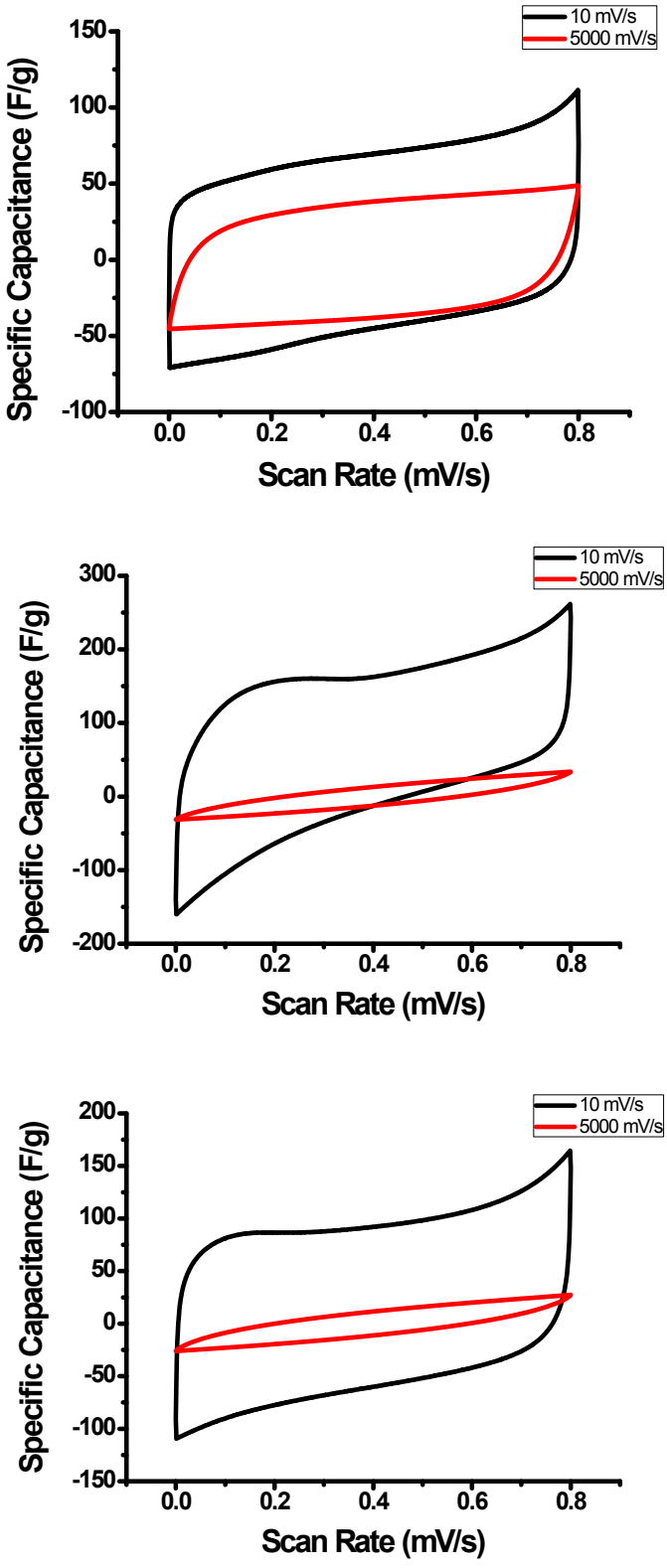


Figure S5. Shape deterioration from 10mV/s to 5000 mV/s of AGF3, RGO and CGF3 (in 1M H_2SO_4).

Table S3 Electrochemical performance comparisons of materials in this articles and reported works.

Ref	Electrode Materials	*Cs* (F/g)	R (%)	*Cs* (F/g)	R (%)	Cs (F/g)	R (%)	Cs (F/g)	R (%)	*Cs* (F/g)	R (%)
This work	GNS (1M H_2SO_4)	1 A/g		10 A/g		20 A/g		50 A/g		100 A/g	
		80.6	100	73.8	91.6	69.6	86.4	61.5	76.3	--	--
This work	CNT (1M H_2SO_4)	1 A/g		10 A/g		20 A/g		50 A/g		100 A/g	
		40.9	100	33.4	81.7	29.4	71.9	--	--	--	--
This work	AGF (1M H_2SO_4)	1 A/g		10 A/g		20 A/g		50 A/g		100 A/g	
		166.8	100	155.2	93.0	149.6	89.7	140.0	83.9	129.1	77.4
This work	CGF (1M H_2SO_4)	1 A/g		10 A/g		20 A/g		50 A/g		100 A/g	
		108.0	100	90.5	83.8	75.6	70.0	44.8	41.5	--	--
This work	RGO (1M H_2SO_4)	1 A/g		10 A/g		20 A/g		50 A/g		100 A/g	
		115.7	100	109.1	94.3	95.8	82.8	61.0	52.7	--	--
48	Carbon nanocages (1M H_2SO_4)	1 A/g		10 A/g		100 A/g		--		--	
		216	100	178	82.4	112	51.9	--	--	--	--
49	Folded structured graphene paper (1M H_2SO_4)	1 A/g		50 A/g		100 A/g		--		--	
		172	100	135	78.5	110	64.0	--	--	--	--
50	Self-stacked solvated graphene film (1M H_2SO_4)	0.108 A/g		100 A/g		--		--		--	
		215	100	180	83.7	--	--	--	--	--	--
51	Vertically oriented graphene (6M KOH)	1 A/g		20 A/g		100 A/g		--		--	
		185	100	165	89.2	156	84.3	--	--	--	--

53	Wrinkle-structured graphene (1M H_2SO_4)	1 A/g		10 A/g		50 A/g		100 A/g		--	
		177	100	170	96.0	152	85.9	140	79.1	--	--
54	Non-stacked reduced graphene oxide (6M KOH)	1 A/g		30 A/g		--		--		--	
		236.8	100	171.2	72.3	--	--	--	--	--	--

Cs and R refer to gram specific capacitance and retention rate, respectively.

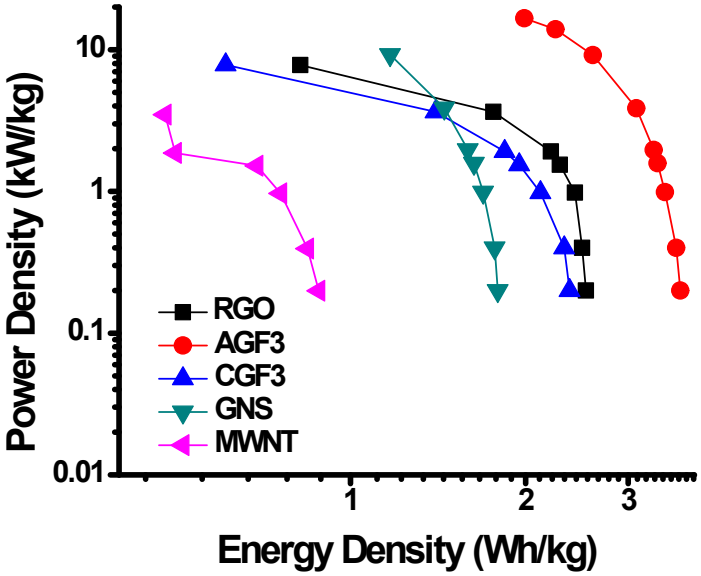


Figure S6. Ragone plots of the materials used in this article (aqueous electrolyte, based on GCD tests).

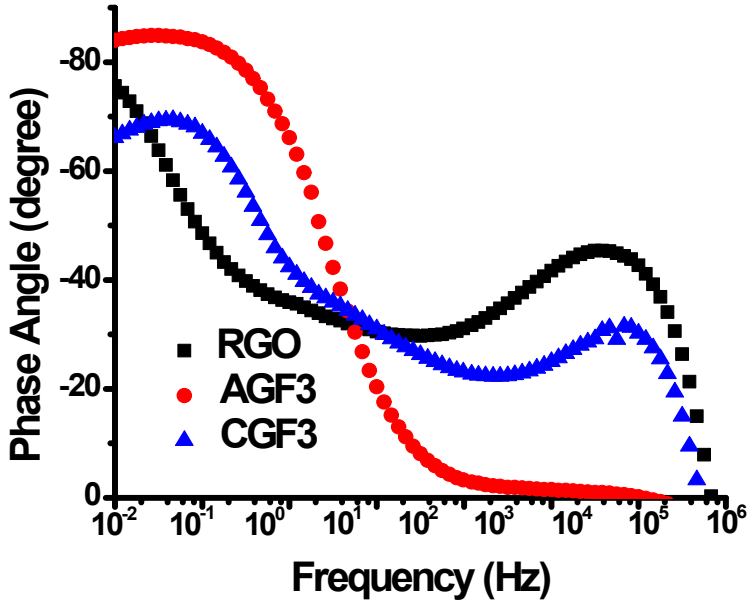


Figure S7. Phase angle-frequency relationship for RGO, AGF3 and CGF3 in 1M H_2SO_4.

Table S4 Specific capacitances and retention rates of RGO film, AGFs and CGFs tested in organic electrolyte. (Calculated by CV)

	RGO		AGF3		CGF3	
Scan rate (mV/s)	***Cs* (F/g)**	**Retention (%)**	***Cs* (F/g)**	**Retention (%)**	***Cs* (F/g)**	**Retention (%)**
10	84.4	100	113.5	100	87.7	100
20	61.8	73.3	87.5	77.0	59.2	67.5
50	35.5	42.0	57.7	50.8	33.3	37.9
80	25	29.6	43.9	38.7	25.2	28.6
100	20.6	24.4	38.1	33.5	22.1	25.2
200	12.1	14.3	25.0	22.0	15.0	17.1
500	6.6	7.9	13.8	12.1	8.9	10.2
800	4.9	5.8	10.2	8.9	6.8	7.8
1000	4.5	5.3	8.8	7.8	6.2	7.0
2000	2.9	3.5	5.7	5.0	3.9	4.5
5000	1.5	1.8	3.1	2.7	2.0	2.3

Table S5 Specific capacitances and retention rates of RGO film, AGFs and CGFs tested in organic electrolyte. (Calculated by GCD)

	RGO		AGF3		CGF3	
Current density (A/g)	***Cs* (F/g)**	**Retention (%)**	***Cs* (F/g)**	**Retention (%)**	***Cs* (F/g)**	**Retention (%)**
1	100.9	100	122.2	100	112.3	100
2	54.1	53.6	88.7	72.6	55.7	49.6
5	15.5	15.4	45.1	36.9	17.2	15.3
8	6.1	6.1	27.6	22.6	8.3	7.4
10	3.8	3.8	14.6	12.0	5.7	5.1
20	0.001	0.001	4.8	3.9	1.4	1.2

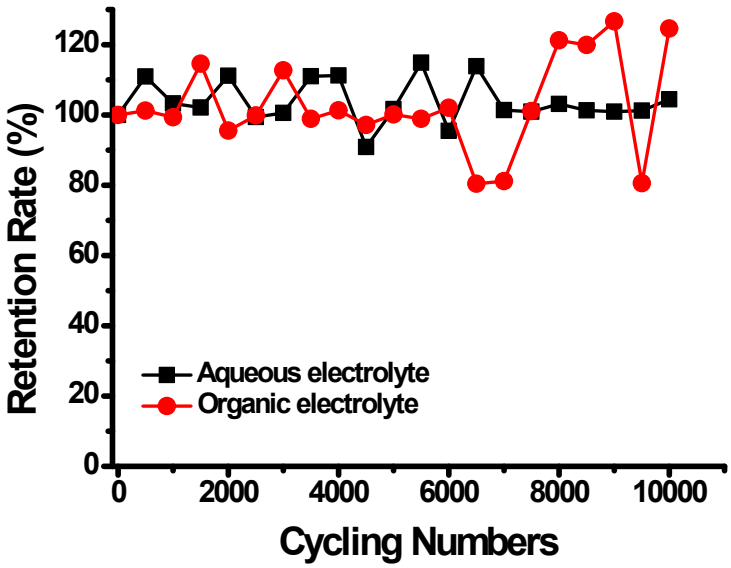
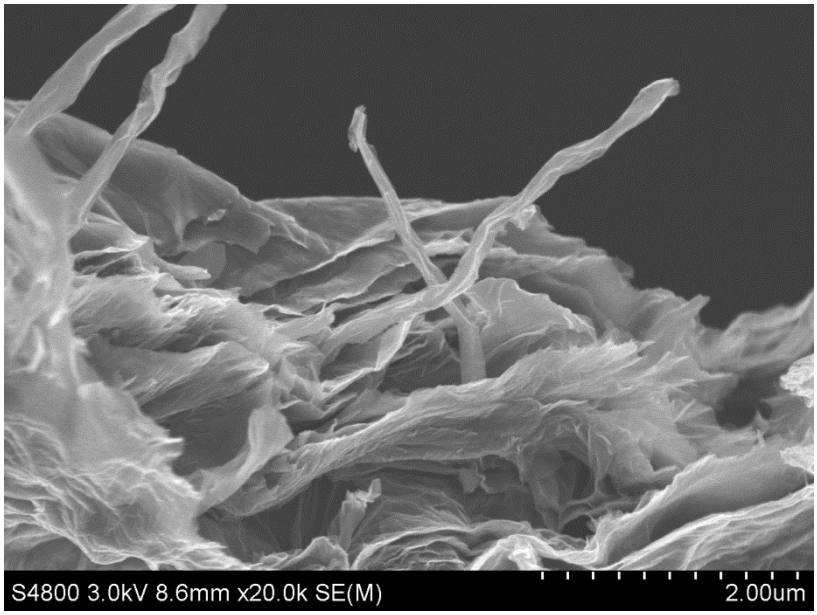


Figure S8. Cycling stability tests of AGF3 in aqueous electrolyte (1M H_2SO_4) and organic electrolyte (1M $EMIM^+$ BF_4^- in AN) at 5 A/g.

(A)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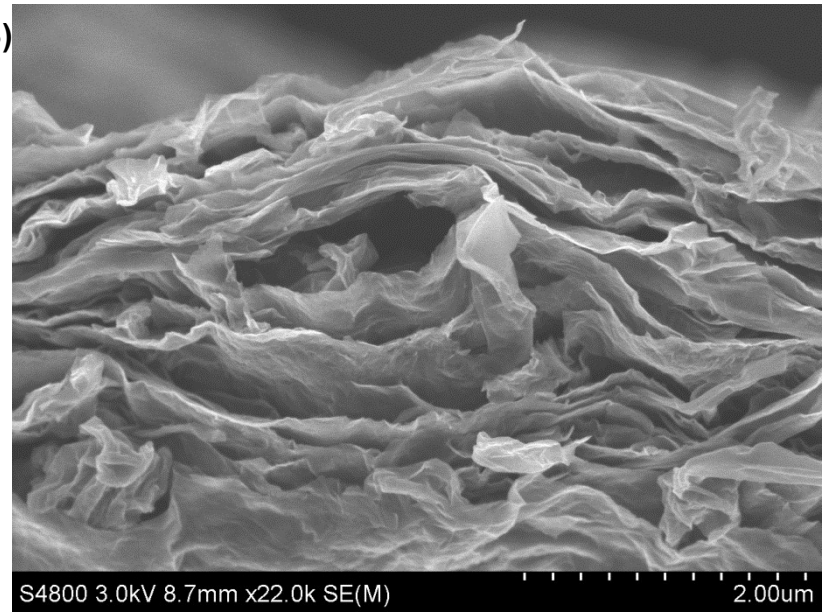
2.00um

(B)


S4800 3.0kV 8.7mm x22.0k SE(M)
2.00um

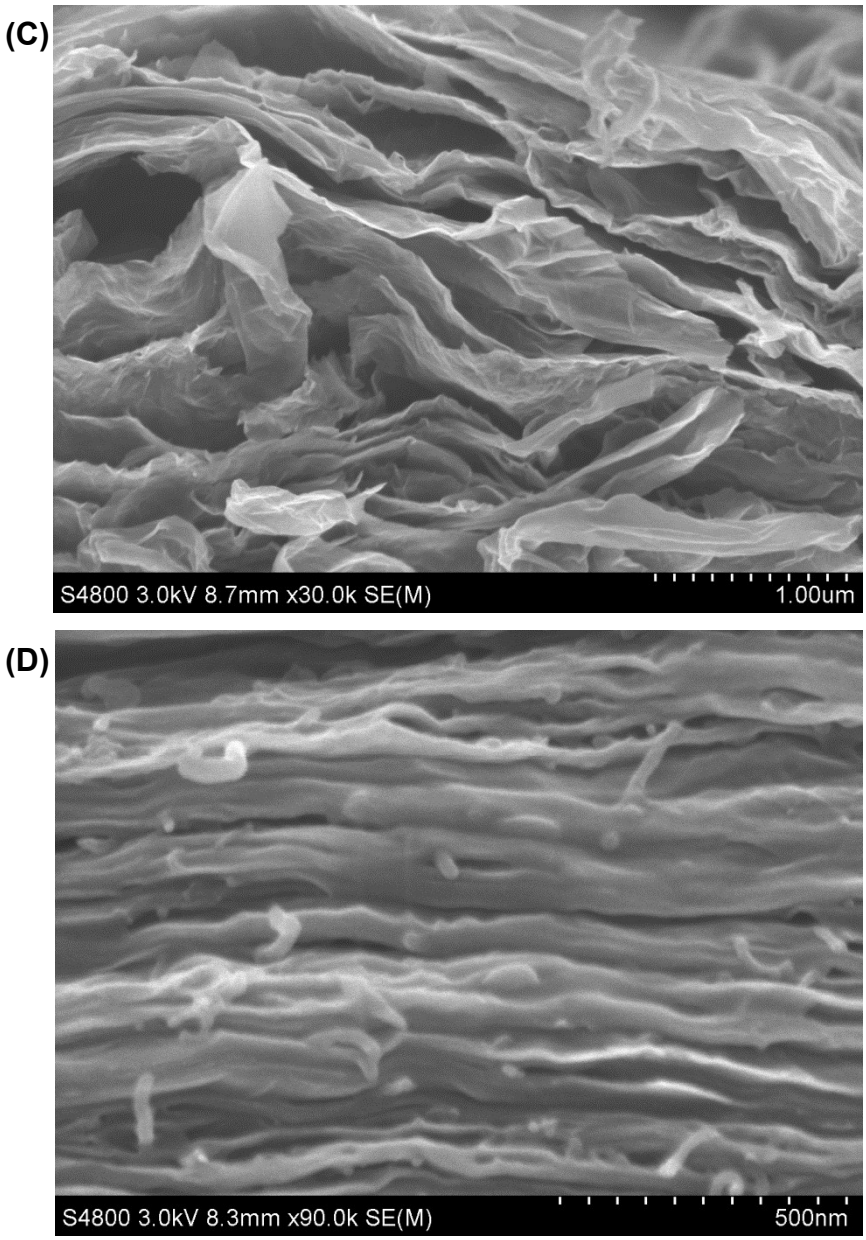


Figure S9. (A) Magnified SEM images of AGF3 in Figure 4B; (B) and (C) SEM images of other AGF3 sample; (D) Magnified SEM images of CGF3 in Figure 4C.