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Supporting Information

Modifying the Electrochemical Performance of Vertically-oriented Few-layered Graphene through Rotary Plasma Processing

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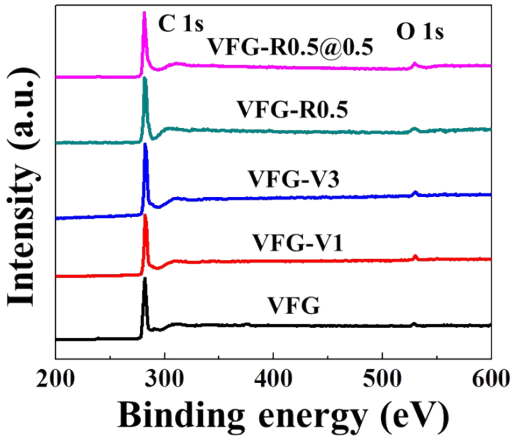

Figure S1. XPS spectra for VFG, VFG-V1, VFG-V3, VFG-R0.5 and VFG-R0.5@0.5

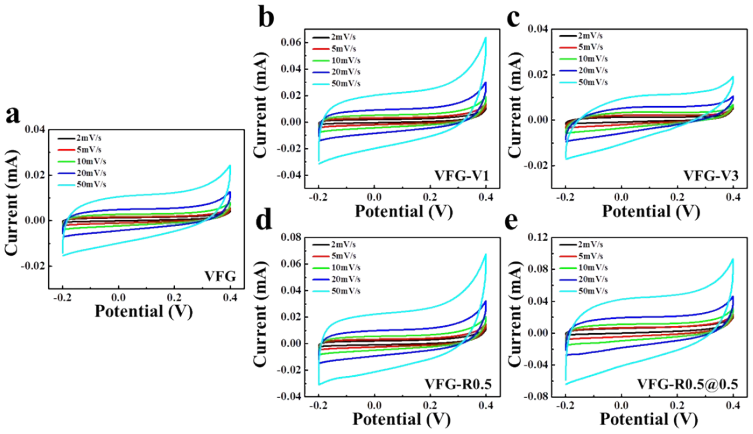

Figure S2. CV curves for VFG, VFG-V1, VFG-V3, VFG-R0.5 and VFG-R0.5@0.5.

Table S1 The contact angle and the capacitance of VFG with different etching angle

	Etching angle (etching time for one side is 30s)				
	5º	10º	15º	20º	30º
Contact angle (º)	79	53	34	59	83
Capacitance ($\mu F/cm^2$)	532	856	1367	703	507

Table S2 The contact angle and the capacitance of VFG with different etching time

	Etching time (etching angle is 15º)			
	15s	30s	1min	3min
Contact angle (º)	53	34	29	25
Capacitance ($\mu F/cm^2$)	633	1367	430	223

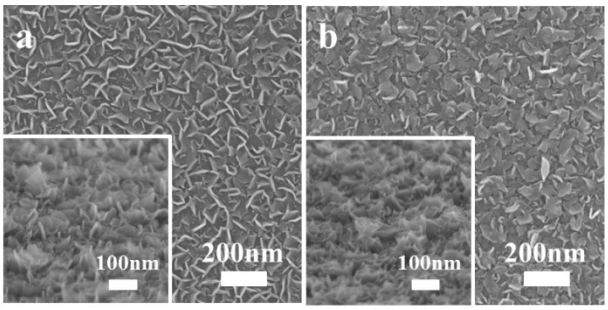

Figure S3 SEM images of VFG after 1min and 3min rotary etching as the etching angle is 15º

We conducted several experiments to optimize the etching angle and etching time. It was noted that all samples was etched to one side and to the other. First, we conducted experiment to optimize the etching angle, and the etching time for one side was fixed at 30s. We investigated the correlation between etching angle, wettability and capacitance, as shown in Table 1. As a result, it can be found that the optimized etching angle is 15º. Further, we conducted experiment to optimize the etching time, and the etching angle was fixed on 15º. We investigated the correlation between etching time, wettability and capacitance. As shown in Table 2, it can be found that VFG after a short time etching, the capacitance could be gradually increased. And the

maximum capacitance can be reached 1367 μF/cm^2 with 30s rotary plasma etching. With prolonging eching time, the capacitance is gradually reduced although the contact angle still decreases. As shown in Figure S3, it coud be found that the majority of edges of VFG after long etching time (1min and 3min) were strongly etched by the Ar plasma. Therefore, vertical etching processes cannot be used for an extended time to modify the surface of VFG without compromising its structural integrity. It can be concluded that the optimal etching angle is 15º and etching time is 30s for both side in the current condition.

Table S3. The volumetric specific capacitance of various carbonaceous nanomaterials.

electrode materials	capacitance	reference
Vertical Graphene by MPCVD	**23.3 F cm^{-3}**	**1**
vertically oriented graphene nanosheets	**3 F cm^{-3}**	**2**
Vertically-oriented graphenes (VGs)	**10.3 F cm^{-3}**	**3**
Vertically oriented graphene sheets (VOGN)	**1.33 F cm^{-3}**	**4**
Ni nanoparticles/CNTs	**1.26 F cm^{-3}**	**5**
vertically oriented Graphene Forest	**3.83 F cm^{-3}**	**6**
vertical graphene nanosheets (VGNs)	**1.67 F cm^{-3}**	**7**
vertically-oriented graphenes	**0.39 F cm^{-3}**	**8**
vertically-oriented graphene (VG)	**53 F cm^{-3}**	**9**
vertically aligned graphene (VG)	**20.9 F cm^{-3}**	**10**
reduced graphene oxide film	**3.06 F cm^{-3}**	**11**
VFG after rotary plasma etching	**137 F cm^{-3}**	**This work**

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