### **Supporting Information**

# Direct hydrothermal reduction of graphene oxide based papers obtained from tape casting for supercapacitor applications

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#### **O1s XPS analyses**



Fig. S1 O1s XPS spectra of (a) P-K, (b) P-KC, (c) P-P and (d) P-PC.

Table S1 Relative contents	of the chemical	bonds derived from	om the O1s XPS spectra
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B. E. (eV)	530.5	531.2	531.9	532.7	533.5
Assignment	Quinone	O-C=O	С=О	C-0	О-Н
P-K	4.17%	28.89%	31.66%	28.74%	6.55%
P-KC	17.74%	16.85%	27.89%	30.31%	7.22%
P-P	7.47%	19.82%	28.54%	36.06%	8.12%
P-PC	9.42%	21.74%	26.32%	33.00%	9.52%

## Reduced graphene oxide (rGO) paper prepared by direct hydrothermal reduction of graphene oxide (GO) paper in 5M NaOH solution

In order to verify the proposed production mechanism of graphene-based paper (GBP) obtained hydrothermally in alkaline solutions, i.e. the electrostatic attraction induced by the intercalated metal cations, an rGO paper was also produced by direct hydrothermal reduction of GO paper in 5M NaOH solution (designated as P-Na) according to the same procedures as in KOH solutions. And a symmetric supercapacitor (SCP-Na) was assembled with the as-prepared P-Na as the two electrodes and 30 wt.% KOH aqueous solution the electrolyte. The following experimental results show that the morphological, structural and compositional characteristics and the consequential electrochemical properties of P-Na are all similar to that of the rGO paper obtained in KOH solution, supporting the proposed electrostatic attraction mechanism.



Fig. S2 Digital photos of a spreading and bended (inset) P-Na.



Fig. S3 SEM images of the (a) cross-section and (b) surface of P-Na.



Fig. S4 SEM image and the corresponding EDX elemental mappings of Na, C and O of P-Na.



Fig. S5 Electrochemical performances of SCP-Na. (a) Cyclic voltammograms with transformed specific capacitance values at different scan rates; (b) Relationship plot between specific capacitance and scan rate. Inset: CV curve at 2 mV s<sup>-1</sup>; (c, d) Galvanostatic charge/discharge curves at different current densities; (e) Relationship plot between specific capacitance and current density; (f) Nyquist plot. Inset: frequency response of capacitance values derived from the imaginary parts of impedance values.

## Reduced graphene oxide (rGO) paper prepared by direct hydrothermal reduction of graphene oxide (GO) paper in 10M KOH solution

An rGO paper was produced in a hydrothermal bath of 10M KOH solution (denoted as P-K-10M), and the SEM image and EDX elemental mappings of the as-obtained paper were measured, as shown in Fig. S6 and S7, respectively. The comparison of the EDX K mappings in Fig. S7 and Fig. 5b demonstrates that the K content in the paper ascends with the increasing KOH concentration of the hydrothermal bath.



Fig. S6 SEM image of the surface of P-K-10M.



Fig. S7 SEM image and the corresponding EDX elemental mappings of K, C and O of P-K-10M.