Supporting information file

Activated graphene as a material for supercapacitor electrodes: effects of surface area, pore size distribution and hydrophilicity.

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1. Characterization of REF material: XPS , XRD and analysis of nitrogen sorption isotherms.

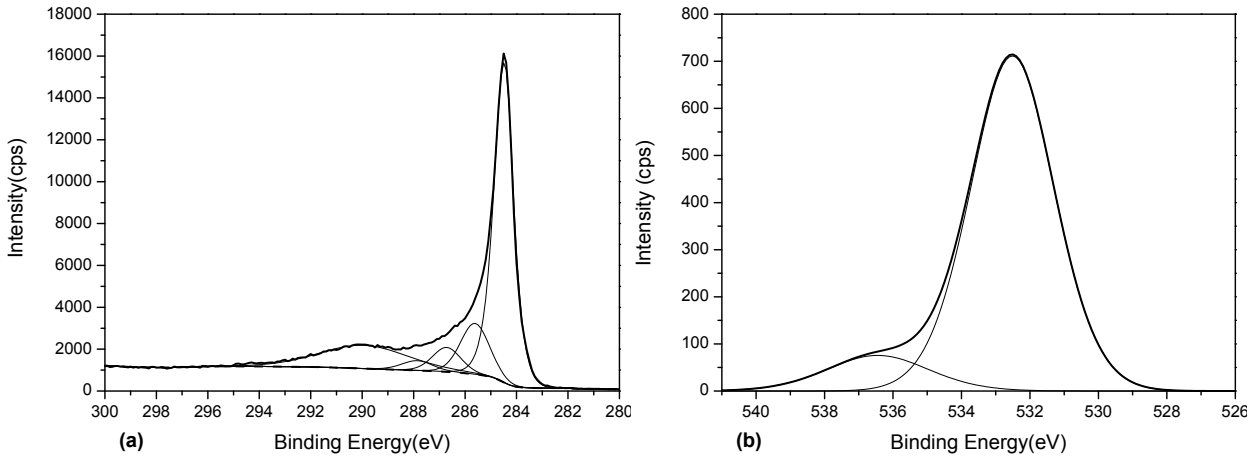


Figure S1 Part of XPS spectrum recorded from REF-sample (a) C1s and (b) O1s. C/O ratio 31.6%

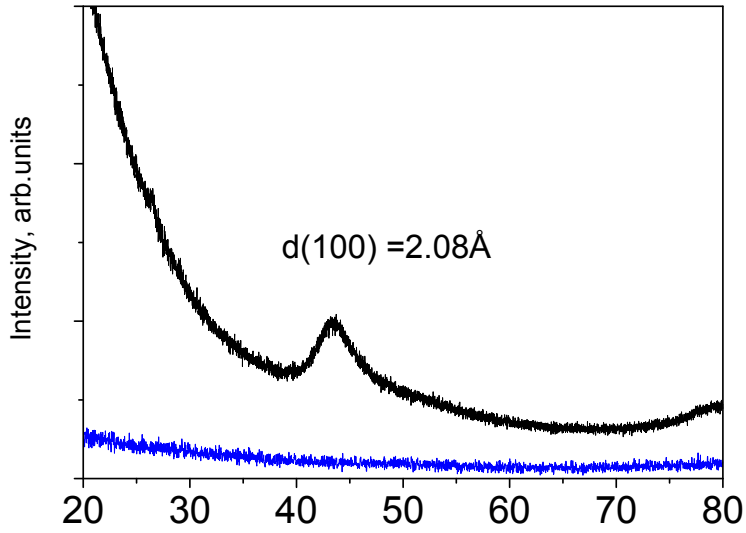


Figure S2. XRD pattern of a-rGO (REF sample) and sample free background. Strong diffuse scattering at low angles is due to disordered structure of porous sample, while presence of in plane (001) reflection confirms that the sample is composed by defect graphene sheets assembled in disordered 3D network.

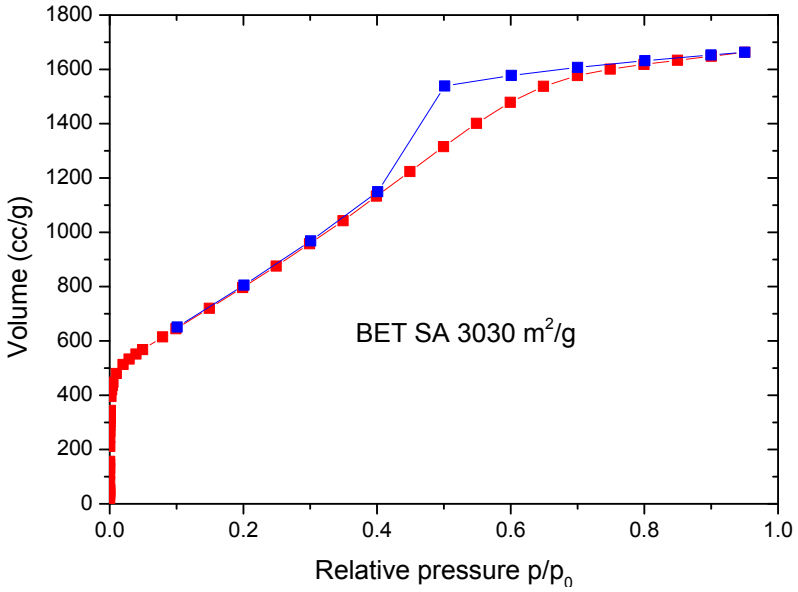


Figure S3 Nitrogen adsorption/desorption isotherm for REF-sample

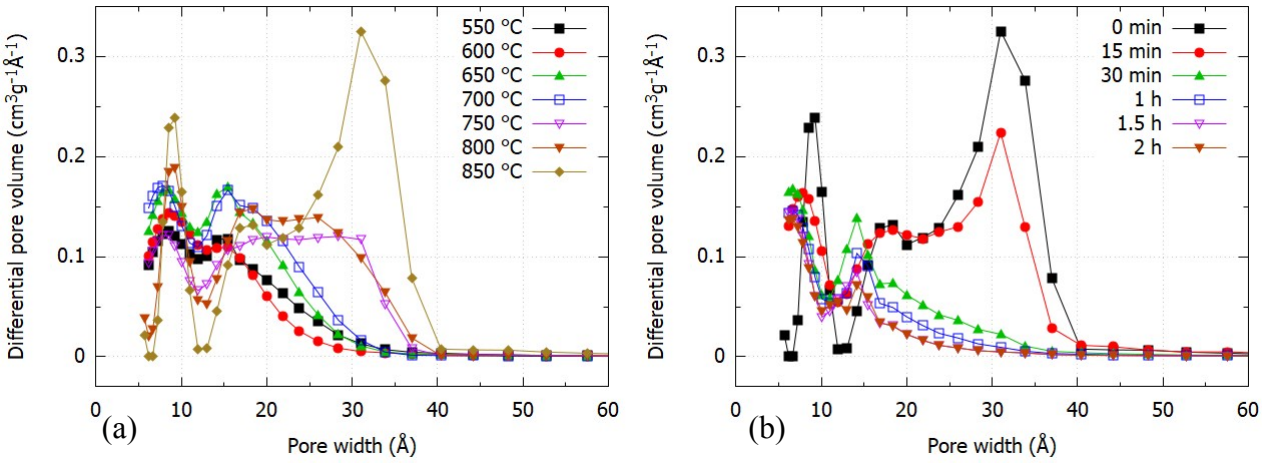


Figure S4 Pore size distribution calculated using QSDFT equilibrium model for N_2 sorption isotherms recorded from set of materials (a) obtained at different activation temperature and (b) after ball milling

2. Additional information on electrochemical performance of electrodes prepared using REF sample in supercapacitor cells.

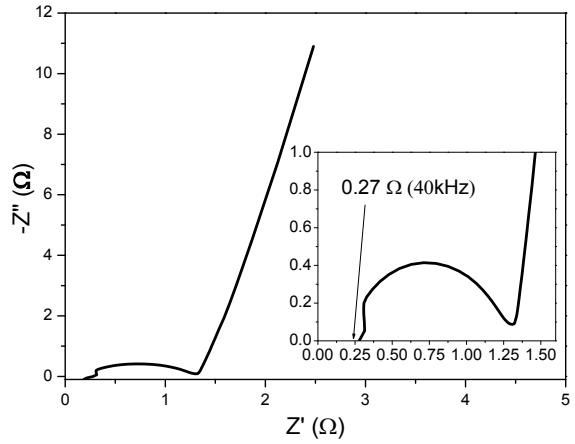


Figure S6 a). Nyquist plot. Electrochemical testing of a-rGO (REF sample) in 6M KOH electrolyte, standard procedure (see experimental part of main text) for preparation of electrodes.

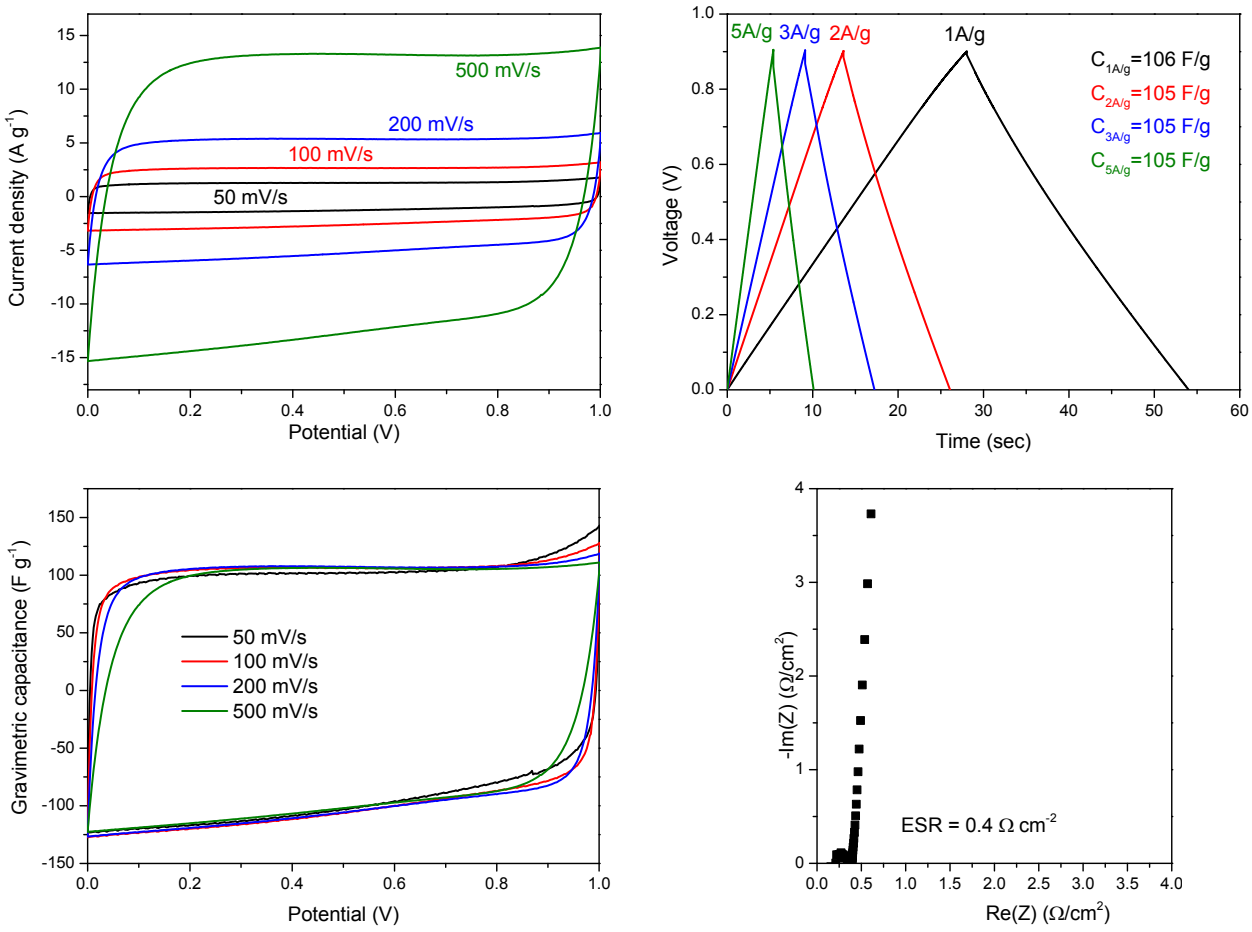


Figure S7 Electrochemical testing of a-rGO (MA sample) in 6M KOH electrolyte. a). CV curves recorded using scan rates 50 mV/s, 100 mV/s, 200 mV/s, 500 mV/s. b) galvanostatic charge-discharge curves (CDCs) recorded with current density 1 A/g, 2 A/g, 3 A/g, 5 A/g., c) gravimetric capacitance calculated from CV curves. d) Nyquist plot.

The electrode was prepared using slightly modified procedure using more hydrophilic separator (Whatman glass separator, thickness of electrode ~100µm, diameter 12mm, mass of one electrode ~3.3mg ~3.0mg, material/binder ratio – 8/1, pressure ~ 7 tons).

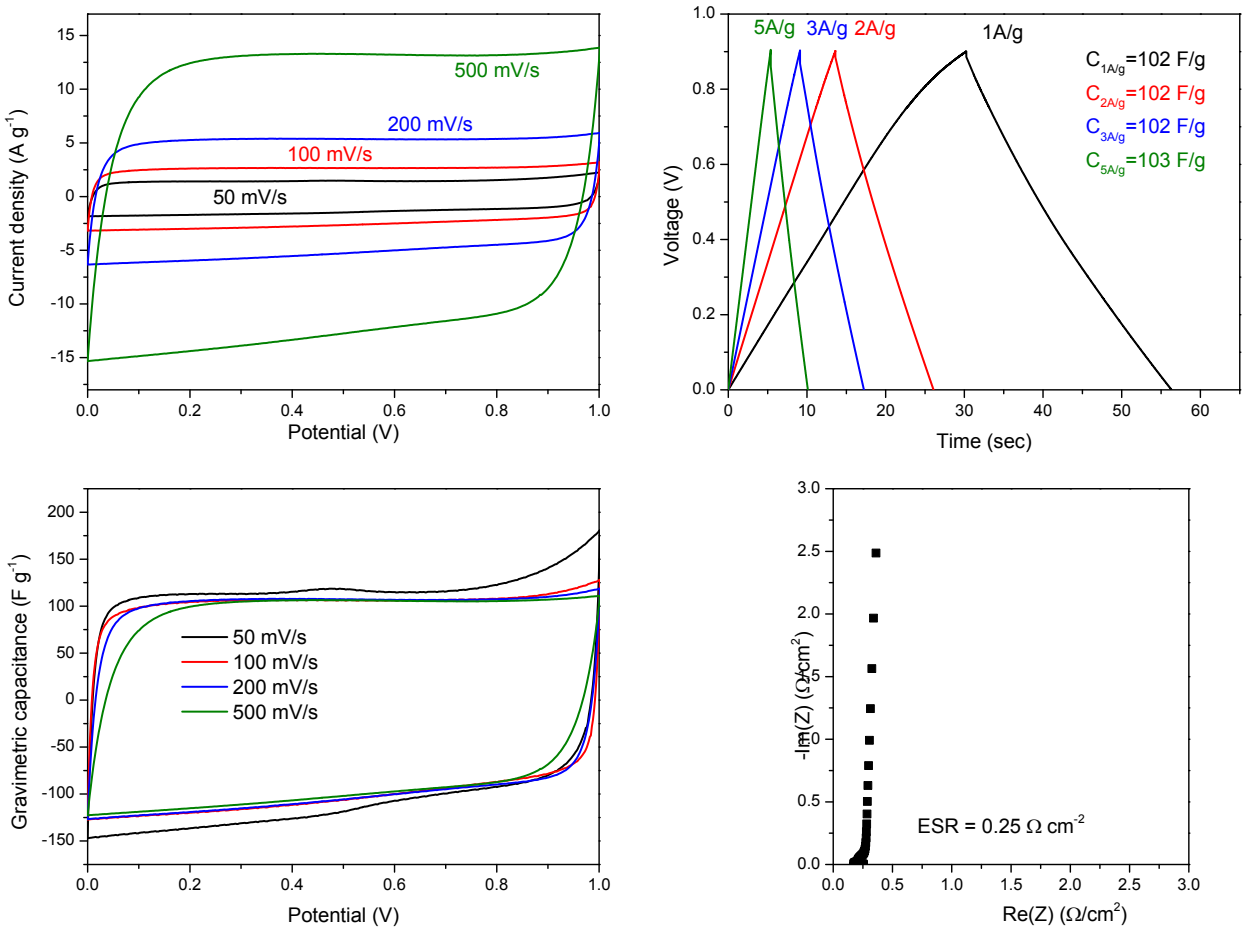


Figure S8 Electrochemical testing of a-rGO (REF sample) in 6M KOH electrolyte. a). CV curves recorded using scan rates 50 mV/s, 100 mV/s, 200 mV/s, 500 mV/s. b) galvanostatic charge-discharge curves (CDCs) recorded with current density 1 A/g, 2 A/g, 3 A/g, 5 A/g., c) gravimetric capacitance calculated from CV curves. d) Nyquist plot.

Slightly modified procedure was used for preparation of this electrode sample to reduce electrical resistance by addition of carbon black: whatman glass separator, thickness ~300µm, diameter 12mm, mass of one electrode ~5.0mg, 81% REF sample, 14% PTEF, 5% Carbon black, used pressure 2 tons.

Summarizing the data shown in Figures S6-S8, the resistance of electrode can be significantly reduced using Whatman glass separator and especially strongly by addition of carbon black.

However, the values of gravimetric capacitance (102-105F/g) are not affected by these variations of preparation procedure.

3. Electrochemical characterization of samples prepared by ball milling for different periods of time.

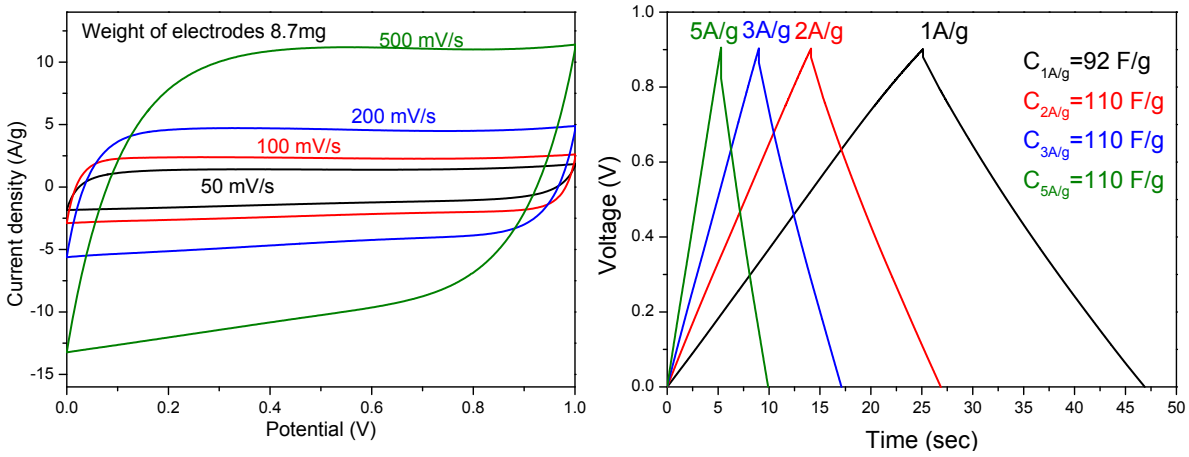


Figure S9 Electrochemical characterization of the REF sample after 15 min ball milling (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

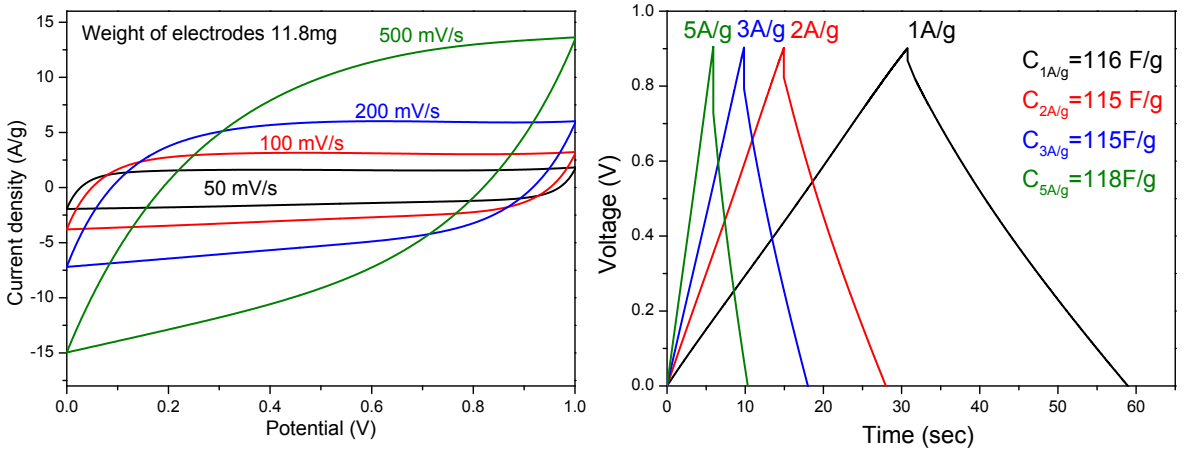


Figure S10 Electrochemical characterization of the REF sample after 30 min ball milling (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

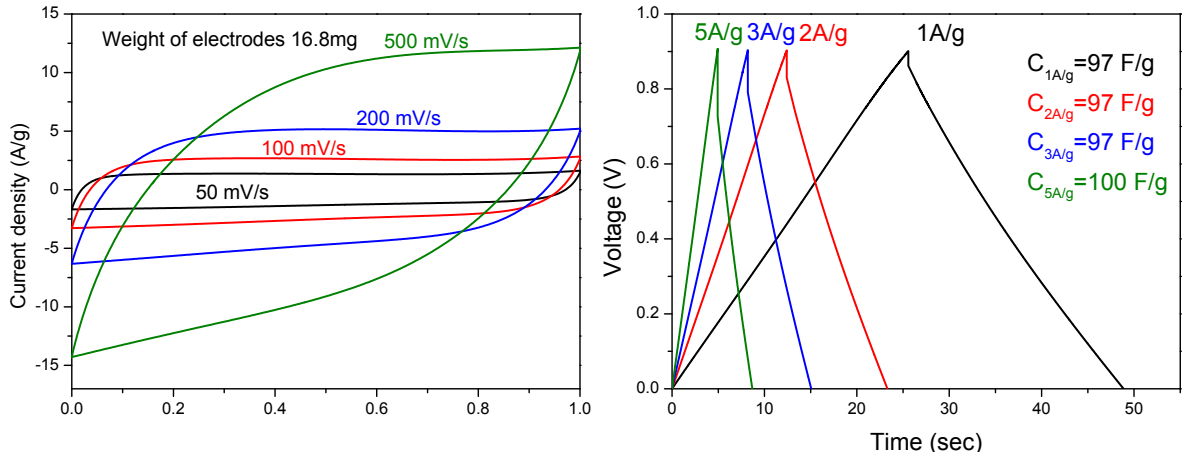


Figure S11 Electrochemical characterization of the REF sample after 60 min ball milling (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density. (c) Nyquist plot.

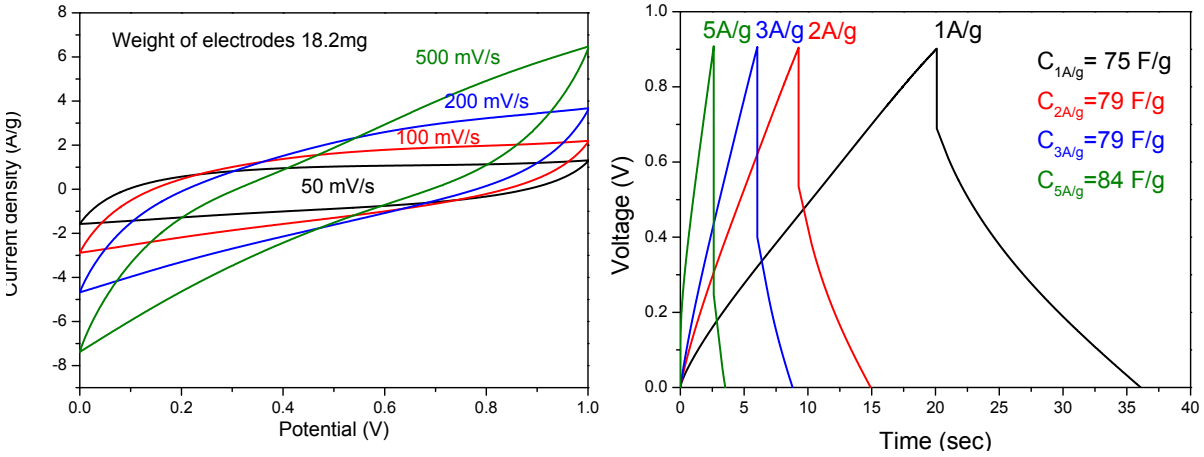


Figure S12 Electrochemical analysis of the MA sample after 90 min ball milling (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

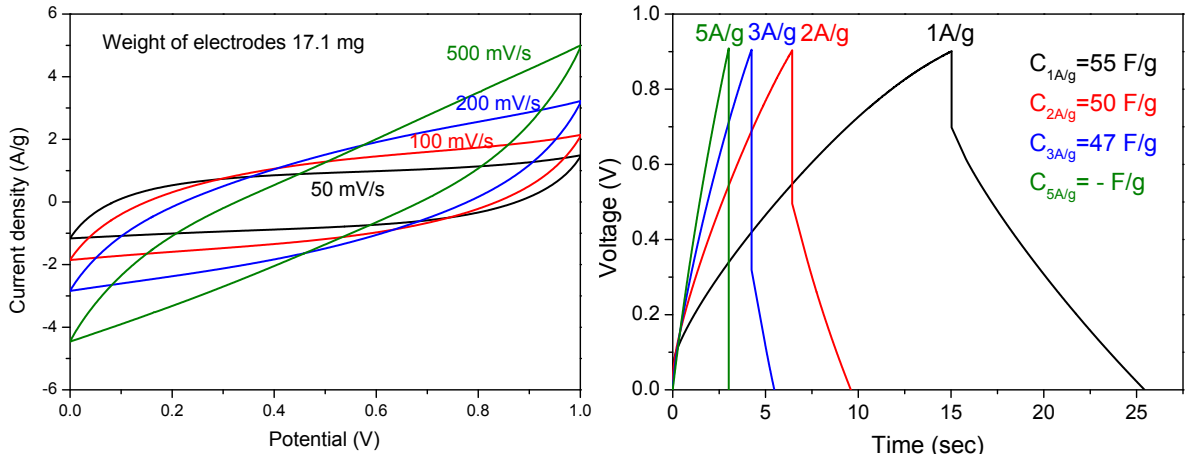


Figure S13 Electrochemical characterization of the REF sample after 120 min ball milling (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

3. Additional information on electrochemical performance of electrodes prepared using different activation temperatures.

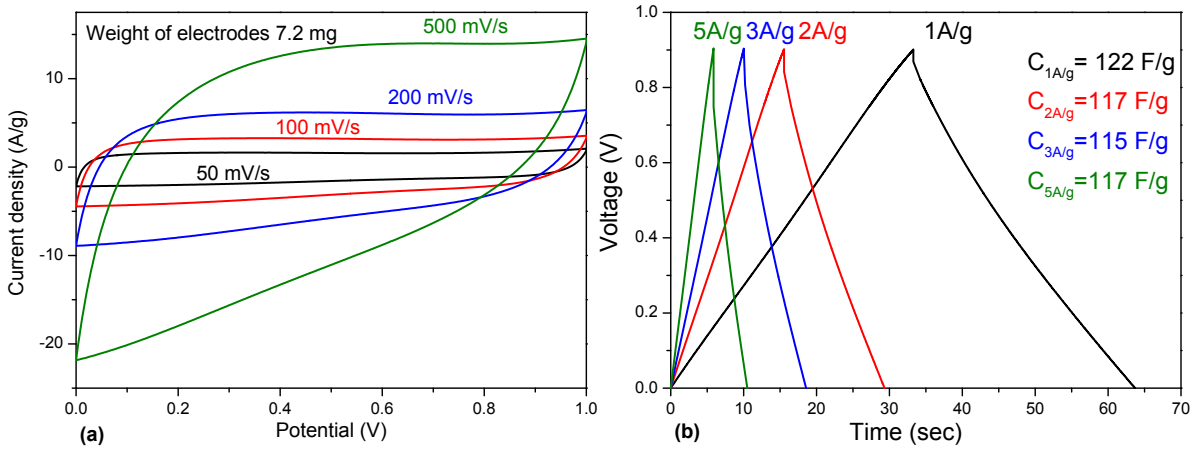


Figure S14 Electrochemical characterization of the material obtained at 800°C (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

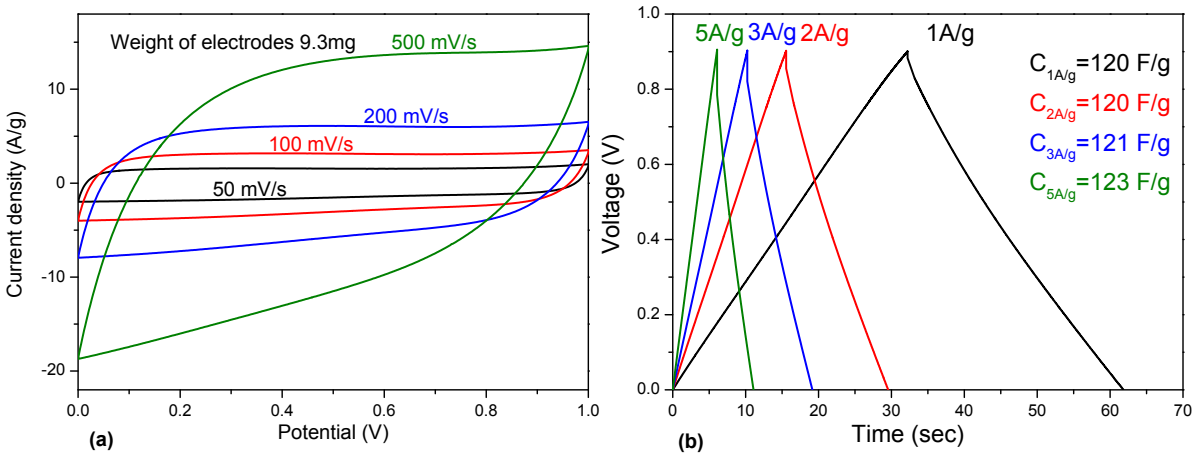


Figure S15 Electrochemical characterization of the material obtained at 750°C (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

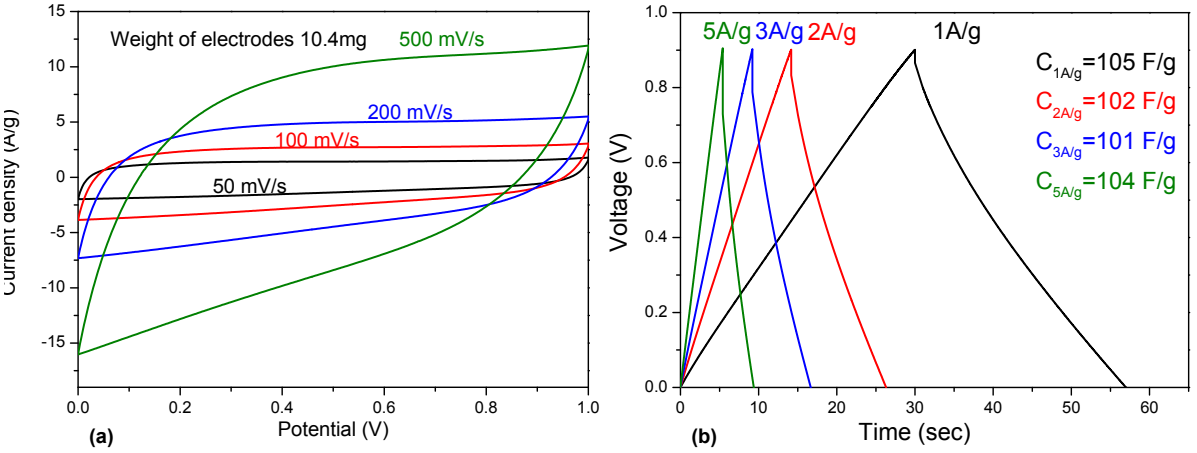


Figure S16 Electrochemical characterization of the material obtained at 700°C (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

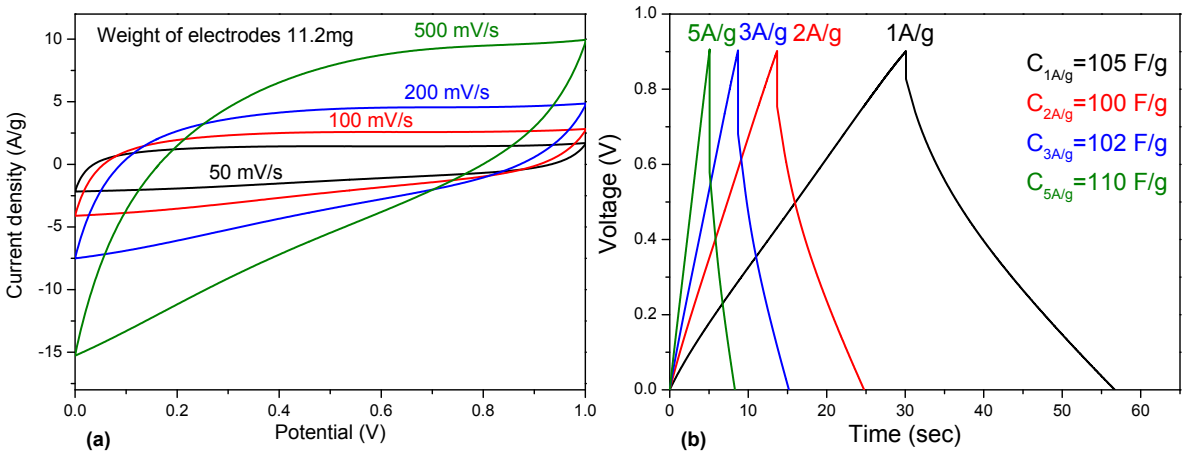


Figure S17 Electrochemical characterization of the material obtained at 650°C (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

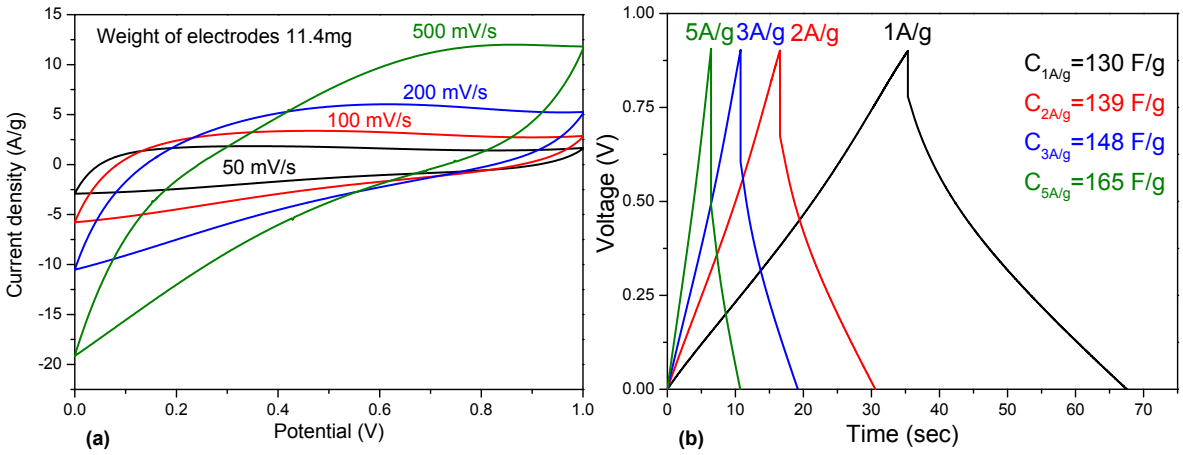


Figure S18 Electrochemical characterization of the material obtained at 600°C (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

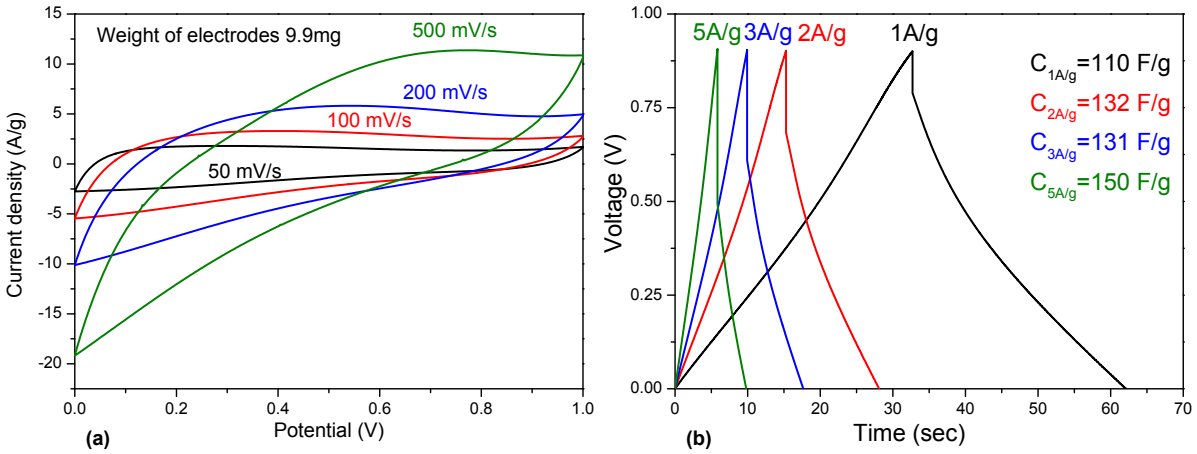


Figure S19 REPEAT Electrochemical characterization of the material obtained at 600°C (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

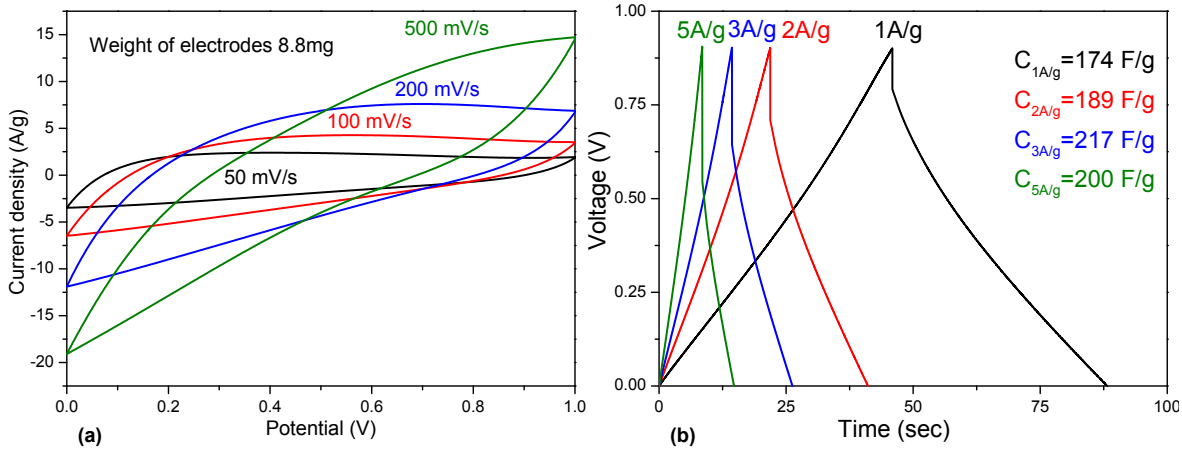


Figure S20 Electrochemical characterization of the material obtained at 550°C (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

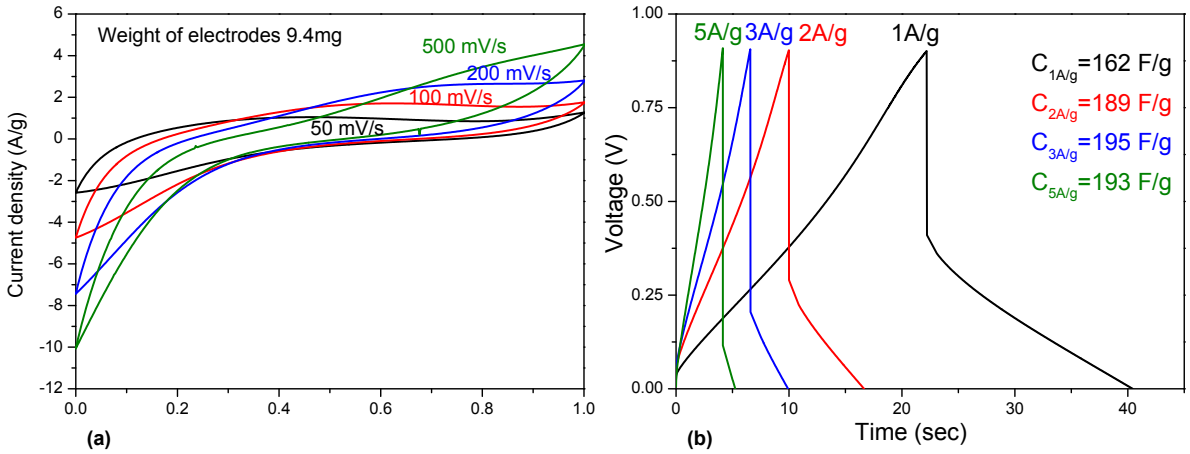


Figure S21REPEAT Electrochemical characterization of the material obtained at 550°C (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

4. Additional data collected using water sorption (DVS method).

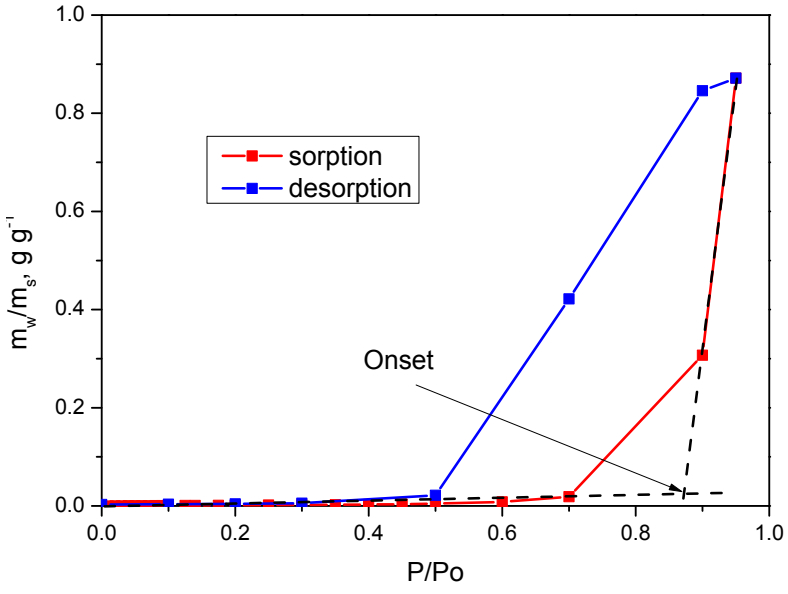


Figure S22 DVS H_2O adsorption/desorption isotherm recorded from REF sample.

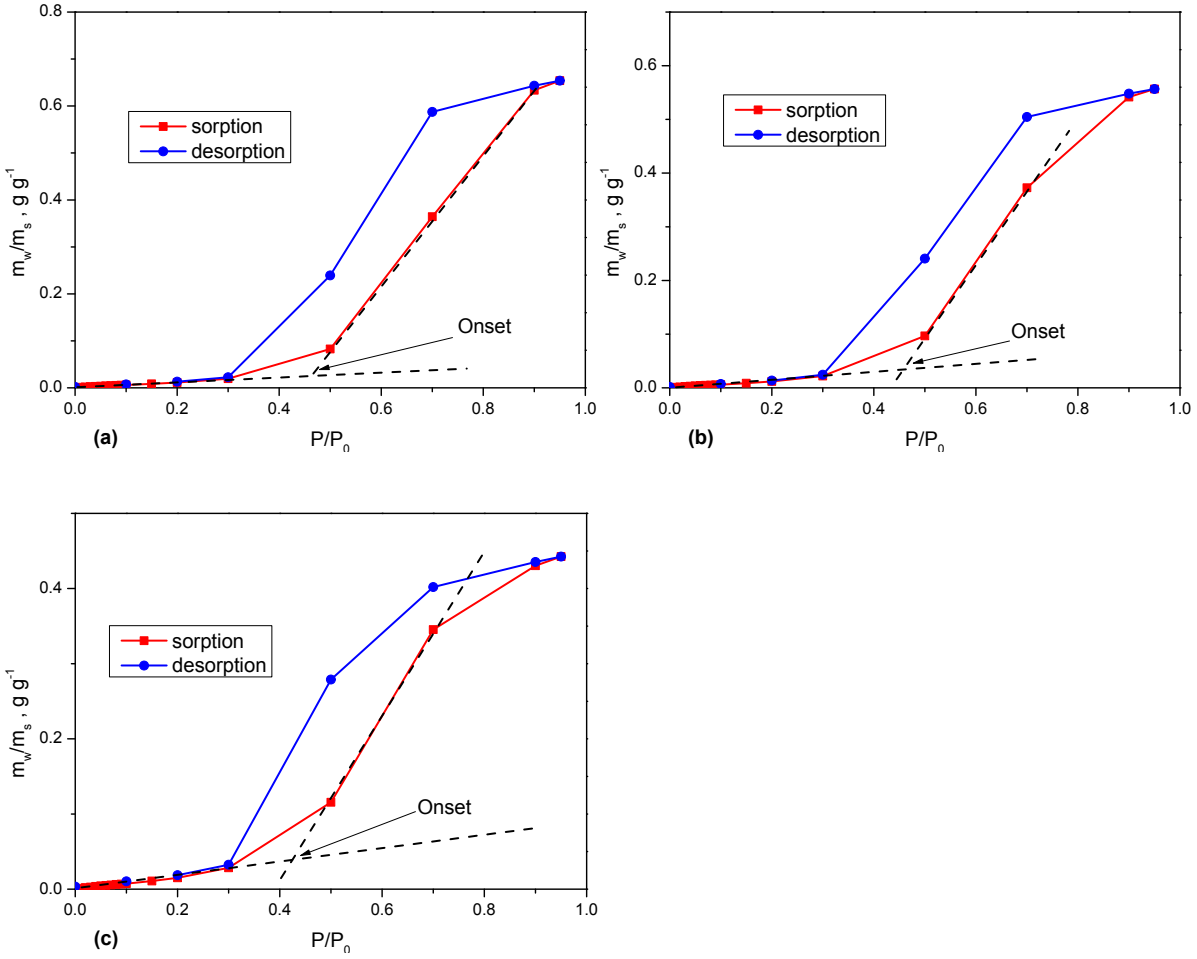
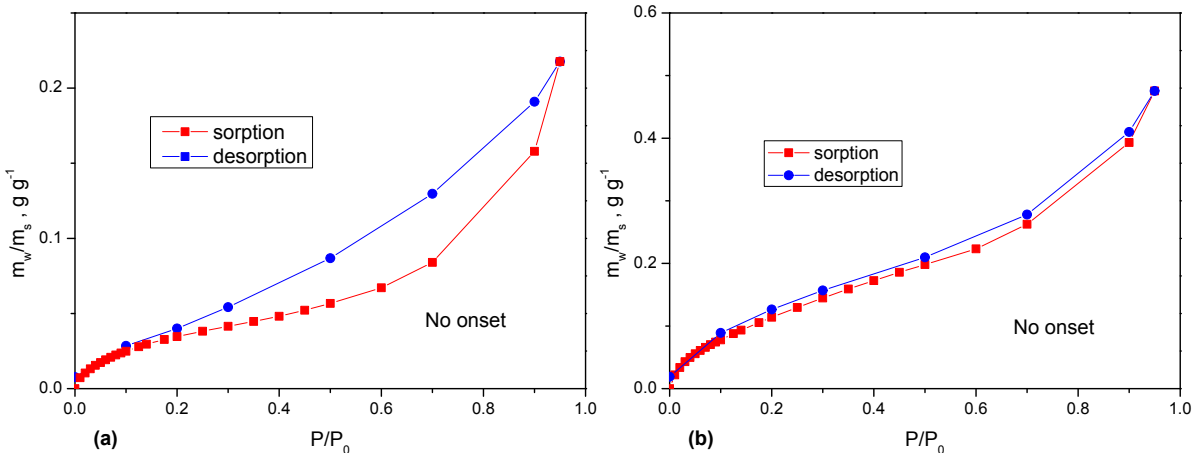


Figure S23 DVS H_2O adsorption/desorption isotherm recorded from REF sample (a) after 30 min milling, (b) after 60 min milling, (c) after 120 min milling



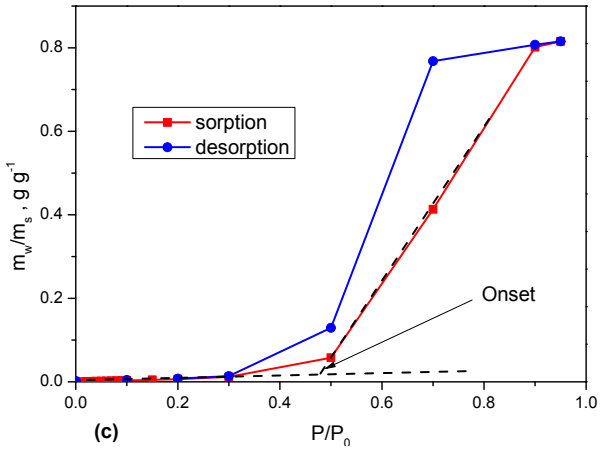


Figure S24 DVS H_2O adsorption/desorption isotherm recorded from (a) rGO, (b) GO used as precursor to prepare all a-rGO samples, (c) reference porous carbon sample

5. XPS spectra recorded from samples activated at different temperatures.

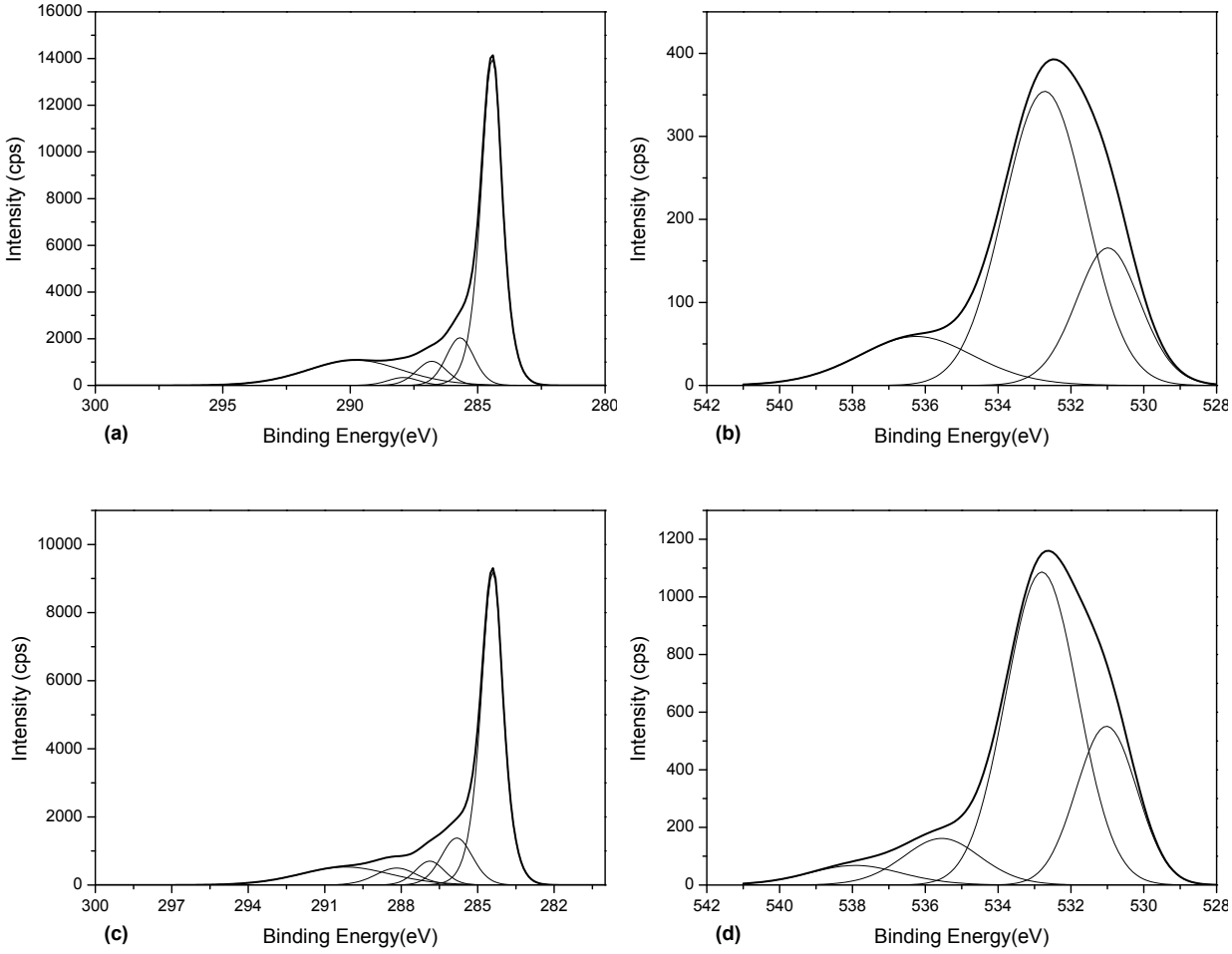


Figure S25 Part of XPS spectra (a) C1s and (b) O1s recorded from the material obtained at 750°C with C/O ratio 48.2 and (c) C1s and (d) O1s of the material obtained at 550°C with C/O ratio 11.6

6. Gravimetric capacitance and micropore volume.

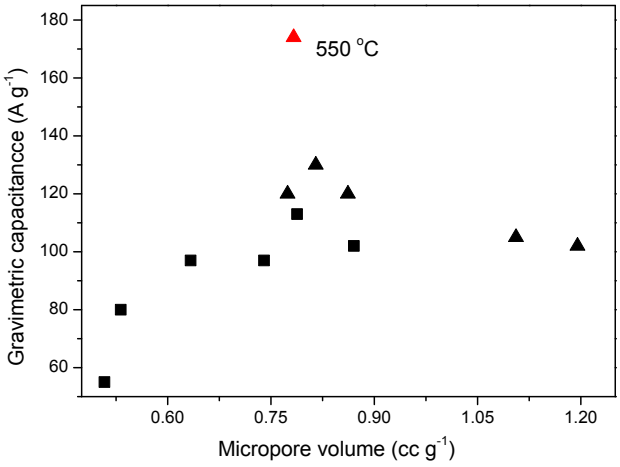


Figure S26 Correlation of gravimetric capacitance and micropore volume for ■ – set of materials after milling and ▲ – set of materials obtained at different activation temperature

7. Analysis of nitrogen sorption isotherms for rGO set of samples and electrodes.

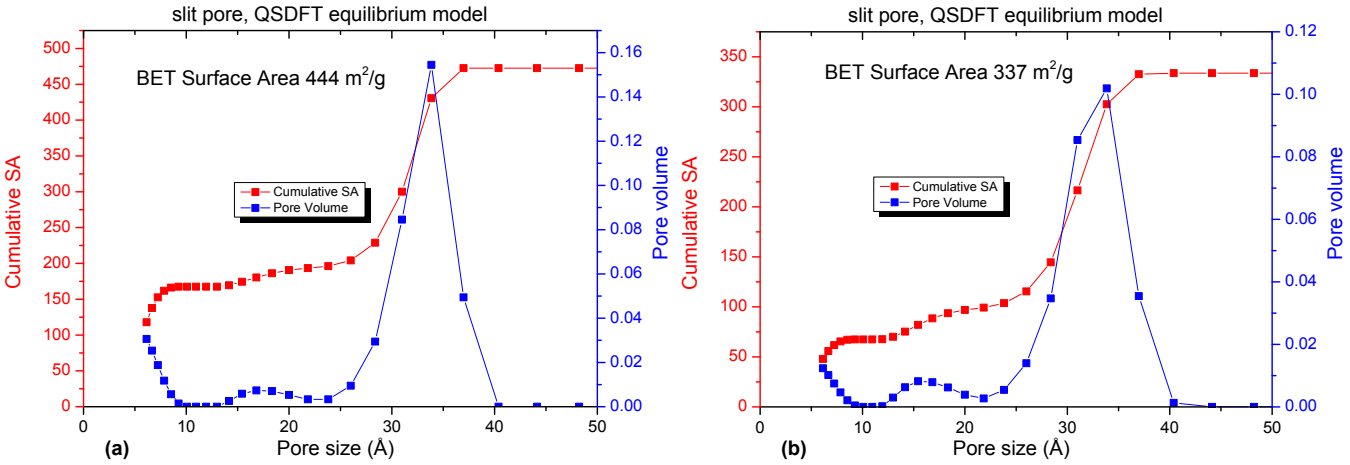


Figure S27 Pore size distribution and cumulative pore volume calculated using QSDFT equilibrium model for N_2 sorption isotherm recorded from (a) rHGO powder and (b) rHGO electrodes.

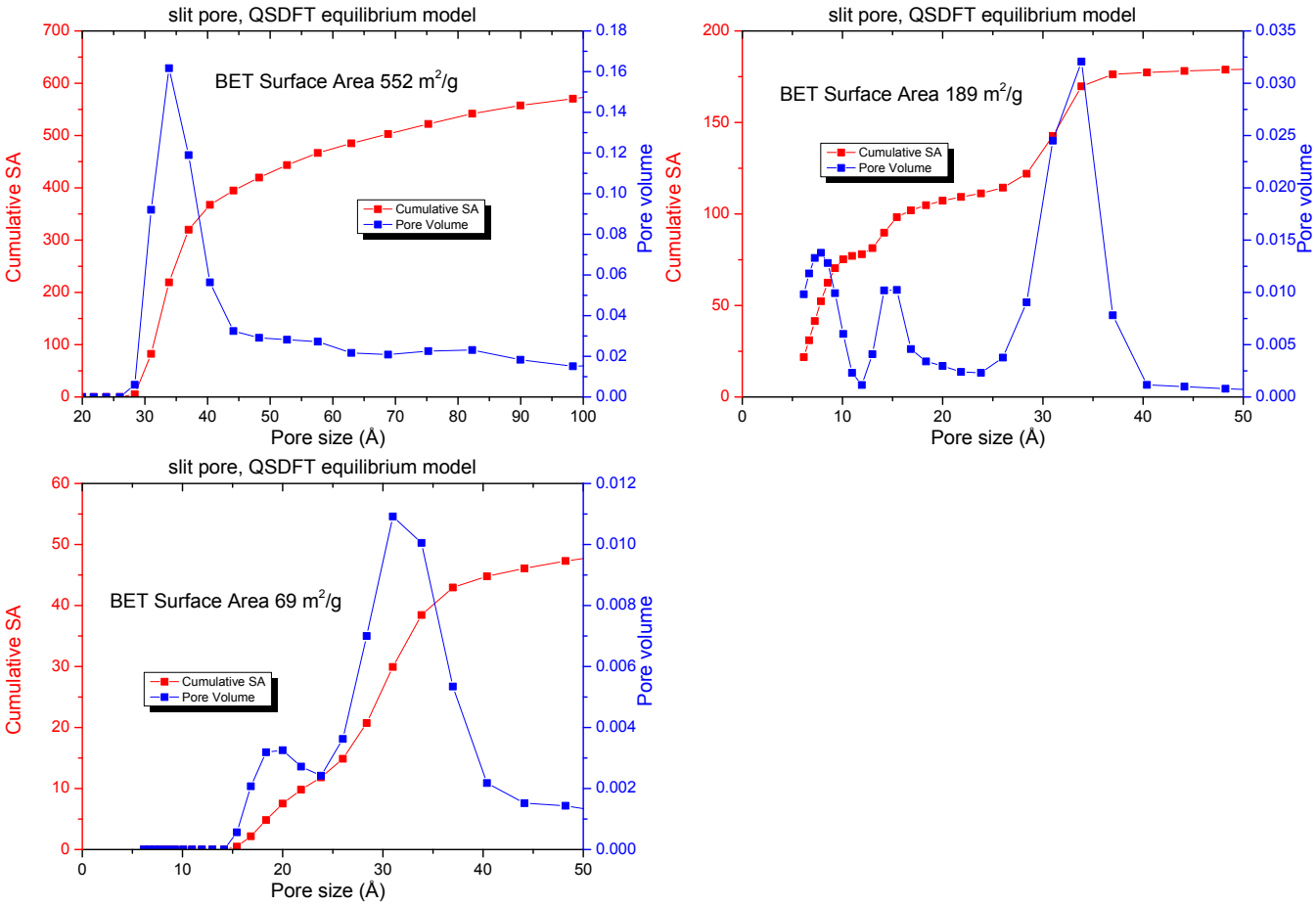


Figure S28 Pore size distribution and cumulative pore volume calculated using QSDFT equilibrium model for N_2 sorption isotherm recorded from (a) rBGO powder and (b) rBGO powder after 5 min milling (c) rBGO after 5 min milling electrodes

8. Electrochemical characterization of the rHGO electrodes.

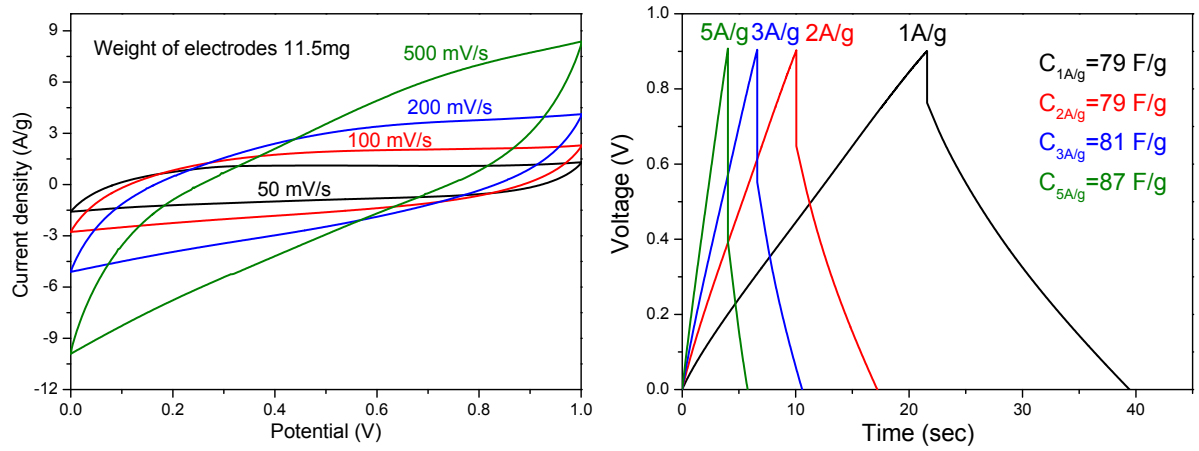


Figure S29 Electrochemical characterization of the rHGO electrodes (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

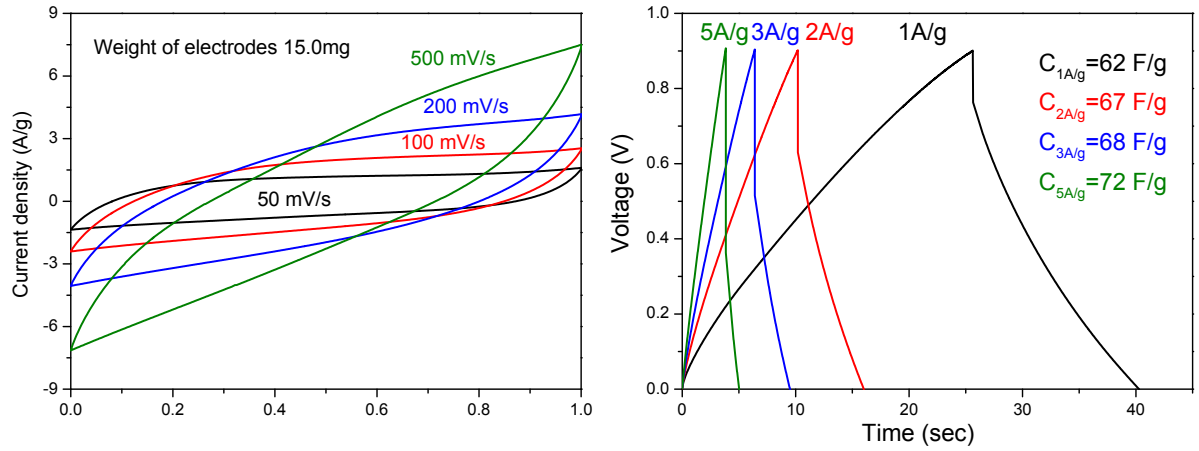


Figure S30REPEAT. Electrochemical characterization of the rHGO electrodes (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

rBGO has an extremely low bulk density after preparation. To be able to prepare electrodes it was necessary to apply ball-milling treatment for 5 min to obtain more dense material.

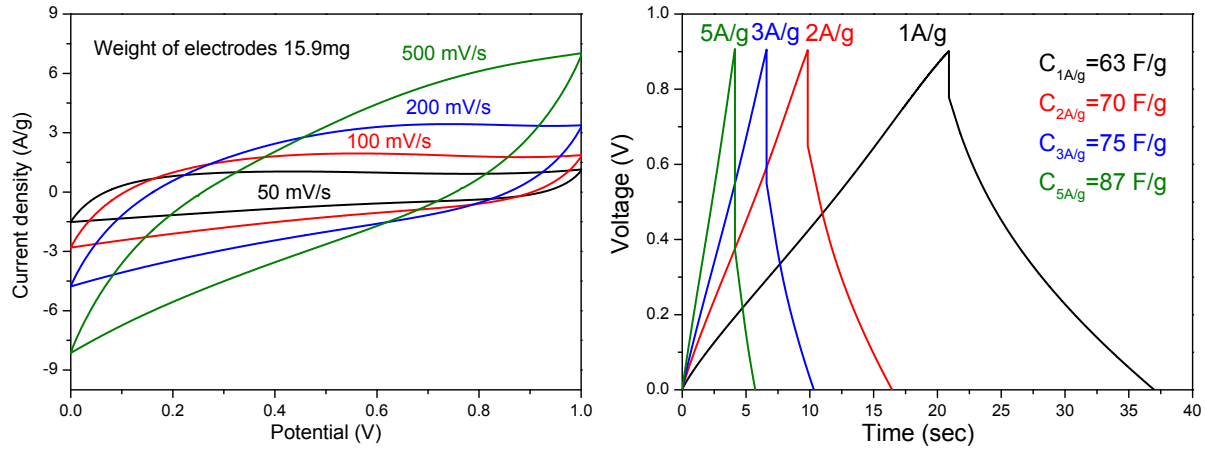


Figure S31 Electrochemical characterization of the rBGO electrodes (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.

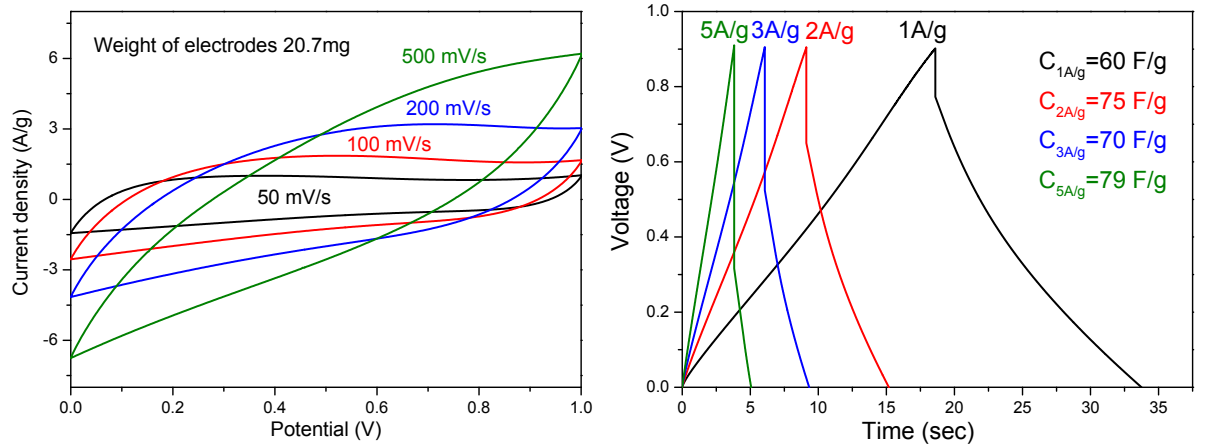


Figure S32 REPEAT Electrochemical characterization of the rBGO electrodes (a) CV analysis at different set of scan rates. (b) Charge/discharge curves recorded at different current density.