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Supporting Information

High Areal and Volumetric Capacity Sustainable All-Polymer Paper-Based Supercapacitors

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Green Energy Storage Device

Figure S1. Cross-sectional schematic illustration of an energy storage device and a photograph of a device.

The energy storage devices are based on self-standing, binder-free, cellulose-PPy paper electrodes, two pieces of graphite foil as current collectors and an acetate cellulose-based separator. As the device uses an electrolyte composed of salt water (i.e. 2 M NaCl) it is clear that these types of devices are metal-free and environmentally friendly. The latter decreases the environmental impact of their manufacturing and renders them more viable for integration into green commercial devices.



Figure S2. A representative TEM image of a part of an APP electrode.



Figure S3. Cross-section SEM images of the compressed APP electrode ($\rho = 1.23 \text{ g cm}^{-3}$).



Figure S4. Charge capacities obtained from the cyclic voltammograms in Figure 3 for the pristine and compressed APP electrodes after normalization with respect to the total weight of the electrodes. The different composite densities are depicted.



Figure S5. Initial charge and discharge curves recorded for a symmetric device containing pristine and compressed APP electrodes, respectively, for a current density of 1 (a), 20 (b), 100 (c) and 200 (d) mA cm⁻².



Figure S6. Gravimetric (a) and volumetric (b) capacitances calculated for different charge and discharge current densities for a symmetric device containing two APP electrodes with a mass loading of ~ 36 mg cm⁻² (uncompressed and compressed using 1 and 5 tons per cm², respectively). The capacitance values in (a) were normalized with respect to the weight of both electrodes.



Figure S7. SEM images with different degrees of magnification showing one of the compressed paper electrode ($\rho = 1.02 \text{ g cm}^{-3}$) of the symmetric device after 8500 cycles (see Figure 3f). The inset in (b) shows a photograph of the electrode.

 Table S1. Summary of the performance of conducting polymer-based energy storage devices.

Electrode	Energy density (Wh L ⁻¹)	Remarks	Ref.
PPy-printing paper	1	Normalized to electrode, separator, electrolyte	1
PANI/graphite paper	0.32	Normalized to electrode, separator, electrolyte	2
CCF- PPy@nanocellulose	0.18	Normalized to the whole cell	3
PVP/rGO	0.85	Normalized to the electrode, and separator	4
CC/GPs/PANI	3.4	Unknown normalization	5
PEDOT-CNT	1.4	Normalized to the active materials	6
This work	3.7	Normalized to the whole device	

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