

## FLEXIBLE AND LOW-COST BINDERLESS CAPACITORS BASED ON P- AND N-CONTAINING FIBROUS ACTIVATED CARBONS FROM DENIM CLOTH WASTES

Juana M. Rosas<sup>1\*</sup>, Tomas Cordero-Lanzac, Francisco J. García-Mateos<sup>1</sup>, José Rodríguez-Mirasol<sup>1</sup>,  
Tomás Cordero<sup>1</sup>

<sup>1</sup> *Universidad de Málaga, Andalucía Tech, Departamento de Ingeniería Química, Campus de Teatinos s/n, 29010 Málaga, España*

\*Presenting author's e-mail: jmrosas@uma.es

### Introduction

Activated carbon cloths (ACCs) show interesting mechanical properties for their use in electric double layer capacitors (EDLCs) or supercapacitors, such as flexibility or toughness. Furthermore, they can be directly used as electrodes, avoiding the addition of binder and/or conductivity promoters, needed in case of granular activated carbons<sup>1</sup>. On the other hand, the presence of some heteroatoms (P and N) into carbon frameworks improves the electrochemical performance of carbon materials<sup>2</sup>. Taking advantage of the high N content of denim waste and the possibility of incorporating P groups by chemical activation with H<sub>3</sub>PO<sub>4</sub>, the goal of this work is the preparation of P- and N-containing fibrous ACCs from denim cloth waste by chemical activation with H<sub>3</sub>PO<sub>4</sub> in a single stage. The effect of the preparation conditions on the porous texture, P and N oxidation state and carbon morphology has been analyzed. These low-cost ACCs have been tested as binderless electrodes.

### Materials and Methods

The fibrous activated carbon cloths (ACCs) were prepared by chemical activation of denim cloth waste (DCWs) with H<sub>3</sub>PO<sub>4</sub>. DCWs were activated using impregnation ratios of 0.5, 1 and 2, at 900 °C. The obtained ACCs were washed with distilled water at 60 °C. The carbon materials were characterized by N<sub>2</sub> adsorption-desorption at -196 °C and CO<sub>2</sub> adsorption at 0 °C; X-ray photoelectron spectroscopy (XPS) analysis; and by scanning electron microscopy (SEM).

ACCs were directly used as electrodes, without the presence of any type of binder and /or conductivity promoter. Electrochemical measurements were performed in a three-electrode cell configuration with an acid electrolyte (1 M H<sub>2</sub>SO<sub>4</sub>). A platinum wire was used as counter electrode and all the potential values were reported versus an Ag/AgCl/KCl 3 M reference electrode.

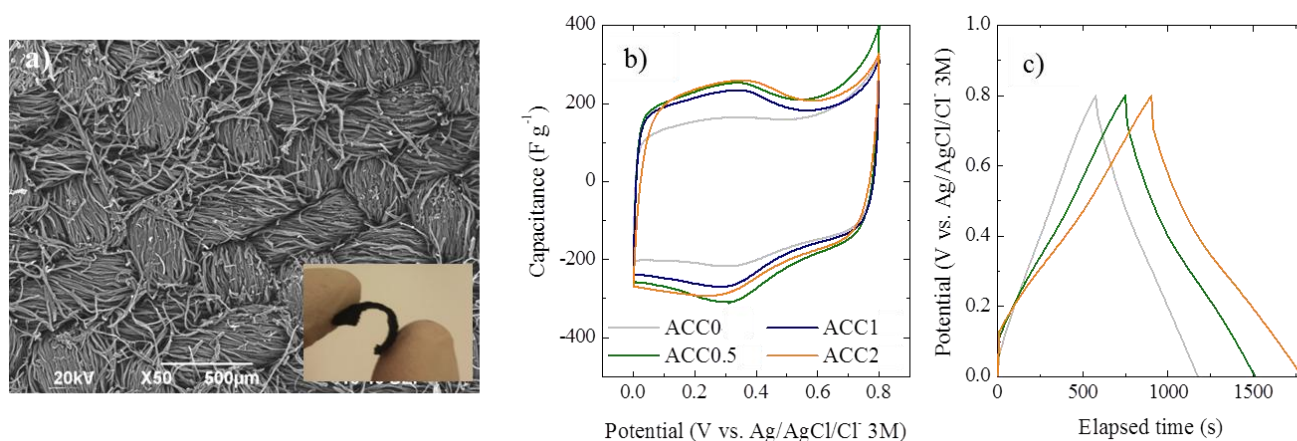
### Results and Discussion

As can be seen in Figure 1.a, the ACCs present a fibrous morphology attributed to the one of the DCWs precursor. The carbon fibers diameter was around 7.4 ±0.7 μm for ACC0.5. These fibers were flexible and easy-to-handle. At low impregnation ratios, ACC0.5 exhibits a type I isotherm attributed to microporous materials, meanwhile a widening of the porous texture is observed at higher impregnation ratios (>1).

The electrochemical characterization was carried out using the ACCs as binderless electrodes by placing a piece of each sample inside a folded stainless steel sheet. The entire ACCs exhibit a rectangular voltammogram shape (Figure 1.b), associated with the development of the electric double layer (EDL). However, chemically activated carbon with H<sub>3</sub>PO<sub>4</sub> also shows a little hump at 0.35 V. This anodic current is attributed to the pseudocapacitive interaction between the electrolyte and the oxygenated surface groups. The electrodes show a relatively low IR drop and the triangle shape suggests their suitable capacitive behaviour (Figure 1.c).

Gravimetric capacitances (C<sub>g</sub>) were calculated from the area of CVs at 1 mV s<sup>-1</sup>. ACC0.5 electrode shows the maximum C<sub>g</sub> value (227 F g<sup>-1</sup>). As expected, there is a clear relationship between the C<sub>g</sub> values and the V<sub>t</sub> values. However, there is a deviation of this tendency for the sample ACC2,

probably associated to the higher P content of this sample.



**Figure 1. a) SEM micrographs of ACC0.5; b) Steady-state cyclic voltammograms and (c) galvanostatic charge/discharge profiles of the ACC electrodes**

The main drawback of binderless electrodes is the inefficient contact between the current collector and the working electrode, which reduces its feasibility of working at high current density. In general, a reduction of  $C_g$  values is exhibited by the electrodes upon increasing the current density, but ACC0.5 electrode presents only a slow decrease of the capacitance and maintains a considerable capacitance at  $5 \text{ A g}^{-1}$ . Specifically, this ACC maintains its flexibility, decreasing the contact resistance between the current collector and the ACC electrode.

## Conclusions

Non expensive activated carbon cloths (ACCs) are obtained from denim clothes waste (DCWs) by chemical activation with phosphoric acid. Low impregnation ratios lead to flexible and microporous activated carbons cloths, whereas more fragile and rigid activated carbon cloths with higher external surface area are produced upon increasing the amount of  $\text{H}_3\text{PO}_4$ . Activation at high temperatures allows for obtaining a more ordered and conductive carbon structure. The activated carbon prepared with an impregnation ratio of 0.5 (w/w) exhibits the best performance as electric double layer capacitor. This electrode shows a specific surface area of  $2016 \text{ m}^2 \text{ g}^{-1}$  and the highest registered gravimetric capacitance ( $227 \text{ F g}^{-1}$ ). Moreover, its flexibility minimizes the ohmic resistance of the electrode, thus increasing the feasibility of working at higher current densities than the other synthesized electrodes.

## Acknowledgment

This work was supported by the Spanish Ministry of Economy and Competitiveness under CTQ2015-68654-R project.

## References

1. Berenguer, R., García-Mateos, F.J., Ruiz-Rosas, R., Cazorla-Amorós, D., Morallón, E., Rodríguez-Mirasol, J., Cordero, T. (2016). Biomass-derived binderless fibrous carbon electrodes for ultrafast energy storage. *Green Chem.* 18, 1506–1515.
2. Hulicova-Jurcakova, D., Seredych, M., Lu, G.Q., Kодиweera, N.K.A.C., Stallworth, P.E., Greenbaum, S., Bandosz, T.J. (2009). Effect of surface phosphorus functionalities of activated carbons containing oxygen and nitrogen on electrochemical capacitance. *Carbon* 47, 1576–1584.