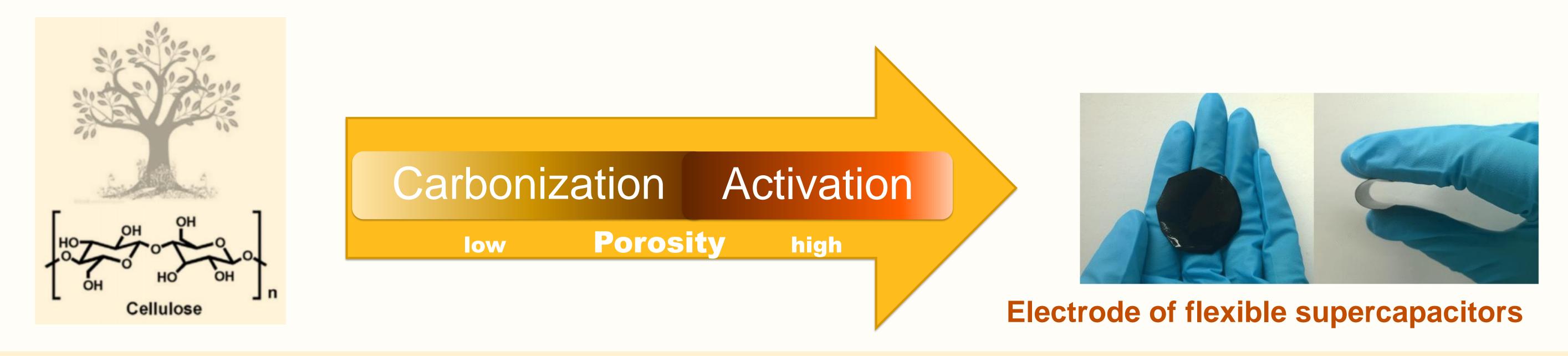
Conversion of cellulose to activated carbons for high-performance supercapacitors

Ekaterina Sermyagina*, Kirill Murashko, Daria Nevstrueva, Arto Pihlajamäki and Esa Vakkilainen LUT University of Technology, Lappeenranta, Finland



Abstract

Activated carbons (**AC**) are fundamental materials for various energy and environmental applications. These materials with high surface area and porosity are widely applied for the **electrical energy storage applications**: lithium-ion batteries and supercapacitors. Adjusting the porosity of the AC to obtain the high capacitance and low impedance is a crucial step for producing the high-performance supercapacitors.

Current work aims to investigate the possibility of substituting the commercial AC with the activated carbon from the cellulose. In two-step procedure, the cellulose is firstly carbonized to produce char (hydrochar) with higher carbon content and a rudimentary porosity. After that, the hydrochar is chemically activated to obtain material with large surface area and developed pores. And these activated carbons are then used to produce flexible self-standing electrodes for an electrical double layer capacitors (EDLCs). Above all, the effect of operational parameters of the carbonization on porous structure evolution during activation and, as a result, on electrochemical characteristics of the produced electrodes was evaluated.

Materials and Methods Activation Carbonization Electrode Hydrochar **Activated carbon** Electrode Hydrothermal **KOH** chemical activation preparation testing **Carbonization (HTC)** - cyclic temperature: 700 °C Phase voltammetry temperature: 250 °C residence time: 1 h inversion curves residence time: 2, 4 h KOH/hydrochar ratio: 4/1 method - electrochemical H₂O/cellulose ratio: 3/1, 6/1 (immersion Cellulose - Burn-off values - Mass yield impedance precipitation) (Domsjö - BET analysis - BET analysis spectroscopy Fabriker AB) - SEM analysis - Scanning Electron

Results and discussion

The increase of the carbonization severity (longer reaction time and higher amount of water in water-cellulose mixture) results in higher decomposition and, consequently, lower mass yields: 44% (HTC-2h-3/1), 38% (HTC-2h-6/1), 40% (HTC-4h-3/1) and 36% (HTC-4h-6/1). The morphology of the hydrochars is significantly affected by the HTC, however the cellulose fibers are still visible. Microspheres resulted from the cellulose decomposition are formed on the surface (Figs. 1A and 1B). Longer residence time leads to the formation of bigger separate spheres of Ø100 μ m each. The BET surface areas for the hydrochar samples are in rather narrow range between 24 - 27 m^2/g .

Microscopy (SEM)

The activation agent acts as a dehydration agent intensifying the decomposition of the feedstock: further mass loss occurs with simultaneous porosity development. The morphology of the ACs differs dramatically from the hydrochars and is characterized with irregular shapes, smooth surfaces and large cavities (Figs. 1C and 1D). Produced activated carbons have high surface areas in the range of $2300 - 2645 \text{ m}^2/\text{g}$.

The electrodes produced from the samples carbonized during 2 h have higher specific capacitance than the samples carbonized for 4 h (Fig. 2a). The variations of the capacity retention with different scan rates are relatively similar for all samples under consideration (Fig. 2b). However, the capacity retention is slightly higher for the samples AC-4h-3/1 and AC-4h-6/1 than other two samples. Also, these samples have higher values of the impedance at low operation frequency. The products of the secondary reactions during HTC may influence on the diffusion of electrolyte ions into the pores of electrode materials that may lead to such results.

Conclusions

- 1. The carbonization severity has a pronounced effect on the morphology of the hydrochars: within the studied HTC parameters, the effect of the residence time is stronger than the effect of the feedstock concentration. Additionally, the surface areas of the derived activated carbons increase with the HTC severity.
- 2. At the same time, electrochemical tests reveal the highest specific capacitances (110.3 F/g (AC-2h-3/1) and 102.5 F/g (AC-2h-6/1) at 50 mV/s scan rate) and the lowest impedance values for the AC carbonized during 2 h. This can be explained by non-complete use of the developed nanopores of the AC during the creation of the electrochemical double layer.

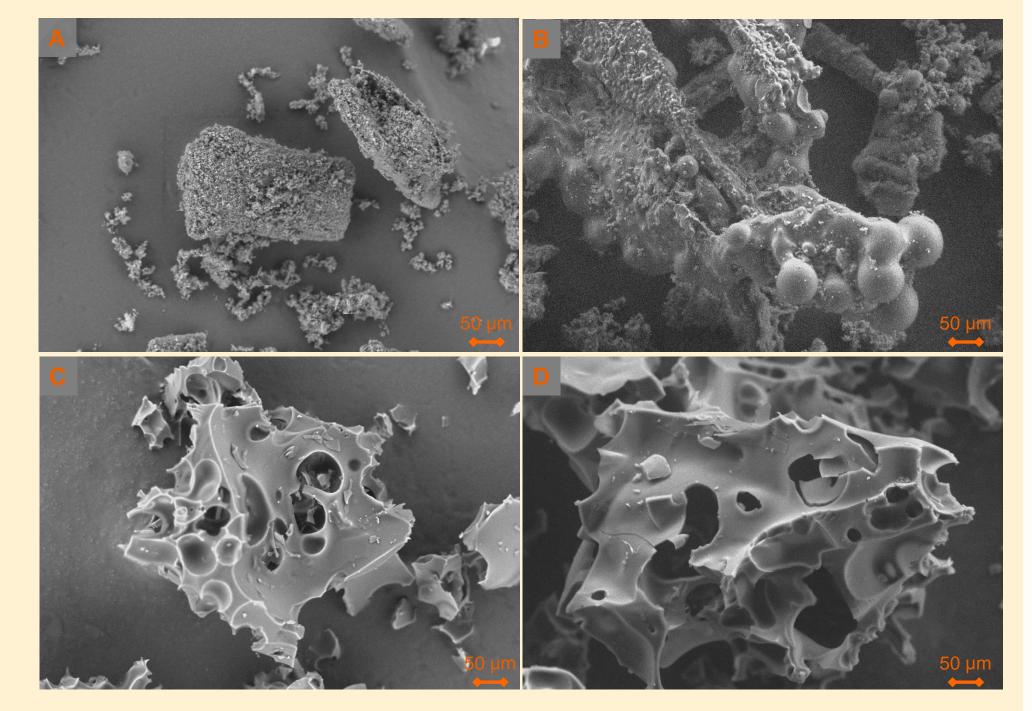


Figure 1. SEM images of the hydrochar samples before activation: **(A)** HTC-2h-3/1; **(B)** HTC-4h-3/1; and after activation: **(C)** AC-2h-3/1; **(D)** AC-4h-3/1.

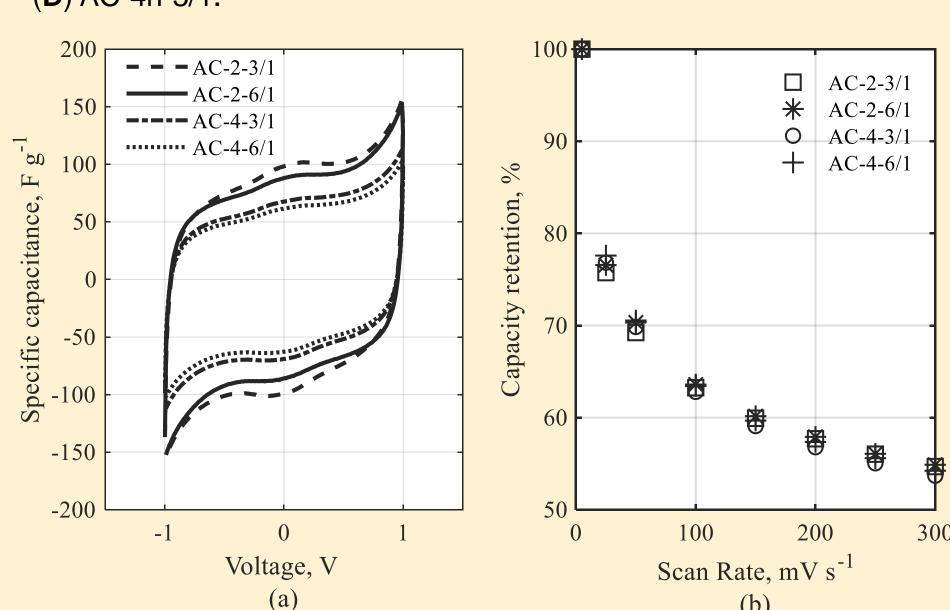


Figure 2. Specific capacitance (**a**) and capacity retention (**b**) of the created EDLCs from prepared composite materials.

CONTACT INFORMATION

Ekaterina Sermyagina, D.Sc., LUT University, Lappeenranta, Finland Ekaterina.Sermyagina@lut.fi, +358469539001

Daria Nevstrueva, M.Sc., LUT University, Lappeenranta, Finland Daria.Nevstrueva@lut.fi, +358451508899

Esa Vakkilainen, Prof., LUT University, Lappeenranta, Finland Esa. Vakkilainen@lut.fi, +358403578684