IMPROVING THE PERFORMANCE OF SUPERCAP ELECTRODES MADE BY SIC

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Introduction

Electrochemical double layer capacitors have found high interest due to their capability as an intermediate between conventional capacitors and batteries. They usually function on the basis of the formation of an electrolytic double layer at the interface of electrodes composed of high specific surface area materials [1]. Our strategy was to produce thin carbonaceous electrodes which are poor in binder content [2] by performing the Substrate Induced Coagulation (SIC) process. The SIC process is an aqueous dip-coating process, applicable for the coating of nano sized materials on various substrates [3]. Unfortunately, the value for specific capacitance of carbon electrodes derived by SIC is not particularly high. Therefore a subsequent activation treatment of the electrodes was performed in order to increase the capacitance.

Experimental

High surface carbons were obtained from Degussa, Germany (XE2) and from Norit, Netherlands (EXP1 and EXP6) with BET surface area in the range of 1000-2000 $m^2.g^{-1}$. The carbons were coated on the current collectors (10 mm $\times$ 10 mm Titanium grid) using the SIC process as described in detail in our previous papers [4]. Therefore the current collector was preconditioned by dipping into a solution of polyelectrolyte in the first coating step. Subsequently, immersion of this preconditioned substrate in a metastable dispersion of carbon black yields coagulation and thus precipitation of carbon on the current collector surface. Repeating this procedure leads to a thin carbon coating on the current collector with a defined thickness [4].

After the coating process of high surface carbons on the current collector was completed, an activation step of the carbon electrodes was performed. Thus the carbon electrode was immersed in nitric acid (30 %, 40 °C) for 1 h [5].

The electrochemical experiments were carried out in a three-electrode glass cell at room temperature using 5.25 M H_2SO_4 as electrolyte. Cyclic voltammogramms (CVs) were recorded at a scan rate of 20 $Mv.s^{-1}$ with a Zahner IM6 measurement system in a potential range of –600 to +200 mV *vs.* Hg/Hg_2SO_4. For carrying out electrochemical impedance spectroscopy (EIS) the same system was used, data were collected in a frequency range of 10 kHz to 5 mHz at a potential of -100 mV *vs.* Hg/Hg_2SO_4.

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Results and Discussion

For the pristine carbon electrodes, CVs with rectangular shape were obtained indicating a capacitive behaviour (Fig. 1a). The specific capacitance was calculated from the CVs using the equation $C = i/v$, where i is the average current and v is the scan rate. The values calculated for the capacitance are shown in Table 1.
After performing the SIC process stable electrodes were obtained with a carbon coating of app. 20 µm thickness. The capacitances were calculated before and after the activation step by CV as well as by EIS. Both methods indicated a significant increase of capacitance due to the successful activation of all different carbon materials as shown in Table 1. After the activation with nitric acid a significant pseudocapacitance occurred which is indicated by the appearance of a broad hump at a potential of app. -100 mV (Figure 1b).

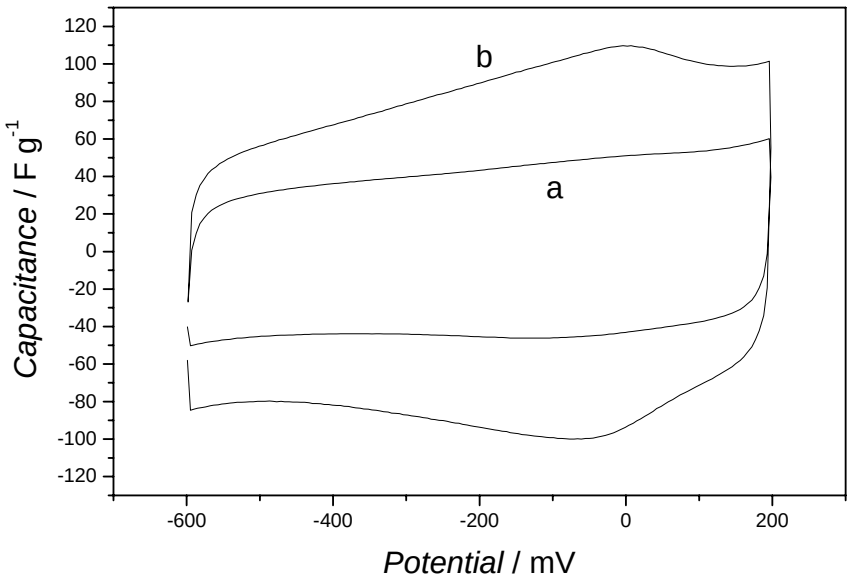

Fig. 1 *CVs (ν = 20 mV s^{-1}) expressed as capacitance vs. potential of a porous carbon electrode (EXP1) made using the SIC process. (a) Before and (b) after the activation with HNO_3.*

Table 1 *Capacitance C of carbons calculated from EIS at a frequency of 5 mHz. (a) before and (b) after the activation with HNO_3*

carbon	BET / m^2 g^{-1}	C_a / F g^{-1}	C_b / F g^{-1}
XE2	965	25	47
EXP1	1550	46	88
EXP6	1710	63	107

The frequency behaviour of all electrodes is excellent indicating that the accessibility of the ions into the pores of the carbon is good. This fact can be concluded by the high values of capacitance at relatively high frequencies (Figure 2).

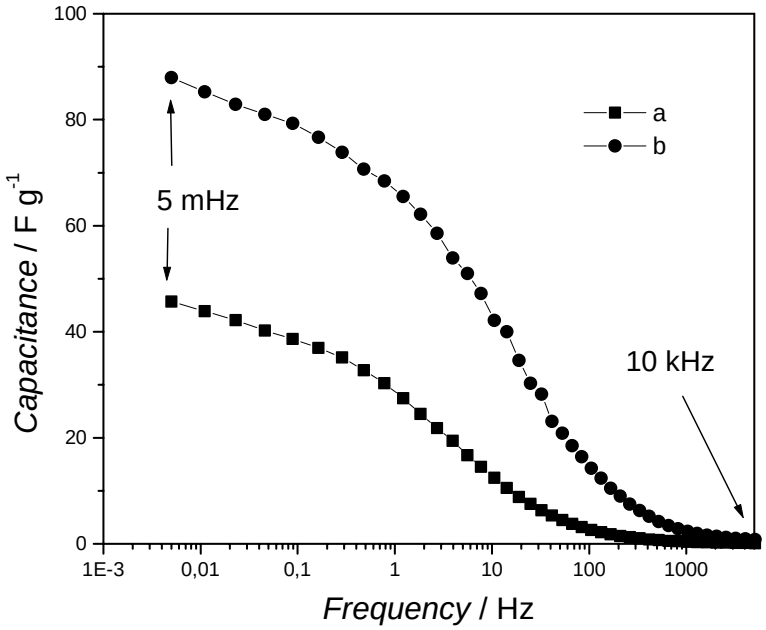

Fig. 2 *EIS expressed as capacitance vs. frequency of a porous carbon electrode (EXP1) made using the SIC process. (a) Before and (b) after activation with* HNO_3.

Conclusions

Mechanically stable supercapacitor electrodes have been successfully made from different carbon materials using the SIC process. Subsequently, the electrodes have been activated with nitric acid. Due to this activation step a significant increase of (pseudo)capacitance was observed. Values of capacitance up to 107 F g^{-1} (carbon EXP 6) have been obtained. The frequency behaviour of the carbon electrodes was excellent indicating that the accessibility of the ions into the pores of the carbon is very good.

Acknowledgements

Support by the Austrian Science Fund in the special research program "Electroactive Materials" is gratefully acknowledged.
We also like to thank Dr. M. Reimerink for supplying us with samples of EXP1 and EXP6 carbons.

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