CARBON BLACK ELECTRODES FOR SUPERCAPACITORS MADE BY SUBSTRATE INDUCED COAGULATION

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When low cost, fine particulate carbon black is used as electrode material, typically a polymer binder is necessary in order to achieve good mechanical integrity and electronic conductivity within the electrode respectively to the current collector. A relatively high binder amount is essential to retain a certain flexibility since too rigid electrodes would be mechanically unstable during electrode manufacturing. However, inevitably the (partial) coverage of carbon particles with binder increases the resistance of the electrode [1] and furthermore interferes the formation of the electrolytical double layer and thus lowers the capacitance.

In order to minimize the binder content in the electrode, we performed carbon precipitation on a current collector substrate by a method which has been developed in our laboratory: Substrate induced coagulation (SIC) [2]. Basically, the SIC process adapted for supercapacitor electrodes proceeds in four steps (Figure 1): (i) The substrate is covered with polyelectrolyte by dipping into a polyelectrolyte solution. (ii) Apart from a monolayer, the polyelectrolyte is removed by rinsing in H_2O. (iii) Immersion of the pre-conditioned substrate in a metastable dispersion of carbon black yields coagulation and thus precipitation of carbon on the substrate surface. (iv) Excess carbon black is removed by rinsing in H_2O. To achieve the desired quantity of active material on the current collector, the steps (i) – (iv) may be repeated as often as requested. As a result, practically binder-free carbon black based supercap electrodes are achieved. By subsequent dipping according to the 4-step SIC process, the mass of coated carbon black precipitated on the substrate increases in a strongly linear manner (Figure 2). It should be emphasized, that only the number of dipping processes and not the duration of immersion of the substrate in the dispersion during each dipping step lead to the observed mass increase of coagulated carbon black. An immersion time of 2 minutes proved to be sufficient.

Figure 3 shows four sweeps of a small carbon black electrode prepared by SIC. The electrochemical and mechanical properties of the electrode remained constant during cycling. The preliminary electrochemical behaviour of the electrode was very capacitive all over the observed potential range. The value of the specific capacitance calculated from the cyclic voltammogram is about 22 F g^{-1}. We expect better capacitance values after we have improved the porosity of the electrodes.

Support by the Austrian Science Fund in the special research program "Electroactive Materials" is gratefully acknowledged.

[1] L. Bonnefoi, P. Simon, J.F. Fauvarque, C. Sarrazin, A. Dugast, *J. Power Sources* **79** (1999) 37.
[2] J.O. Besenhard, O. Claussen, H.-P. Gausmann H. Meyer, H. Mahlkow, US. Pat. 5 705 219.

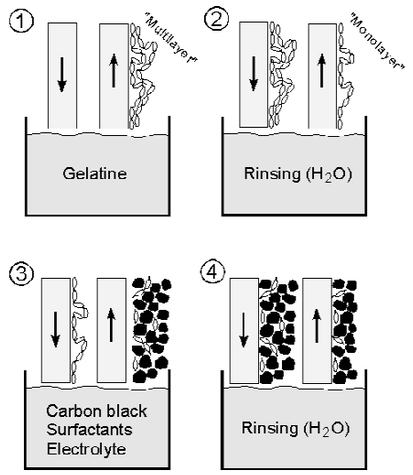

Figure 1: The four-step process for manufacturing "binder-free" electrodes for supercapacitors.

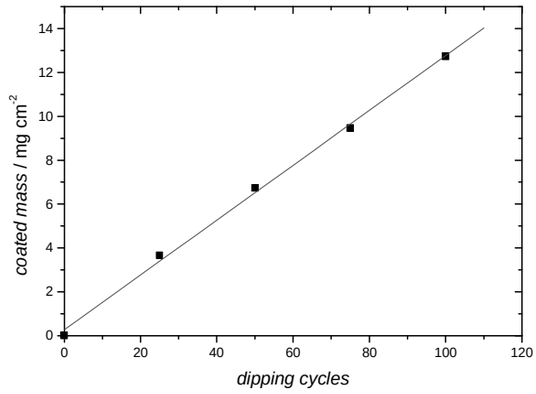

Figure 2: Coated carbon black mass versus number of dipping cycles. A 10 mm × 10 mm titanium grid was used as substrate (current collector), gelatine as polyelectrolyte, hexadecyl trimethylammonium bromide as surfactant, and carbon black (Degussa, type XE2, specific surface area: 960 m^2 g^{-1}) as carbon material. The thickness of a carbon layer obtained by one single dipping process was judged from SEM data and was found to be 0.5 - 1 µm.

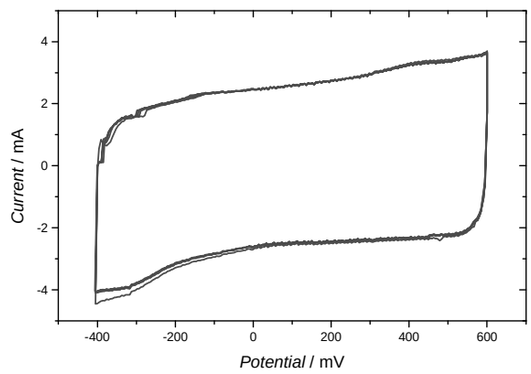

Fig. 3: Cyclic voltammogram of a carbon electrode (10 mm × 10 mm Ti grid) made by the SIC process (sweeps 1, 10, 20, 30, 40 and 50 are shown). Scan rate ν = 20 mV s^{-1}, reference electrode: SCE, electrolyte: 0.5 M H_2SO_4. Specific capacitance: C = 22 F g^{-1}.