Use of electrode-bonded paper separators in non-aqueous electric double-layer capacitors and Li-ion batteries

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As both the weight and thickness of portable electronic devices continue to decrease, novel types of thin and flat Li-ion cells were developed that use flexible and lightweight multi-layer foil packaging materials.[1] Two types of separators are used in such cells: the more traditional microporous polyolefin separators (MPSs) and plasticized-polymer composite separators. The main disadvantage of the better-known MPSs is their cost, which makes their use quite expensive in other types of electrochemical energy storage devices, such as electric double-layer capacitors (EDLCs). This is especially true for the newest bondable, surface-coated MPSs.[2]

Traditionally, non-woven fabrics or special paper separators are used in metal can-packaged EDLCs due to their low cost and high porosity. However, to the best of our knowledge, the use of such materials has not been reported in thin, bonded-electrode systems, such as those investigated by us. In the present communication, we report the results of our successful search for low-cost, high-performance bonded-electrode paper separators, a method to fabricate non-aqueous EDLCs using such separators, and characteristics of the resulting devices.

We evaluated a number of capacitor paper samples from a variety of sources in terms of their bond strength to the plasticized carbon electrode and the overall stability of the EDLC. In addition, we have fabricated a bonded-electrode high-performance rechargeable Li-ion battery based on the $LiCoO_2$/MCMB graphitic carbon system, which used the optimized paper separator. The battery exhibited excellent long-term cycling performance even after several hundred charge/discharge cycles. Of course, the use of such separators is limited to small-capacity cells, where the thermal shutdown of the separator in case of cell malfunction is not required.

The bonded-electrode EDLCs with paper separators were fabricated as follows: an electrode slurry was prepared by mixing at an elevated temperature a high-surface-area carbon (ASupra, 1,800 m^2/g) with a polymeric binder (Kynar PowerFLEX LBG, AtoFina) and plasticizer (propylene carbonate) in acetone as a casting solvent. The electrode tape was cast on a polyester carrier tape using a doctor-blade apparatus and acetone was evaporated to give an electrode tape weighing between 0.04 and 0.15 g/in^2. An aluminum grid current collector coated with an electrically conductive adhesive was laminated to one or two such electrode sheets using a double-roll heated laminator and then cut into coupons ca. 15-80 cm^2 in area. Two such sheets were again laminated to two sides of an untreated paper separator, the EDLC preform extracted in ether or methanol, dried under vacuum at 70-80°C, packaged in a glove box in an aluminum laminate bag, activated with a 1.5 M Et_4BF_4 in acetonitrile, sealed and tested. For comparison, identical procedure was used to fabricate EDLCs using a surface-treated MPS instead of paper.[2,3]

The results shown in Table I and Figs. 1-2 indicate that high-performance EDLCs and small Li-ion batteries can be fabricated by using judiciously selected paper separators that can be bonded without the use of external adhesives to plasticized high-surface-area or graphitic carbon electrodes, as well as to $LiCoO_2$-based electrodes. We expect that these results may lead to substantial materials cost savings during the manufacture of bonded EDLCs and small Li-ion batteries.

Paper sample	Material	Adhesion to PLiON electrodes	Adhesion to supercap electrode
A	regenerated cellulose	good	good
B	regenerated cellulose	fairly good	good
C	regenerated cellulose	good	fair
D	manila longfibre	poor	partial
E		poor	poor
F	kraft/manila mix	partial	fair
G	?	partial	poor
H	?	no	no

Table I. Effect of fiber type in paper on its adhesion to plasticized electrodes

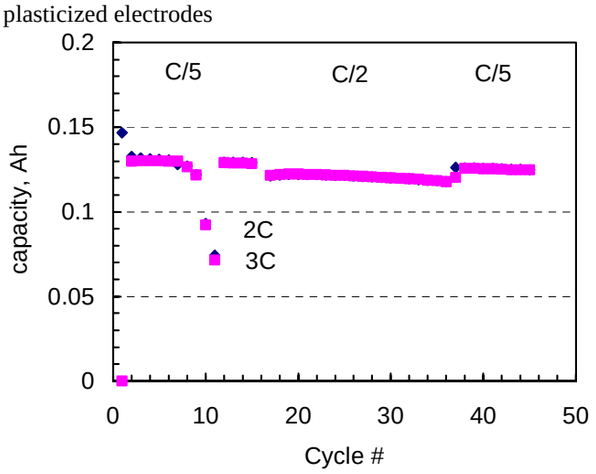

Fig. 1. Capacity of a $LiCoO_2$/MCMB Li-ion battery with a bonded paper separator

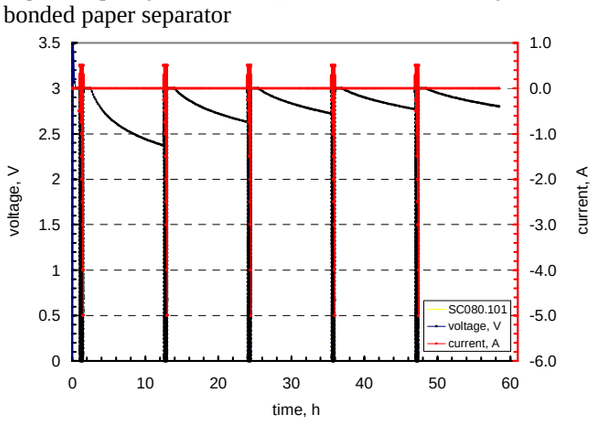

Fig. 2. OC voltage stabilization of a non-aqueous EDMC with a bonded paper separator during subsequent cycles

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