

## Hybrid capacitor with activated carbon electrode, Ni(OH)<sub>2</sub> electrode and polymer hydrogel electrolyte

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Received 29 January 2005; received in revised form 7 July 2005; accepted 8 July 2005

Available online 6 September 2005

### Abstract

A new hybrid capacitor (HC) cell was assembled using an activated carbon (AC) negative electrode, an Ni(OH)<sub>2</sub> positive electrode and a polymer hydrogel electrolyte prepared from crosslinked potassium poly(acrylate) (PAAK) and KOH aqueous solution. The HC cell was characterized compared with an electric double layer capacitor (EDLC) using two AC electrodes and the polymer hydrogel electrolyte. It was found that the HC cell successfully worked in the larger voltage range and exhibited ca. 2.4 times higher capacitance than the EDLC cell. High-rate dischargeability of the HC cell was also superior to that of the EDLC cell. These improved characteristics strongly suggest that the HC cell can be a promising system of capacitors with high energy and power densities.

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**Keywords:** Hybrid capacitor; Activated carbon; Ni(OH)<sub>2</sub> electrode; Polymer hydrogel electrolyte; Electric double layer capacitor

### 1. Introduction

Electric double layer capacitors (EDLCs) have been commercially used as power sources of various devices because of their high power density and long cycle performance. In order to increase the energy and power densities, extensive works on activated carbon (AC) electrodes and electrolytes in EDLCs have been carried out. However, the increase in the energy density is limited because the charge–discharge mechanism is based on the adsorption and desorption of ions on the AC electrode surface without charge transfer. Pseudocapacitance electrodes using metal oxides [1,2] or conducting polymers [3,4] have been reported to show much higher capacitance than AC electrodes, although the kinetics and/or cycle stability are generally poor due to the redox reaction. For the purpose of gaining both high energy and power densities, several types of hybrid capacitors (HCs) composed of the AC and pseudocapacitance electrodes have recently

been proposed and investigated [5–7]. If the pseudocapacitance electrode with very small polarization is used instead of one of the AC electrodes in EDLCs, the voltage change of the electrode scarcely occurs during charging and discharging, leading to a remarkable increase in the capacitance and energy density of the cell.

Ni(OH)<sub>2</sub> has been also used as electrode material of HCs [8,9]. Park et al. assembled an HC cell using an AC negative electrode, an Ni(OH)<sub>2</sub>/AC composite or Ni(OH)<sub>2</sub> positive electrode and 6 M KOH aqueous solution [8]. The HC showed much higher capacitance than the EDLC due to the high capacitance of Ni(OH)<sub>2</sub>. Previously, we reported that an alkaline polymer hydrogel electrolyte prepared from crosslinked potassium poly(acrylate) (PAAK) and KOH aqueous solution [10,11] exhibited excellent performance in EDLCs and increased the capacitance of the AC electrode probably due to the pseudocapacitance [12–14]. The electrolyte also has the significant advantage of showing very slow electrolyte creepage [15]. Furthermore, it was found that charge–discharge reactions occurred smoothly and reversibly at the interface between the electrolyte and Ni(OH)<sub>2</sub> elec-

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trode in nickel/metal hydride cells [16]. In this work, a new HC cell was assembled replacing one of two AC electrodes in the EDLC cell with the polymer hydrogel electrolyte by an  $\text{Ni}(\text{OH})_2$  electrode, and the charge–discharge characteristics were investigated compared with those of the EDLC cell.

## 2. Experimental

The polymer hydrogel electrolyte was prepared from PAAK and 10 M KOH aqueous solution and impregnated into a nonwoven fabric separator as described in our previous paper [14]. An AC fiber cloth (Kuraray Co., Ltd.) with a specific surface area of  $2000 \text{ m}^2 \text{ g}^{-1}$  was used as a negative electrode material of the HC cell. The AC cloth ( $10 \text{ mm} \times 10 \text{ mm}$ ) was dried at  $120^\circ\text{C}$  for 24 h in order to remove impurities such as adsorbed organic compounds. The AC cloth was impregnated with the polymer hydrogel electrolyte in the same manner as the separator. The resulting AC cloth with the electrolyte was stuck on a nickel sheet with the carbon paste. On the other hand, a commercial sintered  $\text{Ni}(\text{OH})_2$  electrode ( $10 \text{ mm} \times 10 \text{ mm}$ ) was used as a positive electrode of the HC cell. Before assembling the cell, the  $\text{Ni}(\text{OH})_2$  electrode was activated by five charge–discharge cycles at a rate of  $0.5\text{C}$  in 10 M KOH aqueous solution, and then fully charged and discharged to the depth of discharge of 50%.

An experimental HC cell was assembled as shown in Fig. 1. The separator with the polymer hydrogel electrolyte was stacked between the AC and  $\text{Ni}(\text{OH})_2$  electrodes. A similar type EDLC cell was also assembled using two AC electrodes for comparison [14]. Thickness of each cell composed of the two electrode and separator was ca. 1.0 mm. In all charge–discharge cycling tests, the HC and EDLC cells were charged and discharged in the voltage ranges of 0.4–1.2 and 0.2–0.8 V, respectively, at a constant current. Cyclic voltammetry for the AC electrode was carried out in the potential range of  $-1.0$  to  $0.2 \text{ V}$  versus  $\text{Hg}/\text{HgO}$  at a scan rate of  $10 \text{ mV s}^{-1}$ . The temperature in all measurements was kept at  $25^\circ\text{C}$ .

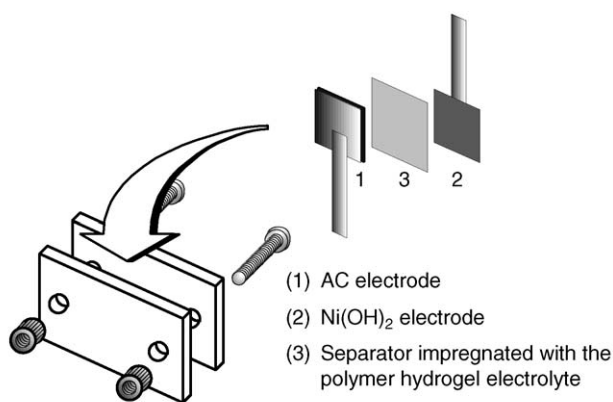


Fig. 1. Schematic representation of the experimental cell assembly.

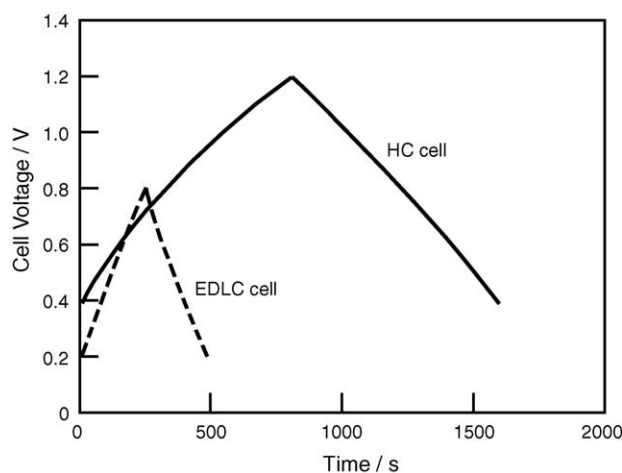


Fig. 2. Charge–discharge curves at  $1 \text{ mA cm}^{-2}$  of the HC and EDLC cells.

## 3. Results and discussion

Fig. 2 shows typical galvanostatic charge and discharge curves of the HC and EDLC cells with the polymer hydrogel electrolyte ( $1 \text{ mA cm}^{-2}$ , 10th cycle). Almost linear charge and discharge curves were observed in each case. In our previous papers, good electrode/electrolyte interface could be constructed using the polymer hydrogel electrolyte and either the AC electrode [12–14] or the  $\text{Ni}(\text{OH})_2$  electrode [16]. It can be considered that the good electrode/electrolyte interface was also formed in the present experimental HC cell and, therefore, the cell successfully worked as a capacitor. IR drop was scarcely observed in each curve under this experimental condition, which is ascribed to high ionic conductivity of the polymer hydrogel electrolyte (ca.  $6.0 \times 10^{-1} \text{ S cm}^{-1}$  at  $25^\circ\text{C}$ ) and its good contact with both the electrodes. From the discharge curve, the discharge capacitance of the HC cell was evaluated to be  $0.97 \text{ F}$ , which was ca. 2.4 times higher than that of the EDLC cell,  $0.40 \text{ F}$ . It is clear that the capacitance of the cell was markedly increased by replacing one of the AC electrode in the EDLC with the  $\text{Ni}(\text{OH})_2$  electrode. In addition, it was found that the HC cell could work until a larger cell voltage such as 1.2 V, compared with that for the EDLC cell, 0.8 V. The coulomb efficiencies were ca. 98 and 95% for the HC and EDLC cells, respectively, in the voltage ranges as shown in Fig. 2. This leads to a large increase in both the power and energy densities of the cell.

In order to elucidate the reason for the improved charge–discharge characteristics of the HC cell, potential changes of the negative and positive electrodes were examined using an  $\text{Hg}/\text{HgO}$  reference electrode during the charge–discharge cycle test of the cell. As can be seen from Fig. 3, both the negative and positive electrodes showed almost linear charge–discharge curves in the HC and EDLC cells. In case of the HC cell, the potential change of the  $\text{Ni}(\text{OH})_2$  positive electrode was very small (ca.  $0.36$ – $0.38 \text{ V}$  versus  $\text{Hg}/\text{HgO}$ ), compared with that of the AC positive electrode in the EDLC cell. On the other hand, the potential of the

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