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Charge storage dynamics in the activated carbon electrode of supercapacitors

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Abstract

This research investigated the electrochemical performance of the alkaline-activated carbon in the EDLC electrode, focusing on the charge storage dynamics in supercapacitors. At the low current density of discharge measurement, models like Ohmic leakage, diffusion-controlled, and activation-controlled Faradaic were applied to capture additional charge storage behaviors. These revealed the influence of parasitic reactions, possibly a decomposition reaction near 1 V. The work bridged gaps in understanding the balance between EDLC and faradaic reactions, contributing to the development of high-performance, next-generation supercapacitors.

1. Introduction

The increasing demand for advanced energy storage in industries has fueled interest in high-performance supercapacitors, known for their high-power density, energy capacity, and long cycle life. Activated carbon is a key material due to its excellent properties, including high surface area and adaptable pore structure. Understanding charge storage dynamics in activated carbon electrodes, which involve both electric double-layer capacitance (EDLC) and faradaic redox reactions, is critical for improving supercapacitor performance[1)]. This study aimed to investigate these mechanisms, applying on the alkaline-activated carbon, to advance next-generation energy storage devices.

2. Experiment

Moso bamboo, steam-treated at 200°C for 2.5 hours, was used as the starting material. Carbonized steam-treated bamboo at 500°C was mixed with KOH or NaOH at a 1:6 weight ratio or with a KOH-NaOH eutectic mixture, then activated at 800°C. After neutral washing, samples were dried at 105°C and labeled (1:0), (NaOH 1:6), (KOH 1:6), and (NaOH + KOH 1:6). Structural properties were characterized using Raman spectroscopy, XPS, and N_2 adsorption isotherm at 77 K with BET and GCMC analyses. Electrodes were prepared by mixing synthesized activated carbon, acetylene black, and PVDF solution in an 8:1:1 ratio with NMP. The slurry was coated onto Al foil, dried, and punched for use in

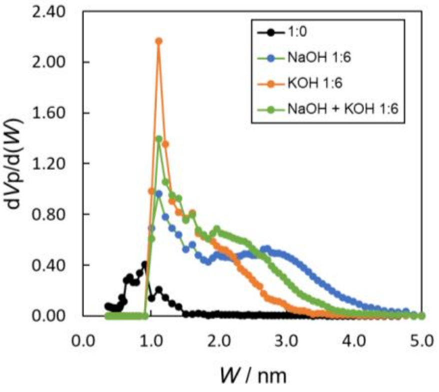

Fig.1 The pore size distribution of the activated samples calculated by the GCMC model.

symmetric cells with 1 M $TEABF_4$/PC electrolyte. Cyclic Voltammetry (CV), electrochemical impedance spectroscopy (EIS) measurement, and charge/discharge tests were performed. Fitting models and 3D Bode plots were used to characterize the charge storage mechanism.

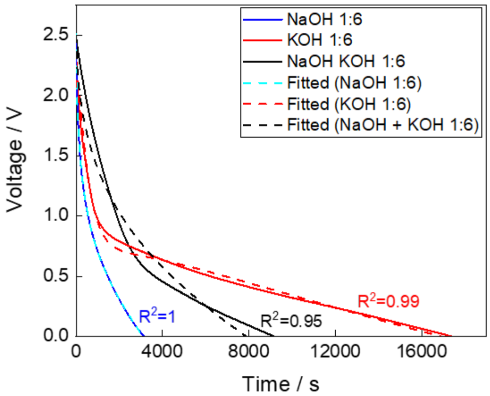

Fig.2 The combination-model fitting with the experiemental results.

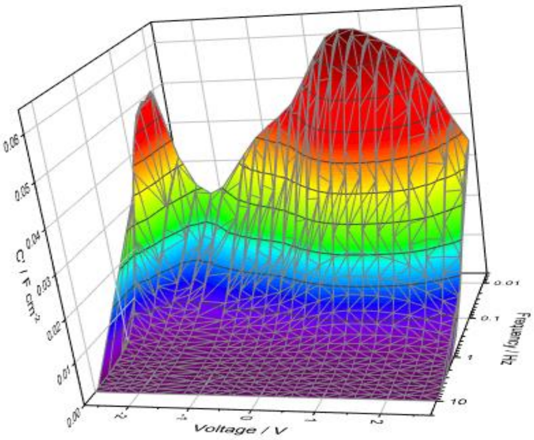

Fig.3 The relationship between real capacitance, potential and frequency expressed through 3D plot of the (KOH 1:6) sample.

3. Results and discussion

3.1 Activated carbon's properties

Raman spectroscopy identified disordered carbon structures. XPS analysis showed varying functional group distributions, with KOH activation preserving more C-C/C=C bonds, while NaOH consumed more carbon content. The pore size distribution (Fig.1) calculated from N_2 adsorption/desorption isotherms indicated that changing activators led to the enlargement in the pore size of the microporous structure.

3.2 Electrochemical mechanism

CV data showed that at high scan rates, samples behaved like ideal EDLCs, with over 90% capacitive contribution, while the mixture-activated sample achieved the best stability. GCD results indicated that the sample acted typical EDLC behavior at high current density; however, it exhibited a unique response at low current density. Therefore, fitting models were applied to understand the underlying phenomenon during the measurement (Fig.2). The charge storage mechanism was explored using models such as the Ohmic leakage, diffusion-controlled, and activation-controlled Faradaic models, revealing contributions from both EDL capacitance and parasitic Faradaic reactions at low current densities$^{2)}$. A combined discharge model and 3D bode plot (Fig.3) from EIS measurement captured the mapping charge storage dynamics, with the decomposition reaction of electrolytic solvent at *ca.* 1 V.

4. Conclusions

The study suggested that the storage and discharge mechanisms of electrodes might involve several processes. The findings also suggested the possible effect of the pore structure on the electrochemical performance of carbon electrodes for supercapacitors.

References

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