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Effects of structural design on the performance of electrical double layer capacitors

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HIGHLIGHTS

- Effect of design parameters on the electrical double layer capacitor (EDLC) performance is studied.
- This study is based on the concentration-dependent mathematical modeling of EDLCs.
- Design parameters studied are the electrode thickness, electrode porosity, and initial electrolyte concentration.
- The results can be applied to optimize the design of EDLCs at broad range of operating conditions.

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ABSTRACT

The effect of different structural designs on the performance of electrical double layer capacitors (EDLCs) has been studied through a mathematical model that considers the mass transfer and conservation of charge equations. The structural design parameters considered in this study are the electrode thickness, electrode porosity, and initial electrolyte concentration. The performance parameters studied are the cell capacitance, specific energy, specific power, and electrolyte concentration for a range of discharge rates. The results of this study can be used to optimize EDLCs at various operating conditions.

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1. Introduction

Electrical double layer capacitors (EDLCs) store energy by the separation of charge at the electrode/electrolyte interface, forming an electrical double layer [1–3]. The reversible nature of this energy storage mechanism and the capability for EDLCs to deliver high power density and long cycle life, results in a wide range of potential applications. For example, they can provide starting power in the start-stop systems used in automobiles, operate as backup power sources in computers, regulate voltage to the energy grid for wind and solar energy generation systems, and open aircraft doors in an emergency [2–5]. However, the most significant application of EDLCs is in electric and hybrid electric vehicles where they can supply energy in the condition of high power demand and capture energy from regenerative breaking systems [6–8]. In order to predict the performance characteristics of EDLCs, developing an appropriate mathematical model that can describe the EDLC behavior during charge or discharge is crucial.

Mathematical models used to analyze the performance and characteristics of EDLCs can be categorized into three groups. The first group consists of models describing EDLCs as equivalent RC circuits [9–13]; Although these models could predict the electrochemical device response with reasonable accuracy, they fail to relate the model parameters to the electrochemical processes that occur in the double layer such as diffusion and migration of ions and electrolyte polarization. In the second group, models take the heterogeneous microstructure of the electrodes into account and are based on solving a Modified Poisson–Boltzman (MPB) equation for electric potential in the electrolyte [14–18]. Such models could be utilized to study the influence of electrode microstructures such as morphology and pore shapes on the surface area and capacitance offered by the electrodes [16–17]. However, similar to RC circuits' models, they could not predict the transport processes in the electrode/electrolyte interface during charge/discharge. This is due to the fact that in both models, it is assumed that the electric potential and ion concentration are at their

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Nomenclature

S_d	maximum surface area (cm^2/cm^3)
c	concentration of electrolyte (mol m^{-3})
C_d	electrical double layer capacitance (F m^{-2})
D	diffusion coefficient (m^2 s^{-1})
E	specific energy (Wh kg^{-1})
F	Faraday's constant, 96,487 (C mol^{-1})
i	current density (A m^{-2})
I	total applied current density to the cell (A m^{-2})
l	thickness (m)
M	molecular weight (g mol^{-1})
$\dot{N}''_x$	rate of molar flux (mol s^{-1} m^{-2})
P	specific power (W kg^{-1})
t	time (s)
T	temperature (K)
t^0_+	transference number of H^+ with respect to the solvent
x	spatial coordinate along the thickness of the cell
z	charge number

Greek letters

ε	porosity
κ	electrolyte conductivity (S m^{-1})
υ	dissociation coefficient
ρ	density (kg m^{-3})
σ	electronic conductivity (S m^{-1})
ϕ	electric potential (V)
ω	mass per unit area of the cell (kg m^{-2})

Subscripts

+	positive electrode
−	negative electrode
0	zero charge/solvent
1	electronic
2	ionic
av	average per area of EDLC
avg	average per gram of EDLC
e	electrolyte
etd	electrode
eff	effective
i	positive or negative charge
init	initial
ref	reference
sep	separator
pos	positive electrode
neg	negative electrode

Superscripts

+	length plus epsilon
−	length mines epsilon

equilibrium in steady-states condition. Hence, they could not depict the electrochemical processes that occurred during the EDLC operation time. For this, a third group of EDLCs models are employed which are based on the governing equation of mass transfer and conservation of charge [19–23]. These models can simulate the electrochemical processes occurring in the EDLC during charge/discharge periods, such as diffusion and migration of ions in the porous mediums and the formation/decay of the electrical double layer. Moreover, they can predict time accurate EDLC voltage and simulate the performance of the EDLC through specific energy and power.

The third group of models was initiated by Johnson and Newman [24]. With the assumption of concentration uniformity and neglecting potential-dependent capacitance, Dunn and Newman [22] found an analytical solution for the governing equations during EDLC charge. In addition, they presented the maximum specific energy achievable by optimizing the electrode thickness, electrode porosity, and the final voltage constrained for constant-current and constant-power discharges. Srinivasan and Weidner [25] studied the importance of electronic and ionic resistance in the design of the EDLC. Their results showed the trade-off between energy and power density, as the physical properties of the cell structural parameters were varied (e.g., electrode thickness). Most of the models in the third group assumed no concentration gradients exist in the EDLC [22,25]. This assumption simplifies the set of partial differential equations to ordinary differential equations which can be solved analytically. However, Lin et al. [26] showed that including concentration polarization in the modeling, could decrease the predicted cell voltage up to 25% compared with a model that ignores concentration polarization for electrode with hydrous ruthenium oxide (RuO_2 xH_2O). In addition, if the concentration polarization is included in the modeling, electrolyte concentration profiles during charge/discharge period are also achievable [27]. Madabattula and Gupta [27] developed their EDLC model based on the transport mechanisms which includes concentration gradient across the cell. They examined the necessity of employing a 2D model and demonstrated that the 1D modeling for a EDLC is quite sufficient.

The aim of present work is to investigate the effect of employing different structural design parameters such as electrode thicknesses, electrode porosities, and initial electrolyte concentrations on the EDLC performance. To do this, first, we have developed a concentration dependent model of a third group as Ref. [27]. The model accuracy is verified by comparing simulated cell voltage profile with the experimental data in Ref. [28]. Then, a set of numerical experiments are conducted to study the effects of utilizing different structural design parameters on the EDLC capacitance, specific energy, specific power, and electrolyte concentration for a range of discharge current densities. The simulation results could also be utilized for practical application, to optimize the cell design of EDLCs. In this study, Comsol 4.3 with PDE coefficient module was employed to simulate the EDLC model.

2. Modeling and computer simulation

The mathematical model employed to simulate electrical double layer capacitance is based on the transport processes in Refs. [24,27]. In this model, the mass transfer and conservation of charge equations are developed for each components of the cell structure, including the two porous activated carbon electrodes and the ion permeable membrane separator. It is assumed that the electrolyte is an asymmetric concentrated solution of sulfuric acid that dissociates in water into two positively charged hydrogen ion, H^+, and one negatively charged sulfate, SO_4^{2-} [27]. The current collectors' resistance is neglected as a result of their high conductivity.

2.1. Mass balance

Mass balance for a dissolved specious j is written as [29]

$$\frac{\partial(\varepsilon c_j)}{\partial t} = -\frac{\partial(\dot{N}''_{x,j})}{\partial x} + R_j \tag{1}$$

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