

High-Energy Electrochemical Capacitors

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1. Introduction

The decarbonization of energy to meet the low-carbon energy strategy set for 2050 has led to a continuous increase in the contribution of electricity generated from renewables to our growing energy demands, where their inherent intermittency of supply must be addressed by a step-change in energy storage. Additionally, the nonstop emerging demands of modern technologies still require the design of versatile energy-storage/power supply systems, ensuring a secure and continuous supply of energy on a large scale with wide ranges of power and energy densities commensurate with each application. In the case of long-term power/energy demands, technologies such as batteries and fuel cells that are capable of storing large amounts of energy and releasing it over a longer period are needed, while, in short-term power needs and energy demands over the timescale of 0.1–100 s, technologies capable of delivering high power to flatten power outputs fluctuations and provide an energy surge in short discharge periods become imperative. Since there is no single energy storage technology to address this wide range of energy/power requirements simultaneously, a variety of storage technologies and systems composed of their combinations must be deployed.

As the key electrochemical energy storage devices in energy-storage/power supply systems, electrochemical capacitors (ECs), also known as supercapacitors, are capable of absorbing and delivering large amounts of energy in the form of electric charge over short timescales. They are increasingly being used as an attractive power solution for a variety of applications, such as automotive acceleration and braking or pulsed power applications in telecommunications, to overcome the problems associated with poor power quality and the inherent intermittency of renewable resources by matching the specification of the primary energy sources closer to that of the average power demand in each particular case.

In order to stimulate the development of ECs and fully exploit their potential in the field of energy storage for a wide range of energy and power supplies, further advances in the engineering of materials, including novel electrode materials, electrolytes, and binders, used for the manufacturing of ECs are needed as well as a better understanding of their energy storage mechanisms, performance analysis, and their integration with other storage technologies.

The objective of this Special Issue is to enhance the knowledge base and to stimulate the development of high-energy supercapacitors for a wide range of energy and power supplies. Therefore, the Special Issue is aimed at providing recent developments in high-energy electrochemical capacitors by exploring a wide range of scientific topics in the field, including, but not being limited to, the following:

- The synthesis, characterization, and properties of electrode materials for electric double-layer capacitors, pseudocapacitors, and hybrid capacitors;
- Novel and benign electrolytes for electrochemical capacitors;
- The understanding of the underlying mechanisms of energy storage in electrochemical capacitors;
- The performance analysis of electrochemical capacitors under different conditions;



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- The integration of electrochemical capacitors with other storage technologies to improve the overall performance of energy systems.

2. Special Issue Content

This Special Issue on “High-Energy Electrochemical Capacitors” reports on some of the most recent findings in electrode materials for supercapacitors and their current state and prospects in energy storage and conversion systems. Six articles were accepted and published in this Special Issue. Most of the articles published here focus on the development of electrode materials for supercapacitors and the method of testing and quality assurance of the device; however, two articles examine the development of anode electrodes in lithium-ion batteries and an electrochemical lithium recovery (ELR) method for the extraction of lithium using a LiMn_2O_4 spinel electrode. These two will help in the performance improvement of the battery and to secure lithium resources for the battery as another key component used in energy-storage/power supply systems to respond to energy demands with different time scales in a wider range as needed.

Farooq et al. [1] developed a multi-channel fast EIS instrument to measure the impedances of the multiple numbers of supercapacitors. This technique is based on a multi-frequency EIS method using multi-sine sweeping that can replace the time-consuming DC tests, such as charge/discharge and self-discharge tests. The method is capable of characterizing and testing multiple supercapacitor cells simultaneously in a short period of time and can be applied to a mass production line of supercapacitors for their quality assurance. This study helps in the classification of supercapacitors based on their performance, which assists in maximizing the performance of the supercapacitor module and reducing its potential performance degradation or failure in mass production lines.

Mirzaeian et al. [2] developed highly porous nanostructured nitrogen-doped carbon and activated carbon aerogels used as electroactive material in an aqueous-electrolyte-based electrochemical cell using a 6 M KOH solution as the electrolyte. Through the performance analysis of the electrochemical capacitors by cyclic voltammetry and electrochemical impedance spectroscopy measurements, they showed that the incorporation of nitrogen and oxygen functional groups shown by the XPS analysis of the carbon materials improves electrode/electrolyte wettability as evidenced by a decrease in the contact angles after nitrogen and oxygen doping. This resulted in a significant increase in the specific capacitance of the cell, with the highest specific capacitance of 289 F g^{-1} attained for the nitrogen and oxygen dual-doped sample. It is argued that this significant increase in the specific capacitance of the cell could be attributed to both EDLC and pseudocapacitive contributions originated from the highest specific surface area of the carbon and the presence of nitrogen/oxygen functional groups within the carbon matrix and on its surface, respectively. The results of this study also showed a strong relationship between the surface chemistry of the carbon used as the electroactive material in the electrode formulation and equivalent series/charge transfer resistances as witnessed by EIS measurements, making nitrogen and oxygen dual-doped carbon an ideal and cost-effective candidate for aqueous-based electrochemical capacitors.

In another study on the “Improvement of the Pseudocapacitive Performance of Cobalt Oxide-Based Electrodes for Electrochemical Capacitors”, Mirzaeian, Abdullin et al. [3] synthesized cobalt oxide nanopowders by the pyrolysis of aerosol particles of a water solution of cobalt acetate as well as a cobalt nanopowder by the subsequent reduction of the obtained cobalt oxide by annealing under a hydrogen atmosphere and investigated the electrochemical characteristics of the materials in an electrochemical cell using a 3.5 M KOH solution as the electrolyte. This study demonstrated a new effect of enhancing the electrochemical activity of the Co_3O_4 electrode as a result of the hydrogen reduction of the oxide to metal and the subsequent formation of a core-shell structure by forming a thin oxide/hydroxide layer obtained by the in situ electrochemical oxidation of the cobalt nanoparticle surface. It is argued that the increased storage capacity of the reduced Co_3O_4 compared to the initial Co_3O_4 is because of the presence of the different forms of oxide

and hydroxide species formed on the surface of metallic Co particles. This new and simple method of enhancing electrochemical activity is mainly related to the redox activity of the electroactive material. In addition to these, the small thickness of the oxide/hydroxide layer provides a high reaction rate, and the presence of a metal skeleton leads to a low series resistance of the electrode. This proposed simple method can be applied to other metal oxides (e.g., nickel or tungsten) that are able to recover under a hydrogen atmosphere, as well as to Co_3O_4 electrodes obtained by other methods.

Lee and Suk [4] investigated the application of laser cutting on the lab-made uncompresses graphite-coated copper anode for lithium-ion batteries by applying the laser cutting on the graphite side and on the copper side of the anode separately and identifying their laser cutting characteristics under different laser powers and cutting speeds in the range of 50–250 W and 500–5000 mm/s, respectively. They measured the absorption coefficients in the ultraviolet, visible, and infrared ranges for the graphite side and for the copper side of the electrodes to understand the physical phenomena of the laser cutting process when applied to different sides of the anode. According to the observations of physical phenomena during laser cutting, they categorized cutting phenomena in five regions: excessive cutting, proper cutting, defective cutting, excessive ablation, and proper ablation. Through the analysis of reflectivity, transmissivity, and absorptivity measurements of the graphite-side-laser-cut and copper-side-laser-cut anode samples, they concluded that the laser cutting applied on the graphite was more efficient than that on the copper and provides an opportunity to understand the material removal mechanism more precisely. Their study helps to understand the effect of electrodes' characteristics on the laser cutting of electrodes to substitute conventional mechanical cutting methods.

Joo et al. [5] studied electrochemical lithium recovery by using spinel LiMn_2O_4 as a positive electrode due to its high Li^+ selectivity and stability. They reviewed different electrochemical lithium recovery (ELR) systems—including the $\lambda\text{-MnO}_2/\text{Pt}$ system with hydrolysis, asymmetric battery system, hybrid supercapacitor system, symmetric rocking chair battery-liked system, and flow-through type reactor system—considering their advantages and limitations, and proposed the $\lambda\text{-MnO}_2/\text{Pt}$ system, $\lambda\text{-MnO}_2/\text{Ag}$ battery system, $\lambda\text{-MnO}_2/\text{AC}$ hybrid supercapacitor, $\text{LiMn}_2\text{O}_4/\text{Li}_{1-x}\text{Mn}_2\text{O}_4$ rocking chair battery-liked, and the flow-type system as representative electrode systems for electrochemical lithium recovery. They also investigated different techniques, including electroanalysis, X-ray measurement, numerical simulation, and selective concentration polarization, for the analysis of the ELR system and understanding its mechanism and behavior. They also reported the practical application of the ELR system in wastewater treatment, battery recycling, and desalination plants.

Abbas et al. [6] reviewed the current state and future prospects of electrochemical energy storage and conversion systems and discussed some of the major examples in this area, condensing their technological developments in recent years with an emphasis on their fundamental working principles and the material compositions of their various components, such as their electrodes and electrolytes. They provided an overview of the technological advances, operational parameters, material compositions, and current/potential applications of electrochemical energy storage and conversion devices along with their technical maturity and commercial practicability and reported a detailed comparison of the technical data of various electrochemical energy storage and conversion devices as well as their advantages in specific applications and the future challenges and perspectives in the applications of these technologies.

3. Closing Remarks and Future Challenges

The research works published in this Special Issue provide new data, information, and findings on: (i) the synthesis, characterization, and properties of different electrode materials for electric double-layer capacitors and pseudocapacitors using aqueous electrolytes as the benign electrolyte; (ii) the underlying mechanisms of their energy storage; (iii) the simultaneous methods of their performance analysis, characterization, and testing

for their fast quality assurance in mass production; and (iv) new methods of electrode fabrication/cutting for Li batteries and also electrochemical lithium recovery (ELR) that help in the integration of electrochemical capacitors with other storage technologies to improve the overall performance of energy systems. I firmly believe that these findings will help the scientific community to deal with aspects of energy storage improvement, energy management, and the problems associated with poor power quality and the inherent intermittency of renewable resources and will stimulate the research community to further contribute to the development of the field.

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