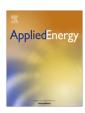


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Characterization of asymmetric ultracapacitors as hybrid pulse power devices for efficient energy storage and power delivery applications *



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HIGHLIGHTS

- An asymmetric NiOOH/C capacitor module was characterized at a number of operating conditions,
- Different capacitor devices for power capture and delivery are summarized for efficient energy conservation.
- Faradaic charge storage in a hybrid ultracapacitor is discussed fundamentally in detailed calculations.
- The capacitor module with a battery was successfully operated up to 150 A as a hybrid power device.
- Modified cold cranking and hybrid pulse power profiles were applied to the device pulse power characterization.

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ABSTRACT

The cost of Ni-MH and Li-ion battery packs is cut considerably for applications in HEV and pHEV onboard energy storage. The operational lifetime of these battery packs is still a cost concern for end users and limits long distance applications. The battery storage devices currently dominate onboard energy storage fields via slow electrochemical reactions. The battery pack experiences an extra burden when absorbing bursts of energy or releasing the required energy to facilitate quick acceleration. The dynamic operation under thermal and mechanical stresses especially during severe weather conditions has adverse effects on the battery operating life due to chemical compounds and electrode materials degradation. To overcome these limitations, an ultracapacitor can quickly polarize electrolyte solutions and accumulate energy via rapid electrostatic charges at the electrode-electrolyte interface. The ultracapacitor combines advantages including long cycle life and high power density of a conventional capacitor with enhanced energy storage capability due to high electrode surface area and low internal resistance. A hybrid ultracapacitor can be designed by using both a double-layer capacitor electrode and a battery electrode. This hybrid structure increases energy conservation and capacitance. It is also recognized as an asymmetric ultracapacitor. A power module consisting of 10 asymmetric ultracapacitors with 300 F at an operating window from 4.0 V to 14.5 V was tested for studying of these unique features. These 10 electrochemical capacitors were connected in series and performed electrical characterizations including Ragone plots, self-discharge, features at high current loads, working voltage, and related energy storage performance. The ultracapacitor module was then operated together with a Pb-acid battery as a hybrid power device. Performance of the hybrid power system was characterized by using an adjusted cold cranking and a modified hybrid pulse power profile. The irreplaceable structures on energy storage and power delivery serve as a potential to further reduce the cyclic burden and thermal stress in advanced battery packs for onboard energy storage applications.

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1. Introduction

Among various energy storage and delivery systems, battery packs (Pb-acid, Ni-MH, and Li-ion cells), fuel cells (PEMs and

SOFCs), electrochemical ultracapacitors (*C/C*, NiOOH/C, *C/Li**), and redox flow batteries (PSBs and VRBs¹) have been acknowledged as the most practical and/or promising candidates for providing a clean and sustainable energy economy, especially some power devices developed and applied to onboard energy conversion and conservation systems [1–3]. Ultracapacitors in the past years have attracted

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 $^{^{\}rm 1}$ PSBs and VRBs refer to polysulfide-bromide and vanadium redox flow batteries, respectively.

considerable attention for various start-stop energy system applications such as heavy-duty trucks, space power, armoured cars, and transit buses [4,5]. These rapid power capture and delivery devices have been built and quickly developed to increase energy efficiency, save fuel, and reduce carbon dioxide emissions for pulse power and energy saving applications [6-9]. Tatarchuk et al. [10-12] utilized metal-carbon fiber composite structures for fabricating electrochemical double-layer capacitors. This fiber composite unique structure was successfully applied to fabricate the ultracapacitors and made an impressive progression on energy densities [7,12]. Chu and Braatz [13] initially characterized and compared the supercapacitors and high-power Li-ion batteries for power-assist applications in hybrid electric vehicles (HEVs). Egido et al. [14] developed a dynamic model for applying ultracapacitor to energy storage systems in order to enhance the frequency stability of the isolated power systems. Ma et al. [15] demonstrated that the hybrid energy storage stabilizes the energy provision for intermittent renewable energy and also alleviates the effect of electric load fluctuations through theoretical analysis and numerical simulation. Chia et al. [16] implemented an energy management system to optimize the energy flow and increase peak power stability by applying the supercapacitor-battery hybrid energy storage system to solar power application. For various hybrid electric vehicles, the ultracapacitor is also considered to work together with the battery energy storage system in order to reduce the battery stress for lifespan requirements [17]. Rambaldi et al. [18] conducted a preliminary analysis and evaluation on batteries plus ultracapacitors as an energy recovery system for HEV powertrain with the regenerative braking system. The ultracapacitor is capable of converting the electrical energy into the electrostatical charge storage inside the capacitor electrodes temporarily and then release this strong power with proper energy for heavy-duty electric vehicles [19], especially for those applications that need repeated stops or frequent speedchanges. Lam et al. [20,21] developed a hybrid valve-regulated ultrabattery to use as a hybrid energy-storage device. It combines an asymmetric ultracapacitor and a Pb-acid battery into a single cell. The capacitor enhances the power and lifespan of the Pb-acid battery as it acts as a buffer during discharge and charge. The lifetime of an energy storage system is predictably prolonged in addition to the ultracapacitor's rapid charge/discharge processes without complicated chemical reactions or significant heat

Various Li-ion batteries have gradually found their way into the hybrid and electric vehicles due to their high energy and power density, although it is necessary to further improve the cycle lifetime and develop more efficient thermal management technology. Jung et al. [22] developed a 42-V HEV ultracapacitor system and tested for 6-kW power boosting/regenerative braking. The thermal stability, charge/discharge efficiency, and cycle life characteristics are reliable except for the ultracapacitor's cost. Burke and Miller [23] discussed the power capability in details for ultracapacitors and lithium batteries for electric and hybrid vehicle applications. The ultracapacitors have high power density, superior cycle life, and cold temperature tolerance [24]. Hochgraf et al. [25] performed an hybrid energy-storage study by using the ultracapacitor and the Li-ion battery in order to reduce the performance degradation of the Li-ion battery pack. Tian et al. [26] conducted a nanosphere material study of nickel hydroxide for asymmetric capacitor and revealed a good cyclic ability. Huang et al. [27] further applied β-Ni(OH)₂ nanosheets onto nickel foam and formed a capacitor electrode with a high specific electric quantity of 790.3 C g⁻¹ approaching the theoretical value. Su and Zhitomirsky [28] synthesized conducting polypyrrole polymers coated with carbon nanotube as asymmetric supercapacitor electrodes. Its capacitor showed low impedance, high capacitance, and excellent cyclic stability. In recent years, researchers [29,30] also explored

the applications of Li-ion capacitors (LICs) assembled with prelithiated graphite anodes and active carbon cathodes. This type of ultracapacitor, as the Generation-I Li-ion capacitor, extended the operating potential among the range of 2.0-4.1 V, and significantly increased the energy density in comparison with the conventional EDLC capacitor in a voltage working window of 0.0-2.5 V. Naoi and Ishimoto et al. [31-33] pointed out additionally that Li⁺ insertion into Li₄Ti₅O₁₂ (LTO) compounds supported by carbon nanofibers in coupled with active carbon positive electrode can be viewed as the Generation-II nanohybrid capacitor (NHC). This nanocomposite structure (LTO/NCF), when used as the capacitor's negative electrode, provides a very fast Li+ insertion and enhances the capacitor's power capture and delivery performance. For better understanding of the ultracapacitor advantages and its hybrid-power device features, the presented work focuses on the concept of the ultracapacitors and various hybrid capacitors for efficient energy storage. This paper also performs electrical characterizations on a NiOOH/C ultracapacitor module, and the pulse power evaluation is then carried out on a hybrid power system using the asymmetric capacitor module and an advanced Pb-acid battery through both of modified cold cranking and hybrid pulse power load profiles.

2. Hybrid electrochemical capacitors for power capture and energy storage

For the power capture and energy storage in a hybrid power system, it is essential to understand the fundamentals of the capacitor power delivery and battery energy conversion as well as storage technologies (Fig. 1). The traditional capacitor has high power densities but low energy densities in comparison with the electric double-layer capacitor (EDLC). The battery is generally capable of storing more energy than a capacitor but with a relatively low power density. This means that a rechargeable battery cannot deliver energy very quickly but an electrochemical capacitor is capable of being discharged rapidly to generate a large amount of power. This is because the battery revolves in the electrochemical reactions. The reaction processes usually have different timeconsuming steps. Most oxidization and reduction processes in the battery electrodes generate heat and also have side-effects on the electrode structure and package materials. This limits the lifetime of the battery operating and cycling for the needs of energy storage and conversion. The ultracapacitor is mostly charged and discharged through transferring charges between the electrode and the electrolyte. This charge accumulation and electric distribution belongs to the physical processes. The very thin double layer and concentrated electrolyte assures that a low-level for the internal resistance for the capacitor. This allows the ultracapacitor to have high power output capability. Much less heat and very small chemical structure changes significantly increase the cycle lifetime during energy storage and power release. The concept of the hybrid power system combines the high power capability of the ultracapacitor and high energy storage ability of the advanced rechargeable battery.

3. Experimental approach

3.1. Asymmetric capacitor module and battery hybrid power device

The asymmetric capacitor module and its hybrid power system with the lead-acid battery was set up for testing and evaluation as shown in Fig. 2. The power module is an EC104 internal bank of 10 NiOOH/C hybrid capacitors connected in series by KAPower with 300 F capacitance. The internal resistance is originally marked as 0.006 Ohm. The capacitor module has a dimension of

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