

Making a High-Performance Dual Carbon Li-Ion Hybrid Capacitor Using a Mass-Balancing Method

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Introduction

Graphite or non-porous hard carbon is typically used in lithium-ion capacitor (LIC) devices as the negative electrode, which frequently fails under high energy demands and power densities. Here, we describe a novel LIC made of hollow carbon spheres (HCS) made of polymers and super activated carbon (AC), which serve as the negative and positive electrodes, respectively. Each of the electrodes exhibits extraordinary capacity values and rate performances as a result of the hollow microstructure of HCS and the extremely large specific surface area of AC. A thorough mass balancing research is also carried out to optimise the LIC's performance both in terms of energy and power densities as well as stability. In the operative voltage range of 1.5-4.2 V, optimised LIC with a 2:1 negative to positive electrode mass ratio has very good reversibility and can still offer a specific cell capacity of 28 mA/h even at a high current density of 10 A/g. At a maximum power density of 30 kW kg⁻¹, this results in an energy density of 68 W h kg⁻¹. Additionally, this LIC device has exceptional cyclability, holding more than 92% of the initial capacity after 35,000 charge-discharge cycles.

Discussion

Due to the rising energy demand from contemporary human activities, the hunt for more potent energy storage technologies has become more intense in recent years. In order to meet the demands of the rapidly expanding markets for electric vehicles, aerospace, and next-generation portable devices, both the scientific and industrial groups must create high power/energy sources [1]. Lithium-ion batteries (LIBs) and supercapacitors (SCs) are the preferred energy sources for high-energy or high-power applications, respectively, among the various energy storage technologies. The key benefits of LIBs over SCs include their higher energy density (200 W h kg⁻¹) and wider working potential window (vs. SCs' 10 W h kg⁻¹). Contrarily, SCs have a longer cycle life and can deliver substantially higher power densities (over 10⁶ cycles). In some applications that require both high-power and high-energy [2], LIBs and SCs that are produced industrially still have their limitations. Due to their ability to deliver high energy densities at high power, hybrid electrochemical capacitors (HECs), which combine a battery-type negative electrode with a capacitive positive electrode, have recently received enormous research and industrial attention. Up to this point, HECs based on various metal ions, such as Li⁺, Na⁺, or K⁺, have been proposed. Particularly, in lithium-ion capacitors (LICs), the intercalation/deintercalation of Li⁺ occurs at the anode side as in a LIB, and the adsorption/desorption of the counter ion (usually PF₆) occurs at the surface of the positive electrode as in an electrical double layer capacitor (EDLC) [3]. Different LIC systems, known as "Dual carbon LICs," have been described in

the literature. These systems combine a high surface area activated carbon as the positive electrode with a Li-ion intercalating carbon (graphite, hard carbons, or soft carbons) as the negative electrode. These systems demonstrate an energy storage capacity that is nearly five times greater than that of EDLCs while maintaining good response at high power demand and stability during long cycling. Due to the asymmetrical arrangement of the anode and cathode, the LIC devices suffer from a significantly reduced self-discharge, similar to Li-ion batteries. This is another advantage over regular EDLC capacitors. Hard carbons have demonstrated encouraging results for the negative electrode, even exceeding the theoretical capacity of graphite [4]. The usage of extra space for Li⁺ ion storage in the cavities and micropores as well as intercalation is made possible by their disordered structure, which contains graphite-like domains with a low degree of crystallinity. Recently, many nanostructured carbon materials, including carbon nanosheets, nanospheres, nanopipes, and nanofibers, have been studied as LIC anodes. In terms of structural stability, transport kinetics, cyclability, and coulombic efficiency, tuning the carbons' microstructure at the nanoscale resulted in notable advancements. Since they serve as electrolyte reservoirs and expedite Li⁺ intercalation/deintercalation processes through the thin carbon walls, the hollow carbon spheres' shape proves to be particularly advantageous. Additionally, due to its extensive interior area, the electrode's mechanical stability is improved by the volume variations that occur throughout the charge/discharge processes [5]. Due to their huge specific surface areas and open porosity, activated carbons are frequently used as positive electrodes because they facilitate quick ionic transport to the entire electrode surface. In a previous paper, we presented a unique and simple synthetic method for the production of activated carbons with extremely high specific surface areas. This simple one-step synthesis method produces carbons with specific surface areas just above 3000 m² g⁻¹ and a hierarchical micro-mesoporous structure. The carbon precursors are simultaneously polymerized, carbonised, and chemically activated. They make a viable alternative for the positive electrode material in LIC systems due to their convenient porous structure and simple manufacturing [6]. Optimizing the mass balance between the positive and negative electrodes can boost the performance of hybrid supercapacitors. In order to increase the device's energy density, varied mass balances translate into various working potential spans and, consequently, a variety in the extent to which each electrode is utilised. In actuality, stability and safety aren't given much consideration in scientific publications; instead, they tend to concentrate on the highest energy/power results.

Conclusion

A simple method has been used to create tiny hollow carbon spheres. This substance performs better during the lithium insertion-extraction process, particularly at very high current rates, making it a good option for use as the negative electrode in lithium-ion capacitors. By combining these micro-structured hollow carbon spheres with a superactivated micro-mesoporous carbon utilising various electrode mass ratios, LICs were created. Hollow carbon spheres can withstand volume changes during repeated lithiation - delithiation cycles, and superactivated carbon's hierarchical porosity, which has a very low barrier to ion diffusion, ensures a good reaction at high current rates. In this LIC system, it was discovered that a 2:1 negative/positive electrode mass ratio produces the highest gravimetric energy density of 117 Wh kg⁻¹ at 0.34 kW kg⁻¹ and still 68 Wh kg⁻¹ at the highest power density of 30 kW kg⁻¹. The LIC's outstanding long-term stability after 35000 cycles with only 8% of capacity loss observed served as confirmation of its reliability. Our

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suggested LIC is a promising energy storage system that stands out from its competitors thanks to this exceptional performance.

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