

## Feature Article

## Three-dimensional carbon architectures for electrochemical capacitors



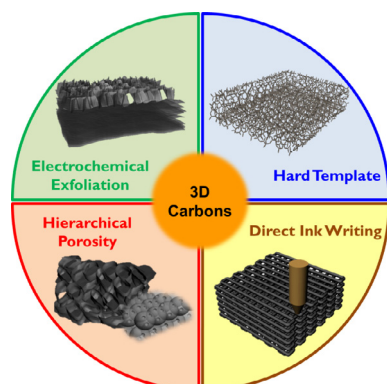
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## GRAPHICAL ABSTRACT



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## ABSTRACT

Three-dimensional (3D) carbon-based materials are emerging as promising electrode candidates for energy storage devices. In comparison to the 1D and 2D structures, 3D morphology offers new opportunities in rational design and synthesis of novel architectures tailor-made for promoting electrochemical performance. The capability of building hierarchical porous structures with 3D configuration can significantly advance the performance of energy storage devices by simultaneously enhancing the ion-accessible surface area and ion diffusion. This feature article presents an overview of recent progress in design, synthesis and implementation of 3D carbon-based materials as electrodes for electrochemical capacitors. Synthesis methodologies of four types of 3D carbon-based electrodes: 3D exfoliated carbon structures, 3D graphene scaffolds, 3D hierarchical porous carbon foams, as well as 3D architectures with periodic pores derived from direct ink writing, are thoroughly discussed and highlighted with selected experimental works. Finally, key opportunities and challenges in which different 3D carbons can significantly impact the energy storage and conversion communities will be provided.

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## 1. Introduction

Depletion of fossil fuels, proliferation of environmental pollution, and climate change are driving the need for shifting energy dependence to clean and renewable sources [1–3]. Practical utilization of sustainable energy requires reliable energy storage devices due to the intermittent nature and the heterogeneous geographical distribution of most sustainable energy sources (e.g., solar energy and wind energy). Therefore, development of energy storage technologies for sustainable energy implementation has received extensive and worldwide research attention [4–6].

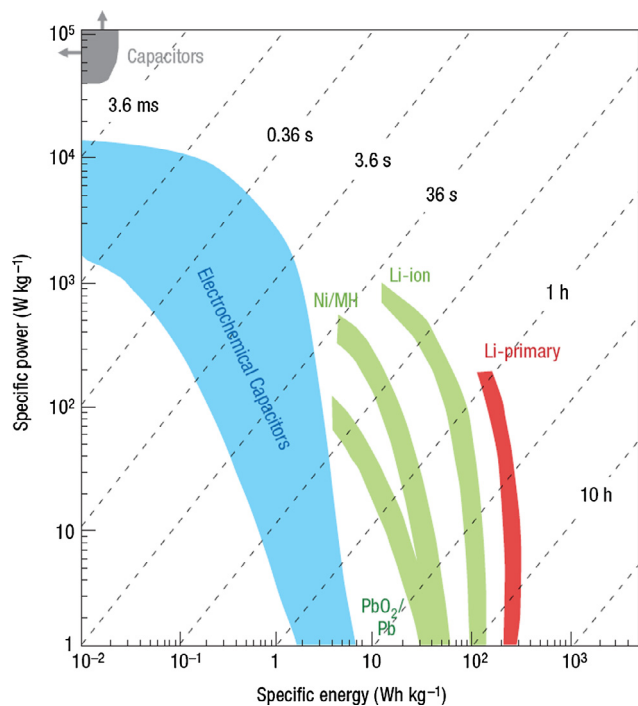
Electrochemical capacitors, *a.k.a.* supercapacitors or ultracapacitors, represent a class of energy storage devices that are capable of storing electrical energy converted from sustainable sources [4,7,8]. As shown in Fig. 1, electrochemical capacitors are distinguished from other energy storage devices by a number of unique

characteristics [2]: they can achieve much higher power densities than rechargeable batteries (*i.e.*, the “Pb/PbO<sub>2</sub>”, “Ni/MH”, “Li-ion” and “Li-primary” in Fig. 1), and they have higher energy densities compared to conventional dielectric capacitors (*i.e.*, the “capacitors” in Fig. 1). Electrochemical capacitors offer a promising solution to meet the increasing demand for rapid energy storage and delivery [9,10]. They have already been incorporated in some electric vehicles (e.g., cars and buses), electronics, and industrial power generation and back-up systems [1,2].

The charge storage mechanism of the electrochemical capacitors is classified into two categories, namely electrical double layer capacitance (EDLC) and pseudo-capacitance. EDLC is correlated to the electrostatic charge accumulation (during charging) or dissipation (during discharging) at the electrode/electrolyte interface. Pseudo-capacitance is associated with fast and reversible redox reactions (e.g., protonation/de-protonation and insertion/de-insertion that involves no phase transition) [2,4].

The performance of electrochemical capacitors largely depends on the structural and electronic properties of the electrode materials, including electrical conductivity, ion-accessible surface area, porosity, and the presence of heteroatoms. Carbon materials, owing to their excellent electrical conductivity, outstanding electrochemical stability, presence of multiple morphologies, and abundance have been extensively explored and implemented as electrodes for electrochemical capacitors [8,11–14]. The typical carbon material for electrochemical capacitors is activated carbon. However, activated carbon is not ideal for next-generation electrochemical capacitors due to the three main disadvantages: (1) its powder form requires the use of electrically insulating binders for electrode preparation, which increases overall device weight and retards electron transport; (2) micropores account about 90% of its porosity [15]. Such a large fraction of micropores impedes fast ion diffusion and limits the capacitance at ultrafast charging rates, because ion diffusion in micropores (especially with pore diameters much smaller than those of ions) is known to be sluggish [16]; (3) The uncontrolled pore distribution of the micropores leads to a tortuous and stochastic porous network that also adds ion diffusion resistance [17].

Currently, three-dimensional (3D) carbon materials are emerging as a promising alternative for electrochemical capacitors [18–21]. 3D architectures usually contain hierarchical pores (e.g., micropores, mesopores and macropores) and are free-standing. These characteristics generally impart the following advantages: (1) ultrahigh surface area for interacting with ions and improving charge storage capability; (2) free-standing nature with interconnected networks requires no insulating additives for electrode preparation; and (3) unique pore network (e.g., hierarchical porous



**Fig. 1.** Plots of gravimetric power density vs. gravimetric energy density (Ragone plot) of various energy storage devices. Values are normalized to the total weight of the device. Dotted lines represent the time needed to fully charge or discharge a device. Reproduced from Ref. [2] with permission, Copyright 2008, Nature Publishing Group.

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