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Development of supercapacitors with 3D porous structures

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The pursuit of supercapacitors with simultaneous high power and energy densities has led to extensive research and successful outcomes through the integration of three-dimensional (3D) electrodes, encompassing both 3D active materials and 3D porous current collectors. This mini review provides a summary of recent developments in supercapacitors featuring 3D structures. The incorporation of both 3D active materials and 3D current collectors proves effective in enhancing the mass loading of active materials without compromising their specific capacitance. The presence of pores in 3D porous current collectors contributes to additional double-layer capacitance by offering a large surface area. Moreover, 3D porous transition metal current collectors can provide both double-layer and faradic capacitances through the *in-situ* surface oxidation mechanism. Beyond materials and geometries, this review also discusses synthesis strategies for electrodes, offering insights into the process-structure-property relationship crucial for supercapacitors. By combining 3D active materials with 3D current collectors, the energy density of supercapacitors could be substantially improved.

1. Introduction

Over the past two decades, the modern intelligent society has witnessed the extensive development of various smart electronic devices, including wearable gadgets and drones. This surge in technological advancements has led to growing demands for reliable and high-performance energy storage devices.[1] Despite significant enhancements in battery performance through rigorous research and development, many batteries still fall short of meeting specific requirements for the next generation of energy storage devices, such as flexibility, safety, and high charging rates.

As an alternative and promising candidate with numerous advantages, supercapacitors have attracted increasing attention.[2] The rapid evolution of nanotechnologies has paved the way for the exploration of various supercapacitors with high power density and energy density. These include carbon-based supercapacitors utilizing a double-layer mechanism,[3] as well as metal oxides- and conducting polymer-based supercapacitors employing a faradic mechanism.[4] The carbon-based supercapacitors have demonstrated excellent properties owing to their high specific surface area and good electronic conductivity. However, they face the inherent limitation of limited energy densities due to their low theoretical specific capacitance of ~550 F/g.[5] On the other hand, transition metal oxides-based supercapacitors boast much larger theoretical capacitance than their carbon-based counterparts but suffer from low electronic conductivity. Nevertheless, nanostructured metal oxides, such as MoS_2 nanosheets,[6] Co_3O_4 nanowires,[7] MnO_2 nanoplates,[8] and $Ni(OH)_2$ nanosheets,[9] have exhibited remarkably high specific capacitances due to their very small dimensions, which shorten electronic and ionic transfer paths.

Despite their impressive performance, nanostructured active materials encounter the challenge of small mass loading on current collectors (usually less than 1 mg/cm^2), leading to low energy density in corresponding supercapacitors. To address this issue and enhance the energy density of metal oxides-based supercapacitors, the mainstream approach is to improve the mass loading of active materials without sacrificing their specific capacitances. The most effective method to tackle this challenge involves synthesizing electrodes with three-dimensional (3D) structures, incorporating both 3D active materials and 3D porous current collectors. This mini review will present recent developments in supercapacitors with 3D structures, covering active materials, current collectors, and synthesis strategies.

2. Active materials with 3D structure

A typical method to synthesize 3D nanomaterials is the sequential synthesis of the vertically aligned primary nanomaterial and the lateral growth of the secondary nanomaterial. Through a two-step nanofabrication process, nanomaterials with a dendrite-like structure could be synthesized. The vertically aligned nanomaterials are usually carbon nanotube array,[10] ZnO nanowire array,[11] Zn–Ni–Co oxide nanoneedle array,[12] and Co_3O_4 nanosheet array.[13] Thus, electrodes with 3D nanostructures such as carbon nanosheets on carbon nanotube array,[14] MnO_2 nanoflakes on carbon nanotube array,[15] MnO_2 nanosheets on ZnO nanowire array,[16] Co-MOF on Zn–Ni–Co oxide nanoneedle array,[12] $Ni(OH)_2$ nanosheets on Co_3O_4 nano-

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sheet array,[17] MnO_2 nanosheets on Co_3O_4 nanowire array[18] were developed. Electrodes with 3D active materials exhibited high specific capacitance, and the corresponding supercapacitors also showed elevated energy density due to the enlarged mass loading of active materials. The 3D Co_3O_4@$Ni(OH)_2$ electrodes showed a high specific capacitance of 1306 F/g with a high mass loading of 4.5 mg/cm^2, resulting in a high energy density of the corresponding supercapacitors (40 Wh/Kg) (Figure 1).[17]

3. 3D porous current collectors

Besides incorporating 3D active materials, enhancing the mass loading of active materials can be achieved more efficiently by utilizing 3D porous current collectors. Ni foam has been widely adopted as a current collector for supercapacitors,[19] demonstrating superior areal capacitance and energy density compared to planar-shaped collectors.[20] However, the limited surface area of Ni foam, dictated by millimeter-scale pore sizes, poses a challenge. To address this, 3D porous carbon current collectors were explored, using Ni foam as a sacrificial template. In this process, a graphene layer was initially deposited on the Ni foam surface through chemical vapor deposition. Subsequently, the Ni foam was removed via etching in concentrated HCl acid or $FeCl_3$ solution[21] (Figure 2), resulting in the fabrication of 3D porous graphene. This material then served as the current collector for depositing nanostructured active materials like MnO_2[21b,22] and $Ni(OH)_2$.[23] The electrodes with 3D porous graphene as the current collector exhibited increased active material loading, leading to a noticeable enhancement in the areal capacitance of the electrodes, e.g., the MnO_2 electrode with a high MnO_2 loading of 9.8 mg/cm^2 had an improved areal capacitance of 1.42 F/cm^2.

Another promising 3D porous current collector is 3D nanoporous gold (NPG), which is commonly fabricated by etching the gold-silver alloy. Lang et al.[24] synthesized and used NPG directly as the electrodes for supercapacitors (Figure 3). Due to the large surface area and high electronic conductivity, the pristine NPG supercapacitors showed high energy densities of 8.31 and 21.7 mWh/cm^3 using KOH and ionic liquid as the electrolytes, respectively. To further enhance the energy density of supercapacitors, Lang et al.,[25] Chen et al.,[26] Kim et al.,[27] Prabhin et al.,[28] Purkait et al.[29] deposited MnO_2, $Co(OH)_2$, $Ni(OH)_2$, Co_3O_4, reduced graphene oxide and polypyrrole, respectively, on the surface of NPG, in which the highest energy density could be up to 98.5 mWh/cm^3. Nevertheless, NPG-based supercapacitors face challenges such as elevated costs and constraints associated with the thickness of electrodes.

4. Alternative electrode synthesis method

In addition to the top-down strategy discussed in Section 2, an alternative approach for fabricating 3D porous electrodes has been recently developed in our lab – the bottom-up strategy. In contrast to the conventional sequence of initially synthesizing 3D porous current collectors followed by the deposition of active materials, this method involves the initial synthesis of nanostructured active materials, followed by the deposition of current collectors. Specifically, Ni was used as the current collector, deposited onto nanostructured active materials through heterogeneous electroless deposition. This process resulted in the simultaneous formation of Ni branches on the final product as shown in Figure 4. Hence, the resultant product exhibits a dendrite structure, allowing it to be compressed within a mold using hydrolytic force. This process forms sturdy nanoporous plates that can function directly as freestanding electrodes. This method is versatile and applicable to a range of nanomaterials. Consequently, our lab successfully synthesized nanoporous CNT@Ni[30] and Fe_3O_4@Ni[31] electrodes in succession. These 3D nanoporous electrodes offer several advantages, including a substantial surface area, incorporation of additional active materials through *in-situ* surface oxidation of the Ni current collector, excellent accommodation to volume changes

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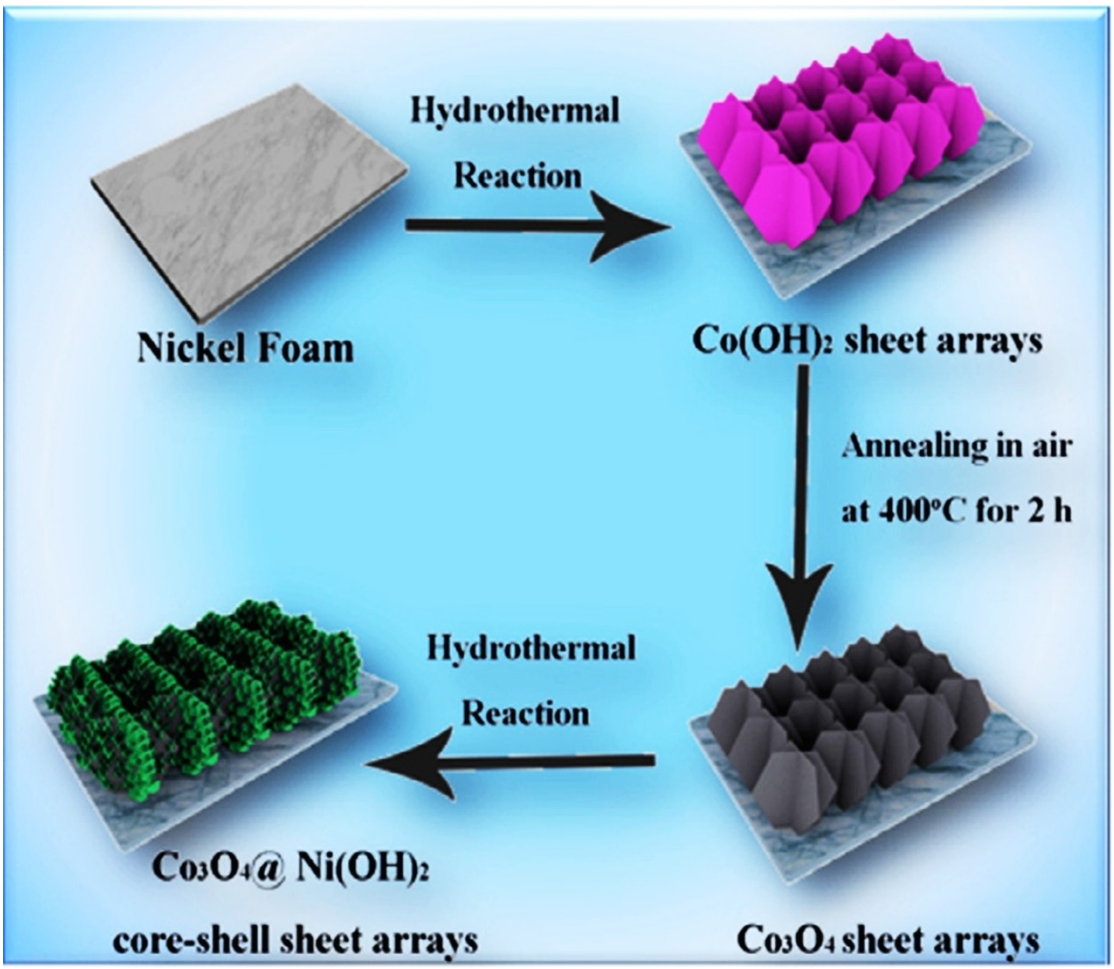

Figure 1. Schematic illustration of synthesis of Co_3O_4@Ni(OH)$_2$ hybrid core-shell nanostructure on a Ni foam.[17]

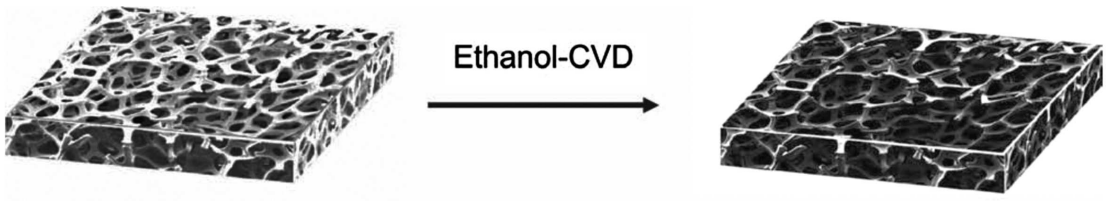

Figure 2. Schematic illustration of synthesis of 3D graphene networks on a Ni foam by ethanol-CVD.[21a]

in active materials owing to the structure of outer current collectors, and high scalability facilitated by the inverse synthesis process. As a result, the Fe_3O_4@Ni electrode showed a high areal capacitance of 82.6 F/cm^2 with the high Fe_3O_4 loading of 79.4 mg/cm^2, and the corresponding symmetric supercapacitor exhibited a high energy density of 94.4 mWh/cm^3. The Fe_3O_4@Ni symmetric supercapacitors also exhibited excellent cyclic capability and a high capacitance retention of 107% after 10,000 cycles. Moreover, the concept of bottom-up strategy can be employed to develop wire-like supercapacitors. Utilizing the "nano reinforced concrete" principle, nanomaterials and Ni can be co-deposited onto thin wires or threads, giving rise to a 3D macroporous structure. Thus, 3D porous Fe_3O_4-GO-Ni[32] and Cu@Ni-GO[33] electrodes were fabricated, and the corresponding wire-like supercapacitors exhibited a high energy density of 30.2 mWh/cm^3.

Table 1 summarizes electrodes with 3D structures, incorporating both 3D nanostructured active materials and 3D nanoporous current collectors. Since the volume of electrodes comprises active materials, current collectors, and internal pores, the capacitance of the total electrodes exhibits a linear relationship with the content of active materials. Enhancing the mass loading of active materials in the electrodes, without compromising their high specific capacitance, stands out as a highly effective approach to elevate both the capacitance of the electrodes and the energy densities of the corresponding supercapacitors. As outlined in Table 1, the adoption of nanostructured current collectors emerges as an efficient method for

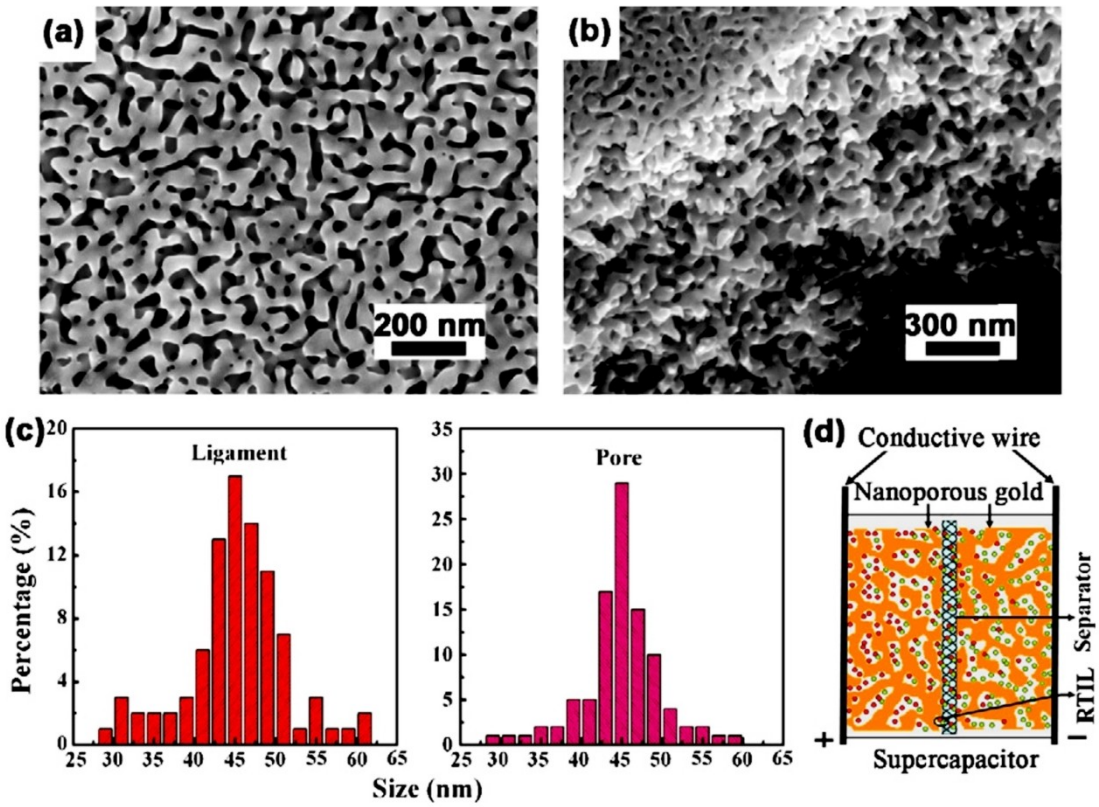

Figure 3. Representative (a) top view and (b) cross-sectional view of SEM images of a NPG electrode. (c) Size distributions of gold ligaments and nanoporous channels of the NPG sample. (d) Schematic diagram of the supercapacitor assembled with NPG as both electrodes and current collectors, and a piece of cotton paper as separator.[24]

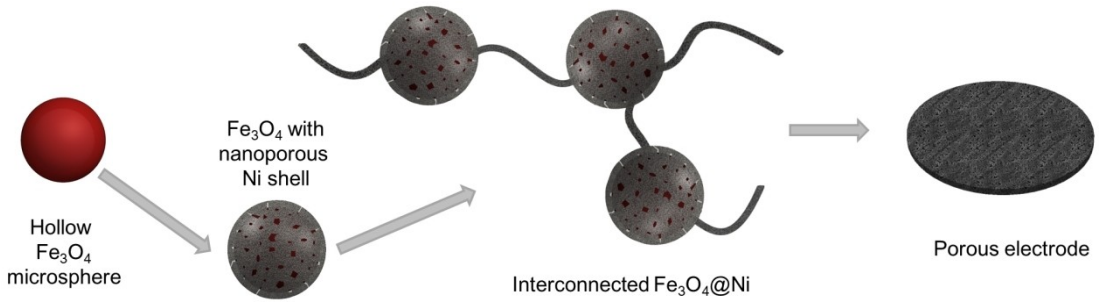

Figure 4. Schematic of the synthesis process of nanoporous electrodes through a bottom-up method.[31]

increasing the mass loading of active materials while maintaining their high specific capacitance. Further studies on nanoporous electrodes are crucial for advancing our understanding and refining the energy density, as well as other comprehensive properties, of supercapacitors.

5. Conclusion and Outlook

The enhancement of supercapacitor energy density has been achieved by incorporating electrodes with 3D structures, encompassing both 3D nanostructured active materials and 3D nanoporous current collectors. These components feature large surface areas and open spaces, facilitating efficient ionic and electronic transfer within the electrodes. The utilization of electrodes with 3D nanostructured active materials and nanoporous current collectors results in elevated energy densities through various mechanisms. These include increased mass loading of active materials, additional double-layer capacitance due to the extensive surface area of current collectors, and additional faradic capacitance arising from the *in-situ* surface oxidation of current collectors.

The fabrication methods for 3D nanostructured active materials involve a combination of vertical growth for primary active materials and lateral growth for secondary active materials. Meanwhile, 3D porous current collectors are produced using either a top-down or bottom-up strategy. In the top-down approach, the etching method is commonly em-

Table 1. Electrodes with 3D structures for supercapacitors.

3D structure	Electrode	Specific capacitance	Capacitance of electrodes	Mass loading of active materials	Reference
3D active materials	Carbon nanosheets on carbon nanotube array	N/A	110 mF/cm^2	N/A	[14]
	MnO_2 nanoflakes on carbon nanotube array	235 F/g	N/A	N/A	[15]
	MnO_2 nanosheets on ZnO nanowire array	654 F/g	N/A	N/A	[16]
	Co-MOF on Zn-Ni-Co oxide nanoneedle array	2,866 F/g	N/A	N/A	[12]
	$Ni(OH)_2$ nanosheets on Co_3O_4 nanosheet array	1,306 F/g	N/A	4.5 mg/cm^2	[17]
	MnO_2 nanosheets on Co_3O_4 nanowire array	480 F/g	N/A	2.3 mg/cm^2	[18]
3D graphene network	NiO on 3D graphene	816 F/g			[21a]
	MnO on 3D graphene		1.42 F/cm^2	9.8 mg/cm^2	[21b]
	$Ni(OH)_2$ on 3D graphite		255 F/cm^3		[23]
Nanoporous gold (NPG)	NPG		24 F/cm^3	N/A	[24]
	MnO_2 on NPG	1,145 F/g	1,160 F/cm^3	1.013 g/cm^3	[25]
	$Co(OH)_2$ on NPG	1,800 F/g	N/A	N/A	[26]
	$Ni(OH)_2$ on NPG	3,168 F/g	2,223 F/cm^3	N/A	[27]
	Co_3O_4 on NPG	1,215 F/g	N/A	N/A	[28]
Nanoporous nickel	CNT@Ni	1,765 F/g	1,232 F/cm^3	N/A	[30]
	Fe_3O_4@Ni	1,734 F/g	1,299 F/cm^3	79.4 mg/cm^2	[31]

ployed, leading to the development of 3D porous graphene and nanoporous gold (NPG). Sacrificial templates such as nickel foam and silver-gold alloy contribute to this process. On the other hand, the bottom-up strategy involves the development of a heterogeneous electroless deposition method, resulting in the synthesis of nano dendrites. These nano dendrites are then utilized to fabricate nanoporous electrodes through hydrolytic force.

Nanoporous electrodes generated through the bottom-up strategy exhibit a unique structure where active materials are enveloped within the current collectors. This configuration positively impacts the electrodes by enhancing the contact between active materials and current collectors, as well as accommodating the volume change of active materials. Consequently, this leads to improved rate capability and cyclic capability of the electrodes.

Supercapacitors typically employ active materials with high theoretical capacitance, allowing the active material content in 3D nanoporous electrodes to be increased to over 50%. Through meticulous calculation, it is possible to achieve energy densities in these supercapacitors that surpass those of conventional batteries. Several crucial parameters, including the structure of active materials, current collectors, and the interactions among active materials, current collectors, and electrolytes, significantly influence the electrochemical performance of nanoporous electrodes. Ongoing research in our lab focuses on optimizing these parameters to enhance the energy density of supercapacitors. The aim is to surpass current limitations and pave the way for 3D porous supercapacitors with heightened energy densities, positioning them as the next-generation energy storage devices applicable in various fields such as electric vehicles and wearable electronics. Achieving this goal requires a systematic exploration of comprehensive supercapacitor properties, including cost, current leakage, and aging characteristics, in future studies.

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Conflict of Interests

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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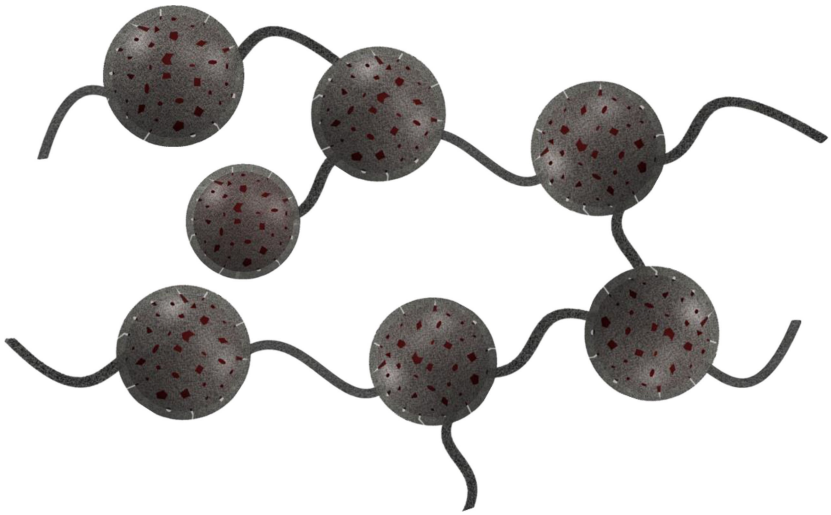

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3D nanoporous electrodes with fully wrapped encapsulation of active materials, interconnected current collectors and the integration of porous channels, facilitates ultrahigh efficiency in both electronic and ionic transfer within electrodes and electrolytes. The resulting nanoporous electrodes exhibit a remarkable combination of high specific capacitance and substantial mass loading of active materials. The 3D supercapacitors crafted from these electrodes demonstrate an ultrahigh energy density, showcasing the potential for advanced energy storage applications.