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Authors: Lili Liu, Zhiqiang Niu, Jun Chen

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Review

# Flexible supercapacitors based on carbon nanotubes

Lili Liu[a,*], Zhiqiang Niu[b], Jun Chen[b]

[a]*Tianjin Key Laboratory for Photoelectric Materials and Devices, National Demonstration Center for Experimental Function Materials Education, School of Materials Science and Engineering, Tianjin University of Technology, Tianjin 300384, China*
[b]*Key Laboratory of Advanced Energy Materials Chemistry (Ministry of Education), College of Chemistry, Nankai University, Tianjin 300071, China*

## Graphical abstract


This review provides an overview of recent progress towards the development of flexible supercapacitors based on macroscopic carbon nanotubes-based electrodes, including one dimensional (1D) fibers, 2D films, and 3D foams, with a focus on electrode preparation and configuration design as well as their integration with other multifunctional devices.

ABSTRACT
Since carbon nanotubes (CNTs) possess unique one dimensional (1D) structure, considerable attention has been paid to constructing CNTs into macroscopic materials with different dimensions, including 1D fibers, 2D films, and 3D foams. Such macroscopic CNT materials exhibit high conductivity, large surface area, as well as good mechanical properties, and thus can be directly used as the flexible supercapacitor (SC) electrodes or the scaffolds for supporting *pseudo*-capacitive electrode materials. Based on these macroscopic CNT electrodes, diverse SCs with different structures, including flexible, stretchable and/or compressible fiber and thin film SCs, have been designed. This review provides an overview of recent progress towards the development of flexible SCs based on macroscopic CNTs-based electrodes, with a focus on electrode preparation and configuration design as well as their integration with other multifunctional devices. Future development and prospects in the CNTs-based flexible SCs are also discussed.

*Keywords:*
Carbon nanotubes
Flexible supercapacitors
Macroscopic materials
*Pseudo*-capacitive materials
Integration

**1. Introduction**

Recently, there is an increasing demand for flexible energy storage systems due to the fast-growing market in portable and wearable electronics, such as roll-up displays and electric papers [1-3]. Among various energy storage devices, supercapacitors (SCs) are attracting much attention because they have high power density, long cycle lifetime, moderate energy density and wide working temperature range [4-16]. Conventional SCs usually include four main components: Current collectors, electrodes, separator, and electrolyte. In general, the separator and electrolyte are usually flexible. However, conventional SC electrodes are often fabricated by mixing active materials with conductive binders and then coating such composites onto metallic current collectors. As a result, these SC electrodes possess limited flexibility and cannot meet the demand of flexible SCs. In addition, the utilization of metallic current collectors would result in heavy configurations and low gravimetric capacitances [5,17-25]. Therefore, the fabrication of flexible electrodes and the simplification of device configurations hold the key to the design of flexible SCs.

As a kind of typical nanocarbon material, carbon nanotubes (CNTs) have a cylindrical structure with a nanometer-scale diameter. CNTs are categorized as single-walled nanotubes (SWCNTs) and multi-walled nanotubes (MWCNTs). The MWCNTs have one or more outer tubes successively enveloping a SWCNT. The unique one dimensional (1D) nanostructure endows individual CNTs with superior physical properties, such as a high thermal conductivity of 3,500 W $m^{-1}$ $K^{-1}$, charge mobility of 10,000 $cm^2$ $V^{-1}$ $s^{-1}$, Young's

* Corresponding author.
*E-mail address*: lililiuhappy@163.com (L. Liu).