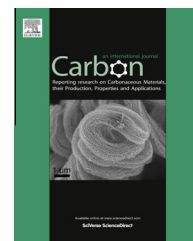


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# Carbon nanotube–nanopipe composite vertical arrays for enhanced electrochemical capacitance

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## ABSTRACT

The newly-designed vertical array structure of carbon nanotubes (CNTs) surrounding carbon nanopipes (CNTs@CNPs) was fabricated, which includes the formation of the primary CNP (ca. 200 nm in diameter) arrays in virtue of the anodic aluminum oxide (AAO) membrane template with infiltration of polymer solution containing metal catalyst precursor, followed by carbonization and CVD growth of the secondary CNTs (20 nm in diameter). The secondary CNTs with controllable density/length are grown both inside and outside the primary CNPs, which not only induce the four times increase of the specific surface areas for the samples, but also largely enhance the interconnection between the vertical CNPs for good electric conductance. The unique CNTs@CNPs arrays significantly improve the electrochemical capacitance per unit area with great long-term stability, indicating the promise as new material platform for development of advanced energy-storage devices.

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## 1. Introduction

Since carbon nanotubes (CNTs) were discovered in 1991 [1], they have been explored for many important applications due to their excellent electrical [2,3], mechanical [4], and optical [5] properties. Among them, vertically-aligned carbon nanotubes (VA-CNTs) have attracted considerable attention of scientists for years due to their unique applications in flat panel displays [6], sensors [7], actuators [8,9], electronic devices [10], smart polymer composites [11,12] and even advanced dry adhesives [13]. Particularly, VA-CNTs provide a well-defined large surface area and are readily incorporated into various device configurations. For certain applications, effort has been made to not only control the structure of individual nanotubes but also their packing state to enhance their performance. In this regard, we have developed a simple multiple contact transfer technique for controllable

fabrication of multilevel, multicomponent microarchitectures of VA-CNTs. Three dimensional (3-D) multi-component micro-patterns of aligned single-walled carbon nanotube (SWCNTs) and multi-walled carbon nanotubes (MWCNTs) have been fabricated [14]. Similarly, we have also managed and tailored the surface morphologies and related surface properties of CNT arrays by selectively removing VA-CNTs from their growth substrate through an extracting approach [15].

CNTs have become an important class of electrode material for various electrochemical devices, including electrochemical capacitors [16,17]. Due to their large mesoporosity and high electrolyte accessibility, it is well-accepted that CNTs are a good choice as high power electrode materials. However, the major limitation is their relatively small specific surface area as compared to activated carbon, which restricts their usage in high energy electric double layer capacitors

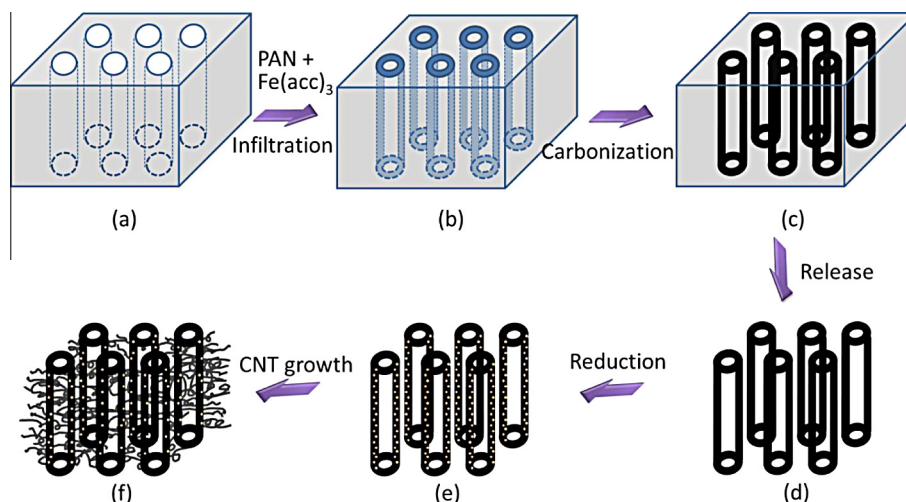
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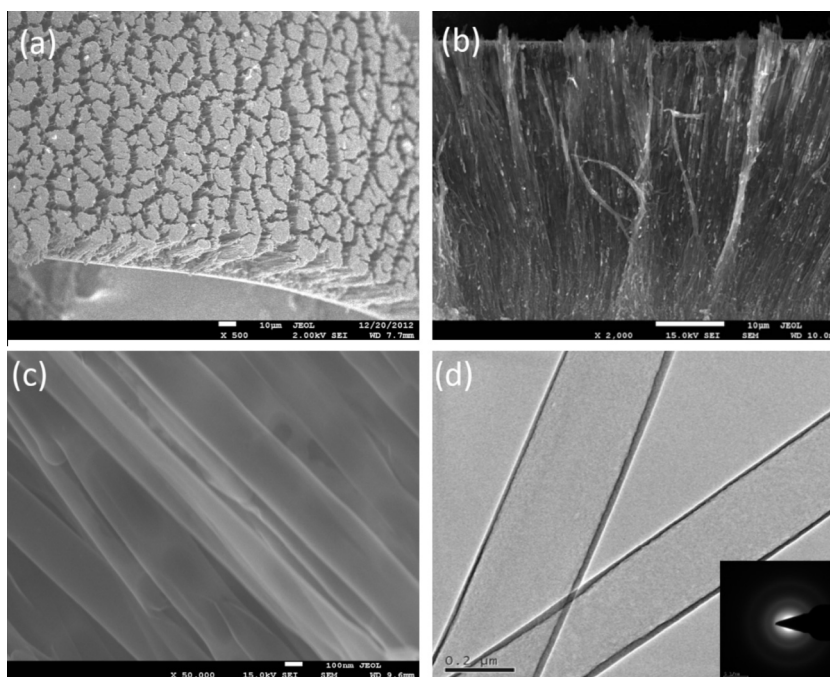
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**Fig. 1** – Schematical illustration of the preparation process for CNTs@CNPs. (a) AAO template, (b) PAN and Fe(acc)<sub>3</sub> infiltrated AAO, (c) Carbonization treatment, (d) Release of the primary CNPs from the AAO template, (e) Reduction by H<sub>2</sub> to form the Fe catalysts within the CNPs, (f) CNT growth by CVD.

(EDLCs). Apart from the surface modification of CNTs with other functional components such as conducting polymer [18] and metal oxide [19], one direct pathway to enhance the capacitance of CNT samples is to densify the CNT arrays for increasing the available surface area within a certain area/volume unit [20]. For this purpose, branching CNTs [21–24] are the good choice since the free space between CNTs could be occupied by CNTs and more importantly, new connections among CNTs would be established through the branched structures, which could thus further improve the electrochemical performance of CNT samples.

Following our recent achievements on high-performance supercapacitors [25–27], including the coaxial composites of polypyrrole and graphene-quantum-dots modified CNT arrays. Herein, we design and fabricate a new array structure of CNTs surrounding CNPs, which we denote as CNTs@CNPs. The primary CNP arrays are prepared by carbonization of polymer nanotubes formed within anodic aluminum oxide (AAO) template, in which the iron precursor has been incorporated in advance. Subsequently, chemical vapor disposition (CVD) is applied for the direct growth of the branched CNTs surrounding the individual CNPs within the arrays. The



**Fig. 2** – SEM (a–c) and TEM (d) images of the primary CNP arrays formed within the AAO templates. The inset in (d) is the corresponding electron diffraction pattern.

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