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Aligned carbon nanotube/zinc oxide nanowire hybrids as high performance electrodes for supercapacitor applications

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Carbon nanotube/metal oxide based hybrids are envisioned as high performance electrochemical energy storage electrodes since these systems can provide improved performances utilizing an electric double layer coupled with fast faradaic pseudocapacitive charge storage mechanisms. In this work, we show that high performance supercapacitor electrodes with a specific capacitance of $\sim$192 F/g along with a maximum energy density of $\sim$3.8 W h/kg and a power density of $\sim$28 kW/kg can be achieved by synthesizing zinc oxide nanowires (ZnO NWs) directly on top of aligned multi-walled carbon nanotubes (MWCNTs). In comparison to pristine MWCNTs, these constitute a 12-fold of increase in specific capacitance as well as corresponding power and energy density values. These electrodes also possess high cycling stability and were able to retain $\sim$99% of their specific capacitance value over 2000 charging discharging cycles. These findings indicate potential use of a MWCNT/ZnO NW hybrid material for future electrochemical energy storage applications. *Published by AIP Publishing.* [http://dx.doi.org/10.1063/1.4979098]

I. INTRODUCTION

High performance, cost-effective, and environmentally friendly renewable energy storage devices are crucial in order to mitigate the present energy challenge.[1–4] Among various energy storage devices, electrochemical capacitors (ECs) or supercapacitors (SCs) are considered as one of the suitable options in order to develop energy efficient, robust, sustainable energy storage systems.[5,6] This is largely due to the fact that SCs exhibit longer life cycles and faster charge/discharge capability than conventional batteries.[7] In general, SC electrodes can be categorized into two groups, primarily based on charge storage mechanisms. These include (1) electrical double-layer capacitors (EDLCs), which employ the ion adsorption/desorption mechanism and typically use carbon-based materials as electrodes and (2) pseudocapacitors (PCs), which undergo rapid surface redox reactions that mainly involve transition metal oxides or electrically conductive polymers as active electrode materials.[8] Carbon-based materials, such as multi-walled carbon nanotubes (MWCNTs), have been actively used as electrodes to fabricate EDLCs because these materials possess higher conductivity, higher chemical stability, larger specific surface area, and are lightweight.[9–11] Most of these properties are attributed due to the high aspect ratio of the MWCNTs and their nanoscopic sizes. The general advantages of the effects of nanostructuring the electrodes in electrochemical applications are well known.[12] However, using only carbon-based materials without much structural or chemical modification can restrict the SC performance.[13] In order to avoid this, researchers in the past have proposed several ways to incorporate various other nanostructures in order to synthesize carbon-based hybrid materials[14,15] that will have improved functionalities. In recent years, rational design of SC electrodes based on hybrids of carbon nanotubes and transition metal oxide has received significant attention since these systems can utilize a combined charge storage phenomenon of electrochemical double layer as well as faradaic pseudocapacitive mechanisms.[16–20]

Several hierarchical and composite structures based on MWCNTs and transition metal oxides supercapacitor electrodes have been widely explored in order to impart significant component redox reaction in them. For example, various transition metal oxides, such as RuO_2,[16] In_2O_3,[17] V_2O_5,[18] ZnO,[19] and NiO,[20] have been successfully used to enhance the capacitive performance of MWCNTs by a few orders of magnitude higher compared to those of typical MWCNTs. Among these transition metal oxides, ZnO based materials have been utilized in many applications, such as light-emitting materials,[21,22] UV-sensors,[23] gas sensors,[24,25] solar cells,[26] and are also shown to have excellent performance as active battery-electrode materials with a high energy density of 650 A/g.[27] Several investigations also indicate the possibility of using composites of carbon based materials and ZnO for energy