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Sulfur-doped carbon nanotubes as a conducting agent in supercapacitor electrodes

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The electrochemical performance of sulfur-doped carbon nanotubes (S-CNTs) was investigated to confirm the S-doping effects and the possibility of their application as conducting agents in supercapacitor electrodes. S-CNTs were successfully synthesized via chemical vapor deposition using dimethyl disulfide as the carbon source. They were purified to obtain purified S-CNTs (P–S-CNTs) with diameters 30–50 nm and S content of 0.65 at%. The doped S atoms were removed partially from the P–S-CNTs by heat treatment in H_2 atmosphere (De-P-S-CNTs). To compare the electrochemical performances of various conducting materials for supercapacitor electrodes, commercial activated carbon (MSP20) was used as the active material and commercial conducting agent (Super-P), commercial multi-walled CNTs (MWCNTs), De-P-S-CNTs, and P–S-CNTs were used as the conducting agents. The electrode with P–S-CNTs exhibited the highest specific capacitance at a high discharge current density of 100 mA cm^{-2} (120.2 F g^{-1}) and the lowest charge-transfer resistance (6.19 Ω) that are significantly superior to those of Super-P (83.9 F g^{-1} and 15.16 Ω), MWCNTs (87.8 F g^{-1} and 17.02 Ω), and De-P-S-CNTs (90.1 F g^{-1} and 22.33 Ω). The superior electrochemical performance of P–S-CNTs can be attributed to the excellent electrical conductivity and pseudocapacitive contribution of the S-doping effect.

1. Introduction

Carbon nanomaterials have been actively studied and applied in various fields such as energy storage, catalysis, chemical sensing, and electronics [1–3]. Among the carbon nanomaterials, carbon nanotubes (CNTs) exhibit great potential for applications in energy-storage fields, owing to their excellent physical, chemical, and electrical properties. Many techniques such as arc discharge, laser ablation, and chemical vapor deposition (CVD) have been developed for CNT synthesis. The CVD synthesis method, in particular, has helped realize the mass production of commercial CNTs [4–6]. Various hydrocarbons such as methane, acetylene, alcohol, ethylene, and toluene are generally used as carbon sources in the CVD process for CNT synthesis [7–13]. Doping carbon nanomaterials is a practical and powerful approach to improve their properties [14,15]. Heteroatom-doped CNTs, in particular, have been studied in many fields such as biotechnology, catalysis, optics, photovoltaics, supercapacitors, and energy storage [16–21]. Among various heteroatoms that can be doped into CNTs, S is a rare dopant that has been studied relatively less than N and B [22–24]. Du et al. reported the synthesis of S-doped CNTs (S-CNTs) by the pyrolysis of S-containing thiophene as a carbon source under optimum synthesis conditions (thiophene concentration of 0.76–1% at 1000 °C) [25]. Zhou et al. reported the synthesis of S-CNTs with an S content of 5.55–7.03% using linear dimethyl sulfide as a carbon source [26]. The electrochemical study of S-CNT supercapacitor electrodes revealed specific capacitance values of 2.07 F g^{-1} and 1.87 F g^{-1} for 1.0 M H_2SO_4 and 0.5 M Na_2SO_4 electrolytes,

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