

Full Length Article

Structural and chemical synergistic effect of NiCo₂S₄ nanoparticles and carbon cloth for high performance binder-free asymmetric supercapacitors

Xiaotong Xu^{a,b}, Xiaodong Tian^a, Xiao Li^{a,b}, Tao Yang^{a,b}, Yiting He^{a,b}, Kai Wang^c, Yan Song^{a,*}, Zhanjun Liu^a

^a CAS Key Lab. of Carbon Materials, Institute of Coal Chemistry, Chinese Academy of Sciences, Taiyuan 030001, China

^b University of Chinese Academy of Sciences, Beijing 100049, China

^c Institute of Science and Technology, Shanxi Coal Import and Export Group Co., LTD, 115 Changfeng Street, Taiyuan 030006, PR China

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ABSTRACT

A facial solvothermal method is used to synthesize NiCo₂S₄ nanoparticles on carbon cloth (CC) to investigate the synergistic effect of transition metal sulfide and carbon substrates on the electrochemical performance in supercapacitors. The results demonstrate that the integrated binder-free NiCo₂S₄/CC electrode achieved a specific capacitance of 1379 F g⁻¹ at 1 A g⁻¹, and high rate capability of 82% when the current density increase to 50 A g⁻¹, which is 13% and 35% higher than that of pristine NiCo₂S₄ counterpart. Detailed electrochemical kinetic analysis reveals that the proportion of surface capacitive effect of the hybrid electrode is higher than that of NiCo₂S₄ electrode at various scan rates due to the introduction of CC. CC substrate can provide a highway for electrolyte ions and electrons, facilitate the reaction kinetics, and shorten the diffusion distances for redox reaction, leading to improved performance of pseudocapacitive NiCo₂S₄. In addition, the as-fabricated asymmetric supercapacitors (ASC) based on NiCo₂S₄/CC//CNF electrodes demonstrate a high energy density of 41.28 W h kg⁻¹ at a power density of 1564 W kg⁻¹, along with long-term cycling performance and high rate stability.

1. Introduction

Portable electronic devices such as flexible sensors, flexible displays or hand-held devices are growing very fast, which have propelled the research for high performance power sources [1]. Due to the attractive power density, superior lifespan, fast charge/discharge rate and excellent reversibility, supercapacitors (SCs) are envisioned to be a promising energy storage device to bridge the gap between traditional dielectric capacitor and batteries. Based on the charge storage mode, SCs can be categorized into two types. One is the electrical double-layer capacitors (EDLCs) whose capacitance is electrostatic arising from charge separation at the interface of electrolyte and electrode [2]. The other is the pseudocapacitance whose capacitance stems from a rapid and reversible redox reaction at the near surface of redox-active materials [3,4]. However, the relatively low energy density of the state-of-the-art EDLCs and poor cycle life of the pseudocapacitors exclude them from wide application. Under such circumstances, it is of great importance to exploit novel electrode materials and develop asymmetric SCs (ASCs) to further fulfill the enhancement of energy density and the cycle life [5].

Recently, binary transition metal sulfides, especially Ni-Co sulfides have been commonly reported as promising pseudocapacitive materials. On one hand, because of the lower electronegativity of S than O, the materials exhibit extra structural flexibility after surface sulfuration which is beneficial to higher stability than metal oxides/hydroxides [6,7]. The smaller band gap also lead to higher conductivity [8]. On the other hand, compared with single component sulfides, binary Ni-Co sulfides (e.g. NiCo₂S₄) display the synergetic effects from both Ni and Co species due to multiple valence transitions, resulting in higher specific capacitance [9,10]. All features mentioned above endow them a great opportunity for next generation electrodes. Nowadays, various NiCo₂S₄ structure, such as nanosheets, nanotubes, nanoparticles, urchin-like structures, nanoellipsoids, nanoneedle, nanosphere-like and nanoflowers, etc. have been synthesized [11–16]. Hou et al. [17] reported hollow mesoporous NiCo₂S₄ ellipsoids with superior capacitance and more flexible structure than its NiCo₂O₄ counterpart, due to the relatively lower electronegativity of S. Zhang et al. [18] prepared four different morphologies NiCo₂S₄ by employing various solvents. Unfortunately, even though tube-like NiCo₂S₄ exhibits the best electrochemical performance than others, the high rate performance is still

* Corresponding author.

E-mail address: yansong1026@126.com (Y. Song).

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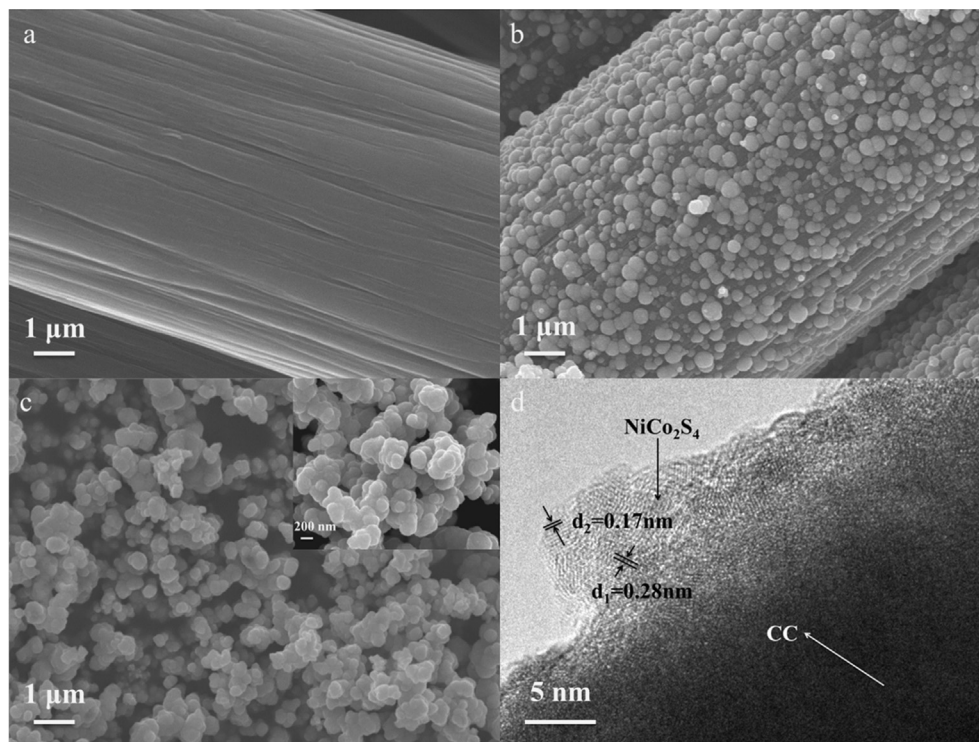


Fig. 1. SEM images of (a) bare CC; (b) NiCo_2S_4 loaded on carbon cloth after the sulfuration process; (c) the NiCo_2S_4 samples at different magnifications; (d) HRTEM images of the $\text{NiCo}_2\text{S}_4/\text{CC}$ sample.

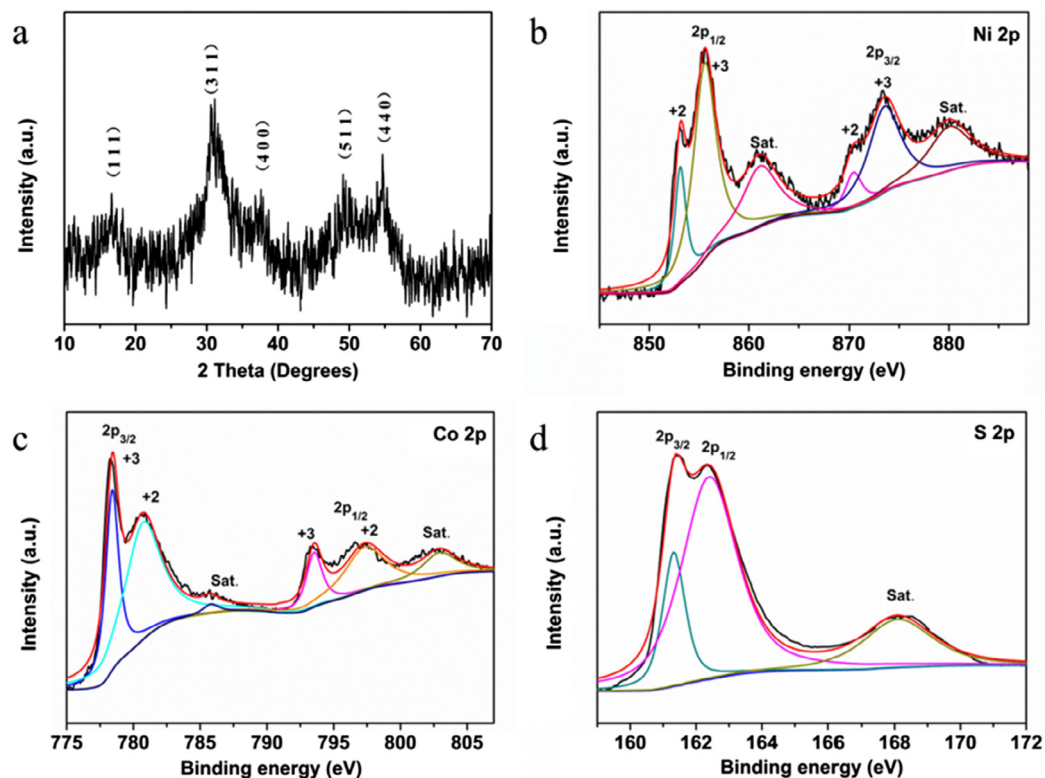


Fig. 2. (a) XRD pattern for the NiCo_2S_4 separated from the carbon cloth substrate; XPS spectra of (b–d) Ni 2p, Co 2p, S 2p.

unsatisfactory.

Therefore, due to the slow reaction kinetics and the high dependence on Faradic redox reactions, NiCo_2S_4 still suffers from low rate capability and poor stability during long term charge/discharge processes [19,20]. A popular approach to solve the problems is combining

with conductive materials. In this regard, nickel foam has been used to construct integrated free-standing electrodes aiming to shorten electron/ion diffusion path, enhance the utilization of electroactive atoms simultaneously [21,22]. Although the electrochemical performance was enhanced, the influence of impurity on the total capacitance cannot be

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