IJARSCT

ISSN (Online) 2581-9429

International Journal of Advanced Research in Science, Communication and Technology (IJARSCT)

Impact Factor: 6.252

Volume 2, Issue 2, March 2022

A Review on the Synthesis of CuCo2O4 Based Electrode Material and their Application in Supercapacitors

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Abstract: *Supercapacitors joined of the most promising energy storage systems are extensively studied due to their unique merits, like long-term cycling stability, fast charge rate, and low maintenance cost. it's widely known that the electrochemical performances of supercapacitors are closely associated with the structure and specific extent of the electrode materials. Therefore, many sorts of research are focused on the planning and synthesis of electrode materials with novel shapes and huge surface areas. CuCo2O4 has recently attracted enormous research interest because of the electrode materials for supercapacitors as a result of its inherent advantages including high theoretical capacity, environmental friendliness, natural abundance, and low cost. In practical applications, the CuCo2O4 still suffers from some drawbacks; for example, poor conductivity, relatively low specific capacity, and poor cycling durability. Hence, a comprehensive summary of the recent progress of CuCo2O4-based materials is critical and significant to higher understand the opportunities and challenges that such materials face. during this work, the progress of preparation methods and electrochemical performances of CuCo2O4-based materials is comprehensively reviewed. The aim of this review is to focus on a number of the advances made by CuCo2O4-based electrode materials for supercapacitors and guide future research toward closing the gap between achieved and theoretical capacity, without limiting the loading mass.*

Keywords: CuCo2O4; Composites; Porous materials; Electrochemical performance; Supercapacitors

I. Introduction

Supercapacitors, also called ultracapacitors or electrochemical capacitors, will be divided into three 4 categories of symmetric supercapacitors, asymmetric supercapacitors, and hybrid supercapacitors based on the device architecture [9]. per the charge storage mechanism of electrode materials, supercapacitors may also be classified into three types including electrical double-layer capacitors (EDLCs), pseudo-capacitors (PCs), and battery-like supercapacitors [10, 11]. The EDLCs store charges mainly through the adsorption and desorption of ions, which occur within the electric double layer formed at the interface between electrode and electrolyte [12]. the whole process is a purely natural action with none occurrence of chemical reactions,which Makes EDLCs possess a high power density. Transition metal oxides (TMOs) -based-materials TMOs like NiO [20], NiCo2O4 [21], Co3O4 5 [22], CuCo2O4 [23], Fe2O3 [24], NiFe2O4 [25], are widely studied as battery-type electrode materials thanks to their multiple valance states for the rich redox reactions. Among many TMOs-based electrode materials with battery-like behaviors, CuCo2O4, a sort of spinel cobaltite, is attractive in Li-ion batteries [26, 27], supercapacitors [28], sensors [29-31], and catalysts [32, 33] because of its inherent advantages like natural abundance, environmentally benign, low cost, and excellent electrochemical properties. During this work, progress associated with the methods for the synthesis of CuCo2O4-based electrode materials, fabrication of pure CuCo2O4 electrode materials, and formation of CuCo2O4-based composites is comprehensively reviewed and discussed thoroughly. The aim of this review is to focus on a number of the advances made by CuCo2O4-based electrode materials on supercapacitors and guide future research direction to shut the gap between the achieved and theoretical capacity of this kind of electrode material.

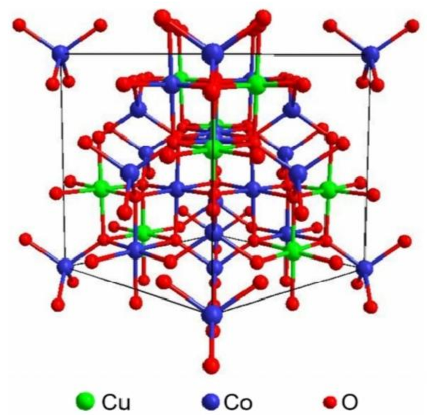


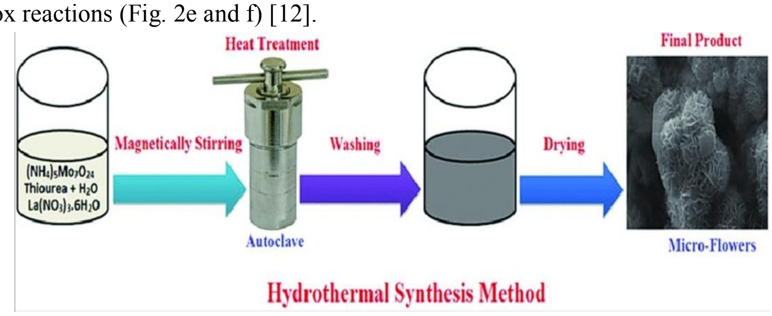
Figure: The crystal structure of $CuCo_2O_4$.

II. Methods for the Synthesis of $CuCo_2O_4$-Based Electrode Materials-

The performance of the supercapacitor is closely related to the electrode material. Advanced $CuCo_2O_4$ electrode materials with unique structure and superior performance are still desired by scientists and engineers. So far, many methods such as hydrothermal, solvothermal, electro-deposition, template-assisted, electro-spinning, and so on, have been used to prepare $CuCo_2O_4$ electrode materials with various structures .

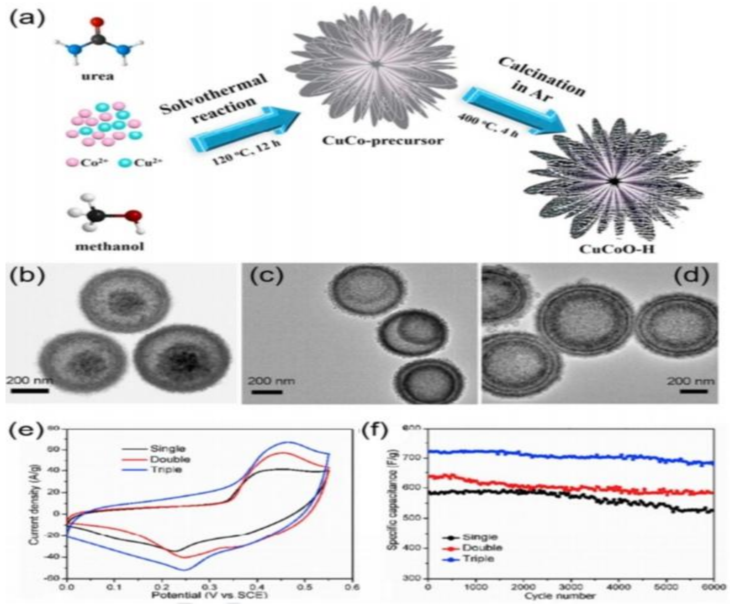
2.1 Hydrothermal Method

Hydrothermal method is the most common method for the preparation of metal oxides at micro and nanoscale. This method uses pure water as a solvent and the reaction temperature is usually higher than 1000C, and just an autoclave is required. It involves many advantages including simple operation and cost-saving, and the size and morphology of materials controlled. For example, Boopathi Raja and co-workers synthesized $CuCo_2O_4$ NPS by employing oxalic acid and NaOH as precipitate agents. The oxalic acid-assisted $CuCo_2O_4$ NPs showed a surface area of 133 m^2 g -1 and exhibited a specific capacitance of 765 F g -1, (0.1-0.4 V) with a cyclic performance of 95% capacity retention after 2000 cycles. However, the NaOH-assisted $CuCo_2O_4$ NPs possessed a surface area of 110 m^2 g -1 and a specific capacitance of 407 F g -1, with 90% capacity retention under the same conditions. Obviously, the oxalic acid-assisted $CuCo_2O_4$ NPs showing better performance are attributed to the good crystallinity and small particles of the oxalate precipitation, which facilitate redox reactions. Liao et al. directly prepared maguey-like $CuCo_2O_4$ NWs on Ni foam by a hydrothermal method. The nickel foam supported magnetically like $CuCo_2O_4$ NWs, exhibited a specific capacitance of 982 F g -1, (0-0.45 V) which provided more open pores for faradaic redox reactions (Fig. 2e and f) [12].



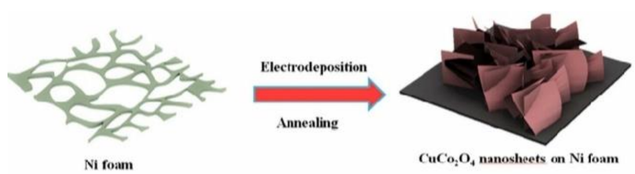
2.2 Solvothermal Method

The solvothermal method is similar to the hydrothermal reaction. The difference is that the solvothermal method uses an organic solvent or mixed solvent rather than pure water, and the reaction temperature is usually higher than the boiling point of the solvent used. For example, Jin et al. prepared copper-cobalt hybrid oxides Cu_2O, and CoO, during which the methanol was used as solvent shown in Fig. Li et al. reported that tripleshelled $CuCo_2O_4$ hollow microspheres were prepared in ethylene glycol and isopropanol mixed solvents. The single shell, double shell, and triple shell hollow structures were formed after 3, 6, and 8 h respectively (Fig. 3b-d).



2.3 Electro-Deposition Synthesis

It is commonly used to synthesize CuCo2O4 nanostructures. Naik et al. synthesized CuCo2O4 NBA no sheets on nickel foam and possessed a specific capacitance of 100 F g -1 with good cycling stability. Abbasi et al. prepared ultrathin CuCo2O4 nanosheets on nickel foam and exhibit the highest specific capacitance reaching 1330 F g -1, with superior 93.6% capacity retention after 5000 cycles (Fig. 4d and e). M. Pawar et al. also obtained CuCo2O4 nanosheets on nickel foam and exhibit a high specific capacitance 1473 F g -1 and a 93% capacity retention after 5000 charge-discharge cycles in 3 M of KOH electrolyte.

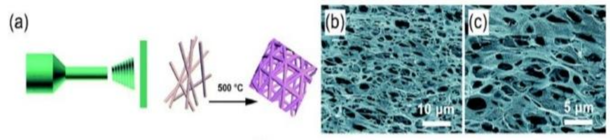


2.4 Template-Assisted Synthesis Method

Pendashteh's group highly ordered mesoporous CuCo2O4 nanowires (NWs) synthesized by a template-assisted method using silica SBA-15 as a hard template. The ordered CuCo2O4 NWs exhibited a high specific capacitance 1210 F g -1, (0-0.5 V) and a good rate capability with 64% capacity retention. However, the disordered CuCo2O4 electrode material only showed a specific capacity capacitance of 270 F g -1, (0-0.5 V). The excellent performance of CuCo2O4 NWs was due to the high order structure, which provided an active site for redox reactions. Kaverlavanini et al. prepared double-shelled CuCo2O4 hollow microspheres via the same method in isopropanol solvent. The whole synthesis process includes two steps. CuCo-glyceride precursor spheres were initially prepared by a simple solvothermal method. Later on, the CuCo-glyceride spheres were transformed into CuCo2O4 spheres with different internal structures by adjusting the heating rate during the calcination process. The specific capacitance of these CuCo2O4 microspheres could reach 1472 Fg -1.

2.5 Other Methods

Urea combustion, molten salts, and electro-spinning have also been employed to prepare CuCo2O4 materials with different structures. Typically, Krishnan et al. synthesized CuCo2O4 NPs using the molten salt method and exhibit a specific capacitance of 133 F g -1. M Silambarasan et al. prepared CuCo2O4 NPS via the chemical precipitation method using KOH as a precipitant and delivered a capacity of 290 F g -1. Pendashteh et al. synthesized cauliflower-like CuCo2O4 nanostructure by a simple urea combustion method and possessed a specific capacitance of 338 F g -1. Wang et al. synthesized CuCo2O4 network structures by an electro-spinning method coupled with thermal treatment (Fig. 6a-c).



III. Fabrication of Pure CuCo2O4 Electrode Materials

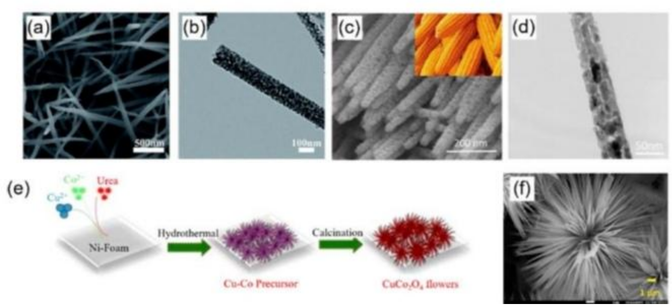
3.1 Powdered CuCo2O4 Electrode Materials

Powdered electrode materials are commonly used because of their advantages including easy synthesis, low cost, and large-scale production. The working electrode is fabricated by mixing the powdered electrode material, conductive reagent, and binder material in an exceedingly solvent then coating on a conductive substrate. as an example, Das et al. obtained porous CuCo2O4 sheets by adopting ammonia to regulate the pH at 14. These porous CuCo2O4 sheets possessed a surface area of 69.44 m2 g -1 and delivered a particular capacity of 449 F g -1 with 94% capacity retention after 5000 cycles.

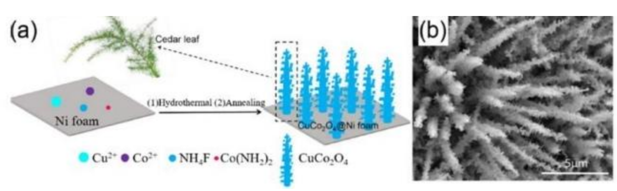
3.2 Binder-Free Structure of CuCo2O4 Materials on a Conductive Substrate

Binder and conductive reagents are employed in the fabrication of powder-modified electrodes. Gu et al. reported the fabrication of CuCo2O4 NWs on nickel wires via a hydrothermal method. The fiber-like supercapacitor consisted of PVA/KOH gel electrolyte and exhibited a particular capacitance of 11.09 F g -1, likewise as well as 93.5% capacity retention after 4000 charge-discharge cycles at 4 mA. Wang et al. prepared flower-like CuCo2O4 nanostructures on Ni foam through a hydrothermal method. The electrode exhibited a selected capacity of 243 F g -1, similarly to 85.3% capacity retention after 3000 cycles. Sequined et al. synthesized CuCo2O4 with different morphologies on nickel foam via changing the quantity ratio of water to ethanol, and it possessed the very best specific capacity of 285.5 F g -1

To further improve the electrochemical performance, Vijayakumar et al. prepared flowerlike CuCo2O4 microstructures on nickel foam for hybrid supercapacitor (Fig. 7a - 7f).



Wang et al. employed the hydrothermal/annealing method to directly grow CuCo2O4 with cedar-leaf like structure on nickel foam (Fig. 8a and b)



IV. Conclusion

In this work, we summarized the synthesis and applications of CuCo2O4 as an electrode material for supercapacitors in detail from the following aspects.

1. The preparation of $CuCo_2O_4$-based materials is realized by different strategies such as hydrothermal/ solvothermal, electro-deposition, electro-spinning, molten salt, and template-assisted method.
2. The synthesis of pure $CuCo_2O_4$ electrode materials is involved in two forms of powders and binder-free structures.

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