

Cite this: *RSC Adv.*, 2020, 10, 19353

Pulsed laser deposited CoFe_2O_4 thin films as supercapacitor electrodes

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The influence of the substrate temperature on pulsed laser deposited (PLD) CoFe_2O_4 thin films for supercapacitor electrodes was thoroughly investigated. X-ray diffractometry and Raman spectroscopic analyses confirmed the formation of CoFe_2O_4 phase for films deposited at a substrate temperature of 450 °C. Topography and surface smoothness was measured using atomic force microscopy. We observed that the films deposited at room temperature showed improved electrochemical performance and supercapacitive properties compared to those of films deposited at 450 °C. Specific capacitances of about 777.4 F g^{-1} and 258.5 F g^{-1} were obtained for electrodes deposited at RT and 450 °C, respectively, at 0.5 mA cm^{-2} current density. The CoFe_2O_4 films deposited at room temperature exhibited an excellent power density (3277 W kg^{-1}) and energy density (17 W h kg^{-1}). Using electrochemical impedance spectroscopy, the series resistance and charge transfer resistance were found to be 1.1 Ω and 1.5 Ω , respectively. The cyclic stability was increased up to 125% after 1500 cycles due to the increasing electroactive surface of CoFe_2O_4 along with the fast electron and ion transport at the surface.

 Received 19th March 2020
 Accepted 11th May 2020

 DOI: 10.1039/d0ra02564j
rsc.li/rsc-advances

1. Introduction

With the endless desire for electricity and drift from the conventional power grid to renewable energy sources, the demand for efficient energy storage devices is rising exponentially. For this, the supercapacitor (SC) is an excellent solution due to its fast charging/discharging rates, high power density, long cycle life, and eco-friendly nature.^{1–4} In general, SCs can be classified into two types based on the operational charge-discharge storage mechanism: electric double-layer capacitors (EDLCs) and pseudocapacitors. EDLCs work on the principle of electrostatics, where the energy is stored by trapping ions at the electrode/electrolyte interface.² A pseudocapacitor is based on the faradaic charge transfer process that utilizes reversible redox reactions and intercalation at electrodes to store charge. In particular, pseudocapacitive materials can achieve excellent specific capacitances and energy densities, and they can thus be

an interesting source for constructing novel energy storage devices.⁵ However, the pseudo capacitor has a comparatively shorter cycle life than an EDLC, due to the not completely reversible redox reactions and the loss of electrical contact resulting from the disintegration of the crystal structure, which reduces electrochemical performance. This can be overcome by combining a pseudo-active material with a conductive support, also known as hybrid supercapacitors.⁶ Transition metal oxides, such as RuO_2 , Co_3O_4 , and Fe_2O_3 have been considered as active materials for supercapacitors on account of their high theoretical capacitance, variable oxidation states, environmentally friendly nature, and low cost.^{7,8} Further, several researches have been focused to improve performance, storage, and energy densities for SCs by either tailoring the material properties^{9,10} and/or altering the surface¹¹ of the electrode materials. Some of the prominent approaches include the electrode made of porous carbon,¹² carbon nanotube,¹³ and metal-organic frameworks.^{14–16}

Recently, mixed metal oxides with spinel ferrite structure, MFe_2O_4 ($\text{M} = \text{Co}, \text{Mn}, \text{Zn}, \text{Mg}, \text{or Ni}$), were reported to have distinct hard or soft magnetic properties or even superparamagnetism, a large range of oxidation states, and chemical stability.¹⁷ In spinel ferrites, the divalent metal ion (M^{2+}) occupies the tetrahedral site, and the trivalent metal ion (M^{3+} or Fe^{3+}) occupies the octahedral position of the cubic close-packed oxygen lattice, see Fig. 1.^{18,19} D. Ravinder *et al.* reported that the ferrites have remarkably high permeability and can attain multiple redox states.²⁰ These properties of ferrites were

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