Study of structure and electrochemical properties of Fe-doped carbon aerogels

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Abstract. A nanocomposite material CAFe was obtained by assembling of Fe and carbon aerogels. The physical characterizations of CAFe were compared using XRD, Raman, XPS, and SEM. The results showed that Fe was successfully loaded onto carbon aerogels. The results of the electrochemical performance revealed CAFe exhibited high specific capacitance (78.7 F g⁻¹) and excellent stability. Therefore, Fe-doped carbon aerogel is a good electrode material.

1. Introduction
Electrical double-layer capacitance (EDLC) supercapacitor, has received increasing attention, due to its greater power density, longer cycle-life, and lower cost compared to other conventional capacitors [1]. EDLC focused on the ion deposition at electrode-electrolyte interface, which relies upon electrode porosity and surface area. Carbon materials, such as carbon aerogels, nanotubes and activated carbons have been widely studied as electrode materials for EDLC supercapacitor, due to their controllable pore structure, great surface area, high chemical stability and cost-effectiveness [2-3].

In the present study, nanocomposite material CAFe and carbon aerogel (CA) were obtained. Their differences are compared by X-ray diffraction (XRD), Raman spectroscopy (Raman), X-ray photoelectron spectroscopy (XPS) techniques. Electrochemical performance of the composite electrode was assessed using cyclic voltammetry (CV), galvanostatic charge-discharge test, and electrochemical impedance spectroscopy (EIS).

2. Methods

2.1. Synthesis of carbon materials
CAFe was prepared by a sol-gel approach. Resorcinol (R), phloroglucinol (P), formaldehyde (F, 37% aqueous solution) and Fe(NO₃)₃ꞏ9H₂O (Fe) were all purchased from Aladin, and then dissolved in distilled water at a R/F/P/Fe molar ratio of 7:16:1:1 and initial concentration of 45% (w/w). All reagents were mixed through 30-min ultrasonication, and then heated at 80 °C in a water bath for 72 h in order to attain wet gels. After that, the obtained wet gels were dried at vacuum condition, followed by pyrolysis at 900 °C in a N₂ atmosphere for 3 h in order to synthesize CAFe.
2.2. Characterization
The surface composition and structure of CAFe and CA was evaluated by XRD (Ultima IV XRD system, Rigaku, Japan) with CuKα radiation (λ=1.54 Å), Raman (DXR Raman microscope, Thermo Fisher, USA) and XPS (PHI5600, Physical ElectronicsInc, USA). Scanning electron microscopy (SEM; Tescan Maia3 MHL) were applied to characterize the morphology of as-prepared samples.

3. Results and discussion
The structures of CA and CAFe were assessed by XRD and Raman. The XRD patterns are summarized in Fig. 1(a). Notably, the XRD patterns indicated 2 peaks at approximately 24° and 44°, accounting for the (002) and (101) diffraction peaks of graphitic carbon, respectively [4]. In addition, the existence of Fe₃O₄ was confirmed.

The Raman spectrum (Fig. 1b) obviously displayed D-band and G-band peaks centered at 1350 cm⁻¹ and 1590 cm⁻¹, respectively. The D band is assigned to edge defects and highly disordered carbon, whereas the G band is attributed to C=C backbone stretching and highly ordered carbon [5]. The relative intensity ratios (R) of D band to G band (R=I_D/I_G) are listed in Fig 1(b). It further noticed that, the R value of CAFe (R=0.90) is lower than that of CA (R=0.98). Such low ratio reflects a high degree of graphitization of CAFe [6], which can markedly increase its electrical conductivity. These findings were consistent with the impedance behavior data (Fig. 4).

To attain more information regarding the elements of CAFe, XPS analysis was carried out and the obtained data are demonstrated in Fig. 2. As presented in Fig. 2, the full XPS spectrum of CAFe confirmed the presence of heteroatoms (Fe, O).

![Fig. 1 (a) X-ray diffraction patterns, (b) Raman spectra of all samples.](image1)

![Fig. 2. Full XPS spectrum of CAFe.](image2)
To further assess the electrochemical performances, CV test and galvanostatic charge–discharge tests were performed in 6 M KOH under a three-electrode configuration, and the results are shown in Fig. 4.

Fig. 4 (a) demonstrates the CV curves of CAFe and CA at 1 mV s\(^{-1}\). The CV curves revealed apparently well-defined redox peaks at approximately -0.004 A and 0.004 A for CAFe electrodes, indicating a major capacitive role of high pseudocapacity. It is noted that the CV curve shape of CA is comparatively different from that of CAFe, which is reasonably symmetrical nature either anodic or cathodic direction. These findings suggest the dominant EDLC behavior [7] of CA, and almost no faradaic contribution from the foreign dopant.

Fig. 4 (b) displays the specific capacitance (C) and galvanostatic charge–discharge curves of CAFe and CA at 1 A g\(^{-1}\). The specific capacitances of 90.8 F g\(^{-1}\) and 78.7 F g\(^{-1}\) were obtained at 1 A g\(^{-1}\) current density for CAFe and CA electrodes, respectively. The galvanostatic charge–discharge curve of CA was almost linear with asymmetrical triangular shape, indicating that this sample displays good electrochemical reversibility and high coulombic efficiency. Considering the pseudocapacitive behavior of CAFe’s functional groups, its galvanostatic charge-discharge curve is relatively unsymmetrical but slightly distorted, as consistent with its CV curve.

**4. Conclusion**

CA and CAFe were prepared. XRD, Raman, XPS, and N\(_2\) adsorption-desorption were adopted to characterize their physical structure. The results showed that Fe, N and O were successfully loaded onto CAFe. Electrochemical performance tests revealed that CAFe exhibited obvious EDLC behavior and faradaic contribution.
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