

BINDERLESS ORDERED MESOPOROUS CARBON MONOLITHS FROM LOW VALUE COAL TAR-DERIVED PRODUCTS

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Introduction

Ordered mesoporous carbons (OMCs) have been obtained by the hard template method using different carbon precursors such as sucrose, furfuryl alcohol, propylene or acetylene, among others.¹ These materials have been used as the active components in electrodes for energy storage devices, such as supercapacitors, taking advantage of their hierarchical porous texture. However, in these applications, the OMCs usually need to be mixed with a binder and a conductive additive to prepare the electrodes. In this work, we have used creosote, a by-product of the distillation of coal tar, as carbon precursor for the synthesis of OMCs, from which binderless monoliths were prepared.

Materials and Methods

Creosote consists of a mixture of aromatic compounds, including phenanthrene, fluorine and acenaphthene as its major components. The infiltration of the mesoporous silica template (SBA-15) was carried out in acidic medium as follows. First, the creosote was mixed with sulfuric acid (creosote:H₂SO₄ 9:1 vol/vol), and the mixture was kept at 40 °C overnight. Two phases were formed, a viscous one at the bottom and a more liquid one at the top. The viscous part was separated, and it was put in contact with the SBA-15 template in a round-bottom flask. The temperature was raised to 110 °C under vacuum, and then the flask was pressurized with argon. The pressure was kept slightly over atmospheric pressure for 3 h. Portions of the resulting composite of creosote and SBA-15 were manually pressed in a screw press to obtain disk-shaped monoliths 10 mm in diameter, which were carbonized in a horizontal furnace at 950 °C under an argon flow (flow rate: 500 mL/min). Finally, the template was removed by washing the composite with 1 M NaOH. The monoliths retained the original disk shape and were named as C/Creo/SBA-15.

The electrochemical performance of the as-obtained carbon monoliths was tested in aqueous electrolyte (1M H₂SO₄) in 3- and 2-electrode cells. A commercial powdered activated carbon (YP-50F from Kuraray) was also tested for comparison purposes. YP-50F was mixed with a conductive additive (carbon black, CB) and a binder (PTFE) to obtain a paste electrode (weight ratio YP-50F:CB:B=90:5:5).

Results and Discussion

After carbonization, the amount of carbonaceous product in the composite was ~36 wt.%, which is close to the value obtained using other carbon precursors with SBA-15 (e.g., sucrose or pitch²), and is sufficient to afford a well structured OMC as suggested by X-ray diffraction (results not shown). The material exhibited a type IV N₂ adsorption isotherm (Figure 1), with a very narrow pore size distribution (main pore size: 5.6 nm), as derived from the DFT method (inset to Figure 1), and a BET surface area of 1037 m²/g.

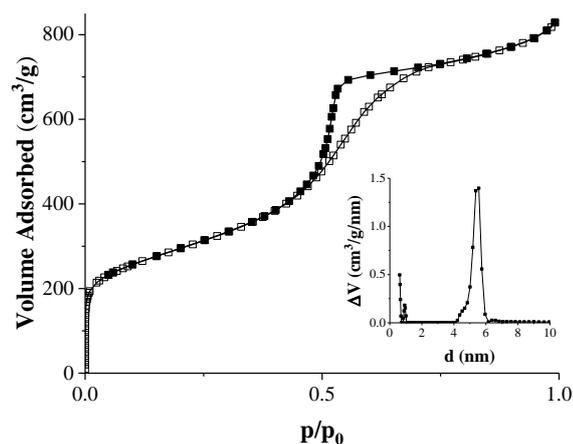


Figure 1. N₂ adsorption-desorption isotherm (−196 °C) of C/Creo/SBA-15, and pore-size distribution obtained by the DFT method (inset).

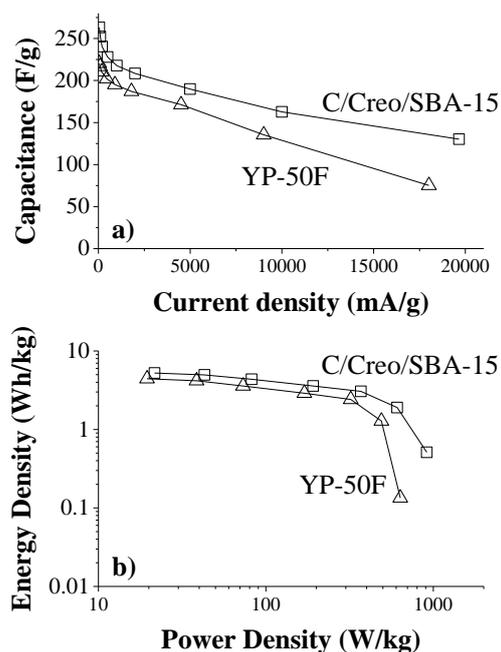


Figure 2. Specific capacitance as a function of current density in a 3-electrode cell configuration (a) and Ragone plot (b)

Capacitance values as high as 263 F/g were recorded at a current density of 50 mA/g in 1 M H₂SO₄ electrolyte in a 3-electrode cell configuration. This value was higher than the one obtained for a paste electrode prepared with YP-50F as the active material, 220 F/g (the capacitance values were normalized to the total weight of the electrodes including the additives used in the preparation of the paste). The capacitance retention with increasing current density was significantly higher in the case of the C/Creo/SBA-15 than for the commercial activated carbon (see Figure 2a). Furthermore, Ragone plots (Figure 2b) indicated that the monolith boasted higher energy and power densities than those of YP-50F. The higher performance of the monolith could be ascribed to the lower inter-particle resistance that was observed in the impedance spectra of the electrodes (not shown here) and also maybe to the well ordered mesoporous channels that allow a more expedient access of the electrolyte ions to the carbon surface.

Conclusions

Binderless monoliths of ordered mesoporous carbon were obtained by the hard template method using a low-value, coal tar-derived carbon precursor. The as-prepared monoliths were used as electrodes for supercapacitors without further components (i.e., no binder and conducting additives were employed), and they exhibited a good performance in acidic electrolyte.

Acknowledgment

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