Advanced Powder Technology xxx (2018) xxx–xxx

Contents lists available at ScienceDirect

Advanced Powder Technology

journal homepage: www.elsevier.com/locate/apt

Original Research Paper

One step synthesis process for fabricating $NiFe_2O_4$ nanoparticle loaded porous carbon spheres by ultrasonic spray pyrolysis

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ARTICLE INFO

Article history:
Received 27 November 2017
Received in revised form 28 February 2018
Accepted 6 March 2018
Available online xxxx

Keywords:
Nano $NiFe_2O_4$
Porous hollow carbon spheres
Ultrasonic spray pyrolysis
Formation mechanism
Potential application

ABSTRACT

In the present work, the porous hollow carbon spheres loaded with $NiFe_2O_4$ nanoparticles have been successfully prepared via ultrasonic spray pyrolysis technique at 700 °C and the associated formation mechanism has been studied. The as-prepared $NiFe_2O_4$/carbon microspheres with the diameter of about 3–5 μm and the specific surface area of 236.6889 $m^2 g^{-1}$ exhibit good monodispersity and an abundance of mesopores of about 40 nm size. Notably, the 20 nm $NiFe_2O_4$ nanoparticles are encapsulated by carbon microspheres and disperse homogeneously inside the carbon matrix. We could tune the relative content of ferrite and carbon sphere via adjusting the composition of the solution used for synthesis and the carbonization temperature. Consequently, some interesting properties can be obtained by combining the magnetic $NiFe_2O_4$ nano powder and the electrically conductive porous carbon, which renders the resulting composite suitable for promising applications in electromagnetic wave absorption, treatment of polluted water, catalyst design, energy storage, batteries and so on.

1. Introduction

Porous carbon sphere based materials have received significant attention over the past few decades because of their promising use in water treatment [1], energy storage [2,3], and lithium-ion batteries [4,5]. In this regard, several methods have been attempted to synthesize porous carbon spheres, such as carbonization of raw materials, hydrothermal synthesis [6], and ultrasonic spray pyrolysis (USP) [7,8]. Among these approaches, the technique of USP is most advantageous. First of all, being a facile and one-step process, it can be easily scaled up for mass production. Besides, the particles possess the same composition corresponding to reactions which occurred within the droplet [9]. Thus, the multicomponent and composite particle could be prepared by adjusting the composition of the precursor [10]. And as the precursor of carbon, block copolymers, sucrose with silicate, and alkali carboxylates is expensive and cumbersome [11], a low cost and environmentally friendly precursor without using a sacrificial template is desirable. Atkinson et al. have reported the synthesis of iron-impregnated porous carbon sphere using sucrose as the precursor [12].

The prepared porous carbon spheres exhibit a relatively inert surface suggesting the need for surface modification [13]. Coating the spheres with oxide nanoparticles could endow them with specific catalytic, magnetic, electronic, optical, or optoelectronic properties and significantly widen their utilities in areas such as electronics, magnetism, optics, and catalysis [14–16]. Li et al. have reported the synthesis of hierarchical MoO_2/C microspheres via a template-assisted process for subsequent application as anode materials for lithium-ion batteries [17]. As for the magnetic oxide nanoparticles, only a few kinds of composites consisting of Fe_3O_4 and carbon sphere have been synthesized for applications in water treatment and lithium-ion batteries [18]. As one of the most important members of the spinel ferrite family, $NiFe_2O_4$ possesses a set of highly specific characteristics such as low coercivity, high saturation magnetization, and excellent thermal and chemical stability [19]. Some interesting properties including electromagnetic wave absorption across a wide frequency band can be obtained by combining magnetic $NiFe_2O_4$ nanopowder and electrically conductive carbon with the porous structure [20]. As for water pollution treatment, the as-obtained $NiFe_2O_4$ nanoparticle plays the role of magnetic separation for removing the organic pollutants from aqueous mixtures while the surface functional groups and porous structure facilitate the process of adsorption[21]. However, few studies have been conducted pertaining to $NiFe_2O_4$ nanoparticles and porous carbon sphere nanocomposites.

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https://doi.org/10.1016/j.apt.2018.03.011
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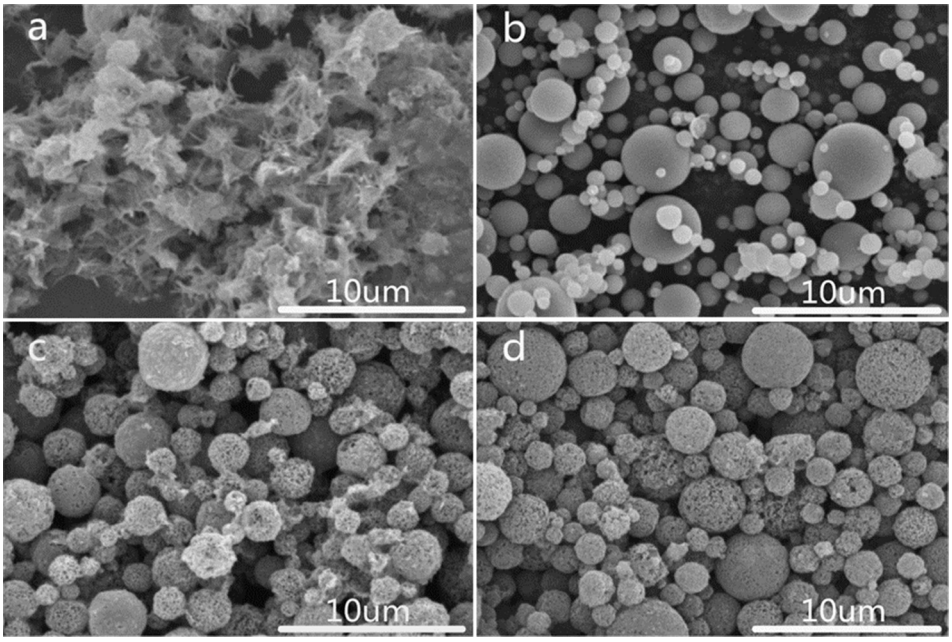

Fig. 1. SEM images of porous hollow carbon spheres resulted from different components, (a) $NiCl_2$ 0.1 M, NaCl 2.4 M and Sucrose 0.6 M, (b) $FeCl_3$ 0.2 M and Sucrose 0.6 M, (c) $FeCl_3$ 0.2 M, NaCl 2.4 M, Sucrose 0.6 M, and (d) $NiCl_2$ 0.1 M, $FeCl_3$ 0.2 M, NaCl 2.4 M and Sucrose 0.6 M.

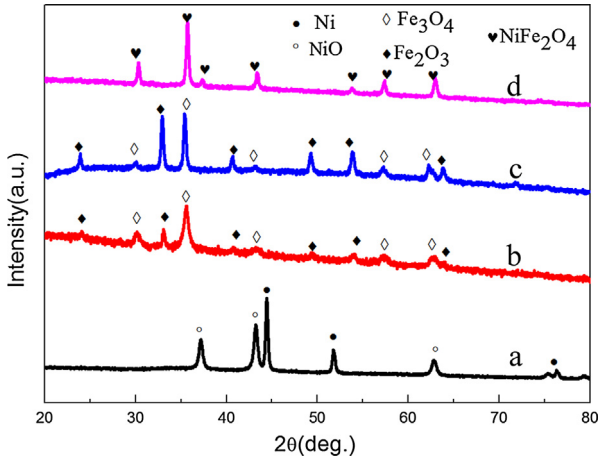

Fig. 2. XRD patterns of porous hollow carbon sphere corresponding to different components, (a) $NiCl_2$ 0.1 M, NaCl 2.4 M and Sucrose 0.6 M, (b) $FeCl_3$ 0.2 M and Sucrose 0.6 M, (c) $FeCl_3$ 0.2 M, NaCl 2.4 M, Sucrose 0.6 M, and (d) $NiCl_2$ 0.1 M, $FeCl_3$ 0.2 M, NaCl 2.4 M and Sucrose 0.6 M.

In this paper, the technique of ultrasonic spray pyrolysis was applied to the precursor of sucrose to synthesize $NiFe_2O_4$ nanoparticles loaded hollow carbon sphere. The mechanism corresponding to the formation of the particles was also proposed.

2. Experimental

2.1. Synthesis of $NiFe_2O_4$ and porous hollow carbon spheres composites

Sucrose (AR), sodium chloride (AR), nickel chloride hexahydrate (AR), and iron (III) chloride (anhydrous, CP) were used without further purification.

In a typical synthesis procedure, sucrose and sodium chloride were dispersed in 200 ml distilled water to obtain molar concentrations of 0.6 and 2.4 M, respectively. In the process of adding nickel chloride and iron (III) chloride into mixture solutions, unless otherwise noted, the molar ratio of nickel chloride to iron (III) chloride was maintained at 1:2. Additionally, the concentration of nickel chloride hexahydrate in the precursor solutions (X) was varied from 0.05 to 0.25 M.

The liquid precursor was atomized into aerosol droplets using a household humidifier (WH-2000, 1.7 MHz). The as-generated droplets were transported by Argon (at a flow rate of 3.0 L/min) into the preheated quartz tube reactor (600–750 °C) inserted inside a tube furnace (length: 100 cm, inner diameter: 8 cm), where solvent evaporation and thermal decomposition occurred. The resulting black product generated from pyrolysis were collected by a Buchner funnel (internal diameter is 9 cm) (as shown in Diagram 1) and washed with deionized water for 12 h. Then, the final products were dried in an oven at 60 °C prior to further material analysis.

2.2. Characterization

The structure was identified using X-ray powder diffraction (XRD, Bruker AXS) with Cu-Kα radiation (λ = 0.15415 nm). Scanning electron microscopy (SEM, Hitachi/SU1510) and transmission electron microscopy (TEM, FEI Tencnai G2 F30) was used to investigate the morphology. The analysis of N2 adsorption isotherms was used to determine the surface area (BET, Micromeritics ASAP 2020C+). The magnetic properties of as-prepared samples were measured by vibrating sample magnetometer (VSM, 7304).

3. Results and discussion

3.1. Morphology and phase composition of $NiFe_2O_4$ loaded porous hollow carbon microsphere

Fig. 1 shows the SEM images of the porous hollow carbon spheres derived from different starting solutions. A range of morphology was observed, including those of not so well defined spheres [Fig. 1(a)], the smooth carbon spheres [Fig. 1(b)], and the porous carbon spheres [Fig. 1(c and d)]. Therefore, it can be concluded that the morphology of carbon sphere produced varies with

Please cite this article in press as: J. Zheng et al., One step synthesis process for fabricating $NiFe_2O_4$ nanoparticle loaded porous carbon spheres by ultrasonic spray pyrolysis, Advanced Powder Technology (2018), https://doi.org/10.1016/j.apt.2018.03.011