Short communication

Hydrothermal deposition and characterization of silicon oxide nanospheres

L.Z. Pei*

School of Materials Science and Engineering, Key Lab of Materials Science and Processing of Anhui Province, Anhui University of Technology, Maanshan 243002, PR China

ARTICLE DATA

Article history:
Received 12 January 2007
Accepted 12 April 2007

Keywords:
Silicon oxide nanospheres
Hydrothermal deposition
Photoluminescence

ABSTRACT

Silicon oxide nanospheres with the average diameter of about 100 nm have been synthesized by hydrothermal deposition process using silicon and silica as the starting materials. The silicon oxide nanospheres were characterized by field emission scanning electron microscopy (FESEM), energy dispersive X-ray spectrum (EDS), transmission electron microscopy (TEM), high-resolution transmission electron microscopy (HRTEM) and photoluminescence (PL) spectrum, respectively. The results show that large scale silicon oxide nanospheres with the uniform size are composed of Si and O showing the amorphous structure. Strong PL peak at 435 nm is observed demonstrating the good blue light emission property.

1. Introduction

There has been an increasing interest in fabrication of inorganic nanosphere structures owing to their important applications in optical, catalytic and sensing devices (such as drug-delivery carriers and nano-reactors) [1–4] in addition to the general core-shell and nanotubular structures [5–8]. Different nanosphere structures such as ZnS, GaP, CdSe, ZrC, Bi_2Se_3, TiO_2 and Ni/SiO_2 nanospheres have been synthesized by different methods [9–15]. Among these nanosphere structures, silicon oxide nanospheres have attracted great interest owing to their novel extremely high surface area catalyst effect, such as the effect of large surface Cu/SiO_2 catalysts which are selective to the conversion of ethanol to acetaldehyde [16]. In the present, silica nanospheres can be synthesized by several kinds of methods, such as a single-step in situ high temperature process or the Stöber–Fink–Bohn method [16–18]. Recently, Lecren et al. [19] reported that multi-wall silica nanospheres were synthesized from non-ionic surfactant and tetraethoxi-orthosilane (TEOS) as precursor under acidic and reflux conditions. However, these methods make this approach complicated and some organic solvent is used. Therefore, the development of less complicated synthesis methods is of great interest for technological applications. More recently, amorphous MoS_2 nanospheres were synthesized by hydrothermal method [20]. Hydrothermal synthesis has many advantages over other methods. It is environmentally benign, inexpensive, and allows for reduction of free energies for various equilibria [21,22]. In this paper, we report a simple hydrothermal deposition route for the synthesis of silicon oxide nanospheres without metallic catalysts with the average diameter of around 100 nm. The products have been characterized by field emission scanning electron microscopy (FESEM), energy dispersive X-ray spectrum (EDS), transmission electron microscopy (TEM), selected area electron diffraction (SAED), high-resolution transmission electron microscopy (HRTEM) and photoluminescence (PL) spectrum.

* Tel.: +86 555 2311570; fax: +86 555 2311570.
E-mail address: lzpei1977@163.com.

1044-5803/$ – see front matter © 2007 Elsevier Inc. All rights reserved.
doi:10.1016/j.matchar.2007.04.011

2. Experimental

The preparation of silicon oxide nanospheres was performed in a reaction kettle. The reaction kettle is composed of stainless steel (1Cr18Ni9Ti). The main detail of the reaction kettle is shown as follows: Maximal pressure is 22 MPa, maximal temperature is 500 °C, volume is 1000 mL, power is 1.5 Kw and stirring velocity of the stirrer is 0–1000 r/min. 1.25 wt.% Si (purity:≥99%, average particle size: ~42 μm) and SiO_2 (purity: ≥99.5%, particle size: 30±5 nm) powders were mixed with 48 ml distilled water. The mole ratio of Si and SiO_2 is 1:1. Then the mixture was put into the reaction kettle. The silicon substrate with the size of 4×2 cm was put into distilled water and cleaned for about 10 min using supersonic wave apparatus before use in order to insure the cleanness in the surface of silicon substrates. Then the cleaned silicon substrate was fixed in the stainless steel bracket in the center of the reaction kettle. After the reaction kettle was sealed safely, it was heated to 470 °C of temperature, 6.85–8.5 MPa of pressure, 200 rpm of the rotating speed for the stirrer of the apparatus and the temperature and pressure were maintained for 24 h. Subsequently the reaction kettle was cooled naturally. Finally, the silicon substrate with bulk light grey deposit was obtained after the experiment. SEM and TEM confirmed that the product was silicon oxide nanospheres existing in the silicon substrate.

SEM observation was performed using JEOL JSM-5600LV FESEM with 1 nm point-to-point resolution operating with a 15-kV accelerating voltage. The surface of the silicon substrate with silicon oxide nanospheres is treated by spraying carbon in order to increase the electrical conductivity before the sample was observed by SEM. The product was separated from the silicon substrate using hardy plastic utensil. TEM and HRTEM sample was prepared by putting several drops of solution with silicon oxide nanospheres onto a standard copper grid with a porous carbon film after the silicon oxide nanospheres were dispersed into distilled water and treated for about 10 min using supersonic wave apparatus. TEM and HRTEM observations were performed using JEOL JEM 3010 transmission electron microscope with 1.7 Å point-to-point resolution operating with a 300-kV accelerating voltage with a GATAN digital photography system, respectively. Photoluminescence (PL) measurement was carried out in the silicon substrate with silicon oxide nanospheres at room temperature using 325 nm as the excitation wavelength with a luminescence spectrometer (Hitatch, F-2500) in the range of 380–650 nm.

3. Results and Discussion

Fig. 1(a) and (b) shows the representative SEM image revealing the general morphology of the silicon oxide nanospheres. A large amount of silicon oxide nanospheres deposited in the silicon substrate are observed from the SEM image of Fig. 1(a). According to SEM image with higher magnification (Fig. 1(b)), it can be seen that the obtained silicon oxide nanospheres have smooth surface and definite diameter distribution. The diameter of the silicon oxide nanospheres is ranging from less than 50 nm to about 200 nm. The average diameter is around 100 nm. The morphology of the obtained silicon oxide nanospheres is similar to that of the silicon oxide nanospheres synthesized by other methods and other kinds of nanosphere structures [9–18]. No other nanostructures are observed besides the nanosphere structure demonstrating that the obtained silicon oxide nanospheres are highly pure. The EDS spectra of the nanospheres are measured in order to determine the composition of nanospheres. Fig. 1(c) and (d) is the EDS spectra of silicon oxide nanospheres with point scanning and plane scanning, respectively showing that the nanospheres are composed of silicon and oxygen. Some amount of element carbon is observed in the EDS spectra because the surface of the silicon substrate with silicon oxide

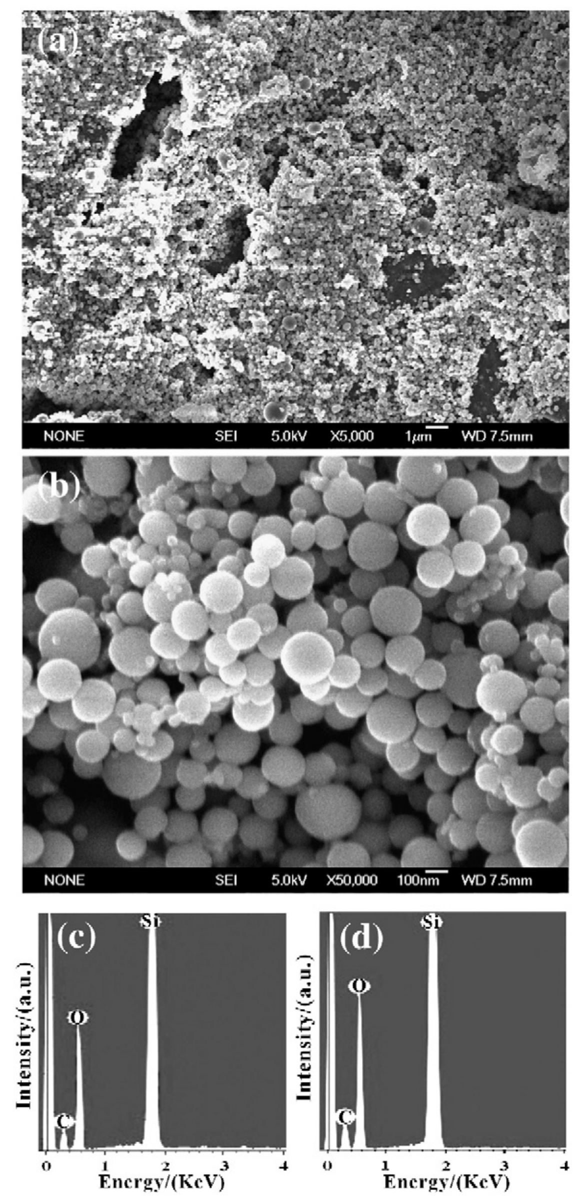

Fig. 1 – (a) and (b) SEM images of silicon oxide nanospheres with different magnifications. (c) and (d) The EDS spectra of silicon oxide nanospheres with point scanning and plane scanning, respectively.