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A simple sonochemical approach for synthesis and characterization of Zn_2SiO_4 nanostructures

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ABSTRACT

Zn_2SiO_4 nanoparticles have been successfully prepared via a simple sonochemical method, for the first time. The effect of various parameters including ultrasonic power, ultrasonic irradiation time and different surfactants were investigated to reach optimum condition. The as-prepared nanostructures were characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM), transmittance electron microscopy (TEM), Fourier transform infrared (FT-IR) spectra and energy dispersive X-ray microanalysis (EDX). The photocatalytic activity of Zn_2SiO_4 nano and bulk structures were compared by degradation of anionic dye methyl orange in aqueous solution under UV-light irradiation. Moreover, the cyclic voltammetry analysis of Zn_2SiO_4 nano and bulk structures were investigated.

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1. Introduction

Recently, much research has been interested in the synthesis and characterization of nanostructures such as sulfides, oxides and nitrides [1–3]. These nanometer-sized inorganic materials can represent an extensive range of optical and electrical properties [4,5]. Zinc silicate (Zn_2SiO_4), as one of the most important family of functional inorganic materials, have wide applications in the fields of phosphors [6,7], lithium-ion batteries [8,9], paints [10], adsorbents [11] and electronic insulators [12] as well as rubber mixtures [13]. For zinc silicates, most of the reported zinc silicate nanomaterials are composed of highly crystalline hemimorphite or willemite units [14–16]. Willemite Zn_2SiO_4 is a natural orthosilicate with a phenacite-like structure in which Zn—O tetrahedra and Si—O tetrahedra share corners to form hollow 'tubes' parallel to [0001] [17,18]. The numerous investigations on the synthesis routes of Zn_2SiO_4 have been carried out, including sol–gel method [19], solid-state route [20], polymer precursor method [21], spray pyrolysis method [22] and hydrothermal method [23,24]. However these methods usually require high pressure or heat-treating at temperatures higher than 180 °C for several hours. These problems can be avoided by using sonochemical method. This method has become an important tool in synthesis chemistry to generate novel nano-sized materials under ambient conditions in recent years [25–30]. The ultrasound effects arise from acoustic cavitation, which is the formation, growth, and implosive collapse of bubbles in a solvent. The growth of the bubble occurs through the diffusion of solute vapor into the volume of the bubble, while the collapse of the bubble occurs when the bubble size reaches its maximum value. According to hot spot theory, very high temperatures (>5000 K) are obtained upon the collapse of a bubble [31,32]. These extreme conditions can drive a variety of chemical reactions to synthesize nano-sized materials. There has not been any report for precipitation of Zn_2SiO_4 using only ultrasonic process, so far. In this work, Zn_2SiO_4 nanostructures were prepared in the presence of ultrasound irradiation applying the low temperature in less than 40 min. The effect of different parameters such as ultrasonic power, ultrasonic irradiation time and different surfactants like cetyltrimethylammoniumbromide (CTAB), sodium dodecyl sulfate (SDS), polyvinylpyrrolidone (PVP) and poly ethylene glycol (PEG) were investigated to reach optimum condition. Moreover, the photocatalytic degradation of anionic dye methyl orange is performed to study effect of ultrasonic irradiation on the catalytic properties of as-produced nanostructures. Also, the cyclic voltammetry analysis of Zn_2SiO_4 nano and bulk structures were compared.

2. Experimental

2.1. Materials and physical measurements

All the chemical reagents for the synthesis of zinc silicate nanostructures such as $Zn(CH_3COO)_2{\cdot}2H_2O$, tetraethyl orthosilicate (TEOS), ammonia hydroxide, methanol, cetyltrimethylammonium-

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Table 1
Preparation conditions for the synthesis of Zn_2SiO_4 nanostructures.

Sample No.	Surfactant	Sonication time (min)	Power (W/cm^2)	Molar ratio (Zn:Si)	Particle size (SEM)/nm
1	CTAB	30	60	2:1	60–300
2	SDS	30	60	2:1	200–400
3	PVP	30	60	2:1	10–55
4	PEG	30	60	2:1	60–300
5	PVP	20	60	2:1	18–250
6	PVP	40	60	2:1	25–100
7	PVP	30	40	2:1	20–140
8	PVP	30	80	2:1	30–90
9	PVP	–	–	2:1	60–450
10	–	30	60	2:1	200–300
11	PVP	30	60	0:1	–
12	PVP	30	60	2:0	–

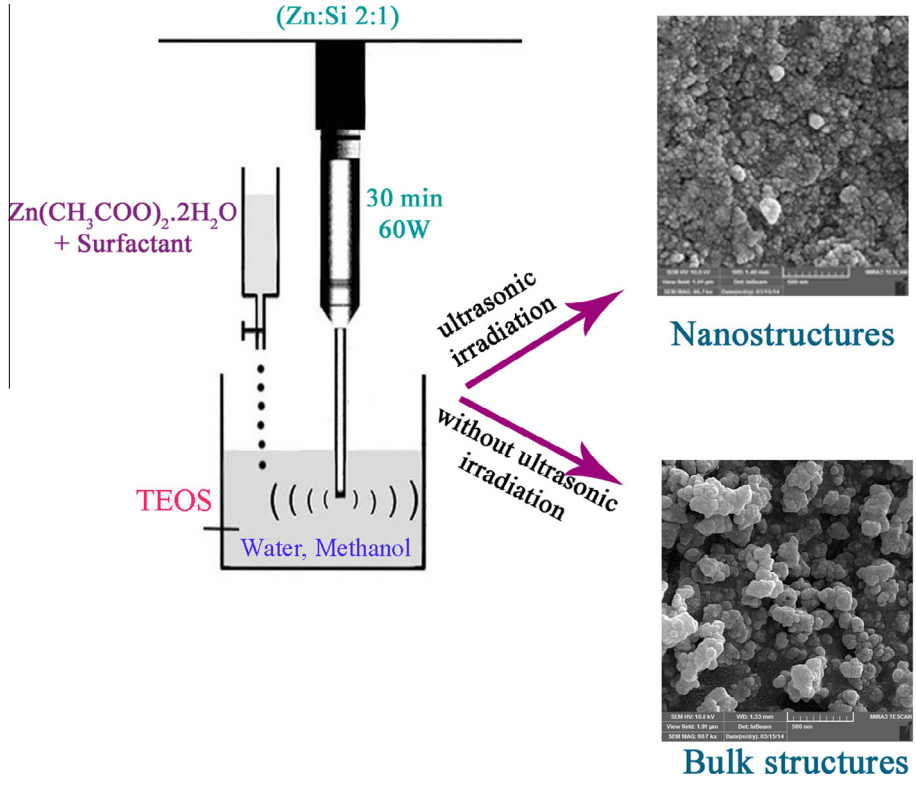
bromide (CTAB), sodium dodecyl sulfate (SDS), polyvinylpyrrolidone (PVP) with molecular weight of 25,000 and poly ethylene glycol with molecular weight of 6000 (PEG) were commercially available and employed without further purification. A multiwave ultrasonic generator (MPI Ultrasonics; welding, 1000 W, 20 kHz, Switzerland), immersed directly in the reaction solution. X-ray diffraction (XRD) patterns were recorded by a Philips-X'pertpro, X-ray diffractometer using Ni-filtered Cu Ka radiation. Fourier transform infrared (FT-IR) spectra were recorded on Nicolet Magna-550 spectrometer in KBr pellets. The electronic spectrum of the sample was taken on Perkin–Elmer LS-55 luminescence spectrometer. Scanning electron microscopy (SEM) images were obtained on LEO-1455VP equipped with an energy dispersive X-ray spectroscopy. The EDX analysis with 20 kV accelerated voltage was done. Transmission electron microscopy (TEM) image was obtained on a Philips EM208 transmission electron microscope with an accelerating voltage of 200 kV.

2.2. Synthesis of Zn_2SiO_4 nanostructures

Zn_2SiO_4 nanostructures were prepared by reaction between Zn $(CH_3COO)_2{\cdot}2H_2O$ and TEOS with molar ratio of 2:1 in the presence of ultrasound irradiation. In a typical experiment, TEOS was dissolved in appropriate amounts of distilled water and methanol under stirrering and a few amount of ammonia hydroxide as a catalyst was added into the mixture. This solution was added dropwise into an aqueous solution including zinc acetate and surfactant (Zn:surfactant 1:1) under ultrasonic waves. The white precipitates obtained under various sonication times were collected by centrifugation at 4000 rpm for 5 min, washed repeatedly with distilled water and ethanol several times. The as-obtained products were dried at 80 °C under vacuum for 2 h and then were calcined at 950 °C for 2 h. The preparation conditions for synthesis Zn_2SiO_4 nanostructures have been illustrated in Table 1. To study the effect of ultrasound irradiation on the morphology and chemical composition of the products, an experiment as blank tests were carried out without sonication at room temperature. In Scheme 1, schematic diagram of formation of Zn_2SiO_4 nano and bulk structures is depicted. Moreover, the effect of ultrasonic irradiation on the morphology and size of structures is seen.

2.3. Photocatalytic measurements

The photocatalytic activity of Zn_2SiO_4 nano and bulk structures was tested by using anionic dye Methyl orange solution. The degradation reaction was carried out in a quartz photocatalytic reactor. The photocatalytic degradation was carried out with 0.05 g of MO solution containing 0.05 g of nano and bulk structures. This



Scheme 1. Schematic diagram of formation of nanostructures and effect of ultrasonic irradiation on the particle size and morphology.

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