



In situ synthesis of ZnO nanostructures on a zinc substrate assisted with mixed cationic/anionic surfactants

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ABSTRACT

Different morphologies of ZnO nanostructures have been successfully prepared on zinc foil via a mild hydrothermal process in the presence of cationic surfactant cetyltrimethylammonium bromide (CTAB) and anionic surfactant sodium dodecyl sulfate (SDS), by using zinc foil as both zinc source and substrate. X-ray diffraction (XRD), filed emission scanning electron microscope (SEM), transmission electron microscope (TEM), selected area electron diffraction (SAED), Raman spectra and room temperature photoluminescence (PL) spectra are used to characterize the obtained products. The observed Raman scattering and PL results confirm that as-obtained ZnO nanostructures have a good crystal quality with a wurtzite hexagonal phase and exhibit good optical property. A reasonable formation mechanism was proposed based on the experimental results.

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1. Introduction

Recently, semiconductor nano-/micro-materials have been extensively studied owing to their potential applications in manufacturing electronic and optoelectronic devices [1,2]. Especially, for the past few years, substantial efforts have been devoted to control the architecture and spatial patterning of metal oxide semiconductor materials. Among various transition metal oxides and their derivatives, zinc oxide (ZnO) have attracted a great deal of interest because of their intriguing optical, electronic, and mechanical properties and potential applications in optoelectronics, photonics, field emission, energy conversion, catalysis, and sensors [3–7].

ZnO is a remarkable semiconductor with its wide band gap (3.37 eV), large excitation binding energy at room temperature (60 meV), and excellent chemical and thermal stability. As a consequence, controllable synthesis of ZnO nanostructures with different morphologies and sizes has attracted considerable attention from the standpoint of basic research and the realization of advanced devices. Up to now, various ZnO nanostructures, e.g. tubes, wires, rods, flowers have been fabricated through a variety of methods, such as hydrothermal method, physical vapor evaporation, thermal decomposition, template-directed growth and surfactant-assisted method [8–12]. Moreover, well-aligned ZnO nanostructures have also been fabricated on a variety of substrates, including glass, GaN

and Si [13–15]. Notably, the direct growth of oriented ZnO nanostructures on a zinc substrate was recently realized by the natural surface-oxidation of zinc foil in solution at room temperature or under hydrothermal conditions, by using Zn foil as both zinc source and substrate [16,17]. In addition, it is to be noted that mixtures of various surfactants, including alkyl amines, alkyl acids, alkylphosphonic acid, and trioctyl phosphine oxide, are frequently used as capping agents to tailor the crystal shape in high-temperature solution phase synthesis. Especially, mixed cationic/anionic surfactants have been widely used to control the morphological evolution of nanostructures and their hierarchical architectures by many groups [18,19]. Inspired by this, here, we report a one-step hydrothermal process for the in situ preparation of different morphologies of ZnO nanostructures on a zinc substrate assisted with mixed cationic/anionic surfactants. The PL and Raman scattering behavior of ZnO nanostructures were also investigated.

2. Experimental

The reagents used in this work, including cationic surfactant cetyltrimethylammonium bromide (CTAB), anionic surfactant sodium dodecyl sulfate (SDS), ammonium peroxydisulfate ((NH₄)₂S₂O₈, APS), NaOH and ethylenediamine (en) were of analytical reagent grade and used without further purification. Zinc foils (10 mm × 10 mm × 0.5 mm, 99.9%) were pretreated by sonication in ethanol for 5 min and dried by using a dry nitrogen stream.

In a typical procedure, the starting solution was prepared by mixing 0.2187 g CTAB and 0.0334 g SDS in 32 ml water, followed by the addition of 3.84 g NaOH and 0.8215 g (NH₄)₂S₂O₈ (or appropriate en) under stirring, giving the molar ratio of CTAB/SDS as 5.7/1.1. After that, the solution was transferred into Teflonlined stainless steel autoclaves, and the previously cleaned zinc foil was then immersed in the

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solution. The autoclaves was sealed and maintained at 150 °C for 18 h. After cooling down to room temperature, the resulting zinc foil was taken out and thoroughly rinsed with ethanol and deionized water.

The obtained products were characterized by X-ray powder diffraction (XRD Shimadzu 6000, Cu K α radiation), field emission scanning microscopy (FE-SEM JEOL JEM-6700F), transmission electron microscope (TEM, HITACHI H-8100), selected area electron diffraction (SAED), Raman (Renishaw 1000, excited with the 514.5 nm Ar⁺ laser) and photoluminescence (PL F-4500, He-Cd laser of 325 nm).

3. Result and discussion

The phase composition and phase structure of as-obtained samples were examined by XRD. As shown in Fig. 1. All of the peaks can be readily indexed to the wurtzite-type phase of ZnO (JCPDS card no. 36-1451) in addition to the peaks marked with Zn that are attributed to the zinc foil substrate (JCPDS 04-0831). The sharp diffraction peaks of the sample indicate that ZnO nanostructures can be easily obtained under the current synthetic conditions. No characteristic peaks of other impurities have been detected, indicating that the products are of high purity.

The morphologies and microstructure of the products were further investigated by SEM. Fig. 2 shows the images of typical products obtained through direct oxidation of zinc foil with APS in the alkali solutions of mixed CTAB/SDS surfactants. An overview SEM image is presented in Fig. 2a, which shows that highly uniform and dense ZnO over a large area were formed on the Zn substrate. Fig. 2b and c are the magnified images, clearly showing that formed structures are composed of sheet-like morphology in high density.

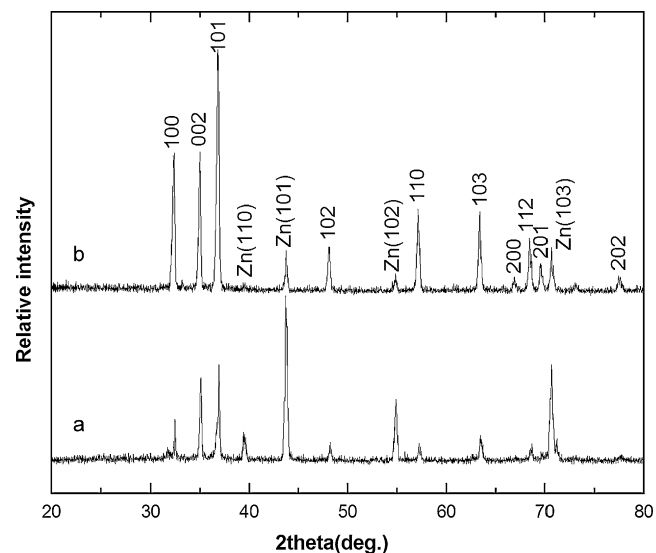


Fig. 1. Typical X-ray diffraction patterns of the products obtained with the addition of: (a) NaOH and APS; (b) en.

According to the careful examination from the enlarged images in Fig. 2d, the nanosheets exhibit crooked anomalous nanosheets morphology. The dimensions are in the range of 400–525 nm and the thickness of 15–25 nm. The ZnO nanosheets have been further

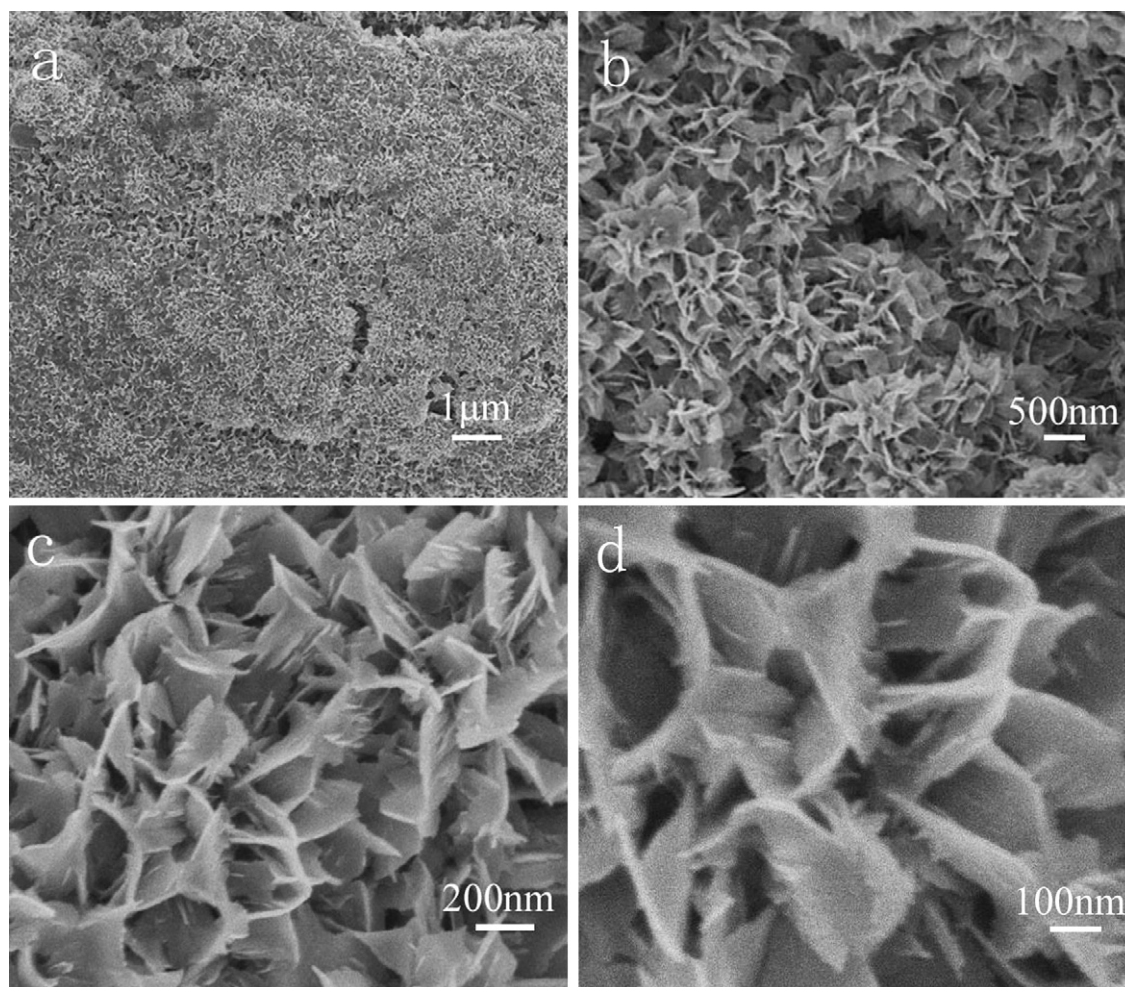


Fig. 2. (a) Overview; (b–d) high-magnification FE-SEM images of ZnO obtained in mixed CTAB/SDS solutions with the addition of NaOH and APS.

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