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Room temperature synthesis of single-crystal Te nanorods from Na_2TeO_4

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Abstract

Single-crystalline tellurium nanorods were synthesized through the reduction of Na_2TeO_4 by hydrazine monohydrate, in the absence of surfactants, in an aqueous ammonia solution at room temperature. X-ray diffraction (XRD), transmission electron microscopy (TEM), and selected area electron diffraction (SAED) were used to characterize the composition and morphology of the products. The concentration of the OH^- ion has a significant influence on the morphology of the products and is found to be responsible for tailoring the crystal growth dynamically: the concentration of Te blocking in the solution is reduced via increasing the concentration of the OH^- ion, and subsequently the nucleation rate of Te is suppressed and Te nanorods gradually grow because of the inherently anisotropic structure of Te. New generation tellurium atoms add to the surface of the particles during the long period of reaction.

Keywords: nanorods; tellurium; room temperature synthesis; single crystal; sodium tellurate

1. Introduction

Since the discovery of carbon nanotubes in 1991 [1], the fabrication of one-dimensional (1D) nanostructure has been the subject of especially intense research because of the great potential of these materials for addressing some basic issues about dimensionality and space-confined transport phenomena, such as quantized conductance and size effects [2-3]. However, the synthesis of 1D nanostructures is a challenge, owing to their extremely small size and their anisotropic structure. Many methods have been applied for the preparation of 1D nanostructures, including microwave-assisted [4], template-assisted [5-6], arc discharge [1], laser ablation [7], and other methods [8-10]. Of late, the solution-phase method has become a promising technique for preparing the 1D nanostructure because of its relatively low cost and potential for large-scale production [11-14].

Elemental tellurium is a narrow band gap semiconductor material, with a highly anisotropic crystal structure of a unique helical-chain conformation. The tellurium exhibits unique properties, such as, nonlinear optical responses, photoconductivity and so on, and is widely used to make highly resistive devices. Therefore, it is attractive to obtain a 1D nanostructure of tellurium for new types of applications or to enhance the performance of the existing devices based on the quantum-sized effects. To date, tellurium nanorods [15], nanowires [16], nanobelts [17], and nanotubes [18] have been fabricated via solution phase methods. However, it is seldom reported that tellurium nanorods are synthesized using Na_2TeO_4 and $N_2H_4{\cdot}H_2O$ as starting materials in the absence of surfactants, by a simple room temperature route.

In this study, the authors have developed a room temperature reduction route for the synthesis of tellurium nanorods through reducing Na_2TeO_4 and using hydrazine monohydrate in an aqueous ammonia solution.

2. Experimental

All of the chemical reagents were purchased from Shanghai Chemical Reagents Company and were used without further purification. In a typical procedure, 1 mmol Na_2TeO_4 was dissolved in 30 mL $NH_3{\cdot}H_2O$ (25 wt.%) under constant stirring to form a homogeneous solution in a 50 mL conical flask at room temperature. Subsequently, 1 mL hydrazine monohydrate ($N_2H_4{\cdot}H_2O$, 85 wt.%) was slowly dropped into the solution under stirring. The conical flask was then sealed and aged at room temperature (about 25°C) for 36 h. The resulting gray solid products were filtered, washed several times with distilled water and absolute ethanol, respectively, and finally dried in vacuum at 60°C

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for 4 h. For comparison, several experiments were carried out just by changing the composition of the solution.

The as-prepared products were characterized by X-ray powder diffraction (XRD) analysis on a Philips Xpert Pro X-ray diffractometer equipped with graphite monochomatized Cu K_α radiation (λ = 0.154178 nm). Transmission electron microscopy (TEM) images and the corresponding selected area electron diffraction (SAED) image were recorded on a Hitachi-800 transmission electron microscope performed at 200 kV.

3. Results and discussion

A typical XRD pattern of the products is shown in Fig. 1. All the strong and sharp reflection peaks in this pattern can be indexed to hexagonal-structure tellurium with lattice constants of a = 0.4451 nm and c = 0.5925 nm, which are consistent with the literature values (JCPDS 79-0736, a = 0.4456 nm and c = 0.5921 nm). No other impurities can be detected.

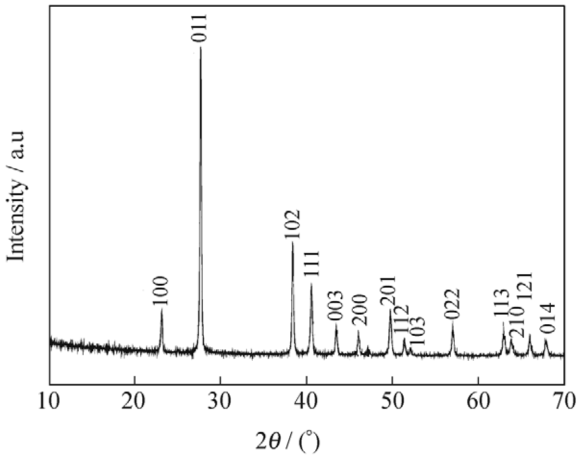

Fig. 1. XRD pattern of the as-prepared tellurium nanorods.

The TEM images and the corresponding SAED image of the as-prepared tellurium nanorods in 25 wt.% aqueous ammonia are shown in Fig. 2. Fig. 2(a) illustrates trigonal tellurium nanorods with 20-50 nm in diameter and 0.3-1.0 μm in length. These nanorods have a uniform size along the axis direction. Fig. 2(b) is an image of a single nanorod with a diameter of 50 nm and a length of 800 nm. Fig. 2(c) shows a SAED image taken from this nanorod, which reveals that the as-prepared nanorods are a single crystalline in structure with a growth direction parallel to the c-axis of the infinite, helical chain of tellurium atoms. The results are consistent with those of XRD. However, the as-prepared products have extremely different morphologies when the mass of aqueous ammonia in the mixture is changed (shown in Fig. 3). Just small irregular tellurium particles can be obtained in pure distilled water instead of aqueous ammonia (shown in Fig. 3(a)). Furthermore, the products obtained from the mixture of 15 mL distilled water and 15 mL of 25 wt.% aqueous ammonia are composed of many nanorods, 30-40 nm in diameter and 0.4-1.0 μm in length, but there are still many irregular tellurium nanoparticles and some inadequate upgrowth nanorods. Obviously, the concentration of aqueous ammonia is a main factor that influences the morphologies of the as-prepared products, without changing other parameters. It is in favor of growing 1D nanorods when the concentration of aqueous ammonia is higher. As is well known, tellurium is a hexagonally structured crystal, and

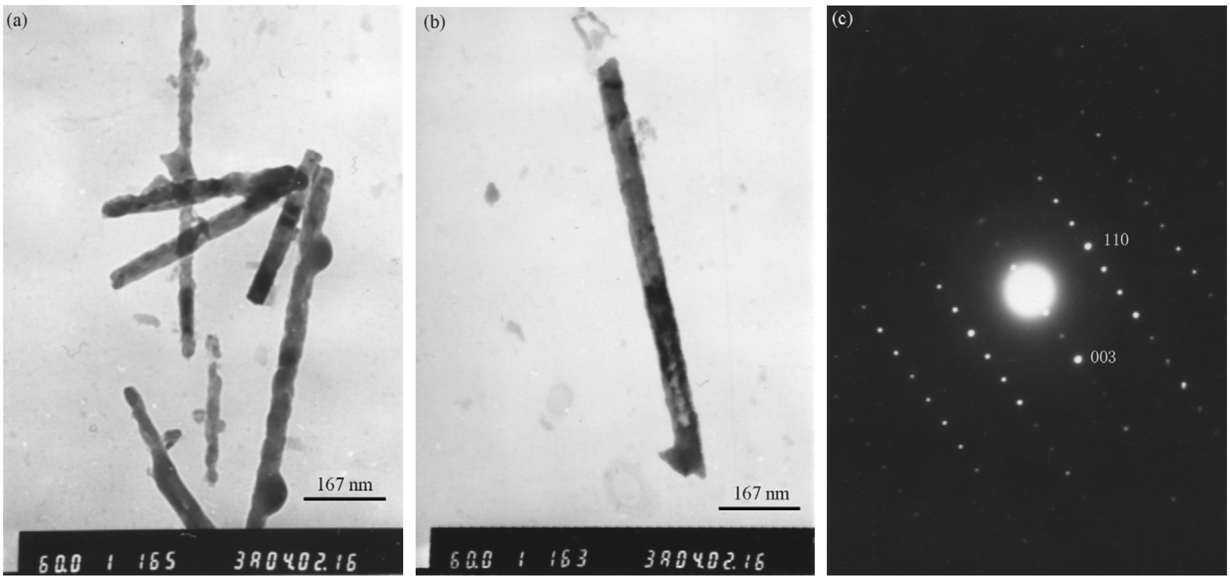

Fig. 2. TEM images of as-prepared tellurium nanorods in 25 wt.% aqueous ammonia: (a) typical TEM image of tellurium nanorods; (b) TEM image of a single tellurium nanorod; (c) SAED image taken from various sites of an individual tellurium nanorod.