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Amorphous lead oxide nanotubes filled partially with single-crystalline lead

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Abstract

Lead oxide nanotubes filled partially with lead were synthesized by a simple thermal evaporation on Si(1 0 0) substrates without any catalysts. The inner diameter, wall thickness, and length of the nanotubes were 10–40 nm, 5–7 nm, and 2–10 μm, respectively. Selected area electron diffraction patterns obtained from these nanotubes revealed that the lead oxide nanotubes were amorphous, but that the lead present inside the nanotubes was single crystalline. During the exposure of an electron beam on these nanotubes, the amorphous lead oxide nanotubes were transformed into single-crystalline lead oxide nanoparticles. A possible formation mechanism of the partially filled nanotubes is suggested in this paper.

Keywords: Lead oxide; Lead; Nanotubes; Nanorods

1. Introduction

Since the discovery of carbon nanotubes, a variety of nanostructures including nanowires, nanobelts, nanocables, and nanotubes have been one of attractive research issues. In particular, there have been many efforts on the fabrication of the one-dimensional metal-oxide nanocables and nanotubes due to their exceptional physical properties and potential applications in nanoelectronics [1,2]. Recently, Zn–ZnO core–shell nanobelts and ZnO nanotubes have been synthesized by solid-vapor decomposition from pure ZnO powders [3], and Sn/SnO_2 core–shell nanowires have been fabricated with kinetic control in thermal oxidation process [4].

Lead (Pb) is a particularly important material because of its superconductivity and high reactivity [5]. Lead oxide has wide industrial applications as the basic material of the electrode active mass in lead-acid batteries. The lead oxide has four basic types of PbO, PbO_2, Pb_2O_3, and Pb_3O_4. In particular, PbO_2 and Pb_2O_3 are commonly used as electrodes in electrochemical devices [6]. Recently, lead nanowires were synthesized by using electrochemical deposition [7,8] or solution-phase method [9]. Nevertheless, to the best of our knowledge, there have been few reports on lead oxide nanostructures. In this study, we report that lead oxide nanotubes filled partially with lead were synthesized by a simple thermal evaporation on Si(1 0 0) substrates without any catalysts. In this paper, the synthesis of the lead oxide nanotubes filled partially with lead is first briefly described. And then the structural properties of the synthesized nanotubes are investigated, and finally a possible formation mechanism of the partially filled nanotubes is suggested.

2. Experimental procedure

PbF_2 and B_2O_3 powders were loaded into the center of a horizontal alumina tube, and 5 mm × 5 mm-sized Si substrates were put at two different temperature regions A (600 °C) and B (400 °C) of the alumina tube. The thermal evaporation of the powders was performed at 1000 °C for 1 h with an argon flow rate of 500 standard cubic centimeters per

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minute (sccm) under a constant furnace chamber pressure of 400 Torr. The morphologies of the as-synthesized products were examined with scanning electron microscopy (SEM, HITACHI, S-4700), and their nanostructures were analyzed with transmission electron microscopy (TEM, JEOL, JEM-2010).

3. Results and discussion

SEM and TEM images of the nanomaterials synthesized from the evaporation of the PbF_2 powders are shown in Fig. 1. The SEM images of Fig. 1a and b reveal the morphologies of the nanomaterials synthesized in the A (600 °C) and B (400 °C) regions, respectively; the nanomaterials synthesized in the A region are single-conical nanorods of which the one-side ends are attached to submicrometer-size single particles (see the inset of Fig. 1a), and the nanomaterials synthesized in the B region are radially directed nanorods. For the careful investigation into their nanostructrures, TEM images were taken for some selected from these nanorods. The TEM image shown in Fig. 1c demonstrates that these nanorods formed in the A region turn out to be partially filled nanotubes, and this image shows also that some droplets are present on the inner walls of the nanotubes. The inset of Fig. 1c is the magnified TEM image of the region drawn circularly by a dotted line, illustrating the ending part with the cap. The TEM images of two selected from the nanorods formed in the B region show a fully filled nanotube (or a nanocable) (Fig. 1d) and a partially filled nanotube (Fig. 1e); the inset of Fig. 1d shows that the fully filled nanotube has a cap. The morphology of these nanotubes is cylindrical. The nanotubes synthesized in this study are typically 10–40 nm in inner diameter and 5–7 nm in wall thickness.

TEM image, selected area electron diffraction (SAED) pattern, energy dispersive X-ray (EDX) spectra of the nanotubes under study are given in Fig. 2. The TEM image of Fig. 2a shows completely hollow and partially filled nanotubes. Fig. 2b and c are the SAED patterns obtained from the parts marked by A and B in (a), respectively. The SAED pattern (Fig. 2b) of the A-marked part in the completely hollow nanotube shows only halo rings; any spots related to crystalline phases are absent in this pattern. The presence of the halo rings in the SAED pattern implies that the nanotubes are amorphous. The SAED pattern (Fig. 2c) of the B-marked part in the partially filled nanotubes shows discrete spots over-

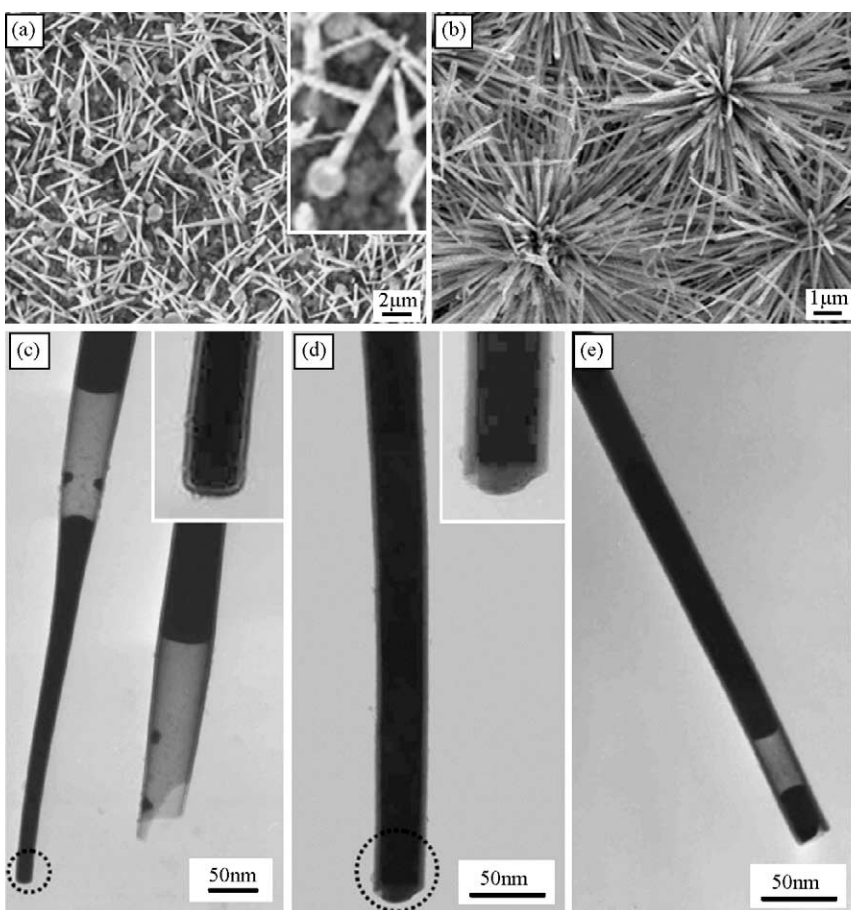

Fig. 1. SEM (a and b) and TEM (c–e) images of partially filled nanotubes. The SEM images of (a and b) exhibit the nanorods synthesized at 600 and 400 °C, respectively; the inset of (a) shows single-conical nanorods attached to single particles. The TEM images of (c–e) demonstrate two partially filled (conic) nanotubes, a fully filled (cylindrical) nanotube with the cap, and a partially filled (cylindrical) nanotube, respectively.