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# Journal of Alloys and Compounds

journal homepage: www.elsevier.com/locate/jalcom

## Growth of ZnO nanorods on graphite substrate and its application for Schottky diode



ALLOYS AND COMPOUNDS

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#### ARTICLE INFO

Article history: Received 28 October 2013 Received in revised form 12 May 2014 Accepted 16 May 2014 Available online 5 June 2014

Keywords: ZnO Nanorods Graphite Hydrothermal method Schottky diode

#### 1. Introduction

ZnO nanorods (NRs) have received much attention due to their direct wide band gap of 3.37 eV, high exciton binding energy of 60 meV, and large piezoelectric coefficient [1,2]. In particular, a preferred directional growth along the *c*-direction of the wurtzite crystal structure enables the ZnO crystals to be a promising constituent of various novel device applications, such as transistors, sensors, and piezoelectric, thermoelectric, electrochemical and optoelectronic devices [1,2]. The fabrication of ZnO NRs has been demonstrated on a variety of substrates including metals, semiconductors, insulators, and polymers [1–4]. Recently, accompanying with active research on the heterostructures between graphene and semiconductors [5,6], graphite also has received much attention as a potential substrate for the growth of semiconductors owing to its excellent mechanical and chemical stability, and high thermal and electrical conductivity [7]. Furthermore, graphite has a potential advantage for transferable substrate since it consists of a multi-layer system with nearly decoupled two dimensional (2-D) graphene planes [8]. There are some reports about the deposition of ZnO nanostructures on graphite substrate by the vapor phase method [9], electrochemical deposition [10], and hydrothermal growth combined with the thin film deposition method [11].

#### ABSTRACT

We report on the growth of ZnO nanorods (NRs) grown on graphite and silicon substrates via an allsolution process and also studied the characteristics of their heterojunctions. Structural investigations indicated that morphological and crystalline properties were not significantly different for the ZnO NRs on both substrates. However, optical properties from photoluminescence spectra showed that the ZnO NRs on graphite substrate contained more point defects than that on Si substrate. The ZnO NRs on both substrates showed typical rectification properties exhibiting successful diode formation. The heterojunction between the ZnO NRs and the graphite substrate showed a Schottky diode characteristic and photoresponse under ultraviolet illumination at a small reverse bias of -0.1 V. The results showed that the graphite substrate could be a good candidate for a Schottky contact electrode as well as a conducting substrate for electronic and optoelectronic applications of ZnO NRs.

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However, there are only a few investigations reporting the growth of ZnO NRs on graphite substrate by the all-solution-process method, especially using the hydrothermal method, even though this method has many advantages including a low temperature process which make it applicable to the integration and in situ fabrication of various devices [12,13]. For a compatible application of ZnO NRs to other devices, all-solution process method at a low temperature on graphite substrate should be possible, and its performance could be compared with conventional substrate, silicon (Si). In this paper, we report on the growth of ZnO NRs on graphite substrate, and on comparative study of the growth of ZnO NRs on Si substrate by using the all-solution process two-step hydrothermal method.

#### 2. Experimental details

#### 2.1. Experimental

The ZnO NRs were grown on highly-oriented pyrolytic graphite and *p*-type Si (100) substrates using an all-solution process two-step hydrothermal method involving the formation of a ZnO seed and main ZnO NR layers. Prior to growth, the substrates were rinsed sequentially in acetone, ethanol, and deionized (DI) water to remove contamination. A seed layer for the ZnO NRs was formed by dipping both substrates into 40 mM zinc acetate dihydrate [Zn(CH<sub>3</sub>COO)<sub>2</sub>·2H<sub>2</sub>O] dissolved in ethanol solution, followed by drying at 100 °C for 5 min. This resulted in around 40 nm-thick ZnO thin layers which are composed of the ZnO small size domains on both substrates. The ZnO NRs were grown by placing the seed layer-grown Si and graphite substrates into a mixed solution of 60 mM zinc nitrate hexahydrate [Zn(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O] and 60 mM hexamine [(CH<sub>2</sub>)<sub>6</sub>N<sub>4</sub>; HMT] in DI water at



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95 °C for 2 h. At a solution temperature above 70 °C, the solution started to become cloudy, indicating that a chemical reaction had begun started. After the reaction, the samples were cleaned in flowing DI water for 5 min.

#### 2.2. Characterizations

A structural characterization of the ZnO NRs was performed by field emissionscanning electron microscopy (FE-SEM) and X-ray diffraction (XRD) using Cu K $\alpha$ radiation. The optical properties of the ZnO NRs were studied using photoluminescence (PL) spectra, which were measured using a 325 nm continuous He-Cd laser (24 mW) at room temperature. The electrical properties of the ZnO NRs on both substrates were measured using a source meter. The current–voltage (*I–V*) characteristics of the ZnO NRs were measured in a dark box to minimize the generation of photocarriers [14].

#### 3. Results and discussion

Fig. 1 shows FE-SEM images of the ZnO NRs grown on the p-type Si (100) and graphite substrates. Without the seed layer, no deposit was observed on both substrates, even for a longer growth time. This indicates that the seed layer plays a critical role in the growth of ZnO NRs on the substrates. The diameter of the ZnO NRs on Si and graphite substrates are 70 nm and 50 nm, respectively. The diameter of the ZnO NRs grown on graphite substrate was slightly smaller than those on Si substrate. And the density of ZnO NRs on graphite is slightly larger than that on Si substrate as shown in Fig. 1(a) and (b). We suggest that this is due to the differences in the substrate used since the thermal conductivity and the surface energy are different for these two substrates. The thermal conductivity values of the Si (100) and graphite are 1.48 and 16–20 W/cm K, respectively [15]. And the surface energy values of the Si (100) plane and graphite are 2130 and 70–80 ergs/cm<sup>2</sup>, respectively [16,17]. These large differences in the thermal conductivity and surface energy of two substrates would affect on the formation of seed layer and growth of the ZnO NRs. The well-faceted six-sided surfaces, as shown in the insets of Fig. 1(a) and (b), were developed, and the top of the ZnO NRs on Si and graphite substrates shows a hexagonal facet. The facet indicates that the ZnO NRs are single crystal grown along the [0001] direction [18]. The lengths of the ZnO NRs on both substrates are similar and  $1.2 \,\mu$ m. The surface morphologies of the ZnO NRs on both substrates are similar. The growth mechanism of the ZnO NRs on the substrates using a solution containing zinc nitrate and HMT can be summarized in the following equations:

$$\begin{split} (CH_2)_6 N_4 + 6H_2 O &\rightarrow \ 6COH_2 + 4NH_3 \\ NH_3 + H_2 O &\rightarrow \ NH_3 \cdot H_2 O &\rightarrow \ NH_4^+ + OH^- \end{split}$$

$$Zn(NO_3)_2 \cdot 6H_2O \rightarrow Zn^{2+} + 2NO_2^- + 6H_2O_2^-$$

$$Zn^{2+} + 2OH^- \rightarrow ZnO + H_2OC$$

In the reaction, HMT plays a very complicated role in the solution by supplying an  $OH^-$  ion to the  $Zn^{2+}$  ion to form a ZnO crystal.

Fig. 2 shows the XRD patterns of the ZnO NRs grown on *p*-Si (100) and graphite substrates. The diffraction peaks in the patterns were indexed to the standard hexagonal wurtzite structured ZnO (JCPDS 36-1451: a = 0.3249 nm, c = 0.5206 nm) and graphite (JCPDS 41-1487: a = 0.2461 nm, c = 0.6708 nm), respectively. All the diffraction peaks from the ZnO NRs on both substrates were identified as corresponding to the (100), (002), (101), (102), and (110) planes of the hexagonal wurtzite ZnO structure and no additional peaks corresponding to carbon-related alloys were observed, as shown in Fig. 2. The main (002) peak intensity of the ZnO NRs on both substrates were almost the same, indicating that there was no significant difference in the crystalline quality.

Fig. 3(a) shows the PL spectra of the ZnO NRs measured at room temperature. The PL spectra of both samples exhibit two main emission peaks: sharp peak in the ultraviolet (380 nm) and broad peak in the visible ranges, which correspond to the band edge emission of ZnO and deep level emission, respectively. The emission band at the visible wavelength range of the ZnO NRs is attributed to the ensemble of radiative recombination through point defects in a ZnO crystal, such as an oxygen vacancy, an oxygen interstitial, a zinc vacancy, a zinc interstitial, and anti-site defects [19,20] The elementary PL spectra shown by dashed lines DL-1 and DL-2 were obtained by deconvoluting the PL spectra from the ZnO NRs on graphite substrate and were mainly attributed to



Fig. 1. FE-SEM images of ZnO NRs grown on (a), (c) p-Si (100), and (b), (d) graphite substrates.

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