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Ga-doped ZnO nanowire nanogenerator as self-powered/active humidity sensor with high sensitivity and fast response

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ABSTRACT

High sensitivity and fast response piezo-humidity sensing have been realized from Ga-doped ZnO nanowire (NW) nanogenerator (NG) as self-powered/active gas sensor. The piezoelectric output generated by Ga-doped ZnO NW NG can not only act as a power source for driving the device, but also as a sensing signal for detecting humidity. Upon exposure to 80% relative humidity (RH) at room temperature, the piezoelectric output of Ga-doped ZnO NWs decreases from 0.56 V (in 45% RH) to 0.12 V, and the sensitivity is up to 358, higher than that of undoped ZnO NWs. In addition, the response time of Ga-doped ZnO NWs is 5 s, much shorter than that of undoped ZnO NWs. Such high performance can be ascribed to that Ga^{3+} ions can provide many donor defects and produce high local charge density/strong electrostatic field. The present results demonstrate a feasible approach for realizing high performance self-powered/active humidity sensor.

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1. Introduction

Humidity sensors have important applications in human's life and industrial production, and metal oxide nanostructures have been widely investigated for their high sensitivity against humidity. Nowadays, working at room temperature and fast response are two important issues for the nanoscale humidity sensors [1–5]. On the other hand, ZnO nanostructures have been widely used in solar cell, fluorescence imaging, photocatalysis and gas sensor duo to its wide-gap and high exciton binding energy [6–9]. In our previous work, by fabricating the piezoelectric and gas sensing characteristics of ZnO nanowire (NW), an unpackaged ZnO NW piezo-nanogenerator (NG) as self-powered/active gas sensor has been firstly established, in which the piezoelectric output of ZnO NW NG acts as both the power source and gas sensing signal [10]. This new device shows the potential for realizing room-temperature humidity sensing. At room temperature, the piezoelectric output of the device is different under different relative humidity (RH). At the current stage, shortening the response time of the device still remains a challenge.

For traditional gas sensors, it has been demonstrated that doping Ga element in ZnO nanostructures is an effective and simple method for enhancing their humidity sensing performance [11]. Ga^{3+} can easily substitute Zn^{2+} without any lattice distortion for their smaller ionic radius [11,12]. The ionic radius of Ga^{3+} and Zn^{2+} is 0.047 and 0.06 nm, respectively [13]. The covalent bond lengths of Ga–O and Zn–O are 0.192 and 0.197 nm, respectively [13]. A slightly smaller bond length of Ga–O than that of Zn–O is expected to make the deformation of the ZnO lattice small. When the Zn atoms are substituted by Ga, the Fermi level shifts from the valence band maximum to the bottom of the conduction band, resulting in a shallow donor level at the bottom of the conduction band [13,14]. It has also been reported that at high doping concentration (>3.8%), the self-compensation mechanism can cause acceptor level in the conduction band, reduce carrier concentrations and decrease the conductivity [15]. At the same time, Ga^{3+} ions have more positive charges than Zn^{2+} [16]. Ga dopant incorporating into ZnO materials can product more shallow/native defects (oxygen vacancies), release free electrons and contribute to the high conducting, and improve gas sensing behavior, such as high sensitivity and fast response [17–19].

In this paper, Ga-doped ZnO NW NG as self-powered/active humidity sensor exhibits high sensitivity and fast response. The sensitivity is higher than that of undoped ZnO NWs, and the response time is shorter. In humid environment, the water molecules can displace the adsorbed oxygen ions on the surface of

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Ga-doped ZnO NWs, which increase the carrier density on the surface, strengthen the piezoelectric screening effect and lower down the piezoelectric output [20–22]. The present results demonstrate a feasible approach for realizing high performance self-powered/active humidity sensor.

2. Experimental

The vertically-aligned Ga-doped ZnO NWs were prepared by a hydrothermal method. Prior to growth, a piece of Ti foil (thickness is 0.1 mm) as the substrate was cleaned with deionized water and alcohol, and dried at 60 °C. 0.5 g of $Zn(NO_3)_2 \cdot 6H_2O$ (40 mM) and 0.0072 g of $Ga(NO_3)_3 \cdot xH_2O$ (0.16 mM) were dissolved in 38 mL of deionized water (The molar ratio of Ga:Zn is 1:250.). After being evenly dissolved, 2 mL of $NH_3 \cdot H_2O$ was added into the solution and stirred for 10 s at room temperature. The solution was then transferred into a Teflon-lined stainless-steel autoclave, and the Ti substrate was immersed in. The autoclave was sealed and maintained at 80 °C for 24 h. After cooling down to room temperature, the Ti substrate coated with vertically-aligned Ga-doped ZnO NWs was removed from the solution, washed with deionized water and ethanol, and dried at 60 °C. The doping concentration is controlled by dissolving different concentrations of $Ga(NO_3)_3 \cdot xH_2O$ in the solution.

As shown in Fig. 1a, the device is made up of three major parts: Ga-doped ZnO NWs on Ti substrate, Al foil (thickness is 0.05 mm) and Kapton boards. Ti foil is as both the substrate of the NWs and the current collector. The Al foil on the tips of Ga-doped ZnO NWs is the counter electrode. To ensure the device, it is tightly fixed between two sheets of Kapton boards as the support frames. Fig. 1b shows the measurement system. The device is connected to a low-noise preamplifier (Model SR560, Stanford Research Systems) through coaxial lines. A compressive force is applied on the device by a hammer, which is actuated by a stepper motor moving along a guide rail (the movement of the motor can be controlled by programming). The area of the hammer (the diameter is about 3 cm) is larger than that of the device. The compressive force applied on the device is 34 N at 1.4 Hz. Under externally applied compressive deformation, the piezoelectric output of the device can act as both the power source and the humidity sensing signal.

3. Results and discussion

Fig. 2a–c are scanning electron microscopy (SEM) images of Ga-doped ZnO NWs on the top view with different Ga doping concentrations (mole ratio). The average diameter of Ga-doped ZnO (1:500, 1:250 and 1:50) NWs is about 163, 241 and 1950 nm, respectively. It can be confirmed that the diameter of Ga-doped ZnO NWs increases with increasing doping concentration. Fig. 2d is SEM image of Ga-doped ZnO (1:250) NWs on the side view, revealing that Ga-doped ZnO NWs are vertically aligned on the substrate and the length of the NWs is about 1.83 μm. Fig. 2e is transmission electron microscopy (TEM) image of one single Ga-doped ZnO (1:250) NW, indicating that the surface of the NW is smooth. Fig. 2f is high resolution TEM image of Ga-doped ZnO (1:250) NW. The lattice spacing of 0.52 nm is consistent with (001) crystal plane of wurtzite structural ZnO, demonstrating that the growth direction of the NW is along (0001). The corresponding select area electron diffraction (SAED) pattern is inserted in Fig. 2f, showing a good single crystalline nature of Ga-doped ZnO.

Fig. 3a is energy dispersive spectrometer (EDS) spectrum of Ga-doped ZnO (1:250) NWs. Three elements (O, Zn and Ga) appear at the selected region, and similar EDS results can be obtained from other regions. From the EDS spectrum, it can be calculated that the atomic percentage of Ga in Ga-doped ZnO NWs (Ga:Zn in the preparation process is 1:250) is 1.18%. These results confirm that the product is Ga-doped ZnO NWs. X-ray powder diffraction (XRD; D/max 2500 V, Cu Kα radiation, $\lambda = 1.5405$ Å) is used to characterize the crystal phase of Ga-doped ZnO NWs, and XRD patterns of undoped ZnO and 1.18% Ga-doped ZnO NWs are shown in Fig. 3b. From the two samples, the peaks corresponding to (100), (002), (101), (102), (110) and (103) planes can be indexed to hexagonal wurtzite structure of ZnO (JCPDS # 36-1451), and the peaks indexed to Ti (JCPDS # 44-1294) are raised from Ti substrate. The (002) peak of the NWs is very strong, further confirming the NWs are vertically aligned on the substrate. As shown in the inset of Fig. 3b, the (002) peak is located at 34.39° for undoped ZnO and shifts to 34.53° for 1.18% Ga-doped ZnO NWs. The peaks shift is related to a reduction of interplanar spaces probably caused by the different ionic charge of the involved ions (Ga^{3+} and Zn^{2+}) [23].

Fig. 4a is the piezoelectric output voltage of 1.18% Ga-doped ZnO NWs at room temperature under the same deformation against different RH. The output voltage is dependent on the humidity, and the piezoelectric voltage dramatically decreases as the RH increases. In different humidity environment, the output voltage is different. As the humidity increases from 45% to 80%, the voltage decreases from 0.56 to 0.12 V. The enlarged views of the output voltage are shown in Fig. 4b–h. Under 45%, 70%, 75%, 80%, 65%, 60% and 50% RH, the piezoelectric output voltage of the device is about 0.56, 0.27, 0.20, 0.12, 0.31, 0.39 and 0.51 V, respectively. The relationship between the RH and the output voltage is shown in Fig. 4i. Similar to the traditional definition of the sensitivity of gas sensors

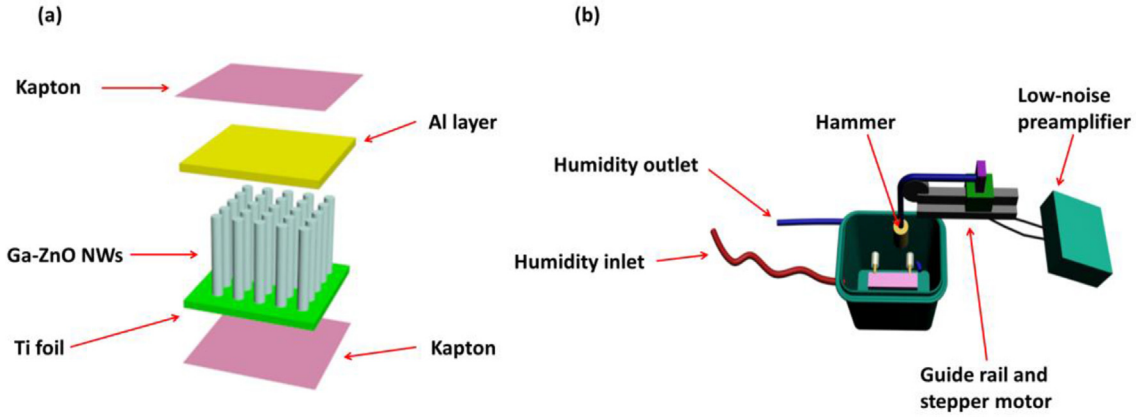

Fig. 1. (a) The device structure of Ga-doped ZnO NW NG as self-powered/active humidity sensor. (b) The measurement system.

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