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Fabrication of Intrinsic Zinc Oxide-Coated, Aluminium-Doped Zinc Oxide Nanorod Array-Based Ultraviolet Photoconductive Sensors

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Abstract. Fabrication and performance of metal-semiconductor-metal (MSM)–type intrinsic zinc oxide (ZnO)-coated, aluminium (Al)-doped ZnO nanorod array-based ultraviolet photoconductive sensors were reported and discussed. The Al-doped ZnO nanorod arrays were prepared using sonicated sol-gel immersion method. The coating process of intrinsic ZnO onto Al-doped ZnO nanorod arrays was performed using radio-frequency (RF) magnetron sputtering at different deposition times varying from 0 to 10 min. We observed that responsivity of the sensors decreased with increasing intrinsic ZnO deposition time, decreasing from 4.81 A/W without coating to 1.37 A/W after 10 min of coating. Interestingly, the sensitivity of the sensors improved with intrinsic ZnO coating, having a maximum value of 19.0 after 1 min coating.

1. Introduction

Nanostructured zinc oxide (ZnO) is one of the most promising semiconductor materials for applications in solar cells, and various sensors [1-5]. Particularly in ultraviolet (UV) sensor application, much attention has been paid to nanostructured ZnO because of chemical stability, large surface area, good oxidation resistibility, great biocompatibility, and high conductivity. Currently, major investigations have focused on the development of one-dimensional (1D) zinc oxide (ZnO) nanostructures, such as nanorod arrays, which have several advantages for ultraviolet (UV) photoconductive sensor applications including wide bandgap, high surface area, and carrier confinement in two dimensions that could improve the sensor performance. The UV sensing properties of ZnO nanorod arrays are mainly dependent on their preparation methods, alignments, sizes, and surface conditions [4, 6-8]. In this study, we fabricated UV photoconductive sensor aluminium (Al)-doped ZnO nanorod arrays, which have been decorated with sputtered intrinsic ZnO at different deposition time and their performances were investigated. Interestingly, the sensitivity and the response of the devices improved with the addition of sputtered intrinsic ZnO on the nanorod surface.

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2. Experimental Procedure

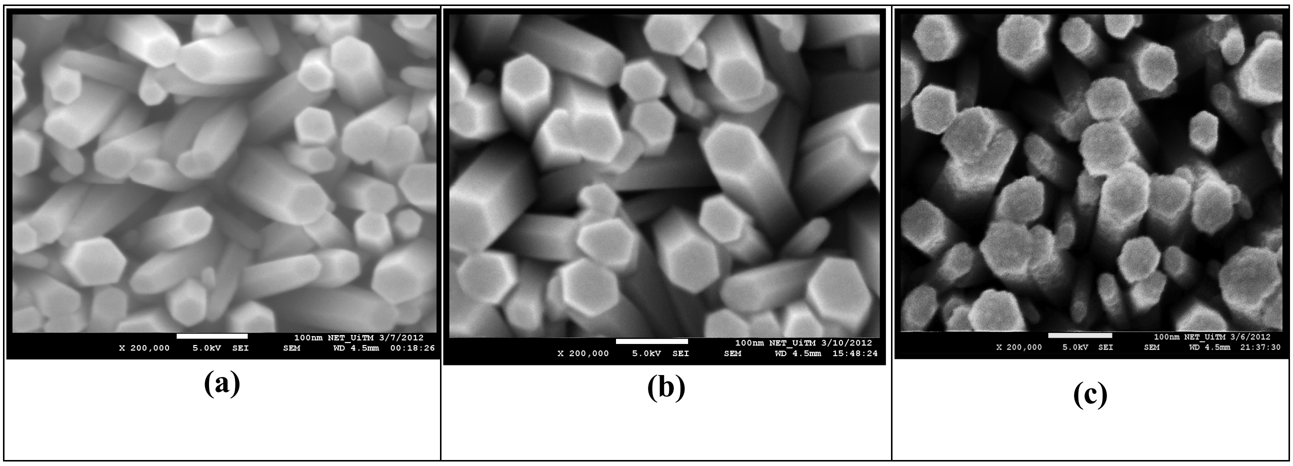
Al-doped ZnO nanorod arrays were grown on a glass substrate coated with an Al-doped ZnO seed layer using a sonicated sol-gel immersion method [9]. The Al-doped ZnO thin film, which was used as a seed layer, was prepared on the glass substrate using sol-gel spin-coating [10]. The Al-doped ZnO nanorod arrays were grown on the seed-layer-coated glass substrate using aqueous solutions of 0.1 M zinc nitrate hexahydrate ($Zn(NO_3)_2{\cdot}6H_2O$; 98 % purity; Systerm), 0.1 M hexamethylenetetramine (HMT; $C_6H_{12}N_4$; 99 % purity; Aldrich), and 0.001 M aluminium nitrate nonahydrate ($Al(NO_3)_3{\cdot}9H_2O$; 98 % purity; Analar). These reagents were dissolved and reacted in a beaker filled with deionised (DI) water using an ultrasonic water bath for 30 min (Hwasin Technology Powersonic 405; 40 kHz). The temperature for the sonication process was 50 ºC. The solution was then stirred and aged for 3 h at room temperature. The resulting solution was poured into vessels with volume capacities of 100 ml, into which the seed-layer-coated glass substrates were placed on the bottom. Next, the sealed vessels were immersed in a hot water bath at 95 ºC. The samples were immersed for 50 min. The nanorods deposited on the substrates were rinsed with DI water and dried in a furnace at 150 ºC for 10 min. Then, Al-doped ZnO nanorod arrays were coated with intrinsic ZnO (4-inch ZnO target: purity 99.99 %; Process Materials Inc.) using radio-frequency (RF) magnetron sputtering (SNTEK) at an RF power of 50 W, before annealing in air at 500 °C for 1 h. The coating process was performed after 10 min of pre-coating in 45 sccm argon (Ar) and 20 sccm oxygen (O_2) which flowed at a deposition pressure of 0.67 Pa with varying deposition times of 1, 5, and 10 min. The coated intrinsic layer thickness was estimated as 5, 18, and 35 nm, respectively. Next, 60-nm-thick Al contacts were deposited on the nanorod array films using a thermal evaporator to produce a metal–semiconductor–metal (MSM)–type UV photoconductive sensor. The deposition of metal contacts was performed at a chamber pressure of 4×10^{-4} Pa. The separation between the metal contacts was fixed at 2.00 mm for all samples using a metal mask.

The surface morphology and cross-sectional images of the samples were taken with a field-emission scanning electron microscopy (FESEM; JEOL; Model: JSM-7600F). The current-voltage (I-V) characteristics of the UV photoconductive sensors were investigated using a two-probe I-V measurement system (Keithley; Model: 2400). The UV photoresponse measurements of the fabricated sensors were taken using a UV photocurrent measurement system (Keithley; Model: 2400) operating at 365 nm with a power density of 750 µW/cm2 and a bias voltage of 10 V.

3. Results and Discussion

The FESEM images of samples coated for 0 (uncoated), 1 and 10 min are shown in Figures 1(a), 1(b) and 1(c), respectively. The image in Figure 1(a) shows that the nanorods grew uniformly on the seed layer substrate with an average diameter of 61 nm. When the nanorods were coated with sputtered intrinsic ZnO for 1 min, no significant change in morphology was observed, as shown in Figure 1(b). However, the surface of nanorods was modified when the nanorod were coated with sputtered intrinsic ZnO for 10 min, which can be seen in Figure 1(c). The image in Figure 1(c) shows that the surface of nanorods became rough with intrinsic coating for 10 min as a result from coated intrinsic ZnO particles through sputtering process. These images in Figures 1 show that the ZnO particles are clearly observed for intrinsic ZnO coatings deposited for longer periods of time. These images also reveal that sputtered, intrinsic ZnO were uniformly deposited on surface of the nanorod arrays.

Figure 2 depicts I-V curves at different deposition times. The I-V curves indicate that the Al metal connections form good ohmic contacts with both uncoated and sputtered, intrinsic ZnO-coated Al-doped ZnO nanorod arrays. The spectra reveal that the current intensity with respect to the voltage decreases when the deposition time of the intrinsic ZnO layer was increased. The resistance of the samples was calculated to be 0.16, 0.27, 0.31, and 0.47 MΩ after the Al-doped ZnO nanorod arrays were coated with intrinsic ZnO at deposition times of 0 (uncoated),1, 5, and 10 min, respectively. The increase in resistance indicates that a thicker, lower-conductivity layer is deposited onto the nanorod surface as deposition time increases [7].



(a) **(b)** **(c)**

Figure 1, The FESEM Images of Al-Doped ZnO Nanorod Arrays Prepared at an Immersion Time of 50 min and Coated with Intrinsic ZnO at Deposition Times of (a) 0, (b) 1, and (c) 10 min.

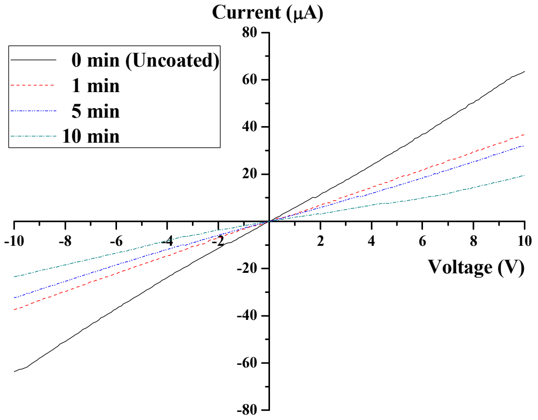


Figure 2, The I-V Characteristic of Al-Doped ZnO Nanorod Arrays Prepared at 50 min After Being Coated With Intrinsic ZnO Using RF Magnetron Sputtering at Deposition Times of 1, 5, and 10 min.

The photocurrent measurement spectra of the Al-doped ZnO nanorod arrays deposited with intrinsic ZnO for 1, 5, and 10 min are shown in Figure 3. The results show that the photocurrent of the nanorod arrays decreased for longer depositions of intrinsic ZnO coatings. The photocurrent (dark current) of the Al-doped ZnO nanorods that were deposited with intrinsic ZnO at deposition times of 0 (uncoated), 1, 5, and 10 min are 2.39 × 10^{-4} (2.26 × 10^{-5}), 1.42 × 10-4 (7.50 × 10^{-6}), 1.03 × 10^{-4} (8.53 × 10^{-6}) and 6.69 × 10-5 (5.53 × 10^{-6}) A, respectively. From the photocurrent spectra, the responsivity of the sensors was estimated using following equation [11]:

$$R = \frac{I_{ph} - I_{dark}}{P_{op}} , \qquad (1)$$

where I_{ph} is the photocurrent, I_{dark} is the dark current, and P_{op} is the optical power of the UV source.The responsivity of the sensors also decreased with ZnO deposition coating times, with values of 4.81, 3.00, 2.10, and 1.37 A/W after 0 (uncoated), 1, 5, and 10 min coatings, respectively. The results show that the responsivity decreases with intrinsic ZnO coatings due to an increase in the resistance of the nanorod arrays. However, the sensitivity of the devices improved with sputtered, intrinsic ZnO coatings. The sensitivity of the sensors, defined as the photocurrent-to-dark-current ratio, are 10.6, 19.0, 12.0 and 12.1 for Al-doped ZnO nanorod arrays coated with sputtered intrinsic ZnO at deposition time of 0 (uncoated), 1, 5, and 10 min.

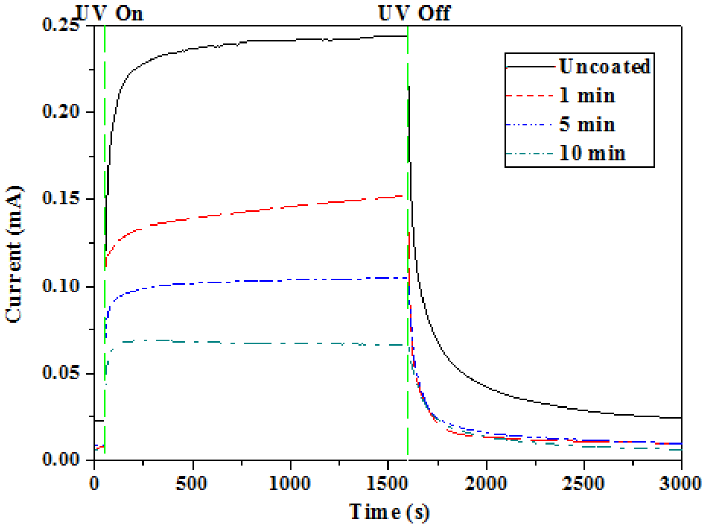


Figure 3, Photoresponse Spectra of the UV Photoconductive Sensor Using Al-Doped ZnO Nanorod Arrays Prepared at an Immersion Time of 50 min and Coated With Intrinsic ZnO Layer at Different Deposition Times Under UV Illumination (365 nm, 750 μW/cm2) and a 10 V Bias.

The rise (decay) time constants of the sensor were calculated from the photocurrent spectra using the following equations:

$$I = I_0\left[1-\exp\left(-\frac{t}{\tau_r}\right)\right] \text{: rise process with UV illumination on} \qquad (2)$$

$$I = I_0 \exp\left(-\frac{t}{\tau_d}\right) \qquad \text{: decay process with UV illumination off.} \quad (3)$$

Here, I is the magnitude of the current, I_0 is the saturated photocurrent, t is the time, τ_r is the rise time constant and τ_d is the decay time constant. It was found that the rise (decay) time constants of the sensors were 10 (26), 4 (15), 7 (20), and 7 (40) s after ZnO coatings made for 0 (uncoated), 1, 5, and 10 min, respectively. Interestingly, these findings show that these rise time constants are lower than those of uncoated Al-doped ZnO nanorod arrays, suggesting that the intrinsic ZnO layer plays an important role in producing a low-conductivity barrier that controls electron flow when switched on and off under UV light. However, we also observed that the increase in deposition time resulted in thicker intrinsic ZnO coatings on the Al-doped ZnO nanorod arrays. For these samples, the rise (decay) time constants increased. This condition could be attributed to defects generated by the intrinsic layers, which trap carriers and increase the response time of the UV sensors.

4. Conclusion

The UV photoconductive sensors were successfully fabricated using intrinsic ZnO decorated, Al-doped ZnO nanorod arrays. FESEM images reveal that the morphology of Al-doped ZnO nanorod arrays were modified after intrinsic ZnO coatings. The I-V measurement indicates that the resistance of the sensors increased after coating process at longer deposition times. By sputtered coating with intrinsic ZnO, we observed that sensitivity and response time of the sensor were improved. The results indicate that performance of the Al-doped ZnO nanorod array-based UV photoconductive sensor could be modulated by intrinsic ZnO layer coating on the nanorod surface, which may controls the electron flow when switched on and off under UV light.

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