



Fabrication of an ultraviolet photoconductive sensor using novel nanostructured, nanohole-enhanced, aligned aluminium-doped zinc oxide nanorod arrays at low immersion times

M.H. Mamat^{a,b,*}, M.F. Malek^a, N.N. Hafizah^b, Z. Khusaimi^b, M.Z. Musa^a, M. Rusop^{a,b}

^a NANO-ElecTronic Centre (NET), Faculty of Electrical Engineering, Universiti Teknologi MARA (UiTM), 40450 Shah Alam, Selangor, Malaysia

^b NANO-SciTech Centre (NST), Institute of Science (IOS), Universiti Teknologi MARA (UiTM), 40450 Shah Alam, Selangor, Malaysia

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ABSTRACT

Novel nanostructured thin-film arrays of nanohole-enhanced aligned aluminium (Al)-doped zinc oxide (ZnO) nanorods were prepared using sonicated sol-gel and immersion methods for metal-semiconductor-metal (MSM)-type ultraviolet (UV) photoconductive sensor applications. These nanorod arrays were grown on a glass substrate coated with ZnO nanoparticle thin film as a seed layer at immersion times ranging from 10 to 120 min. Notably, the nanoholes appeared on the nanorods after the annealing process due to the evaporation of water and impurities. The photocurrent properties for the Al-doped ZnO nanorod arrays were significantly improved, increasing by more than five-fold compared to the seed layer when the sensor was illuminated with 365 nm UV light at a density of 750 $\mu\text{A}/\text{cm}^2$. Interestingly, the highest responsivity using the Al-doped ZnO nanorod arrays was 1350.84 A/W with an improved photocurrent-to-dark current ratio of 22.7, achieved for the samples prepared with a 50 min immersion time and a gap between metal contacts of 0.07 mm. Our results show that high-performance UV photoconductive sensors can be achieved using a novel structure of nanohole-enhanced aligned Al-doped ZnO nanorod arrays prepared at low immersion times which reduces fabrication time and cost.

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1. Introduction

Nanostructured zinc oxide (ZnO) is currently receiving a large amount of attention in device fabrication due to its morphology variations and superior physical and chemical properties. Particularly as an ultraviolet (UV) photoconductive sensor, nanostructured ZnO is promising because of its wide band gap of 3.3 eV, high surface-to-volume ratio, excellent radiation hardness, high electron mobility, and surface chemistry [1,2]. One of the advantages of using the nanostructured ZnO is that numerous types of preparation methods can be used. These preparation techniques including sputtering [3], chemical vapour deposition [4,5], metal-organic chemical vapour deposition (MOCVD) [6,7], and solution-based methods [8–12].

Recently, ZnO nanorod arrays have emerged as potential nanostructured materials for UV sensor applications due to their surface area availability and carrier transport properties. To increase the

performance of the UV sensor, the surface area must be increased and the surface conditions of the nanorods must be enhanced. The surface area of the sensor can be improved using nanorod arrays with small diameter sizes and high nanorod densities [13,14]. High-surface-to-volume-ratio nanorod array-based UV sensors have shown improved UV sensing performance [13]. In addition, the surface area of nanostructures can be increased using strategies such as using branched nanostructures or introducing pores into the film. For example, Mridha et al. demonstrated that nanopores in a ZnO nanoparticle thin film improve the photoresponse and photocurrent of a sensor [15]. On the other hand, the surface condition of the nanorods could be improved with surface treatments such as annealing [16,17], metal coatings [18], passivation layer coatings [19], and chemical treatments [20]. These treatments are reported to give different surface chemistries, which influence the surface interaction with different environments, including oxygen adsorption and desorption processes. Park et al. reported that having a rough nanowire surface induced by a chemical treatment significantly improved the photocurrent of the ZnO due to the enhanced oxygen trapping efficiency of the rough surface compared with a smooth surface [20]. These oxygen adsorption and desorption processes are reported to have a strong influence on

* Corresponding author. Tel.: +60 3 55211883; fax: +60 3 55443870.

E-mail addresses: hafiz_030@yahoo.com, mhmamat@salam.uitm.edu.my (M.H. Mamat).

the performance of UV photoconductive sensors [21–23]. Therefore, the surface characteristics of nanorods are of great importance to the UV photoconductive sensor.

In this paper, we discuss the fabrication of a UV photoconductive sensor using nanohole-enhanced aligned ZnO nanorod arrays via a simple technique using ultrasonic-assisted sol-gel and immersion methods at low immersion times. The importance of this study is the amplification of sensor performance using thin ZnO nanorod arrays optimised at low immersion times using a hydrothermal process, resulting in a low-cost, time-saving fabrication process. Notably, and for the first time, nanoholes were produced on the nanorod surface simply using an annealing process for 1 h in ambient without further chemical treatment. We also discuss the performance of a UV sensor fabricated using Al-doped ZnO nanorod arrays prepared at different low immersion times up to 120 min, resulting in different thicknesses and surface conditions. These results have rarely been reported for Al-doped ZnO nanorod array-based UV sensors fabricated using solution-based techniques. Our study shows that nanohole-enhanced aligned ZnO nanorod arrays prepared at immersion times of 50 min produce the highest responsivity as fabricated UV photoconductive sensors when compared with other samples. However, we found that the UV photoconductive sensor using Al-doped ZnO nanorod arrays prepared with an immersion time of 20 min showed a high photocurrent-to-dark-current ratio, which may be attributed to the greater quantity of nanoholes on the nanorod surfaces. To the best of our knowledge, the performance of a UV sensor using nanohole-enhanced aligned Al-doped ZnO nanorod arrays prepared using hydrothermal methods at low immersion times, yielding extremely thin nanorod arrays, has not yet been discussed elsewhere.

2. Experimental details

The UV photoconductive sensors were fabricated using Al-doped ZnO nanorod arrays prepared using the sonicated sol-gel immersion method [8]. The fabrication process consists of two steps: Al-doped ZnO nanoparticle thin film preparation or seed layer preparation and Al-doped ZnO nanorod preparation. First, the Al-doped ZnO nanoparticle thin films were prepared on glass substrates using a sol-gel spin-coating method [24]. Next, 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 ($\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$; 98% purity; System), 0.1 M hexamethylenetetramine (HMT; $\text{C}_6\text{H}_{12}\text{N}_4$; 99% purity; Aldrich), and 0.001 M aluminium nitrate nonahydrate ($\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$; 98% purity; Analar). These reagents were dissolved and reacted in a beaker filled with distilled water using an ultrasonic water bath for 30 min (Hwasin Technology Powersonic 405; 40 kHz). The temperature for the sonication process was 50 °C. 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 °C. The samples were immersed for different lengths of time, from 10 to 120 min. The nanorods deposited on the substrates were rinsed with distilled water and dried in a furnace at 150 °C for 10 min. Subsequently, the samples were annealed for 1 h in ambient conditions at 500 °C. The samples prepared for 50 min were also annealed at different annealing temperatures (300–500 °C in ambient conditions for 1 h), annealing times (0.5–2 h at 500 °C in ambient conditions), and oxygen flow rates (0.2–1.0 L/min at 500 °C for 1 h). In addition, undoped ZnO nanorod arrays were prepared at immersion time of 50 min and were annealed in ambient condition at 500 °C for 1 h. Next, 60-nm-thick aluminium (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. Using a similar procedure, silver (Ag) and gold (Au) contacts were also deposited on the Al-doped ZnO nanorods immersed for 50 min. The thickness of these metal contacts was fixed at 60 nm. The separation between the metal contacts was fixed at 2.00 mm for all samples using a metal mask. For the Al-doped ZnO nanorods immersed for 50 min, the separation between metal contacts (for Al contacts) was varied between 0.07 mm, 0.50 mm, and 2.00 mm.

The surface morphology and cross-sectional images of the samples were taken with a field-emission scanning electron microscope (FESEM, JEOL JSM-7600F). The crystallinity of the nanorod arrays was characterised using an X-ray diffractometer (XRD, Panalytical X'pert PRO). The photoluminescence (PL) properties of the synthesised nanorods were investigated using a PL spectrophotometer with a helium–cadmium (He–Cd) excitation laser at 325 nm (Horiba Jobin Yvon-79 DU420A-OE-325). The Raman spectra of the Al-doped ZnO nanorod arrays were measured using micro-Raman spectroscopy with an argon (Ar) laser operating at 514 nm as the excitation source (Horiba Jobin Yvon-79 DU420A-OE-325). The current–voltage (I – V) characteristics of the UV photoconductive sensors were investigated using a two-probe I – V measurement system (Keithley 2400). The composition analysis of the Al-doped ZnO nanorod arrays was performed using an energy-dispersive X-ray spectroscopy (EDS, Oxford Instruments X-Max) attached to the FESEM. The UV photoresponse measurements of the fabricated sensors were taken using a UV photocurrent measurement system (Keithley 2400) operating at 365 nm with a power density of $750 \mu\text{W}/\text{cm}^2$ and a bias voltage of 10 V.

3. Results and discussion

Surface morphologies of Al-doped ZnO nanorod arrays prepared at different immersion times are shown in Fig. 1(a–g). Fig. 1(a) depicts a field-emission scanning electron microscopy (FESEM) image of a nanorod array after 10 min of immersion. The image shows that the nanorods grew uniformly on the seed layer substrate with an average diameter of 51 nm. When the immersion times were increased to 20, 30, 40, 50, 60, and 120 min, as shown in Fig. 1(b), (c), (d), (e), (f), and (g), respectively, the morphology remained the same with a slight increase in average diameter from 57 to 63 nm. The average diameter for the nanorods immersed at 20, 30, 40, 50, 60, and 120 min are 57, 60, 60, 61, 62 and 63 nm, respectively. We also observed the growth of the nanorods along the lateral direction, or c -axis, through the observation of tilted nanorods with improved lengths after longer immersion times.

To confirm the lateral direction growth, we observed cross-sectional images using FESEM. Fig. 2(a–g) shows cross-sectional images of Al-doped ZnO nanorod arrays at different immersion times. The images show that all of the nanorod arrays were deposited on the seed-layer-coated glass substrate with good uniformity. Fig. 2(a) shows cross-sectional images of Al-doped ZnO nanorods after 10 min of immersion. The image shows that the sample contains ultra-thin nanorod arrays that are approximately 78 nm in length. The overall film thickness was 259 nm, and the seed layer had an average thickness of 181 nm. The nanorods grew rapidly after 20 min of immersion, after which the film thickness reached 513 nm, as shown in Fig. 2(b). The continuity of nanorod growth in the lateral direction was also observed for nanorod arrays immersed for 30, 40, 50, 60 and 120 min, with increased nanorod film thicknesses of 545, 585, 603, 637, and 688 nm, respectively. The cross-sectional images of nanorod arrays after 30, 40, 50, 60, and 120 min of immersion are shown in Fig. 2(c), (d), (e), (f), and (g), respectively. This finding indicates that the nanorod growth is mainly along the c -axis as immersion time is increased.

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