Al_2O_3/ZnO NON-PLANAR HETEROSTRUCTURES FOR UV RADIATION SENSOR APPLICATIONS

Nicolae MAGARIU [1], Alexandr SEREACOV [1], Mani Teja BODDULURI [2], Anna GAPEEVA [3]

[1] *Technical University of Moldova, Faculty of Computers, Informatics and Microelectronics, Department of Microelectronics and Biomedical Engineering, Center for Nanotechnology and Nanosensors, PhD School Computer Science, Electronics and Energy, Chisinau, Republic of Moldova*
[2] *Materials and Processes for Nanosystem Technologies, Institute for Materials Science, Kiel University, Kaiserstrasse.2, D-24143 Kiel, Germany*
[3] *Chair for Functional Nanomaterials, Faculty of Engineering, Institute for Materials Science, Kiel University, Kaiser Str. 2, D-24143, Kiel, Germany*

Magariu Nicolae: nicolae.magariu@mib.utm.md

Abstract. *In this work the characteristics of aluminum oxide/zinc oxide heterostructures based sensors used for UV radiation detection were investigated. The UV response dependent on the distance of the UV source versus sample and relative humidity changes was studied. The dependency between the response time and the distance to the UV source was determined.*

Keywords: *heterostructures, aluminum oxide, zinc oxide, UV sensor, humidity.*

Introduction

Zinc oxide (ZnO) is a *n*-type semiconductor that has a bandgap of between 3.1 and 3.3 eV [1-4]. In order to modify the bandgap, various impurities such as titanium oxide, palladium [2], cadmium, europium, silver as well as functionalization with noble metals such as Au and Pt can be used. All these changes are performed to develop gas sensors or ultraviolet (UV) radiation detectors, which can be incorporated into various portable devices for the detection of gases such as ammonia, methane, ethanol, hydrogen, carbon dioxide, butanol, propanol as well as for UV radiation detection in different ambient.

Experimental

In order to obtain zinc oxide, one of the most effective and inexpensive methods of manufacturing a sensor based on semiconductor oxide was chosen, namely the synthesis from chemical solutions (SCS) method [10]. This is a straightforward method that also allows impurity doping during the zinc oxide production. The substrate glass, on which the zinc oxide is deposited, is cleaned preventively and then sensitized in a $SnCl_2 \cdot 2H_2O$/HCl solution. Zinc sulphate is used as a cationic precursor and sodium hydroxide is used as an initial reagent to form a complex solution. All solutions of chemical reagents were of purity higher than 99.9% with no further purification [5]. The $ZnSO_4$ solution was gradually added to the NaOH solution which was continuously stirred with a magnetic stirrer until the solution became transparent. The impurity doping can be achieved by introducing impurity ions into the complex solution [6].

Low temperature thermal atomic layer deposition (ALD) of Al_2O_3 deposition was carried out using Picosun's R-200. Trimethylaluminum (TMA) was used as aluminum source and H_2O to oxidize chemisorbed TMA. Alternating pulses of aforesaid precursors pulsed into reactor chamber for 0.1 s per pulse to grow monolayers of Al_2O_3. Nitrogen was used to transport the precursor vapors into the reaction chamber and to purge byproducts from the reactor.

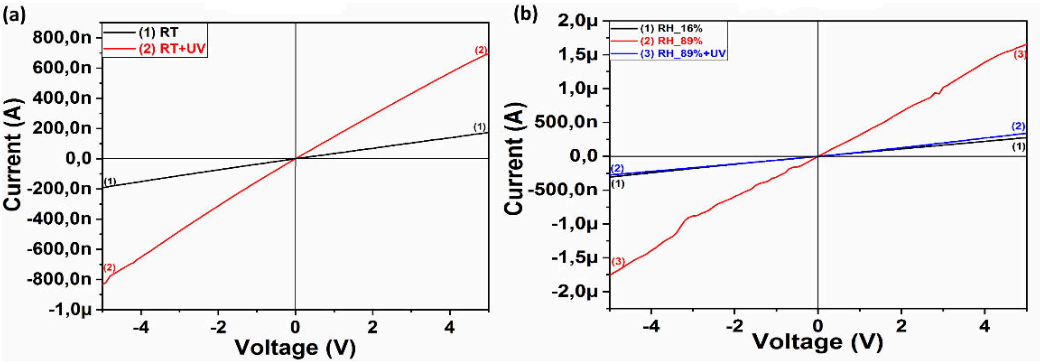

Figure 1. Current-Voltage characteristics of aluminum oxide/zinc oxide heterostructures: a) with and without UV radiation application; b) at different relative humidity (RH) levels

Cleaning of the glass substrate surface was performed by a successive washing in acids HCl 2% with decontamination in distilled water. This cleaning method is widely described in previous papers. [10]. After the deposition of the films by the SCS method, rinsing in deionized water and drying in a hot air stream for 1 min at t≈150 ºC was carried out. Rapid thermal annealing (RTA) or post-growth heat treatment is mandatory for SCS materials in order to improve the structural, chemical, optical properties and to diversify the scope of the respective nanomaterials.

In this paper all experiments were performed at room temperature. The current-voltage characteristics were investigated under different conditions: in Figure 1.a the curve (1) indicates the characteristic without the application of UV radiation, while the curve (2) shows the characteristic with the application of UV during the measurement. As can be observed, there is a greater increase of current under UV illumination, which demonstrates that the developed sensor can be used for UV detection. In Figure 1.b curves (1) and (2) indicate the sensor's response depending on the humidity. Curve (3) shows the volt-ampere characteristic at the humidity level of 89% and with UV illumination. The analysis of the curve (3) shows that this structure senses UV light at the humidity levels higher than the medium room humidity level.

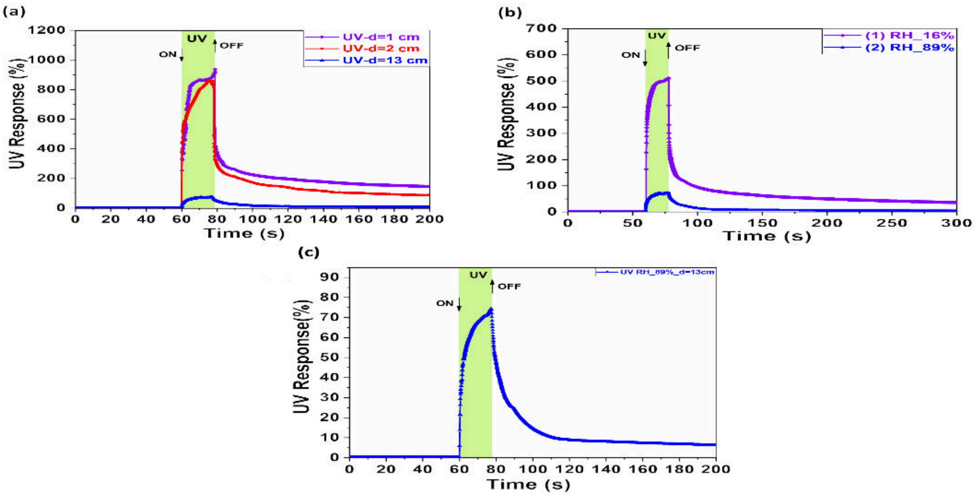

Figure 2. Response to UV radiation: (a) response depending on the distance to UV source at RH-89% humidity; (b) the response with the 13 cm distance to the UV source and at the 16% and 89% relative humidity levels; (c) the response with RH-89% at a distance of 13 cm to the UV source

Figure 2.a shows the response to UV measured at humidity level of 89%, depending on the distance from the UV source. It is observed that together with the change of distance, the change of answer takes place. The response at the distances of 1 cm and 2 cm does not differ too much, but at the distance of 13 cm the response to UV is much smaller. To determine the response to UV radiation the following equation was used: Eq (1):

$$S = \frac{G_{UV} - G_{dark}}{G_{dark}} * 100\% \quad (1)$$

where $$G = \frac{1}{R} \quad (2)$$

In order to demonstrate that the developed sensor, being mounted at a distance of 13 cm to the UV source, has a high response, it was tested in ambient air with a relative humidity (RH) level of 16%. The result of this measurement is shown in Figure 2.b. It can be concluded that the humidity level has an influence on the sensor's response to UV light.

The response time τ_r and the recovery time τ_d of the Al_2O_3/ZnO heterostructure at different distances from the UV source are shown in Figure 2.a. For calculation purposes, the response time τ_r was defined as time which is needed to reach 90% of the complete response, while the recuperation time τ_d was defined as the time to reach the value of 10% of the initial response [8]. From the figure it can be determined that the response times at the distance of 1 cm and 2 cm to the UV source are $\tau_r = 4{,}35s$ and $\tau_r = 5{,}07s$ while for the distance of 13 cm the response time is $\tau_r = 8{,}24s$. In all cases a partial recovery takes place.

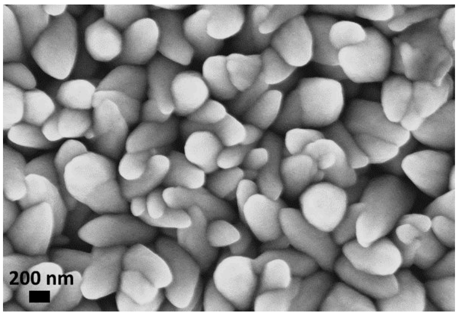

Figure 3. SEM image of sensible surface of Al_2O_3/ZnO heterostructure

The UV sensing mechanism that describes the response to ultraviolet radiation can be explained on the basis of a depletion region forming at the surface of Al_2O_3 and can be described by the "Eq.(3)". Thus, the dark conductivity appears in the inner layers which have low resistance. Under UV illumination the excess of photo generated charge carriers is formed [7].

$$hv = h^+ + e^- \quad (3)$$

Further, electrons and holes are separate and electrons migrate through the inside of the film, while holes remain on the surface and migrate to the surface forming a depletion region. At the same time, the oxygen photo-desorption takes place, which is described by the Eq.(4) [7].

$$h^+ + O_2^- \rightarrow O_2 \quad (4)$$

leads to a narrowing of the depletion region and to the increasing of through the inner layer current. As a consequence, the current gets saturated due to the reaction described in equations (3) and (4) taking place.

Conclusion

Measurement results showed that presented Al_2O_3/ZnO heterostructure has a high potential as a sensor for the UV radiation detection of at both, normal and high humidity levels. It was observed that at the humidity level of 89% the response is dependent on the distance to the UV source: the response changes from 825% at 1 cm distance to 78% at 13 cm distance. At the humidity level of 16% and at 13 cm distance, the value of the UV response was 525%. It can be concluded that the sensor is able to detect UV at different distances from the UV source. Sensing properties might be further enhanced by increasing the surface area of Al_2O_3/ZnO structure, e.g. by using a highly porous network made of tetrapodal shaped ZnO particles as a substrate material [9].

Acknowledgements

Magariu Nicolae, PhD student year 3, and Sereacov Alexandr PhD student year 2, PhD School of Computer Science, Electronics and Energy gratefully acknowledges for the active support and guidance to the head of PhD project to Prof.univ, Dr.hab Lupan Oleg and Professor Adelung. This research was sponsored in part by the NATO Science for Peace and Security Programme (SPS) under grant G5634 „Advanced Electro-Optical Chemical Sensors" AMOXES.

References

1. SRIKANT, V., CLARKE D. R. *On the optical band gap of zinc oxide*. Journal of Applied Physics, 83(10), 1998. pp.5447–5451.
2. LUPAN, O., POSTICA, V., HOPPE, M., WOLFF, N., POLONSKYI ,O., PAUPORTÉ, T., VIANA, B., MAJÉRUS, O., KIENLE, L., FAUPE,L F. and ADELUNG, R. PdO/PdO2 functionalized ZnO : Pd films for loweroperating temperature H2 gas sensing Nanoscale, 2018.
3. XU, C.K., LIU, Y.K., XU, G.D., WANG, G.H. Preparation and characterization of CuO nanorods by thermal decomposition of CuC2O4 precursor. Materials ResearchBulletin, 37, 2002 pp.2365–2372.
4. ORTEGA J.J., AGUILAR-FRUTIS, M.A., ALARCO´N, G.,FALCONY, C., ME´NDEZ-GARCI´A, V.H., ARAIZA, J.J. Band gap engineering of indium zinc oxide by nitrogen incorporation. Mater. Sci. Eng. B 187, 2014 pp.83–88.
5. LUPAN, O., CRETU, V., POSTICA, V., POLONSKYI, O., ABABII, N., SCHÜTT, F., KAIDAS, V., FAUPEL, F., ADELUNG, R., Non-Planar Nanoscale p-p Heterojunctions Formation in ZnxCu1−xOy Nanocrystals by Mixed Phases for Enhanced Sensors. Sensors and Actuators B: Chemical, 230, 2016 pp.832-843 2016.
6. LUPAN, O., CHOW, L., SHISHIYANU, S., MONAICO, E., SHISHIYANU, T., ŞONTEA, V., ROLDAN CUENYA, B., NAITABDI, A., PARK, S., SCHULTE A., , Nanostructured zinc oxide films synthesized by successive chemical solution deposition for gas sensor applications. Mater. Res. Bull., 44, 2009 pp.63-69.
7. CHEN, H., LIU, Y., XIE, C., WU, J., ZENG, D., LIAO, Y. A comparative study on UV lightactivated porous TiO2and ZnO film sensors for gas sensing at roomtemperature, Ceram. Int. 38, 2012 pp.503–509.
8. POSTICA, V., HÖLKEN, I., SCHNEIDER, V., KAIDAS, V., POLONSKYI, O., CRETU, V., TIGINYANU, I., FAUPEL, F., ADELUNG,R., LUPAN, O. Multifunctional device based on ZnO:Fe nanostructured films with enhanced UV and ultra-fast ethanol vapour sensing Materials Science in Semiconductor Processing, Volume 49, 2016 pp.20-33.
9. GAPEEVA, A., BODDULURI, M.T., KAPS, S., RASCH, F., WAGNER, B., ADELUNG, R., LUPAN, O. Mechanical and Wetting Properties of Three-Dimensional Flexible Tetrapodal ZnO Networks ALD-Coated with Al2O3. International Conference on Nanotechnologies and Biomedical Engineering. Springer, Cham, 2019.
10. LUPAN, O., CHOW,L., SHISHIYANU, S., MONAICO, E., SHISHIYANU, T., ŞONTEA, V., CUENYA, B.R., NAITABDI, A., PARK, S., SCHULTE, A. Nanostructured zinc oxide films synthesized by successive chemical solution deposition for gas sensor applications, Mater. Res. Bull. 44, 2009 pp.63-69