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Deposition of Zinc Oxide Based Binary Thin Film Doped With Dyes and Its Applications in Solar Devices

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Abstract
ZnO thin films doped with dyes from Eupatorium Odorata, Ageratum Conyzoides and Pucro pueraria was successfully deposited on a glass substrate using solution growth techniques. The grown films were characterized using spectrophotometer to determine the optical and solid state properties. Spectrophotometric analysis of the grown films shows very high absorbance in the UV region, moderate in the VIS region and low in the NIR region for all the films. The optical properties studied include absorbance, transmittance, reflectance, absorption coefficient and band gap. This properties were affected greatly by thermal annealing. The optical analysis of ZnO thin films indicate that absorbance is ≤ 2.3, transmittance is ≤ 78%, absorption coefficient varied from 0.25 x $10^6 m^{-1}$ to 6 X $10^6 m^{-1}$, band gap decreases from 4.01eV for Dye C to 3.90eV for Dye B to 3.86eV for Dye A and 3.84eV for As-deposited. High transmittance of ZnO thin films in the infrared region exhibited by the films suggest that the films can be used for screening off UV portion of electromagnetic spectrum which is dangerous to human health and as well harmful to domestic animals. The films can be used for coating eye glasses for protection from sunburn caused by UV radiation. It can also be used for coating poultry roofs and walls. This films are promising materials for fabrication of optoelectronic devices and window layers in solar cell fabrication.
Key Words *Dye, Doping and Spectrophotometer*

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I. Introduction

ZnO is one of the most promising chalcogenide semiconductors due to its great variety of applications. It is one of the principal semiconductor in thin films that can be substituted with effectively no toxic elements in a buffer layer in solar cells such as CdTe and copper indium gallium selenide, nanosized sensors, photodetectors, photodiodes, flat-panel displays, spectrally selective filters, electroluminescent devices and photocatalysts (Koch, Timbrell and Lamb, 1995; Lin et al, 2000; Chen, Bagnall and Yao, 2000; Shinde, Gujar and Lokhande, 2007).

Recently, interest in the use of natural dyes has been growing rapidly due to the result of stringent environmental standards imposed by many countries in response to toxic and allergic reaction associated with some compound (Neha and Vidya, 2011). The utilization of the dyes which are natural and locally available is a step towards producing green energy and reducing the climatic and environmental problems associated with fossil fuel production.

Research has shown that natural dyes are environment-friendly, locally available and a means of weed control.

Hopefully, the growth of ZnO oxide thin films doped with dyes in which the oxide is made up of group II - VI element will enable the production of thin films which will combine the unique properties of group II - IV element. The survey of literature shows that ZnO thin films have been deposited by various deposition techniques such as chemical bath deposition, spray pyrolysis, spin coating technique to mention but a few. However, there are few report on the doping of ZnO thin films with organic dye extracts. Among the few, none has used dye extracts from fresh leaves of Eupatorium Odorata, Ageratum Conyzoides and Pucro Pueraria. Thus, we report on the deposition of ZnO thin films doped with Eupatorium Odorata, Ageratum Conyzoides and Pucro Pueraria Pheseoloides.

II. Experimental Details

2.1 Materials

The following materials were used for the deposition of the films: Zinc chloride (ZnCl), Ammonia (NH_3), Distilled water, Extract of fresh leaves of Eupatorium Odorata, Ageratum Conyzoides and Pucro

Pueraria , Glass beakers, Magnetic stirrer, Glass slides, Electric oven, Measuring cylinders, Syringe, Synthetic foams , Digital weighing balance etc.

2.2 Methods

Chemical bath deposition technique was used to deposit the films on plane glass substrates. The dyes were extracted by preparing an aqueous solution of plant (10g in 100 ml distilled water) and the extraction process was carried out at a temperature range of 80-85^oC for 1hour. Colouring materials from the plant was used for the deposition of the desired compound in a reaction bath. The reaction bath was kept undisturbed under a temperature range of 80-85^oC for 1hour. After deposition, the films was rinsed in distilled water and drip dried in air.

Before deposition, the substrates were degreased in hydrochloric acid treatment, rinsed with distilled water and left to dry in dust-free environment. All reagents were of analytical grade and used without further purification. 1M Zinc chloride (ZnCl) was weighed exactly and dissolved in distilled water. With the solution being continuously stirred, the ZnCl aqueous solution was mixed with an appropriate amount of ammonia and aqueous dye extracts from fresh plants. After stirring, this clear and homogeneous aqueous solution was used as the precursor solution for ZnO thin film. The pre-treated substrates were floated on the surface of the solutions to prevent particles formed in the solution from accumulating on the substrate surface. The beakers containing the precursor solution were maintained at desired temperature for different deposition duration in order to obtain the proposed films. The substrates were dipped vertically into the centre of the reaction baths in such a way that they did not touch the bottom or walls of the bath containers. Several variations of the bath constitutions of each compound whose films were to be deposited was employed. For a chosen standard reaction bath for such a compound the substrates was allowed to stay in the bath for different dip times. This is to allow the growth condition to be optimized and the baths standardized. After deposition, the films were washed in distilled water and drip dried in air after which four of them were annealed at different temperature while one was left un-annealed to serve as the control.

III. Results and Discussion

Fig. 1 depicts the plot of absorbance against wavelength. The absorbance decreased with wavelength in the neighbourhood of 300-350 nm and remained fairly constant within 350-1000 nm. All the samples exhibited maximum absorbance in the UV region and different absorbance peaks in the VIS-NIR region. The absorbance for as-grown ZnO, ZnO+dye A, ZnO+dye B and ZnO+dye C thin films, in the UV region are 2.35, 2.15, 2.50 and 2.00 respectively. In the VIS-NIR region, the maximum absorbance of 0.25, 0.23, 0.60 and 0.15 were observed for as-grown ZnO, ZnO+dye A, ZnO+dye B and ZnO+dye C respectively. Clearly, the doping of the as-grown ZnO films with dyes altered the absorbance spectra. Our range of values for ZnO thin films are in the same order of magnitude with the values reported elsewhere (Igweoko et al, 2018). The transmittance spectra for as-grown and dye doped ZnO thin films are shown in Fig. 2. It turns out that the transmittance increased with wavelength exhibiting a maximum in the infrared region which agrees with the report of other authors (Agbo *et al.*, 2017; Igweoko *et al.*, 2018). Transmittance transparency varies from 1.5-60%, 2-63%, 1-27% and 2-77% for As-grown ZnO, ZnO+dye A, ZnO+dye B and ZnO+dye C respectively. ZnO+dye C transmitted higher compared to other film samples. The transmittance of the as-grown ZnO films was greatly modified by the decoration of same with dyes A and C while dye B reduced the transmission of as-grown ZnO films significantly. The enhancement of transmittance by dyes A and C could be attributed to the reduction in the thickness of the films.The relationship between the optical transmission and thickness is given by the Beer-Lambert equation as follows (Reddy *et al.*, 2006):

$$T = \frac{I}{I_o} = e^{(-\alpha t)} \qquad (1)$$

where I is the transmitted intensity at a particular wavelength, I_o is the incident light intensity, α is the absorption coefficient, and t is the film thickness. The equation shows that the optical transmission of the ZnO films will increase inversely proportional to the film thickness. The optical transmission is inversely proportional to the thickness of the films. With respect to the reduction in the transmittance of the as-grown ZnO films by decoration with dye B, it can also be explained on the basis of the thickness vis-à-vis concentration of the reagents used. Reduction in transmittance implies increase in film thickness in accordance with lambert-Beer's law. At higher concentration, the constituent atoms will be more and more, as well as the light-particle collisions with atoms and the more difficult light can pass through it (Kamran, 2012; Saleem *et al.*, 2012; Nagayasami *et al.*, 2013). It should be noted that when the concentration is high, the ions in solution would be high as well, leading to high rate of film formation. As such thicker films are grown at higher metallic precursor concentration. The transmittance of thin films can be greatly modified by various growth parameters. In the literature, several authors have reported on the variation of transmittance with parametric investigation involving annealing temperature (Chikwenze and Nnabuchi, 2010; Augustine and Nnabuchi, 2017; Igweoko *et al.*, 2018), concentration (Nnabuchi, 2005; Chikwenze and Nnabuchi, 2010; Agbo *et al.*, 2017; Onyishi *et al.*, 2018; Kalu *et al.*, 2018), deposition time (Nnabuchi, 2006; Ekuma *et al.*, 2011; Nnaemeka *et al.*, 2014; Chizomam *et al.*, 2014; Onyishi and Nnabuchi, 2014; Kalu *et al.*, 2018) and P^H (Odezue *et al.*, 2014). The relatively high transmittance of the film samples coated with dyes A and C compared to other film samples in this work, indicate their suitability for coating the interiors of poultry houses to admit infrared radiation (heat) into the building for the purpose of warming young chicks. The absorption coefficient generally varies from (0.5-5.1) x 10^6m^{-1} , (0.45-5.00) x 10^6m^{-1}, (1.25-5.70) x 10^6m^{-1} and (0.25-4.50) x 10^6m^{-1} for as-grown, dye A, B and C samples of ZnO thin films respectively (Fig. 3). From Fig. 4, the energy band gap (E_g), of the films was obtained from extrapolation of the straight portion to the energy axis at $(\alpha h\nu)^2 = 0$. It was found to be 3.90eV, 4.00eV, 3.85eV and 4.05eV for as-grown, dye A, B and C samples of ZnO thin films respectively. Values for ZnS thin films as widely reported is 3.29-3.95 eV (Chen *et al.*, 2000; Lin *et al.*, 2000; Mahalingam *et al.*, 2005; Shinde *et al.*, 2007; Baruah and Dutta, 2009; Ajuba *et al.*, 2010). It can be seen that the doping of as-grown samples of ZnO films with dyes shifted the fundamental absorption edge of the as-grown ZnO thin films thus providing tuning effect to the band gap for special applications. The high transparency in the visible region and wide direct band gap energy exhibited by our film samples make them ideal for use as window layer in heterojunction solar cells. The primary function of a window layer in a heterojunction is to form a junction with the absorber layer while admitting a maximum amount of light to the junction region and the absorber layer (Chikwenze and Nnabuchi, 2010). Yet, another advantage is that recombination in wide band gap semiconductor is usually low and is a desirable attribute for use as buffer layer in heterojunction solar cell.

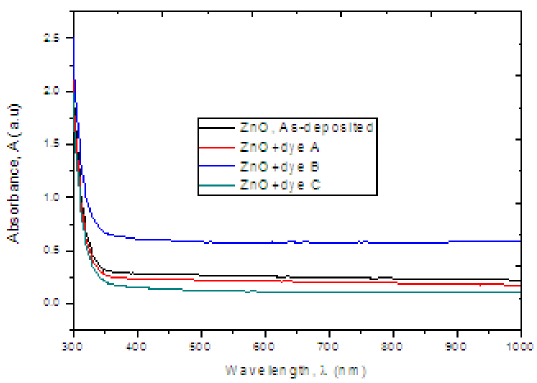

Fig. 1: Plot of absorbance against wavelength

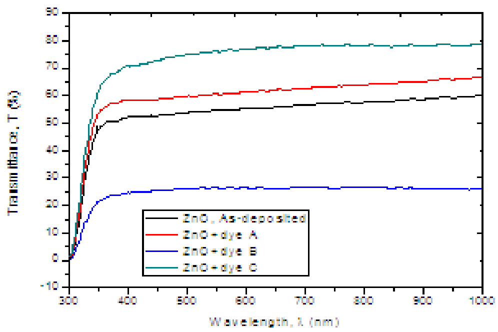

Fig. 2: Plot of transmittance against wavelength

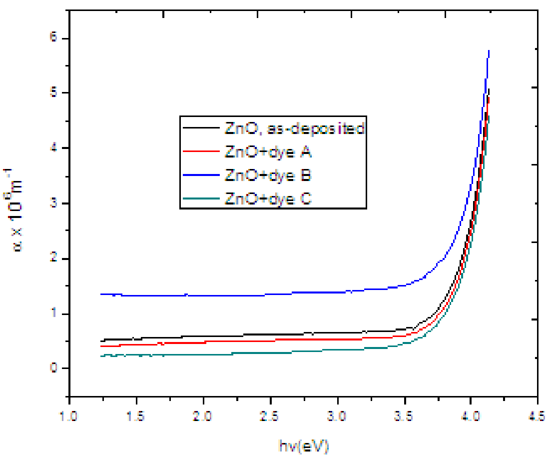

Fig. 3: Plot of absorption coefficient (α) versus hν

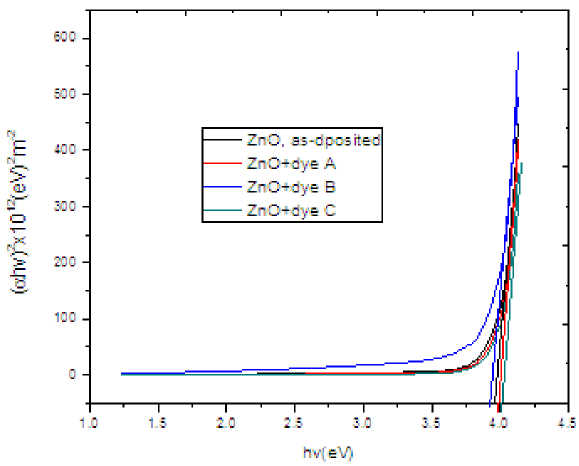

Fig. 4: Plot of $(\alpha h\nu)^2$ as a function of hν

IV. Conclusion

Solution growth process has been employed for the deposition of ZnO films doped with aqueous dye extract from flesh leaves of Eupatorium Odorata, Ageratum Conyzoides and Pucro Pueraria. Spectrophotometer was used to measure the transmittance from which the absorption coefficient and band gap were computed using relevant mathematical relations. The optical analysis showed band gap enhancement when dye was introduced into ZnO matrix thus tailoring the band gap for special applications. Based on the exhibited properties of the film, it can be concluded that it is a promising material for selective coatings for solar cells; effective coatings for poultry houses; and for fabrication of optoelectronic devices.

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