Photoresponse Characteristics of Aluminum Doped Zinc Oxide Nanostructure Thin Films.

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***Abstract*- The paper investigates the influence of different Al doping concentration treatments on the photoresponse of photocurrent formed ZnO nanostructure thin films. Al doped ZnO (AZO) thin films have been prepared by dip-coating technique on glass substrate. The annealing process is used to prepare the ZnO thin films. The influence of doping concentration of aluminum on the surface morphologies was characterized by Scanning Electron Microscopy (SEM). The electrical properties of thin films were characterized using 2-probe Current-Voltage (I-V) measurement system (Advantest R6243). The optical properties were studied using UV-Vis-NIR spectrometer. The "as prepared" ZnO nanostructure thin films layers have a grain structure and show a comparably low photocurrent magnitude under dark to illumination. We obtained that higher conductivity under illumination. Al doping concentration proportional increased with electrical conductivity.**

Keywords: Photoresponse; Al doped ZnO; dip-coating; dark; illumination.

I. INTRODUCTION

Aluminum doped zinc oxide (AZO) coatings exhibit high transparency and low resistivity and these materials are suitable for fabricating transparent electrodes in solar cells, gas sensors and ultrasonic oscillators [5]. Zinc oxide (ZnO) is a direct wide-band gap II-VI semiconductor with Eg =3.37 eV and exciton binding energy of ~60 meV at room temperature [1, 2, 4, 5]. Doping of ZnO with various elements has been reported to improve their electrical conductivity for use in optoelectronic device [12]. There are many type of dopants have been used to enhance the conductivities of ZnO. The group III in periodic table (B, Al, In, Ga) is the type of dopants can make a better conductivity to ZnO thin films. From group III, Al-doped ZnO thin films have been widely studied and are considered as candidate materials for organic electroluminescence display [2].

From the literature studies it is well known that Al is conductive material and have many advantages when doping with ZnO. Al is an excellent heat and electricity conductor and in relation to its weight is almost twice as good a conductor as copper. This has made Al the most commonly used material in major power transmission lines and semiconductor devices [12]. When increasing the doping of Al-doped ZnO, we aspect the electrical properties will increased and became less resistivity.

ZnO thin films have been prepared by a variety of thin film deposition techniques, such as pulsed-laser deposition, RF magnetron sputtering, chemical vapor deposition, spray pyrolysis, chemical bath deposition and the sol–gel process. Among the preparation technique, the combination of ZnO thin films with dip-coating is offered at the lower cost for technological application. It's used grips to hand tool for example pliers or clippers. This technique also known as a traditional method [13].

Focusing on the title, this research has been done to characterize the photoresponse characteristic. It's more involving the photoluminescence (PL) and UV-Vis to characterize optical properties. Beside this ZnO have a larger photoresponse to manufacture solar blind UV detector. In electrical properties under dark and illumination, the analysis of photoresponse more to sensitivity of the thin films toward lights [6].

II. METHODOLOGY

ZnO solution will be prepared using zinc acetate dehydrate as a precursor, monoethanolamine (MEA) as a stabilizer and zinc acetate dehydrate ($Zn(CH_3COO)_2.2H_2O$), 2-methoxyethanol as the solvent and aluminum nitrate nanohydrate ($Al(NO_3)_3.9H_2O$) as dopant source. Glass was use as a substrate. The molar ratio of MEA to zinc acetate dehydrate will be maintained at 1:1. 2-methaoxyethanol will be use to dissolved reactants and reagents. The use of 2-methaoxyethanol also provides temperature control, either to increase energy of the colliding particles or to absorb heat that will generate during an exothermic reaction. MEA used to avoid colloids from aggregating and increase the conductivity of the film. In this research, the dopant level determined by 100 x [Al]/ [Al+Zn], was varies from 0, 1,3,5,7 and 9at. %. By sol-gel method, all material will be mixed together and will be stirred using hot plate magnetic stirrer at 80°C for 3hours to yield a clear homogeneous solution. The solution then will be aged for 24 hours at room temperature to assure the chemical process is completed and ready for deposition process.

Al doped ZnO thin films will be deposited by dip-coating technique. Clean glass substrate with tape at the back (use one side) were dip in 30seconds into precursor solution with fixed withdrawal speed in 1mm/s at 1 cycle. After the dip coating, the film will be dried at 200°C in 10 minutes to evaporate the

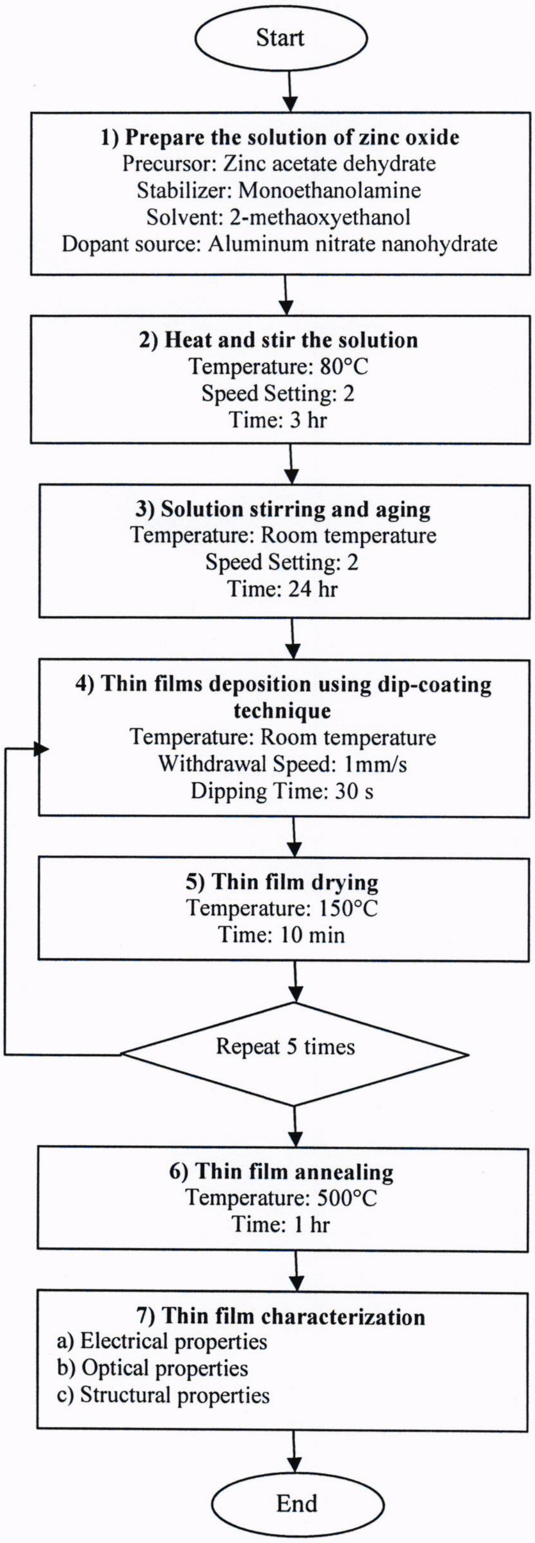

Fig. 1. Methodology of the research.

solvent and to remove organic residuals. This dip-coating and drying process will repeat for 5 times to yield required thickness.

Then, the prepared thin film will be annealed at 500°C for 1 hour to obtain a quality thin film. The electrical properties were characterized using 2-probe Current-Voltage (I-V) measurement system (Advantest R6243) in dark and illumination (photoresponse). Optical transmittance measurements were carried out using a UV–VIS–NIR spectrophotometer. The Scanning Electron Microscopy (SEM) use to characterize the surface morphology.

III. RESULT AND DISCUSSION

A. Electrical Properties

Fig. 2 show current-voltage (I-V) graphs under of undoped and Al doped ZnO thin films under room illumination and dark at room temperature. It is known that pure ZnO is a photosensitive and n-type semiconductor. Therefore, ZnO shows a small electrical current under the influence of a small voltage (10v). The electrical properties of ZnO thin films under illumination are closer to each line compared to the dark room. This occurred because of ion of ZnO thin films receive more energy from the light and vibrate then the ion will collide and produce the higher energy and became high current. This case is due to the photoresponse characteristic to show the differentiation under illumination and dark room.

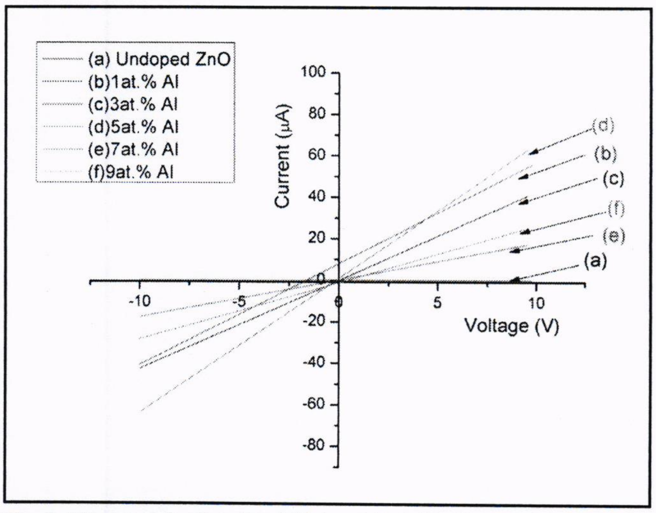

Fig. 2. I-V curve of undoped and Al doped ZnO thin films at different doping concentration under dark.

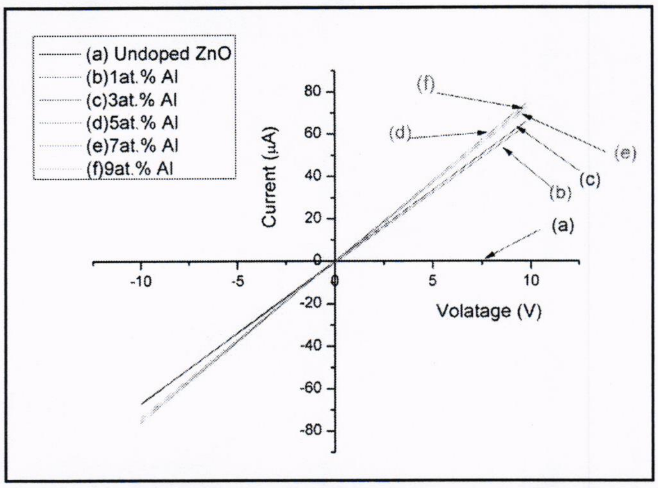

Fig. 3. I-V curve of undoped and Al doped ZnO thin films at different doping concentration under illumination

Al doping enhances, in general, the electrical conductivity of ZnO films. Fig. 3 shows the electrical dark conductivity and illumination conductivity increased with higher doping concentration of Al. The electrical illumination conductivity is higher than dark conductivity. This relation inversely proportional to current resistivity and were found that the decreasing of resistivity due to increasing of Al doping concentration. The average of resistivity of every point was taken from the I-V measurement data. Undoped ZnO thin film exhibits high resistivity but the resistivity significantly reduced with Al doping to indicate Al doping effectively improved ZnO thin film electrical properties.

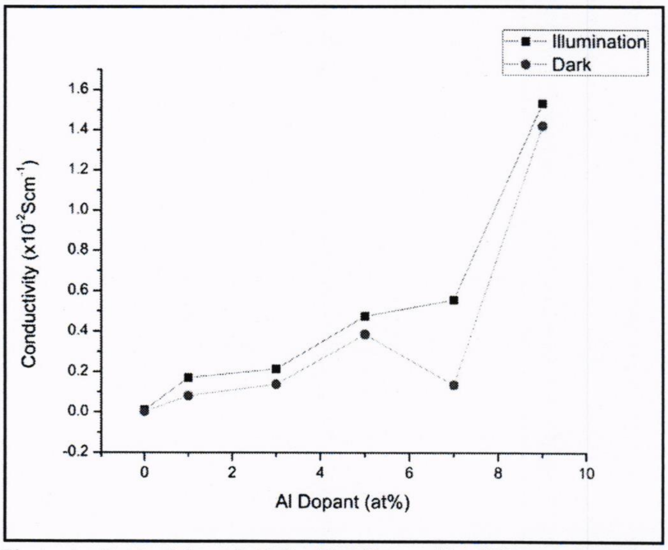

Fig. 4. Conductivity of ZnO thin films with different Al doping concentrations with consider under dark and illumination.

B. Optical Properties

Fig. 5 shows the transmittance spectra of the undoped and Al-doped ZnO thin films derived through the sol-gel process within the visible range of 200-800nm.

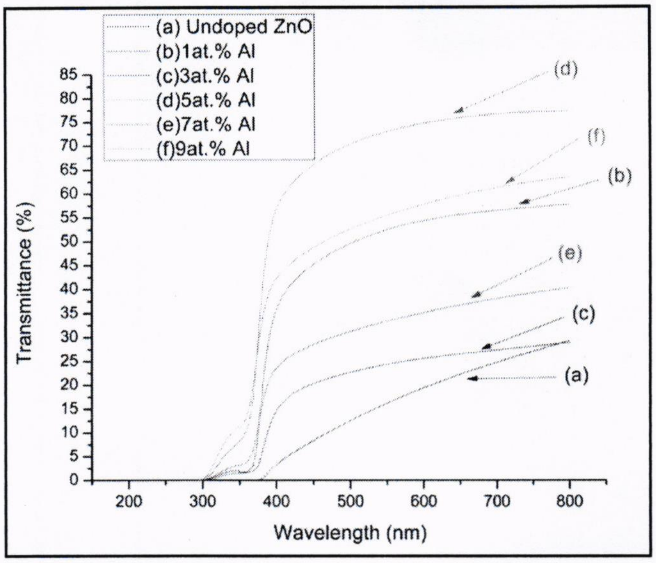

Fig. 5. Transmission spectra of undoped and Al doped ZnO thin films.

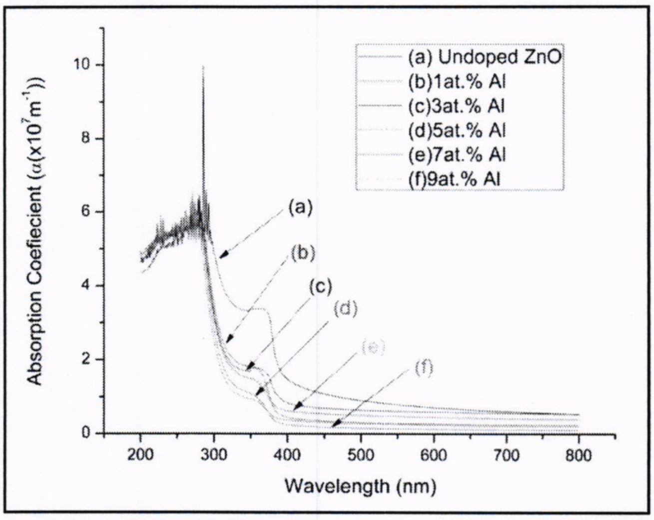

Fig. 6. Absorption coefficient of Al doped ZnO thin films at various doping concentration.

The absorption coefficient of undoped and Al doped ZnO thin films as a function of the wavelength at different doping concentration is shown in Fig. 6. The absorption coefficient was obtained through Lambert's Law which is indicated by following relation Eq. (1):

$$\alpha = \left(\frac{1}{t}\right) ln \left(\frac{1}{T}\right) \quad (1)$$

Where t is the thin film thickness and T is the transmittance spectrum of thin films. The result shows all films exhibit very low absorption in the visible and near infra red (NIR) range (380-800 nm) but exhibit high absorption in the ultraviolet (UV) range. The properties indicate transparency characteristic of ZnO thin films in the visible and NIR region [3]. High absorbance characteristic in the UV range are due to its wide band gap properties. The optical band gap energy was calculated using transmittance data and Tauc's plot to give the values as indicates in Fig. 7. When photon energy which is equal or higher than optical band gap energy is supplied, the electron will have adequate energy to jump from valence band to conduction band. The situation explained the strong absorbance properties of undoped and Al doped ZnO thin films in the UV region where the photon energy is used for electron excitation from valence band to conduction band. The situation explained the strong absorbance properties of undoped and Al doped ZnO thin films in the UV region where the photon energy is used for electron excitation from valence band to conduction band. As Al concentration is increased, the carrier concentration increased resulting in the optical band gap widening with doping concentration. The Burstein-Moss effect explained the broadening of band gap energy with the increasing of carrier concentration as given by a relation shown in Eq. (2):

$$\Delta E^{BM} = \left(\frac{h^2}{8m^*}\right)\left(\frac{3N}{\pi}\right)^{2/3} \quad (2)$$

Where ΔE^{BM} is the energy band gap broadening, h is the Planck constant, m^* is the effective mass of the electron and N is the carrier concentration [3]. As Al concentration is increased, the carrier concentration increased resulting in the optical band gap widening with Al concentration. This are supported the I-V measurement result, which conductivity is change due to increasing carrier concentration.

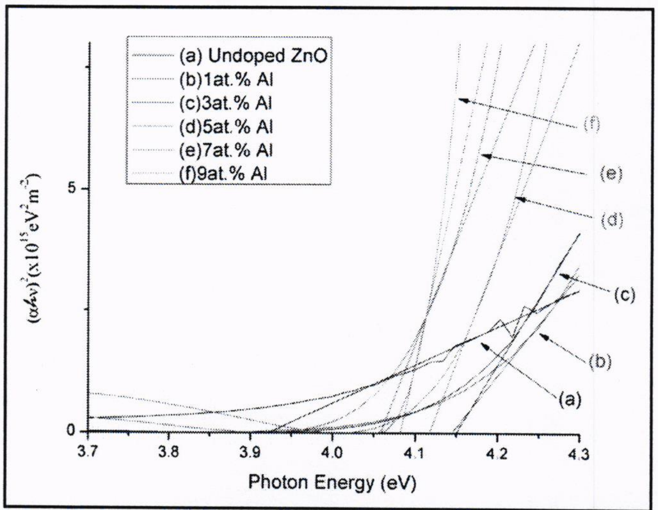

Fig. 7. Estimation of optical bandgap energy of undoped and Al doped ZnO thin films.

The direct band gap of ZnO thin films is estimated by plotting $(\alpha h\upsilon)^2$ versus photon energy and extrapolating the linear portion near the onset of absorption edge to the photon energy axis as shown in Fig. 7. From Fig. 7 and Fig. 8, we observe that undoped and Al doped ZnO at.1%, 3at.%, at.5%, 7at.% and 9at.% have a band gap of 3.93eV, 4 .14eV, 4.15eV, 4.12eV, 4.06eV and 4.08eV respectively. At 5at. % the optical band gap value is down from 3at. %. This occurred because of band tailing effect [14]. Band tailing occurs due to donor levels broadening into impurity bands which merge with the conduction band [14]. When increasing doping concentration of Al that deteriorates the crystalline films, which may be due to the formation stress induced by ion size difference between Zn and Al and the segregation of Al in grain boundaries for high doping concentrations [7].

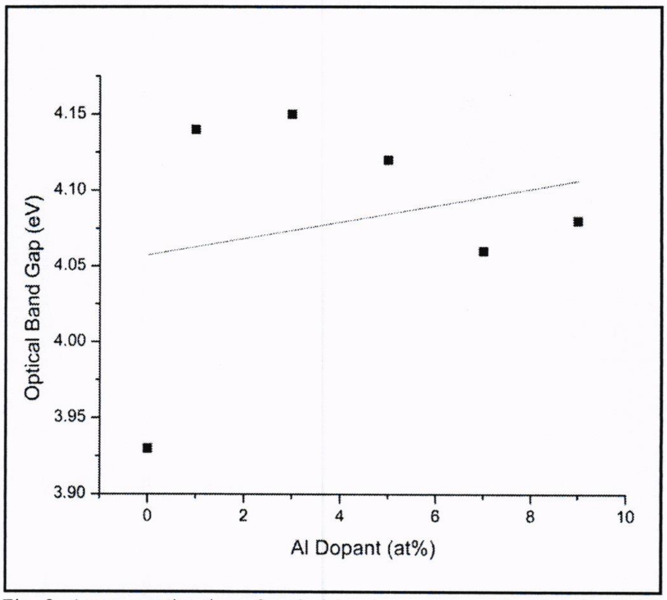

Fig. 8. Average estimation of optical band gap energy of undoped and Al doped ZnO thin films.

C. *SEM Characterization*

Fig. 9 shows the SEM images of the sol-gel-coated ZnO:Al thin film with different Al doping concentrations. The image show microsized polycrystalline microstructes. The undoped ZnO thin film showed many pores. Compared with the undoped ZnO thin film containing 1at.% Al. The grain size decreased with increasing Al concentration. The pores between grains dissapeared, as can be seen in Fig. 9, showing that the film became denser with increasing Al concentration. For Al doped ZnO thin films at doping concentration of 3at.% and 5at.%, there are crack areas presented which might have been caused by lattice strain due to difference of ionic radius between Zn^{2+} and Al^{3+}.

IV. CONCLUSION

We have investigated ZnO nanostructures are suitable doping with different Aluminum to increased current condcuctivity. Focus on photoresponse, the result current conductivity proved that the more response and high under illumination room. Its definitely logic to accept that under illumination the more conductive compared to dark conditon. The obtical band gap energy found to be increased with doping concentration. The surface morphology are investigated by using the SEM. According to the SEM studies, the result shows that the Al doping concentration influemce

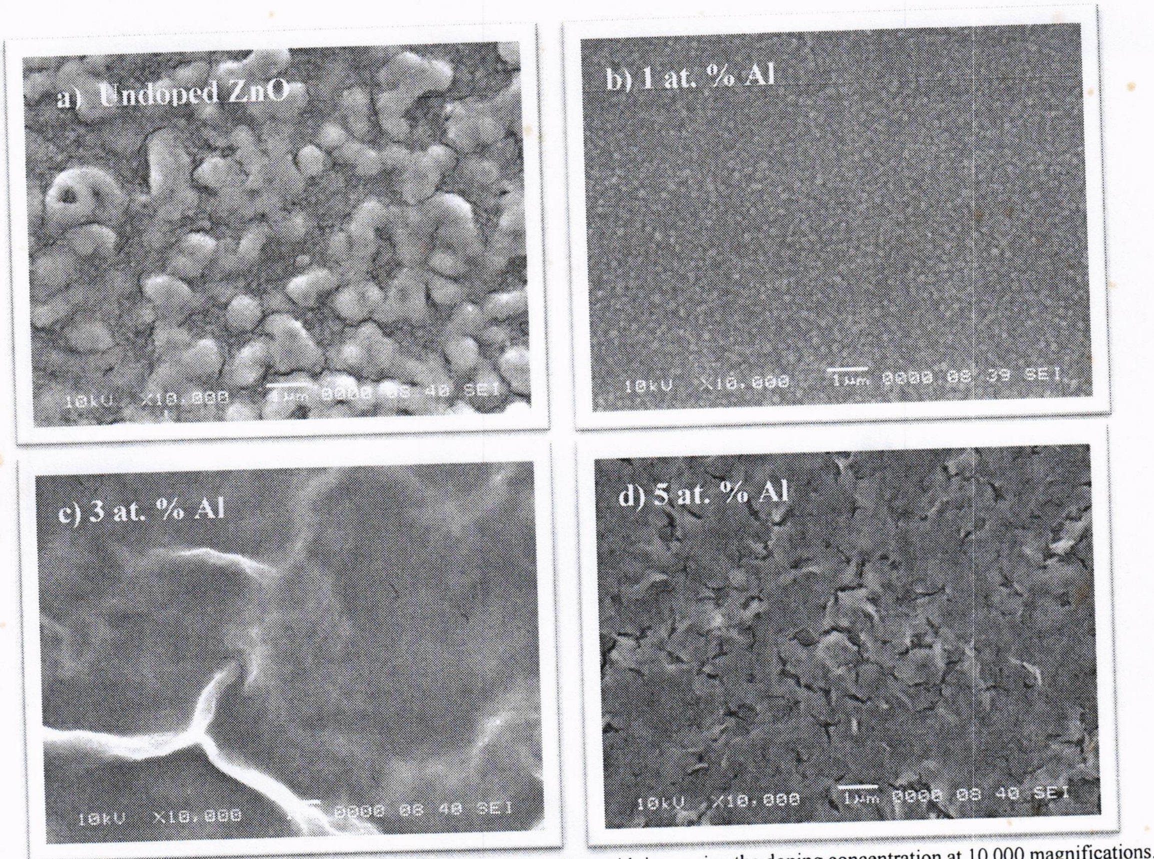

Figure. 9. Plain-view SEM micrographs of the Al doped ZnO thin films with increasing the doping concentration at 10,000 magnifications.

the surface of ZnO thin films. SEM images reveal reduction of ZnO particles size with doping concentrations.

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