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INVESTIGATION OF VOLTAGE MODULATION ON THE OPTICAL AND STRUCTURAL PROPERTIES OF ELECTRODEPOSITED ZINC SULFIDE (ZnS) THIN FILMS

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Abstract: Zinc sulfide thin films were successfully deposited onto a fluorine-doped tin oxide (FTO) glass substrate by employing electrodeposition method in this work. The basic materials used were; zinc acetate dihydrate which served as zinc source, thiourea (H_2NCSNH_2) served as sulfur source, FTO was the working electrode, silver/silver chloride served as the reference electrode, while platinum rod was used as the counter electrode. Deposition voltages of 2.0 volts, 4.0 volts and 6.0 volts from a DC supply unit (model Long Wei: PS-305D) were optimized to investigate their effects on the optical and structural properties of the thin films for applications. The films were characterized using UV-Vis spectrophotometer and X-ray diffractometry analysis to investigate the properties of the films. The results showed that the absorbance of the films is low but was influenced by the deposition voltage with the film deposited at 6.0 volts having the highest values in the range of 0.15 to 0.30 while the film deposited at 2.0 volts has the lowest value in the range of 0 to 0.10 within the visible (VIS) and near infrared (NIR) regions. The percentage reflectance of the films was also found to be low but increased with an increase deposition voltage throughout the VIS and NIR regions. The films have high percentage of transmittance with the values in the range of 80% to 100% for the film deposited at 2.0 volts but decreased to the values in the range of 50% to 70% for the film deposited at 6.0 volts within the VIS and NIR regions. The refractive index of the films was also influenced by the deposition voltage by increasing its values from 1.0 to 1.9 for the for film formed at 2.0 volts but increased to the range of 2.15 to 2.6 for the film formed at 6.0 volts. The direct bandgap energy was found to be 2.13 eV, 2.65 eV and 2.90 eV for the film deposited at 2.0 volts, 4.0 volts and 6.0 volts respectively. The X-ray diffraction analysis showed that the films have trigonal (hexagonal axes) structure and influenced by deposition voltage variations. The average crystallite size for the films formed at 2.0 volts and 6.0 volts are 29.68 nm and 20.64 nm

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respectively while the micro-strain and dislocation density are 1.15×10^{-3} and 3.31×10^{-3} nm^{-2} for the film deposited at 2.0 volts and 2.36×10^{-3} and 4.92×10^{-3} nm^{-2} for 6.0 volts film.
These properties possessed by the deposited thin films of ZnS in this work position them as suitable material for many optoelectronic device applications such as blue LEDs, solar cells and for antireflection coating applications

Keywords: Zinc Sulfide, Electrodeposition, Semiconductor, Bandgap, Solar cells, LEDs

1.0 INTRODUCTION

Compound semiconductors have become the focus of researchers in the recent time. This is largely as a result of their potential applications in solar cells and other optoelectronics devices to replacing the elemental semiconductors such as silicon and germanium based solar cells which are highly expensive in nature, [1]. This shift to the relatively available and cost-effective semiconductor materials is as a result of advancement in science and technology which have expanded the requirements for energy consumption around the globe, and various countries have resorted to renewable energy sources to meet up their energy demands, [2]. Zinc Sulfide (ZnS) is one of the compound semiconductor materials which has attracted large attention owning to their potential applications in the new generation of nano-electronics and optoelectronics devices, [3]. Zinc sulfide (ZnS) is an important group II–VI semiconductors and it has been intensively studied due to its rich morphologies at the nanoscale, excellent physical properties, and unique photocatalytic properties [4]. ZnS semiconductors have shown a remarkable versatility and high promise for diverse applications such as light-emitting diodes (LEDs), electroluminescence, flat panel displays, infrared window, sensors, LASERS and biodevices, photonics, nonlinear optical devices, catalysis etc. [5, 6]. The material has been known to occur in two distinct polymorphs of cubic sphalerite and hexagonal wurtzite structures and have been attributed to the varieties of potential applications credited to it, [7]. The sphalerite phase is believed to be more stable at low temperatures compared to the hexagonal wurtzite phase and is a fairly common trace mineral in sulfidic sediments, [8]. The bandgap energy of ZnS has been noted to be about 3.67 eV and 3.90 eV for cubic sphalerite and hexagonal wurtzite ZnS respectively which is suitable for visible-blind ultraviolet (UV)-light based devices such as sensors/photodetectors, [9, 10]. Reports have shown that the characteristic features/properties of ZnS are highly dependent on the methods of its film deposition, deposition parameters and conditions, hence it has become very interesting to investigate the properties of the material with respect to the different deposition methods and conditions for optimum performance, [11]. In this regard, the properties of ZnS thin films deposited at different power range of 70 to 100 W and subjected to thermal annealing treatment at temperatures of 350 °C, 400 °C, and 450 °C for 20 minutes have been reported by, [12]. The effect of SILAR deposition cycle on the optical and structural properties of ZnS thin films have been reported by [13]. It was reported that the thickness of the films can be controlled by deposition cycles which has direct impact on the grain size but little on the optical properties of the films. The influence of deposition temperature on the properties of chemically sprayed ZnS thin films have been studied by [14]. The authors discovered that the structural properties of the films such as crystallite size and micro-strain increased with an increase temperature while the optical properties like bandgap was found to decreased slightly as deposition temperature increased. The micro and nano tribology analysis of ZnS and CdS materials through combination of Tomlinson's model with Sang's equation and bond-orbital model have been reported by [15]. It was observed that ZnS exhibits better tribological properties at nano level than CdS and the efficiency of the material will increase if micro and nano- electromechanical systems are designed to operate normally at slightly high temperatures. The effect of doping concentration and annealing temperature on the properties of Cu doped ZnS thin films deposited by electron beam evaporation on glass substrates have been reported by [16]. It was observed that the bandgap energy for the Cu:ZnS films with x = 0 and 3wt% Cu contents decreased slightly with increase in annealing temperature up to 400◦C but the values for the films with x = 6wt% and x = 9wt% contents of Cu increased slightly. These behaviors were attributed to increase in crystallinity and change in chemical composition like oxidation of the films. The influence of the Cu^{2+} ions concentration on the optical and structural properties of Cu:ZnS thin films fabricated by chemical bath method have also been investigated by [17]. It was discovered that the photoluminescence intensity of the films is strongly dependent on the concentration of Cu ions by increasing significantly with increasing the Cu:Zn molar ratio. The bandgap energy of the films were however, found to decrease with an increase in Cu ions concentration. The authors concluded that the deposited Cu;ZnS thin films can offer good potential applications in optoelectronic devices fabrications such as light-emitting diodes. In this report, the effect of deposition voltage on the optical and structural properties of electrodeposited ZnS thin films was investigated for possible optoelectronics applications.

2.0 EXPERIMENTAL PROCEDURE AND METHOD

ZnS thin films were prepared on the fluorine-doped tin oxide (FTO) glass substrates by electrodeposition technique. The substrates were first cleaned ultrasonically using acetone, for 10 minutes, and distilled water for another 10 minutes to ensure complete cleanness of the substrates. The ZnS thin films were deposited using the following materials: zinc acetate

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dihydrate as zinc source, thiourea (H_2NCSNH_2) as sulfur source, fluorine-doped tin oxide (FTO) conductive glass substrate: - used as working electrode, silver/silver chloride (Ag/AgCl):- served as reference electrode, platinum rod:- used as the counter electrode, 50 ml glass beaker:- used as reaction bath, distilled water:- used as the reaction medium for the precursors, voltage supply (model Long Wei: PS-305D):- used as DC supply unit and magnetic-stirrer. Electrodeposition method with three electrodes configuration system was used to deposit the thin films of ZnS. To deposit the thin film on FTO substrate through this method, aqueous electrolytic bath solution containing 1.0 M of 20 ml of zinc acetate and 0.5 M of 20 ml of thiourea were used.

The three electrodes were immersed into the bath containing the electrolytic solution and 2 volts was applied from the DC supply setup for 60 seconds. This process was repeated for two more times by varying the applied voltage of deposition from 4 volts to 6 volts at interval of 2 volts. Other parameters including concentration and time of deposition were kept constant as shown in Table 1.

Table 1: Stoichiometric parameters for the preparation of ZnS thin films.

1.0 M $Zn(C_2H_3CO_2)_2{\cdot}2H_2O$ Volume (ml)	**0.5 M H_2NCSNH_2 Volume (ml)**	**Deposition Time (secs)**	**Deposition Potential (V)**
20.00	20.00	60.00	2.0
20.00	20.00	60.00	4.0
20.00	20.00	60.00	6.0

3.0 RESULTS AND DISCUSSIONS

3.1 Optical Properties of the deposited ZnS thin Films

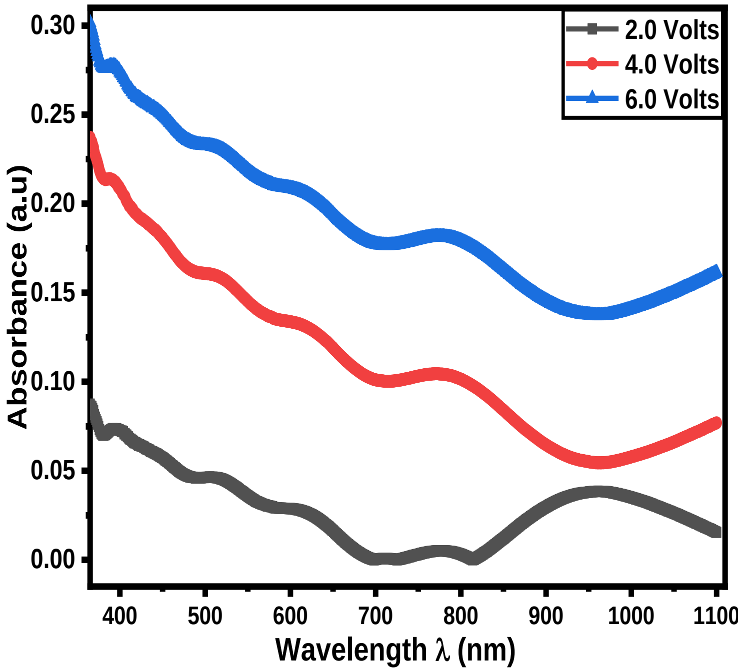

Figure 1: Graph of absorbance against wavelength for the deposited thin films of ZnS

Figure 1 is the graph of measured absorbance of the deposited thin films of ZnS against wavelength to determine the absorbance property of the films at different wavelength range. The figure showed that the deposited thin films of ZnS generally have low absorbance value but were influenced by deposition voltage variation. The absorbance however, increased with an increase in the deposition voltage. The film deposited at 6.0 volts has the highest absorbance value in the order of 0.15 to 0.30 within the visible (VIS) and near infrared (NIR) regions of electromagnetic spectrum while the film deposited at lowest voltage of 2.0 volts has the lowest value in the range of 0 to 0.10 also in the two regions. The figure also showed that the absorbance of the films decreases within the VIS range and tend to increase in the NIR range. This result is in close agreement with the materials reported by [18].

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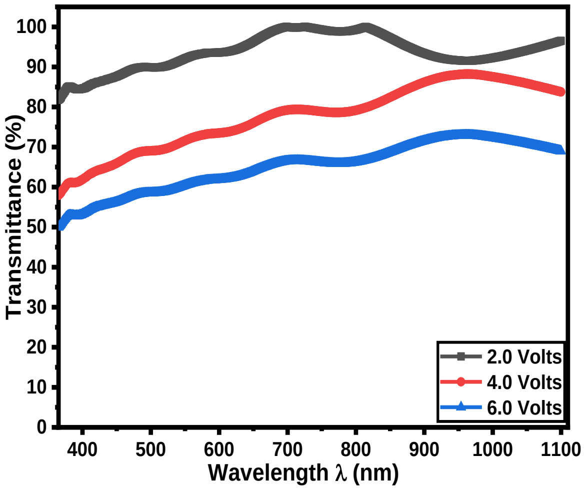

Figure 2: Graph of percentage transmittance against wavelength for the deposited thin films of ZnS

The plot of percentage transmittance of the films as a function of wavelength is shown in figure 2 to study the transmittance of the deposited thin films of ZnS for their possible device applications. The transmittance of the films was calculated using formula as given by [19].

$$T = 10^{-A} \qquad 1$$

Where A is the measured absorbance of the films. The figure indicates that the transmittance of the films is quite high but decreased with an increase in deposition voltage with film formed at voltage of 6.0 volts having the lowest value in the range of 50 to 70% throughout the VIS and NIR regions. The film deposited at 2.0 volts has the highest value in the order of 80 to 100% within the VIS and NIR regions. The high transmittance exhibited by the deposited thin films of ZnS makes them good material for cold window coating application in the low temperate regions of the World, since maximum amount of visible infrared rays can be allowed to pass through the material.

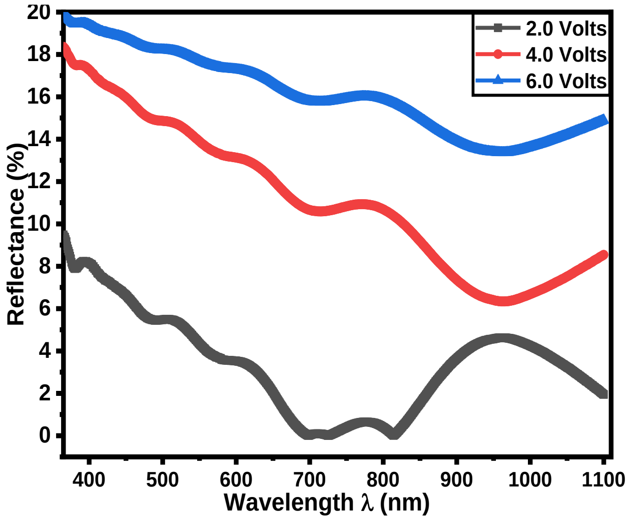

Figure 3: Graph of percentage reflectance against wavelength for the deposited thin films of ZnS

The graph of percentage reflectance of the films against wavelength is displayed in figure 3. The reflectance of the films was evaluated from the conservation relation as provided by [20, 21].

$$A + T + R = 1 \qquad 2$$

Where A represents the measured absorbance, T is the transmittance of the films. From the relation, the percentage of the films was calculated via equation 3.

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$$R = (1 - A - T) \times 100\% \quad 3$$

The figure showed that the deposited thin films have low percentage reflectance and decreased with wavelength in the VIS region. The reflectance of the films however increases with an increase in the deposition voltage with the film deposited at highest voltage of 6.0 volts having the highest reflectance percent in the range of 14% to 20% throughout the VIS and NIR regions. The low values of percentage reflectance exhibited by the films position them for suitable material for antireflection coating applications.

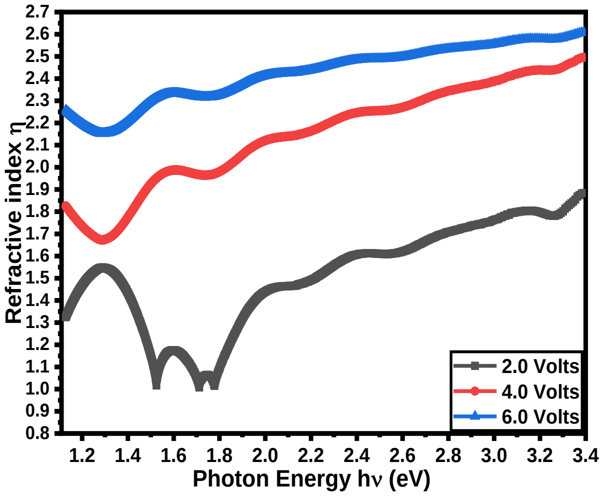

Figure 4: Graph of refractive index against photon energy for the deposited thin films of ZnS

The graph of refractive index of the films as a function of photon energy for the films is displayed in figure 4. The refractive index was evaluated using the relation as provided by [22].

$$n = \frac{1+\sqrt{R}}{1-\sqrt{R}} \quad 4$$

Where R is the reflectance of the films. The figure showed that the refractive index of the films increased as deposition voltage increased with the film deposited at higher voltage of 6.0 volts having the highest value in the range of 2.15 to 2.65 while the film formed at 2.0 volts has the lowest value in the range of 1.0 to 1.8. The figure also showed that the refractive index of the films increased as the photon energy increased except for the film 2.0 volts which showed slight decrease in value at lower photon energy range of 1.1-1.8. The increase in the value of refractive index of the films as a result of variation in the deposition voltage is significant for device applications such as in optical waveguide, optical reflector, filter and fibers, [23].

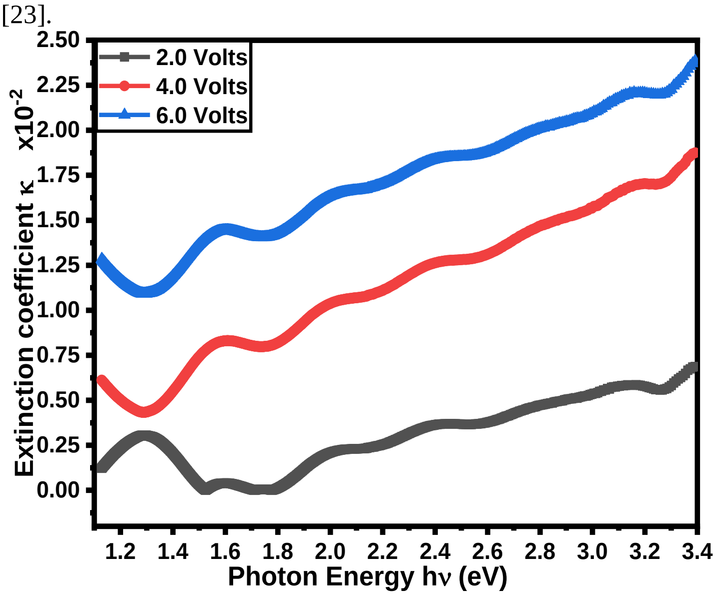

Figure 5: Graph of extinction coefficient against photon energy for the deposited thin films of ZnS

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Figure 5 is the graph of extinction coefficient of the deposited thin films against photon energy to determine the rate at which the interaction of electromagnetic radiation got extinct in the material at different photon energy range. This property of the films was calculated using the formula given by [24, 25].

$$k = \frac{\alpha\lambda}{4\pi} \qquad 5$$

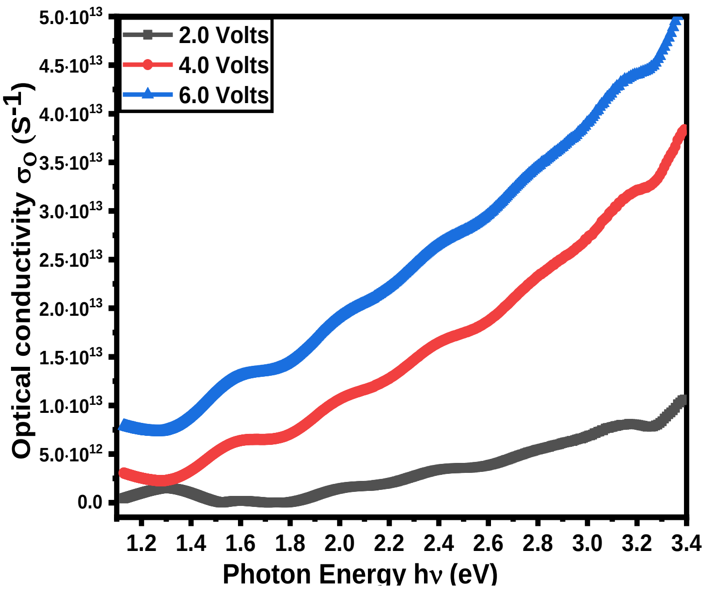

Figure 6: Graph of optical conductivity against photon energy for the deposited thin films of ZnS

The plot of optical conductivity of the deposited thin films of ZnO as a function of photon energy is presented in figure 6. The optical conductivity of the films which determine the ability of the films to conduct an electric current in response to electromagnetic radiation was evaluated using the relation as provided in [26].

$$\sigma_o = \frac{\alpha n C}{4\pi} \qquad 6$$

From the figure, it can be seen that the optical conductivity of the films increased with an increase photon energy as well as with deposition voltage. The film deposited with higher voltage of 6.0 volts has the highest optical conductivity value between 7.5×10^{12} S^{-1} and 5.0×10^{13} S^{-1} while the film deposited at lower voltage, 2.0 volts has the lowest values in the range of 0 to 1.3×10^{13} S^{-1} within the photon energy range as depicted in the figure.

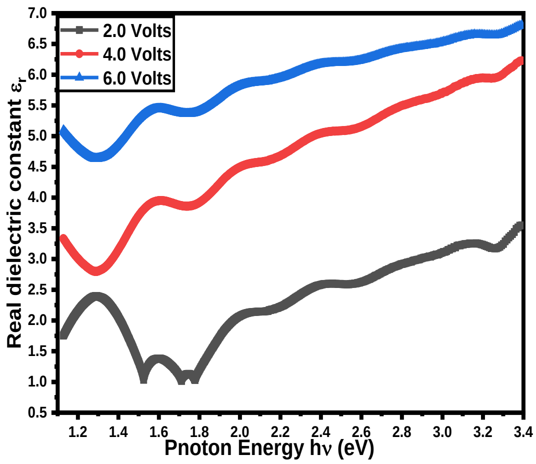

Figure 7: Graph of real dielectric constant against photon energy for the deposited thin films of ZnS

The graph of real dielectric constant of the deposited thin films of ZnS as function of photon energy is depicted in figure 7. This property was evaluated using the relation as provided by [27]

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$$\varepsilon_r = n^2 - k^2 \qquad 7$$

The figure showed that the deposited thin films of ZnS increased as photon energy increase except for the film deposited at 2 volts which initially decreased from the value of 2.5 to 1.0 in the lower photon energy range 1.1 eV to 1.8 eV and thereafter increased with an increase in photon energy. The figure also showed that the real dielectric constant of the films increases with increase in deposition voltage with the film deposited at higher voltage of 6.0 volts having the highest values within the range of 4.5 to 6.8 while the film deposited with lower voltage of 2.0 volts has the lowest values in the range of 1.0 to 3.5 within the photon energy of interest. These results showed that deposition voltage variation has significant influence on the dielectric constant of the films for device applications.

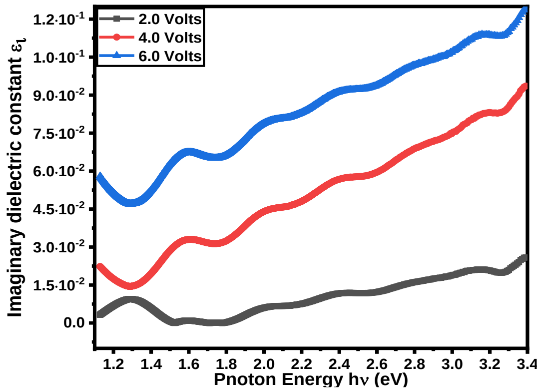

Figure 8: Graph of imaginary dielectric constant against photon energy for the deposited thin films of ZnS

Figure 8 represents the graph of imaginary dielectric constant against photon energy for the deposited thin films of ZnS. The imaginary dielectric constant of the films which represent the dielectric losses for the films was calculated using the formula as provided by [28].

$$\varepsilon_i = 2nk \qquad 8$$

The figure showed that the imaginary dielectric constant of the films is quite low but increased with an increase in photon energy. The figure also showed that the imaginary dielectric constant of the films increased with deposition voltage with the film deposited at 6.0 volts having the highest value in the order of 0.045 to 0.12 while the film formed at 2.0 volts has the lowest value in the order of 0 to 0.02. The low value of dielectric loss indicates the suitability of the films for applications in solar cells.

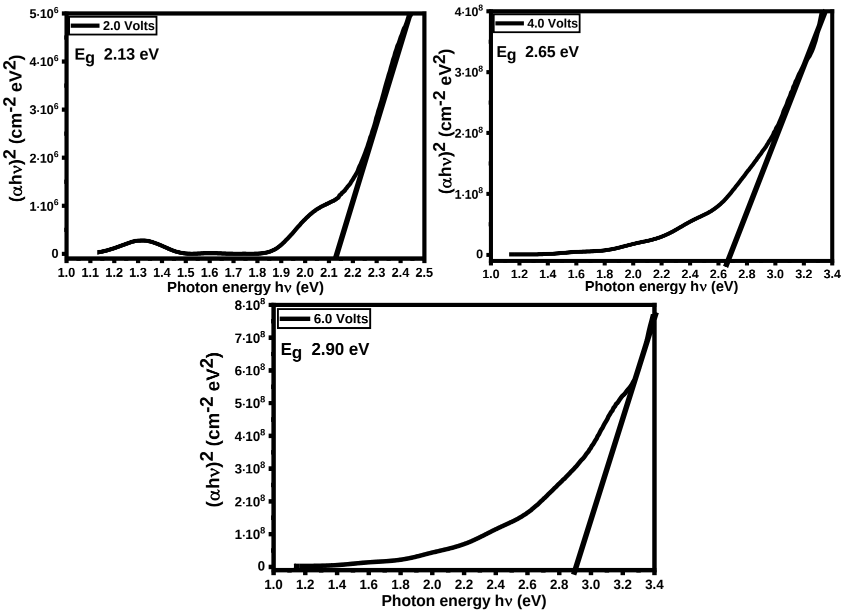

Figure 9: Plots (αhν)^2 as a function of photon energy for the deposited thin films of ZnS

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The plots of $(\alpha hv)^2$ as a function of photon energy are displayed in figure 9 in order to determine the bandgap energies of the deposited thin films of ZnS. The bandgap energies of the films were estimated based on the formula as given by [29].

$$(\alpha hv)^n = \beta(hv - E_g) \qquad 9$$

Where α is the absorption coefficient of the films, h is the Planck's constant, v frequency, E_g is the bandgap energy, β is a constant while n is the transition form factor which determine the band nature of the films; with the values of 2 for direct bandgap, 0.5 for indirect bandgap transition. The figure showed that the direct bandgap energy of the films increased with an increase in the deposition voltage. The obtained values of the bandgap energies of the films through extrapolation on the photon energy axes as shown in the plots are 2.13 eV for the film deposited at 2.0 volts, 2.65 eV and 2.90 eV for the films deposited at 4.0 volts and 6.0 volts respectively. This range of bandgap energy value is suitable for blue LEDs device fabrication.

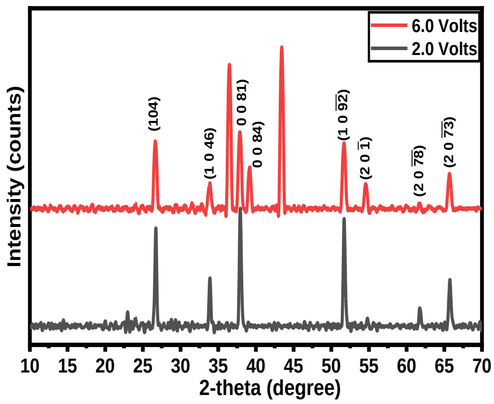

Figure 10: X-ray diffraction pattern of the deposited thin films of ZnS

The graph of x-ray diffraction (XRD) patterns of the deposited ZnS thin films at 2.0 volts and 6.0 volts is shown in figure 10. The figure revealed that the deposited thin films exhibited crystalline structure with preferential planes as depicted in the XRD pattern. The patterns matched almost well with the standard JCPDS Card No: 00-231-0814 with space group R3m:H and crystal structure of Trigonal (hexagonal axes) for ZnS. The film deposited at higher deposition voltage however has extra diffraction sharp peaks as compared to the film formed at low voltage of 2.0 volts. This showed that deposition voltage variation has effect on the crystal orientation of the deposited ZnS thin films. The crystallite size of the films was calculated using the Debye-Scherrer relation as provided by [30, 31].

$$D = \frac{k\lambda}{\beta Cos\theta} \qquad 10$$

Where k is a shape factor with constant value 0.9, λ is the x-ray Cu-k-α radiation wavelength, β is the full weight at half maximum (FWHM) and θ is the Bragg angle. The obtained value of the crystallite size for the films are 29.68 nm and 20.64 nm for the films deposited at 2.0 volts and 6.0 volts respectively. The micro-strain and dislocation density of the films were also calculated using the formula given by [32, 33].

$$\varepsilon = \frac{\beta}{4.tan\theta} \qquad 11$$

$$\delta = \frac{1}{D^2} \qquad 12$$

The calculated values of the micro-strain and dislocation density for films are 1.15×10^{-3} and 3.31×10^{-3} nm^{-2} for the film deposited at 2.0 volts while the film deposited at 6.0 volts has the values 2.36×10^{-3} and 4.92×10^{-3} nm^{-2}. This variation in these crystal parameters confirmed the effect of deposition voltage on the deposited ZnS thin films for applications.

CONCLUSION

The effect of deposition voltage modulations on the optical and structural properties of electrodeposited thin films of ZnS for device fabrication was studied in this research work. Spectrophotometric and X-ray diffraction techniques were employed to investigate the properties. As a result, the optical properties such as absorbance was observed to be low and

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decrease with wavelength in the VIS region. The absorbance of the films however increased as deposition voltage increased within the VIS and NIR regions. The transmittance of the films was observed to be in the range of 80% to 100% for the film deposited at 2.0 volts but decreased to the values in the range 50% to 70% for the film formed at 6.0 volts within the VIS and NIR regions. The refractive index of the films was found to be in the range of 1.0 to 1.9 for the film formed at 2.0 volts but increased to the values in the range of 2.15 to 2.6 for the film formed at 6.0 volts. The direct bandgap energy of the films was also found to increase with deposition voltage with values 2.13 eV, 2.65 eV and 2.90 eV for the film deposited at 2.0 volts, 4.0 volts and 6.0 volts respectively. The X-ray diffraction analysis revealed that the thin films exhibited crystalline structure with trigonal (hexagonal axes) and the structure is influenced by deposition voltage variations. The crystallite size obtained for the films formed at 2.0 volts and 6.0 volts are 29.68 nm and 20.64 nm respectively while the values of the micro-strain and dislocation density for films are 1.15×10^{-3} and 3.31×10^{-3} nm^{-2} for the film deposited at 2.0 volts while the film deposited at 6.0 volts has the values 2.36×10^{-3} and 4.92×10^{-3} nm^{-2} respectively. These properties and others exhibited by the deposited thin films of ZnS position them for many optoelectronic device applications such as blue LEDs and solar cells.

Conflict of Interest
Authors declared that they do not have any conflict of interest during the period of the work.

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