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Effect of Nitrogen Doping on the Optical and Dielectric Properties of Zinc Oxide Thin Film

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ABSTRACT

The optical and dielectric properties of chemically deposited Zinc Oxide (ZnO) and Nitrogen doped Zinc Oxide (N-ZnO) thin films were investigated using UV-Vis Spectrophotometer within the wavelength of 330 nm – 700 nm. The absorbance decreases with increasing wavelength. The transmittance was found to increase within the UV-Visible region as wavelength increases. The reflectance was observed to decrease with increasing wavelength for all samples. Band gap values obtained are 3.55 eV, 3.42 eV, 3.35 eV and 3.15 eV for for 0 %N, 2 %N, 4 %N and 6 %N concentrations. The extinction coefficient decreases gradually within the visible region with increasing wavelength except for 2 %N where it value increases with increasing wavelength from 540 nm to 700 nm. The value of the refractive index decreases sharply between 345nm to 380 nm for all the samples. Its value was low within the visible region. Low values of the real and imaginary parts of the dielectric constant within the visible region are as a result of the dielectric losses at high wavelengths.

Keywords: Synthesis, Z-Doping, Chemical Bath Deposition, Optical Properties, Dielectric Properties, and Visible Region.

1.0 Introduction

Semiconductor materials and devices are the consequent of the advancement in thin film synthesis. This is the deposition of molecules on a 2D-material deposited by either molecule-by-molecule or atom-by-atom condensation method [23, 32]. Optical absorption measurements are used to obtain the band structure and the energy gap of thin films, because the analysis of the optical absorption spectra is one of the most useful tools for understanding and developing the band structure of many materials [11].Transparent conducting oxides (TCOs) have drawn much attention because of their potential applications in different devices, from transistors to optical detection and emission [26].

The wide optical band gap (~3.5 eV) and large exciton binding energy (60 meV) of Zinc oxide (ZnO) makes it a potential material for different applications. It has an enormous potential for use in optoelectronics and electroluminescent devices [24]. It has attracted a lots of interest because of their applications [8, 18, 37]. It is a promising photonic materials in ultraviolet and visible region due to its direct band gap and large exciton binding energy. ZnO-based materials are primary candidate in optical devices production, high density optical memories, photovotaics and solid layer devices etc. Therefore, to better manufacture high quality ZnO-based optoelectronic materials, fundamental study about optical and dielectric behavior of these materials are important. Band gap engineering a potential approach in designing modern optoelectronic devices. Different theoretical and experimental techniques has been used to synthesize ZnO thin flms by diferent researchers to modify and improve its properties by both chemical and physical processes, the techniques include; chemical spray pyrolysis (CSP) [20, 25, 30], chemical bath deposition (CBD) [6, 34], chemical vapor deposition (CVD) [3, 22], electrochemical deposition [2, 26], pulse laser deposition (PLD) [4, 19], thermal evaporation [9 ,26, 12] and sol–gel [15, 21, 35], because of excellent features. However, researches are still ongoing due to a wide range of topics [10]. Many studies have been carried out in rder to change the properties of ZnO semiconductor thin films by incorporating metallic elements such as, as well as non-metallic dopants such as F, N. Nitrogen is seen as an element which possesses the potential to produce a shallow acceptor region in ZnO as a result of the similarities in the radius and electrical behaviour between oxygen and nitrogen. So it [13] . It is in the fifth group and therefore considered to be a better dopant for p-type conductivity in ZnO, which possess the smallest ionization energy. Nitrogen doping is a suitable approach to fitune the optical properties of several materials [36]. In the present work, chemical bath deposition (CBD) technique was used to investigate the effects of Nitrogen doping on the optical and dielectric properties of ZnO thin film because of the simple and cost effective nature of technique [7]. Many researchers in the field of material science been given much interest to CBD because it is easy, inexpensive and utilizes simple materials [34]. Additionally, CBD technique improves uniformity, ease selection of substrates and many samples can be deposited in one setup [31]. Thin films produced by CBD and SP techniques find applications in optical imaging, solar window coating, and photovoltaics [31]. The advantages of the chemical techniques involves relatively low temperature and absence of any stringent conditions requiring very basic equipment like a hot plates, magnetic stirrers, hot water bath, and water-soluble salts [14].

Methods

The materials are use as received without further purifications. Zinc Oxide (ZnO) was prepared by dissolving 0.091M Zinc acetate in 100ml of distilled water and stirred for 10 minutes. The Ammonium Chloride (NH_4Cl) precursor was prepared by dissolving 0.037M Ammonium Chloride powder in 100ml of distilled water and stirred for 10 minutes. The Nitrogen doped Zinc Oxide was prepared by mixing ZnO solution with the NH_4Cl solution in the order of increasing NH_4Cl concentration from 0 %, 2 %, 4 % and 6 %. 0.2 ml of TEA which serves as a complexing agent was added to each of solutions and stirred for 20 minutes using a magnetic stirrer to ensure a homogeneous mixture.

Prior the deposition, 75x25x1 mm dimension glass slides were dipped inside acetone for 10 minutes, washed with detergent and raised with distilled water. The already cleaned glass substrates were immersed vertically in the solutions inside beakers and left for 24 hours at room temperature. The substrates were removed and washed with distilled water before drying in an oven at 150ºC for 30 minutes.

The samples were annealed at 350^0c for one hour using muffle furnace after drying. Optical characterization was performed on the samples using UV-vis spectrophotometer to measure the absorbance and transmittance within the wavelength range of 350 nm to 700 nm.

2.0 Results

2.1 Absorbance

The absorbance is at its peak value at about 340 nm for all samples (Fig. 1) it was observed that the 2 %N concentration sample exhibits the highest peak absorbance and the 6 %N concentration sample exhibits the lowest peak absorbance. The absorbance decreases with wavelength from 370 nm to 700 nm. The absorbance was observed to increases by the introduction of Nitrogen into Zinc Oxide, while its high percentage decreases its absorbance as seen for sample with 6 %N (Fig. 1). This is as a result of the size confinement effect for nanoparticles and introduction of impurity into the ZnO [16].

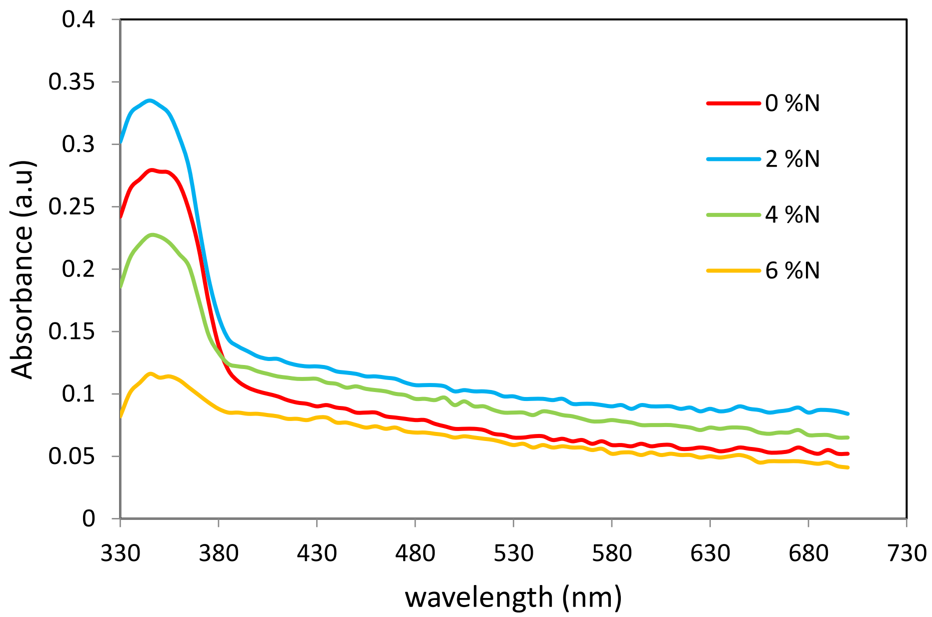

Figure 1. Plot of absorbance against wavelength (nm) for ZnO and N- ZnO Thin Films.

2.2 Transmittance

The transmittance decreases sharply at 340 nm for all the samples with 2 % N concentration sample exhibits the lowest transmittance and the highest transmittance was observed for 6 % N concentration (Fig. 2). The transmittance increases gradually from 370 nm to 700 nm. The addition of Nitrogen into Zinc Oxide reduces its transmittance in the visible region (27). This is as a result of the size confinement effect for nanoparticles and introduction of impurity into the ZnO as seen in the absorbance plot which is the reverse form of transmittance. The incorporation of high percentage increases the transmittance as seen for sample with 6 %N this could be due to alignment between the donor and acceptor atoms.

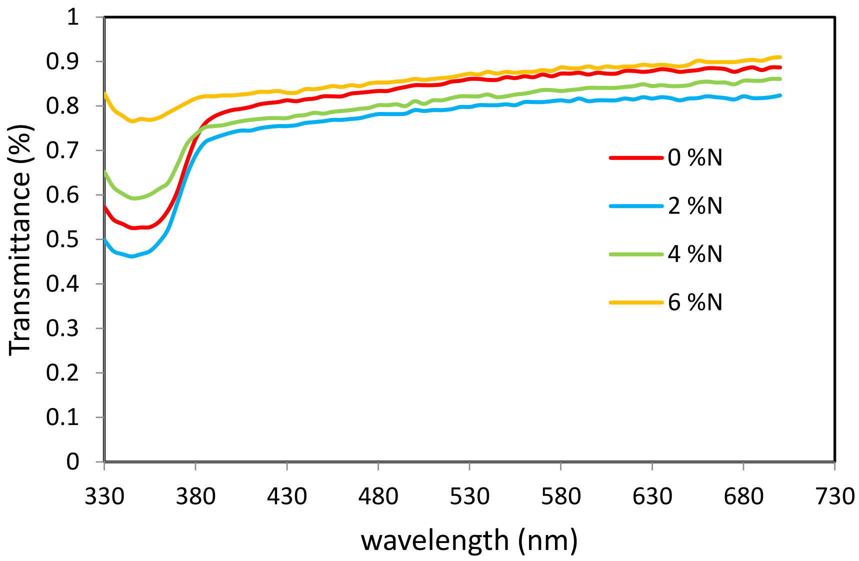

Figure 2: Plot of Transmittance (%) against wavelength (nm) for ZnO and N- ZnO Thin Films.

2.3 Reflectance

The reflectance was calculated by using the equation (1)

R = 1- (A+T) (1)

The highest values of the reflectance of 0.095 %, 0.119 %, 0.110 % and 0.085 % were observed at 345 nm for 0 %N, 2 %N, 4 %N and 6 %N respectively. The reflectance was observed to decrease continuously within the visible region for all the samples, which is in agreement with [34]. Samples with 2 %N and 4 %N exhibit high reflectance compare to the sample with 0 %N. While the sample with 6 %N show lower reflectance (Fig. 3). This implies that the addition of small quantity of Nitrogen into Zinc Oxide increases its reflectance, while the addition of large quantity decreases its reflectance similar to what was observed for absorbance.

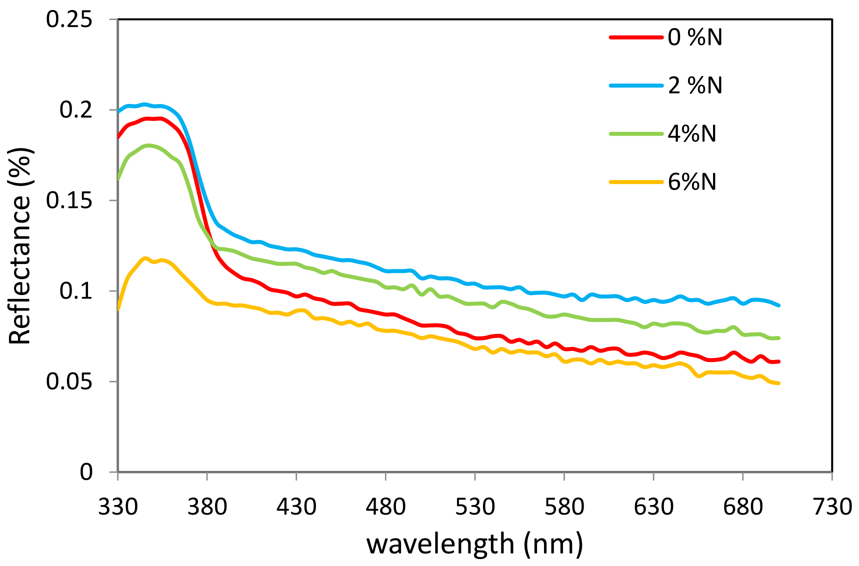

Figure 3: Plot of Reflectance (%) against wavelength (nm) for ZnO and N- ZnO Thin Films.

2.4 Optical Bandgap

The band gap of the material was determined by plotting the graph of $(\alpha h\nu)^2$ against hf, and extrapolating the linear portion of the curve to $(\alpha h\nu)^2 = 0$ as shown in Fig. 4. Where α represent the absorption coefficient and $h\nu$ represent the photon energy. The values of the band gap obtained are 3.55 eV, 3.42 eV, 3.35 eV and 3.15 eV for 0 % Nitrogen concentration, 2 % Nitrogen concentration, 4 % Nitrogen concentration and 6 % Nitrogen concentration as presented in table 1. It shown that an increase Nitrogen concentration leads to an increase film thickness and consequently leads to a decrease in band gap as shown in Table 1, Fig. 5 and Fig. 6 which is in agreement with [29]. The decrease in optical band gap can be attributed to the formation of compensating energy states that arise owing to the nitrogen dopant elements, consequently shifting the conduction band edges [36, 33, 17]. A band gap narrowing was observed due to the effect of electron–electron and electron-impurity scattering [27].

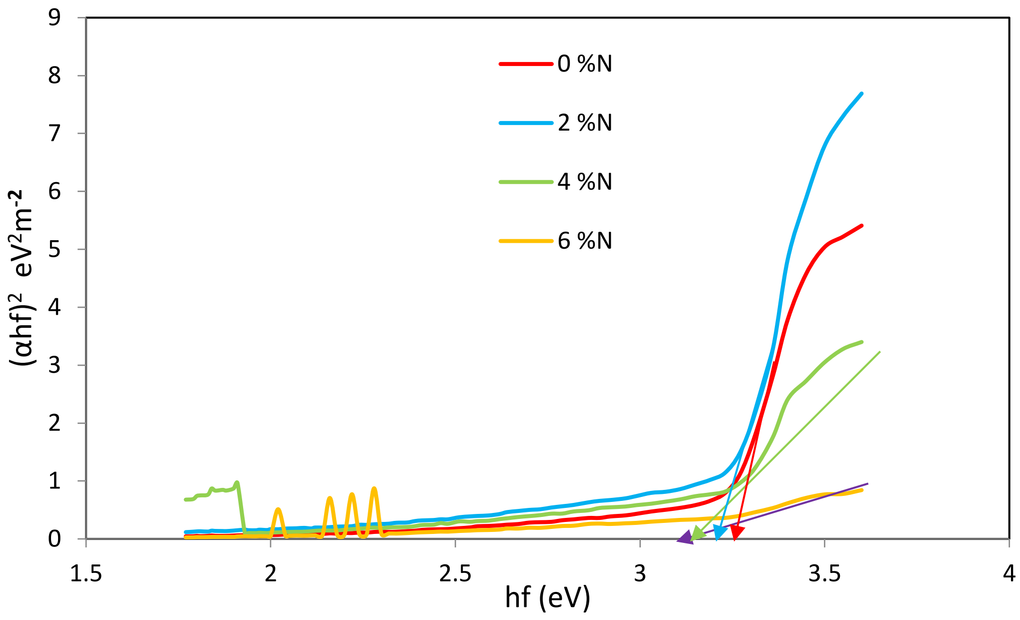

Figure 4. Plot $(\alpha h\nu)^2$ eV^2m^{-2} against hν (eV) for ZnO and N- ZnO Thin Films.

Table 1: Samples and Measured Parameters of Zno and N-Zno

Sample	Band gap (eV)
0 % N	3.55
2 % N	3.42
4 % N	3.35
6 % N	3.15

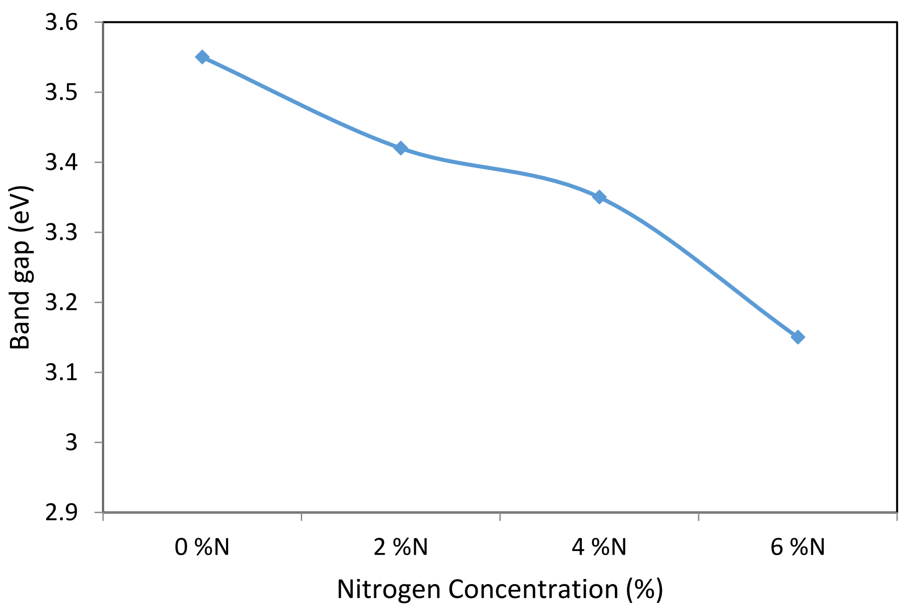

Figure 5: Plot of the Variation of Band gap (eV) with Nitrogen Concentration (%) for ZnO and N- ZnO Thin Films.

2.5 Extinction Coefficient and Refractive Index

The extinction coefficient k is described as ratio of the fraction of light lost due to scattering and absorption to unit distance of the penetration medium. It can be estimated using equation (2), [1].

$$k = \frac{\lambda\alpha}{4\pi} \qquad (2)$$

Where λ represents wavelength, α represents the absorption coefficient. The extinction coefficient has it peak at 355 % nm for 0 %N and 6 %N, at 350 nm for 2 %N and 4 %N. The value of the extinction coefficient decreases sharply between 350 nm to 380 nm which is in agreement with [34]. Its value remained almost unchanged within the visible region as the wavelength increases. The samples investigated show similar patterns except for 2 %N concentration where the extinction coefficient increases as the wavelength increases from 540 nm to 700 nm, which is as a result more charge carrier generation attributed to introduction Nitrogen dopant. The introduction of Nitrogen increases the extinction coefficient of Zinc Oxide within the visible region, but high concentration reduces its value as seen for sample with 6 %N concentration (Fig. 6).

The refractive index n of thin films can be calculated from equation (3), [5].

$$n = (1+R/1-R)+(4R(1-R)^2- k^2)^{1/2} \qquad (3)$$

Where R is the reflectance, k is the extinction coefficient. The peak values of the refractive index were found at 345 nm for 0 %N, 2 %N, 4 %N and 6 %N respectively. The value of the refractive index decreases sharply between 345nm to 380 nm for all the samples. It decreases gradually within the visible region as the wavelength increases (Fig. 7). As observe in the plot of extinction coefficient, the doping of Zinc Oxide with Nitrogen increases the refractive index, but it value decreases with high concentration.

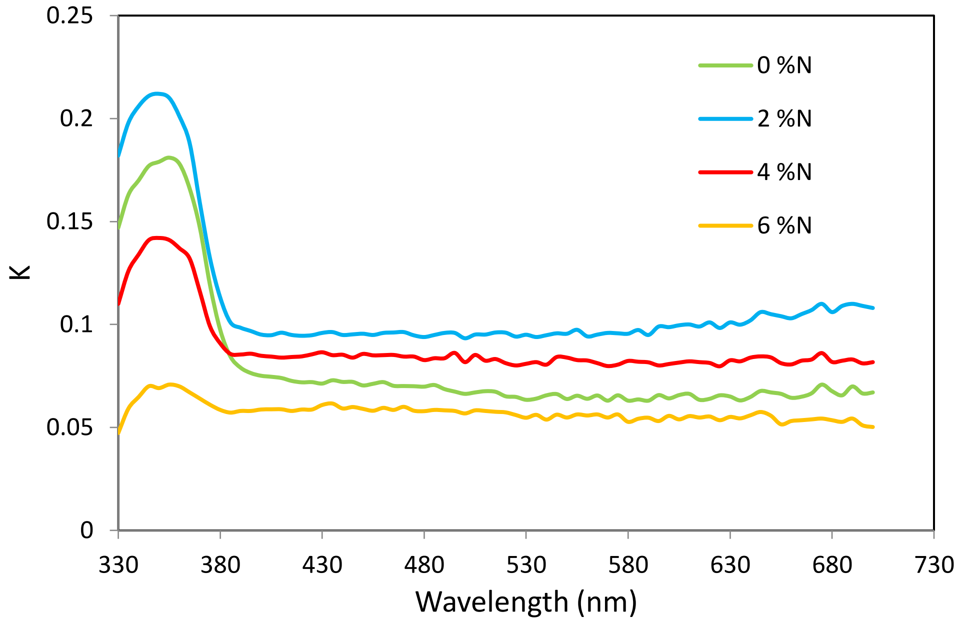

Figure 6: Plot of Extinction Coefficient against Wavelength (nm) for ZnO and N- ZnO thin films.

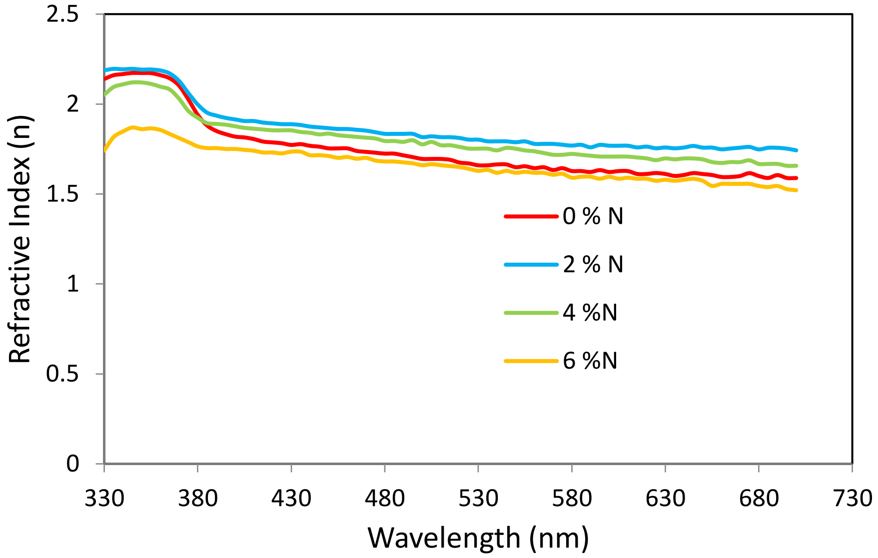

Figure 7: Plot of Refractive Index against Wavelength (nm) for ZnO and N- ZnO Thin Films.

2.7 Dielectric constant

The real ε_r and imaginary ε_i parts of dielectric constant were determined using equation 5 [34].

$\varepsilon_r = n^2 - k^2$ and $\varepsilon_i = 2nk$ (5)

Fig. 8, shows that the real part of the dielectric constant has it peak value at about 360 nm for all the samples. Its value decreases sharply between 360 nm and 380 nm. The values continue to decrease gradually increases in wavelength increases. It has low value within the visible region of the electromagnetic spectrum in which the 2 %N sample has the highest value and the 6 %N sample has the lowest value.

A similar pattern was also observed in the imaginary dielectric constant (Fig. 9). higher values were observed for the imaginary part of the dielectric constant compared to that of the real part within the visible region.

The low values of the real and imaginary parts of the dielectric constant within the visible region is as a result of the dielectric losses at high frequencies and wavelengths [28].

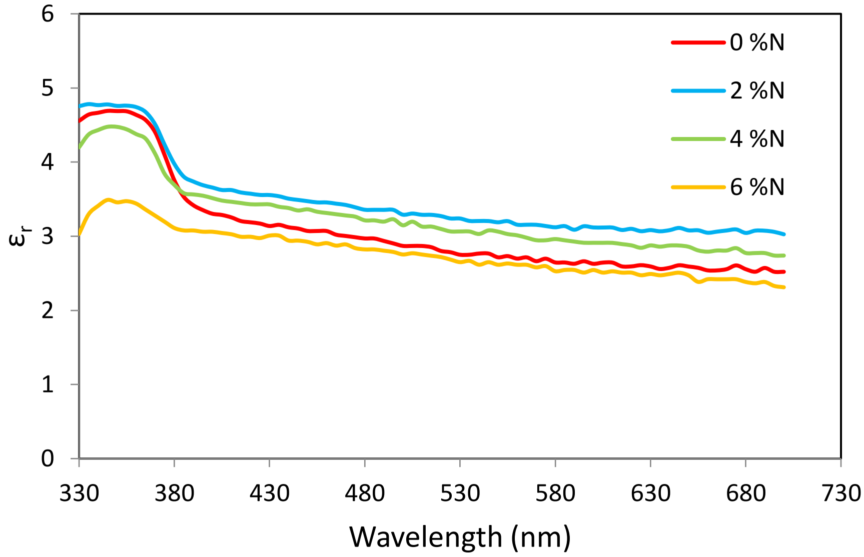

Figure 8: Plot of Real Part of Dielectric Constant against Wavelength (nm) for ZnO and N- ZnO Thin Films.

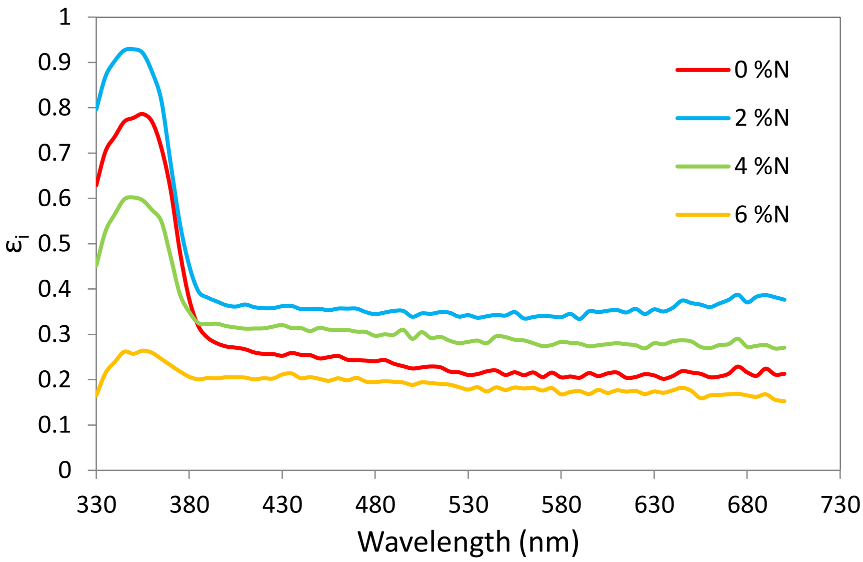

Figure 9: Plot of Imaginary Part of Dielectric Constant against Wavelength (nm) for ZnO and N- ZnO Thin Films.

3.0 Conclusion

The optical and dielectric properties of Nitrogen doped Zinc Oxide thin film deposited by chemical bath method were investigated using UV-vis Spectrophometer within the wavelength range of 330 nm to 700 nm. The peak absorbance was recorded at 340 nm, while the least transmittance was observed at the same wavelength of 340 nm. The 2 % Nitrogen concentration sample was found to have the highest absorbance, while the 6 % Nitrogen concentration was found to have the least absorbance.

An inverse pattern is observed for transmittance at this particular wavelength of 340 nm. The 6 % Nitrogen concentration has the highest value of transmittance, while the 2 % Nitrogen concentration has the least value of transmittance.

The highest values of the reflectances were observed at 345 nm for 0 %N, 2 %N, 4 %N and 6 %N respectively. A continuous decrease in the reflectance was observed within the visible region for all the samples which is in agreement with [34].

The optical band gap decreases from 3.55 eV for undoped to 3.15 eV for 6 % Nitrogen concentration. This is in agreement with **[**29] where the band gap decreases from 3.29 eV for undoped ZnO to 3.15 eV for 0.4 concentration of Nitrogen.

The introduction of Nitrogen increases the extinction coefficient of Zinc Oxide within the visible region, but high concentration reduces it value as seen for sample with 6 %N concentration (Fig. 6). As observed in the plot of extinction coefficient, the doping of Zinc Oxide with Nitrogen increases the refractive index, but it value decreases with high concentration.

The optical conductivity curve has similar pattern to that of refractive index.

Low values of the real and imaginary parts of the dielectric constant were observed within the visible region which is as a result of the dielectric losses at high frequencies and wavelengths [28]..

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Conflict of Interest

The authors declare that there was no conflict of interest.

Availability of data and materials: The data and materials for this research will be made available on request.

Ethics: Consent to Participate, and Consent to Publish declarations: not applicable.

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