Study of the Irradiation Effect by α-Particles on Optical Properties of ZnO:6%In Thin Films

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Abstract. This study investigates the effect of radiation doses of 4.78MeV energy of alpha particles emitted from radium radioactive source (Ra²²⁶), which gives an equivalent dose rate of (5mrem/h) with an activity of (60KBq), on the optical properties of 350nm thickness of the ZnO thin films doped 6% rate of indium (In) that prepared by spray pyrolysis technique. The study of the optical properties included energy gap, reflectivity, absorption coefficient, refractive index, extinction coefficient, real and imaginary insulation constant by recording the transmission and absorption spectra of photons have wavelength range of 300-1000nm before and after exposing the thin film to α-particles to radiation doses of 125, 250 and 500mSv by exposing them to different time periods. When ZnO:6%In thin films exposed to radiation dose, the results generally showed decreasing of the energy gap value, a slight variation in the values of reflectivity, absorption coefficient, refractive index, extinction coefficient, real and imaginary insulation constant at the low dose (125mSv), and a significant variation in the values of these properties at higher doses (250, 500mSv) with a variation in behavior of the curves with the increasing of doses values.

1. Introduction
When the alpha particles (α), which represents the nucleus of He²⁺, pass through a medium, they mainly interact with electrons of this medium due to the Coulomb's force between the charged particles (α-particles) and the electrons, Thus, the dominant mechanism for the loss of α-particle energy is Coulomb scattering [1]. The radiation effect on the thin film represents a transferred linear energy rate which directs proportional with the square of charge value and inversely with the square of speed value [2].

Thin films easily absorb α-particles, and the last can lose all energy in this thin film, then α-particle gains two electrons from the substance that passes through in to turn into a neutral helium atom [3]. The total absorbed radiation dose received by the thin films represents the dose rate multiplied by the exposure time, so increasing the exposure time means increasing the amount of absorbed dose [4]. The number of decay that occurs per second and measured in the unit of Becquerel (Bq) or disintegrations per second (dis/sec) [5]. Radium radioactive source (Ra²²⁶) was used as α-particles radioactive source with the energy of 4.78MeV which gives an equivalent dose rate of 5mrem/h and the activity of 60KBq.

Thin films are defined as one or several layers of material atoms with a thickness of one micron or several nanometers [6]. They Produced by the intensification or deposition of atoms, particles or ions on substrates made of glass, quartz, silicon, aluminum, and other metals according to the nature of use and the types of study [7]. Zinc oxide thin films are semiconductors with a wide direct energy gap and
are used in the manufacture of electronic parts and optical and electro-optical applications such as display panels, solar cells, and manufacturing of Transparent Conductive electrodes [8]. The exposed of the thin films to the radiation doses may improve their behavior, where it was found that the value of the energy gap decreases as a result of the generation of additional energy levels within the energy gap [9]. The increased exposure of thin films to these doses may lead to a breakdown of chemical bonds, increasing interstitial distances and a decrease in density, leading to defects in a crystalline structure, and for further increase of exposure, this leads to the breakdown of crystalline structure [10].

2. Experimental Side and Measurements

The thin films of Zinc Oxide (ZnO) doped Indium (In) in the rate of 6% were prepared by spray pyrolysis technique on thin bases of glass heated at a temperature of 400°C at spraying rate of 5ml/min and pressure of $10^{-3}$ bar as shown in Figure 1.

Where the spray solution was prepared using Anhydrous Indium Chloride (InCl$_3$), which is a white color solid and quick to dissolve in water, Where Indium Chloride substance (InCl$_3$) with 6% ratio was added to Anhydrous Zinc Chloride substance (ZnCl$_2$) with molecular weight of 136.29gm after gradually dissolved it in 100ml of distilled water using a Magnetic Stirrer. Then put the spray solution in the tank of the spray device and make the spray distance of 30cm between the glass bases and the nuts after placing the glass bases over the electric heater until it reaches the required temperature (400°C), Then we sprayed for time of 5sec followed by a stopping time for 1min to ensure that the heat returned to the original value and to complete the process of crystalline development, This process is repeated several times until the desired thickness is achieved. After the deposition process, the film is left on the surface of the heater after it is extinguished to cool down without attempting to lift it. To complete the chemical reactions on the surface of the film, the thickness of the samples was measured and its values were 350nm by using the FT-650 film thickness probe system device. The absorption and transmission spectra of the thin films were measured for the range of the 300-1000nm wavelengths using the spectrometer, and optical constants were studied before and after the exposed of the ZnO:6%In thin films to the radioactive doses of 125, 250 and 500mSv using radium radioactive source (Ra$^{226}$). Optical transmission (T) is defined that it is the ratio of the transmitted rays (I) to the incident rays ($I_o$) intensity on the substance as shown in the following equation: [11]
\[ T = \frac{I}{I_0} \]  

(1)

The absorption (A) can be calculated as follows: [12]

\[ A = -\log(T) \]  

(2)

The value of reflectivity (R) can be calculated from the knowledge of the values of both absorption (A) and transmission (T) according to the following equation [13]:

\[ R + A + T = 1 \]  

(3)

By determined of the absorbent substance thickness (t), the values of the absorption coefficient (\( \alpha \)), which represents the decrease in the flux of the incident rays for the distance unit towards the propagation of the wave within the medium, are given in the following equation [14]:

\[ \alpha = 2.303 \times \frac{A}{t} \]  

(4)

Also, the value of the energy gap (\( E_g \)) is calculated by the following equation [14]:

\[ a\hbar = A(h\nu - E_g)^r \]  

(5)

where \((h\nu)\) represents the absorbed photon energy, and \((r)\) represents an exponential factor dependent on the type of transition.

Extinction coefficient (\( K_o \)) is defined as an amount of the energy absorbed in the film, or other words represents the inertia of the electromagnetic wave within the substance. The extinction coefficient is calculated through the following equation [15]:

\[ K_o = \frac{a\lambda}{4\pi} \]  

(6)

where \(\lambda\) represents the absorbed photon wavelength.

So, the refractive index is defined as the ratio between the speed of light in the vacuum and its velocity within the substance, where it is given by the following equation [13]:

\[ n_0 = \left[ \frac{(1 + R)^2}{(1 - R)^2} - (K_0^2 + I) \right]^\frac{1}{2} + \frac{(1 + R)}{(1 - R)} \]  

(7)

The interaction between the light and medium charges occurs because of the absorption of energy in the substance and thus polarization of the charge of that medium. This polarization is usually described by the complex electrostatic insulation constant (\( \epsilon \)) of the medium and can be calculated as follows [13]:

\[ \epsilon = \epsilon_1 - i\epsilon_2 \]  

(8)

where \(\epsilon_1\) and \(\epsilon_2\) represent the real and imaginary insulation constant respectively whose values are given in the following mathematical equation [13]:

\[ \epsilon_1 = n_0^2 - k_0^2 \]  

(9)

\[ \epsilon_2 = 2n_0k_0 \]  

(10)
3. Results and discussion

3.1. Transmittance spectrum (T)

The transmission spectrum of ZnO:6%In in the thin film was studied before and after irradiation of α-particles with a radiation dose of 125, 250 and 500mSv using equation (1) where, in general, it was found that the transmission increases with increasing wavelength, as shown in Figure 2. The behavior of the transmission spectrum of thin films exposed to low dose (125mSv) is fully approached pre-irradiation as shown in figure 1, but when thin films exposed to higher doses (250 and 500mSv) it was observed that there is a variation in the behavior of curves then it was before irradiation, and the reason is that the increase of radiation doses led to breakdown the chemical bonds among thin film atoms and this leads to increase distances between the atoms and decrease in the density and the transmission increases accordingly [16], where the transmission reached as high as possible at the dose 250mSv and then the thin film reached the saturation state by absorption of most of the α-particles by acquiring two electrons from the thin film atoms and converted to a neutral helium atom [17]. After that, the transmission value slightly decreases at the highest dose (500mSv). The variation in transmission curve behavior at higher doses (250 and 500mSv) than its behavior at low dose (125mSv) indicates that the total dose increases by increasing the exposure time, therefore the defects caused by radiation within the thin film will increase, which could lead to a flaw in the crystal structure [18].

3.2. Absorption Spectrum (A)

The absorption spectra of the ZnO:6%In thin film before and after irradiation by different radiation doses of α-particles, that were calculated using equation (2) and as shown in Figure 3, show opposite behavior of transmission spectra. Absorption decreases with increasing wavelength because the incident photons cannot lift the electrons from the valence band to conduction band where the energy of incident photons is less than the value of the energy gap of the ZnO:6%In in the thin film, where the relationship is inverse between the wavelength and photon energy [19]. So, the absorption decreases with increasing wavelength. We also note the variation of the absorption curve behavior at radiation doses (250 and 500mSv) than its behavior at a lower dose (125mSv), and this confirms the effect of α-particles on the absorption spectrum.
3.3. Reflective Spectrum (R)

The reflectivity was calculated from transmission and absorption spectra according to the equation (3). Figure 4 shows the reflectivity spectrum of ZnO:6%In thin film before and after irradiation with α-particles with radiation doses mentioned above as a function of wavelength, where the reflectivity value is found to be as high as possible at the wavelength of 500nm and decreases with increasing wavelength, this figure shows that α-particles didn’t change the nature of the curve at low radiation dose (125mSv), but in the higher doses (250 and 500mSv), one can observe a variation in the behavior of the reflective curve by shifting the reflective curve peaks to the wavelengths below 500nm and this explains that the α-particles led to the breakdown of the bonds and thus caused a change in the crystalline structure of the lattice [20].

Figure 4: Reflectance spectrum (R) as a function of the wavelength (λ) of Zn:6% In thin film before and after irradiation by α-particles.
3.4. Absorption Coefficient ($\alpha$)

The absorption coefficient represents the decreasing in the energy flux of the incident ray relative to the distance unit towards the propagation of the wave within the material, it was calculated by equation (4). Figure 5 shows the variation in the absorption coefficient of the thin films (ZnO:6%In) as a function of the photon energy before and after exposure the thin film to irradiation with $\alpha$-particles in the radiation dose mentioned above, the absorption coefficient is increased by increasing the energy of the incident photons, and there is a gradual increase in the values of the absorption coefficient at the range ($h\nu<3eV$). Then we observe a fast increase of the absorption coefficient at the energies range ($h\nu>3eV$), this shows direct electronic transitions between the valence and conduction bands, and help us to predict the edge of exponential absorption [21]. The absorption coefficient was reduced after exposed the thin film to $\alpha$-particles, due to increasing of transmission, where the variation in the behavior of the absorption coefficient was slightly variation at low doses (125mSv), while one can notice that there is a significant decrease in the coefficient of absorption coefficient at the higher doses (250 and 500mSv), which confirms the presence of the effect of $\alpha$-particles on this optical constant [22].

![Figure 5: Variation of absorption coefficient ($\alpha$) as a function of photon energy ($h\nu$) of Zn:6%In thin film before and after irradiation by $\alpha$-particles.](image)

3.5. Energy Gap ($E_g$)

The optical energy gap of the ZnO:6%In thin films was calculated for the direct allowed electron transitions by the equation of (5) before and after exposure the thin films to irradiation with $\alpha$-particles in the different radiation doses identified in the current research, it is determined by the value of the energy gap from the point of intersection of straight line which clear in Figure 6, which is obtained at the value of $(\alpha h\nu)^2=0$. Figure 6 shows that the energy gap value of the ZnO:6%In thin films before exposed the thin film to irradiation with $\alpha$-particles was 3.207eV and after exposed the thin films to 125, 250 and 500mSv values of radiation doses of $\alpha$-particles were 3.192, 3.204 and 3.198eV respectively. In general, there is a decrease in the energy gap values as a result of the generation of additional energy levels within the forbidden region between the valence and conduction bands [23].
3.6. Extinction coefficient ($K_o$)

The extinction coefficient was calculated by equation (6). Figure 7 represents the behavior of the extinction coefficient as a function of photon energy for ZnO:6%In thin film before and after exposed it to $\alpha$-particles with the identified different radiation doses, the extinction coefficient behaves very similar to the behavior of the absorption coefficient because they are related to each other according to the equation (6) that relate the extinction coefficient with the absorption coefficient. One can notice from figure 7 that there is a gradual increase in the values of the extinction coefficient at the photon energies less than the absorption edge and followed by a clear increase in energies of $h\nu>3$eV, this increase indicates that direct electronic transitions between the valence and conduction bands led to an increase in the absorption coefficient and thus an increase in the extinction coefficient. Figure 7 shows that $\alpha$-particles didn’t change the nature of the curve at low radiation dose (125mSv), and at the higher doses (250, 500mSv), there is a variation in the behavior of the curve extinction coefficient, this is because the increased doses increase the defects caused by radiation on the thin film, which leads to breakdown the bonds between the atoms of thin film and increases the distances between the interface and decrease in density [24]. Thus, the increase of transmission and decrease of absorption leads to a decrease in the extinction coefficient.

Figure 6: Variation of $(a\nu)^2$ as a function of photon energy ($h\nu$) of Zn:6%In thin film before and after irradiation by $\alpha$-particles.

Figure 7: Variation of extinction coefficient ($K_o$) as a function of the photon energy ($h\nu$) of Zn:6%In thin film before and after irradiation by $\alpha$-particles.
3.7. Refractive index ($n_o$)

The refractive index was calculated by equation (7). Figure 8 shows the variation in refractive index as a function of the photon energy of the ZnO:6%In thin films before and after exposing it to the radiation doses of 125, 250 and 500mSv. In general, figure 8 shows that the refractive index increases with the energy of the photon increasing until it reaches its highest value (2.644) at energies near the energy gap and then after declining sharply [8].

![Figure 8: Variation of reflection index ($n_o$) as a function of photon energy (hν) of the Zn:6%In thin film before and after irradiation by α-particles.](image)

The behavior of the refractive index at the irradiation with a low dose (125mSv) identical to its behavior before irradiation of thin film with α-particles, but at higher doses (250 and 500mSv), one can observe the deviation of the refractive index peaks to high photon energies without significant variation in the refractive index value.

3.8. Real and Imaginary Insulation Constant ($\varepsilon_1$, $\varepsilon_2$)

The real and imaginary insulation constant was calculated using equations (9) and (10). Figure 9 shows the variation of the real insulation constant as a function of the photon energy of the ZnO:6%In thin films before and after exposure of the thin film to the studied radiation doses, it is generally observed in Figure 9 that the real insulation constant behaves similarly to the refractive index because of their correlation according to the equation (9). Also, one can notice that the real insulation constant increases with the energy of the incident photons until it reaches its highest value at energies close to the energy gap value and then decreases sharply [8]. The behavior of the real insulation constant at a lower dose of irradiation (125mSv) corresponds to its behavior before irradiation with α-particles, but at higher doses (250 and 500mSv), a deviation of the real insulation curve peaks towards high photon energies can be observed without significant variation in their value.
Figure 9: Variation of real insulation constant ($\varepsilon_1$) as a function of photon energy ($h\nu$) of the Zn:6%In thin film before and after irradiation by α-particles.

Figure 10: Variation of imaginary insulation constant ($\varepsilon_2$) as a function of photon energy ($h\nu$) of Zr:6%In thin film before and after irradiation by α-particles.

Figure 10 shows the variation of the imaginary insulation constant as a function of the photon energy of the ZnO:6%In thin films before and after exposing it to the radiation doses above. One can observe from figure 10 that the imaginary insulation constant curve is similar to the real insulation constant curve but its value is less than the real insulation constant, i.e. $\varepsilon_1 > \varepsilon_2$. The behavior of the imaginary insulation constant at lower irradiation dose (125mSv) corresponds to its behavior before irradiation of the thin film with α-particles. Either at higher doses (250 and 500mSv), one can observe the increase of curve peaks of the imaginary insulation constant towards high photon energies.

4. Conclusions
Variation of the values and behavior of the spectral distribution curves which have been studied in the present research as a function of photon energy after exposed the ZnO:6%In thin film to the relatively low dose (125mSv) was slight and close to what it was before irradiation, but, But when increasing of
the radiation dose value to 250 and 500mSv had led to varying the values and curves of the spectral distribution of the ZnO:%6In this thin film. Radiation doses generally had led to a decrease in the energy gap values in thin films due to the generation of additional energy levels within the restricted area between valence and conduction bands.

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References
[1] Leybold Didold Didactic Gmbtt 2006 Atomic and Nuclear Physics.
[2] S Krane Kenneth 1988 Introductory Nuclear Physics, John Wiley & Sons Inc. 246–269.
[3] Yu A Zaykin, B A Aliyev 2002 Radiation Physics and Chemistry. 227-230.
[4] Young H D and Freedman R A 2006 University Physics. 1664-1647.
[5] Gh A Eid, A I Kany, M M El-Toony, I I Bashiter and F A Gaber 2013 Arab Journal of Nuclear Science and Applications. 46(2) 226-233.
[6] K D Lever 1971 Thin Films. Wykeham Pub, London 32.
[7] R W Berry, P M Hall 1979 Thin Films Technology. New York: McGraw-Hill 63.
[8] G Singh, S B Shrivastava, Deepjitain, Swati Pandya 2010 India Academy of Science. 33(5) 581-587.
[9] C C Regimol, C S Menon, 2007 Materials Science (Poland). 25(3).
[10] Olga Budenkova, M. Vasiliev, V. Yuferev, V. Kalaev 2007 Journal of Crystal Growth. 303(1) 156-160.
[11] H H Wieder 1970 Intermetallic Semiconducting Films. Oxford: Bergamo Press Inc 402.
[12] A N Donald 1992 Semiconductor Physics and Devices. USA: Irwin 321.
[13] O Stenz et. al. 2005 The Physics of Thin Film Optical Spectra.
[14] S S AL- Rawi, S J Shakir, Y M Hasan 1990 Solid State Physics. AL-Mosul University Arabic Version 134.
[15] T Seiyama 1992 Analytical Chemistry. 34 p1052.
[16] Aliaa H Ali, Adwan N Hameed, Sabah J Fathi 2016 Tikrit Journal of Science Exchange. 21(4) 131-136.
[17] R L Clough 2001 Nuclear Instruments and Methods in Physics Res. Sec. B 185 8-33.
[18] J W Allen 2007 Journal of Crystal Growth. 303(1) 156-160.
[19] KB Sundaram and G K Bhagavat 1981 J. Phys. D:App. Phys. 14 921-925.
[20] A G Nilens 1973 Deep unparity in Semiconductors (Wiley-Interscience Publication).
[21] Arun Poudel et. al. 2017 Applied science. 10(7) 1094-1104.
[22] Asma Ahmed Aziz 2012 Nahrain University Journal of Science 15(3) 27-31.
[23] BL Mattes, L Kazmarsk 1980 Polycrystalline and Amorphous Thin Films and Devices (U.S.A.: Academic Press) 489.
[24] R Plugaru, A Istrate, I Mihalache and R Gavrila 2015 International Conference on Defects in Semiconductors 31(2) 143-146.