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Optical properties of ZnO thin films under impact of alpha particles

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Original Research

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Abstract:

This paper investigates the impact of alpha particle irradiation on the optical characteristics of zinc oxide (ZnO) thin films, which were produced using various concentrations. A zinc oxide thin film was fabricated using the chemical bath deposition (CBD) technique, and glass was selected as the substrate material. A 5 MeV alpha particle beam was emitted during the irradiation process, created by a 241-Americium source. The optical properties of ZnO films within the wavelength range of 200 to 800 nm was measured using a UV-Vis spectrophotometer. The Fourier transform infrared (FTIR) spectrum exhibits a distinct peak at 490 cm^{-1}, characteristic of zinc oxide (ZnO). Increasing the duration of alpha particle irradiation leads to a significant increase in intensity while causing only a slight alteration in the peak at 2500 cm^{-1}. Optical measurement revealed a decrease in the direct energy band gaps as the radiation doses increased. This trend was observed for different durations 20, 40, 60, 80, and 100 minutes and across various concentrations 0.05, 0.1, 0.2, 0.3, 0.4, and 0.5 M. The energy band gaps ranged from (3.48 - 3.12) eV, (3.02 - 2.61) eV, (3.25 - 2.84) eV, (3.01 - 2.69) eV, (2.65 - 2.32) eV. Similarly, the band gaps diminish as the irradiation doses increase at various durations and concentrations. Additionally, the energy is likewise characterized as indirect. The energy ranges are as follows: (3.74 – 3.52) eV, (3.63 – 3.37) eV, (3.58 – 3.26) eV, (3.49 – 3.25) eV, (3.47 – 3.20) eV, and (3.42 – 3.20) eV. Our research revealed that the most favorable and smallest direct energy gap value ranges from 2.65 to 2.32 eV, while the indirect energy gap value ranges from 3.42 to 3.20 eV. These optimal values were seen at a concentration of 0.5 M, and the ideal irradiation time was determined to be 100 minutes. The energy gap for direct and indirect transitions can be reduced by increasing the concentration of alpha particle irradiation during the growth of ZnO nanotubes using the CBD method. These nanotubes have potential use in nano-photodetectors.

Keywords: CBD; ZnO thin films; Band gap; Alpha particles; FTIR

1. Introduction

White ZnO powder is cheap and easy to work with, making it ideal for sensors and ultraviolet lasers [1]. ZnO, an n-type semiconductor, was suitable for short wavelength emitting applications due to its 3.37 eV direct band gap, 60 eV excitation-binding energy at room temperature [2], 100 cm^2/V.s electron mobility [1], high visible spectrum clarity, low chemical toxicity, good bioefficiency, and high thermal stability near room temperature [3]. CBD can produce ZnO films from aqueous solutions quickly, cheaply, and in huge quantities without a vacuum or high-temperature apparatus. Controlled solution chemical precipitation produces high-quality deposits on suitable surfaces [4]. CBD-produced ZnO film characteristics depend on precursors, additives, solvents, CBD time, temperature, and substrate type [5]. ZnO suffers from photo corrosion, faster electron-hole pair recombination, and UV light dependency. There are many uses for zinc oxide. Dual transistors, laser diode clear electrodes, transparent flat screen electrodes for plasma and liquid crystal displays, thermal solar cell aggregations, low radiation-strength glass, luminous magnetic semiconductors, and microwave lines enhance chemical visualization [6]. B. Taunk et al. 2015 [7] zinc oxide thin films were deposited on a glass slide from an aqueous solution of $ZnCl_2$ and NaOH by a chemical bath deposition method. The films of various thicknesses have been obtained by varying the concentration of TEA (1 - 0.01) M. Optical properties, surface morphology, and particle size of the deposited thin film have been studied by U.V spectrophotometer (Varian) SEM and XRD. The optical band gap of the ZnO thin film was found in the range of (2.59 - 3.57) eV.

Mursal et al. 2018 [8] ZnO thin films were successfully deposited from zinc acetate utilizing spin coating and annealing at various temperatures in this job. The qualities of ZnO thin films were discovered to be that they were highly sensitive to annealing temperature. ZnO films have an optical band gap in the region of (3.82 – 3.69) eV. With increasing annealing temperatures, the optical band gap of ZnO films decreases.

In 2018, Monalisha Goswami et al. [9] the zinc oxide nanoparticles were synthesized by the chemical deposition method, the UV-visible The absorption spectra show a modest red shift with a reducing in the optical band gap ranging from 3.84 to 3.56 eV an temperature rise during heat.

In 2019, Amna Raad Dahham et al. [10] created a zinc oxide coating in a vacuum using a closed oven on glass platforms at room temperature. In the wavelength range of (-250110) nm, the optical characteristics of the produced films were investigated. The transmission spectra revealed extremely transparent coatings with energy gaps ranging from 3.47 eV to 3.3 eV.

In 2019, B. Abdallah et al. [11] an investigation was carried out into the effect of exposure to alpha particles on the structural and optical properties of ZnO thin films produced by RF-magnetron sputtering. After alpha irradiation, the XRD data did not reveal any significant changes in grain size. The UV-vis transmittance spectra determined the optical band gap of the ZnO films formed on the glass. For both 300 nm and 700 nm thicknesses, the band gap was found to be marginally decreased after alpha irradiation from 3.37 eV to 3.36 eV.

In 2020, Amanullah Fatehmulla et al. [12] the current study focused on the fabrication of ZnO thin films using the sol-gel method approach, both undoped and doped with various Cu concentrations (0.0%, 0.8%, 3.0%, 5.0%, 10.0%, and 20.0%). The optical band gap (E_g) for undoped films is 3.239 eV and increases slightly to 3.248 eV when doped with 0.8% Cu concentration but decreases further with increasing Cu doping concentration, from 3.248 eV to 3.107 eV for 20% Cu. The rise in Cu doping concentrations improves the E_U (Urbach Energy). The PL (photoluminescence) spectra show ultraviolet (UV) and blue-band emissions. UV's maximum intensity. It decreases as the concentration of copper doping increases, but the intensity of the blue peaks increases. These findings show that the Cu-doped ZnO films meet the requirements for use in various blue emission devices.

In 2020, Awatif Sami Abbas et al. [13] chemical bath deposition (CBD) thin films of zinc oxide (ZnO) with ammonia solution to 300 mL of aqueous solution $Zn(NO_3)_2.6H_2O$ (0.3 M) are formed on commercial glass substrates with bath temperature $(80 + 5)$ °C and annealing temperatures varying from (373, 473, and 573) K at constant time of 1 h. The structural feature examined by X-ray diffraction (XRD) is polycrystalline with hexagonal wurtzite structure for all samples. The optical measurements revealed that the nature of the optical transition was directly allowed, with average band gap energies increasing from 3.77 eV to 3.78 eV with increasing annealing temperature at (373 and 473) K, except at 573 K, where it decreased to 3.73 eV.

R. A. AL-Wardy et al. 2021 [14] have studied the zinc oxide films were prepared by using a(CBD). The optical properties were measured, and it was found that the energy gap increased slightly from through the increase in the annealing temperature (373, 473) K, while the band gap reduced to 3.73 eV at an annealing temperature of 573 K.

Noubeil Guermat el at. 2021 [15] in this work, the fabrication and characterization of thin films of zinc oxide (ZnO) the spray pyrolysis technique was used on glass substrates. The optical gap measurements calculated based on optical measurement were found to be in the range of (3.26 – 3.30) eV.

In 2022, Z. Th. Abdulameer et al. [16] zinc oxide nanorods (ZnO NRs.) film prepared by hydrothermal method ultraviolet-visible absorption spectra showed the optical band gap ultraviolet-visible absorption spectra showed the optical band gap, the energy gap of ZnO NRs. was 3.2 eV and 2.5 eV for ZnO/CdZnS film.

The purpose of this study is to prepare ZnO thin films using a chemical bath technique and to investigate the effect of alpha irradiation on their optical properties that can be utilizing in nano-photodetector applications.

2. Experimental work

We fabricated zinc oxide films on glass substrates using chemical bath deposition. After washing the substrate with distilled water for 5 minutes, it is immersed in acetone for an additional 5 minutes to remove any impurities. Next, we rinsed it with distilled water and dried it in an oven at 50 °C. The zinc oxide film was created using six different concentrations by dissolving pure aqueous zinc nitrate, a white, water-soluble solid with a molecular weight of 297.5 g/mol. We measured the material using a Mettler scale with an accuracy of 10 mg. We dissolved the chemical in 200 mL of distilled water using a magnetic stirrer (700 Hz) for approximately 15 minutes until we obtained a clear, uniform, transparent, and colorless solution. After completing the dissolution process and obtaining the appropriate solution, a 25% ammonium hydroxide solution is added gradually by distillation using a buret until the solution undergoes a noticeable change in color and turns into a milky color. We continue the addition process until the solution again obtains a clear, uniform, and transparent color. We increase the temperature of the solution to (75 - 80) °C and then immerse the glass in an upright position inside the solution for thirty minutes. All solutions should have a pH ranging from 9 to 10.5. Depositing a zinc layer on glass slides results in a white coating. After that, the slices were dried, and the thickness of these films was measured by optical interference and was 79 nm. Next, the slices are exposed to alpha particles for several periods 20, 40, 60, 80, and 100 minutes. The samples are then analyzed using a UV spectrophotometer. Samples are also analyzed using Fourier

transform infrared (FTIR) spectroscopy. As shown in Figure 1.
From Figure 2 shows the effect of alpha particles on the optical properties of ZnO thin films, where the figure shows the relationship between the optical absorption spectrum versus the wavelength of ZnO film at different concentrations 0.05, 0.1, 0.2, 0.3, 0.4, and 0.5 M with different irradiation time of alpha particles 20, 40, 60, 80, and 100 min. Shows from the graph that the absorbance increases, and this is due to the process of crystallization of water, which works to close the pores in the gaps, this increases absorption because the crystalline water fills the holes, causing a new distribution of particles to work where the light does not pass directly and thus increasing absorption that the maximum value of absorption in the UV is 100 minutes, As shown in Fig. 2 (a, c, e, and f), the minimum value of the UV absorption choice is 20 minutes. In comparison, the maximum value is 60 minutes, as indicated in Fig. 2 (b,d).
Figure 3 shows the relationship between the direct vertical band gap energy of the ZnO film and the horizontal axis photon energy exhibit histograms of the direct energy band gap of ZnO nanoparticles exposed to various quantities of alpha radiation. As indicated in Eq. (1), the optical band gap (E_g) was calculated using a Tauc equation [17].

$$\alpha h\nu = A(h\nu - E_g)^n \tag{1}$$

where α is the absorption coefficient, $h\nu$ is photon energy, A is constant, E_g is the optical energy band gap, and n is transmission type dependent (equals 1/2 for permitted direct transmission and 2 for permitted indirect transmission). The optical band gap in non-irradiated ZnO is 3.48, 3.02, 3.25, 3.20, 3.01, 2.65 eV for a concentration of 0.05, 0.1, 0.2, 0.3, 0.4, 0.5 M, as shown in the black-colored drawing.
Figure 3 (a) explain the varied irradiation times of zinc oxide are irradiated for 20, 40, 60, 80, and 100 minutes, the resulting band gaps are 3.39, 3.33, 3.28, 3.21, and 3.12 eV. As for Figure 3 (b), the values are 2.96, 2.89, 2.80, 2.70, 2.61 eV and 3.18, 3.08, 2.99, 2.90, 2.84 eV Figure 3 (c). And Figure 3 (d) are 3.15, 3.06, 3.01, 2.90, 2.28 eV. As for Figure 3 (e and f), For the identical doses, the energy gaps are 2.91, 2.83, 2.78, 2.75, 2.69 eV and 2.61, 2.55, 2.47, 2.40, 2.32 eV, respectively. As a result, we see that the band gap energy drops with increasing doses of alpha radiation. We discovered that the optimal irradiation time is 100 minutes. B. Abdullah [11] reported similar behavior, as seen in Table 1. It was also determined who was the best and who had the worst values of the direct energy gap (2.65 - 2.32) eV for a concentration of 0.5 M. This means that the molar concentration influences the value of the energy gap.
Increasing molarity decreases the energy gap value of this ZnO thin film, It supports the findings of the previous study [18] that increasing molarity results decreases the energy gap of ZnO thin film. This result can be used in transistors, capacitors, and solar cells.
Figure 4 shows the relationship between the ZnO film's indirect band gap energy and photon energy by displaying histograms of the indirect energy band gap of ZnO nanoparticles exposed to various quantities of alpha radiation. As demonstrated in Eq. (1), a Tauc diagram was used to compute the optical band gap (E_g).
The optical band gap in non-irradiated ZnO is 3.74, 3.63, 3.58, 3.49, 3.47, and 3.42 eV at a concentration of 0.05, 0.1, 0.2, 0.3, 0.4, and 0.5 M, as shown in the black-colored drawing. Figure 4 (a) When different dosages of zinc oxide are irradiated for 20, 40, 60, 80, and 100 minutes, the resulting band gaps are 3.69, 3.66, 3.62, 3.58, and 3.52 eV.

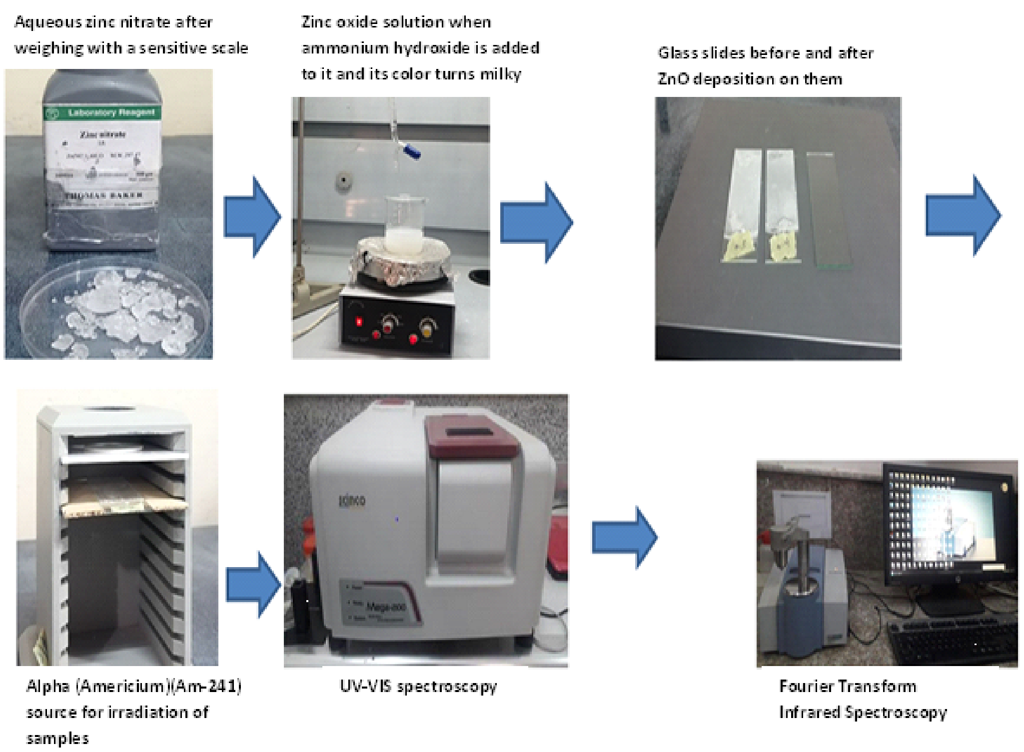

Figure 1. Steps of work.

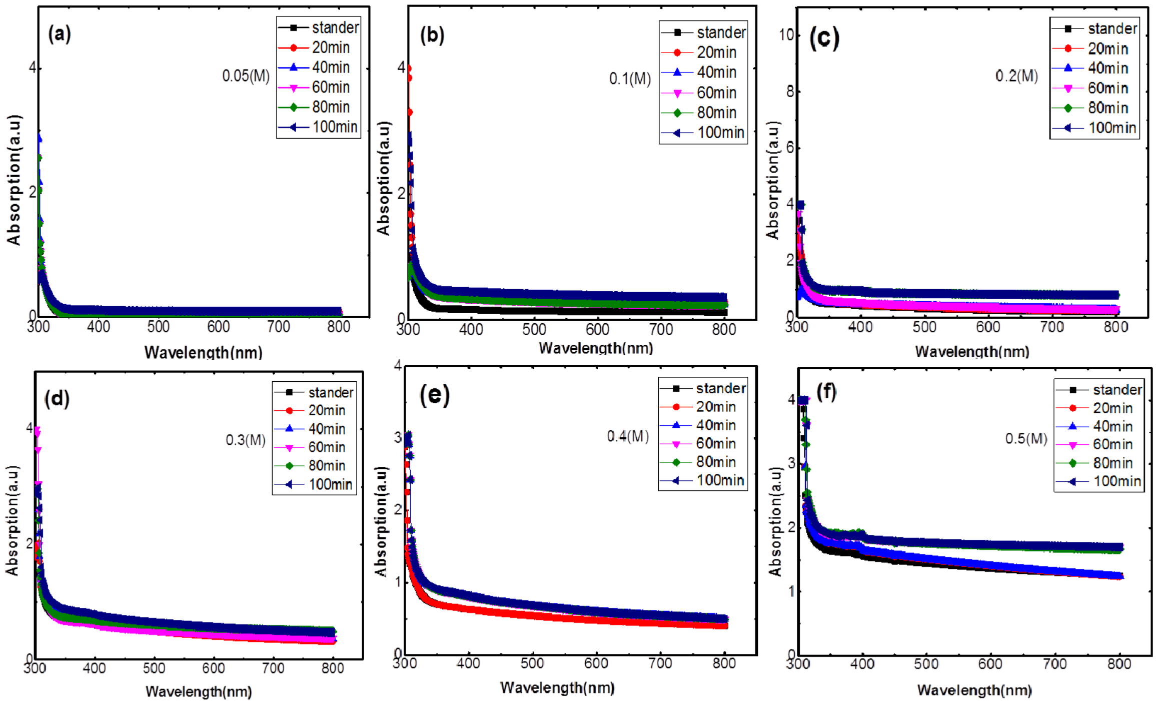

Figure 2. Represent absorption spectrum versus wavelength of a zinc oxide film.

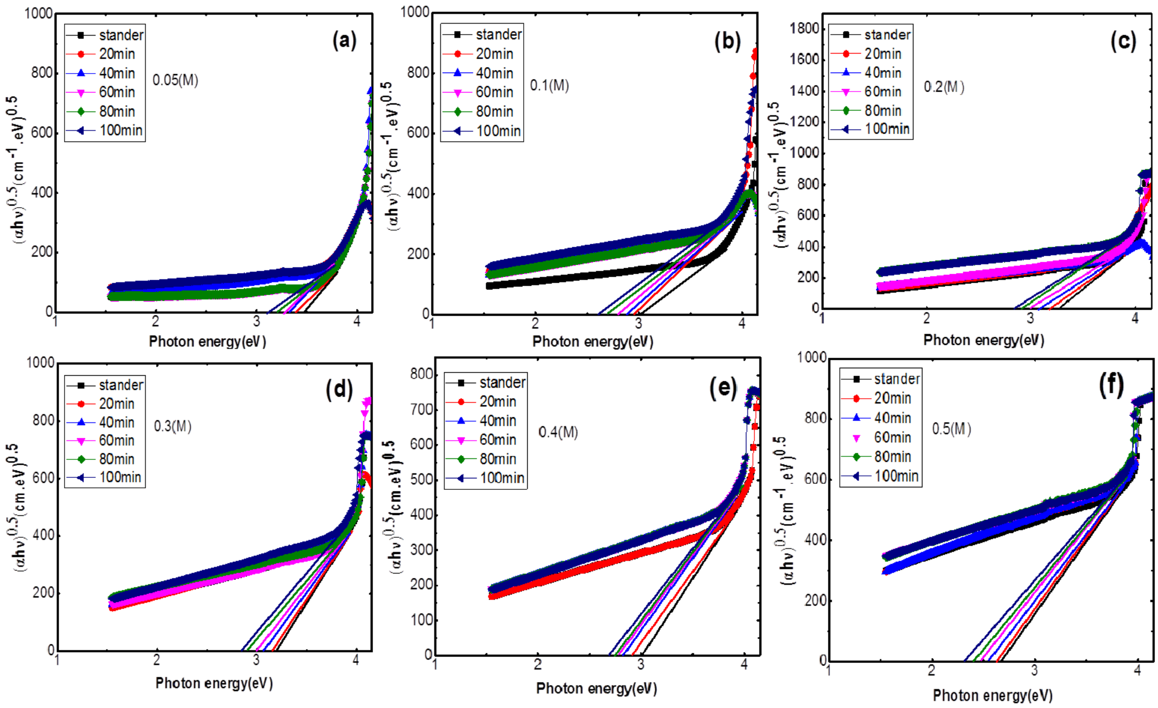

Figure 3. Tauc plot $(\alpha h\nu)^{0.5}$ corresponds to the energy gap $(h\nu)$ of the ZnO film for different irradiation concentrations and times.

Figure 4 (b) is 3.54, 3.51, 3.49, 3.42, and 3.37 eV, while Figure 4 (c) is 3.52, 3.47, 3.39, 3.32, and 3.26 eV. And the values are 3.45, 3.40, 3.36, 3.30, and 3.25 eV (Figure 4 (d)). As for Figure 4 (e and f), For the identical doses, the energy gaps are 3.41, 3.37, 3.31, 3.25, and 3.20 eV; and 3.37, 3.34, 3.30, 3.25, and 3.20 eV, respectively. As a result, we notice that the band gap energy goes down when different doses of alpha radiation are used. We found that the best irradiation time is 100 min. Similar behavior was reported by B. Abdullah [11]. obtained the best and least value of the indirect energy gap (3.42 – 3.20) eV for a concentration of 0.5 M, as shown in Table 1. The results showed that there is an effect of the solution concentration on the band gap value. With increasing solution concentration, the band gap decreases [19].

Figure 5 shows that the largest energy gap for passive direct

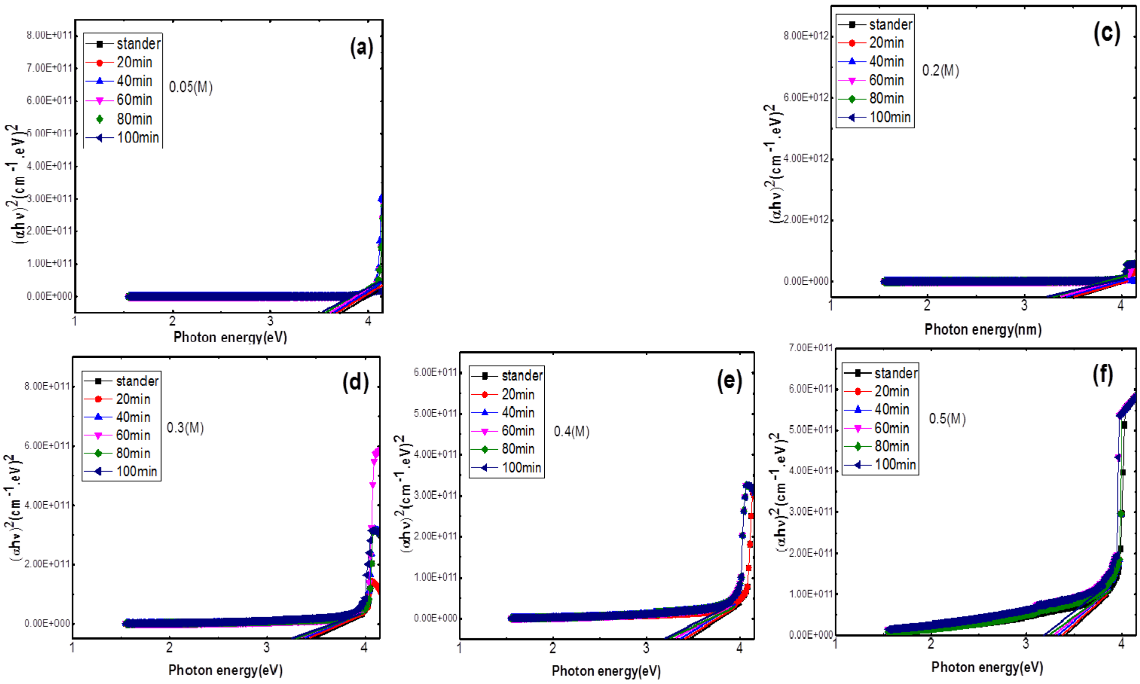

Figure 4. The tauc plot $(\alpha h\nu)^2$ corresponds to the energy gap $(h\nu)$ of the ZnO film for different irradiation concentrations and times.

commute and passive indirect transition occurs at time zero, without irradiation, and that the energy gaps start to narrow with increasing duration of irradiation with alpha particles. As shown in the figure, the best irradiation time is 100 min, during which we obtained the best and lowest direct and indirect transport energy gaps.

Figure 6 shows SEM images of the films showing zinc. The images show a fine structure of nanoparticles distributed on the surface in the form of nanotubes with good adhesion, which is consistent with researcher Y. J. Xing [20]. We notice from the figures that when the ZnO layer is exposed to radiation, these nanotubes are destroyed, and their destruction increases the more time the membrane is exposed to radiation. This is the effect of alpha rays on the membrane. Because zinc oxide materials are characterized by reconfiguration, the molecules of the material reassemble, and as a result, the energy gap decreases, and this is what researcher B. Abdullah agrees with [11].

Figure 7 shows FTIR spectra of pure ZnO nanoparticles irradiated with alpha particles at different times. The spectra demonstrate that the Zn-O bond exists appears at approximately (450 – 490) cm^{-1}. The band at around 490 cm^{-1} may be connected to oxygen deficiency or anoxia of ZnO. This Hypoxia should result in increased green emission in the UV-vis spectrum of absorption. The highest points at 1350 cm^{-1} correspond to the adsorption of CO to the surface made of ZnO. The zone of low absorption at 2367 cm^{-1} indicates carbonates Which is most likely due to ambient carbon dioxide during synthesis. The wide peaks of about 3429 cm^{-1} are ascribed to the mode of O-H stretching of the hydroxyl group and 1634 cm^{-1} (bending) to the asymmetric the zinc carboxylate is stretched Table 2 [21]

Table 1. Displays the direct and indirect energy gaps for zinc oxide films at various concentrations and irradiation periods.

The irradiation time	0.05 M		0.1 M		0.2 M		0.3 M		0.4 M		0.5 M	
	dir. (eV)	Indir. (eV)	dir. (eV)	Indir. (eV)	dir. (eV)	Indir. (eV)	dir. (eV)	Indir. (eV)	dir. (eV)	Indir. (eV)	dir. (eV)	Indir. (eV)
Stander	3.48	3.74	3.02	3.63	3.25	3.58	3.20	3.49	3.01	3.47	2.65	3.42
20 min	3.39	3.69	2.96	3.54	3.18	3.52	3.15	3.45	2.91	3.41	2.61	3.37
40 min	3.33	3.66	2.89	3.51	3.08	3.47	3.06	3.40	2.83	3.37	2.55	3.34
60 min	3.28	3.62	2.80	3.49	2.99	3.39	3.01	3.36	2.78	3.31	2.47	3.30
80 min	3.21	3.58	2.70	3.42	2.90	3.32	2.90	3.30	2.75	3.25	2.40	3.25
100 min	3.12	3.52	2.61	3.37	2.84	3.26	2.84	3.25	2.69	3.20	2.32	3.20

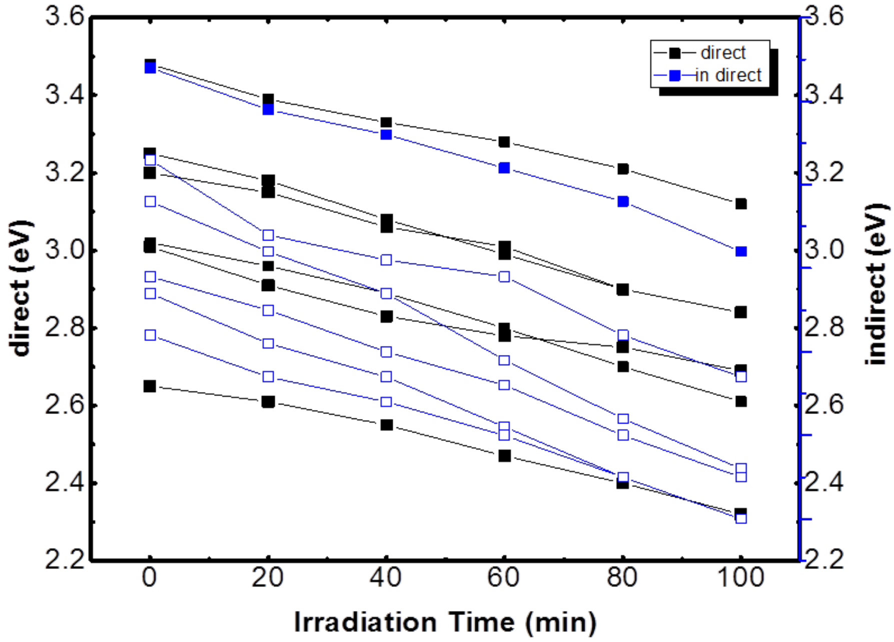

Figure 5. The direct and indirect band gaps as a function of exposure time.

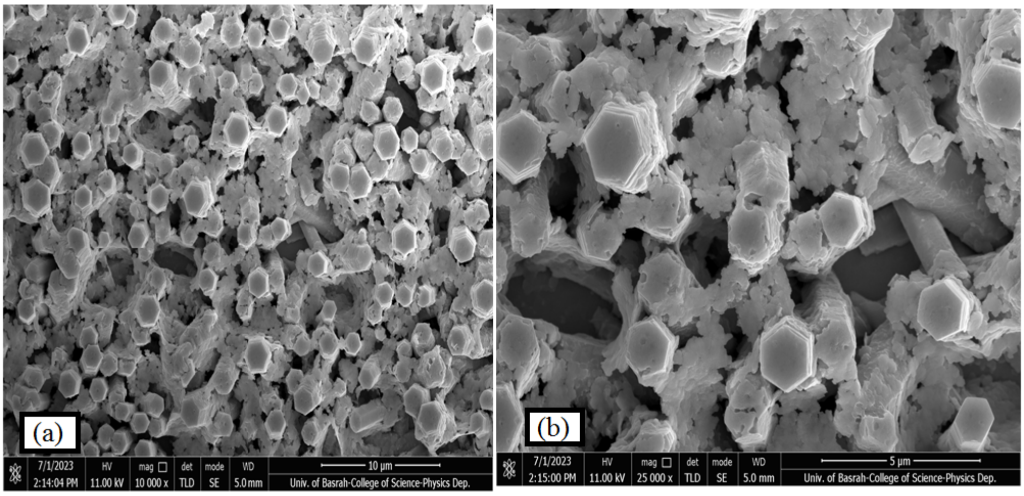

Figure 6. SEM image of ZnO nanotubes; (a) Shows images of zinc oxide films irradiated for 20 min, (b) irradiated for 40 min.

Table 2. The bond type for each concentration is shown in this table.

Band Type	0.05 M	0.1 M	0.2 M	0.3 M	0.4 M	0.5 M
Zn-O	479.13	479.13	458.61	448.35	439.12	448.35
C-C	680.23	659.71	629.95	659.71	640.21	659.45
C–O–C	1011.62	1072.39	1011.62	1021.88	1002.39	-
stretching	1122.43	1082.42	112.43	1061.90	1021.88	-
C=C aromatic	1505.12	1565.66	1505.12	1515.38	1515.38	1534.88
	1676.47	1695.96	1685.70	1695.96	1606.22	1606.22
C=N	2340.28	2350.54	2350.54	2359.71	2350.54	2350.54
O-H	3557.11	3577.63	3597.12	3557.11	3587.89	3597.12

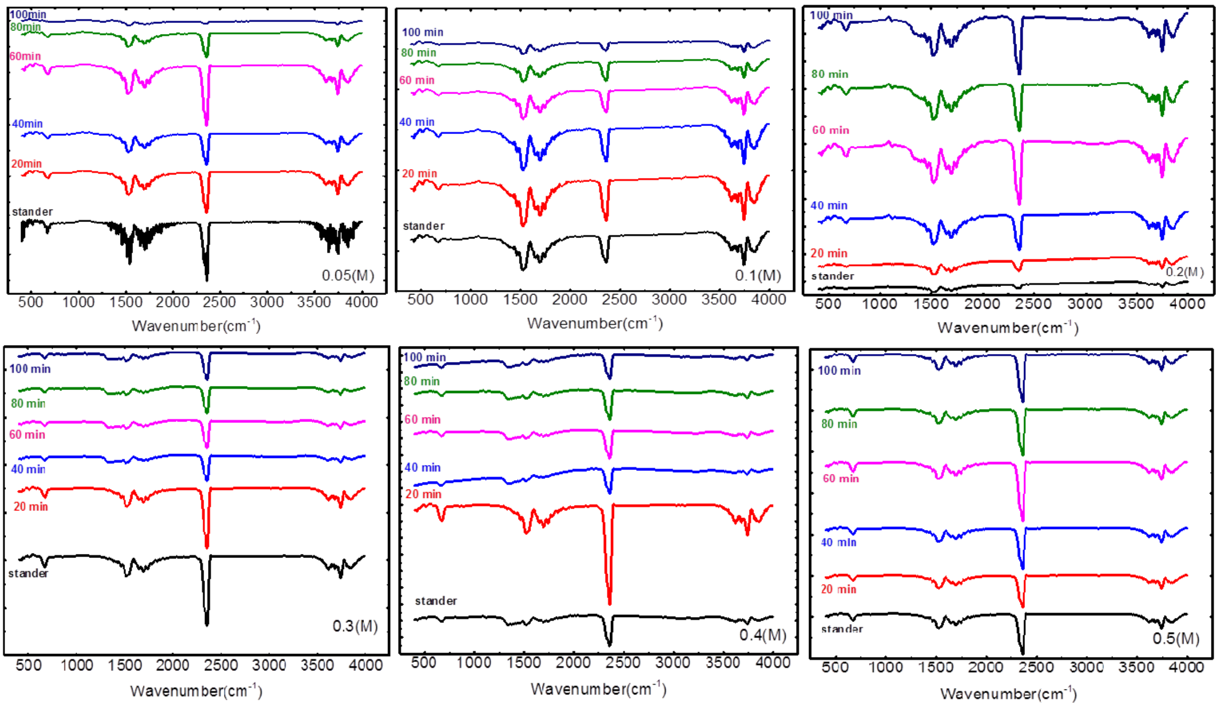

Figure 7. FTIR spectra of zinc oxide (ZnO) film for different concentrations and different irradiation times.

shows this.

3. Conclusion

The optical properties of ZnO thin films generated using a CBD chemical bath were examined through alpha particle irradiation. We used the UV-vis absorption spectra to determine the direct transmission optical band gap and the indirect band gap of ZnO films deposited on glass. The largest disparity in energy levels was seen during the unirradiation period, with concentrations ranging from 0.05 M to 0.5 M. The energy gaps are 3.48 to 2.65 eV for the direct transition, and 3.74 to 3.42 eV for the indirect transition. The indicated concentrations caused a reduction in the energy gap after an irradiation time of 100 minutes. The study revealed that the band gap, both direct and indirect, decreases as the duration of exposure to alpha particles of varying concentrations increases. Notably, the lowest values for the direct energy gap 2.65 eV and the indirect energy gap 3.42 eV were observed at a concentration of 0.5 M. These values are considered to be among the most favorable compared to the other concentrations tested. The maximum duration of irradiation was 100 minutes. The FTIR spectrum displays a distinct peak at 490 cm^{-1}, which corresponds to the typical feature of ZnO. As the duration of exposure to alpha particles grows, there is a notable rise in intensity and a modest displacement in the peak location at 2500 cm^{-1}. According to the provided information, the CBD method has the potential to be used for growing ZnO nanotubes, which can be applied to nano photodetectors.

Authors Contributions
Authors have contributed equally in preparing and writing the manuscript.

Availability of data and materials
Data presented in the manuscript are available via request.

Conflict of Interests
The author declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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