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Effect of Irradiation on the Electronic Transitions of CuO Thin Films Prepared by SPD Technique

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Abstract
Investigates this study the effect of gamma irradiation (E= 662 KeV from a Cs^{137} source) on the optical properties specifically the absorption coefficient and band gap of Copper Oxide (CuO) thin films prepared by spray pyrolysis deposition (SPD) technique on glass substrates at (400°C) with a thickness of (300nm). Irradiation durations of (18 and 24) days were applied. XRD analysis the dominant diffraction peaks correspond to characteristic Copper Oxide (CuO) reflections, indicating preferential growth orientation consistent with monoclinic Copper Oxide (CuO), confirms that the CuO films are polycrystalline and crystallize in the monoclinic C2/c phase.The films exhibit both direct and indirect allowed electronic transitions. The direct optical band gap ranges from (2.4–2.7) electron Volt, while the indirect band gap ranges from (1.8–2.1) electron Volt for non-irradiated and irradiated samples.Gamma irradiation results in a measurable decrease in the absorption coefficient and an apparent increase in both direct and indirect optical band gaps, indicating irradiation-induced modification of electronic states.
Keywords: Thin Films, Copper Oxide (CuO), Electronic Transitions, Spray Pyrolysis Deposition (SPD), Irradiation by Gamma Ray.

1. Introduction

CuO is an important transition metal oxide and semiconductor that plays a structural role in cuprate high-temperature superconductors. It is a chemical compound of copper (also known as cupric oxide). Insoluble in water or alkalis, (CuO) material is dark brown in color, odorless, and can be obtained by the oxidation of copper [1]. CuO simplicity of preparation, abundant in nature, low cost and nontoxic, this material is considered today as one of the most used in photovoltaic module-technologies for the manufacture of solar cells [2]. CuO material has been studied because of its good properties in applications, including super capacitors, magnetic storage, photo detectors, and superconductors, photo catalysis [3]. CuO is unique as it has a square planar coordination of copper by oxygen in the monoclinic structure, the lattice parameters of CuO are a = 4.684 Å, b = 3.425 Å, c = 5.129 Å and b = 99.28° [4]. The copper oxide family includes two stable oxides: cuprous oxide (Cu_2O) and cupric oxide (CuO) [5]. CuO is an intrinsic p-type semiconductor with a monoclinic crystal structure. It has good conductivity, a good range of stability, and high light absorption in the visible region. It has many applications in various fields, so it is used in solar energy applications including photovoltaic cells and thermoelectric sensors [6].

There are several techniques have been employed for depositing CuO thin films, each offering unique advantages and suitability for specific applications, thermal evaporation [7], oxidation [8], molecular beam epitaxy [9], and electro deposition [10], pulsed laser deposition [11].

The Optical Energy gap value depends mainly on the prepared method, the energy band gap value of thin oxide typically ranges from (1 to 1.9) eV, and A band gap near 1.5 eV is

theoretically favorable for photovoltaic conversion under ideal Shockley–Queisser conditions, although practical efficiencies depend on material quality and device architecture [12].
This work aims to evaluate how gamma irradiation influences the electronic transitions and optical properties of CuO thin films prepared by spray pyrolysis (SPD) technique.

2. Materials and Methods

Spray pyrolysis deposition technique was chosen due ability to produce high-purity copper oxide (CuO) thin films with high cohesion at varying substrate temperatures, thus offering good properties for use in solar cell and detector applications [13].
Copper oxide [Cu $(NO_3)_2.3H_2O$] with a molecular weight of (241.60gm/mol), a blue powder supplied by the British company (BHD) with a purity of (98%), was used to prepare the solution used to produce the copper oxide (CuO) film. The solution was prepared at a molar concentration of (0.1 mol/L) by adding (2.146g) of it to (100 ml) of distilled water and gradually dissolving it using a magnetic stirrer for (15-20 minutes). The molar concentration was calculated using the **Equation (1)** [14].

$$M = \left(\frac{Wt}{M_{Wt}}\right)\left(\frac{1000}{V}\right) \quad \textbf{(1)}$$

Where:
M: Molar concentration.
W_t: Weight to be dissolved.
M_{Wt}: Molecular weight of the substance.
V: Volume of distilled water in which solution was dissolved.
After dissolving, the solution is left for an appropriate period of time to ensure that no sediment remains. It is then placed in the sprayer tank. After the spraying process is complete, the gases evaporate due to heat, leaving a copper oxide film on the glass substrate.
Study the effect of irradiation by gamma rays with energy (E = 662 keV) on copper oxide films the emitted from a radioactive the cesium (CS^{137}) source for an irradiation (18, 24) days. The optical interference method using a helium-neon (He-Ne) laser used to calculated thickness of the films. The thickness (t) was calculated by the interference of the laser light resulting from the phase difference between the reflected rays from the back and front surfaces of the thin film, using the following **Equation (2)**: [15]

$$t = \frac{\Delta L}{L} \times \frac{\lambda}{2} \quad \textbf{(2)}$$

Whereas:
λ: is the laser wavelength.
L: is fringe spacing.
ΔL: is fringe shift.
The **Figure 1** Show the diagram of workflow and measurements used in the preparation and analysis of copper oxide (CuO) thin films.
The absorption coefficient is calculated according to the following **Equation (3)** [16]:

$$\alpha = 2.303\ \frac{A}{t} \quad \textbf{(3)}$$

Whereas:
A: Absorbance spectrum.
Optical energy gap:
A. The direct Energy gap can be calculated using the following **Equation (4)** [17]:

$$\alpha h\nu = A\,(h\nu - E_g)^r \quad \textbf{(4)}$$

Whereas:
$\alpha h\nu$: Photon energy.
A: Constant.
r: is an exponential coefficient that depends on the type of transition.

Equation (4) can be rewritten by substituting the value of the constant (r =1/2) in the following form: [18]

$$(\alpha h\nu)^2 = A\left(h\nu - E_g\right) \quad \textbf{(5)}$$

By drawing the relationship between $(\alpha h\nu)^2$ and photon energy $(h\nu)$, extending the straight part of the curve to intersect photon energy axis $(h\nu)$ at $(\alpha h\nu)^2 = 0$ determining value direct optical Energy gap.

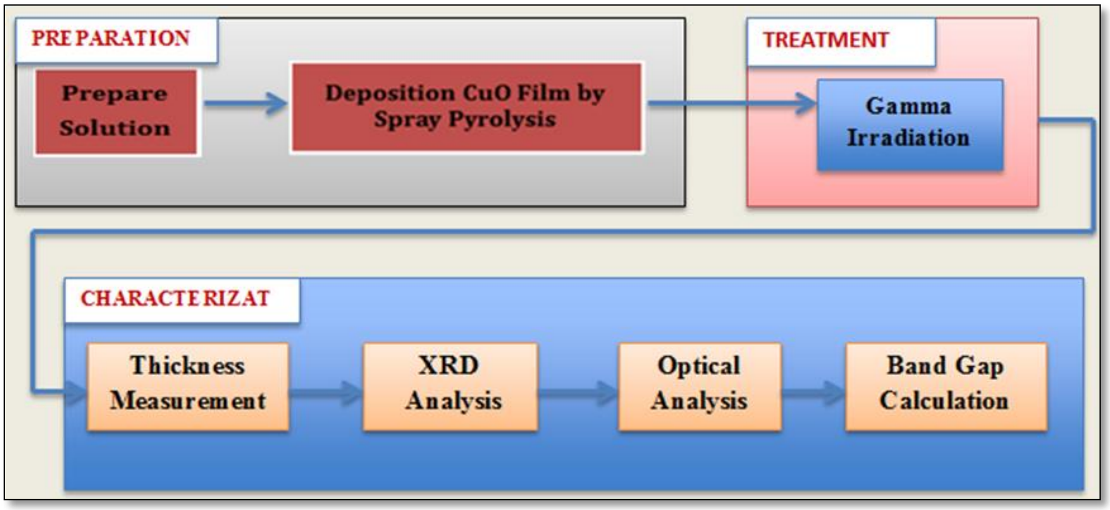

Figure 1. diagram the workflow and measurements used in the preparation and analysis of copper oxide (CuO) thin films.

B. The indirect optical energy gap can be calculated using the following **Equation (6)**: [19]

$$\alpha h\nu = A\left(h\nu - E_{g} \mp E_P\right)^r \quad \textbf{(6)}$$

Whereas:

$+E_P$: Phonon absorption process,

$-E_P$: phonon emission process,

Equation (6) can be rewritten by substituting the value of the constant (r =2) in the following form: [20]

$$(\alpha h\nu)^{1/2} = A\left(E - E_{g} \mp E_P\right) \quad \textbf{(7)}$$

By drawing the relationship between $(\alpha h\nu)^2$ and photon energy $(h\nu)$ we notice the presence of two straight parts, as follows:

The first part: has a supply interruption $(h\nu)$ at low energy values and represents the transitions that are accompanied by phonon absorption.

When: $(\alpha h\nu)^{1/2} = 0$

When the **Equation (7)** leads to:

$$h\nu = E_g - E_P \quad \textbf{(8)}$$

The Second part: has a supply interruption $(h\nu)$ at high energy values and represents the transitions that are accompanied by phonon emission.

When: $(\alpha h\nu)^{1/2} = 0$

When the **Equation (7)** leads to:

$$h\nu = E_g + E_P \quad \textbf{(9)}$$

This means that phonon emission from the lattice requires minimum photon energy of $(h\nu)$. The forbidden optical energy gap for the permitted indirect transitions $\left(E_g\right)$ can be calculated from the difference between the two intercept values.

3. Results

The XRD analysis in **Figure 2** shows confirms that the CuO thin films are polycrystalline and crystallize in the monoclinic C2/c phase; consistent with standard diffraction data (JCPDS card No. 00-045-0937). **Figure 2** also the dominant diffraction peaks correspond to characteristic CuO reflections, indicating preferential growth orientation consistent with monoclinic CuO.

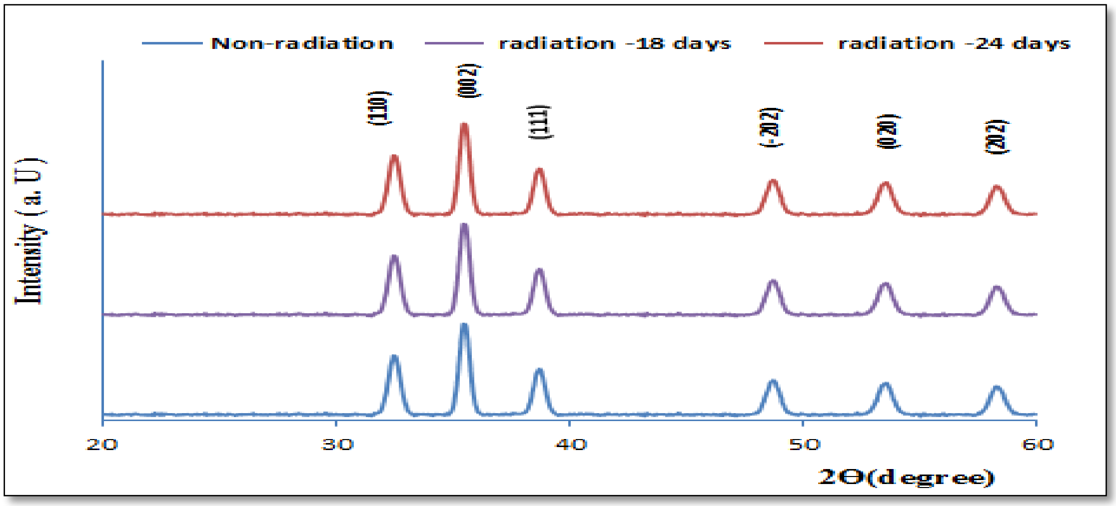

Figure 2. The x-ray diffraction patterns of copper oxide thin films of Non irradiation and irradiation (18, 24) days at temperature (400˚C) and thickness (300 nm).

The optical properties include calculating both the absorption coefficient and the optical energy gap of non-irradiated and irradiated by gamma ray of copper oxide (CuO) thin films for (18, 24) days with thickness (300 nm).

3.1. Absorption Coefficient (α):

Figure 3 shows that all absorption coefficient curves for copper oxide (CuO) thin films Non irradiated and irradiation (18, 24) days with thickness (300nm), with behave similarly, we note an increase value absorption coefficient value as photon energy increases within the range of (1.4 -4.2) eV. **Figure 3** also shows decrease absorption coefficient value for copper oxide (CuO) thin films with increases period irradiation value, Also **Figure 3** can see from the shift's absorption toward higher photon energies, indicating a slight increase of energy gap, as shown in **Tables 1** and **2.**

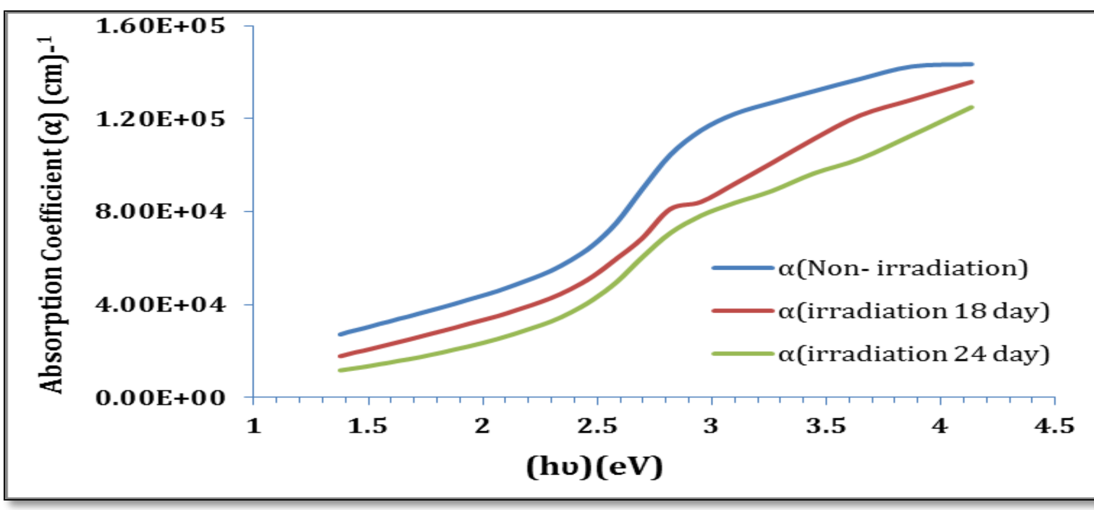

Figure 3. Absorption coefficient (α) as a function of photon energy for (CuO) thin films of Non irradiation and irradiation (18, 24) days with thickness (300 nm).

3.2.Optical energy gap (Eg):

Figure 4 and **Table 1** show that increasing the irradiation time results increase in the forbidden direct optical energy gap value.

Table 1. Direct optical energy gap of (CuO) thin films of Non irradiation and irradiation (18, 24) days.

Energy gap (eV)		
Non- irradiation	**Irradiation (18day)**	**Irradiation (24day)**
2.4 eV	2.6 eV	2.7 eV

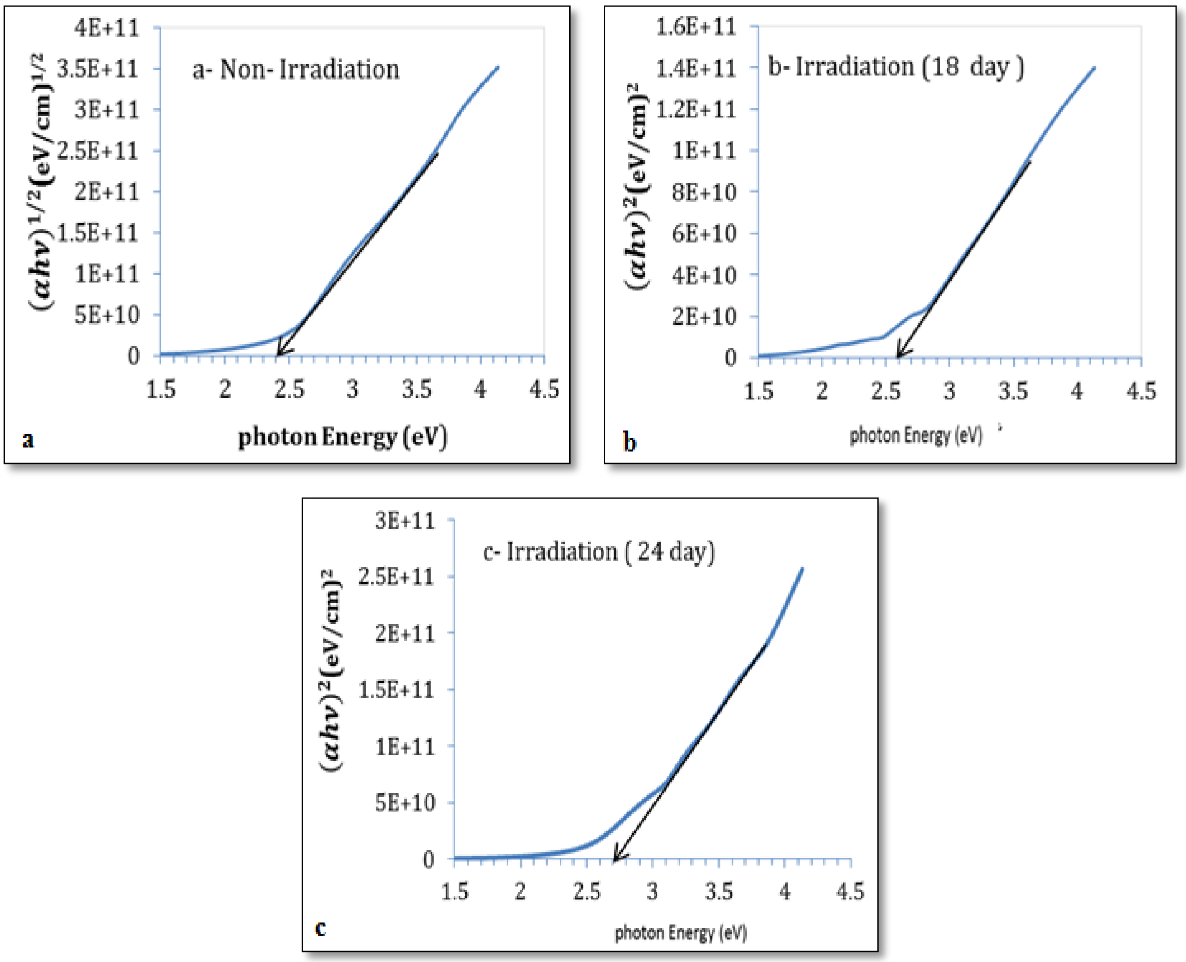

Figure 4. The relationship between the changes in $(\alpha h\nu)^2$ as a function of photon energy for (CuO) thin films of (a-non-irradiation, b- irradiation18days, c- irradiation24days).

Figure 5 show the indirect optical energy gap values for all (CuO) thin films after irradiation for (18, 24) days were greater than their values before irradiation. We also observed an increase in the optical energy gap value with increasing irradiation time, as shown in **Table 2.**

It can also be noted from the **Figure 5** that the behavior of the non-irradiated curves was similar to their behavior before irradiation (18, 24) days.

Table 2. Indirect optical energy gap of (CuO) thin films of Non irradiation and irradiation (18, 24) days with thickness (300nm)

Energy gap (eV)		
Non- irradiation	**Irradiation (18day)**	**Irradiation (24day)**
1.8 eV	1.9 eV	2.1 eV

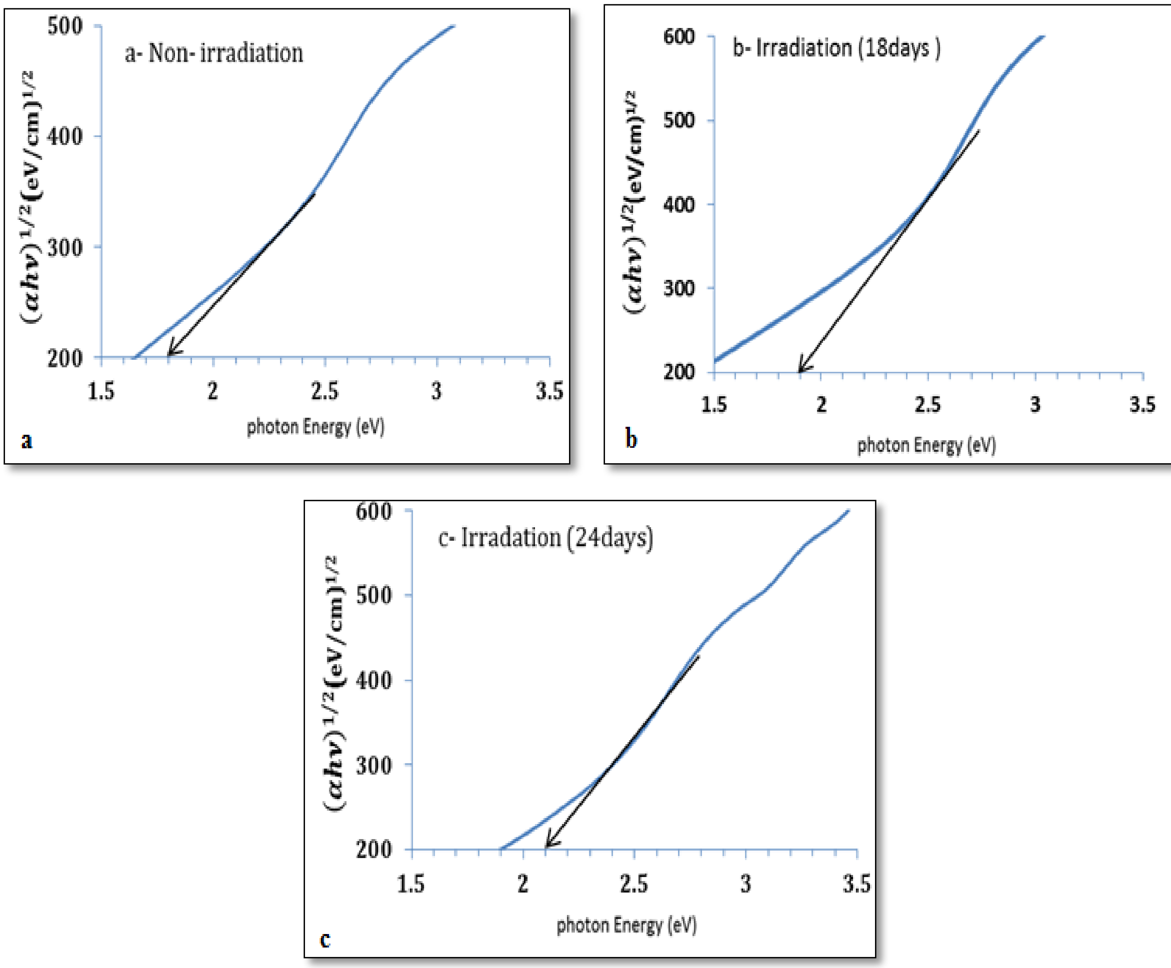

Figure 5. The relationship between the changes in $(\alpha h\nu)^{1/2}$ as a function of photon energy for (CuO) thin films of (a- non-irradiation, b- irradiation18days, c- irradiation24days).

4.Discussion

Structural investigations of copper oxide thin films using X-ray diffraction are crucial for understanding the relationship between the optical properties and structural changes for thin films, as each material has its own crystal structure. The XRD analysis in **Figure 2** shows confirms that the CuO thin films are polycrystalline and crystallize in the monoclinic C2/c phase; consistent with standard diffraction data (JCPDS card No. 00-045-0937). **Figure 2** also the dominant diffraction peaks correspond to characteristic CuO reflections, indicating preferential growth orientation consistent with monoclinic CuO[21].

Optical properties of (CuO) thin films are of great importance in evaluating these films and determining their potential applications. The optical properties include calculating both the absorption coefficient and the optical energy gap of non-irradiated and irradiated by gamma ray of copper oxide (CuO) thin films for (18, 24) days with thickness (300 nm).

Absorption Coefficient (α): Electronic transitions depend on absorption that occurs based on the energy band of the semiconductor material between (valence band and conduction band). High absorption coefficient values ($\alpha > 10^4$ cm^{-1}) indicate possibility of a direct electronic transition, while low absorption coefficient values ($\alpha < 10^4$cm^{-1}) indicates the occurrence of an indirect electronic transition [22]. **Figure 3** shows that all absorption coefficient curves for copper oxide (CuO) thin films Non irradiated and irradiation (18, 24) days with thickness (300nm), with behave similarly, we note an increase value absorption coefficient value as photon energy increases within the range of (1.4-4.2) eV. **Figure 3** also shows decrease absorption coefficient value for copper oxide (CuO) thin films with increases period irradiation value, the reduction in absorption coefficient after irradiation may result from radiation-induced modification of defect states and carrier scattering mechanisms. A shift of the absorption edge toward higher photon energies corresponds to band gap widening, while a shift toward low energies indicates band gap narrowing[23]. Also **Figure 3** can see from the shift's absorption toward higher photon energies, indicating a slight increase of energy gap, as shown in **Tables 1 and 2**. Absorption edge is not sharp, but rather curved; indicating that the films prepared in this way have non-sharp absorption edge reflects band tailing caused by localized defect states and structural disorder typical of polycrystalline film [24].

Optical energy gap defined is the minimum energy required for an electron to move from top of valence band to bottom of conduction band [25]. The current study revealed that two types of electron transitions occur: direct and indirect, and value of the optical energy gap was calculated based on this.

Direct optical energy gap**: Figure 4** shows that increasing the irradiation time results increase in the forbidden direct optical energy gap value. This may be attributed to the fact that the irradiation the apparent increase in band gap may result from redistribution of localized states or band tail modification induced by gamma irradiation[26, 27], as shown in **Table 1**. It can also be noted from the **Figure 4** that the spectral behavior of the irradiated films remains qualitatively similar to the non-irradiated samples, with differences primarily in band edge position.

Indirect optical energy gap**: Figure 5** illustrates the possibility of determining the forbidden optical energy gap and the phonon energy associated with this process from the best tangents to the straight lines in the curve. We found the optical energy gap values for all (CuO) thin films under study after irradiation for (18, 24) days were greater than their values before irradiation. We also observed an increase in the optical energy gap value with increasing irradiation time, as shown in **Table 2**. It can also be noted from the **Figure 5** that the behavior of the non-irradiated curves was similar to their behavior before irradiation (18, 24) days.

5. Conclusion

The x-ray diffraction CuO thin films exhibit polycrystalline monoclinic structure. The optical properties all copper oxide (CuO) thin films possess both direct and indirect allowed electronic transitions. Gamma ray irradiation resulted in a decrease in the absorption coefficient for all copper oxide (CuO) thin films. Gamma irradiation modifies the absorption behavior and results in an apparent increase in extracted optical band gaps.

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Conflict of Interest

The authors declare that they have no conflicts of interest

Funding

None

Ethical Clearance

There were no human or animal subjects in the study, and no personal information was gathered in any way. Thus, informed consent and ethical clearance were not necessary.

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