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Optical Properties of Indium-Doped Tin Dioxide Thin Films Irradiated by Alpha Particles

This study investigates the effect of α-particle irradiation (5.48 MeV) on the optical properties of pure and indium-doped (1.5 and 3 at.%) SnO_2 thin films prepared by the spin-coating technique. Results of the UV–visible spectroscopy in the range 300–1100 nm show that increasing the indium content reduces the optical transmittance and narrows the direct band gap. The initial band gaps were 3.84 eV (pure), 3.80 eV (1.5% In), and 3.74 eV (3% In). After irradiation, the band gaps decreased slightly to 3.82 eV, 3.78 eV, and 3.73 eV, respectively. This reduction is attributed to radiation-induced defects that introduce localized states near the band edges.

Keyword: Tin dioxide; Thin films; Alpha radiation; Optical properties; Spin coating
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1. Introduction

Thin film physics is one of the important branches of solid-state physics, which deals with very small thickness systems that consist of tens of nanometers and a few micrometers, and are deposited on the solid materials known as "substrates, like glass, silicon, and some salts of minerals and polymers [1-2]. Thin films are used to replace many of the electronic circuitry with the same properties and more efficiently, such as rectifiers, capacitors, transistors, and digital computing. As a result of the wide and huge use in industrial applications and research of thin films and the remarked importance of electrically conductive and photoconductivity transparent films, thin films of SnO_2 became very interesting. These films have a direct energy gap, also, it is one of the transparent conductive films and it is (n-type), this is mean that the electrons are the main carriers of charges [3].

In 1942, McMasters prepared the film of SnO_2 for the first time, and it has been extensively studied. This film has many different applications, which allow it to be used as a Windows-connected photovoltaic technology; also, it has several applications that are used in electro-optical devices and direct energy conversion devices (conductive-insulator-conductive) in solar panels [4,5]. SnO_2 is a sensitive material for various gases, which has led to much extensive research in the field of gas sensors [6]. Recently, it has been used in the design and production of ultraviolet light-emitting devices and laser diodes [7].

Alpha particles are nuclear radiation that is characterized by its ability to ionize or irritate the atoms of the material that are falls on it. When alpha particles (helium atom nucleus) pass through a medium, they mainly interact with the electrons of the medium due to the Coulomb energy, which affects the interaction between the charged particle and electrons. Thus, the dominant mechanism for the massive loss of alpha particles is Coulomb's scattering [8]. The radiation effect in the films represents the linear energy flow rate (linear energy transfer), which is directly Proportional to the square of the charge and inversely proportional to the velocity square [9]. The films easily absorb alpha particles, and alpha particles can lose all of their energy in these films. When alpha particles pass through a certain material, they can obtain two electrons from it and turn into neutral helium atoms. Alpha particles can obtain two electrons from the material that they pass through and turn into neutral helium atoms.

2. Experimental Part

In this work, tin(II) chloride dihydrate ($SnCl_2{\cdot}2H_2O$) was used as the main source for preparing SnO_2 thin films. It is a white powder that dissolves easily in water, with a molecular weight of 225.6456 g/mol and a purity of 99.5%. The spin coating technique prepare the films. Indium chloride ($InCl_3$) was added as a dopant material with weight ratios of 2.5%, 5.5%, 7.5%, and 15%. The amount of hydrated tin chloride was determined by weight without any change in the titer. The solution was prepared with a molar concentration of 0.5 mol/L by dissolving 4.58 g of $SnCl_2{\cdot}2H_2O$ in 30 mL of methanol, according to the standard calculation formula [10]

$$M = \frac{W_t}{M_{Wt}} \times \frac{1000}{V} \qquad (1)$$

where M is the molar concentration (mol/L), W_t is the weight required to be dissolved (g), M_{wt} is the molecular weight of the material (g/mol), and V is the volume of distilled water in which the solution is solved (mL)

The precursor materials and doping ratios were precisely weighed using a sensitive electronic balance (Metler HK-160) with an accuracy of 10^{-4} g. Solutions for pure SnO_2, 1.5% In-doped SnO_2, and 3% In-doped SnO_2 films were all based on a 0.5 M molarity. The specific quantities of all chemicals used are listed in table (1). The mixture was stirred magnetically for 60 minutes until a colorless, homogeneous solution was obtained. The doping solution was prepared by adding the tin chloride solution to 0.1–0.2 g of indium chloride dissolved in 30 mL of methanol, according to the required doping ratio.

Table (1) Weight ratios and precursor concentrations for the preparation of pure and In-doped SnO_2 thin films

Molarity (M)	Weight Ratio (%)	SnO_2 (g)	$InCl_3$ (g)	CH_3CO_2H (g)	CH_3OH (mL)
0.5	0.0	4.5	0	1.2	40
0.5	1.5	4.5	0.10	1.2	40
0.5	3	4.5	0.11	1.2	40

During spin coating, a speed of 3000 rpm was used for 30 s to deposit uniform thin films. The films were subsequently dried at 100 °C for 10 minutes, followed by 150 °C for 20 s. Film thickness was measured using an optical microscope and the Pro-Axel software. The sample edges were examined under magnification, and the film thickness was recorded in pixels. This measurement was repeated three times at different positions, and the average value was converted to nanometers using the relation: 1 µm = (pixel) / 50.205 (where 1 µm = 1000 nm). This process confirmed that the deposited films had a consistent thickness of approximately 0.5 µm. Post-deposition, the films were annealed for 60 minutes to enhance crystallinity. Annealing was tested at 355 °C, 455 °C, 555 °C, and 655 °C. The most uniform and stable films were obtained at 500 °C, which was identified as the optimal temperature. Glass substrates were carefully selected and cleaned before coating to ensure strong adhesion and a smooth surface.

The alpha particle source was Americium-241 (^{241}Am), with a half-life of 432.6 years and a radioactivity of 330 kBq. During irradiation, the source was positioned 0.29 cm from the sample surface, delivering particles with an energy of 5.37 MeV. The samples were irradiated for 27 minutes, receiving a total dose of 5 Gy. All irradiation experiments were conducted in air at room temperature under controlled conditions in the Nuclear Laboratory, Department of Physics, College of Science, University of Kirkuk. To ensure result reliability, four identical samples were prepared and irradiated for each film type: pure SnO_2, 1.5% In-doped SnO_2, and 3% In-doped SnO_2.

3. Results and Discussion

In this study, the optical properties of the SnO_2 film were investigated before and after irradiation, as discussed in the next paragraph.

Transmittance stands for the ratio between the passing light intensity (I_T) to the falling light intensity (I_0) on the sample [11]

$$T = \frac{I_T}{I_0} \qquad (2)$$

The values of the transmittance of the prepared films in the wavelength range of 300-1100 nm were calculated. Figures (1) and (2) show that the SnO_2 film transmittance decreases with the increase of doping. The transmittance curve for all films (pure and In-doped) shows similar behavior, which shows a sharp increase in the wavelength range of 300-350 nm and then tends to stabilize for a certain range of wavelength 400-1100 nm. The film is used as an optical window for solar cells. The increase of the doping ratio leads to the creation of inter-planar levels under the conduction band, and these levels are ready to receive the electrons and generate tails in the optical energy gap. These tails tend to reduce the energy gap. It is one of the crystalline defects, and this finding agrees with the study results of [12].

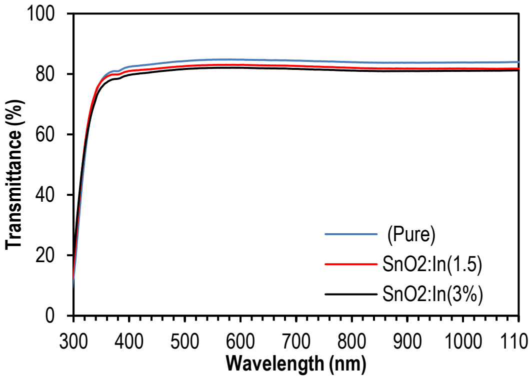


Fig. (1) Transmission spectra of SnO_2 films before irradiation

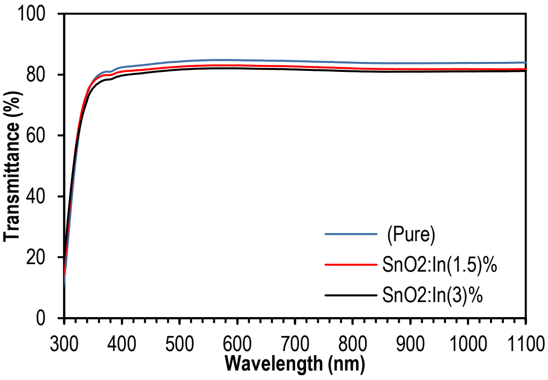


Fig. (2) Transmission spectra of SnO_2 films after irradiation

Whereas, the absorption coefficient (α) for all the films prepared, including both doped and pure films, was determined using the following relation [13]:

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

Figure (3) shows that the absorption coefficient before irradiation as a function of photon energy. On the other hand, the results illustrated that the absorption coefficient increases with the increase of photon energy, as well as increases with indium doping. Figure (4) shows the absorption coefficients of pure and In-doped SnO_2 after irradiation with the same doping ratio. In fact, the alpha radiation caused a change in the absorption coefficient, which resulted in reduction in its value, and this confirms that there is a relationship between the absorption coefficient and the energy gap during direct electronic transmissions and these results are consistent with previous research [14]. Table (2) shows the values of absorption coefficient of the pure and In-doped SnO_2 samples after irradiation with alpha particles.

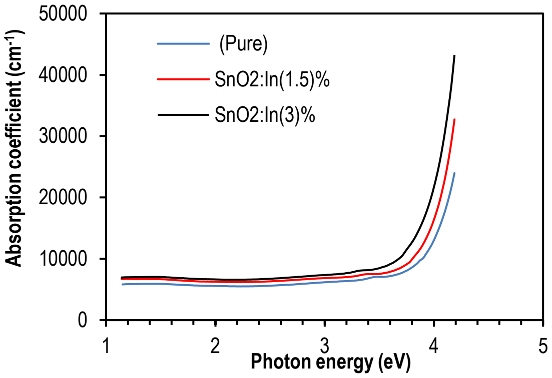


Fig. (3) Absorption coefficients of pure and doped SnO_2 films before irradiation

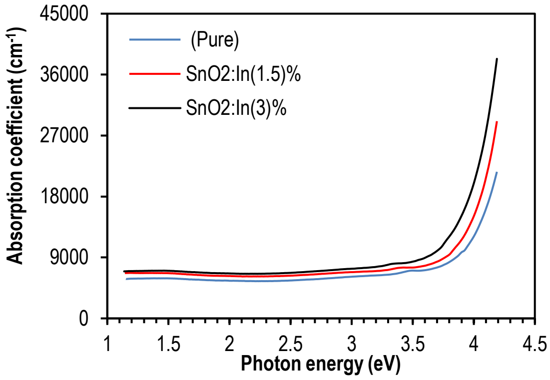


Fig. (4) Absorption coefficients of pure and doping SnO_2 films after irradiation

Table (2) Absorption coefficient of pure and irradiated SnO_2 films with alpha particles

Sample	α x10^4 (cm^{-1})	
	Pure	After doping
SnO_2:In (0.0%)	2.2	2
SnO_2:In (1.5%)	3	2.97
SnO_2:In (3%)	4.2	3.81

The optical direct band gaps of pure and In-doped SnO_2 films prepared by the spin coating technique were calculated by the Tauc relation is represented as [15]:

$$\alpha h\upsilon = B_o (h\upsilon - E_g^{opt})^r \quad (4)$$

where r is the exponential coefficient that determines the type of transition, B_o is a constant that depends on the material nature, E_g^{opt} represents the optical band gap in (eV), and $h\upsilon$ represents the photon energy (eV)

By extrapolating the straight line of the relationship between $(\alpha h\upsilon)^2$ and photon energy $(h\upsilon)$ to cross the photon energy axis at the point $(\alpha h\upsilon)^2 = 0$, the optical energy gap (E_g^{opt}) is determined for the allowed direct transitions, as shown in Fig. (5).

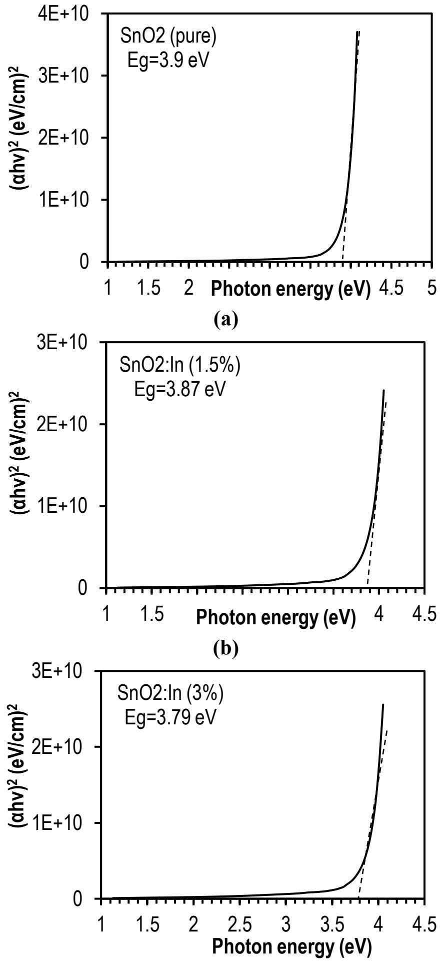
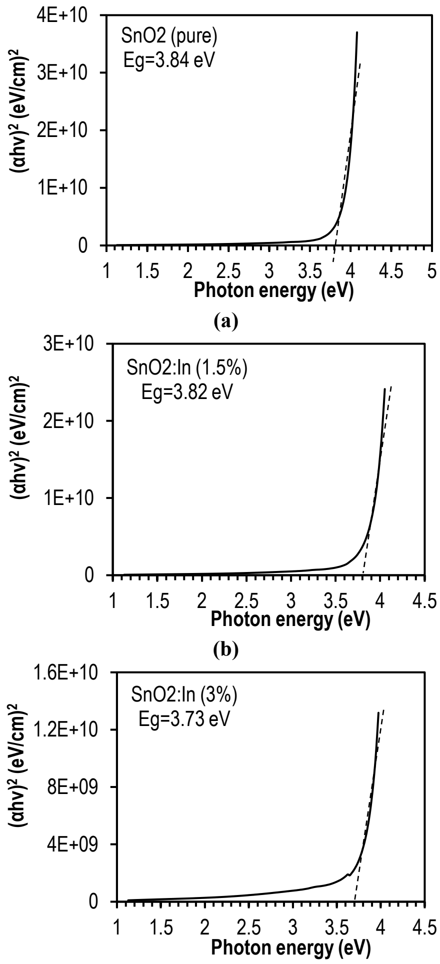


Fig. (4) Determination of optical allowed direct band gap of pure and In-doped SnO_2 films before irradiation

There is a decrease in the value of E_g^{opt} of SnO_2 film due to doping with indium. This confirms that alpha particles have an effect on the energy gap value, and the increase of the doping with indium leads to the appearance of new inter-band levels within the conduction band, which are ready to receive electrons, and produce tails in the optical energy gap; these tails tend to reduce the energy gap. This finding is consistent with previous reports [16-19]. After irradiation, the optical energy gap (E_g^{opt}) for the allowed direct transition was decreased. This confirms the effect of alpha particles on the optical energy gap, as they have generated additional inter-band energy levels within the energy gap as shown in Fig. (6) and table (3).

Table (3) Initial band gap values and band gap values after irradiation for all three types of films

Sample	Energy band gap Before irradiation	Energy band gap After irradiation
SnO_2:In (0.0%)	4.00	3.94
SnO_2:In (1.5%)	3.84	3.82
SnO_2:In (3.0%)	3.84	3.73



(a)

(b)

(c)

Fig. (5) Determination of optical allowed direct band gap of pure and In-doped SnO_2 films after irradiation

4. Conclusions

This work demonstrates that indium doping and alpha-particle irradiation significantly modify the optical properties of SnO_2 thin films. Increasing the indium concentration (up to 3 at.%) systematically reduces the optical band gap and transmittance, due to defect states from In^{3+} substitution and oxygen vacancies. Alpha irradiation further slightly narrows the band gap, indicating the creation of radiation-induced localized states. The 3% In-doped films showed the most pronounced response, highlighting their potential for use in radiation-sensing devices, gas sensors, and solar cell applications where controlled tuning of optical properties is essential.

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