Optical Characteristics of Tin Oxide Thin Films Doped with Indium and Aluminum Using the Sol-Gel Spin Coating Technique

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ABSTRACT
The aims of this research are to understand the optical characteristics of SnO₂ doped with Indium and Aluminum (SnO₂:In+Al) using the sol-gel spin coating technique. Optical characteristics of SnO₂:In+Al thin films were measured by UV-Vis Spectrophotometer. The optical characterization results showed that the SnO₂:In+Al thin films had an increase in transmittance from (68.6 – 78.3)% at a wavelength of 300 – 470 nm and an increase in absorbance at a wavelength of 295 nm from 4.34 – 5.00 with the increase in the percentage of doping. This shows the thin layer absorbs the maximum waves at a wavelength of 295 nm. The increase in the doping percentage causes the energy gap of SnO₂ thin films is decreasing. The direct energy gap decrease from 3.58 to 3.54 eV and the indirect energy gap decrease from 3.90 to 3.87 eV. The energy of optical activation of the SnO₂ thin films decreases from 0.97 to 0.86 eV when increasing the doping percentage. This research has indicated that SnO₂:In+Al thin films has a low energy gap and high transmittance so that it belongs to be high-quality thin films.

Keywords: Aluminum, Indium, Optical Characteristics, Sol-Gel, Spin-Coating, Thin films, Tin oxide.

1. INTRODUCTION
Current technological developments cannot be separated from the support of the existence of semiconductor materials. The use of semiconductor materials is mostly found in electronic devices. Tin oxide (SnO₂) is a semiconductor material that is often used. This SnO₂ semiconductor is included in an N-type semiconductor. The size of semiconductor material can be made from micrometer to nanometer, which is in a thin layer form. A gap of thin layer energy made from SnO₂ is large for a semiconductor, which is 3.60 – 3.98 eV [1, 2]. Modifications made to semiconductor materials can make the semiconductor materials using more diverse.

Thin-film semiconductors are widely used as touch screens, solar cells [3], and sensors of gas [4]. To make this thin layer function better, an additional substance (doping) is needed to decrease the energy gap. Some doping that has been added to SnO2 is Aluminum [5], Fluoride Dehydrate [6], Cesium [7], Aluminum-Zinc [8], Indium [9], and Fluorine [10]. Some research results that add doping to SnO₂ thin films have shown that the conductivity of doped thin films has increased. In other words, the energy gap is decreasing.

Indium is used as doping in SnO₂ because Indium reflexes to temperature [11], has fairly high transparency [12], and the synthesis process can be carried out at low temperatures [13]. Meanwhile, Aluminum is used as doping because Aluminum produces high transparency in the range of visible light [14]. The availability of Al material in nature is very abundant. Besides, Aluminum has a resistance to oxidation (not corrosive), strong and lightweight.

Some techniques in making thin films include RF sputtered [15], pulsed laser deposition [16], reactive...
magnetron sputtering [17], RF plasma enhanced reactive thermal evaporation [18], DC and RF sputtering [19] which are efficient in electric power [20], a spin-coating technique [21], and dip-coating [22]. The spin-coating is the most economical, simplest, and efficient technique [23]. This method can produce thin films with high-quality with a thickness that can be adjusted through playback time and rotational speed [24].

2. EXPERIMENTAL

In principle, the process of synthesizing thin films with the sol-gel spin-coating technique uses centrifugal force to spread the solution used as the base material evenly on the surface of the substrate used. The solution used as a base for making thin films is dripped on a glass substrate and then rotated until the solution is spread evenly on the surface of the glass. The rotating speed used is 2000 rpm. The playback time used is 3 minutes. That speed and playback time have produced a thin layer with a thickness of about 60 nm. This result can be obtained from Figure 1.

![Figure 1](Image)

**Figure 1** Graph of the relationship between film thickness and rotation speed [25].

The basic material used as a thin layer in this study was SnO$_2$:2H$_2$O which was doped with InCl$_3$.4H$_2$O and AlCl$_3$ which was dissolved using C$_2$H$_5$OH. The main ingredient of SnO$_2$ thin film was doped with a mixture of Indium and Aluminum with a percentage of doping mass of 0, 5, 10, 15, 20, and 25%. The optical characteristics of the resulting thin film consist of transmittance and absorbance. Measurement of transmittance and absorbance of thin films using UV-Vis Spectrophotometer in a wavelength range from 200 nm to 800 nm.

The energy gap amount is determined from the value of the absorbance. The energy gap is the lowest energy absorbed by material so that electrons can move to the conduction band from the valence band. The energy gap can be determined from the slope of the graph $(\alpha h\nu)^n$ to the photon energy $(h\nu)$. The energy gap consists of direct optical energy gap $(n = 1/2)$ and indirect optical energy gap $(n = 2)$ [26].

The energy gap can be determined by using the equation

$$\alpha(\nu)h\nu = C(h\nu - E_g)^n$$

Note: $h\nu$ is the initial energy of the photon, $h$ is the Planck’s constant, $E_g$ is the energy gap, and $C$ is constant.

3. RESULT AND DISCUSSION

Figure 2 below shows the thin films SnO$_2$:In+Al with doping percentages of 0, 5, 10, 15, 20, and 25% with the number of layers 1 at a heating temperature of 100 °C.

The optical characteristics obtained from the UV-Vis Spectrophotometer are the absorbance and transmittance of the SnO$_2$ thin film doped with a mixture of Indium and Aluminum.

![Figure 2](Image)

**Figure 2** SnO$_2$:In+Al thin films with Doping (a). 0:100, (b). 5:95, (c). 10:90, (d). 15:85, (e). 20:80, (f). 25:75%.

3.1. Measurement of Transmittance and Absorbance

The results of absorbance and transmittance of SnO$_2$:In+Al thin films with the number of layers 1 at a heating temperature of 100 °C were tested using UV-Vis Spectrophotometer shown in figure 3.

![Figure 3](Image)

Figure 3 (a) shows the transmittance obtained from SnO$_2$:In+Al thin films with doping concentrations of 0, 5, 10, 15, 20, and 25% respectively have an impact in increasing the transmittance of SnO$_2$ thin films. This phenomenon has similar results to previous studies on thin films doped with Indium [27, 28]. Besides, this study also has results similar to previous studies using Aluminum doping [29].
The transmittance increases at a wavelength of 300 – 470 nm. The maximum value of transmittance at 470 nm wavelength from the lowest doping concentration to the highest is 74.5; 68.6; 71.6; 78.3; and 74.8%. This shows that the movement of electrons occurs due to the presence of photon energy at a wavelength of 300 – 470 nm. Whereas at wavelength 470 – 600 nm the graph of transmittance looks flat, which means that the wavelength of 470 – 600 nm photons does not occur in the movement of electron jumps.

Figure 3 (b) shows the increase of the doping percentage causes the absorbance of the thin film to increase at wavelength 290 – 295 nm. The absorbance at a wavelength of 295 nm for doping percentage 0, 5, 10, 15, 20, and 25% is 4.67; 4.34; 5.00; 4.58; 4.50; and 4.59. Absorbance value decrease after reach a wavelength of 295 nm. In general, the increase of doping percentage makes the transmittance of SnO$_2$:In+Al increases, and the absorbance of SnO$_2$:In+Al thin films decreases. This is due to the presence of In and Al doping in the SnO$_2$ structure which causes the density of the crystal structure to increase so that electrons from light can move more rapidly.

![Graphs](a) Transmittance of SnO$_2$:In+Al thin films with doping percentage (%) 0, 5, 10, 15, 20, and 25%, (b) Absorbance of SnO$_2$:In+Al thin films with percentage of doping 0, 5, 10, 15, 20, and 25%.

### 3.2. Calculation of Energy Gap

The energy gap of the thin layer is determined from the analysis of the direct optical energy gap and the indirect optical energy gap. The absorbance amount of SnO$_2$:In+Al thin films is used to find the energy gap of the thin films. The energy gap is determined from the graph slope of the energy of the photon ($hν$) to $(αhν)^n$.

#### 3.2.1. Direct Optical Energy Gap ($n = 1/2$)

Figure 4 shows the doping of Indium and Aluminum to the SnO$_2$ thin layer reduces the direct optical energy gap from 3.58 eV to 3.54 eV. The higher percentage of doping: 0, 5, 10, 15, 20, and 25% results in an energy gap being reduced from 3.58; 3.56; 3.55; 3.54; 3.56; and 3.55 eV.

#### 3.2.2. Indirect Optical Energy Gap ($n = 2$)

Figure 5 shows the slope graphs of each doping variation. The addition of doping causes the gap of indirect energy to thin film to decrease, starting from 3.90; 3.88; 3.88; 3.88; 3.88; and 3.87 eV.

Figure 6 shows that the direct optical energy gap is lower than the indirect optical energy gap. The direct optical energy gap decreasing from 3.58 eV to 3.54 eV. The indirect optical energy gap decreased from 3.90 eV to 3.87 eV.

The position of the valence band and conductivity of the direct optical energy gap is in one phase, whereas the valence band and conductivity for the indirect energy gap are in different phases. In the direct optical energy gap, electrons move to the conduction band from the valence band in a straight line so that the electrons arrive at the conduction band faster. Meanwhile, in the indirect optical energy gap, there is a jump of electrons to the conduction band from the valence band in a non-straight line, so the electrons arrive at the conduction band slower.

This causes the direct optical energy gap to be smaller than the indirect optical energy gap. The reduction in the energy gap is caused by the presence of impurities in the SnO$_2$ structure, namely doping given in the form of Indium and Aluminum which makes a new recombination center with lower emissions of energy.
Figure 4  Allowed transition $(\alpha h\nu)^{1/2}$ versus $h\nu$ on SnO$_2$:In+Al thin films (a) 0%, (b) 5%, (c) 10%, (d) 15%, (e) 20%, and (f) 25%.
Figure 5 Allowed transition $(ahv)^2$ versus $hv$ on SnO$_2$:In+Al thin films (a) 0%, (b) 5%, (c) 10%, (d) 15%, (e) 20%, and (f) 25%.

Figure 6 The direct and indirect optical energy gap of SnO$_2$:In+Al thin films.

3.3. Optical Activation Energy

The value of the optical activation energy is determined from the slope value ($m$) of the photon energy graph for $\ln(\alpha)$. The amount of optical activation energy $E_a = 1/m$ [30].

Gradient graph ($m$) of each percentage of doping (0, 5, 10, 15, 20, and 25%), namely 1.063 each; 1.027; 1.070; 1.072; 1.168; and 1.130. From the value of gradient, the activation energy of optic in figure 6 is...
0.94; 0.97; 0.94; 0.93; 0.86; and 0.88 eV. The reduced activation energy causes electrons to move faster than the valence band to the conduction band in the material so that the material becomes more semiconductor [31, 32].

![Graph](image)

**Figure** 7 The graph of the doping percentage versus activation energy of SnO$_2$:In+Al thin films.

### 4. CONCLUSION

The addition of doping percentages of Indium and Aluminum to SnO$_2$ thin films causes the transmittance of thin films to increase from 68.6 – 78.3% at a wavelength of 300 – 470 nm, and the absorbance is decreased after reach a wavelength of 295 nm. The gap between the SnO$_2$ thin film energy bands decreases with increasing doping concentrations of Indium and Aluminum. The reduction in energy gap from 3.58 eV to 3.54 eV and optical activation energy from 0.97 eV to 0.86 eV makes high-quality thin films.

### AUTHORS’ CONTRIBUTIONS

Aris Doyan has contributed to making articles and data analysis. Susilawati contributed to the data analysis. Haris Munandar has contributed to data collection and data processing for making complete articles.

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