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MORPHOLOGY AND RESISTIVITY VALUES OF FLUORINE-DOPED TIN OXIDE (FTO) USING INDONESIAN LOCAL DIMETHYL TIN DICHLORIDE (DMTC) PRECURSORS

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Abstract

TCO (transparent conductive oxide) is the main component for solar cell fabrication. One of the promising types of TCO is FTO (fluorine-doped tin oxide). The method used in depositing the conductive layer of FTO is spray pyrolysis with an ultrasonic nebulizer. The precursor used is a local Indonesian product, DMTC (dimethyltin dichloride) with doping ammonium fluoride (NH_4F). The variable that used in this study were variations in deposition time (5, 10, 15, 20, and 25 minutes) with a fixed substrate temperature at 300 °C and doping variations (un-doped, 2 wt.% doped, and 8 wt.% doped) to see the effect of adding F doping to the precursor solution. The resistivity values with a variation of deposition time 5, 10, 15, 20, and 25 minutes (2 wt.% doped) are $0.218x10^{0}$; $0.449x10^{-1}$; $1,567x10^{-2}$; $0.676x10^{-2}$ $0.377x10^{-2}$ Ω.cm. For doping variations (un-doped, 2 wt.% doped, and 8 wt.% doped) the values are $0.883x10^{-2}$; $0.377x10^{-2}$; $0.506x10^{-3}$ Ω.cm. The resistivity values tend to decrease with an increase in deposition time and doping addition, resulting in enhanced conductivity. The grain size will increase as deposition time and doping are both increased. In this study, the optimal resistivity value of 0.377x10-2.cm was obtained at a deposition time of 25 minutes with 2 wt.% doping.

Keywords: *FTO (fluorine-doped tin oxide), DMTC (dimethyltin dichloride), deposition time, doping, resistivity*

1. INTRODUCTION

Indonesia is an archipelagic country located on the equator and in a ring of fire, so it has potential sustainable power such as wind, solar, and geothermal. The angle of the sunlight is shallow on the equator, so the solar energy that falls per unit area is more extensive. The utilization of solar energy can be used as an alternative energy source in overcoming the energy crisis because it enhances sustainability and reduces pollution. The radiant light from the sun can generate electricity by utilizing solar cell technology. DSSC (dye-sensitized solar cell) is among the solar cells that can be used which is simple and flexible so that it can be used in various applications, and its fabrication requires low costs [1].

In DSSC manufacturing, one of several main components needed is a TCO (transparent conductive oxide) or transparent conductive glass. Tin oxide (SnO_2) is a semiconductor with good electrical and optical performance, so it is extensively utilized as a transparent conductor for various applications [2]. SnO_2 has been developed as a result of its low production costs, its resistance to atmospheric conditions [3], and responsiveness to several gase [4]. The optical and electrical performance of the tin oxide film could be improved thru doping with ITO (indium

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tin oxide) [5], ATO (antimony tin oxide) [6], or FTO (fluorine tin oxide) [7]. The most common tin oxide film commercialized is ITO. However, the material cost for ITO is extremely limited and expensive, so FTO is an alternative to ITO because it is less expensive [4] and requires more accessible raw materials. The electrical properties of tin oxide film are highly dependent on oxygen stoichiometry [3]. Various types of deposition techniques are chemical vapor deposition [8], sputtering [9], sol-gel method [10], spin coating [11], and spray pyrolysis [12].

The most widely used precursors in FTO manufacturing are $SnCl_2.2H_2O$ (dihydrated tin (II) chloride) [13]-[14] and $SnCl_4.5H_2O$ (Tin (IV) chloride pentahydrate) [15]. Several previous studies have conducted experiments using DMTC (dimethyltin dichloride) precursors, but most used CVD (chemical vapor deposition) method in their research. Mannie, G.J.A studied the surface characteristics of tin oxide thin films through CVD method utilizing TTC (tin tetrachloride) precursors, DMTC, and MBTC (monobutyl tin trichloride) [8]. Van Mol et. al., [16] studied the decomposition of DMTC in pure N_2 using a CSTR reactor at T = 370-630 °C and p = 1 bar. He observed the production of methane, ethane, ethylene, and propane by FTIR and QMS during the decomposition of DMTC at 500 °C and found no evidence of the presence of HCl and CH_3Cl.

This research used a local product precursor produced in Indonesia as DMTC as an alternative precursor to make FTO conductive layer utilizing the ultrasonic spray pyrolysis nebulizer technique. In this investigation, the influence of deposition time and fluorine (NH_4F) doping on the morphology and physical properties of the FTO conductive layer was determined.

2. MATERIALS AND METHODS

The first stage of this research was to wash the substrate glass (soda lime glass) gradually. First, the substrate is cleaned using detergent to eliminate grease and dirt. Then it is placed within a beaker filled with acetone and vibrated using an ultrasonic cleaner for 15 minutes, dried, and put in a storage container.

The precursor solution was made by mixing 0.9 M of Indonesian $C_2H_6Cl_2Sn$ (dimethyl tin dichloride) with 86 ml of distilled water for 30 minutes. The solution was then treated for 30 minutes with various concentrations of ammonium fluoride (98%, Merck Ltd., Germany). The method chosen for depositing the solution in this study was spray pyrolysis using an ultrasonic nebulizer (GEA Medical 402A1) at a speed of ± 30 ml / 10 minutes and a distance of 10 cm. Spray pyrolysis, in particular, is one of the promising deposition techniques because it is simple, reliable, and economical [17]. The variations used in this study were deposition times of 5, 10, 15, 20, and 25 minutes at 300 °C temperature. The morphology of this FTO (fluorine tin oxide) thin layer was characterized utilizing a SEM-EDS (scanning electron microscope-energy dispersive spectrometry) (JEOL-JSM 6390A) while the electrical parameter, such as resistivity, was measured using a four-point probe (FPP5000).

3. RESULT AND DISCUSSION

3.1 Deposition Time Effect

SEM (scanning electron microscope) analysis in Fig. 1 shows that adding deposition time will result in significant morphological changes.

Figure 1(a) reveals that after 5 minutes of deposition, the surface morphology of the layer is flat and smooth. It consists of small refined grains with a particle size diameter in the range of 39.40-53.81 nm. The particle size diameter increased to 46.65-100.32, 82.07-34.40, and 99.04-150.31 nm as deposition time was increased to 10, 15, and 20 minutes. The granules become denser and more interconnected as the porosity is significantly decreased. The largest particle size was obtained at the deposition time of 25 minutes. According to Table 1, the diameter of the particles was between 604.37 and 832.01 nm. The particle size will grow as deposition time increases. After allowing new nucleation to form, adjacent grains start to collide, causing an increase in internal stress that is reduced by the forming of a more excellent crystalline structure [18].

Table 1. The particle size of the FTO conductive layer with variations in deposition time with a doping ratio of 2 wt.% and a substrate temperature of 300 °C using SEM analysis

Time (minutes)	Particle Size (nm)
5	37.95 - 53.81
10	46.65 - 100.32
15	82.07 - 134.40
20	99.04 - 146.48
25	604.37 - 832.01

According to the semi-qualitative and semi-quantitative EDS (energy dispersive spectrometry) analysis, the conductive FTO (fluorine tin oxide) layer depicted in Figure 2 contains O and Sn, which are obtained from the precursor solution of dimethyl tin dichloride. ($C_2H_6Cl_2Sn$). In contrast, Mg, Ca, Na, Si are derived from substrate constituent elements (glass soda-lime) [19]. The presence of a glass element in the film indicates that the positioning process has not occurred completely. Figure 2 depicts a semi-quantitative

analysis showing that the FTO thin layer consists primarily of SnO_2.

Figure 2 indicates that the concentrations of Sn and O fluctuate. Sn elements are dominant from time to time because Sn elements with larger molecular weights tend to stick more to the substrate than the F dopant when given heating treatment and at a longer deposition process. This phenomenon proves that as the deposition time getting longer, the higher the probability that the F dopant in the form of gas will experience two conditions when the substrate is heated, namely, some of the F dopant sticks to the substrate and the other evaporates into the air [20]. The F content appears undetectable by the semi-quantitative EDS test because the F content is minimal. This finding is in agreement with the experiment performed by Bilgin et. al., [21], that the F doping element only functions as a catalyst to produce SnO_2 compounds with better structure and quantity.

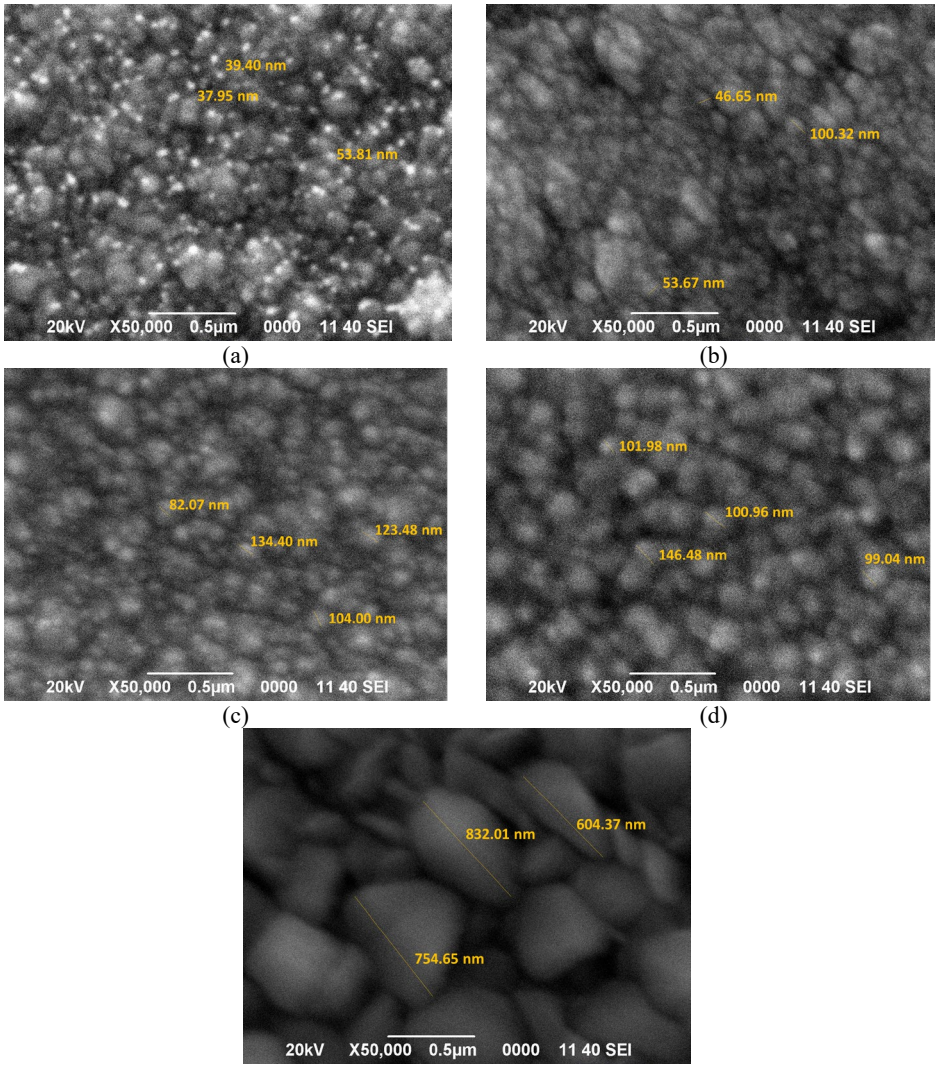


Figure 1. Morphology of SnO_2 thin layer, doping ratio 2 wt.% and substrate temperature 300 °C with variation of deposition time; (a) 5, (b) 10, (c) 15, (d) 20, and (e) 25 minutes

Figure 3 is a cross-sectional SEM image of the FTO conductive layer (top side) and glass substrate (bottom side), resulting from deposition on the substrate within 20 and 25 minutes. From Figure 3, it can be seen that the layer thickness is 560-600 nm at 20 minutes and the layer thickness is 1.28-1.36 µm at 25 minutes. With increasing deposition time, the collision of elemental fluorine ions on SnO_2 will continue. The longer the collision, the thicker the surface layer will be [22]. There was a linear relationship between the thickness and the deposition time between 20 and 25 minutes. This result is consistent with the report from previous authors [23]-[24].

Table 2. Resistivity values with varying deposition time, doping ratio 2 wt.% and substrate temperature 300 °C

Time (minutes)	Resistivity (Ω.cm)
5	0.218×10^{0}
10	0.449×10^{-1}
15	1.567×10^{-1}
20	0.676×10^{-2}
25	0.377×10^{-2}

The resistivity of the SnO_2 thin layer versus deposition time is shown in Table 2. The resistivity value decreases as deposition time increases. This finding suggests that the conductivity value improves with increasing deposition time. Generally, an increase in the thickness of a thin layer causes the resistivity to decrease [25]. Conductivity is the ability of a material to conduct electric current. It is the inverse of electrical resistivity. Therefore, increasing the thickness of the thin layer increases its conductivity. As deposition time increases, the SnO_2 particles become denser, thereby lowering porosity, and the interconnection becomes more interconnected, electronic transfer between particles becomes smoother, and the resistivity is considerably decreased [26].

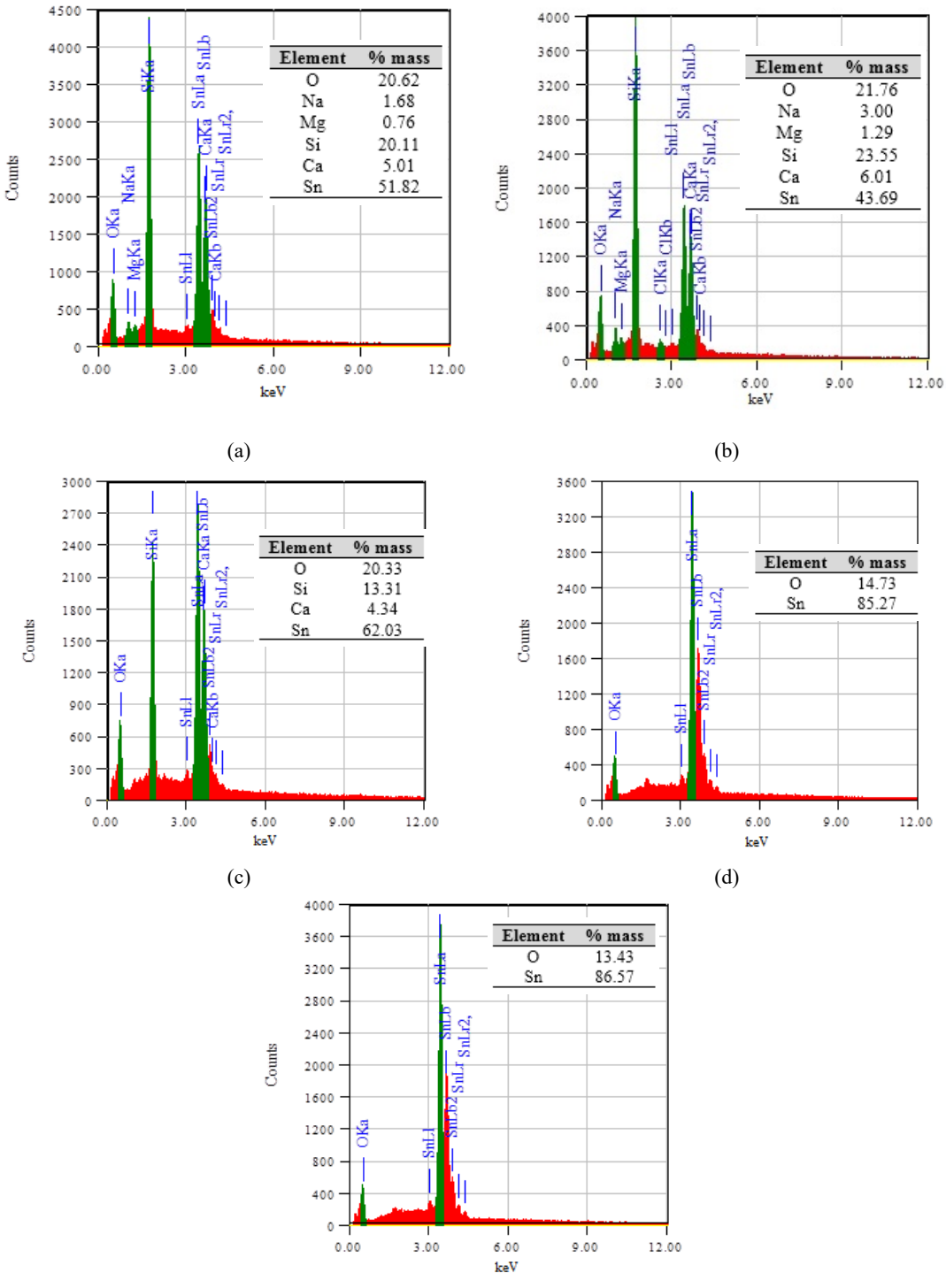
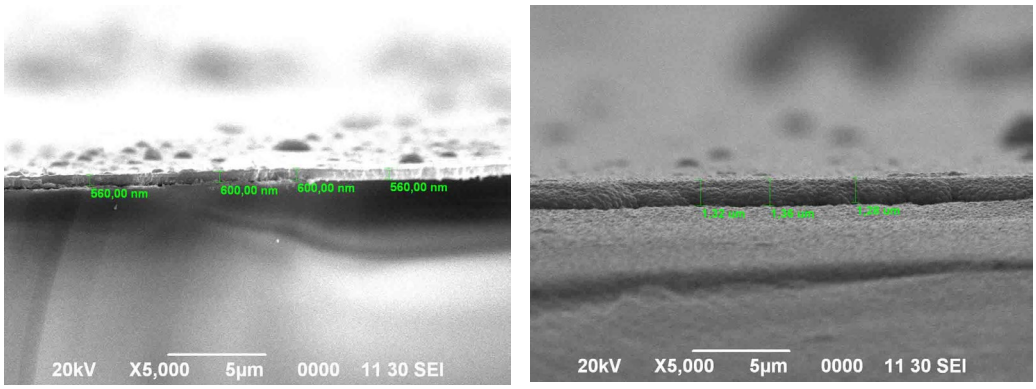


Figure 2. Semi-qualitative graph of EDS of the FTO conductive layer at deposition time variations with a doping ratio of 2 wt.% and a substrate temperature of 300 °C using EDS-SEM analysis (a) 5, (b) 10, (c) 15, (d) 20, and (e) 25 minutes

3.2 Doping Effect

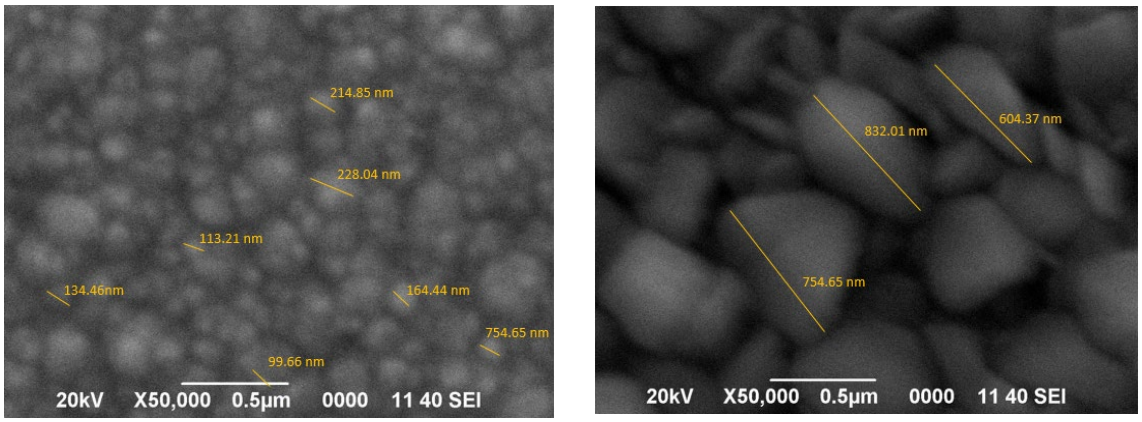
Figure 4 depicts the surface morphology of FTO thin films with varying doping levels, coated for 25 minutes and 300 °C substrate. The surface morphology of the non-doped layer has a particle size diameter of 99.66-228.04 nm. There was an increase in the particle size diameter to 496.06-596.01 and 868.00-1290.00 nm with the addition of 2 wt.% and 8 wt.%, respectively, as shown in Table 3.

According to the SEM analysis, the surface morphology of the undoped thin layer consists of small grains, whereas the particle size increases with the addition of doping. According to research conducted by P. Yao [27] and D. Tatar [28], the grain size increases as the doping level rises. The results are consistent with these findings. The morphology of the crystals gets bigger with increasing doping levels because the crystals are getting denser, and there is an agglomeration process [29]. The agglomeration process involves the accumulation of nanoparticle-sized solids in order to produce larger crystals.

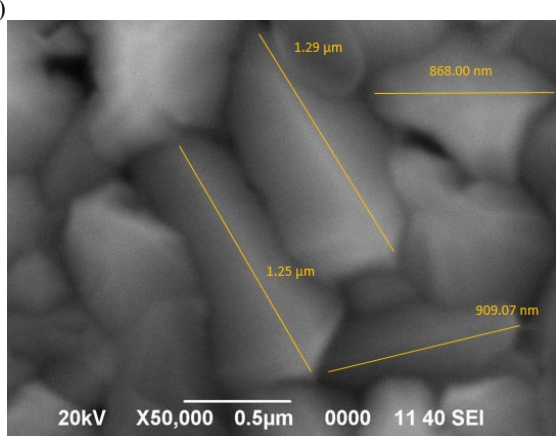


(a) (b)

Figure 3. Cross-sectional view of the FTO thin layer, doping ratio 2 wt.% And substrate temperature 300 °C with varying deposition time (a) 20, and (b) 25 minutes



(a) (b)



(c)

Figure 4. Morphology of the SnO_2 thin layer, deposition time of 25 minutes and substrate temperature of 300 °C with doping variations; (a) undoped, (b) 2 wt.%, (c) 8 wt.%

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EDS analysis of chemical content of thin films in Fig. 5 shows that the elements of Sn and O are more dominant in thin films. However, element F was not detected in the thin layer. Gaseous Element F is difficult to identify with EDS. Furthermore, the small concentration of this element in the original solution may limit the detection of this element. Figure 5 shows that the incorporation of F increases the number of Sn elements. Since the diameter of the fluorine ion (F^-: 0.133 nm) is nearly identical to oxygen (O^{2-}: 0.132 nm), a rise in the Sn/O ratio shows the effective doping of F through to the lattice as either an interstitial or an oxygen substitution [7], [21].

According to Table 4, NH_4F addition will impact the resistivity of the thin layer. The undoped film has a greater resistivity value than the doped layer. As doping increases, the resistivity value lowers. According to prior studies, the doping amount effects the stoichiometric of the SnO2 thin layer. As the doping level increases, the thin film becomes nonstoichiometric, wherein fluorine atoms with ionic surface mobility scattered on the specimen surface increase with doping, causing the grain size to increase, grain boundaries decrease so that the electrical properties increase [28].

Table 3. The particle size of the conductive FTO layer with the effect of adding doping, deposition time of 25 minutes, and substrate temperature of 300 °C using SEM analysis

Doping	Particle Size (nm)
Undoped	99.66 – 228.04
2 wt.%	496.06 – 596.01
8 wt.%	868.00 – 1290.00

This is in accordance with previous studies, which demonstrates that the resistance drops with increasing doping concentrations after doping with 30 F/Sn atomic% but then increases at doping levels greater than 70 F/Sn atomic% [30]-[31].

Table 4. The results of the resistivity test for doping variations with a deposition time of 25 minutes and a substrate temperature of 300 °C

Doping	Resistivity (Ω.cm)
0 wt.%	0.883×10^{-2}
2 wt.%	0.377×10^{-2}
8 wt.%	0.506×10^{-3}

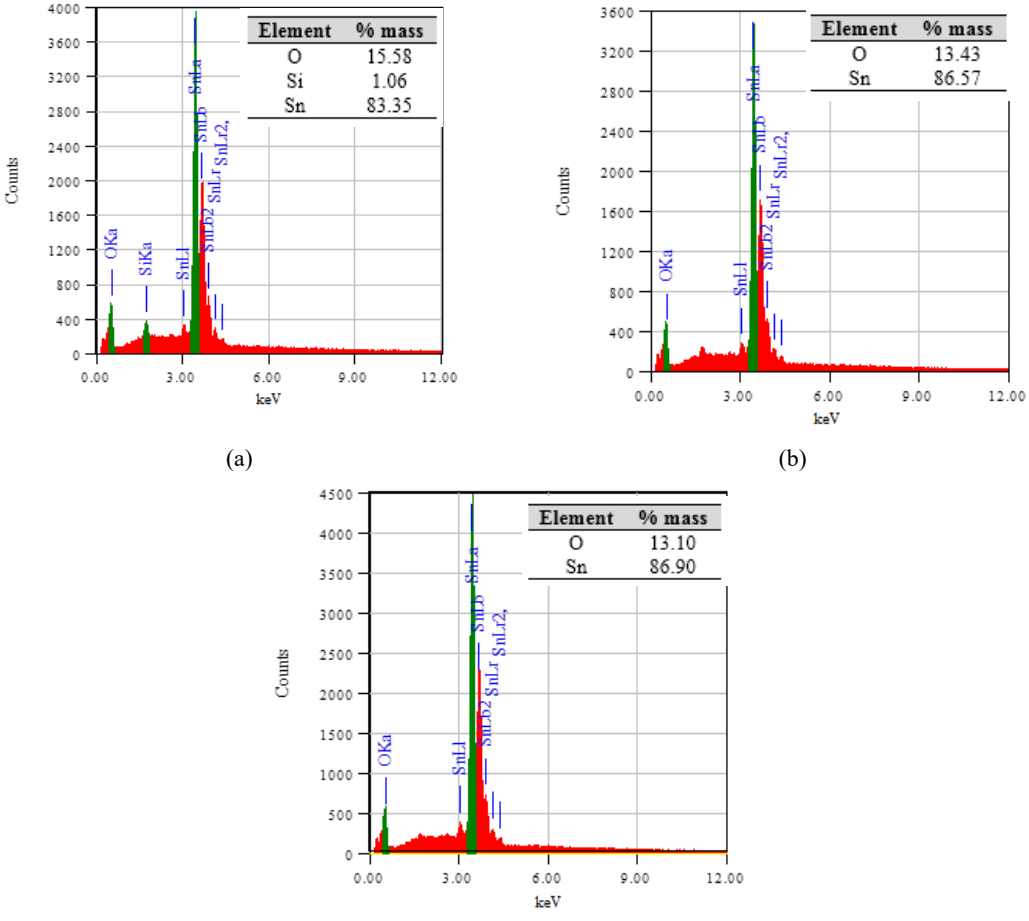

Figure 5. Semi-qualitative graph of EDS of the conductive FTO layer at doping variations with a deposition time of 25 minutes and a substrate temperature of 300 °C using EDS-SEM analysis (a) Undoped, (b) 2 wt.%, and (c) 8 wt .%

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4. CONCLUSION

FTO (flourine tin oxide) conductive layer production by spray pyrolysis method utilizing an ultrasonic nebulizer generated a comparatively good layer with the smallest resistivity value of $0.377x10^{-2}$ Ω.cm at a time deposition of 25 minutes and 2 wt.% doping. As deposition time increases, the larger the grain size will be, which causes the conductive layer to be thicker. The effect of increasing the thickness of the conductive layer will result in a decrease in the resistivity value so that better conductive properties will be obtained.

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