Semiconductor Properties of Nanostructured Al doped ZnO thin film Annealed at Different Temperatures

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***Abstract*- The material Aluminium doped Zinc Oxide was used as a thin film with doping concentration of 1%.Thin film was deposited by using sol-gel spin coating method. Different annealing temperature were varied as a parameter from 400, 450 500, 550 °C. The thin film were characterized by using 2- point probe I-V measuremet, UV-VIS spechtrophotometer (Jasco/V-670 Ex), Surface Profiler, FESEM. I-V measurement results revealed improvement in electrical properties of the thin films with annealing temperatures. Annealing temperature increase carrier concentration in the nanostructures arrays and improves the semiconductor properties of sample.**

Keywords: Al doped Zn; Sol Gel Spin-Coating Method; Doping Concentration; Electrical Properties; carrier concentration

I. INTRODUCTION

Currently, intense researches are being done on zinc oxide (ZnO) due to its unique properties; such as non-toxicity, high chemical stability, high abundance, and controllable electrical conductivity. ZnO has wide direct band gap of 3.37 eV and higher exciton binding energy than GaN, of 60 meV which makes it suitable for optoelectronic devices [1]. For sol-gel derived ZnO thin films, annealing temperature was set parameters in order to optimize the properties of the thin film. Zinc oxide (ZnO) as a n-type II-VI inorganic semiconductor material can be used for sensor, electronic, optoelectronic, and photovoltaic applications [2]. The main advantage of the sol gel method is its simplicity, low cost and ability to obtain uniform films with good adherence and reproducibility [3]. In this paper we report on the preparation of high-quality AlZnO thin film by the solgel deposition process. The solgel deposition technique is an easy and very cheap method to metal oxide film [4]. By doping impurities such Al resulted high conductivity in electrical properties[5]. Al doped ZnO thin films at concentration of 1% through sol-gel spin-coating method. The doping concentration is fixed at 1 at.% because it have been reported that the value is the optimum parameter to obtain low resistance thin film [6]. The effect of annealing temperature were observed towards the semiconductor properties of nanostructured AlZnO thin film.

II. METHODOLOGY

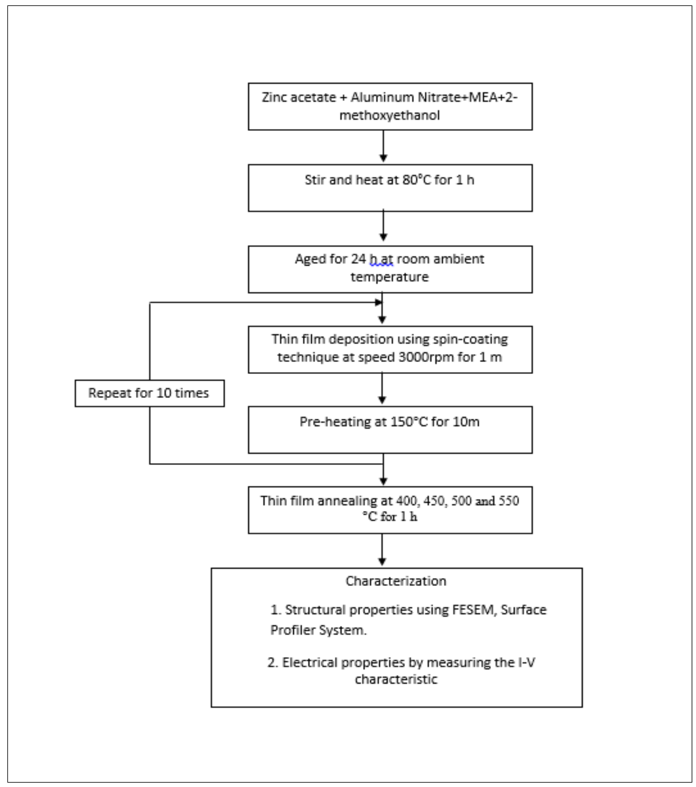

Fig. 1: Al-doped ZnO thin films preparation using sol- gel spin-coating technique.

Al doped ZnO thin film-coated glass substrates were prepared using sol-gel spin-coating method. A solution for thin films fabrication was prepared at concentration of 0.8 M using zinc acetate dihydrate as a precursor,monoethanolamine (MEA) as a stabilizer, aluminum nitrate and 2-methoxyethanol as a solvent. The solution was stirred and heat for 1 hours before aged for 24 hours at room ambient. Then, the solution was spin-coated on glass substrates at a speed of 3000 rpm for 1 minute. The as-deposited

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films were dried at 150 °C for 10 minutes to evaporate the solvent. The coating procedure was repeated for a 10 times to increase the film thicknesses. After the process, the thin films were annealed at different temperature at 400, 450, 500 and 550 °C for 1 hour in a furnace.

The surface morphologies of Al doped ZnO thin film were characterized using field emission scanning electron microscope (FESEM). The thicknesses of the samples were measured using surface profiler (VEECO/Dektak 150+). The electrical properties of the samples were investigated using two-probe current-voltage (1-V) measurement system (Advantest R6243).

III. RESULT AND DISCUSSION

A. Electrical properties

By using 2 probe *I-V* measurement, the result was plots for Al doped ZnO thin films at different annealing temperatures are shown in Fig. 4. Gold (Au) was used as the metal contacts in the measurement. The result indicates the Ohmic behavior of thin films with linear *I-V* curve observed for all annealed thin films. The supply voltage that supplied to the different type of sample is same from -5v to 5v. The graph on figure 4 shows that current value with fixed temperature increase when the annealed temperature increase [6]. From the Fig 4, we can see that the slope of graph is different between different temperatures. It is because of effect annealed temperature at 400, 450, 500 and 550 °C. The thickness of a thin film not affected the resistivity. Refer from the *I-V* curve on figure 4, we observed the resistivity by using following equation[6] (1):

$$\rho = \left(\frac{V}{I}\right)\left(\frac{wt}{x}\right) \qquad (1)$$

Where is resistivity, V is voltage, I is current, w is width of Au, t is thin film thickness and x is distance between electrodes. Hence, we seen that conductivity is inversely proportional to the resistivity. Referred from the following equation (2):

$$\sigma = \left(\frac{1}{\rho}\right) \qquad (2)$$

The increase in electrical conductivity brought in by the aluminum doping can be explained as follows: the concentration of free charge carriers in ZnO increases by the aluminum doping because aluminum has one valence electron more than zinc. We may consider that Aluminum substitutes the zinc atom or it occupies interstitial sites [2].

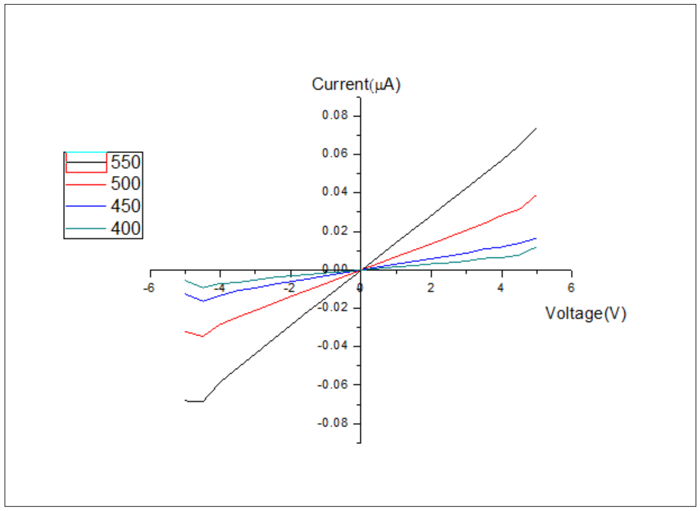

Fig. 4: I-V curves of Al doped ZnO thin film deposited on glass substrates with different of Annealing temperature.

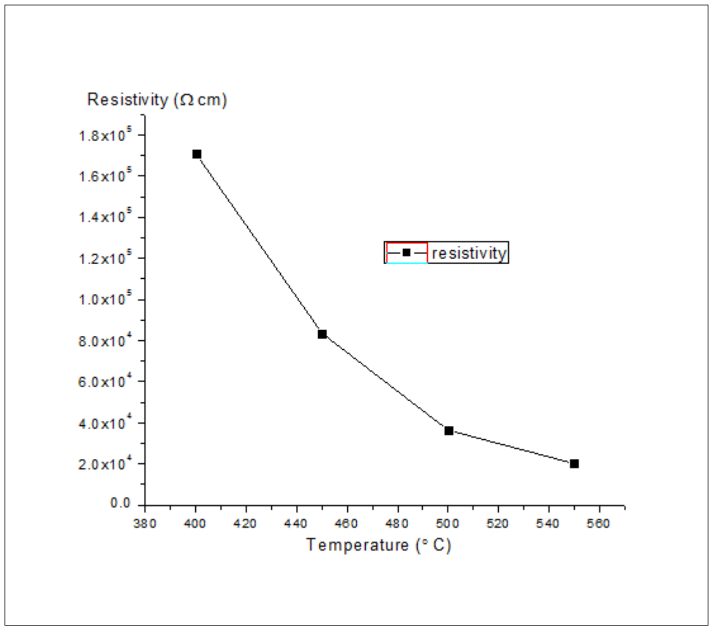

Fig. 5: Electrical resistivity of AlZnO thin films at different annealing temperature.

The graph of resistivity from the fig. 5 shows that the resistivity of thin film decrease when the temperature is increase. The calculated result for resistivity obtained in this project to be 1.71×10^5, 8.36×10^4, 3.64×10^4, 2.02×10^4 (Ω cm) for AlZnO thin film annealed at 400, 450, 500, 550 (°C). The crystallinity of the thin film is one of the role that affected the resistivity. The crystallinity of the thin film varies due to the annealed temperature. Figure 5 shows that the higher annealing temperature contributed to the decreasing of electrical resistivity.

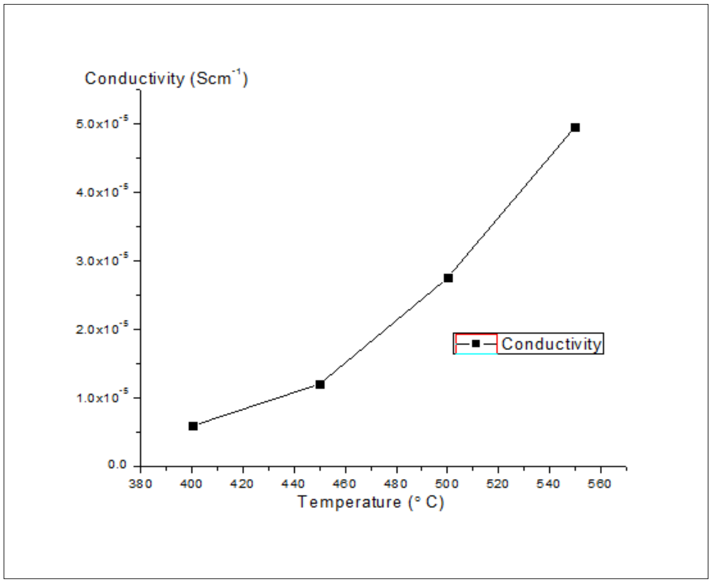

Fig. 6: Electrical conductivity of AlZnO thin films at different Al doping concentrations.

Annealing Temperature [°C]	Thickness [μm]	Resistance, R [Ω]	Resistivity, ρ [Ω cm]	Conductivity, σ [Scm^{-1}]
400	0.192	1.60x10^{9}	1.71x10^{5}	5.854x10^{-6}
450	0.178	6.65x10^{6}	8.36x10^{4}	1.196x10^{-5}
500	0.168	4.43x10^{6}	3.64x10^{4}	2.245x10^{-5}
550	0.172	1.87x10^{7}	2.02x10^{4}	4.953x10^{-5}

Table 1: Properties of AlZnO thin films at different annealed temperature obtained I-V measurement.

The diffusion of Al dopant in ZnO lattice during annealing process also contributed to the decrement of electrical resistivity in the nanostructures arrays at higher annealing temperatures. This process increases carrier concentration in the nanostructures arrays and improves the electrical properties of sample. The reduction of defects concentration might be also contributed to the lower resistivity value at higher annealing temperatures.

$$\sigma = q\mu_n N$$

q= charge
$\mu_{n=}$ Electron mobility
N= Electron majority carrier

B. Structural Properties

The FESEM images of the AlZno thin film annealed at different temperature for one hour were shown in figure 7. The figure 7 reveals all films were uniformly deposited on the substrate as the particles fully covered the substrate. The images also verified that prepared AlZnO materials in this research were within nanometer range (<100nm).In addition, fig. 6 proved that the particle size increase varies the temperature from 400, 450, 500,550 (°C) [6][7]. We also found that at 550 (°C) annealed temperature, the particles in the thin film start to combine each other and increase the grain size[5]. It is because of the particle attempt to release surface energy. As the annealing temperature increased, the grains became large and the grain boundary became densely packed. Thin film resistivity decreased as the annealing temperature increased because of a decrease in defects in the grain structure [8].

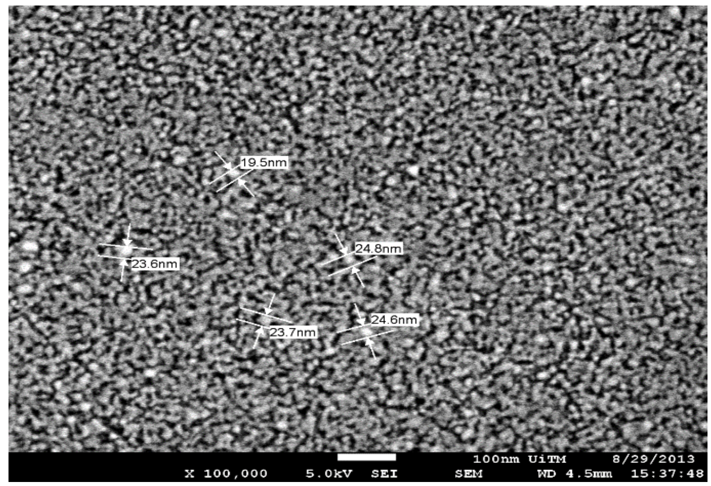

Fig. 7(a): Structural properties of AlZnO thin films at 400 °C. 23.24nm.

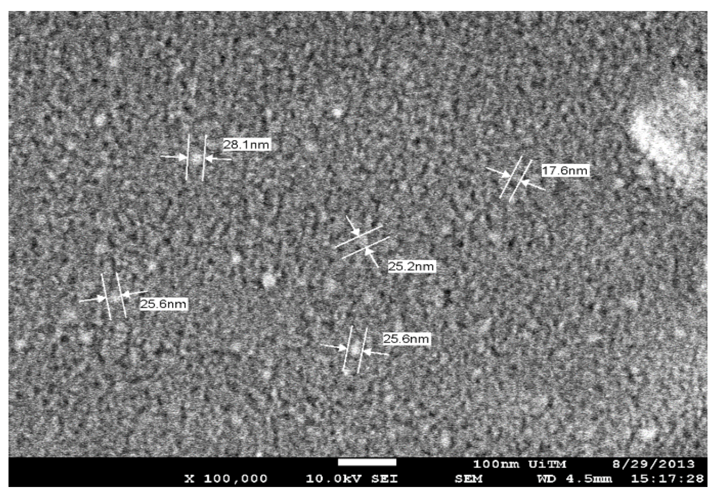

Fig. 7(b): Structural properties of AlZnO thin films at 450 °C. 24.54nm.

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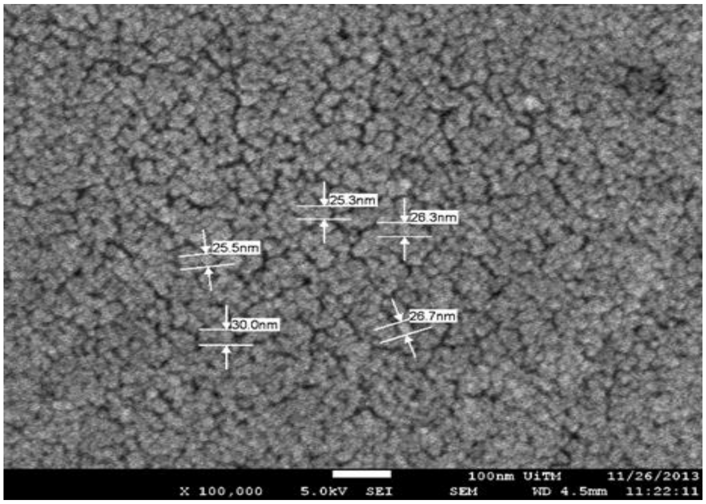

Fig. 7(c): Structural properties of AlZnO thin films at 500 °C. 26.67 nm.

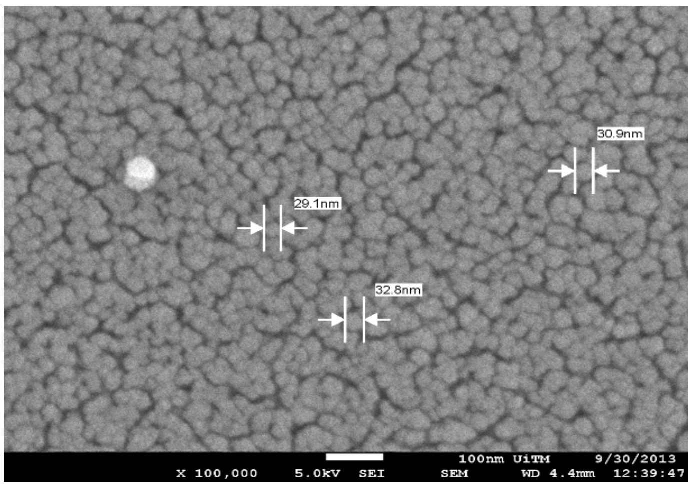

Fig. 7(d): Structural properties of AlZnO thin films at 550 °C. 30.93nm.

C. *Optical Properties*

As shown in Fig. 8 the transmittance spectrum of AlZnO thin film at annealing temperature. It could be observed that all films have high transparency in the visible and near infrared region which is good for transparent electronic device fabrication. From the figure 8, it shows that thin film at all variable temperature transmitted at more than 80% [4][9].Result show that when the annealing temperature increase, the transmittance of AlZnO increase [10].

The absorption coefficient was found by using Lambert`s law equation[9] (3). Hence, the graph of absorption coefficient was plotted on the figure 9.

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$$\alpha = \frac{1}{t}\ln\left(\frac{1}{T}\right) \qquad (3)$$

t = thickness of the thin film
T = transmittance spectrum of thin film

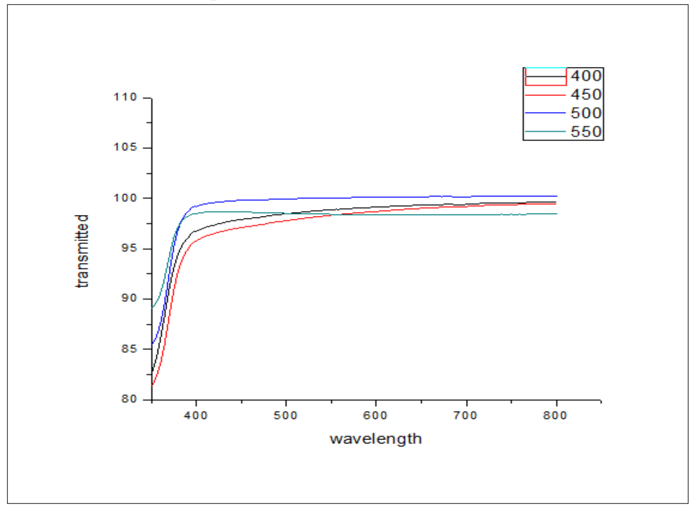

Fig. 8: Transmittance spectrum of AlZnO thin film at annealing temperature.

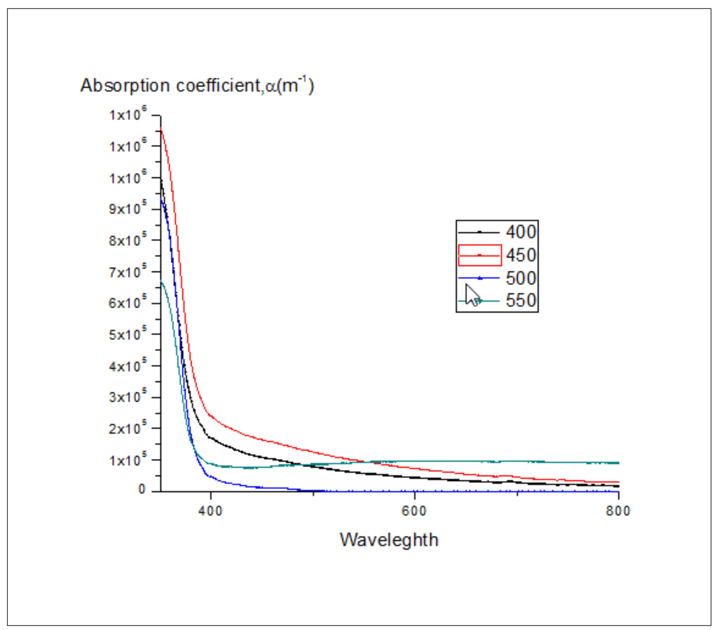

Fig. 9: Absorption coefficient of AlZnO thin film at annealing temperature.

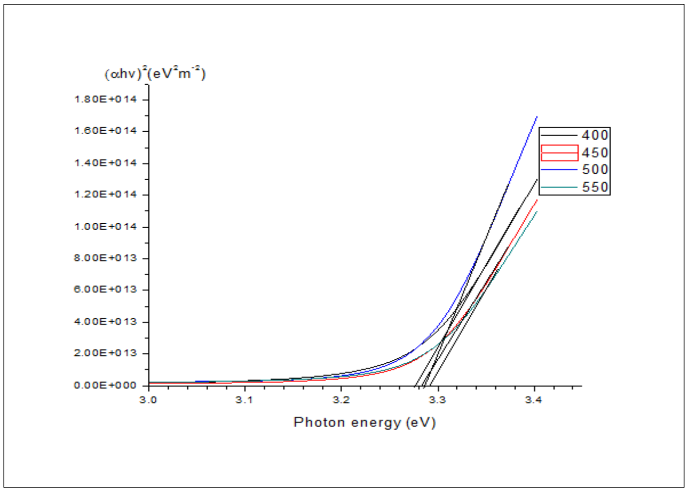

Fig. 10: Optical band gap energy of AlZnO thin film at annealing temperature.

The optical band gap values of AlZnO films were obtain from transmission measurements by plotting $(\alpha h\nu)^2$ versus photon energy graphs where α is the absorption coefficient and hv is the photonic energy. From the figure 10, the optical band gap is found to be 3.278, 3.282, 3.286, 3.29 (eV) for AlZnO thin film annealed at 400, 450 500, 550 (°C). In semiconductors the relation connecting the absorption coefficient *a*, the incident photon energy *hv* and the optical band gap E_g takes the form[2] .The graph on figure 10 was plotted using following equation(4)

$$ah = A(h\nu - E_g)^{1/2} \qquad (4)$$

IV. CONCLUSION

The AlZnO successfully prepared on a glass substrate by sol-gel spin-coating method.A homogeneous and stable sol have been prepared using zinc acetate (Zn(CH3COO)2.2H2O) as precursor, aluminum nitrate nanohydrate (Al(NO3)3.9H2O) as dopant source 2-methoxyethanol as solvent, monoethanolamine (MEA) as stabilizer. The effects of Annealing temperature on the AlZnO thin films properties have been investigated. The electrical properties have been characterized using I-V measurement while the optical properties have been studied using UV-Vis spechtometer The surface morphology of the thin films have been investigated using FESEM. The conductivity of thin films increased with increase of annealing temperature. Additional, the resistivity of thin film decrease varies temperature. The transmitted on UV-Vis showed that the thin film transmitted more than 80%, it also stated in literature. The FESEM research show that the nanoparticles size becomes larger as the increase of annealing temperature. At 550°C the particle start to merge each other because the particles attempt to release surface energy. Moreover, surface profiler showed that the thickness of a thin film decreasing from 400°C to 500 °C (0.192µm to 0.168µm). Only at 550°C the thickness of a thin film slightly increase to 0.172µm because of softening point, glass substrate expand when the temperature more than 500 (°C). It can be concluded that the semiconductor properties of AlZnO thin films fixed at 1.0% are optimum when annealed at 550 °C due to the highest conductivity and low resistivity.

FUTURE RECOMMENDATION

In the future, the surface topology of AlZnO can be investigate by using Atomic Force Microscopy. The sol-gel spin coating method can be replaced by using other method. Such as RF magnetron sputtering. It is because RF magnetron sputtering can produce smooth thin films. In addition, fabricate thin films for device application. Such as electronic application and gas sensor.

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