

## Electrical and optical properties of dip coated Al-doped ZnO thin films: Effect of Al-concentration, starting solution and sample ageing

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### ABSTRACT

Undoped and Aluminum doped Zinc oxide thin films were synthesized by dip coating technique. The electrical properties of the films were studied due to the Aluminum doping, starting solution aging and sample aging. The sheet resistance of ZnO : Al films was minimum at 2.5 at % whereas carrier concentration is maximum. Both undoped and aluminum doped Zinc oxide thin films were found to be highly transparent lying in between 65 - 79% in the wavelength range 367 nm to 1038 nm. The band gap of deposited films changed slightly from 3.22 eV to 3.27 eV.

**Keywords:** Zinc oxide thin films, dip coating technique, sample aging, solution ageing, band gap

### INTRODUCTION

Zinc oxide (ZnO) thin film is a II - VI group compound semiconductor [1-3] which is technologically important due to their range of electrical and optical properties [4] and these make them suitable for a variety of applications like in gas sensor and in optical waveguide along with the solar cells, thermoelectric gas sensitive devices and transparent electrodes in optoelectronic device [1-5].

It is known that aluminum is one of the effective dopants to ZnO to enhance its conductivity. Compared with indium tin oxide (ITO) and tin oxide (TO), aluminum doped zinc oxide thin films have a higher optical transmittance and lower resistivity. In addition, these films are more resistant against hydrogen and hydrogen plasma treatments, and they do not degrade active solar cell materials by inter-diffusion of constituents as it occurs with ITO or TO [6]. Hence, aluminum-doped ZnO films are an important alternative to tin doped indium oxide layers, which are widely used as transparent conductive oxide (TCO) layers in solar cells due to their superior electrical and optical properties. Besides the good electrical and optical parameters, zinc oxide is less toxic and relatively cheaper than its competitors [6, 7].

There are various techniques to synthesize doped and undoped ZnO films. They are spray pyrolysis [8], magnetron sputtering [9-11], chemical vapor deposition [12, 13], Electrochemical deposition [14], Atomic layer deposition [15], Sol-gel technique [16 -18] etc.

Out of all above mentioned technique, Sol-gel technique is an attractive technique for obtaining thin films and has an advantage of easy control of the film composition and fabrication of small as well as large area thin films with low cost [19]. Generally, there are three methods used in sol-gel technique. They are spin-coating, dip coating and spray coating. The properties of ZnO films are not only influenced by the deposition technique but also the post deposition sample age and ageing-time of the starting solution [20, 21, 22]. There are many studies such as effect of ageing-time of starting solution [22], aluminum (Al) concentration [13], aging and annealing [20], optical properties [19], structural, electrical and optical properties [23], optical transmittance and photoconductivity [24] of ZnO films deposited by various techniques were found. However, detailed study on electrical properties such as effects of sample age, solution age and Al-concentration on sheet resistance as well as optical properties of both undoped and aluminum doped zinc oxide (ZnO : Al) thin films deposited onto glass substrate by dip coating technique, using 0.1 M solution of zinc acetate and methanol as precursors, has not been reported.

Since dip coating technique is both simple and economic [25], so, in this work, we have synthesized ZnO and ZnO : Al thin films by dip coating technique using methanol as precursor solution and then electrical properties due to the effects of sample age, solution age, Al-concentration in parent solution and optical properties of the thin films are investigated.

## EXPERIMENTAL

The sol-gel technique is a versatile solution process for making ceramic and glass materials. In general, Sol-gel process involves the transition of liquid "Sol" to solid "Gel" phase. The films were prepared on a piece of glass substrate by dip coating. When the "Sol" is cast into a mold then a net "Gel" will form. With further drying and heat treatment, the "Gel" is converted into dense ceramic or glass materials. For the sample solution, 0.1 M solution using zinc acetate  $[Zn(CH_3COO)_2]$  (Ava Chemicals Private Limited, India, 98% Purity) as a precursor and pure methanol ( $CH_3OH$ ) (Sisco Research Laboratories Private Limited, India, purity 99%) as solvent, was made and stirred by magnetic stirrer to get homogeneous solution. The chemical reaction for the formation of ZnO is as follows:  $Zn(CH_3COO)_2 \rightarrow ZnO + CO_2\uparrow + CH_3COCH_3\uparrow$  and hence the film is deposited on the substrate. The acetone ( $CH_3COCH_3$ ) is a volatile substance which is evaporated. The prepared solution was allowed to age for few days before deposition. For deposition of ZnO thin films, the commercial grade glass substrate was thoroughly cleaned with cleaning liquid soap by using ultrasonic cleaner (PS-60A, ROHS) for 15 minute and it was kept inside oven for 5 hours to dry it at room temperature. After that this glass substrate was dipped inside the beaker containing precursor solution then it was taken out. The deposition rate can be varied by varying the withdrawing velocity. A standard withdrawing velocity of 0.2 cm/s was selected for the present case. After deposition, it was allowed to dry in air and then it was heated at the temperature about 490 °C to crystallize the ZnO film for 2 minutes. Then it was allowed to cool in air at room temperature for further characterization. Now, to prepare Al-doped ZnO (ZnO : Al) thin film, aluminum chloride ( $AlCl_3$ ) was used as dopant source. For deposition of ZnO : Al, 0.1 M solution of  $AlCl_3$  with  $CH_3OH$  was prepared and it was added to the main zinc acetate  $[Zn(CH_3COO)_2]$  solution with different volume to vary doping concentration and repeated the same dip coating method to fabricate

ZnO : Al thin films to increase number of coating. We prepared both undoped and ZnO : Al thin films with 16 number of coating. In this way, the smooth, transparent and uniform thin film of both undoped and ZnO : Al deposited on glass substrate were obtained.

## RESULTS AND DISCUSSION

In order to study effect of Al concentration on electrical properties of ZnO : Al thin film, samples with different values of Al doping concentration (*i.e.* 0, 1.25, 1.875, 2.5, 3.75 and 5 at %) in the starting solution were used. For carrier concentration determination, laboratory purpose Hall effect measurement apparatus was used. Furthermore, to study optical properties spectrophotometer (USB2000 (photonics), Ocean Optics Singapore) was used to measure transmittance in wavelength range 367 to 1038 nm.

The transmittance data was further used to determine the absorbance and band gap energy of the deposited films.

**Electrical properties:** For electrical characterization, the sheet resistance and Hall coefficient were measured by using four probe methods in linear and vander paw configuration respectively.

Sheet resistance of the thin films can be determined by using the relation:

$$(Rs) = \frac{\pi}{\ln 2} \frac{V}{I} \quad (1)$$

Where current I is passed through the outer two probes and voltage V is measured between inner two probes using four probe method in linear configuration.

**Effect of aluminum concentration on sheet resistance:** Figure 1(a) shows the variation of normal (in light) and dark sheet resistance of ZnO : Al film having number of coating 16 with different % of Al content in ZnO. From this curve, it is seen that dark sheet resistance is higher than normal (in light) sheet resistance of both undoped and aluminum doped ZnO thin films measured at different sample age and solution age.

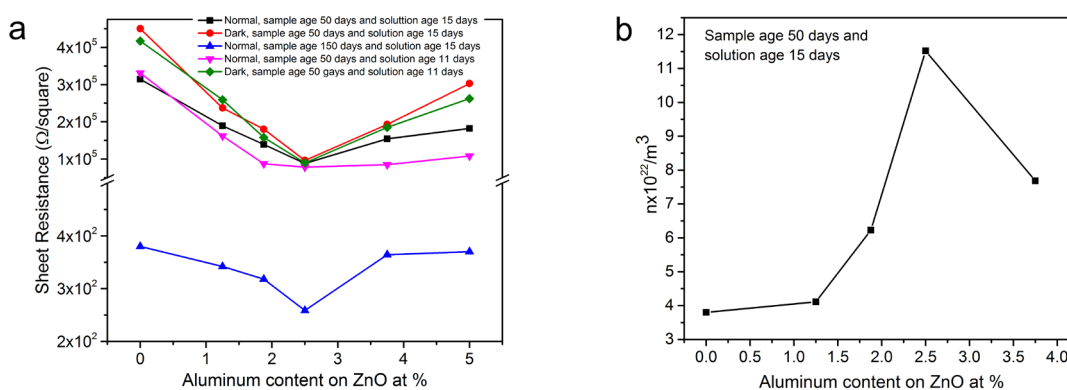


Fig. 1(a) Effect of different aluminum content on electrical sheet resistance of ZnO : Al thin films prepared with different solution age and sample age, (b) Effect of aluminum content on carrier concentration of ZnO : Al thin films with sample age 50 days and solution age 15 days composites catalyst, respectively.

In addition to this, both normal and dark sheet resistance of the films decreases gradually with increasing Al concentrations and attains minimum value at 2.5 at %. Beyond this, sheet resistance is observed to increase gradually with increasing doping concentration. The minimum value of dark and normal sheet resistance of the deposited films with sample age 50 days and solution age 15 days were found to be 96.3 K $\Omega$ / $\square$  and 88.37 K $\Omega$ / $\square$  respectively. Similarly, the minimum value of dark and normal sheet resistance of the deposited films with sample age 50 days and solution age 11 days were found to be 89.6 K $\Omega$ / $\square$  and 78 K $\Omega$ / $\square$  respectively. Furthermore, we have also taken sheet resistance measurement of the samples prepared with solution age of 15 days and sample age 150 days. It is seen that the variation of sheet resistance with aluminum content show similar trend in which the minimum sheet resistance with value 258.57  $\Omega$ / $\square$  is found at same aluminum content (ZnO : Al) = 2.5 at %.

Similar observation has also been reported by Mondragon-Suarez [13] for Al-doped ZnO films deposited onto glass substrates by the chemical spray technique. They found that the lowest resistivity values were obtained at same aluminum content (ZnO:Al) = 2.5 at % using ethanol and isopropanol as solvent with 0.2 M concentration.

In the case of sample age 150 days, drastic decrease in the value of sheet resistance is observed even though the same solution age *i.e.* 15 days. In figure 1(a), it is also seen that the variation of sheet resistance takes place not only with aluminum content but also with the difference in solution age as well as sample age. Irrespective to the deposition condition, solution age and sample age, the variation of sheet resistance with Al-concentration shows similar trend and minimum sheet resistance is observed at same aluminum content (ZnO : Al) = 2.5 at %. The decrease in sheet resistance up to 2.5 at % Al-content is due to the fact that the aluminum atom occupy into ZnO lattice acts as donor by supplying a single free electron when Al<sup>3+</sup> ion occupy Zn<sup>2+</sup> ion sites. One of the reasons for such configuration is that the ionic radius of Al (0.530 Å) is smaller than ionic radius of Zn (0.60 Å) in tetrahedral configuration [4]. So aluminum atoms occupy into ZnO lattice effectively act as donor by supplying a single free electron when Al<sup>3+</sup> ion occupies Zn<sup>2+</sup> ion sites. But at higher concentration, Al<sup>3+</sup> do not contribute free electron and acts as scattering source for charge carriers [26]. Hence, at higher doping level greater than 2.5 at %, the sheet resistance of film increases with increasing Al-concentration.

Furthermore, to study the effect of aluminum doping on carrier concentration, we have measured the value of carrier concentration of thin films by using the relation:

$$n = -\frac{1}{(R_H \times e)} \quad (2)$$

Where Hall coefficient RH is obtained as:

$$\text{sheet coefficient: } (R_H/d) = \frac{1}{B} \left[ \frac{R_{31,42} + R_{42,31}}{2} \right] \quad (3)$$

$$\text{where, } R_{31,42} = V_{42}/I_{31}$$

Where, current I is fed through two probes 1,3 and the voltage V generated across other two probes 2,4 is measured.

Using the above equations, we have also measured carrier concentration of both undoped and Al-doped ZnO thin films. The thickness of the films obtained by optical method was found to be 217 nm. Fig. 1(b) shows the relation of carrier concentrations and the Al content on ZnO film with sample age 50 days and solution age 15 days having number of coatings 16. It is found that carrier concentration increases with increase of aluminum concentration and becomes maximum at 2.5 at %. This causes sheet resistance of films decreases as shown in Fig. 1(a) and becomes minimum at 2.5 at % whereas at higher Al-concentration above 2.5 at %, carrier concentration decreases. This is due to fact that increasing dopant atoms above 2.5 at % form some kind of neutral defects and these neutralized Al-ions and do not contribute free electrons. The amount of electrically active Al in the film decreases when doping is high *i.e.* more Al-ions are neutralized at higher Al-concentration [26]. So, further increasing aluminum concentration, carrier concentration decreases which enhance the value of sheet resistance.

#### **Effect of the ageing–time of the starting solution on sheet resistance:**

To study the effect of ageing-time of the starting solution on sheet resistance, we have deposited the ZnO : Al film on the glass substrate with different solution ages, which are 1, 2, 3, 5, 6, 9, 11, 15 days in the (starting) parent solution. All the samples are deposited by the same process and condition with same number of coating 16. Fig. 2(a) shows the value of normal and dark sheet resistance of ZnO : Al film deposited at different ageing of starting solution having sample age 50 days. From this, it is seen that dark resistance is higher than normal resistance measured at same time and temperature. It can also be seen that as the ageing-time of solution increases, the dark sheet resistance of ZnO : Al film decreases and attains minimum value at nine-days and after that sheet resistance increases gradually. This is due to the chemical evolution of starting solution with time [22]. Similar observation has also been reported by Guillen-Santiago [22] for Fluorine-doped (F-doped) ZnO films deposited onto glass substrates by the chemical spray technique. They found that the lowest resistivity value obtained for 9 days aged methanolic-solution.

We have also measured the carrier concentration of ZnO : Al thin film with different solution age having sample age 50 days and number of coating 16.

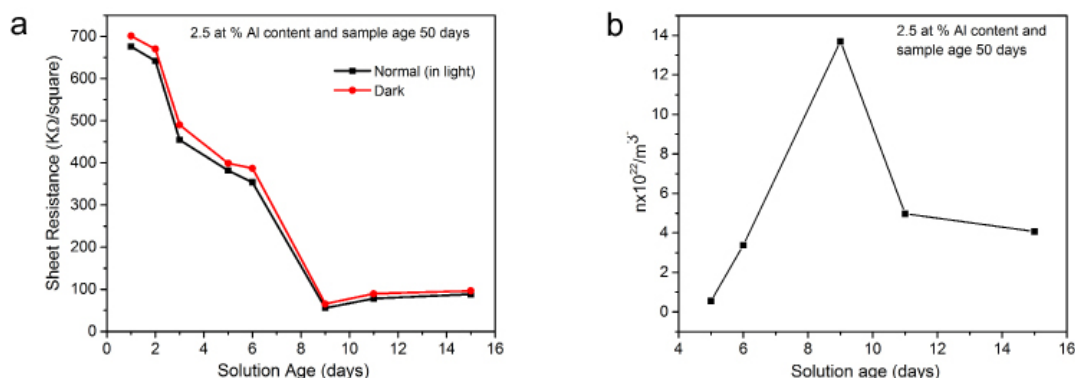


Fig. 2 (a) Effect of ageing time of starting solution on electrical sheet resistance of ZnO:Al thin films, (b) Effect of ageing time of starting solution on carrier concentrations of ZnO:Al thin films

The thickness of the films in this case was found to be 201 nm. Fig. 2(b) shows that as solution aging increases, the carrier concentration of ZnO : Al film increases up to 9 days aged solution and above this limit it starts to decrease. Therefore the sheet resistance of ZnO : Al film decreases as the ageing-time of solution increases and attains minimum value at 9 days and after that sheet resistance increase gradually.

**Effect of sample age on sheet resistance:** To study the effect of sample age on its sheet resistance, we have taken 2.5 at % aluminum content ZnO film prepared with 15 days aged solution with number of coating 16. The measured sheet resistance at different time interval is shown in Fig. 3.

From figure it can be clearly seen that the sheet resistance of the sample is lowered faster with increasing the ageing time of the sample within 60 days and then becomes almost approximately constant. Similar observation has also reported by Bandyopadhyay [21] for Al-doped ZnO films deposited onto glass substrates by the dip coating technique using zinc acetate-2-hydrate and dehydrated isopropyl alcohol to prepare solution with concentration of 0.6 M. According to them, the observed change in resistance of sample with time is due to some surface related phenomenon. It might be due to desorption of the surface H<sub>2</sub>O or negatively charged O<sub>2</sub> on the surface.

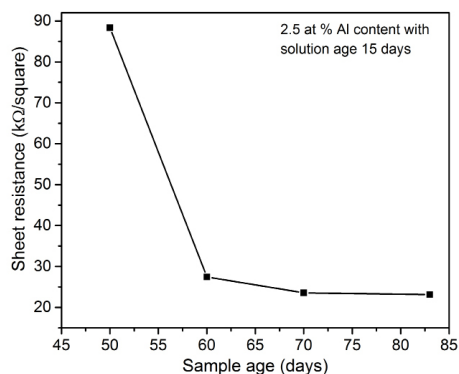


Fig. 3. Effect of sample ageing on electrical sheet resistance of ZnO : Al thin films

**Optical properties:** To study the optical properties of both undoped and aluminum doped ZnO thin films, the optical band gap obtained from measured transmittance has been determined by using the Tauc relation [27]

$$\alpha = A (h\nu - E_g)^{1/2}/h\nu, \quad (4)$$

Where  $A = \ln(100/T)$  is called optical absorbance.

By plotting  $(\alpha h\nu)^2$  versus  $h\nu$ , we found the possible transition occurring and corresponding energy gap for the material.

Fig. 4(a) and (b) shows the variation of optical transmittance and absorbance with wavelength for ZnO :Al film prepared by dip coating technique, with 50

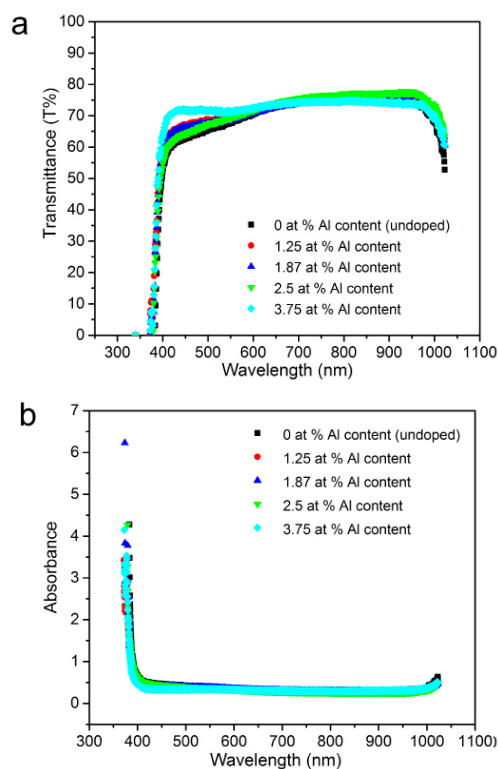


Fig. 4. (a) Transmission spectra of both undoped and Al-doped ZnO thin films, (b) Absorption spectra of both undoped and Al-doped ZnO thin films.

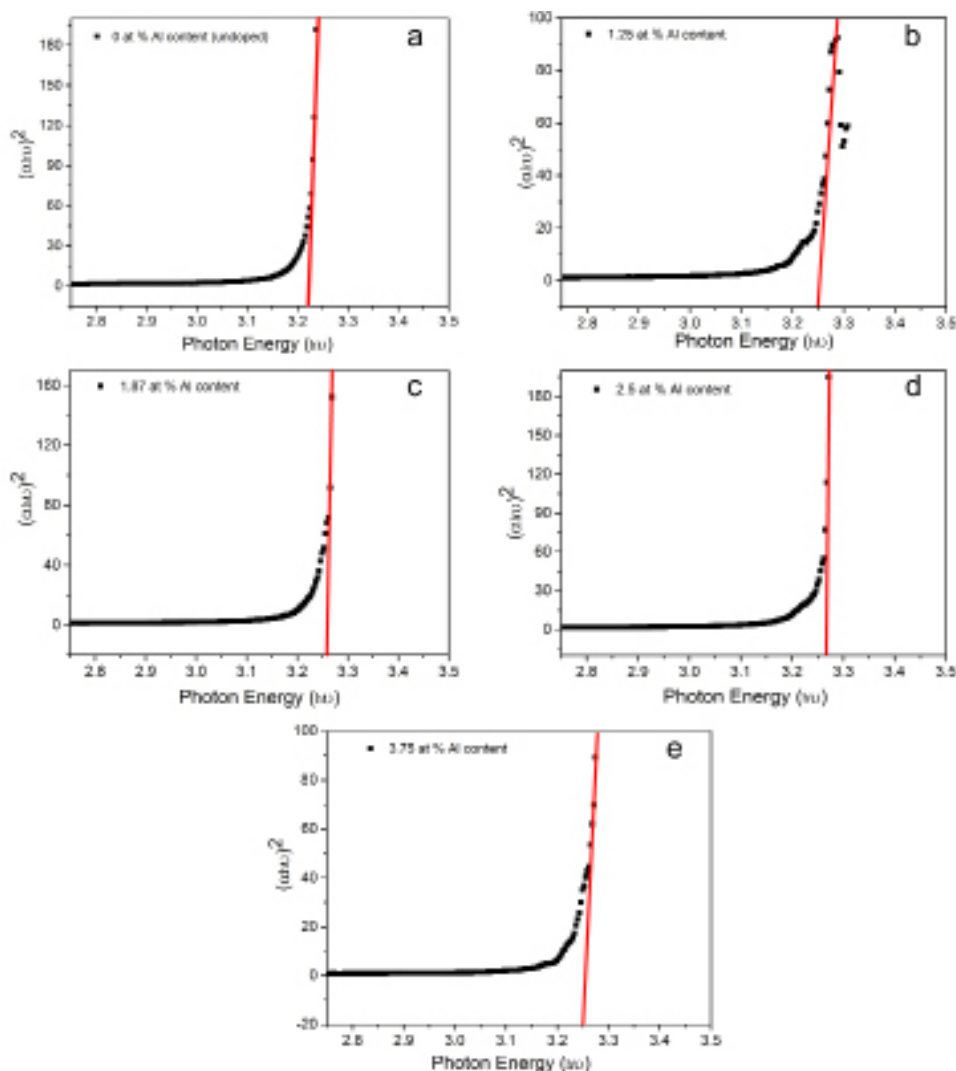


Fig. 5(a) Band gap of the undoped ZnO thin film, (b) Band gap of Al-doped ZnO thin film deposited at 1.25 at %, (c) at 1.875 at %, (d) at 2.5 at %, and (e) at 3.75 at %.

days sample age, 15 days solution age and number of coating 16. The transmittances of films are observed to be in between 65 to 79% in the wavelength range 367 to 1038 nm. We observed superimposed optical interference in the transmittance spectra, indicating good homogeneity of the film. Fig. 4(b) shows that the sharp cut off is around wavelength 385 nm for ZnO : Al and undoped samples. In this case, absorption edges shift to smaller wavelength values when aluminum doping takes place suggesting an increasing in band gap [24]. Fig. 5(a) shows the band gap of undoped ZnO thin film and Figs. 5(b), (c), (d) and (e) show the band gap of the ZnO thin films with Al content at 1.25, 1.875, 2.5, 3.75 at % respectively. Table 1 shows the band gap energy of undoped ZnO and Al doped ZnO at different Al-content on ZnO.

From Table 1, it is seen that band gap of ZnO : Al film is greater than ZnO film deposited in the same time and temperature. The optical band gaps of deposited films are observed to change very slightly with the Al-concentration. The increase in band gap of ZnO : Al films may be attributed to the Burstein-Moss effect

Table 1. Band gap of ZnO : Al films deposit with different Al-content

| Doping concentration, (Al/ZnO) at % | Band gap (eV) |
|-------------------------------------|---------------|
| 0.00                                | 3.22          |
| 1.25                                | 3.25          |
| 1.87                                | 3.26          |
| 2.50                                | 3.27          |
| 3.75                                | 3.25          |

(B-M effect). The band gap can be expressed as:  $E_g = E_{g0} + \Delta E^{B-M}$ , where,  $E_{g0}$  is the intrinsic energy gap of material and  $\Delta E^{B-M}$  is the B-M shift [28] due to filling of lying levels in the conduction band. Since the B-M shift (energy shift) is directly proportional to  $n^{2/3}$ , where  $n$  is the number of the carrier concentration. The enhancement of band gap ensures that aluminum was successfully doped in the ZnO thin films. It is further observed that a decrease in band gap occurs for 3.75 at % Al-doped ZnO film. Similar observation has also been reported by Mondal [29] for Al-doped ZnO film

deposited on glass substrates by successive ion layer adsorption and reaction. They found that a decrease in band gap occurs for 2% Al-doped ZnO film.

## CONCLUSIONS

Transparent and conductive ZnO : Al and undoped ZnO thin films on glass substrates were prepared by sol-gel method using dip-coating technique. The effects of different aluminum concentration, ageing-time of starting solution and sample ageing on electrical sheet resistance and optical properties of the films were presented in this work. It was found that as aluminum concentration is increased, the value of sheet resistance decreases and becomes minimum at 2.5 at % but on further increasing doping concentration sheet resistance starts to increase. The variation of sheet resistance with Al doping shows similar trend irrespective to the solution aging and sample aging. However, sheet resistance changes drastically with sample ageing for first few days and then it becomes almost constant. As the doping concentration is increased, the value of carrier concentration also increases and becomes maximum at 2.5 at % of Al doping but on further increasing doping concentration decreases the value of carrier concentrations. As the aging of parent solution increases, the sheet resistance of thin film decreases gradually and becomes minimum at 9 days and after that it starts to increase. On the other hand, carrier concentration of thin film increases until aging of solution reaches up to 9 days and after it starts to decrease. All the doped and undoped thin films are found to be highly transparent with transmittance lying in between 65% to 79% in the wavelength range 367 to 1038 nm and an increase in aluminum concentration caused the band gap to become broader.

## AUTHOR CONTRIBUTIONS

\*R.P.Yadav and K.B.Rai contributed equally to this work.

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