

# Study of Transparent Conducting Oxide (TCO) Based thin Films Grown by Different Growth Techniques; Review

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Submitted: 08 Aug 2022; Accepted: 19 Aug 2022; Published: 29 Aug 2022

**Citation:** Emmanuel Ifeanyi Ugwu, Usman Rilwan, Ibrahim Abdullah Danasabe, Rafiu Adewale Busari .(2022). Study of Transparent Conducting Oxide (TCO) Based thin Films Grown by Different Growth Techniques; Review. *J App Mat Sci & Engg Res*, 6(2), 14-24.

## Abstract

Various Transparent Conducting Oxide based (TCO) Thin Films have been Developed using different deposition techniques such as SILAR, RF, Chemical Bath, DC magnetron sputtering, chemical organic vapor deposition, etc with their properties well analyzed and each found to have unique properties peculiar in accordance with its mechanism. In this work, we intend to ascertain the influence of the deposition mechanisms on some of the relevant properties of the Oxide based TCO thin films such as CdO, SnO<sub>2</sub>, ZnO etc so that one can be able to infer the growth technique that would give the optimum properties such as the structural, optical, solid state properties that would make the thin films well amenable to appreciable high quality applications in field of opto-electronic and other solid state electronic devices. However from this review, it has been observed that no deposition technique has shown clear cut different characteristics of the properties of this thin film. Therefore it is assumed that all the deposition method is unique on its own.

**Keywords;** Review, Oxide Thin film, Deposition Techniques, Srstructural and Optical Properties

## Introduction

A review on the influence of deposition growth mechanism on ZnO thin film has been looked at with a focus on the structural and optical properties [1]. It is well known that ZnO thin film is one of the oxides based thin films that belong to group III-IV type of semiconductors that are known for high technological applications in the field of optoelectronics and other solid-state devices [2, 3]. And is based on these reasons that many researchers have developed interest on them coupled that they are well known to be highly transparent conducting oxide (TCO) thin film that are amenable for use as transparent electrodes in solar cells and other electronic devices [4, 5]. In fact, it has been made clear that in order to realize the applicability of modern electronic and to make such realizable, these transparent electrodes have been found to be first of all meet the primary requirement be fulfilled by TCO thin films based on their unusual unique properties of high visible wavelength transparency and metal like conductive characteristics [6]. However apart from the aforementioned unique properties possessed by them, they can be tailored into n-type semiconductor materials by doping them with other elements that may enhance the already possessed property

of visible transparency and conductivity to higher percentage [7]. These materials however have been discovered to have their several significant problems due to difficulties encountered especially during the growth of the thin films and achieving a large area deposition of it on a surface for their applications which is particularly due to the scarcity and high cost of the materials in growing them. They have again problem of significant optical absorption in blue- green region, chemical and temperature instability coupled with their likelihood of fracturing on flexible substrates.

However, these problems and challenges posed by scientists of these thin films in the synthetic chemists in particular during the growth are highlighted here to provide an incentives to others in order to spur them into action of working in that area to be cautious and to widen their horizon in their quest to the way of overcoming these challenges and focus on improving the properties of these thin films in other to optimize their applicability either by varying the parameters used for their deposition such as lingands or by annealing them at various temperature or doping or by exploring a better result yielding deposition

methods or conditions of preparation. For instant, various deposition technique techniques such DC magnetron reactive sputtering Metal-organic Chemical Vapour deposition (MOCVD) Vacuum evaporation, electrochemical Deposition, Pulse Laser Deposition, PLD, Spray Pyrolysis Electron beam evaporation (Ali 2009; Mwathe, 2015) [8-14]. Chemical Vapor deposition and Chemical Bath Deposition .sol-gel [16-20]. Ionic Layer Adsorption and Reaction (SILAR) and Atomic Layer Deposition have been used to prepare some of the oxide based thin films especially CdO SnO<sub>2</sub>, ZnO, TiO<sub>2</sub> etc. And there has not been any report of a better one among these deposition techniques [21-23]. There must have an expectation that out of one of these techniques one may have advantage over others in depositing a particular one of these oxide based thin films in terms of cost effect, simplicity, reproducibility and optimization of the properties as it is often generally inferred that the precursor, such as anion and cation in one technique may offer a better control over the deposition parameters such as PH, temperature and time of deposition over other, but various deposition methods have been used to grow some of these oxide based thin films without any glaring report on which one is the best technique of depositing the films and more so which one has more influence in optimizing structural and optical properties of these thin film even if different parameters were used. Sequel to this one deemed it necessary to carry out this review on the exploration of the effect of the deposition techniques on the above three enlisted oxide-based films especially as regards their structural, optical, properties and energy band gap of the thin films for three cases such as as-deposited, annealed and doped irrespective other varied deposition parameters used during the process.

### Materials and Methods

In this section, we briefly outline some of these deposition techniques that have been employed in the synthesis of some of the oxide based thin films that are being considered here briefly, and the next after this consideration is the presentation of the results, featuring properties of the thin films produced based on the deposition mechanisms used.

#### Cadmium Sulphide Thin Film by SILAR Method

The CdO thin film was grown by SILAR growth technique. First the glass substrate was prepared by washing and cleaning it with hot chromic acid after which it was rinsed in distilled water and again cleaned with alkali and acetone. The mechanism used here is a double dip technique with two- step modified SILAR involving cadmium acetate dihydrate with varied molarities at the temperature of 90° C under using ammonium hydroxide as a complexing agent that made the medium to have pH value of (7.0±0.2). 20 dip cycles technique was used after which the film deposited on the substrate and dried in the air and subsequently annealed at temperature of 200° C [24]

#### Deposition of CdO Thin Film by RF Power Technique

In this case for the same CdO, the deposition technique used is radio frequency power mechanism in which the thin film was deposited on a well cleaned microscopy glass substrate by RF magnetron sputtering in a pure argon atmospheric condition with ambient temperature using a well-blended CdO, Sm<sub>2</sub>O<sub>3</sub> alloy target in polyvinyl alcohol (PVA) and then well prepared

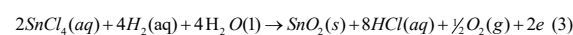
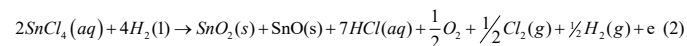
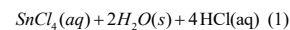
such that it could be used as target material for RF sputtering system as required. The prepared target (cathode) was placed inside the highly evacuated sputtering chamber and well arranged to ensure optimum deposition of the CdO on the prepared substrate. During the process, the temperature of the chamber was varied with the adjustment of RF power from 100W to 250W in steps of 50W allowing the deposition time up to 30 minutes [25].

#### Deposition of SnO<sub>2</sub> Thin Film by SILAR Technique

The microscopic glass slides used for the deposition were first of all prepared by soaking them into soap solution, washed and rinsed using distilled water, after which they were left to dry and stored in desiccation for some time ready for use.

The as-deposited thin films were carried out by using precursor solution consisting of Tin (IV) chloride prepared along with stannic chloride, after which palladium chloride solution was included in conjunction with lead chloride and ammonium fluoride for which the concentration of stannic chloride was varied from 0-22.74 % in order to ensure realization of F:SnO<sub>2</sub> thin film to be at correct ratio as required for optimum deposition [26-30]. In these processes, the prepared translucent precursor as contained in the ionized atoms were proportionally predetermined [31]. The report showed that the deposition underwent three major stages during the course of spray pyrolysis before the final formation and deposition of the thin film on the substrate.

The reaction equation for the entire process are as follows; [32, 33]



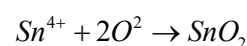
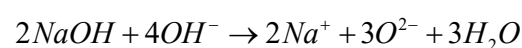
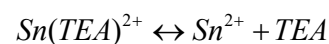
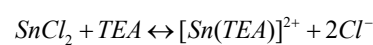
And the deposited films were annealed at the temperature of 450° C [34-37]

#### Deposition of SnO<sub>2</sub> Thin Film by Chemical Bath Technique

The chemical bath system was prepared using SnCl<sub>2</sub>·2H<sub>2</sub>O Sodium hydroxide NaOH with the presence of (TEA or NH<sub>3</sub>) as a complexing agent in which the mix were carried out in a well ordered ratio and subsequently stirred magnetically before the deposition proper.

The glass substrates used were prepared ultrasonically by first of all cleaning them with acetone, washed ethanol and then rinsed with de-ionized water after which they were allowed to dry in the air before insertion inside beaker containing the solution which was at temperature of 60° for deposition process [38].

The reaction equations are as follows;



After the deposition, the deposited thin films were suspended in the air for some time to dry before the samples were subjected to annealing temperature ranging from 100° to 400° .

Just like others, the analysis of the structural, Morphological and optical properties and energy band gap using XRD, SEM and Spectral-Photometer or other analytical equipment meant for that process.

### ZnO Thin Films

Various methods have been used to grow ZnO thin film at different time and places such as Chemical bath deposition, Sol-gel growth techniques, radio-frequency magnetron sputtering and Chemical vapour deposition under low vacuum condition, e.t.c. In the case of CBD, different complex agents such as ammonia, hydrazine, ethanolamine, methylamine, triethanolamine etc has been used to deposit the thin film both at room temperature and at annealed temperature ranging from 150°c to 400°c while in the case of Spray pyrolysis using aqueous methanolic solution zinc acetate as spraying solution has been used to develop the thin film apart from the use of ultrasonic spray pyrolysis technique that was carried out at 200° to 500° by many researchers [17]. Apart from atomic Layer deposition technique that has been reported a good candidate for the growth of high performance n-type growth of ZnO at low temperature Sol-gel technique has not been has been employed using  $Zn(CH_3COO)_2 \cdot 2H_2O$  as a starting material to prepare an acetone gas sensor for use for the growth of the thin film temperature ranging between 700°c to 800°c. ZnO thin film has been doped with different elements such copper, Tellurium Aluminum etc using different growth techniques such as Sol-gel, Magnetron sputtering, CBD, Spray CVD etc with the analysis of morphological structures for both as-deposited, annealed and doped ZnO based thin films using different machines carried out.

### Results/Discussion

In this section, we present the results obtained when some of these deposition technique as outlined in the literature in reviewed are presented as given below according to the growth mechanism of the thin films for which the major properties in the focus were structural and optical properties as obtained from the XRD and Spectral photometer of different models.

Subsequently, the results are presented in accordance with the outlined thin films and the growth technique.

### Ultrasonic Spray Deposited $SnO_2$

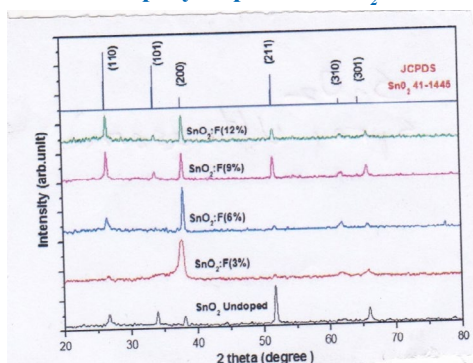


Figure 1: XRD of  $SnO_2$  Thin film grown by Spray ultrasonic

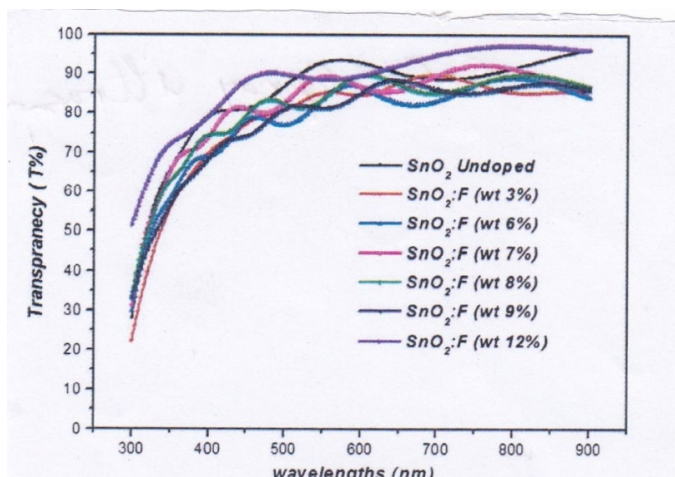


Figure 2; Transmittance of  $SnO_2$  Thin film grown by Spray ultrasonic

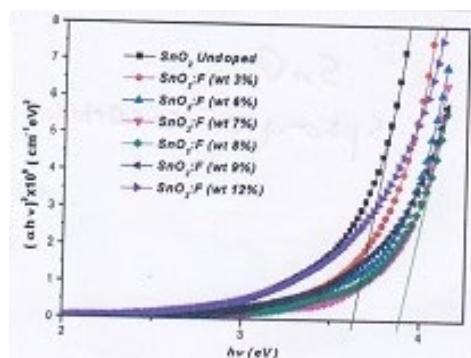


Figure 3; Graph of  $(ahv)^2$  as a function Photon Energy of  $SnO_2$  Thin film grown by Spray ultrasonic

For  $SnO_2$  thin films grown using Ultrasonic Spray, The XRD analysis for both as-deposited, doped were presented as in fig.1 with the diffraction pattern observed to peak at the plane and angle as shown in table 1.

Figure 1: XRD angle and plane of the preferred orientation where the peaks occurred for  $SnO_2$  thin film deposited by Spray Ultrasonic

$2\theta^\circ$	26.68	33.97	37.80	51.79	61.72	61.72
Planes	(110)	(101)	(200)	(211)	(310)	(301)

The optical Transmittance and band gap were as presented in fig- and fig----, respectively as a function of wavelength where it could be easily noticed that as –deposited  $SnO_2$  thin film transmittance spectra have a high transmittance within the visible region with average transmittance range of 92% coupled with a distinct exhibition of oscillatory characteristics within the long wavelength, ie within infrared region.

Generally, the transmittance decreased in the visible region for the doped thin films reducing to the average range of 81.3% and then the only suspected situation that might have affected the transmittance is the effect of the doping element that must have



affected the structure of the thin films and for the optical band gap, the estimation was done due to Tuac's relation as in fig.-from where it could be seen that the average energy band gap for both fluorine doped and as-deposited  $\text{SnO}_2$  thin films, ranged within 2.51eV to 3.842eV [39].

### SnO<sub>2</sub> Deposition using Spray Pyrolysis Technique

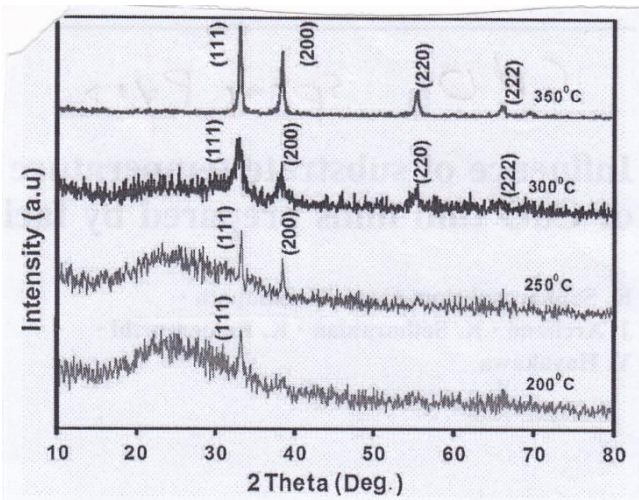


Figure 4; XRD of SnO<sub>2</sub> Thin film grown by Spray ultrasonic

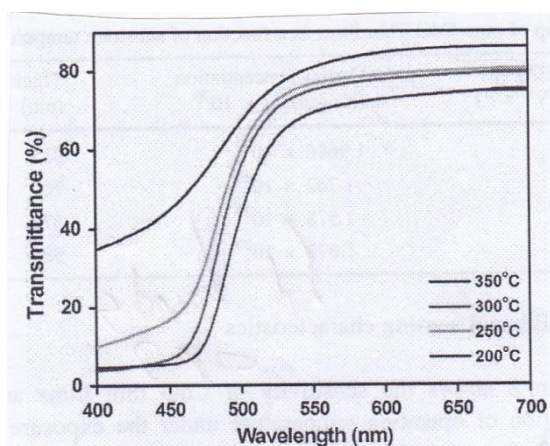


Figure 5; Transmittance of SnO<sub>2</sub> Thin film grown by Spray Pyrolysis

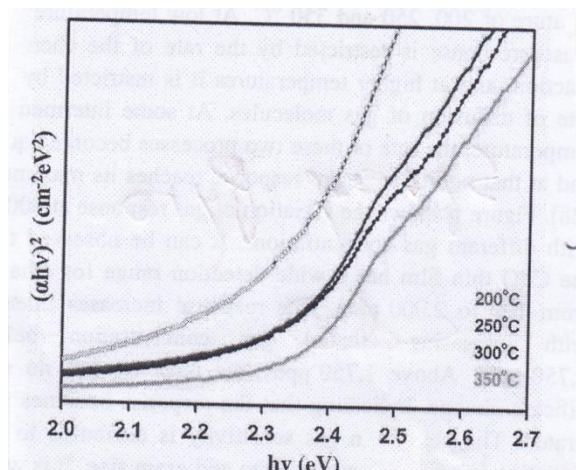


Figure 6; Graph of  $(ahv)^2$  as a function Photon Energy of SnO<sub>2</sub> Thin film grown by Spray Pyrolysis

On the other hand, when it comes to  $\text{SnO}_2$  thin film prepared by Spray Pyrolysis technique, as presented in fig.5, with table2, showing where the peak occurred at the preferential orientation planes and angles as in XRD.

Figure 2: XRD angle and plane of the preferred orientation where the peaks occurred for SnO<sub>2</sub> Thin film deposited by Spry Pyrolysis

$2\theta^\circ$	26.51	38.00	51.96	27.38	38.70
Planes	(110)	(200)	(211)	(110)	(211)

Form the preferential peaks as seen in tables 1 and 2 for these two growth techniques, the pane show that the  $\text{SnO}_2$  thin films are polycrystalline in nature exhibiting tetragonal structure.

For the spectral analysis and the energy band gap, it could be observed that the film depicted high transmittance within the visible region ranging from 81% to 85% with little decrease with the infrared region just like the Spray ultrasonic developed counterpart. The Transmittance and the graph of energy band gap and shown in figures 6 and 7 respectively where the energy band gap was also estimated due to Tuac's relation with the average value 3.97eV and with a close look, the value is closely in line with that obtained in Spray ultrasonic because what it implies is that the energy band gap in all the cases lie between 3.60eV to 4.00eV.

### Sol-gel Deposited SnO<sub>2</sub>

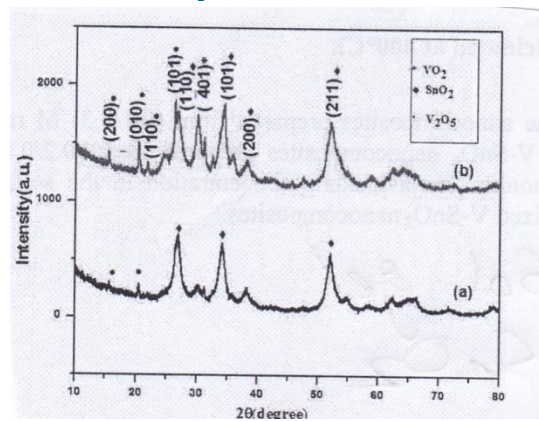


Figure 7; XRD of SnO<sub>2</sub> Thin film as deposited grown by Sol-gel Technique

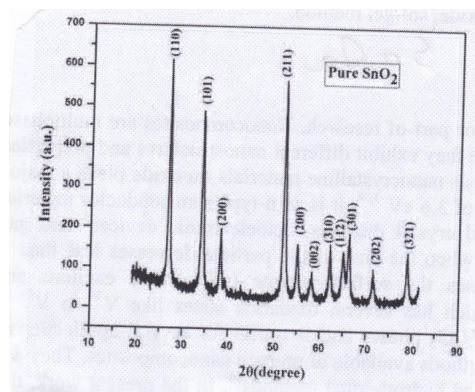


Figure 8; XRD of SnO<sub>2</sub> Annealed Thin film grown by Sol-gel Technique



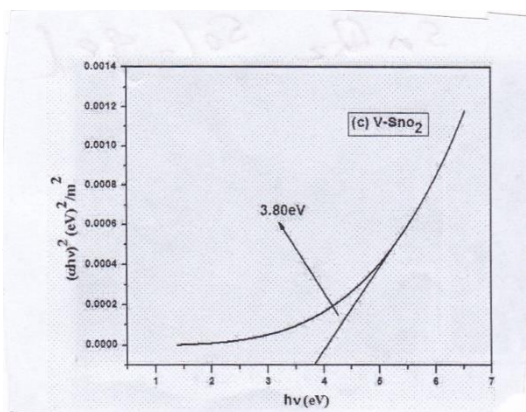


Figure 9; Graph of  $(ahv)^2$  as a function Photon Energy of  $SnO_2$  Thin film grown by sol-gel

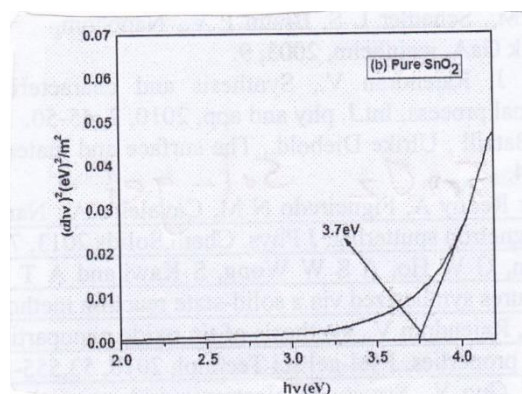


Figure 10; Graph of  $(ahv)^2$  as a function Photon Energy of  $SnO_2$  Thin film grown by sol-gel

The Sol-Gel deposited  $SnO_2$  thin film XRD pattern is presented in fig.7 and fig.8 the with the preferential orientation based on XRD analysis found to peak at the plane as in table 3.

Table 3: XRD angle and plane of the preferred orientation where the peaks occurred for  $SnO_2$  Thin film deposited by sol-gel technique

$2\theta^\circ$	27.50	34.50	38.50	64.50	65.00	72.00	79.50
Plane	(110)	(101)	(200)	(211)	(310)	(202)	(321)

The peaks as observed here when compared with the JCPDS file (CardNo41-1445) reflects the tetragonal structure nature of  $SnO_2$  and this is in absolute agreement the result of the two previous techniques already discussed. The Tauc graphs for the deduction of band gap in which Tauc's method was also used to estimate ranged between were shown in figure10 and fig.11 respectively for as-deposited and annealed csae from where it was seen that the band gap ranged from 3.70eV to 3.80eV for both doped and as-deposited with just a slight increase when it was doped or annealed culminating with our earlier assertion on the range of the energy band gap of  $SnO_2$  thin films following the observation from the other first two methods of deposition techniques already presented.

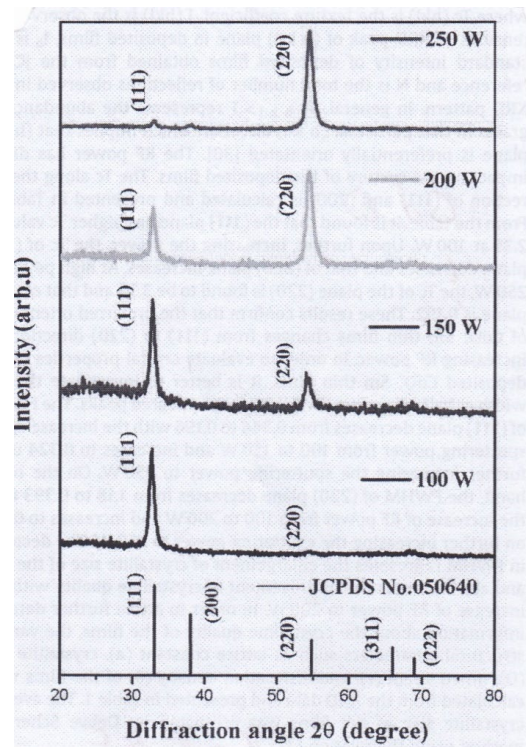


Figure 11; XRD of CdO Thin film deposited by radio Frequency Power Sputtering Technique

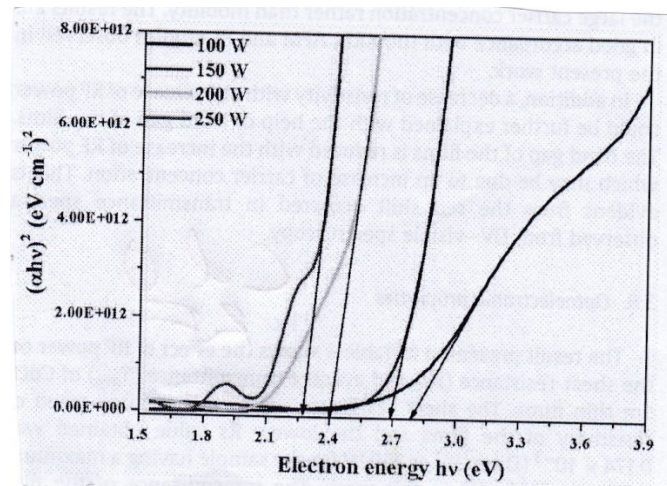


Figure 12; Graph of  $(ahv)^2$  as a function Photon Energy of deposited by radio Frequency Power Sputtering for CdO Technique

The XRD pattern of CdO thin film deposited by radiofrequency power sputtering is presented in fig.12 while the preferential orientation plane and the angle where they occur are at table 3.

Table 4: The XRD diffraction angle and plane of the preferred orientation showing where the peaks occurred as seen for CdO using this deposition technique are as in the table5 while the graph for band gap deduction is in fig.13.

**Table5; XRD Pattern for RF power deposited CdO Thin film.**

$2\theta^\circ$	33.75	39.50	55.50	69.00	69.00
Plane	(111)	(200)	(220)	(311)	(222)

From the table5 above, it is seen that the peak at the preferred plane of orientation (111) decreased with increase in RF power and doping followed with increase in in the peak at the plane at (220) with other peak at the plane of preferred orientation (200), (311) and (222) become extinct with increase in RF power while the location remains the same.

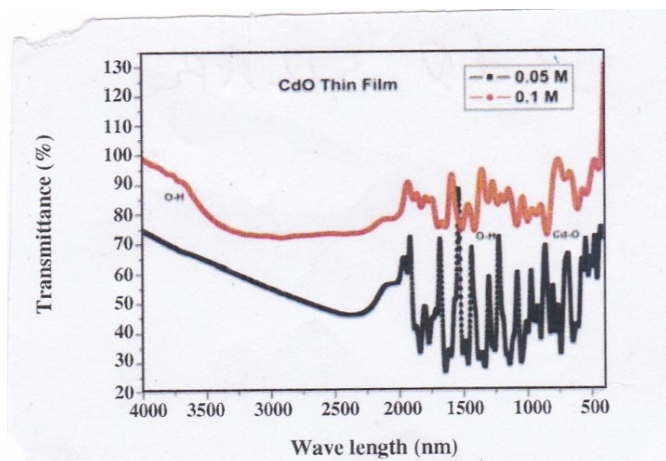
Significantly, with two prominent peaks cited at the preferred plane of orientation (111) and (200), it simply points out that CdO thin films structurally exhibits a polycrystalline nature that appears as a cubic lattice structure with clear observation that as RF power increases, there was a shift of the plane to smaller angle due to lattice expansion coupled with a decrease in the plane intensity without a noticeable change in location of the diffraction peaks.

This serves as confirmation of the report given by that DC magnetron sputtered CdO thin films changes there preferred orientation from (111) to (200) with increase in DC power [40]. The energy band gap of the thin film is as shown the figure ranged from 1.8eV to 2.8eV.

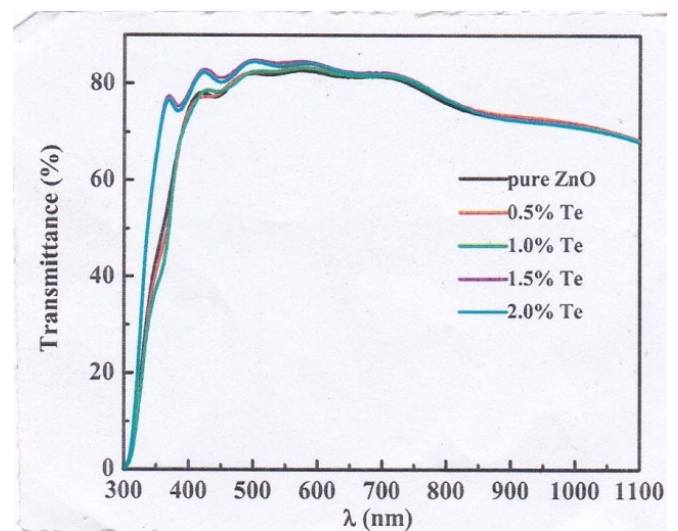
The transmittance of CdO is notable in its characteristic of having low value within the UV and visible region while the value increased within the long wavelength region that is within the infrared region which goes further to illustrate that CdO has strong intense emissive power spectra within the UV and visible regions corresponding to blue and green emission.

On the other hand, the energy band gap was estimated using Tauc's plot as in fig.12 show that in this particular case the RF power used during the deposition were indicated in each case and was found to have influenced the band gap, though in all the cases, the energy band gap value lied between 2.26eV to 2.78eV with a clear indication that there was a decrease in the band gap with increase in RF power.

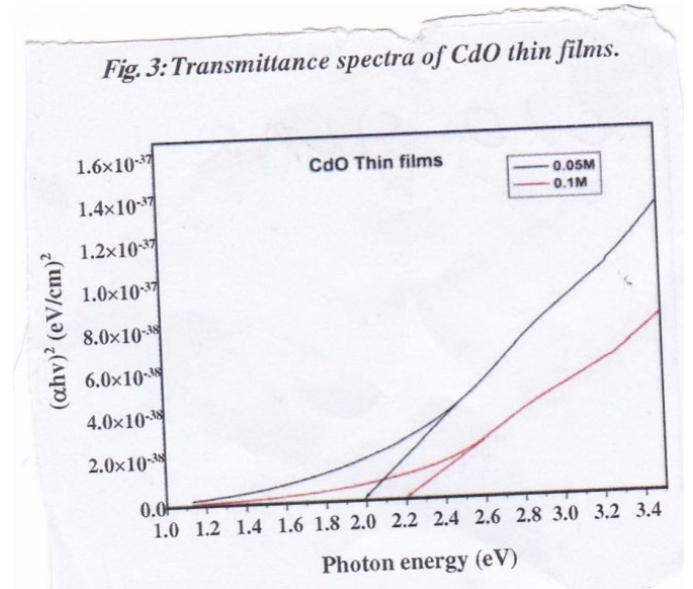
**SILAR Deposited CdO**



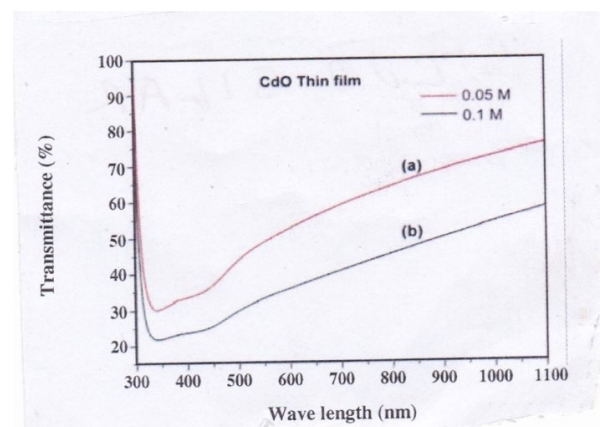
**Figure 14;** Transmittance of CdO Thin film deposited by SILAR Technique



**Figure 15;** Transmittance of CdO Thin film deposited by SILAR Technique



**Figure 16;** Graph of  $(ahv)^2$  as a function Photon Energy of deposited by SILAR Technique for CdO



**Figure 17;** Transmittance of CdO Thin film deposited by SILAR Technique

For Spray pyrolysis deposited CdO thin films, the XRD pattern is showcased in fig.----- and table 4.



**Table5: XRD angle and plane of the preferred orientation where the peaks occurred for CdO Thin film deposited by Spray Pyrolysis**

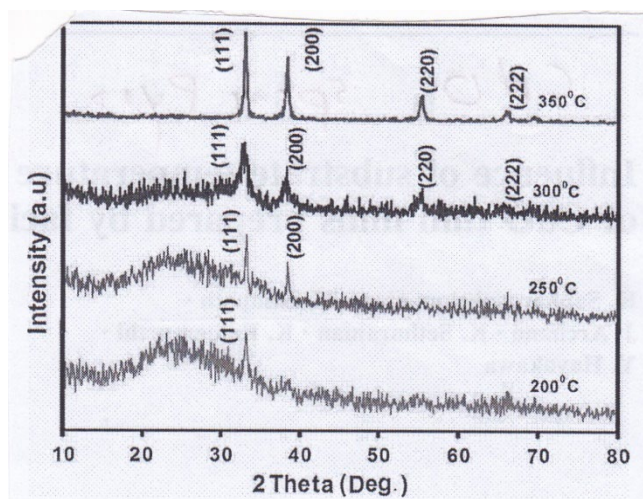
$2\theta^\circ$	33.00	38.50	55.00	64.50	69.00
Plane	(111)	(200)	(220)	(222)	(222)

The table4 above indicates that CdO developed by this technique has strong orientation along (111) and (200) direction with increase in the intensity of the peaks with increase in the temperature of the substrate. It was significantly noticed that the diffraction angle of (111) shifts slightly to lower angle as there was lattice shrinkage and from the analysis based on this XRD pattern, the thin film appear to be polycrystalline in nature just like that developed using Rf power and thus it exhibits cubic lattice structure when compared with JCPDS file no. 75-0591.

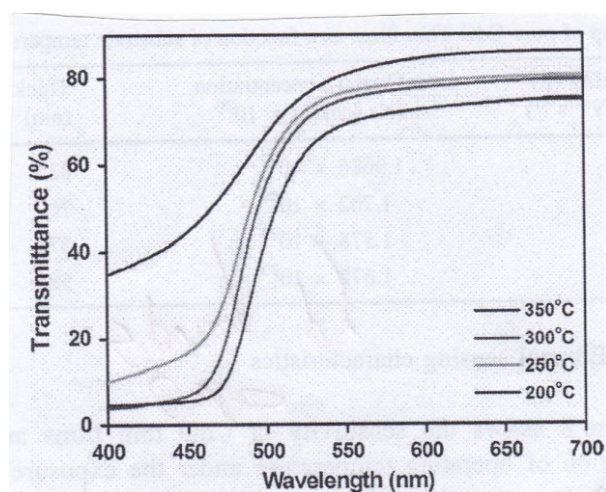
The spectral transmittance is shown in fig. 17 and the energy band is showcased in fig.16. Here, it was also observed that the transparency is lower within the UV and visible region of the spectra with increase as it moves from visible to infrared region.

The band gap was estimated due to Tauc's plot from where it was observed that the band gap ranged between 2.24eV to 2.34eV depending on the substrate temperature, and point out that the band gap is generally narrow lying just between 2.0 eV and 2.3 eV and it is found to have direct band gap whereby culminating the result of the band gap obtained from the other method of deposition of CdO discussed here.

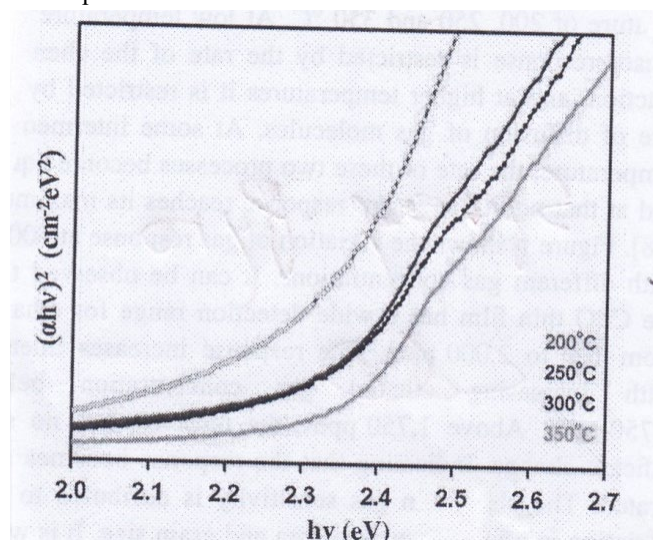
### 3.6 Deposition CdO Thin Films by Radio Frequency deposition technique



**Figure 18;** XRD of CdO Thin film deposited by RF technique



**Figure 19;** Transmittance of CdO Thin film grown by RF technique



**Figure 20;** Graph of  $(\alpha hv)^2$  as a function Photon Energy of deposited by RF Technique for CdO

The XRD pattern of CdO deposited using RF deposition technique as shown in fig.18 and table6

**Table 6; XRD angle and plane of the preferred orientation where the peaks occurred for CdO Thin film deposited by SILAR Technique**

$2\theta^\circ$	32.50	39.00	69.00
Plane	(110)	(200)	(310)

The intensity of the peaks occurred as indicated in the table5 which could be indexed to cubic structure since these three major peaks are associated with polycrystalline material having cubic structure which is in agreement with standard data for CdO thin film for (JCPDS) data file (No. 78-0653) because from this study it is clear that the thin film exhibited a preferential orientation along with the (111) diffraction plane.

The optical spectral Transmittance and the energy band gap are as shown in the fig.19 and fig.20 respectively from where it



was glaring that it has low spectral transmittance within the UV and Visible regions just like the other two deposition technique already discussed. The varied from 55% to 80% within the range of wavelength 350nm to 1100nm which implies that the transmittance is higher in the infrared region. As for the energy band gap, the same as in the other case was used to deduce the band gap was found be between 2.17eV and 2.21 eV which appear to be narrow also as observed in the other two methods of CdO deposition technique.

### Atomic Layer Deposited of ZnO Thin films

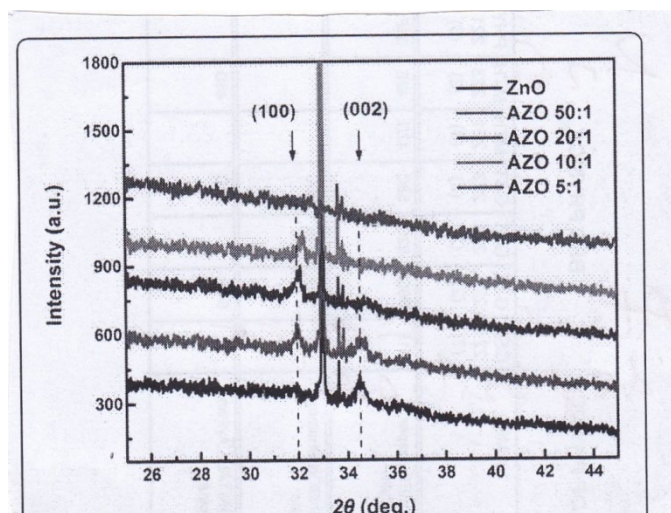


Figure 21; XRD of ZnO Thin film grown by Atomic Layer Technique both for as- deposited and Doped

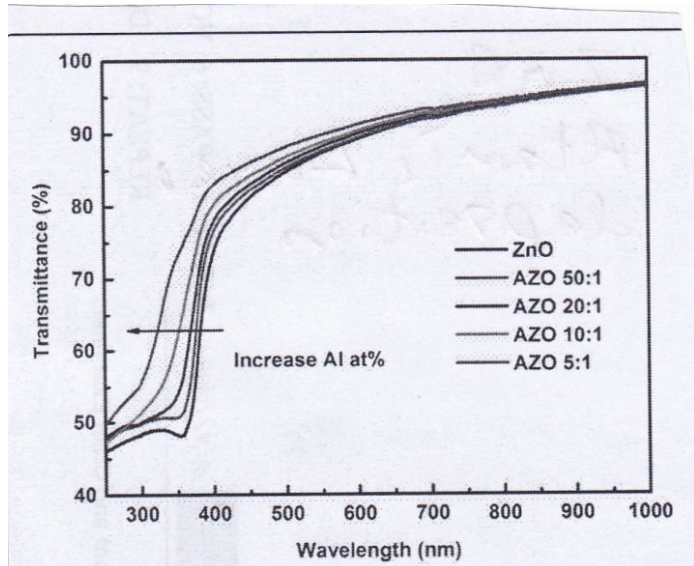


Figure 22; Transmittance of ZnO Thin film grown by Atomic Layer Technique

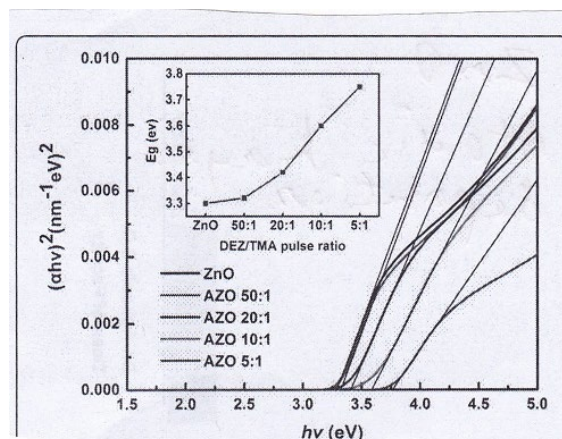


Figure 23; Graph of  $(ahv)^2$  as a function Photon Energy of deposited by Atomic Layer Technique for ZnO Thin film

Table 7; XRD pattern Atomic layer deposited ZnO based thin films is presented in fig21 and table is as in table7

Table7;.XRD pattern of ZnO deposited using Atomic Layer technique

$2\theta^\circ$	33.00	35.00	37.50	48.00	57.00	64.00	68.50
Plane	(100)	(002)	(101)	(102)	(110)	(103)	(112)

From the pattern, it was revealed that the samples appear as polycrystalline in nature and exhibits single-phase ZnO hexagonal Wurtzite structure [JCPDS (36-1451)] as indicated in peaks recorded as shown in the table 6. The optical transmittance is shown in fig.22 while the energy band gap graph is as in fig.23 In this case, it could be observed that there is increase in transmittance as doping concentration is increased for all the element used for doping but however it depicted a trend of low transmittance within the UV region while it increased within the visible to near infrared region.

### 3.8; Chemical Bath Deposited ZnO Thin Film

From the XRD pattern as indicated here, it was obviously shown that the preferred orientation in ZnO thin film lies with 30 degree and 35 degree and is given as (100) and (002)

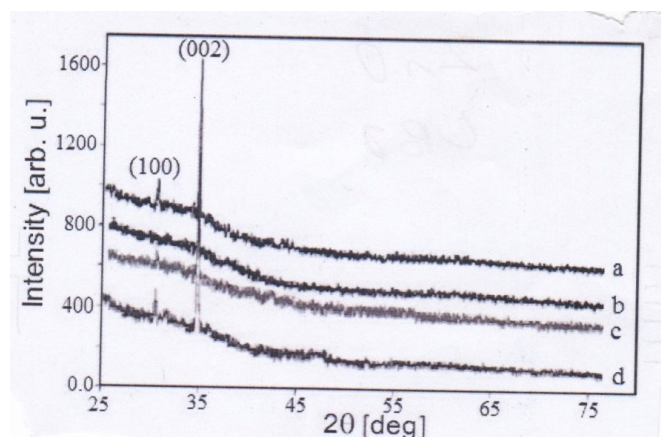


Figure 24; XRD of ZnO Thin film grown by Chemical Bath,



both for as- deposited and Doped

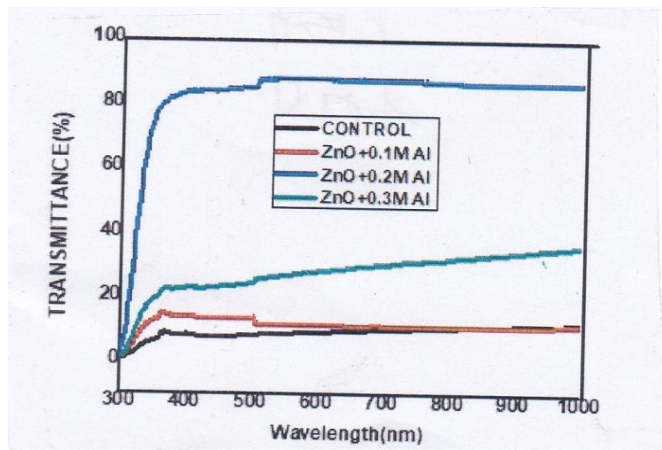


Figure 25; Transmittance of ZnO Thin film grown by Chemical Bath Technique

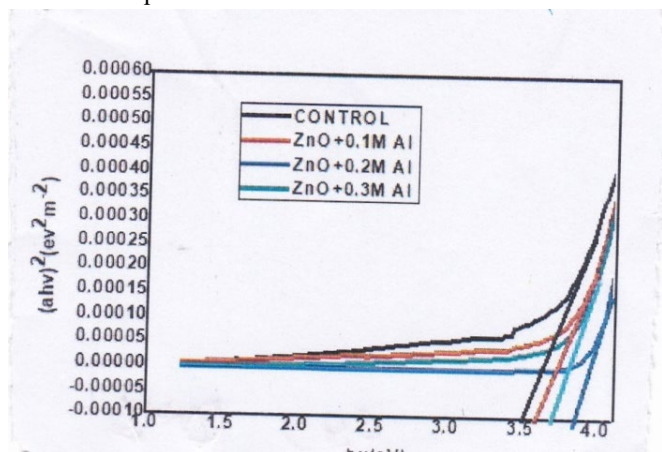


Figure 26; Graph of  $(ah\nu)^2$  as a function Photon Energy of deposited by Chemical Bath for ZnO Thin film

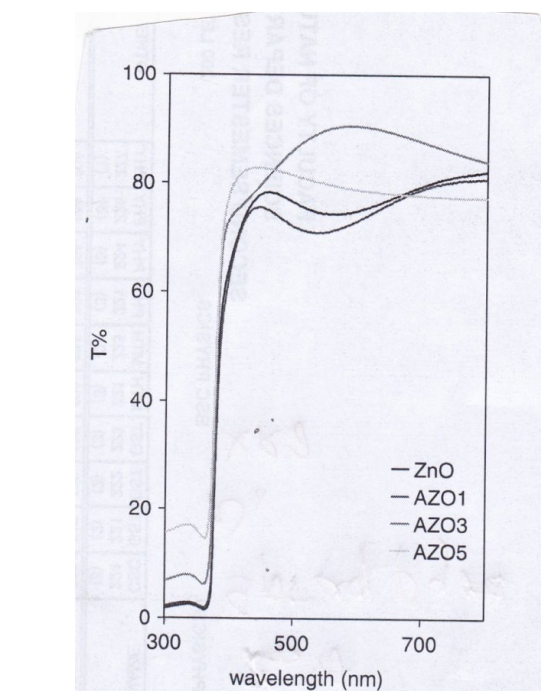


Figure 27; Transmittance of ZnO Thin film grown by Spray

Pyrolysis

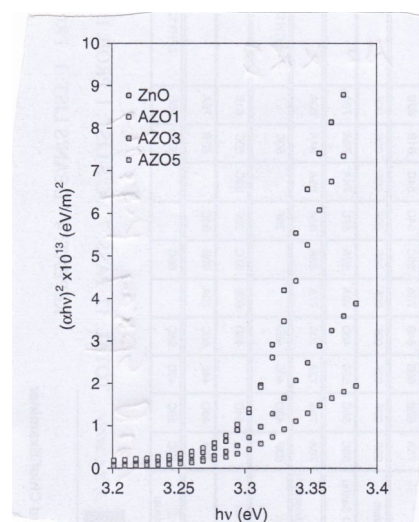


Figure 28; Graph of  $(ah\nu)^2$  as a function Photon Energy of

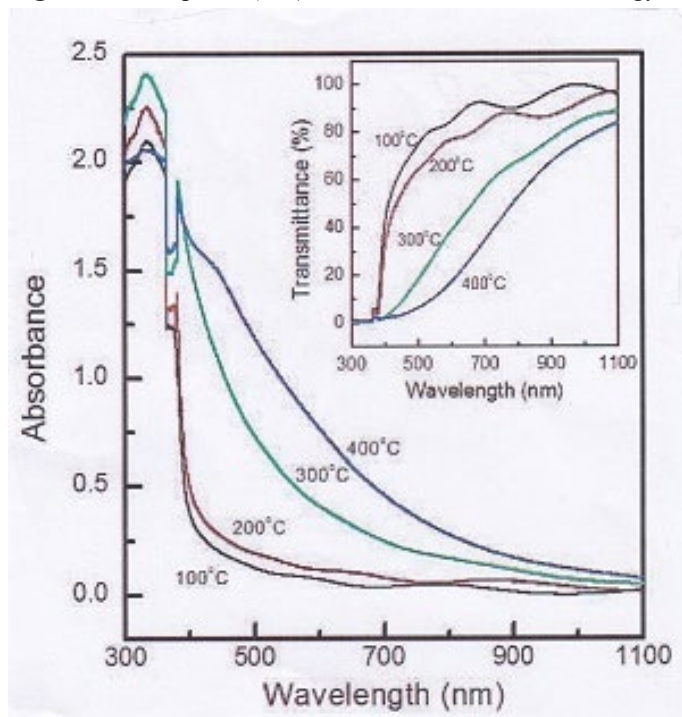


Figure 29; Transmittance of ZnO Thin film grown by Atomic Layer Technique

The diffraction pattern for ZnO grown using chemical spray pyrolysis deposition technique is depicted table 7

Table.7 XRD Pattern of ZnO Thin film

$2\theta^\circ$	32.00	35.00	38.00	48.00	59.00	63.00	67.00
Plane	(100)	(002)	(101)	(102)	(110)	(103)	(200)

also here clearly revealed that both as-deposited and Al-doped ZnO are indexed to the hexagonal crystal structure in accordance with (JCPDS) card file no. 36-1451 also with peak intensity decreasing with increase in Al. doping [41]. For the duo, it must be observed that all the case for both as-deposited and doped

ZnO thin film, there is a manifestation of polycrystalline nature with c-axis having preferential orientation normally to the substrate irrespective of deposition technique and the element used in doping [42, 43].

Coming for the optical properties and energy band gap in all the deposition techniques, it was clearly upheld that ZnO based thin films has low transmittance within the UV and visible region of electromagnetic wave spectra with increase in its transmittance within the near infrared region as in fig25, fig. 27 and fig. 29 The energy band gap appears to lie within the range 3.00eV and 4.00eV all cases as in fig 26 and fig.28 [17].

### Summary

Based on the observation from the review of some of the properties of these oxide based thin films as studied here in this work, it was clearly revealed that the depositions technique did not play any major significant variation on some the properties of the Oxide thin films considered here [44]. The factors that were observed to be paramount in influencing the properties were doping annealing and Concentration of doping elements because it was clearly noticed that these were the factor that contribute in modification of structural properties of the thin films due to the fact that they contribute in re-orientation of the lattice structure of the system which invariably brings about adjustment in XRD diffraction characteristics, spectral transmittance and energy band gap [17, 42, 45].

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