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Review on the effect of preparative parameters of spray pyrolysis technique on optical and electrical properties of Zinc Oxide thin film

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ABSTRACT

Spray pyrolysis technique is one of the most widely used, simple and inexpensive technique to deposit thin films of various materials. The physio-chemical properties of these thin films are depending upon preparative parameters such as concentration of precursor solution, dopants, substrate temperature, spray rate, post annealing treatments, etc. In this paper an intensive review on effect of preparative parameters on optical and electrical properties of ZnO thin films deposited by spray pyrolysis method have been presented. These properties of thin film can be precisely controlled by optimizing the preparative parameters. ZnO due to its attractive optical and electrical properties plays an important role in various electronics, optoelectronics, biomedical and sensing applications.

Keywords: *Spray Pyrolysis Technique, Zno Thin Film, Preparative Parameters, Optical And Electrical Properties*

1. INTRODUCTION

Zinc oxide due to its low cost, abundant availability and non-toxic nature and having verity of nanostructures with enhanced structural, optical and electrical properties the material of interest for many researchers. It is a II-VI compound of n-type semiconductor having wurtzite structure with a direct wide band gap of about 3.7 eV and large binding energy of about 60 meV at room temperature [1]. Due to which efficiently emits light in UV spectral region and effectively used for optoelectronic application[2]. ZnO has excellent optical and electrical properties hence its thin films are widely used in sensor, solar cell, LED and laser systems[3]. Due to highly conductive and transparent employed in transparent electrodes also. Beside that the different nanostructures of zinc oxide are used in verity of applications such as microelectronics, magnetic devices, photocatalysis, photovoltaics, optics, energy storage, thermoelectricity, piezoelectricity, electrochemistry, temperature sensing, etc. Zinc oxide can be synthesized in the form of various nanostructures such as nanotubes, nanowalls, nanorods, nanofibers, nanowires, nanoflakes. The different methods used for this purpose are sol gel technique, spray pyrolysis technique, chemical bath deposition, chemical vapour deposition technique, PVD, electrospinning, hydrothermal, solvothermal, PLD, etc.

The thin film of ZnO nanoparticles were prepared by using PLD method for investigation of structural, electrical and optical properties [4]. By using simple chemical sol gel method is possible to control the particle size and morphology with optimizing the reaction parameters [5]. Zinc oxide nanowires and nanosheets are formed by using PVD method for solar cell application [6]. Nanofibers were formed by using electrospinning method which are useful in various applications such as self-cleaning, gas sensing, water purification, air purification, and hydrogen production by water splitting, etc [7]. Nanorods, nanoflowers and nanospheres were synthesized by hydrothermal method [8]. Chemical bath deposition method was used to form ZnO thin films of different nanostructures such as fibrous nanoflakes, nanobeads, nanoparticles, cactus, nanoneedles and hexagonal nanorods [9]. Spray pyrolysis technique used for deposition of thin films [10]. Laser chemical vapor deposition (LCVD) used for synthesis ZnO nanoparticle array [11]. Molecular beam epitaxy for growth of p-type epitaxy layer [12]. ZnO nanostructures can be synthesized by using physical, chemical or biological route and their physicochemical properties are depends upon the synthesis technique used [13].

In this paper more emphasis is given on spray pyrolysis technique for synthesis of Zinc oxide material and effect of its preparative parameters on electrical and optical properties.

2. SPRAY PYROLYSIS TECHNIQUE

Spray pyrolysis is one of the chemical methods for synthesis of nanomaterials and thin film deposition. Due to its attractive features such as inexpensive, simple, operating at low temperature, flexibility in substrate selection, useful for large area deposition, etc widely used technique [14]. The ultrasonic spray-pyrolysis [15], Chemical spray pyrolysis [16], flame spray pyrolysis [17] these are some techniques used in spray pyrolysis method. Fig 1. shows the basic set up for spray pyrolysis technique.

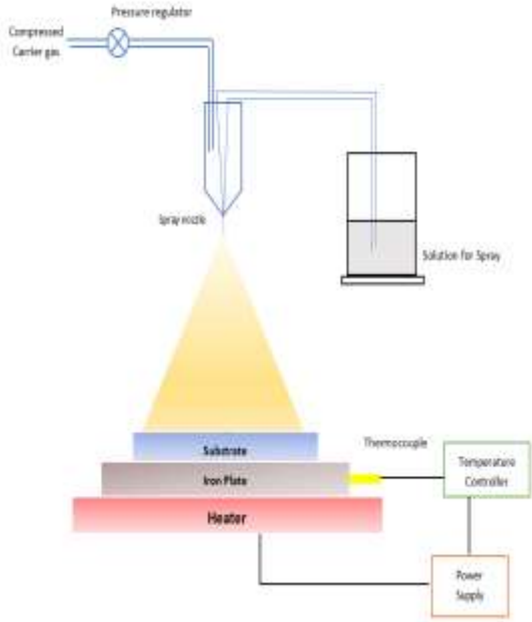

Fig. 1 spray pyrolysis technique

It consists of substrate heater, temperature controller, precursor solution, spray nozzle, carrier gas compressor and regulator system. In Spray pyrolysis technique precursor solution is sprayed on preheated substrate in the form of small droplets (aerosols) through spray nozzle, which further thermally decomposes to form thin film of material with desired properties. In this method solution and carrier gas flow rate are controlled by automizer system. Properties of deposited film are depending upon concentration of precursor solution, spray rate, substrate temperature, carrier gas, distance between spray nozzle and substrate, etc. This technique can provide uniform deposition of thin films. Quality and physicochemical properties of thin films are maintained by controlling these preparative conditions.

Advantages of spray pyrolysis technique:
1) Simple and inexpensive, 2) uniform and high-quality deposition over large surface area, 3) deposition can be takes place at low temperature about 500℃, 4) films are reproducible, 5) by changing the precursor composition layering of films can be easily possible. 6) preparative parameters can be easily controlled and hence the properties of material.

3. EFFECT OF PREPARATIVE PARAMETERS ON PROPERTIES OF ZnO Thin Film

Zinc oxide thin films prepared by spray pyrolysis method and its structural, optical and electrical properties were reported by various researcher. It is observed that film properties are get affected by changing substrate temperature, precursor concentration and dopant used, spray rate, post annealing temperature, etc.

3.1 Effect of substrate temperature

The substrate temperature is main parameter which greatly affects the film morphology, optical and electrical properties. F. Zahedi *et. al.* [18] studied effect of substrate temperature on structural, optical and electrical properties of ZnO thin film. The film was deposited on glass substrate by varying substrate temperature from 200-500℃. Resistivity of film prepared at 400 ℃, 450 ℃ and 500 ℃ was 2.56 Ωcm, 2.86 Ωcm and 2.93 Ωcm and it is too high at 200 - 300℃. It has high optical transmission above 80% in visible region for substrate temperatures above 400℃. Increasing optical band gap from 3.19 to 3.25 eV with increasing substrate temperature from 300-500℃ due to Burstein-Moss effect were reported [19]. B V Rajendra *et. al.* [20] studied effect of substrate temperatures and subsequent annealing on optical properties of the ZnO thin films. It is observed that with increase in temperature from 473 K – 673 K band gap decreases from 3.28 eV to 3.17eV. Optical transmittance increases with increasing substrate temperature and it is about 95% and 95.5% at 673K without and with annealing respectively. Georgi Marinov *et. al.* [21] deposited polycrystalline ZnO thin films on silicon substrates by electrospray method at different substrate temperature. Refractive indexes for films at wavelength of 600 nm, are 1.67 at 150℃, 1.97 at 200℃ and 1.93 at 250℃. It shows that refractive index is very low at lower temperature. Pure ZnO thin films were deposited on glass substrate for different substrate temperatures varying from 573K to 723K. It shows increasing band gap from 3.14 to 3.20 eV due to improved crystallinity with increasing temperature. Average optical transparency for all deposited films were about 80%, highest transmittance is about 95% obtained at 673K but it is slightly decreasing at 723K due to powdery nature of material deposited [22]. Morphological and optical properties were studied for ZnO thin film deposited on glass substrate at different temperature for solar cell application. At 550℃ the ratio of the PL emission intensities in UV and in visible is maximum which indicate that less defects are generated and quality of film increases with increasing temperature [23].

3.2 Effect of solution spray rate:

Solution Spray rate is another one parameter which greatly affect morphological, optical and electrical properties of thin film. It particularly affects grain size and thickness of sample. Merike Kriisa *et. al.*[24] deposited ZnO:In thin film on glass substrate at substrate temperature 400^0C with varying spray rate from 0.5–6.7 mL/min. resistivity of the ZnO:In films decreases from 6.2x 10^{-2} Ωcm to 3.7x10^{-3} Ωcm with increasing spray rate from 0.5 mL/min to 3.3 mL/min, respectively irrespective of film thickness. It is because of carrier mobility increases with increasing spray rate. Yacine Aoun *et.al.*[25] deposited ZnO thin film on glass substrate at 350 ^{0}C with varying spray rate from 10 ml to 35 ml. Optical band gap increases at 3.28eV and 3.279eV for film deposited at 25ml and 35 ml. Average optical transmittance is about 95%. Resistivity of film decreased from 0.394 to 0.266 Ω.cm with increasing spray rate from 20 – 25 ml and then increases upto 0.509 Ω.cm for 35 ml. The film was prepared by using ultrasonic spray pyrolysis technique for spray rate changing from50ml/hr to 150 ml/hr and optimizing other parameters substrate temp at 350 ^{0}C, substrate to nozzle distance at 5 cm. The transmittance of films was about 80% and band gap increases from 3.274 and 3.282 eV with increasing flow rate from 50 to 150 ml/h.[26]. S. Zargou et.al. deposited ZnO thin film on glass substrate by using chemical spray pyrolysis method at 300°C at different spray rates i.e. 5, 10, 15 and 20 ml/h. It is found that electrical resistivity decreases from 4.048×10^3 to 0.781×10^3 Ω.cm respectively with increasing spray rate from 5ml/h to 20 ml/h. The film deposited at spray rate 15 ml/h was highly transparent (99.1%) with maximum dark electrical conductivity hence can be useful in optoelectronic applications [27].

Table 1: The deposition parameters of ZnO thin-film deposited using the spray pyrolysis deposition technique

Sr. No.	Precursors	Deposition parameters					Remark	Ref.
		Substrate temp.	Substrate used	Solution concentration / quantity	Spray rate	Nozzle to substrate distance		
1	aqueous zinc nitrate solutions	500 °C	Sapphire, glass	0.2M	3-5 ml per minute		Resistivity 110 Ωcm, 140 Ωcm for Sapphire and glass respectively. Band gap: 3.27eV and 3.29 eV for Sapphire and glass respectively.	[28]
2	Zinc acetate and Aluminium Pentanedionate (ZnO:Al film)	420 °C	glass	0.2M	2 ml/min	28 cm	Lowest resistivity 2.4 x 10^{-2} Ωcm at 2 at.% doping concentration and then after goes on increasing with increasing doping concentration 6.67 x 10^{-1} Ωcm at 6 at.%. Irrespective of dopping concentration of aluminium transmittance was above 80%. Band gap increases from 3.18eV to 3.35eV with increasing doping concentration from 0 at.% to 10 at.%	[29]
3	zinc acetate dihydrate	573 K	fused silica and Pt		50 ml/hr.	10 cm	Band gap energy 3.2 eV, Electron concentration 5 x 10^{23}m^{-3}	[30]
4	Zinc acetate	360^0 C	glass				Undoped ZnO film has poor conductivity where as vaccum heat treated films are good transparent conductors.	[31]
5	Zinc acetate dehydrate, ammonium acetate (ZnO:N thin films)	450^0 C	glass	0.1 M to 0.4 M		25 cm	Optical transparency decreases in visible region as molar concentration goes on increasing, electrical resistivity decreases with increasing molar concentration, a p-type Hall effect behaviour is observed. Use full for TCO material and CMOS devices.	[32]
6	Zn $(CH_3CO_2)_2$	623 K, 673 K and 723 K	glass	0.1 M	3 ml/min	20 cm	About 85% transmittance in visible region, PL measurement shows that emission peak intensity decreases with increasing substrate temperature, electrical resistivity also decreases with increasing temperature.	[33]
7	Zinc acetate	400^0 C	glass	0.2 M	8 ml/min	25 cm	Highly transparent ZnO thin film was prepared. Over 90% transmittance observed in visible region and sharp absorption edge near 380 nm, optical bandgap 3.27eV	[34]
8	aqueous zinc acetate	723 K	glass	0.4 M	5 ml /min	22 cm	Hydrobhobic and transparent thin film of ZnO was prepared. Average	[35]

							transmittance about 85 % with sharp absorption edge near 381 nm, optical bandgap 3.25eV. Applications: transparent self-cleaning surfaces, anti-fog, fluid microchips, microreactor, etc.	
9	zinc acetate, Aluminium chloride hexahydrate ($AlCl_3.6H_2O$)	450^0 C		0.2 M	2.5 ml/min	35 cm	undoped and Al doped ZnO thin films are deposited to study optoelectronic properties. Undoped ZnO thin film has transmittance about 90%, while Al doping cause it increasing upto 92%. Optical bandgap increases, from 3.25 to 3.32eV for Al doping. Electrical resistivity decreases with Al doping. The dark resistivity of Al:ZnO film was 1.33 x 10^{-3} Ω.cm whereas 1.10 x 10^{-2} Ω.cm for undoped ZnO films. Al:ZnO films has application in TCO layers for photovoltaic cell.	[36]
10	zinc acetate dehydrated, cuprous acetate (ZnO, Cu:ZnO films)	450^0 C	ITO coated glass substrate	0.4M			Cu:ZnO thin film was prepared. Absorption edge shows red shift i.e. band gap decreases with increasing doping concentration. Electochemical properties shows that fill factor (FF) and open circuit voltage values increase with increasing Cu doping concentration	[37]
11	zinc nitrate	500 °C	glass	0.05M to 0.2 M	2 ml/min	45 cm	It shows decreasing transmission with increasing molar concentration. Energy band gap decreases from 3.64 to 3.06 eV with increasing molar concentration from 0.05M to 0.2M.	[38]
12	zinc acetate dehydrate, aluminium nitrate nanahydrate $Al(NO_3)_3.9H_2O$	180 °C	Teflon substrates			35 cm, 40 cm and 45 cm	Aluminium Zinc Oxide thin film were prepared by varying nozzle to substrate distance and its effect on structural and optical properties has been studied. Optical band gap varies	[39]

M ARDYANIAN *et. al.* deposited ZnO thin film on glass substrate by using spray pyrolysis method and studied effect of spray parameters such as spray rate, solution volume, substrate temperature on properties of ZnO thin film. It is found that from XRD patterns films has polycrystalline wurtzite structure and preferred direction along (0 0 2) planes, about 90% transparency in visible region, optical band gap at 3.27 which is not significantly affected. The electrical properties show carrier density 4.28 x 10^{-13} cm^{-3}, sheet resistance 8.1 MΩ/sq and resistivity 102 Ω.cm which are obtained at best optimized parameters conditions at 3 ml/min deposition rate, 450 °C substrate temperature, for different solution volume [40]. R S DARIANI *et. al.* studied effect of precursor concentration on properties of ZnO thin film. The films were deposited on glass substrate at 450°C for different concentrations 0·1 to 0·4 M. Films are polycrystalline in nature with (002) plane on preferred direction. Transmission decreases with increasing concentration it is about 95% for film deposited with 0·1 M and decreases upto 75% for 0.4 M. The band gap energy also decreases from 3·37 to 3·19 eV with increasing precursor concentration. Electrical resistivity decreases from 49·80 to 2·09 Ω cm with increasing precursor concentration from 0.1 to 0.3 M and then again increases up to 3·41 Ω cm for 0.4 M. The values of photocurrent and photosensitivity are increases with increasing precursor concentration due to increasing grain size, porosity and quality of thin film [41]. E. Muchuweni *et.al.* prepared ZnO thin film with micro-ring structures by spray pyrolysis method on glass substrate from 0.1 M of zinc acetate dihydrate at 623 K substrate temperature, solution spray rate 2ml/min and nozzle to substrate distance maintain at 15cm. XRD shows hexagonal wurtzite structure with plane (002) on preferred direction. SEM shows that rod-shaped and spherical nanoparticles combined to form micro-ring like clusters on film. Average transmittance is about 75-80% and optical bandgap 3.28 eV were reported. Electrical properties show low sheet resistivity about 6:03 × 10^1 Ωcm and high figure of merit 4:35 × 10^{-6} $Ω^{-1}$ which reveals the film applicable in optoelectronics applications [42]. Amol Bhavsar studied structural, electrical and optical properties of ZnO thin film deposited by intermittent spray pyrolysis technique for photovoltaic application. To get the maximum conversion efficiency of photovoltaic cell or solar cell transparent conducting material with wide band gap and high conductivity are required [43]. Above all discussions reveals that by optimizing process parameters of spray pyrolysis technique one can acquire desire optical and electrical properties as well. Olusegun J. Ilegbusi *et.al.* studied the properties of single and mixed oxide thin films deposited by spray pyrolysis method. Both single and mixed oxide thin films were deposited on alumina substrate. Single oxide films of ZnO and SnO_2 are found to be nonhomogeneous at low temperature below 450°C. The films of mixed oxides of SnO2 + In2O3 and ZnO + In2O3 found to be more homogeneous. The synthesis parameters were greatly affecting the film morphology and optoelectronic properties. For highly sensitive and selective conductometric gas sensors precise microstructures of thin films are required which could be achieved with the help of optimizing the preparative parameters of thin films [44].

3.3 Applications of Zinc oxide thin film:

Zinc oxide thin films has numerous applications in different field. Its high optical transmission in visible region and wide band gap makes it useful for optoelectronics applications. It's electrical, optical and structural properties can be optimized as per application requirement by optimizing preparative parameters of spray pyrolysis technique. Some of the applications of ZnO thin films are High-Performance Metal-Oxide Field-Effect Transistors [45], gas sensing applications [46][47], Thin film transistor and flexible electronics [48], transparent electrodes for solar cell and photovoltaic applications [49], dye sensitised photoelectrolytic degradation [50], Transparent conducting flexible display[51]. The promising optical and electrical features of zinc oxide thin film makes it more useful in new electronic devices.

4. CONCLUSION AND FUTURE SCOPE

In this review effect of preparative parameters on optical and electrical properties of ZnO thin films deposited by spray pyrolysis method were reported. Spray pyrolysis technique is a simple and cost-effective method for large area deposition. Its preparative parameters such as Spray rate, substrate temperature, distance between spray nozzle and substrate, solution concentration and quantity, are greatly affect structural, optical and electrical properties of ZnO thin film. ZnO thin film shows maximum transmittance upto 95%, optical bandgap about 3.27eV. Electrical resistivity gets affected due to solution molar concentration, dopant, spray rate, nozzle to substrate distance due to which mobility of charge carrier's changes. To obtain precise optical and electrical properties of zinc oxide thin films for particular application preparative conditions of spray pyrolysis technique must be optimized. For that purpose, instead of trail method it is proposed to design a expert logic system which can predict about preparative conditions and corresponding optical and electrical properties of thin film.

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