ZNSE THIN FILMS- A BRIEF REVIEW

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ABSTRACT
ZnSe thin film deposition and their properties are briefly reviewed in this article. This includes analysis of structural (crystallinity, grain size and surface texture), electrical, corrosive and optical properties (absorbance, transmittance, refractive index and optical band gap). It has been observed that the deposition of ZnSe films dependent on various parameters like deposition technique, deposition temperature, bath composition, deposition time, thickness of the film etc. ZnSe thin films had the best preferential orientation of crystallinity to (111) plane of f.c.c. structure; however hexagonal wurtzite structure is also seen. The optical band gap of ZnSe films makes them suitable for various optoelectronic applications.

KEYWORDS: *Thin film, Deposition technique, Optical band gap, Transmittance*.

I. INTRODUCTION

Today the complicated electronic device structures can easily be constructed by thin films. Thin films are commonly used as deposit on a substrate for integrated optical circuits, capacitors, transistors etc.
Metal thin films can be fabricated by various methods for example chemical vapour deposition (CVD), electrodeposition, chemical bath deposition (CBD), thermal evaporation, successive ion layer adsorption and reaction method (SILAR) etc. According to the literature electrodepositon is more than 150 years old, thin film preparation technique which has been largely used to deposit metal films [1]. Electrodeposition by passing current through an electrochemical cell from an external source was first observed by Alessandro Volta in 1800. Bunsen and grove obtained metal films in 1852 by chemical bath deposition [2]. Faraday made metal films in 1857 by thermal evaporation.
The synthesis and properties of II-VI compounds i.e. Metal chalcogenide thin films are widely studied by various researchers because of their optical and semi conducting properties.
Metal chalcogenide thin films are important because of their numerous applications. The studies of these thin films provide directly or indirectly, a way for new areas of research in thin film chemistry and optoelectronic devices.
Zinc Selenide (ZnSe) is one of the most interesting metal chalcogenide due to its properties like direct band gap, transparency over a wide range of visible spectrum and relatively large value of non linear optical co-efficient. These properties of ZnSe thin films make it useful for semiconductor devices.
The ZnSe thin films have various applications like;

- In light emitting diodes and laser diodes.
- Photovoltaic solar cells [3].
- As Buffer layers on solar cell [4].
- In luminescent devices [11].
- In manufacturing lenses for high power IR laser [24]

II. EFFECT OF DEPOSITION TECHNIQUE AND DEPOSITION PARAMETERS

2.1 Suitable deposition potential for electrodeposition:

The cyclic voltammogram results obtained by R. Chandramohan *et al* [2005] on electrodeposition of ZnSe thin films on glassy carbon electrode as working electrode indicated a deposition potential between -700 mV to -1100 mV [6]. Similar results about deposition potential of ZnSe thin films were obtained by T. Mahalingam *et al* [2007] on using fluorine doped tin oxide coated glass and titanium substrate [7] and by Remiquisz *et al*[2009] on using Cu substrate as working electrode[8]. Influence of mechanical agitation on electrodeposition of ZnSe from aqueous solution of H_2SeO_3-H_2SO_4, was studied by J.L. Yang *et al* [2008]. They found that cubic ZnSe films could only be obtained at -900 mV from stirring solution [9].

It is concluded that stoichiometric ZnSe thin films can deposit around -900 mV (-0.9 V) Vs standard calomel electrode (SCE).

2.2 Structural properties:

By X-ray diffraction studies A.P. Samantilleke *et al*[1998] reported that the preferred orientation of ZnSe layers is (111) [5]. R. Chandramohan *et al*[2005] studied XRD pattern of ZnSe thin films electrodeposited onto a pre cleaned titanium and SnO_2 coated glass substrate and confirmed that the preferred orientation of these films was along (111), (100) and (222) planes. They observed (hhh) and (h00) type orientation, dominating in all the deposits. The thin films were found to be polycrystalline in nature having cubic crystal structure. SEM micrograph confirmed the uniform and needle like growth of film and the estimated grain size was 0.002 microns. The study of Raman spectra of as grown and annealed thin film showed increase of crystallite size due to annealing [6]. A similar conclusion was given by T. Mahalingam *et al* [2007] by studying 2Θ values of diffraction pattern and confirmed that the electrodeposited ZnSe thin films possess cubic zinc blende structure and the deposited film was polycrystalline in nature. SEM studies done by them showed that the film had a smooth surface and the substrate was well covered by grains. The crystalline size was estimated 9.5 nm for (111) plane of ZnSe thin film using Debye-Scherrer formula [7].

A.R. de Moraes *et al* [2002] studied the structural properties of electrodeposited ZnSe-Fe thin films on stainless steel substrate at 65 ^oC and by XPS analysis they observed an excess of Se near the surface of deposits. They claimed that this excess of Se was due to electrochemical kinetics. On XRD analysis of ZnSe-Fe thin films diffraction peaks at $2\theta = 31.7^o$, 53.2^o and 63^o were observed associated with ZnSe. The observation leads to a conclusion that deposited sample films were polycrystalline having zinc blende structure with predominant (111) plane corresponding to the $2\theta = 31.7^o$. No diffraction peak corresponding to Fe was observed in XRD pattern. They claimed that this may be due to the small particle size of Fe and their intrinsic structural disorder [10].

K. R. Murali *et al* [2009] observed the XRD pattern of pulse electrodeposited ZnSe thin films and found similar conclusion about the structure as observed earlier i.e. Cubic structured films with (111) preferred orientation. By this technique a nanocrystalline thin film of crystalline size of 15 nm can easily be obtained so they concluded that these films can be used in luminescent devices [11].

Pradip KR Kalita *et al* [2000] deposited ZnSe thin films in vacuum using thermal evaporation method by ZnSe powder within substrate temperature range from 303 K-623K. The thickness of the films was found to be 2000Å. By XRD analysis it was observed that the deposited films have f.c.c. zinc blende structure with (111) preferred plane. The lattice constants were estimated by Nelson-Riley plots which were found within the range of 5.6764 Å to 5.7074 Å. The lattice constant first increases upto 473 K then shows a decrement with in the substrate temperature range, this change indicated the strain in film grains deposited by thermal evaporation method. The grain size of films did not show any significant dependence on substrate temperature (T_S) and found to be very small within the range of 170 Å to 200 Å [12]. M. Öztaş *et al* [2005] studied the effect of 0.2:1, 0.4:1, 0.6:1, 0.8:1 and 1:1 Zn:Se ratio on the properties of ZnSe thin films synthesized on glass substrate at 430^oC by spray pyrolysis technique. On XRD analysis they observed that thin films were nanocrystalline with most preferred orientation along (200) plane as observed by earlier scientists using different deposition techniques. They also observed the increased grain size and crystallinity of the films after annealing at 400 ^oC for 60 min. by the results obtained by them, they reported a inverse relationship between Zn:Se ratio and grain size for both unannealed and annealed thin films. As the ratio of Zn:Se decreases the grain size increases. For annealed films the calculated grain size was 33.4 Å for 0.2:1 Zn:Se ratio while for 1:1 Zn:Se ratio it was 23.2 Å [13].

S. Venkatachalam *et al* [2007] synthesized vacuum evaporated zinc selenide thin films of different thicknesses (d=70 to 130 nm) on glass substrate. The SEM analysis showed that films had smooth

surface. The percentage of Zn and Se in ZnSe thin film (70 nm) was analyzed by EDAX spectrum and found to be 34.35% and 65.65% respectively. They observed that the films of lower thickness (70 to 100 nm) were amorphous in nature while the film of thickness 130nm was crystalline. The XRD analysis of higher thickness film showed the preferred orientation was along with (111) plane of cubic phase as mentioned by the earlier researchers [14].

Sunyang Ham *et al* [2008] demonstrated the photochemical deposition of CdZnSe thin films on Au substrate and by the study of EDX, Raman and UV reflectance spectroscopy they found that the ratio of Cd/Zn within the films can be controlled by controlling electrolyte composition [15].

Charita Mehta *et al* observed that the chemical bath deposited ZnSe thin films had a mixture of hexagonal and cubic phase [16]. M. M. Ivashchenko *et al* synthesized ZnSe thin films by CSVS (close-spaced vacuum sublimation) method and found that the films had zinc blend phase (sphalerite) along with low number of hexagonal phase (wurtzite). The evaluated lattice constant was 0.56 nm similar as reported earlier [17].

Pandurang C. Pingale *et al* analyzed the compositional and structural properties of chemical bath deposited ZnSe thin films of various Zn/Se ratio (0.993 to 0.57) and observed a decrement in Zn-content from 49.82at% to 36.47% with decreasing Zn/Se ratio from 0.993 to 0.57. The XRD analysis showed the hexagonal wurtzite phase along with(101) plane and for the different ratios of Zn/Se, the crystallite size decreases from 67 nm to 55 nm. By the AFM analysis they observed that the surface for higher Zn/Se ratio had spherical crystallite with bigger size and hillocks and valleys, so the surface was rough while for lower Zn/Se ratio hillocks collapsed and surface became smooth [18]. The ZnSe thin films with aluminum i.e. ZnSe:Al thin films were synthesized by M. G. M. Choudhury *et al* by laser-assisted evaporation; which had zinc blend cubic structure with preffered orientation (111) similar as ZnSe thin films. The lattice constant of ZnSe:Al thin films calculated from diffraction pattern was 0.56 nm which is slightly smaller than the lattice constant of ZnSe thin Films reported by the other workers [19].

Taj Muhammad Khan *et al* demonstrated the synthesis of ZnSe thin films on quartz by closed surface sublimation process at room temperature and by the analysis of XRD,AFM, FT-IR and Raman spectra they found the synthesized films suitable for optoelectronic applications [20]. The diffraction pattern of ZnSe:Mn/ PVA (Poly vinyl alcohol) nanocomposites showed that they are cubic in nature and the calculated particle size was in the range of 2 to 4 nm. [21]

2.3 Optical properties:

A.P. Samantilleke *et al* observed the optical absorbance of electrochemically deposited ZnSe thin film and found it maximum around 460 nm which indicates band gap of 2.7 eV [5].

T. Mahalingam *et al*[2007] reported that electro synthesized ZnSe thin films in aqueous solution of $ZnSO_4$ and SeO_2 have a high transmission of 90% in IR region while a low transmission of 40% at 400 nm. They found the optical band gap value 2.66 eV [7]. Optical band gap of thin films is in range of 2.64-2.68 eV was found through pulse electroplating technique which is close enough with that of reported earlier. The photoluminescence spectum exhibit peaks at 675 nm [11].

On increasing the concentration of silicotungustic acid the PL emission peaks shift to shorter wavelength, may be due to increase in band gap. By increasing the concentration of Silicotungustic acid, the band gap also increases in the range of 2.7- 3.1 eV. The chemically deposited ZnSe thin films have a direct band gap of 3.5 eV [charita Mehta *et al* 2009].

S. Chalina *et al* observed the transmission spectra of thermally evaporated ZnSe thin films and claimed that ZnSe thin films had high transmittivity to visible light. They also concluded that for heat treated sample the transmission co-efficient was greater than the value that obtained before heat treatment. The shape of transmission spectra was found to be very much influenced by the preparation conditions. The calculated energy gap was in the range between 2.6-2.7 eV. The refractive index calculated using swanpoel method was 2.487 at 756 nm while 2.658 at 505 nm [22]. Umesh Khairnar *et al* found the dependency of band gap on the thickness of thermally evaporated ZnSe thin films. He noticed an increment in band gap with increase in thickness. Refractive index for these films of different thickness(500 Å to 5000 Å) had values in the range from 1.75 to 2.05 [23].

But N.A. Okereke *et al* found that Optical band gap had an inverse relationship with film thickness. Band gap decreased with increase in film thickness. This conclusion obtained for 1.64 µm and 1.60 µm thick ZnSe thin films which were deposited on glass substrate from a mixture of $Zn(NO_3)_2$, and $SeSO_3$ solution by chemical bath deposition. The optical spectra showed that the ZnSe films had very

low absorbance in VIS-IR region while had high absorbance in UV region. The transmittance spectra indicated high transmittance (80%) in VIS-IR region which made it suitable for optoelectronic devices. The refractive index was also found to be decreased with increase in film thickness and had value in the range of 2.0-2.4 [24]. Same results were obtained by S. Venkatachalam *et al* [2007], they also found that optical band gap decreased with increase in film thickness(t=70,100 and 130 nm). Crystalline ZnSe films had band gap of 2.70 eV while amorphous ZnSe film had a band gap of 2.74 eV.

Comparison of Band gap energies of ZnSe thin films obtained by different synthesis techniques are given below

Table 1: Comparison of Optical Band Gap energy of ZnSe Thin Films

Scientists	**Methods**	**Optical band GAP (eV)**
M. Öztaş *et al [*2005*]*	Spray Pyrolysis	2.63-2.70
S. Venkatachalam *et al* [2007]	Vaccum Evaporation	2.70-2.74
R. Chandramohan *et al* [2004]	Electrodeposition	2.82
K.R. Murali *et al* [2009]	Pulse Electrodeposition	2.64-2.68
S.Chaliha *et al* [2008]	Thermal Evaporation	2.60-2.70
S. Anatohe *et al* [2013]	Thermal Vacuum evaporation	2.70
F.I. Ezema *et al* [2006]	Chemical Bath Deposition	1.60-1.75
N.A. Olereke *et al* [2011]	Chemical Bath Deposition	2.70-2.75
P.C. Pingale *et al* [2013]	Chemical Bath Deposition	2.52-2.69

The transmittance of ZnSe nanodots embedded in SiO2 thin films prepared by sol-gel dip coating process was decreased with the increase of molar ratio of ZnSe/SiO2 and the film thickness. 6% ZnSe/SiO2 had the maximum transmission of 54.58% and reduced to 36.71% for 14% ZnSe/SiO2 thin film. The films had band gap in range of 2.64-3.59 eV [27].

On adding 3.0 wt% Al in ZnSe film a decrement in band gap was observed (E_g of ZnSe without Al was 2.62 eV while while E_g of ZnSe:Al was 1.85) [19].

M. Öztaş *et al* [2005] and Pandurang C. Pingale *et al* [2013] found the optical band gap dependent on Zn:Se ratio. M. Öztaş *et al* [2005] studied the transmission spectra of sprayed ZnSe thin films and the optical band gap were estimated in the range of 2.65- 2.71 eV for unannealed thin films while for annealed thin films up to 400 ^oC for 60 min in an open air furnace the E_g was found in the range of 2.70-2.63 eV . On annealing the estimated band gap was reduced and an increased blue green response was observed [13].

F.I. Ezema *et al* also studied the effect of thermal annealing on the optical properties of CBD ZnSe thin films. Decreased transmission was observed after annealing the film at 333 K and 473 K. The as-deposited ZnSe films had E_g 1.60 eV while the band gap for annealed thin films was found to be 1.70 eV and 1.75 eV which indicated a strong red shift in optical spectra. They claimed that this was due to the morphological dependent properties of optical band gap and deposition condition of the films [26].

Anuar kasim *et al* claimed that the band gap value increases as the deposition time is reduced from 120 to 30 min while the stoichiometric ZnSe was obtained at 12 min [28].

2.4 Electrical properties:

S. Chalina *et al* [2008] observed the electrical properties of thermally evaporated ZnSe thin films. The films were highly resistive and their conductivity at room temperature was found to be in order of 10^{-7} ohm^{-1} cm^{-1}. The electrical behavior of these ZnSe thin films was found to be unstable. The resistivity of films decreased with increase in substrate temperature upto 180^0c while the doping of the films enhanced the conductivity [22]. M. Öztaş *et al* [2005] studied the photoconductivity of ZnSe thin films having different Zn:Se ratio. They found the photoconductivity in order of 10^{-6} ohm^{-1}. An increase in conductivity was seen with decreasing Zn:Se ratio. They found 0.2:1 Zn:Se ratio more conductive among all the Zn:Se ratio taken [13]. Electrical properties of thermally evaporated ZnSe thin films were studied by Jeewan sharma *et al* [29]. The films were deposited onto glass substrates at different pressures of Ar gas. He concluded that the photoconductivity of these films was due to two factors carrier modulation i.e. resultant increase in the photo generated carriers and barrier modulation i.e. increase of effective mobility. In this case, the photocurrent was controlled by trapping centers.

Both shallow and deep traps were found in these films and they observed a quasi-continuous distribution of various traps.

2.5 Corrosive properties:

The electrodeposited ZnSe thin films synthesized using H_2O-DMSO mixtures exhibit improved resistance toward corrosion [30]. R.K. Pathak *et al* also studied the corrosion behavior of ZnSeHg thin films and concluded that the films exhibit increased corrosion resistance on addition of Hg content [31].

III. CONCLUSION

This article has surveyed the various methods of preparation of ZnSe thin films and their effect on properties of prepared ZnSe films. Almost similar data was observed for the films synthesized by different synthesis techniques. On incorporating other metals like Fe or Hg, an enhancement in optical and corrosive properties has been seen respectively.

IV. FUTURE WORK

In future, it is possible to develop higher corrosion resistance thin films containing zinc and selenium with other suitable metal particles. It is also possible to reduce manufacturing cost of metal chalcogenide thin films by developing a cheap preparation technique with the use of other suitable starting materials.

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