International Journal of Thin Film Science and Technology

Volume 10
Issue 1 *Jan. 2021*

Article 7

2021

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Ho Soonmin
Center for American Education, INTI International University, Putra Nilai, 71800, Negeri Sembilan, Malaysia, soonmin.ho@newinti.edu.my

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http://dx.doi.org/10.18576/ijtfst/100108

Deposition of Metal Sulphide Thin Films by Chemical Bath Deposition Technique: Review

*Ho Soonmin**

Center for American Education, INTI International University, Putra Nilai, 71800, Negeri Sembilan, Malaysia

Received: 21 Feb. 2021, Revised: 22 Mar. 2021, Accepted: 24 Mar. 2021.
Published online: 1 Jan. 2021.

Abstract: Chemical bath deposition technique has many advantages if compared to other deposition methods. This deposition technique is simple, inexpensive and convenient for large area deposition at low temperature as highlighted by many researchers. In this work, binary, ternary, and quaternary thin films have been produced by using this technique. An extensive survey of thin films synthesized during the past twenty years (2000 to 2020) is reported. The properties of thin films such as structural, optical, compositional, morphological and electrical were described based on literature review.
Keywords: band gap, chemical bath deposition, semiconductor, solar energy, thin films

1 Introduction

Preparation and characterization of metal chalcogenide thin films widely reported by many researches [1-3]. Because of the very unique physio-chemical, electronics and optical characteristics [4-6]. There are many deposition techniques including chemical and physical deposition technique were used to synthesize thin films [7-9]. As highlighted by researcher, each deposition method has its own advantages and imitations as well [10, 11]. The choice of deposition method strongly depended on production cost, available resources, and specific application [12]. Chalcogenide compounds [13] contain at least one chalcogen anion (sulfur, selenium and tellurium) and at least one more electropositive element.

Chemical bath deposition method has many advantages including low cost, easy set-up, suitable for large deposition at low bath temperature [14,15]. This technique does not use any toxic volatile constituents, could produce binary, ternary, quaternary and pen ternary metal chalcogenides. The growth of thin films onto suitable substrates [16-20] such as indium tin oxide coated glass, soda lime glass, microscope glass slide, titanium, mica, stainless steel, and fluorine doped tin oxide coated glass. The properties of obtained films can be controlled under different conditions such as pH, bath temperature, solution concentration, complexing agent, and deposition time. The film prepared at low concentration of solution showed very thin and non-uniform due to insufficient supply of ionic species during the deposition process. The presence of complexing agent during the formation of films will reduce the deposition rate because of higher complexation. These complexing agent including sodium tartrate [21], ammonia [22], tartaric acid [23], disodium ethylene diamine tetra-acetate (Na_2EDTA) [24], triethanolamine [25], acetic acid [26], hydrazine [27], ammonium sulphate [28], hexamine [29], nitrilotriacetic acid [30] and ethylenediaminetetraacetic acid (EDTA) [31]. In terms of deposition temperature, the films prepared at higher temperature indicated larger granule growth. Meanwhile, the films produced at lower bath temperature will sluggish the re-dissolution stage. Deposition time is one of the factor that influence the structural, optical, morphological, compositional of films. The films prepared at longer deposition time produced thicker films, low absorbance value in higher wavelength region. In this work, deposition of metal sulphide thin films by chemical bath deposition technique was reported. The properties of chemical bath deposited films were discussed according to literature review from 2000 to 2020. Several keywords such as “chemical bath deposition”, “metal sulphide thin films”, “thin films”, “binary film”, “ternary films” and “quaternary films” were searched in SCOPUS, Web of Science and goggle scholar database.

2 Literature survey

2.1 Binary thin films

*Corresponding author E-mail: soonmin.ho@newinti.edu.my

The hexagonal phase SnS_2 thin films were prepared onto glass slide in the presence of thioacetamide, citric acid and tin (IV) chloride pentahydrate. Under the optimum conditions, these films showed band gap of 2.4 eV, homogeneous and well adhered to the substrate [32]. Tin disulfide films produced for 90 minutes showed shuttle shaped grains, good crystallinity and low resistivity of 11.2 Ωcm. Power conversion efficiency was about 0.91 % [33]. The SnS_2 films were synthesized in the presence of ammonia solution. Visual observation showed that thin films were reddish brown, uniformly thick and adherent to the surface of substrate. The intensity of the (001) peak, film thickness, and particle size increased with the increase of ammonia concentration in the solution [34]. The growth of films in the presence of triethanolamine and ammonia (complexing agent). The films showed major peak in (001) plane and warm like grain in XRD and SEM studies, respectively. Band gap values are in the range of 3.3 eV to 3.7 eV, and optical transmittance was more than 80 %. Chemical bath deposited Sn_2S_3 thin films exhibited orthorhombic structure as reported [35]. There are four Raman peaks (66.3, 111.7, 224.7 and 308.9 cm^{-1}) could be observed. Band gap increased (2.03 eV to 2.12 eV) when the deposition time was reduced (24 to 20 hours). Deposition of films by using ammonia, ammonium chloride and triethanolamine as complexing agent [36]. SEM and XRD results showed films were densely packed surface coverage and orthorhombic phase, respectively. Researchers have reported that the electrical and optical behaviors of films could be easily tailored by modifying the experimental conditions [37]. So that, these materials could be used in sensor, battery, solar cell and biomedical sciences [38].

The amorphous copper sulfide thin films were deposited onto commercial glass slide at 40 °C for 24 hours in acidic conditions. Optical properties revealed that the band gap values are in the range of 2.13 to 2.35 eV [39]. The growth of CuS films onto glass substrate in alkaline solution, by using tartaric acid, copper sulphate, thiourea. Visual observation showed that the colour changed from blue to dark indigo blue during the experiment [40]. Activation energy increased reducing bath temperature, time, pH value, solution concentration. The polycrystalline and hexagonal phase of CuS films could be synthesized under different copper salts at 50 °C, 7 hours [41]. The band gap value (2.31 eV to 2.46 eV) and optical transmission (2 to 57 % in visible region) were reported. The CuS films have been deposited onto ITO glass in alkaline conditions (pH 11), for 5 hours at 300 K. Morphology studies revealed that these films have high roughness, compact and uniform grain size. These films could be used in dye sensitized solar cell, the power conversion efficiency and fill factor were found to be 0.34% and 27.1%, respectively [42]. The influence of solution concentration on the chemical bath deposited CuS films was studied [43]. The homogeneous grains with hexagonal crystal could be observed for the films prepared onto microscope glass slide in the presence of tartaric acid (complexing agent).

The bismuth sulfide (Bi_2S_3) films were deposited onto ITO glass, for 30 minutes at 40 °C. The films showed band gap of 1.8 eV with thickness of 50 nm. The open circuit voltage and short circuit current density are 440 mV and 0.022 mA/cm^2, respectively [44]. The ammonium citrate was employed as complexing agent during the experiment [45] and the films were well adhered to the substrate. As-deposited showed amorphous structure, while the annealed films exhibited excellent crystallization of films. Soda lime was used as substrate, while ammonium hydroxide and ethylenediamine as complexing agent during the deposition of films. Transmission electron microscopy (TEM) analysis revealed that interplanar distance of 3.11 Å, which corresponded to orthorhombic phase in sample [46].

The CoS films were deposited onto glass slide in the presence of cobalt (II) sulfate, thiourea and triethanolamine. The average crystalline size was 14 nm and revealed hexagonal wurtzite structure. The thickness was 2 nm and the band gap estimate to be 1.6 eV [47]. Synthesis of CoS films from chemical bath containing cobalt acetate, disodium EDTA and thioacetamide. Deposition was carried out onto soda lime glass at pH of 4.6, at temperature of 85 °C. XRD data showed that the films consisted of mixed phases of hexagonal (CoS), face centered cubic (Co_3S_4) and cubic (Co_9S_8) phases with the deposition time from 2 hours to 3.5 hours. The resistivity ($1.7X10^6$ to $1.3X10^4$ Ωm) and band gap (1.75 to 1.3 eV) reduced with increase in the crystallite size as the deposition time increases [48]. Production of cobalt sulphide thin films was carried out in the chemical bath containing cobalt chloride, thiourea, ammonia and EDTA solutions. Experimental results showed that the obtained films are good absorber of UV radiation and exhibited poor reflectance of solar reflection [49]. Preparation of CoS films onto glass slide in alkaline medium. The obtained films were dark green, tightly adhered and uniform based on visual observations. The film thickness, crystallite size, absorption coefficient and band gap were 537 nm, 15 nm, 10^4 to 10^5 cm^{-1}, and 1.13 eV, respectively. The obtained films exhibited n-type material, with activation energy about 0.67 eV [50].

The pyrite phase of FeS_2 thin films have been prepared onto soda lime glass by using ferrous sulphate, Na_2EDTA and sodium thiosulphate. The films showed uniform and adhered well to the substrate [51]. The growth of tetragonal FeS films onto glass slide in acidic conditions. XRD data showed the major peak corresponded to (001) plane with crystallite size about 14 nm. EDAX analysis confirmed the iron to sulfur ratio was 1:1. Band gap energy was 1.87 eV [52]. The influence of bath temperature (50 °C to 80 °C) on FeS_2 films was studied. The films prepared at 80 °C indicated most homogeneity, higher crystallinity and higher absorption [53]. The obtained films showed yellowish brown and orthorhombic phase. The Fourier transform infrared (FTIR) measurements were conducted and highlighted the presence of OH, CH, SO stretching vibration modes [54]. The FeS thin films have been produced onto substrate in chemical bath contained iron (II) chloride, urea,

thioacetamide. Experimental results showed that increase of temperature (25 to 75 °C) and immersion time (24 to 72 hours) cause increase in the corrosion rate. However, reduce the corrosion rate with increasing the pH value (2 to 5) [55]. The FeS_2 thin films were synthesized at pH 2.5, 90 minutes and bath temperature of 90 °C. Morphology studies revealed that the surface of substrate was covered with irregular grains. Band gap was 1.85 eV [56]. Iron disulphide (FeS_2) films could be used a solar absorber material due to abundance, less toxicity of the elements, high optical absorption coefficient and suitable band gap. The FeS_2 films were produced in the presence of EDTA at 50 °C. The Raman spectra confirmed marcasite phase with peak at 217 cm^{-1}. Morphology investigations showed inhomogeneous distribution of grains on substrate with small grain size [57].

Silver sulphide films were classified as I-VI compounds. The silver sulphide (Ag_2S) thin films were deposited onto glass slide by using silver nitrate, thiourea, EDTA (complexing agent) and ammonia solution. The films showed small grain size, exhibited high absorbance in ultra violet region. The band gap was 3 eV [58]. The influence of deposition time on the chemical bath deposited silver sulphide (Ag_2S) thin films was reported. The optical transmission of the films decreased as the deposition time was increased. The obtained n-type films showed monoclinic structure, and band gap values are in the range of 1.8 eV to 2.17 eV. The highest responsivity of the photodetector about 0.5 A/W at 850 nm based on the experimental results [59]. Silver sulfide films were used in IR detectors, electrochemical storage, photoconductor because of unique photoelectric and thermoelectric properties [60]. Deposition of silver sulfide thin films at 40 °C, 2 hours at pH (pH 2-3) by using sodium thiosulphate and silver nitrate. The band gap and grain size were 1.3 eV to 1.83 eV and 17 nm to 120 nm, respectively [61].

Cadmium sulphide thin films have been prepared using $CdCl_2$, $(NH_2)_2CS$ and ammonia solution. The structural changes from cubic to hexagonal phase because of varying cadmium ions during the experiment [62]. The 800 nm thick cadmium sulfide thin films were prepared onto ITO glass. XRD data confirmed cubic (111) and hexagonal (002) reflections [63]. Ethanolamine was used as complexing agent during the formation of CdS films [64]. Optical properties revealed that the films have high transparency (60 to 80%), as the wavelength more than 600 nm. The cubic phase CdS films have been prepared at 90 °C and for 50 minutes in the presence of ammonium hydroxide (complexing agent). Higher presence of organic compounds could be observed by using low ammonium hydroxide [65]. Resistivity increased (10^2 to 10^7 Ωcm) with increasing the concentration of complexing agent (0.18M to 0.36 M). The influence of deposition time on the properties of CdS films was studied [66]. Hexagonal structure, uniform morphology and good adherent to substrate were reported for all samples. The crystallite size was found in the range between 11 to 15 nm. EDAX results confirmed that the ratio of cadmium is more compared to sulfur. The band gap increased from 2.23 eV to 2.27 eV with increasing the deposition time (40, 60, 80 minutes). The effect of bath temperature on the chemical bath deposited CdS films was investigated [67]. The optical transmittance reduced, but thickness increased with increasing bath temperature from 40 °C to 80 °C. Electrical characterization confirmed that high Voc and ISc values for the films prepared at 80 °C.

Arsenic trisulphide thin films have been deposited onto glass substrate in the chemical bath containing arsenic trioxide, disodium ethylenediaminetetra acetic acid and thioacetamide. The obtained films are uniform, and indicated n-type conductivity. Experimental results showed that band gap and electrical resistivity reduced as the film thickness increased [68]. The As_2S_3 films have been prepared in chemical bath, at pH 2, containing sodium thiosulfate [69]. The XRD patterns showed mixture of As_2O_3 and As_2S_3 in as-deposited films, however changed to As_2S_3 in annealed films (heated at 150 to 250 °C). Deposition of films at pH 3, for 40 minutes under various bath temperatures [70]. Optical studies indicated that band gap varies (2.2 eV to 2.6 eV) with film thickness (67 nm to 265 nm). Variation in activation energy (0.23 to 0.11 eV) because of increase in defect levels in nanosize films. Preparation of As_2S_3 films in the presence of various types of complexing agents was reported [71]. Experimental findings showed the variation of film thickness with complexing agent such as EDTA (301 nm), tartaric acid (287 nm), acetic acid (217 nm), and oxalic acid (207 nm). Different activation energy values (0.3 to 0.39 eV) could be observed depending on the complexing agent.

ZnS films have been deposited onto quartz substrate. The films prepared under self-catalyst growth process showed smooth, flat morphology and showed excellent crystallinity [72]. ZnS films were synthesized onto glass substrate in the chemical bath containing $ZnCl_2$, NH_4NO_3 and $CS(NH_2)_2$ solutions [73]. The films showed band gap of 3.84 eV, transmittance more than 85% in the 300-800 nm, and stoichiometry ratio of 49:51 (Zn:S). The influence of thioacetamide concentration on the chemical bath deposited ZnS films was studied [74]. Crystallinity and surface roughness reduced, band gap increased (3.7 eV to 3.95 eV) with reducing in concentration of thioacetamide. The annealed chemical bath deposited ZnS films showed excellent crystallinity, produced larger grain size and enhanced the absorption in the visible region [75]. The 100 nm thick films were deposited onto soda lime glass in chemical bath containing high concentration ratio of thiourea. The growth rate (0.88 nm/minute) and transmittance of films (more than 80%) were reported [76]. Tri sodium citrate was used as complexing agent during the formation of zinc sulphide films. An increase in the concentration of complexing agent leads to improvement of uniformity of films, excellent crystallinity, reduced in the grain size, surface roughness (RMS) value, and band gap value [77]. ZnS films were synthesized using a mixture of Na_3-citrate and EDTA in basic medium [78]. The electrical resistivity, average transmittance and band gap were 105 Ωcm, more than 70% and 3.84 eV, respectively.

Indium sulfide thin films were synthesized in chemical bath containing indium chloride, thioacetamide and citric acid (complexing agent). Surface morphology changes from nanospheres to network like with increasing the deposition time [79]. The influence of bath temperature on the properties of films was studied [80]. Crystalline structure and roughness value were changed with increasing bath temperature from 30 °C to 85 °C. The highest transmittance of more than 90% and the highest band gap (3.4 eV) could be observed for the films prepared at 85 °C. The effect of thioacetamide on the properties of films was investigated [81]. The crystallite size varied (15 – 25 nm), granular density decreased with the increase of thioacetamide concentration. The photovoltaic parameters were investigated and the power conversion efficiency was 0.6 %. The growth of thin films onto glass substrate in the pH 2.6, bath temperature of 35 °C and under various deposition times [82]. The films showed electrical conductivity of 10^{-3} to 10^{-7} $(\Omega m)^{-1}$ in all preparation conditions. The annealed films (350 °C) showed an open circuit voltage of 630 mV and short circuit current density of 0.6 mA/cm^2.

The PbS films have been deposited onto glass substrate at room temperature. The films prepared for 30 minutes showed adherent to substrate, preferential XRD diffraction peak (200) orientation, crystallite size of 40.4 nm [83]. The formation of PbS films using lead acetate and thiourea. Crystallite size was determined by using Scherrer formula, and showed 57 nm [84]. Optical properties revealed that thermal treatment (100, 200, 300 °C) strongly influence the band gap energy (1.68 eV to 2.12 eV) of films [85]. The growth of PbS films onto glass substrate in the presence of TEA (complexing agent). XRD showed face centre cubic crystal structure and photoluminescence spectra indicated a well-defined peak at 428 nm in all samples. The electrical resistivity and band gap (0.98 eV to 0.68 eV) decreased with increasing the concentration of complexing agent [86]. Deposition of PbS films in chemical bath containing thiourea and lead nitrate. Visual observation showed that homogeneous and well adhered to the substrate with dark surface [87]. XRD data confirmed that cubic phase, preferential (111) orientation with lattice constant (5.936 Å). The obtained films exhibited high absorbance and high reflectance in the visible region.

The cubic phase of MnS_2 thin films were synthesized in chemical bath containing sodium thiosulfate, manganese sulfate and sodium tartrate. Experiment results indicated that more diffraction peaks could be detected as the deposition time was increased from 3 days to six days. AFM investigations supported that the film thickness, grain size and surface roughness reduced with decreasing the deposition time [88]. The trisodium citrate was used as complexing agent during the formation of manganese sulphide thin films. The band gap was 3.23 eV [[89]. The synthesis of various phases (metastable form, hexagonal γ-MnS, cubic β-MnS) of manganese sulfide thin films via chemical bath deposition method. Scanning electron microscopy (SEM) studies revealed hexagons, spheres, cubes or flowers like in obtained films [90]. The hexagonal structure of MnS films have been prepared onto glass slide. The lattice parameters a=b=3.9 Å and c=6.4Å. The crystallite size was 8.35 nm based on Scherrer formula. EDAX showed the obtained films were in the best stoichiometry. Optical properties revealed that direct (3.67 eV) and indirect (2.67 eV) band gap values [91]. The formation of MnS films by using manganese (II) acetate tetrahydrate (manganese source) at 60 °C. The band gap was 3.1 eV [92]. AC electrical conductivity studies were conducted. The conductivity increased with increasing in the frequency and temperature. The activation energy was 0.28 eV [93].

The Ni_4S_3 thin films were deposited onto indium doped tin oxide (ITO) glass in the chemical bath containing nickel sulphate, sodium thiosulfate and triethanolamine. The films prepared at pH 2.5 showed preferentially (111) plane based on XRD pattern [94]. The influence of triethanolamine on the properties of films was studied. The XRD results showed that the number of diffraction peaks depended on the amount of complexing agent [95]. Deposition of NiS thin films onto glass slide by using sodium sulphide and nickel sulphate at room temperature [96]. The black colour of thickness of 0.3 μm was observed. Morphology studies revealed that film surface has holes, indicating these films could be used in electrochemical capacitive performance. The growth of hexagonal phase of nickel sulphide thin films in alkaline condition (pH 10). The crystalline size and band gap were 22 nm (Scherrer equation), and 0.4 eV (Tauc plot), respectively [97]. The obtained materials could be used in optoelectronic applications due to high refractive index (1.82 at λ=800 nm). The Ni_3S_2 thin films have been deposited onto chemical bath containing thiourea, EDTA and ammonia solution. Optical properties showed that the band gap was 3.6 to 3.8 eV, reflective index of 1.56 with high transmittance of 80% [98].

The molybdenum disulphide thin films have been deposited onto different substrates such as glass slide and quartz substrate by using ammonium tetrathioolybdate. XRD data showed amorphous character in as-deposited films [99]. Formation of rhombohedral and homogeneous ultrathin nanoflakes of MoS_2 films could be used in high performance supercapacitors [100]. These films indicated good electrochemical performance (maximum specific capacitance of 576 F/g at 5 mV/s) and excellent cycling stability (82% over 3000 cycles). The brown colour of MoS_2 thin films were deposited onto glass slide in chemical bath containing ammonium molibdat and sodium sulphide. Thin layer and bulky structure could be observed when the bath temperature were 30 °C and 60 °C, respectively. SEM images confirmed that the formation of microdomes could be seen with increasing the temperature from 60 °C to 90 °C [101].

Deposition of HgS films in chemical bath in the presence of complexing agent (iodine). Results showed that formation of $[HgI_4]^{2-}$ complex ions and reduced the amount

of sulfur when the concentration of iodine increased [102]. The influence of deposition time on the properties of thin films was studied. The films thickness directly proportional to the deposition time [103]. The obtained films could be used in photovoltaic systems due to unique optical and electrical properties. Mercury sulfide (HgS) films have been synthesized onto glass substrate in chemical bath containing mercury (II) nitrate, thiourea and trisodium citrate. Deposition was carried out in pH 7.3, 10 minutes and 80 °C. XRD data supported the formation of films with trigonal modification. The optical transmission spectra indicated sharp increase of the light transmittance in wavelength more than 350 nm [104]. The band gap values were 3 eV to 3.06 eV. These materials could be used in photoconductors, solid state solar cell, storage cell and photo electrochemical cell.

Calcium sulphide thin films were used in window coating due to low absorbance (0.01 to 0.04), low reflectance range (1 to 15%) in infrared, visible and ultraviolet region. Deposition of thin films in bath containing calcium sulphate, EDTA (complexing agent) and sodium thiosulphate for 2 days, at room temperature. The obtained films showed high transmittance (70-85%) in the UV visible region and the band gap about 3.9 eV [105]. On the other hand, microscope glass slide was used a s substrate during the formation of films [106]. The experimental results indicated film thickness (2.64 to 2.98 µm), refractive index (1.37 to 1.43), optical conductivity (1.71 to 2.1 $X10^{12}$ S^{-1}), and absorption coefficient ($0.05X10^{6}$ m^{-1}).

Barium sulphide thin films were synthesized in chemical bath containing BaCl2, EDTA and thiourea under various deposition times. The optical properties showed that the films prepared for 48 hours indicated the highest absorption of electromagnetic radiation (0.8 to 0.85 A) and highest refractive index peak (value=2.205). The films prepared for 24h, 48h and 60 hours exhibited low reflectance value (14%), indicating these samples could be used for anti-reflection coating for solar cells. The band gap of the films prepared for 24 hours and 48 hours are 1.35 eV and 1.25 eV, respectively [107].

Chemical bath deposition was used to produce beryllium sulphide films in the presence of EDTA (complexing agent). Deposition was carried out onto glass slides, by using beryllium nitrate, thiourea, in alkaline conditions. The obtained showed higher transmittance within the far infrared regions, indicating these materials could be used for thermal control window coatings for cold climates [108]. A small variation of the film thickness could be observed (0.068 to 0.085 µm) when the pH was increased from 11.3 to 13.7. The band gap of the films prepared for 18 hours and 24 hours, at pH 11.3 were 2.3 eV and 4.4 eV, respectively.

The tungsten disulfide (WS_2) thin films have been deposited onto substrates in alkaline conditions, at room temperature, by using chemical bath deposition method [109]. The obtained films are yellow in colour, polycrystalline in nature, show uniform, and well adherent. Experimental results showed n-type conduction mechanism, band gap was 1.36 eV and specific electrical conductivity of $10^{-3}(\Omega cm)^{-1}$. On the other hand, synthesis of WS_2 films in the presence of complexing agent (triethanolamine). XRD showed the existence of hexagonal wurtzite type phase and the crystallite size was 37.4 nm. The power conversion efficiency, open circuit voltage and short circuit current were 1.29%, 428 mV and 367 µA [110].

Magnesium sulphide films have been deposited onto stainless steel under various deposition times [111]. Chemical bath contained $MgCl_2$, NaOH, EDTA and thiourea. Thermal emittance (0.146 to 0.18) increased with increasing the deposition time from 2.3 hour to 5 hours. The film thickness (1790 to 9937 µm) and mass of MgS (0.07 to 0.11 g) increased when the deposition time was increased from 2.3 hours to 4 hours. However, film thickness and mass of MgS were 6323 µm and 0.07g, respectively when the deposition time was 5 hours.

2.2 Ternary thin films

Copper doped cadmium sulfide thin films were reported. The formation of hexagonal crystalline structure was detected. These materials could be used as a field effect transistor [112]. The $Cd_{0.96}Cu0._{04}S$ thin films with crystalline size of 2.81 nm were synthesized at 80 °C on glass substrate. These films indicated direct allowed transition with band gap of 2.28 eV, could be used as suitable window material in fabrication for solar cell [113].

The copper iron sulfide thin films have been deposited onto glass substrate in acidic conditions. The nano-sized crystallites indicated band gap of 1.7 eV [114]. The synthesis of FeCuS films onto glass substrate by using thiourea, iron (III) trioxonitrate, copper chloride, TEA, EDTA. Thicker films (0.9 µm) could be observed for the films prepared at pH 7.9. Optical properties showed high absorbance could be seen in the UV region, and band gap values are in the range of 2.5 to 2.8 eV [115].

The hexagonal phase of copper manganese sulfide (CuMnS) thin films has been deposited onto glass substrate under different bath temperatures. The crystallite size and electrical conductivity increased with increasing the bath temperature. The films prepared under different bath temperatures have lesser particle size and the UV-visible spectroscopy indicated red shift in the optical band gap [116].

The CdZnS films have been prepared onto glass slide in the presence of complexing agent (ammonia). During the formation of thin films, experimental findings showed that the rate of cadmium ions integrated inti system is faster than the rate of zinc ions. Optical properties revealed that the band gap was 3.1 eV [117]. CdZnS films were synthesized onto soda lime in the bath containing cadmium chloride, zinc acetate, thiourea. Formation of CdZnS with wurtzite structure based on XRD pattern. The band gap (2.83 eV to 2.93 eV) and resistance increased, crystallinity reduced as the zinc content was increased [118]. Ternary

semiconductor material such as cadmium zinc sulphide thin films have been deposited onto glass slide, in alkaline conditions for 90 minutes, at bath temperature of 343 K. Morphology studies showed nearly uniform and homogeneous films without pin holes based SEM images. Band gap (3.2 to 4.1 eV) and grain size (6.63 nm) were investigated [119]. Deposition of CdZnS films in chemical bath containing $CdCl_2$, NH_2CSNH_2 and $Zn((C_2H_3O_2)_2.2H_2O)$. The grain sizes were 6-25 nm, and indicated transmittance from 50 % to 80% in the visible region [120]. Zinc cadmium sulfide films were used in chalcopyrite solar cells. XRD pattern confirmed that cubic ZnS changed to hexagonal CdS by changing the initial concentration of zinc and cadmium salts in chemical bath [121]. Synthesis of thin films onto soda lime glass substrate under various concentrations of zinc content. The resistivity, and band gap (2.4 eV to 2.7 eV) increased as the zinc content increased [122].

The $Cd_{0.825}Pb_{0.175}S$ thin films have been produced under optimum conditions such as deposition time (1 hour), bath temperature (80 °C), pH (pH 10.5). XRD data showed mixture of hexagonal and cubic phases. The obtained power conversion efficiencies are in the range of 0.184% to 0.245% [123]. The CdPbS thin films have been deposited onto glass substrate. Optical properties showed direct band to band transition and absorption coefficient of 10^4 cm^{-1}. The obtained films exhibited n-type behavior, and activation energies in the range of 0.168-0.71 eV [124]. The particle size of annealed CdPbS films was 12 nm based on XRD measurements. Optical studies revealed band gap about 2.61 eV [125]. Deposition of PbCdS films in chemical bath containing thiourea, ammonia, cadmium acetate and lead acetate. SEM analysis confirmed the presence of a compact layer with a surface composed of platelike shaped nanocrystals with well-defined boundaries [126]. The band gap was 1.9 eV.

The $AgInS_2$ thin films were heated in nitrogen gas, for 60 minutes at 400 °C [127]. Band gap and electrical conductivity were 1.86 eV and $1.2X10^{-3}$ $(\Omega m)^{-1}$. Experimental results indicated that the highest refractive index about 2.6 in the visible region, showing that these films could be used for photovoltaic applications and solar thermal conversion.

The $AgAlS_2$ thin films were prepared in the chemical bath containing silver nitrate, aluminium sulphate , thiourea and EDTA (complexing agent). The thickness increased (0.03 to 0.52 µm) with deposition time (18 hours to 48 hours). The band gap values are in the range of 2.15 eV to 2.4 eV [128]. Optical properties revealed that the obtained films showed transmittance value of 98% and 75% in NIR and VIS-NIR region, respectively. The films exhibited low reflectance values (7% to 20%), indicating these materials could be used for anti-reflection coating.

The Al-doped ZnS films were deposited onto glass substrate under various concentrations of aluminum [129]. Band gap was 3.66 eV and transmittance of 75 % in the visible range for the films prepared using Al-doping content of 6 %. The elemental composition analysis showed zinc =44.9%, Sulphur=49.8% and aluminium=5.3 %. The hexagonal wurtzite structure of aluminum doped zinc sulfide thin films have been prepared at 85 °C, onto glass substrate. FESEM analysis confirmed the dense mosaic like nanostructure in obtained films [130]. UV-visible spectrophotometric measurement indicated that the highest transmittance of 80% was obtained for 6 wt % Al-doped ZnS films. The n-type electrical conductivity with the band gap value (3.52 eV to 3.76 eV).

The synthesis of lead silver sulphide thin films at room temperature in the bath containing lead nitrate, silver nitrate, EDTA (complexing agent), TEA (complexing agent), thiourea. The absorbance and film thickness (0.62 µm to 1.18 µm) increased with increasing the deposition time (6 hours to 48 hours). The highest refractive index (2.1 to 2.64) could be observed for the films prepared for 36 hours. The band gap values are in the range of 2.2 eV to 2.4 eV [131].

Deposition of $CuInS_2$ thin films onto glass slide in alkaline conditions. XRD showed chalcopyrite and wurtzite structure with grain size (22.8 nm). Band gap was 2 eV and showed strong emission peak in IR region [132]. The lattice constants a=5.517 Å and c=11.11 Å have been detected for the $CuInS_2$ thin films produced at 45 °C. The transmittance is high in visible region (600-650 nm). The photoconductive has been analyzed and was observed about 10^4 [133]. The copper indium disulfide thin films have been deposited onto ITO glass in acidic conditions. Experimental results highlighted that the crystal degree and electrical property were improved with increasing the heat treatment temperature [134]. As-deposited chemical bath deposited $CuInS_2$ films were heated for 1 hour, at 350 °C and 400 mPa. The obtained films indicated elongated shape and length 40 nm. The electrical conductivity values are in the range 64.9 to $4.11x10^{-3}$ $\Omega^{-1}cm^{-1}$. The band gap was 1.4 eV, and could be used as absorbing layer in photovoltaic structure [135].

The growth of Ni doped Sb_2S_3 thin films onto glass slide [136]. The films showed orthorhombic phase and the grains were spherical based on XRD and SEM studies. Optical properties highlighted the band gap of 1 eV to 2.6 eV with absorption coefficient of 10^4 cm^{-1}.

Preparation of $Zn_{0.8}Mn_{0.2}S$ thin films onto microscope glass slide by using zinc sulphate hepta hydrate, manganese sulphate and thioacetamide in alkaline conditions. Optical studies showed that shoulder at 315 nm based on the UV-visible absorption spectrum. Magnetic properties revealed that these films showed a ferromagnetic behavior at 300 K [137].

Deposition of $Ni_3Pb_2S_2$ thin films onto microscope glass slide in acidic conditions (pH 1.5), at bath temperature of 70 °C [138]. The film thickness increased (131.9 nm to 633.4 nm), band gap decreased (1.9 eV to 1.6 eV) with increasing of tartaric acid (complexing agent) from 0.1 M to 0.15 M. The influence of bath temperature on the chemical bath deposited $Ni_3Pb_2S_2$ thin films was studied [139]. SEM analysis revealed that smooth and dense with average

diameter of 2.5 µm could be observed for the films deposited at 70 °C. Bigger grains (4 to 12 µm) with irregular shape could be detected when the bath temperature was 75 °C and above. The growth of thin films onto soda lime glass was reported [140]. Morphology investigations highlighted that pinhole free, uniform surface with different grain sizes. XRD data showed the existence of rhombohedral and major peak corresponded to (012) plane. The power conversion efficiency was conducted [141]. The photovoltaic parameters such as fill factor (0.47), power conversion efficiency (2.7%), open circuit voltage (0.61 V) and short circuit current density (9.9 mA/cm^2) were reported.

The PbMoS thin films have been deposited onto glass slide at room temperature. The crystallite size and band gap were 24.12 nm and 1.91 eV, respectively. The photovoltaic properties were investigated and the power conversion efficiency was 2.11 % [142].

The growth of $CuSbS_2$ films onto glass substrate at room temperature in the chemical bath containing antimony trichloride, sodium thiosulphate and copper chloride [143]. XRD data exhibited the formation of crystalline films with major peak corresponded to (400) plane and the crystallite size was 19.54 nm. Optical properties showed low transmittance in the visible region, indicating these materials could be used as anti-dazzling coatings in the car windscreen.

Nano crystalline $CdCoS_2$ films have been deposited onto glass substrate in the chemical bath containing $CoCl_2.6H_2O$, $CdCl_2.2H_2O$, thiourea, TEA and ammonium solution (complexing agent). Optical studies revealed that the band gap values are in the range of 2.5 eV to 2.7 eV, showed average transmittance of more than 55 % in the visible and near infrared region [144].

The growth of copper zinc sulphide thin films have been deposited onto glass slide in chemical bath containing copper chloride, EDTA, TEA, ammonia, thiourea and zinc nitrate. These films have low absorbance in visible and infrared region, but indicated high absorbance in UV wavelength. The obtained films could be used for eye glasses coating, solar thermal conversion, anti-reflection coating and coating poultry buildings [145].

Ternary iron zinc sulphide films have been synthesized onto glass slide by using ferrous nitrate, zinc chloride, EDTA, ammonia, thiourea. Optical properties showed that the high and low transmittance could be observed in visible & infrared regions, and UV region, respectively. These materials could be used photo thermal application, solar cell industry [146].

Cadmium silver sulphide thin films were produced at room temperature. The chemical bath containing cadmium chloride, silver nitrate, thiourea and TEA (complexing agent). The optical properties (low absorbance in visible region), particle size (94.5 nm to 723.7 nm) and band gap (1.9 to 2.4 eV) were reported [147].

The CrPbS films have been synthesized under various concentrations of chromium. The grain size reduced (71.5 nm to 18.9 nm) with increasing Cr doping concentration. The band gap (1.49 eV), Hall mobility (59.6 $cm^2V^{-1}s^{-1}$) and volume carrier concentration (20.7 x 10^{16} cm^{-3}) of the films prepared using 2% Cr-doping concentration [148]. The p-type Cr-doped PbS films have been deposited via chemical bath deposition technique. The obtained films showed polycrystalline in nature, low transmittance in the UV-visible region [149].

The Ho doped Ag_2S films have been deposited onto glass substrate in chemical bath containing silver nitrate, EDTA (complexing agent), thiourea and holmium nitrate. The photoluminescence emission peak was detected at 294 nm. SEM image showed the flower type structure [150].

The mercury chromium sulphide (HgCrS) thin films were produced onto glass slide in the chemical bath containing EDTA (complexing agent), $HgCl_2$, CrO_3 and NH_2-CS-NH_2 solutions. The influence of bath temperature, complexing agent, thiourea concentration and deposition time was investigated. The best conditions were 120 minutes, 10 mL of thiourea, 4.5 mL of EDTA, 65 °C based on the experimental results. The XRD data indicated cubic crystal structure and the major peak corresponded to (220) plane. EDAX spectra confirmed the obtained films were homogenous and stoichiometric [151].

The strontium doped lead sulphide thin films were successfully deposited onto glass substrate [152]. Experimental results showed that crystallite size reduced (17.9 nm to 9.14 nm), band gap increased (1.73 eV to 2.19 eV) with increasing Sr-doping concentration from 0% to 5%.

The iron doped PbS films have been synthesized onto glass substrates at room temperature [153]. The concentration of iron influenced the band gap and crystal size based on the experimental results. The photovoltaic properties revealed that the power conversion efficiency increased with increasing the concentration of iron in samples.

The cobalt doped PbS films have been produced via chemical bath deposition method under various cobalt concentrations [154]. The photovoltaic properties of obtained films were investigated. The research findings confirmed that the cobalt doped PbS films significantly improved the power conversion efficiency value.

The high purity in composition of Mn-doped CdS films have been synthesized using chemical bath deposition method [155]. The best conditions were temperature=80 °C, pH=6, time=300 minutes, precursors molar ratio of Cd:Mn:EDTA:S =1:3:0.4:5.

Preparation of Mn-doped PbS films have been deposited onto glass substrate by using lead acetate, thiourea and manganese acetate. XRD data confirmed that broadening of peaks, average crystallite size (21 nm to 11 nm) and reducing of peak intensity could be observed when the concentration of manganese was increasing from 0% to 2%. SEM investigations revealed the the films were smooth, and uniform. HRTEM showed lattice fringes with d-spacing value of 0.25 nm to (200) plane [156]. Selected area electron diffraction (SAED) images showed the nearly spherical nanoparticles were detected.

The stoichiometric $CuBiS_2$ films have been synthesized onto glass substrate in the presence of EDTA (complexing agent). The films showed n-type semiconductivity [157] and band gap of 1.82 eV. Excellent crystallinity could be observed in the annealed films (at 250 °C, in an inert atmosphere).

Deposition of lead silver sulphide (PbAgS) films onto non-conducting microscopic glass substrate at room temperature, in the presence of complexing agent (EDTA and TEA). The properties of thin films such as reflectance (1.48% to 20.35%), energy band gap (2.3 eV to 1.89 eV), film thickness (493 nm to 945 nm), electrical conductivity ($1.33X10^{-3}$ to $9.33X10^{-3}$ S/cm), transmittances (3.76% to 86.7%) were reported [158].

2.3 Quaternary thin films

The Cu_2ZnSnS_4 (CZTS) thin films have been deposited by using chemical bath deposition technique. The annealed films (at 673 K) showed tetragonal structure and band gap of 1.5 eV [159]. Production of CZTS film solar cell via chemical bath deposition and annealing route was reported. Photovoltaic parameters were studied, conversion efficiency of 3.79 % was obtained [160]. Chemical bath deposited CZTS films showed a mixture of Cu_2ZnSnS_4 and $Cu_{2-x}S$ phases based on the XRD patterns [161]. The soda lime glass was used as substrate during the formation of CZTS films [162] at 60 °C. The chemical bath containing copper sulfate, zinc sulfate heptahydrate, tin sulfate dihydrate, and sodium thiosulfate. The as-deposited films prepared in the presence of ammonia and TEA solution exhibited kesterite structure and band gap of 1.6 eV.

The CdZnSeS thin films [163] have been synthesized onto glass substrates in the presence of TEA (complexing agent) and cetyl trimethylammonium bromide (capping agent). The band gap reduced (2.21 eV to 2.01 eV) with increasing annealing temperature (100 °C to 500 °C). The tetragonal phase of Ag_2ZnSnS_4 films have been deposited onto ITO glass in chemical bath at 70 °C [164]. The obtained films showed n-type semiconductor, and band gap values are in the range of 2.08 eV to 2.56 eV. Formation of films in the presence of EDTA (chelating agent) indicated the highest photoelectrochemical response (0.65 mA/cm^2 at an external potential of 1 V versus Ag/AgCl).

4 Conclusions

Deposition of binary, ternary and quaternary metal sulphide thin films have been successfully prepared by using chemical bath deposition method. This deposition technique is simple, inexpensive and convenient for large area deposition at low temperature. The physical, optical, and electric properties of films have been characterized via various tools. Experimental results showed that the obtained materials could be used in the solar cells and optoelectronic devices.

Acknowledgement

The author is grateful to the INTI International University for providing the financial assistance to carry out this work.

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