

International Journal of Thin Films Science and Technology

# A Comprehensive Review of Preparation Methods, Parameters and Deposition Analysis of CuO/ZnO Thin Films

T. Nimalan<sup>\*</sup> and M. Rigana Begam

Department of Physics, Vel Tech Rangarajan Dr.Sagunthala R& D Institute of Science and Technology, Avadi, Chennai-600062, Tamil Nadu, India

Received: 14 Mar. 2024, Revised: 24 Jun. 2024, Accepted: 28 Jul. 2024 Published online: 1 Sep. 2024

Abstract: The purpose of this review paper is to present an analysis of the various thin film deposition methods, parameters, advantages and disadvantages and deposition analysis of copper and zinc oxide thin films. A thin film is a very thin effective layer of material ranging from a few nanometers to several micrometers in thickness. Thin film deposition methods are classified commonly in two categories. The first one is physical deposition methods, and the second one is chemical deposition methods. This research paper describes various physical and chemical deposition methods and conditions for copper and zinc oxide thin film preparations. From our review work, various deposition parameters are analyzed which are useful for deposition of high-quality low-cost metal oxide thin film.

Keywords: Thin films, Deposition conditions, Physical methods, Chemical methods, Copper oxide, Zinc oxide.

#### **1** Introduction

Over the past century, thin films have become an intrinsic part of everyday life. The visible applications are wideranging across many fields; from microelectronics to automobile parts, and from windows on skyscrapers to the metallic coatings on the insides of bags of potato chips [1]. Nowadays, the use of thin films to enhance the physical and chemical properties of materials is the most common practice in almost all the fields. Thin solid films have been used in many types of engineering systems and have been adapted to fulfill a wide variety of functions. For example, important and great developments in thin film technology were achieved allowing a rapid development of miniaturization of electronic devices [2].

Category	Applications
	Reflective and anti-reflective coatings; Anti corrosive films;
Optics	Interference and band filters; Polarizer; Protective glasses;
_	Building and airplane smart windows; Beam splitter.
	Active and passive thin film elements (Transistors / Resistors);
Flootropics	Rectifiers; Semiconductor devices; Insulation; Conduction;
Electronics	Integrated circuits; Wireless communications;
	Telecommunications; Flat-panel displays; Computer chips.
	Photo detectors; Photo conductors; Liquid crystal display (LCD);
Optoelectronics	Thin film transistors (TFT); Optical memories; Solar cells;
	Light-emitting diodes (LED); Electro-optic coatings.
Crystochnics	Superconducting switches; Superconducting memories;
Cryotechnics	Superconducting interference devices (SQUIDS).
	Electro and Bio and Photo catalysis; Diffusion barriers;
Chemistry	Protection against oxidation and corrosion; Gas and liquid
	sensors.
	Tribological coatings; Hard coatings; Adhesion providers;
Mechanics	Friction reducers; Micro- electromechanical systems (MEMS);
	Multifunctional emerging coatings.
	Audio and video systems; Computer memory discs; Magnetic
Magnetics	
Magnetics	read and write heads; compact discs; Magneto optic discs;

Table 1: Applications of Thin Film Technology



Thin films have been used in a wide range of applications in a wide range of fields. It can be summarized as shown in Table 1.

Thin film deposition methods are classified commonly in two categories. The first one is physical deposition methods, and the second one is chemical deposition methods. It can be summarized as shown in Figure 1.

Thin-Film Deposition Techniques						
Physical Methods	Chemical Methods					
Thermal evaporation	Sol-gel					
Electron beam evaporation	Chemical bath deposition					
Molecular beam epitaxy	Spray pyrolysis					
Pulsed laser deposition	Chemical vapor deposition					
Ion plating evaporation	Electrodeposition					
Radio frequency (RF) sputtering	Atomic layer deposition					
Direct current (DC) sputtering	Spin coating					
Ion beam sputtering	Successive Ionic Layer Adsorption and Reaction(SILAR)					
Arc evaporation	Electroless deposition					
Magnetron sputtering	Ion implantation					

Fig. 1: Classifications of Thin-Film Deposition Technique

# 2. Preparation Analysis of Copper Oxide and Zinc Oxide Thin Films by Physical and Chemical Methods:

#### 2.1 Copper oxide:

In recent years, copper oxide thin films have attracted much interest due to their potential applications for solar cells and gas sensor. Cuprous oxide (Cu<sub>2</sub>O) and cupric oxide (CuO) are the two main semiconductor phases of copper oxide with narrow band gap [3].



Cupric oxide (CuO) is a *p*-type semiconductor having a band gap of 1.21–1.51 eV and monoclinic crystal structure. Cuprous oxide (Cu2O) is also a *p*-type semiconductor having a band gap of approximately 2.0 eV and a cubic crystal structure [4]. In general, the copper oxides are studied for applications such as solar cells, high temperature superconductors, solar light modulation, window coatings for solar control, gas sensors, photo catalyst, electrochromism, lithium batteries, hetero junction for transparent conducting oxides, low friction coatings, and other electronic devices [5].

#### 2.2 Zinc oxide:

ZnO is a bio-safe material available in the form of white powder, which is insoluble in water. Its high thermal and mechanical stability at room temperature makes it a potential material for multipurpose applications [6].

Recently, zinc oxide (ZnO) has attracted substantial attention due to its superior physical properties and potential technological applications. ZnO is an II–VI compound semiconductor with a wide direct-band gap of 3.37 eV. ZnO has a large exciton binding energy of 60 meV, which favors efficient excitonic emission processes at room temperature and enables devices to function at a low threshold voltage [7].

Zinc oxide has been successfully included in the transparent conducting oxides used in modern solid-state technology (e.g. Solar cells, optoelectronic devices, sensors, and heat reflecting mirrors) [8].

The parameters of the copper and zinc oxide thin film can be listed in the following table 2.

Table 2: The various properties of CuO & ZnO

Parameters	Cuprous oxide	Cupric oxide	Zinc oxide
IUPAC	Copper (I) oxide	Copper (II)	Zinc oxide
Name		oxide	
Definition	Cuprous oxide is an inorganic chemical compound composed of the metallic cuprous ions $Cu^{+1}$ and the nonmetallic oxygen ion $O^{-2}$ .	Cupric oxide is an inorganic chemical compound composed of the metallic cupric ion Cu <sup>+2</sup> and the nonmetallic oxygen ion O <sup>-2</sup> .	Zinc oxide is an inorganic chemical compound composed of the metallic zinc ion $Zn^{+2}$ and the nonmetallic oxygen ion O <sup>-2</sup> .
Chemical	Cu <sub>2</sub> O	CuO	ZnO
Formula			
Appearance	Brownish-red	Black to brown	White solid
	solid	solid	
Molar mass	143.09 g/mol	79.545 g/mol	81.406 g/mol
Melting point	1,235 °C	1,326 °C	1,974°C
Boiling point	1,800 °C	2,000 °C	2,360 °C
Density	6 g/cm <sup>3</sup>	6.31 g/cm <sup>3</sup>	5.606 g/cm <sup>3</sup>
Solubility	Insoluble in	Insoluble in	Insoluble in
	water	water	water
Band gap	2.137 eV	1.2 eV	3.2 eV
Crystal	Cubic	Monoclinic	Wurtzite
structure			

#### 2.3 Deposition analysis of CuO and ZnO thin films:

Various physical and chemical methods have been used for copper oxide and zinc oxide thin films depositions namely: Thermal evaporation, Electron beam evaporation, Molecular beam epitaxy, Pulsed laser deposition, Ion beam sputtering, Radio frequency (RF) sputtering, Direct current (DC) sputtering, Sol–gel, Chemical bath deposition, Spray pyrolysis, Chemical vapor deposition, Electro deposition, Atomic layer deposition, Spin coating and SILAR. These methods are explored and discussed in tables 3, 4, 5 and 6 below.

Physica	Physical deposition methods (PDM) / Copper oxide thin film deposition						
Thin film	Methods	Source material / Substrate	Conditions for deposition parameters	Quality of film / Cost	Ref.		
CuO film	Thermal evaporation	Red Cu <sub>2</sub> O Powder / Glass tantalum SiO <sub>2</sub>	Base pressure: $5 \times 10^{-4}$ Pa Substrate temperature: 300 °C Annealing temperature: 500 °C for 3 h deposition rate : 0.1 nm/s.	Moderate / High	9		
Cu2O film	Thermal evaporation	CuO powder / sapphire	Base pressure: $1.5 \times 10^{-5}$ mm Hg Substrate temperature: 700 °C oxygen partial pressure: $1 \times 10^{-4}$ mm Hg Deposition time: 3 h	Moderate / High	10		
CuO and Cu <sub>2</sub> 0 films	Ultrahigh vacuum molecular beam epitaxy (UHVMBE)	Cu, O <sub>2</sub> / MgO	Incident O <sup>+</sup> beam energy: 50 eV Substrate temperature: 100–400 °C Cu flux: 2.5 × 10 <sup>13</sup> to 1.6 × 10 <sup>14</sup> atoms/cm <sup>2</sup> s Base pressure: $3 \times 10^{-10}$ Torr Total pressure: $3 \times 10^{-9}$ to $2 \times 10^{-8}$ Torr O <sup>+</sup> flux: 2.7 × 10 <sup>14</sup> atoms / cm <sup>2</sup> s O <sup>+</sup> ion current: 45 micro ampA	Excellent / High	11		
CuO film	Plasma- assisted molecular beam epitaxy (PAMBE)	Cu, O <sub>2</sub> / MgO	oxygen partial pressure: $2 \times 10^{-5}$ mbar Deposition time: 90 min O <sub>2</sub> flux : $2 \times 10^{-5}$ mbar Temperature: 500 °C	Excellent / High	12		
CuO and Cu <sub>2</sub> 0 films	Pulsed laser deposition	Cu2O / Glass	Substrate temperature: 300 and 500°C Chamber vacuum pressure: 10 <sup>-3</sup> mbar target-substrate distance: 4 cm Deposition time: 10 to 20 min	Excellent / High	3		
Cu2O film	Pulsed laser deposition	Cu, O <sub>2</sub> / Quartz glass, ITO, NaCl, Si	Substrate temperature: 25–400 °C oxygen partial pressure: 0–10 mTorr target-substrate distance: 5 cm Base vacuum of the chamber : $\leq 10^{-5}$ Torr	Excellent / High	13		
CuO film	Radio frequency (RF) magnetron sputtering	Cu, O <sub>2</sub> / Glass	Chamber base pressure: $1 \times 10^{-6}$ Torr DC power supply: 200 W Deposition time: 600 to 1800 sec with an step size of 600 sec Ar gas flow rate: 10 sccm O <sub>2</sub> gas flow rate: 15 sccm Sputtering pressure :5 × 10 <sup>-3</sup> Torr target-substrate distance: 14cm	Excellent / High	14		
CuO, Cu <sub>2</sub> O, and Cu <sub>4</sub> O <sub>3</sub> films	Radio frequency (RF) magnetron sputtering	Cu, O <sub>2</sub> / Glass	Chamber background pressure: $3 \times 10^{-4}$ Pa RF power supply: 50 W Deposition time: 60 min Substrate temperature: 300 °C Ar gas flow rate: 10 sccm O <sub>2</sub> gas flow rate: 0–2 sccm Sputtering pressure :1.7–1.8 Pa target-substrate distance: 80 mm	Excellent / High	15		

	Table 3: Physical de	position methods of copper oxide thin films
--	----------------------	---



216

Cu2O film	Radio frequency (RF) magnetron sputtering	Cu, O <sub>2</sub> / Glass	Deposition time: 5 min Base pressure: $10^{-6}$ Torr Substrate temperature: room temp. Ar gas flow rate: 50 sccm O <sub>2</sub> gas flow rate: 1-4 sccm RF power supply: 150,200 and 200W Chamber pressure : 2,3,6 and 12 mTorr	Excellent / High	5
Cu2O film	Reactive direct current (DC) magnetron sputtering	Cu, O <sub>2</sub> / Glass	Deposition pressure: $6.3 \times 10^{-3}$ torr Base pressure: $6 \times 10^{-6}$ Torr DC power supply : $60$ W Ar gas pressure: 20 sccm oxygen partial pressure: $1.1, 1.5, 1.8 \times 10^{-3}$ and $8.0 \times 10^{-4}$ Torr Substrate temperature: 473K	Excellent / High	16
CuO and Cu <sub>2</sub> 0 films	Reactive direct current (DC) magnetron sputtering	Cu, O <sub>2</sub> / Corning glass	O <sub>2</sub> gas flow rate: 10 sccm DC sputtering power : 10-40 W Ar gas flow rate: 15 sccm target-substrate distance: 500 mm Substrate temperature: 300 °C Deposition time: 222-57 min for 10-40 W	Excellent / High	17
CuO film	Direct current (DC) magnetron sputtering	Cu, O <sub>2</sub> / Glass	Substrate temperature: 623K Deposition time: 20 min target-substrate distance: 5 cm DC sputtering power : 600 V at 1.2 mA / cm <sup>2</sup> System pressure: $5.2 \times 10^{-3}$ Torr	Excellent / High	18
CuO and Cu <sub>2</sub> 0 films	Io beam sputtering	Cu, O <sub>2</sub> / Si and glass	Working and background pressure: $4 \times 10^{-3} \& 8 \times 10^{-4} Pa$ Ar ion beam current: 400 micro Amp Deposition time: 20 min to 3 h target-substrate distance: 6 cm Annealing temperature: 200- 600 °C for 1-7 h in a 1 h step	Excellent / High	19

Table 4: Chemical deposition methods of copper oxide thin films

Chemica	Chemical deposition methods (CDM) / Copper oxide thin film deposition					
Thin film	Methods	Source materials / Substrate	Conditions for deposition parameters	Quality of film / Cost	Ref.	
Cu2O film	Sol-gel	Copper (II) acetate, Isopropyl alcohol, Diethanolamine, Glucopon, polyethylene glycol, ethylene glycol / Indium tin oxide (ITO) coated glass	Temperature: 60 °C for 10 min Rotating speed: 3000 rpm for 40s Annealing temperature: 350 °C for 1 h in 5% H <sub>2</sub> + 95% N <sub>2</sub> atmosphere.	Good / Low	20	
CuO film	Sol–gel	Cupric chloride, methanol, diethanolamine, Monoethylene glycol, glucopone / TiO <sub>2</sub>	Rotating speed: 2000 rpm for 40s Annealing temperature: 200 - 400 °C for 7 h air and N <sub>2</sub> atmosphere Temperature: 25-30 °C	Good / Low	21	
CuO film	Chemical bath deposition	Copper (II) chloride, ammonia, water / Glass	Temperature: 90 °C Deposition time: 7 min Annealing temperature: 400,500°C for 2 h pH: 3.80-10.0	Excellent / Low	22	



Cu <sub>2</sub> O film	Chemical bath deposition	copper sulphate, sodium sulfate, Sodium hydroxide / Glass	Temperature: 70 °C Deposition time: 20 sec Annealing temperature: 200- 400°C for 1 hr	Excellent / Low	23
CuO film	Chemical spray pyrolysis	Copper (II) chloride / Glass	Substrate temperature: 300°C Air pressure : 0.4 kg cm <sup>-2</sup> Precursor flow rate : 10 ml min <sup>-1</sup>	Excellent / Low	24
CuO film	Ultrasonic spray pyrolysis	Copper (II) chloride / Glass	Substrate temperature: 300°C Deposition time:20 min Solution flow rate: 10 to 30 ml/h with a step of 5 ml/h.	Excellent / Low	25
CuO ,Cu2O films	Atmospheri c-pressure chemical vapor deposition	Copper dipivaloylmethanate, oxygen / Borosilicate glass plate	O <sub>2</sub> flow rate: 1 and 300 cm <sup>3</sup> min <sup>-1</sup> N <sub>2</sub> flow rate: 599 and 300 cm <sup>3</sup> min <sup>-1</sup> Temperature: 300 and 500 °C oxygen partial pressure: $1.689 \times 10^2$ and $5.07 \times 10^4$ Pa	Excellent / Low	26
Cu2O film	Low temperature chemical vapor deposition	(N, N'-di-sec butylacetamidinato)copper (I), water / silicon wafers or glassy carbon or silica glass	Substrate temperature: 125– 225°C Process pressure: 1–10 torr N <sub>2</sub> flow rate: 40 sccm	Excellent / Low	27
CuO ,Cu2O films	Hot wall chemical vapor deposition	Copper (II) acetylacetonate, oxygen / sapphire and MgO	Reactor pressure &temperature: 20 mbar & 175°C Substrate temperature: 350– 500°C O <sub>2</sub> flow rate: 5-200 sccm	Excellent / Low	28
Cu2O film	Metal organic chemical vapor deposition	Copper (II) hexafluoroacetylacetonate, oxygen gas, water vapor/ ZnO coated glass	Substrate temperature: 300– 400°C Process pressure: 2 torr Ar flow rate: 5 sccm O <sub>2</sub> flow rate: 300 sccm H <sub>2</sub> O vapor flow rate: 120 sccm	Excellent / Low	29
CuO ,Cu2O films	Electro deposition	copper sulphate, lactic acid, Sodium hydroxide / Copper	Temperature: 40 and 60 °C pH: 11.5-12.5 Applied potential: -0.45 to -0.65V Deposition time: 10-50 min Copper sulphate con. : 0.15-0.25 M	Excellent / Low	30
Cu2O film	Electro deposition	Copper(II) acetate, Sodium acetate trihydrate / indium- doped tin oxide (ITO) glass	Temperature: 20–80 °C Deposition time: 2–80 min NaCl con.: 0–10 mM Potential: –0.1 to –0.4 V	Excellent / Low	31
Cu2O film	Atomic layer deposition	Copper(II) acetate monohydrate, Copper(II) acetate anhydrous, water vapor / soda lime glass (SLG), Si	Temperature: 180–220 °C N <sub>2</sub> , H <sub>2</sub> O and O <sub>2</sub> flow rate: 400 sccm Deposition cycles: 500–7000 Rector pressure: 10 mbar	Excellent / Low	32
CuO film	Atomic layer deposition	copper (II)-bis-(- dimethylamino-2- propoxide), ozone / SiO <sub>2</sub> /Si	Substrate temperature: 112– 165°C Deposition cycles: 500–10000 oxygen partial pressure: 34 Pa Rector pressure: 200-260 Pa	Excellent / Low	33



CuO film	SILAR	Copper chloride, ammonia/ Glass	Annealing temperature: 200 - 400 °C for 30 min SILAR cycle: 80 times Immersion Time: 30&60 s for anionic and cationic precursor Rinsing time: 7&30 s for anionic and cationic precursor pH: 3 Volume of precursor: 100 mL for anionic and cationic con of cationic precursor: 0.1M	Excellent / Low	34
CuO ,Cu2O films	SILAR	copper sulphate pentahydrate, sodium thiosulfate, Sodium hydroxide / Glass	SILAR cycle: 80 times Immersion& Rinsing Time: 20&10 s for anionic and cationic precursor con of cationic & anionic precursor: 1M Annealing temperature: 200 - 400 °C for 1 to 4 hr	Excellent / Low	35
Cu <sub>2</sub> O film	Spin coating	Copper (II) acetate, Ethanol/ Soda-lime glass	Annealing temperature: 275 to 500°C for 2 h in N <sub>2</sub> atmosphere Temperature: 150 °C Spin coating speed:1500&3000 rpm for 15&30s	Excellent / Low	36
CuO film	Spin coating	Cupric acetate, methanol/ Glass	Temperature: 300 to 700°C for 1 h in air Annealing temperature: 100 °C for 10 min Spin coating speed: 3000 rpm for 40s	Excellent / Low	37

## Table 5: Physical deposition methods of zinc oxide thin films

Physical	Physical deposition methods (CDM) / Zinc oxide thin film deposition						
Thin film	Methods	Source materials / Substrate	Conditions for deposition parameters	Quality of film / Cost	Ref		
ZnO film	Thermal evaporation	ZnO Powder / Glass	target-substrate distance: 6 cm Chamber base pressure: 10 <sup>-7</sup> mbar Thermal Oxidation Temperature: 200 - 500 °C for 2 h Annealing temperature: 523K, 623K and 723K Film Thickness: 60 nm & 130 nm	Moderate / High	38		
ZnO film	Thermal evaporation	ZnO Powder / polyethylene terephthalate	Chamber vacuum pressure: $3 \times 10^{-5}$ Torr Film Thickness: 100 to 300 nm Heated Direct Current: 3.0 to 8.0 A	Moderate / High	39		
ZnO film	Laser- molecular beam epitaxy (L- MBE)	ZnO Powder, polyvinyl alcohol binder, water / SI- InP	target-substrate distance: 6 cm Laser pulse energy: 250mJ Repetition rate: 10 Hz Substrate temperature: RT, 300 and $300^{\circ}$ C Chamber evacuated pressure: 1 × 10 <sup>-6</sup> Torr O <sub>2</sub> pressure: 1 × 10 <sup>-5</sup> Torr	Excellent / High	40		
ZnO film	Plasma- assisted	Zn, O <sub>2</sub> / Si and porous	Substrate temperature: 350 °C Deposition time: 5 to 30 min	Excellent / High	41		

SP	219

	molecular beam epitaxy (PA-MBE)	silicon (PS)	Film Thickness on Si: 50,85,220 and 320 nm Film Thickness on PS: 50,100,150 and 270 nm		
ZnO film	Pulsed laser deposition	ZnO Powder / ITO coated glass	target-substrate distance: 50 nm Chamber background pressure: $4 \times 10^{-3}$ Pa Oxygen pressure: 5 or 50 mTorr Deposition Temperature: 50–650°C Incident Laser pulses: 5000 Laser frequency: 8 Hz Laser beam power density: 0.85 J cm <sup>-2</sup>	Excellent / High	42
ZnO film	Pulsed laser deposition	ZnO ceramic plate / Glass	target-substrate distance: 6 cm Chamber base pressure: 10 <sup>-7</sup> Torr oxygen partial pressure: 50 and 100 mTorr Substrate temperature: 250 °C Energy per pulse: 300 mJ Incident Laser pulses: 6000 and 24000	Excellent / High	43
ZnO film	Radio frequency (RF) magnetron sputtering	Zn, O <sub>2</sub> / Corning Glass	RF power supply: 150 W target-substrate distance: 50 mm Deposition Temperature: 500°C Sputtering pressure : 1.0 Pa Chamber evacuated pressure: $5 \times 10^{-4}$ Pa Argon: oxygen ratio: 2sccm:18sccm, 6sccm:14sccm, 10sccm:10sccm and 14sccm:6sccm.	Excellent / High	44
ZnO film	Direct current (DC) magnetron sputtering	ZnO / Glass	Deposition Temperature: RT to $450^{\circ}$ C Deposition pressure : 12 mTorr to 25 mTorr Film thicknesses: 150 nm to 700 nm target-substrate distance: 70 mm Chamber base pressure: $2 \times 10^{-6}$ Torr Deposition time: 10 to 70 min	Excellent / High	45
ZnO film	Direct current (DC) magnetron sputtering	ZnO / Corning Glass	Target-substrate distance: 7 cm Ar Flow Rate: 26 -28 sccm Substrate Temperature: 27 °C Sputtering Power: 100 W Deposition Time: 10 min Deposition pressure : 12 mTorr to 20 mTorr	Excellent / High	46
ZnO film	Ion beam sputtering	ZnO / Si and FSD	Working and background pressure: $6.5 \times 10^{-4} \& 3 \times 10^{-6}$ Torr Total Ar & O <sub>2</sub> gas flow rate: 15 sccm Ar & O <sub>2</sub> ratio: 10/0 to 5/5 Substrate Temperatures: 125,190,250 and 300 °C Ion beam voltage & current: 800V & 150 mA Deposition Time: 90 min	Excellent / High	47
ZnO film	Ion beam sputtering	Zn, O <sub>2</sub> / Si	Chamber base pressure: $5 \times 10^{-6}$ Torr Ar Flow Rate: 10 sccm Ion beam voltage & current: 1000V & 20 mA oxygen partial pressure: 2 to 20 $10^{-5}$ Torr	Excellent / High	48



Chemical deposition methods (CDM) / Zinc oxide thin film deposition									
Thin film	Methods	Source materials / Substrate	Conditions for deposition parameters	Quality of film / Cost	Ref.				
ZnO film	Sol–gel	zinc acetate dihydrate, ethanol, ethanolamine / Glass	Rotating speed: 3000 rpm for 60s Pre & post Annealing temperature: 300 & 800°C for 1 h in air atmosphere precursor concentrations: 0.5 M to 1 M	Good / Low	49				
ZnO film	Sol–gel	zinc acetate dihydrate, ethanolamine, ethanol / Si	Rotating speed: 1000, 2000, 3000 rpm precursor concentrations: 0.75 M pre Annealing temperature: 300°C for 10 min in air atmosphere Post Annealing temperature: 400°C for 3 hr in N <sub>2</sub> and air atmosphere	Good / Low	50				
ZnO film	Chemical bath deposition	zinc chloride, ammonia / Glass	Bath solution concentrations: 0.1 M pH: 8.24 Temperature: 80 to 85 °C Annealing temperature: 400°C for 1 h Bath time: 15 min to 1 hr	Excellent / Low	51				
ZnO film	Chemical bath deposition	zinc nitrate, ammonia / Sodium Glass	Temperature: 80 °C Deposition time: 30 to 120 min zinc nitrate & ammonia concentrations: 0.1M & 0.1M,0.05M,0.01M Annealing temperature: 500°C for 1 hr in 1°C / min rates	Excellent / Low	52				
ZnO film	spray pyrolysis	Zinc acetate dehydrate / Glass	solution concentrations: 0.05M Substrate nozzle distance: 30 cm Spraying rate: 0.5 ml/min Spraying time: 10 min Substrate temperature: 473K to 673K Annealing temperature: 673K for 2 hr	Excellent / Low	53				
ZnO film	spray pyrolysis	Zinc acetate dehydrate / Glass	Substrate temperature: 350°C solution concentrations: 0.1 M Substrate nozzle distance: 28 cm Spraying rate: 3 ml/min Annealing temperature: 450°C for 1 hr in 10°C / min rates	Excellent / Low	54				
ZnO film	chemical vapor deposition	ZnO & graphite powders, O <sub>2</sub> / Silicon, sapphire	Substrate temperature: 720, 820 and 920°C Deposition time: 1 hr O <sub>2</sub> flow rate: 100,500 & 1000 sccm	Excellent / Low	55				
ZnO film	Metal organic chemical vapor deposition	Diethylzinc, O <sub>2</sub> / sapphire	Substrate temperature: 300 to 600°C Reactor pressure: 30 & 60 Torr Growth pressure: 50 Torr Rotation speed: 600 to 1200 rpm	Excellent / Low	56				



	(MO- CVD)				
ZnO film	Electro deposition	Zinc nitrate hexahydrate, sodium thiosulfate pentahydrate / Cu and ITO-coated glass	Temperature: 90 °C Film Thickness: 350 nm pH: 5.74 Applied potential: -0:60 V for 30 min Annealing temperature: 100°C in air	Excellent / Low	57
ZnO film	Electro deposition	zinc chloride, potassium chloride / FTO glass	Temperature: 70 °C Film Thickness: 350 nm pH: 5.74 Applied potential: -0.1 V for 10 min Annealing temperature: 400°C for 1 hr	Excellent / Low	58
ZnO film	Atomic layer deposition	Diethylzinc, water / Si or SiO <sub>2</sub> /Si	Growth temperatures: 150 to 400 °C Rector pressure: O.6 Torr Ar flow rate: 1600sccm Deposition cycles: 100–300	Excellent / Low	59
ZnO film	Atomic layer deposition	Diethylzinc, water / Si	Deposition temperature: 100 to $280^{\circ}$ C Deposition cycles: 10–1200 N <sub>2</sub> flow rate: 20 sccm	Excellent / Low	60
ZnO film	SILAR	zinc sulphate heptahydrate, ammonia / Glass	Annealing temperature: 100 °C for 16 hr Deposition cycles: 25,50,75 and 100	Excellent / Low	61
ZnO film	SILAR	zinc sulphate, sodium hydroxide / FTO coated glass	Rinsing temperature: 70 °C pH: 6 Number of cycles: 150 con of cationic & anionic precursor: 0.1M Immersion Time: 30 s	Excellent / Low	62

## 3. Conclusion:

Thin film is an important branch of emerging technology and science. In this review, the preparation of copper and zinc oxide films by using various physical and chemical deposition methods will be investigated and the deposition conditions of each method have been analyzed in detail. From our research work we are able to gain a clear understanding of the deposition of low-cost high-quality copper and zinc oxide thin films. Finally compared to physical methods, it is clear that chemical methods are used to produce high quality thin films at low economic cost.

## **References:**

- Rossnagel, S. M. (2003). Thin film deposition with physical vapor deposition and related technologies. Journal of Vacuum Science & Technology A: Vacuum, Surfaces, and Films, 21(5), S74– S87. <u>https://doi.org/10.1116/1.1600450</u>.
- [2] Benelmekki, M., & Erbe, A. (2019). *Nanostructured thin films–background, preparation and relation to*

the technological revolution of the 21st century. Nanostructured Thin Films, 1– 34. <u>https://doi.org/10.1016/B978-0-08-102572-</u> 7.00001-5.

- [3] Jawad, M. F., Ismail, R. A., & Yahea, K. Z. (2011). Preparation of Nanocrystalline CU2O thin film by pulsed laser deposition. *Journal of Materials Science: Materials in Electronics*, 22(9), 1244–1247. https://doi.org/10.1007/s10854-011-0294-0.
- [4] Papadimitropoulos, G., Vourdas, N., Vamvakas, V. E., & Davazoglou, D. (2005). Deposition and characterization of Copper Oxide Thin Films. *Journal* of *Physics: Conference Series*, 10, 182–185. <u>https://doi.org/10.1088/1742-6596/10/1/045</u>.
- [5] Darma, T. H., Ogwu, A. A., & Placido, F. (2011). Effects of sputtering pressure on properties of copper oxide thin films prepared by RF magnetron sputtering. *Materials Technology*, 26(1), 28–31. https://doi.org/10.1179/175355511x12941605982226.
- [6] Sreedev, P., Rakhesh, V., Roshima, N. S., & Shankar,



B. (2019). Preparation of zinc oxide thin films by SILAR method and its optical analysis. *Journal of Physics: Conference Series*, *1172*, 012024. https://doi.org/10.1088/1742-6596/1172/1/012024.

- [7] Lu, J. G., Kawaharamura, T., Nishinaka, H., Kamada, Y., Ohshima, T., & Fujita, S. (2007). Zno-based thin films synthesized by atmospheric pressure mist chemical vapor deposition. *Journal of Crystal Growth*, 299(1), 1–10. <u>https://doi.org/10.1016/j.jcrysgro.2006.10.251</u>.
- [8] Raidou, A., Benmalek, F., Sall, T., Aggour, M., Qachaou, A., Laanab, L., & Fahoume, M. (2014). Characterization of zno thin films grown by Silar Method. OALib, 01(03), 1–9. <u>https://doi.org/10.4236/oalib.1100588</u>.
- [9] Al-Kuhaili, M. F. (2008). Characterization of copper oxide thin films deposited by the thermal evaporation of cuprous oxide (cu2o). *Vacuum*, 82(6), 623–629. <u>https://doi.org/10.1016/j.vacuum.2007.10.004</u>.
- Gevorkyan, V. A., Reymers, A. E., Nersesyan, M. N., & Arzakantsyan, M. A. (2012). Characterization of cu2O thin films prepared by evaporation of Cuo Powder. *Journal of Physics: Conference Series*, 350, 012027. <u>https://doi.org/10.1088/1742-6596/350/1/012027</u>.
- [11] Kawaguchi, K., Kita, R., Nishiyama, M., & Morishita, T. (1994). Molecular beam epitaxy growth of Cuo and CU2O Films with controlling the oxygen content by the flux ratio of Cu/O+. *Journal of Crystal Growth*, *143*(3–4), 221–226. <u>https://doi.org/10.1016/0022-0248(94)90059-0</u>.
- [12] Yang, K. G., Hu, P., Wu, S. X., Ren, L. Z., Yang, M., Zhou, W. Q., Yu, F. M., Wang, Y. J., Meng, M., Wang, G. L., & Li, S. W. (2016). Room-temperature ferromagnetic Cuo thin film grown by plasmaassisted molecular beam epitaxy. *Materials Letters*, *166*, 23–25. https://doi.org/10.1016/j.matlet.2015.11.128.
- [13] Farhad, S. F., Cherns, D., Smith, J. A., Fox, N. A., & Fermín, D. J. (2020). Pulsed laser deposition of single phase N- and p-type cu20 thin films with low resistivity. *Materials & amp; Design, 193*, 108848. https://doi.org/10.1016/j.matdes.2020.108848.
- [14] Al-Ghamdi, A. A., Khedr, M. H., Shahnawaze Ansari, M., Hasan, P. M. Z., Abdel-wahab, M. Sh., & Farghali, A. A. (2016). RF sputtered Cuo thin films: Structural, optical and photo-catalytic behavior. *Physica E: Low-Dimensional Systems and Nanostructures*, 81, 83–90. https://doi.org/10.1016/j.physe.2016.03.004.
- [15] Patwary, M. A., Saito, K., Guo, Q., & Tanaka, T. (2019). Influence of oxygen flow rate and substrate positions on properties of Cu-oxide thin films

fabricated by radio frequency magnetron sputtering using pure Cu target. *Thin Solid Films*, 675, 59–65. https://doi.org/10.1016/j.tsf.2019.02.026.

- [16] Chu, C.-L., Lu, H.-C., Lo, C.-Y., Lai, C.-Y., & Wang, Y.-H. (2009). Physical properties of copper oxide thin films prepared by DC reactive magnetron sputtering under different oxygen partial pressures. *Physica B: Condensed Matter*, 404(23–24), 4831–4834. https://doi.org/10.1016/j.physb.2009.08.185.
- [17] Shukor, A. H., Alhattab, H. A., & Takano, I. (2020). Electrical and optical properties of copper oxide thin films prepared by DC magnetron sputtering. *Journal* of Vacuum Science & Compton Proceedings 10, 38(1), 012803. <u>https://doi.org/10.1116/1.5131518</u>.
- [18] Dolai, S., Dey, R., Das, S., Hussain, S., Bhar, R., & Pal, A. K. (2017). Cupric oxide (Cuo) thin films prepared by reactive D.C. magnetron sputtering technique for photovoltaic application. *Journal of Alloys and Compounds*, 724, 456–464. <u>https://doi.org/10.1016/j.jallcom.2017.07.061</u>.
- [19] Horak, P., Bejsovec, V., Vacik, J., Lavrentiev, V., Vrnata, M., Kormunda, M., & Danis, S. (2016). Thin copper oxide films prepared by ion beam sputtering with subsequent thermal oxidation: Application in chemiresistors. *Applied Surface Science*, 389, 751– 759. <u>https://doi.org/10.1016/j.apsusc.2016.07.143</u>.
- [20] Halin, D. S., Talib, I. A., Daud, A. R., & Hamid, M. A. (2014). Characterizations of cuprous oxide thin films prepared by sol-gel spin coating technique with different additives for the photoelectrochemical solar cell. *International Journal of Photoenergy*, 2014, 1–6. https://doi.org/10.1155/2014/352156.
- [21] Halin, Dewi Suriyani, Talib, I. A., Daud, A. R., & Hamid, M. A. (2013). Effect of annealing atmosphere on the morphology of copper oxide thin films deposited on tio2 substrates prepared by sol-gel process. *Key Engineering Materials*, 594–595, 113– 117.

https://doi.org/10.4028/www.scientific.net/kem.594-595.113.

- [22] Sadiq, Z. M., Hassan, M. A., & Hassoon, K. I. (2022). Preparation and characterization of Cuo nanostructured thin films by chemical bath deposition. *Journal of Physics: Conference Series*, 2322(1), 012088. <u>https://doi.org/10.1088/1742-6596/2322/1/012088</u>.
- [23] Saadaldin, N., Alsloum, M. N., & Hussain, N. (2015). Preparing of copper oxides thin films by chemical bath deposition (CBD) for using in environmental application. *Energy Procedia*, 74, 1459–1465. <u>https://doi.org/10.1016/j.egypro.2015.07.794</u>.
- [24] Dhas, C. R., Alexander, D., Christy, A. J., Jeyadheepa, K., Raj, A. M., & Raja, C. S. (2014).

Preparation and characterization of Cuo Thin Films prepared by spray pyrolysis technique for ethanol gas sensing application. *Asian Journal of Applied Sciences*, 7(8), 671–684. https://doi.org/10.3923/ajaps.2014.671.684.

- [25] Lamri Zeggar, M., Chabane, L., Aida, M. S., Attaf, N., & Zebbar, N. (2015). Solution flow rate influence on properties of copper oxide thin films deposited by ultrasonic spray pyrolysis. *Materials Science in Semiconductor Processing*, 30, 645–650. <u>https://doi.org/10.1016/j.mssp.2014.09.026</u>.
- [26] Maruyama, T. (1998). Copper oxide thin films prepared by chemical vapor deposition from copper dipivaloylmethanate. Solar Energy Materials and Solar Cells, 56(1), 85–92. https://doi.org/10.1016/s0927-0248(98)00128-7.
- [27] Chua, D., Kim, S. B., Li, K., & Gordon, R. (2019). Low temperature chemical vapor deposition of cuprous oxide thin films using a copper(i) amidinate precursor. ACS Applied Energy Materials, 2(11), 7750–7756. <u>https://doi.org/10.1021/acsaem.9b01683</u>.
- [28] Jeong, S., & Aydil, E. S. (2010). Structural and electrical properties of cu20 thin films deposited on zno by metal organic chemical vapor deposition. *Journal of Vacuum Science & amp; Technology A: Vacuum, Surfaces, and Films, 28*(6), 1338–1343. https://doi.org/10.1116/1.3491036.
- [29] Eisermann, S., Kronenberger, A., Laufer, A., Bieber, J., Haas, G., Lautenschläger, S., Homm, G., Klar, P. J., & Meyer, B. K. (2011). Copper oxide thin films by chemical vapor deposition: Synthesis, characterization and electrical properties. *Physica Status Solidi (a)*, 209(3), 531–536. https://doi.org/10.1002/pssa.201127493.
- [30] Rahman, A. S. M. S., Islam, M. A., & Shorowordi, K. M. (2015). Electrodeposition and characterization of copper oxide thin films for solar cell applications. *Procedia Engineering*, 105, 679–685. <u>https://doi.org/10.1016/j.proeng.2015.05.048</u>.
- [31] Zhao, W., Fu, W., Yang, H., Tian, C., Li, M., Li, Y., Zhang, L., Sui, Y., Zhou, X., Chen, H., & Zou, G. (2011). Electrodeposition of CU2O Films and their photoelectrochemical properties. *CrystEngComm*, *13*(8), 2871. <u>https://doi.org/10.1039/c0ce00829j</u>.
- [32] Iivonen, T., Heikkilä, M. J., Popov, G., Nieminen, H.-E., Kaipio, M., Kemell, M., Mattinen, M., Meinander, K., Mizohata, K., Räisänen, J., Ritala, M., & Leskelä, M. (2019). Atomic layer deposition of photoconductive cu2O thin films. *ACS Omega*, 4(6), 11205–11214. https://doi.org/10.1021/acsomega.9b01351.
- [33] Tamm, A., Tarre, A., Verchenko, V., Seemen, H., & Stern, R. (2020). Atomic layer deposition of

superconducting cuo thin films on three-dimensional substrates. *Crystals*, *10*(8), 650. https://doi.org/10.3390/cryst10080650.

- [34] ÇAYIR TAŞDEMİRCİ, T. (2020). Copper oxide thin films synthesized by silar: Role of varying annealing temperature. *Electronic Materials Letters*, 16(3), 239– 246. <u>https://doi.org/10.1007/s13391-020-00205-4</u>.
- [35] Lee, W.-J., & Wang, X.-J. (2021). Structural, optical, and electrical properties of copper oxide films grown by the SILAR method with post-annealing. *Coatings*, *11*(7), 864. <u>https://doi.org/10.3390/coatings11070864</u>.
- [36] Eskandari, A., Sangpour, P., & Vaezi, M. R. (2014). Hydrophilic CU2O nanostructured thin films prepared by facile spin coating method: Investigation of Surface Energy and roughness. *Materials Chemistry* and Physics, 147(3), 1204–1209. https://doi.org/10.1016/j.matchemphys.2014.07.008.
- [37] Patil, V., Jundale, D., Pawar, S., Chougule, M., Godse, P., Patil, S., Raut, B., & Sen, S. (2011). Nanocrystalline Cuo Thin films for H2S monitoring: Microstructural and optoelectronic characterization. *Journal of Sensor Technology*, 01(02), 36–46. <u>https://doi.org/10.4236/jst.2011.12006</u>.
- [38] Ahmed, G., Mahmoud, T., Mohammed, H., & Hussein, A. (2021). Fabrication and study of zno thin films using thermal evaporation technique. *Egyptian Journal of Chemistry*, 64(9), 5183–5191. <u>https://doi.org/10.21608/ejchem.2021.71184.3563</u>.
- [39] Faraj, M. G., & Ibrahim, K. (2011). Optical and structural properties of thermally evaporated zinc oxide thin films on polyethylene terephthalate substrates. *International Journal of Polymer Science*, 2011, 1–4. <u>https://doi.org/10.1155/2011/302843</u>.
- [40] Ramamoorthy, K., Sanjeeviraja, C., Jayachandran, M., Sankaranarayanan, K., Bhattacharya, P., & Kukreja, L. M. (2001). Preparation and characterization of zno thin films on INP by lasermolecular beam epitaxy technique for solar cells. *Journal of Crystal Growth*, 226(2–3), 281–286. https://doi.org/10.1016/S0022-0248(01)01393-8.
- [41] Kim, M. S., Yim, K. G., Leem, J.-Y., Kim, S., Nam, G., Lee, D.-Y., Kim, J. S., & Kim, J. S. (2011). Thickness dependence of properties of zno thin films on porous silicon grown by plasma-assisted molecular beam epitaxy. *Journal of the Korean Physical Society*, 59(3), 2354–2361. https://doi.org/10.3938/jkps.59.2354.
- [42] Franklin, J. B., Zou, B., Petrov, P., McComb, D. W., Ryan, M. P., & McLachlan, M. A. (2011). Optimised pulsed laser deposition of zno thin films on transparent conducting substrates. *Journal of Materials Chemistry*, 21(22), 8178. https://doi.org/10.1039/c1jm10658a.



- [43] Villanueva, Y. Y., Liu, D.-R., & Cheng, P. T. (2006).
   Pulsed laser deposition of zinc oxide. *Thin Solid Films*, 501(1–2), 366–369.
   <u>https://doi.org/10.1016/j.tsf.2005.07.152</u>.
- [44] Dave, P. Y., Patel, K. H., Chauhan, K. V., Chawla, A. K., & Rawal, S. K. (2016). Examination of zinc oxide films prepared by magnetron sputtering. *Procedia Technology*, 23, 328–335. <u>https://doi.org/10.1016/j.protcy.2016.03.034</u>.
- [45] Hoon, J.-W., Chan, K.-Y., Krishnasamy, J., Tou, T.-Y., & Knipp, D. (2011). Direct current magnetron sputterdeposited zno thin films. *Applied Surface Science*, 257(7), 2508–2515. <u>https://doi.org/10.1016/j.apsusc.2010.10.012</u>.
- [46] Krishnasamy, J., Kah-Yoong Chan, Jian-Wei Hoon, Sharul Ashikin Binti Kamaruddin, & Teck-Yong Tou. (2010). Direct current magnetron sputter-deposited zno thin films. *International Conference On Photonics 2010*. <u>https://doi.org/10.1109/icp.2010.5604395</u>.
- [47] Tsai, H.-Y. (2007). Characteristics of zno thin film deposited by Ion Beam sputter. *Journal of Materials Processing Technology*, 192–193, 55–59. <u>https://doi.org/10.1016/j.jmatprotec.2007.04.029</u>.
- [48] Hsu, J.-C., & Chiang, Y.-S. (2013). Influence of oxygen on zinc oxide films fabricated by ion-beam sputter deposition. *ISRN Materials Science*, 2013, 1–7. <u>https://doi.org/10.1155/2013/710798</u>.
- [49] Aryanto, D., Jannah, W. N., Masturi, Sudiro, T., Wismogroho, A. S., Sebayang, P., Sugianto, & Marwoto, P. (2017). Preparation and structural characterization of zno thin films by Sol-Gel Method. *Journal of Physics: Conference Series*, 817, 012025. <u>https://doi.org/10.1088/1742-6596/817/1/012025</u>.
- [50] Ben Moussa, N., Lajnef, M., Jebari, N., Villebasse, C., Bayle, F., Chaste, J., Madouri, A., Chtourou, R., & Herth, E. (2021). Synthesis of zno sol-gel thin-films CMOScompatible. *RSC Advances*, *11*(37), 22723–22733. <u>https://doi.org/10.1039/d1ra02241e</u>.
- [51] Sreedev, P., Rakhesh, V., & Roshima, N. S. (2018). Optical characterization of zno thin films prepared by chemical bath deposition method. *IOP Conference Series: Materials Science and Engineering*, 377, 012086. <u>https://doi.org/10.1088/1757-899x/377/1/012086</u>.
- [52] Fu, Y. P., & Chen, J. J. (2014). Characteristics of zinc oxide film prepared by chemical bath deposition method. *Key Engineering Materials*, 602–603, 871–875. <u>https://doi.org/10.4028/www.scientific.net/kem.602-603.871.</u>
- [53] Rajendra, B. V., Bhat, V., & Kekuda, D. (2014). Optical properties of zinc oxide (zno) thin films prepared by spray pyrolysis method. *Advanced Materials Research*, 895, 226– 230. https://doi.org/10.4028/www.scientific.net/amr.895.226.
- [54] Komaraiah, D., Radha, E., Vijayakumar, Y., Sivakumar, J., Reddy, M. V., & Sayanna, R. (2016). Optical, structural and morphological properties of photocatalytic zno thin films deposited by pray pyrolysis technique. *Modern Research in Catalysis*, 05(04), 130–146. https://doi.org/10.4236/mrc.2016.54011.

- [55] Zhuo Chen, Shum, K., Salagaj, T., Wei Zhang, & Strobl, K. (2010). ZnO thin films synthesized by chemical vapor deposition. 2010 IEEE Long Island Systems, Applications and Technology Conference. <u>https://doi.org/10.1109/lisat.2010.5478331</u>.
- [56] Fenwick, W. E., Woods, V. T., Pan, M., Li, N., Kane, M. H., Gupta, S., Rengarajan, V., Nause, J., & Ferguson, I. T. (2005). Metal organic chemical vapor deposition of zinc oxide. *MRS Proceedings*, 892. <u>https://doi.org/10.1557/proc-0892-ff18-07-ee09-07</u>.
- [57] Rahal, H., Kihal, R., Affoune, A. M., Ghers, M., & Djazi, F. (2017). Electrodeposition and characterization of zno thin films using sodium thiosulfate as an additive for photovoltaic solar cells. *Journal of Semiconductors*, 38(5), 053002. <u>https://doi.org/10.1088/1674-4926/38/5/053002</u>.
- [58] Saravanan, S., & Dubey, R. S. (2019a). Optical and structural studies of zno thin film prepared by electrodeposition method. 7TH NATIONAL CONFERENCE ON HIERARCHICALLY STRUCTURED MATERIALS (NCHSM-2019). https://doi.org/10.1063/1.5114581.
- [59] Min, Y.-S., An, C.-J., Kim, S.-K., Song, J.-W., & Hwang, C.-S. (2010). Growth and characterization of conducting zno thin films by atomic layer deposition. *Bulletin of the Korean Chemical Society*, 31(9), 2503–2508. <u>https://doi.org/10.5012/bkcs.2010.31.9.2503</u>.
- [60] Hussin, R., Hou, X. H., & Choy, K. L. (2012). Growth of zno thin films on silicon substrates by atomic layer deposition. *Defect and Diffusion Forum*, 329, 159–164. <u>https://doi.org/10.4028/www.scientific.net/ddf.329.159</u>.
- [61] Santhamoorthy, A., Srinivasan, P., Krishnakumar, A., Rayappan, J. B., & Babu, K. J. (2021). Silar-deposited nanostructured zno thin films: Effect of deposition cycles on surface properties. *Bulletin of Materials Science*, 44(3). <u>https://doi.org/10.1007/s12034-021-02465-8</u>.
- [62] D. Dhaygude, H., K. Shinde, S., B. Velhal, N., Lohar, G. M., & J. Fulari, V. (2016). Synthesis and characterization of zno thin film by low cost modified silar technique. *AIMS Materials Science*, *3*(2), 349–356. <u>https://doi.org/10.3934/matersci.2016.2.349</u>.