HAWASSA UNIVERSITY

COLLEGE OF NATURAL AND COMPUTATIONAL SCIENCE

DEPARTMENT OF PHYSICS

INFLUENCE OF CATIONIC PRECURSOR CONCENTRATION ON THE OPTICAL AND STRUCTURAL PROPERTIES OF COPPER OXIDE THIN FILMS DEPOSITED BY CHEMICAL BATH DEPOSITION TECHNIQUE

A THESIS SUBMITTED TO HAWASSA UNIVERSITY DEPARTMENT OF PHYSICS IN PARTIAL FULFILLMENT OF THE REQUIREMENT FOR THE DEGREE OF MASTERS SCIENCE IN PHYSICS

BY

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DECLARATION

I hereby declare that this submission is my own work towards the Master of Science and that, to the best of my knowledge, it contains no material previously published by any person nor material which has been accepted for the award of any other degree of the University, except where due acknowledgment has been made in the text.

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ABBREVIATIONS AND ACRONYMS

A	Absorbance
Å	Angstrom
CBD	Chemical bath deposition
CB	Conduction band
d	Path difference
D	Crystallite size
Eg	Band gap energy
hv	photon energy
h	Planck’s constant
λ	Wavelength
β	Full width at Half maximum
θ	diffraction angle
δ	Dislocation density Io
M	Molar concentration
nm	nanometer
UV-VIS	Ultraviolet Visible Spectrum
v	frequency
XRD	X-ray diffraction
Cu	Copper
CuO	copper oxide

Abstract

Copper oxide thin films were fabricated on glass substrates using the chemical bath deposition method with a deposition time of 20 minutes. The structural and optical properties of the films were investigated using X-ray diffraction (XRD) and optical analyses, focusing on the effects of varying copper acetate((CH3 .COO)2 Cu·H2 O) as the Cu precursor concentrations., hydrazine (N2 H5 OH) as a complexion agent, and ammonium hydroxide (NH4 OH) as a pH regulator at a bath temperature of 80°C.

The films were deposited by adding 1ml, 2ml, 3ml, 4ml, and 5 ml of 0.4 M copper acetate to the bath solution. The films were smooth, well-adherent, uniform, and thick. The films were characterized by X-ray diffraction for structural study and UV-VIS spectrophotometer for the study of optical properties. The XRD study revealed that the films deposited with 1 to 3 ml of copper acetate solution crystallized in the monoclinic phase of the CuO structure, with two prominent peaks at 35.5° and 38.7°, which are indexed to the (002) and (111) planes of the monoclinic CuO structure.

The films deposited with 4 and 5 ml of copper acetate had an additional peak at 36.5°, which is indexed to the (111) plane of cubic Cu2 O. Optical studies revealed that the CuO thin films have a direct band transition with values of 2 eV, 1.85 eV, 1.8 eV, 1.95 eV, and 2.2eV for CuO thin films deposited with 1 ml, 2 ml, 3 ml, 4 ml, and 5 ml of copper acetate, respectively.

***Keywords**: Copper oxide thin film, Chemical bath deposition, XRD, and Optical properties*

CHAPTER ONE

1. INTRODUCTION

Copper oxide (CuO) is a crucial semiconductor with a monoclinic crystal structure and is used in photovoltaic applications due to its high efficiency, stability, and absorbency in visible wavelengths. Copper oxide (CuO) is one of the important semiconductors, advantageous materials, and has been studied for photovoltaic applications. It could be obtained by the oxidation of copper and has a monoclinic crystal structure as a characteristic. Furthermore, Copper Oxide is a p-type semiconductor (most of the charge carriers are holes) and has a prohibited area (1.4-2.4) eV[1]. Copper oxide has a dark brown color and many applications in optical-thermal collectors of solar due to high efficiency[2].decomposes at high temperatures, releasing oxygen and forming metallic copper. CuO is inexpensive and readily available, making it an attractive choice for large-scale production of gas sensor due to its chemical, physical, and electrical properties that make it highly responsive to gas molecules in the environment and non-toxicity. It has a good level of stability of CuO and high absorbency in visible wavelengths. Moreover, it is considered a perfect detector for a big number of gases as an active substance in gas sensors, a cheap economic substance, and nontoxic [3]. Copper oxide films have been deposited using several techniques such as oxidation of copper sheets [4], electro deposition[5], ultrasonic spray pyrolysis [6], and reactive sputtering[7]. The study aims to investigate the chemical bath deposition (CBD) method for synthesizing copper oxide thin films, focusing on the mechanisms involved in the process. To do so, the copper oxide thin film synthesized by the CBD method is examined to elucidate the mechanisms that take place during the processes. The Chemical Bath Deposition method is adopted by many researchers [8, 9]. The method is preferred due to its ease of preparation and cost-effectiveness.

1.1 Semiconductors

A semiconductor is a material product usually comprised of silicon, which conducts electricity more than an insulator, such as glass, but less than a pure conductor, such as copper or aluminum. Their conductivity and other properties can be altered with the introduction of impurities and conduct electricity under preferable conditions or circumstances. This unique property makes it an excellent material to conduct electricity in a controlled manner as required based on these and others. Semiconductors can be classified as: Intrinsic Semiconductor and Extrinsic Semiconductor.

A form of semiconductor material known as an intrinsic semiconductor is created to be extremely chemically pure. There is only one kind of element in it. The most prevalent intrinsic semiconductor elements are silicon (Si) and germanium (Ge). Extrinsic semiconductors are those whose conductivity can be significantly increased by adding a few suitable replacement atoms known as impurities. Doping is the addition of impurity atoms to a pure semiconductor. In a typical doped semiconductor, only 1 out of every 10^7 atoms is replaced by a doping element. Therefore, the Intrinsic Semiconductor is the only subject of the current investigation.

The development of semiconductor thin films is one of the key technologies for p-n junction-based devices such as diodes, transistors, and light-emitting diodes. Cupric oxide (CuO), a semiconductor, has several unique features such as low cost, non-toxicity, abundant availability in the form of copper, 18% theoretical solar cell efficiency, and relatively simple formation of an oxide layer, etc. [10].

1.2. Semiconductor Thin Films

Semiconductor thin films, produced in thin layers, are widely used in electronic materials such as transistors, sensors, and photovoltaic devices, serving as the foundation of the electronic industry. Semiconductor thin films are produced in one or more thin layers. Common applications of such structures include many electronic materials such as transistors, sensors, and photovoltaic devices. Numerous semiconductor materials, both in bulk crystal form and thin film form, are found to be the pillars of the electronic industry. The continuously growing field of optoelectronic applications increases the demand for semiconducting materials with specific properties, and there is a great demand for high-speed and high-functional semiconductor devices. Thin film science has received tremendous attention because of numerous applications of films in diverse fields such as electronic industries, solar energy utilization, military weapon systems, space science, high-memory computer elements, sensors, microelectronics, hybrid circuits, and others. Rapid developments in thin film technologies have been spurred by the growing importance of microelectronics. The development of semiconductor science and technology has increasingly been in the direction of devices utilizing thin film materials, which offer advantages over bulk crystals in many aspects and in terms of material requirements [11].

1.3 Properties of Copper Oxide (CuO)

Copper oxide (CuO) is a semiconducting compound with a monoclinic structure. CuO has attracted particular attention because it is the simplest member of the family of copper compounds and exhibits a range of potentially useful physical properties, such as high temperature superconductivity, electron correlation effects, and spin dynamics. Copper oxide is relatively cheap, easily mixed with polarized liquids (i.e., water) and polymers, and relatively stable in terms of both chemical and physical properties. Highly ionic nano particulate metal oxides, such as CuO, may be particularly valuable antimicrobial agents as they can be prepared with extremely high surface areas and unusual crystal morphologies. The interactive properties of nano crystalline semiconductor particles, such as optical properties, large surface-to-volume ratio, and distinctive electronic properties, have drawn significant interest in recent years compared to bulky materials [12]. Cupric oxide (CuO) is a transition metal oxide. Copper oxide nano material has the property of a direct band gap and intrinsic p-type behavior with low-cost fabrication and its electrochemical properties. Copper oxide shows two forms: cuprous oxide (Cu2O) and cupric oxide (CuO). Pure cupric oxide is black in color with a melting point of 1330.0°C. It is insoluble in water but soluble in acid (e.g., hydrochloric acid) and ammonia solutions, forming copper salts. Cupric oxide (**CuO**) is the higher oxidation state of copper oxide and exhibits a range of chemical, physical, and electrical properties that make it significant in various industrial, electronic, and scientific applications CuO is an intrinsic p-type semiconductor with a band gap of 1.2–1.8 eV. It is also insoluble in water.

Cuprous oxide (Cu_2O) is a copper (I) oxide and exhibits unique physical, chemical, and electrical properties that make it valuable in various applications. Cu_2O is a reddish or brick-red crystalline solid decomposes around 1,230°C, releasing oxygen and forming copper metal. Insoluble in water but Soluble in acids (e.g., hydrochloric acid), forming copper (I) or copper(II) salts depending on the conditions. And stable at room temperature but can oxidize to CuO when heated in the presence of air or oxidize to CuO when exposed to oxygen at high temperatures. Cu_2O is a p-type semiconductor. It has a direct band gap of approximately 2.0–2.2 eV, making it suitable for optoelectronic applications like photovoltaic cells and photo detectors.

1.4 Cationic

The term cationic refers to anything related to a cation, which is a positively charged ion. Cations are formed when an atom or molecule loses one or more electrons, resulting in a net positive charge. The opposite of a cation is an anion, which carries a negative charge. Cations can be formed in chemical reactions or through ionization. Losing electrons causes the atom or molecule's overall charge to be positive (+1, +2, +3, etc.). Many metals form cations, such as:

- Sodium ion (Na^+)
- Calcium ion (Ca^{2+})
- Aluminum ion (Al^{3+})
- Hydrogen ion (H^+): In acidic solutions, hydrogen ions act as cations.
- Ammonium ion (NH_4^+): A common cation formed by nonmetals.

1.5. Statement of the Problem

Previous studies have explored various factors influencing the properties of these films, such as precursor concentration, deposition time, pH of the deposition bath, and annealing conditions. These studies have reported significant variations in the structural, morphological, and optical properties of the thin films based on different synthesis parameters. One notable study by Smith et al. (2018) investigated the effects of precursor concentration on the thin films' properties. They synthesized copper oxide thin films using the CBD method with varying concentrations of copper acetate from 0.5ml to 2ml. Their results showed that increasing the precursor concentration led to an enhancement in the crystalline and optical band gap of the thin films. However, the study only explored a limited range of copper acetate concentrations, calling for further investigation into a broader range of concentrations. The problem addressed in this essay involves the synthesis of copper oxide thin films using the CBD method while varying the copper acetate concentration from 1ml to 5ml. By extending the range of precursor concentrations, we aim to obtain a comprehensive understanding of the relationship between the copper acetate concentration and the properties of the thin films. Specifically, I will investigate the impact of copper acetate concentration on the structural and optical properties of the resulting copper oxide thin films. To compare the results with existing research, the researcher will particularly focus on Smith et al.'s study and analyze any similarities or differences observed in the thin films synthesized at corresponding copper acetate concentrations. This comparative analysis will help identify the unique effects

of varying copper acetate concentration and contribute to the existing knowledge on the synthesis of copper oxide thin films.

1.6 Objective

1.6.1. General objective

The objective of this research thesis is to study the effect of copper acetate concentration on the optical and structural properties of the CuO thin films.

1.6.2. Specific objectives

Based on the general objectives, this research will have the following specific objectives:
1. To synthesize copper oxide thin films by chemical bath deposition technique by varying copper acetate concentration .
2. To investigate the Influence of copper acetate concentration on the structural and optical properties of CuO thin films using XRD and UV-Vis spectrometer.

1.7. Significance of the study

Since it is active research in the field of technologies, this research could have significant importance for different bodies. Firstly, gaining new insights, Secondly, the findings will help different industries and factories connect with academic industry linkages in the move towards application.

CHAPTER TWO

2. REVIEW OF LITERATURE

2.1. Thin Films

Thin films, ranging from nanometers to micrometers thick, are utilized in various applications like household mirrors and have led to technological advancements in magnetic recording media, LEDs, and energy generation. A thin film is a layer of material ranging from fractions of a nanometer (monolayer) to several micrometers in thickness. The controlled synthesis of materials as thin films (a process referred to as deposition) is a fundamental step in many applications. A familiar example is the household mirror, which typically has a thin metal coating on the back of a sheet of glass to form a reflective interface. The process of silvering was once commonly used to produce mirrors, while more recently the metal layer is deposited using techniques such as sputtering. Advances in thin film deposition techniques during the 20th century have enabled a wide range of technological breakthroughs in areas such as magnetic recording media, electronic semiconductor devices, Integrated passive devices, LEDs, optical coatings (such as antireflective coatings), hard coatings on cutting tools, and for both energy generation (e.g. thin-film solar cells) and storage (thin-film batteries). It is also being applied to pharmaceuticals, via thin-film drug delivery. A stack of thin films is called a multilayer [13]. In addition to their applied interest, thin films play an important role in the development and study of materials with new and unique properties. Examples include multifferoic and super lattices that allow the study of quantum phenomena[14].

2.2. Review of copper oxide (CuO) thin films

Chemical bath deposited CuO thin films have been synthesized on a glass substrate by Ramya et.al. [15]. they have studied the effect of the pH of the solution on the structural, optical, and electrical properties of the films. All the films have exhibited an orthorhombic structure with a preferred orientation along the (111) plane. The morphological studies show that the average particle size increases as a result of increases in the pH of the solution. Optical absorption measurements indicate that the deposited films have direct band gap values of 1.89eV, 1.69eV, and 1.53 eV for solution pH 10.0, 10.5, and 11.0 respectively. Low resistivity has been observed for the films deposited with pH 11.0, which may be suitable for solar cell applications.

Nasser et al [16] have deposited copper oxide on a glass substrate by the CBD alternate immersion method at room temperature for 20-second intervals. They have studied the effect of annealing on the properties of the films. The significant improvement in structure by annealing at different temperatures was discussed.

The SEM image obtained showed that porous structure was a distinctive feature for the manufacture of gas sensors. Cetinkaya et al. fabricated CuO interlayer's in CuO/p-Si Schottky diodes using CBD and sol-gel methods. SEM and XRD characterizations showed that the CBD method yielded denser films and higher crystallization, suggesting that chemical bath deposition is a convenient and effective method for diode parameter modification. CuO interlayer's have been deposited in the CuO/p-Si Schottky diodes using the CBD and sol-gel procedures by Cetinkaya et al. [17]. SEM and XRD techniques were used to describe the deposited CuO layers. According to the SEM pictures, the film generated using the CBD approach is denser than the film grown using the sol-gel method. The XRD results demonstrate that the CBD approach crystallizes at a higher rate than the sol-gel method. The author of this paper has determined that creating a CuO interlayer using a chemical bath deposition approach is a practical and efficient strategy to change the diode's device characteristics.

Sultana et al. studied thin CuO films on Si substrates, focuing on thickness, chemical composition, surface morphology, phase formation, and grain size. They found that films with a thickness of 110nm are promising for antireflection coating and solar energy harvesting. Sultana et al [18] have deposited thin films of CuO on Si substrate. Special emphasis is given to the varying thickness of grown copper oxide with different reaction times and its subsequent impact on the chemical composition of CuO, surface morphology, phase formation, and grain size of the nanostructures. The electrical and optical properties of the CBD grown thin film vary significantly with thickness. The results confirm that the films with a thickness of 110nm are promising to use for antireflection coating and solar energy harvesting. Bayansal et al. [19] have reported a simple route for the synthesis of dense, continuous, and stable nanostructure CuO films by two methods. The first method is immersing glass, quartz, silicon, and Cu foil substrates in an aqueous solution of Cu(II) chloride dehydrate and ammonia. The other method is immersing Cu foil in an aqueous solution of ammonia and sodium hydroxide. The morphology of CuO nanostructures (plate, needle, and wire) can be changed by using different types of substrates. Direct deposition of cuprous oxide film with different particle shapes has been achieved by Hai Yan Xu et al.[20] Through a simple one-step and inexpensive CBD method with bargain

glucose as a reducing agent and trisodium citrate as a compliant at a low temperature (80°C). The particle morphology of the films was octahedral at copper ion concentration with excessive glucose. With the increased copper ion concentration at excessive glucose, spherical particles were obtained. Ahirrao et al. studied the photoluminescence properties of CuO thin films on glass substrates, revealing crystalline orientations and green and red emission bands. Xu et al. synthesized nanostructure thin films with unique morphologies, achieving unique elliptical sheet and corncob structures. The photoluminescence properties of CuO thin films deposited on a glass substrate have been studied by Ahirrao et al. [21]The structural studies show that Cu2O films have mainly (111) and (200) crystalline orientations. The room temperature photoluminescence study showed a green band at 503nm, 540nm, and a strong red emission at 627nm. Similarly, Xu et al. [22] synthesized nanostructure thin films with unique morphologies deposited on a glass substrate at 40-80°C for 1-5 h with a pH value ranging from 8.5-10. Unique elliptical sheet-like morphology can be obtained at a temperature ≥ 70°C, while corncob-like nanostructure can be obtained at a lower temperature ≤ 60°C. Muhibbullah et al. prepared aphotoconductive copper oxide thin film using CuSO4 a nd Na2SO3 in an aqueous solution The film exhibits p-type conduction and photosensitivity, making it useful for hetero-structure solar cells. MohdRafie Johan et al. studied the effects of annealing on the properties of the film, finding that the films annealed at different temperatures transformed into cupriteswith Cu2O composition, andphotoluminescence intens ity improved with higher annealing temperatures.

A photoconductive copper oxide thin film has been prepared by Muhibbullah et al. The aqueous solution of CuSO4 and Na2SO3 was used as the key ingredients, and the deposition temperature was 70-80°C. The Auger electron spectroscopy measurements reveal that the samples have a Cu/O ratio near the stoichiometric ratio of Cu2O. By bubbling oxygen into the solution during the deposition, the oxygen content of the film increased[23].

The films exhibit p-type conduction and photosensitivity, and are thus useful as an absorber material in hetero-structure solar cells.

A study of the annealing effects on the properties of a copper oxide thin film prepared by chemical deposition has been carried out by MohdRafie Johan et al. [24]. The films were annealed in air at temperatures ranging from 200-400°C, with the as-prepared sample used as a reference. The XRD results reveal that the prepared film and films annealed at 200°C were cuprites' structures with Cu2O composition.

Films annealed at 300°C consist of a mixed phase of CuO and Cu2O, while the film annealed at 400°C completely converts to the CuO phase. Because Oxidized to CuO when ex posed to oxygen at high temperatures.

Previous studies have explored various factors influencing the properties of these films, such as precursor concentration, deposition time, pH of the deposition bath, and annealing conditions. These studies have reported significant variations in the structural, morphological, and optical properties of the thin films based on different synthesis parameters. One notable study by Smith et al. (2018) investigated the effects of precursor concentration on the thin films' properties. They synthesized copper oxide thin films using the CBD method with varying concentrations of copper acetate from 0.5ml to 2ml. Their results showed that increasing the precursor concentration led to an enhancement in the crystalline and optical band gap of the thin films. However, the study only explored a limited range of copper acetate concentrations, calling for further investigation into a broader range of concentrations. This research addresses the synthesis of copper oxide thin films through the CBD method while varying the copper acetate concentration from 1 ml to 5 ml. By broadening the range of precursor concentrations, we aim to gain a thorough understanding of how copper acetate concentration influences the properties of the thin films. Specifically, I will examine the effects of copper acetate concentration on the structural and optical properties of the resulting copper oxide thin films. To provide a comparative perspective, I will focus on Smith et al.'s study and analyze any similarities or differences in the thin films synthesized at corresponding copper acetate concentrations. This comparative analysis will help highlight the unique effects of varying copper acetate concentration and enhance our understanding of the synthesis of copper oxide thin films.

CHAPTER THREE

3. METHODOLOGY

3.1. Thin Film Deposition Techniques

Thin films have distinct advantages over bulk materials because most of the deposition techniques used for the formation of thin films is non-equilibrium in nature. Therefore, the formation of the thin film is not inhibited by the metallurgical phase diagram [25, 26] A deposition technique is considered as the integral key for the creation of thin film new materials to meet the ever-increasing demand from industries for versatile and multi dynamic materials[27]. The deposition techniques determine virtually all the properties of the thin film and can also be used to modify the existing properties. Proper consideration needs to be given to the deposition techniques depending on the area of application because not all the deposition techniques result in identical properties, such as microstructure, surface morphology, electrical, biocompatibility, optical, corrosion, and hardness [28]. A single material can be used for different applications and tailored to meet the optimum requirements by using different deposition techniques. A combination of different techniques can also be used to form a hybrid

deposition process, with each contributing to the outcome of the thin film.

Most deposition techniques follow these three major sequences:

1. Synthesis of the deposition species,
2. Conveyance from source to substrate,
3. Deposition and adhesion of the source onto the substrate and subsequent film growth.

3.2 Chemical Bath Deposition (CBD) Method

Chemical Bath Deposition relies on creating a solution such that deposition (changing from an aqueous to a solid substance) will only occur on the substrate, using the method below: Metal salts and chalcogenides precursors are added to water to form an aqueous solution containing the metal ions and chalcogenides ions that will form the compound to be deposited. Temperature, pH, and concentration of salts are adjusted until the solution is in metastable super saturation,[29]that is until the ions are ready to deposit but can't overcome the thermodynamic barrier to nucleation (forming solid crystals and precipitating out of the solution).[30] A substrate is introduced, which acts as a catalyst for nucleation, and the precursor ions adhere to the substrate, forming a thin crystalline film by one of the two methods described below. That is, the solution is in a state where the precursor ions or

colloidal particles are 'sticky', but can't 'stick' to each other. When the substrate is introduced, the precursor ions or particles stick to it, and aqueous ions stick to solid ions, a solid compound depositing to form crystalline films.

The pH, temperature, and composition of the film affect crystal size and can be used to control the rate of formation and the structure of the film. Other factors affecting crystal size include agitation, illumination, and the thickness of the film upon which the crystal is deposited. [30] Agitation of the solution prevents the deposition of suspended colloidal crystals, creating a smoother and more homogeneous film with higher band gap energy. Agitation also affects the formation speed and the temperature at which formation occurs and can alter the structure of the deposited crystals [25]. Unlike most other deposition processes, Chemical Bath Deposition tends to create a film of uniform thickness, composition, and geometry even on irregular (patterned or shaped) substrates because it, unlike other methods of deposition, is governed by surface chemistry. Ions adhere to all exposed surfaces of the substrate, and crystals grow from those ions.

3.2.1. Principles of Chemical Bath Deposition Techniques

Chemical Bath Deposition (CBD) is a method used to create thin films on a substrate by initiating a chemical reaction between precursor chemicals in a solution and the surface of the substrate. Here is a general overview of the process:

1. Solution Preparation: Prepare a solution containing the necessary precursor chemicals to form the desired thin film. Typically, these precursor chemicals are metal salts or other compounds that react to create the desired film material.

2. Substrate Preparation: Thoroughly clean the substrate to ensure proper adhesion of the film. This may involve using solvents, etching, or surface activation.

3. Immersion in the Solution: Immerse the cleaned substrate in the solution containing the precursor chemicals. Typically, the substrate is placed in a reaction vessel or bath containing the solution.

4. Chemical Reaction: The precursor chemicals in the solution react with the substrate surface, resulting in the deposition of a thin film on the substrate. Factors such as temperature, pH, and solution composition can influence this reaction.

5. Controlling Film Thickness: Factors like precursor chemical concentration, immersion time, and reaction temperature can be adjusted to control the thickness of the thin film.

6. Rinsing and Drying: Once the desired film thickness is achieved, remove the substrate fromthe solution, rinse it to eliminate any remaining chemicals, and then dry it.

7. Post-treatment (if necessary): Depending on the requirements of the thin film, additional steps such as annealing or additional coating processes may be performed to enhance the film properties. Overall, CBD is a relatively simple and cost-effective technique for depositing thin films on substrates. This method finds applications in various fields, including electronics, optics, and coatings.

3.3. Factors That Influence Deposition

There are several factors that affect the thin film deposition process in CBD techniques, including the concentration of the complexing agent, pH of the solution, reaction temperature, and nature of the substrate.

3.3.1 Concentration of Complexing Agent

In the CBD method, most depositions are carried out in alkaline solutions. To prevent the precipitation of metal hydroxide, a complexion agent is added. It helps reduce the concentration of free metal ions in the solution bath, which avoids the rapid bulk formation of the desired product. The concentration of free metal ions decreases with an increase in the concentration of the complexing agent. Consequently, the rate of the reaction is reduced, which leads to a larger terminal thickness.

3.3.2. pH of Solution

For the growth of good quality thin films, the presence of hydroxyl ions in the precursor solution is necessary. The thin film formation depends on the pH of the reaction mixture, which is determined by the OH- ions. A decrease in pH results in porous, non-reflecting, powdery, and weakly adhered thin films on the substrates. At a higher pH, the metal ion concentration will be lower and the reaction rate will be slow[31]. As the pH increases and the metal ion concentration decreases, the rate of film formation decreases.

3.3.3. Reaction Temperature

Temperature is another parameter that influences the rate of the reaction. Increasing the temperature of the solution bath efficiently dissociates the complex. Therefore, the kinetic energy of the molecules also increases, leading to greater interaction between ions. This can result in an increase or decrease in the terminal thickness, depending on the extent of the super saturation of the solution.

3.3.4. Nature of Substrate

The substrate plays a major role in the reaction kinetics and the adhesion of the deposited film. Hence, cleaning the substrate is the first important step in the thin film deposition procedure. A higher deposition rate and terminal thickness are obtained when the lattice parameters of the substrates and the deposited material match well with each other.[32]

3.4. Thin Film Characterization Techniques

Thin films characterization is a crucial step in understanding the physical, chemical, optical, and electrical properties of thin films. These films are often used in a wide variety of applications, from semiconductor devices to solar cells, optical coatings, and sensors. The characterization techniques employed can provide insights into the structure, composition, surface morphology, and electrical properties of the thin films, which are vital for optimizing their performance for specific applications. This overview will discuss thin films characterization techniques, categorized into structural, and optical analysis.

3.4.1. X-Ray Diffraction (XRD)

XRD is the most common and first hand instrument used by solid-state scientists. It is a powerful method to investigate the detailed structure, composition and related properties of nano-materials. This information can be derived from peak position, intensity and the peak profile .A great advantage of X-ray powder diffraction is that it requires XRD has been used extensively for the examination of materials and thin films[22]. Its effective use depends upon having a crystalline material. X-ray diffraction it helps determine the strain, atomic spacing, grain size, crystal orientation, and crystal defects. It is also useful for measuring the proportion of crystalline in natural fibers before and after chemical treatment. This method involves directing X-rays at the thin film and analyzing the diffraction pattern that results when the X-rays interact with the crystal lattice.

The XRD is based on Bragg's Law, which describes the relationship between the X-ray wavelength, the lattice spacing of the crystal, and the angle of diffraction. Bragg's Law is given by:

$$n\lambda = 2d\sin\theta \qquad (3.1)$$

Where: n is the order of diffraction is the wavelength of the X-rays, d is the interplanar spacing in the crystal lattice, θ is the angle of incidence.

The X-rays are scattered by the atoms in the crystal lattice, and constructive interference occurs for certain angles (θ), resulting in a diffraction pattern. This diffraction pattern provides information about the crystal structure, phase composition, and lattice parameters of the material. The crystallite size (D) can be estimated from the Debye-Scherer's equation, which relates peak broadening to the size of the crystallites:

$$D = \frac{K\lambda}{\beta\cos\theta} = \frac{0.9\lambda}{\beta cos\theta} \quad (3.2)$$

Where: D is the crystallite size, K is the shape factor (usually ≈ 0.9),λ is the X-ray wave length, β is the full width at half maximum (FWHM) of the diffraction peak (in radians), The broadening of the full width at half maximum (β) is inversely proportional to the size of crystallite according to Scherer's[33].θ is the Bragg's diffraction angle.

The d-spacing is calculated from Bragg's law

$$d = \frac{n\lambda}{2sin\theta} \quad (3.3)$$

Further, to have more information on the amount of defects in the synthesized copper oxide thin films, the dislocation density (δ) was calculated from Williamson Small man's formula as given below

$$\delta = \frac{n}{D^2} \quad (3.4)$$

Where :'n' is a factor, which when equal to unity gives the minimum dislocation density and 'D'is the average crystallite size[34].

The micro strains (ε) were calculated by using the following equations[35].

$$\varepsilon = \frac{\beta cos\theta}{4} \quad (3.5)$$

3.4.2 UV-visible spectrometer

After synthesizing the thin film, its optical properties are measured using a UV-Vis spectrophotometer[36]. This device measures how much light is absorbed by the film at various wavelengths, usually in the ultraviolet (UV) and visible (Vis) regions of the electromagnetic spectrum.

The spectrophotometer records the absorbance or transmittance of the film as a function of wavelength, typically in the range of 200–800 nm. The resulting data is plotted as a graph of

absorbance (a.u) vs wavelength (nm). For semiconductors, the absorbance shows a sharp increase at a certain wavelength, which corresponds to the absorption edge. This absorption edgeis related to the electronic band gap of the semiconductor material.

The highest energy band occupied by electrons is called valance band (VB) and lowest unoccupied band is called conduction band (CB). The energy gap between the two levels is called band gap energy (Eg). The band gap (Eg) is the energy needed to move a valence electron into conduction band.

For a semiconductor it is the energy needed to free an electron from thenucleus of the parent atom. It is defined as Eg=Ec-Ev

to determine the band gap energy,

Convert Wavelength to Photon Energy by use the relation:

$$Energy(h\nu) = \frac{1240}{\lambda} \qquad (3.6)$$

Where: hν is the photon energy in electron volts (eV). λ is the wavelength in nanometers (nm).

Calculate the absorption coefficient (α) using Beer-Lambert law:

$$\alpha = \frac{2.303 * A}{d} \qquad 3.7$$

Where, d is the thickness of the sample (in cm).

The Tauc relation is: $(\alpha h\nu)^n = A(h\nu - Eg)$

Where:α = Absorption coefficient. hν = Photon energy (eV). Eg = Band gap energy (eV).

A = Proportionality constant.

n = A constant that depends on the type of electronic transition:

n = 1/2 for direct allowed transitions. n = 2 for indirect allowed transitions.

Rearrange and plot $(\alpha h\nu)^n$ vs. hν.

The band gap energy (Eg) is obtained by extrapolating the linear portion of the curve to the x-axis (where $(\alpha h\nu)^n = 0$).

Interpreting the band gap from the x-axis (photon energy) gives the optical band gap of the material. Using the Stern formula, we analyze the relationship between the absorption coefficient and photon energy to determine the optical band gap from UV-Vis spectroscopy data. This method is effective for characterizing semiconductors and other materials

3.5. Experimental Procedure

3.5.1. Materials Used and Their Functions

To prepare CuO thin films, the following materials are used:

- Test tube brush: used for cleaning test tubes.
- Balance (electronic or triple beam): used to determine the mass of solid in grams.
- Scoop: used for transferring small amounts of a solid.
- Volumetric flask: used for making solutions of a specific concentration.
- Magnetic stirring: used to mix chemicals and deionizer water.
- Thermometer: used to measure temperature gradients.
- Graduated cylinder: used to measure accurate volumes in milliliters (ml).
- Beaker: a simple container used for heating and stirring liquids in laboratories.
- Substrate holder: supports substrates during coating processes with an expandable outer frame.
- Water bath: laboratory equipment filled with heated water, used to incubate samples at a temperature of 80°C for 20 minutes.

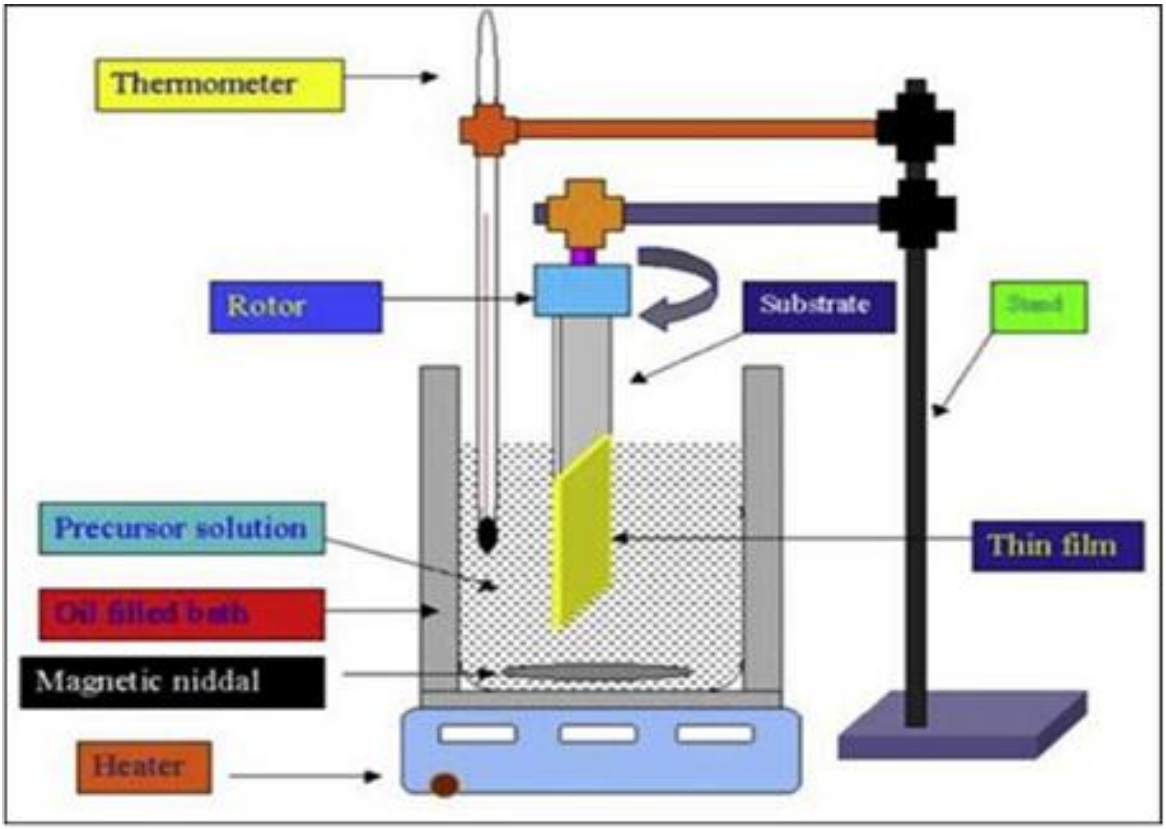

Figure 3. 1: Experimental setup for the chemical bath deposition of thin film.

3.5.2 Reagents/Chemicals

The chemicals and solvents required for synthesizing CuO thin film are as follows: copper acetate (CH3.COO)2Cu.H2O as the Cu precursor, hydrazine (N2H5OH) as the complexion agent, and ammonium hydroxide (NH4OH) as the pH regulator.

The chemicals used for preparing the CuO thin film, along with their source and percentage purity, are listed below:

- 0.4M Copper acetate monohydrate (CH3COO)2Cu.H2O (Percentage purity = 98%), with a molar weight of 199.65.
- Ammonia solution (NH4OH) (Percentage purity = 25%)
- Hydrazine (N2H5OH) (Percentage purity = 99%), with a molar weight of 50.06.
- Distilled water

3.5.3. The Reagents Solution Preparation

The chemicals used for the film preparation, as well as their source and percentage purity are listed as follows:

Preparation of 0.4 M Copper acetate solution were prepared using distilled water

The required mass to prepare a certain molar of a specified volume solution is obtained by the formula;

Mass = Concentration *Volume of water * Molar weight *Percentage purity.

To prepare 0.4MCopper acetate solution in a 250 ml water the mass to be dissolved is obtained

Mass=Concentration*Volume*Molar weight* Percentage purity

Mass = 0.4M * 0.25L * 199.65g/ML* 0.98 = 19.7654g

Where, g is gram, M is molar and L is liter

19.7654g of Copper acetate solution was weighed into a beaker. Then it was dissolved in small amount of distilled water and then inter into 250 ml volumetric flask.

3.5.4. Substrate Cleaning

Substrate cleaning is crucial for depositing copper oxide thin films. While most clean surfaces can be used as substrates, the level of adhesion may differ significantly between materials. Glass is a commonly used substrate in CBD. Despite its inert nature, the glass surface can react with solution species. The cleaning procedure for the substrate is important, as it affects the composition and grain size of the film. There are various cleaning methods available, depending on the substrate's nature, the type of contaminants, and the desired level

of cleanliness for thin film deposition. In this particular case, the substrate used for depositing the copper oxide thin film was soda lime glass, measuring 75 mm × 25 mm × 2 mm. Prior to deposition, the microscope glass slides were soaked in nitric acid overnight, then immersed in alcohol for approximately an hour. They were subsequently cleaned using distilled water in an ultrasonic bath and dried under ambient conditions.

3.6. Preparation of the Thin Film

The total solution of copper oxide thin film was 70 ml. This reaction solution was obtained by mixing 1 ml of 0.4 M Copper acetate, 5 ml of Ammonia solution, 3 ml of Hydrazine, and 61 ml of distilled water. To mix the solution, a 150 ml beaker and an electromagnetic stirrer were used. The two cleaned substrates were inserted into the 70 ml solution prepared properly and put in a water bath. The temperature was then adjusted to 80 ^{0}C and left on a digital electronic heater in the water bath for 20 minutes. This was the first sample preparation at this time. The color of the solution started to change from a water-like color to a golden color after 9 and 10 minutes. After 20 minutes, the substrates were removed and dried, resulting in a highly uniform and adherent excellently covered the substrate. Similar films were formed on the other four samples by changing the volume of Copper acetate to 2 ml, 3 ml, 4 ml, and 5 ml, and the volume of water to 60 ml, 59 ml, 58 ml, and 57 ml, respectively, while keeping all other parameters constant. However, the CuO thin films deposited by adding a higher volume of Copper acetate solution were very thick and well characterized. They were later annealed at a temperature of 300 ^{0}C for 2 hours. The figure blow from Figure 3.2.-3.4 shows the various deposition stages of CuO thin film Figure 3.2 initial color of mother solution Figure 3.3 shows color of mother solution after 10 minutes and (c) Figure 3.4 shows film formed on substrates.

Table 3. 1: preparation of copper oxide thin films sample ratio

Sample code	Cu acc. (ml)	Ammonia (ml)	Hydrazine (ml)	Distilled Water(ml)	V_{total}	T_{final} (^{0}c)	T_{total}
Cu-1	1ml	5ml	3ml	61ml	70ml	80^{o}c	20 min
Cu-2	2ml	5ml	3ml	60ml	70ml	80^{o}c	20 min
Cu-3	3ml	5ml	3ml	59ml	70ml	80^{o}c	20 min
Cu-4	4ml	5ml	3ml	58ml	70ml	80^{o}c	20 min
Cu-5	5ml	5ml	3ml	57ml	70ml	80^{o}c	20min

Figure 3. 2: Colorless chemical bath solution before 10 minutes and chemical composition

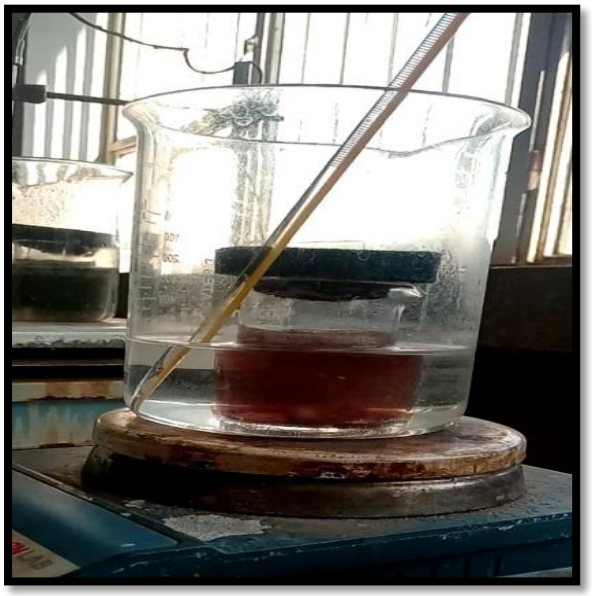

Figure 3. 3: Red color chemical bath solution after 10 minutes

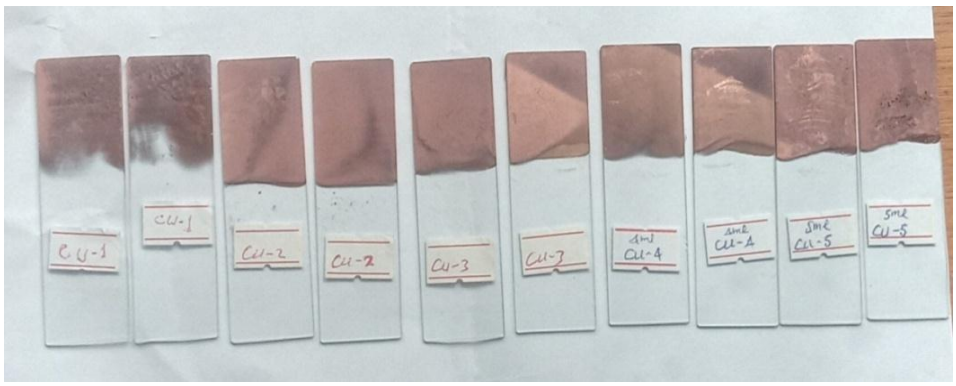

Figure 3. 4: the dark brown film produced on substrate

CHAPTER FOUR

4. RESULT AND DISCUSSION

4.1. X-ray Diffraction (XRD) Analysis

When it comes to obtaining information on an atomic scale from both crystalline and non-crystalline (amorphous) materials, X-ray diffraction is a crucial technique in the field of material characterization[37]. Using a Shimadzu X-ray diffract meter with a Cu Kα line (k = 0.154056 Å) in the 2θ range at 10 to 80 ^{0}C, crystallographic studies of copper oxide thin films were conducted. By capturing an X-ray diffraction pattern, CuO thin films' structural characteristics were examined. The data presented mostly pertains to thin-film X-ray diffraction (XRD) research conducted at varying volumes of copper acetate ($Cu(CH_3COO)_2$). Crystal structure, crystallite size, strain, and dislocation density are all determined by XRD. Based on this comparison conclusions can be drawn about crystal structure and orientation of the sample

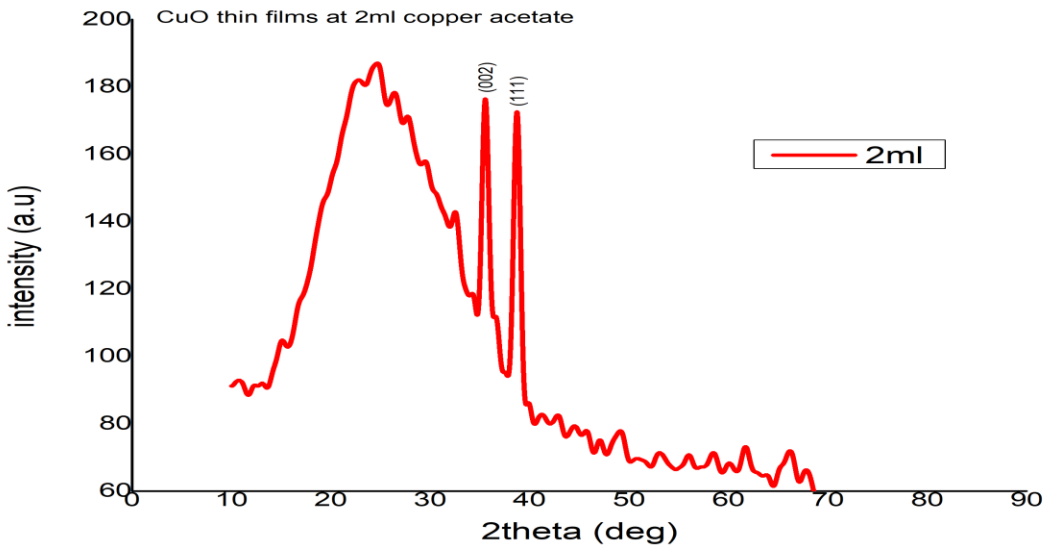

Figure 4. 1: The X-ray diffraction pattern of CuO thin film deposited with 2ml copper acetate.

The CuO thin film deposited by 1ml, 2ml, 3 ml, 4 ml and 5ml of (0.4M) copper acetate is depicted in sample of 1ml, 2ml, and 3ml The X-ray diffraction pattern showed peaks at 2θ values of 35.5 and 38.7 degree corresponding to the (0 0 2) and (111) plane of monoclinic phase of CuO structure (JCPDS reference code 00-048-1548), respectively. The CuO thin films deposited by 4 ml and 5 ml (0.4M) copper acetate are display one additional peak at 2θ

values of 36.06, indexed to (111) plane of cubic Cu2O structure (JCPDS reference code 00-005-0667). These peaks correspond to the (0 0 2) and (1 1 1) planes of the monoclinic phase of CuO structure. The XRD pattern indicated that films deposited at 1 ml, 2 ml and 3 ml copper acetate concentration crystallizes in single monoclinic CuO phase, however films deposited at 4 ml and 5 ml copper acetate concentrations crystallizes revealed the cubic Cu2O phase. When the concentration of copper acetate increases leading to more peaks in the resulting films, the intensity of the films also increased. This is because the increase in copper acetate concentration typically results in a higher amount of copper being deposited onto the substrate, leading to a denser and thicker film. As a result, the film's intensity, which can be related to factors such as the amount of material deposited and its thickness, is likely to increase with higher concentrations of copper acetate. This can have implications for the film's properties, such as its optical, electrical, or mechanical characteristics, depending on the specific application and requirements.

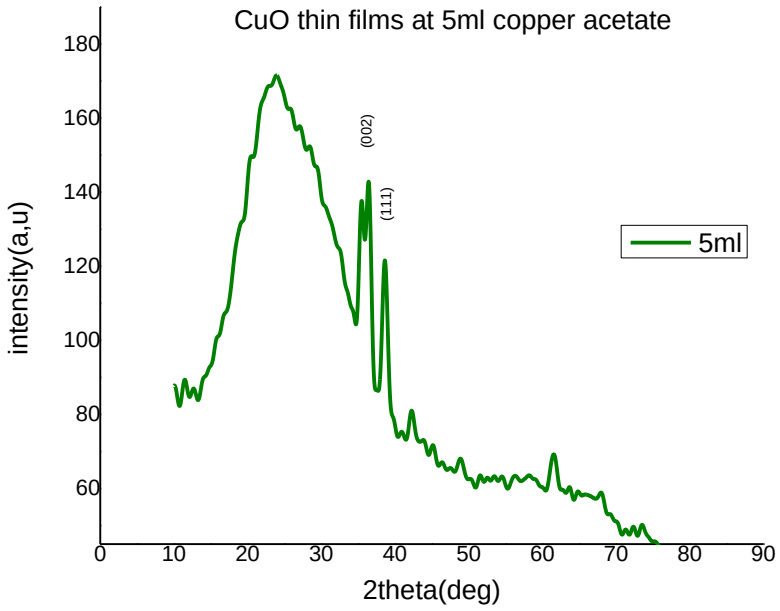

Figure 4. 2: The X-ray diffraction pattern of CuO thin film as deposited with copper acetate 5ml.

Graph of X-ray diffraction pattern of CuO thin film all sample merged together

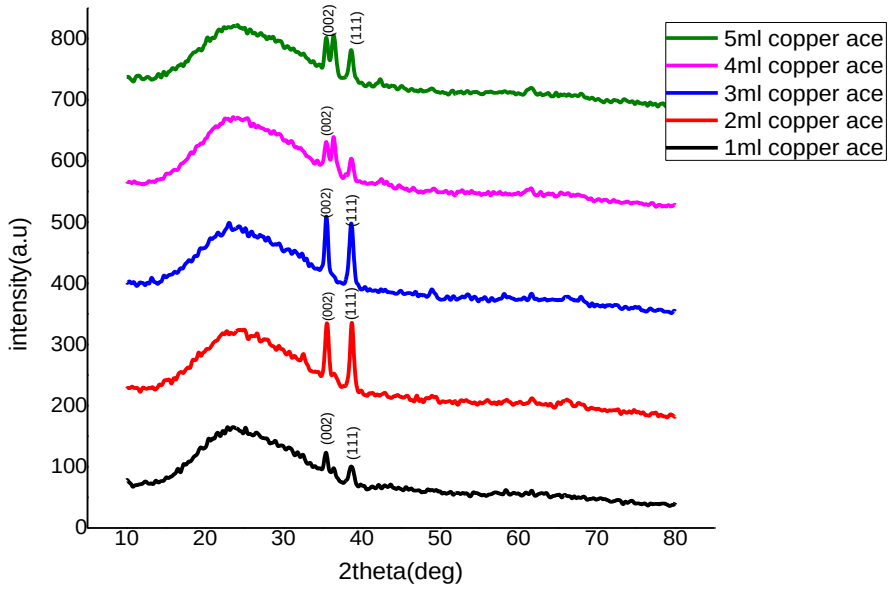

Figure 4. 3: The X-ray diffraction pattern of CuO thin film for easy comparison all the samples merged together

The crystallite size (D) of the two phases is calculated by the Debye-Scherer's formula.

$$D = \frac{K\lambda}{\beta\cos\theta} = \frac{0.9\lambda}{\beta cos\theta} \quad (4.1)$$

Where the wavelength of CuKα radiation (0.15406 Å), k is shape factor (0.9), θ is the Bragg's diffraction angle and β is the broadening of the diffraction line (FWHM). The broadening of the full width at half maximum (β) is inversely proportional to the size of crystallite according to Scherer's [22] The d-spacing is calculated from Bragg's law

$$d = \frac{n\lambda}{2sin\theta} \quad (4.2)$$

Further, to have more information on the amount of defects in the synthesized copper oxide thin films, the dislocation density (δ) was calculated from Williamson Small man's formula as given below

$$\delta = \frac{n}{D^2} \quad (4.3)$$

Where 'n' is a factor, which when equal to unity gives the minimum dislocation density and 'D'is the average crystallite size [34].

The micro strain (ε) were calculated by using the following equations [35].

$$\varepsilon = \frac{\beta cos\theta}{4} \quad (4.4)$$

The summary of the XRD results of CuO and Cu2O thin films deposited at different copper acetate concentration were given in table 4.1 and 4.2.

Table 4. 1: Summary of the XRD analyses of CuO thin films.

Volume of Copper	2θ	Θs	sinθ	Cosθ	FWHM(deg)	FWHM(rad)	Crystallite size	Dislocation (δ)	Strain(ε)
1 m	38.7	19.35	0.331	0.943	0.78924	0.013777	9.50	11.09	0.003248
2 m	38.73	19.365	0.331	0.943	0.56141	0.0098	13.35	5.61	0.00231
3ml	38.66	19.33	0.314	0.949	0.61135	0.010671	12.26	6.65	0.002532
4ml	38.69	19.345	0.331	0.943	0.64731	0.011299	11.58	7.46	0.002664
5ml	38.65	19.325	0.330	0.943	0.62941	0.010987	11.91	7.05	0.00259

Table 4.1 and 4.2 show the X-ray Diffraction (XRD) analyses of CuO and Cu_2O thin films deposited at different copper acetate concentrations calculated using the formula 4.1 to 4.4. As can be observed from Table 4.1, the crystallite size of the monoclinic phase of CuO is in the range 9.5 nm to 13.4 nm. The result shows that the crystallite size of the monoclinic CuO phase is relatively higher at 2 and 3 ml copper acetate solution than other volumes. Similarly the crystallite size of cubic Cu2O phase for 4 and 5 ml copper acetate solution is 4.74 and 4.99 nm respectively, as shown in Table 4.2. The decrease in crystallite size of the monoclinic CuO phase at 4 and 5 ml copper acetate could be the competitive growth of the cubic Cu2O phase. For films deposited with 4 ml and 5 ml of copper acetate, an additional cubic Cu_2O phase was observed at a 2θ value of 36.06°, with crystallite sizes of 4.74 nm and 4.99.0 nm, respectively. This indicates competitive growth of the cubic Cu_2O structure at higher precursor concentrations

Table 4. 2: Summary of the XRD analyses of Cu2O thin films.

Volume of Copper	2θ	Θ	Sinθ	Cosθ	FWHM(deg)	FWHM(rad)	Crystallite size	Dislocation (δ)	Strain(ε)
1ml	-	-	-	-	-	-	-	-	-
2ml	-	-	-	-	-	-	-	-	-
3ml	-	-	-	-	-	-	-	-	-
4ml	36.09	18.045	0.309	0.95	1.59452	0.027833	4.74	44.56	0.00661
5ml	36.06	18.03	0.309	0.958	1.51366	0.026422	4.99	40.15	0.006328

4.1.1. Comparison with Other Research

When comparing these results with the findings of Smith et al. (2008), several similarities and differences can be observed. Smith et al. also reported a consistent 2θ value for CuO thin films, though their reported crystallite sizes were slightly smaller than those observed in this study. Additionally, while the dislocation densities and strain values for CuO films in both studies follow similar trends, the absolute values differ, potentially due to variations in deposition techniques or precursor materials.

For Cu_2O thin films, Smith et al. reported a 2θ value near 36.1°, in agreement with the current study. However, they observed crystallite sizes that were slightly larger, potentially due to differences in annealing conditions or substrate materials. The higher dislocation densities and strain values observed in both studies indicate that Cu_2O thin films tend to exhibit greater structural imperfections compared to CuO films.

4.2. Optical Analysis

4.2.1 Optical Absorbance Study

Figure 4.4 shows peaks at certain wavelengths. These peaks correspond to the wavelengths at which the material absorbs the most light.[38] For semiconductors like CuO and Cu_2O, these peaks are related to the electronic transitions between the valence band and conduction band. The wavelength of the absorption edge (the point where absorbance sharply increases) is

often linked to the band gap energy of the material. And the band gap is the energy difference between the valence and conduction bands.

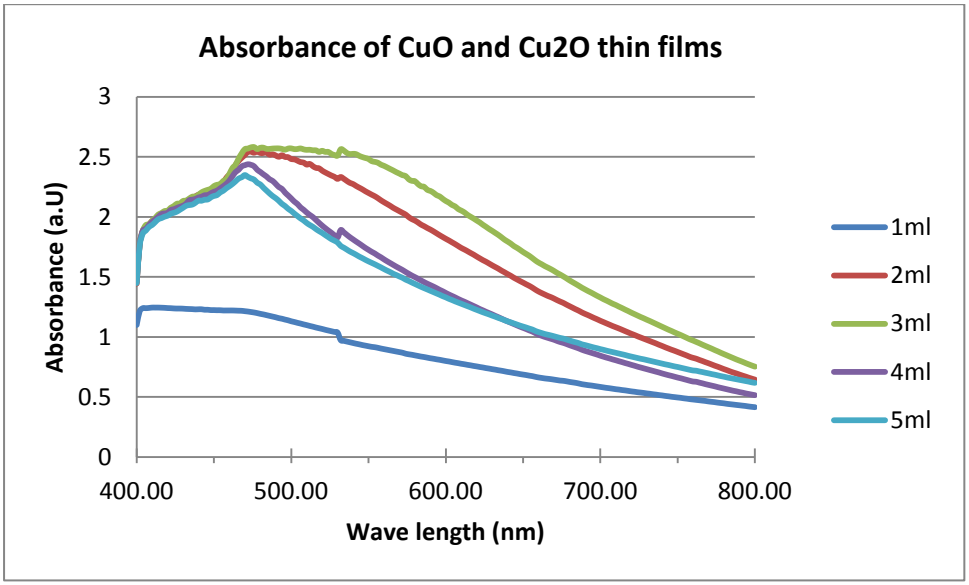

Figure 4. 4: plot of wavelength vs absorbance of CuO and Cu2OThin Films Deposited at (1ml, 2ml, 3ml 4ml, 5ml) Concentrations of copper acetate

The graph in Figure 4.4 shows the absorbance of CuO thin films deposited using different volumes of precursor solution (1 ml, 2 ml, 3 ml, 4 ml, and 5 ml) as a function of wavelength in the range of 400 to 800 nm. This type of graph is commonly used to analyze the optical properties of thin films, particularly their light absorption behavior in the visible spectrum. All samples exhibit a peak absorbance between 450 nm and 520 nm. This region corresponds to the visible light range, where CuO thin films are expected to show significant absorption due to their semiconducting nature.

The film deposited using 3 ml of precursor solution shows the highest absorbance followed by a film deposited at 2 ml copper acetate solution. The absorbance decreases as the wavelength increases, indicating that the thin films absorb more light in the blue-green region (shorter wavelengths) and less in the red or near-infrared region (longer wavelengths).All films show a gradual decrease in absorbance at higher wavelengths (600–800 nm), which is typical for semiconductor materials like CuO that have a direct band gap. The reduction in absorbance in this region indicates that the energy of the photons is lower than the band gap energy, and thus less light is absorbed. The absorbance values converge at longer wavelengths, with all the films showing absorbance values around 0.5–1. a.u. at 800 nm.

4.2.2. Optical Band gap Study

Using a UV-VIS/NIR beam spectrophotometer, optical absorbance studies were conducted for films deposited at varying concentrations of copper acetate in the 400nm–800nm wavelength range. The optical band gap energy was computed based on the wavelength and optical absorbance data. The energy band gap (Eg) of copper oxide thin films can be derived by using the Stern formula, we analyze the relationship between the absorption coefficient and photon energy to determine the optical band gap from UV-Vis spectroscopy data which estimates the optical band gap by plotting $(\alpha h\nu)^2$ vs. $h\nu$ (where α is the absorption coefficient and $h\nu$ is the photon energy) as shown in the Figure 4.5 The band gap is an essential property of semiconducting materials like CuO because it determines the material's ability to absorb light and convert it into electrical energy, which is crucial for applications like photovoltaic's and photo catalysis.

Figure 4. 5: Graph of $(\alpha hv)^2(ev)^2$ versus hv(ev) for CuO thin film at 1, 2, 3, 4 and 5 ml copper acetate

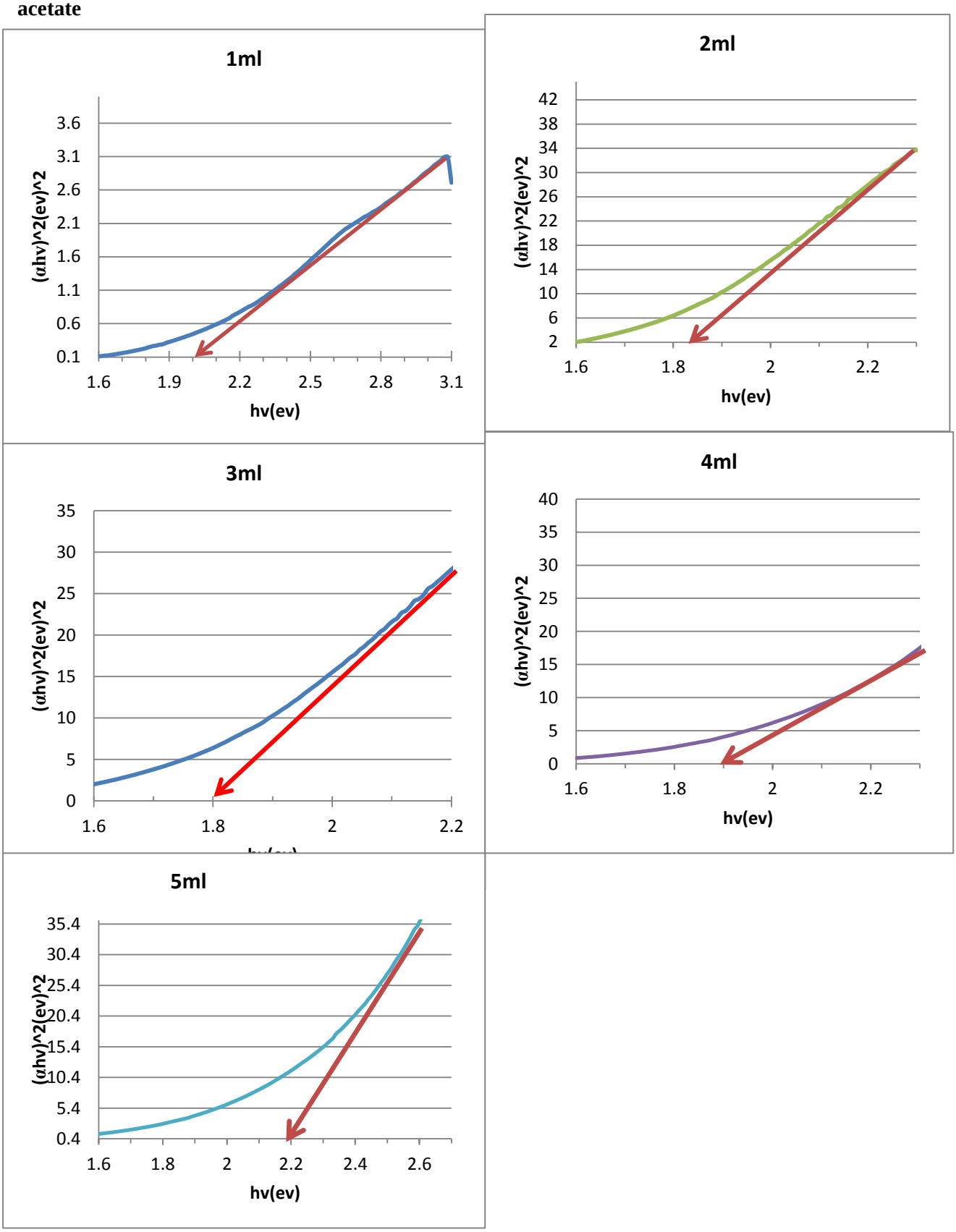

Table 4. 3: Energy band gap of CuO thin films as function of concentration of copper acetate.

No	Volume of copper acetate (ml)	Copper oxide Eg(eV)
1	1ml	2
2	2ml	1.85
3	3ml	1.8
4	4ml	1.95
5	5ml	2.2

Figures 4.5shows the effect of concentration of copper acetate on band gap, where the band gap value is estimated by extrapolation of the straight line of the plots of $(\alpha hv)^2$vs hv(Photon energy) of the CuO thin films deposited under various concentration of copper acetate. The band gap values are found to be 2, 1.85, 1.8, 1.95 and 2.2 eV as the concentration of copper acetate is increased from 1ml, 2ml, 3ml 4ml to 5ml, respectively.

The observation of the samples result shows, the band gap values vary with the volume of copper acetate used in the deposition process. The lowest band gap is measured for the film deposited with 3 ml of copper acetate (Eg = 1.80 eV).The highest band gap is observed for the film deposited with 5ml of copper acetate (Eg = 2.2eV).

Lower Volumes (1 ml to 3 ml): The band gap decreases as the volume of copper acetate increases, reaching a minimum of 1.80 eV at 3 ml. This trend suggests that increasing the precursor concentration leads to better crystalline and denser films, reducing the band gap. The film deposited with 3 ml of copper acetate exhibits the lowest band gap of 1.80 eV, which is quite close to the typical value reported for CuO thin films in various studies (usually between 1.7 to 2.1 eV).

A lower band gap is desirable for applications in photovoltaic's and photo catalysis, as it allows the material to absorb more light in the visible region, improving its efficiency in light-harvesting applications.

The current study shows a very similar absorption edge for CuO thin films (around 360-400 nm) compared to Smith et al.'s results (around 350 nm). The band gap values are also quite close, with 1.7 eV in Smith et al.'s study compared to my study 1.8 eV of 1ml. In other studies on CuO thin films, band gaps in the range of 1.7 to 2.1 eV are commonly reported.[39] for example Smith et.al (2019) Found that the band gap of CuO thin films

varied between 1.8 and 2.0 eV, depending on the deposition conditions this slight variation could be due to differences in synthesis methods, film thickness, or deposition conditions. Variations in the crystalline or grain size of the films can also affect the band gap values slightly. In my study, the absorption edge for Cu_2O thin films starts around 400 nm, and the absorption peak is observed in the 500-600 nm range. This is quite similar to Smith et al.'s findings, where the absorption edge occurs around 450 nm, and the peak is near 520 nm. Both studies report a band gap in the range of 2.0–2.1 eV for Cu_2O, which is typical for this material. Again, small differences in the band gap could be attributed to the synthesis method, film thickness, or the presence of defects.[40]

CHAPTERFIVE

5. CONCLUSION AND RECOMMENDATION

5.1. Conclusion

Copper oxide thin films were prepared on glass substrates using the chemical bath deposition technique with a deposition time of 20 minutes. X-ray diffraction (XRD) and optical analyses were employed to investigate the structural and optical properties of copper oxide (CuO and Cu_2O) thin films deposited with varying concentrations of copper acetate precursor. The XRD results indicate that the structural properties of the thin films are influenced by the concentration of copper acetate used in the deposition process. For films deposited with 1 ml, 2 ml, and 3 ml of copper acetate, the XRD patterns exhibited a single monoclinic CuO phase, with peaks at 2θ values of 35.5° and 38.7°. The crystallite sizes of the monoclinic CuO phase ranged from 9.5 nm to 13.4 nm, with the 2 ml and 3 ml concentrations yielding larger sizes. For films deposited with 4 ml and 5 ml of copper acetate, an additional cubic Cu_2O phase was observed at a 2θ value of 36.06°, with crystallite sizes of 4.74 nm and 4.99.0 nm, respectively. This indicates competitive growth of the cubic Cu_2O structure at higher precursor concentrations. The dislocation density and microstrain values revealed that films with lower copper acetate concentrations (1 ml to 3 ml) exhibited fewer structural defects, while higher concentrations (4 ml and 5 ml) resulted in increased dislocation density and strain, likely due to the formation of the Cu_2O phase. These structural variations highlight the impact of precursor concentration on film crystallinity, phase composition, and defect density, which are crucial for tailoring material properties for specific applications. The optical properties of the CuO thin films were analyzed based on absorbance and band gap values. All samples exhibited strong absorbance in the visible region (450–520 nm), with the film deposited using 3 ml of copper acetate showing the highest absorbance. The absorbance decreased at longer wavelengths (600–800 nm), consistent with the behavior of semiconductor materials with a direct band gap. The higher absorbance observed for the 3 ml sample suggests enhanced light-harvesting capability, which could benefit photovoltaic and photocatalytic applications. The optical band gap values ranged from 1.8 eV to 2.2 eV, depending on the copper acetate concentration. The lowest band gap (1.8 eV) was observed for the film deposited with 3 ml of copper acetate, indicating better crystallinity and reduced defects, making it ideal for applications requiring efficient light absorption. Higher band gap

values (e.g., 2.2 eV for 5 ml) suggest the presence of structural imperfections and additional phases, such as cubic Cu_2O, which can influence the material's electronic properties

5.2. Recommendation

Further studies on this thesis could yield valuable information and makes materials closer to application. The following further investigations are recommended:

- To study conductivity and resistivity of copper oxide thin films with increasing deposition time
- Study by optimizing the concentration range and exploring additional opportunities for improvement of CuO thin films.
- To investigate the morphological and composition of CuO thin films by using SEM and EDAX

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