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Title:

Thermoélectrique système pour la Conversion de l'Énergie Thermique en Énergie Électrique

Presented by:

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Dedication

First and foremost, I praise Allah, my Creator, for granting me the strength and perseverance to complete this modest work.

First and foremost, I would like to express my profound gratitude to my parents, whose unconditional love, sacrifices, and constant encouragement have been my greatest source of strength throughout my academic journey. Their support has been the foundation of all my achievements. I am equally thankful to my friends for their companionship, encouragement, and for being a constant source of motivation and positivity during challenging times.

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I would like to express my deepest gratitude to my supervisor, **Dr.Mohammedi Abdelkader**, for his invaluable guidance, support, and advice throughout the entire process of this research.

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General Introduction

Thermoelectric materials are important scientific innovations that help improve the efficiency of energy use. These materials stand out for their ability to convert the temperature difference into electrical energy, as well as to convert electrical energy into thermal energy, opening up vast opportunities for innovative applications in many fields.

This conversion process is based on the thermoelectric effect, which comprises three main physical phenomena: the Seebeck effect, which generates an electric current when there is a temperature difference between the two ends of the material; the Peltier effect, which occurs when an electric current passing through a thermoelectric material heats or cools its ends, depending on the direction of the current; and finally, the Thomson effect, which occurs when the electric current interacts with the thermal gradient inside the material, causing thermal variations along the material. These materials, which are characterized by their availability and ease of deposition, represent a revolutionary solution for improving the efficiency of the use of thermal energy.

The performance of such devices depends heavily on the structural integrity, optical behavior, electrical conductivity, and thermoelectric efficiency of the materials. Moreover, optimizing operating conditions, such as the intensity of applied electric current and annealing temperature, is crucial to enhancing energy conversion capabilities. This study focuses on the synthesis and characterization of ZnO and CuO thin films, the evaluation of their thermoelectric properties, and the simulation of their performance within a microscale thermoelectric module. Through experimental analysis and numerical modeling, the work aims to provide insights into the viability of ZnO/CuO-based thin film systems for next-generation thermoelectric devices.

Our idea is to use these materials as a smart coating for glass, so as to regulate the heat transfer between the indoor and outdoor environment according to seasonal variations. In summer, they reduce the intake of outdoor heat, and in winter, they keep the heat indoors, thus significantly contributing to improve the efficiency of energy consumption and offer thermal comfort.

This innovation is not just an idea, but a step towards a more sustainable future. It has great potential to become an innovative start-up or patent, capable of bringing a positive change in our daily lives as well as in the industrial sector.

Chapter 1: Basic Concepts for Thermoelectric Materials

This chapter will provide a theoretical introduction to thermoelectric materials, including their definition and their types, physical principles of the electrothermal phenomenon, and practical applications, Whit our thin layers.

The three main phenomena will be explained: the Seebeck effect (generation of electric current with a temperature difference), the Peltier effect (heating or cooling of the edges of a material under the effect of an electric current), and the Thomson effect (interaction between the electric current and the thermal gradient inside the material).

In addition, the concept of a thermoelectric merit number (ZT) will be discussed, as well as its importance for evaluating the effectiveness of these materials, and like principle of a thermoelectric generator.

Chapter 2: Thin layer deposition

This chapter will focus on the practical steps of COMSOL Multiphysics, whit definition and our applicated system in simulation

This chapter contents all parts of the system in 3D whit some results to our condition using

This chapter will present the Preparing thin layers with centrifuge technology, like substrate preparation, focusing on techniques for studying the properties of thin films

Finally, suggestions will be made to improve performance and morphology our thin films

Chapter 3: Conclusion and discussions

In this chapter, we present the results and discussions of a theoretical and experimental study on a thermoelectric generator. The chapter is divided into two main parts: a simulation of the thermoelectric generator using COMSOL Multiphysics, and an investigation of the effect of current intensity on temperature and heat distribution.

- The geometric structure of the model and the properties of the materials used
- Effect of electric current on temperature variability
- The effect of the intensity of the current on the difference of the potential of the object

- The effect of the intensity of an electrical current on conducting inside the stereo

Whit trial results, specifying properties of zinc oxide and copper oxide whit electrothermal constants determination

Chapter 4: Explanation of the proposed project

This chapter presents a smart energy solution based on thermoelectric materials, specifically ZnO and CuO thin films. The project proposes a coating system for glass surfaces that can regulate heat flow according to seasonal changes while simultaneously generating electricity from temperature differences. By combining heat management and energy harvesting in one system, the project aims to reduce energy consumption in buildings, promote environmental sustainability, and offer a practical, scalable innovation for modern energy challenges.

Chapter I: Basic Concepts on Thermoelectric Materials

I.1. Introduction

This chapter provides an overview of the existing work in this field, which serves as the foundation for our research. We will divide it into four main sections. First, we will introduce the fundamentals of thermoelectric materials, discussing their definition, physical and chemical properties, and the last we will show real-world applications.

The second part is based on the three point we will talk at our thin films haw it depends because it is the important part of this project, we will focus on thin films, examining how their characteristics depend on various factors. This is a crucial aspect, as the performance and efficiency of our system relies heavily on the properties of these films.

In addition, we will show the concept of a Thermoelectric generator whit their principal coefficient, as well as its importance for evaluating the effectiveness of these materials.

I.2. Thermoelectric materials

I.2.1 thermoelectric materials definition

The thermoelectric effect is a physical phenomenon that allows a direct conversion between thermal energy and electrical energy. This conversion can occur in two main directions: on the one hand, transforming a temperature difference into an electrical potential difference, called Seebeck effect, and on the other hand, producing a temperature difference from an electrical current, known as Peltier effect. These effects have many applications, especially in the precise measurement of temperatures (thermometry) and refrigeration. In addition, thermoelectric devices play a key role in recovering heat lost in exothermic processes, such as engines or energy-intensive industries. This area is particularly relevant in the current context, where the scarcity of fossil fuels and the need for environmentally friendly alternative energies are driving the development of innovative solutions such as thermoelectricity [1].

I.2.2. Physical principles of the Thermoelectricity phenomenon

a. **Electrical conductivity**

Thermoelectric materials must have a high electrical conductivity to minimize energy losses in the form of heat generated by the electrical resistance. This allows an efficient flow of the electrons that transport energy.[2]

b. **Thermal conductivity**

A low thermal conductivity is necessary to maintain a significant temperature difference between the two ends of the material. This promotes a better conversion of thermal energy into electrical energy (or vice versa).[2]

c. **High Seebeck effect**

An ideal material has a high Seebeck coefficient, expressing its ability to produce an electrical voltage in response to a temperature gradient. Metals often show a low Seebeck effect, while semiconductors show higher values

d. **Operating temperature**

Each material has an optimal temperature range where it offers the best performance. For example, bismuth-telluride alloys work well at low temperatures, while silicon-germanium materials work well at high temperatures.

I.2.3. Seebeck effect

Figure (I.1).Seebeck effect Generates an electrical voltage when there is a temperature difference between two points in a material. This effect is used, for example, in thermocouples to measure temperatures.[3]

In 1821 Thomas Seebeck, a German physicist discovered that when two dissimilar metal (Seebeck used copper and bismuth) wires are joined at two ends to form a loop, a voltage is developed in the circuit if the two junctions are kept at different temperatures. The pair of metals forming the circuit is called a

thermocouple. This effect is due to conversion of thermal energy to electrical energy.[3]

The valence electrons in the warmer part of the metal play a crucial role in this process, primarily due to the influence of thermal energy. With higher kinetic energy, these electrons migrate more rapidly toward the colder end compared to the movement of electrons from the colder region toward the warmer part. On the hot side, the Fermi distribution is broader, meaning there is a higher concentration of electrons above the Fermi energy, whereas on the cold side, the Fermi distribution is sharper, indicating fewer electrons above the Fermi energy. Since electrons naturally move toward regions of lower energy, they migrate from the warmer end to the colder end, facilitating energy transport and ultimately leading to temperature equilibrium [3].

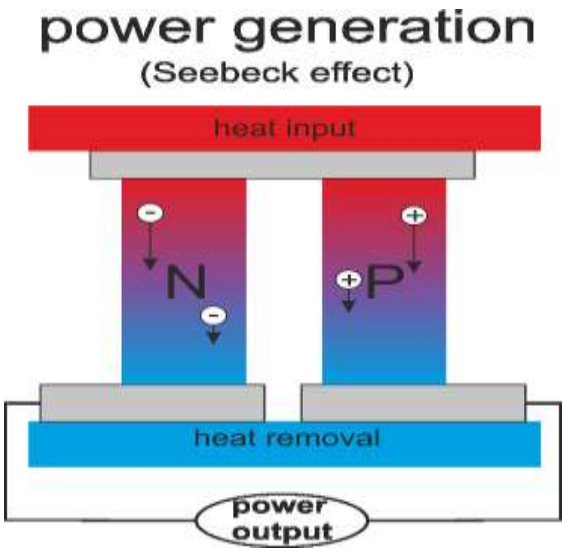

Figure (I.1): Seebek effect.

This movement results in the more negative charge at colder part than warmer part, which Leads to the generation of electrical potential. If this pair is connected through an electrical circuit. It results in the generation of a DC.

I.2.4. Peltier effect

When an electrical current flows through a junction between two materials, it causes a temperature variation at that junction, which is used for cooling in Peltier modules.

In 1834 Jean Peltier, a french watch maker, discovered a second thermoelectric effect. If a current flows through a circuit containing junction of two dissimilar metals, it leads to an absorption or liberation of heat at the junctions. Heat is given out or absorbed depending on the pairs of metals and the direction of the current. The phenomenon of heat evolution is different from the Joule heat as Peltier effect is a reversible process while Joule loss is irreversible. If the direction of the current at the junction is the same as the direction of the Seebeck current, heat is liberated if the Seebeck junction is a hot junction or is absorbed if the junction is cold. Thus, for a copper.[4]

Constantan thermocouple, if the current flow at the junction is from copper (+) to constantan (-), heat is absorbed. On changing the direction of the current, heat will be liberated at the same junction, showing that the phenomenon is reversible as shown in **Figure (I.*2*)**.

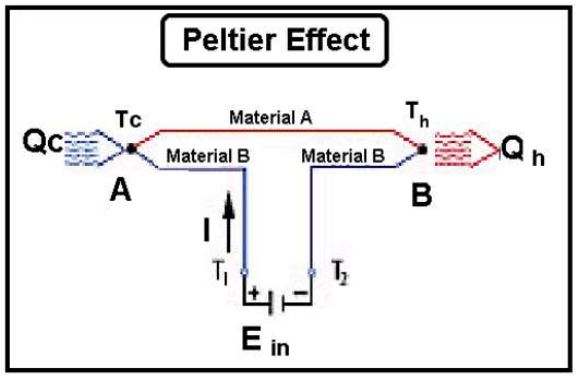

Figure (I.2): Peltier effect.

I.2.5. Thomson effect

Describes how heat is generated or absorbed when an electrical current flows through a material that is subject to a temperature gradient as showing in **Figure (I.3)**.

William Thomson (later well known as Lord Kelvin) discovered a third thermoelectric effect which provides a link between Seebeck effect and Peltier effect. Thomson found that when a current is passed through a wire of single homogeneous material along which a temperature gradient exists, heat must be exchanged with the surrounding in order that the original temperature gradient may be maintained along the wire. (The exchange of heat is required at all places of the circuit where a temperature gradient exists.[5]

The Thomson effect is the result of the combination of the Seebeck and Peltier effects. Discovered by William Thomson in 1854, it describes the phenomenon in which a conductor carrying an electric current and subjected to a temperature difference between two points absorbs or releases heat depending on the nature of the material. This heat, known as Thomson heat (Q), is determined by **Equation (I.1):**

$$Q = \rho J 2 - u * J \frac{dT}{dx} \qquad \textbf{(I.1)}$$

Where ρ is the resistivity of the material, and dT/dx is the temperature gradient along the conductor.

J represents the current density, and μ is the Thomson coefficient. The first term ρJ corresponds to Joule heating (irreversible), while the second term represents Thomson heating, whose sign changes with the direction of the current[4].

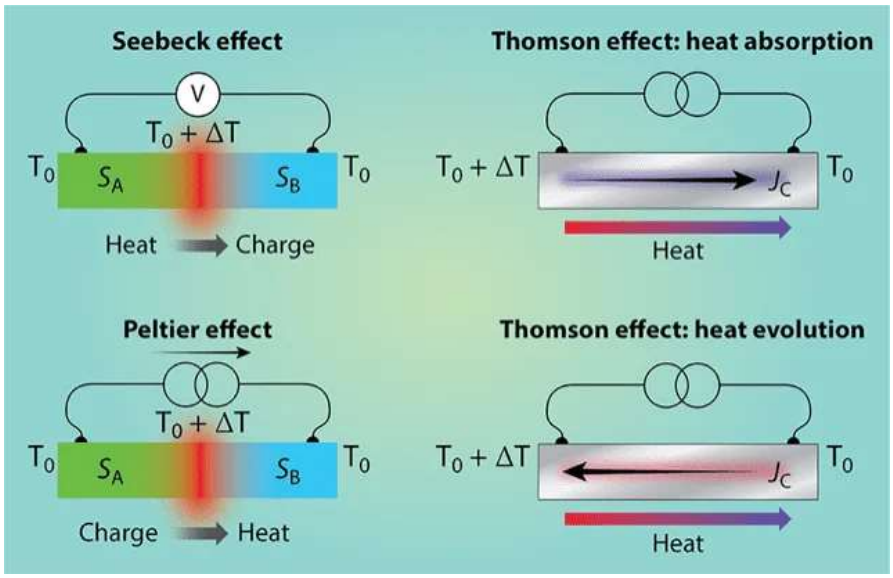

Figure (I.3): Peltier effect.

The performances of thermoelectric materials are measured by a parameter called an adidimensional merit factor (often denoted *ZT*), which depends on their thermal and electrical conductivity and their capacity to generate a voltage.

In practice, devices such as thermocouples exploit the Seebeck effect to measure temperatures: a temperature-proportional potential difference appears between the ends of two different metals connected in a weld. On the other hand, the Peltier modules, widely used for cooling, consist of assemblies of semiconductors of the *n* and *p* type connected. When an electrical current flows through these

semiconductors, one side becomes cold while the other becomes hot, allowing effective cooling in various applications.

I.3. Thin films

I.3.1 Define thin films

A thin film is a delicate layer of material deposited on a substrate, with one of its dimensions greatly reduced, usually measured in nanometers (nm). This extremely small thickness between the two boundary surfaces alters most of its physical properties [6]

A small number of atomic layers often exhibit properties that differ significantly from those of the same element in their bulk state. This difference arises because, in the bulk state, the influence of boundaries (such as the substrate or surrounding molecular environment) is generally negligible. In contrast, for a thin film, boundary effects play a dominant role in determining its properties. As the thickness decreases, the material's two-dimensional behavior becomes more pronounced. Conversely, when the thickness exceeds a certain threshold, the influence of thickness diminishes, and the material gradually regains the characteristics of its bulk counterpart. [6]

A thin film is always attached to a substrate, regardless of the preparation method used. Therefore, the nature of the substrate must always be considered when fabricating thin films. In other words, a thin film of the same material and thickness may exhibit significantly different physical properties depending on the substrate on which it is deposited.

I.3.2. Thin films deposition

Thin film depositions can be achieved using two primary methods: Physical Vapor Deposition (PVD) and Chemical Vapor Deposition (CVD).

PVD encompasses a range of surface coating technologies used for decorative coatings, tool coatings, and various equipment applications. It is fundamentally a vaporization process, where material is transferred atom by atom from the solid phase to the vapor phase and then back to the solid phase, gradually forming a thin film on the target surface. In reactive deposition, the depositing material interacts with a

gaseous environment or co-deposited material to create a compound film, such as nitride, oxide, carbide, or carbonitride.[7]

Physical evaporation is one of the earliest techniques for depositing metal films. Metals like aluminum and gold are heated until they vaporize, after which the vapor condenses onto the substrate to form a thin film. All film deposition processes occur in a vacuum or a precisely controlled atmosphere to ensure the desired film quality.

Chemical Vapor Deposition (CVD) is a technique in which chemical processes, such as reactions occurring at the substrate surface or in its surrounding environment, facilitate film deposition. According to Bhat (2006), CVD can be formally defined as a system where a combination of gases reacts with the substrate surface at a relatively high temperature, causing the decomposition of certain gas constituents and the formation of a solid film, depositing either a metal or a composite onto the substrate.

There are various types of CVD processes; however, this project focuses on one significant method: Plasma-Enhanced Chemical Vapor Deposition (PECVD) [8].

Compared to PVD, this technique offers higher deposition rates, better conformance in rough substrates, easy deposition onto complex surfaces, and high throughput. However, some disadvantages, such as the use of high substrate temperatures, and the toxicity and flammability of the reactive gases have prevented it from being used in low-scale developments but is well justified in applications where high-throughput is required, i.e. semiconductor industry.

I.3.3. Thin films Properties

In this section a brief description of the mechanical, electrical, and optical properties in connection with the morphological features will be provided.

1- Structural properties

Thin films are widely used in electronics, optics, and energy applications due to their unique structural and functional properties. The structural properties of thin films significantly influence their performance, including electrical conductivity, optical transparency, and mechanical strength. These properties depend on deposition techniques, substrate type, and processing conditions.

Thin films can be amorphous, polycrystalline, or single-crystalline, depending on the material and deposition method, The surface roughness of a thin film affects optical and electronic performance. Techniques like Atomic Force Microscopy (AFM) and Scanning Electron Microscopy (SEM) are used to analyze morphology.

Thin films may contain vacancies, dislocations, or impurities, which can degrade performance or enhance specific properties. Controlled doping can tailor electronic and optical characteristics [9] .

2- Electrical properties

The electrical properties of thin films cover a wide field, considering the diverse microstructures that arise depending on whether they are metallic, semiconductor, or insulating films, as well as the type of substrate used for deposition. However, many of these films share common morphological characteristics that lead to similar transport mechanisms, allowing conductivity to be analyzed from a broader, unified perspective.

One of the primary reasons for the deviation in conductivity of thin films compared to bulk materials is the size effect. As the thickness of the material decreases, the electron means free path is reduced due to the activation of additional scattering mechanisms. In a simplified approach, where conductivity in bulk materials is directly proportional to the electron means free path, this results in a decrease in conductivity. This relationship holds for epitaxial, polycrystalline, and amorphous structures, as the maximum crystallite size is constrained by the film thickness, thereby maintaining the size effect assuming all other factors influencing conductivity remain unchanged. However, as discussed in previous sections, the microstructure may contain numerous structural defects and grain boundaries, which serve as additional scattering centers for charge carriers, further reducing conductivity [10].

Grain boundaries are considered disordered regions where mobile carriers are scattered as they travel between crystallites. For instance, the electrical conductivity of polycrystalline and nanocrystalline materials is significantly lower than that of their bulk single-crystalline counterparts. This reduction is primarily due to decreased carrier mobility, despite having a similar carrier concentration. Additionally,

microstructural defects such as voids, dangling bonds, and localized defects—predominantly found at grain boundaries—further contribute to the decline in conductivity. These defects can trap mobile carriers in doped semiconductors, creating potential energy barriers that hinder charge carrier movement between crystallites[11, 12]

3- Optical properties

The optical behavior of thin films is primarily determined by their microstructure, which in turn depends on the deposition parameters. Before exploring the relationship between optical properties and microstructural features, it is essential to define the optical coefficients. The optical response of thin films is characterized by their reflection and transmission coefficients.

In a typical configuration, a thin film of thickness d and refractive index n_1 is deposited onto a substrate made of either the same or a different material with a refractive index n_2. When a beam of light interacts with the system, multiple reflections and transmissions occur at both the air-film and film-substrate interfaces, leading to interference effects. The total amplitude of these interactions is determined by summing the individual reflections directed back into the incident medium and the individual refractions passing into the substrate.

A key distinction between non-absorbing and absorbing layers lies in the complexity of the refractive index. In absorbing layers, the refractive index incorporates an additional term, the extinction coefficient k, which quantifies energy absorption within the film. Consequently, the optical coefficients are strongly influenced by the refractive indices of each medium, the extinction coefficient, and the film thickness [13].

I.4. Thermoelectric material types

I.4.1. n-type thermoelectric materials

Bismuth telluride (Bi_2Te3) A widely used thermoelectric material with high efficiency in converting heat into electricity, commonly used in cooling and power generation applications, when doped with elements like selenium (Se) or bromine (Br), it becomes an n-type semiconductor, meaning it has an excess of free electrons that contribute to electrical conductivity.[14]

Telluride selenium (SeTe): A semiconductor material with thermoelectric properties, often explored for energy conversion applications, Selenium can act as an electron donor, making SeTe behave as an n-type material, thermoelectric performance of tin and selenium co-doped bismuth telluride system has been investigated in the temperature range 10–300 K. Structural investigation reveals that the prepared samples have hexagonal crystal structure [15].

Zinc oxide (ZnO): A semiconductor material with high thermal and electrical conductivity, used in various electronic and thermoelectric applications, Intrinsic defects (like oxygen vacancies) or doping with elements like aluminum (Al) or gallium (Ga) introduce extra electrons, making it an n-type material, it is confirmed that both the Al doping concentration and film thickness control the thermoelectric, optical and structural properties of these films. Seebeck coefficients up to −134 μV K^{-1} and electrical conductivities up to 4×10^4 $(\Omega\ m)^{-1}$ lead to power factors up to 4×10^{-4} W mK^{-2} [16].

I.4.2. p-type thermoelectric materials

Bismuth telluride (Bi_2Te_3) The p-type counterpart of Bi_2Te_3, often alloyed with other elements to enhance its thermoelectric performance, The introduction of Sb creates a deficiency of electrons, resulting in hole conduction. This is the first instance of combining these materials in such a way, achieving a device output voltage of 4.7 mV for a temperature difference of ~4°C. This innovative approach demonstrates significant potential for developing efficient and versatile wearable thermoelectric energy harvesting technologies [14].

Tin telluride (SnTe) A thermoelectric material with promising properties, particularly in mid-temperature energy conversion applications, Tin vacancies create an abundance of holes, making it a p-type material.[15]

Potassium oxide (K_2O) A ceramic material with potential applications in thermoelectric, though mainly used in glass and catalysis, when A work function of 1.35 eV is measured which and the expected efficiency of the corresponding converter is discussed. It is generally assumed that the decrease of the work function in the alkali/oxygen/silicon system is attributed to the creation of a surface dipole resulting from a charge transfer between the alkali metal and oxygen.[17]

Copper oxide (CuO) is attractive materials for device applications in energy conversion and storage because of their earth abundancy, nontoxicity, low environmental impact, and low production cost. The CuO phase is of further interest due to its high stability and environmental friendliness. It finds a diverse field of applications in solar energy conversion, the metastable Cu_4O_3 has also been found to be attractive for its superior electrochemical performance, photodetectors, and photonic filters [18].

The properties of the selected thermoelectric materials are summarized in the following **Table (I.1):**

Table (I.1): Types of thermoelectric materials.

Material Type	**Name**	**Chemical Formula**	**Key Properties**	**Figure of Merit (ZT)**	**Generator Efficiency (η)**
n-type	Bismuth telluride	Bi_2Te_3	High thermoelectric efficiency, good stability	~1.0 – 1.2 (300 K)	~5 – 8%
n-type	Selenium telluride	SeTe	Good electrical conductivity, used in thin films	~0.4 – 0.8	~2 – 5%
n-type	Zinc oxide	ZnO	High thermal stability, wide-bandgap semiconductor	~0.3 – 0.6	~1 – 4%
p-type	Bismuth telluride	Bi_2Te_3	High efficiency, typically alloyed with Sb_2Te_3	~1.0 – 1.2 (300 K)	~5 – 8%
p-type	Tin telluride	SnTe	Good electrical conductor, used in thermoelectric devices	~0.6 – 1.2	~3 – 7%
p-type	Potassium oxide	K_2O	High ionic conductivity, used in ceramics and batteries	~0.1 – 0.3	~0.5 – 2%
p-type	Copper oxide	CuO	Good thermal absorber, used in nanostructures and photovoltaics	~0.2 – 0.5	~1 – 3%

Notes :

- The thermoelectric figure of merit (ZT) is given at room temperature (~300 K) but may vary with temperature and doping.
- Efficiency (η) is an approximate range and depends on the temperature gradient (ΔT) and system design.
- Higher ZT values correlate with higher efficiency, but practical efficiencies are often lower due to heat losses and contact resistance.
- State-of-the-art thermoelectric generators (TEGs) can achieve ~5 – 8% efficiency with optimized materials and design.

I.5 Thermoelectric generator

I.5.1. Thermoelectric generator operation principle

Thermoelectric generators (TEGs) are solid-state devices that convert thermal energy into electrical power through the thermoelectric effect. This phenomenon is based on the Seebeck effect, which occurs when a temperature gradient is applied across dissimilar semiconductor materials, generating an electric voltage. TEGs have gained significant attention in various applications, including waste heat recovery and portable power generation. Their ability to harness and convert otherwise lost heat makes them particularly valuable in industrial manufacturing and electricity generation processes [19].

The Seebeck phenomenon, in which a temperature difference between two dissimilar materials causes a voltage potential difference, is the basis for thermoelectric generators' operation. A TEG module is made up of a series or parallel connection of many thermocouples, each of which is made up of p-type and n-type semiconductors with opposite charge carriers. A temperature gradient is created throughout the module when one end of the thermocouple is placed in contact with a heat source (hot side), and the other end is placed in contact with a heat sink (cold side). The voltage potential difference between the hot and cold ends of each thermocouple is directly proportional to the temperature gradient between them.

I.5.2. Thermoelectric Systems Configuration: Order of Materials p and n

Thermoelectric generator (TEG) consists of several essential components, with thermoelectric modules forming its core. These modules comprise a series of interconnected thermoelectric devices made from p-type and n-type thermoelectric materials, which are placed between ceramic substrates. The ceramic substrates provide electrical insulation and mechanical support while also limiting heat transfer between the hot and cold surfaces.

The transfer of heat from the heat source to the TEG is facilitated by heat exchangers, which are designed with a large surface area to optimize heat transfer efficiency. These devices come in various forms, such as finned heat sinks, plate heat exchangers, and tube-and-shell arrangements. Proper insulation is crucial to minimize heat dissipation, thereby maintaining a greater temperature gradient for improved efficiency. Insulation materials such as ceramic fiber, fiberglass, and aerogel are commonly used due to their low thermal conductivity [20].

For power transmission from the TEG to an external load, efficient electrical connections are required. To minimize power losses, these connections must have low resistance. Metallic interconnects are typically used to establish reliable electrical pathways between thermoelectric modules and external loads or power management systems.

The n- and p-type thermoelectric materials with the same chemical composition BSTS have been creatively designed and prepared, in which the formation for different types of antistites defects and vacancies is manipulated by using the heavy plastic deformation and various hot-pressing processes. The band structure, meanwhile, is also modified by point defect engineering to optimize the electrical properties and transport properties for TE materials. In addition to the strategies, materials possessing special crystal structures such as three-dimensional holes and weak interlayer coupling in two-dimensional act as promising candidates for TE materials.

I.5.3. Calculating Merit Factor (ZT)

This relation of merit, presented by **Equation (I.2)**, depends on the transport properties of the two materials constituting the thermoelectric torque.

Thus, for maximum conversion efficiency, similar materials should be chosen to form the torque.

$$Zab = \frac{(SA-SB)^2}{(\sqrt{(\rho aKa)}+\sqrt{(\rho bKb)})} \qquad \textbf{(I.2)}$$

Where:

- Sx, Seebeck coefficient of material X expressed in V/K
- ρx, electrical resistivity of material X expressed in Ω·m
- Kx, thermal conductivity of material X expressed in W/m·K
- Zab, figure of merit of the AB pair expressed in T^{-1}

However, in practice, it is easier to focus on the couple's legs individually, so that they can be optimized separately. For this purpose, an individual dimensional merit figure is defined as **Equation (I.3):**[21]

$$ZT = \frac{S^2}{\rho K} T = \frac{PF}{K} T \qquad \textbf{(I.3)}$$

Where:

- PF=S2/ρ, power factor expressed in W/m·K²
- T, temperature expressed in K

According to the I-1 relationship, a high-performance thermoelectric material must have properties optimized to maximize its efficiency, as described in the given relationship. The following is a detailed explanation of these criteria:

1. High Seebeck coefficient

The Seebeck coefficient is a key parameter that measures the ability of a material to convert a temperature difference (ΔT) into an electrical potential difference (ΔV).

- A high Seebeck coefficient means that the material is effective at generating an electrical voltage from a thermal gradient.

- It is essential to increase the amount of usable electrical energy generated by the material.

2. Low electrical resistivity

Electrical resistivity is the ability of a material to pass electrical current. Low resistivity is essential for:

- Reduce energy losses due to the Joule effect ($P=I^2R$), where III is the current and R is the resistance.
- Allow an efficient flux of electrons, which are the main charge carriers in thermoelectric conversion.

These two parameters (Seebeck coefficient) and (electrical resistivity) together determine the **power factor** of the material (PF).

3. Low thermal conductivity K

Thermal conductivity K measures the ability of a material to transmit heat.

- Low thermal conductivity is necessary to maintain a stable temperature gradient between the two ends of the material.
- If the material conducts heat too well, the temperature gradient dissipates quickly, reducing the efficiency of thermoelectric conversion.

4. Optimization of properties: the ZT merit factor

These three properties (Seebeck coefficient, electrical resistivity, and thermal conductivity) are combined into an dimensionless parameter called the ZT figure of merit, which determines the overall efficiency of a thermoelectric material by **Equation (I.4)** [21]**:**

$$ZT = \frac{S^2 \cdot \sigma \cdot T}{K} \qquad \textbf{(I.4)}$$

- $S^2\,\sigma$ corresponds to **the power factor**.
- T is the absolute temperature in kelvins.
- A low K (thermal conductivity) maximizes ZT, as does a high PF.

I.5.4. Calculate the efficiency of the generator η

The ZT factor characterizes the effectiveness of the material, and the ZT criterion ≥ 1 is generally used for applications. The efficiency η of the module depends on the thermoelectric properties of the materials of which it is made, as well as on the temperature difference ΔT = Tc - Tf to which it is subjected, via the **Equation (I.5):**

$$\eta_{max} = \frac{Tc-Tf}{Tc} \times \frac{\sqrt{1+ZTm}-1}{\sqrt{1+ZTm}+(Tc/Tf)} \qquad \textbf{(I.5)}$$

Where Tm is the average temperature and ZTm is close to the average material merit factor. This efficiency may be greatly reduced in the case of poor electrical or thermal contacts, which limit the establishment of thermal gradients, or which generate dissipation by the Joule effect, The three quantities S, ρ and κ are linked via the carrier concentration (FIG. E3), and a compromise must be found between them to maximize ZT. In a metal, ρ is very low, but S is typically a few μV/K, making the magnitude S2/ρ (called the "power factor") very low. Conversely, in a semiconductor or an insulator, S is very high (a few mV/K), but the very high value of ρ also makes the ratio S 2 /ρ practically zero [21].

I.5.5. Factors influencing the performance of thermal generators.

With the growing concerns over energy shortages and environmental pollution, countries worldwide are actively seeking alternative energy sources to replace traditional fossil fuels. As a result, nuclear, solar, and wind energy have been extensively developed and deployed on a global scale. However, challenges persist, including safety concerns with nuclear energy, the low efficiency of solar energy, and the susceptibility of wind energy to environmental factors [22].

Thermoelectric generator (TEG) technology offers a promising solution by directly converting heat energy into electrical energy through the Seebeck effect. This technology boasts several advantages, including a simple power generation structure, noiseless operation, and a long service life. Significant research efforts have been dedicated to enhancing TEG efficiency, focusing on material design and optimization, internal structural improvements, and advanced heat transfer mechanisms.

Among these research areas, optimizing heat dissipation structures and mechanisms is particularly crucial. Traditional cooling methods, such as fin-based or

metal plane heat dissipation, have been extensively studied, with research exploring flow patterns, fin structure optimization, and different cooling fluids. However, heat dissipation is inherently constrained by the physical properties of air, affecting thermoelectric performance, while metal plane cooling is less effective at high power levels.[23].

In contrast, heat pipes, known for their exceptionally high thermal conductivity, have emerged as highly effective heat transfer elements. They are widely utilized in various applications requiring enhanced heat dissipation, including aerospace, electronic refrigeration, heat exchangers, and solar heating systems[24]

I.6. Thermoelectric materials applications

At present, thermoelectric modules are used in niche areas, such as space or areas far from conventional electrical distribution circuits (in mountains, for communication systems on pipelines, etc.). These generators are widely used in space applications, in particular those based on PbTe or SiGe onboard from 1961 onboard by NASA, or more recently for the Curiosity mission to Mars. In this field, thermoelectric technology has shown its full effectiveness, since thermogenerators have been operating for 30 years, with low power losses, for example in Voyager probes. Moreover, one advantage of this technique is that the modules are not subjected to any mechanical movement and do not use any fluid (unlike conventional refrigeration).

making this technology very reliable and robust. Another large-scale application is the air-conditioning of car seats in the United States and Canada using Peltier chillers: only the seats used are active, which avoids air-conditioning the entire passenger compartment of the vehicle.

And so, we can make a precise classification for the application of thermoelectric materials under the applied effects

A. Sensors: Uses of heat sensing

Thermoelectric devices play a crucial role in energy recovery, converting waste heat into electricity using the Seebeck effect. This technology is particularly useful in industries and automobiles, where significant heat is lost during operation, industrial Applications Industries such as steel manufacturing, cement production, glass

processing, and power plants generate high amounts of waste heat Thermoelectric generators (TEGs) can be installed to recover this heat and convert it into usable electricity, leading to Reduced energy consumption by reusing lost thermal energy Lower CO_2 emissions, improving environmental sustainability Enhanced system efficiency in high-temperature industrial processes Automotive Applications In automobiles, a large percentage of fuel energy is lost as heat through the exhaust system and engine components Thermoelectric devices can be integrated into vehicles to Recover exhaust heat and convert it into electricity. Improve fuel efficiency by reducing the load on the alternator Power auxiliary systems (e g, air conditioning, lights, battery charging) without increasing fuel consumption [25].

B. Cooling systems

Peltier modules are commonly used to create cooling systems without mechanical parts electronic Cooling of processors, sensitive electronic components or lasers to maintain their performance Portable Refrigerators Compact and quiet refrigeration systems for travel or domestic applications [25].

C. In renewable energy

Renewable Energy Hybrid Solar Systems Coupling solar panels with thermoelectric devices to convert both sunlight and heat into electricity. Geothermal Direct exploitation of the earth's heat in thermoelectric generators to produce electricity without turbines.

Thermoelectric materials, while promising, are still limited by their efficiency (*ZT*), high cost and, sometimes, durability. However, recent advances open the door to even wider applications.

I.7. Conclusion

The study of thermoelectric materials and the physical principle of thermoelectricity highlights the crucial role of Seebeck, Peltier and Thomson effects in the conversion of heat into electricity. Thermoelectric materials, including telluride compounds and oxides, are classified into n and p types, forming the basis of thermoelectric generators. Thin films, with their specific properties and advanced deposition techniques, greatly improve the performance of thermoelectric devices by optimizing the merit factor (ZT) and the efficiency of the generators. The configuration of the thermoelectric systems and the selection of the materials directly influence their efficiency. Thermal technology applications range from thermal sensors to renewable energy conversion and cooling systems. They play a key role in optimizing energy efficiency and recovering lost heat sources. Advances in this field are thus paving the way for innovative solutions for energy production and management.

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Chapter II: Thin films deposition

II.1. Introduction

This chapter will present the Preparing thin films with centrifuge technology, like substrate preparation, Preparation of thin layers by spin coating and spray pyrolysis focusing on techniques for studying the properties of thin films

Thermoelectric energy conversion offers a promising approach to harnessing waste heat and generating sustainable electrical power. This project focuses on developing thin-film thermoelectric materials using centrifuge technology, specifically zinc oxide (ZnO) and copper oxide (CuO) thin films. The fabrication process includes solution preparation, substrate treatment, spin coating, spray pyrolysis, and thermal processing to enhance material properties.

Finally, suggestions will be made to improve the performance of our thin films. Various characterization techniques are employed, including morphological analysis, optical property assessment, and electrolyte studies. Furthermore, electrothermal properties such as the thermoelectric figure of merit (ZT), SEPAC S-factor, and generator efficiency (η) are calculated to determine the viability of the materials for thermoelectric applications.

II.2. Fabrication of Thin Films Using the Spin Coating Technique

II.2.1. Preparing zinc oxide solution and - copper oxide

II.2.1.1. Preparing zinc oxide solution

All the chemicals were of analytic grade show the **Figure (II.1)**, To prepare the sol solution for 10 ml whit concentration 0.5 mol/L, we make 1.075 g zinc acetate dihydrate Zn $(CH3COO)_2.2H_2O$, 0.302 ml monoethanolamine (MEA) and adequate deionized water were added to 9.072 ml isopropanol, and then heated to 50 ºC with continuous stirring for 1 h, and heated to 250°C for 4 minutes to every single one, finely 1 h 30 min in 500°C temperature operation and It then cools directly into the air. The physical quantities of the elements required to prepare the solution are shown in the following **Table (II.1)**.

Table (II.1): the physical quantities of the elements required to prepare the solution.

Finely concentrated mol/L	zinc acetate dihydrate g	Monoethanolamine ml	Isopropanol ml	TOTAL
0.5	1.075	0.302	9.072	10

We prepared Ct=0.5 Mol/L solution whit volume Vf=10 ml , that we make a zinc acetate dihydrate [$Zn(CH_3COO)_2.2H_2O$] in a size of isopropanol ($CH_3CH(OH)\ CH_3$), then heated to 50 ºC with continuous to mix it whit magnetic mixer for 15 minutes we stopped when we seeing the white solution, and we add to that a size of MEA ($NH_2CH_2CH_2O$) whit [n(MEA) = 1 mol] it change their colors to Transparent Color

And we mix it for 2h than we make it for 24 hours in ambient temperature for the stabilization.

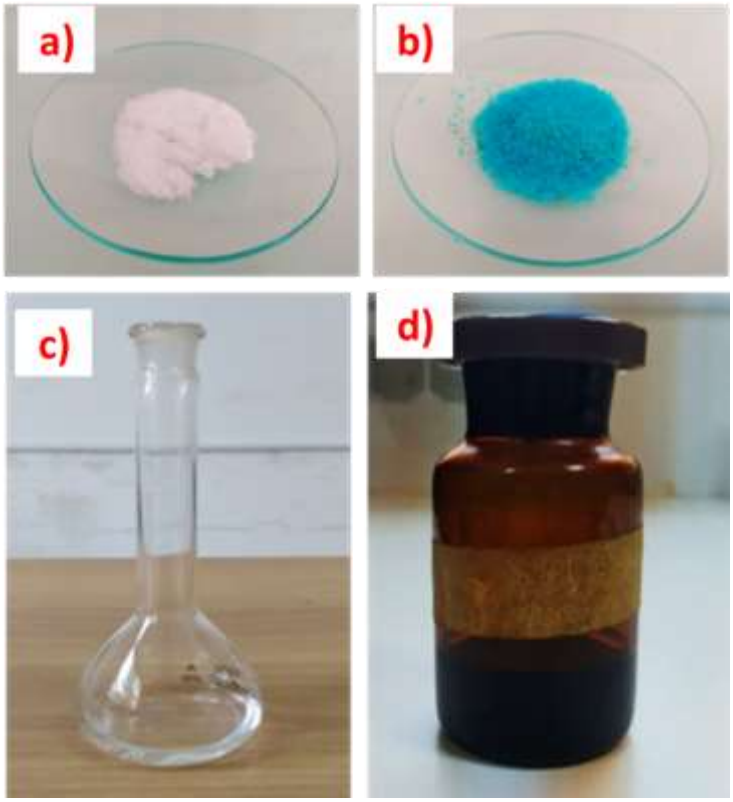

Figure (II.1): Materials used in preparing the solution**.**

II.2.1.2. Preparing copper oxide solution

A methanolic solution of cupric chloride ($CuCl_2$) $2H_2$ O) is taken as a starting solution for the deposition of copper oxide thin films in this technique. The solution was prepared using 0.25 g cupric chloride in one litre methanol. A clean substrate was then dipped into the starting solution and withdrawn vertically at a controlled speed (1.33 mm/s), under atmospheric conditions using this setup. After withdrawal, the substrate with the liquid film adhering to it is baked at 360-500 °C for 5 min. Cupric chloride ($CuCl_2$) $2H_2O$) hydrolysed and reacted on the heated substrate inside the furnace at a high temperature in open air and formed thin solid films according the following reactions [1]:

$$4CuCl_2 + 4H_2O \xrightarrow{360°C} 2Cu_2O + 8HCL + O_2 \quad (1)$$

$$CuCl + H_2O \xrightarrow{400\text{-}500\ °C} CuO + HCL \quad (2)$$

Figure (II.2): Solution of ZnO and CuO**.**

II.2.2. Substrate preparation

In general, the dynamics of spin coating can effectively be demonstrated by separating the complete process into four stages deposition, spin-up, spin-off and evaporation of solvents [2].

II.2.3. 3. Preparation thin films by spin coating

ZnO thin films were deposited by sol-gel spin coating method in **Figure (II.3)** onto glass substrates. Zinc acetate dehydrates and monoethanolamine (MEA) were used as a starting material, solvent and stabilizer, respectively. The molar ratio of MEA to zinc acetate dehydrate was maintained at 1.0 and the concentration of zinc acetate was 0.5 M. The glass substrate was precleaned detergent and then cleaned in methanol and acetone for 10 min and then cleaned with deionized water and dried. The coating solution was dropped onto glass substrate, which was rotated at 3000 rpm for 30 s by using a spin coater. After depositing by spin coating, the film was dried at 300 o C for 10 min in a furnace to evaporate the solvent and remove organic residuals. The procedures from coating to drying were repeated ten times. The film was then inserted into a tube furnace and annealed in air at 550 o C for 1 h.

Figure (II.3): Spin coqting device used**.**

II.2.3.1. A. Deposition

In this step, the substrate is placed on the spin coater machine and the coated material as a solution is dropped on the substrate surface [2], as illustrated in **Figure (II.4a)**.

II.2.3.2. B. Spin-Up

After the deposition of materials the substrate will begin to spin until it reaches a certain speed. This step is typically regarded as destructive fluid ejection from the surface of substrate by the rotational process **Figure (II.4b)**

II.2.3.3. C. Spin-Off

This stage is considered as the third stage among the spin coating technique. It is started once the wafer is spinning at a continuous ratio and liquid viscous forces control solution thinning conduct, which is also called stable solution thinning. Through this step, the fluid spreads out through the surface and at that time the edge of the substrate is removed as the film thins out, as displayed in **Figure (II.4c).**

II.2.3.4. D. Evaporation

In this phase, the substrate spin at a steady rate and the coating thinning conduct is dominated by solvent evaporation. It is important to point out that the thicker film will be obtained if the time of the evaporation process is increased **Figure (II.4d)**

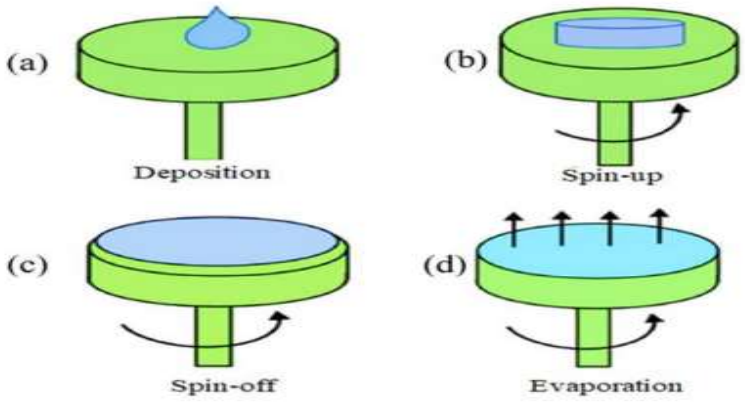

Figure (II.4): Four stages of spin coating process.

II.2.4. Ennaling Thermal Processing

The final stage is thermal treatment to complete the preparation of the thin films. The temperature of the substrate has a significant impact on the characteristics of the produced films. Higher substrate temperatures often result in the production of better crystalline films [3]. The samples are placed in a furnace at a temperature of 500 °C for one and a half hours. This process eliminates the organic compounds present in the starting solution and promotes the densification and crystallization of the material.

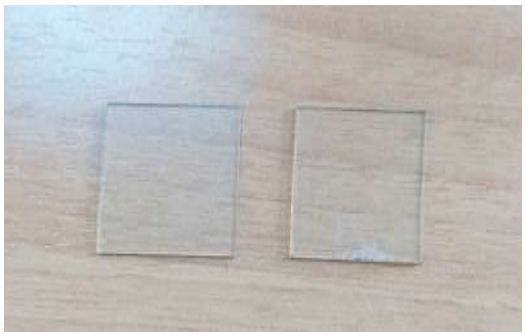

Figure (II.5): Represents the samples prepared after the annealing process.

II.3. Techniques for characterizing thin films

This study explores the structural, surface morphological, electrical, and optical properties of ZnO/CuO thin films. Various advanced characterization techniques are employed to analyze these properties. The crystal structure is examined using X-ray diffraction (XRD), Optical properties are assessed via UV-Visible spectrophotometry by analyzing transmittance and absorbance spectra. Electrical resistivity measurements are conducted using the four-point probe technique and thermoelectric properties.

II.3.1. Structural properties

A thorough structural characterization is essential to understand the properties of these nanostructures. XRD techniques enable the investigation of internal layer structures and the quality of interfaces in multilayer assemblies showing in **Figure(II.6).**

Figure (II.6): XRD Machin.

The principle of X-ray diffraction is based on the ability of X-rays to interact with the atomic structure of a crystalline material. When an X-ray beam is directed at a crystal, the X-rays interact with the electrons of the crystal atoms. This interaction modifies the direction and amplitude of the beam, so that similar to the refraction of light by a lens.

The electrons in a crystal are arranged in a regular and periodic structure, which means that they are aligned at regular intervals in each direction. X-rays that interact with these electrons are subjected to constructive and destructive interference which creates a characteristic diffraction pattern [4]. This pattern is recorded on a detector, which can be used to determine the crystalline structure of the sample.

The principle of X-ray diffraction is based on Bragg's theory, developed by British scientists William Henry Bragg and his son William Lawrence Bragg in 1912. According to this theory, X-ray diffraction is produced when X-rays interact with the atoms of a crystal to create wave network stationery. The resulting diffraction pattern

is determined by the geometry of the crystal structure and the X-ray wavelength like present the **Figure (II.7)**.

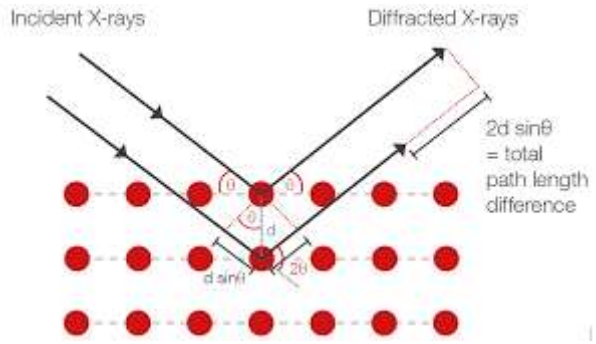

Figure (II.7). X-ray reflection from two planes pf atoms in a solid

The reflection follows Bragg's law as follows **Equation (II.1)**:

$$\mathbf{2}\,\boldsymbol{d}\,\boldsymbol{sin}\,\boldsymbol{\theta} = \boldsymbol{n\lambda} \qquad \textbf{(II.1)}$$

Where:

d is the distance between crystal planes

θ is the X-ray incidence angle

λ is the X-ray wavelength

n is a positive integer

According to Bragg's rule, diffraction is only feasible when (λ < 2d), the set of network planes (h k l) is identified from the standard data using the 'd' values and the lattice parameters of a crystal structure [5]. can be calculated for ZnO and for CuO using the **Equation (II.2) Equation (II.3)**:

$$\mathbf{D} = \frac{\mathbf{0.9\lambda}}{(\boldsymbol{\beta^2}_{\mathbf{obsv}} - \boldsymbol{\beta^2}_{\mathbf{inst}})^{1/2}\mathbf{cos\theta}} \qquad \textbf{(II.2)}$$

$$\boldsymbol{\delta} = \frac{\mathbf{1}}{\mathbf{D^2}} \qquad \textbf{(II.3)}$$

Where λ is the X-ray wavelength (0.154 nm), θ is the Bragg's diffraction angle, β_{obv} and β_{inst} are respectively the full width at half maximum (FWHM) of the experimental peak and the instrumental broadening (in radians).

$$\varepsilon_c = \frac{C_{film} - C_{bulk}}{Cbulk} \times 100\% \quad \textbf{(II.4)}$$

$$\sigma_c = -2.33 \times 10^{11} \times \left(\frac{C_{film} - C_{bulk}}{C_{bulk}}\right) \quad \textbf{(II.5)}$$

Where c_{film} and c_{bulk} the lattice parameters of prepared thin films and bulk ZnO, respectively.

II.3.2. Optical characterization

Optical characterization techniques are becoming important tools for in-situ or real time monitoring in materials research whit advances particularly in the areas of non-intrusive thin films characterization and process control. However, optical probes have serval well-kwon limitation regarding the spectral wavelength range accessible, sensitivity towards surface processes, or their complexity in implementation or interpretation.

The sensitivity of optical probe techniques is determined approximately by the ratio of the penetration depth of light to the thickness of the surface layer, the energy absorbed by light causes disturbances in the electronic structure of the thin layer, resulting in electrons moving from a lower energy level to a higher level, the light beam produces a photon beam at a given wavelength and is divided into two beams, one for reference and the other for linear optical constants of the our piece as shown in **Figure (II.8).**

Figure (II.8): machine of optical characterization

From the latter curve, band gap energy (E_g) can be calculated by Tauc's model [43] using the following **Equation 6** :

$$(\alpha h\upsilon)^n = B(h\upsilon - E_g) \qquad \textbf{(II.6)}$$

Where α represents the absorption coefficient[44], obtained by **Equation 7** :

$$\alpha = \frac{\mathrm{Ln}(\frac{1}{T})}{t} \qquad \textbf{(II.7)}$$

Where T is transmittance, t thickness of films, B is constant and n = 2 for direct band gap energy.

II.3.3. Electrical characterization

The four-point probes are placed collinearly with equal spacing between them on the sample. The current is passed through the two outer probes, and the potential is measured between the two inner probes. The errors due to electrical contacts are absent because the current and voltage leads are separate. This is a widely used technique to measure electrical resistivity by superficial contact.as shown in **Figure (II.9)**.

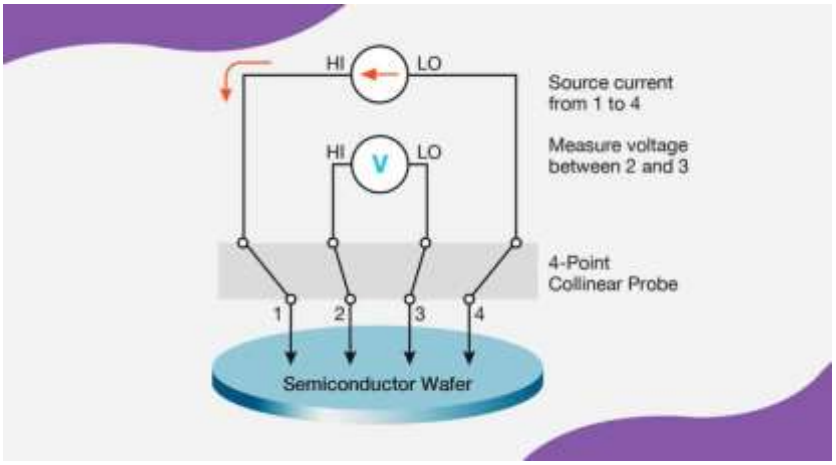

Figure (II.9): Diagram of the principle of the four-point method

This technique is mainly used in the semiconductor industry, research and manufacturing field. In the first experiment, the resistivity of semiconductor crystal is given by **Eq. (8-9):**

$$\rho = (v / I) \times 2\pi S \quad = (V/I) \times (\pi/\ln(2)) \qquad \textbf{(II.8)}$$

$$R = C_f \times (V/I) \qquad \textbf{(II.9)}$$

C_f is the factor that reflects the dispersion of lines between two points, in general situation C_f =4.532, If the four points form a square C_f =9.06.

Resistance and transferability of thin layers can be calculated by the following **Equation (II.10-11)**, And are notably related to the size and thickness of the sample.

$$\rho = R\frac{A}{L} \qquad \textbf{(II.10)}$$

$$\sigma = \frac{1}{\rho} \qquad \textbf{(II.11)}$$

II.3.4. Thermoelectric proprieties

II.3.4.1. Calculate Merit Factor (ZT)

Merit Factor Based on relaxation times and band alignments calculated by the first-principles calculations, transport coefficients (S, σ, and k), which have been defined previously, are acquired by solving the semi-classical Boltzmann equation in the framework of the relaxation time approximation method. In addition, κl can also be calculated with some approximate models, or by solving the Boltzmann transport equation for phonon [7] using the following **Equation (II.12-14)**.

$$ZT = \frac{s^2\sigma T}{k} \qquad \textbf{(II.12)}$$

Seebeck coefficient s/m

Temperate k

Thermal conductivity w/m.k.

$$K = K_{electron} + K_{lattice} \qquad \textbf{(II.13)}$$

$$K_{lattice} = Cv * Vt * Zl \qquad \textbf{(II.14)}$$

Cv heat capacity

Vt photon speed in matter

Zl average photon time in reception

II.3.4.2. Seebeck coefficient Factor Calculation

The ratio between the voltage generated and the temperature gradient to which a material is exposed is known as the Seebeck coefficient. A measurement system to determine the Seebeck coefficient must, therefore, allow for a temperature difference to be established between the two ends of a material. This can be accurately achieved by using two Peltier modules connected in series with reverse bias: when the modules are powered the temperature at one border of the sample increases while it decreases at the other border. In the system reported here, the Peltier modules are mounted on a copper block and are powered by a voltage source that controls the temperature difference applied to the sample, as shown as in **Equation (II.15)**

$$\mathbf{s = \frac{dv}{dT}} \qquad \textbf{(II.15)}$$

dv = voltage difference

dt = change in temperature .

II.3.4.3. Calculate efficiency of the generator η

The ZT factor characterizes the effectiveness of the material, The efficiency η of the module depends on the thermoelectric properties of the materials of which it is made, like calculating in **Equation (II.16-17)**

$$\mathbf{PF = s^2\sigma} \qquad \textbf{(II.16)}$$

$$\boldsymbol{\varphi} = \frac{\mathbf{T^{10-\log T}}}{\mathbf{R_{S_i}}} \qquad \textbf{(II.17)}$$

II.4. Conclusion

In this chapter, the fundamental steps for the deposition of thin films using the sol-gel spin coating technique were presented. The preparation process of the thin films was described, along with a detailed discussion of the methods used to analyze their morphological, structural, and linear optical properties. Furthermore, relevant equations for the calculation of the electrical properties were introduced.

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Chapter III: Results and Discussions

III.1. Introduction

In this chapter, we present the results and discussions of a theoretical and experimental study on a thermoelectric generator. The chapter is divided into two main parts: a simulation of the thermoelectric generator using COMSOL Multiphysics, and an investigation of the effect of current intensity on temperature and heat distribution.

The experimental part focuses on the fabrication and characterization of the thermoelectric materials. Zinc oxide (ZnO type n) thin films are deposited on glass substrates using the spin coating technique, while copper oxide (CuO type p) thin films are deposited separately. We then study the structural, optical, electrical, and finally the thermoelectric properties of these layers to evaluate the overall performance of the thermoelectric generator.

III.2. Simulated Thermoelectric Structure 3D

III.2.1. The geometric structure of the shape and the properties of the materials used

Using a simulation program to develop a prototype model of a thermoelectric generator system, **Figure (III.1a-c)** illustrates a 3D model of a thermoelectric unit simulated using the finite element method (FEM). The geometry consists of a vertical cuboid structure composed of multiple layers and components, each with precise dimensions.

The generator includes a thin film of copper oxide (CuO) deposited on a glass substrate, and on the opposite side, thin films of zinc oxide (ZnO) with a thickness of 0.5 mm. The glass substrate has a thickness of 1 mm, and a copper element is used as the electrical conductor.

Figure (III.1c) shows the temperature distribution within the generator, where the blue color represents lower temperatures and the red color indicates higher temperatures.

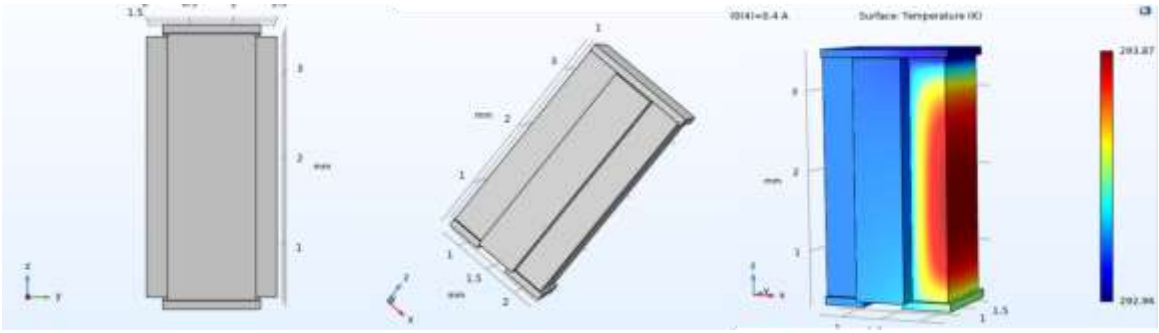

Figure (III.1a-c): Represents thermoelectric materials component.

III.2.2. The effect of electric current on temperature variation

Figure (III.2) illustrates the effect of varying electric current on the temperature distribution across the thermoelectric unit. Four simulation scenarios were conducted with different current intensities: 0.1 A, 0.2 A, 0.3 A, and 0.4 A. The surface temperature patterns for each case are presented, highlighting the system's thermal response to increasing electrical input. As the electrical current increases, the thermal gradient across the thermoelectric element becomes more pronounced. This behavior is attributed to both the Joule heating effect and the Peltier effect, both of which are directly influenced by the magnitude of the current.

- At 0.1 A, the temperature difference is minimal, with surface temperatures ranging from approximately 293.16 K to 293.19 K. A slight thermal accumulation is observed on one side of the unit.
- At 0.2 A, the thermal gradient becomes more noticeable. The hot side reaches around 293.19 K, indicating the early onset of temperature asymmetry across the surface.
- At 0.3 A, heat concentration increases significantly, and the temperature difference becomes more distinct, with surface temperatures rising to approximately 293.28 K.
- At 0.4 A, the most prominent thermal gradient is observed, with surface temperatures ranging between 292.96 K (cold side) and 293.87 K (hot side). The color transition across the surface clearly reflects the enhanced heat flow from one end to the other.

When an electric current flows across the interface between two different materials ZnO type-n and CuO type-p electrons move from a lower energy level in the

ZnO side to a higher energy level in the CuO side. To complete this transition, the electrons must absorb thermal energy from the surrounding medium, which results in localized cooling at the interface. This phenomenon is a manifestation of the Peltier effect.

As the current increases, the amount of heat absorbed or released at the junction correspondingly increases, thereby enhancing the temperature gradient across the interface. This effect is clearly observed in **Figure (III.3)**, where the temperature at the ZnO type-n interface is higher compared to the CuO type-p interface, reflecting the directional flow of heat and the system's thermoelectric response to increased current.

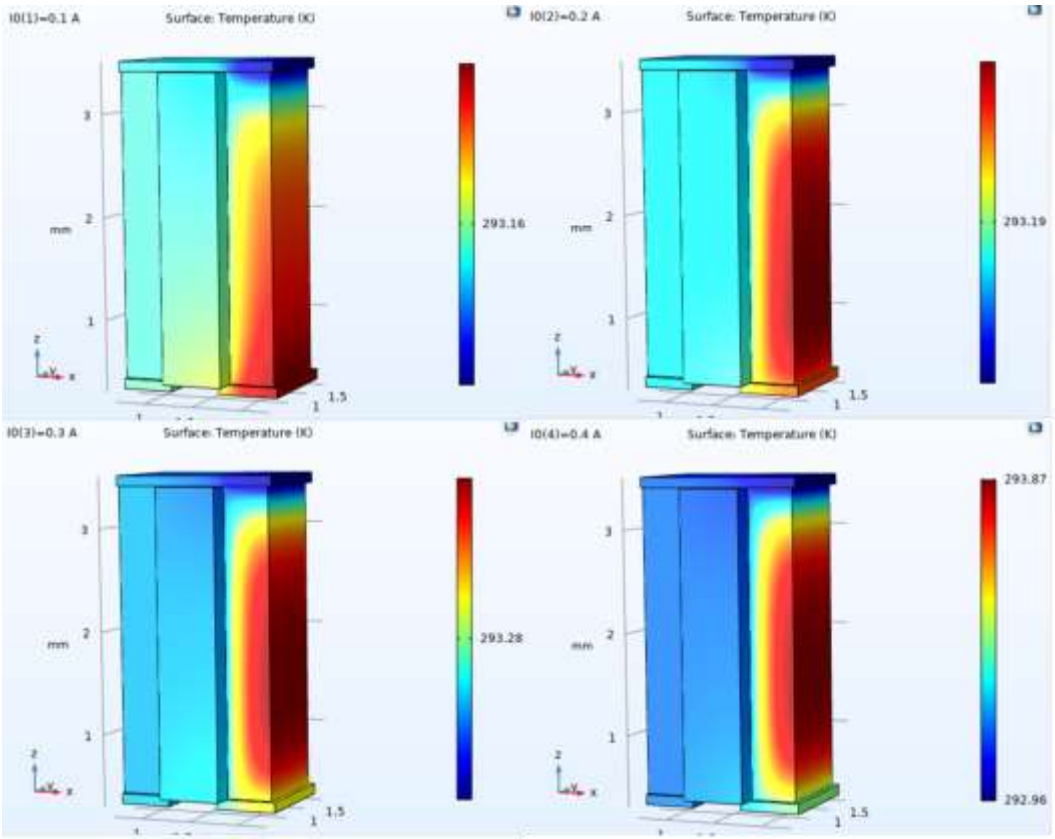

Figure (2.III): Represents images of temperature gradient as a function of current intensity.

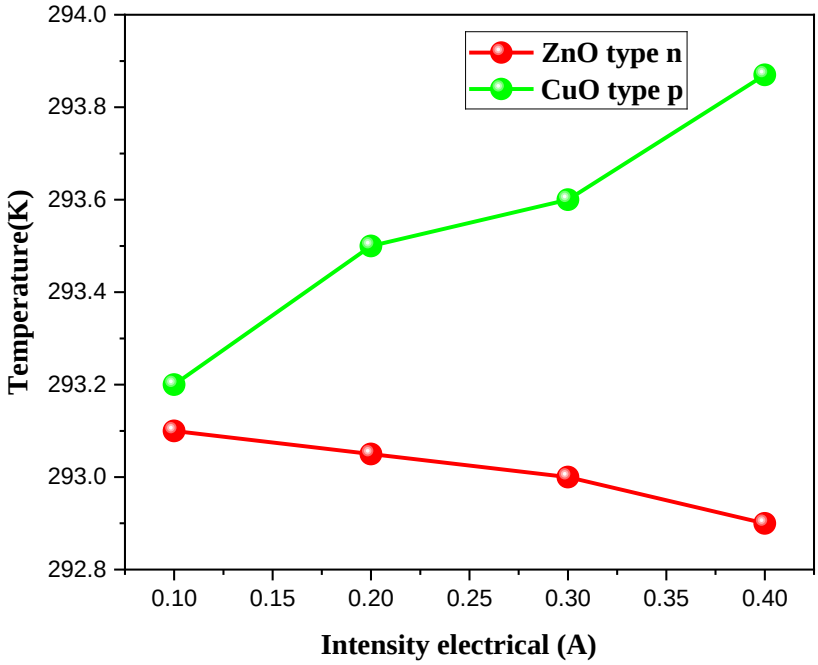

Figure (3.III): values of temperature gradient as a function of current intensity.

III.2.3. Effect of electric current intensity on the junction within the object

As a continuation of **Figure (III.4)**, the isothermal surface Quantitative data (K distribution in volume) visualizations illustrate the spatial distribution of temperature within the thermoelectric generator at different current levels (ranging from 0.1 A to 0.4 A). A gradual increase in both the size and intensity of the hot regions is observed as the current increases:

- At 0.1 A, the temperature rise is slight and remains localized.
- At 0.2 A, a noticeable increase in temperature occurs, accompanied by an expansion of the hot zones.
- At 0.3 A and 0.4 A, heating becomes more intense and widespread, particularly around the core of the junction.

The figure allows us to identify regions of heat concentration within the thermoelectric generator, typically occurring at the interface between different materials. Notably, the areas near the junction between CuO and ZnO exhibit the highest thermal response, clearly indicating the influence of the Peltier effect at the

material interface. This insight is valuable for guiding the design of more efficient thermoelectric systems.

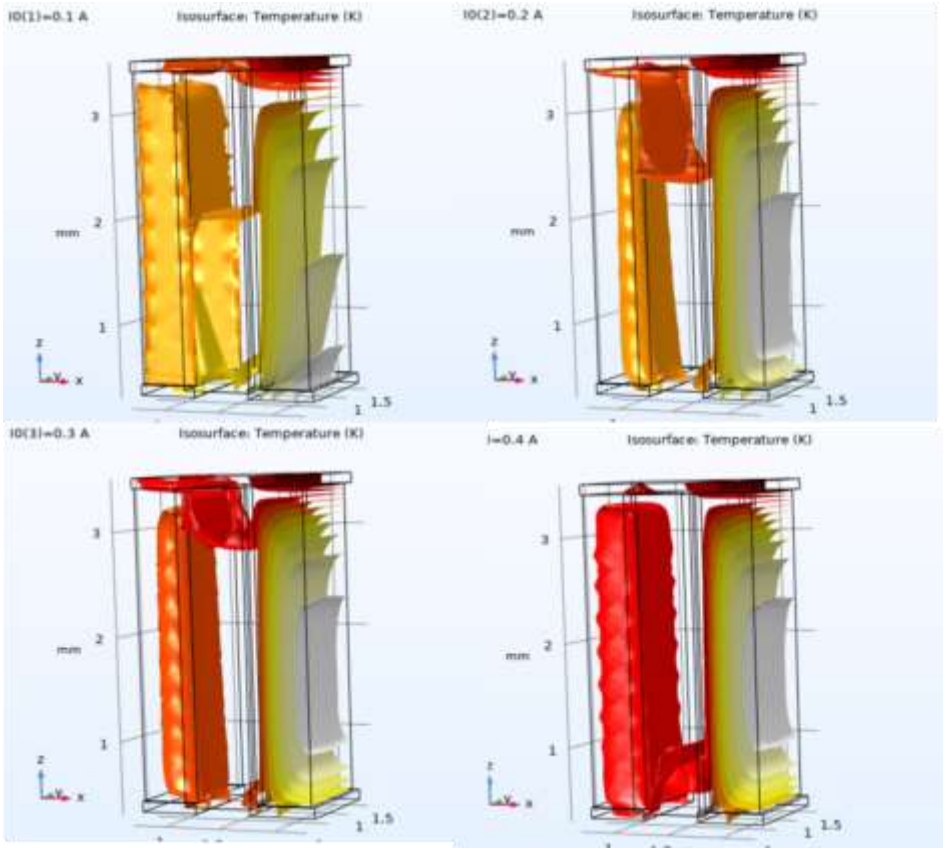

Figure (4.III): Represents the internal junction temperature gradient distribution as a function of current intensity.

III.3. Fabrication and Characterization of CuO/ZnO Thermoelectric Materials

III.3.1. Characterizing of thin films

III.3.1.1. Structural properties

Figure (5a.III) presents the X-ray diffraction (XRD) patterns of the prepared ZnO thin films. The spectra indicate that all films are monocrystalline, with prominent diffraction peaks corresponding to the (002) plane. This peak matches well with the standard ZnO diffraction pattern indexed to the hexagonal wurtzite structure (JCPDS No. 36-1451). The dominance of the (002) peak suggests that the films exhibit a preferred growth orientation along the [002] direction, which is perpendicular to the substrate surface. This orientation is thermodynamically favored, as the (002) plane of

ZnO possesses the lowest surface energy, promoting vertical growth along this axis [1].

In addition **Figure (5b.III)** presents the X-ray diffraction (XRD) patterns of the prepared CuO thin films. The recorded spectra exhibit distinct diffraction peaks characteristic of the monoclinic phase of CuO, consistent with the standard crystallographic data (JCPDS No. 00-048-1548). Prominent peaks are observed at the (002), (−111), (202), and (−311) planes. Notably, the (002) plane, which is oriented perpendicular to the substrate surface, is considered the most thermodynamically stable due to its low surface energy. This preferential orientation aligns well with previously reported results in the literature [2-4].

The XRD analysis reveals clear structural differences between the two films. The ZnO film shows a strong preferential orientation along the (002) plane, indicating a highly oriented crystalline structure with phase purity and vertical growth relative to the substrate. In contrast, the CuO film displays multiple peaks corresponding to various planes, confirming its polycrystalline nature with no dominant orientation. This contrast highlights the structural simplicity and uniformity of ZnO films compared to the more complex and multi-oriented nature of CuO films.

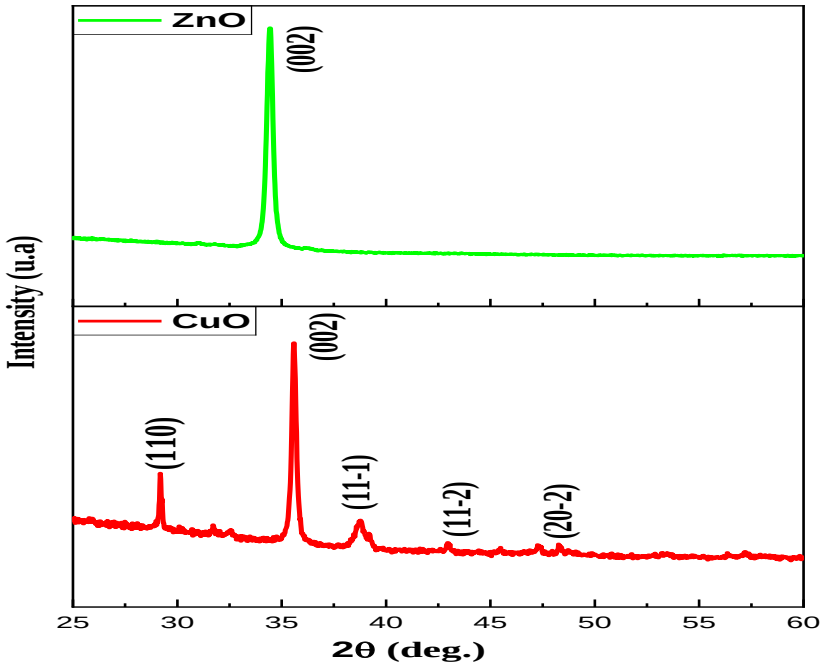

Figure (5a-b.III): XRD patterns of ZnO and CuO thin films.

To gain deeper insight into the structural properties of the ZnO and CuO thin films, the XRD patterns were analyzed to extract several key parameters. The peak position (2θ) and full width at half maximum (FWHM) were directly obtained from the diffraction data. Using these values, additional structural characteristics were calculated the parameters are summarized in the following **Table 1**

Table 1: The peak position 2θ, FWHM, distance between crystal d_{hkl}, crystallite size (D), dislocation density(δ), strain (ε_c) stress (σ_c) of ZnO and CuO thin films.

Samples	2θ(deg.)	FWHM	d_{hkl}(nm)	D(nm)	δ(10^{-3}/nm^2)	ε_c (10^{-3})	σc(GPa)
CuO	35.048	0.126	0.256	66.62	0.225	1.732	0.241
ZnO	34.415	0.331	0.251	25.140	1.582	0.109	0.255-

III.3.1.2. Optical characterization

The optical transmittance spectra of ZnO and CuO thin films are shown in the **Figure (III.6)** above, measured in the UV-Visible range. The transmittance curves exhibit distinct behavior for each material, reflecting differences in optical properties and electronic structure.

- The ZnO thin film (green curve) shows high optical transmittance, exceeding 80% in the visible range (400–800 nm). This high transparency is characteristic of wide-bandgap semiconductors and makes ZnO suitable for applications in transparent conductive oxides (TCOs), UV photodetectors, and solar cells.
- The CuO thin film (red curve) shows lower transmittance, which gradually increases with wavelength. This behavior is expected due to CuO's narrower band gap, resulting in stronger absorption in the visible region. CuO is more suited for photothermal and photocatalytic applications.

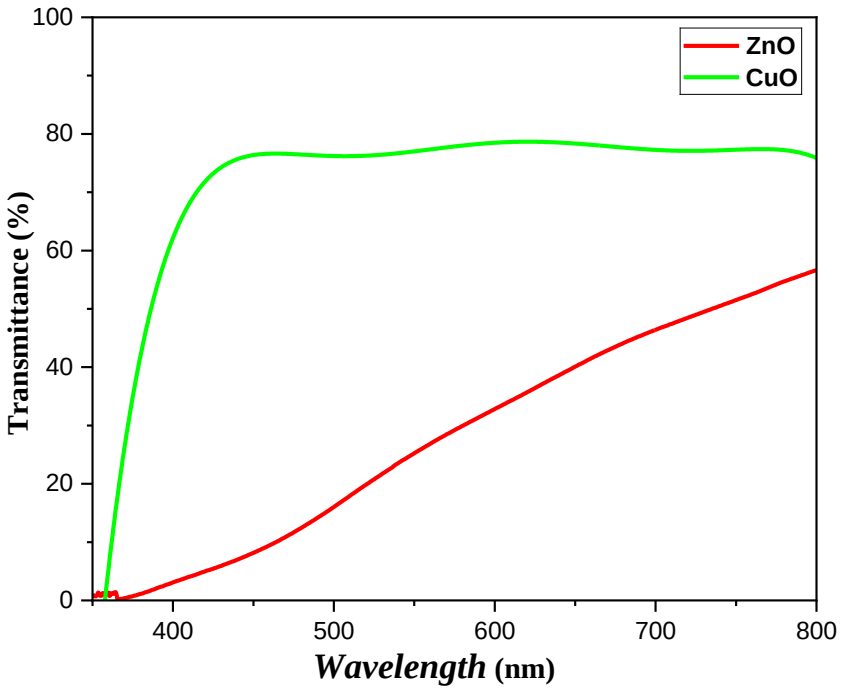

Figure (6.III): Optical transmittance of ZnO and CuO thin films as a function wavelength.

The optical band gap energy of the films was showing in **Table (2.III)**,These values are consistent with literature and confirm the semiconducting nature of the materials. The sharp absorption edge observed in ZnO further supports its crystalline quality, whereas the broader absorption edge of CuO may suggest structural defects or smaller crystallites.

Table (2.III): Band gap energy of ZnO and CuO thin films.

Samples	Eg(eV)
CuO	2.75
ZnO	3.35

III.3.1.3. Electrical characterization

The electrical properties of ZnO and CuO thin films were evaluated through measurements, and the results are presented in **Figure (7-9.III)** above. The graphs illustrate the variation of sheet resistance, resistivity, and electrical conductivity as a function of thermal annealing.

ZnO Thin Films (Red Curve)

The sheet resistance decreases significantly with annealing, indicating improved electrical conductivity. This enhancement can be attributed to better crystallinity, reduced defect density, and increased carrier mobility after annealing. Additionally, the formation of oxygen vacancies during annealing may contribute to an increase in free carriers, further reducing resistance.

CuO Thin Films (Green Curve)

A moderate decrease in sheet resistance is observed after annealing. The effect is less pronounced compared to ZnO, which could be related to the distinct electronic structure of CuO and the dynamics of carrier concentration. Nevertheless, annealing still contributes to improved grain connectivity and a reduction in surface defects.

TEG (Blue Curve)

The heterojunction exhibits relatively low and stable sheet resistance throughout the annealing process. This stability may be attributed to the synergistic conduction behavior of the two oxides and the formation of a stable interface that maintains efficient electrical pathways.

Overall, the data highlight the positive impact of thermal annealing on the electrical performance of both single and hybrid oxide films, particularly ZnO. Such improvements are critical for enhancing the efficiency of these materials in our thermoelectric power generation system.

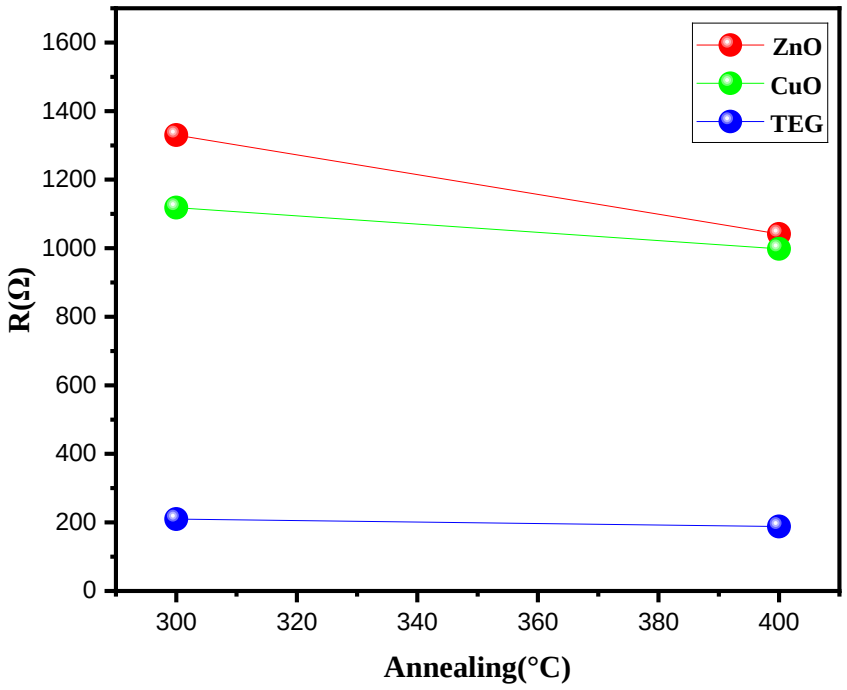

Figure (7.III): Sheet resistance of ZnO and CuO thin films as a function annealing.

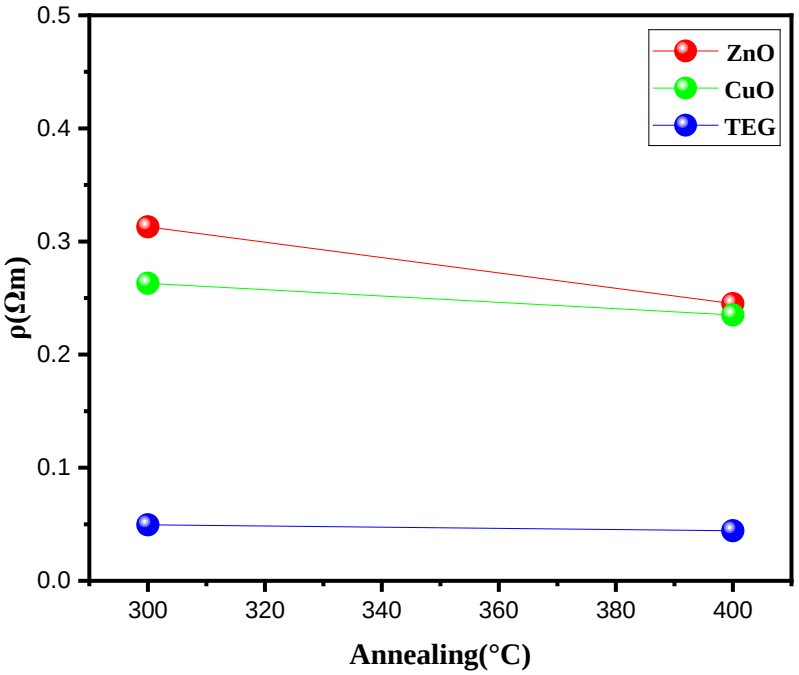

Figure (8.III): Electrical resistivity of ZnO and CuO thin films as a function annealing.

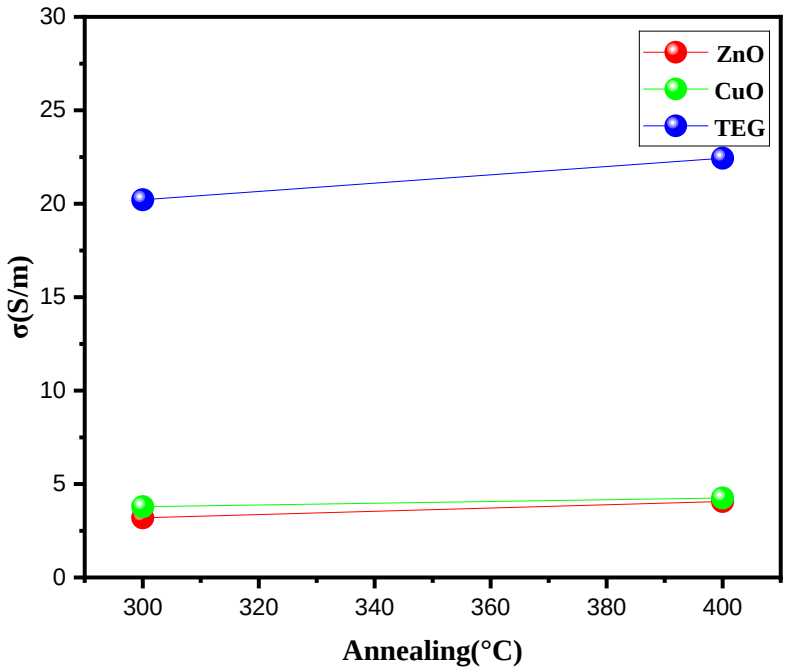

Figure (9.III): Electrical conductivity of ZnO and CuO thin films as a function annealing.

III.3.1.4. Thermoelectric proprieties

Figure (III.10) shows the basic structure of a thermoelectric module based on a CuO/ZnO thin-film junction deposited on a glass substrate. In this configuration:

CuO, type-p semiconductor, is placed on the exterior (hot side) where it absorbs heat energy, while ZnO, type-n semiconductor for example [5], is positioned on the interior (cold side), establishing a temperature gradient across the structure. As heat flows from the exterior to the interior, a thermoelectric voltage is generated through the Seebeck effect [6], with electrons in type-n and holes in the type-p moving in opposite directions. This movement of charge carriers closes the electrical circuit, enabling current generation and allowing the device to convert waste heat into electrical power. A glass layer serves as both a structural support and a thermal barrier, ensuring thermal separation between the two materials and enhancing the device's efficiency.

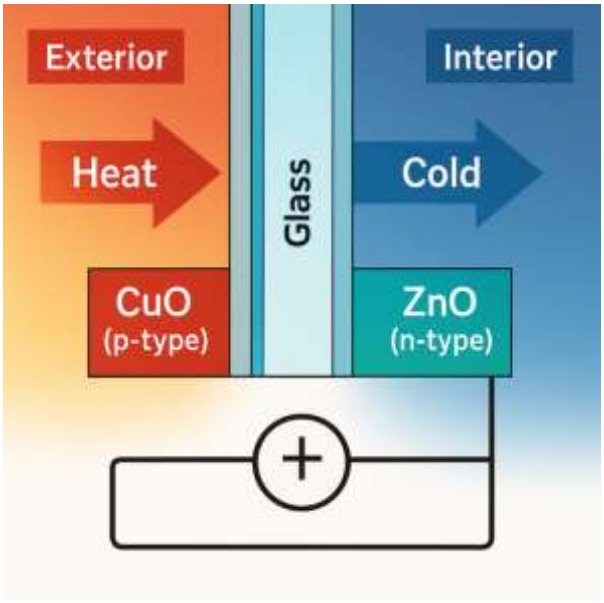

Figure (10.III): Structure of a Thermoelectric Module: p-type and n-type Material films for Heat Transfer.

Table (2.III). show Thermoelectric properties were evaluated for ZnO, CuO, and thermoelectric generator (TEG), thin films annealed at 300°C and 400°C. Key parameters analyzed include electrical conductivity **(σ)**, Seebeck coefficient **(S)**[6], thermal conductivity **(K)**[7], the dimensionless figure of merit **(ZT)**[8], and the conversion efficiency **(η)**[9].

Individual ZnO and CuO thin films exhibit relatively low electrical conductivity, and no Seebeck coefficients were measured for these samples within the current dataset. Consequently, the thermoelectric figure of merit (ZT) and power generation efficiency (η) were not calculated separately for each oxide, indicating limited thermoelectric performance of the individual oxide films under the present conditions. In contrast, the ZnO/CuO thin-film heterojunction structure (TEG) demonstrates a significant enhancement in thermoelectric performance. The electrical conductivity increases markedly, reaching values of 20.21 S/m at 300 °C and 22.44 S/m at 400 °C. This improvement is attributed to the increased concentration of free charge carriers generated by thermal excitation, which enhances electron mobility in ZnO and hole mobility in CuO. Additionally, annealing improves crystalline quality and reduces structural defects, particularly at the interface between the two materials, facilitating charge transfer and boosting electrical conductivity.

Conversely, the Seebeck coefficient decreases from 285.7 μV/K at 300 °C to 200 μV/K at 400 °C. This decline is due to the increased carrier concentration at higher temperatures, which reduces the energy difference across the material and thus lowers the thermoelectric voltage despite improved conductivity. The thermal conductivity remains constant at 1.5 W/m·K, a value assumed to simplify the calculation of the thermoelectric figure of merit (ZT), implying stable heat transport within the studied temperature range. The ZT value is 0.00033 at 300 °C and decreases to 0.00018 at 400 °C, primarily because of the reduction in the Seebeck coefficient with temperature rise, despite the increase in electrical conductivity. This results in a slight decline in overall thermoelectric performance. The power generation efficiency (η) is estimated at 1.649×10^{-6} W/m·K² at 300 °C and decreases to 8.976×10^{-7} W/m·K² at 400 °C, following the trend of ZT, confirming the direct dependence of conversion efficiency on the figure of merit.

These results indicate that annealing significantly improves electrical conductivity but causes a decrease in the Seebeck coefficient, slightly reducing overall thermoelectric performance at higher annealing temperatures. Nevertheless, the TEG structure remains promising for thermoelectric applications [10]. compared to the individual oxide films, due to its enhanced electrical and thermoelectric properties.

Table (3.III): Sheet resistance **Rs,** electrical resistivity **ρ,** electrical conductivity **σ,** Seebeck coefficient **S,** Thermal conductivity **K,** Merit Factor **ZT,** efficiency of the generator **η** as a function annealing**.**

Annealing	Rs ohm	ρ Ωm	σ S/m	S V/K	K W/mK	ZT	η W/mK^2
ZnO 300°C	$12.547*10^{6}$	0.313	3.19	-	-	-	
ZnO 400°C	$9.824*10^{6}$	0.245	4.07	-	-	-	
CuO 300°C	$10.55*10^{6}$	0.263	3.79	-	-	-	
CuO 400°C	$9.417*10^{6}$	0.235	4.25	-	-	-	
TEG 300°C	$1.979*10^{6}$	0.0495	20.21	$285.7*10^{-6}$	1.5	0.00033	$1.649*10^{-6}$
TEG 400°C	$1.774*10^{-6}$	0.0443	22.44	$200*10^{-6}$	1.5	0.00018	$8.976*10^{-7}$

III.4. Conclusion

In this chapter, we conducted both theoretical and experimental investigations to assess the performance of a thermoelectric generator (TEG) based on CuO (p-type) and ZnO (n-type) thin films. The theoretical simulation using COMSOL Multiphysics allowed us to visualize and analyze the influence of current intensity on the temperature and heat distribution across the TEG structure. The results highlighted the critical role of current in shaping the thermal behavior and efficiency of the system.

On the experimental side, ZnO and CuO thin films were successfully deposited using the spin coating technique. Comprehensive structural, optical, electrical, and thermoelectric characterizations were carried out. Structural analyses confirmed improved crystallinity and reduced defects with annealing, particularly at 400°C. Optical measurements showed ZnO's higher transparency and a suitable band gap for energy applications, while electrical studies revealed enhanced conductivity with thermal treatment.

Thermoelectric measurements of the assembled TEG structure showed modest but functional performance, with an increase in Seebeck coefficient and power factor, and a maximum ZT of 0.00033 at 300°C. These findings confirm the potential of CuO/ZnO thin films for low-cost, eco-friendly thermoelectric applications. The integration of simulation and experimental approaches in this study provides a solid foundation for future optimization and development of oxide-based thermoelectric generators.

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Chapter IV: Explanation of the proposed project

IV.1. Introduction

In the context of growing global energy demands and environmental concerns, there is a pressing need for innovative technologies that can reduce energy consumption while maintaining thermal comfort in buildings. Thermoelectric materials offer a promising solution by enabling direct conversion between thermal and electrical energy. Chapter 4 focuses on a novel application of these materials, specifically ZnO (n-type) and CuO (p-type) thin films, as smart coatings for glass surfaces. This chapter aims to provide a comprehensive explanation of the proposed project, which integrates thermoelectric principles into a practical and sustainable energy-saving system.

The proposed system is designed to regulate heat transfer across building envelopes, adapting to seasonal variations by either conserving indoor heat in winter or minimizing heat gain during summer. Simultaneously, the thermoelectric films are capable of generating electricity from ambient temperature gradients, contributing to the energy independence of the building. The chapter is structured to present the project's conceptual framework, its specific objectives, the operational mechanism of the system, its cost-effectiveness, practical applications, environmental benefits, and long-term vision. Through this multifaceted approach, the project aims to bridge the gap between advanced materials research and real-world energy solutions, ultimately contributing to the development of smarter, greener, and more efficient buildings,it will be divided into several sections as follows:

IV.2. Idea of the project and its importance

- The project aims to develop a system based on thermoelectric materials to regulate the consumption of thermal energy in buildings whit generated electrical energy from the heat sources.
- The innovation lies in the use of CuO type p and ZnO type n thin films as an intelligent coating on glass, allowing control of the heat flux between the interior and the exterior depending on the seasons. In addition, it generates electrical energy for daily use.
- The project seeks to reduce energy use for heating and cooling, thereby helping to reduce the carbon footprint and promote environmental sustainability.

IV.3. Project objectives

- Achieve high efficiency in heat conversion and transfer using thermoelectric materials to generate electrical energy.

- Develop a practical and easy-to-manufacture solution that can be applied on a large scale.
- Provide an innovative product that improves thermal comfort and reduces energy costs.

IV.3.1. System operation

- P-type CuO and n-type ZnO films will be deposited on glass substrates using advanced techniques.
- When the temperature difference between the indoor and outdoor environment is present, these films generate an electric current or transfer heat reciprocally, resulting in optimal thermal equilibrium.
- System performance will be adjusted to suit ambient temperature and seasons in the **Figure (IV.1)**.

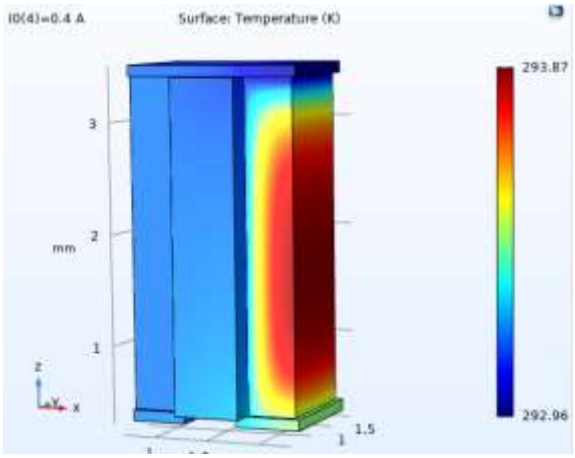

Figure (IV.1): Represents the thermoelectric material component of the generator.

IV.3.2. Cost-effective and convenient applications

- **Economic profitability:** The project will result in substantial energy savings, making the system economically viable in the long term.
- **Practical applications:** This system can be used in residential and commercial buildings, vehicles, and even in electronic devices requiring efficient cooling.

The cost to elaborate a $1m^2$ of system: ..

$1m^2$ Matérielle: ..

$1m^2$ Elaboration: ..

IV.3.3. Environmental and social impact

- The project helps reduce reliance on traditional energy sources, thereby reducing greenhouse gas emissions.
- It also improves thermal comfort and reduces energy costs for consumers, thus having a positive impact on quality of life.

IV.3.4. Future Vision

The project aims to develop even more advanced solutions, such as the integration of thermoelectric materials with solar systems or thermal storage devices, to further improve efficiency and broaden the scope of application.

IV.4. Conclusion

This chapter presents an innovative thermoelectric solution aimed at improving energy efficiency and environmental sustainability in modern buildings. By leveraging the unique properties of zinc oxide and copper oxide thin films, the proposed system offers a dual-functional approach—regulating heat exchange and generating electrical power from temperature gradients. Its integration into glass surfaces enables both passive and active energy management, adapting to seasonal changes and building needs. With potential applications in both the residential and commercial sectors, the system promises not only to reduce energy consumption and costs, but also to lower carbon emissions and enhance occupant comfort. Looking to the future, this innovation lays the foundation for more advanced hybrid systems that combine thermoelectric coatings, solar panels, and thermal storage technologies. These developments will further support the global transition towards smart, energy-independent, and environmentally friendly infrastructure.

General Conclusion

This study highlights the promising potential of thermoelectric materials specifically CuO and ZnO thin films as innovative solutions for efficient energy management and sustainable development. Through a comprehensive exploration of their structural, optical, electrical, and thermoelectric properties, we have demonstrated their viability for use in smart systems designed to convert thermal gradients into electrical energy and vice versa. Theoretical analysis and simulation using COMSOL Multiphysics provided valuable insights into the behavior of these materials under different conditions, confirming the influence of electric current intensity on thermal and electrical performance. The proposed application, which integrates these materials as intelligent coatings on glass surfaces, offers a dual benefit: dynamic regulation of heat flow between indoor and outdoor environments, and generation of electrical power. This approach addresses both energy efficiency and environmental impact, making it a strong candidate for commercialization as a startup or patented technology. The project not only meets technical and practical objectives but also aligns with global goals for reducing energy consumption and carbon emissions. Looking forward, the integration of thermoelectric materials with renewable energy systems such as solar or thermal storage could further enhance their effectiveness, marking a significant step toward smarter, greener buildings and devices.

This work presents an in-depth exploration of thermoelectric materials—particularly ZnO (zinc oxide) and CuO (copper oxide) thin films—and their application in energy-efficient systems. Thermoelectric materials hold immense promise due to their dual capability to convert heat into electricity (via the Seebeck effect) and electricity into heat (via the Peltier effect), making them ideal candidates for smart thermal management systems. The experimental and simulation-based approach adopted in this study has provided comprehensive insight into the behavior of these metal oxide semiconductors under various thermal and electrical conditions.

Structurally, the ZnO and CuO thin films synthesized and deposited using spin-coating techniques exhibited good crystallinity, as confirmed through X-ray diffraction (XRD) analysis. The calculated parameters such as crystallite size, dislocation density, and strain indicate improved structural quality with annealing. Optically, ZnO displayed high transparency and a wide bandgap of 3.35 eV, whereas CuO, with a narrower bandgap of 2.75

eV, showed good light absorption properties, suggesting their complementary roles in optoelectronic and energy harvesting applications.

Electrically, both materials demonstrated improved conductivity after annealing, with CuO serving as the p-type and ZnO as the n-type component of the thermoelectric junction. The measurements of sheet resistance and Seebeck coefficients further confirmed their thermoelectric behavior. Simulation results using COMSOL Multiphysics clearly illustrated the impact of increasing electric current intensity on temperature gradients and electric potential across the thermoelectric module, validating the role of Joule heating and thermoelectric effects in device performance.

Importantly, this study goes beyond characterization to propose a practical and sustainable application: using CuO/ZnO thin films as a smart coating for glass surfaces. This concept enables dynamic thermal control by regulating heat transfer between the interior and exterior environments based on seasonal conditions reducing heat gain in summer and retaining warmth in winter. Additionally, it allows for the generation of small-scale electrical power, contributing to energy independence in buildings.

From a socio environmental perspective, this innovation offers several benefits: it reduces dependency on traditional heating and cooling systems, lowers energy bills, and decreases greenhouse gas emissions. The solution is scalable and cost-effective, with potential applications in residential buildings, commercial infrastructures, vehicles, and consumer electronics.

Looking ahead, the integration of these thermoelectric materials with renewable energy technologies such as photovoltaic systems or thermal energy storage could further enhance their efficiency and broaden their scope. This positions the project as a promising candidate for patenting or commercialization, with the potential to launch an innovative start-up focused on sustainable energy solutions.

ملخص

تهدف هذه الدراسة إلى استكشاف وتوظيف المواد الكهروحرارية، وخاصة أغشية ZnO (نوع n) وCuO (نوع p)، كحلول ذكية لتحسين كفاءة استخدام الطاقة في المباني. تعتمد هذه المواد على تحويل الفرق الحراري إلى طاقة كهربائية والعكس، من خلال تأثيرات سيبيك، بيلتييه، وطومسون. تم تحضير الأغشية الرقيقة ودراسة خصائصها الفيزيائية والكهروحرارية، إلى جانب محاكاتها باستخدام برنامج COMSOL Multiphysics. يقترح المشروع استخدام هذه الأغشية كطلاء ذكي للنوافذ الزجاجية لتنظيم انتقال الحرارة حسب الفصول وتوليد الكهرباء من الفروق الحرارية، ما يسهم في تقليل استهلاك الطاقة، وتحقيق راحة حرارية، ودعم الاستدامة البيئية.

Abstract

This study aims to explore and employ thermoelectric materials, specifically ZnO (n-type) and CuO (p-type) thin films, as smart solutions to enhance energy efficiency in buildings. These materials rely on converting thermal differences into electrical energy and vice versa through the Seebeck, Peltier, and Thomson effects. The thin films were prepared, and their physical and thermoelectric properties were studied, along with simulations using COMSOL Multiphysics software. The project proposes using these films as smart coatings for glass windows to regulate heat transfer according to seasons and generate electricity from thermal gradients, thereby reducing energy consumption, improving thermal comfort, and supporting environmental sustainability.

Résumé

Cette étude vise à explorer et à utiliser des matériaux thermoélectriques, en particulier des couches minces de ZnO (type n) et de CuO (type p), comme solutions intelligentes pour améliorer l'efficacité énergétique des bâtiments. Ces matériaux reposent sur la conversion des différences thermiques en énergie électrique et vice versa, grâce aux effets Seebeck, Peltier et Thomson. Les couches minces ont été préparées et leurs propriétés physiques et thermoélectriques ont été étudiées, ainsi que des simulations réalisées avec le logiciel COMSOL Multiphysics. Le projet propose d'utiliser ces films comme revêtements intelligents pour les fenêtres en verre afin de réguler le transfert de chaleur selon les saisons et de générer de l'électricité à partir de gradients thermiques, réduisant ainsi la consommation d'énergie, améliorant le confort thermique et soutenant la durabilité environnementale.