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TITANIUM DOPED COPPER (II) OXIDE THIN FILMS FOR SOLAR CELL APPLICATION

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Abstract

There has been a need for a more convenient, greener, and efficient material for energy conversion and electronic applications many years ago. Cu_2O thin films, produced using the spray pyrolysis method, offer a solution that meets economic viability and cost requirements. It is widely believed that these films will enable the development of functional technologies. In this study, spray pyrolysis was used to add titanium to copper (I) oxide thin films. The deposition temperature was set at 200°C, and the films were annealed for 2 hours at the same temperature. Investigation of optical, surface morphology, and photovoltaic characteristics of the resulting Ti-doped Cu_2O films were thoroughly carried out. The best features were observed in the Cu_2O films doped with 3% titanium. One notable change observed in the Ti-doped Cu_2O films was a shift in the near-band emission from 385 nm to 400 nm. Additionally, the band gap of the films decreased from 2.35 to 1.98 eV when doped with 3% titanium. These changes resulted in significant improvements in the short circuit current density and open circuit voltage of Cu_2O (Ti)-based solar cells. Overall, the addition of titanium to p-CuO has shown promising results in enhancing the optical and photovoltaic properties of Cu2O thin films. This research paves the way for the development of more efficient and cost-effective energy conversion and electronic devices.

Keywords: *Characterization, Spray pyrolysis, Open circuit voltage, Deposition temperature, thin film, and Solar cell.*

1. Introduction

Research is being conducted on photovoltaic systems to develop sustainable and clean energy solutions [1]. One material being studied is cuprous oxide (Cu_2O), which is a low-cost semiconductor with high absorption capabilities [2]. Cu_2O has several advantages including non-toxicity, low cost, and a high absorption coefficient [3]. In sunlight, Cu_2O exhibits p-type semi conductivity and has a direct band gap range of 2.10 to 2.60 eV, making it efficient for power conversion. However, there are still challenges to overcome in harnessing the full potential of Cu_2O [4, 5].

Homojunction solar cells made with Cu_2O have not been performing effectively, with lower efficiency than predicted [6]. This is due to the poor electrical properties of Cu_2O films, such as low conductivity and short minor carriers of diffusion length [7]. These properties make it difficult for electrons and electrodes to move to holes, resulting in significant recombination in the bulk. Work have been on enhancing the optical and electrical properties of Cu_2O by using dopant elements such as Mn, Fe, Ni, F, and Zn [8].

Yu et al [6] used an electrochemical deposition method to create F-doped Cu_2O thin films with varying levels of fluorine. These films exhibited excellent electrical and optical properties, particularly when the F/Cu ratio was 1:2. Another study by Oluyamo et al. [7] explored the effects of doping Cu_2O thin films with carbon nanotubes (CNTs) using spray pyrolysis at low temperatures. The doping of CNTs resulted in changes in the optical properties of the thin films, such as their absorption and extinction coefficients, as well as the refractive index. Doped samples showed reduced absorption in the ultraviolet region and lighter absorption in the visible range of the electromagnetic spectrum.

It has been found that heating samples increases both the absorbance and extinction coefficients, making them suitable for use as an absorbance layer in device manufacturing. One study achieved a conversion efficiency of 1.06% in p-n homojunction Cu_2O solar panels by improving the front contacts. Another study reported a conversion efficiency of 0.89% in the synthesis of Cu_2O homojunctions. A homojunction Cu_2O solar cell with a 2.05% efficiency was developed using Cl-doped and Na-doped Cu_2O films. Additionally, a p-n homojunction Cu_2O cell with a 0.1% conversion efficiency was produced by electrochemically developing a p-type Cu_2O film on an n-type Cu_2O film.

Ti-doped Cu_2O has gained attention for its advantageous properties, including being lightweight, having high strength and elastic modulus, and being corrosion-resistant. Lavanya et al [9] conducted experiments using the DC magnetron sputtering process to dope Ti on Cu_2O thin films. By increasing the bias voltages from 360 V to 390 V, they observed a decrease in electrical resistivity and optical band gap values. Additionally, varying the Argon/Oxygen gas flow ratios affected the magnitude of the (111) peak in the produced Ti-doped Cu_2O thin films. These findings highlight the potential of Ti-doped Cu_2O for various applications.

Also advancements have been made in the development of p-n homojunction Cu_2O solar panels through successive electrochemical deposition of p-doped and n-doped Cu_2O layers. McShane and colleagues [10] achieved a conversion efficiency of 0.29%, which was increased to 1.06% by strengthening the front contacts [11]. Wijesundara et al. [12] reported a 0.89% efficiency, while Elfadill et al. reached 2.05% efficiency using optimized films [8]. Han et al [13] achieved a 0.1% conversion efficiency by electrochemically developing a p-type Cu_2O film on an n-type Cu_2O film. Jayathilaka et al. [14] varied the thickness of the p- and n-type Cu_2O films to produce p-n homojunctions.

Ti-doped Cu_2O thin films have attracted attention due to the favorable properties of titanium, such as lightweight, high strength, and corrosion resistance [15]. Lavanya et al. [5, 9] have used the DC magnetron sputtering process to dope Ti on Cu_2O films and observed that increasing bias voltages from 360 V to 390 V resulted in reduced electrical resistivityof the films from 1.4×10^{-4} to 0.88×10^{-4} m and optical band gap valueswere also reduced from 3.71 to 3.58 eV. Additionally, the researchers varied the Argon/Oxygen gas flow ratios and found that the (111) peak magnitude increased with a decrease in oxygen concentration. These findings highlight the potential of Ti-doped Cu_2O films for various applications.

The spray pyrolysis method was chosen to synthesize thin films in this study [16 -18] due to its advantages, such as low cost, perfect mixing, ease of use, and uniformity. The research aimed to investigate the impact of titanium doping on p-type copper (I) oxide thin films using this method, which has not been previously explored. The optical properties of the Ti-doped Cu_2O thin film were examined using a spectrophotometer, while the chemical bonds were analyzed using Fourier transform infrared (FTIR) scanning. Scanning Electron Microscopy (SEM) and X-ray diffraction (XRD) were respectively used to investigate the surface morphology and crystallinity of Ti: Cu_2O.

The heterojunction solar cells of n-Si/p-Cu_2O and 3%Ti-*n*-Si/p-Cu_2O were evaluated.

2. Materials and method

2.1. Preparing the precursor solution

The Ti: Cu_2O films were created using the spray pyrolysis method. Various chemicals were used, including copper chloride, hydrochloric acid, acetone, hydrogen peroxide, sodium hydroxide, ammonia, and titanium trichloride were all purchased from Jeo-Chem Ventures, Nsukka, Enugu State. To prepare the precursor solution, copper chloride was dissolved in distilled water and stirred until a clear solution formed. Titanium chloride solution was then added to the copper chloride solution, and ammonia was added as a complexing agent. The solution was stirred for an hour using a magnetic stirrer.

2.2. Thin-film preparation

Titanium doped thin films were fabricated using the spray pyrolysis technique. The precursor solutions were deposited on a microscopic glass substrate. The spray pyrolysis machine atomizes the precursor solution into tiny droplets, which are then sprayed onto the heated substrate. The machine was powered by an electric motor that converts electric energy into mechanical energy. This energy was conveyed to a pulley system via a conveyor belt. The rotating rod causes the pulley to spin, transferring motion to the compressor. The compressor collects air from the atmosphere through a suction valve and transfers it through a discharge valve.

The compressed air, at a pressure of approximately 3 bar, was released from the air tank and goes through a filter to remove impurities like oil, water, and dust. The filtered air then enters the atomizer, where a precursor solution was turned into small droplets. These droplets are sprayed onto a heated glass substrate. Afterward, the coated glass slides undergo post heat treatment by placing them on ceramic plates in 200◦C furnaces for 2 hours. Once the annealing time was complete, the furnace was turned off, and the slides were allowed to cool to room temperature. The cooled glass slides are then analyzed for their crystallinity, morphology, and optical properties.

2.3. The characterization of thin films

The microstructure of the films was examined using VEGA 3 TESCAN scanning electron microscopy. X'Pert Pro model X-ray diffraction was used to analyze the film's phases. The optical properties were assessed using UV-visible spectroscopy model HP 8453, Agilent. The photoluminescence of the samples was measured using a Fluoromax-4 spectrofluorimeter. The carrier concentration, hall mobility, and electrical resistivity were determined using Kaise insulation model SK5010. The I-V characteristics of the heterojunction solar cells of n-Si/p-Cu_2O and 3%Ti-n-Si/p-Cu_2O were evaluated using the solar cell meter model Keithley 2450.

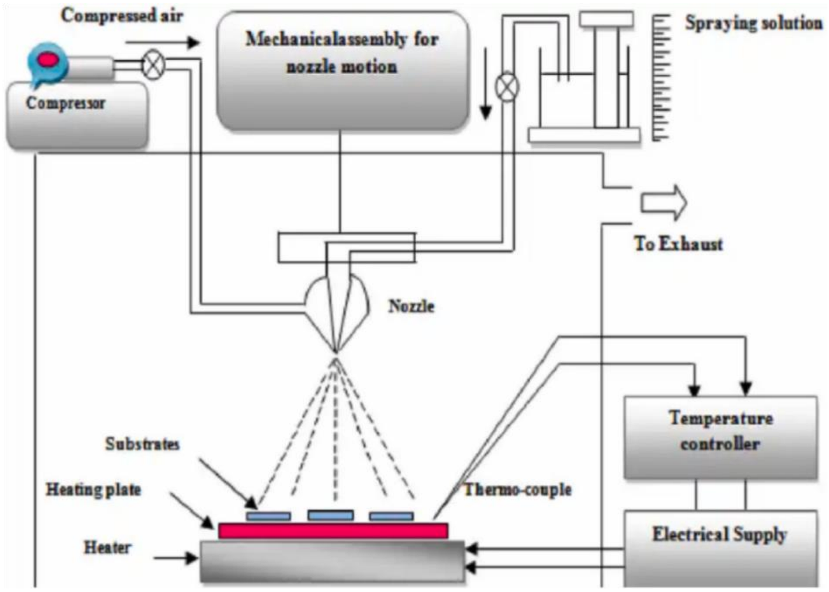

Fig. 1. Diagram showing the systematic operation of the spray pyrolysis technique.

3. Results and discussion

3.1. Analysis of the UV-VIS spectrum

The microstructure of the films was examined using VEGA 3 TESCAN scanning electron microscopy. X'Pert Pro model X-ray diffraction was used to analyze the film's phases. The optical properties were assessed using UV-visible spectroscopy model HP 8453, Agilent. The photoluminescence of the samples was measured using a Fluoromax-4 spectrofluorimeter. The carrier concentration, hall mobility, and electrical resistivity were determined using Kaise insulation model SK5010. The I-V characteristics of the heterojunction solar cells of n-Si/p-Cu_2O and 3%Ti-n-Si/p-Cu_2O were evaluated using the solar cell meter model Keithley 2450. The results of the data gotten from absorption spectra for Ti: Cu_2O thin films are shown in Fig. 2.

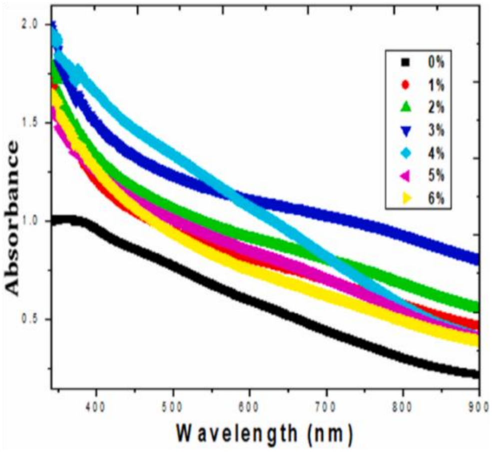

Fig. 2: Absorption spectra for Ti: Cu_2O

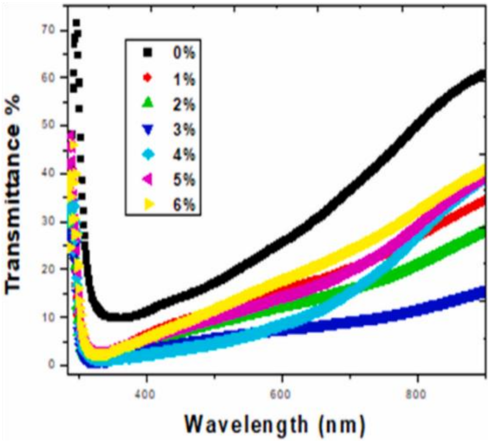

Fig. 3: The transmission spectra for Ti: Cu_2O

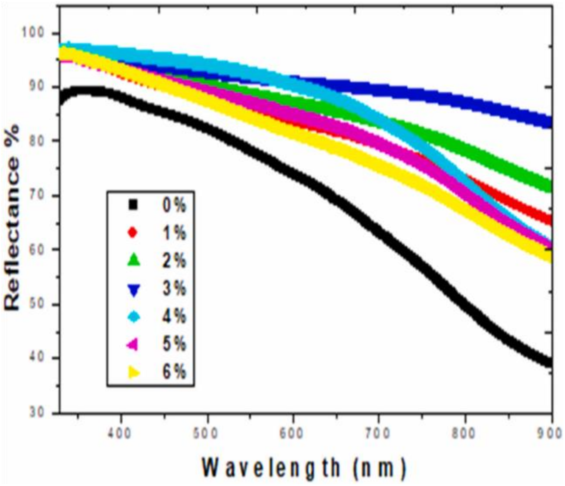

Fig.4: The reflectance spectra for Ti: Cu_2O

The transmittance graph shows how much light was transmitted through a sample. In this case, the undoped Cu_2O film allows more light to pass through compared to the doped region. The graph also considers the effectiveness of reflecting photon radiant energy in the visible light wavelength range for 0%, 1%, 2%, 3.4%, 5%, and 6% Ti doping, respectively [Fig. 3].

The undoped thin-film shows a regular pattern in this relationship. The drop in transmittance was attributed to the crystallinity of the deposited thin films, which makes it difficult for light waves to flow through [13,14]. This reduction in transmittance suggests that the Ti: Cu_2O film can be used as an anti-reflection coating in solar cells.

Additionally, a graph of reflectance versus wavelength is presented in Figure 4.

Thin films deposited with 3% Ti have the highest percentage reflectance in the visible range but the reflectance decreases as the wavelength increases. The reduction of thermal stress contributes to the increase in reflectance. The fraction of incident light also increases with wavelength.

The main focus of this work is to increase the bandgap of Ti: Cu_2O below the value reported in previous studies. The addition of Ti to Ti/Cu_2O samples results in a slight decrease in the bandgap as well as a decrease in deep level emission and near-band-edge band energy. This was attributed to Ti enhancing the carrier concentration and mobility of the material, creating an electronic pathway, and lowering resistivity. Doped Ti ions fill the interstitial vacancies of Cu_2O, creating free electrons and improving conductivity while reducing grain boundary scattering.

Doping the Cu_2O lattice with Ti affects the bandgap, was calculated using the Tau'c plot. The optical bandgap values for different doping percentages of Ti in Cu_2O were found to be 2.35, 2.24, 2.15, 1.98, 2.04, 2.00, and 1.99 eV [Fig. 5].

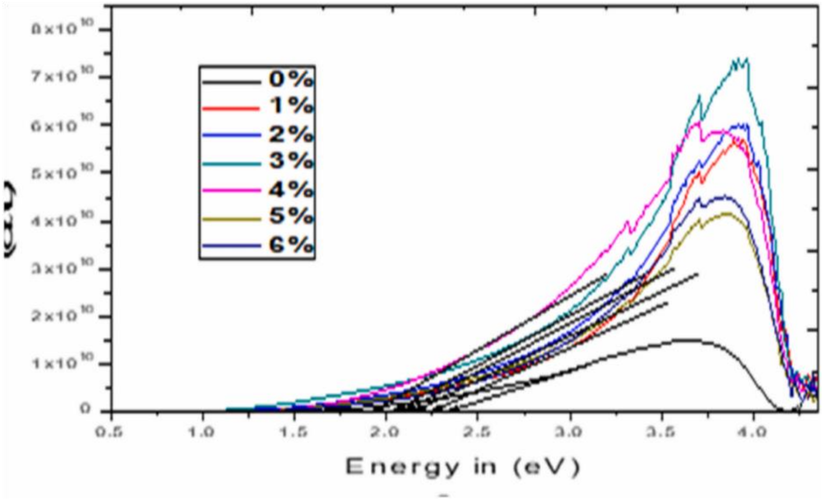

Fig. 5. Band Gap Energy (Eg) for Cu_2O thin film.

The development of 3-D dense structures and conductive network configurations in the Cu_2O lattice by Ti has resulted in the creation of conductive paths that allow for quick movement of charge carriers. The direct contact of Ti in Cu_2O leads to both non-ohmic and ohmic conduction, while indirect contact of Ti in Cu_2O also occurs. Further analysis was conducted on 3%Ti/Cu_2O, which exhibited the most promising properties, including the lowest bandgap.

Doping Cu_2O with Ti improves its electrical conductivity by increasing carrier concentration and mobility. The presence of Ti ions fills vacancies in the Cu_2O lattice, resulting in the formation of a conductive network. This leads to the reduction of resistivity and the promotion of free electron movement within the material. The direct contact between Ti and Cu_2O enables both non-ohmic and ohmic conduction, facilitating the swift travel of charge carriers. Overall, the addition of Ti enhances the electrical properties of Cu_2O thin films.

3.2 AFM surface

The surface roughness of doped Ti: Cu_2O was measured using the Bruker Dimension Icon AFM instrument. The AFM pictures in Figure 6 show that the surface roughness of the thin films remains nearly the same regardless of the Ti content. Increasing the Ti content to 3% has only a minimal impact on the surface roughness. The presence of Ti and Cu_2O in the formulation leads to the formation of islands, which actually decreases the surface roughness [12]. This was because the island construction helps to release stored energy and reduce tension. Further increases in Ti content beyond a certain point were not discussed in the given text.

The addition of titanium (Ti) to copper oxide (Cu_2O) films has been found to enhance their electrical properties without compromising their crystal clarity, optical qualities, and surface form. Specifically, the incorporation of Ti into Cu_2O films leads to an increase in surface roughness and a decrease in surface mobility in extensively doped materials. This was due to the expansion of grain laterally when more Tiwas added to the film. Overall, the inclusion of Ti in Cu_2O films offers a promising approach to improve their electrical performance while maintaining their desired characteristics [13].

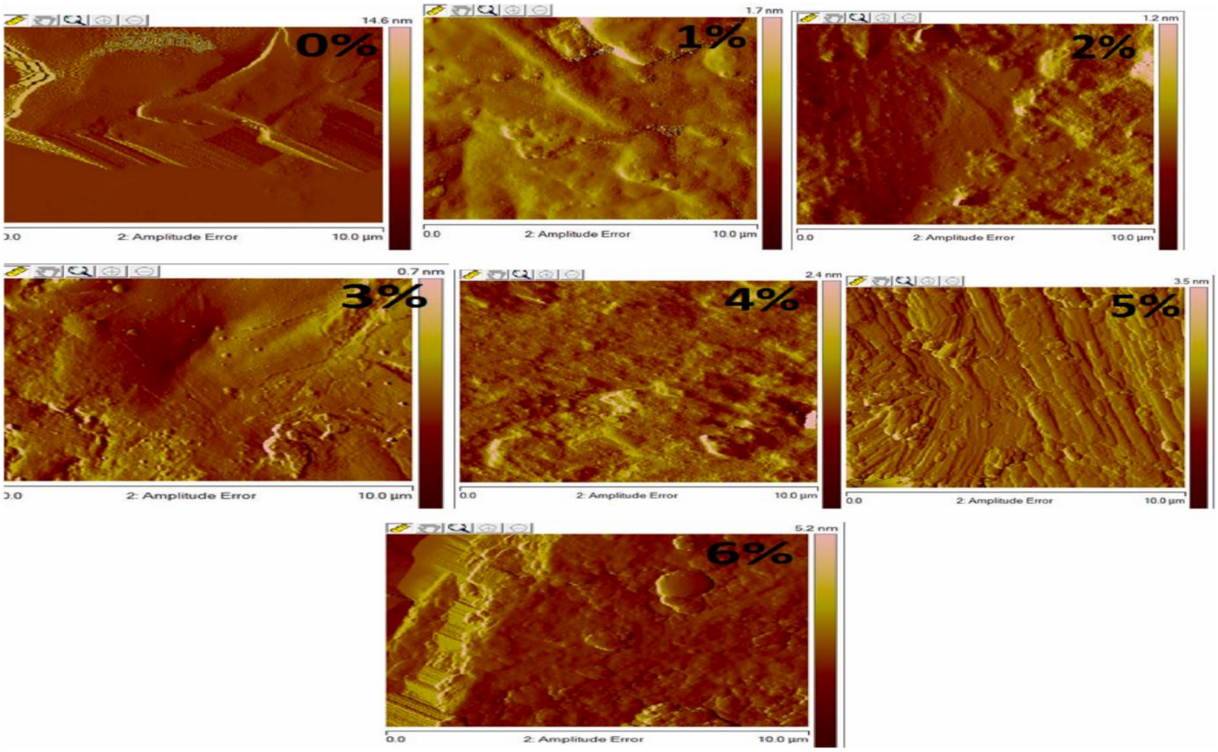

Fig. 6. AFM surface morphology of Ti: Cu_2O thin films.

3.3 FTIR analysis

The functional group of the deposited thin films was analyzed using FTIR. The results [Fig.7] showed significant changes in the peaks after doping with titanium. Several peaks were observed that could be attributed to the addition of Ti. These peaks included stretching modes OH and NH, C–O symmetric stretching, C–H stretching, C–N stretching, C–O stretching, and N–H wagging. The analysis also revealed that at 0%Ti concentration, the ratio of light energy falling on the thin films to that transmitted through them increased as the wavelength increased from 500 to 4000 cm^{-1} [13].

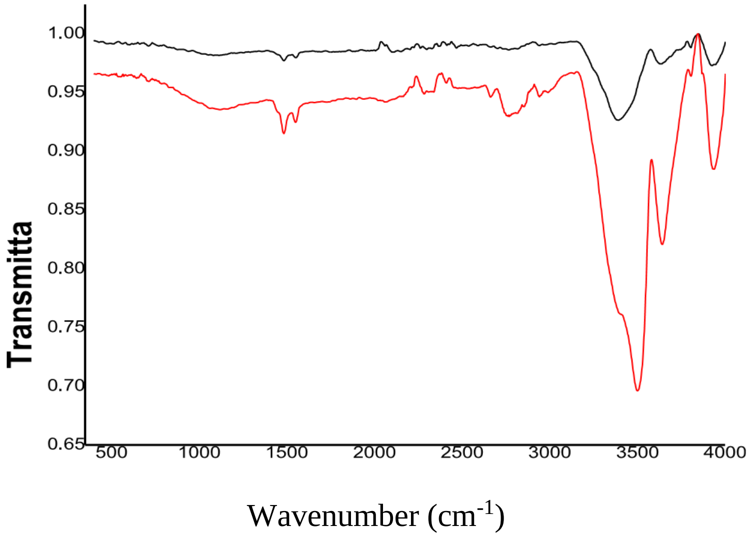

Fig. 7: FTIR spectra of the thin films

3.4 XRD analysis

The XRD analysis result is represented in figure 8. The analysis was conducted to study the phase purity and crystallinity of 3%Ti doped Cu_2O compared to undoped Cu_2O. The XRD spectra revealed that the doped thin film exhibited higher peak intensity and more peaks compared to the undoped film, indicating a more crystalline structure with better arrangement of nano molecules. The diffraction peaks of Cu_2O were observed at 2 = 30.11◦, corresponding to (111) planes. With the addition of Ti, sharp peaks were observed at 2 = 45◦ and 52.1◦, corresponding to (100) and (102) planes, confirming the presence of Ti. These results were consistent with the standard XRD database, confirming the crystallized hexagonal close-packed structure of Ti.

Table 1 provides information on the dislocation density, crystallite size, interplanar spacing, standard Bragg angle, and microstrain of deposited thin films. The table clearly demonstrates that reducing microstrain leads to a decrease in dislocation density, which aligns with previous research. Conversely, decreasing the average crystalline size increases both microstrain and dislocation density due to the presence of more grain boundaries and grains. Increasing grain boundaries further enhances dislocation density and microstrain. In this study, the incorporation of Ti in Cu_2O results in smaller crystallite size, leading to higher microstrain and dislocation density. The doped sample also exhibits a lower energy band gap due to its crystallized hexagonal closed-pack structure and smaller crystallite size, which is consistent with previous research [14].

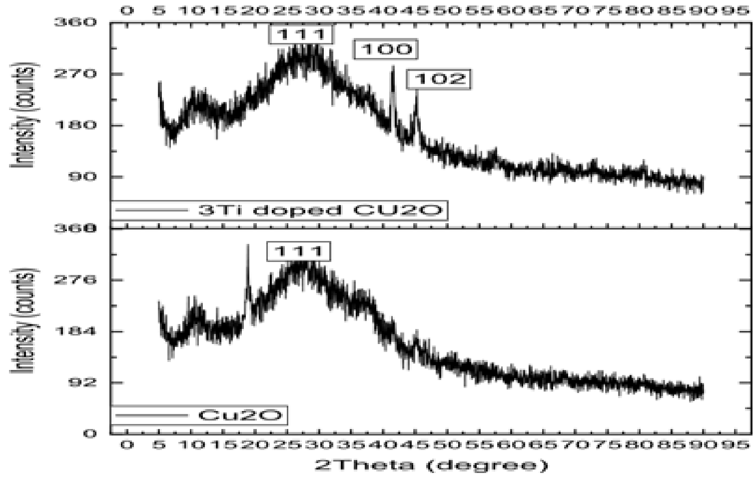

Fig. 8: XRD Spectrum of the sample

Table 1: The thin film structural parameters

Sample	(hkl)	Crystalline size (G) (nm)	Dislocation density (δ) (Lines/$m^{2)}$ x 10^{15}	Micro strain (ε) x 10^{-3}
100	5.11	38.29	4.153	
3Ti-Cu_2O	102	7.51	17.73	2.34
CuO	111	9.56	10.94	1.455

3.5 SEM analysis

Ti-doped Cu_2O thin film has more nanoparticles compared to undoped Cu_2O thin film, as shown in Fig. 9a and b. The presence of Ti influences the morphology of pure Cu_2O, leading to small differences in particle sizes. This can be attributed to disorder in the molecular structure and lattice strain, resulting in a uniform microstructure as observed in the XRD spectrum. The doped sample exhibits a dense, compact, and flake-like nanocluster particle structure, which is responsible for its lower bandgap. Similar findings have been reported in previous studies

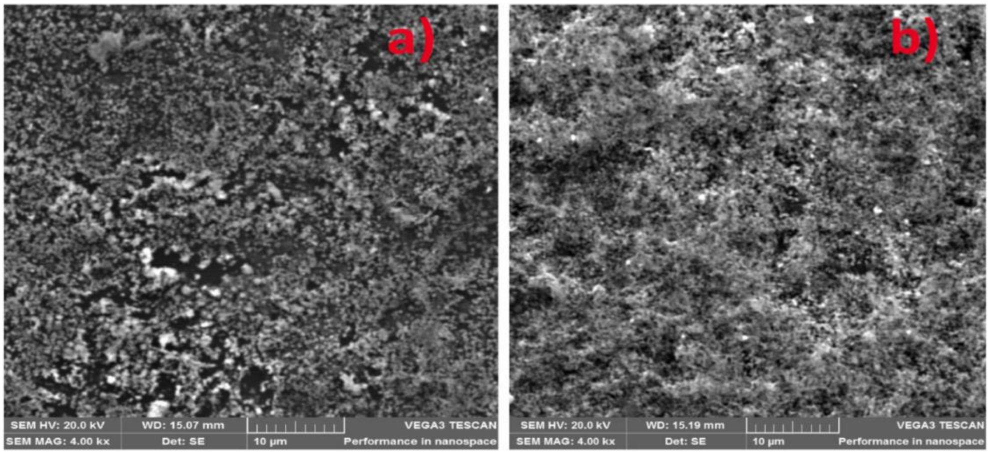

Fig. 9(a) 0% Titanium concentration in Cu_2O thin films and (b) 3% Titanium concentration in Cu_2O thin films.

3.6 Current-voltage features

Adding Ti to Cu_2O thin films improves their electrical properties without affecting their optical qualities, surface form, and crystal clarity. In the production of heterojunction solar cells, a 50 nm thick Cu_2O: Ti absorber layer was used. The PV cell properties were determined using the PV simulator model XES-151 S and the dark J-V performance was characterized using the Hewlett-Packard 4140 B semiconductor analyzer model. The dark current density of the solar cells was compared for n-Si/p-Cu_2O and 3% Ti/n-Si/p-Cu_2O samples and it was found that the Ti:Cu_2O sample had a higher potential but lower current density**.**

Doping 3% Ti into Cu_2O thin films increased the polarization values (Pr) from 7.50 C/cm^2 to 25.20 C/cm^2. This was because Ti substitution reduces oxygen vacancies and leakage currents, resulting in higher polarization switching values [20]. The doped samples also showed dense and compact grain boundaries, leading to good electrical properties [23]. The lattice distortion caused by Ti doping trapped ions at the interface and produced electron-hole pairs. Higher Pr values can be used to efficiently separate the hole and electron in photovoltaic devices.

The I-V characteristics of a single-sun n-Si/p-Cu_2O and 3% Ti/n-Si/pCu_2O PV device were studied under AM 1.5 G and sun illumination. The results showed three distinct regions labeled (i), (ii), and (iii) in the graph. Region (i) exhibited ohmic behavior with high electrical resistance due to few injected electrons. Regions (ii) and (iii) were dominated by conduction mechanisms related to space-charge and recombination of tunneling effects, respectively. The inclusion of Ti led to higher recombination rates and a larger voltage region in (ii) compared to the undoped sample.

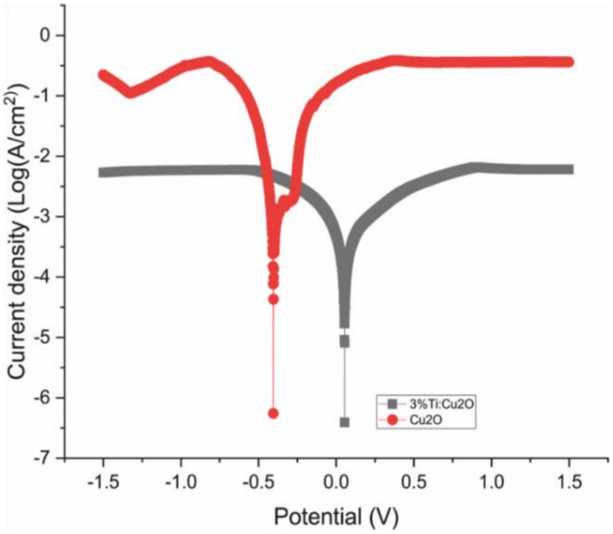

Fig. 10a: Dark current of heterojunction solar cells p-Cu_2O/n-Si and 3%Ti/p-Cu_2O/n-Si solar cells

The study found that the introduction of 3%Ti: Cu_2O doping improved the VOC and JSC values compared to pure Cu_2O samples. The power conversion efficiency also increased to 1.55% and 2.48% for the doped samples. The improved PV response was attributed to the higher polarization yield in the doped samples. On the other hand, the low values obtained in Cu_2O were due to excessive leakage caused by oxygen vacancies [22, 23]. The JSC and VOC values obtained were higher than those reported in previous studies [5, 9, 19, 24] on PV devices. The degradation of the p-CuO/n-Si interface properties caused by the formation of an amorphous isolation and Cu-rich interfacial layer between the Si substrate and copper oxide absorber layer was identified as a contributing factor to the increased leakage current and reduced Voc [5, 6, 21, 23].

The addition of Ti to the absorber layer of Cu_2O improves its electrical conductivity. This leads to better charge collection efficiency and lower resistance in the solar cells. As a result, the short circuit current density and open circuit voltage of the Cu_2O (Ti)-based solar cells are enhanced. This research confirms that the properties of these solar cells, produced with Ti/p-Cu_2O/n-Si, meet the recommended standards for photovoltaic solar cell production.

4. Conclusions

The study explored a new method of improving the efficiency of solar cells by adding titanium to copper oxide.

1. The nebulizer spray pyrolysis approach was used to create thin films of titanium-doped copper oxide, with up to 6% titanium.

2. They found that the addition of titanium shifted the near-band emission and bandgap of the copper oxide, resulting in improved electrical conductivity, charge collection efficiency, and reduced resistance.

3. The solar cells made with titanium-doped copper oxide showed enhanced short circuit current density and open circuit voltage.

4. Overall, the study concluded that solar cells with 3% titanium addition met the recommended standards for photovoltaic production.

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