



# Fabrication NiO: Cu / Si Heterojunction by the Aerosol-Assisted Chemical Vapor Deposition (AACVD)

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## Abstract

In this research, as the thin films were formed by an AACVD process, copper doped nickel oxide was used to prepare the Cu doped Ni thin films by ratio doping (Cu/Ni = 0, 7.5, 10 and 12.8 at w.t %). Thin films of Cu doped NiO were heated at a crystallization temperature of 400 °C for 2 hours. The thin films obtained by the AACVD method have a film thickness of the order (45-62 nm). Promising solar cells that could be created by NiO film as the absorber using Cu doping. The NiO:Cu film has promising optical characteristics; about (3.5-2.8 eV) energy gap band and a high absorption coefficient, which means that the most suitable absorber can be commercially developed using the NiO:Cu film. Furthermore, there are no rare metals in the NiO: Cu film. The best conversion efficiency with the heterojunction of NiO: Cu/Si and NiO:Cu was 2.8571429% which showed the possibility of a very low cost solar cell.

**Key Words:** Thin Film, Aerosol-Assisted Chemical Vapor Deposition, Hall Effect, I-V Characteristics, Heterojunction.

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## Introduction

NiO film is a promising material for transparent conducting electrodes to be applied to p-type. Various kinds of techniques, including spray pyrolysis, plasma enhanced chemical vapor deposition, and sputtering, etc., Due to its spontaneously formed nickel vacancies in the lattice, nickel oxide (NiO) is one of the most investigated p-type transition metal oxide semiconductors. [Thimsen; et. Al 2012]. Nickel oxide (NiO) is one of the important transition metal oxides that, due to a large number of spins and surface atoms, has fascinating optical and electronic properties with novel morphologies. [Wang; et. al 2019; Abzieher et. al.2019]. Due to its excellent chemical stability, electrical, optical and magnetic properties, the NiO film is a promising substrate to be applied photoelectrolysis [Koffyberg et. al 1981] and Electrochromic device [Lampert 1984] and positive electrode batteries [Vincent 1987]. The resistivity of NiO films can be

reduced by the application of O<sub>2</sub> gas when depositing NiO films. [Nandy et. al 2007]. It can also increase the electrical conductivity of the films, such as Li-doped NiO films [Jang et. al. 2010], by adding monovalent atoms to the NiO film. Increased p-type conductivity was registered, on the other hand, by doping Al atoms into NiO film [Nandy et. al 2009].

## Experimental Details

Aerosol-assisted chemical vapor deposition (AACVD) is a technique that includes the atomization into fine droplets of a precursor solution that are dragged through a mobile nozzle by a hot air flow to a heated reaction zone where chemical reactions occur. This work provides a description of the mechanical, electrical and electronic components for AACVD implementation. [Hincapié-Zapata et. al 2019].

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Due to the low instrumentation costs needed for its implementation and the numerous applications in the field of solid state research, the aerosol-assisted chemical vapor deposition (AACVD) technique is becoming increasingly popular [Choy 2003]. Via installing their own equipment to make depositions with different types of coatings, these two features allow companies and universities to advance in research and application processes. For example, Monárrez et al., using this technique, synthesized magnetite nano-particles in a carrier gas reactor [Monárrez-Cordero et. al 2018, and Monárrez-Cordero et. al 2014], But if the idea is to obtain thin films, the precursor solution, as described by a carrier gas and a nozzle, must be nebulized and transported to the substrate. P Lim et al. To deposit TiO<sub>2</sub> on tin oxide glass-coated surfaces [Lim et. al 2014]; or, as in the study by Jagdeep et al., which deposited nanostructured CoFe<sub>2</sub>O<sub>4</sub> thin films [Sagua et. al 2017]. The method used in this work follows experimental information close to those used by Pizá-Ruiz et al, where CuFeO<sub>2</sub> thin films were deposited on borosilicate glass coated with a thin film of TiO<sub>2</sub> and ZnO [Pizá-Ruiz et. al 2019]. The AACVD method consists essentially of a nebulizer that atomizes the precursor solution which, with the assist of hot air, rises to a given flow through a nozzle and is deposited on a film-generating substrate. The physical characteristics of this film (thickness, uniformity, structure, etc.) depend on the substrate temperature, the carrier gas and the nozzle movement speed.

The AA CVD method involves atomizing a precursor solution into fine aerosol droplets of sub-micrometer size that are delivered to a heated reaction zone and are undergone to evaporation, decomposition, and to form the desired products by homogeneous and/or heterogeneous chemical reactions. AACVD tackles the availability and distribution concerns of chemical precursors as a variation of traditional CVD processes. [Hou and Choy 2006].

In this study, NiO:Cu were Prepare by The Aerosol-Assisted Chemical vapor deposition (AACVD) method using nickel chloride hex water NiCl<sub>2</sub>6H<sub>2</sub>O (molecular weight of 237.7048 g/mole), Prepare the solution with 0.35M molarity by dissolving 33.27g of nickel chloride salt in 400 ml of water. Slowly stir the solution for an hour in the magnetic stir device to ensure that it is completely dissolved. Prepare the solution with 0.1M molarity by dissolving 1.34g of copper chloride salt (CuCl<sub>2</sub>) in 100 ml of water. Slowly stir the solution for an hour

in the magnetic stir device to ensure that it is completely dissolved. The amount of volume was controlled according to the weight ratio between Ni and Cu. In the Aerosol-Assisted Chemical vapor deposition (AACVD), Then Cu-doped NiO thin films were prepared by AACVD method Silicon wafer substrates for 1 h. The Silicon wafer substrates had been ultrasonically cleaned by ethanol, acetone, and distilled water for 30 min. An ultrasonic nebulizer was used to form an aerosol from the material to be deposited and to form thin films, and the aerosols were transferred from a device by a hose to the chamber.

This chamber contains a heater, as well as two openings, one for entering the material to be deposited and the other exit of unwanted products and easy passage of the fumigant.

The substrate is placed on the heater. At a temperature (200 °C ± 10), and at a distance (12 cm ± 2), the sample was deposited with the doping ratio shown in the table (1). After that, thin films were annealed for two hours at 400 °C, to obtain crystalline films and got rid of the defects that may have occurred in the deposition process.

**Table 1.** Show the volumes taken from proportions necessary to prepare

Sample	0.35M NiCl <sub>2</sub> ·6H <sub>2</sub> O (ml)	0.1M CuCl <sub>2</sub> (ml)	Deposition time (hour)	Cu/Ni WT%
NiO	100	0	1	0
NiO:Cu	80	20	1	7.5
NiO:Cu	75	25	1	10
NiO:Cu	70	30	1	12.8

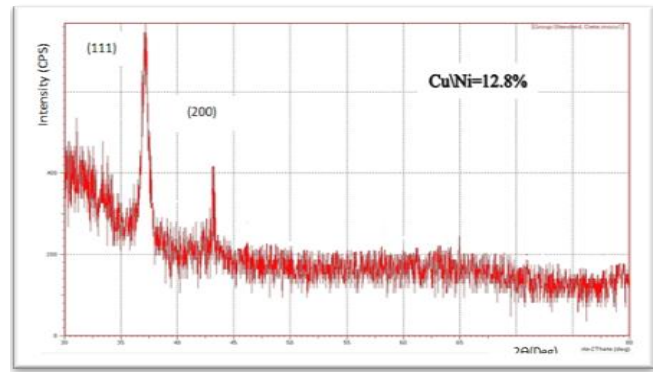
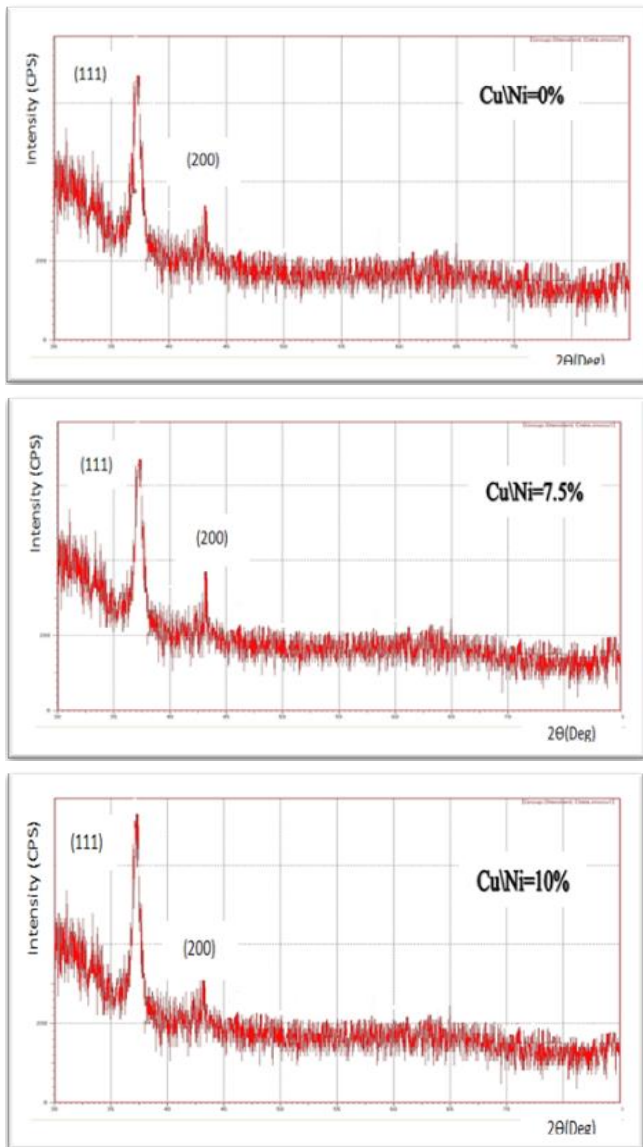
## Results and discussion

### Structural Properties (XRD)

The synthesis of the prepared films (NiO: Cu) was identified by AACVD and different doping of copper. It was found that the film has a polycrystalline and cubic polycrystalline structure of crystalline planes (111) and (200) which are corresponding angles (2θ = 37.2 °) and (2θ = 43.2 °) respectively. When the doping is increased, noticed a rise in the peaks and became sharp for the crystal planes and the appearance of the peak at the angle (2θ = 37.2°) which returns to the crystal plane (111) the results obtained from X-ray diffraction were confirmed with ICDD (International Center for Diffraction Data) card number (00-152-2025) observed that the preferred directions (111) when doping change from (0-12.8 %) and other preparation conditions are constant. When increasing the doping, we



observed an increase in the peaks relative to the crystalline planes with a decrease in the value of FWHM this indicates an increase in crystallite size. The increase in doping results in a small increase in the value of the lattice constants this is due to an increase in the value of the distance between crystalline levels ( $d_{hkl}$ ) with the increase of doping. Because the relationship between them is inverse and that means an improvement in the crystal structure, i.e. in the grain size as shown in Figure 1.

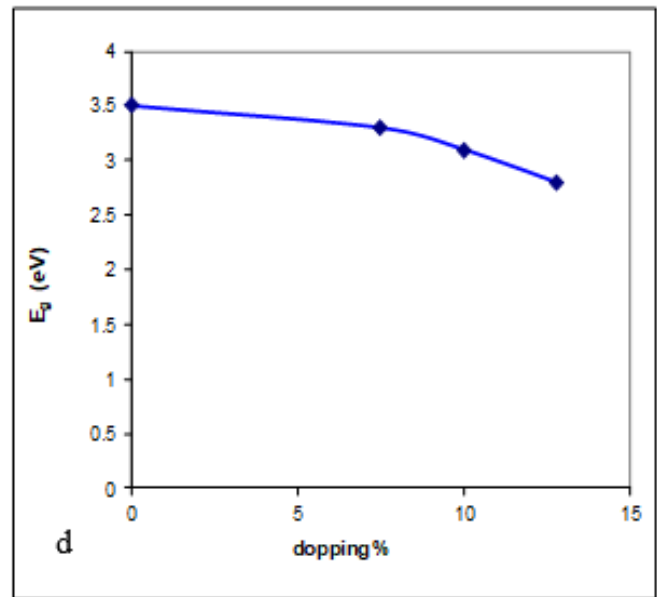
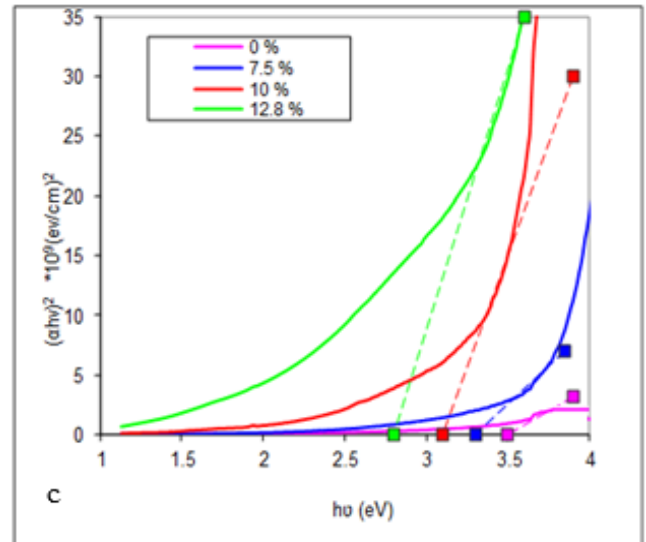
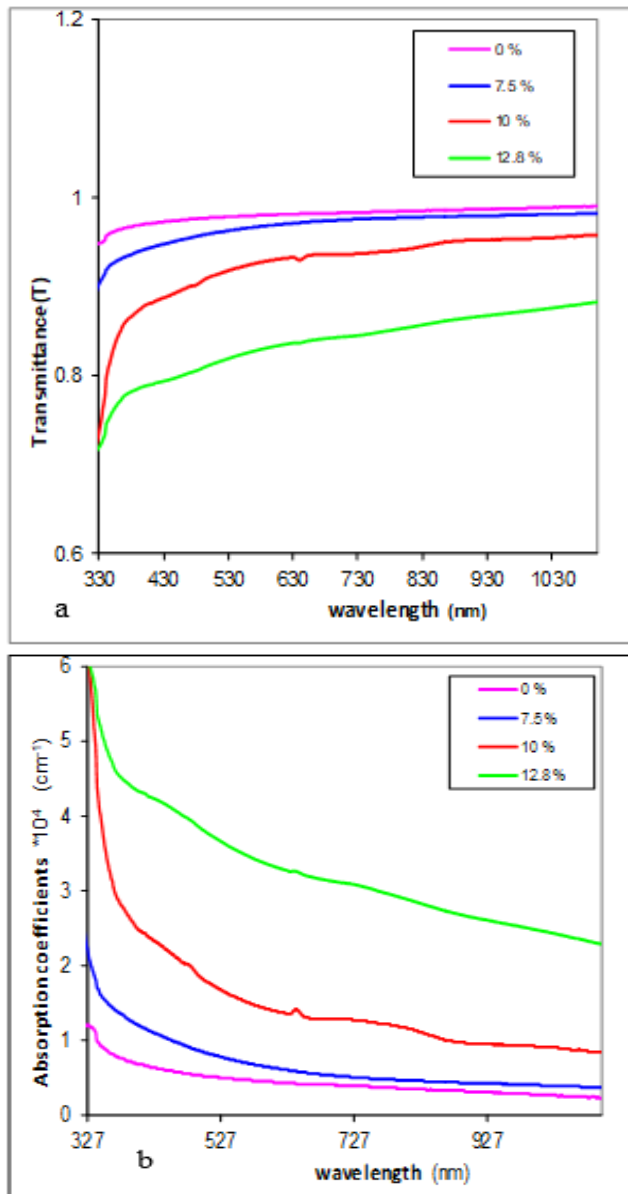


**Figure 1.** X-ray diffraction of fabricated thin films of Cu doped NiO deposited at several Cu concentrations.

### Optical Properties

The effect of Cu-doping on some NiO optical properties: thin film NiO. With the Cu doping in the prepared films, the transmission is decreased, while the absorption coefficient and skin depth are increased with the Cu-doping. Figure 2(a) explain the relationship between transmittance and wavelength for NiO:Cu films prepared with different doping from copper. Shown transmittance of prepared films decreases with increasing doping and increased thickness of films. The variation of optical transmittance of NiO:Cu films prepared with different doping from copper. As seen from these spectra's the transmittance of Cu doped NiO thin films were decreased with increasing Cu concentration in the visible region, the maximum transmittance of NiO thin films were obtained for NiO pure. The transmission of Cu doped NiO thin films at higher concentrations in the visible region was decreased due to the increase in absorption. Figure 2(b) shows the optical absorption coefficient as a function of the incident wavelength of NiO:Cu films prepared with different doping agents and it has been observed that all the thin films prepared have an absorption coefficient that increases with increasing doping and decreases with increasing wavelength and this is consistent with [Ijeh et. al 2019]. The figure shows that the absorption coefficient of the membranes gradually increases with doping. This is because the absorption value parameter indicates the ability of the film material to absorb the energy of the incident radiation and the increase in doping increases the thickness of the prepared films. From Figure 2 (c), the optical energy gap of NiO films deposited on glass substrates and different doping from copper, The figure shows the since intersection point represent the optical energy gap of the allowed direct electronic transitions and was found to decrease as

doping copper increased, This means that the optical energy gap values can be controlled by changing the doping when prepared NiO films. Observed that when calculating the energy gap, found that its value ranges between (2.8 –3.5 eV) this corresponds to what the researcher mentioned [19]. The energy gap decreases with increasing the doping copper when preparing the films. The Figure 2 (d) indicates that the band gap energy, decreased with the increase in Cu concentration to the maximum value of 2.8 eV- 12.8 in. % Cu, this decrease can be explained by the interstitial site company between Ni and Cu, which was seen for the vacancy in oxygen. [Benramache et. al 2020]



**Figure 2(a).** Transmittance spectrum as a function of wavelength for at different of Cu doped NiO thin films (b): Absorption coefficients as a function of wavelength for different of Cu doped NiO thin films (c): The optical energy gap in the allowable direct transition for at different of Cu doped NiO thin films. (d): The variation of band gap energy of NiO doped thin films at several Cu concentrations

### Electrical Properties

It is necessary to know the electrical properties of the Hall coefficient (R<sub>H</sub>). Depending on the value of the incident magnetic field (R<sub>H</sub>), the film is vertically placed in front of that field. Hall effect measurements of NiO films were analyzed for different doping copper in order to determine the form of charge carriers, their concentration, Where it was found that the carrier form of the NiO films is a positive type (p-type) of the positive, Hall coefficient (R<sub>H</sub>) value, the NiO:Cu (were when doping rate Ni/Cu = 12.8 %) films were converted from p-type to n-type from hall coefficient (RH)





value as the doping copper increases, the NiO:Cu (were when doping rate Ni/Cu = 12.8 5) value was converted from hall coefficient ( $R_H$ ) value, The explanation for this change is the increase in doping, which has resulted in an increase in the

film's grain size. It is noted from Table (2) by using a computer program that the values of conductivity, resistivity, concentration ( $n_H$ ) and hall coefficient ( $R_H$ ) for the type of carrier (n) are different, but the values of mobility increase as the doping increases.

**Table 2.** Hall parameters for NiO films at different doping for copper

Sample	Concentration of Carriers $n_H \text{ cm}^{-3}$	Conductivity $\sigma(\Omega.\text{cm})^{-1}$	Resistivity $\rho(\Omega.\text{cm})$	Mobility $\mu_H(\text{cm}^2/\text{V.s})$	Hall Effect $R_H(\text{cm}^{-3})$	Type of Carriers
NiO	$3.749 \times 10^{13}$	$1.642 \times 10^{-13}$	$6.09 \times 10^{12}$	$1.9 \times 10^{-5}$	$4.933 \times 10^4$	p-type
NiO:Cu Cu/Ni =7.5%	$5.287 \times 10^{12}$	$2.315 \times 10^{-11}$	$4.319 \times 10^{10}$	$1.26 \times 10^{-3}$	$1.523 \times 10^5$	p-type
NiO:Cu Cu/Ni =10%	$1.952 \times 10^{12}$	$3.138 \times 10^{-8}$	$3.187 \times 10^7$	0.1	$3.198 \times 10^6$	p-type
NiO:Cu Cu/Ni =12.8%	$-3.81 \times 10^{11}$	$3.863 \times 10^{-5}$	$2.589 \times 10^4$	$6.325 \times 10^2$	$-1.637 \times 10^7$	n-type

It is important to study current-voltage properties in the dark and the lighting state because it is a clear indication of the possibility of using the compound NiO:Cu as a light-sensitive material (solar cell). Where we observe the effect of light is evident in the change of current and voltage values for the samples prepared for the NiO compound at different doping for copper. Where Figures shown that values the current in the dark state are less than the lighting state in the forward and reverse biases of all films. As it is noted that the current values are disparit for the forward and reverse bias when changing the doping. It is noted from Figure (3) that the electrical current of the Si (n) sample flow at NiO pure in the light and dark state because of the low potential barrier formed at the junction (p-n) of the sample. The less current pass through the heterojunction in the dark state. Figure (4) show that the electrical current of the Si (n) sample decrease flow at NiO:Cu in the light and dark state because of increase the potential barrier formed at the junction (p-n) of the sample. The Figure (5) that the electrical current of the Si (n) sample increase flow at NiO:Cu in the light and dark state because of increase the grain size and decrease the potential barrier formed at the junction (p-n) of the sample. The increase doping turns NiO:Cu semiconductor from p-type to n-type is evident. when the (n) film is deposited on the Si (n) substrate, as in Figure (6) for type (n-n) the difference in the concentration of carriers, The current does not pass through the heterojunction in the dark state when increase the doping because the n- type carriers are not generated while the current resistance in the lighting state is higher in the forward bias than the reverse bias because changed the carriers

concentration of n- type of the Si (n) substrate and deposited n- type has on the substrate of the sample. It is noted from Figure (7) that the electrical current of the Si (p) sample more flow the NiO pure in the dark state because of the low potential barrier formed at the junction (p-p) of the samples. And The increase doping increase the current of the Si (p)/ NiO:Cu (p) flow at in the light and dark state because of the low potential barrier formed at the junction (p-p) of the sample. As in Figure (8). The Figure (9) shows decrease the current of the Si (p)/ NiO:Cu (p) flow at in the light and dark state because the increase of the grain size formed at the junction (p-p) of the sample. The Figure (10) show increase the current of the Si (p) / NiO:Cu (n) flow at in the light and dark state at the junction (p-n) of the sample. By comparison of the Si (n)/ NiO:Cu (n) at the junction (n-n) of the sample because the n- type carriers are not generated while the current resistance in the lighting state is higher in the forward bias than the reverse bias because changed the carriers concentration of n- type of the Si (n) substrate and deposited n- type has on the substrate of the sample. Whether it is the type (p-p) or the similar type (n-n) have the same values of density, but the second side of the heterojunction may have the same density as the carriers of silicon type (p) and silicon type (n) but they are different in density for carriers of the compound, which relatively generates a different potential barrier, due to the difference in density values then it is note that the current values are dispersed, which reduces its sensitivity to darkness. It is also noted from the previous figures that the current values are higher for the same voltage values in the lighting state due



to the excitation of the heterojunction (p-n) when photons incident, which generate more carriers because of the breaking of the bonds that occur as a result of the incident, which increases the current values. As for the efficiency of the heterojunction prepared, it is noted that it increases at the doping Si (n) / NiO:Cu (p) (a ratio doping Cu/Ni =10%) then it starts to decrease when the (n) film is deposited on the (n) substrate. Because the electron density increases when the doping increases and Table (3) indicates open circuit voltage (Voc) and short circuit current (Isc), maximum voltages, current, fill factor, and efficiency of samples from heterojunctions of the Si/NiO:Cu compound. Using a computer program, while it was noticed that the highest value of open circuit voltage is (78mV) for the Si (p) sample with a (p) type film deposited for the NiO pure sample. It was noted that the best efficiency is a value of (2.8571429%) when depositing the type (p) film of the NiO:Cu (at ratio doping Cu/Ni =10%) sample of the Si (n) substrate.

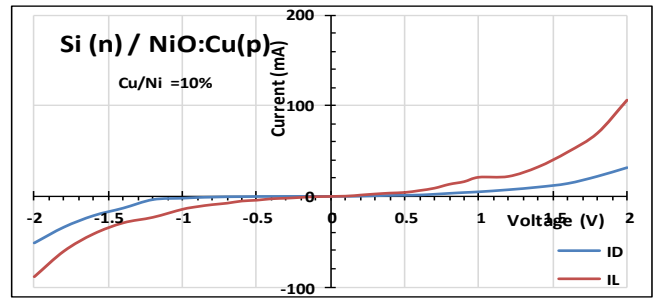


Figure 5. I-V Characteristics in Dark and Light for film Si (n) /NiO:Cu Cu/Ni =10% (P)

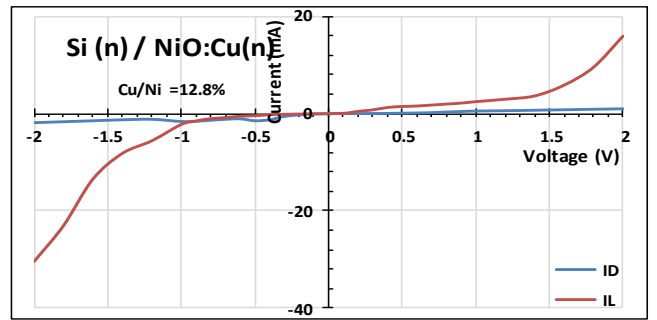


Figure 6. I-V Characteristics in Dark and Light for film Si (n) /NiO:Cu Cu/Ni =12.8% (P)

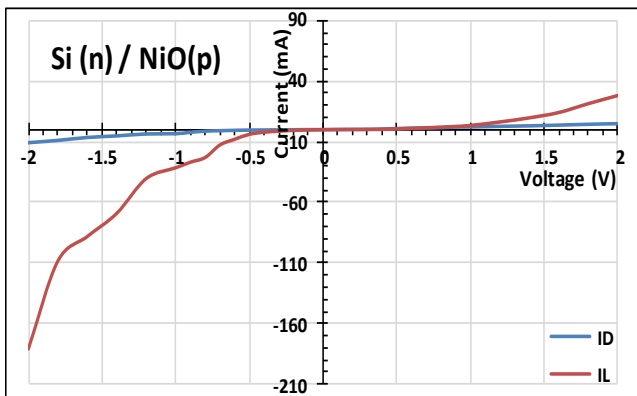


Figure 3. I-V Characteristics in Dark and Light for film Si (n) /NiO (P)

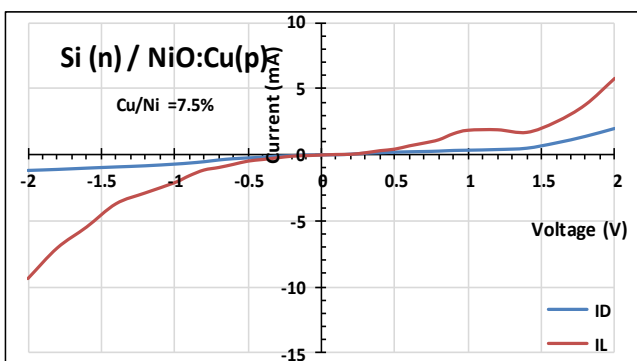


Figure 4. I-V Characteristics in Dark and Light for film Si (n) /NiO:Cu Cu/Ni =7.5% (P)

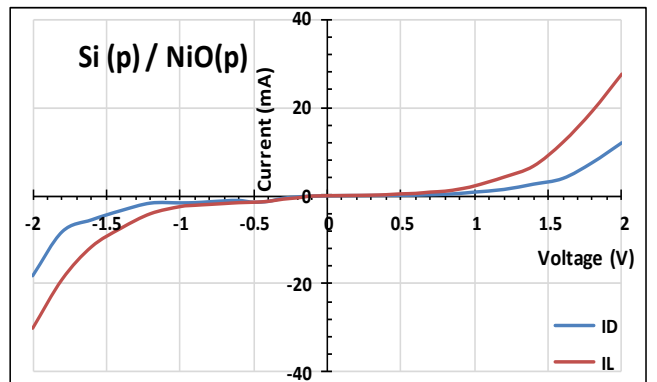


Figure 7. I-V Characteristics in Dark and Light for film Si (p) /NiO (P)

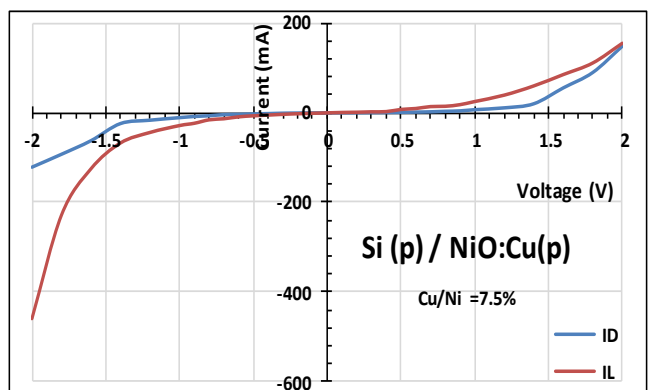


Figure 8. I-V Characteristics in Dark and Light for film Si (p) /NiO:Cu Cu/Ni =7.5% (P)



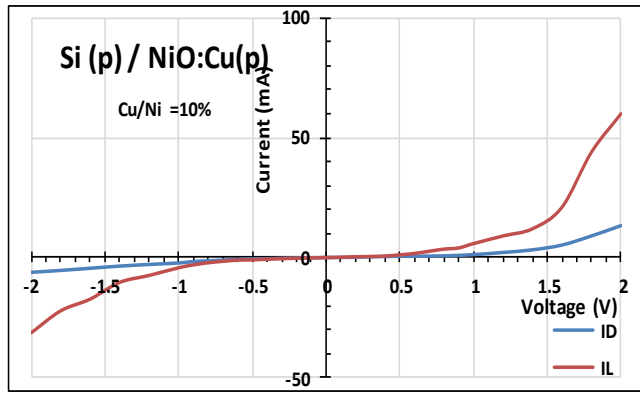


Figure 9. I-V Characteristics in Dark and Light for film Si (p) /NiO:Cu Cu/Ni =10% (P)

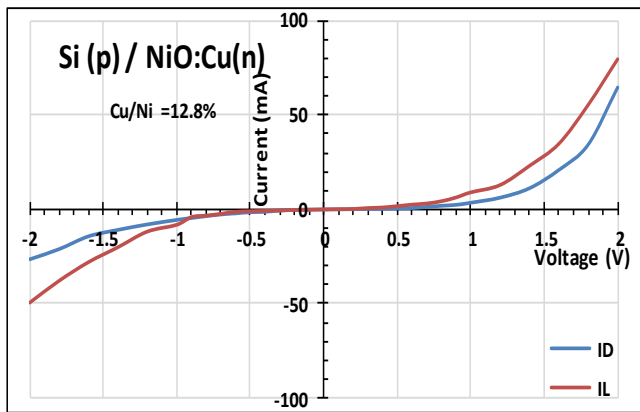


Figure 10. I-V Characteristics in Dark and Light for film Si (p) /NiO:Cu Cu/Ni =12.8% (n)

Table 3. Values Open Circuit Voltage, Closed Circuit Current and values Maximum Voltage and Current, Fill Factor, and Efficiency for samples of heterojunction of Si(p,n) / NiO at different doping copper

sample	Cu/ Ni (wt %)	V <sub>oc</sub> ( mV)	I <sub>sc</sub> ( mA)	V <sub>max</sub> ( mV)	I <sub>max</sub> ( mA)	F. F	η%
Si(n)/NiO (p)	0	42	0.082	18	0.05	0.26	1.530 6122
Si(n)/NiO :Cu(p)	7.5	35	0.015	20	0.009	0.34	0.306 1224
Si(n)/NiO :Cu(p)	10	50	0.13	28	0.06	0.26	2.857 1429
Si(n)/NiO :Cu(n)	12.8	21	0.09	10	0.005	0.03	0.093 6563
Si (p) / NiO(p)	0	78	0.022	28.00	0.015	0.26	0.766 0714
Si(p)/NiO :Cu(p)	7.5	12	0.12	6.5	0.06	0.26	0.663 2653
Si(p)/NiO :Cu(p)	10	35	0.058	20	0.03	0.30	1.020 4082
Si(p)/NiO :Cu(n)	12.8	32	0.05	15	0.03	0.28	0.765 3061

**Conclusion**

The following conclusion was reached:

1. Due to the increase in absorption, the decrease in the transmission of Cu doped NiO thin films at a higher concentration. At a

crystallization temperature of 400 °C for 2 h, the Cu doped NiO thin films were heated.

2. The obtained thin films have a cubic structure peak (111) and (200) by using AACVD method. As doping increases, the carrier type changes the NiO: Cu compound films from p-type to n-type.
3. The carrier density increases, conductivity increases with increasing doping and resistivity decreases, while mobility increases with increasing doping. With the disparity of current value in the dark state.
4. Decrease and an increase in the sensitivity of the samples in the forward and reverse bias lighting. It was noted that the best efficiency heterojunction is at a doping ratio of Cu/Ni= 10% with a value of (2.8571429) when depositing the type (p) film of the NiO: Cu sample of the Si (n) substrate.
5. As for the efficiency of the prepared heterojunction, it is noted that NiO:Cu (p) /Si (n) increases at the doping (Cu/Ni=10 %) then it starts to decrease when the (n) film is deposited on the (n) substrate, because when the doping increases, the electron density increases.

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