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ANNEALING TEMPERATURE ENHANCED THE CONVERSION EFFICIENCY OF (ITO/ZNO/CUO) HETEROJUNCTION

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Abstract

Copper oxide (CuO) thin film was deposited on glass substrates using pulsed laser deposition by fundamental wavelength (1064 nm) Q-switched Nd:YAG laser with 900 mJ peak energy at RT and annealed at 200 and 300 °C respectively. The Structural and optical properties of the prepared copper oxide thin films were studied using X-ray diffraction (XRD) and energy UV-visible spectroscopy. The copper oxide films exhibited Cu₂O phase of cubic structure converted gradually to CuO in a monoclinic crystal structure when annealed to 300 °C. Direct energy gap 2.25 eV value at RT sample, While the annealed samples have 1.8 and 1.6 eV indirect transitions. The I-V Characteristics of the ZnO/CuO heterojunction were studied under illumination revealed that the fabricate samples exhibit photovoltaic properties. The best efficiency appeared at 200 °C annealing temperature.

Keywords: Copper (II) Oxide, XRD, UV-Visible.

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Introduction

With the increase in the global need for energy in all fields, the urgent need for alternatives to renewable energy sources began instead of using fossil fuels harmful to the environment. The available and unused sun energy, as the energy supplied to the Earth in one hour is sufficient to meet the needs of the whole world in one year. Therefore, exploiting a small portion of the solar radiation is sufficient to meet the increasing request for energy [1].

One of the emerging strategies to address the current energy crisis in the world is photovoltaic cells, which specifically convert sunlight into electrical energy. Crystalline silicon solar cells have exceptional high conversion efficiency, but it is limited due to high cost [2].

Cupric oxide (CuO) and cuprous oxide (Cu₂O) are contenders for use in photovoltaic cells. They are P-type semiconductors that have an indirect power gap ~ 1.5 eV and direct ~ 2.0 eV, respectively. This value is almost perfect in solar cell fabrication due it allows good solar spectral absorption. In addition, it is a low-cost material and has low toxicity [3]. Zinc oxide (ZnO) is a wide band gap (3 –3.5 eV), high free carrier concentrations, and very similar electron affinity. These properties suggest for use as window in hetero junction [4]

Although copper oxide is one of oldest known semiconductor, no sufficient studies has been achieved to improve the efficiency of the tested solar cells manufactured from them, as the highest efficiency of solar CuO cells is less than 2% [5], which is still much lower than the theoretical efficiency limit of the Cu- O solar cell [6].

In this research, ZnO/CuO solar cell fabricated by simple pulsed laser deposition technique. The influence of annealing temperature on the synthesized films and the heterojunction properties was investigated..

Experimental Part

Copper (II) oxide (CuO) of purity (99.99 %) powder and zinc oxide (ZnO) of purity (99.99 %) powder by Merck Company were pressed under piston to make a capsules as targets. Thin films were prepared by Q switched Nd:YAG pulsed laser (DIAMOND-288 pattern) $\lambda = 1064$ nm, 9 ns pulse width, 10 Hz pulse repetition and 900 mJ peak power under vacuum 10-3 Torr vacuum. Thin films were deposited on glass substrates process were done by laser-target interaction. The laser beam enter the chamber through a window and making 45° angle with the target surface. The glass slide as substrate is located 2 cm above the target surface. Thin film thickness was measured using a reflectivity spectrometer model (TFProbe TM / Angstrom Sun Technology Inc.), which about 250 ± 10 nm for all samples.

Structural analysis for CuO powder and thin film at different annealing temperature were done by X-ray diffraction (using Shimadzu XRD 6000 system) and the optical properties are analyzed by UV-visible spectroscopy.

The schematic for fabricated solar cell was shown in Fig. 1. A layer of Cu-O thin film was deposited on ZnO thin film, and annealed at 200 and 300 °C. Aluminum electrodes where used on back of solar cell.

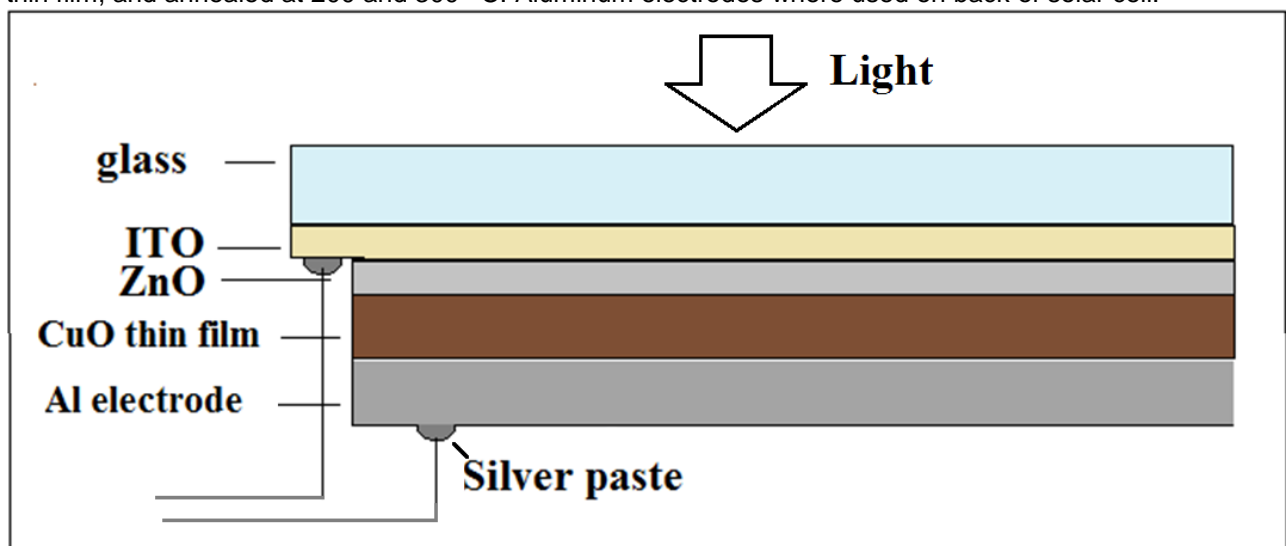


Fig. 1: Schematic of solar cell layers

The I-V characteristics for the fabricated solar cells were measured under illumination by Halogen lamp with 100 mW/cm² light intensity using Keithley Digital multi-meter and voltmeter for the forward bias to study the effect of annealing temperature on their characteristics.

Results and Discussion

Fig. (2) shows the XRD for CuO pellet used in deposited thin films. The pure sample pattern have many peaks corresponding monoclinic structure compared with standard card No 96-101-1195.

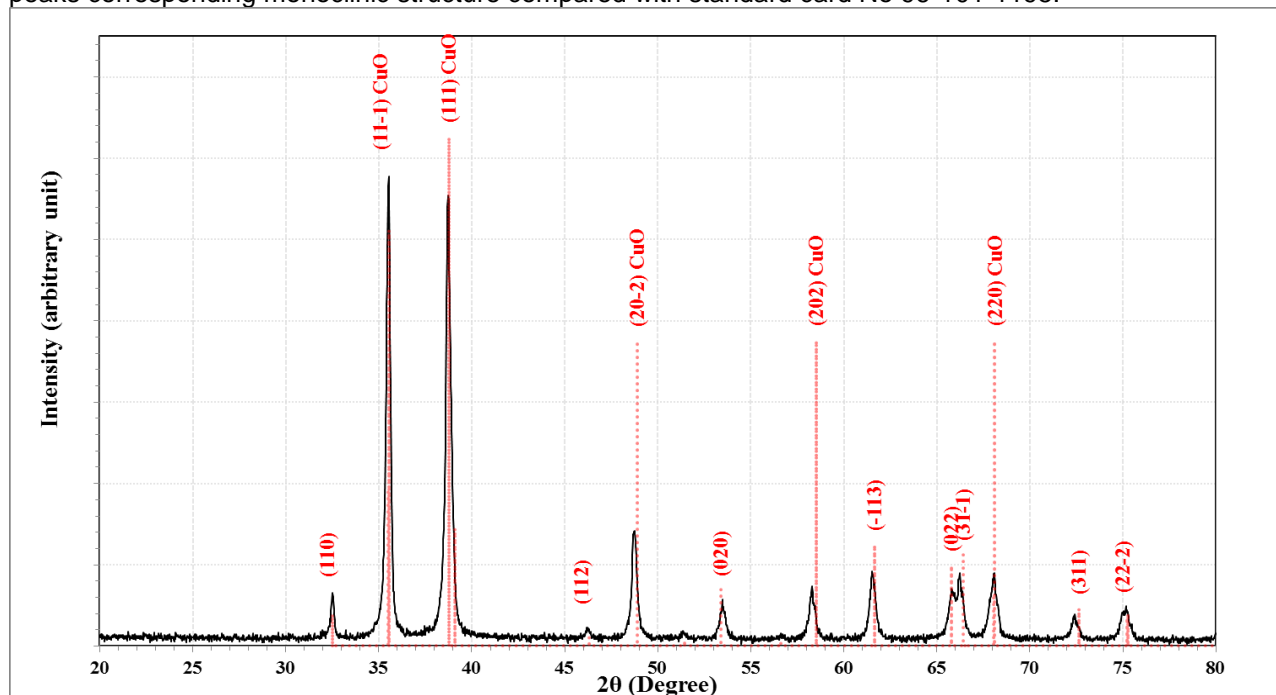


Fig.2: XRD patterns CuO powder

Fig. (3) showed the XRD for the as-deposited thin film under vacuum and annealed at 200 and 300 °C respectively, prepared on a glass substrate. The XRD pattern of as deposited thin film sample has low crystalline with peaks located at 36.4162 and 42.2915° assigned to (111) and (200) for Cu₂O cubic structure due to the lack of some oxygen atoms, in addition to peak at 46.3250° assigned to (-112) corresponding to monoclinic CuO structure.

The peaks intensities corresponding to CuO were increased with increasing annealing in exist of oxygen, while the Cu₂O peaks decreases when sample annealed at temperature of 200 °C and vanished at 300 °C annealing temperature. increasing temperature to 300 cause to increase sample crystallinity of pure CuO phase. This result agree with Prabu et al [7]

The full width at half maxima (FWHM) for assigned peaks decreases with increasing annealing temperature, indicate increasing the crystalline size as shown by Scherrer formula [8]. Table (1) shows all XRD peaks parameters compared with the standard inter-planer of lattice distance in card No. 96-900-7498 and 96-101-1195 for Cubic Cu₂O and Monoclinic CuO respectively

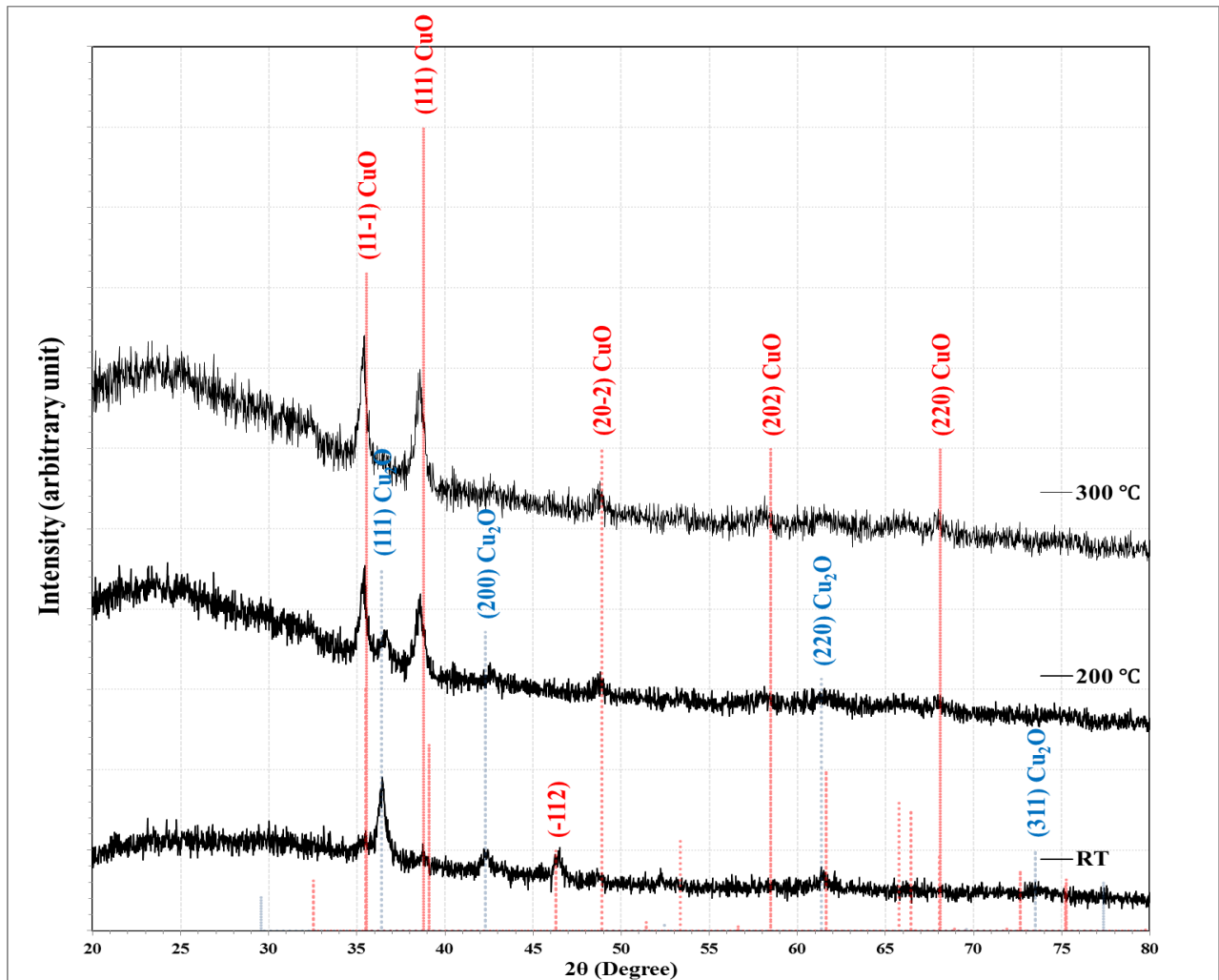


Fig. 3: XRD patterns as deposited and annealed CuO thin films at 200 and 300 °C temperature on glass substrates

Table 1: XRD peaks parameters for CuO thin films annealed at different temperatures..

Ta (°C)	2θ (Deg.)	FWHM (Deg.)	dhkl Exp.(Å)	C.S (nm)	dhkl Std.(Å)	hkl	Phase	card No.
RT	36.4162	0.4701	2.4652	17.8	2.4644	(111)	Cub.Cu2O	96-900-7498
	42.2915	0.8277	2.1353	10.3	2.1342	(200)	Cub.Cu2O	96-900-7498
	46.3250	0.4701	1.9584	18.4	1.9591	(-112)	Mono.CuO	96-101-1195
200	35.3621	0.3271	2.5362	25.5	2.5228	(11-1)	Mono.CuO	96-101-1195
	36.4082	0.4701	2.4657	17.8	2.4644	(111)	Cub.Cu2O	96-900-7498
	38.5821	0.4701	2.3316	17.9	2.3212	(111)	Mono.CuO	96-101-1195
	42.3530	0.6846	2.1324	12.5	2.1342	(200)	Cub.Cu2O	96-900-7498
300	35.3583	0.3271	2.5365	25.5	2.5228	(11-1)	Mono.CuO	96-101-1195
	38.5764	0.4701	2.3320	17.9	2.3212	(111)	Mono.CuO	96-101-1195
	48.6660	0.6131	1.8695	14.2	1.8617	(20-2)	Mono.CuO	96-101-1195
	58.1100	0.4701	1.5861	19.3	1.5764	(202)	Mono.CuO	96-101-1195
	67.9643	0.4701	1.3782	20.4	1.3755	(220)	Mono.CuO	96-101-1195

Fig. (4) displays the transmission curves for the prepared thin films. The transmission decreased, their behavior changed, and the band edge shifted toward higher values due to phase conversion from Cu₂O to CuO with annealing [9].

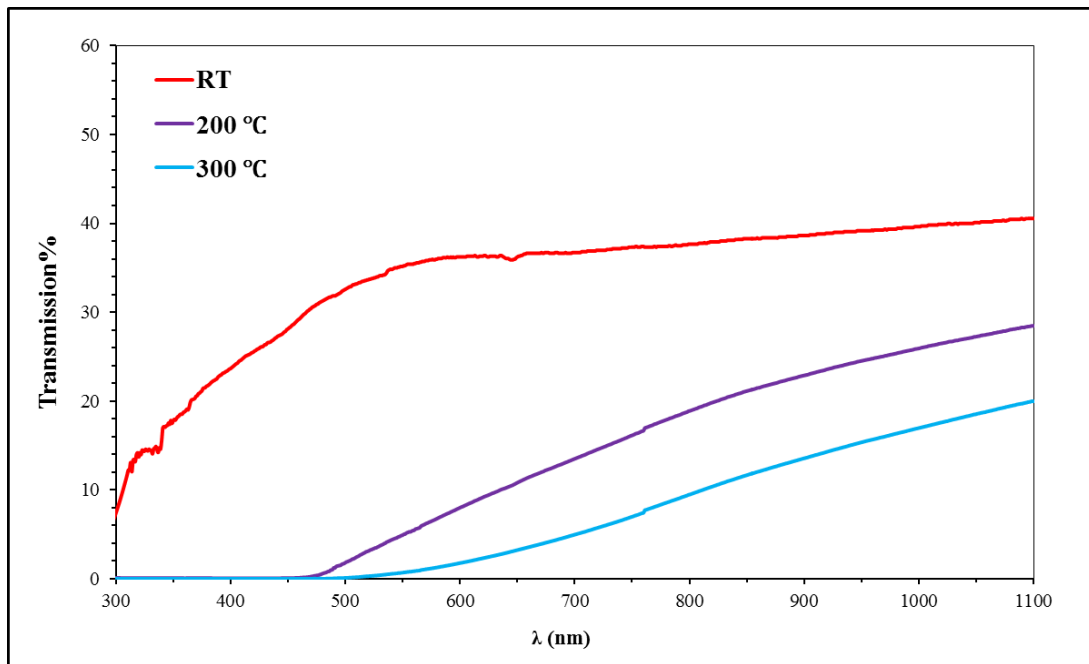


Fig. 4: Transmission curves for CuO thin films annealed at different temperatures.

The direct optical band gap (E_{gopt}) for CuO thin films prepared at room temperature has been determined by Tauc plot $(\alpha h\nu)^2$ vs. $h\nu$ as shown in Fig. (5). The Cu₂O thin film sample has allowed direct electronic transition of 2.25 eV energy gap, which agree with previous results such as Kidowaki et al [3]. Figure (6) reveals that the band gap of annealed sample at 200 oC (1.8 eV) greater than the annealed sample at 300 oC which has (1.6 eV) indirect transitions band gap corresponding to CuO structure, which agree with Shaban el al [10].

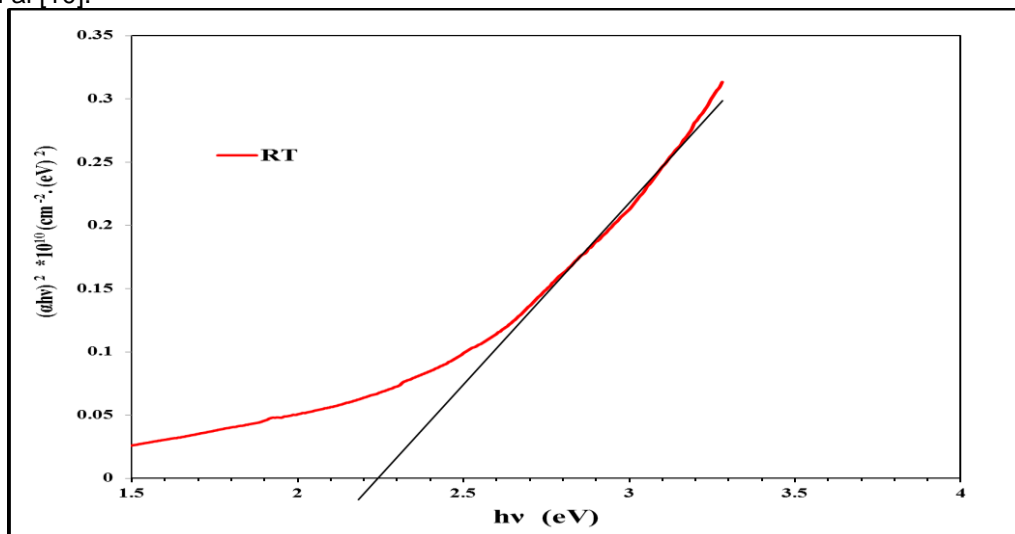


Fig. 5: Variation of $(\alpha h\nu)^2$ vs. $h\nu$ for as deposited thin film at RT

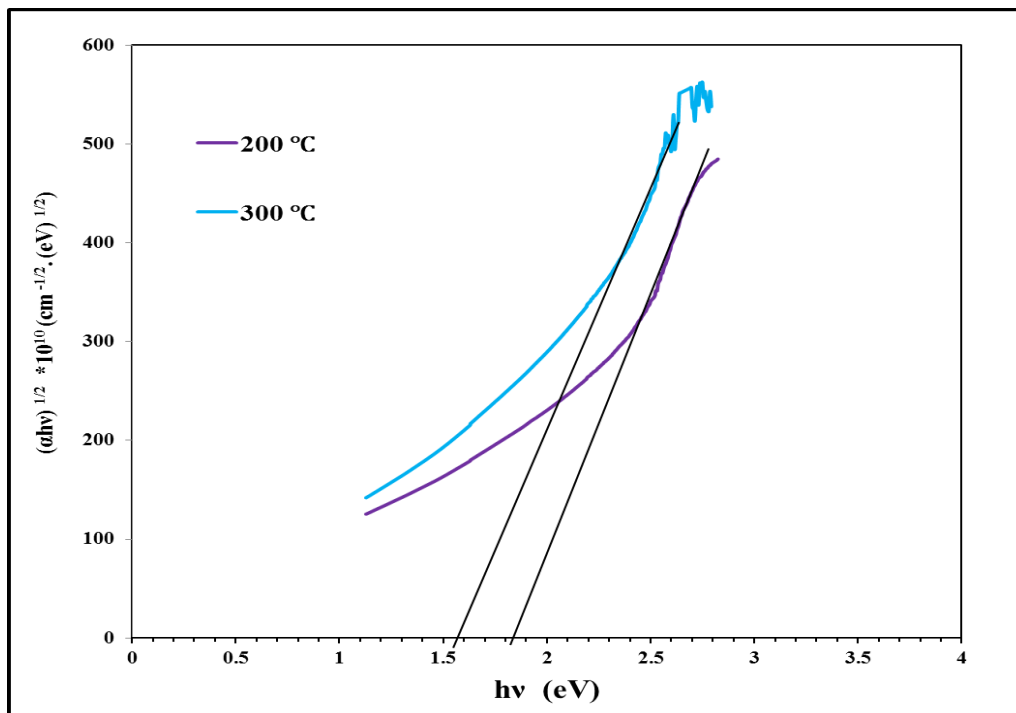


Fig. 6: Variation of $(\alpha h\nu)^{1/2}$ vs. $h\nu$ for annealed thin films at 200 and 300 °C

The I-V characteristics of the fabricated solar cells prepared from films annealed at different temperatures (RT, 200 and 300 °C) under 100 mW/cm² light intensity at the range of 0 to 0.6 Volt applied voltage are illustrated in Fig. (7). It is appeared that the maximum power area increased at 200 °C annealing temperature and decrease at 300 °C temperature. The solar cells parameters were shown in Table (4) the best annealing temperature is the 200 °C of higher short current, open circuit voltage, filling temperature and efficiency.

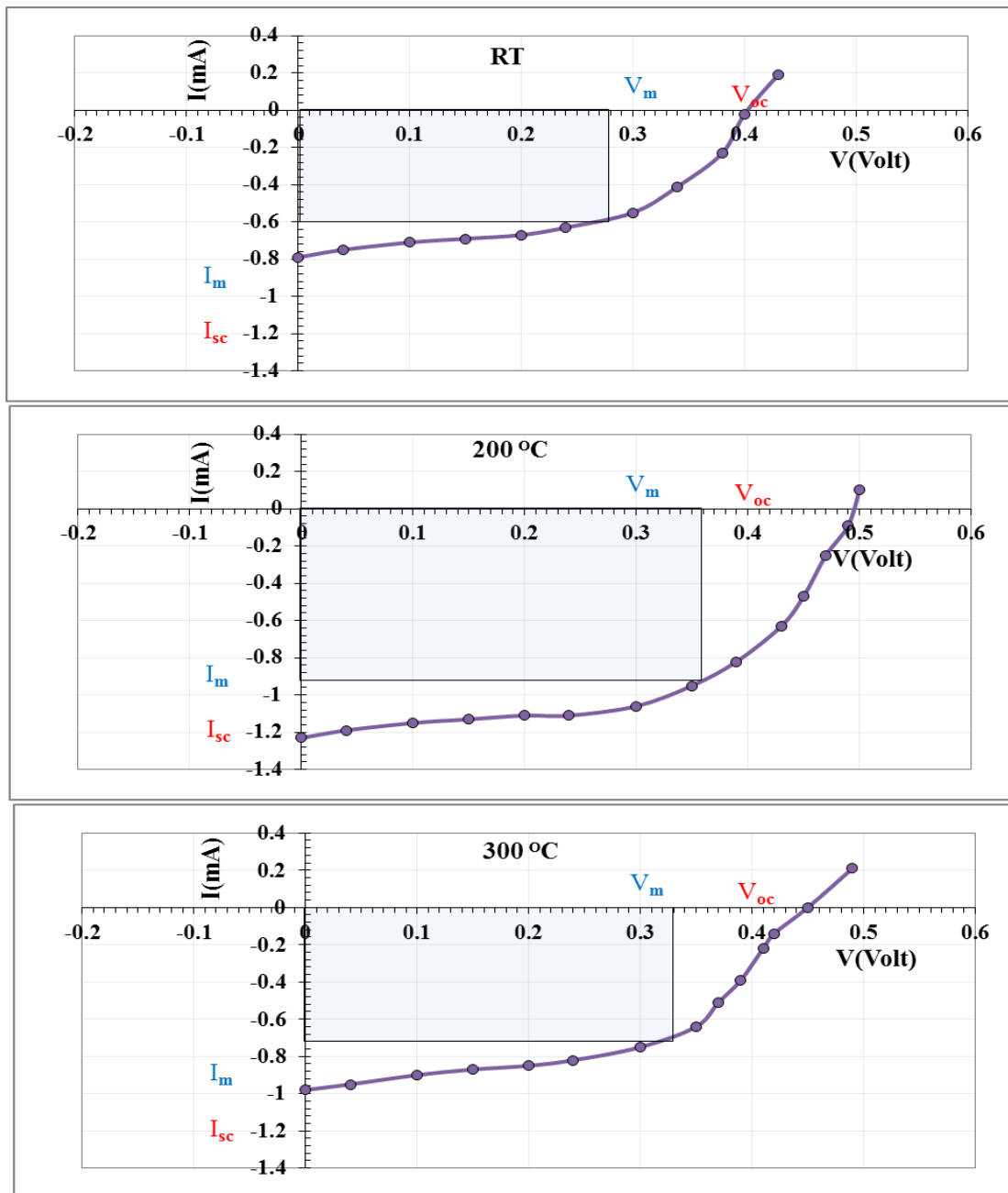


Fig. 7: I-V characteristics for fabricated solar cells annealed at different temperatures

Table 3: I-V Parameters for fabricated solar cells (ZnO/CuO/ITO) Heterojunction annealed at different temperatures

T (°C)	I _{sc} (mA)	V _{oc} (V)	I _m (mA)	V _m (V)	F.F	η%
RT	0.80	0.40	0.60	0.28	0.53	0.56
200	1.22	0.50	0.92	0.36	0.54	1.10
300	1.00	0.46	0.72	0.33	0.52	0.79

Conclusions

The structural properties results for copper oxide deposited by pulse laser deposition on glass substrates show that annealing convert cubic Cu₂O phase prepared at room temperature to monoclinic CuO phase at 300 °C. The crystallinity and the crystallite size enhanced with increasing annealing temperature.

The optical properties indicate the phase transition, where annealing causes to varying the energy gap from 2.2 eV of direct transition to 1.6 eV of indirect transition.

The results for fabricated solar cells based on copper oxide shows that the best annealing temperate is 200 °C, which better than the RT and 300 °C annealing temperature samples.

Although the efficiency obtained is small compared to that achieved in other materials, it can be used as a low-cost photovoltaic sensor.

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