Characterization of ZnO/GaAs heterojunction

Eman M. Nasir and Asmaa N. Mohammed Ali

Department of Physics, College of Sciences, University of Baghdad

E-mail: eman.itabi@gmail.com

Abstract
Thin films of ZnO are prepared by pulse laser deposition. These films were deposited on GaAs substrate to form heterojunction ZnO/GaAs solar cell. The effect of annealing temperatures at (RT, 373, 473) K on properties of structural and optical of ZnO thin films has been studied. The Diffraction analysis by X-ray indicated that all films have hexagonal polycrystalline structure. AFM shows that the grains uniformly distributed with homogeneous structure. The optical absorption spectra showed that all films have direct energy gap. The band gap energy of these films decreased with increasing annealing temperatures. From the electrical properties, the carriers have n-type conductivity. From C-V measurement of ZnO/GaAs heterojunction at frequency 100, 200 kHz, it is found that built-in potential (Vbi) increases with increased frequency. As well as, from I-V characteristic it is observed that the ideality factor is 2.7. Short-circuit current (Isc) is 4.0 mA/cm², open circuit voltage is 0.5 V, fill factor is 0.7 and the efficiency is about 6.0 %.

Key words
Thin films, optical properties, electrical properties, semiconducting, solar cell.

Introduction
Semiconductor thin films are important in industry due to their optical and electrical properties that used in many optoelectronic applications. zinc oxide have wide band gap (~3.3 eV) II-VI because showed several characteristics suited for several apparatus like transparent electrodes which work with the solar devices and antireflection coatings [1], it used in solar cell fabrication and steps of thermal treatment which required for improvement the photovoltaic properties. First step Range of temperature (350-5000) °C,
then deposition of the window substances [2]. It is necessary to detect effect of the post-thermal annealing on properties of the window thin films, wherever, it use as heterojunction materials in solar cells. Caban et al. have fabricated ZnO/GaAs heterojunction solar cells by the ALD method and found that ZnO films doped with Al effect on the photovoltaic properties of solar cell [3]. Makableh et al. have used ZnO as anti-reflection coating in GaAs solar cell; they observed a great enhancements in the properties of solar cell [4]. Eman et al. have studied the morphology and electrical properties of ZnO-CuO and found that roughness and conductivity increased with x content and decreased with annealing temperatures [5].

At present study, we studied effect temperature factor in annealing stage on material structural, electrical and optical properties of ZnO and fabricated of ZnO/GaAs heterojunction thin film solar.

**Experimental**

Pure zinc oxide (ZnO) were used as start materials to prepare films by pulsed laser deposition (PLD) technique on glass, GaAs substrates under vacuum of $3 \times 10^{-3}$ mbar with 500nm of thickness, at different annealing temperatures using laser Nd:YAG at $\lambda=1064$ Nanometer, the pulse duration was 15 ns and the frequency was 6 Hz and at energy 800 mJ. The material was pressed into pellets (2 cm) in diameter and (0.25 cm) thickness, using hydraulic type (SPECAC), under pressure of 15 tons. finally the pellets were sintered in air at (500 K) for 2 h. 500 laser pulse is incident on the target surface by angle of 45°. 10 cm is the distance between the target and the laser, and 1.5 cm between the target and the substrate.

The composition structure of the thin films was determined by using (Philips X-ray diffractometer). Atomic force microscopy by AA3000 Angstrom advanced lnmtd is used for studying morphology of surface of the prepared films, while the optical charterers is studied by spectrophotometer (Optima-3000). The electrical measurements include D.C conductivity; Hall Effect for ZnO films with different annealing temperatures has been studied.

The ability of the heterojunction was calculated by the reverse bias voltage at (0–1.2) V with two frequencies of (100 and 200) kHz by unit model (HP–R2C) 4275A and 4274A LRC meter. The current-voltage measurements in the dark were done using Keithly digital electrometer type 616 and D.C power supply and used light of halogen lamp at 25 mW/cm² (Philips Co).

**Results and discussion**

Determination of the structure of the ZnO films was done by using XRD to ensure the stoichiometry of our material. The Fig.1 shows XRD of thin films (ZnO) on glass substrates in many temperatures (as deposited, 373, 473) K for 1h, It can be noticed that they are three peaks for polycrystalline hexagonal ZnO phase located at 31.75°, 34.4° and 36.15° with hkl (100), (002) and (110), respectively that is have damae with (JCPDS 96-900-4180) data as in Table 1. The crystallite size (grain diameter), D, of the ZnO films have been calculated by using the Scherrer's formula,

$$D = \frac{0.9 \lambda}{\beta \cos \theta} \quad (1)$$

where:

$\lambda$ is wavelength of X-ray, $\theta$ is half the angle between $\beta$ and the scattered and incidents X-ray and $\beta$ represent full width at half maximum. The values of grain size (G.S) decrease with increasing annealing temperatures and then decreased as given in Table1.
Similar result has been emphasized by Eman et al. [5] and Gupta et al. [6]. The increases in intensity and sharp peak in X-ray types showed the perfect crystallinity.

![XRD patterns of ZnO thin films at different annealing temperatures (as deposited, 373, 473) K.](image)

**Table 1: XRD parameters of ZnO thin films at different annealing temperatures (as deposited, 373, 473) K.**

<table>
<thead>
<tr>
<th>Ta (K)</th>
<th>2θ (Deg.)</th>
<th>FWHM (Deg.)</th>
<th>d&lt;sub&gt;hkl&lt;/sub&gt; Exp.(Å)</th>
<th>G.S (nm)</th>
<th>d&lt;sub&gt;hkl&lt;/sub&gt; Std.(Å)</th>
<th>hkl</th>
</tr>
</thead>
<tbody>
<tr>
<td>RT</td>
<td>31.75</td>
<td>0.2130</td>
<td>2.8160</td>
<td>38.8</td>
<td>2.8174</td>
<td>(100)</td>
</tr>
<tr>
<td></td>
<td>34.4</td>
<td>0.2340</td>
<td>2.6049</td>
<td>35.5</td>
<td>2.6037</td>
<td>(002)</td>
</tr>
<tr>
<td></td>
<td>36.15</td>
<td>0.3256</td>
<td>2.4827</td>
<td>25.7</td>
<td>2.4780</td>
<td>(101)</td>
</tr>
<tr>
<td>373</td>
<td>31.8</td>
<td>0.4103</td>
<td>2.8112</td>
<td>20.13</td>
<td>2.8174</td>
<td>(100)</td>
</tr>
<tr>
<td></td>
<td>34.5</td>
<td>0.5275</td>
<td>2.5976</td>
<td>15.76</td>
<td>2.6037</td>
<td>(002)</td>
</tr>
<tr>
<td></td>
<td>36.25</td>
<td>0.4689</td>
<td>2.4761</td>
<td>17.82</td>
<td>2.4780</td>
<td>(101)</td>
</tr>
<tr>
<td></td>
<td>56.6</td>
<td>0.7034</td>
<td>1.6248</td>
<td>12.82</td>
<td>1.62661</td>
<td>(110)</td>
</tr>
<tr>
<td>473</td>
<td>31.65</td>
<td>0.4778</td>
<td>2.8196</td>
<td>17.28</td>
<td>2.8174</td>
<td>(100)</td>
</tr>
<tr>
<td></td>
<td>34.5</td>
<td>0.2986</td>
<td>2.6095</td>
<td>27.83</td>
<td>2.6037</td>
<td>(002)</td>
</tr>
<tr>
<td></td>
<td>36.25</td>
<td>0.2986</td>
<td>2.4801</td>
<td>27.98</td>
<td>2.4780</td>
<td>(101)</td>
</tr>
<tr>
<td></td>
<td>56.65</td>
<td>0.4181</td>
<td>1.6274</td>
<td>21.56</td>
<td>1.62661</td>
<td>(110)</td>
</tr>
</tbody>
</table>

Parameters of AFM are included (Peak-Peak, average roughness and diameter of size) of ZnO films deposited on glass substrates at RT and different temperatures have been determined using AFM analysis. Fig.2 shows 3D-AFM images for ZnO thin films at RT and many temperatures. It is observed from Table 2, that the average grain size values become more with elevating temperature and then decreased at 473 K and the ZnO film at 373 K have maximum values of Peak-Peak and roughness value. The results were a good agreement with El Sayed [7].
Fig. 2: 3D-AFM images for ZnO thin films at RT and different annealing temperatures.

Table 2: Average grain size, average roughness and Peak- Peak for ZnO thin films with different annealing temperatures (as deposited, 373 and 473) K.

<table>
<thead>
<tr>
<th>Ta(K)</th>
<th>Ave. grain size (nm)</th>
<th>Ave. Roughness (nm)</th>
<th>Peak-Peak (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RT</td>
<td>350.16</td>
<td>0.478</td>
<td>5.16</td>
</tr>
<tr>
<td>373</td>
<td>644.4</td>
<td>2.84</td>
<td>28.7</td>
</tr>
<tr>
<td>473</td>
<td>230.12</td>
<td>1.46</td>
<td>17.5</td>
</tr>
</tbody>
</table>

Fig. 3 Transmittance spectrum of ZnO thin film on the glass at (200-1100) nm. It is observed from this figure that the values of transmittance decreases and shifting to higher wavelength with increasing annealing temperatures. The average optical transmittance is (75) %. As well as, results in to excellent optical quality that is agreed with the studies [8, 9].

Edge of the fundamental absorption in the semiconductors is depending on the exponential law [10].

\[ \alpha h\nu = A (h\nu-E_g)^r \]  \hspace{1cm} (2)

\( A \) is constant factor
\( E_g \) is energy of optical band gap
\( r \) is any value between (1/2) and (3)\( \alpha \) is absorption coefficient and \( h\nu \) is photon energy

The absorption data is measured depended on Eq.(2) [10].

\[ \alpha = \ln(1/T)/d \]  \hspace{1cm} (3)

A is calculated based on equation [11]:
d refer to film thickness. 

\((ahv)^2\) vs \(hv\) refer to plot of the graph. By the optical methods, the direct band gap value of the produced ZnO films has been provided from \((ahv)^2 = 0\). From Fig.4, \(E_g\) is decreased from 2.53-2.2 eV with increasing annealing temperatures from as depositing to 473K. The optical constant can be shown in Table 3 at wavelength 600 nm. It can be deduced that all optical constants become more with elevating temperatures [12].

![Fig.4: \((ahv)^2\) as a function of \(hv\) for ZnO thin films at different annealing temperatures (as deposited, 373, 473) K.](image)

### Table 3: Optical properties parameters of ZnO films at different annealing temperatures.

<table>
<thead>
<tr>
<th>(T_a) (K)</th>
<th>(E_g) (eV)</th>
<th>(10^4(a^*cm)^{-1})</th>
<th>(n)</th>
<th>(K)</th>
<th>(\varepsilon_i)</th>
<th>(\varepsilon_r)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RT</td>
<td>2.5</td>
<td>3.280</td>
<td>2.177</td>
<td>0.130</td>
<td>4.724</td>
<td>0.569</td>
</tr>
<tr>
<td>373</td>
<td>2.3</td>
<td>4.009</td>
<td>2.307</td>
<td>0.159</td>
<td>5.297</td>
<td>0.736</td>
</tr>
<tr>
<td>473</td>
<td>2.2</td>
<td>5.331</td>
<td>2.486</td>
<td>0.212</td>
<td>6.136</td>
<td>1.054</td>
</tr>
</tbody>
</table>

The change of electrical conductivity with temperature depending on the relation:

\[\sigma = \sigma_0 \exp(-E_a/ k_B T) \]  \(\text{(4)}\)

where \(E_a\): is the thermal activation energy, \(T\): is the absolute temperature, \(k_B\): is the Boltzmann constant \(\sigma_0\): is the minimum electrical conductivity at 0 K.

Fig.5 demonstrates the plots of \(\ln\sigma\) versus \(10^3/T\) for ZnO films deposited at R.T and different annealing temperatures (100, 200) C in the range (273-473) K. From d.c measurements the conductivity \((\sigma_{d.c})\) become low with increasing of temperature in annealing stage and then increased, because the rearrangement is activated during the high temperature of annealing stage.

The activation energy conduction mechanism \((E_{a2})\) is activated at temperatures (443-473) K, but the activation energy \((E_{a1})\) is activated in lower temperatures (303-433) K.

Influence of temperature on \(E_{a1}\) and \(E_{a2}\) for ZnO films is cleared as in Table 4. It is founded the energies activated increased with increasing of annealing temperatures.
The hall coefficient ($R_H$) is determined by measuring the Hall voltage that generates the Hall field across the sample of thickness ($t$), by:

$$R_H = \frac{V_H}{I} \frac{t}{B} \quad (5)$$

Carriers concentration can be determined by using the relation

$$n_H = \frac{-1}{qR_H} \quad \text{for electrons,}$$

$$p_H = \frac{1}{qR_H} \quad \text{for holes} \quad (6)$$

Hall’s mobility ($\mu_H$) can be written by

$$\mu_H = \frac{\sigma}{nq} \quad (7)$$

$$\mu_H = \sigma |R_H| \quad (8)$$

the mobility of electron and hole, respectively,

$$\mu = \frac{V_d}{E} \quad (9)$$

where $V_d$ is the drift velocity. So that we have

$$\sigma = (nq^2\tau)/m^* \quad (10)$$

where $\mu$: is the mobility, $\tau$: is the carrier’s lifetime, $n$: is the carrier’s concentration, $m^*$: is the effective mass of the carrier, and $q$: is the electron charge. Carriers concentration can be determined by using the relation

$$n_H = \frac{-1}{qR_H} \quad \text{for electrons,}$$

$$p_H = \frac{1}{qR_H} \quad \text{for holes} \quad (11)$$

and mean free bath can be represented as

$$L = \frac{V_d}{\tau}$$

Hall mobility ($\mu_H = \sigma |R_H|$), drift velocity ($V_d$), carriers concentration ($n_H$), lifetime ($\tau$) and mean free path ($\ell$) of charge carriers is calculated from Hall examinations. It can be noticed the whole films has negative Hall coefficient as shown in Table 5, the carrier’s concentration reduce with elevating temperatures, but the Hall mobility become more when temperatures is increased. Also, the parameters lifetime, drift velocity $V_d$ and mean free path $\ell$ of the carriers have been determined as in Table 5. It is observed that all parameters approximately increase with increasing of annealing temperatures [11, 12].

![Fig.5: ln $\sigma$ versus $10^3/T$ for ZnO films at different annealing temperatures.](image)

**Table 4: D. C. conductivity parameters for ZnO films at different annealing temperature.**

<table>
<thead>
<tr>
<th>$T_a$ (K)</th>
<th>$\sigma_{d.c.} \times 10^{-5}$ (Ω.cm)$^{-1}$</th>
<th>$E_{a1}$ (eV)</th>
<th>Temp. Range (K)</th>
<th>$E_{a2}$ (eV)</th>
<th>Temp. Range (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RT</td>
<td>2.61</td>
<td>0.056</td>
<td>303-433</td>
<td>0.42</td>
<td>443-473</td>
</tr>
<tr>
<td>373</td>
<td>1.69</td>
<td>0.061</td>
<td>303-383</td>
<td>0.59</td>
<td>393-473</td>
</tr>
<tr>
<td>473</td>
<td>1.78</td>
<td>0.064</td>
<td>303-443</td>
<td>0.60</td>
<td>443-473</td>
</tr>
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The hall coefficient ($R_H$) is determined by measuring the Hall voltage that generates the Hall field across the sample of thickness ($t$), by:

$$R_H = \frac{V_H}{I} \frac{t}{B} \quad (5)$$

Carriers concentration can be determined by using the relation

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$$\mu_H = \frac{\sigma}{nq} \quad (7)$$

$$\mu_H = \sigma |R_H| \quad (8)$$

the mobility of electron and hole, respectively,

$$\mu = \frac{V_d}{E} \quad (9)$$

where $V_d$ is the drift velocity. So that we have

$$\sigma = (nq^2\tau)/m^* \quad (10)$$

where $\mu$: is the mobility, $\tau$: is the carrier’s lifetime, $n$: is the carrier’s concentration, $m^*$: is the effective mass of the carrier, and $q$: is the electron charge. Carriers concentration can be determined by using the relation

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$$p_H = \frac{1}{qR_H} \quad \text{for holes} \quad (11)$$

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Hall mobility ($\mu_H = \sigma |R_H|$), drift velocity ($V_d$), carriers concentration ($n_H$), lifetime ($\tau$) and mean free path ($\ell$) of charge carriers is calculated from Hall examinations. It can be noticed the whole films has negative Hall coefficient as shown in Table 5, the carrier’s concentration reduce with elevating temperatures, but the Hall mobility become more when temperatures is increased. Also, the parameters lifetime, drift velocity $V_d$ and mean free path $\ell$ of the carriers have been determined as in Table 5. It is observed that all parameters approximately increase with increasing of annealing temperatures [11, 12].

![Fig.5: ln $\sigma$ versus $10^3/T$ for ZnO films at different annealing temperatures.](image)

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<th>$E_{a1}$ (eV)</th>
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<th>$E_{a2}$ (eV)</th>
<th>Temp. Range (K)</th>
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<tr>
<td>RT</td>
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<td>0.056</td>
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<td>0.42</td>
<td>443-473</td>
</tr>
<tr>
<td>373</td>
<td>1.69</td>
<td>0.061</td>
<td>303-383</td>
<td>0.59</td>
<td>393-473</td>
</tr>
<tr>
<td>473</td>
<td>1.78</td>
<td>0.064</td>
<td>303-443</td>
<td>0.60</td>
<td>443-473</td>
</tr>
</tbody>
</table>
Table 5: Hall parameters for ZnO films at different annealing temperatures.

<table>
<thead>
<tr>
<th>T₀(K)</th>
<th>σ₀ × 10⁻⁵ (Ω cm)⁻¹</th>
<th>n₀ × 10¹⁴ (cm⁻³)</th>
<th>μ₀ × 10¹ (cm²/V sec)</th>
<th>v₀ (cm/s)</th>
<th>τ × 10⁻⁹ (s)</th>
<th>1 × 10⁻⁹ (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RT</td>
<td>3</td>
<td>2</td>
<td>18</td>
<td>0.93</td>
<td>1.30</td>
<td>1.20</td>
</tr>
<tr>
<td>373</td>
<td>0.22</td>
<td>0.13</td>
<td>60</td>
<td>1.58</td>
<td>1.47</td>
<td>2.33</td>
</tr>
<tr>
<td>473</td>
<td>0.02</td>
<td>0.01</td>
<td>83</td>
<td>0.03</td>
<td>1.65</td>
<td>2.93</td>
</tr>
</tbody>
</table>

Measurements (C-V) determines several factors parameters like junction capacitance (C), junction type and built-in potential (Vbi). The capacitance voltage of versus reverse bias was (0-1.2) volt with two frequencies 100 kHz and 200 kHz for ZnO /GaAs heterojunction as Fig. 6. The capacitance reduces voltage of the reverse bias depending on equation:

\[ C = \frac{dQ}{dV} \]  

(12)

The width of the junction can be deduced from the following equation:

\[ W = \frac{\varepsilon_s}{C_o} \]  

(13)

where \( C_o \) is the capacitance at zero biasing voltage, and \( \varepsilon_s = \frac{\varepsilon_e \varepsilon_p}{\varepsilon_n + \varepsilon_p} \).

The concentration of carriers was calculated from the relation [11]:

\[ \frac{1}{C^2} = \left[ \frac{2(\varepsilon_p N_p + \varepsilon_n N_n)}{qN_p N_n \varepsilon_p \varepsilon_n} \right] (V_D - V) \]  

(14)

where \( [2(\varepsilon_p N_p + \varepsilon_n N_n)/qN_p N_n \varepsilon_p \varepsilon_n] \) represent the slope. Fig. 7 is showing contrast \( C^{-2} \) with voltage. These data reveal the linear dependence in the reverse bias direction. The voltage axis provided the built-in potential value. It is observed that junctions have abrupt type (linear relation). The behavior is belonging to increasing width of depletion region (W) that results in increase in built-in potential value and the Vbi value as Table 6. The effect of frequency was showed in Fig. 6. It is noticed that the capacitance is reduce with elevating of frequency. Capacitance of the depletion layer indicates for charging increment on unit area depend on the changing with applied voltage. This means that behavior of the charge carriers transition was found as abrupt, that is determine by the relation between reverse bias and \( 1/C^2 \) as a straight line. This result is in agreement reported by [13].

![Fig. 6: The variation of capacitance as a function of reverse bias voltage for ZnO/GaAs heterojunction at frequencies 100 kHz and 200 kHz.](image-url)
**Fig. 7:** Variation of \((1/C)^2\) with voltage for ZnO /GaAs at frequencies 100 kHz and 200 kHz.

**Table 6: C-V parameters for ZnO /GaAs heterojunction at f=100 kHz and 200 kHz.**

<table>
<thead>
<tr>
<th>Frequency (kHz)</th>
<th>(C_0 \times 10^9) (F/cm²)</th>
<th>(W) (µm)</th>
<th>(n \times 10^{14}) (cm⁻³)</th>
<th>(V_{bi}) (volt)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>48</td>
<td>0.10</td>
<td>2.5</td>
<td>0.1</td>
</tr>
<tr>
<td>200</td>
<td>39</td>
<td>0.12</td>
<td>2.1</td>
<td>0.15</td>
</tr>
</tbody>
</table>

Using p-type GaAs substrate, ZnO/GaAs heterojunction device was prepared, the results of the current-voltage (I-V) measurements of the ZnO/GaAs heterojunctions were estimated under light and under dark conditions respectively as demonstrated in Fig. 8. This figure shows that the variation of the current at dark condition is approximately a typical of ohmic conduction. Also it can be recognized two regions in this figure the first one represent the recombination current, while the second region represents the tunneling current. The first region could be approximated by an expression:

\[I = I_o \exp\left(\frac{q V}{\beta k_B T}\right),\]

where \(\beta\) is the ideality factor. So the ideality factor has been calculated as in Table 7 and its about 2.7 which indicate that the junction have recombination tunneling mechanism.

**Fig. 8:** I-V characteristics under dark and illumination for ZnO/GaAs.

Fig. 8 demonstrates voltage of the open circuit \((V_{oc})\) and current of the short circuit \((J_{sc})\) were determined by intersects of photocurrent curve with x and y axis. Also maximum voltage \((V_m)\) and current solar cell \((I_m)\) were found. \(I_m\) and \(V_m\) are the current and voltage corresponding to the maximum power point. This is the point where maximum power can be
generated by the device. Full factor $F.F$ and efficiency had been calculated as shown in Table 7. The fill factor given by the equation [11].

$$FF = \frac{V_m I_m}{V_{oc} J_{sc}} \times 100 \quad (15)$$

The efficiency($\eta$) is another important parameter. It is a measure of the amount of light energy that is converted into electrical energy and is given by:

$$\eta = \frac{p_m}{p_i} = \frac{FF \times I_{sc} \times V_{oc}}{p_i} \times 100\% \quad (16)$$

<table>
<thead>
<tr>
<th>$T_d$(K)</th>
<th>$J_{sc}$ (mA/cm$^2$)</th>
<th>$V_{oc}$ (V)</th>
<th>$I_m$ (mA)</th>
<th>$V_{m}$ (V)</th>
<th>F.F</th>
<th>$\eta%$</th>
<th>$\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>RT</td>
<td>4.10</td>
<td>0.5</td>
<td>4</td>
<td>0.4</td>
<td>0.7</td>
<td>6.0</td>
<td>2.9</td>
</tr>
</tbody>
</table>

Conclusions
All the prepared films by pulsed laser deposition technique exhibits $n$-type conductivity and have good structural and morphology and have two activation energies. The Electrical and photovoltaic characteristic of ZnO/GaAs heterojunction are strongly dependent on the illumination and frequency, while the C-V measurement revealed that both prepared device are abrupt. The photocurrent is important parameter that have effective role in manufacturing solar cells.

References