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INVESTIGATION OF OPTICAL PROPERTIES OF ZnO NANORODS GROWN ON DIFFERENT SUBSTRATES

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ABSTRACT:
ZnO nanorods arrays are synthesized over the different substrates namely; Indium Tin Oxide (ITO), Kapton Tape (KT), Polyethylene terephthalate (PET), Porous Silicon (PS) and Silicon (Si) using modified chemical bath deposition (MCBD) method at 95 ºC for 4 h. The MCBD is the air bubbles inside growth solution during CBD process. The ZnO nano-seed layers are coated on different substrates using RF magnetron sputtering technique. The optical properties (transmittance, reflectance and energy band gap) and surface morphology of ZnO nanorods grown on different substrates have been investigated in details by using UV-Visible Spectrometer and Field emission scanning electron microscopy (FESEM), respectively. The results found that the morphology and diameter of ZnO nanorods is closely concerned with the nature of substrates. Also it is indicated that the substrate has strong and important impact on the growth, optical properties, E_g and quality of synthesized ZnO nanorods (NRs). The higher transmittance has been observed for ZnO NRs grown over KT substrates and is about (~ 33 %). The average transmittance decreases sharply near UV region at wavelength around 393 nm for ZnO nanorods grown on ITO substrate. However, for PET and KT substrates, the transmittance decreases sharply near visible region around 401nm and 498 nm, respectively. Besides, the ZnO NRs grown on PS substrate have the strong reflectance characteristics after approximately 395 nm, and then decreases in the wavelength range of 410 nm to 700 nm. On the other hand, the strong reflectance property of ZnO NRs grown on Si substrate is observed at 400 nm. Also, the minimum and maximum E_g are obtained for ZnO nanorods that fabricated on the KT substrate and porous silicon substrate, respectively.

KEYWORDS: ZnO, Nanorods, Substrates, Modified Chemical bath Deposition, Optical Properties

1. INTRODUCTION

In semiconductor, the optical properties are mostly depended in the crystal structure on both the intrinsic and the extrinsic impurities. It was received that the optical properties of zinc oxide was studied in the first time since 1960 (Thomas, 1960). The optical properties of ZnO are significantly influenced by the structure of energy gap and lattice dynamics (C.J. and S.P., 2006). The optical properties of 1-D ZnO structures is remarkable for abundant of their technological implementations in nano-photonic devices, LED devices, UV sensors and lasers (Ryu et al., 2006; Choy et al., 2003).

At room temperature (RT), the ZnO has the extensive exciton binding energy of 60 meV, which is larger than twice that of thermal energies (25 meV) and Gallium Nitrate (25 meV). Also, the ZnO has high optical gain which is about 300 cm^{-1} (Wong & Searson 1999; Meulenkamp, 1998). These unique optical properties at RT gives ZnO to equipping the very stable band edge ultraviolet emission by the exciton recombination process as opposed to the importantly less efficient electron–hole (e-h) plasma process employed in current devices based GaN (Li et al., 2008). In addition, the optical properties depend strongly on texture, nano-microstructure, and surface topography on the existence of impurities which in turn depend on the deposition (growth) technique doping and modification of the surface of the substrates (Ryu et al., 2006).

In the crystal structure of Zinc Oxide, there are various deep levels of impurities, which are affected on the optical and electrical properties of ZnO (C.J. and S.P., 2006). As a result, in the blue-UV region the ZnO is one of the promising photonic materials (C.J. and S.P., 2006). In the semiconductors, the electronic band structure is very important to be understood for its usefulness in devices and for further improving these devices performance (Özgür *et al.,* 2005; Rossler, 1969). ZnO is a direct optical energy gap (Eg) semiconductor with Eg = 3.37 eV, lack of poisoning, transparency in the visible domain, resistivity control over range 10^{-3} to 10^{5} Ωcm, high thermal, mechanical and chemical stability, high electrochemical stability, unique electrical and optical properties (Shinde *et al.,* 2005; Lupan *et al.,* 2007). These properties make ZnO semiconductor useful for nano-optoelectronic devices such as, gas sensors, UV light sensors, transparent conducting layers, dye sensitized solar cells, photocatalysts, blocking layer in flexible organic solar cell, thin film transistors, light emitting diode (LED) devices (Gimenez, 2010; Jha *et al.,* 2013). Due to the large advantage mentioned of the ZnO, the ZnO is currently the topic of among numerous groups research. This benefit is due in part to the availability of ZnO pure materials at relatively numerous unique qualities, low cost and high levels of lucidity and in part due to a great number of potential implementations for which it has been suggested that ZnO will have a wide benefit over other types of semiconductors

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(Wang, 2009; Desgreniers, 1999). In tandem with the increasing advantage in ZnO as a semiconductor, the nanotechnology field has also raised in popularity due to claims for increased application performance, device miniaturization, reduced power exhaustion and materials with excellently properties (Desgreniers, 1999). ZnO is belonging to the group of II-VI semiconductor materials and is an inorganic binary compound (Wang, 2009). ZnO different nanostructures materials have received huge benefit, chiefly in electronics, optics, and a large assortment of photonic usage, due to their novel and enchanting mechanical, optical, electrical, thermo, and chemical properties as well as their potential technology implementations (Wang, 2004; Alias & Mohamad, 2014). Different nanostructures of ZnO have different morphology such as, nanorods/nanowires, nanotubes, nanodisks/nanorings, nanopropellers, nanobelts, mesoporous nanowires, nanohelixes, nanocages with shell structure, nanohierarchical with propeller structure, nanocombs, tetraleg structure, spiral of nanobelts, nanosprings, nanorings formed by loop by loop coiling, nanoarchitecture, nanobows and rigid helix (Wang, 2004; H.M. & U. Ozgur, 2009).

These various ZnO nanostructures can be produced by different grown technique, such as chemical vapor deposition (CVD), hydrothermal synthesis, vapor phase transport (VPT), sol–gel, electrodeposition, sonochemical method, spray pyrolysis, chemical bath deposition (CBD) and modified chemical bath deposition (MCBD) method. Every growth technique has its own advantage and can create several ZnO nanostructures (Wang, 2004; Wang, 2007).

The MCBD method is attracting a great attention, high performance, most effective, and efficient growth method for synthesis various nanostructures and nanomaterial due to its advantages such as; reproducibility, low temperature (95 ºC), low cost, starting chemicals are commonly cheap and available, simplicity, it does not require complex growth system, use of environmentally friendly chemicals, non-hazardous, formation of high density arrays, high quality of obtained crystal and large capacity of growth vessel used (Shabannia, & Abu-Hassan, 2013; Ryu, 2006). Also, in MCBD method, for growing different nanostructures it can be using the large number of different substrates (flexible, nonflexible, organic and inorganic) that are kept in the aqueous solutions (Shabannia, & Abu-Hassan, 2013), it does not require the electrical conductivity of the substrates.

In previous study, researchers were essentially concentrated on the ZnO NRs preparation, parameters of growth solution and growth conditions such as precursor concentration, growth time, growth temperature, and pH value on the optical properties, morphology and the structure of the ZnO nanorods. Also, substrates kind able to influence on the ZnO nanorods growth because the substrates structure, substrate surface topography and their mismatch of lattice with nanostructure are certain significant parameters since these factors control the morphology (topography) and nature of ZnO nanorods fabrication (Ahmed *et al.,* 2016). The investigation of a substrate effect can help us to help growth mechanisms. In addition, an understanding of the substrate type impact has the potential parameter to realize the selective growth of nanomaterials, which are very useful for future nano sensors, nano electronics, and nano-devices (Wang, 2006).

In this investigation study, the effects of different types of substrates (flexible, non-flexible, organic, and inorganic) on the optical properties (transmittance, reflectance and energy gap) and surface morphology of ZnO nanorods have been investigated. The different substrates are Indium Tin Oxide (ITO) coated glass, polyethylene terephthalate (PET), Silicon (Si), Porous Silicon (PS) and Kapton Tape (KT). These substrates have been chosen in this study to understand the growth mechanism and optical properties of ZnO nanorods. This article study gives an indication that the substrate type has strong and remarkable effect on the optical properties, energy band gap and quality of fabricated ZnO nanorods.

2. EXPERIMENTAL DETAILS

In this study, all chemicals without further purification that used are from Company of Sigma Aldrich. Also the Deionized water with resistivity of 18.2 MΩ*cm is utilized for all preparation, synthesis and treatment process. The different substrates such as; ITO, PET, Kapton Tape, Silicon and Porous Silicon are employed the substrates for growing ZnO nanorods by MCBD. The cleaning process for all substrates mentioned has been described in details in our previous studies (Ahmed *et al.,* 2016; Ahmed *et al.,* 2016; Ahmed *et al.,* 2017; Ahmed *et al.,* 2017; Ahmed *et al.,* 2017). The porous silicon substrates are prepared from n-type Si (100) wafer by the photoelectrochemical Etching (PECE) method (Ahmed *et al.,* 2016). The PECE process was conducted in a Teflon cell. The electrolyte consisted of 1:4 volume ratio mixtures of hydrofluoric acid (48%) and ethanol (96%). At room temperature, the layer of PS was created for 20 min with a constant current density of 2 mA using the platinum (Pt) wire and silicon as the cathode and anode respectively (Ahmed *et al.,* 2016). During process of etching, the visible lamp with 60 W is used to illuminate the samples. The produced porous silicon substrates are swill with Deionized water and then dried with nitrogen gas after finishing the etching process. The ZnO seed layer with 100 nm thick is deposited over the cleaned various substrates by employing RF magnetron sputtering using (99.999% purity) of ZnO target with argon gas with pressure 5.5 $\times 10^{-3}$ mbar and RF sputtering power 150 Watt for a quarter hour. After that ZnO seed layer coated on the various substrates are placed in an annealing tube furnace at 200 ^{o}C for 1 h under atmosphere to stress relief the coated ZnO seed layer and improve quality ZnO seed layer.

The MCBD method has been employed for synthesis the high-goodness vertically well-aligned ZnO NRs on different substrates. The detail of the MCBD is described in our previous study (Ahmed *et al.,* 2016). The deionized water is used as the solvent and each of the Hexamethylenetetramine (HMTA) ($C_6H_{12}N_4$) and Zinc Nitrate Hexahydrate (Zn $(NO_3)_2.6H_2O$) are utilized as precursors. A 0.05 M of both ($C_6H_{12}N_4$) and (Zn $(NO_3)_2.6H_2O$) was dissolved separately in deionized water at 80 ^{o}C and two solutions mixed with each other under magnetic stirrer to get the homogenous growth solution. The ZnO seed layer coated various substrates types were vertically inserted inside a beaker containing a mixture of the two growth solutions. To investigate the impact of the different substrate's types on the optical properties of ZnO nanorods, the beakers were putted inside an oven for 4 h at 95 ^{o}C. After finishing the require time of growth of the ZnO nanorods, the all various substrates were taken first out from the solution of growth and washed by using the deionized water to take off the remaining salt, and the nitrogen gas is used to dry the produced samples because it is the inert gas not reacted with produced samples (Ahmed *et al.,* 2016). The chemical reactions of the formation or growth of ZnO nanorods grown over different substrates types can be summarized as below equation (1) (Tripathi & Rath, 2013; Wahab *et al.,* 2010; Polsongkram *et al.,* 2008; Strano *et al.,* 2014; McPeak *et al.,* 2013):

$$C_6H_{12}N_4 + 6H_2O \rightarrow 6HCHO + 4NH_3$$
$$NH_3 + H_2O \rightarrow NH_4^+ + OH^-$$

$$Zn(NO_3)_2^- \rightarrow Zn^{2+} + 2NO_3^-$$

$$Zn^{2+} + 4NH_3 \rightarrow Zn(NH_3)_4^{2+}$$

$$Zn^{2+} + 4OH^- \rightarrow Zn(OH)_4^{2-}$$

$$Zn(NH_3)_4^{2+} + 2OH^- \rightarrow ZnO + 4NH_3 + H_2O$$

$$Zn(OH)_2 \overset{\Delta}{\rightarrow} ZnO + H_2O$$

The Field Emission Scanning Electron Microscope (FESEM) model (FEI Nova a nano SEM 450 Netherlands and Leo-Supra 50 VP, Carl Zeiss, Germany) is used to examined the surface morphology, size, shape, density, aligned and distribution of ZnO nanorods. Also, the high-performance Cary 5000 UV-Vis- NIR Spectrophotometer in range (175-3300) nm-based transmittance and reflectance spectrums is used to investigate and study the optical properties of ZnO nanorods synthesized over the different substrates.

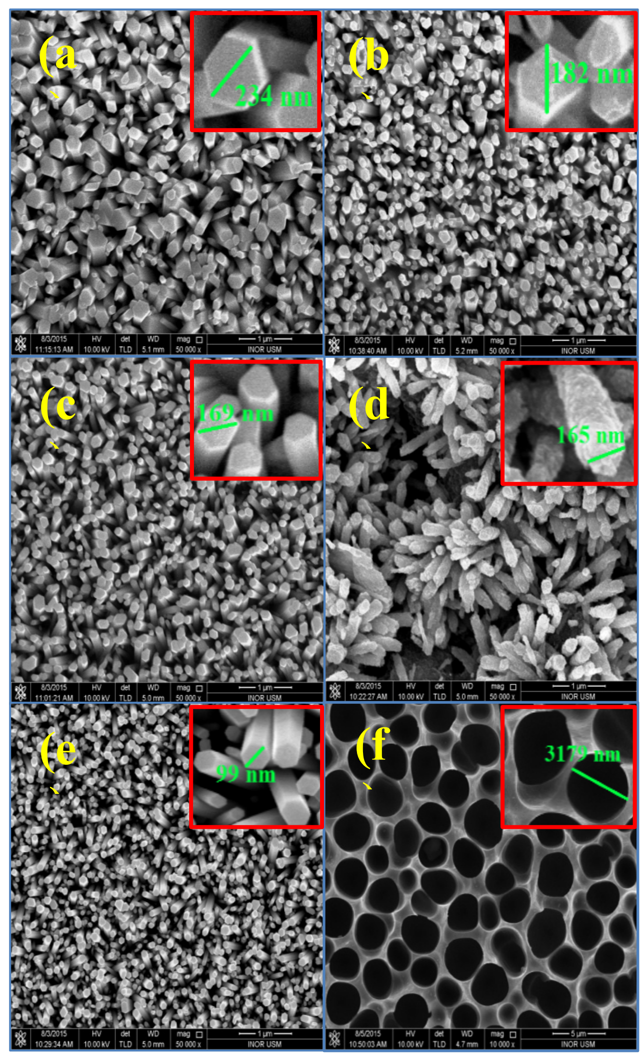
3. RESULTS AND DISCUSSION

The field emission scanning electron morphology (FESEM) images top view of the fabricated ZnO nanorods over different substrates types are shown in Figure (1). Figure (1a) shows the ZnO nanorods grown on ITO substrate, it was observed that the fabricated ZnO nanorods was randomly oriented, have different size and shape, non-homogenous and no vertically well-aligned over the entire ITO substrate with average diameter of 234 nm. The vertically aligned ZnO nanorod along the c-axis with a high distribution density of nanorods over the entire KT substrate is shown in Figure (1b). It can be indicated that ZnO nanorods orientation is random and has nanotube and nanorods. Also it can clearly see the raindrop over the most of produced ZnO nanorods. This is may be due to effect of KT substrate on the growth way of nanorods on this type of substrate. The average diameter of fabricated ZnO nanorods is about 182 nm. However, the average diameter of produced ZnO nanorods on PET substrate is decreased to 169 nm as shown in Figure (1c). It was observed that the fabricated ZnO nanorods over the PET substrate are vertically well- aligned, same shape, uniform shape, and uniformly orientated with high distribution. The ZnO well-defined nanorods arrays were successfully fabricated vertically over PS substrate as shown in Figure (1d). It was observed that the produced ZnO nanorods like nanoflowers with average diameter are about 165 nm and no hexagonal shape. The bottom of nanorods is smaller than the top of nanorods. This is due to the porous silicon layer produces the large mismatch in the thermal expansion coefficients and lattice constant, thereby producing large stress between the PS substrate and ZnO nanostructures (Wang, 2014). In addition, the vertically well-aligned ZnO nanorods with high density distribution over the Si substrate are shown in figure (1e). It can clearly observed that the obtained ZnO nanorods have uniform size, uniform shape, uniform orientation and have hexagonal shape with average diameter of 99 nm. The surface morphology of produced PS layer substrate for growing the ZnO nanorods over it is shown in Figure (1f). It was observed that the porosity has been done for the silicon before coating ZnO seed layer over it. This indicated that the PS is used for growing the ZnO nanorods. The obtained PS layer has almost uniform circle shape of porosity and different size with average diameter about 3179 nm.

From the all images in Figure (1), one can conclude that the substrate plays a main role in obtaining the nanorods morphology. This nanorods morphology depends on the substrate type which can be concerned to the fact that the structures of substrates, surface topography and their lattice mismatch with nanostructure have particular important effect on nanorods morphology. These factors control the nature and morphology of synthesis ZnO nanorods. The investigation of a substrate effect can help us to probe growth mechanisms (Wang, 2006).

Figure (2) shows the optical transmittance spectrum of fabricated ZnO nanorods on different substrate such as ITO, PET and Kapton Tape with range of wavelength from 300 nm to 800 nm. It was observed that the all fabricated ZnO nanorods samples in visible region have high transmittance and in the UV region have low transmittance. The higher transmittance is found for ZnO NRs produced over polymer Kapton Tape (KT) substrate which is decreased from (~ 33 %) to (~ 7 %) for ZnO nanorods prepared on PET substrate. But the average transmittance is about (~ 16 %) for ZnO nanorods grown on ITO substrate. This may basically be due to enhanced scattering effect in ZnO nanorods films grown on PET substrate. The transmittance decreases sharply near UV region at wavelength around 394 nm due to the optical energy band gap absorption when ZnO nanorods grown on ITO substrate. But for PET and KT substrates, the transmittance decreases sharply near visible region around 401 nm and 498 nm, respectively. The substrates topography (nature), ZnO nanorods thin film thickness and shape of ZnO nanorods may be increasing the optical scattering, which decreases (reduce) the transmittance of ZnO nanorods thin films.

Figure 1. Top view FESEM Images of ZnO Nanorods for Different Substrate Types (a) ITO, (b) KP, (c) PET, (d) PS, (e) Si & (f) PS Substrates



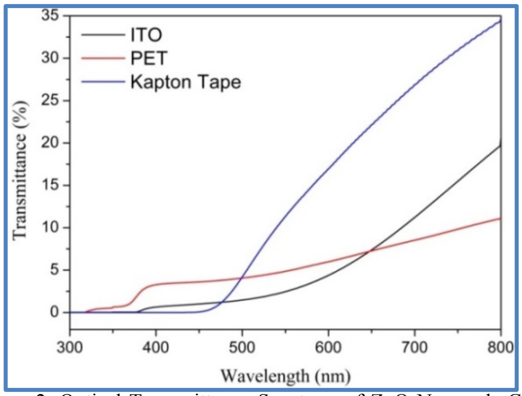


Figure 2. Optical Transmittance Spectrum of ZnO Nanorods Grown on Different Substrates Types (ITO, PET, and Kapton Tape)

Both Si and PS substrates are not transparent, and then the diffuse reflectance spectrum is used to study the optical properties of ZnO nanorods grown on these two substrates. Figure (3) shows the diffuse reflectance spectrum of the ZnO nanorods grown on the silicon and porous silicon in the range of wavelength of 300 nm to 800 nm. The sharp increase in reflectance spectrum or absorption brim of produced ZnO nanorods on silicon and porous silicon substrates at 380 nm and 355 nm due to the energy band gap. The ZnO nanorods grown on PS have the strong reflectance characteristics after approximately 395 nm, and then decreased in the domain of wavelength of 410 nm to 700 nm. But the strong reflectance property of ZnO nanorods grown on Si substrate is noted at 400 nm. This may be due to the substrate type topography, size and length of obtained ZnO nanorods.

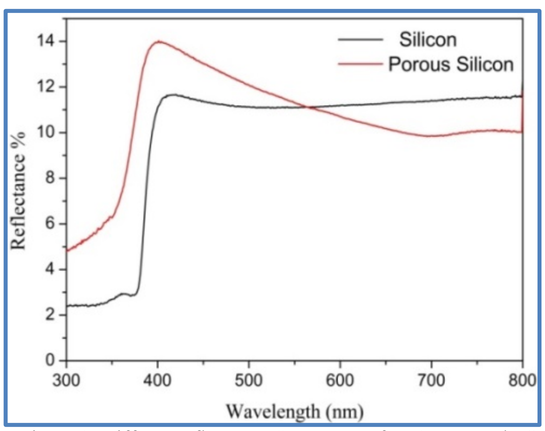


Figure 3. Diffuse Reflectance Spectrums of ZnO Nanorods Grown on Silicon (Si) and Porous Silicon (PS) Substrates

In order to investigate the optical band gap (Eg) of ZnO nanorods through transmittance spectrum and diffuse reflectance spectrum as shown in Figure (4), an Tauc formula by the extrapolation of the linear portion of $(\alpha h\nu)^2$ versus hν plots is utilized to calculate the energy band gap of ZnO nanorods grown on transparent substrate (ITO, PET and KT) by following (Akhiruddin *et al.,* 2015):

$$(\boldsymbol{\alpha h\nu})^2 = \mathbf{A}(\mathbf{h\nu} - \mathbf{E_g})^{\mathbf{n}} \quad 2$$

Where α is the coefficient of absorption, hν is the energy of photon, A is constant, Eg is the optical energy gap and n depends on the type of transmission (equals to 1/2 for allowed direct transmission). For the transmittance spectrum the (α) coefficient can be calculated by (Akhiruddin *et al.,* 2015):

$$\boldsymbol{\alpha} = \frac{\ln\left(\frac{1}{T}\right)}{d} \quad 3$$

Where T is the ZnO films transmittance and d is the film thickness. Also the (α) coefficient can be obtained from the absorbance spectrum.
However, for non-transparent substrates (Silicon and Porous Silicon), the Kubelka-Munk functions by multiplying the F(R) function by hv is used to calculate the energy band gap by following equations (10-11) (López & Gómez, 2012):

$$\mathbf{F(R)} = \frac{(1-\mathbf{R})^2}{2\mathbf{R}} \quad 4$$

Where F(R) is proportional to the coefficient of extinction (α) and R is the reflectance (López & Gómez, 2012). This equation is commonly utilized to absorbing particles in a matrix and highly light scattering materials. The basic Kubelka-Munk functions suppose the illumination of diffuse of the particulate coating. A modified (K-M) function can be investigated by multiplying the F(R) function by hv, utilizing the corresponding coefficient (n) related with an electronic transition as follows:

$$(\mathbf{F(R)} * \mathbf{h\nu})^{\mathbf{n}} \quad 5$$

By plotting above equation as a function of the energy gap in eV, the E_g of semiconductor particles of can be determined (López & Gómez, 2012).

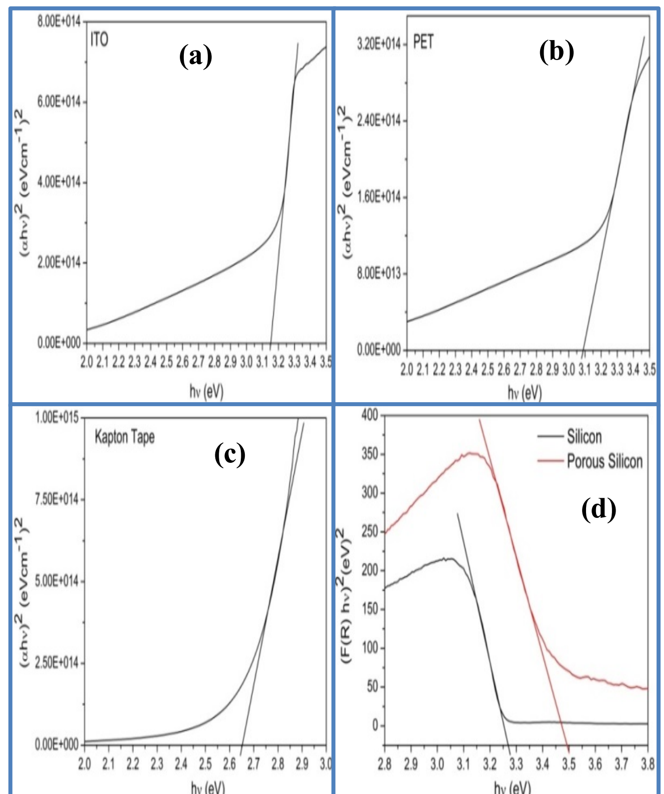


Figure 4. Tauc plot $(\alpha h\nu)^2$ and Kubelka-Munk $(F(R)\ h\nu)^2$ Versus Energy Band Gap (hν) of the ZnO Nanorods Fabricated for Different Substrate Types, (a) ITO (b) PET (c) KT, (d) Silicon and Porous Silicon

The obtained values of direct E_g of produced ZnO nanorods on different substrate such as ITO, PET, Kapton Tape, Silicon, and Porous Silicon are listed in Table (1). The minimum and maximum energy band gaps are noted for ZnO nanorods that fabricated on the polymer (Kapton Tape) substrate and porous silicon substrate, respectively. This may be related to the topography of substrate, grain size, aligned, length, morphology, and crystal structure of obtained ZnO nanorods grown on these substrates (Ahmed *et al.,* 2016).

Table 1. Energy Band Gap (Eg) of ZnO Nanorods Grown on Different Substrates Types

Substrate Type	E_g (eV)
ITO	3.15
PET	3.09
Kapton Tape	2.65
Silicon	3.27
Porous Silicon	3.5

4. CONCLUSION

In conclusion, high quality vertically well-aligned ZnO nanorods were fabricated successfully on different substrates types using modified chemical bath deposition method for 4 h at 95 ºC. The effect of several substrates types on the optical properties based the transmittance and reflectance of ZnO NRs has been obtained. One can concluded that the substrate topography or type consider a significant growth or preparation parameters for controlling the morphology, size, shape, quality, optical properties and energy band gap of ZnO nanorods. Our studies on different substrate effect indicate that by choosing appropriate growth condition, one can tune the light emission, which might be potential candidates for nano-optoelectronic device implementation. The selective growth of ZnO nanorods is possible through the choice of the type of substrates.

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