Photocatalytic Degradation Characteristics of ZnO Thin Films: Investigation on Solvent's Influence

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Research Article

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Abstract

Using a sol-gel dip coating process, this research created on glass substrates, ZnO thin films are formed with a range of solvents (2-methoxyethanol and ethanol). The goal of this study is to determine how solvent type affects structural, photocatalytic, morphological, optical, and electrical characteristics of ZnO films. Hall effect, AFM, XRD, and UV–vis spectroscopy has been employed to characterize the films. The findings indicate that the solvent used greatly affects the characteristics of ZnO films. Photocatalytic effectiveness of the produced photocatalysts by Methylene blue (MB) photodegrades in a dilute aqueous solution when exposed to UV-A light have been determined. The findings indicate the sample created using ethanol is more efficient than the sample made using 2-methoxyethanol.

1. Introduction

Industrial and commercial activities, as well as contemporary lifestyles, have exacerbated environmental pollution concerns. Numerous factors contribute to this pollution, most notably industrial waste, heavy metals, and others that are hazardous to individuals and the environment. As a result, it is critical to develop a technique capable of removing the hazardous chemical compounds that poison the environment. as a consequence, for this reason, significant emphasis is placed on the development of methods for converting harmful organic contaminants into simple, harmless substances in order to reduce environmental contamination. Photocatalytic activity is one of the known ways in this domain. ZnO, Fe_2O_3, MgO, CuO and TiO_2, and other metal oxides have been examined as photocatalyst materials for organic pollutants in the environment are decomposed [1, 2]. ZnO is one of the most researched materials among them due to its high photocatalytic effectiveness and stability [3].

ZnO films have been extensively explored in recent years and are usually regarded as the most appropriate semiconductor materials for a variety of possible applications. Zinc oxide is a widely used substance with a broad band gap. Take into account their 3.37 eV band gap, at ambient temperature, their 60 meV excitonic binding energy [4], their powerful emission, their high velocity of saturation ($3.2x10^7$cm.s^{-1}), and their considerable breakdown voltage [5]. Due of it's optical, physical, and electrical properties, as well as its high thermal and chemical stability, abundant natural supply and nontoxicity. ZnO is ideally suited for a range of applications [3]. These include, light emitting diode [13], acoustic devices [7], gas sensing [9], thin film transistors [8], solar cells [10], and photodetectors [6], lasers [12] and ceramics [11].

Numerous techniques, including magnetron sputtering [16], spray pyrolysis [15], sol gel approach [17], and deposition by pulsed laser [14], is utilized for the purpose of fabricating ZnO films. Several of these techniques, the sol–gel technique displays a number of distinct benefits, including simplicity, accurate composition control, cheap cost, great homogeneity, simple thickness control, and a low crystallization temperature [18, 19]. Many reviews were centered on ZnO thin film photocatalytic activity [20–23].

Zinc oxide thin films were produced in this work utilizing the sol–gel dip coating process using a variety of solvents. The effect of solvent type on the optical, structural, electrical, photocatalytic and morphological characteristics of ZnO films have been investigated.

2. Experimentation

ZnO Films Fabrication

The host precursor was zinc acetate dehydrate ($Zn(CH_3COO)_2.2H_2O$), and the stabilizing agent was monoethanolamine (MEA). All precursor solutions were prepared using the organic solvents ethanol and 2-methoxyethanol. All compounds were analytic grade and were used unpurified.

At room temperature, 30 ml of zinc acetate dehydrate was dissolved in 30 ml of each solvent individually in a beaker to create ZnO solutions. Zinc Acetate dehydrate was held at a constant concentration of 0.3 M. The resultant mixed solution was agitated at 333 K for 1 hour with a magnetic stirrer, and the milky solution was then mixed dropwise with MEA and stirred for another 1 hour to obtain a homogenous mixture.

Following that, at ambient temperature, the solution was kept covered for 24 hours. Ultrasonically cleaned glass substrates in methanol, acetone, and deionized water. At ambient temperature, the films were produced using the dip coating process at withdrawal speeds of 100 mm/min. Following each deposition, all samples were dried at 200°C for ten minutes to remove solvents. Finally, films were annealed for 2 hours at 500°C.

Characterization

Numerous characterization approaches were employed to investigate the characteristics of films. X-ray diffraction was used to determine the structural properties of films (XRD, Bruker AXS-8D) with Cu K radiation (Cu Kα = 0.1541 nm), the morphology of films was determined using atomic force microscopy (A 100-AFM), the optical characteristics were examined via a UV-visible spectrophotometer (Jasko V-630), and the hall effect (HMS-3000) was used to determine the electrical characteristics.

Catalytic Activity Through Photocatalysis

The photocatalytic activity of ZnO thin films was determined by measuring the breakdown of methylene blue (MB) in an aqueous solution under visible light. Thin film ZnO were placed in a beaker containing 5×10^{-5} M MB of MB solution. The photocatalytic degradation of MB solution was examined at 665 nm via UV–Vis spectrophotometer type (Jasco V-630) to determine the absorbance of the solution every 30 minutes. The relationship was used to calculate the efficiency of MB deterioration is [24],

$$\text{Degradation (\%)} = \frac{C0 - C1}{C0} \times 100 = \frac{A0 - A1}{A0} \times 100 \quad (1)$$

where: C_0 denotes the initial concentration and C_1 denotes the concentration after t minutes. A_0: initial absorbance, whereas A_1: the absorbance after 't' minutes.

3. Results And Discussion

Rate of Growth

The thicknesses have been approximated and are shown in Table 1and the calculated growth rates of the films for the two solvents used. The thicknesses of the ZnO thin layers vary between (180 and 230) nm. It is observed that the maximum thickness corresponds to the film prepared by 2-methoxyethanol and the minimum thickness for the film prepared from ethanol, different films' growth rates vary (7.2 to 9.2) nm/min, the growth rate of the film fabricated by 2-methoxyethanol is higher than that is made by ethanol. Variation in the rate of growth as a function of the nature of the solvent may have its origin in the variation in the viscosity of the solution with the nature of the precursor. The use of 2-methoxyethanol leads to a more viscous solution than the case of ethanol.

Table 1
Thicknesses, growth rate, Crystallite's size and surface roughness of ZnO thin films.

Solvents	Thicknesses (nm)	growth rate (nm/min)	Crystallite's size (nm)	surface roughness (nm)
Ethanol	180	7.2	20.53	12.1
2-Methoxyethanol	230	9.2	32.23	17.6

Structural Properties

The crystalline structure of films produced with two solvents was reported XRD. The spectrums of the two samples are shown in Fig. 1 with different solvents. The spectrum of the film prepared with 2-methoxyethanol contains intense peaks on the other hand the spectrum of the film prepared with ethanol is a little diffuse with wide peaks of low intensities. The difference in the intensities of the peaks is probably due to the difference in the thicknesses of the films because the film prepared with 2-methoxyethanol has a thickness of the order of 230 nm, Alternatively, the thickness of the created film with ethanol is thinner, it is of the order of 180 nm.

The prepared films essentially composed the ZnO polycrystalline phase proven by the peaks relating to the planes (100), (002), (101), (102), (110), (103) and (112). This indicates zinc oxide films have a hexagonal wurtzite structure and are preferred to be orientated perpendicular to the surface of the substrate along c-axis. It should be mentioned that the film's spectrum was produced with 2-

methoxyethanol shows the appearance of new peaks corresponding to the diffraction planes (201) and (004). The width at half maximum (β) is about 0.41 for the film prepared with 2-methoxyethanol, but in the case of the film prepared with ethanol, it is larger, it is l order of 0.74. The size of the crystallite can be approximated using Scherer's formula from the whole width at half maximum (β) of the (0 0 2) peak of diffraction [25]:

$$D = {}^{0,9 \times \lambda}/_{\beta \times \cos\theta} \qquad (2)$$

where: λ: wavelength of X-ray, β: XRD peak's entire width at half maximum, θ: Bragg diffraction angle.

The films' crystallite sizes produced by two solvents are set out in Table 1. As may be observed the crystallites in the film manufactured using 2-methoxyethanol are larger than those produced with ethanol. This difference is mainly due to the thicknesses of the films because the film prepared by 2-methoxyethanol has a thickness of the order of 230 nm, while the film obtained by ethanol, the thickness is of the order of 180 nm. The results acquired are contradict the results of Srivastava et al. [26] who state that regardless of the solvent used, the size of the crystallites remains the same at around 30 nm.

Surface Morphology

The AFM images, presented in Fig. 2 **(a, b, c, and d)**, reveal that deposited ZnO thin films have a continuous and dense surface morphology. It is interesting to see that the morphology of the films' surfaces varies on the composition of the solvent utilized. ZnO film's prepared surface from ethanol has smaller grains than those of the film's surface is prepared using 2-methoxyethanol. Table 1 reports the surface roughness values for the two solvents studied. It is noted that the films prepared with ethanol have smoother surfaces than those prepared with 2-methoxyethanol. This difference in surface roughness is mainly due to the size of the grains formed in the films prepared based on 2-Methoxyethanol compared to that of the grains formed in the films prepared with ethanol.

3.1. Optical Characteristics

Transmittance

The transmission spectrum of the films taken are examined using a UV-Vis spectrometer. Figure 3 illustrates the transmittance variation as a function of the film's wavelength made using two solvents. The measurements of the transmittance measurements were made in the wavelength region (300–1100) nm where the films possess a high transmittance of more than 80% in the visible spectrum, suggesting a high optical quality of the film deposit. Ethanol-deposited films are more transparent than 2-methoxyethanol-deposited films which give transmissions of the order of (90 and 82.4) % respectively. This difference in transmission is due to the difference in the thicknesses of the films. Indeed, the film obtained with 2-methoxyethanol has a higher thickness than that of the film prepared with ethanol. the

results obtained do not agree with the results found by Srivastava et al. [26] who found that the highest transmittance factor for the film prepared with 2-methoxyethanol is 80%, whereas that prepared with 2-methoxyethanol. ethanol is 62%.

Optical Band Gap

Using the Tauc equation, the energygap of ZnO thin films produced with different solvents (ethanol and 2methoxyethanol) was calculated [27–50]:

$$(\alpha h\square) = B(h\square - E_g)^{\frac{1}{2}}$$

3

where: B: a constant, $h\upsilon$: photon energy, and E_g: energygap.

The fluctuation of $(\alpha h\upsilon)^2$ of the ZnO films formed by two solvents as a function of the energy of the incoming photon $h\nu$ is shown in Fig. 4. The linear component of these curves may be extrapolated to get the energy gap E_g, it is of the order of (3.35 and 3.34) eV respectively for ethanol and 2-methoxyethanol. The result obtained allows us to deduce that the gap energy depends slightly on the nature of the solvent. These results are comparable with those obtained by Srivastava et al [26]. The increase in the optical gap between films deposited with 2-methoxyethanol and those deposited with ethanol may be due to the films' microstructure and surface morphology, which alter the films' interatomic bonding. It is noted that all the films show a band gap close to that of bulk ZnO (3.37 eV).

3.2. Electrical Characteristics

Electrical Conductivity

The electrical conductivity values of ZnO films made in variety solvents (ethanol and 2-methoxyethanol) are shown in Table 2. In this Table, the results indicate that the conductivity of ZnO films deposited by 2-methoxyethanol is higher than that obtained from ethanol. This increase in conductivity in the film made from 2-methoxyethanol compared to that of the film prepared with ethanol is due to the size of the grains as suggested by the AFM images Fig. 2, the films deposited from of 2-methoxyethanol have relatively large grains which causes an increase in carrier mobility, this results in an increase in film conductivity.

Table 2
Optical band gap, electrical conductivity, figure of merit and MB photocatalytic degradation of ZnO thin films.

Solvants	Optical band gap (eV)	Electrical conductivity $(\Omega.cm)^{-1} \times 10^{-3}$	Figure of merit $(\Omega^{-1}) \times 10^{-8}$	Degradation (%)
Ethanol	3.35	9.1	4.06	81.3
2-Methoxyethanol	3.34	7.3	6,42	73.9

Figure of Merit

Table 2 summarizes the change in the figure of merit of ZnO films fabricated using two solvents. It is self-evident that the ZnO films deposited with 2-methoxyethanol has a figure of merit greater than that of the film produced with ethanol. It is concluded that the preparation of the films with 2-methoxyethanol proves to be an optimal solvent for ZnO thin films produced via dip coating technique.

3.3. Photocatalytic Activity

At ambient temperature, ZnO thin films' photocatalytic activity was investigated by measuring the degradation of methylene blue (MB) solution to visible light in both with and without ZnO films.

The impact of the solvents utilized (ethanol and 2-methoxyethanol) on the photocatalytic degradation of MB presence of visible light is seen in Fig. 5. As shown in this illustration, MB does not degrade without ZnO films. However, if ZnO films are present, the ethanol-fabricated sample degrades more rapidly than the 2-methoxyethanol-prepared sample have been observed.

The degrading efficiency or conversion rate of thin ZnO films produced with various solvents is shown against time in Fig. 6 for a 300-min irradiation duration. These graphs demonstrate that ethanol and 2-methoxyethanol have a degradation efficiency of 81.3% and 73.9%, respectively (Table 2). The variation in the efficiency of MB degradation demonstrates that the solvent type has an effect on the photocatalytic effectiveness of ZnO thin films. This discrepancy has been linked to differences in the size and shape of the cristallites in ZnO films [51–54]. As a result, the film formed with ethanol is more uniform and hexagonal in shape (20.53 nm of cristalittes size), but the film made with 2-methoxyethanol as the solvent has crystallites size of 32.23 nm and is less effective in MB photocatalytic degradation.

4. Conclusions

In this study, On glass substrates, ZnO thin films were produced utilizing the sol gel dip coating technique and the influence of the solvent on the structural, morphological, optical, electrical, and photocatalytic characteristics of ZnO films was examined. The findings indicate that the solvent utilized has a

noticeable impact on the characteristics of ZnO films. The photocatalytic degradation of MB in the presence of various fabricated materials and UV light irradiation revealed that film made using ethanol as the solvent exhibited a greater degree of photodegradation than the film prepared using 2-methoxyethanol.

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Figures

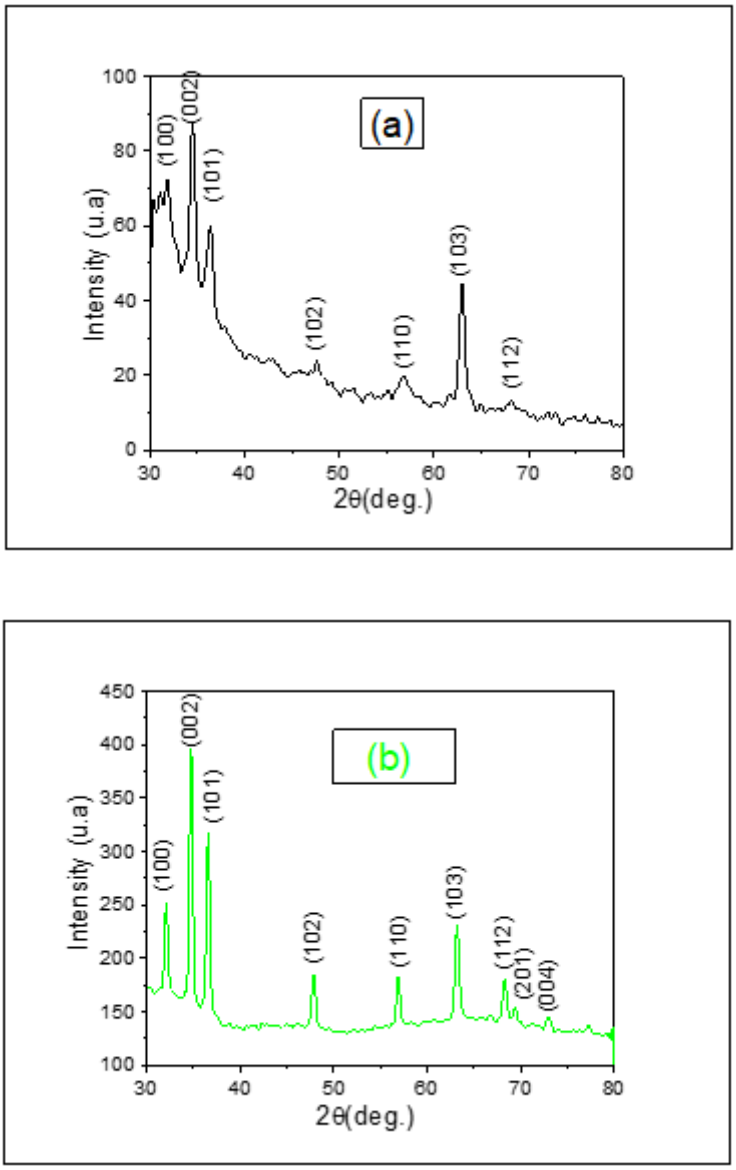

Figure 1

XRD patterns of ZnO thin films prepared from (a) Ethanol and (B) 2-Methoxyethanol.

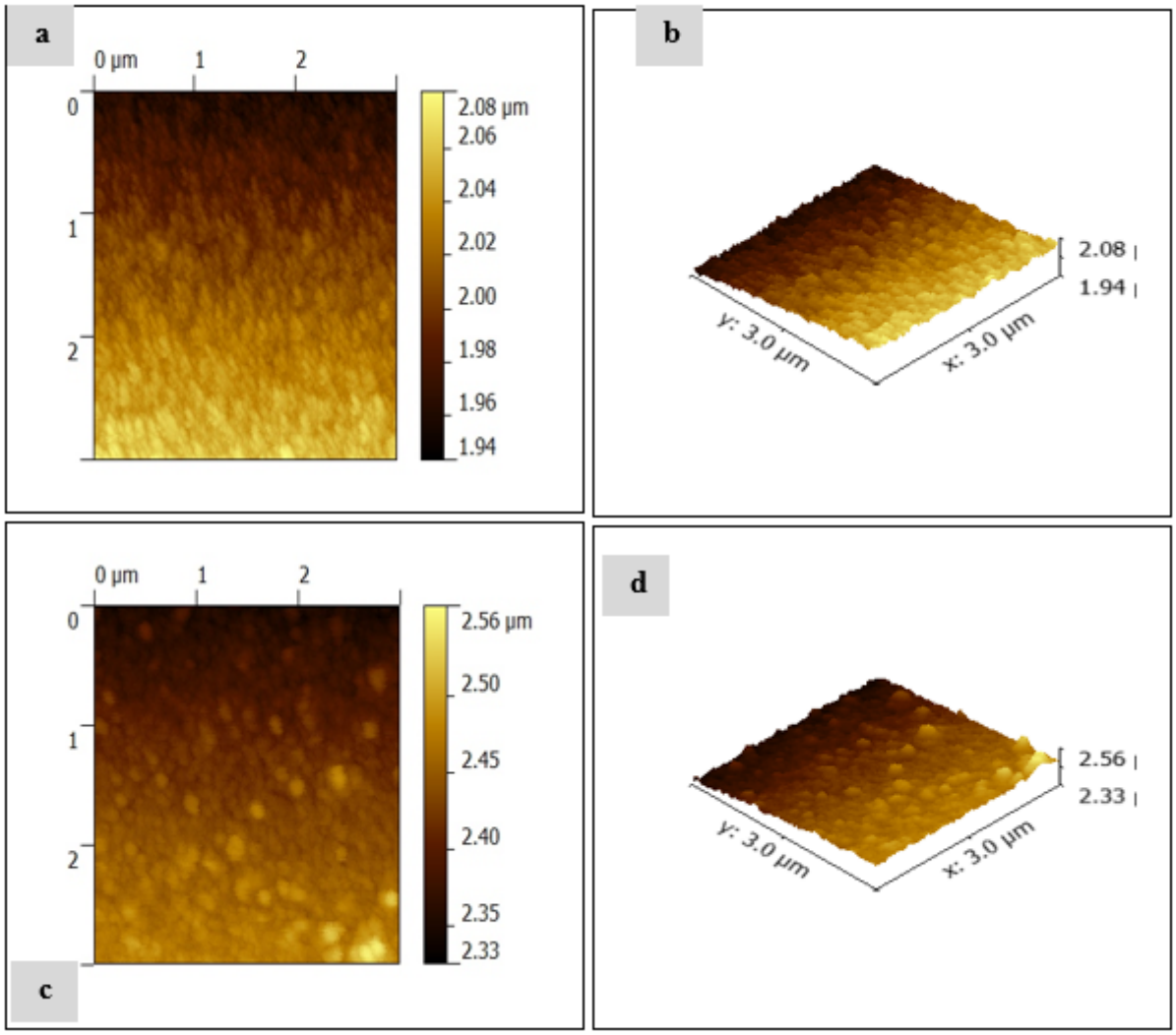

Figure 2

Surface morphology images of ZnO thin films prepared from (a, b) ethanol, (c, d) 2-Methoxyethanol.

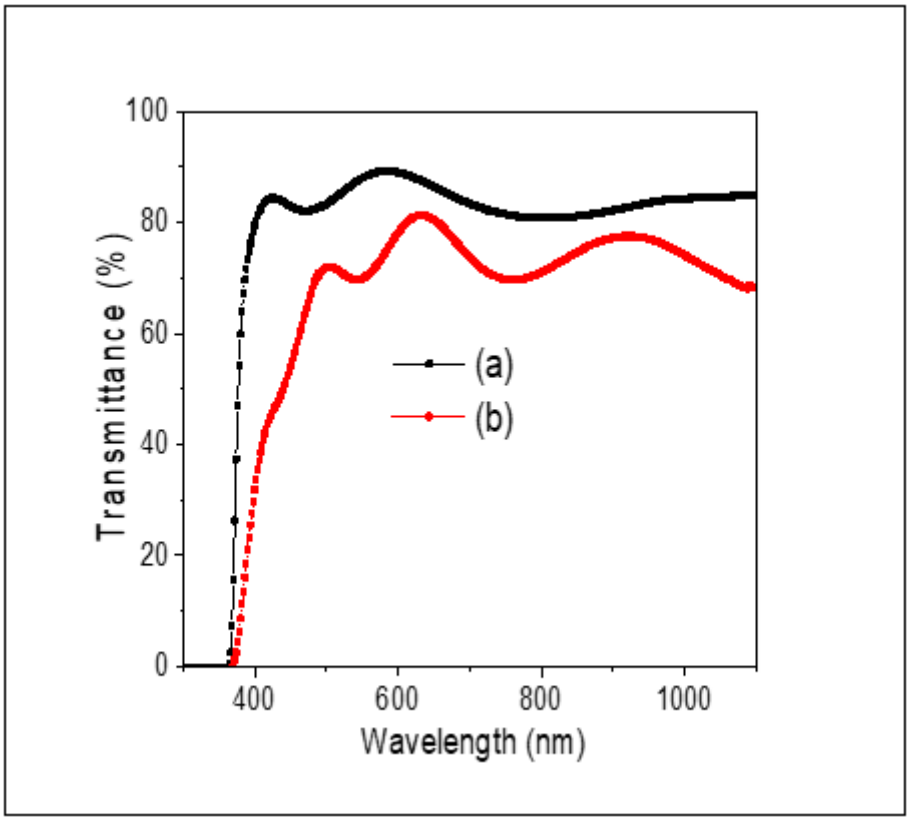

Figure 3

Transmittance spectra of ZnO thin films prepared from (a) Ethanol and (b) 2-Methoxyethanol.

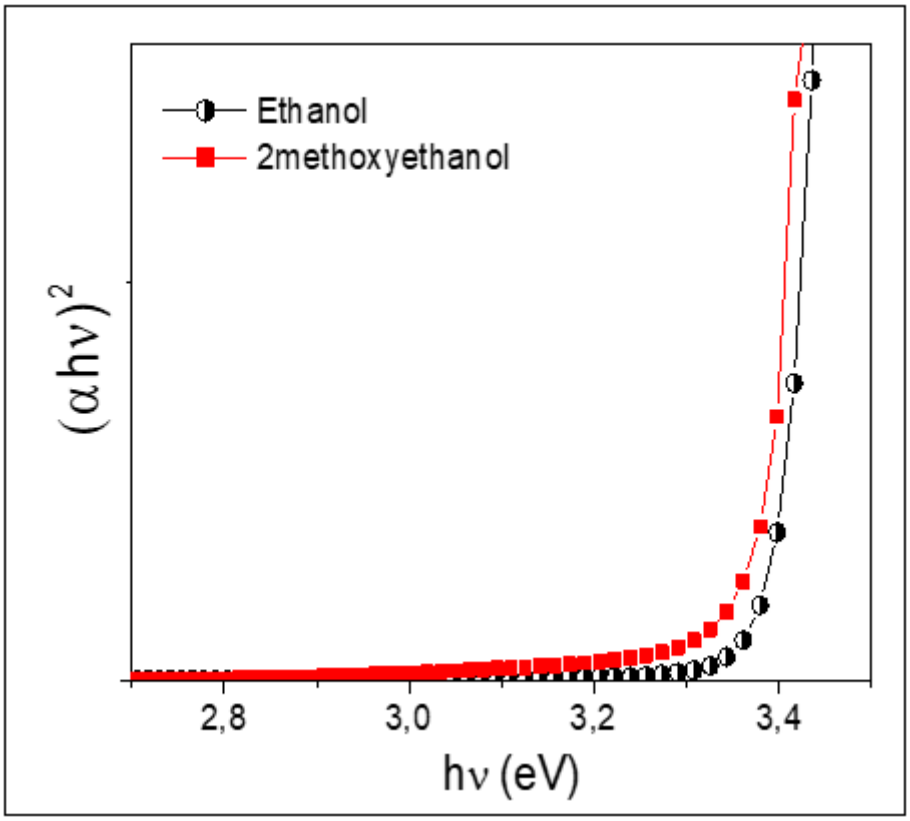

Figure 4

Curves of $(ahu)^2$ ersus photon energy for ZnO thin films prepared from (a) Ethanol and (b) 2-methoxyethanol.

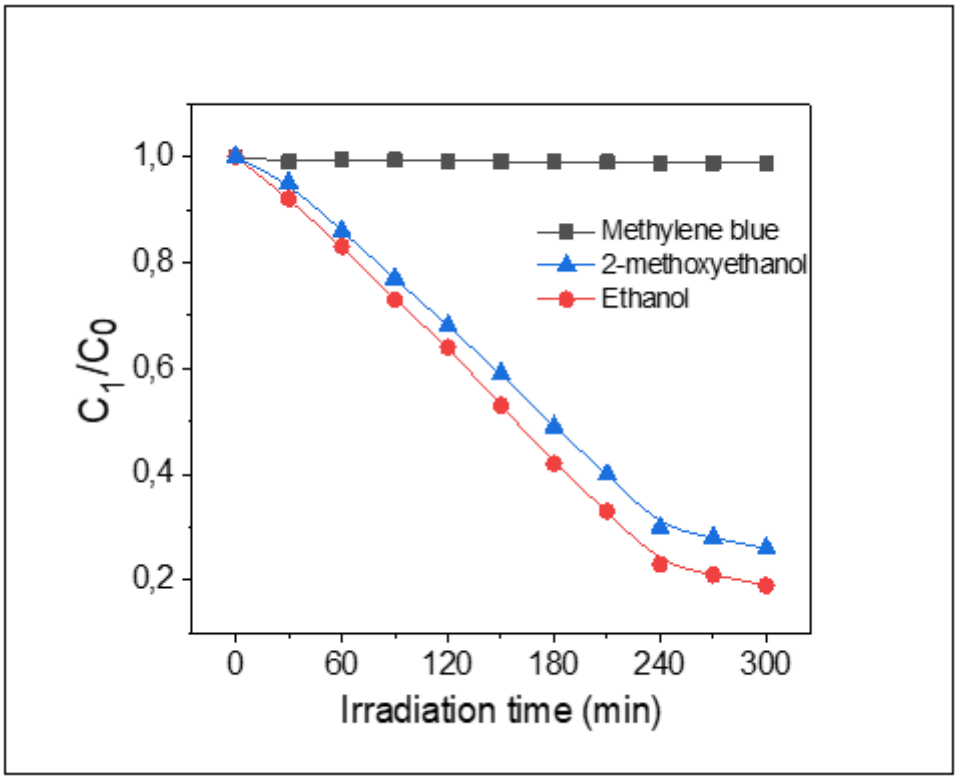

Figure 5

Photocatalytic degradation of MB measured in: ZnO thin films prepared from ethanol and 2-methoxyethanol.

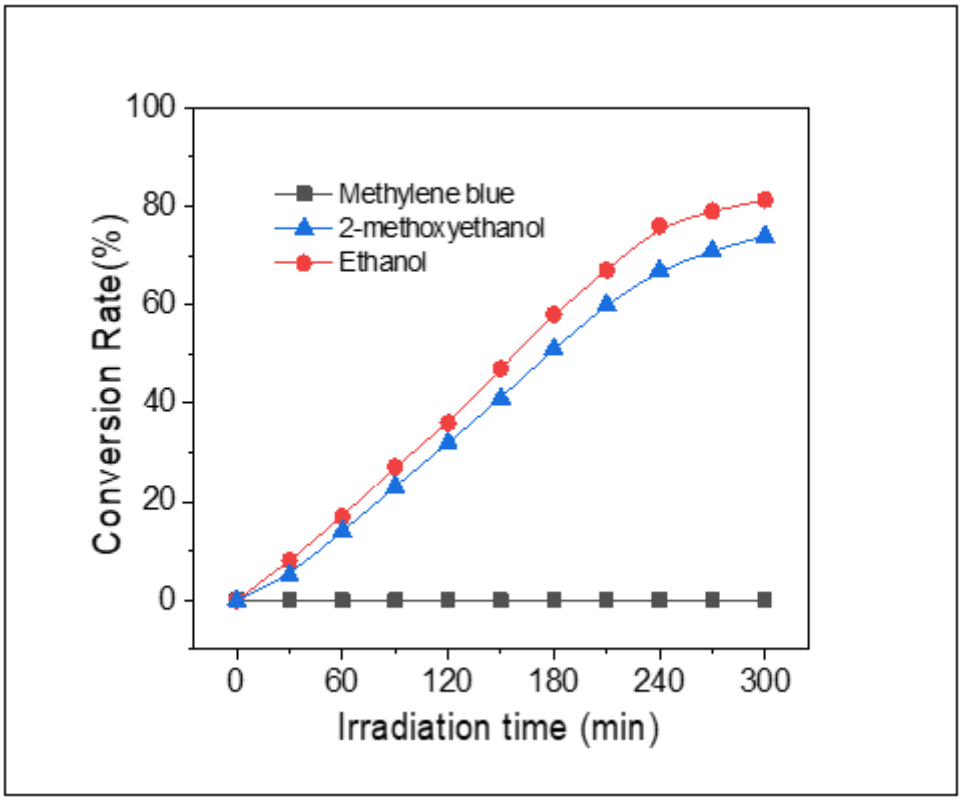

Figure 6

Conversion rate of MB degradation measured in: ZnO thin films prepared from ethanol and 2-methoxyethanol.

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