

Analyses the Scope of Solar Radiation Transmittance and Physical Properties of Polymethyl Methacrylate/Zinc Oxide Nanocomposite Films

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ABSTRACT

Films of poly (methyl methacrylate) PMMA and PMMA doped with zinc oxide nanoparticles (ZnONPs) with different amounts were created using the solution casting technique. Different amounts of zinc oxide ranging from (0.001-0.005) g in step 0.001g were investigated and added to a polymer of constant amount (0.5) g of PMMA in 15ml Tetrahydrofuran (THF). It is known that increasing the amount of ZnONPs in this technique contributes to the appearance of ZnONP peaks within the polymer. The differences in the XRD spectrum show that the nanomaterial changed the microstructure of the polymer. This was discovered using a nanoscale scanning electron microscope (SEM). SEM revealed that the PMMA polymer surface exhibit an amorphous nature with a smooth surface. Fourier Transform Infrared (FTIR) spectroscopy demonstrates that there was no chemical reaction between the PMMA polymer and ZnONPs. Spectrophotometric measurements of absorbance and transmittance in the wavelength range 200- 900 nm were used to evaluate the optical characteristics of all films. Increased amounts of ZnONPs improve the absorbance, absorption coefficient, and extinction coefficient of PMMA polymer. With increasing ZNO content, the variance in computed optical energy gap values has been interpreted, the optical energy gap dropped from 4.65 eV to 4.10 eV. The solar radiation intensity in Baghdad for the pure PMMA films and PMMA/ZnO nanocomposites was measured at a rate of 1 hour for seven days in beginning in early September 2021, from 6 A.M to 6 P.M (13 h). As can be observed, almost all films have the same ratio of intensity of transmitted radiation to intensity of sunlight for all hours and days.

KEYWORDS: PMMA polymer; ZnO nanoparticle; physical properties; transmitted sunlight.

الخلاصة

تم تحضير أغشية بولي (ميثيل ميثاكريلات) PMMA و PMMA مطعمة مع جزيئات أكسيد الزنك (ZnONPs) بكميات مختلفة باستخدام تقنية صب المحلول. كميات مختلفة من أكسيد الزنك تتراوح بين (0.001-0.005) غم وتضاف إلى البوليمر بكمية ثابتة (0.5) غم من PMMA في 15 مل تتراهيدروفوران (THF). من المعروف أن زيادة كمية ZnONP في هذه التقنية يساهم في ظهور قمم أكسيد الزنك ZnONP داخل البوليمر. تظهر الاختلافات في طيف XRD أن المادة النانوية قد غيرت البنية المجهرية للبوليمر. تم اكتشاف ذلك باستخدام المجهر الإلكتروني الماسح بالقياس النانوي (SEM). كشفت SEM أن سطح بوليمر PMMA يظهر طبيعة غير متبلورة مع سطح أملس. يوضح التحليل الطيفي لتحويل فوريير للأشعة تحت الحمراء (FTIR) أنه لا يوجد تفاعل كيميائي بين بوليمر PMMA و ZnONPs. تم استخدام القياسات الطيفية للامتصاصية والنفاذية في نطاق الطول الموجي 200-900 نانومتر لتقييم الخصائص البصرية لجميع الأغشية. تحسين الامتصاصية ومعامل الامتصاص ومعامل الخمود لبوليمر PMMA. مع زيادة كمية ZnO، تم تفسير التباين في قيم فجوة الطاقة البصرية المحسوبة، وانخفضت فجوة الطاقة البصرية من 4.65 إلكترون فولت إلى 4.10 إلكترون فولت. تم قياس شدة الإشعاع الشمسي في بغداد لأغشية PMMA النقية ومترابكات PMMA / ZnO النانوية بمعدل ساعة واحدة لمدة سبعة أيام في بداية سبتمبر 2021، من الساعة 6 صباحًا حتى 6 مساءً (13 ساعة). كما يمكن ملاحظته، فإن جميع الأغشية تقريبًا لها نفس نسبة شدة الإشعاع النافذ إلى شدة ضوء الشمس لجميع الساعات والأيام.

INTRODUCTION

New materials emerge as a result of the need to increase the efficiency and performance of structures, and new materials provide new

opportunities to produce updated structures and technology. Composite materials, which are one of the best manifestations of new materials related with the process of developing materials,

structures, and technologies [1]. Poly methyl methacrylate is an amorphous thermoplastic that can be delivered as clear as glass. The material is polymerization of methyl methacrylate according to the chemical structure as showing in Figure 1 [2], [3] Poly methyl methacrylate (PMMA) has found extensive applications in polymer industry as alternative polycarbonate. PMMA possesses versatile properties such as high impact strength, low ductility, high thermal stability and transparency in visible light [3–5]. PMMA as a polymeric waveguide has attracted much attention for use as optical components and in optoelectronics devices due to their low cost and volume productivity. Recently, some researchers reported optical components such as an optical switch, a coupler, a splitter, and a transceiver [6-8]. PMMA is considered as an excellent host material for doping due to their good transparency (the transmission for visible light is very high), resistivity, mechanical strength, and optical homogeneity which can play an important role in building up advanced optical materials [9].

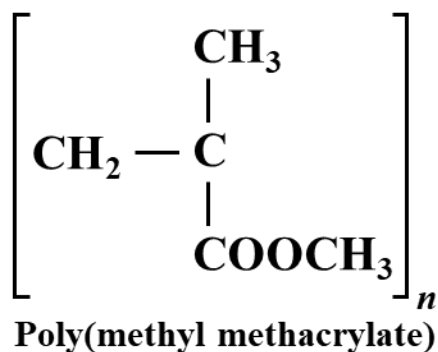


Figure 1. Structure of Poly methyl methacrylate [2].

ZnONPs have intriguing features for a variety of applications, including blue light emitting devices (LEDs), photo catalysis, and solar cells. Figure 2 depicts the structure of ZnO. Zinc Oxide represents semiconductor it is a metallic compound with energy gap about 3.4 eV in the near UV and crystallizes preferentially in the hexagonal wurtzite type structure [10]. Khalid et al. investigated the optical characteristics of Poly(methyl methacrylate) and Chromium chloride PMMA/CrCl₂ composites using samples manufactured with various CrCl₂ concentrations and thicknesses [11]. When the number of CrCl₂ particles inside the polymer rose, the optical constants increased and the forbidden energy gap reduced.

E. Soleimani and R. Moghaddami investigated the optical properties of PMMA with copper oxide (CuO) nanocomposites by casting method for preparing these nanocomposites with different concentrations of CuO and discovered that the absorption coefficient, extinction coefficient, refractive index, and real and imaginary dielectric constants of (PMMA/CuO) nanocomposites increased with increasing concentrations of copper oxide nano particles [12].

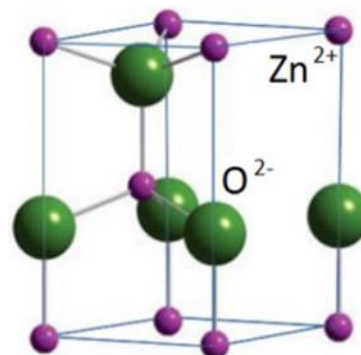


Figure 2. Shape of ZnONPs [10].

With increasing copper oxide nanoparticle concentrations, the energy band gap of (PMMA/CuO) nanocomposites reduced. R. Balen et al. created films and fibers of a zinc oxides/poly methyl methacrylate nano composite to investigate its structural and optical properties. They discovered that near 370 nm, the absorption was high and broad, implying that the optical characteristics of PMMA enhanced as a result of the addition of ZnO nanoparticles within the composite [13]. M. K. Poddara, et al. were characterized physical properties and structural morphology for nanocomposites using standard techniques such as XRD, FTIR, particle size analysis, UV–Visible spectroscopy, electrical conductivity [14]. M. I. Mohammed, et al. studied of a new solution casting method for producing zinc oxide/poly(methyl methacrylate) (ZnO/PMMA) nanoparticles with different content zinc oxide nanoparticles. It has been proved that ZnO/PMMA is a promising material in electronic, optoelectronic, and photocatalytic applications [15]. According to our knowledge, few studies include investigations for measure the transmission of PMMA/ ZnO nanocomposite films under sunlight and studied their structural and optical properties. So that, the aim of this work is to improve the physical properties of PMMA polymer by filling matrix with ZnONPs

and measure the transmission of these films under sunlight by simple and low cost.

THEORETICAL PART

Beer-Lambert's law explains the relationship between absorption and the properties of the absorbing medium. Therefore, it states that the intensity of light passing through a sample decreases according to its concentration and thickness within a specified wavelength. The absorption coefficient may be described as the ratio of the reduction in radiation energy flow to the unit of distance towards wave propagation in the material, which varies depending on the kind of absorbent material and incoming wave wavelength [16]. The absorption coefficient can be computed using Lambert's law, which asserts that the absorbance of a material sample is proportional to its thickness (path length) (t), and is stated as:

$$\frac{I}{I_0} = e^{-\alpha t} \quad (1)$$

Where I/I_0 the ratio of the incident wave's transmittance intensity to the incident wave's incidence intensity, which represents the transmittance of the electromagnetic wave (T), and since the absorbance (A) is represented as:

$A = \text{Log}\left(\frac{I_0}{I}\right)$ The absorption coefficient (α) can thus be expressed mathematically as [17]:

$$\alpha = \frac{2.303}{t} A \quad (2)$$

Where: α is the absorption coefficient. and the thin film transmittance (T) is given by the relationship:

$$1 = A + R + T \quad (3)$$

Wherever: R: Reflectance and A: Absorbance.

The equation which associates the absorption coefficient with energy gap is [18].

$$\alpha h\nu = B(h\nu - E_g)^r \quad (4)$$

Where E_g : the energy gap, B: constant depending on the nature of the material, $h\nu$: the energy of the incident photon, r: a constant whose value is determined by the presence of the transformation, with $r=(1/2, 3/2)$ for allowed and forbidden direct transitions, respectively, and

$r=(2,3)$ for allowed and forbidden indirect transitions, respectively.

The extinction coefficient is the amount of attenuation of an electromagnetic wave going through a material, and its value relies on the density of free electrons in the material as well as the structure's nature. It may be computed using the equation below [19].

$$K = \frac{\lambda\alpha}{4\pi} \quad (5)$$

Using the Debye-Scherrer equation, the average Crystalline size of ZnO nanoparticles was calculated from equation below.

$$D = 0.94\lambda/\beta \cos\theta \quad (6)$$

Where, λ is the wavelength, β is the full width at half the maximum intensity (FWHM) in radians, and θ is the diffraction Bragg angle.

EXPERIMENTAL PART

Poly methyl methacrylate (PMMA) is a powder supplied by Didactic Barcelona Espana, its chemical formula $(C_5O_2H_8)_n$ and molecular weight 200000 g/mol . Zinc oxide (ZnO) NPs supplied from China. This work used a solution casting method to prepare pure PMMA and PMMA/ZnO nanocomposite films. As a solvent, Tetrahydrofuran (THF) with a purity of 99.8% was obtained from (LAB-SCAN, Poland). By dissolving 0.5g of polymer in 15ml of THF, pure PMMA film can be made. The solution was then thoroughly stirred with a magnetic stirrer until a homogenous mixture was achieved. Pour this mixture into a clean glass petri dish (diameter 6 cm) and dry for 24 hours to obtain a film. The amounts (0.001, 0.002, 0.003, 0.004, and 0.005g) from ZnONPs were added to PMMA polymer in order to obtain PMMA / ZnO nanocomposite films. The thickness of the as-prepared films was (156-161) μm , and the measuring accuracy of the measurement range (0-150) mm was (0.001) mm using an electronic Digital micrometer (Tesda) made in Japan. The amounts of light reaching the earth's surface changes according to season, time of day, location, and other factors. As a result, the location of this work was set in Baghdad. Table 1 shows the temperatures for seven days in a row (1).

Table 1. The temperatures for seven consecutive days.

Time (o'clock)	Saturday 1/9/2021	Sunday 2/9/2021	Monday 3/9/2021	Tuesday 4/9/2021	Wednesday 5/9/2021	Thursday 6/9/2021	Friday 7/9/2021
	Temperature (°C)						
6 am	27	26	27	26	25	24	24
7 am	29	28	28	29	27	26	27
8 am	30	29	30	31	30	29	29
9 am	32	31	32	33	32	31	32
10 am	34	33	34	35	36	34	34
11 am	36	36	37	36	37	36	36
12 am	38	38	39	38	38	38	38
1 pm	39	39	39	39	39	38	39
2 pm	39	39	39	39	39	39	39
3 pm	39	39	39	39	39	39	39
4 pm	39	38	38	39	39	39	38
5 pm	38	37	38	38	38	37	37
6 pm	37	36	37	36	36	36	36

From 1 September 2021 to 7 September 2021, the illuminance levels can be tested using a (Auto Digital Luxmeter) made in China. The transmitted intensity of solar radiation measured from 6 O'clock (A.M.) to 6 O'clock (P.M.). The transmitted intensity of Sunlight through the films denoted by (symbol I_{sun}) and the illuminance of Sunlight denoted by (symbol I_0). The optical properties of all samples are assessed using a UV spectrophotometer (T80 Series

UV/VIS spectrometer), which measures ultraviolet and visible light in the (200-1100) nm range. All films were subjected to Fourier transformation infrared spectroscopy (FTIR) using a (Bruker-Tensor 27 with ATR unit). In transmission mode, the wavenumber range is (400-4000 cm^{-1}). The structure of PMMA polymer and PMMA/ZnO nanocomposite films was studied using an X-ray diffractometer (XRD; X'Pert PRO, PANalytical, the Netherlands). The surface composition of all films was characterized using high-resolution scanning electron microscopy (ZEISS SIGMA VP Field Emission Scanning Electron).

RESULTS AND DISCUSSION

XRD Analysis

Figure 3A-D illustrates an XRD analysis of ZnONPs, pure PMMA polymer, and PMMA/ZnO nanocomposite films, respectively to explore the crystalline structure and phase purity of the material. ZnONPs have a diffraction pattern with several peaks corresponding to reflection planes the peaks at 2θ of 31.8° , 34.48° , 36.32° , 47.6° , 56.65° , 62.9° , 66.4° , 67.9° , 69.1° corresponding to the (100) (002) (101) (102) (110) (103) (200) (112), (201), (004) and (202) and the average crystallite size is 16.54 nm was obtained from equation (6), as indicated in table 2.

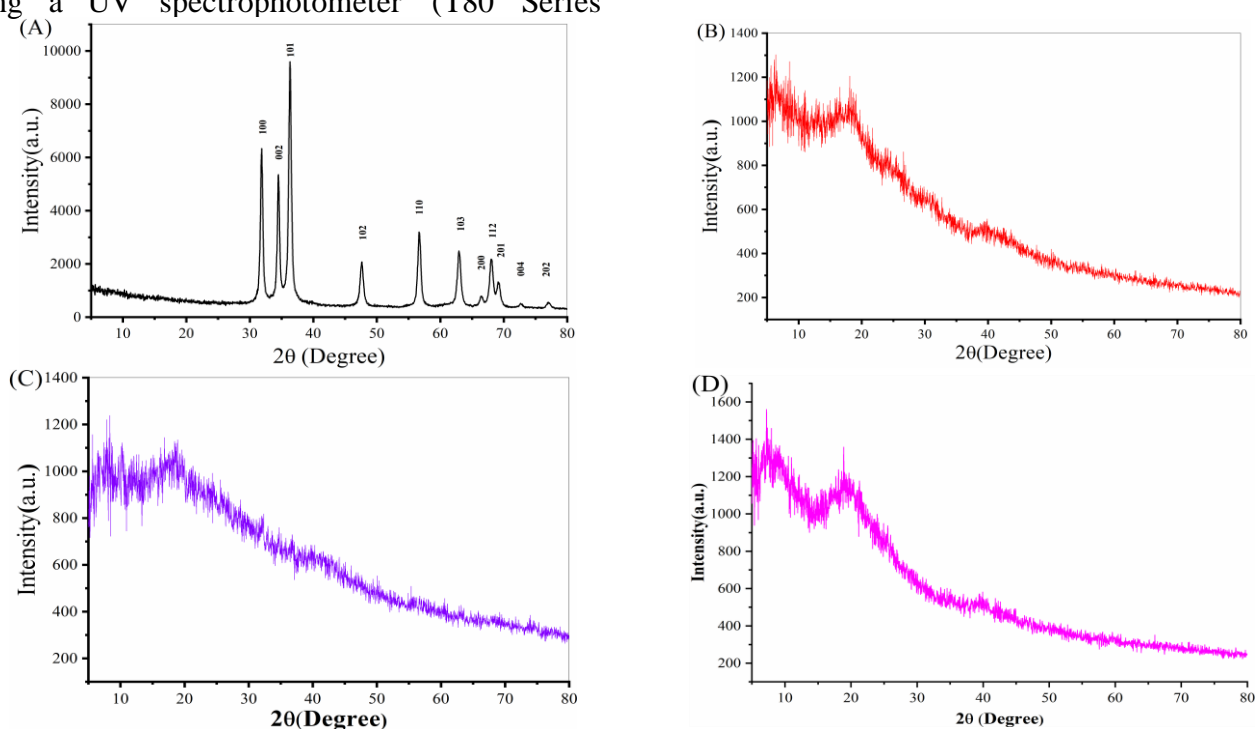


Figure 3. X-ray diffraction spectra of the A) ZnONPs, B) pure PMMA polymer, C) PMMA/ZnO (0.001g), D) PMMA/ZnO (0.003g).

Table 2. Analysis of XRD and the assignments of various reflections of ZnONPs.

2θ°	FWHM (deg)	Intensity (counts)	d(A°)	(hkl)	D (nm)
31.83	0.456	62.65	2.80911	(100)	18.12
34.48	0.40	52.52	2.59870	(002)	20.79
36.31	0.468	100.00	2.47274	(101)	17.84
47.59	0.57	19.14	1.90930	(102)	15.23
56.64	0.51	32.56	1.62385	(110)	17.70
62.93	0.64	24.44	1.47537	(103)	14.55
66.43	0.56	30.92	1.40614	(200)	16.95
67.92	0.59	22.68	1.3770	(112)	16.23
69.12	0.70	23.34	1.3581	(201)	13.69
72.74	0.60	10.60	1.3004	(004)	16.40
76.96	0.70	20.48	1.237	(202)	14.50

At both angular direction and strength, these results correspond with the International Center of Diffraction Data (ICDD) card number (01-089-0510) shows that ZnONPs have a hexagonal crystalline structure that agree with research[20]. The small peaks detected for the pure PMMA films suggest that they were fully amorphous in nature, increasing the amount of ZnONPs in this technique contributes to the appearance of ZnONP peaks within the polymer PMMA. From Figure 3D, the XRD pattern of was shown PMMA doped with ZnONPs the

result shows four high and clear peaks in the polymer (as a result of increasing the amount of nanomaterial) at $2\theta = 39.781$ which indicate ZnONPs.

SEM Analysis

Figure 4 A-C shows typical SEM images of (PMMA, ZnO, and PMMA/ZnO) nanocomposites films, respectively. Figure 4A shows SEM revealed that the PMMA polymer surface exhibit an amorphous nature with a smooth surface containing particles of different shapes. Figure 4B shows the morphology of ZnONPs, which shows an aggregation with homogeneous size and a multidimensional structure. This corresponded to planes generated by its hexagonal structure, as revealed by XRD pattern data. As the ZnO level varied from (0.001 to 0.003 g) Figure 4C and Figure 4D with more agglomerated Zinc Oxide, white patches of ZnO on the surface were seen, indicating segregation in the polymeric system. As a result, there was an adhesion between the PMMA polymer and the ZnO nanoparticles [21].

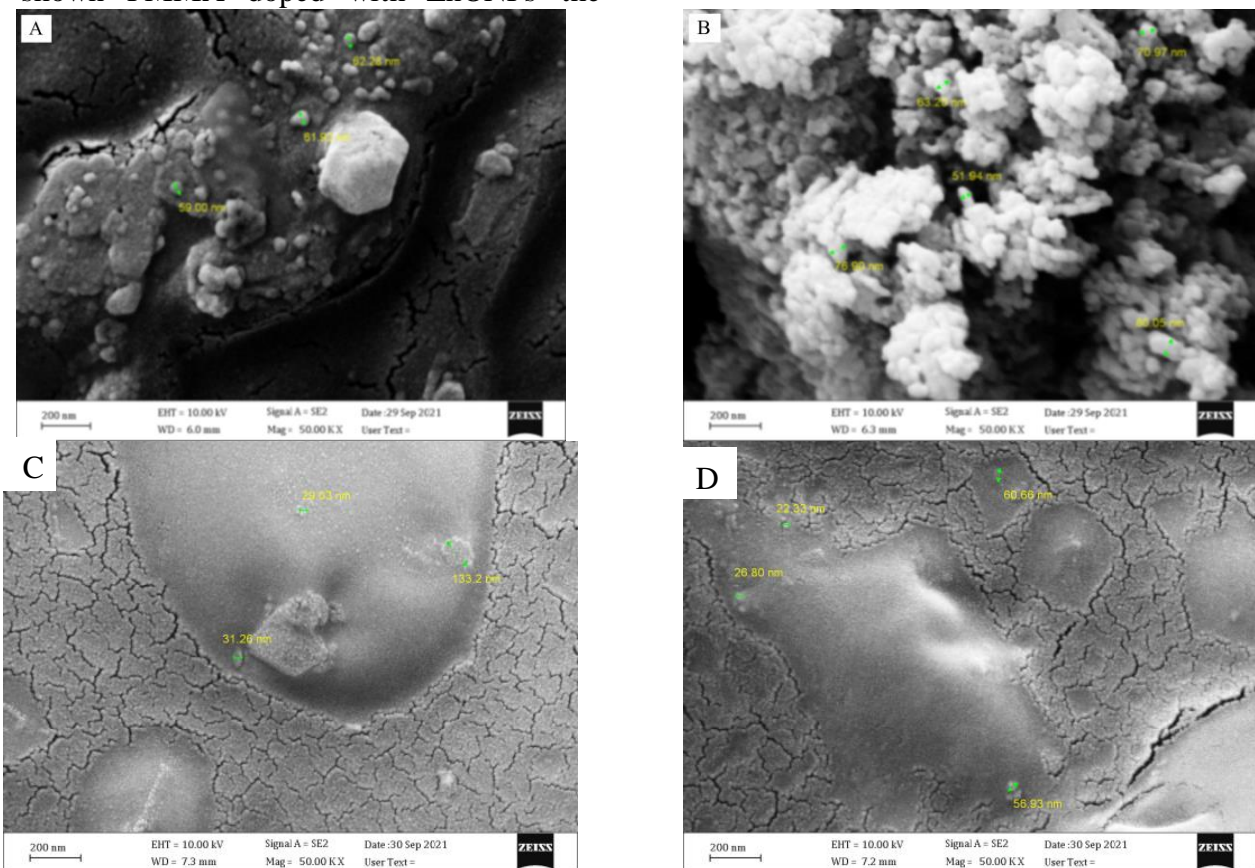


Figure 4. SEM images for A) pure PMMA, B) pure ZnONPs, C) PMMA/ZnO (0.001g), D) PMMA/ZnO (0.003g).

FTIR Spectra

Figure 5A shows the FTIR result of ZnONPs, which indicated 664.51cm^{-1} for Zn-O and 905.32cm^{-1} for Zn-OH. Zn-OH is also represented by the band at 1007.41cm^{-1} . CO_2 was represented by the peaks at 2139.89cm^{-1} this is supported by research [22]. The FTIR spectra of PMMA are shown in Figure 5B. The bands (2993.63 and 2949.81cm^{-1}) correspond to C-H symmetric and asymmetric stretching peaks. The peaks attributed to the C-H stretching vibrations occur from the methyl carbonyl and the chain methyl pendent groups as well as due to the main chain methylene (CH_2) groups. An intense band at 1722.21cm^{-1} represents the carbonyl $\text{C}=\text{O}$ stretching vibration of PMMA, that is found in agreement with the earlier reported results[23]. Several intense bands are observed lower than 1500cm^{-1} . Mainly, this region contains strong transmission features as a result of various (C-H) deformation modes of methyl subunit between ~ 1481.90 to 1306.23cm^{-1} .

The (C-C-O) stretching vibrations of the methyl carbonyl group are perceived between 1269.25 and 1142.98cm^{-1} , whereas (C-O-C) vibrations are established between 1189.70 and 1142.98cm^{-1} . The transmission features below 1000cm^{-1} has been apportioned as a result of C-H rocking modes and C-C skeletal mode [24]. When ZnO is added to the PMMA matrix, the charts exhibit the same behavior as seen in figure 5C. Except for slight changes in the intensities of some transmission peaks, the bonds oscillate at the same wave number regions, which may be due to wagging oscillations in the covalent bonds of the functional groups of PMMA. This demonstrates that no chemical reaction occurred between the PMMA and ZnO, resulting in a composite with the PMMA serving as the matrix and the ZnO serving as reinforcing additives, this is agreed with researcher[25].

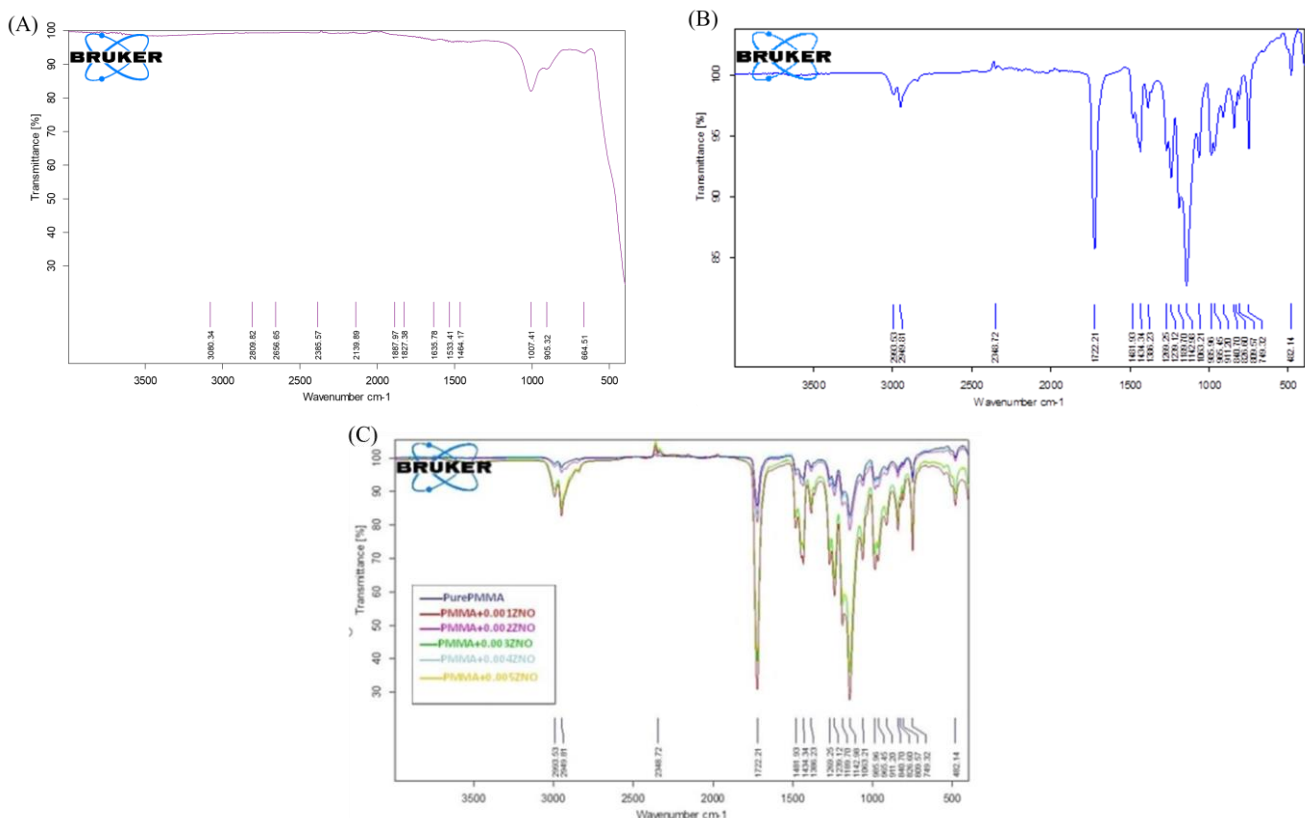


Figure 5. FTIR Spectra for A) Pure ZnO, B) Pure PMMA film, C) PMMA/ZnO with different amounts of ZnONPs.

OPTICAL PROPERTIES

Figures 6 A, B show the absorption and transmission spectra of pure PMMA and PMMA/ZnO nanocomposites films. The PMMA polymer has high transmission above $\sim 400\text{nm}$ wavelength, that is to be expected given as

PMMA has numerous uses as an optically transparent window material. As-prepared sample has a high absorption edge (reduction in transmission) below 400nm . The electronic stimulation of the carbonyl chromophores ($\text{C}=\text{O}$) found in the PMMA structure results in the

formation of this high absorption edge Figure 6A. It is revealed that the $\pi \rightarrow \pi^*$ transition in the C=O system that's what causes the most powerful absorption band seen in the spectra between 250 and 280 nm. There is an extra absorption band perceived at 360 nm. However, this peak offers an enhanced saturated absorption peak from 286 to 362 nm which may be due to an increased number density of PMMA molecules in the increased absorption path length. These peaks are attributed to the forbidden $n \rightarrow \pi^*$ transition that are less intense in comparison to $\pi \rightarrow \pi^*$ transition. These findings are consistent with the preceding findings [24]. The highest absorption

wavelength of ZnONPs is (370nm), which is comparable to intrinsic band-gap absorption in ZnO, that according to studies[26]. The presence of ZnONPs leads to -an increase in the absorbance and to a decrease in the transmittance as ZnONPs amounts increases in the region below 400 nm. The excitation of electrons from the valence band to the conduction band of ZnO causes UV absorption in nanocomposites. The UV shielding capability of nanocomposites can be easily adjusted by modifying the ZnO content in the PMMA matrix, while transparency in the visible region can be maintained since ZnO particles are nanometer scale and well dispersed in the PMMA matrix, according to the findings.

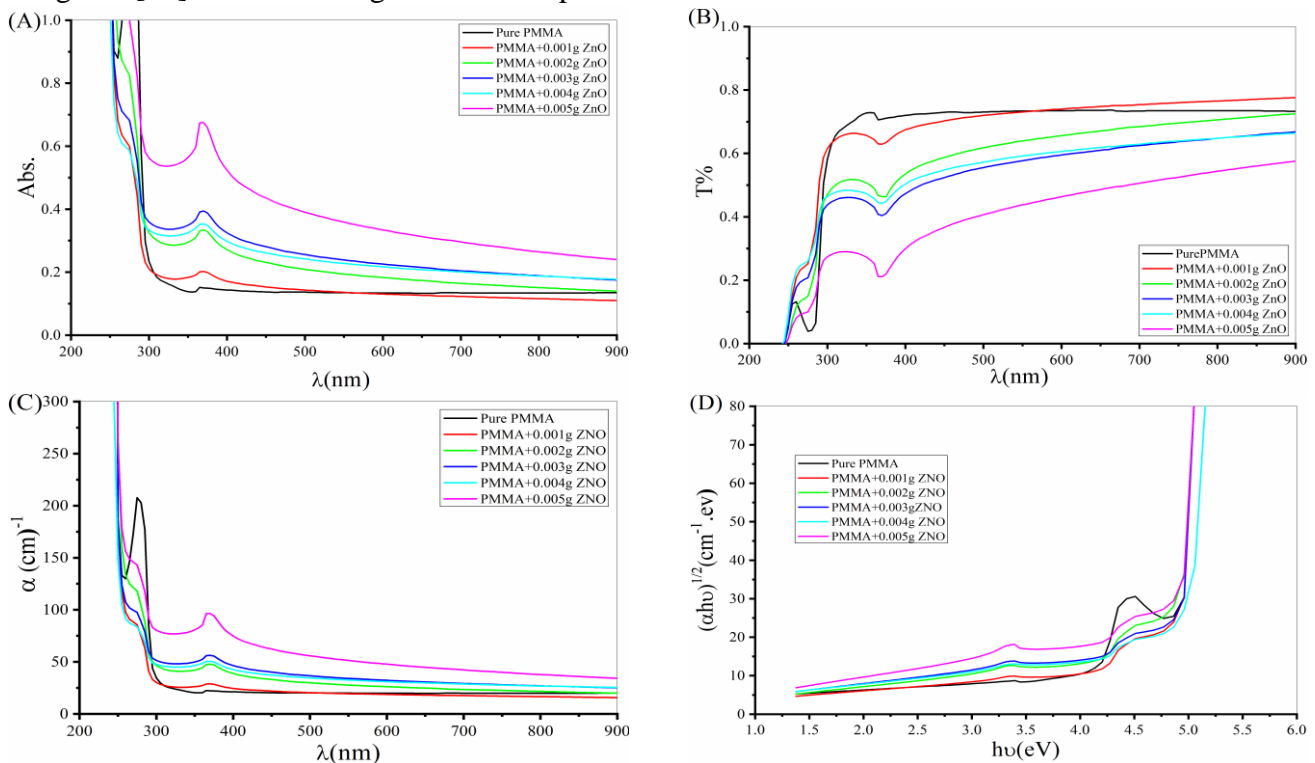


Figure 6. A) Absorption spectrum for PMMA/ZnO nanocomposite, B) Transmittance variation with wavelength to PMMA/ZnO nanocomposite, C) Absorption coefficient (α) variation with wavelength to PMMA/ZnO nanocomposites, D) $(\alpha h\nu)^{1/2}$ Variation with photon energy to PMMA/ZnO. Nanocomposites.

These optical qualities could allow nanocomposites to be used in optical fields[27]. The transmittance of pure PMMA polymer film is higher than that of PMMA/ZnO nanocomposite films. Pure polymers and nanocomposites films have a lower transmittance in the UV region than in the visible range. The effect of appended ZnO nanoparticles is evident; they have a very low UV transmittance, (high absorbance and low transmittance) Figure 6B these findings are

consistent with those of other studies[28]. Additionally, the measurement of the absorption coefficient (α), which characterizes each absorber molecule or ion, was done to further analyze the optical characteristics of PMMA/ZnO nanocomposites films. It describes a substance's ability to absorb light with a particular wavelength per unit length. The absorption coefficient can be calculated using Equation (2).

The values of the absorption coefficient, as shown in Figure 6C, were less than (10^4 cm^{-1}), indicating that the indirect electronic transition was investigated [29]. From the figure, it is clear that for both the PMMA and the PMMA/ZnO nanocomposites, the absorption coefficient decreases at high wavelength regions due to the low energy of the incident photon (hf), which is insufficient to cause electron transitions from the valence band to the conduction band, but increases at low wavelength. Another finding revealed the value of the absorption coefficient; these results agree with [30]. Moreover, Figure 6D shows the optical energy band gap (E_g) for as-prepared films. The value of energy band gap decreased as the number of nanoparticles increases, as in Table 3.

Table 3. Energy band gap value to PMMA/ZnO nanocomposites.

Samples	E_g (eV)
Pure PMMA	4.65
PMMA- 0.001g ZnO	4.52
PMMA- 0.002g ZnO	4.41
PMMA- 0.003g ZnO	4.30
PMMA- 0.004g ZnO	4.19
PMMA- 0.005g ZnO	4.10

This may be explained as increases in localized levels of prohibited energy band gap cause decreases in energy band gap value [31]. The extinction coefficient (k) is proportional to the

absorption coefficient (α), and k may be calculated using the equation (5). The extinction coefficient as a function of wavelength is shown in Figure 7. According to a graph, the extinction coefficient increases as the ZnO ratio increases, indicating an increase in the number of charge carriers [32]. The sun radiation intensity transmitted from pure polymer PMMA and PMMA/ZnO nanocomposite films was measured in Figure 8A-F. The transmitted sun radiation intensity in the samples is found to be almost identical or slightly different at times. The chart clearly shows that the largest transmitted sun radiation strength occurs at 12 o'clock, following which the intensity steadily decreases [33].

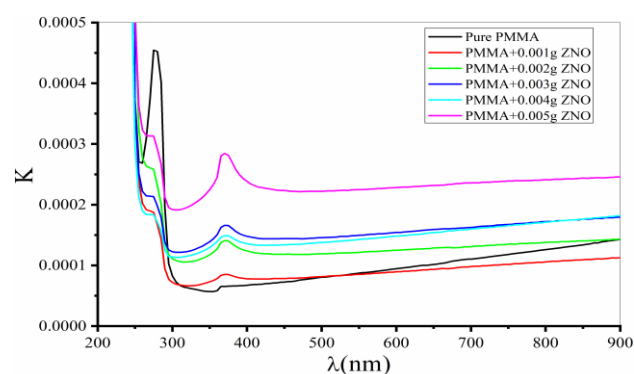


Figure 7. Extinction coefficient variation with wavelength to PMMA/ZnO nanocomposites.

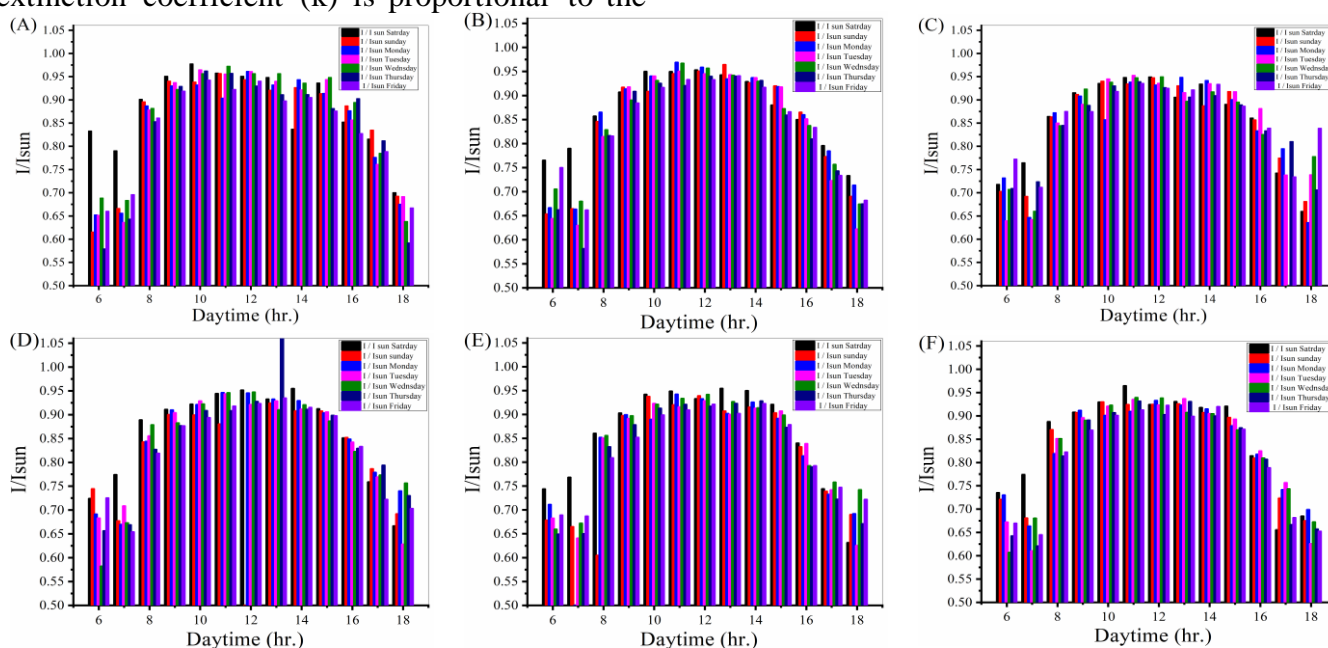


Figure 8. Measurement of radiation intensity before weathering of A) polymer PMMA, B) PMMA/ZnO (0.001g), C) PMMA/ZnO (0.002g), D) PMMA/ZnO (0.003g), E) PMMA/ZnO (0.004g), F) PMMA/ZnO (0.005g).

The temperatures in the seven consecutive days from the beginning of September 2021 are

roughly equal for the time from six o'clock in the morning to six o'clock in the evening, according

to Table 1. The solar radiation intensity in Baghdad for the pure PMMA films and PMMA/ZnO nanocomposites was measured at a rate of 1 hour for seven days in beginning in early September 2021, from 6 in the morning until 6 in the evening (13 h). As can be observed, almost all films have the same ratio of intensity of transmitted radiation to intensity of sunlight for all hours and days[34].

CONCLUSIONS

According to the findings, nanoparticle zinc oxide ZnO can effectively dope PMMA and improve its optical properties. Increasing the amount of ZnONP contributes to the appearance of ZnONP peaks within the polymer. Differences in the XRD spectrum show that nanostructures have altered the microstructure of the polymer and the XRD results also showed the hexagonal crystal structure of ZnONPs. The conclusion was made using nanoscale scanning electron microscopy (SEM). That the surface of the PMMA polymer shows an amorphous nature with a smooth surface. By Fourier Transform Infrared (FTIR) spectroscopy it was shown that there is no chemical reaction between the PMMA polymer and ZnONPs, which leads to the creation of a composite material in which the PMMA polymer acts as a matrix and the ZnONPs acts as an additive to the reinforcement. The presence of ZnO causes an increase in absorbance and a decrease in transmittance as the amount of ZnO increases. The extinction coefficient and absorption coefficient are dependent on ZnO amounts, increasing after doping and with increasing ZnO amounts. The optical band gap decreased from 4.65 eV to 4.10 eV as the filler amount increased. The intensity of solar radiation was measured at a rate of one hour for pure PMMA and PMMA/ZnO nanocomposites films for seven consecutive days, from 6 a.m. to 6 p.m. (13 hr.). It is reasonable to deduce that the ratio of transmitted radiation strength to solar radiation intensity for all films is the same at all times and on all days.

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