ISSN 2347 - 3983

Volume 7, No.8 August 2019

International Journal of Emerging Trends in Engineering Research

Available Online at http://www.warse.org/IJETER/static/pdf/file/ijeter04782019.pdf

https://doi.org/10.30534/ijeter/2019/04782019

Structural, Electronic, Optical Properties and Antibacterial Application of Novel (PMMA-Al_2O_3-Ag) Nanocomposites for Dental Industries Applications

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ABSTRACT

In this paper, the effect of adding Ag nanoparticles on the optimized geometrical parameters, electronic and optical properties of the (PMMA-Al_2O_3) nanostructures for antibacterial application. The electronic properties included electrochemical hardness and electronic softness while the optical properties include absorbance**,** transmittance, absorption coefficient**,** extinction coefficient, refractive index, real and imaginary parts of dielectric constants and optical conductivity. The properties calculated by using Gaussian 0.9 program with Gaussian View 0.5 using density function theory (DFT) with local spin density approximation B3LYP level, 6-31G and STO-3D basis sets. The results showed that the addition of Ag nanoparticles lead to decrease the chemical hardness and increase in the softness. The optical properties for (PMMA-Al_2O_3-Ag) nanocomposites showed that the absorbance**,** absorption coefficient**,** extinction coefficient, refractive index, real and imaginary parts of dielectric constants and optical conductivity of (PMMA-Al_2O_3) nanocomposites increase while the transmittance and energy band gap decrease with increase in Ag nanoparticles concentrations. The results of application showed that the (PMMA-Al_2O_3-Ag) nanocomposites have good antibacterial for positive and negative gram organisms bacteria.

Key words: Antibacterial, PMMA, nanocomposites, silver, optical properties, Al_2O_3.

1. INTRODUCTION

Polymers are a part of everyday life and examples can be found almost anywhere. Many people think of polymers simply as plastics used for packaging, in household objects, and for making fibers, but this is just the tip of the iceberg. Polymers are used in all sorts of applications we might not have thought much about before. Polymers and composites (materials made by combining two or more materials) arevital to modern dentistry. Currently, the acrylic resin polymethylmethacrylate (PMMA) is used almost universally for denture base and orthodontic devices fabrication[1].Discovered and commercialized many years ago, PMMA is one of the most widely used industrial polymeric materials and still remains an active material for research at the cutting edges of science. Due to its good degree of biocompatibility with human tissue, reliability, dimensional stability, absence of taste, odor, tissue irritation and toxicity, teeth adhesion, insolubility in body fluids, relative ease of manipulation, good aesthetic appearance, and color stability, PMMA based materials are widely used as biomaterials. Nowadays, PMMA finds applications not only in dentistry but also in areas such as transparent glass substitutes, interior design, transparent dielectric films, acrylic paints, and microcellular foams[2-8]. Still, one of the most attractive applications of PMMA based materials is in various biomedical applications such as intraocular lenses that are implanted in the eye when the original lens is removed in treatment of eye opacity, in particular acrylic contact lenses are useful for patients with recurrent eye inflammation (inflammation of the iris), as the acrylic material causes less inflammation[9], bone cement in orthopedic surgery, and removable partial denture. Although PMMA denture base materials have a lot of qualities, they are often subject to intense criticism because oftheir inherent drawbacks such as residual monomer toxicity and its effect on the oral tissues, mechanical properties that are not always perfect[10-17], and susceptibility to distortion. The cumulative effect of these properties ma ylead to residual monomer leakage into adjacent oral tissues and the generation of cracks and the other structural damage of denture base that, apart from leading to denture base breakage, can also form a point of entry for various bacteria, yeasts, and moulds. The physical properties of the final polymer are important in the fabrication of polymeric denture bases as the cured polymer should be stiff enough to hold the teeth in occlusion during mastication and to minimize the uneven loading of the mucus. The denture material should not creep under masticatory loads if good occlusion is to be maintained. Not only does the material have to have sufficient strength and resilience to withstand normal masticatory forces but it must also be able to withstand the sudden shock caused by impact forces. The application of PMMA as an ideal denture base material is still restricted by a few limitations. One of them is the difficulty in achieving intrinsic radio paucity in the material, which is due to the constituent elements of PMMA. The nanocomposites applications are quite promising in the fields of microelectronic packaging, medicine, automobiles, optical integrated circuits, drug delivery, injection molded products, sensors, membranes, aerospace, packaging materials, coatings, fire-retardants, adhesives, consumer goods .. etc. Polymers have been used as insulators in early works because of their dielectric properties and high resistivity. Polymer-based insulators are used in electrical devices to separate conductors without passing current through themselves. The insulator applications of polymers include corrosion protective electronic devices, printed circuit boards and cable sheathing materials. Polymers have several advantages, such as low cost, easy processing, flexibility, good mechanical properties and high strength. In the microelectronic fabrication industry, it are used in the photolithography process. Polymeric nanocomposites consisting of inorganic nanoparticles and organic polymers represent a class of materials that have motivated considerable interest in recent years [18-26]. The aims of this study, the effect of addition silver nanoparticles on electronic, optical properties of (PMMA-Al_2O_3) nanocomposites for antibacterial applications

2. THEORETICAL PART

The density functional theory (DFT) provides the ground state properties of a system, and the electron density plays a key role. It is presently the most successful approach to compute the electronic structure of matter. Its applicability ranges from atoms, molecules and solids to nuclei. It predicts a great variety of molecular properties, molecular structures, vibrational frequencies, atomization energies, ionization energies, electric, magnetic properties and reaction paths, etc.

The fundamental concepts of DFT rely on the ground state energy and all other ground state electronic properties are uniquely determined by the electron density. DFT is today one of the most important tools for calculating the ground state properties of metals, semiconductors, and insulators[26].The chemical hardness (H) is a measure of the resistance to charge transfer. The theoretical definition of chemical hardness has been provided by the density functional theory as the second derivative of electronic energy with respect to the number of electrons N, for a constant external potential V (r) [27,28]:

$$H = \frac{1}{2}\left[\frac{\partial^2 E}{\partial N^2}\right]_V = \frac{1}{2}\left[\frac{\partial \mu}{\partial N}\right]_V = -\frac{1}{2}\left[\frac{\partial X}{\partial N}\right]_V \text{....……… (1)}$$

Finite difference approximation to chemical hardness gives:

$$H = \frac{IP-EA}{2} \text{……….(2)}$$

The global chemical softness, S, is a property of molecules that measures the extent of chemical reactivity. It is the inverse of the chemical hardness H[29]:

$$S = \frac{1}{2H} = \left[\frac{\partial^2 N}{\partial E^2}\right]_V = \left[\frac{\partial N}{\partial \mu}\right]_V \text{…………(3)}$$

Optical properties of materials are very important due to it can obtain information about the internal structure ,the nature of the bonds and their employment by knowing the amount of absorbance, reflectance and transmittance of these materials[30].

3. RESULT AND DISCUSSION

The geometry of a molecule determines several of its physical and chemical properties. It is necessary to find the relaxation of the molecule, in which the optimized structure of the molecule is the structure at minimum energy. The optimized structures of pure polymethylmethacrylate (PMMA),(PMMA-Al_2O_3) and (PMMA-Al_2O_3-Ag) nanocomposites were initially designed at Gauss View 5.0.8 program. The relaxation of PMMA was done by employing the hybrid functional B3LYP density functional theory (DFT) with basis sets 6- 31G and STO-3D level. Figures (1-3) show the optimized of pure polymethylmthacrylate, (PMMA-Al_2O_3) nanocomposites and (PMMA-Al_2O_3-Ag) nanocomposites. Table 1: includes some electronic properties for pure polymethylmethacrylate (PMMA) and (PMMA-Al_2O_3-Ag) nanocomposites, such as the chemical hardness (H) and chemical softness (S) these properties are calculated in (eV) using Koopman's theorem. Figure 4 and 5 show the chemical hardness and chemical softness of PMMA and (PMMA-Al_2O_3-Ag) nanocomposites correspondingly, the hardness decreases and softness increases due to addition nanoparticles to pure polymethylmethacrylate (PMMA). The decrease of hardness and increased in softness is the main future as a sign for that band gap goes to be rather soft and lowering the resistance of a species to lose an electron. Figures (6-8) show the variation of absorbance for (PMMA-Al_2O_3-Ag) as a function of wavelength. Figure 6 shows the absorption of pure material (PMMA) at UV region while figures 7 and 8 show that the absorption for all samples of nanocomposites at visible region, due to increase number of atoms per volume unit is caused excitement of electrons from valance band to the conduction band , the high absorption of samples for nanocomposites at visible region credited to the energy of photon sufficient to interact with atoms. generally, It has been discovered that the absorbance reduces with increasing wavelength intended for all samples of nanocomposites. This physically means that will an incident photon has been able in order to excite the electron and even transfer it from valence band to the conduction band because of the energy gap value of the semiconductor. Generally typically the absorbance of all samples of nanocomposites have reduced values in the visible and near infrared region. The absorbance will increase when the wavelength reduced, due to the interaction between incident photon and material will occur[30-33]. Also it can be observed increase the absorbance increases with nanoparticles ratio increases, due to decrease the energy gap of all samples of nanocomposite and increase the number of charges carries [34-39]. The optical transmission values decrease along with the increases of Ag nanoparticles. This kind of behavior may be as a result of the increase in free electrons with the addition of Al_2O_3 and Ag nanoparticles[40], as shown in figures(9-11). The figures show, that absorption coefficient of most samples for (PMMA-Al_2O_3-Ag) nanocomposites is large at high energies. Figure(12-14) indicate the variation of absorption coefficient for (PMMA-Al_2O_3-Ag) nanocomposites as a function of photon energy of the incident light correspondingly, the energy of incident photon will be sufficient to transit typically the electron from the donor levels to the accepter levels which as the consequence the energy of the incident photon is larger than the energy band gap, due to the electron transition has high probability. The absorption coefficient contributions in order to know the nature involving electron transition . When the values in the absorption coefficient of material $\alpha > 10^4$ cm^{-1}, the electron transmission is probable to be direct transition of electron but the electron transmission will be probable to be indirect transition if the values of the absorption coefficient of material are usually minimal $\alpha < 10^4$ cm^{-1}; the transition of electron is indirect. The energies gaps for allowed indirect transitions of pure (PMMA), (PMMA-Al_2O_3) and (PMMA-Al_2O_3-Ag) nanocomposites are displayed in figures (15-17). The energy gaps for forbidden indirect transitions of pure (PMMA), (PMMA-Al_2O_3) and (PMMA-Al_2O_3-Ag) nanocomposites are shown in figures (18-20). As is shown in the figures, the energies gaps for allowed and forbidden indirect transitions of nanocomposites are decreased with the increasing of the Ag nanoparticles, this performance is due to the making of levels in the energy gap; the transition of electron in this instance is conducted in two stages that include the transition from the valence band to the local levels in energy gap and to the conduction band as a result of increasing the Ag nanoparticles, the electronic conduction depends on addition of nanoparticles[41-44]. Figures(21-23) show the variation of extinction coefficient (k) with wavelength for the (PMMA-Al_2O_3-Ag) nanocomposites , in general, it will be clear that the extinction coefficient (k) decreases with increasing of wavelength (λ) for all samples of (PMMA-Al_2O_3-Ag) nanocomposites which is related to increase the absorbance[45-47]. Furthermore the figure shows that will the extinction coefficient rises with increasing of the Ag nanoparticles. This kind of behavior can be acknowledged to the increasing associated with carrier density which approves the increasing within the absorption coefficient with Ag nanoparticles and that lead to increase the extinction coefficient along with addition of nanoparticle (Ag) [48-52]. The extinction coefficient associated with nanocomposites has great values at visible region, this kind of behavior attributed to higher absorbance of all samples of nanocomposites. The refractive index of (PMMA-Al_2O_3-Ag) nanocomposites as a function of wavelength is shown in figures (24-26), the refractive index of nanocomposites increases with the increasing of the Ag nanoparticles. Refractive index is decreased with the increase of the wavelength. This performance ascribed to addition (Ag) nanocomposites which is related to increase the density[53-56].

Figure 27: shows the antibacterial activity application for (PMMA-Al_2O_3-Ag) nanocomposites. The disks of nanocomposites were placed over the media and incubated at 37°C for 24 hours. The figure illustrations that the inhibition zone rises with increase of the silver nanoparticles (Ag) concentration, the microorganisms carry negative charges while attributed to the metal oxides carry the positive charge , due to electromagnetic attraction between microorganisms and the metal oxides which leads to oxidization and finally death of microorganism[57-63].

4. CONCLUTION

Novel (PMMA-Al_2O_3-Ag) nanocomposites for dental industries applications have been synthesized. The results showed that the chemical hardness decreases and the softness increases with increase in Ag nanoparticles concentrations. The optical properties for (PMMA-Al_2O_3-Ag) nanocomposites showed that the absorbance**,** absorption coefficient**,** extinction coefficient, refractive index, real and imaginary parts of dielectric constants and optical conductivity of (PMMA-Al_2O_3) nanocomposites increase while the transmittance and energy band gap decrease with increase in Ag nanoparticles concentrations. The results of application showed that the (PMMA-Al_2O_3-Ag) nanocomposites have good antibacterial for positive and negative gram organisms bacteria.

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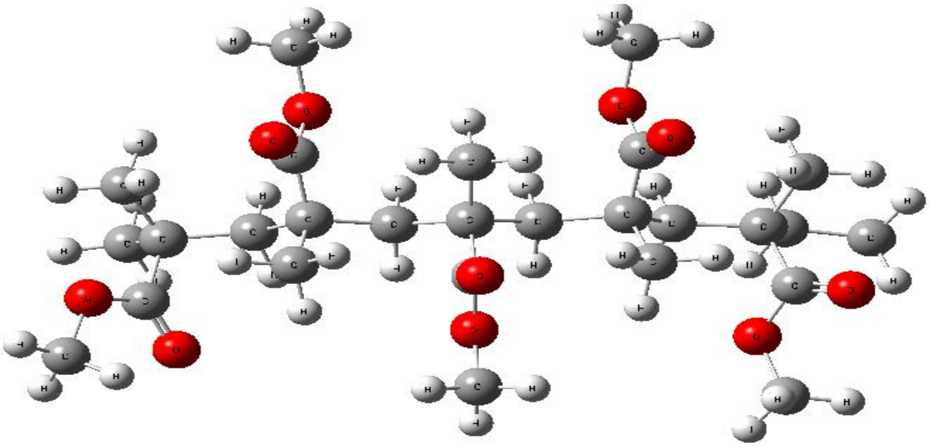

Figure 1: The relax structures of the pure polymethlmethacrylate (PMMA).

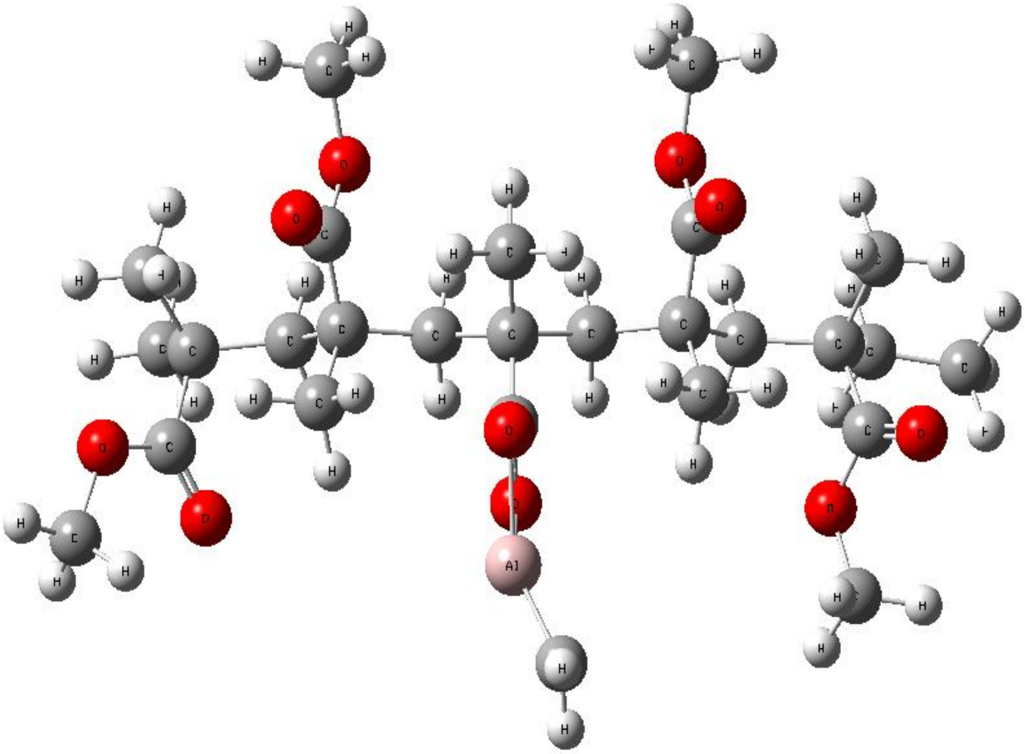

Figure 2: The relax structures of the (PMMA-Al_2O_3) nanocomposites.

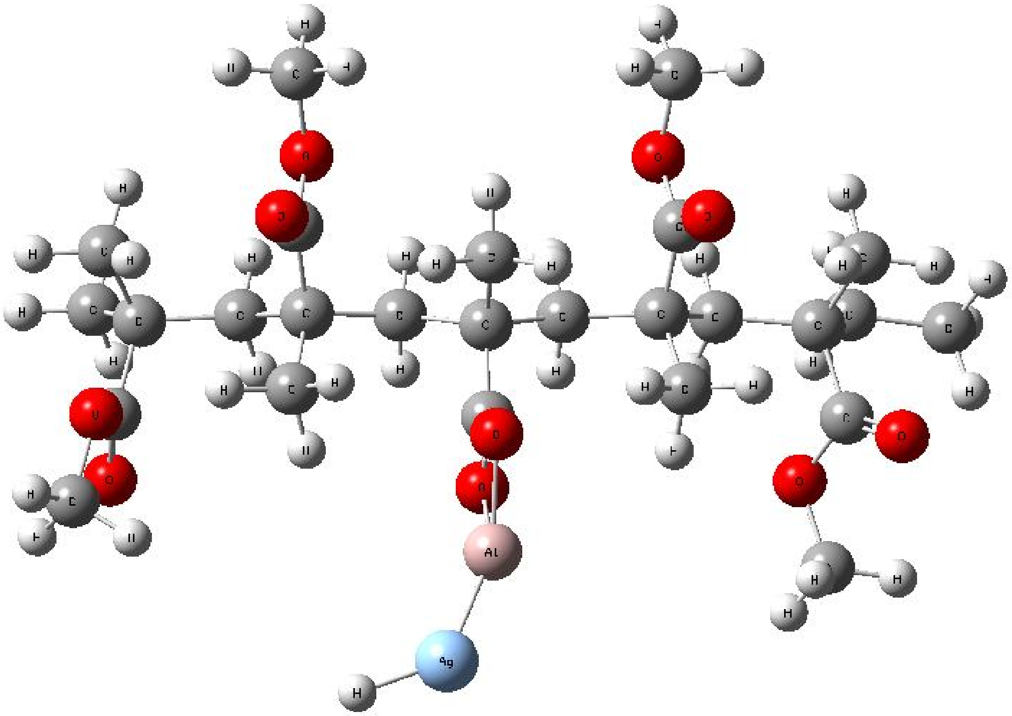

Figure 3: The relax structures of the (PMMA-Al_2O_3-Ag) nanocomposites

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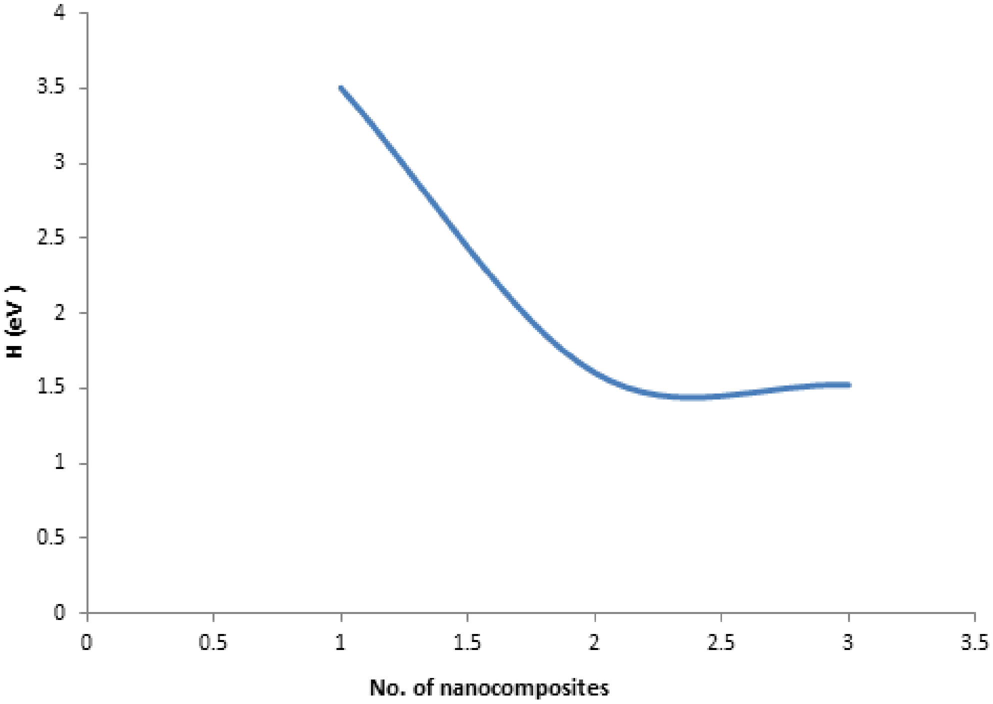

Figure 4: The electrochemical hardness H for pure (PMMA) and nanocomposites.

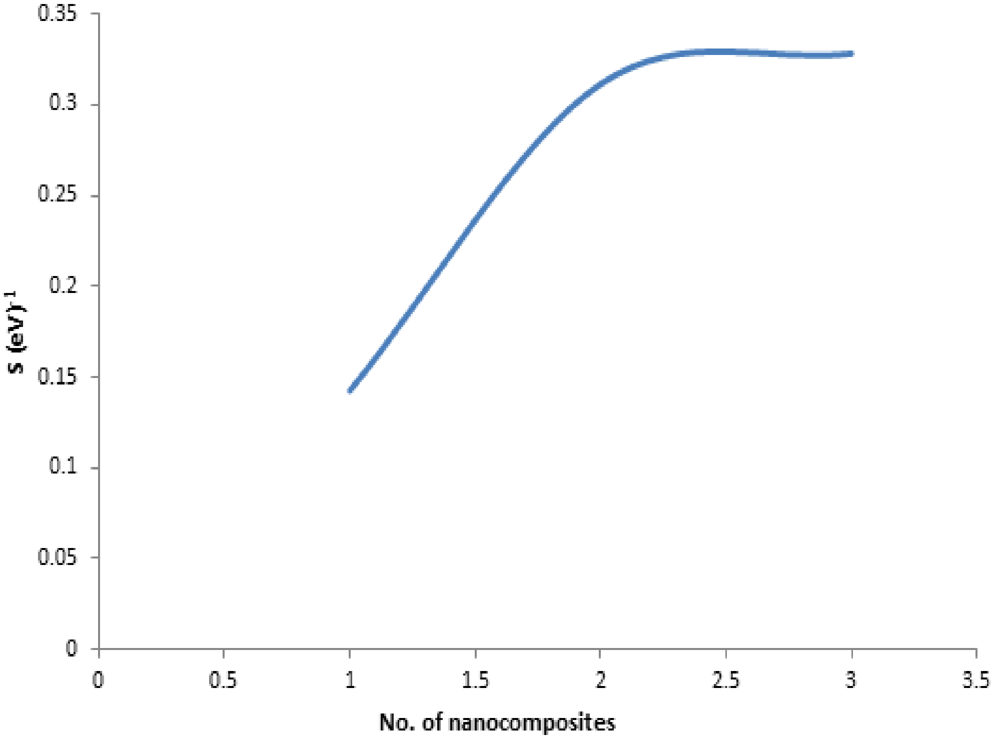

Figure 5: The electronic softness S for PMMA and nanocomposites.

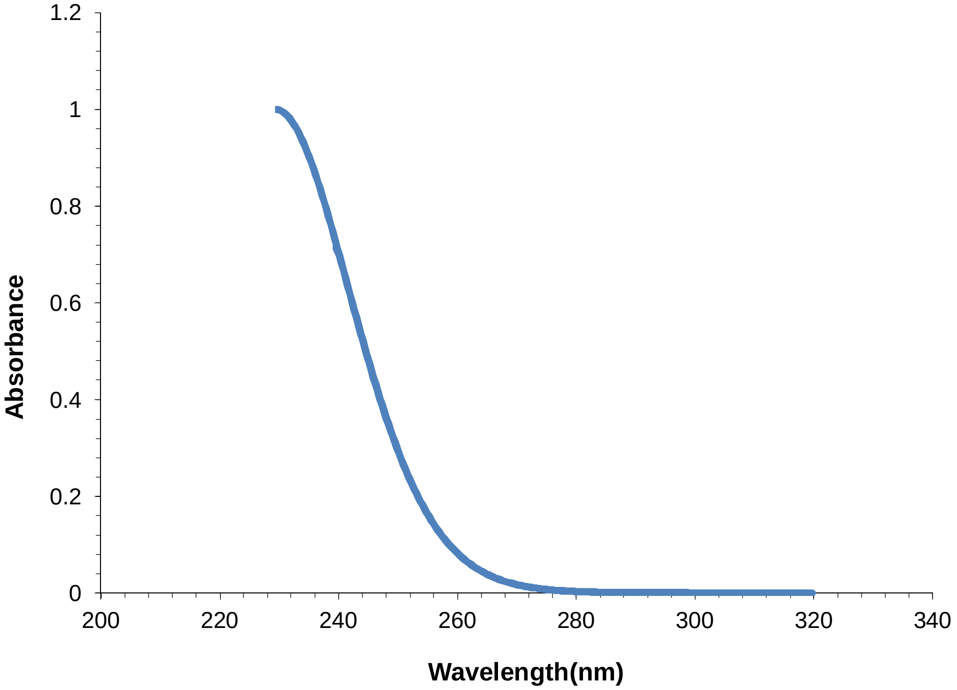

Figure 6: Absorbance as a function of wavelength for pure polymethylmethacrylate.

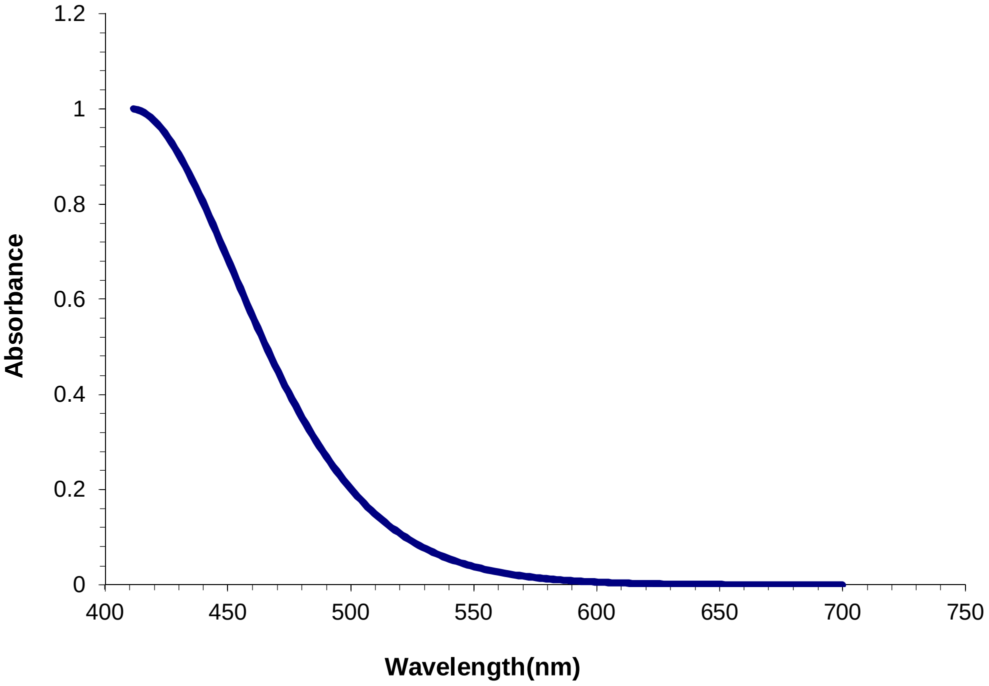

Figure7: Absorbance as a function of wavelength for (PMMA-Al_2O_3) nanocomposites.

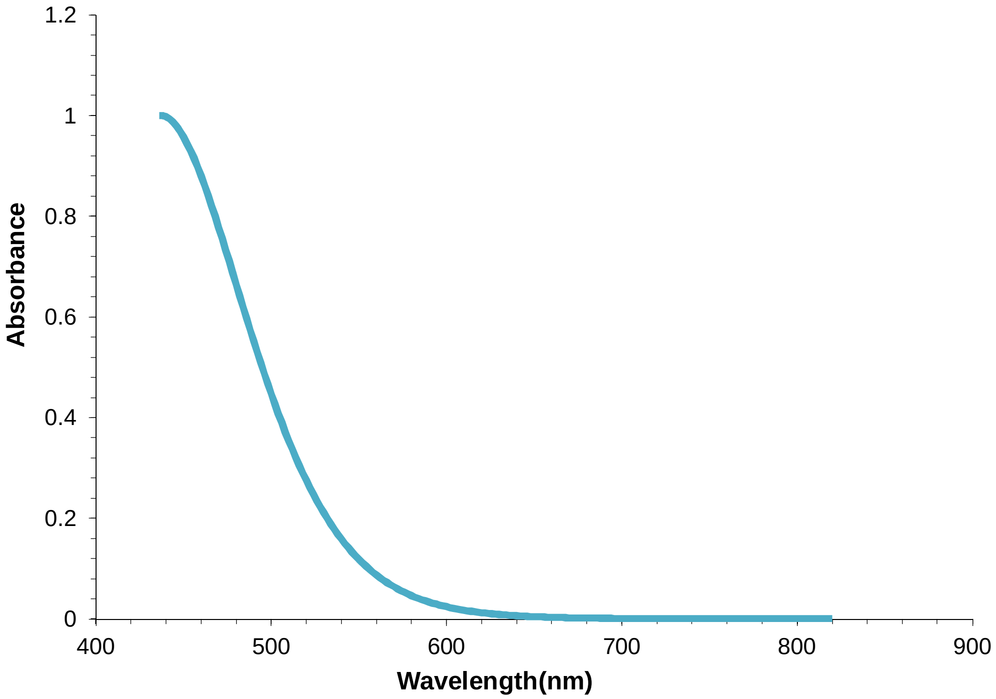

Figure 8: Absorbance as a function of wavelength for (PMMA-Al_2O_3-Ag) nanocomposites.

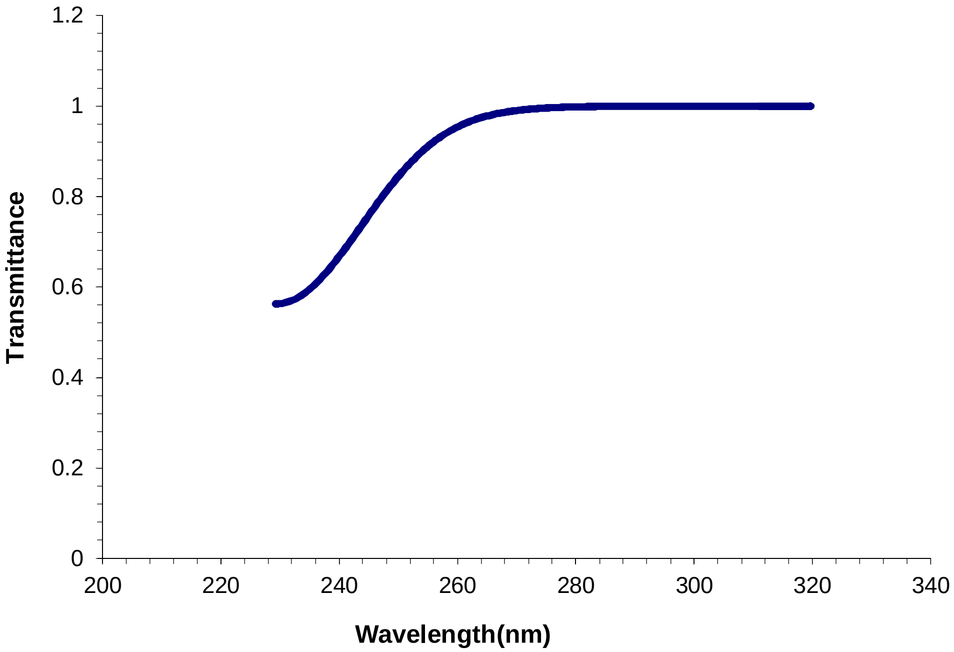

Figure 9: Transmittance as a function of wavelength for pure polymethylmethacrylate.

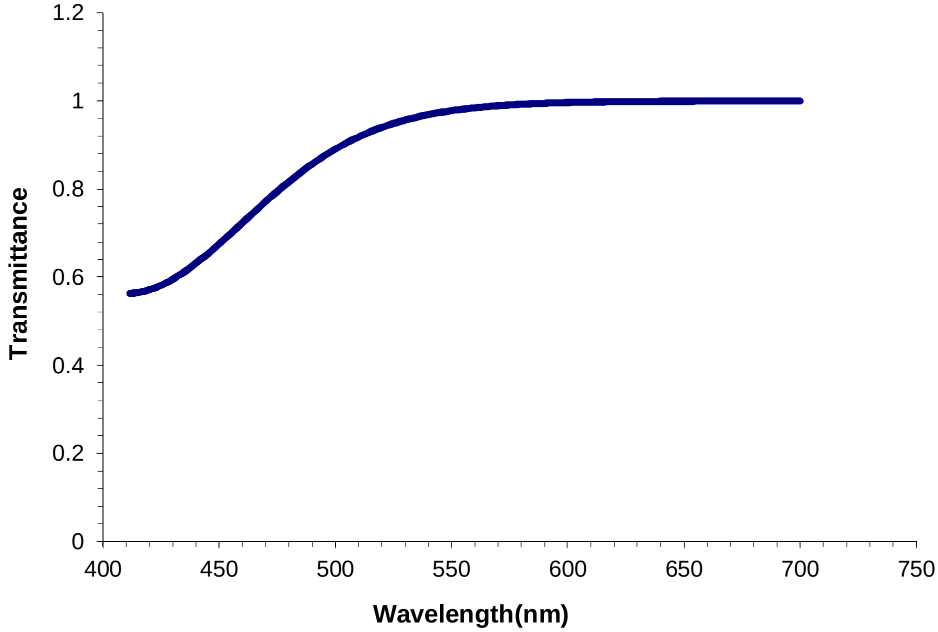

Figure 10: Transmittance as a function of wavelength for (PMMA-Al_2O_3) nanocomposites.

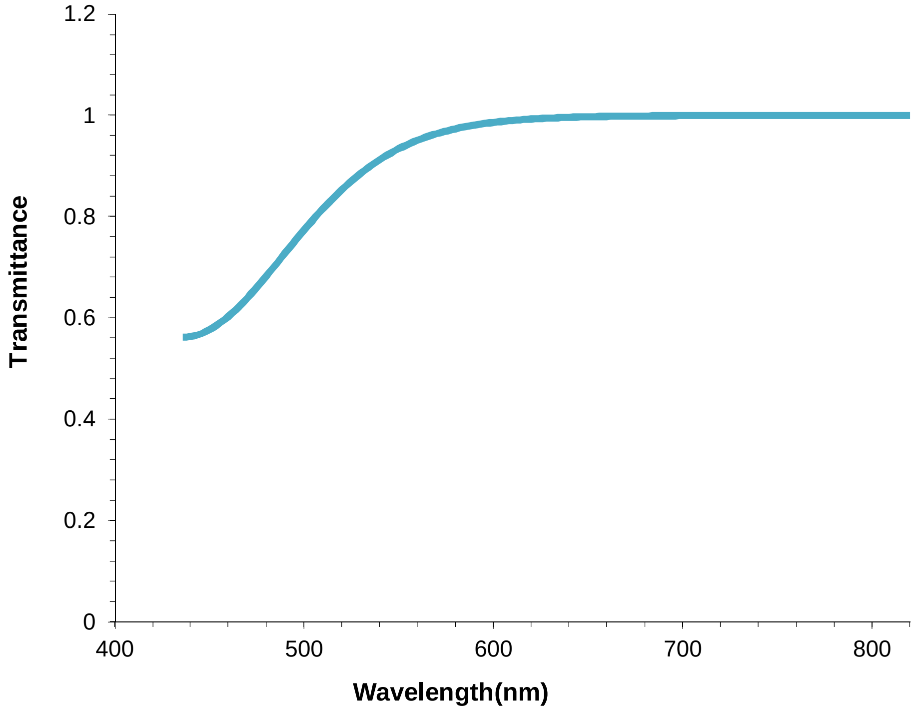

Figure 11:. Transmittance as a function of wavelength for (PMMA-Al_2O_3-Ag) nanocomposites.

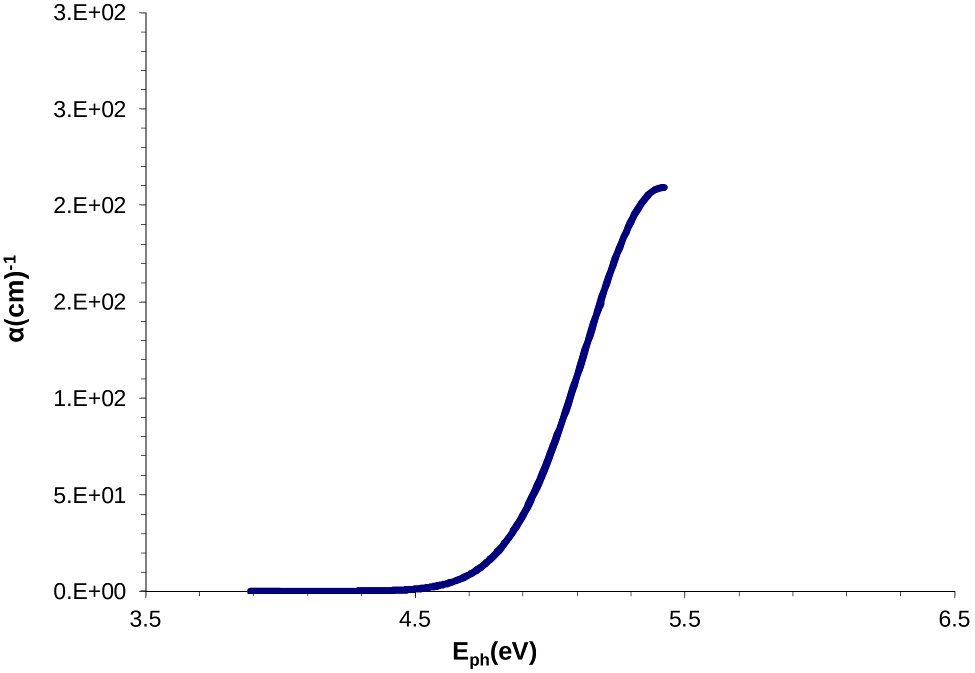

Figure12: Variation of absorption coefficient (α) for polymethylmethacrylatewith photon energy.

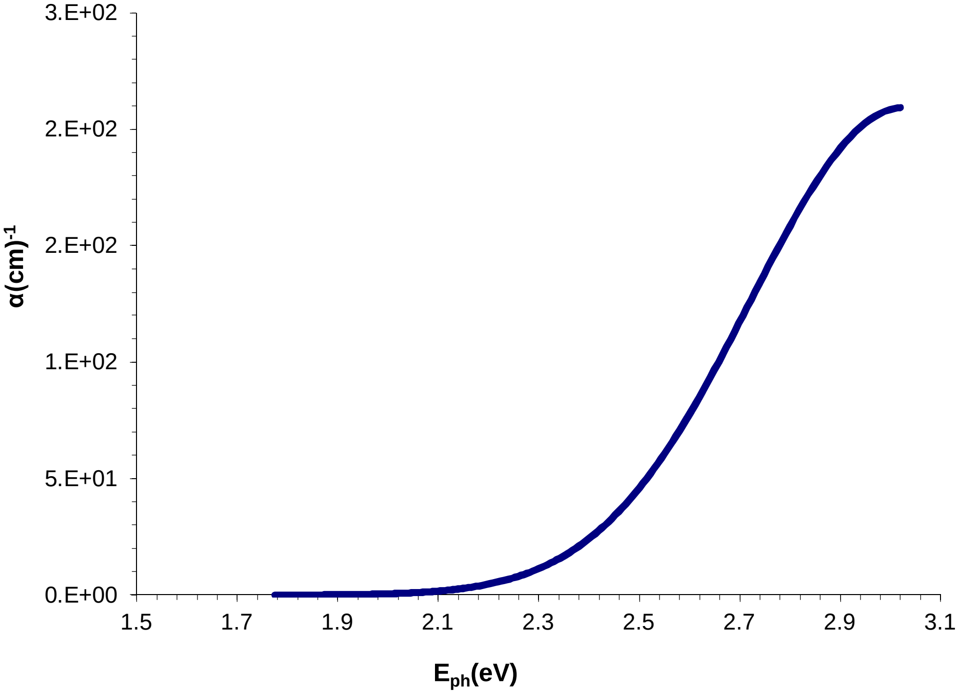

Figure 13: Variation of absorption coefficient (α) for (PMMA-Al_2O_3) nanocomposites with photon energy.

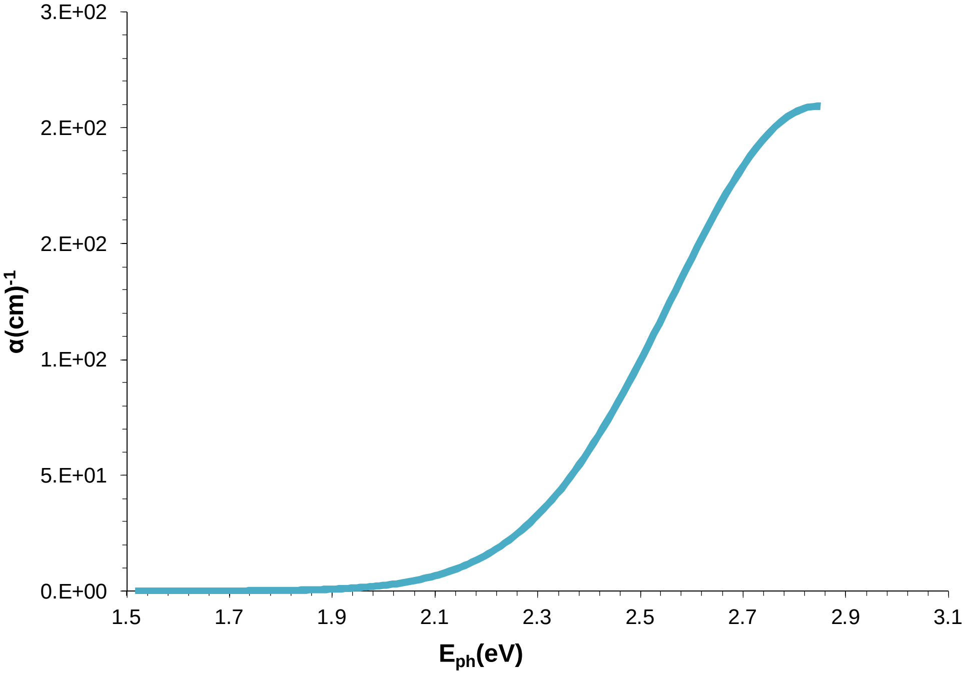

Figure 14: Variation of absorption coefficient (α) for PMMA-Al_2O_3-Agnanocomposites with photon energy.

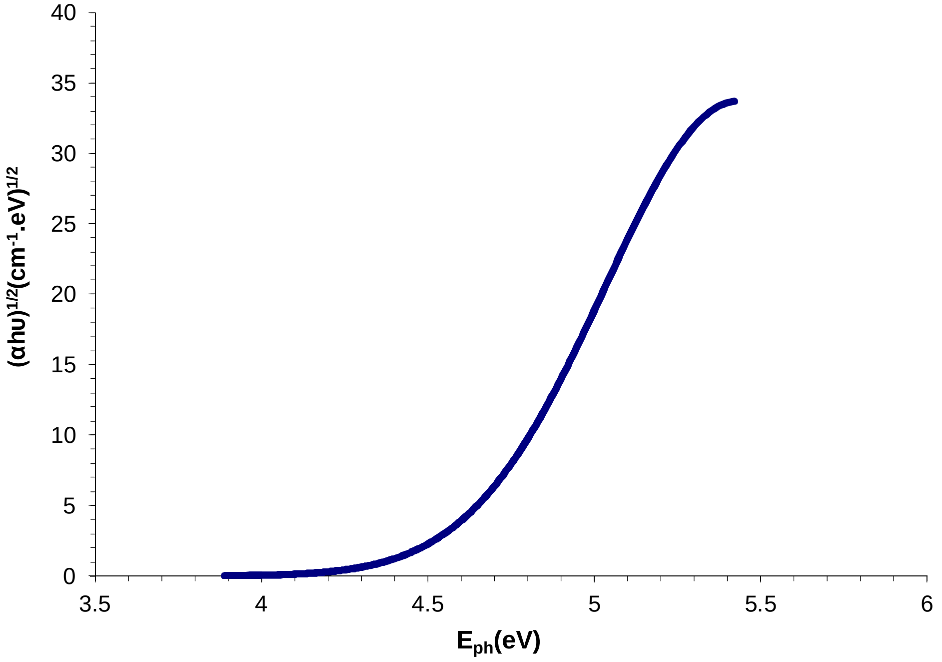

Figure 15: Variation of $(\alpha h\upsilon)^{1/2}$ for polymethylmethacrylate with photon energy.

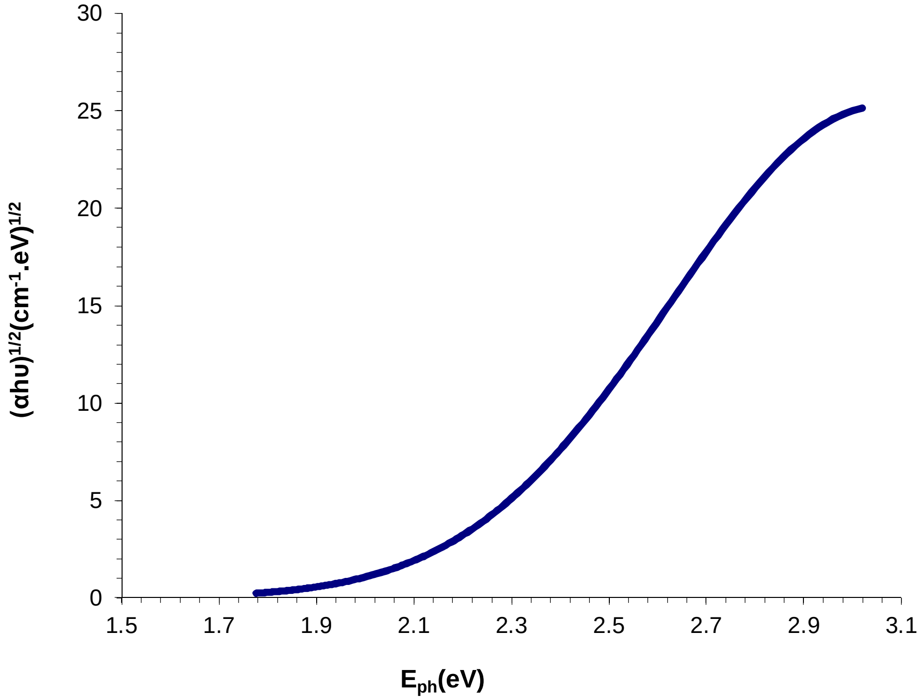

Figure 16:. Variation of $(\alpha h\upsilon)^{1/2}$ for (PMMA-Al_2O_3) nanocomposites with photon energy.

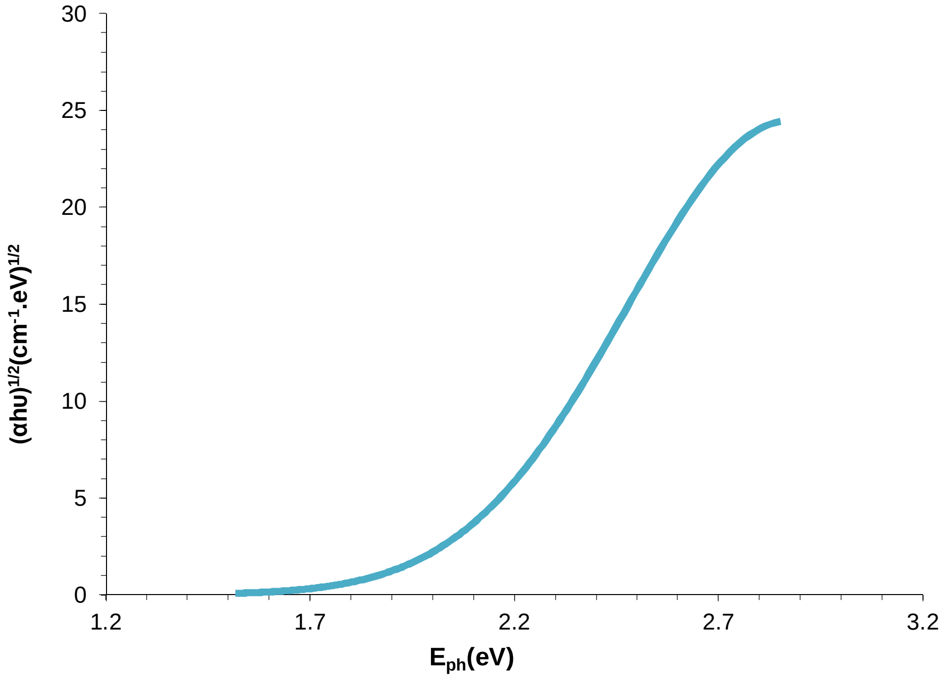

Figure 17: Variation of $(\alpha h\upsilon)^{1/2}$ for (PMMA- Al_2O_3–Ag)nanocomposites with photon energy.

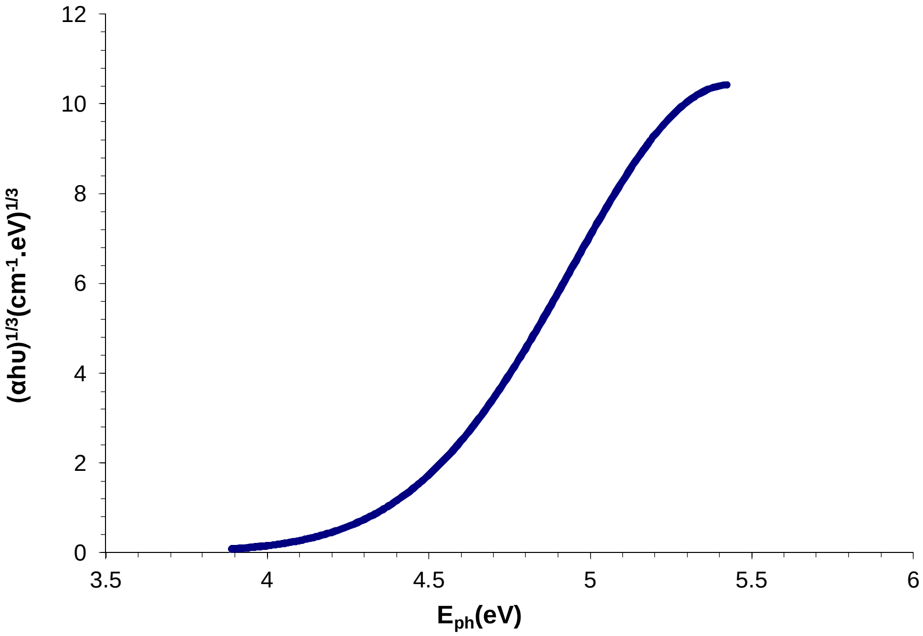

Figure 18: Variation of $(\alpha h\upsilon)^{1/3}$ for polymethylmethacrylatewith photon energy.

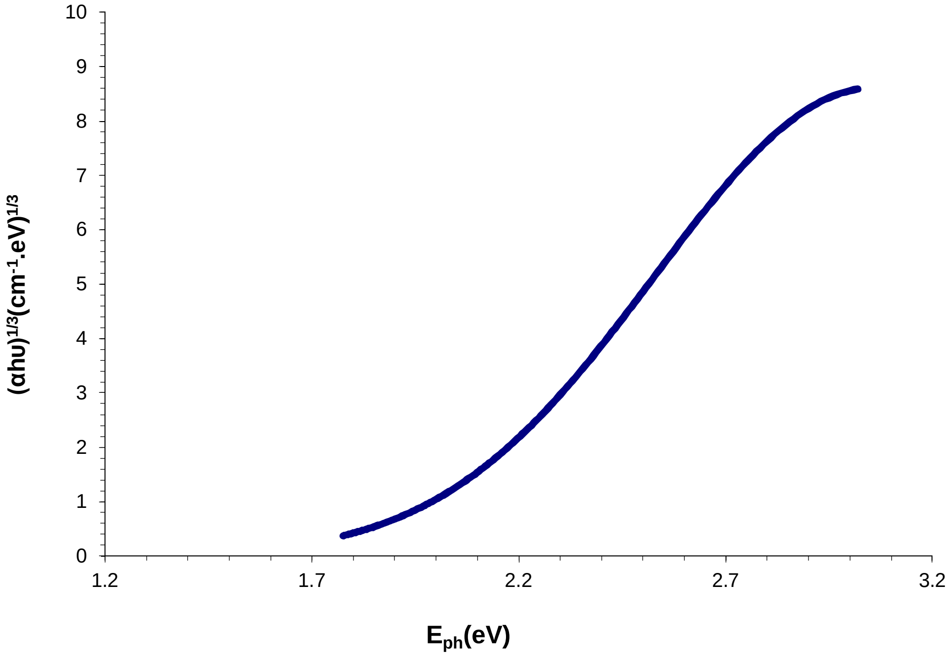

Figure 19: Variation of $(\alpha h\upsilon)^{1/3}$ for (PMMA-Al_2O_3) nanocomposites with photon energy.

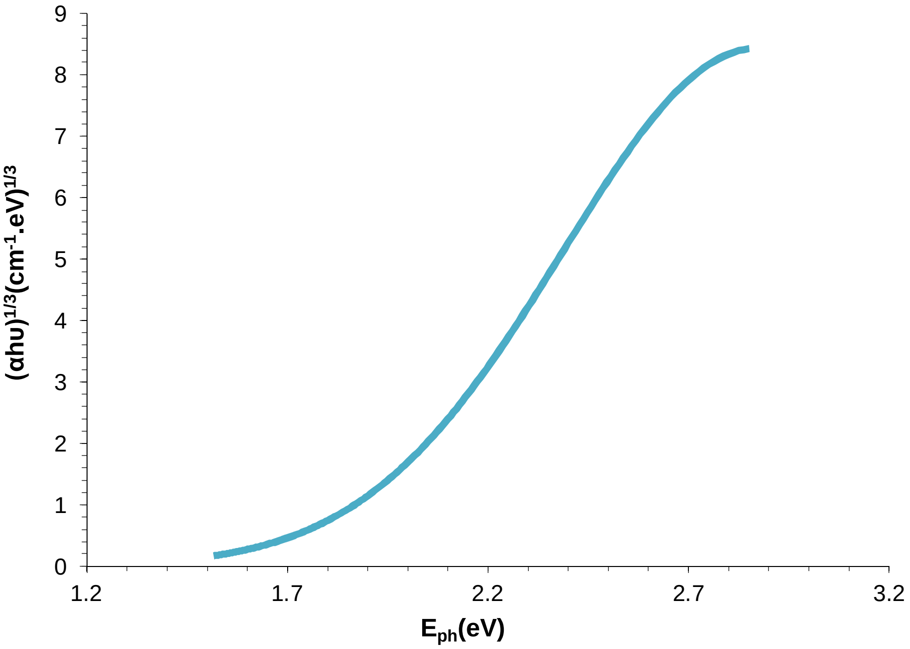

Figure 20: Variation of $(\alpha h\upsilon)^{1/3}$ for (PMMA-Al_2O_3-Ag) nanocomposites with photon energy.

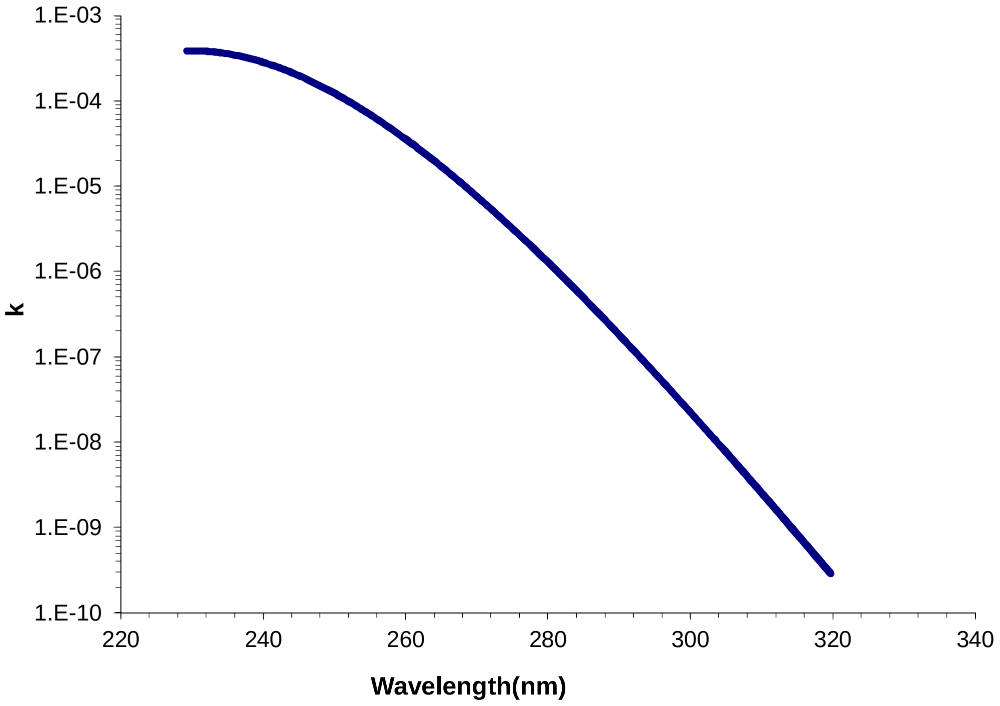

Figure 21: Variation of extinction coefficient for (PMMA) polymethelmethacrylate with wavelength.

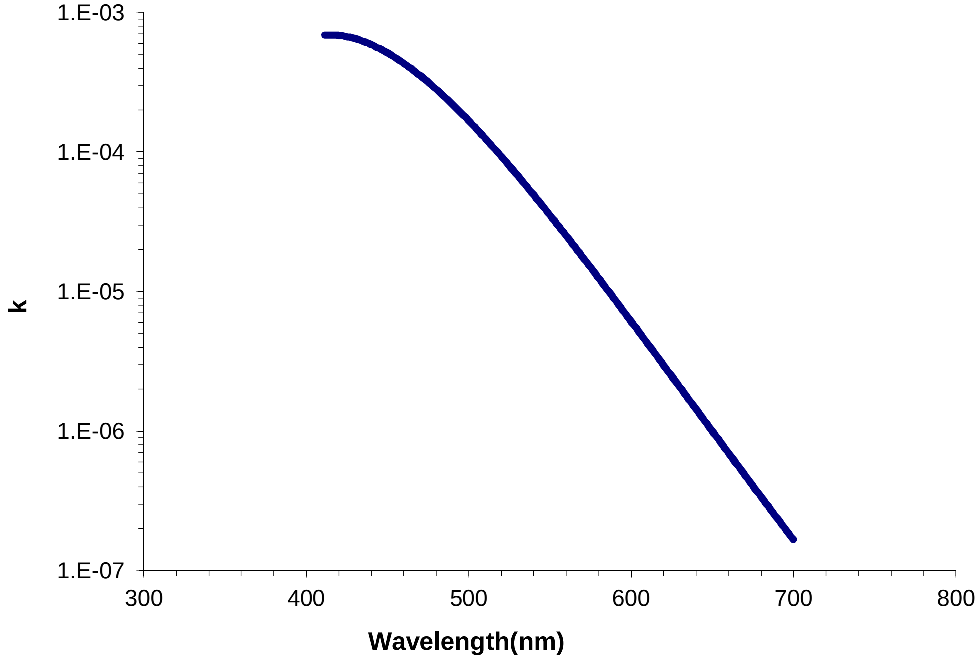

Figure 22: Variation of extinction coefficient for (PMMA-Al_2O_3) nanocomposites with wavelength.

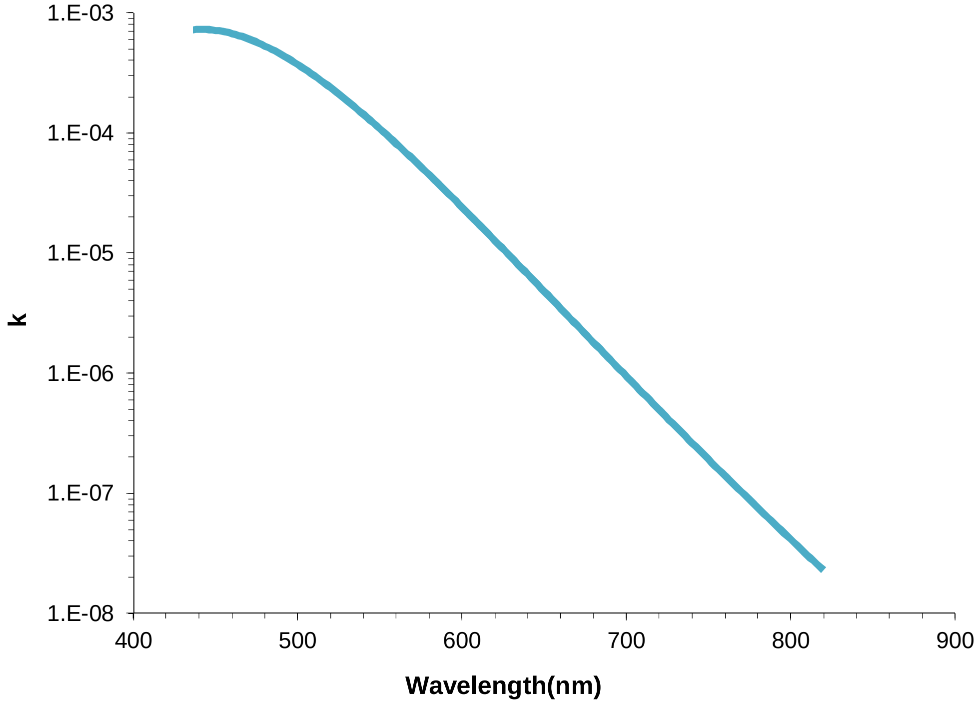

Figure 23: Variation of extinction coefficient for (PMMA-Al_2O_3-Ag) nanocomposites with wavelength.

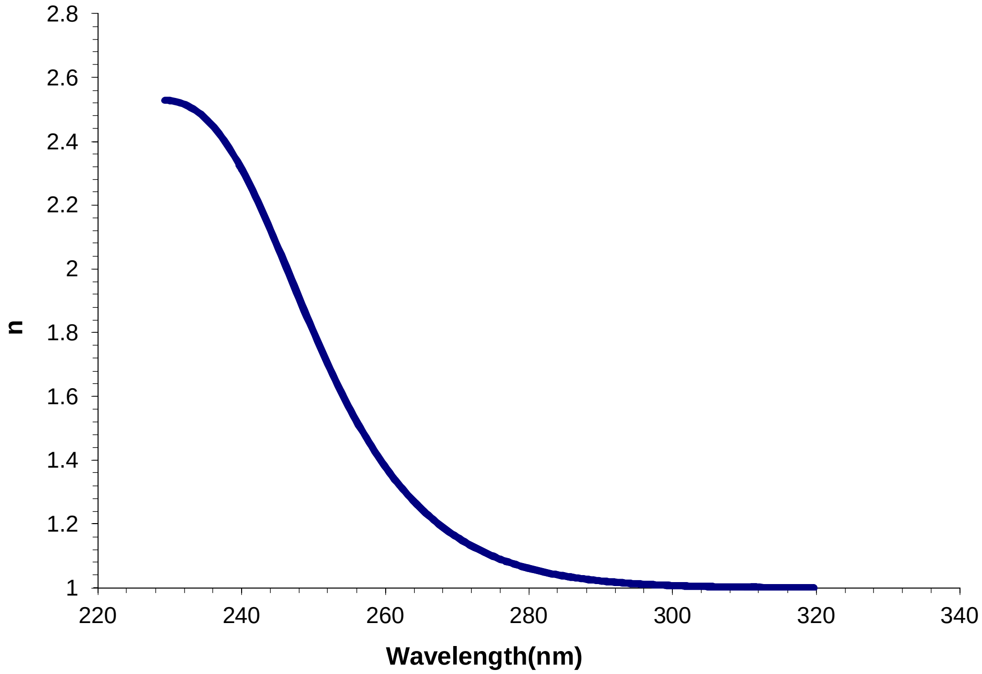

Figure 24: Variation of refractive index for polymethylmethacrylate with wavelength.

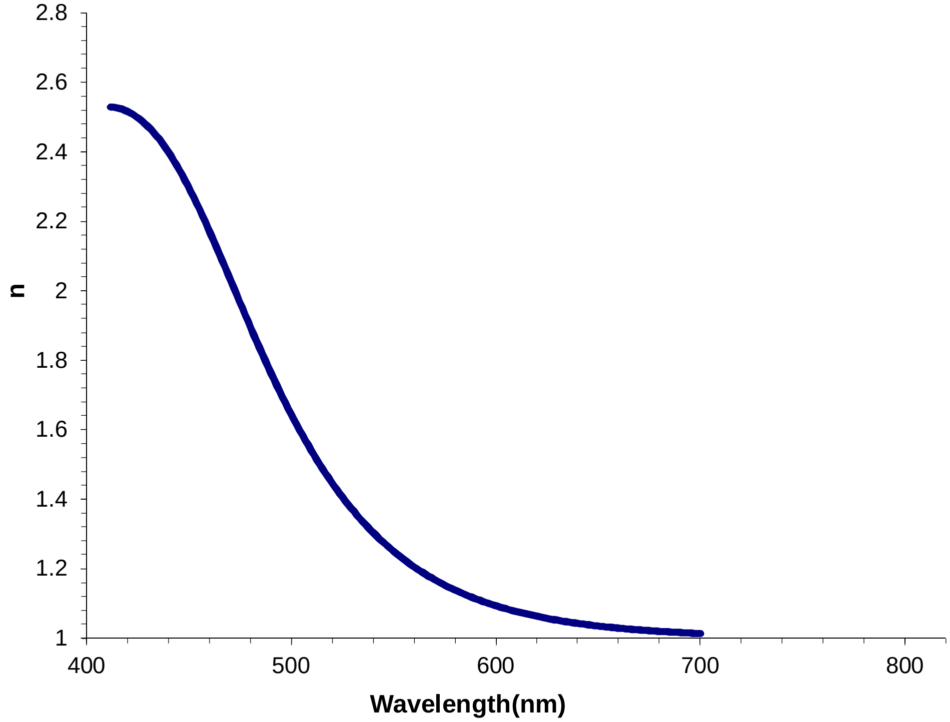

Figure 25: Variation of refractive index for (PMMA-Al_2O_3)nanocomposites with wavelength.

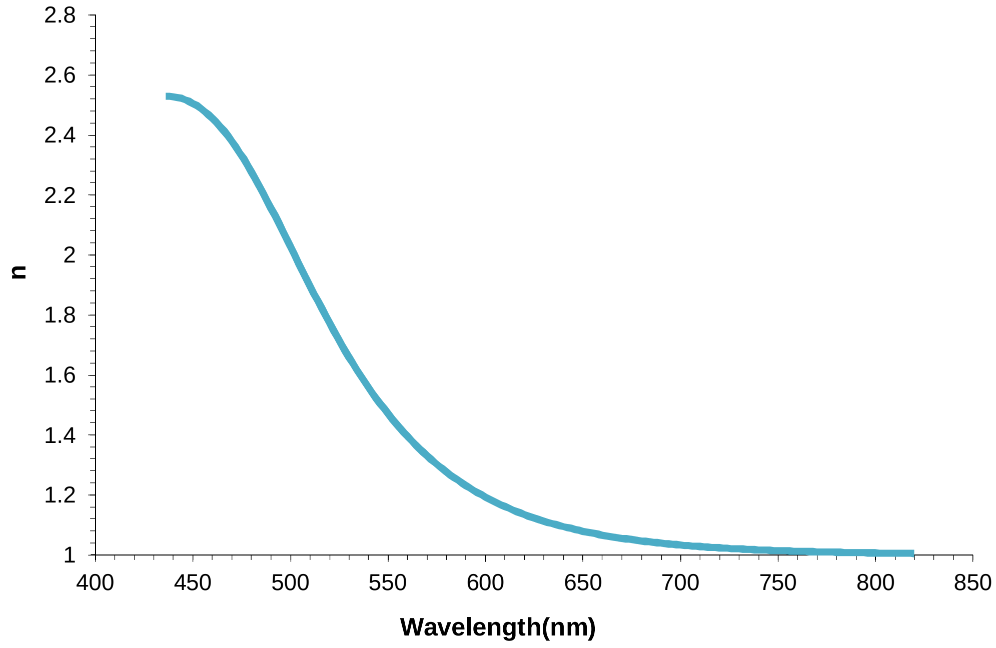

Figure 26: Variation of refractive index for (PMMA-Al_2O_3-Ag) nanocomposites with wavelength.

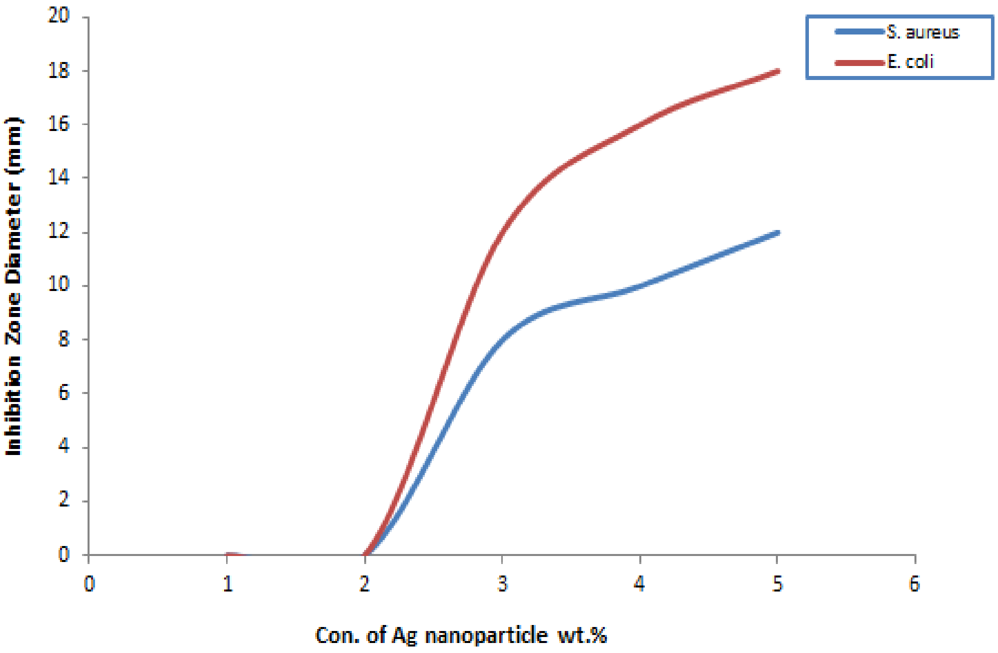

Figure 27: Antibacterial Effect of (PMMA-Al_2O_3-Ag) as a function of Agnanoparticles concentrations on S. aureus and **E. coli.**

Table 1: Some electronic variables of the pure PMMA and nanocomposites.

Materials	H (eV)	S $(eV)^{-1}$
PMMA	3.5049	0.1426
PMMA-Al_2O_3	1.6046	0.3115
PMMA-Al_2O_3-Ag	1.5234	0.3282