



Urbach Energy Estimation of Biosynthesized CMC+PEG/Ag Nanocomposites for Improving Photocatalytic Activity

Mothana Jassem Kadhim^{1,2*}  and Raghad Subhi Abbas¹ 

¹Department of Physics, College of Education for pure science (Ibn Al-Haitham), University of Baghdad, Baghdad, Iraq

²Physics Department, General Directorate of Education Diyala, Ministry of Education, Diyala, Iraq

*Corresponding author

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Abstract

Silver nanoparticles (Ag) were filled at different ratios (6, 12, 18, 24, 30, and 36%) into a polymer matrix consisting of carboxymethyl cellulose (CMC) and polyethylene glycol (PEG) using the solution casting method. The biopolymer (CMC) was prepared from rice husks (RH), which are locally available. AgNPs were prepared by green syntheses method using orange peel extract. Silver nanoparticles were calcined at 300°C for (1 hour) and filled into the polymer matrix (CMC+PEG) to determine their effect on the Urbach energy (E_u). The absorption coefficient and Urbach energy of the produced specimens were measured using a UV-Vis spectrometer at wavelengths ranging (200 to 1000nm). The results studied of specimens showed that the Urbach energy of the (CMC) polymer that was derived and extracted from rice husks (RH) is ($E_u=0.7132\text{ev}$), and ($E_u=1.2096\text{ev}$) for pure (CMC+PEG) polymer matrix, as for the (CMC+PEG/Ag) nanocomposite results the Urbach energy values raised from the ($E_u=2.2393\text{ev}$) to the ($E_u=3.2733\text{ev}$) when silver nanoparticles were filled into the (CMC+PEG) polymer matrix.

Keywords: Urbach energy, Optical properties, Green synthesis, Silver nanoparticle (Ag), Carboxymethyl cellulose (CMC), Polyethylene glycol (PEG).

1. Introduction

Polymeric nanocomposites (NCs) have attracted considerable interest because of their unique industrial properties, such as optical, mechanical, and electrical properties. It is well recognized that polymer NCs exhibit superior qualities compared to pure polymers, which might be due to the flaws and stresses created during the manufacturing of polymer-based NCs¹.

The characteristics of polymer-based NCs are influenced by their microstructural surfaces and interfacial regions, in addition to the concentration of the component elements. Because nanomaterials have a high aspect ratio and large surface area, the wide interfacial area between the polymer matrix and integrated components is crucial to improving the various characteristics of polymer-based nanocomposites².

The use of polymer, nanofiller (the nanomaterial to be filled), and their uniform mixing during the synthesis process are linked to the various characteristics of nanocomposite³. Many researchers have been used various nanomaterials with the polymer matrix to improve their different properties among these studies are, graphene oxide (GO) have been loaded in Polyvinyl Alcohol (PVA) polymer by using simple solution casting technique to created (PVA/GO) nanocomposite⁴. The polymers polyvinyl alcohol (PVA), polyethylene glycol (PEG), and polyvinylpyrrolidone (PVP) were mixed with deferent concentrations of titanium dioxide (TiO_2) nanoparticles and study the optical properties of (PVA+PEG+PVP/ TiO_2) nanocomposite⁵. Gold nanoparticles (AuNPs) were prepared by green syntheses method using the leaves of the Stevia rebaudiana plant and using the casting process, different concentrations of AuNPs were added to

polyvinyl alcohol/carboxymethylcellulose (PVA+CMC) and study the structural, optical, and electrical properties of the (PVA+CMC/AuNPs) nanocomposite⁶. In other study copper oxide (CuO) nanoparticles have been mixed with polymethyl methacrylate (PMMA) and polyvinylidene fluoride (PVDF) by using the casting technique and study the structural, optical, and electrical properties of the (PMMA+PVDF/CuO) nanocomposite⁷. Also, there is study about investigation the structural and optical properties of the (PEO+CMC/AuNPs) nanocomposite when the gold nanoparticles (AuNPs) are loaded in 50/50wt% blend of the polyethylene oxide (PEO) and carboxymethyl cellulose (CMC). The main benefits of using nanomaterial polymer/metal nanocomposites provide new options for frequently filled polymers, enabling blending of the organic phase (polymer) with inorganic NPs to create novel and exceptional materials with exceptional properties⁸.

Carboxymethyl cellulose (CMC) is an important industrial polymer with several uses, such as textiles, flocculation reduction, drag, detergents, paper, oil well drilling operations, medications, and foods. Three variables affect the characteristics of CMC: molecular weight of the polymer, average amount of carboxyl content per anhydro glucose unit, and arrangement of carboxyl substituents along the polymer chains. Through chemical modification, natural cellulose is converted into CMC, which was combined with silver nanoparticles in the current study, a naturally occurring biodegradable and biocompatible anionic polymer^{9,10}.

Owing to the expanded applications of these hybrid materials, silver nanoparticles and polymers have garnered a lot of interest¹¹. Two different fundamental approaches, top-down and bottom-up, have been examined in the literature to create nanoparticles with the right shape. Grinding machines, sputtering, lithographic procedures, and etching are examples of the conventional techniques for creating nanoparticles. The bottom-up approach uses simpler materials to generate particles. and medical research on the effects of nano-oxides on the human body, including how they affect the kidneys and liver^{12,13}.

Accordingly, this study aims to improve the optical properties by calculate and analysis the Urbach energy of the (CMC+PEG/AgNPs) nanocomposite and to study the effect of loading silver nanoparticles (Ag) into the (CMC+PEG) polymer matrix in order to potentially improve the photocatalytic process.

2. Materials and Methods

One easy and environmentally friendly way to obtain the required qualities of nanoparticles is green synthesis using living organisms, as shown in **Figure 1**. Both types of living organisms participate in various biological processes. Plants are well-known producers of natural chemicals, which are both affordable and environmentally friendly. The synthesis of nanoparticles from plant extracts offers several advantages over other biosynthetic methods, such as those utilizing bacteria, fungi, actinomycetes, and algae^{14,15}.

The crushed rice husk (RH) in the potassium hydroxide (KOH) solution were treated with a concentration of 6% under the constant stirring for (1.5 hours) at 70°C, then filtered with a filter paper and washed three times (the washing process is also repeated in subsequent steps for the purpose of until the arrival of PH = 7), then the precipitate is taken after drying (drying is done by placing the precipitate in an electric oven at 60°C for two hours, then leaving it at room temperature for three days ,this process is repeated for subsequent steps as well). Then the (RH) that was treated by alkaline process is dissolving it in solution of sulfuric acid H₂SO₄ with a concentration 4% for (1.5 hours) then filtering, washing and drying to obtained on the cellulose. Cellulose is dissolving it in the sodium chlorite solution at a concentration of 3% and at PH= (3-4) under the constant stirring for (3 hours) at 70°C. After bleaching and drying the cellulose, a 20% NaOH solution was prepared and distilled into 100 ml of ethanol. The cellulose was then dissolved in this solution at 60°C under continuous stirring for (1.5 hours). Then, a solution of monochloroacetic acid was added to the mixture, filtered and washed several times with ethanol to obtained the CMC.

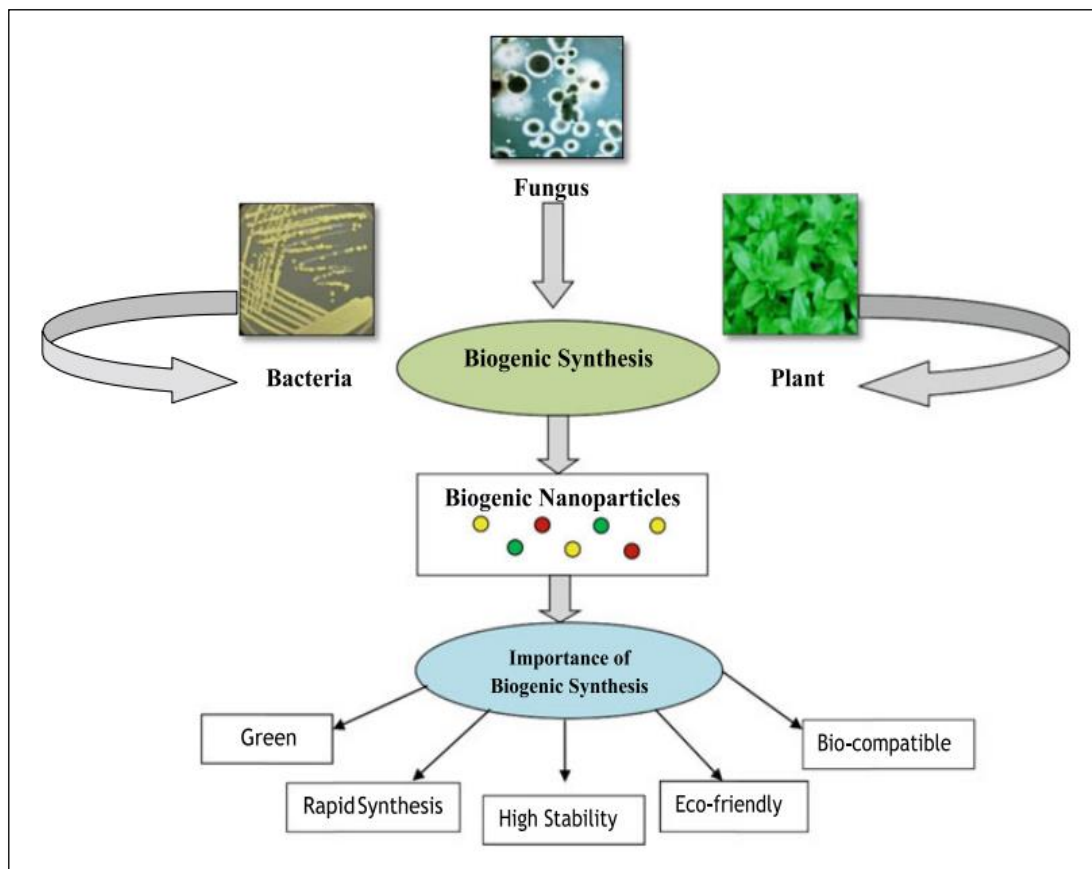


Figure 1. Green Synthesis/ Biological of Nanoparticles¹⁶.

Silver nanoparticles were prepared from orange peel extract by dissolving 20g of silver nitrate (AgNO_3) in 75 ml of deionized water. Then, 50 ml of orange peel extract was distilled into the solution under continuous stirring at a temperature of 70 °C. After the extract was distilled, the PH neutralized. Then it is separated by centrifugation and washed several times with deionized water and then with ethanol, then dried and calcined at 300°C for (1 hour).

Nanocomposite NCs were prepared by solution casting method by filling silver nanoparticles calcined at 300°C into the polymer matrix (CMC+PEG) with different concentrations of Ag NPs (6, 12, 18, 24, 30 and 36%) as shown in **Table 1**. Moreover, 15% of the sample's total weight should be added to citric acid monohydrate in order to crosslink the components. After pouring the uniform polymer nanocomposite mixture into glass molds and drying for two hours at 80°C to strengthen the mechanical link between the components, the steps involved in preparing the (CMC+PEG/AgNPs) nanocomposite are illustrated in **Figure 2**.

Table 1. The Material ratios and weights in the preparation of (CMC+PEG/AgNPs) nanocomposites.

Code of the sample	CMC Wt%	PEG Wt%	Ag Wt%
A0	60	40	0
A1	60	34	6
A2	60	28	12
A3	60	22	18
A4	60	16	24
A5	60	10	30
A6	60	4	36

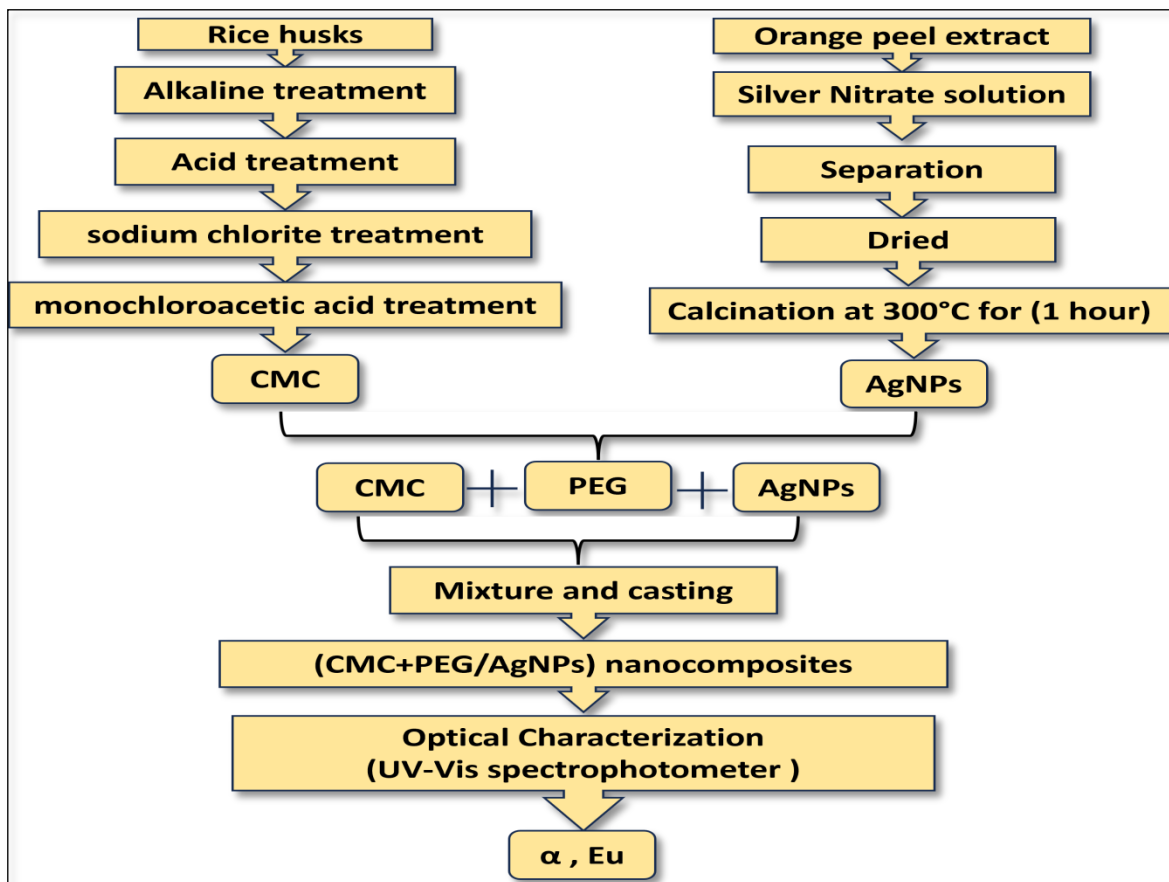


Figure 2. Flow chart for the preparation of (CMC+PEG/AgNPs) nanocomposites.

3. Results

Both photon energy and band gap energy have a substantial correlation with the absorption coefficient of a material. It may be characterized as the attenuation that occurs for the incident photon energy by the material, or it can be described as the ratio of the decrease in the intensity of the incident radiation per unit distance in the direction of the propagation of the wave within the medium. It is dependent on the type of electronic transition that occurs between the energy bands in the material and the energy ($h\nu$) of the incident photon. The absorption mechanisms are responsible for the attenuation of incoming photon energy. A UV-Vis spectrophotometer with wavelengths between (200 and 1000nm) was used to record the optical absorbance of the nanocomposite from the absorbance (A) results. Based on this relationship, the absorption coefficient (α) was calculated (**Equation 1**):

$$\alpha = (2.303 \frac{I_0}{I})/d = (2.303) \times (\frac{A}{d}) \quad (1)$$

whereas: (α) represent is the absorption coefficient and (d) the thickness of the medium through which the radiation passes. During their transition between the valence band and the conduction band, electrons encounter a density of states, which is expressed by $\rho(h\nu)$, the ($h\nu$) is representation of the photon energy, and the (h) refers to Planck's constant approximately and (ν) the frequency of the electromagnetic wave of photon. This interaction frequently leads to anomalies that generate disruptions inside the energy gap. The Urbach energy E_u and absorption tails near the band edge are indicative of this phenomenon. The Urbach tail is the term used to describe the expansion of ($h\nu$) into the energy band gap. The energy linked to this tail is thus called the Urbach energy. The exponential decline of the absorption coefficient $\alpha(h\nu)$ is the cause of these phenomena.

Determining Urbach energy can help explain the phenomena of structural disorderliness in a (CMC+PEG) polymer matrix upon (Ag) nanoparticle loading. This relates to the magnitude of expanding of localized states inside the energy gap. The optical absorption coefficient (α) was fitted to a function of energy ($h\nu$). The $(\ln(\alpha))$ diagram is first produced against of ($h\nu$), which can be used to calculate the breadth of the band tail, known as the Urbach energy (E_u). The value of the Urbach energy is then determined by computing the reciprocal of the slope of the linear part of the graphs, as shown in **Figure 3**.

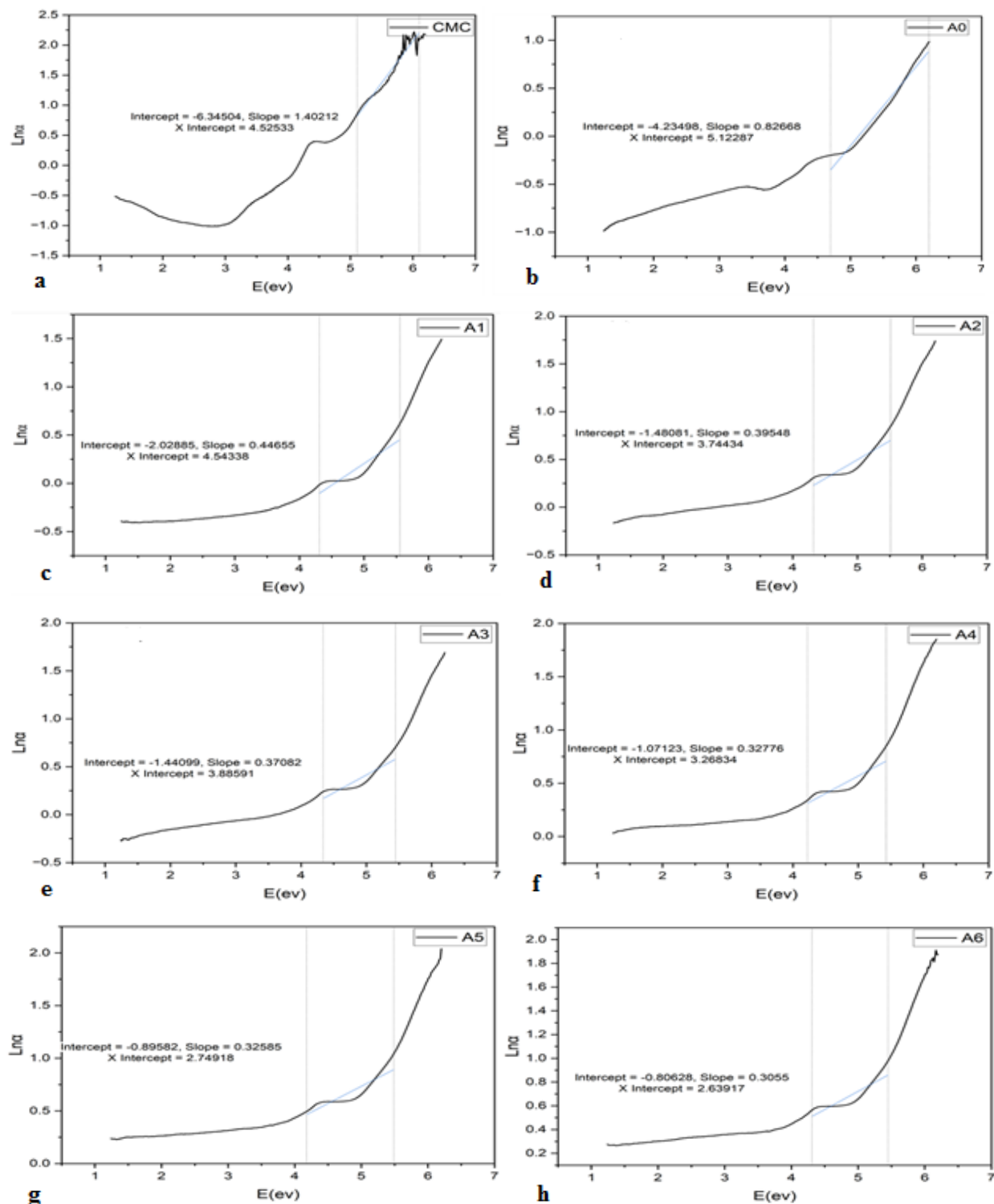


Figure 3. The Urbach energy (E_u) for (CMC) and (CMC+PEG/Ag) nanocomposites. A. CMC, b. A0, c. A1, d. A2, e. A3, f. A4, g. A5, h. A6.

The estimated Urbach energy (E_u) magnitudes are shown in **Table 2**, which yields the following **Equation 2**.

$$\alpha = \alpha_0 \exp\left(\frac{h\nu}{E_u}\right) \quad (2)$$

where E_u is the Urbach energy tail, and (α_0) represents a constant. Based on **Table 2**, the data indicate that the calculated Urbach energy (E_u) for the pure polymer (CMC) without any additives is (0.7132eV), while the value increases to (1.2096eV) for the (CMC+PEG) polymer matrix. On the other hand, the results from **Table 2** shows that the value of Urbach energy increases from (2.2393eV) to the (3.2733eV) for the (CMC+PEG/Ag) nanocomposites when loading silver nanoparticles (Ag) into the (CMC+PEG) polymer matrix.

Table 2. Urbach energy (E_u) for CMC and (CMC+PEG/Ag) nanocomposites.

Code of the sample	$E_u(\text{eV})$
CMC	0.7132
A0	1.2096
A1	2.2393
A2	2.5285
A3	2.6967
A4	3.0510
A5	3.0688
A6	3.2733

4. Discussion

Disordered and amorphous materials have an exponential tail in their electronic band structure because they contain localized states that extend within the energy band gap. This exponential tail makes it easier to comprehend how electrons move through the composite materials¹⁷. The level of disturbance in the energy band gap between the valence band and the conduction band for (CMC+PEG) polymer matrix is measured by (E_u), which is a measurement of the extent of the band tail of localized states before and after loading by (AgNPs). Urbach provided an empirical correlation between the photon energy and the absorption coefficient (α), which is represented by **Equation (2)** of amorphous^{10,18}. Based on **Figure 3** and as included in **Table 2** the Urbach energy for the pure (CMC) polymer derived from (RH) is 0.7132 eV. Additionally, it is evident that the filling of the nanomaterial (nanosilver) in the polymer matrix (CMC+PEG) increased the Urbach energy, which began to increase from 2.2393 eV for sample A1 until it reached 3.2733 eV for sample A6. The difference in the value of (E_u) as a function of the AgNPs concentration indicates the introduction of defects in the band structure of the (CMC+PEG) polymer matrix. Because of the polymer and AgNPs interactions, the addition of AgNPs nanoparticles to the polymer matrix causes extra defect states in the polymer host, which causes disorderliness in the polymer chain packing of (CMC+PEG)¹⁹. The data imply that the increase in E_u and the concentration of the nanofiller are proportionate. It was discovered that the distribution of the nanofiller and the process used to prepare the polymer nanocomposites both had an impact on the structural flaws observed in the NCs²⁰. The filling process of AgNPs has a substantial impact on the value of E_u , where it generates more defects which is represented as band tails of localized states, crystal defects and disorderliness of microstructural lattice in the (CMC+PEG) polymeric matrix¹⁸. As seen by the variance in the (E_u) magnitude in these (CMC+PEG/AgNPs) nanocomposites, which may be understood by taking into account the mobility concept given by Davis and Mott²¹. It was discovered that the density of localized states was related to the concentration of these flaws (band tails of localized states, crystal defects and disorderliness of microstructural lattice) and, hence, to the AgNP content. The current experimental findings on the Urbach energy tail E_u and the optical energy band gap E_g suggest that filling has a major impact on these optical absorption characteristics⁴.

5. Conclusion

The solution casting method was utilized as it is easy and inexpensive to prepare (CMC+PEG/AgNPs) nanocomposites by different concentrations of (Ag) nanofillers. (CMC) biopolymers have been synthesized from the rice husks (RH), which are considered waste agricultural products, and orange peel extract is used as a green synthesis technique to create (AgNPs). It has been shown from the result that the Urbach energy of the (CMC+PEG/AgNPs) nanocomposites is significantly impacted by the addition of AgNPs to the (CMC+PEG) polymer matrix. The results demonstrated that the addition of AgNPs increases the charge carrier density inside the (CMC+PEG) polymer matrix, which is reflected in an increase in Urbach energy. These characterization findings show that the produced polymer nanocomposites have potential applications in the photocatalysis process as adsorbent surfaces for purification of wastewater from the pollutants that are present there.

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Conflict of Interest

The authors declare that they have no conflicts of interest.

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