

Synthesis and characterization of polypyrrole polymer thin film and the effect of adding silver nanoparticles (PPY/AgNPs)

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ARTICLE INF.

Article history: Received: 3 SEP., 2024 Revised: 30 NOV., 2024 Accepted: 11 DEC., 2024 Available Online: 29 DEC.

Keywords:

2024

Pyrrole (py), Ammonium persulphate (APS), silver nano particles, m- cresols, thin films.

ABSTRACT

In this research, a chemical oxidative polymerization process was used to prepare polypyrrole (PPY). The characterization of polypyrrole thin films pure and dopped with silver nanoparticles (AgNPs) at different weight ratios of AgNPs (i.e., 5, 10, and 15 wt%) was studied. The thin films were prepared via the drop casting method and flung down on a clean glass substrate at room temperature. Using FT-IR, XRD, and AFM, the system and facet morphology of the thin films were investigated. FTIR analysis of undoped PPY powder revealed distinctive vibrational frequencies of PPY undoped. Strong absorption bands at 3653 cm-1 (N-H stretching), 2342 cm⁻¹ (nitridation), and peaks at 1542, 1459 cm⁻¹ (C=C, C=N vibrations) were observed. Other peaks at 1176 cm⁻¹ (C-N stretching), 1039, 964 cm-1 (C-H vibrations), 893, 782 cm-1 (C-H bend deformation), 670 cm⁻¹ (C-C deformation), and 590 cm⁻¹ (polypyrrole presence) were identified. The results showed that the FT-IR spectra of pure ppy and the spectra of PPY/AgNPs composites were similar. The single XRD data also showed that ppy is amorphous and The distinctive peak of polypyrrole was found at (2θ) 24.35°, indicating the short-range arrangement of PPy chains the interlayer distance (d) between the measured value of spacing for PPY was 3.6599 A^0 . Additionally, By using AFM, the parameters Ra, R.M.S, and G.S. were determined for PPY doped AgNPs thin films. These values increased with higher doping percentages. For undoped PPY thin film, values were (2.557nm, 3.299nm, 33.40nm) while for doped films with 5%, 10%, and 15% AgNPs, values were (3.397nm, 4.859nm, 46.12nm), (9.328nm, 11.90nm, 66.33nm), and (13.87nm, 19.28nm, 86.89nm) respectively. the AFM results for all thin films of PPY undoped and composites doped with silver nanoparticles (PPY/AgNPs) demonstrated that the values of grain size (G.S.) and root-mean-square (R.M.S.) roughness average (Ra) increase as the number of AgNPs increases.

DOI: <u>https://doi.org/10.31257/2018/JKP/2024/v16.i02.17377</u>

تحضير وخصائص الاغشية الرقيقة للبوليمير بولي بايرول وتاثير اضافة جسيمات الفضة النانوية (PPY/AgNPs)

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الخلصة

في هذا البحث ، بوليمير بولي بايرول (PPY) تم تحضيره بواسطة طريقة البلمرة التأكسدية الكيميائية تم دراسة خصائص الاغشية الرقيقة للبولي بايرول النقى والمشوب مع جسيمات الفضية النانوية (AgNPs) عند نسب تشويب وزنية مختلفة 15, 10, and 15 (wtw على التوالي. تم تحضير اغشية رقيقة على قواعد زجاجية نظيفة باستخدام طريقة الصب المتساقط عند درجة حرارة الغرفة (R.T.) .تم استخدام مطياف تحويل فورير-للأشعة تحت الحمراء (FTIR) ، حيود الاشعة السينية (XRD) و مطياف الأشعة المرئية -فوق البنفسجية (UV-VIS) spectroscopy)) لدراسة خصائص العينات ومور فولوجية السطح للأغشية الرقيقة. نتائج FTIR للمسحوق الغير مطعم للبوليمير PPY يشير الى وجود ترددات اهتزازية مميزة تعود الى PPY الغير مشوب. حيث تم ملاحظة حزم امتصاص قوية عند 3653 cm⁻¹ والتي تعود الي حزمة المط (N-H) ، 2342 cm⁻¹ والتي تعود الي النترات وظهور قمم امتصاص عند¹-m 1459 والتي تعود الى الاهتزازات (C=C, C=N). تم ملاحظة وجود قمم مط واهتزاز عند 1176 cm⁻¹ والتي تقابل حزمة مط 893, 782 cm⁻¹ ، (C-H) والتي تعود الى حزمة الاهتزاز (C-H) ، (C-H) ، (C-N) والتي تقابل اهتزازات التمدد (C-H) ، ¹ - 670 cm والتي تعود الي (C-C) و ¹ - 590 cm كل هذه القمم تشير الى تكوين البولى بايرول. نتائج FT-IR الطيفية كانت متماثلة للبولى بايرول النقى والمطعم بالجسيمات الفضية النانوية (PPY/AgNPs). اشارت نتائج XRD بان البولى بايرول النقى يمتلك طبيعة عشوائية وظهور قمة مميزة للبولى بايرول عند زاوية (20) هي (<24.35) ، والتي تشير إلى ترتيب متسلسل قصير المدى للبولي بايرول، المسافة البينية (d) المقاسة بين المستويات الداخلية للبولي بايرول هي تساوي (A⁰ A. 3.6599). علاوة على ذلك تم استخدام AFM لتحديد المعاملات والمتمثلة بمعدل الخشونة R ومعدل الجذر التربيعي R.M.S والحجم الحبيبي G.S للأغشية الرقيقة للبولي بايرول النقية والمشوبة بالجسيمات الفضة النانوية. هذه المعاملات تزداد مع زيادة نسبة التشويب. حيث كانت القيم للأغشية النقية للبولى بايرول (2.557nm, 3.299nm, 33.40nm) بينما القيم و (9.328nm, 11.90nm, 66.33nm) • (3.397nm, 4.859nm, 46.12nm) و (13.87nm, 19.28nm, 86.89nm) هي للأغشية المشوبة مع جسيمات الفضة النانوية عند (AFM and 15%) على التوالي. اظهرت نتائج AFM لجميع الأغشية الرقيقة النقية من PPY والمطعمة بالجسيمات الفضة النانوية (PPY/AgNPs) أن قيم معدل الخشونة R_a ، الحجم الحبيبي (G.S) ومتوسط الجذر التربيعي (R.M.S) تزداد مع زيادة مقدار جسيمات الفضية النانوية (AgNPs).

الكلمات المفتاحية:

بيرول (py) ، بيرسولفات الأمونيوم (APS)، جسيمات نانوية من الفضة. الأغشية الرقيقة.

1. INTRODUCTION

Due to the remarkable qualities of conductive polymers and their common application in a variety of fields, amendment and the creation of new polymers have drawn great attention in the past ten years [1]. Extensive research has been conducted over the past 20 years to produce electrical conductivity in backbone polymers with polyaromatic chains. Of them, polyaniline (PANI), polypyrrole (PPY), polythiophene (PT), etc. [2]. PPY has attained significant attention among conducting polymers owing to some properties, and at room temperature, conducting polymer (CP)based gas sensors showed excellent sensitivity and quick reaction times [3, 4]. A polymer as such as PPY has extensive use due to its numerous possible applications [5, 6]. Figure (1) shows the chemical structure of polypyrrole. Many properties of its polymer can be changed via the doping process [7,8]. The synergistic benefits of combining conducting polymers (CPs) and silver nanoparticles (AgNps) have garnered significant interest [9]. Nano silver has perfect characteristics [10]. These nanoparticles are beneficial in many different contexts. specifically, silver nanoparticles are a multipurpose element that can be used in biological sensors, semiconductors, antivirals, and other fields [11]. Researchers recognize the desirable high-conductivity qualities of Ag nanoparticles [12]. Various techniques, such as wet chemical oxidative processes and electrochemical polymerization, can prepare the PPY. Generally, oxidation methods are employed to prepare PPY

which results in a polymer that is highly conductive [13, 14] PPY. Generally, oxidation methods are employed to prepare PPY, which results in a polymer that is highly conductive [13, 14]. In (2020) P. Mrunalini, et al.,[14] studied the synthesis of polypyrrole nanopowder using an oxidant solution of ammonium persulfate and pyrrole in an aqueous solution via chemical in-situ polymerization The polypyrrole was dissolved in m-cresol to prepare thin films Polypyrrole thin films were described using Fourier transform infrared spectroscopy (FTIR), and Xray diffraction (XRD) are used. The amorphous nature of the materials is revealed by the XRD spectrum. All of are PPY the peaks primary to FTIR characteristics, according analysis .In(2023) Muataz S, et al.,[15] studied the synthesis of Poly(pyrrole) by using FeCl3 with pyrrole monomer in alcoholic and aqueous solutions. The structural properties were analyzed using diagnostic tests like FESEM, XRD, and FTIR. UV-VIS spectroscopy was used to measure the energy band gap. AFM studies were conducted to impact of explore the reaction conditions on thin film morphology.

2. Materials and methods

2.1Materials

Pyrrole (py) with (a purity, 98%) and Ammonium Peroxidisulphate(Aps) with (a purity, 98%)and silver Nano particles (Ag) with (a purity, 99.9%) and solvent m- cresols with (a purity, 98%), all are fromSigma-Aldrich Chemicals products.

2.2 Preparing of Polypyrrole (PPY)

The polymer polypyrrole (PPY) powder was synthesized by the chemical oxidative polymerization method at the interface of two solutions prepared the pyrrole solution. It was taken a 0.2 M fresh pyrrole monomer (C4H5N) solution. weighing approximately 13.42 g/mol. It was dissolved it in 50 mL of deionized water in a bottom flask and stirred it for 10 minutes at ambient temperature. It The aim in this study we prepared polypyrrole by chemical oxidative polymerization method and doped with silver nanoparticles at various ratios To improve the properties of the polymer in order to obtain better conductivity. and studied the structural properties of polypyrrole thin film un doped and doped with silver nanoparticles prepared by drop casting mechanics.



Fig. 1. The chemical structure of PPY [7].

was taken the ammonium persulfate (APS) solution by dissolving (0.02 M) of fresh ammonium persulfate solution (NH4)2S2O2 at ambient temperature, weighing approximately 4.56 g/mol, in 100 mL of deionized water. The two solutions were mixed by adding ammonium persulfate (APS) solution. The two solutions it mixed by adding ammonium persulfate (APS) solution to the pyrrol mixture and gradually distilling through the use of a burette (25 ml), where the polymerization

continued using an ice bath at a temperature less than 5 °C for 4-5 hours .Then leave the solution for a whole day until it settles. It was filtered the solution using filter paper and cleansed it multiple times with deionized water to eliminate any reacting contaminants and monomers. It was allowed the polypyrrole to dry at day. ambient temperature for a Eventually. a black powder of polypyrrole emerged.

2.3. characterization of undoped and doped thin films.

PPY powder was doped with different weights of silver nanoparticle powder (PPY/AgNPs) (i.e. 5, 10, and 15 wt%). Table 1 below displays the sample materials weight fraction. Impurity samples were obtained by adding the weight ratios during the preparation process and with the same steps described in the synthesis of PPY undoped. The preparation process was repeated three times according to the addition of the weight percentages of the impurity material (5, 10, and 15 wt%). Composite films undoped and doped with different weights of nanosilver (5, 10, and 15) wt% were synthesized by the drop casting method [16]. 60 mg of polypyrrole powder, undoped and doped, was placed in 5 ml of m-cresol solvent. We stirred the solution for a full day to ensure uniformity [17]. Doped and undoped thin film of PPY/AgNPs was deposited on clean glass substrates using drop casting. This was done by placing the glass substrate on a hot plate stirrer at a temperature of less than 60°C. We dripped the undoped PPY and PPY/AgNPs solution onto the clean glass

substrate. After that , the substrate was heated at less than $60 \circ C$ to eliminate any remaining solvent. Figure (3). explains the steps for preparing thin films.

Table 1. Samples materials weightfraction.

Sample	PPY	Ag	Percenta
	gm	gm	ges for
			doping
			Wt%
Pure	13.42	0	0%
PPY			
PPY/Ag	13.42	0.67	5%
NPs			
PPY/Ag	13.42	1.3	10%
Nps			
PPY/Ag	13.42	2.01	15%
NPs			



Fig. 2. The steps for PPY thin films preparation.

2.4 Analyzing structure

Using solid KBr discs with a Shimadzu FT-IR 8000 series fourier transform infrared spectrophotometer, FTIR spectra were captured. All materials XRD spectra were obtained using an automatic X-ray diffractometer (Philips PW - 3050/60 (Theta/Theta), with a continuous scan rate of 20/min and an accuracy of 0.01 at 40 kV and 30 mA .the device used Cu-K α radiation with a wavelength of 1.5406 Å.

An Instruments, Inc. BY3000 digital microscope was used in conjunction with an atomic force microscope (AFM) to evaluate the deposited roughness average (Ra), grain size (G. S.), and root-mean-square (RMS) are typical data obtained from images of rising AFM.

3. Results and discussion

3.1 FTIR Analysis for PPY (pure) and PPY/AgNPs

Using infrared spectroscopy, the newly synthesized PPY/AgNPs were obtained. The wavelength range in which the samples were conducted was 4000-400 cm-1. The synthetic pure materials FT-IR spectrum PPY and PPY / AgNPs powder at different concentrations of AgNPs (5, 10, and 15) wt% are presented in Figures 3,4, 5, and 6. below The (ppy undoped) FTIR spectrum that was prepared can be seen in Figure 3. Strong absorption bands were seen at about (3653 cm-1) in line with N-H stretching. The peak at 2342 cm-1 corresponds to the fragments partial nitridation and the cleavage of pyrrole molecules. These peak locations closely correspond to those documented in the literature [18, 19]. At The distinctive bands of protonated PPY were visible at (1542. and 1459. cm-1) due to C=C and C=N vibrations that stretch of the ring, in that order (1176. cm-1). C-N stretching the peaks at (1039 and 964 cm-1) were ascribed to vibrations caused by C-H in-plane displacement and outof-plane deformation vibration, respectively; the C-H out-of-plane bend deformation is responsible for the peaks (893 and 782 cm-1). The C-C out of plane ring deformation, also known as C-H rocking, is responsible for the minor peak at (670 cm-1). This yields a result that is consistent with literature reports [20, 21]. The signal (590 cm-1) indicates that polypyrrole is present.[19] The FT-IR spectrum after adding PPY and (PPY/AgNPs) Ag nanocomposites at different doping levels (5, 10, and 15 wt%) is shown in Figures 4, 5, and 6. These changes in emission intensity clearly show that each bond has a different value, but note no changes in bond positions. The table below displays information about the spectra. The PPY/AgNps composite's FTIR spectra match those of pure PPY, indicating a strong resemblance between Figures 3, 4, 5, and 6.



Fig. 3. FTIR of pure PPY Fig. 4. FTIR of PPY/5%



Fig. 5. FTIR of PPY 10%

Fig. 6. FTIR of PPY/15%

Table 2. FT-IR Spectral of synthetically produced pure polypyrrole (PPY) and PPY /AgNPs.

Sample	Vibraton Bondrange (1/cm)	Observed Postion(1/c m)	Expected vibrations
	500-600 600-900 1000 – 1188	(590) (893, 782,670) (1039, 1176)	presence of polypyrole Out-of-plane bending vibration of aromatic C-H in-plane bending vibration of aromatic C-H
PPY pure	1400 -1480 1500-1600 2000-3000	(1459) (1542) (2120)	stretching vibration C=C stretching C=N Pyrrole Cleavage
	3000-3500	(3653)	The second ary amines N–H stretching vibrations

Sample	Vibration Bondrange (1/cm)	Observed Postion(1/cm)	E spected vibrations
	500-600 600-900 1000 1188	(564) (860, 777,670) (1035, 1196)	presence of polypyrole out-of-plane bending vibration Aromatic C-H in-plane bending vibration aromatic C-H
PPY /AgNps 5%	1400 - 1480	(1459)	stretching vibration C=C
	2000-3000 3000-3500	(2153) 3669	stretching C=N Pyrrole Cleavage The secondary amine's N–H stretching vibrations

Sample	Vibration Bondrange (1/cm)	Observed Postion(1/cm)	Expected vibrations
	500-600 600-900 1000 - 1188	(511) (771,670) (1063, 1166)	presence of polypyrrole out-of-plane bending vibration Aromatic C–H in-plane bending vibration Aromatic C–H
PPY/AgNps 10%	1400 -1480 1500-1600 2000-3000 3000-3500	(1459) (1536) (2125) (3698)	stretching vibration C=C stretching C=N Pyrrole Cleavage The secondary amine's N–H stretching vibrations

Sample	Vibration Bondrange (1/cm)	Observed Postion(1/cm)	Expected vibrations
	500-600 600-900 1000 1188	(525) (719 ,672) (1039)	presence of polypyrole out-of-plane bending vibration Aromatic C-H in-plane bending vibration Aromatic C-H
PPY/AgNps 15%	1400 - 1480	(1442)	stretching vibration C=C
	1500-1600 2000-3000 3000-3500	(1535) (2156) (3754)	stratching C=N Pyrrole Cleavage The secondary amine's N-H stratching vibrations

3.2 XRD Analysis for (purePPY) and AgNPs

One popular experimental method for figuring out lattice parameters is Xray diffraction. The XRD pattern of the synthesized (PPY) undoped powder is shown in Figure 7. This indicates that the material is amorphous. The distinctive peak of polypyrrole was found at (2θ) 24.35°, indicating the short-range arrangement of PPy chains. We used Braggs law to determine the interlayer spacing (d) for each sample. [22].

$$n\lambda = 2dSin\theta \tag{1}$$

The wavelength of XRD, represented by n, is

0.15406 nm. and the angle θ between the scattering planes and the incident light. To estimate the amount of interlayer spacing (d), the 2θ value of the peak with the highest prominence was utilized. From equation (1), the interlayer distance (d) between the measured value of spacing for PPY was 3.6599 A0. The XRD pattern of the Ag's four characteristic peaks of the face-centered cubic (fcc) crystalline structure of AgNps is shown in Figure 8, which is consistent with previously published data[23].namely (111),(020), (022), and (131) at $2\theta = 38.11,44.3, 64.4$, and 77.40 (JCPDS file no. 04-0783).



Fig.7. X-Ray diffraction for PPY (undoped)

3.3 XRD Analysis for PPY/AgNPs thinfilm

Figures (9, 10, and 11) display the XRD spectra of the PPY/Ag thin films synthesized at various addition ratios (5, 10, and 15) in weight percent. Following addition, showed peaks for the (5%) ratio centered at 24.595, 38.778, 44.930, 65.027, and 77.928 with d-spacing to be (0.36217nm), (0.23235nm) ,(0.20186nm) ,(0.14350nm) and (0.12266nm), respectively . The 10% ratio displayed peaks at 23.963, 38.612, 44.783, 64.887, and 77.795, with dspacing values of (0.37158 nm), (0.20249)nm), and (0.12283),respectively. Likewise, the (15%) ratio showed peaks centered at 39.449, 45.611, 65.656, and 78.519, with dspacing to be (0.22855 nm), (0.19900 nm), (0.14228 nm), and (0.12188 nm), respectively. The (15%) ratio showed peaks centered at 39.449, 45.611, 65.656, and 78.519, with d-spacing to be (0.22855 nm), (0.19900 nm), (0.14228 nm), and (0.12188 nm), respectively. Figures 9, 10, and The crystalline size (D) can be calculated using the Debye-Scherer formula [3].

$$D \frac{k\lambda}{\beta cos \theta}$$

(2)

Where D is the crystallite's size (nm), k is the average crystallite's form factor (0.9), and these values vary depending on the shape of the crystal and the miller index of the reflecting plane. λ stands for the XRD wavelength in



Fig. 8 X – Ray for (AgNPs)

for nanometers, and β the crystallite peak's width at half maximum (FWHM), which is given in radians. In addition, θ is the XRD peak's Bragg's angle (deg). We found that the crystal size increased as the amount of silver nanoparticles in pure polypyrrole increased. This was unmistakable proof that PPY coatings may develop on silver nanoparticles. Figures 9, 10, and 11 show PPY/AgNps thin films with different amounts of doping (5, 10, and 15 wt %). The centers see fewer high intensity diffraction peaks as the doping ratio goes up. Table (3) displays the XRD data for PPY /AgNps thin films.







Fig.10. X-Ray diffraction for PPY /AgNps 10%



Fig.11. X-ray diffraction for PPY /AgNps 15%

Table 3. XRI) measurements a	are used to	calcu	ılate	the crystallin	e size (D) a	and interlayer
			•				

Sample	2θ(Deg.)	θ(Deg.)	dhkl A ⁰	β(Deg.)	D (nm.)
	24.595	12.297	0.36217	3.9729	2.0458
	38.778	19.389	0.23235	0.5036	16.717
ΡΡΥ/ΔσΝΡς	44.930	22.465	0.20186	0.5062	16.976
5%	65.027	32.513	0.14350	0.5279	17.837
570	77.928	38.964	0.12266	0.42	24.312
		11.981	0.37158	3.7314	2.1757
	23.963	19.306	0.23331	0.3905	21.548
PPY/AgNPs	38.612	22.391	0.20249	0.3974	21.612
10%	44.783	32.443	0.14378	0.317	29.681
	64.887	38.897	0.12283	0.3141	32.4787
	77.795				
	39.449			0.2951	
PPY/AgNPs	45.6118	19.7245	0.22855	0.3468	28.587
15%	65.6563	22.8059	0.19900	0.2397	24.840
	78.519	32.82815	0.14228	0.2413	39.421
		39.2595	0.121884		42.494

spacing of various samples.

3.4 AFM Analysis for PPY (undoped) and PPY/AgNPs thinfilm

The average Granular size (G.S.), root-mean-square (R.M.S.), and exterior surface roughness average (Ra) have been measured using atomic force microscopy (AFM), also known as surface morphology analysis. These measurements are thought to be among the most significant surface characteristics. All undoped and doped thin-film deposits on glass substrates with different doping percentages (5, 10 and 15)Wt% of AgNPs, produced by drop casting, show a regular granular surface nature in the 2D and 3D views, respectively, according to the AFM test. The pure PPY thin film simple figure (12) has a smooth and regular surface with small spherical nanoparticles, or globules, on it. The surface's (Ra) was 2.557 nm, its (R.M.S.) was 3.299 nm, and its (G.S.) was 33.40 nm, dispersed over the 2000 x 2000 nm2 scanned region, as well as its analytical capability (pixels = 416,424). According to table (3), these values rise as the weight ratio of AgNps. Using different weight ratios (5, 10, and 15%) of AgNPs for AFM analysis of doped PPY thin film samples figure (12) and in 2D and 3D view), it can be observed that the surface samples consisted of small clusters with (Ra) in the range of (3.397 -13.87) nm, (R.M.S.) in the range of (4.859-19.28) nm, and (G.S.) in the range of (46.12-86.89) nm,

respectively. Table (3) displays the (G.S.) for each thin film; it is observed that (G.S.) increases with increasing additional percentages, leading to surface uniformity; this observation is consistent with XRD testing. Additionally, Table (3) indicates that Ra increases with increased adding ratios due to high surface area for both pure and doped thin films as well as R.M.S. However, the as adding percentages rise. it becomes apparent that the surface is coarse. The rises in (R.M.S.) relate to crystalline development that occurs more in a vertical direction than a horizontal one. these result was in good agreement with literature [15]



Fig. 12. AFM pictures of synthetic, PPY undoped thin films in (A) 2 and (B) three dimensions, AFM photos of the PPY /AgNps thin films that were made at a ratio of 5% for (C) 2-D and (D) 3-D, . AFM

photos of the ppy/ AgNps thin films that were made at a ratio 10% for (E) 2-D and (F) 3-D and AFM photos of the PPY/AgNps thin films that were made at a 15% for (G) 2-D and (H) 3-D.

Sample	Ra(nm)	R.M.S (nm)	G.S(nm)
PPY (pure)	2.557	3.299	33.40
PPY/AgNPs 5%	3.397	4.859	46.12
PPY/AgNPs 10%	9.328	11.90	66.33
PPY/AgNPs15%	13.87	19.28	86.89

4.Conclusions

The chemical oxidative polymerization method successfully synthesized PPY by using pyrrole as a monomer. We can dope PPY with AgNPs at various additional ratios (5, 10, and 15 percent) using the drop casting process, an effective method for producing thin films. We use FT-IR, XRD, and AFM to

estimate the surface shape and characteristics structural of the produced thin films. The PPY/AgNPs composites' FTIR spectra resemble those of PPY(undoped), yet some of the bands have wave number shifts because of the interaction between PPY and Ag. If the amount of doped PPY/AgNps thin films is increased, the high-intensity diffraction peaks in the middle get weaker. This indicates that the presence of AgNPs lowers the

proportion of ppy in the composite, which in turn weakens the PPY diffraction peaks. Through the use of (AFM), the (PPY/AgNPs) thin films showed an increase in the values of (R.M.S.), (Ra.S.), and (G.S.) as the doped ratio increased. The addition of silver nanoparticles most effectively affects surface topography features. In fact, the best Ag addition is 15%, compared to 5% and 10% for all characteristics. the results of these studies indicate that the obtained material is polymer PPY. A great technique for preparing polymeric thin films that are both undoped and doped with AgNPs at different weight percentages (5, 10, and 15 Wt%) over glass substrates is the drop coasting approach.

5. References

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