

Research Article

An Investigation into the Influence of Silver Nanoparticles on the Refractive Index of AgNO₃-Doped PVP/PVA Nanocomposites via the Refractive Index-Band Gap Energy Paradigm

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Article Info	Abstract
Article History	A nanocomposite consisting of a Polyvinylpyrrolidone (PVP) and polyvinyl alcohol (PVA) blend doped with silver nitrate (AgNO ₃) was prepared using a combination of chemical reduction and solution casting, with varying silver nanoparticle (Ag-NP) contents. Polymer nanocomposites were prepared by adding AgNO ₃ (0, 10, 20, and 30 wt.%) to a PVP/PVA host polymeric matrix with a weight ratio containing 60% of (PVP) and 40% of (PVA). The UV-Vis-NIR absorption spectroscopy was employed to examine the optical properties of the prepared films. The impact of various AgNO ₃ concentrations on the direct and indirect band gap energies was obtained from Tauc's plots, and the refractive index was determined using the optimal formula, as reported in previous studies, which provides an accurate correlation between refractive index and band gap energy. The structural and crystallinity degree were examined by XRD analysis. SEM images showed the nanoparticle formations. The discussion focused on the discovery that the refractive index change increased substantially with increasing salt concentration, consistent with the formation of silver nanoparticles.
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1. Introduction

Polymeric materials are used in various applications, including energy storage and conversion devices such as supercapacitors, batteries, and fuel cells, as well as display technologies like electrochromic displays. Nowadays, scientists have closely studied polymeric materials owing to their wide range of uses [1-3].

An extensive effort was undertaken to improve the optical characteristics of polymers. One such method is the blending of several polymers, which has the potential to enhance the effectiveness of polymeric materials [4-6], and nanoparticle formation is a particularly relevant way to enhance the optical, thermal, and electrical properties of polymeric materials [7-9].

Polyvinyl alcohol (PVA) and Polyvinylpyrrolidone (PVP) are well-known polymers used in optical devices due to their convenient fabrication processes, flexibility, biodegradability, environmental stability, transparency, non-toxicity, and resistance to ultraviolet (UV) radiation [10, 11]. PVA is a semi-crystalline polymer [12]. The excellent water solubility of PVP makes it a valuable component in casting solutions for preparing PVP-PVA polymer blends [13]. A thorough analysis was carried out on blends containing varying proportions of the two polymers to assess modifications in their physical, electrical, and optical characteristics, and to identify the best blend for a given purpose [14-16].

2. Literature Review

In a study conducted by Abou_Taleb [17], the addition of PVA to PVP resulted in changes to the crystalline structure and optical characteristics, and in an enhancement of the infrared absorption spectrum range. Rajeswari, et al. [18] studied the ionic conductivity of electrolytes comprising polymer blends based on PVA-PVP-NH₄NO₃. Also, an investigation by Al-Muntaser, et al. [19] focused on the electrical and optical characteristics of PVA/PVP/CMC/ZnO₂ nanocomposite films with varying ZnO₂ content. Badawi [20] prepared polymeric nanocomposite films of PVA/PVP doped with SnS at various ratios and determined the optical characteristics for each addition of SnS.

Silver nanoparticles (AgNPs) possess remarkable antimicrobial, electrical, and optical properties, making them suitable for photonics and electronic devices [21-25]. On the other hand, Duraikkan, et al. [26] and Abd El-Kader, et al. [27] investigated the effect of AgNO₃ doping on a PVP/PVA polymer blend by examining its structure, electrical properties, and antibacterial activity. Furthermore, the band gap energy and refractive index are two essential physical factors that determine the medium's electrical and optical characteristics [28]. In addition, prior knowledge of the refractive index and band gap energy is necessary for devices such as photonic crystals, waveguides, solar cells, and detectors [29].

The refractive index is a fundamental parameter across a diverse array of applications, including optoelectronics, linear and nonlinear optics, optical switching, photovoltaic devices, photodetection, laser technology, optical sensing, and integrated photonic circuits [30-32]. Extensive research efforts have been dedicated to formulating and establishing correlations between the optical refractive index and the energy band gap, with a focus on developing simplified relationships such as those of Hervé and Vandamme [33] and Akaltun, et al. [34] and Ravindra, et al. [35].

Polymeric materials with high refractive indices are extensively employed in optical components and apparatus, including anti-reflection coatings, optical waveguides, and lenses [36-39]. For these applications, a transparent polymeric material with a refractive index (RI) of at least 1.8, ideally exceeding 1.9, across the entire visible spectrum, is required. Unfortunately, there isn't a polymer like that, but the polymer with the highest refractive index is now 1.76, investigated by Naccarato, et al. [40].

One approach to enhance the refractive index (RI) of polymers involves incorporating nanoscale high-refractive-index (HRI) inorganic semiconductors, such as ZnS, TiO₂, ZrO₂, PbS, and CdS, into the polymer matrix to form nanocomposites [41]. In contrast, Jalal [42] previously worked on a PVP/PVA nanocomposite based on AgNO₃. The author focused on analyzing the structures and optical properties, specifically the absorption, band gap and Urbach energies, and the absorption coefficient, but not the refractive index.

In contrast to our earlier work, which addressed the absorption edge, Urbach energy, and absorption coefficient, this study examines direct and indirect band gaps. It evaluates the refractive index using the Goma correlation to assess the potential of PVP/PVA–AgNP nanocomposites as high-refractive-index materials. This study aims to investigate the refractive index of PVP/PVA–AgNP nanocomposites and evaluate their potential as high-refractive-index polymer materials for optical applications.

3. Materials and Methods

The used materials were PVA (31,000 g/mol) and PVP (24,000 g/mol) powders, supplied by Carl Roth GmbH + Co. KG, Karlsruhe. High-purity AgNO₃ (>99.5%) was supplied by BIOCHEM Chemo pharma, France. For the experiments, PVP, PVA, and AgNO₃ were dissolved in distilled water, sourced from the Physics Research Laboratory at the University of Halabja.

3.1. Synthesis of Nanocomposite Polymer Films

A solution-casting method was used to prepare PVP/PVA–AgNO₃ nanocomposites, in which separate solutions containing 1 g of PVP and 1 g of PVA in 25 mL of distilled water were initially prepared. Following separate dissolution, the PVP solution was magnetically stirred at ambient temperature, whereas the PVA solution was stirred at 90° C at 800 rpm for approximately 24 hours to ensure complete homogeneity. Subsequently, a blend incorporated by mixing the solutions in a 0.6:0.4 weight ratio of (PVP: PVA) was prepared, with continuous stirring on a magnetic stirrer hot plate for one hour to achieve a

homogeneous mixture.

The final step involved Ag nanoparticle (Ag-NP) formation; it was completed by gradually adding AgNO₃ solutions (10, 20, and 30 wt.%) to the PVP/PVA blend at 90 °C under continuous stirring. A change to brown within 30 minutes indicated successful nanoparticle synthesis [42, 43]. Film formation was achieved by casting the solutions onto Petri dishes (8 cm in diameter), which were then left to dry at room temperature.

The obtained samples were coded as (BSN0, BSN1, BSN2, and BSN3) according to the (0, 10, 20, and 30 wt.%) concentration of AgNO₃, respectively. The samples ranged in thickness from about 75-150 μm , the thicknesses were measured at several locations on the films under examination, since they do not possess a uniform thickness throughout. This variability is attributed to the casting procedure used and the subsequent drying at ambient temperature. Double beam UV-Vis spectrophotometer (Model: Lambda 25) from Perkin Elmer was used to record the absorbance spectrum of samples in the wavelength range between 190 nm and 1100 nm.

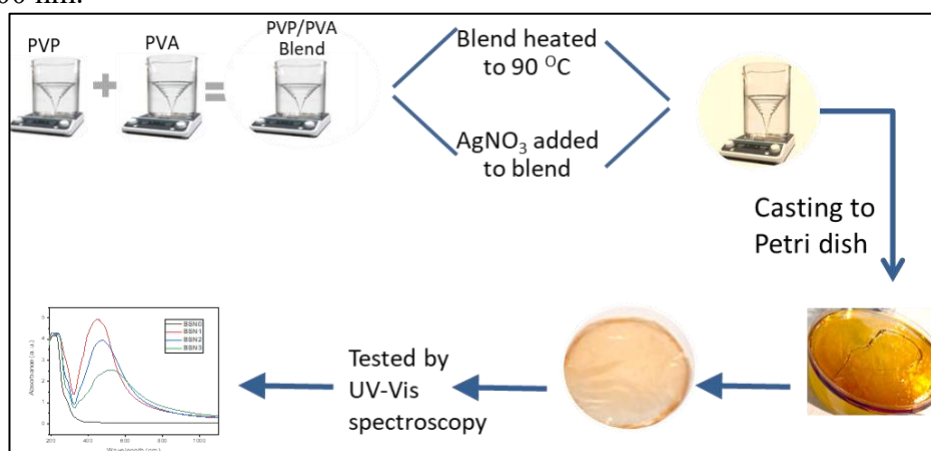


Figure 1. Schematic illustration of the preparation of PVP/PVA–AgNO₃ nanocomposites

4. Results and Discussion

Optical absorption spectra are a fundamental technique employed in optics to investigate the band-structure characteristics of polymer nanocomposite films [44, 45]. Figure 2 shows the absorbance spectra of the prepared samples, as recorded by the UV-Vis-NIR spectrometer. A peak at 219 nm appears, corresponding to $n-\pi^*$ transitions, characteristic of unsaturated bonds, consistent with findings by Ragab, et al. [46] and Jalal [42]. The absorption peak at 219 nm exhibits increased intensity and broadening with higher AgNP content, attributed to AgNP-PVP/PVA interactions. This observation aligns with existing research [47-51], reflecting the impact of particle size, dielectric environment, and chemical conditions on

absorption properties.

By analyzing the fundamental absorption edge, where absorption rises significantly, we can determine the band gap energy and identify the type of electronic transitions [42, 52, 53]. The absorption coefficient α quantifies the attenuation of light as it passes through a material, and its value can be evaluated from the optical absorbance (A) spectrum by using the famous Beer-Lambert law [52, 54-56].

$$\alpha = 2.303 \left(\frac{A}{t} \right) \quad (1)$$

where t is the sample thickness, the Tauc model [57] provides a framework for determining the band gap energy by correlating the absorption coefficient (α) with the photon energy ($h\nu$).

$$\alpha = \beta (h\nu - E_g)^\gamma / h\nu \quad (2)$$

The coefficient β is structure-dependent, while E_g corresponds to the optical band gap energy. The exponent γ values (1/2, 3/2, 2, 3) determine the electronic transition types: direct-allowed, direct-forbidden, indirect-allowed, and indirect-forbidden, respectively, and h denotes Planck's constant [3, 46, 58-60]. From Figure 3, the direct band gap energy was determined, and from Figure 4, the indirect band gap energy was determined. The relationship between optical absorption and electronic transitions across the energy gap E_g is widely recognized.

From this perspective, the relationship between the index of refraction and the optical band gap became apparent [34, 35]. The optimal formula, as determined by [28], was selected because a meticulous examination of previous research correlating the optical refractive index and band gap concluded that the Reddy approximation [61] was adopted to produce a new figure that offers an exceptional fit to the experimentally reported data. Consequently, it is employed directly as an accurate formulation in theoretical studies. Also, it provides an excellent match to the experimental data, including both low and high energy ranges, which is a notable success compared to prior relationships.

$$n = \sqrt{\frac{A}{E_g^{0.5}} - B} \quad (3)$$

where $A = 3.44^2$ and $B = \sqrt{3.44}$.

Table 1 summarizes the effect of Ag-NPs on optical parameters of PVP/PVA nanocomposites, specifically the direct band gap energy (E_{gd}), indirect band gap energy (E_{gi}), and the refractive indices. The addition of 30 wt.% AgNO₃ to the PVP/PVA blend results in a substantial decrease in E_{gd} (from 4.52 eV to

1.74 eV) and E_{gi} (from 4.05 eV to 1.03 eV), consistent with prior studies demonstrating salt-induced band gap reduction in polymer nanocomposites [11, 38, 46, 55, 62, 63]. A clear trend emerges: increasing AgNO₃ concentration decreases both the direct and indirect band gap energies, accompanied by a corresponding increase in refractive index. The refractive index of the PVP/PVA blend rises from 1.93 to 2.67 and from 2 to 3.13 upon addition of 30 wt.% AgNO₃, correlating with the E_{gd} and E_{gi} values, respectively.

The high refractive index exhibited by PVP/PVA-Ag-NPs composites with 30 wt.% AgNO₃ makes them promising materials for diverse applications, including LED and OLED coatings, antireflective films for display devices, optical storage disc cover layers, and CCD image sensor micro-lens arrays, highlighting their potential utility in cutting-edge optoelectronic technologies [36, 40, 41, 64, 65].

Table 1. Direct E_{gd} and indirect E_{gi} band gap energies, refractive index, and degree of crystallinity of PVP/PVA nanocomposite samples with varying AgNO₃ contents (wt%)

Sample code	E_{gd} (eV)	Refractive index $n(E_{gd})$	E_{gi} (eV)	Refractive index $n(E_{gi})$	Crystallinity degree (X_c) %
BSN0	4.53	1.924886	4.06	2.004543	53.5
BSN1	2.12	2.504521	1.5	2.794167	45.2
BSN2	1.89	2.598645	1.19	2.998853	43.7
BSN3	1.74	2.66764	1.03	3.131337	34.2

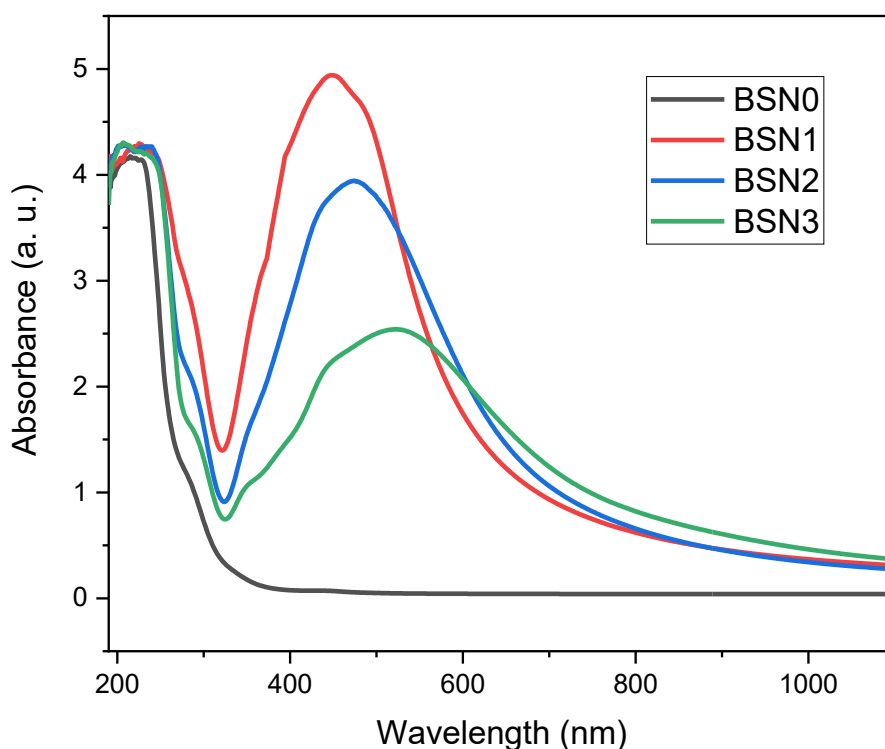


Figure 2. Absorption spectra of PVP/PVA-AgNO₃ nanocomposites with varying AgNO₃ concentrations

The high refractive index values obtained in this study (up to approximately 3.1) are derived from an empirical model and may therefore differ from values measured directly through dispersion experiments. Consequently, future work employing direct optical characterization techniques, such as spectroscopic ellipsometry or prism-coupling methods, would be beneficial for validating the refractive index values inferred from the band gap energy (E_g).

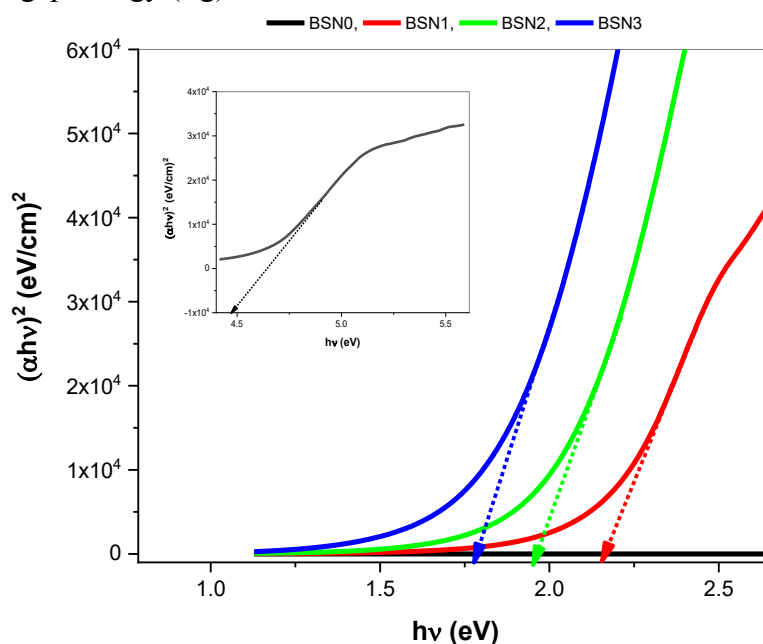


Figure 3. Tauc plots of $(\alpha hv)^2$ versus hv used to determine the direct band gap energy; the inset shows the pristine sample (BSN0)

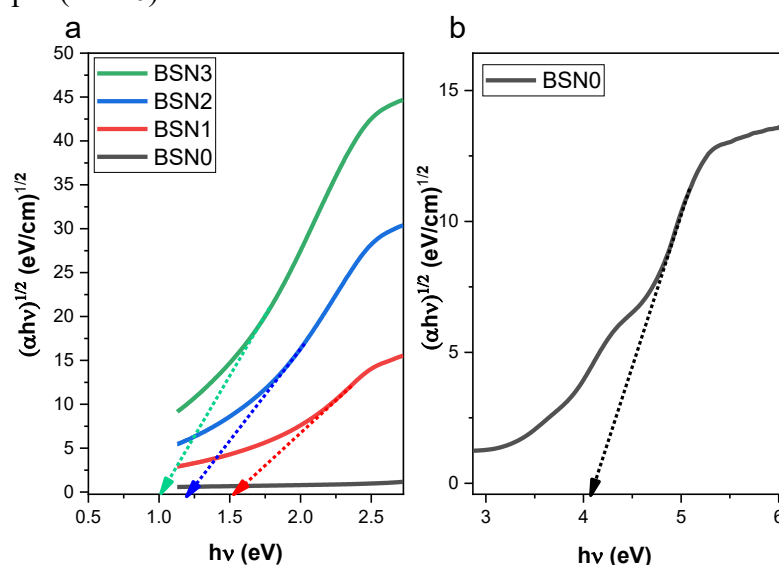


Figure 4. Tauc plots of $(\alpha hv)^{1/2}$ versus hv are used to determine the indirect band gap energy

4.1. XRD Analysis

XRD spectra confirmed the Ag-NPs formation. Figure 5 shows the XRD pattern for the pure PVA/PVP blend (BSN0). The diffraction peak is at $2\theta=20.19^\circ$, which is in line with other reports [10, 46,

66], and the blend doped with AgNO₃ at different concentrations exhibits a semi-crystalline structure. According to the reduction of inter-planar spacing, the diffraction peaks from $2\theta = 20.19^\circ$ changes to $2\theta = 22.15^\circ$. This variation could appear by interactions between Ag-NPs and PVA/PVP blend, which leads to an increase in the amorphous region. The same results appeared by Ragab, et al. [46], Jalal [42], Yang [67]. The crystallinity degree X_c can be determined by using the Hermans and Weidinger equation [68]:

$$X_c = \left(\frac{A_c}{A_T} \right) \times 100 \quad (4)$$

where A_T represents the area of both crystalline and amorphous regions and A_c represents only the crystalline area. The crystallinity degrees of the prepared samples are shown in Table 1. A decrease in crystallinity from 53.5% to 34.2% indicates a good reaction between Ag-NPs and the polymer blend. The peaks obtained between $2\theta = 35^\circ - 40^\circ$ describe the formation of silver nanoparticles [27, 69].

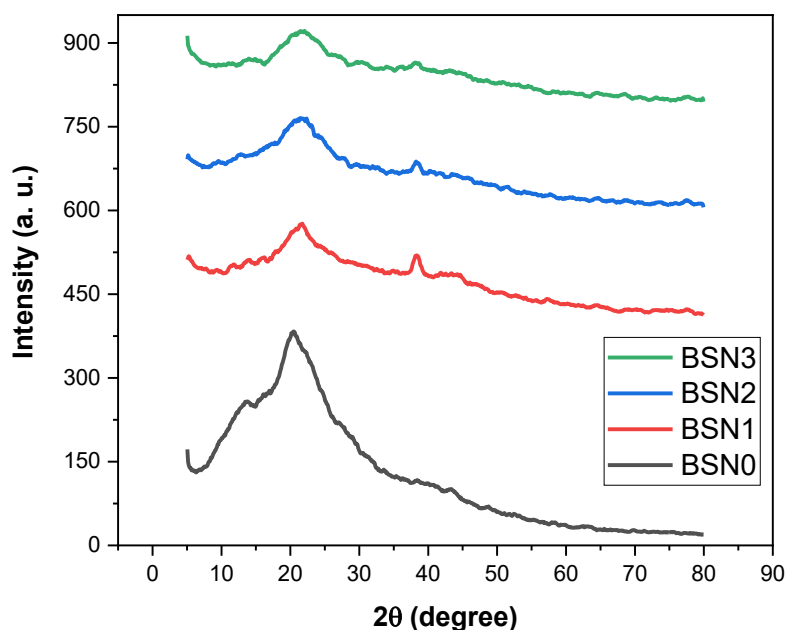


Figure 5. XRD patterns for PVA/PVP-AgNO₃ nanocomposites

4.2. Scanning Electron Microscopy (SEM)

The SEM analysis provided a detailed examination of the film's surface structure, phase separation, and defects. SEM images of the nanocomposite thin films were taken at 10kx magnification and are presented in Figure 6. As shown in Figure 6a, the PVP/PVA blend (BSN0) exhibited a smooth surface and a uniform structure, consistent with prior research [27, 70]. The incorporation of Ag-NPs into PVP/PVA films (Figure 6b-d) resulted in a uniform surface distribution of nanoparticles, forming additional particles

with notable roughness. The improvement is attributed to the Ag nanostructures embedded in the blend matrix, with optimal results at 30% AgNO₃ concentration (Figure 6d). The synthesis of Ag-NPs within polymer matrices via the introduction of AgNO₃ has been widely explored and improved [71-74]. SEM images were obtained to complement the XRD results and provide insight into the optical properties observed in the UV-Vis analysis.

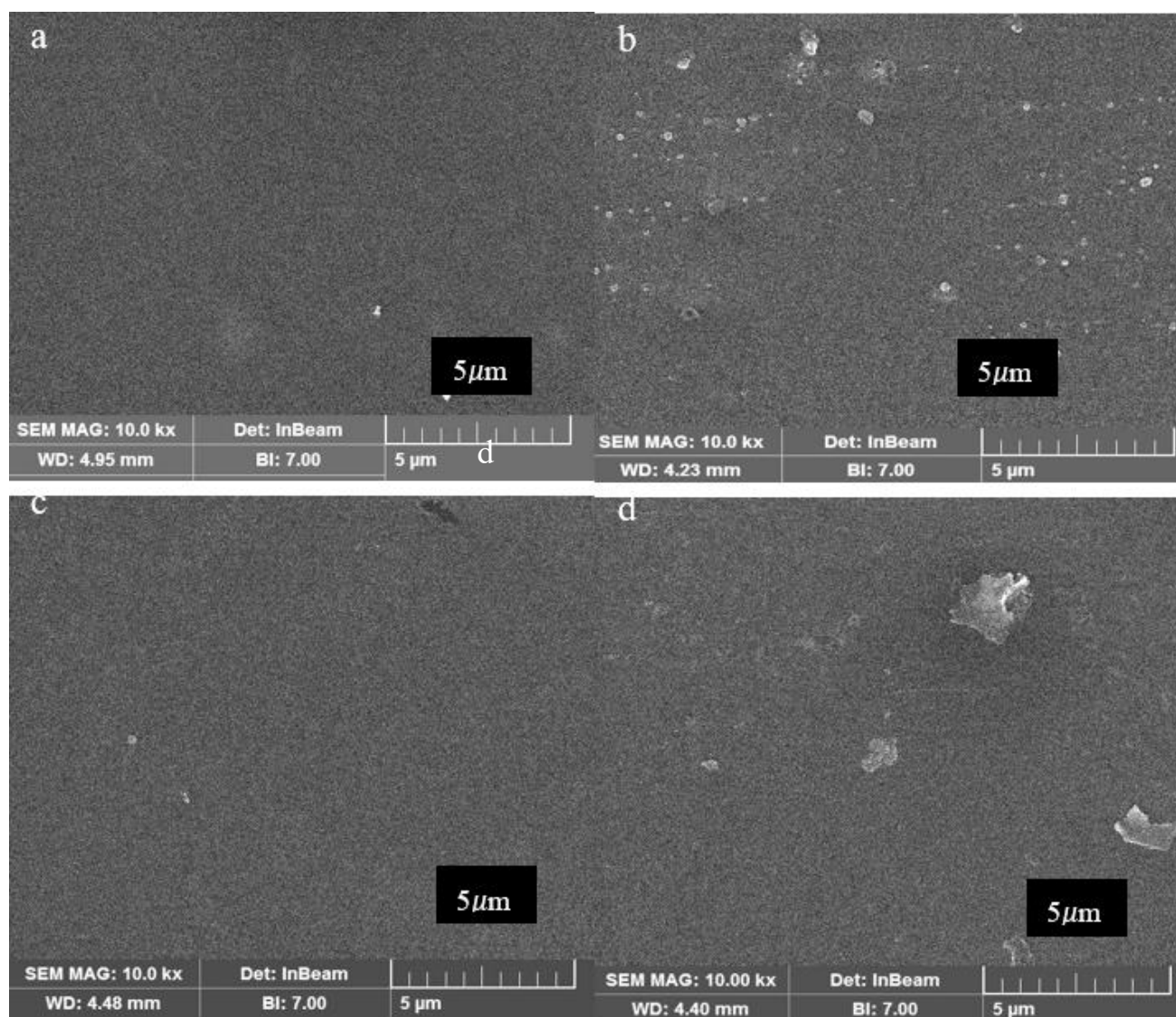


Figure 6. SEM images of PVP/PVA blend (a), PVP/PVA-AgNO₃ 10 wt. % (b), 20 wt. % (c), 30 wt. % (d)

5. Conclusion

In this study, PVP/PVA-Ag-NPs nanocomposite thin films were synthesized using the casting method at ambient temperature. The optical characteristics of PVP/PVA: AgNO₃ nanocomposites, specifically the band gap and refractive index, were determined by UV-Vis absorbance spectroscopy. The direct band gap energy decreased from 4.53 eV to 1.74 eV, and the indirect band gap energy decreased from 4.06 eV to

1.03 eV, as the AgNO₃ content increased to 30 wt.% in the PVP/PVA blend matrix, due to the introduction of additional energy levels. Moreover, the refractive index for PVP/PVA blend was about 1.93 for direct transition, and 2.67 for 30 wt.% AgNO₃-loaded PVP/PVA nanocomposite. Furthermore, for indirect transitions, the refractive index of the PVP/PVA blend was about 2, whereas that of the 30% AgNO₃-reinforced PVP/PVA nanocomposite was about 3.13. The XRD patterns indicate that the crystallinity of the PVA/PVP blend decreased from 53.5% to 34.2% with increasing AgNO₃ content to 30 wt.%, indicating the formation of Ag-NPs. The developed system shows potential for use in various optical applications, including LED/OLED encapsulation, display device antireflective coatings, optical storage disc protection, and CCD image sensor micro-lens arrays, etc., due to its high refractive index in both direct and indirect electron transitions. Future research could focus on nanoparticle formation to maintain high refractive index materials.

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