



Optical and Structural Modification of Polyvinyl Alcohol Induced by MoS₂ and WS₂ Nanomaterial Incorporation

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التعديل البصري والتركيبى لبوليمر بولي فينيل الكحول الناتج عن دمج MoS₂ و WS₂ المواد النانوية

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ABSTRACT

As a semi-crystalline polymer, polyvinyl alcohol (PVA) has been extensively used in optical/opto-electronic applications owing to its excellent transparency, chemical stability, and processability. However, the large optical band gap of PVA limits efficient light absorption in the visible region. The optical, structural, and morphological properties of PVA thin films have been systematically tuned by embedding semiconductor MoS₂/WS₂ and its hybrid forms via solution casting to obtain uniform ≈ 150 nm-thick films. X-ray diffraction studies indicated that the hexagonal crystalline nature of MoS₂ and WS₂ was retained in the PVA matrix and that the hybrid nanocomposite exhibited improved ordering. Field-emission scanning microscopy revealed that well-dispersed GNPs and MWCNTs are induced by strong interactions with the nanomaterials, as evidenced by rough surfaces and larger grain sizes on the PVA film with increasing amounts of nanomaterial, especially dual-filler. Optical studies using UV-Vis spectroscopy showed a substantial increase in absorption and the absorption coefficient, and a decrease in transmittance of the nanocomposite films. The optical band gap as calculated from Tauc plots corresponding to direct transitions went down monotonically from 4.1 eV for neat PVA to 3.8 eV for PVA/MoS₂, 3.68 eV for PVA/WS₂, and finally 3.5 eV for the PVA/MoS₂/WS₂ hybrid, reflecting the emergence of localized electronic states and synergy between the two nano-materials.

Key words: Polyvinyl alcohol (PVA), Nanomaterial, molybdenum disulfide (MoS₂), tungsten disulfide (WS₂)



INTRODUCTION

Polyvinyl alcohol is an atactic, semicrystalline polymer with a wide range of uses due to its high biocompatibility, biodegradability, favorable mechanical properties, excellent optical properties, and non-toxicity [1–3]. Non-corrosiveness, high optical transmission, water solubility, and thermal stability are some of polyvinyl alcohol's other outstanding qualities [4]. These characteristics, particularly its optical properties such as the energy gap and refractive index, support its industrial and technical applications as a sensor component, supercapacitor, solar cell, optoelectronic device, and coating [5,6]. The presence of hydroxyl groups on the polyvinyl alcohol backbone facilitates hydrogen bonding with other materials, aiding composite formation [6,7]. Because it is widely available, reasonably priced, possesses various volatile functional groups, and is hydrophilic, polyvinyl alcohol is of great interest. It provides consistent, high-optical-quality films for nonlinear optical sensors and instruments, and it has a strong dielectric. The flexibility of polyvinyl alcohol chains is increased by temperature dependence and intramolecular or intermolecular connections [8]. These characteristics fortify the polyvinyl alcohol matrix, making it a feasible composite for optoelectronics and electrical devices. To alter and customize polyvinyl alcohol's optical characteristics for specific uses, fillers were added as dopants [8].

To achieve improved mechanical, chemical, electrical, optical, and physical characteristics, a polymeric composite incorporates reinforcements and a filler into its fundamental polymer matrix [9]. The characteristics of the filler amount, the original polymer, the interaction between the filler and the polymer, and the type, shape, and size of the modified composite all significantly impact the properties of these polymeric composites [8]. Natural fibers, polymeric fibers, glass fibers, volcanic minerals, chalk, carbon fiber, and metal powders are some examples of fillers [10]. Polyvinyl alcohol's hydroxyl groups on its carbon backbone facilitate the formation of composites with various materials and provide a practical means to improve its optical properties (energy gap and refractive index) [11]. Because it affects and is related to magnetic, optical, and electrical features, the index of refraction represents one of the most significant optical characteristics of any polymer [12]. For the production of optical fibers and waveguide films, among other uses, it is crucial to have a sufficient understanding of the refractive indices of polymers [13]. Because they may boost light output by decreasing reflection losses, polyvinyl alcohol compounds with high refractive indices are much sought after in photonics and optics [14].

Transition metal dichalcogenides (TMD) have gained interest as nano-filler materials for devices that store energy and have begun to be utilized for the manufacturing of catalysis and optoelectronic devices [15]. Molybdenum disulfide (MoS_2) was a particularly significant TMD material among the others due to its exceptional mechanical and electrical characteristics [16,17]. Tungsten disulfide (WS_2) represents another example of this TMD category. Because of its size-dependent variable energy band gap, WS_2 may be employed for advanced uses, including photocatalysis and photovoltaics. Since both WS_2 and MoS_2 have an energy band gap of around 1.2 eV, they are both categorized as semiconductor materials. In its bulk state, MoSe_2 exhibits an indirect bandgap of 1.1 eV. MoSe_2 may be exfoliated into a few layers to provide a straight band gap of 1.5 eV [18]. As-grown MoS_2 flakes from the process of chemical vapor deposition have a modulable energy band gap between 1.86 eV and 1.57 eV [19]. Additionally, it has been found that TMDs have band gap energies between 1.5 and 1.8 eV [20,21]. For several electrical uses, nevertheless, the size of this energy band gap was inadequate [22,23]. An energy band gap of at least 1.8 eV is necessary for technological uses. One may switch between both active and inactive states via the conduction of MoS_2 at a limit that is equal to 1.8 eV for the band gap energy [24].



Auda and Idan [25] used the casting method to produce thin films of PVA doped with WS₂ nanoparticles at 0.5, 1, and 2%. The films were applied to 150 nm-thick glass substrates. The semi-crystalline nature and optical behavior of the composites have been investigated by optical and structural characterizations utilizing UV-Vis spectroscopy and X-ray diffraction (XRD). According to the XRD examination, pure PVA is semi-crystalline, and when the doping ratio increases, crystalline WS₂ phases emerge. Increased light absorption in the visible spectrum was observed in UV-Vis measurements, with a steady red shift in the absorption edge. For 2% WS₂-doped films, the direct band gap dropped from 4.1 eV for pure PVA to 4.03 eV, while the Urbach Energy rose, suggesting that structural disorder increased with doping. By lowering the band gap and enhancing light absorption, the addition of WS₂ nanoparticles to PVA dramatically alters its structural and optical properties. The findings demonstrate the potential of PVA/WS₂ nanocomposites for optoelectronic and photonic applications.

Although PVA has been considered a transparent polymer matrix, and the reinforcement of nanomaterials has been widely reported as a successful method for tuning polymer properties, the literature lacks clear insights into sulfide-based nanomaterials, such as MoS₂ and WS₂, embedded in PVA systems. The available reports in the literature are scarce and mainly focus on individual nanofillers, which contribute to an incomplete explanation of the optical or structural variations of those materials, but not all parameters at once within the same system. In addition, previous studies lack a systematic comparison of MoS₂ and WS₂ mixed in the same polymer matrix, prepared, and characterized under identical conditions. Therefore, the comparative efficiency of these nanomaterials for modifying optical absorption, bandgap behavior, and structural arrangement in PVA has not yet been fully explored. Furthermore, the correlation between nanomaterial concentration and interfacial interactions with changes in polymer chain ordering and morphology is currently poorly understood. Thus, a systematic comparative study of the synergistic effects of MoS₂ and WS₂ on the optical, structural, and morphological characterization of PVA thin films prepared using a well-established fabrication process is limited to date. This gap must be suitably bridged to form a basis for reliable structure–property relationships and for future progress towards PVA-based nanocomposites for optical/functional applications.

The objective of the present work is to improve the optical, structural, and morphological properties of polyvinyl alcohol (PVA) by adding MoS₂ and WS₂ nanomaterials and to systematically explore their effects on PVA thin films prepared by the solution-casting method. The purpose of this study is to determine the changes in photophysical behaviour (absorption spectra, bandgap modification) and the structural order (supramolecular organization, surface morphology) that occur as a result of the addition of nanomaterials into the polymer matrix. In addition, the work attempts to build a clear structure–property relationship that connects nanofiller dispersion and interfacial interactions with the optical and structural properties of PVA-based NC films, providing a theoretical reference for their further application in advanced optical and functional materials.



EXPERIMENTAL WORK

Utilized materials

- In this study, PVA was selected as the matrix due to its good film-forming properties, high optical transparency, and strong hydrophilicity. PVA is a semi-crystalline polymer containing a high density of hydroxyl ($-OH$) functional groups along the molecular main chain, leading to hydrogen bonding and effective interaction with nanofillers. These features render PVA especially suitable for introducing nanomaterials to modify its optical and structural properties. Moreover, PVA is chemically stable, mechanically flexible, and easy to process in aqueous solutions to form a uniform thin film without defects.
- The two-dimensional transition-metal dichalcogenide (TMDC) nanomaterial used was molybdenum disulfide (MoS_2). Schematic of the structure of MoS_2 : its single layers are bound by weak van der Waals forces in each plane. MoS_2 behaves as a semiconducting material at the nanoscale, with an adjustable bandgap, strong optical absorption in the visible region, and a large surface-to-volume ratio. These features enable MoS_2 to provide local electronic states and enhance light-matter interactions when incorporated into a polymer matrix.
- Tungsten disulfide (WS_2) was also used as a 2D nanomaterial with an S–W–S hexagonal structure similar to that of MoS_2 . It is chemically inert, thermally stable, has a high refractive index, and exhibits strong excitonic effects, resulting in strong optical absorption and room-temperature photoluminescence. Attributed to its well-behaved semiconducting properties and strong interfacial compatibility with polymers, WS_2 can be considered as an efficient nanofiller for adjusting the optical and structural properties of PVA.

Preparing method

First, a polyvinyl alcohol polymer liquid was prepared by dissolving 10 grams of polymer powder in 250 mL of distilled water using a stirrer and a magnetic stirrer at $30^\circ C$ for 30 minutes. The liquid was then cooled to room temperature. Second, 0.5 g of nanomaterial was added by weight to 20 mL of the prepared polymer solution and mixed for 15 minutes to homogenize the solution. Third, another 0.5 grams of nanomaterial was added to 20 ml of the prepared polymer solution, and the mixture was mixed for 15 minutes to homogenize the solution. Fourth, the nanomaterials were added to 20 ml of the prepared polymer liquid in the same proportions as above, and the mixture was stirred for 30 minutes. The casting method was used to prepare thin films with a thickness of 150 nm.

Tests

- **X-Ray Diffraction (XRD) analysis**

The crystalline structure, phase composition, and structural transformations of pure PVA and PVA-based nanocomposite thin films were examined by X-ray diffraction (XRD). Diffraction patterns were scanned over an appropriate 2θ range to identify the characteristic diffraction peaks of the polymer matrix and the encapsulated nanomaterials.

- **Field Emission Scanning Electron Microscopy (FESEM)**

Field Emission Scanning Electron Microscopy (FESEM) was utilized to examine the surface morphology, dispersion state, and microstructural evolution of pure PVA and nanocomposite



films. The FESEM images provided a high-resolution visualization of surface texture and nanofiller distribution at the nanoscale.

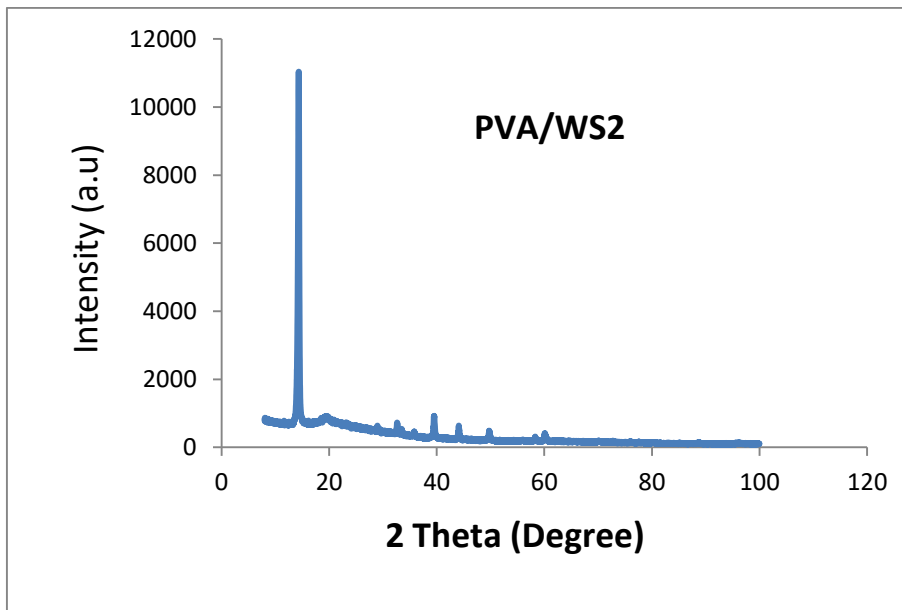
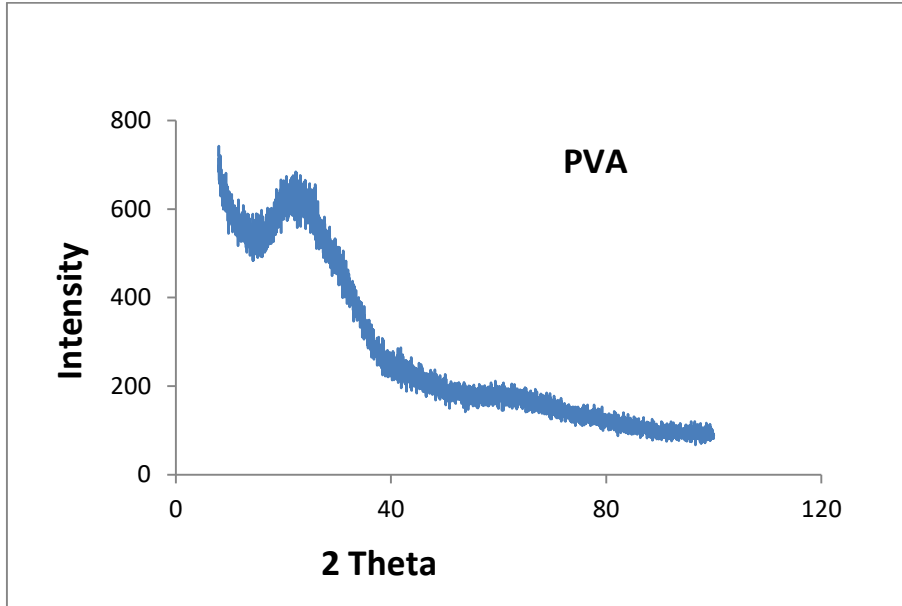
• Optical Properties

The optical properties of pure PVA and PVA-based nanocomposite thin films were investigated using UV–Visible spectroscopy to evaluate absorption behavior, optical transmittance, absorption coefficient, and optical band gap energy. The absorption spectra were recorded over the ultraviolet and visible regions of the spectrum.

RESULTS AND DISCUSSION

X-ray diffraction (XRD)

Fig. 1 (a-d) shows the XRD patterns of pure PVA and composites. PVA consists of (WS₂/PVA, MoS₂/PVA, and PVA/MoS₂/WS₂), respectively, showing a gradual change in the diffraction pattern due to the incorporation of nanomaterials. The diffraction pattern of pure PVA shows a halo at $2\theta \approx 19-22^\circ$, indicating its semi-crystalline nature. This feature is attributed to the restricted molecular ordering imposed by hydrogen bonding between hydroxyl groups along the polymer chain, with no well-defined reflections indicative of an essentially amorphous character [26]. On the contrary, in PVA/WS₂ nanocomposite, a well-defined diffraction peaks due to the hexagonal phase WS₂ are superimposed on the background of polymer. These peaks indicate the successful insertion of crystalline WS₂ into the PVA host matrix, with no disruption as a result of film casting [27,28]. The observation of attenuation in the PVA halo indicates interactions at the polymer–nanofiller interface, which would likely modify chain packing and reduce amorphous dominance, in line with earlier work on polymer nanocomposites filled with lamellar sulfide nanostructured materials [29,30]. In the PVA/MoS₂ sample, a further structural change is noticed since very sharp and intense diffraction peaks of hexagonal MoS₂ are present [31]. Much stronger and sharper peak intensities are observed in the case of WS₂ (compared to film containing WS₂), suggesting relatively higher crystallinity and preferential orientation of MoS₂-crystallites inside the polymer matrix. This behaviour is usually associated with the increased interfacial affinity and nucleation potential of MoS₂, which favours local ordering among successive polymer chains. These diffraction peaks are characteristic of both nanomaterials and confirm the simultaneous presence and uniform embedding of this material within the PVA matrix [32]. The additional attenuation of the amorphous PVA contribution and the more pronounced peak nature indicate a constructive structural cooperation between the two dual nanofillers. This kind of synergy improves the architecture and packing efficiency, thereby creating a finer ordered system. Such results are similar to those reported in studies of hybrid polymer nanocomposites, wherein complementary nanofillers enhance interfacial bonding and structural integrity.



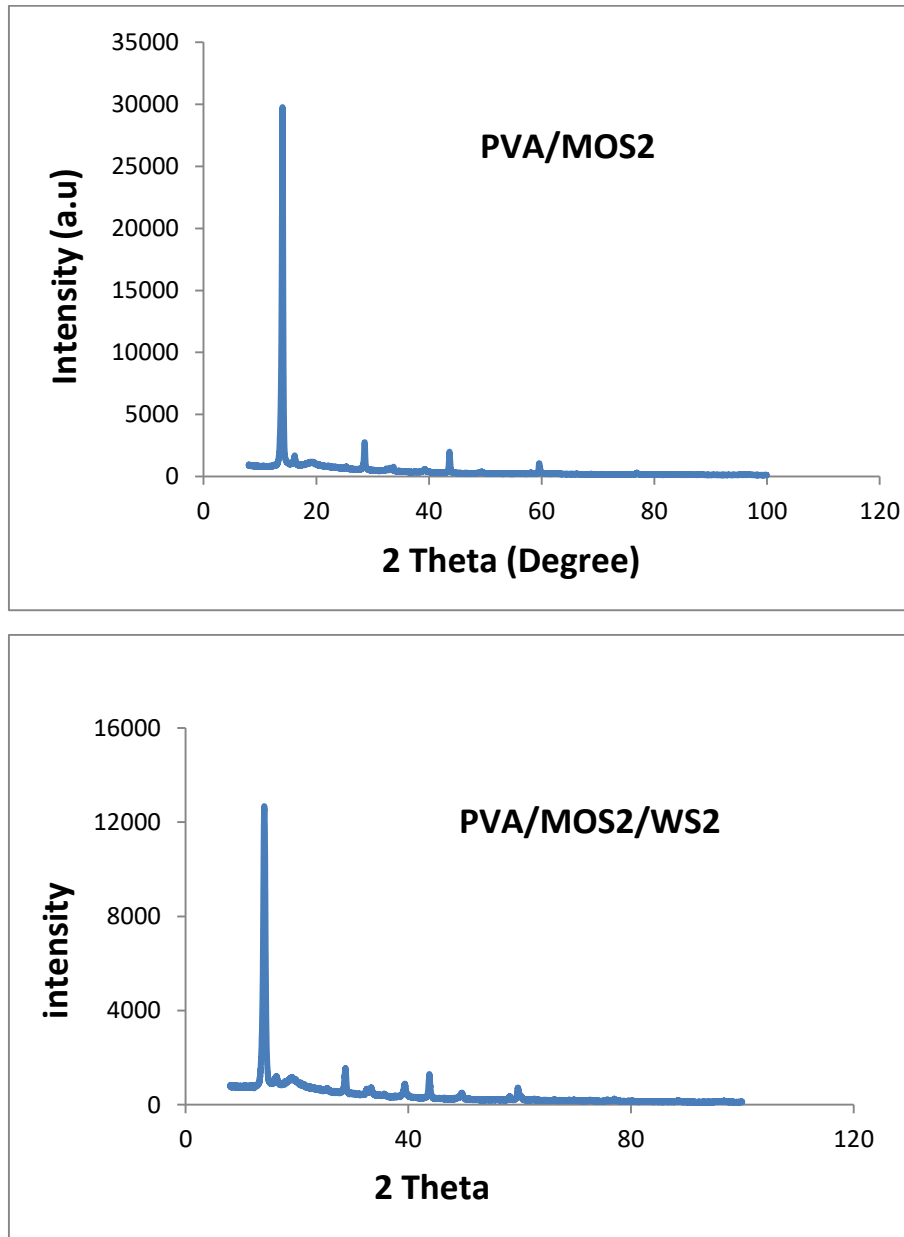


Figure 1: XRD pattern for samples consists of: a) PVA; b) PVA/ WS₂; c) PVA/ MoS₂; d) PVA/ MoS₂/ WS₂.

Field Emission Scanning Electron Microscopy (FESEM)

Figure 2 depicts FESEM images of pure PVA and its nanocomposites with WS₂ and MoS₂, as well as their respective hybrid forms, showing apparent variations in surface morphology attributed to the addition of the nanofillers. The pristine surface of PVA (Figure 2a) exhibits several smooth, compact features with few texture features, indicating that the solution-casting route can deposit a uniform film [33]. The low average grain size (51.39 nm) implies that the polymer matrix is essentially amorphous, but molecular stacking was determined by intermolecular hydrogen bonding, a characteristic of unfilled PVA films. After incorporation of WS₂ (Figure 2b), a rough surface can be observed, and some discrete granular features were

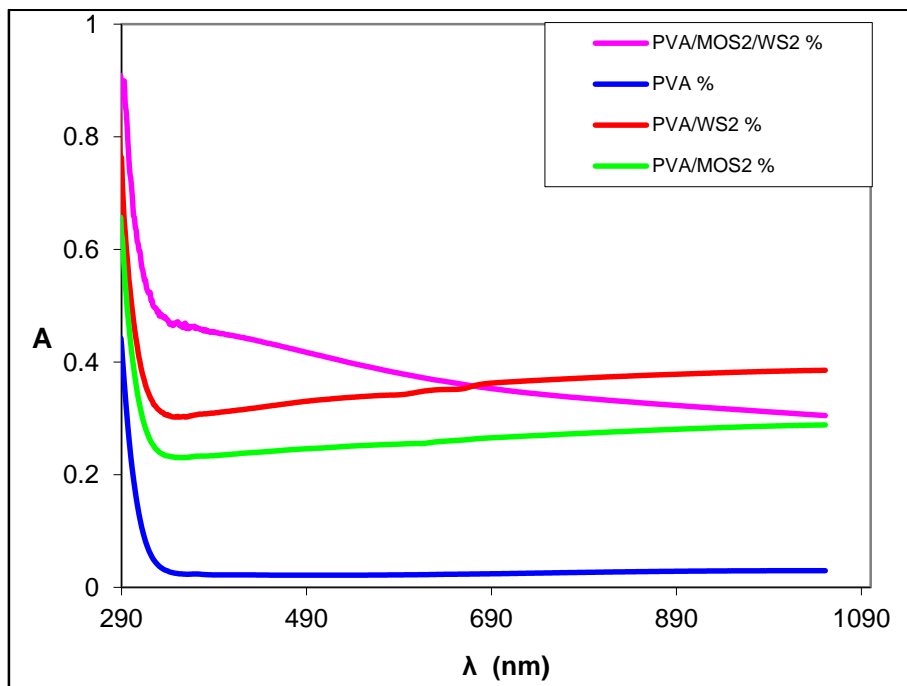


Figure 3: The absorption behavior for all selected sample mixtures (Pure PVA, PVA/WS₂, PVA/MoS₂, and PVA/ MoS₂/ WS₂).

Figure 4 shows the optical transmittance spectra of PVA and PVA-based nanocomposites containing WS₂, MoS₂, and their hybrid composite films in the wavelength range from 290 to 1100 nm. The pristine PVA film shows the highest transmittance, exceeding 85–90% in the visible region, which can be ascribed to its wide band gap, uniform microstructure, and the intrinsic transparency of the polymer matrix. In contrast, the presence of nanomaterials results in a notable decrease in transmittance across both the UV and visible regions. However, PVA/WS₂ and PVA/MoS₂ films exhibit moderate attenuation, as strong light absorption/scattering can occur with increased semiconducting character and a stratified morphology of WS₂ or MoS₂. These nanofillers generate additional electronic and excitonic states, thereby increasing photon–matter interactions within the polymer matrix. The hybrid PVA/MoS₂/WS₂ nanocomposite shows the smallest transmittance at shorter wavelengths, followed by an increase with wavelength, with a positive slope, reaching values closer to 1 in the near-IR region, demonstrating that considering both nanomaterials' presence provides a deeper understanding of their optical behavior together. This may be due to higher interfacial complexity and the density of optically active centers, which strengthen absorption and scattering mechanisms at shorter wavelengths with partial transmission at longer wavelengths.

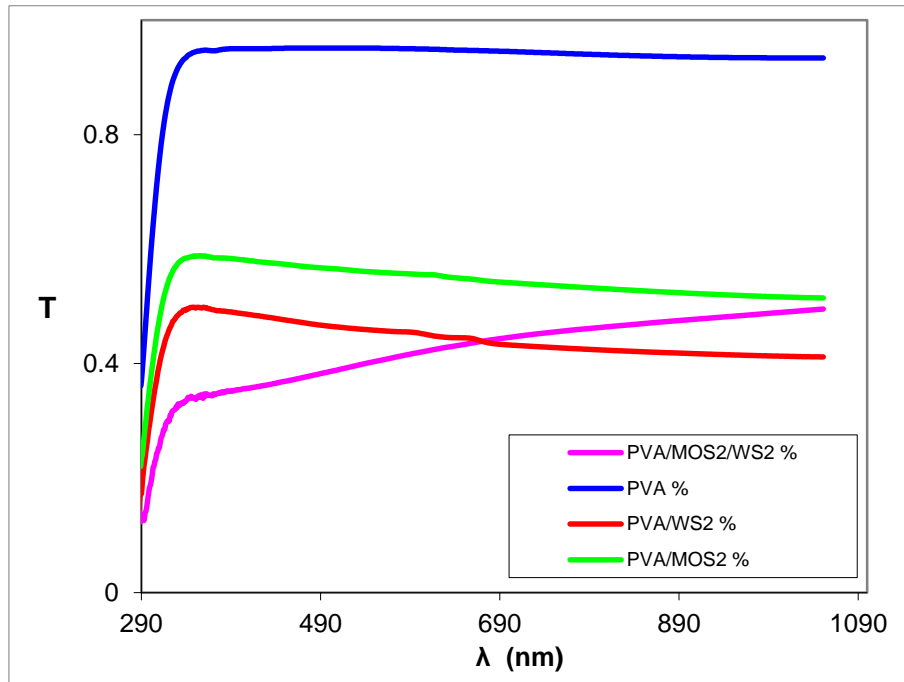


Figure 4: The optical transmittance for all selected sample mixtures (Pure PVA, PVA/WS₂, PVA/MoS₂, and PVA/ MoS₂/ WS₂).

The spectral dependence of the optical absorption coefficient (α) for pure PVA and its nanocomposites containing WS₂, MoS₂, and their hybrid system is shown in Figure 5. PVA pristine has the lowest α across the entire wavelength range studied, reflecting its large band gap, amorphous nature, and high optical transparency, as evidenced by a rapid decrease in α in the UV region, followed by upwardly constant, low values at longer wavelengths. In comparison, all the nanocomposite films have significantly higher absorption coefficients, especially at lower wavelengths, due to the addition of semiconducting nanoparticles to the polymer matrix, which can provide additional electronic states and increase light-matter interaction. The α values of the PVA/WS₂ sample are larger than those of the PVA/MoS₂ film, which may be ascribed to the strong excitonic transitions and efficient absorption characteristic of WS₂. The PVA/MoS₂ nanocomposite also has a higher value of α than that of the pure PVA, indicating that MoS₂/polymer chains possess high interfacial interaction and charge transfer. Crucially, the absorption coefficient of the hybrid PVA/MoS₂/WS₂ nanocomposite exhibits the highest value across a wide range of wavelengths, indicating that the enhanced optical response is due not only to the combined effect of the two individual nanomaterials but also to frequent defects and strong interfacial interactions. This systematic improvement in the absorption response, it is suggested, agrees with findings for PVA nanocomposites based on layered transition-metal dichalcogenides and indicates the better photon-absorbing performance of the developed films.

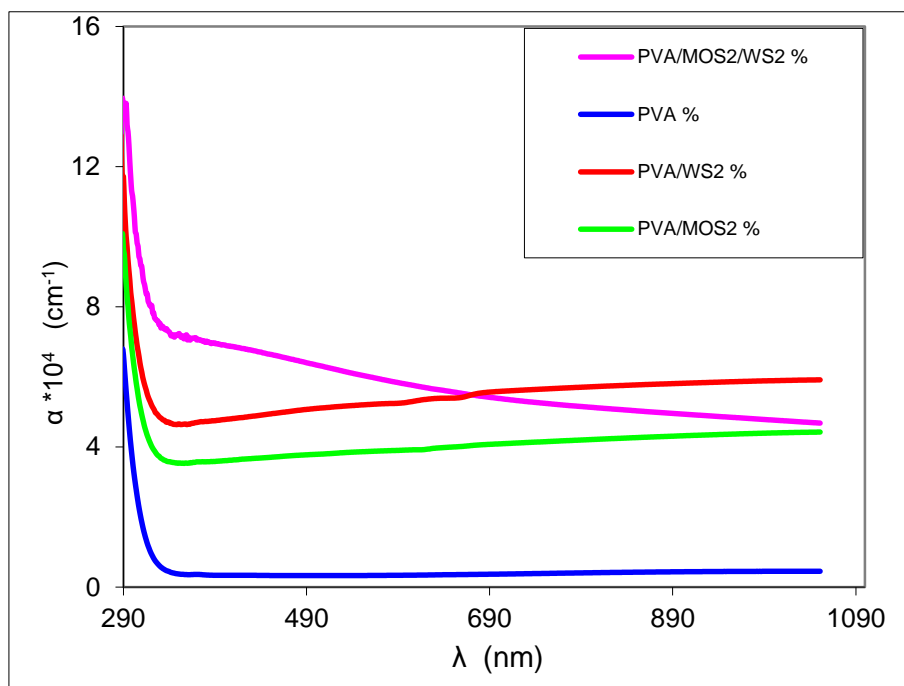


Figure 5: The absorption coefficient for all selected sample mixtures (Pure PVA, PVA/WS₂, PVA/MoS₂, and PVA/ MoS₂/ WS₂).

Figure 6 shows the analysis of the optical band gap energy (E_g) determination for pure PVA and PVA-based nanocomposites comprising WS₂, MoS₂, and their hybrid system, along with Tauc plots for allowed direct electronic transitions as a function of incident energy. The neat PVA film has a band gap energy of around 4.1 eV, characteristic of a wide-bandgap polymeric material, confirming its high optical transparency and limited conjugation. With the introduction of MoS₂ and WS₂ nanomaterials, a gradual decrease in the optical band gap is observed, with corresponding E_g values of 3.8 eV for PVA/MoS₂ and 3.68 eV for PVA/WS₂. This reduction in the band gap is ascribed to the formation of localized electronic states in semiconducting nanomaterial/PVA polymer chains, stemming from strong interfacial interactions that catalyze the tail states near band edges and stimulate electronic transitions at lower photon energies. It is worth noting that the band gap of the hybrid PVA/MoS₂/WS₂ nanocomposite (3.5 eV) is the smallest, indicating a synergistic effect arising from the coexistence of both nanomaterials. This more pronounced drop in E_g can be attributed to increased structural disorder, higher defect density, and the coexistence of electronic states in MoS₂ and WS₂, which together lead to pronounced band tailing and a smaller energy difference between the ground and excited states for electrons. This gradual reduction in the band gap by incorporating nanomaterials has been commonly observed in polymer nanocomposites containing layered transition-metal dichalcogenides. It is generally associated with enhanced light absorption and charge transfer.

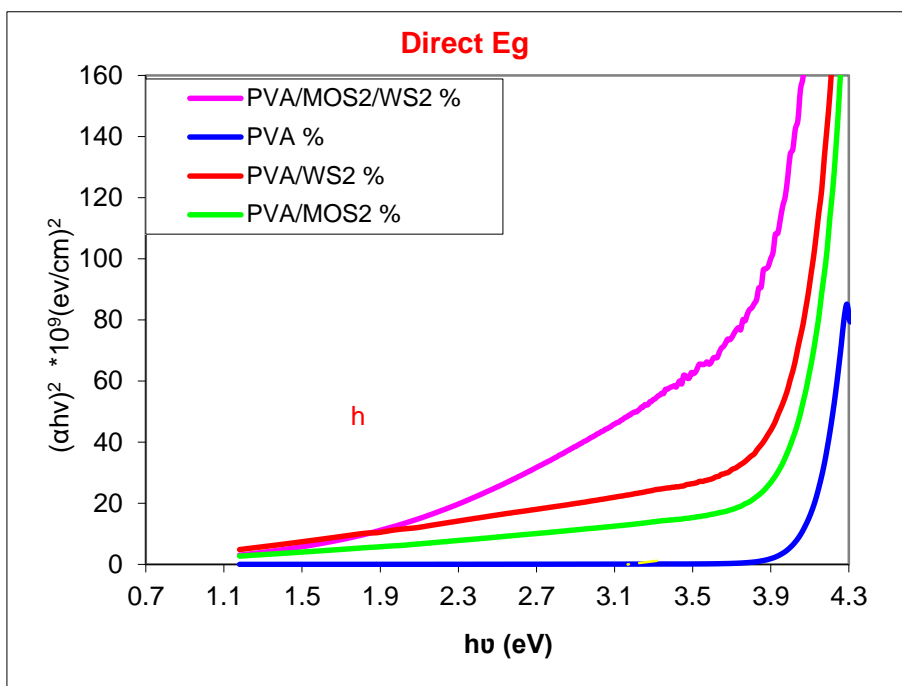


Figure 6: The optical band gap energy for all selected samples mixtures (Pure PVA, PVA/WS₂, PVA/MoS₂, and PVA/ MoS₂/ WS₂).

CONCLUSION

This work demonstrates that incorporating MoS₂ and WS₂ nanomaterials in to PVA thin films is an efficient means to modify their structural, morphological, and optical characteristics. According to the XRD pattern, the guest WS₂ and MoS₂ retained their hexagonal structure within the polymeric PVA matrix. At the same time, the mixed system of both revealed relatively enhanced structural ordering because of the combined type. The FESEM observation clearly shows a change in surface morphology from a smooth, uniform PVA film to more irregular nanocomposites with larger grain sizes, suggesting good interfacial adhesion and efficient dispersion of nano-fillers. In contrast, an incremental trend in average grain size was observed for both composites, without a single discontinuity (dual or single fillers). Optical studies indicated significant enhancements in absorbance and absorption coefficient, and a decrease in transmittance with nanomaterial incorporation, which can be attributed to stronger light–matter interaction and more optically active states. The optical band gap decreased gradually from 4.1 eV for neat PVA to 3.8 and 3.68 eV for MoS₂- and WS₂-loaded films, reaching a minimum of 3.5 eV for the hybrid nanocomposite, reflecting the synergistic effect of the dual nanofillers on the electronic properties of the polymer material. Taken together, our results illustrate a pronounced structure-property relationship and show that hybrid MoS₂/WS₂ reinforcement provides superior optical tuning relative to single nanofillers, rendering PVA/MoS₂/WS₂ nanocomposites appealing materials for optoelectronic, photonic, and light-modulating applications.



Conflict of interests.

The authors decelerate that there is no conflict of interest.

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الخلاصة

المقدمة: باعتباره بوليمراً شبه بلوري، يُستخدم بولي فينيل الكحول (PVA) على نطاق واسع في التطبيقات البصرية والإلكترونية-الضوئية نظراً لشفافيته العالية، واستقراره الكيميائي، وسهولة معالجته. ومع ذلك، فإن فجوة الطاقة البصرية الكبيرة لـ PVA تحدّ من كفاءة امتصاص الضوء في نطاق الطيف المرئي.

طرق العمل: في هذا السياق، جرى ضبط الخصائص البصرية والتركيبية والمورفولوجية لأغشية PVA الرقيقة بصورة منهجية من خلال تضمين أشباه الموصلات النانوية MoS₂ و WS₂ وكذلك أنظمتها الهجينة، باستخدام طريقة الصب بالمحلول للحصول على أغشية متجانسة بسماكة تقارب 150 نانومتر. أظهرت دراسات حيود الأشعة السينية أن الطبيعة البلورية السداسية لكل من MoS₂ و WS₂ قد حُوِّظ عليها داخل مصفوفة PVA، كما بيّنت أن المترابك النانوي الهجين يتمتع بدرجة أعلى من الانتظام البلوري.

الاستنتاجات: كشفت صور المجهر الإلكتروني الماسح بانبعثات المجال عن توزيع جيد للجسيمات النانوية داخل المصفوفة البوليمرية نتيجة التفاعلات القوية مع المواد النانوية، وهو ما انعكس في زيادة خشونة السطح وكبر حجم الحبيبات في أغشية PVA مع زيادة محتوى المواد النانوية، ولا سيما في حالة الإضافة المزدوجة. وأظهرت الدراسات البصرية باستخدام مطيافية الأشعة فوق البنفسجية-المرئية زيادة ملحوظة في الامتصاص ومعامل الامتصاص، يقابلها انخفاض في النفاذية للأغشية المترابكة. كما انخفضت فجوة الطاقة البصرية، المحسوبة من مخططات تاوك للانتقالات المباشرة، بشكل تدريجي من 4.1 إلكترون فولت لـ PVA النقي إلى 3.8 إلكترون فولت لـ PVA/MoS₂، و 3.68 إلكترون فولت لـ PVA/WS₂، وصولاً إلى 3.5 إلكترون فولت لمترابك PVA/MoS₂/WS₂ الهجين، وهو ما يعكس نشوء حالات إلكترونية موضعية وتأثيراً تأزرياً واضحاً بين المادتين النانويتين.

الكلمات المفتاحية: بولي فينيل الكحول (PVA)، المواد النانوية، ثنائي كبريتيد الموليبيدينوم (MoS₂)، ثنائي كبريتيد التنغستن (WS₂).