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Surface Functionalization and Plasmonic Interactions in PVA–TiO_2–Silver Nanoparticle Hybrid Nanocomposites

Georges Martin Genius

georgesmartingenius@gmail.com

Pwani University

Research Article

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Abstract

Hybrid polymer nanocomposites comprising poly(vinyl alcohol) (PVA), titanium dioxide (TiO_2), and silver nanoparticles (AgNPs) were systematically designed and synthesized to investigate the combined influence of surface functionalization and plasmonic interactions on structural, optical, dielectric, and thermal properties. TiO_2 nanoparticles (~ 25 ± 5 nm) were chemically modified using 3-aminopropyltriethoxysilane (APTES) to enhance interfacial compatibility and dispersion within the hydrophilic PVA matrix. Silver nanoparticles (~ 15 ± 3 nm) were synthesized via sodium borohydride reduction and stabilized using citrate ions to ensure colloidal stability and controlled particle size. The nanocomposites were fabricated via solution casting with varying filler concentrations, and comprehensive characterization was performed using X-ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR), scanning and transmission electron microscopy (SEM/TEM), atomic force microscopy (AFM), UV–Vis spectroscopy, dielectric spectroscopy, and thermogravimetric analysis (TGA). Structural analysis confirmed the coexistence of semicrystalline PVA, anatase TiO_2, and metallic Ag phases, while spectroscopic results verified successful surface functionalization and strong interfacial interactions. Morphological studies revealed uniform dispersion of TiO_2 and well-distributed AgNPs with minimal aggregation, contributing to increased surface roughness and interfacial heterogeneity. Optical measurements demonstrated enhanced absorption in the 400–500 nm region due to localized surface plasmon resonance (LSPR) of AgNPs, alongside intrinsic TiO_2 bandgap absorption in the UV region. Dielectric analysis showed a substantial increase in dielectric constant from 9.6 for pristine PVA to 45.2 at 1 kHz for the optimized composite (15 wt% TiO_2, 2 wt% AgNPs), with low dielectric loss (0.038), attributed to strong interfacial polarization mechanisms. Thermal analysis further indicated improved stability, with decomposition onset shifting from 275°C to 308°C. These findings highlight the synergistic role of surface functionalization and plasmonic coupling in tailoring multifunctional polymer nanocomposites, offering significant potential for applications in advanced dielectric materials, optoelectronic devices, and energy storage systems.

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1. Introduction

Polymer-based hybrid nanocomposites have emerged as a versatile class of materials capable of combining the mechanical flexibility and processability of polymers with the functional properties of inorganic nanostructures. Among various polymer matrices, poly(vinyl alcohol) (PVA) has attracted considerable attention due to its excellent film-forming ability, hydrophilicity, chemical stability, and ease of processing. However, its relatively low dielectric constant and limited optical functionality restrict its applicability in high-performance electronic and optoelectronic devices (1–8). To address these limitations, the incorporation of inorganic nanofillers such as titanium dioxide (TiO_2) and silver nanoparticles (AgNPs) has been widely explored. TiO_2, particularly in its anatase phase, is known for its high dielectric permittivity, chemical stability, and photocatalytic properties (9–19). Meanwhile, AgNPs exhibit unique optical characteristics arising from localized surface plasmon resonance (LSPR), which leads to strong absorption in the visible region and enhanced electromagnetic field localization. A critical challenge in developing such hybrid systems lies in achieving homogeneous nanoparticle dispersion and strong interfacial interactions within the polymer matrix. Surface functionalization of nanoparticles provides an effective strategy to overcome this challenge. In this context, silane coupling agents such as 3-aminopropyltriethoxysilane (APTES) can form covalent bonds with the nanoparticle surface while simultaneously interacting with polymer chains, thereby improving compatibility and dispersion. Furthermore, the combination of high-permittivity ceramic nanoparticles and plasmonic metallic nanoparticles introduces synergistic effects that can significantly enhance both dielectric and optical properties. In particular, interfacial polarization mechanisms, such as Maxwell–Wagner–Sillars polarization, play a crucial role in enhancing dielectric response in heterogeneous systems (20–29). In this study, PVA–TiO_2–AgNP hybrid nanocomposites were synthesized with controlled compositions to systematically investigate the effects of surface functionalization and plasmonic interactions. A comprehensive set of characterization techniques was employed to elucidate the structure–property relationships governing the performance of these materials.

2. Materials and Methods

Poly(vinyl alcohol) (Mw ≈ 89,000–98,000 g/mol, 99% hydrolyzed) was used as the polymer matrix without further purification. A 10 wt% PVA solution was prepared by dissolving the polymer in deionized water under continuous magnetic stirring at 90°C for 4 hours until a homogeneous and transparent solution was obtained. Titanium dioxide nanoparticles (anatase phase, ~ 25 ± 5 nm) were dispersed in ethanol and subjected to ultrasonication at 40 kHz for 45 minutes to minimize agglomeration. Surface functionalization was achieved by adding 2 vol% APTES to the dispersion, followed by refluxing at 70°C for 3 hours. This process facilitated the formation of Ti–O–Si bonds, enabling effective grafting of silane groups onto the nanoparticle surface. The functionalized nanoparticles were subsequently washed with ethanol and dried at 80°C for 12 hours. Silver nanoparticles were synthesized via chemical reduction by introducing aqueous silver nitrate ($AgNO_3$) solution into deionized water, followed by the dropwise addition of freshly prepared sodium borohydride ($NaBH_4$) under ice bath conditions to control nucleation and growth. Sodium citrate was added as a stabilizing agent to prevent aggregation. The resulting nanoparticles were collected by centrifugation, washed, and redispersed in water. Hybrid nanocomposites were fabricated by incorporating functionalized TiO_2 (5, 10, and 15 wt%) and AgNPs (0.5, 1.0, and 2.0 wt%) into the PVA solution. The mixtures were subjected to probe sonication at 200 W for 30 minutes to ensure uniform dispersion. The resulting suspensions were cast onto glass substrates and dried at 60°C for 24 hours to obtain free-standing films with a thickness of approximately 110 ± 5 μm.

3. Characterization

The structural, morphological, optical, dielectric, and thermal properties of the prepared nanocomposites were systematically investigated using a suite of advanced analytical techniques. X-ray diffraction (XRD) analysis was conducted using Cu Kα radiation to determine the crystalline phases and structural integrity of the composite components. Fourier transform infrared (FTIR) spectroscopy was employed to identify functional groups and confirm successful surface modification of TiO_2 nanoparticles. Surface morphology and nanoparticle dispersion were examined using scanning electron microscopy (SEM), while transmission electron microscopy (TEM) provided high-resolution imaging of nanoparticle size and distribution within the polymer matrix. Atomic force

microscopy (AFM) was used to quantify surface roughness and topographical variations. Optical properties were analyzed using UV–Vis spectroscopy over a wavelength range of 300–800 nm, enabling the identification of plasmonic absorption features. Dielectric properties were measured using an LCR meter across a frequency range of 100 Hz to 1 MHz, with gold electrodes deposited on both sides of the films to ensure accurate measurements. Thermogravimetric analysis (TGA) was performed under a nitrogen atmosphere to assess thermal stability and decomposition behavior.

4. Results and Discussion

The structural, morphological, optical, dielectric, and thermal properties of the synthesized PVA–TiO_2–AgNP hybrid nanocomposites were systematically analyzed to elucidate the role of surface functionalization and plasmonic interactions. The X-ray diffraction patterns presented in Fig. 1 confirm the successful incorporation of all composite constituents without compromising their crystalline integrity. The characteristic diffraction peak of semicrystalline PVA observed at $2\theta \approx 19.5°$ is retained across all samples, indicating preservation of the polymer matrix structure. Distinct reflections at 25.3°, 37.8°, and 48.0° correspond to the (101), (004), and (200) planes of anatase TiO_2, respectively, confirming its phase purity. Additionally, the presence of a weak but discernible peak at 38.1° is attributed to the (111) plane of face-centered cubic silver, verifying the successful incorporation of AgNPs. The relatively low intensity of the Ag peak is consistent with its lower concentration and nanoscale dispersion within the matrix.

Further insights into chemical interactions and surface modification are provided by the FTIR spectra shown in Fig. 2. The broad absorption band around 3300 cm^{-1} corresponds to O–H stretching vibrations of PVA, while the peak near 1087 cm^{-1} is associated with C–O stretching. Importantly, the emergence of bands in the 1120–1140 cm^{-1} region is indicative of Ti–O–Si linkages, confirming successful silanization of TiO_2 nanoparticles via APTES. A subtle band around 1650 cm^{-1} is attributed to citrate groups stabilizing the AgNPs. Slight shifts and broadening of the O–H band suggest strong hydrogen bonding interactions between the PVA matrix and the functionalized nanoparticles, highlighting improved interfacial compatibility.

The morphological features observed in the SEM micrographs (Fig. 3) reveal a relatively smooth and homogeneous surface for pure PVA, while the nanocomposite films exhibit a more textured morphology due to the incorporation of inorganic fillers. TiO_2 nanoparticles appear to be uniformly dispersed throughout the matrix, while AgNPs form small, well-distributed clusters without significant agglomeration. This uniform dispersion is attributed to the effectiveness of surface functionalization in reducing nanoparticle–nanoparticle interactions and enhancing compatibility with the polymer matrix.

High-resolution TEM images in Fig. 4 further confirm the nanoscale dispersion and size distribution of the fillers. TiO_2 nanoparticles are observed with an average size of approximately 25 nm and are well anchored within the polymer matrix. AgNPs, with sizes around 15 nm, are distributed as discrete particles with minimal aggregation. The clear distinction between the two types of nanoparticles and their intimate contact with the polymer matrix supports the formation of effective interfacial regions, which are critical for enhanced functional properties.

Surface topography analysis using AFM (Fig. 5) provides quantitative evidence of increased surface roughness upon nanoparticle incorporation. The root mean square (RMS) roughness increases significantly from 8.7 nm for pristine PVA to 32.5 nm for the composite containing 15 wt% TiO_2 and 2 wt% AgNPs. This increase in roughness reflects enhanced interfacial heterogeneity and the formation of nanoscale protrusions associated with embedded nanoparticles. Such surface features are known to contribute to improved interfacial polarization and dielectric performance.

Dielectric properties, presented in Fig. 7, demonstrate a substantial enhancement in dielectric constant with increasing nanoparticle loading. At 1 kHz, the dielectric constant increases from 9.6 for pure PVA to 45.2 for the optimized composite. This enhancement is primarily attributed to interfacial polarization mechanisms, particularly Maxwell–Wagner–Sillars (MWS) polarization, arising from charge accumulation at the interfaces between PVA, TiO_2, and AgNPs. The presence of conductive AgNPs further facilitates charge carrier mobility, contributing to polarization under an applied electric field. Notably, despite the significant increase in dielectric constant, the dielectric loss remains low (0.038), indicating minimal energy dissipation and efficient charge storage behavior. This balance between high permittivity and low loss is essential for practical dielectric applications.

Thermal stability, as evaluated by thermogravimetric analysis in Fig. 8, shows a clear improvement in the nanocomposite system. The onset of thermal decomposition shifts from approximately 275°C for pure PVA to 308°C for the hybrid nanocomposite. This enhancement is attributed to strong interfacial interactions between the polymer matrix and the functionalized nanoparticles, which restrict polymer chain mobility and delay thermal degradation. The presence of inorganic fillers also acts as a thermal barrier, further improving resistance to heat-induced decomposition.

Overall, the results demonstrate that the combined effects of surface functionalization and plasmonic interactions lead to significant improvements in structural integrity, optical absorption, dielectric performance, and thermal stability. The formation of strong interfacial regions and the synergistic interaction between TiO_2 and AgNPs within the PVA matrix are key factors governing the enhanced multifunctional properties of these hybrid nanocomposites.

5. Conclusion

In this study, multifunctional PVA–TiO_2–AgNP hybrid nanocomposites were successfully developed through a combination of surface functionalization and controlled nanoparticle incorporation. The use of APTES-modified TiO_2 significantly improved dispersion and interfacial adhesion, while AgNPs introduced strong plasmonic effects that enhanced optical absorption. The synergistic interaction between the polymer matrix and nanofillers resulted in remarkable improvements in dielectric performance, optical response, and thermal stability. The optimized nanocomposite demonstrated a high dielectric constant, low dielectric loss, and enhanced thermal resistance, highlighting its potential for advanced applications in dielectric devices, sensors, and optoelectronic systems. Overall, this work underscores the importance of interfacial engineering and nanoscale design in tailoring the multifunctional properties of hybrid polymer nanocomposites.

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Figures

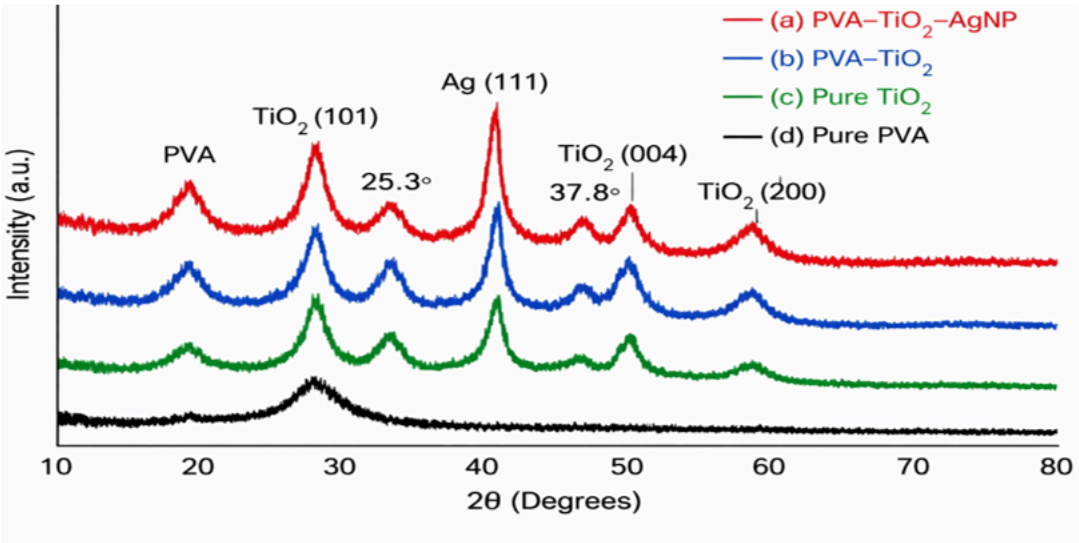


Figure 1

X-ray diffraction (XRD) patterns of pristine PVA, TiO_2 nanoparticles, and PVA–TiO_2–AgNP hybrid nanocomposites showing the coexistence of semicrystalline PVA, anatase TiO_2, and metallic Ag phases.

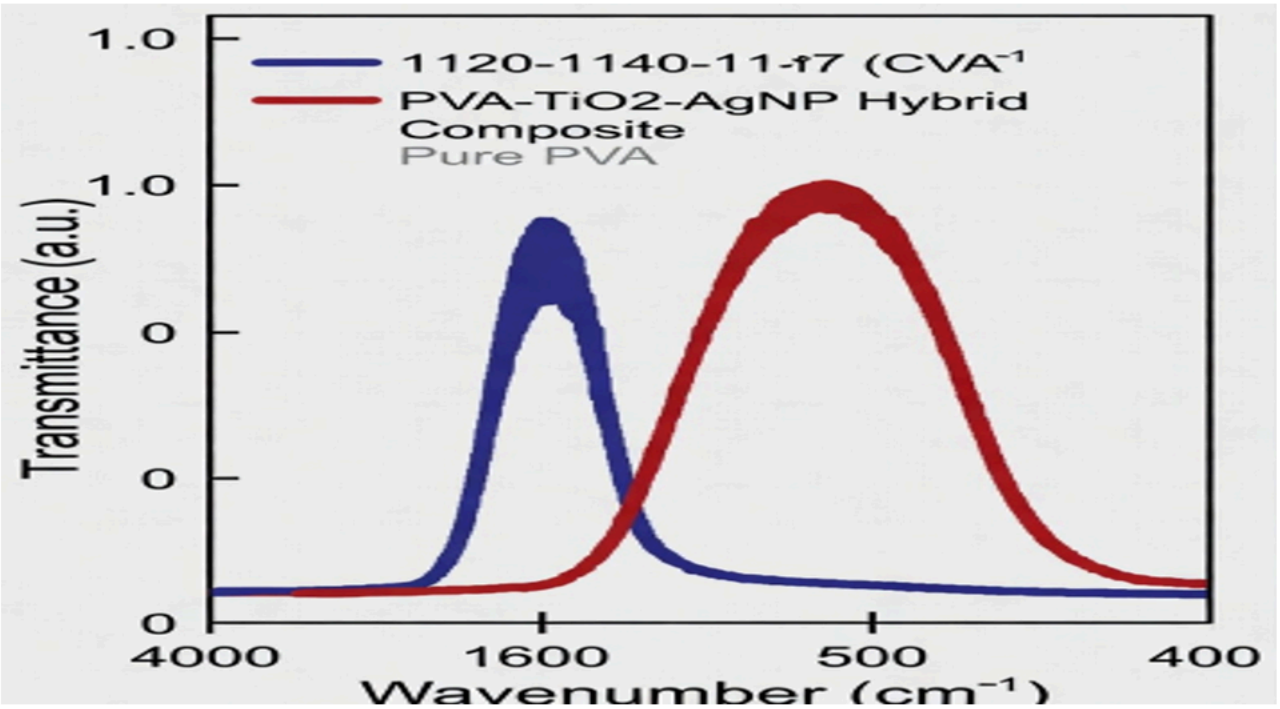


Figure 2

FTIR spectra of pristine PVA and PVA–TiO_2–AgNP nanocomposites illustrating Ti–O–Si bond formation and enhanced interfacial interactions due to surface functionalization.

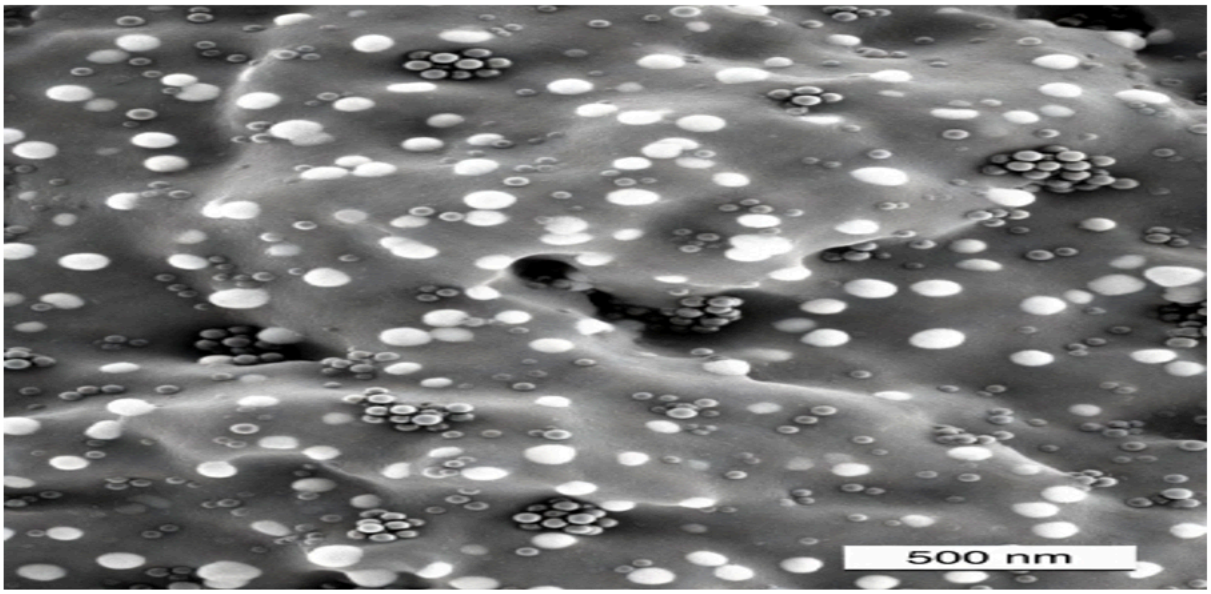


Figure 3

Scanning electron microscopy (SEM) images of PVA–TiO_2–AgNP nanocomposites demonstrating uniform nanoparticle dispersion and modified surface morphology.

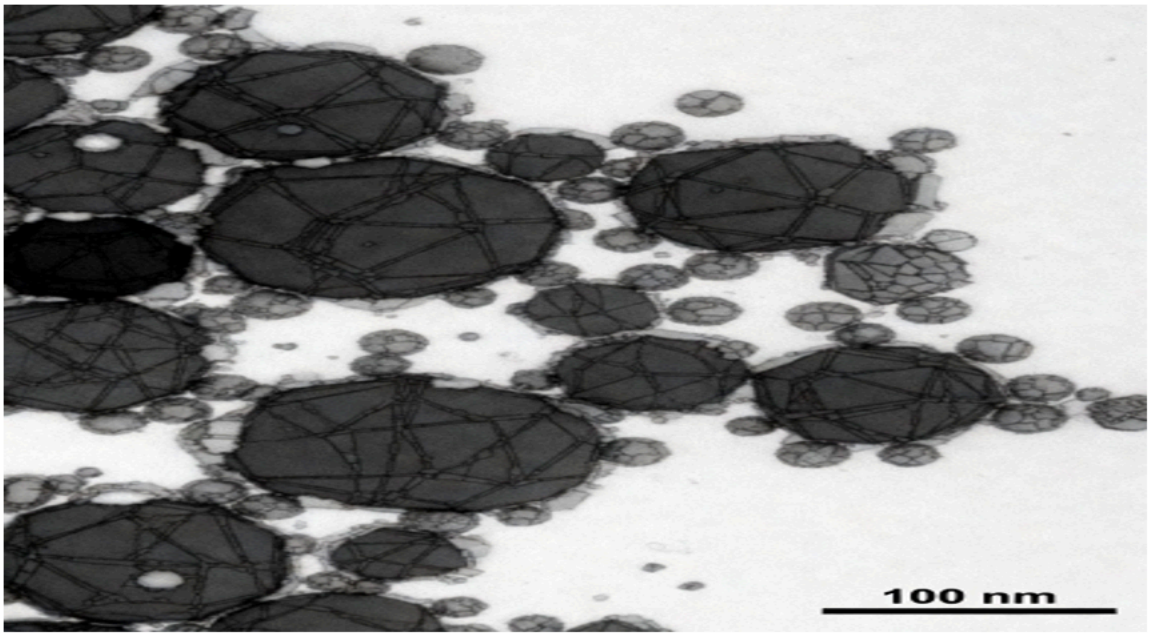


Figure 4

Transmission electron microscopy (TEM) images showing nanoscale distribution of TiO_2 nanoparticles and AgNPs within the PVA matrix with minimal aggregation.

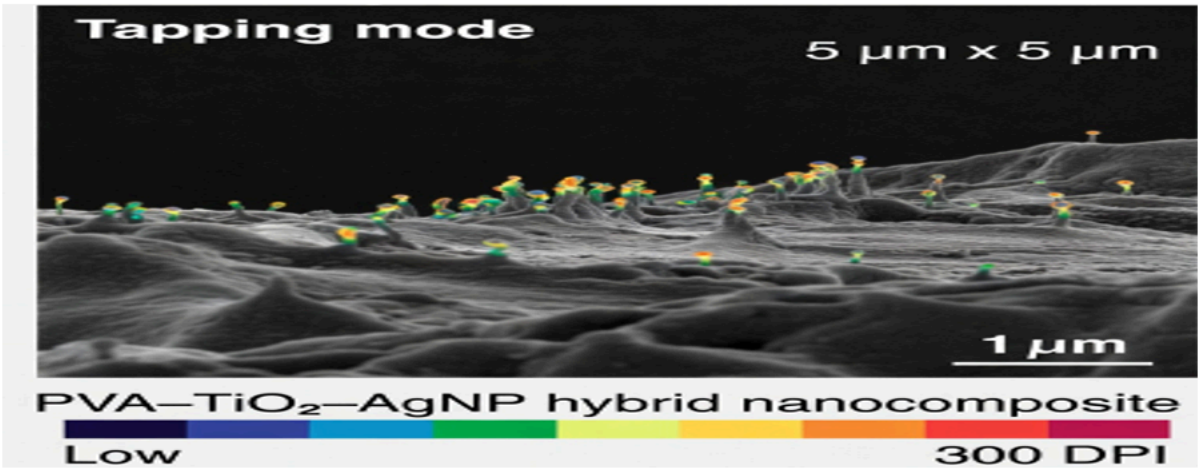


Figure 5

Atomic force microscopy (AFM) surface topography of PVA–TiO_2–AgNP nanocomposites indicating increased surface roughness and interfacial heterogeneity.

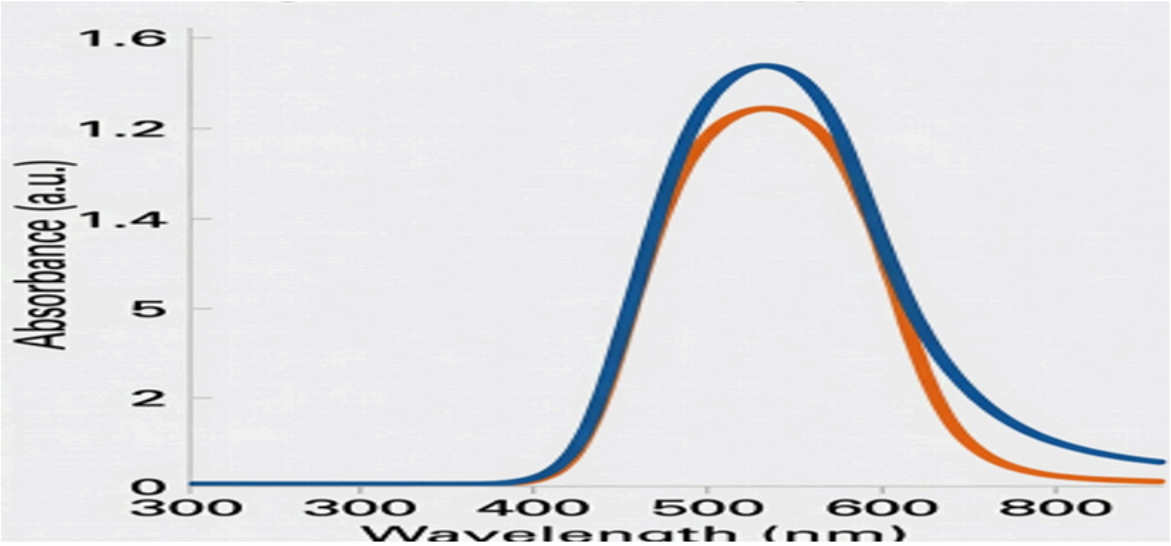


Figure 6

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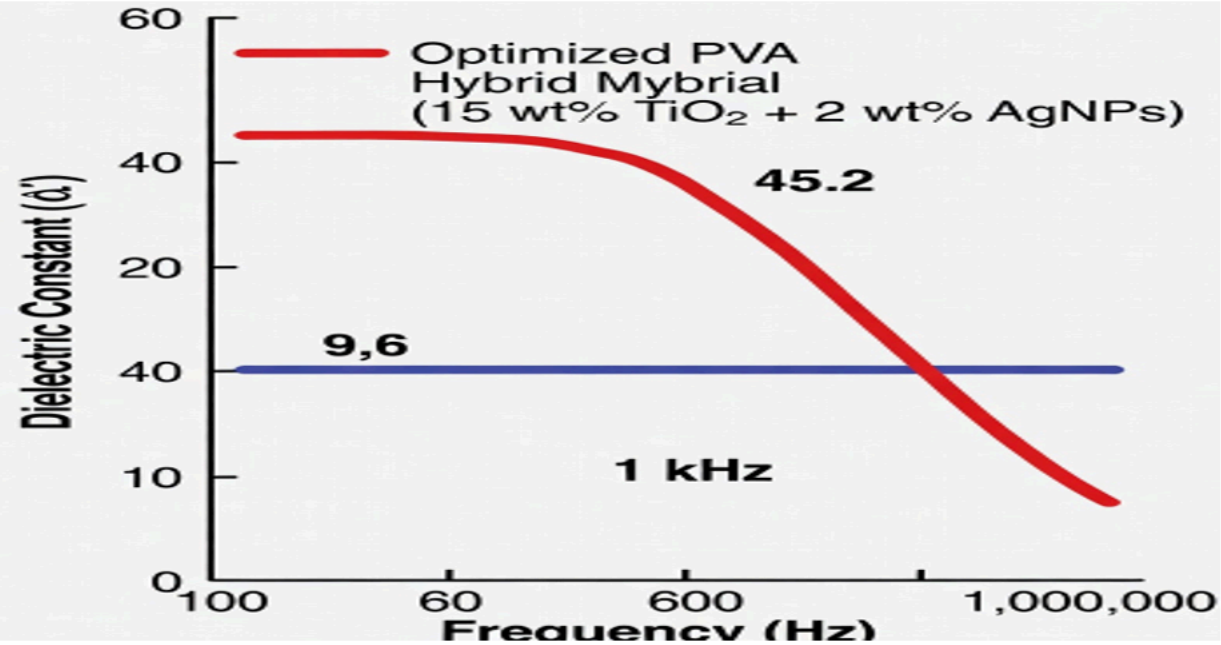


Figure 7

Frequency-dependent dielectric constant and dielectric loss of PVA–TiO_2–AgNP nanocomposites showing enhanced permittivity and low dielectric loss due to interfacial polarization.

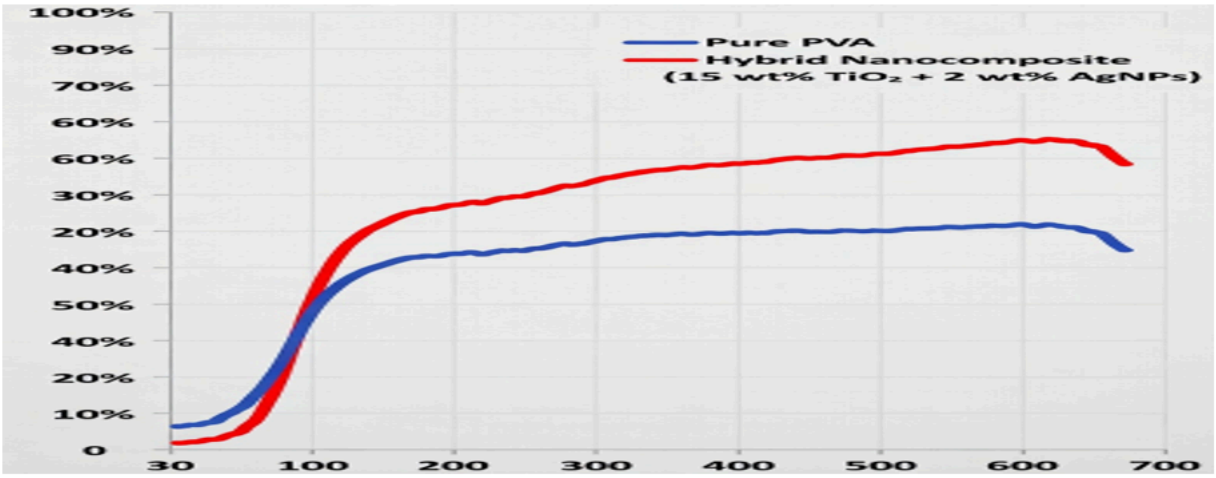


Figure 8

Thermogravimetric analysis (TGA) curves of pristine PVA and PVA–TiO_2–AgNPs nanocomposites demonstrating improved thermal stability and delayed decomposition.