

Review

Synthesis, Rheology, Morphology, and Mechanical Properties of Biodegradable PVA-Based Composite Films: A Review on Recent Progress

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Abstract: Biodegradable polymers play an important role in environmental concerns compared to non-biodegradable polymers. Polyvinyl alcohol (PVA) is a biodegradable polymer with film-forming properties with antimicrobial and antioxidant activities and are considered for numerous practical applications in the industry, like food packaging, pharmaceuticals, and so on. The synthesis of PVA with promising properties like rheology, morphology, and mechanical performance is significant from the application point of view in industrial sectors. It is vital to realize the drawbacks and promising prospects associated with PVA rheology, morphology, and mechanical properties and how to address the problems concerning these properties. The present review describes the contemporary advancement of numerous synthesis approaches of PVA-based composite films and their rheology, morphology, and mechanical properties. This comprehensive review offers a comprehensive discussion of various strategies to enhance the rheology, morphology, and mechanical properties of composite films. It emphasizes modifications using environmentally friendly materials such as nanoparticles, metal oxides, polymers, and others. Additionally, existing challenges and the potential for forthcoming advancements in the properties of such composite films are discussed. The correlation between the PVA-based composite films and their promising properties like rheology, morphology, and mechanical performance may provide a reference for new insights into their applications in industrial sectors.

Keywords: synthesis; PVA-based composites; rheology; morphology; mechanical properties; biodegradability



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1. Introduction

Among the biodegradable polymers, PVA possesses great potential owing to its considerable mechanical properties, safety, elevated swelling properties, good optical transparency, superior biocompatibility, non-toxicity, exceptional film-forming characteristics, and biodegradability [1–3]. For these unique features, PVA was utilized in biomedical and pharmaceutical sectors, electronics, optical sensors, and packaging applications [4–6]. Further, in various applications, PVA can exist in various forms, including films, hydrogels, fibers, scaffolds, and membranes [7].

Although PVA has potential and prospects in different areas of application, its utilization in several applications is limited due to its poor mechanical properties. For instance, researchers used metal oxides (for example, CuO, CaO, Al₂O₃, ZnO), nanolignin, ZnS, chitin, cellulose, and graphene oxide (GO) to improve the thermal, morphological, mechanical, and barrier properties and to apply to wastewater treatment [8–15]. To enhance biodegradability, biopolymers like starch, carboxymethyl cellulose (CMC), gelatin, and

chitosan were used by the researchers [16–22]. Not only the metal oxides and biopolymers but other inorganic functional nanomaterials, including ZnO nanoparticles (NPs), TiO₂NPs, AgNPs, carbon nanotube (CNT), and halloysite nanotubes (HNTs), were also utilized for the synthesis of PBC [23–27]. Similar to the addition of these materials, the addition of bioactive compounds to PVA to prepare composites is useful for their antibacterial and antioxidant applications. Such bioactive compounds include curcumin, gallic acid, and essential oils, which are very effective in enhancing the UV-barrier properties and antioxidant and antibacterial activities of PVA [28–33]. Polyaniline (PANI) was introduced to fabricate PVA/PANI films for energy storage applications like supercapacitors [34]. The time–temperature indicator was developed by incorporating chitosan into PVA and using anthocyanins for food quality checking [34]. On the other hand, a well-defined morphological structure is vital for enhancing the mechanical and thermal properties of PVA hydrogels, as the internal architecture significantly influences these attributes. Additionally, controlled morphology enhances thermal stability by ensuring even heat distribution and reducing structural degradation at elevated temperatures. This structural optimization not only improves the hydrogel's performance but also broadens its potential applications in biomedical engineering and environmental remediation.

However, PVA-based polymeric films are widely recognized for their excellent biodegradability, making them highly attractive for environmentally sustainable applications. Enhancing the rheology and morphology of PVA films plays a pivotal role in optimizing their functional properties while maintaining their eco-friendly nature. In this context, a well-developed morphological structure is crucial as it directly influences key characteristics such as mechanical strength and thermal stability, ensuring the films' suitability for diverse applications. Moreover, improved rheological behavior facilitates better processing and uniformity, contributing to the overall performance and durability of PVA-based materials. These advancements not only expand the applicability of PVA films but also reinforce their potential as biodegradable alternatives in various industries.

Therefore, most of the recent review work was conducted focusing on the structure and chemical properties of PVA and PBC for biomedical, food packaging, and fuel cell applications [7,32,35,36]. Enhancing their rheological, morphological, and mechanical characteristics through environmentally friendly modifications, such as incorporating nanoparticles, metal oxides, and polymers, can further expand their potential in these sectors. This review focuses on exploring advanced strategies for tailoring PVA-based films to meet the demands of these targeted applications. The current study also aimed to discuss the numerous synthesis approaches for the PVA-based composite preparation by the addition of numerous functional materials. Further, this review explained the recent progress on the numerous PBCs along with their significance connecting to rheological, morphological, and mechanical properties, emphasizing their different applications.

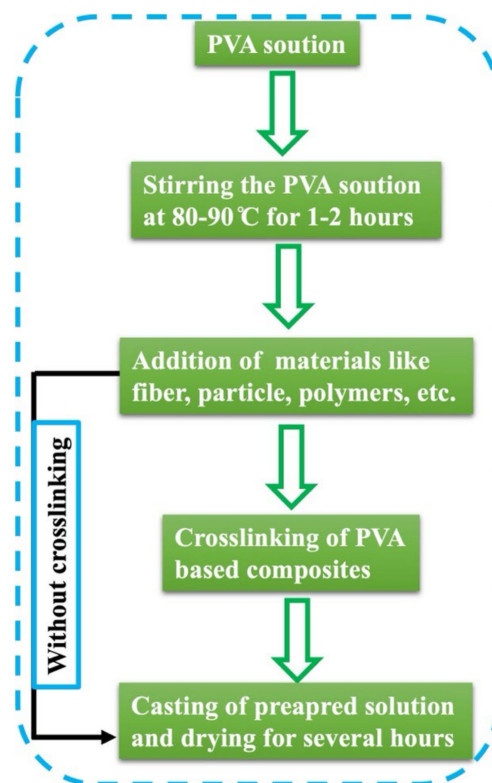
2. Synthesis

2.1. Synthesis of PVA-Based Composite Films

The synthesis of PBC was performed by the researchers with numerous approaches for different applications along with their distinct properties, including rheological, morphological, mechanical, and so on. The different synthesis approaches of PBC are listed in Table 1. For the synthesis, researchers utilized different materials like nanotubes, nanoparticles, nanofibers, polymers, and so on, as presented in Figure 1 [32]. For example, Sankar et al. [37] synthesized PVA-based composites by the addition of tungsten disulfide nanotubes with enhanced thermal and mechanical properties. Kuljanin et al. [38] synthesized PVA–PbS composites by the incorporation of PbS nanoparticles with improved mechanical and thermal properties. In this section, recent reports on the synthesis of various PVA-based composites and their key formation features and characteristics will be reviewed.

Table 1. Recent reports on the numerous synthesis methods of PVA–metal oxide-based nanocomposites for different applications.

Nanocomposites	Synthesis Method	Remarks	Applications	Ref.
PVA–ZnS	Solvent casting	Continuously stirred for 5 h at 60 °C	MB removal	[9]
PVA–ZnO–Al ₂ O ₃	Solvent casting	Sonicated for 2 h; temperature 80–90 °C	MB removal	[10]
PVA–CaO–CuO	Solvent casting	Acid-catalyzed polymerization, ultrasonication	MB removal	[8]
PVA–ZrO ₂	Solution casting	Synthesized at room temperature	Membrane for filtration	[39]
Chitosan/PVA/MeOx (Cu ₂ O, ZnO)	Plasma	DC power supply maintaining voltage up to 5 kV and resistor 0.5 k Ohm	Antimicrobial activity	[40]
PVA–Fe ₂ O ₃ –TiO ₂	Chemical blending process	Continuous stirring	Biomedical	[41]
PVA–ZnO	Sol–gel	Stirring at ~115 °C for about 15 min	Antimicrobial activity	[42]
PVA/PVP/Al ₂ O ₃ /SiO ₂	Dip-coating	Ultrasonication, pH ~7 at 70 °C for 3 h	Probable applications in optoelectronic devices	[43]

**Figure 1.** Synthesis of PVA-based composite materials [32].

Owing to the sustainability and environmental impact issues, researchers are focusing on the synthesis of environmentally friendly materials like PBC. Figure 2 displays the synthesis procedure of PBC materials. The synthesis of numerous PBCs is explored by the researchers as such materials are easy to biodegrade and have low cost and simple fabrication approaches. For example, PVA–ZnS composite films were synthesized by

changing the composition of the PVA addition ranging from 1 to 5 wt% following the solvent casting approach [9]. The authors demonstrated better photocatalytic depiction of composites along with enhancement of photoluminescence (PL), morphology, and thermal properties [9]. PVA–ZnO–Al₂O₃ composite films were fabricated with the addition of different amounts of ZnO and Al₂O₃ into PVA during the synthesis process [10].

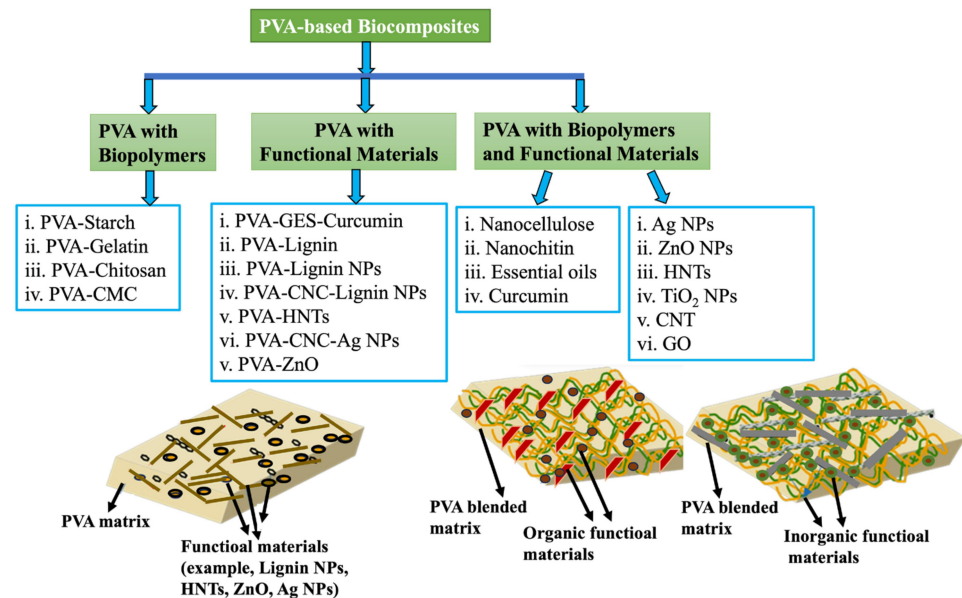


Figure 2. Formation of PVA-based bio nanocomposite films with numerous biopolymers, organic and inorganic functional materials, and their mixed conditions [1].

The addition of metal oxides was able to improve the luminescence and thermal properties of the fabricated composites [10]. Very recently, the same research group prepared PVA–CaO–CuO composites with distinct morphological topography and thermal stability [8]. The synthesized composites showed better photocatalytic performance compared to PVA alone. Mehto et al. synthesized PVA–ZrO₂ composites with distinct optical features, morphological properties, and improved crystallinity [39]. Chitosan/PVA/MeOx (Cu₂O, ZnO) composites were synthesized using discharge plasma for antibacterial applications [40]. The discharge plasma was exciting using a 0.5 k Ohm ballast resistor (R_b) and a handmade DC power source with an output voltage of up to 5 kV. PVA-supported ZnO-based nanomaterials were synthesized for antibacterial applications along with porous morphology and semi-crystalline properties [42]. Ahmed I. Ali et al. [43] synthesized PVA/PVP/Al₂O₃/SiO₂ composite hydrogel, which can be used as a membrane for industrial applications. The authors claimed that the prepared composites are polycrystalline structures with grains in nanoscale and homogenous features of membranes [43]. PVA–Fe₂O₃–TiO₂ composites were fabricated for biomedical applications along with distinct morphological features and better optical transparency [41]. The authors showed that the optical band gap of prepared PVA–Fe₂O₃–TiO₂ nanocomposite was better for the higher content of metal oxide (Fe₂O₃–TiO₂) nanoparticles [41].

2.2. PVA-Based Biopolymer/Inorganic Nanoparticles/Functional Composite Films

In this section, focus will be given to PBC fabrication by the addition of different functional nanofillers or bioactive compounds to enhance the functional properties of PVA. These materials include ginger nanofibers, lignin, nanocellulose, nanochitin, halloysite nanotubes (HNTs), etc. The inclusion of these types of functional materials has the potential to improve the morphological, mechanical, and thermal properties of PVA [1,15,44,45]. For example, the blending of PVA with ginger nanofibers enhanced the tensile strength, thermal stability, and moisture resistance by 65.6%, 7%, and 18.7%, respectively, compared to PVA

alone [44]. Moreover, the addition of non-toxic, low-cost, and superior biodegradability, like nanocellulose incorporation, can improve the thermal and mechanical properties of PVA. In this context, Espinosa et al. [46] demonstrated that the incorporation of nanocellulose increased the tensile strength of PVA films from 43.6 MPa to 58.8–72.7 MPa, varying the type of cellulose nanocrystals [46]. Further, the addition of functional materials is also able to enhance the sensing, antimicrobial, and antioxidant activities of PVA. These types of materials include lignin in nanoparticle form, essential oils, curcumin, AgNPs, and so on (Figure 2). The antioxidant activities of PVA films were greatly improved by the incorporation of lignin in their nanoparticle form [46,47]. Further, the addition of lignin is also able to develop the UV-shielding properties of PVA films [46,47]. To enhance the antimicrobial properties of PVA, researchers introduced essential oils and metal nanoparticles such as CuNPs, AgNPs, etc. [23,31,48–50]. The PbS nanoparticles (4.6 and 2.3 mass%) were incorporated with PVA by colloidal chemistry technique to fabricate nanocomposites by Kuljanin et al. [38]. It was observed that the increasing amount of PbS in PVA was able to enhance the tensile strength value of 53.0 ± 3.1 MPa. This outcome was obtained for the highest content of PbS (4.6 mass %) in PVA. PVA/rGO (reduced graphene oxide) nanocomposites, vastly amorphous in nature, were fabricated through an in-situ solution-blending approach. The prepared composites exhibited a low percolation threshold (~ 1.7 wt%) and elevated electrical conductivity value of 11 S/m [51].

Microfibrillated cellulose (MFC)/PVA composites were synthesized by varying the amount of MFC in the synthesis process [32,52]. The authors demonstrated that the fracture strength of PVA was increased from 34.1 MPa to 89.9 MPa for the 40 wt% addition of MFC. The cellulose/PVA biocomposites were prepared by the treatment of *n*-butyl-3-methylimidazolium chloride with superior tensile strength [52]. PVA/cellulose composites were also prepared by Zhang et al., where the authors demonstrated that the tensile strength of the composite materials increased to 16.4 ± 0.2 MPa [53]. The introduction of chitosan into PVA to prepare blend membrane and crosslinking with Trimesoyl chloride (TMC)/hexane were able to enhance the tensile strength of crosslinked Chitosan (CS)-PVA films up to a maximum value of 74.5 ± 2.7 MPa [54].

3. Morphology of PVA-Based Composite Films

The PBC shows distinct morphological features such as the spherical-shaped structure with nanoscale diameter [8], agglomerated inorganic particles with flowerlike structure on the surface of the PVA matrix [9], uniformly dispersed nanofibers on PVA [55], and so on. For example, the TEM images of PVA/cellulose nanofibers (CNF) composites are displayed in Figure 3, where the aggregation of CNF can be appreciated in PVA/CNF0 (Figure 3a) [55]. However, comparatively uniformly distributed white parts of CNF without significant aggregation on PVA can be seen for CNF10 and CNF20, respectively (Figure 3b,c) [55]. Composite samples, CNF10 and CNF20, are relatively regularly distributed (white parts) without remarkable agglomeration on PVA [55,56]. Between CNF10 and CNF20, the CNFs are more uniformly dispersed into the PVA matrix for PVA/CNF20 composites (Figure 3c). Thus, these outcomes indicate that the higher the content of CNF (vol%) in the composite sample, the better the compatibility, which can ultimately improve the mechanical properties of PVA/CNF [55,57].

The morphological features of the crack surface cross-section were also identified for PVA and PVA/CNF samples, as displayed in Figure 4a–d. In this case, the surface morphology of PVA was smooth and elongated with incessant flow, which revealed distortion of PVA with a definite load (Figure 4a) [55]. On the contrary, PVA/CNF composites exhibited an irregular surface morphology owing to CNF addition. For PVA/CNF0 composites, the CNF particles were agglomerated on the PVA surface (Figure 4b). However, less agglomeration and comparatively homogeneously dispersed CNFs were identified for the higher amount of CNF addition (Figure 4c,d). These results support the TEM data presented in Figure 3. Thus, good adhesion can be appreciated between PVA and CNF in the composite samples, resulting in the improvement of the tensile modulus of composites [55].

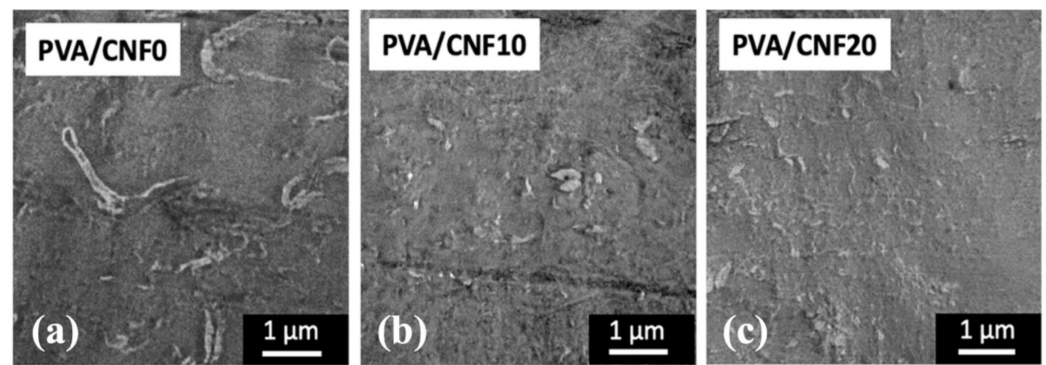


Figure 3. (a–c) TEM micrographs of PVA/cellulose nanofiber (CNF) composites synthesized at various compositions of CNF (vol%) [55].

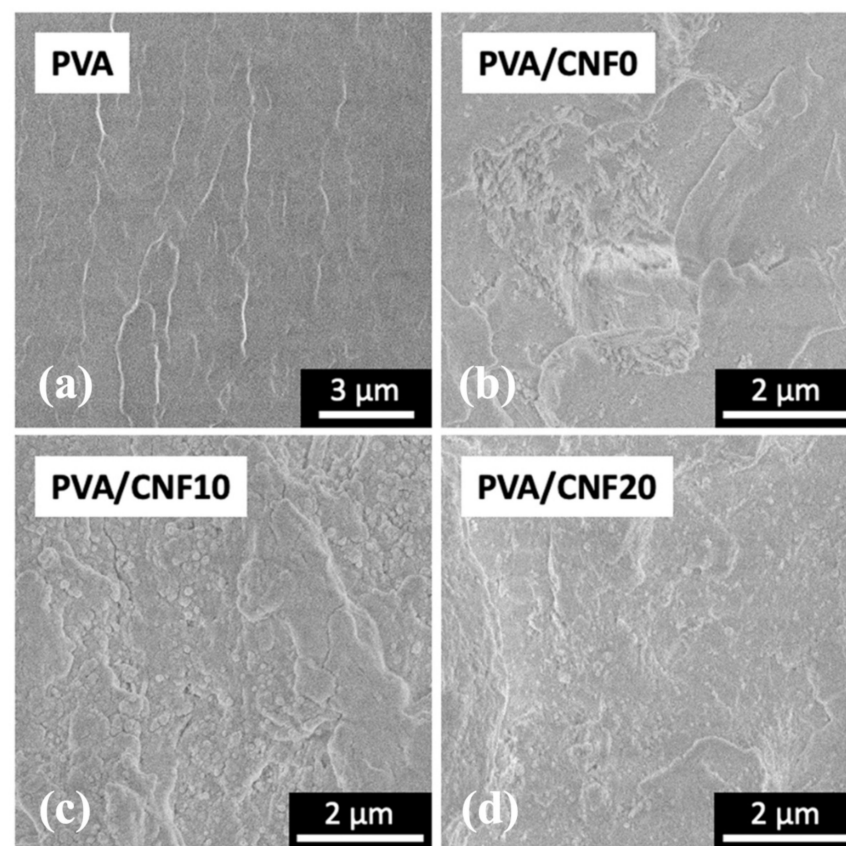


Figure 4. FESEM micrographs of tensile fractured surface of PVA and PVA/CNF composites (a–d). The PVA/CNF composites were synthesized at various compositions of CNF (vol%) [55].

The SEM micrographs showing morphological features of PVA and PBC are presented in Figure 5a–i. Figure 5a–i shows the “honeycomb” morphology of PVA, while in the composite’s large grains like HA and Se-doped TiO₂, nanoparticles are erratically distributed in the PVA matrix. Thus, the structure of PVA clearly varies from composite samples, where hydroxyapatite (HA) creates a fibrous structure among the pores and particles (Figure 5a–i). These types of morphological outcomes demonstrate that the existence of HA in the PVA matrix intensely influences composite morphology, generating fibrous and porous forms. Such morphological features are useful for the applications of these composites for soft tissue replacement [58]. Moreover, the incorporation of HA in the “honeycomb” structure of PVA may play a vital role in the adhesion capability of osteogenic and chondrogenic precursor cells, maintaining their proliferation [58].

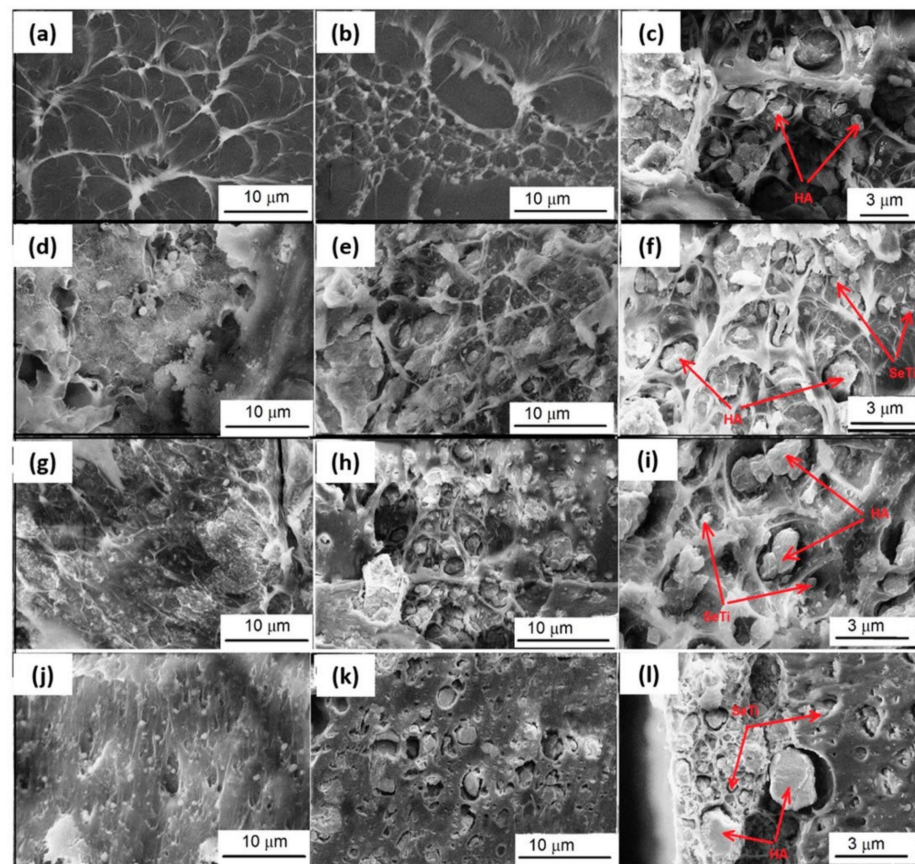


Figure 5. SEM micrographs (cross-section) of PBC reinforced with Se-doped TiO_2 particles and hydroxyapatite (HA), with distinct particulars (different PVA samples with and without HA): (a) PVA 10%; (b,c) PVA HA; (d) PVA 400; (e,f) PVA 400HA; (g) PVA 600; (h,i) PVA 600HA; (j) PVA 800; (k,l) PVA 800HA [58]. Copyright, 2020, MDPI.

4. Rheological Properties of PVA-Based Composite Films

The rheological properties of polymers are crucial for understanding and optimizing their processing behavior, as they directly impact both manufacturing efficiency and material performance [59–63]. This is particularly true in systems exposed to high shear stresses, such as PVA-based nanocomposites. In many studies, PVA is utilized as a filler material, and its interaction with other components significantly influences the rheological characteristics, which, in turn, affect the final composite's mechanical and processing properties [64–66]. A thorough understanding of these properties is critical for optimizing the formulation and processing conditions to achieve the desired performance characteristics [67,68]. By carefully selecting components and adjusting processing parameters, it is possible to tailor the material's behavior to meet specific requirements, whether for biomedical, electronic, or material science applications. This knowledge enables precise control over attributes such as mechanical toughness, swelling behavior, and responsiveness, ensuring improved functionality and stability in the final product. The rheological characteristics, such as viscosity, shear thinning, and elasticity, of these composites can significantly affect their processability and final properties. Despite numerous studies on some polymers, the unique rheological behaviors of nanocomposite remain less well understood and have only recently gained focused experimental attention. Notably, polymers exhibit near-frequency-independent storage (G') and loss (G'') moduli at low filler concentrations and frequencies, indicating solid-like behaviors even in dilute systems [69]. According to the theoretical network model proposed by Picu and Sarvestani [70], the overall viscoelastic response is significantly influenced by the duration of polymer–filler junctions, where solid-like characteristics are attributed to network-forming bridging seg-

ments among fillers. However, the fluid behaviors of polymeric materials are primarily categorized as Newtonian or non-Newtonian based on their flow behavior. In Newtonian fluids, a consistent relationship exists between shear stress and shear rate, meaning that viscosity remains unchanged regardless of the applied shear conditions [71]. Conversely, non-Newtonian fluids exhibit a viscosity that varies depending on their flow history and can change over time when subjected to a constant shear rate or stress [71]. Shear-thinning fluids, for instance, show a reduction in apparent viscosity as the shear rate increases, while shear-thickening fluids exhibit the opposite trend [72]. Furthermore, plastic fluids need a minimum yield stress to initiate movement.

Many polymer systems display both viscous and elastic qualities, defined as viscoelastic materials [73]. For example, silicone rubber exhibits notable viscoelastic properties, allowing it to deform under stress and then gradually return to its original shape when the force is removed [74]. Oscillatory tests are commonly utilized to examine these characteristics. The G' defines the material's ability for resistance to elastic deformation under stress; a larger G' indicates less elastic deformation under equivalent stress, showing the degree to which molecular chains can revert following external forces [75,76]. In contrast, the G'' measures viscous behavior, gaining energy lost from irreversible deformation often caused by molecular friction and slippage during stress [77,78]. For viscoelastic solids, δ approaches 0° at zero frequency, while in viscoelastic liquids, it nears 90° at the same frequency. Elastic gels typically exhibit a strong elastic response, where G' is much greater than G'' and remains steady across varying frequencies [79,80]. In Newtonian fluids, however, the viscous response dominates, with G'' significantly larger than G' .

The non-Newtonian properties of various polymers, especially shear-thinning, where the viscosity remains nearly invariant to filler concentration at high shear rates, paralleling that of unfilled polymers [81]. This effect is thought to arise from the alignment of polymer layers in response to shear deformation and relatively weak interactions among the different layers of polymers. Interestingly, a reduction in viscosity is observed with the introduction of filler materials, deviating from Einstein's classical model of viscosity enhancement in particle-laden suspensions [82]. This viscosity decrease is often attributed to an increase in free volume rather than a decrease in polymer entanglement density [83,84]. For systems with attractive interactions, the viscosity is substantially higher than that of the pure melt [85,86]. In contrast, polymers with neutral filler-polymer interactions show a modest increase in viscosity, while polymers with repulsive interactions exhibit reduced viscosities compared to pure melts [87–89]. Notably, the normalized polymer viscosity remains unaffected by chain length within measurement uncertainties, suggesting minimal nanoconfinement effects in these systems [78].

Mirela Teodorescu et al. [90] conducted a study where the viscoelastic moduli of the solutions vary with oscillation frequency in a manner typical of Maxwell fluids, where G'' is proportional to ω and G' scales with ω^2 . Solutions containing entangled PVA, Polyvinylpyrrolidone (PVP), and their mixtures underwent multiple freeze–thaw cycles, frozen at -20°C for 16 h, followed by thawing at room temperature for 8 h. Rheological properties were examined after various freeze–thaw cycles. After thermosetting the samples at 37°C , their viscoelastic properties were analyzed. The impact of aging was also assessed at both room temperature and 37°C . Following only two freeze–thaw cycles, samples with a high PVA concentration exhibited a gel-like character, indicated by $G' > G''$ and $\tan\delta < 1$. For mixtures with a high PVP content, gelation required more cycles, and no gel formed in pure PVP samples, even after 20 cycles [90]. Additionally, in the same study, the temperature-dependent aging effect was also discussed. However, an aging effect became evident when the temperature was raised from room temperature to near 37°C , resulting in time-dependent changes in viscoelastic properties after heating. In this work, the progression of the elastic modulus during frequency sweep tests was conducted at various aging intervals for a sample with 25% PVP after four freeze–thaw cycles [90]. The frequency's influence appeared minimal, but the effect of aging time was substantial. Specifically, the G' value for the sample with 25% PVP increased by two orders of mag-

nitude after resting for 5600 s, suggesting that the network structure developed through freeze–thaw cycles continued to strengthen at 37 °C as interactions between PVP and PVA chains intensified.

Yang Feng et al. [91] investigated the linear viscoelasticity of PVA and gellan gum (GG)-based hydrogels, where the increment of the GG concentrations on the composite enhanced the G'' value (Figure 6). The outcomes represent the average values from multiple measurements taken within the linear viscoelastic range. The mean G' values for pure PVA and PVA/GG composites with GG contents of 0.1%, 0.3%, and 0.5% are 6.4 ± 1.3 , 9.3 ± 1.5 , 10.0 ± 1.1 , and 12.1 ± 0.8 kPa, respectively. This trend demonstrates a progressive rise of 45%, 62%, and 89% in average G' with increased GG concentration. Although the mean G' of pure PVA is 45% lower than that of the 0.1% PVA/GG hydrogel, the large statistical variations prevent a decisive interpretation. However, the 0.3% PVA/GG composite shows a minimum 37% increase in G' , accounting for statistical fluctuations. Increasing the GG content to 0.5% brings a significant boost in G' , with the 0.5% PVA/GG samples averaging an 89% enhancement in G' . Even with statistical variations, there is at least a 48% rise in G' . The storage modulus G' measures the energy retained in a deformed material, indicating the elastic aspect of its viscoelastic properties. Notated, a higher G' value indicates that the hydrogel can retain more energy during deformation, signifying greater material stiffness. These results suggest that elevating GG content within PVA/GG hydrogels enhances their stiffness [91].

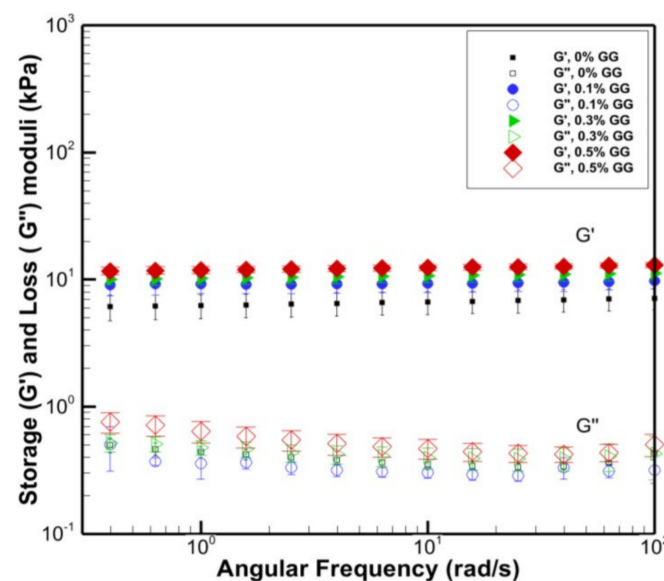


Figure 6. Graphs depicting the storage modulus (G') and loss modulus (G'') of pristine PVA hydrogels compared to PVA/GG hydrogels with varying concentrations of GG. The figure is adapted from ref. [91]. Copyright 2022, MDPI.

Caitlin et al. [92] studied the rheological characters of PVA and cellulose nanocrystal (CNC)-based nanocomposite suspensions. As the molecular weight of PVA increases, the viscosity of the suspension also rises due to the network formation within the CNC/PVA nanocomposite. This network may form from hydrogen bonding between the hydroxyl groups of PVA molecules or through crystallite formation within the PVA chains when they interact with water. Higher molecular weights in PVA result in increased viscosity, crystallinity, and elasticity. The findings indicate that using low-molecular-weight PVA with an optimal CNC concentration promotes stronger reinforcement, leading to a more rigid nanocomposite gel.

Nonlinear behaviors, such as critical strain, healing capacity, and responses to varying strain amplitudes and cyclic loading, are essential for effective material design. Moud et al. [93] examined the influence of CNC on the rheological behavior of PVA in the exis-

tence of NaCl, assessing performance in both small amplitude oscillatory shear and large amplitude oscillatory shear regions. In the linear region, the G' across all frequencies showed two significant jumps as CNC concentration increased. The first rise is attributed to the linking of CNC particles by primary and secondary polymer bridges, while the second reflects the formation of a secondary network between CNC–CNC and CNC clusters, leading to improved mechanical stability. This enhanced modulus value ranges between 10 and 30 kPa, which is considered suitable for supporting osteogenic cell differentiation. Additionally, studies show that the elastic modulus of individual chondrocytes is approximately 9.3 ± 0.8 kPa. The modulus of chondrons, which consists of the chondrocyte and its surrounding pericellular matrix, is around 12 ± 1 kPa [93]. On the other hand, Yang Feng et al. [91] investigated the elastic moduli of the composite PVA/gellan gum (GG) gels that were in close alignment with those of the chondrogenic matrix, positioning them as promising candidates for cartilage regeneration and related tissue engineering applications. Figure 6 represents the G'' for PVA and PVA/GG hydrogels with varying GG concentrations. The loss modulus reflects the viscous component of the viscoelastic material's behavior [91]. The data indicate that G' exceeds G'' , with both values remaining constant across different frequencies. The mean values for G'' are 0.43 ± 0.05 , 0.40 ± 0.08 , 0.47 ± 0.09 , and 0.61 ± 0.12 kPa for GG contents of 0%, 0.1%, 0.3%, and 0.5%, respectively. No significant variation in loss modulus was observed with the addition of 1% and 3% GG to PVA.

However, a slight increase in G'' was noted when the GG content reached 0.5 wt%. G'' represents the viscous component of the viscoelastic properties, which is associated with the liquid-like behavior of the material. Previous research has shown that when higher GG concentrations (from 0.5 to 10 wt%) were incorporated into PVA/GG composites, the water content of the hydrogels increased. This effect was attributed to GG's role in preventing the entanglement of PVA polymer chains and maintaining the integrity of microcrystalline regions within PVA. Furthermore, higher GG concentrations enhanced the hydrogel's ability to retain water. These characteristics enable PVA/GG hydrogels to absorb and hold more water, which, in turn, contributes to an increase in the loss modulus [91].

The sol–gel transition temperature can be effectively determined through dynamic oscillatory testing, which involves measuring the changes in both viscous and elastic moduli with temperature [94,95]. This method distinctly highlights the rheological alterations associated with the sol–gel transition. The point at which the moduli intersect $G'' = G'$ marks the structural transformation within the gel, clearly pinpointing the transition temperature. Notably, for thermally responsive polymers, such as poly(N-isopropylacrylamide) (PNIPAm) and PVA–PNIPAm composites, a pronounced volumetric phase shift occurs near their lower critical solution temperature (LCST), which also influences their viscosity and elasticity [94]. For instance, a study conducted by Zhang and colleagues developed a thermoresponsive PNIPAM-based interpenetrating polymer network (IPN) hydrogel incorporating PVA [76]. While the LCST remained unchanged, the responsiveness of the IPN was significantly improved. The addition of only 10 wt% PVA led to a rapid loss of approximately 95% of the water within one minute at 40 °C, whereas the PNIPAM gel only released 50% of its water after two hours under similar conditions [96].

Bhasha Sharma et al. [97] investigated the viscoelastic properties of PVA nanocomposites using dynamic mechanical analysis (DMA), revealing that the 1.5% GO-PVA nanocomposite exhibited a significantly higher storage modulus at 25 °C compared to pristine PVA, as shown in Figure 7 [97]. The addition of GO nanofillers results in an increase in the storage modulus, which can be attributed to the interaction between the hydroxylic groups of PVA and the epoxide bonds on GO, thereby enhancing the material's mechanical strength. As the temperature rises, a slight reduction in modulus occurs, but the modulus remains superior to that of PVA due to the thermal relaxation of PVA's polymer chains. The presence of GO nanoparticles limits the mobility of the PVA chains, transferring mechanical stress and thereby enhancing the rigidity and strength of the composite. At lower concentrations of GO, this effect was either not observed, or the samples were not sufficiently

homogeneous, possibly due to inaccuracies in the dispersion process, as GO has a high surface area-to-mass ratio. The increased storage modulus correlates with a higher degree of crosslinking, suggesting that more energy is stored and recovered during deformation. Moreover, the network formed within the polymer backbone facilitates the accumulation of elastic energy. As temperature increases, the reduction in storage modulus is likely due to the weakening of covalent bonds. In the glass transition region, the decrease in storage modulus is linked to the fact that molecular motions become limited to short-range vibrational and rotational movements in the glassy state.

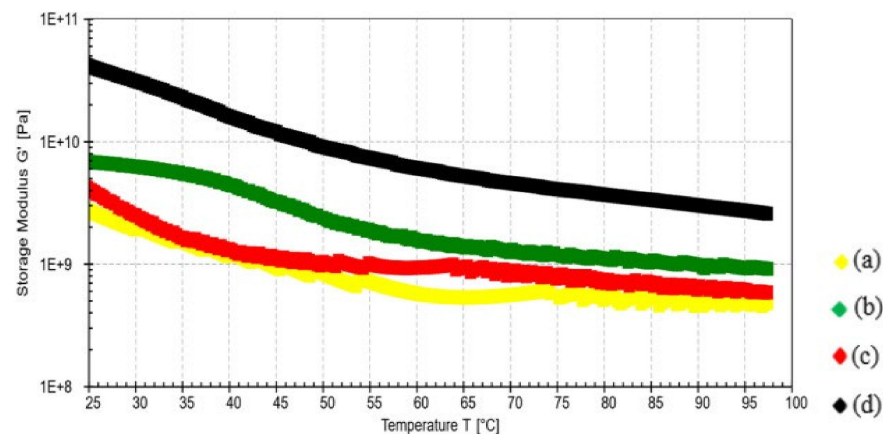


Figure 7. DMTA shows the G' as a function of temperature for (a) pure PVA, (b) 0.5% GO, (c) 1% GO, and (d) 1.5% GO-reinforced PVA nanocomposite films. The figure is adapted with permission from ref. [97]. Copyright © 2020 Elsevier Ltd.

PVA-based composite films exhibit rheological properties that are highly dependent on the type and concentration of fillers or additives incorporated. These properties, including viscosity, shear-thinning behavior, and storage/loss moduli, are critical for understanding their processability and mechanical performance. The tunability of rheological characteristics makes PVA composites versatile for applications such as flexible electronics, packaging, and biomedical devices.

5. Mechanical Properties of PVA-Based Composite Films

Human load-bearing tissues, particularly cartilage, consist of a structure of collagen fibers, proteoglycans, and a significant amount of interstitial fluid [98–100]. Together, these components provide the tissue with mechanical integrity and rigidity to withstand substantial loads [101]. The biphasic nature of cartilage contributes to its viscoelastic behavior, primarily driven by the resistance of interstitial fluid as it flows through a dense collagen-proteoglycan network with low permeability [102,103]. This process, known as fluid pressurization, plays a key role in enhancing cartilage stiffness under dynamic loading conditions [104]. Additionally, fluid pressurization is essential for providing cartilage with its characteristic low friction and wear resistance [105–107]. In everyday activities, knee cartilage is subjected to mechanical loads ranging from 1.2 to 7.2 times an individual's body weight [108–111]. These loads include compressive forces perpendicular to the cartilage surface, tensile stresses along the surface, and shear stresses. As such, materials designed for cartilage injury repair must be capable of withstanding external mechanical loads while restoring normal function [112,113].

The mechanical properties of cartilage have been explored extensively using diverse testing methodologies [114–116]. Compression properties are often quantified using the biphasic theory and nanoindentation techniques, revealing the aggregate modulus of human femoral head cartilage to range between 0.53 and 1.82 MPa [117]. Through uniaxial unconfined compression analysis, the instantaneous modulus of human cartilage is found to vary between 8.4 and 15.3 MPa [118]. Similarly, tensile properties are evaluated, with

Young's modulus measured in a direction parallel to the cartilage surface, spanning from 0.68 to 12.49 MPa [119]. Compared to natural cartilage, the mechanical performance of physically crosslinked PVA hydrogels remains insufficient [120]. Therefore, scaffolds intended for cartilage repair must closely replicate the mechanical properties of native cartilage to effectively bear joint loads [121]. Recent advancements have explored various strategies to strengthen hydrogels mechanically [122]. In the case of physically crosslinked PVA hydrogels, introducing reinforcing networks such as secondary gel matrices or nanoparticles, optimizing fabrication protocols, and employing post-treatment processes have shown significant improvements.

Electrospun nanofibers based on PVA and chitosan nanoparticles loaded with *Artemisia ciniformis* extract have been developed as effective wound dressings [122–125]. Additionally, co-electrospun PVA/chitosan/silk biocomposite nanofibers have been tailored to facilitate mesenchymal stem cell differentiation into keratinocytes, enabling their use as skin substitutes for wound repair [126]. Despite these advances, PVA/chitosan nanofiber-based dressings face challenges in clinical applications due to limited mechanical strength, suboptimal water solubility, and moderate antibacterial properties [126–129]. To address these issues, tetracycline-loaded PVA/chitosan nanofibers have been fabricated to enhance antimicrobial activity [130]. Wang et al. [131] successfully synthesized electrospun chitosan/cellulose nanocrystal membranes with improved mechanical attributes. Leveraging nanotechnology and incorporating metallic nanoparticles into PVA/chitosan nanofibers has further enhanced both mechanical and antibacterial performance [132]. For instance, a dual-layer nanofiber system comprising Cu nanoparticle-loaded PVA/chitosan as the base layer and polyvinylpyrrolidone (PVP) as the upper layer has demonstrated superior antimicrobial efficacy, mechanical robustness, and wound-healing capabilities; electrospun scaffolds with ZnO nanoparticles loaded into PVA/chitosan have been fabricated, exhibiting antioxidant properties and accelerated healing in diabetic wound [133]. Kharaghani et al. [134] innovated antibacterial nanofibers using Cu/Ag nanoparticles in a PVA/chitosan composite, which exhibited pronounced antibacterial activity against both Gram-positive and Gram-negative bacteria.

The incorporation of reinforcing fibers into the PVA matrix significantly enhances the mechanical strength and durability of PVA composites compared to unmodified PVA films, expanding their potential applications. For instance, Kiro et al. [135] developed a sericin–PVA composite impregnated with ZnO nanoparticles, achieving Vickers hardness values increasing from 15.11 to 20.09 kg/mm² with incremental additions of ZnO. Similarly, Sonker et al. [37] fabricated structurally modified PBC using glutaric acid and reinforced it with tungsten disulfide nanotubes, achieving tensile strengths up to 139.9 MPa and Young's modulus values reaching 7.1 GPa. Guzman-Puyol et al. [136] explored the plasticizing effect of trifluoroacetic acid on PVA–cellulose composites, noting increased elongation at break (693%) with reduced Young's modulus (25 MPa).

Liu et al. [137] developed a conductive PVA hydrogel using a simple method. First, PVA was dissolved in water, followed by the addition of the ionic liquid 1-ethyl-3-methylimidazolium acetate (EMImAc) to the PVA solution [137]. The mixture was then incubated at room temperature without any additional treatment. The resulting physically crosslinked PVA/EMImAc/H₂O hydrogel exhibited flexibility, excellent ionic conductivity, and anti-freezing properties. Notably, the hydrogel retained its exceptional mechanical performance at low temperatures. As shown in Figure 8A, the PVA/EMImAc/H₂O hydrogel retained its ability to endure significant distortions, including knotting, stretching, and twisting, even after being stored at −50 °C for 1 h [137]. Moreover, the hydrogel's transparency remained almost unchanged after storage at −50 °C for 1 h, as seen in Figure 8, in contrast to the PVA/H₂O hydrogel, which froze into a white, ice-like solid and lost its flexibility (Figure 8B) [137]. Additionally, the PVA/EMImAc/H₂O hydrogel demonstrated excellent compression resilience at −50 °C (Figure 8C) [137]. Further mechanical testing of the PVA/EMImAc/H₂O hydrogel across a broad temperature range and cyclic compressive loading–unloading curves is shown in Figures 8D and 8E, respectively [137].

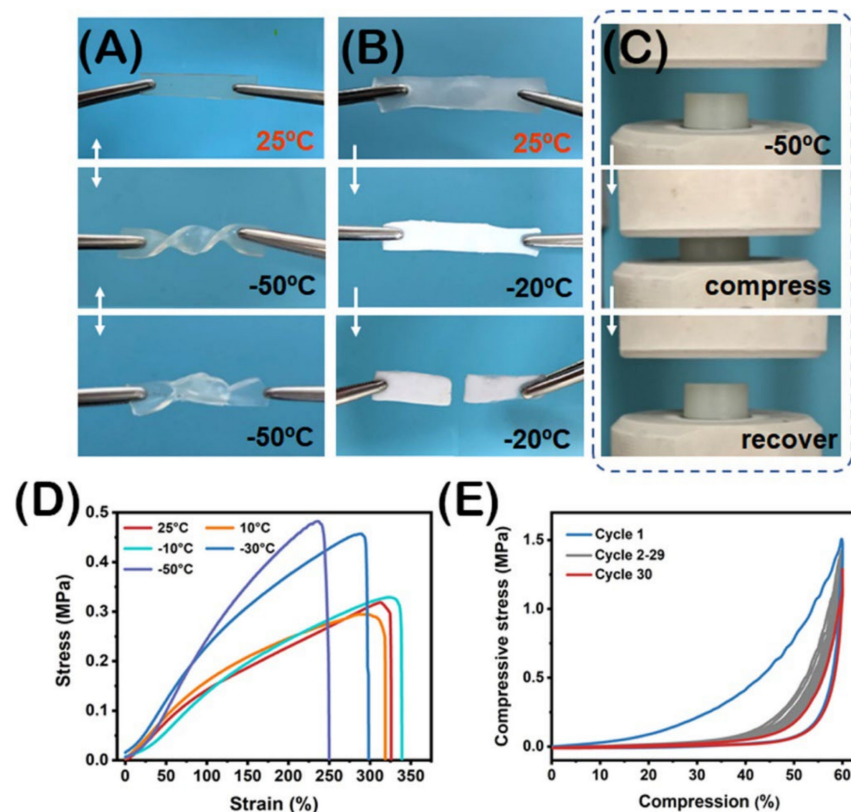


Figure 8. Mechanical behavior of the PVA/EMImAc/H₂O Hydrogel at Subzero Temperatures. (A) Demonstration of twisting and knotting of the PVA/EMImAc/H₂O hydrogel at 25 °C and −50 °C. (B) Visual form of PVA/H₂O hydrogel at 25 °C and −20 °C. (C) Compression and retrieval behavior of PVA/EMImAc/H₂O hydrogel at −50 °C. (D) Stress–strain curves representing the PVA/EMImAc/H₂O hydrogel at various temperatures. (E) Cyclic compressive loading–unloading curves of PVA/EMImAc/H₂O hydrogel at −50 °C. The figure is adapted with permission from the ref. [137], Copyright © 2021 American Chemical Society.

Further studies have demonstrated how GO enhanced PVA composite properties through ultrasonication. Li et al. [138] reported a tensile strength increase of 12.6% for GO-PVA composites, peaking at 82.5 MPa after 30 min of ultrasonication. Ibrahim et al. [139] investigated nanospherical cellulose reinforcement in PVA films, revealing that tensile strength improved with the incorporation of bleached linen particles but declined with cotton linter. Ching et al. [140] demonstrated that nanocellulose and nanosilica reinforcements optimized tensile strength in PVA films, with the highest Young's modulus achieved at 15% nanocellulose content. Other studies have evaluated the addition of lignosulfonate particles, fly ash, and microfibrillated cellulose, all of which substantially improved the mechanical properties of PVA composites [141]. The inclusion of cellulose nanofibers (CNF) in PVA composites enhances both stiffness and strength [142,143]. The tensile modulus and strength of CNF-reinforced PVA increased by approximately 50%, with adhesion between the CNF and PVA matrix facilitating load transfer to the rigid CNF structure [144,145]. Additional processing techniques such as ultrahigh-pressure homogenization and Starburst cycles further enhance tensile properties, underscoring the versatility of PVA composites.

Li et al. [146] reported a straightforward method for preparing a dual physically crosslinked hydrogel composed of nanosized PVA crystallite networks and a hyaluronic acid-Fe³⁺ network [146]. The process does not involve any polymerization reactions, making it environmentally friendly and appropriate for large-scale fabrication. Through annealing, hydrogel achieves potent mechanical properties, with a maximum toughness of 19.6 MJ/m³ and an elastic modulus of 10 MPa. Notably, the viscous precursor solution exhibiting good viscoelastic features can be processed via 3D printing to form tough

gel morphology with various patterns. This 3D printability greatly facilitates consequent applications, including in soft devices. The HA content was optimized at 1.5 wt%, as shown in Figure 9, which illustrates the effect of HA on hydrogel's mechanical performance [146]. It is evident that the overall mechanical properties considerably improve as the HA content increases from 0 to 1 wt%, owing to the formation of the HA-Fe³⁺ network. Compared to the pure PVA hydrogel, the toughness of the PVA/HA hydrogel increased by approximately 400%, from around 3.8 to 19.6 MJ/m³, while the elastic modulus rose by approximately 170%, from about 1.8 to 4.9 MPa. However, when the HA content exceeded 1 wt%, the mechanical properties sharply declined since the additional HA polymer chains disrupted PVA crystallization, resulting in a less dense PVA network. Consequently, the mechanical performance showed a nonmonotonic dependence on HA content, with the finest value observed at about 1 mg/mL.

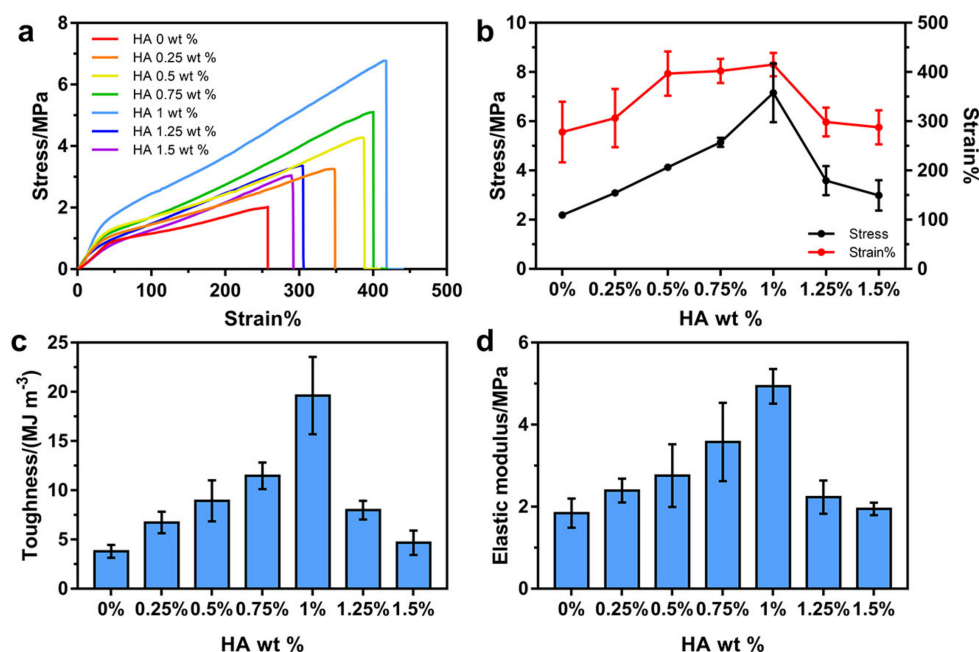


Figure 9. (a) Representative stress–strain curves for PVA/HA-Fe³⁺ hydrogels synthesized with varying HA concentrations. (b) Stress and strain of hydrogels as a function of HA content. (c) Toughness and (d) elastic modulus of PVA/HA-Fe³⁺ hydrogels fabricated with different HA concentrations. The figure is adapted with permission from the ref. [146]. Copyright © 2019 American Chemical Society.

PBC films exhibit excellent mechanical properties, including high tensile strength and elasticity, due to strong hydrogen bonding within the polymer matrix. Incorporating reinforcing agents, such as nanofillers or crosslinkers, further enhances their mechanical performance by improving stiffness, toughness, and thermal stability. These attributes make PVA composites highly suitable for biomedical and industrial applications requiring durable and flexible materials.

6. Summary and Future Perspectives

This review provides a general idea of the recent progress of synthesis, rheology, morphology, and mechanical properties of biodegradable PVA-based composite films. Numerous properties of the prepared composites depend on the approaches of modification, and the incorporated materials into the PVA matrix. These PVA-based composite materials possess a special type of materials and exhibit the unique features of rheological and mechanical properties, adhesion, and biocompatibility, which are suitable for antimicrobial, antioxidant activity, and biomedical applications. Although considerable progress has already been made on PVA-based composite materials, there are still potential prospects to utilize them for large-scale applications. In this concern, the applications of PVA alone

are challenging owing to its several limitations, including poor rheological and mechanical properties, agglomerated morphology, and so on. Thus, the researchers are searching for suitable materials to introduce with the PVA for better characteristics, which can facilitate its numerous applications. In these areas, several trials have already been performed, such as the introduction of organic or inorganic materials (for example, essential oils, metal oxides, metal nanoparticles, etc.), biopolymers (for example, cellulose, etc.) has enhanced rheological, mechanical, and morphological features. However, numerous challenges such as morphological complexity and enhancement of rheological and mechanical properties are still challenges affecting their applications. Thus, more focus is required on the PVA-based composite materials on their design and easy and low-cost synthesis to functionalization with controllable morphological features and rheological and mechanical properties. Future research will aim to use their various properties for clinical applications in areas such as wound healing, drug delivery, and water purification, where cost-effectiveness and functionality are essential. By optimizing PVA-based hydrogels, this work seeks to address the growing demand for sustainable and efficient solutions in these fields. In future work, the properties of PVA can be improved by introducing suitable inorganic, organic, and polymeric functional materials. Moreover, for large-scale usages of PBC and more technical enhancements, cost-effective applications are highly expected.

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Abbreviations

PVA	Poly vinyl alcohol
PBC	PVA-based composites
GO	Graphene oxide
CMC	Carboxymethyl cellulose
CNT	Carbon nanotube
HNTs	Halloysite nanotubes
PANI	Polyaniline
PL	Photoluminescence
rGO	Reduced graphene oxide
CS	Chitosan
MFC	Micro fibrillated cellulose
TMC	Trimesoyl chloride
CNF	Cellulose nanofibers
HA	Hydroxyapatite
GG	Gellan gum
CNC	Cellulose nanocrystal
PNIPAm	Poly(N-isopropylacrylamide)
EMImAc	1-ethyl-3-methylimidazolium acetate
LCST	Lower critical solution temperature
DMA	Dynamic mechanical analysis
GO	Graphene oxide

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