

Review

Poly(vinyl alcohol) nanocomposites: Recent contributions to engineering and medicine

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Abstract: Poly(vinyl alcohol) is a water soluble hydrophilic synthetic polymer. As a matrix it can be mixed with fillers having nano dimensions to form polymer nanocomposites with interesting properties, different than those of PVA. From the chemical point of view nanofillers can be elements such as Au, Ag, Se, oxides such as CuO, SrO, TiO₂, graphene oxide, minerals like hallosite, montmorillonite, tubes such as carbon nanotubes, hallosite nano tubes, etc. As presented in this review, some of the nanofillers are able to change and improve poly(vinyl alcohol)'s mechanical, thermal, and electrical properties, and can find uses in medicine. In the latter, due to the fact that poly(vinyl alcohol) is biocompatible, biodegradable, and bioabsorbant, it is possible to produce poly(vinyl alcohol) nanocomposites to be used like antibacterial, tissue engineering, drug carrier, wound dressing, etc. The present article retains the most recent (2019–2020) studies done on poly(vinyl alcohol) nanocomposites structure and contributions to engineering and medicine.

Keywords: poly(vinyl alcohol); nanocomposites; applications in engineering and medicine

Abbreviations: AA: Acrylic acid; AC: Alternating current; Ag: Agarose; C: Carbon dot; CAP: Cold atmosphere plasma; CNC: Cellulose nano crystal; CNT: Carbon nano tube; CS: Chitosan; D: Diamond; DC: Direct current; FA: Folic acid; g: ginger rhizome extract; G: Graphene; GEO: Ginger essential oil; GMA: Glycidyl methacrylate; GO: Graphene oxide; Gr: Graphite; HAP: Hydroxy apatite; HEC: Hydroxy ethyl cellulose; HNT: Hallosite nano tube; MMT: Montmorillonite; nD: Nano diamond; nDO: Oxidized nano diamond; PAA: Poly(acrylic acid); PANI: Polyaniline; PE:

Polyethylene; PEG: Poly(ethylene glycol); PVA: Poly(vinyl alcohol); PVAc: Poly(vinyl acetate); PVP: Poly(vinyl pyrrolidone); RGO: Reduce graphene oxide; SA: Sodium alginate; SEM: Scanning electron microscopy; SS: Silk sericin; St: Starch; TEM: Transmission electron microscopy

1. Introduction

Poly(vinyl alcohol) PVA is a synthetic polymer obtained by the alcoholysis of poly(vinyl acetate) (PVAc). The alcoholysis can be done with either an alkaline or acid catalyst. PVA is a hydrophilic polymer and therefore it dissolves in water. As a matrix, PVA can be reinforced with nanofillers to form PVA nanocomposites.

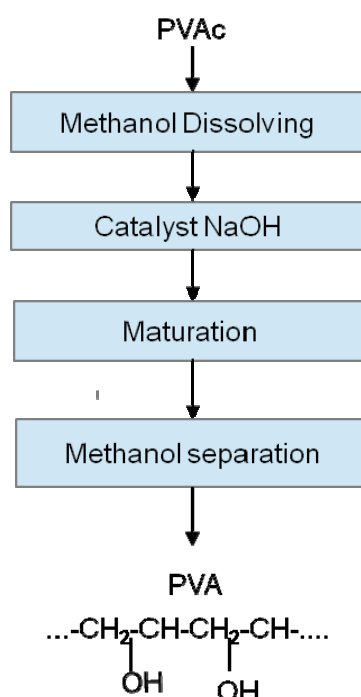


Figure 1. PVA synthesis.

Nanocomposites have become one of the most important fields in the contemporary materials science. The nanofillers are able to modify and improve some of the polymers properties leading to some new products. Being biocompatible, biodegradable, and bioabsorbant make PVA nanocomposites promising products for biomedical applications.

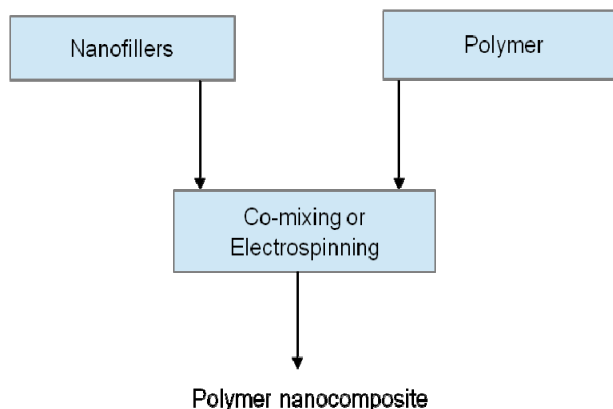


Figure 2. Polymer nanocomposite synthesis.

The diffusion of nanoparticles such as gold (nAu) within entangled solutions and gels formed by high MW PVA in water, using fluctuation correlation spectroscopy, has been studied. The result qualitatively agrees with the scaling theory prediction of hopping motion for particles within entangled polymer solutions. The data obtained will help to understand the size-dependent nanoparticle and molecular diffusion in PVA network [1].

This review will show how different nanofillers such as Se, Ag, Au, C dots, CuO, SrO, TiO₂, SrTiO₃, graphite (Gr), graphene (G), graphene oxide (GO) hallosite nanotube (HNT), carbon nano tube (CNT) montmorillonite (MMT), kaoline, hydroxyapatite (HAP), and others confer new properties to PVA.

The most pronounced enhancement of properties such as elastic modulus, yield stress, creep, and durability were found for individually dispersed nanoparticles due to the larger effective surface and the smaller inter element distance [2]. CNT/SiO₂ dispersed in PVA matrix acted as a strong nucleating agent and led to the enhanced crystallization of PVA in nanocomposites through heterogeneous nucleation [3].

Several series of hydrogel nanocomposites with different PVA/MMT ratios were prepared. MMT caused steric hindrance to the PVA crystallization and promoted the formation of a rigid amorphous phase. Its presence corroborated with the data of swelling in water. Since comparable results were obtained with MMT-org and MMT-Na for swelling, it shows that the swelling degree depends on the amount of MMT loaded in the hydrogel [4].

Polyaniline (PANI) nanoparticles and PVA/PANI nanocomposite films were produced in a study. Scanning electron microscopy (SEM) images show that PANI has nano fibers structure and the irradiated PVA/PANI nanocomposites have a mixture of nanorode and nanosphere structures. The transmission electron microscopy (TEM) shows that the irradiated PVA/PANI has core-shell structure [5].

2. Contributions to engineering

2.1. Mechanical properties

Using MMT, PVA/MMT nanocomposite showed a higher crystallinity, higher storage modulus, and higher thermal stability as compared with pure PVA films [6].

Nanocomposite films of surface functionalized HNT and PVA with improved mechanical and thermal properties were produced. Although the boronic acid used on the HNT surface decreased the crystallinity of the composite, the HNT modified with boronic acid (via aminopropyltriethoxysilane) exhibited superior mechanical and thermal properties. The increase is due to the fact that amine groups performed as base catalysts for the boronic acid–PVA interaction [7].

A report discusses the reinforcement of mechanical and thermal properties of polymer nanocomposites with nanodiamond (nD) as filler. According to the researchers' opinion, nanocomposites with nDs will be able to compete as a nanofiller against conventional nanofillers for polymer nanocomposites [8].

PVA/poly(vinyl pyrrolidone) PVP/nTiO₂ nanocomposites were prepared by solvent casting. The mechanical and dielectric properties, as well as fluorescence, were studied; the data obtained shows that the nTiO₂ doped PVA/PVP nano films are an appealing product for potential engineering application [9].

Bionanocomposite films made of PVA and gelatin were cross-linked with glutar aldehyde in the presence of kaolin. The increased addition of kaolin into bionanocomposite film shows a greater microhardness, increase of Young's modulus, tensile strength, and a significant reduction in water vapor transmission. It also offers a lower water sorption in comparison with gelatin/PVA film. This bionanocomposite is recommended for application in food packaging [10].

The aim of a paper is to investigate the effect of processing conditions for fabrication PVA nanocomposites by varying the exfoliation span, and to study its surface and the mechanical strength by using graphite (Gr) and graphene oxide (GO) as nanofillers. Through thermomechanical analysis it is concluded that GO is a more powerful and effective nanofiller when compared with Gr. As well as having application in packaging and many other areas, PVA nanocomposites also have great potential applications in energy storage devices [11].

For selective separation of CO₂ from methane, a PVA/graphene (G) nanoplate dense membrane was developed. The presence of G nanoparticles improved mechanical and chemical properties when G was below 1.5%. Compared to the pristine PVA, the CO₂/CH₄ selectivity of the nanocomposite membrane (PVA/1.5G) was higher by about 19% [12].

The morphology and the physical and mechanical properties of PVA/GO nanocomposites were also studied. With a 4% filler, the tensile strength and Young's modulus of the nanocomposite increases, the presence of the nanofiller changes the mechanism of deformation and leads to the decrease of the elongation at break. Cold rolling suppresses the tendency of the nanocomposite to brittle fracture [13].

The tensile strength of PVA/nSrO as compared with that of PVA increased from 85.2 to 112.8 MPa. DC electrical conductivity of PVA/nSrO also increased, thus indicating a potential application of these nanocomposites in optoelectronics due to their semiconducting properties and in biomedical field because of their mechanical properties [14].

Films consisting of composites of PVA with 3-aminopropyltriethoxysilane and 4-phenylphormil boronic functionalized HNT were prepared. They exhibited superior mechanical and thermal properties because the amine groups performed as base catalyst for PVA–boronic acid interaction [15].

2.2. Electrical properties

PVA/Cadmium sulphide and PVA/CdS nanocomposite films having 0–3% CdS nanofiller have

been produced. The good interaction PVA-CdS led to improvement in the dielectric properties as evidenced by the homogeneous distribution of the nanofiller within PVA, thus making nanocomposite films a good candidate for electrical charge storage devices [16].

In an article, GO/PVA/H₃PO₄/H₂O gel electrolyte was prepared and studied. Due to its high electrochemical and mechanical properties it is a great choice for use in flexible energy storage devices and wearable electronic devices [17].

Nanocomposite films based on PVA and reduce graphene oxide (RGO) have been prepared. The dielectric properties of PVA/RGO nanocomposite films were explored in the frequency range 100–25 Hz, which showed increase in dielectric permittivity and AC conductivity with the increase of RGO amount [18].

nAgs were used to prepare PVA/nAg composite films and their electrical conductivity was investigated. Different shapes and sizes of nAg were obtained by changing the concentration of sodium borohydrate solution. Depending on the shape and size of nAg, colloidal solutions had different color. High resolution TEM confirmed the spherical and the triangular shapes of yellow and blue nAg respectively. The latter, owing to their uniform distribution in PVA, show better charge carrier generation and enhanced surface plasmon resonance exhibited superior electrical conductivity as compared to yellow nAgs [19].

PVA/CaAl₂ZnO₅ nanocomposites were prepared by solvent intercalation. By studying the optical, cyclic voltammogram, and electrical properties, it is evident that the investigated nanocomposites are promising materials for energy storage and electrical applications [20].

A functional thin film of PVA/strontium titanate (PVA/SrTiO₃) hybrid nanocomposite was sandwiched between Ag electrodes for the application of random access memory. Flexible resistive switching showed excellent bipolar resistive switching behavior when subjected to both electrical and medical endurance tests [21].

The inertness of Gr when using it as a nanofiller was improved through a Diels–Alder reaction with acrylic acid (AA). Analyses done on PVA/AA@Gr nanocomposite show that it possessed excellent thermal stability which might push the scope of enhancing thermal conductivity of applied materials in flexible electronics with excellent combination property [22]

2.3. Other properties

Folic acid (FA) has been used as a biosafe and biodegradable product for the surface modification of ZnO nanoparticles. The PVA/ZnO/FA nanocomposite film is highly recommended in the production of food packaging and textile industry. It showed good resistance to *Escherichia coli* and *Staphylococcus aureus* [23].

PVA based active food packaging product with nAg was prepared. Ginger rhizome extract (g) was used for in situ reduction of AgNO₃ to nAg (gnAg) within PVA matrix. The presence of nanofiller gnAg in the PVA/gnAg film provided UV and light barrier with antibacterial activity against the food borne pathogens *Salmonella Typhimurium* and *Staphylococcus aureus*. Such film can expect to have promising food packaging applications [24].

The results of a study rank the PVA/ZnO₂ and PVA/carbon dot ZnO₂ (CZnO₂) as candidate adsorbent products for wastewater management, desalination and catalytic application [25].

A novel fabrication of durable, fluorine free and self-healing super hydrofobic cotton textiles by using boric acid as a cross linker to silica nano particles and PVA followed by polydimethyl-siloxane

modification by grafting is reported. The simple fabrication technique has promise in large scale superhydrophobic textile manufacturing applicable to accidental oil spills in the ocean and oily industrial waste water [26].

The double polyblend PVA/poly(ethylene glycol) (PEG) networks are hydrophilic smart materials that possess shape memory phenomenon by thermal stimuli-responsive. The researchers believe that the concept of 3D/4D printing through the double-network is worthy of additional exploration in additive manufacturing use to the application of shape memory hydrogels in a variety of technological fields [27].

Surface concentration of oxidized nanodiamond (nDOs) and nDOs modified with oleylamine in PVA/nDOs composite films has been determined by autoradiography using tritium-labeled nDOs. The study has shown that the hydrophobization of nDOs led to significant change in the nanoparticle distribution over the composite film and that the surface tension is determined by PVA due to the adsorption of PVA at the nanoparticles; the addition of nanofiller does not affect its value [28].

3. Contributions to medicine

3.1. Antibacterials

Thin biofilms of PVA/sodium alginate (SA) doped with selenium nanoparticles (nSe) were produced. UV/vis. optical absorption data shows that the dependence of nSe concentration with the optical energy gap PVA/SA polyblend with 8mL and 16mL had the highest antimicrobial activity [29].

PVA/nAg and Chitosan (CS/nAg nanocomposite films were obtained by augmentation with bio-synthesized using *Enterobacter cloacal* Ism26. nAg showed homogeneous dispersion through the two polymer matrices. These nanocomposites showed very low toxicity against Huh-7 liver cells and IC_{50} . Antibacterial, antibiofilm, and cytotoxic activities of the films were studied against multi drug resistant bacteria. The results of this study confirm the use of nontoxic PVA/nAg and CS/nAg nano films in many bio-medical and engineering applications [30].

PVA nanofibers with Fe-doped ZnO nanoparticles were obtained by electrospinning and studied for their antibacterial with cytotoxic properties. The cytotoxic studies have shown good compatibility of Fe-doped nZnO@PVA nanofibers to MCF-7 cell lines. The results confirm the fact that such nanofibers have the potential to be used in various biomedical applications [31].

3.2. Wound dressing

The ion cross linked poly(acrylic acid)-co-acrylamide/PVA/cloisite hydrogel film with 5.33 mg/mL allicin has the best efficiency as a hydrogel wound dressing on inhibition of the fungus species [32].

PVA/CS hydrogels containing Cerium oxide (CeO_2) nanoparticles were produced for medical applications. These PVA/CS hydrogels incorporated with CeO_2 nanoparticles could be a potential candidate as a suitable wound dressing product which can decrease wound infections without the necessity of antibiotics [33].

A wound dressing made of PVA and starch (St) with graphite carbon nitride $g-C_3N_4$ as a filler and Ag deposited titania nanoparticles ($Ag@TiO_2$) as an antibacterial agent was produced. On the basis of the results of this research it can be claimed that the obtained hydrogel membranes based on

PVA/St have the potential to be used as wound dressings effective for second and third degree burn wounds [34].

The encapsulation of ginger essential oil (GEO) within CS nano particles was studied. Semi solid PVA hydrogels containing different amounts of GEO-loaded CS nano particles were prepared, and characterized for mechanical properties, water content, and release characteristics such as release rate and amount of GEO. The semi-solid hydrogels were non-toxic to both NCTC clone 929 and NHDF cells [35].

3.3. Tissue engineering

Methacrylated PVA-glycidyl methacrylate/hydroxyapatite (PVA/GMA/HAP) has been developed. PVA/GMA hydrogel and PVA/GMA/HAP nanocomposite hydrogels exhibit excellent performance in compression tests. The photocrosslinked PVA/GMA/HAP nanocomposite hydrogels were cytocompatible to L929 cells and could promote the L929 cells attachment and proliferation. The nanocomposite hydrogel with HAP had enhanced mechanical strength and cell adhesion, showing their potential as tissue engineering scaffolds [36].

A porous 3D scaffold of hydroxyethyl cellulose HEC/PVA and HEC/PVA/cellulose nano crystal (CNC) HEC/PVA/CNC were successfully produced. Their study revealed that the melting temperatures of HEC/PVA/CNC scaffold were slightly shifted to a higher value. Scaffolds containing CNC showed an improvement in properties, thus promising a great potential for application in tissue engineering, mainly in bone cell culture [37].

Random and aligned PVA/poly(acrylic acid) PAA nanofiber scaffolds fabricated by electrospinning and treated with cold atmospheric plasma (CAP) were investigated. Antimicrobial tests have shown that electrospun PVA/PAA scaffolds can acquire strong antibacterial activity when treated with CAP that has no toxic effects on the fibers. The proposed strategy for plasma surface modification of electrospun nanofibers may break new ground for various biomedical applications [38].

Silk sericin (SS) has been combined with PVA and electrospun to create scaffolds with enhanced physical and mechanical properties, nontoxicity, good water stability, and biodegradability. PVA/SS nanofiber mats have the potential for use as skin tissue regenerative scaffolds [39].

A novel PVA/HAP/C dual network hydrogel scaffold with excellent fluorescence and bio-compatibility has been produced. It can be used as fluorescence-visible scaffold in biomedical engineering [40].

3.4. Drug delivery

Bio-films for potential orthopedic application have been obtained based on CS/PVA/GO/HAP/Au. These films were found to be bio-compatible with mouse mesenchymal cells. The result of the study suggests that the nanocomposite films have osteogenic potential for treating bone and bone diseases. The films also show a high inhibition against gram positive and gram negative bacteria (*Escherichia coli*, *Streptococcus mutans*, *Staphylococcus aureus* and *Pseudomonas aeruginosa*). The nanocomposite bio-films obtained are highly bio-compatible and can be used for bone regeneration application [41].

pH-responsive biodegradable hydrogel nanocomposites were synthesized using PVA/agarose

(AG) with the introduction of C. The researchers assumed that C integrated AG and PVA/AG hydrogel nanocomposites act as double drug carrier which are suitable for controlled drug delivery of norfloxacin (NFX) [42].

PVA nanocomposite hydrogels containing CuO nanoparticles (nCuO) were synthesized. Loading and releasing behavior of ibuprofen in nanocomposite hydrogels showed that compared with nanocomposite hydrogel, pure hydrogel had more drug loading. The drug release from pure hydrogel was more than that from nanocomposite hydrogels and an increase in nCuO nanoparticles reduced the drug release activity [43].

Coupling chitosan-mediated assembly of poly(vinyl alcohol-co-ethylene) (PVA-co-PE) nanofiber with the electrochemically controlled system provides great possibilities for multi functional coatings and drug elution on conductive implants [44].

3.5. Other applications

A research focused on the role of nanoparticle shape in targeted drug delivery to various organs including liver, spleen, lung, brain and also tumor sites of different tissues [45].

A study follows the diffusion of n Au within entangled solutions and gels of PVA in water. The researchers consider that their results will be important in the area of biological transport [1].

Polypyrrole nanoparticles were encapsulated into PVA hydrogel during its reticulation with glutar aldehyde. The hydrogels produced were tested for metoprolol a β -blocker used to treat angina, hypertension, and to prevent heart attacks. The application of electrical stimuli changes the release of the drug. This hydrogel can be considered a potential stimuli-responsive product for biomedical applications [46].

4. Conclusions

The present article is a literature review of recent studies (2019–2020) concerning the contributions of poly(vinyl alcohol) nanocomposites to engineering and medicine. For most of the studies the interesting synthesis and applications are discussed. They show how various nanofillers (chemical elements, oxides, clays, etc.) contribute to improve mechanical, thermal, electrical, medical and other PVA properties. Conducting PVA hydrogels found a wide variety of applications mainly in medicine to deliver drugs and cells for tissue engineering. The presented research results are a strong impulse for the continuation of the investigation in the above-mentioned fields.

Conflict of interest

The author declares there is no conflict of interest.

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