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Research Progress in Mechanical Properties of Polyvinyl Alcohol-Based Hydrogel Composites

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Abstract

Polyvinyl alcohol (PVA) hydrogels have broad application prospects in biomedical engineering, flexible electronics and other fields due to their excellent biocompatibility and chemical stability. However, traditional PVA hydrogels generally suffer from poor mechanical strength, low toughness and unsatisfactory fatigue resistance, which seriously restrict their practical applications. This paper systematically reviews the research progress in mechanical properties of PVA based hydrogel composites, focusing on the reinforcement mechanisms of strategies such as double network structure, nanoparticle composite, cross linking mode regulation, natural polymer composite and topological structure design. It summarizes the effects of cross linking density, polymer concentration and environmental factors on mechanical properties, deeply discusses the energy dissipation mechanisms and constitutive relationships, and prospects the future development directions in this field.Keywords.

Keywords

Polyvinyl alcohol, hydrogel, composite, mechanical property, strengthening and toughening.

1. Introduction

As a kind of soft and wet material with three dimensional network structure, hydrogels show great application potential in tissue engineering, drug delivery, flexible electronics and other fields due to their high similarity to biological tissues[1]. As an important base material for synthetic hydrogels, polyvinyl alcohol (PVA) has become one of the most widely studied hydrogel systems by virtue of its excellent film forming property, chemical stability and biocompatibility[2]. Nevertheless, traditional PVA hydrogels generally have insufficient mechanical properties: the fracture energy is usually only 10–100 J/m^2, far lower than that of biological tissues such as articular cartilage (10^2–10^3 J/m^2)[3]. This shortcoming seriously limits their practical applications in load bearing tissue repair and other fields.

In recent years, researchers have carried out a lot of research on the strengthening and toughening of PVA hydrogels, and developed a variety of strategies including double network structure, nanocomposite and cross linking regulation[4]. Although these strategies have achieved remarkable results in improving mechanical properties, they also have different limitations. Double network hydrogels often face irreversible damage, nanocomposites have the problem of uneven dispersion, and chemical cross linking may bring potential biotoxicity[5]. Therefore, how to realize the synergistic optimization of multiple properties such as high strength, high toughness, rapid recovery and fatigue resistance is still a research hotspot in this field[6].

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This paper systematically reviews the research progress of PVA based hydrogel composites in mechanical property enhancement, and conducts an in depth analysis from three dimensions: reinforcement strategies, influencing factors and action mechanisms, aiming to provide theoretical reference for the rational design of high performance PVA hydrogels.

2. Basic Characteristics and Cross-Linking Modes of PVA Hydrogels

2.1. Molecular Structure and Properties of PVA

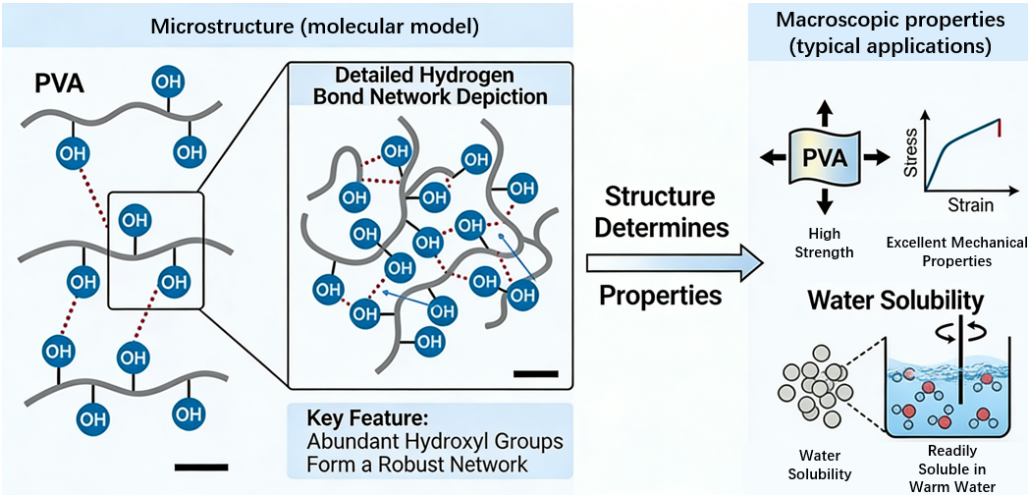

Figure 1. Schematic diagram of polyvinyl alcohol (PVA) hydrogen bonding network and its macroscopic properties

PVA is a water-soluble polymer prepared by alcoholysis of polyvinyl acetate. There are a large number of hydroxyl groups on its molecular chains, which can form intramolecular and intermolecular hydrogen bonds[7]

The properties of PVA are mainly affected by two key parameters: polymerization degree and alcoholysis degree. High-molecular-weight PVA can form longer molecular chains, which is conducive to the construction of high-density network structures. The alcoholysis degree determines the content of hydroxyl groups, thereby affecting the water solubility and crystallization behavior of PVA. Generally speaking, the higher the alcoholysis degree, the more regular the PVA molecular chains, the stronger the crystallization tendency, and the higher the mechanical strength of the formed hydrogel, but the solubility decreases accordingly[8].

3. Characteristic Comparison of Different Cross-Linking Modes

The cross-linking modes of PVA hydrogels can be divided into physical cross-linking and chemical cross-linking, which have their own characteristics and applicable scenarios.

Physical cross-linking is typically represented by the freeze-thaw method. Its principle is to make PVA molecular chains arrange locally regularly to form microcrystalline regions through repeated freezing-thawing cycles, and these microcrystalline regions act as physical cross-linking points to build a three-dimensional network[9]. The advantage of the freeze-thaw method is that no chemical cross-linking agent is needed, so the obtained hydrogel has high biological safety. Moreover, the formation of microcrystalline regions is reversible, endowing the material with certain self-healing ability. However, physically cross-linked hydrogels have relatively low mechanical strength, mainly relying on hydrogen bonds and microcrystalline regions to maintain the network structure, and are prone to irreversible chain slippage under external force. Studies have shown that increasing the number of freeze-thaw cycles can

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improve the compression performance of hydrogels due to the increase in the number of microcrystalline regions[10].

Chemical cross-linking connects PVA molecular chains through covalent bonds formed by cross-linking agents such as glutaraldehyde, boric acid and epichlorohydrin. Chemically cross-linked hydrogels have stable network structure and high mechanical strength, but the residue of cross-linking agents may cause biotoxicity problems. In addition, the formation of chemical cross-linking points is often irreversible. Once the network structure is damaged, the material is difficult to self-heal. Liu Huaqing et al.[11] prepared chemically cross-linked PVA hydrogels using dimethylol dihydroxy ethylene urea as cross-linking agent. Results showed that cross-linker dosage significantly affected the viscosity of the gel system, with an optimal dosage range, beyond which the viscosity decreased.

Radiation cross-linking is a clean cross-linking method, which cross-links PVA molecular chains by generating free radicals under γ-ray or electron beam irradiation[12].This method does not need to add cross-linking agents, avoiding chemical residue problems. The cross-linking degree can be accurately controlled by irradiation dose, but it has high requirements for equipment.

4. Strengthening and Toughening Strategies for PVA-Based Hydrogels

4.1. Double Network Structure Reinforcement

The design concept of double network (DN) hydrogels originates from the imitation of biological tissue structure. By interpenetrating and compounding two networks with complementary mechanical properties, the "1+1>2" reinforcement effect is achieved[13].This concept was first proposed by Gong Jianping's research group in 2003. The first network is usually composed of hard and brittle polyelectrolytes, which act as "sacrificial bonds" to preferentially break and dissipate energy under stress. The second network is composed of soft and tough neutral polymers, responsible for maintaining the overall structural integrity of the gel[14].

The reinforcement effect of the double network structure depends on the property matching of the two networks and the interaction between the networks. Studies have found that hydrogels can achieve the best mechanical properties when the first network is tightly cross-linked and the second network is loosely cross-linked[15]. This is because the tightly cross-linked first network can effectively bear load and break to dissipate energy, while the loosely cross-linked second network has sufficient segmental movement ability to continue to bear stress through chain stretching and rearrangement after the first network breaks. He Kaining et al.[16] systematically introduced the deformation and failure behaviors of high-strength and tough hydrogels, pointing out that DN hydrogels first exhibit macroscopic yielding and necking during stretching, followed by mechanical softening, accompanied by the gradual fracture of the first network and stress bearing of the second network.

In recent years, the development of double network hydrogels has evolved from fully chemical cross-linking to physical/chemical hybrid cross-linking, and then to fully physical cross-linking. Although fully chemically cross-linked double networks have excellent mechanical properties, the fracture of the first network is irreversible, leading to permanent softening of the material after large deformation. Replacing part of chemical cross-linking with reversible physical cross-linking makes double network hydrogels have both high strength and self-recovery ability[17]. Mi Zhiyuan et al.[18] systematically reviewed the formation and self-healing mechanisms of double network hydrogels, pointing out that dual physically cross-linked double network (DPC-DN) hydrogels have better self-healing and self-recovery abilities than physically-chemically hybrid cross-linked DN hydrogels.

The double network system constructed by cellulose nanomaterials and PVA has become a research hotspot in recent years. Ma Sensen et al.[19] prepared dialdehyde microfibrillated

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cellulose (DAMFC)/PVA composite hydrogels, using the acetal reaction between aldehyde groups on DAMFC surface and hydroxyl groups of PVA to form covalent connections. Such covalent connections ensure efficient stress transfer from the matrix to the reinforcement. After soaking in sodium sulfate solution, the compressive stress can reach 5.819 MPa, which is 35 times that of pure PVA hydrogel. Studies have shown that DAMFC content and oxidation time have significant effects on mechanical properties, and there is an optimal ratio range.

Zheng et al.[20] prepared sodium alginate/polyacrylamide double physically cross-linked network hydrogels by one-pot method, with fracture stress of 1.16 MPa, fracture strain of 2604%, and toughness of 14.20 MJ/m^3. This study endows hydrogels with both high strength and self-recovery properties by introducing dynamic reversible physical networks.

5. Nanoparticle Composite Reinforcement

Nanoparticles have become ideal candidates for reinforcing PVA hydrogels due to their high specific surface area, high modulus and abundant surface functional groups[21]. The reinforcement mechanisms of nanoparticles mainly include: acting as physical cross-linking points to increase network density; transferring and dissipating stress through interfacial interaction; and bearing part of the load by virtue of their own high modulus.

Cellulose nanomaterials are the most representative bio-based reinforcements[22]. The abundant hydroxyl groups on the surface of cellulose nanofibers can form a dense hydrogen bond network with PVA, significantly improving the mechanical properties of composite hydrogels. Cui Yexuan et al.[23] systematically reviewed the construction and applications of cellulose-based hydrogels, pointing out that the mechanical properties of cellulose hydrogels can be effectively improved through interpenetrating network (IPN) and semi-interpenetrating network (semi-IPN) structures. Studies have shown that the larger the aspect ratio of cellulose nanofibers, the more obvious the reinforcement effect, which is related to the ability of fibers to form a penetrating network in the matrix.

As a layered silicate clay, montmorillonite nanosheets can be dispersed in PVA matrix to form intercalated or exfoliated structures[24].PVA molecular chains can be inserted into montmorillonite layers. This structure not only increases physical cross-linking points, but also strengthens the gel skeleton through the high modulus characteristics of montmorillonite sheets. The introduction of montmorillonite can simultaneously improve the hardness, thermal stability and strength of hydrogels, but it should be noted that the dispersion state of sheets has a decisive impact on the reinforcement effect.

Nano-SiO_2 mainly interacts with PVA molecules through hydrogen bonds to strengthen the three-dimensional network structure. Its reinforcement effect comes from two aspects: nano-SiO_2 acts as a physical cross-linking point to increase cross-linking density; the interfacial interaction between nanoparticles and matrix can effectively transfer and dissipate stress. Zhang Shiling et al.[25] reviewed the research status of nano-micron particle reinforcing agents in polyacrylamide gel systems for oilfield chemistry, pointing out that nano-SiO_2 can effectively improve the storage modulus and compressive strength of gel systems.

Graphite materials such as graphene oxide have abundant oxygen-containing functional groups on the surface that can form hydrogen bonds with PVA. At the same time, the high modulus of graphene itself makes it an ideal reinforcement[26]. The introduction of graphene oxide can simultaneously improve the toughness and strength of hydrogels, but there is an optimal addition range. Excessive addition may lead to agglomeration and reduce mechanical properties.

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6. Synergistic Regulation of Physical/Chemical Cross-Linking

Cross-linking mode is the core factor determining the mechanical properties of hydrogels. Li et al.[27] systematically studied the effect of hydrogel cross-linking degree on adhesion properties, and found that the adhesion strength of hydrogels decreased significantly with the increase of cross-linking degree. Taking PVA hydrogel as an example, when the freezing time was extended from 2 h to 8 h, the adhesion strength decreased from 1600 Pa to 400 Pa. This phenomenon is related to the mechanism that hydroxyl groups are "fixed" after participating in cross-linking and are difficult to participate in surface interaction, suggesting that some free functional groups need to be reserved for interfacial interaction when designing high-adhesion hydrogels.

Regulating the aggregation state of polymer chains by using the Hofmeister effect is an important progress in recent years[28]. Wu et al.[29] studied the Hofmeister effect of PVA hydrogels. By regulating the aggregation state of polymer chains through soaking in different salt solutions, the mechanical strength of PVA hydrogels can be adjusted in a wide range. The salting-out effect of different salt ions on PVA hydrogels follows the Hofmeister sequence: the salting-out ability of anions is SO_4^{2-}>CO_3^{2-}>Cl^-. After soaking in sodium sulfate solution, the compressive stress of PVA hydrogel can reach 5.819 MPa, which is 35 times that of deionized water soaking, 15 times that of NaCl solution and 5 times that of Na_2CO_3 solution. This phenomenon originates from the polarization of hydrated water molecules by SO_4^{2-}, which destroys the hydrogen bonds between PVA and hydrated water molecules, and promotes the aggregation of PVA molecular chains and the formation of crystalline domains.

The universality of the Hofmeister effect is reflected in that no matter what cross-linking method is used to prepare PVA hydrogels, soaking in salt solution can further regulate its mechanical properties[30]. This provides a simple and effective method for adjusting the mechanical strength of PVA hydrogels in a wide range.

7. Natural Polymer Composite Reinforcement

Natural polymers have good biocompatibility and degradability. Compounding with PVA can prepare hydrogel materials with both excellent mechanical properties and biocompatibility[31-32].

The "egg-box" structure formed by sodium alginate and Ca^{2+} has high mechanical strength. After compounding with PVA, a double network system can be constructed. Zhang Xinlong et al.[33] introduced the preparation methods of sodium alginate hydrogels in detail, pointing out that sodium alginate network acts as a rigid skeleton to provide structural support, and PVA network provides toughness and deformation recovery ability. The synergistic effect of the two achieves strengthening and toughening. Wu Zhe et al.[34] prepared sodium alginate/microcrystalline cellulose composite hydrogels, with maximum adsorption capacities of 331.25 mg/g and 253.31 mg/g for methyl orange and methylene blue respectively, indicating that the composite system has both functional properties while maintaining mechanical properties.

As the only alkaline polysaccharide existing in nature, the amino groups on the molecular chains of chitosan are protonated under acidic conditions, which can form hydrogen bonds and electrostatic interactions with PVA[35]. The introduction of chitosan can simultaneously improve the mechanical properties and antibacterial properties of hydrogels. Ladeira et al.[36] prepared chitosan and carboxymethyl chitosan hydrogels using hexamethylene diisocyanate (HDI) as cross-linking agent. The swelling ratio of CMC-based hydrogels could reach 64 g/g, much higher than 15 g/g of chitosan hydrogels. Yang et al.[37] reviewed the structure and properties of physically cross-linked hydrogels based on natural polymers, pointing out that chitosan and its derivatives have unique advantages in constructing high-strength hydrogels.

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Protein/polypeptide materials can form fibrous structures through self-assembly, providing a multi-level structural basis for hydrogels[38]. Xue et al.[39] prepared high-strength, high-toughness, rapid-recovery and fatigue-resistant hydrogels based on picot peptide fibers, with fracture stress of 4.1 MPa, toughness of 25.3 kJ/m^2, and fatigue threshold of 424 J/m^2, far exceeding conventional synthetic hydrogels. Such excellent performance stems from the "hidden length" design in the fibrous structure: under stress, the β-sheet structure in the fiber gradually unfolds, releasing the hidden length to dissipate energy while maintaining network connectivity.

8. Topological Structure Hydrogels

Topological structure hydrogels optimize mechanical properties through unique network topology design[40]. Slip-ring hydrogels are designed based on rotaxane structure. Cross-linking points can slide along polymer chains, avoiding stress concentration caused by uneven dispersion of chemical cross-linking points. Okumura and Ito[41] first reported slip-ring hydrogels based on rotaxane structure. When subjected to external force, cross-linking points slide to dissipate energy, and at the same time, local stress concentration is eliminated through segment rearrangement to prevent polymer chain breakage. This design enables the elongation at break of slip-ring hydrogels to reach more than 20 times.

Homogeneous network hydrogels are prepared by end-group coupling reaction of four-arm polyethylene glycol. The network structure is uniform and regular, avoiding local stress concentration around short chains in traditional hydrogels[42]. Sakai et al.[43] prepared homogeneous network hydrogels through aminolysis reaction of four-arm polyethylene glycol. Even at 90% water content, the compressive strength can still reach 60 MPa, far exceeding that of natural cartilage. This performance indicates that improving the uniformity of network structure is an effective way to achieve high mechanical properties.

9. Influencing Factors and Reinforcement Mechanisms of Mechanical Properties

9.1. Systematic Analysis of Key Influencing Factors

Cross-linking density has a dual effect on the mechanical properties of hydrogels: moderate increase in cross-linking density can improve the strength and modulus of gels, but excessive cross-linking density will limit segment movement ability and make gels brittle[44]. Guo Shoucheng et al.[45] prepared poly(p-dioxanone)/polyvinyl alcohol (PPDO/PVA) composite hydrogels. By adjusting PPDO content (optimal at 3%), the tensile strength reached 0.63 MPa and elongation at break reached 481%, about 3 times that of pure PVA hydrogels. Studies have shown that there is an optimal cross-linking density range, within which hydrogels can obtain the best comprehensive mechanical properties.

Increasing polymer concentration can improve network density and mechanical strength, but there is also an optimal range. Kang et al.[46] prepared p-DN hydrogels. When the solid content was 19%, the compressive strength reached 37.80 MPa, but when the solid content increased to 23%, the compressive strength decreased to 7.18 MPa, which was due to the excessive PAM molecular chains hindering the stress transfer efficiency.

Environmental factors such as pH, temperature and ionic strength have significant effects on the mechanical properties of hydrogels. Wu Ying et al.[47] prepared poly(N-isopropylacrylamide)/polyacrylic acid hydrogels with both temperature and pH dual responsiveness. The swelling ratio was the highest at pH=7, and showed high-temperature shrinkage when the temperature was higher than 50 ℃. This is related to the protonation/deprotonization of ionizable groups in the network and hydrophobic interaction.

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9.2. Theoretical Understanding of Reinforcement Mechanisms

The toughening mechanism of double network hydrogels comes from the fracture and energy dissipation of the first network as "sacrificial bonds"[48]. Brown et al.[49] and Tanaka et al.[50] proposed a crack tip local yielding and hardening model. They believed that in the crack tip region with stress concentration, the first network preferentially breaks to form a softened damage zone, and cracks need to pass through the damage zone to further expand, which significantly improves the fracture energy. Studies have shown that the size of the damage zone and energy dissipation density are key parameters determining the toughening effect[51].

Hydrogels with entanglements far exceeding cross-links achieve strengthening and toughening through dense physical entanglements[52]. Kim et al.[53] prepared hydrogels in which the number of entanglements far exceeds the number of cross-links. When subjected to external force, tension is transmitted along the chains and dispersed to surrounding chains through entanglement points. Even if chain breakage occurs, energy will not be released intensively. This mechanism endows hydrogels with both high toughness and low mechanical hysteresis.

The mechanisms of nanoparticle composite reinforcement include: nanoparticles act as physical cross-linking points to increase network density; interfacial friction between nanoparticles and matrix dissipates energy; high-modulus nanoparticles themselves bear part of the load[54]. The three mechanisms work together to achieve the synergistic effect of strengthening and toughening.

9.3. Constitutive Relationships and Models

The mechanical behavior of hydrogels can be described by various constitutive models[55]. Rubber elasticity theory is suitable for describing elastic behavior under small strain, correlating stress with the entropic elasticity of network chains. According to rubber elasticity theory, shear elastic modulus $\mu_x=\nu_x kT$, where ν_x is the effective chain density[56]. For large strain behavior, the non-Gaussian characteristics and finite extensibility of chains need to be considered.

The constitutive relationship of double network hydrogels is more complex, and the independent contributions and interactions of the two networks need to be considered simultaneously. Studies have shown that the stress-strain curve of double network hydrogels can be described by a two-network parallel model. The first network contributes to yielding and strain softening behaviors, and the second network contributes to strain hardening behaviors under large strain[57].

10. Conclusion and Prospect

Great progress has been made in the mechanical property enhancement of PVA-based hydrogel composites. Various reinforcement strategies have their own characteristics: double network structure is ingeniously designed to achieve toughening through sacrificial bond energy dissipation, but faces the problem of irreversible damage; nanocomposite has significant reinforcement effect and can transfer stress through interfacial interaction, but has the challenge of uneven dispersion; cross-linking regulation is simple and effective, and the Hofmeister effect can adjust properties in a wide range, but needs to balance other functions; natural polymer composite is environmentally friendly and safe, and can synergistically improve biocompatibility, but interfacial compatibility needs to be improved; topological structure design is novel, and slip-ring and homogeneous networks avoid stress concentration, but the preparation process is complex.

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