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A Review of Hydrogels: Types, Formulation Mechanisms, and Applications

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ABSTRACT

A hydrogel is a three-dimensional polymer network that is insoluble in water and can absorb fluids from the body in a biological setting. Hydrogels absorb water through hydrophilic groups, capillary action, and osmotic pressure. There are generally three types of hydrogels dependent on the polymer used: natural, synthetic, and natural-synthetic. Polymer networks develop by physical and chemical cross-linking. The preferred crosslinking process is determined by the hydrogel's intended properties and applications. Stimulus sensitive hydrogels (smart hydrogels) are 3D network materials that change physically or chemically in response to environmental stimuli such as temperature, pH, light, enzyme activated, and particular chemicals. Hydrogel is employed in many medical applications, including wound healing, contact lenses, tissue engineering, 3D bioprinting, and the release of medicinal substances. Its structure is comparable to that of the extracellular matrix (ECM) and its capacity to absorb water. This review article focuses on the types of hydrogels, the mechanism of hydrogel formulation, and their applications.

Keywords: Hydrogels, Polymer networks, Stimuli-responsive (smart) hydrogels, Wound healing, Tissue engineering.

1.Introduction

Hydrogels are three-dimensional networks of polymer chains that can absorb and retain water in their interstitial spaces [1]. Hydrogels absorb water due to their hydrophilic groups (e.g. -NH2, -OH, -COOH, -SO_3H), capillary action, and osmotic pressure [2]. They also resist dissolution due to cross-linking between the polymer chains. Hydrophilic groups produce secondary contacts between hydrogels and biological tissues, including the mucous membranes and epithelial tissues. Hydrogels can be cross-linked, either physically or chemically, preventing them from dissolving even after absorbing significant volumes of water or physiological fluids [3]. In physical hydrogels, cross-linking occurs through secondary hydrogen bonding between polar groups on the polymer chains, while in chemical hydrogels, cross-linking occurs through covalent bonds between functional groups on the polymer chains, which are facilitated by special cross-linking agents [4].

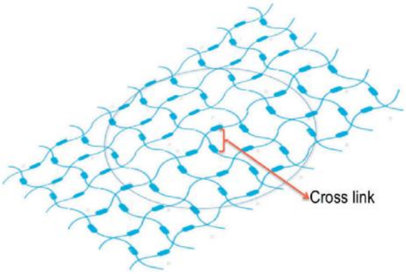

Figure 1. Structure of hydrogel [5]

Hydrogels are suitable for biomedical applications such as tissue engineering and drug delivery due to their softness, hydrophilicity, superabsorbancy, viscoelasticity, biodegradability, biocompatibility, and similarity to extracellular matrix [5,6]. Hydrogels have minimal toxicity, tissue injury, and no inflammatory or thrombotic reactions. Hydrogels' reversible responses to stimuli such as pH, temperature, electric field, magnetic field, solution ionic strength, and biological molecules make them valuable for various biomedical applications [6].

2. Types of hydrogels

There are three types of hydrogels based on the polymer used: natural, synthetic, and natural-synthetic polymer. Natural or synthetic polymers that have been hydrogenated are regarded as raw materials for use in medicine. Hydrogels are made of natural and synthetic polymers that must be biocompatible, biodegradable, and blood compatible in some applications where the hydrogel comes into contact with blood [7].

2.1 Natural hydrogels

Natural hydrogels contain polymers derived from natural sources. Hydrogels made using natural polymers are biocompatible, biodegradable, and non-toxic. The usage of natural polymers in hydrogel manufacturing relies on the biomaterial's intended function. Hydrogels employed for controlled material release should be biocompatible, biodegradable, and informal [8]. Natural polymers, such as polysaccharides and proteins, are commonly employed to release chemicals. Polysaccharides are more appropriate because of their biocompatibility, enzymatic breakdown, high durability, and non-toxicity, according to the results of in-body testing of these polymers. Natural hydrogels utilized in clinical settings include alginate, collagen, gelatin, and fibrin. Alginate, for example, has been used to normalize left ventricular activity following a heart attack. Collagen has been utilized as a substitute to vascular bundles. Gelatin and fibrin have several applications, including artificial constructions, tissue engineering, adhesives, and anticoagulants in surgery [9].

2.2 Synthetic Hydrogels

Synthetic hydrogels are made from synthetic polymers like polyamides and polyethylene glycol (PEG). Synthetic polymers have lately been used to replace natural polymers in hydrogel construction due to benefits such as longer life, gel strength, and increased ability to absorb water. Hydrogels made from synthetic polymers have a variety of medical applications. Synthetic polymers are superior to natural polymers due to their hydrophobic properties, mechanical structure, and chemical composition. These polymers include polyacrylamide, poly vinyl alcohol (PVA), and PEG[10].

PEG is a common polymer for synthetic hydrogenation in a wide range of medical applications, including medication release, tissue engineering, bone prostheses, and wound dressings. This polymer's characteristics, including biocompatibility, immune system non-stimulation, and resistance to protein adsorption, make it ideal for medical applications. PEG alone can form insoluble network structures. Adding factor groups increases crosslinking in the hydrogen network [8].

2.3 Hybrid Hydrogels

Hybrid hydrogels, which combine natural polymers like alginate and xanthan gum with synthetic polymers like PAM and PVA, provide unique alternatives for pesticide encapsulation in agriculture. These hydrogels improve biodegradability and biocompatibility, reduce soil and water pollution hazards, and offer high mechanical and chemical stability. Pesticides are effectively protected in various farm situations through controlled release under predetermined parameters [7]. Despite obstacles in balancing material ratios, eco-friendly synthesis, and cost reduction, these hydrogels are a significant advancement in pesticide encapsulation technology, supporting effective pesticide usage and sustainable agricultural practices [8].

3. Mechanism of hydrogel formulation

3.1 Physical Cross Linking

The interactions between polymer chains in physically cross-linked hydrogels are based on physical interactions rather than covalent ones. Coordination bonds, hydrogen bonds, van der Waals forces, and hydrophobic interactions are a few examples of these interactions [11,12]. Physical cross-linking, as opposed to chemical cross-linking, is reversible in specific circumstances, allowing the hydrogel to change structurally without rupturing any covalent bonds. Because of this property, physically cross-linked hydrogels react more strongly to outside stimuli such as ionic strength, pH, and temperature [13]. They might have special qualities, like "self-healing" behavior, in which the gel can mend itself after breaking. These interactions create hydrogels, which are special physical gels with great temperature reversibility and water sensitivity [14,15]. Hydrogels have a limited lifespan in physiological media, lasting from a few days to a month. Hydrogels are typically utilized for short-term medication release [16]. These hydrogels are safe to employ in therapeutic settings as they do not require hazardous covalent crosslinking chemicals. This method involves producing hydrogels via non-covalent methods such as electrostatic, hydrogen bonding, and hydrophobic forces between polymer chains [17,18]. Hydrogel preparation methods offer advantages including high water sensitivity and thermal reversibility. Hydrogels generated with this technology are safe for therapeutic use as they don't require harmful covalent crosslinking chemicals. Chitosan can be combined with small anionic molecules like sulphates, phosphates, and citrates of Pt, Pd, and Mo to create hydrogels using physical techniques. Hydrogel synthesis is influenced by anions' charge and size, as well as chitosan deacetylation concentrations [19,20].

3.2 Chemical Cross linking

Clustering of molecules in physical gels creates free chain loops and inhomogeneity, indicating network defects with short lifespan. Hydrogel networks are easier to prepare and use than physical hydrogels since they are not affected by pH. Chemical crosslinking can modify the physical properties of hydrogels [21]. Chemically cross-linked hydrogels form covalent connections between their polymer chains. The strong and stable covalent bonds form a 3D network of interconnected polymer chains. Cross-links are often produced through chemical events like polymerization or agent-induced reactions.The hydrogel structure is stronger and more resilient to variations in environmental factors like pH and temperature when covalent bonds are present [22,23]. As a result, chemically cross-linked hydrogels often have higher long-term stability and mechanical strength. Since the preparation process and applications of chemically crosslinked hydrogels are independent of pH, they are simpler to manage than physical hydrogels. Tan et al. used a Schiff base approach to manufacture N-succinyl chitosan that functions as an alized hyaluronic acid injectable composite hydrogel [24]. As the proportion of N-succinyl chitosan in the hybrid hydrogel increased, it was found that the compressive modulus a crucial component for cartilage tissue engineering improved. The identical hydrogel fabrication technique employing cellulose and alginate was also reported by Ito et al. [25]. The amino group's derivatives create hydrogels by Michael's addition reactions. The amino groups interact with the vinyl groups of other polymers. Hydrogels improved mucoadhesive qualities in certain formulations. This approach has some drawbacks, such as the multiple-step preparation and purifying process. Polymers may become cytotoxic after functionalization with reactive groups [26].

4. Stimulus responsive hydrogels (Smart hydrogels)

4.1 Thermoresponsive hydrogels

Thermoresponsive hydrogels (also known as thermogels) are polymers that produce gels when swollen in water. Thermogelation in aqueous solutions occurs through multiple methods, some of which are still debated for specific polymers. Many thermoresponsive polymers exhibit a decrease or increase in solubility due to changes in the hydrophilicity of their chains in response to temperature fluctuations. The behavior of thermoresponsive hydrogels in aqueous solutions depends on three types of interactions [27]: (1) between polymer molecules, (2) between polymer and water molecules, and (3) between water molecules themselves. A rise in temperature causes a net unfavorable free energy for polymer–water interactions for polymers with a lower critical solution temperature (LCST), which encourages associations between polymers and water. Thermogels can undergo a sol–gel phase transition in response to temperature changes because they are often made of amphiphilic polymers with both hydrophilic and hydrophobic regions [28]. Unlike traditional polymers that melt at greater temperatures within a certain range of temperatures, certain thermogels gel when heated and return to a liquid condition when the temperature drops.

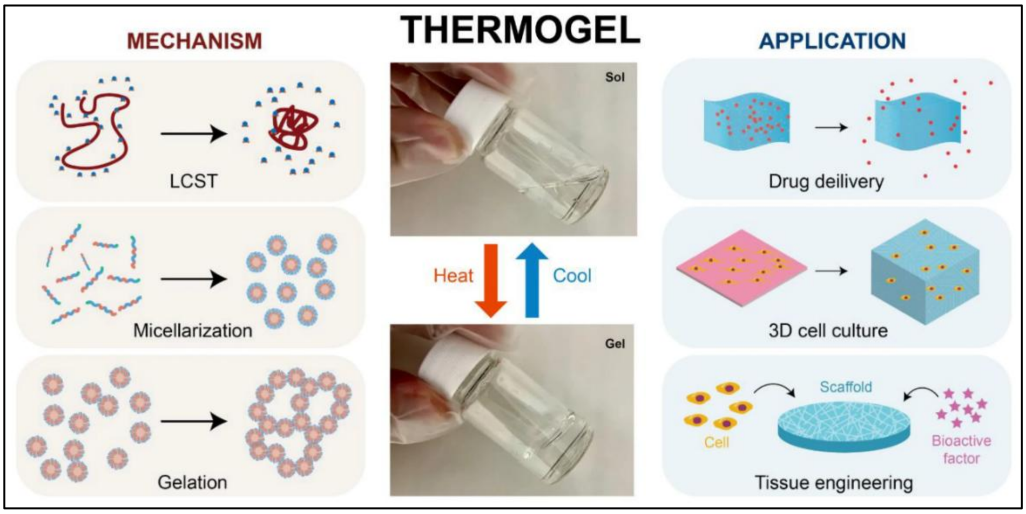

Figure 2. A schematic summary of the thermogelling behaviour, illustrating the combined effects of multiscale thermoresponsive mechanisms, as well as the various applications of thermogels [29].

4.2 pH-responsive hydrogels

pH-sensitive hydrogels are under intense development, with numerous parameters impacting their swelling and deswelling behavior. Factors influencing network properties include ionic charge, ionization degree, pH, monomer type, polymer concentration, and hydrophilicity. Anionic hydrogels swell when weakly acidic functional groups (e.g., COO−, SO−, and PO−) ionize at high pH levels [30]. At lower pH, cationic hydrogels swell due to ionization of basic functional groups such amines. pH-responsive hydrogels have significant environmental applications such as water treatment and purification, wastewater management, controlled pesticide release, environmental sensing, and soil remediation [31,32].

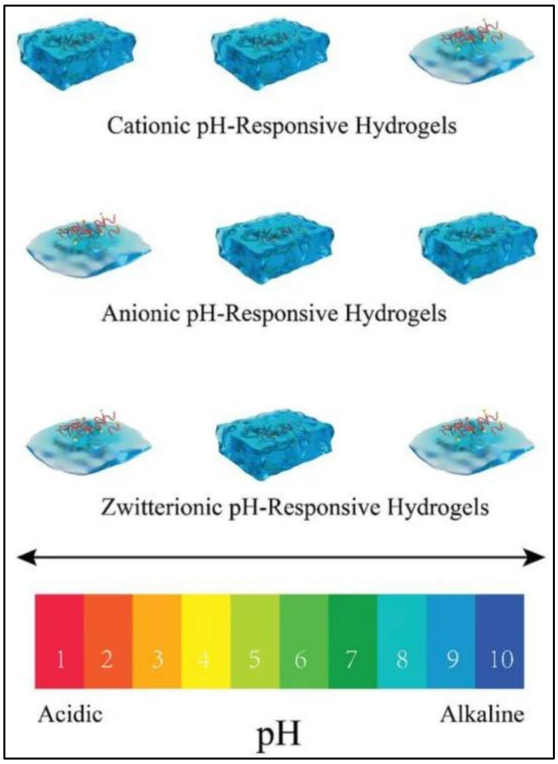

Figure 3. General working mechanism of pH responsive hydrogels [33]

4.3 Photo-responsive hydrogels

Photo-responsive hydrogels are typically made up of a polymer network and a photoreactive component, which is usually a photochromic chromophore that serves as the functional element. Initially, photochromic molecules absorb the optical signal [34]. The chromophores within the photoreceptor then convert the light signal into a chemical response by photoreactions like isomerization, bond cleavage, or dimerization. This chemical signal is then conveyed to the hydrogel's functional component, which controls its properties. The reaction of the chromophores to light is substantially dependent on their molecular structure, and hence, the needed irradiation varies appropriately. Photo-responsive hydrogels are typically composed of a polymer network and a photoreactive component, which is commonly a photochromic chromophore that functions as the functional element. Initially, photochromic molecules absorb the optical signal [35]. The chromophores within the photoreceptor subsequently translate the light signal into a chemical response by photoreactions such as isomerization, bond breaking, or dimerization. This chemical signal is subsequently routed to the hydrogel's functional component, which regulates its properties [36].

4.4 Enzyme-triggered hydrogels

Enzyme-triggered hydrogels gel or disintegrate through specific enzymatic processes, such as proteolysis or glycosylation, allowing for focused responses in wound beds rich in enzymes like MMPs. These systems use enzyme-specific substrates (e.g., MMP-cleavable peptides like GPQGIAGQ) as crosslinkers in networks such as PEG or HA; enzyme action cleaves bonds, resulting in gelation from cyclic-to-linear peptides or degradation for payload release. HRP/H2O2 oxidizes tyramine phenols in HA-Tyr, resulting in fast coupling; MMPs hydrolyze peptide linkages in PEG hydrogels, allowing for remodeling [37].

5. Applications

5.1 Wound Healing

Super absorbent hydrogels, also known as super porous hydrogels, may absorb up to 90% of their weight in water without breaking down. This makes them suitable for wound healing dressings, including alginate-based hydrogels. They offer bioadhesive and sealing mechanisms as alternatives to staples and sutures. Wound healing involves four phases: homeostasis, inflammation, proliferation, and remodelling [38,39]. Delays in healing, particularly after three months, might result in chronic wounds due to inflammation.

Hydrogel dressings are helpful for chronic wounds as they may be tailored with growth factors, biomolecules, and medications. Adding anti-inflammatory chemicals, such as phenolic compounds from honey or plant extracts, can improve hydrogels and promote healing by lowering inflammation. Xin et al. [40] created a hydrogel for diabetic wound healing by mixing oxidized Gastrodia elata polysaccharide (OGEP) with a gastrodin-chitosan combination (GAS/CS) via a Schiff base reaction. The hydrogel containing EGCG microspheres was evaluated for its effectiveness in treating diabetic wounds, as well as its biosafety, rheology, and hemostasis features.

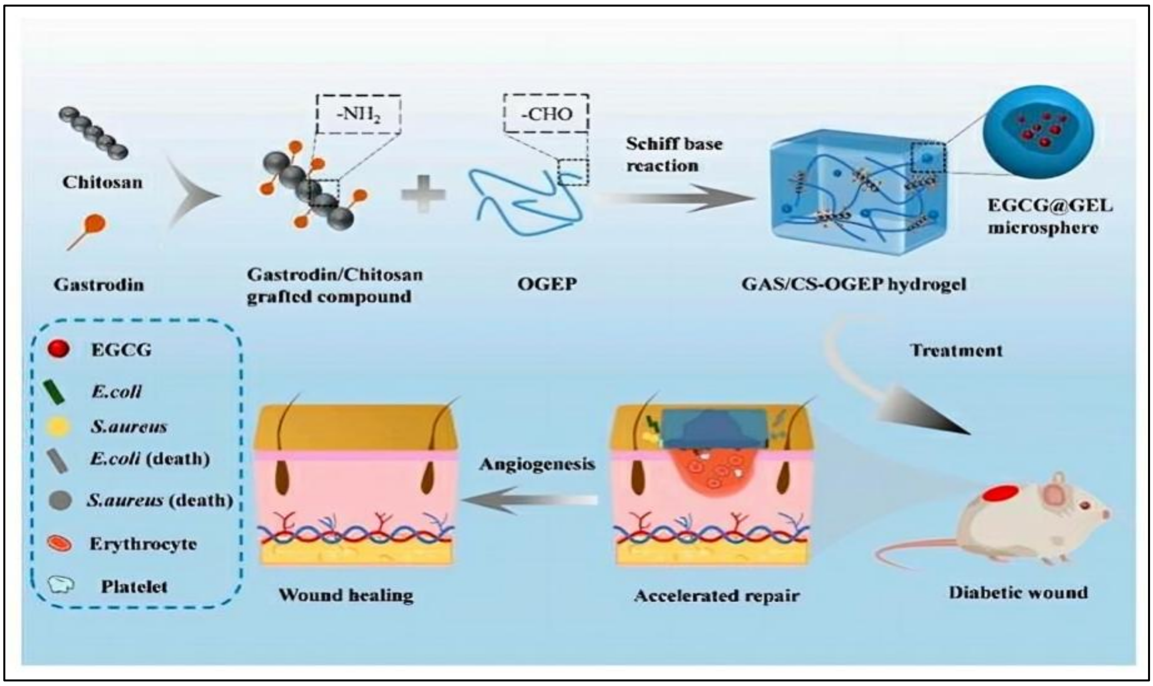

Figure 4. Schematic illustration of EGCG@GEL/GAS/CS-OGEP hydrogel prepared and applied to diabetic wound healing [40].

5.2 Contact Lenses

Contact lenses are mostly used as an alternative to spectacles for vision correction. They are being developed for drug administration, ocular defect correction, post-surgical wound repair, and cosmetic applications [41]. Traditional therapies like ocular drops and ointments have limitations, including limited permeability and frequent dosage. Contact lenses are being investigated as a viable alternative for delivering ophthalmic drugs.

Contact lens materials should be robust, stable, transparent, and provide sufficient oxygen to the cornea. Mechanical, optical, and chemical elements all contribute to lens properties. Wettability helps maintain a stable tear film, while mechanical testing affects comfort, fit, and durability. Optical qualities promote good vision. Hydrogels are advised as they are very compatible with corneal cells.

Hydrogels have advanced clinical ophthalmology by being used in SCL, IOL, and drug-eluting hydrogels that target the eye's anterior parts [42]. Polymeric hydrogels, like as pHEMA, are widely used for SCL and drug delivery [43]. When compared to rigid lenses, SCL have higher water content, softness, elasticity, and oxygen permeability. In cataract surgery, IOLs, specifically hydrogels, are utilized to replace the native lens. Drug-loaded IOLs can improve drug delivery by lowering losses, adverse effects, and patient compliance concerns [44]. Yang et al. [45] found that electrodeposition allows for the creation of chitosan-based hydrogel contact lenses with improved transparency, mechanical strength, oxygen permeability, and Staphylococcus aureus resistance (Figure 5). This process also results in efficient cross-linking of chitosan and epichlorohydrin, presenting a fresh approach to contact lens production.

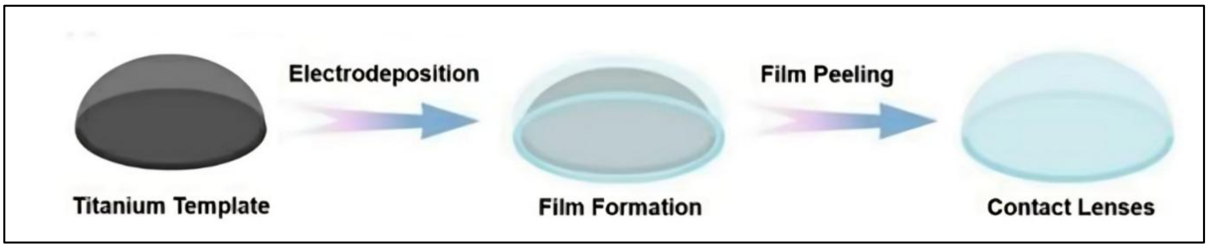

Figure 5. Electrofabrication of chitosan-based hydrogel contact lenses [45]

5.3 Tissue Engineering

Hydrogels stand out for their versatility in drug delivery and tissue engineering [46].

Tissue engineering regenerates tissue by integrating cells into a scaffold under physiological settings. Cells, scaffolds, and growth factors make up the "triad" of critical ingredients. Hydrogels as scaffolds promote cell adhesion, growth, and tissue regeneration. Their 3D polymer networks effectively store water, making them suitable for this application. Hydrogels can replicate the extracellular matrix (ECM) of native tissues via crosslinking methods. ECMs are often porous networks made up of fibrous proteins in a matrix of GAGs and proteoglycans, comparable to hydrogels [47].

Hydrogels are essential in tissue engineering for regenerating dermal tissue, cartilage, blood vessels, bone, the cornea, and other soft tissues. They imitate the ECM, making them excellent for repairing damaged tissues. Cartilage, lacking lymphatics, blood arteries, and nerves, relies on the extracellular matrix (ECM) to maintain cell microenvironment homeostasis. Cartilage scaffolds require high biocompatibility, porosity, and mechanical strength, similar to Type II collagen. Elastic hydrogels with smooth surfaces and high-water content imitate the ECM, making them excellent for cartilage regeneration. According to Mei Liu et al. [48], Figure 6 depicts a schematic of injectable hydrogels used for cartilage and bone tissue engineering.

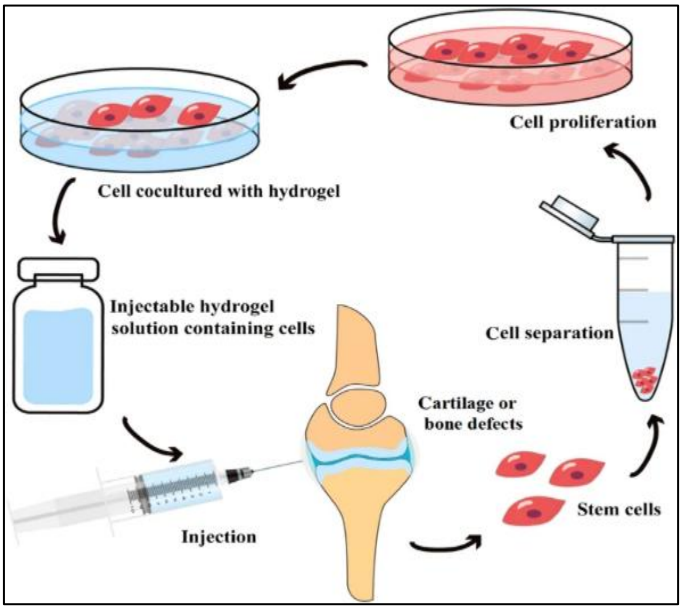

Figure 6. Schematic illustration of approaches to make injectable hydrogels for cartilage and bone tissue engineering applications [48].

5.4 3D Bioprinting

Bioprinting with hybrid hydrogels combines the advantages of both technologies, enabling layer-by-layer printing with solid filament structure and physical gelation during extrusion [49]. Hydrogels improve cell processes necessary for tissue engineering, such as proliferation, migration, and differentiation. Extrusion-based bioprinting is preferred due to its compatibility with low-viscosity hydrogel precursors. Hydrogel bioinks, made from cells and precursor solutions, play a crucial role in bioprinting tissues and organs due to their optimum rheological and biochemical properties [50].

Various crosslinking methods and bioink formulations have been devised to imitate tissue structures, resulting in effective printing using materials such as HA-based bioink crosslinked with calcium solutions [51]. Boons et al. [52] developed a 3D bioprinting platform to manufacture hydrogels containing live dialgae, which serve as biological monitors for water quality. These hydrogels enabled the visual evaluation of diatom proliferation to identify water contaminants when subjected to varying amounts of salt, herbicides, and antibacterial agents. Pollutant sensitivity testing was made possible by the diatoms' continued metabolic activity in the hydrogels.

5.5 Biosensors

Hydrogels have numerous advantages when used in electronic skin and wearable biosensors. Because they are elastic, flexible, and effectively adhere to human skin, they provide comfort.

Their water absorption maintains the skin moisturized and stable, and their conductivity ensures reliable signal transmission. Building complex hydrogels with conventional methods is challenging because they need combining multiple components to improve various properties. AI technology enables efficient screening and optimization of hydrogel components. High-throughput screening has led to the development of polysulfobetaine hydrogels with superior mechanical and self-healing properties, expanding their use in flexible electronics and enhancing intelligent features like wireless communication and real-time health monitoring. Because AI-integrated hydrogels can accurately recognize handwriting, they are ideal for intelligent human-device interfaces. To guarantee the stability, security, and privacy of these applications, more investigation is required. Laccase from Trametes versicolor was immobilized by Diaconu et al. [53] using a MWCNT-CS solution containing 25 U/mL laccase to create a nanocomposite film. Compared to existing polyphenol biosensors, the Au/Lacc-CS-MWCNT biosensor was five times more sensitive to micromolar concentrations due to the conducting MWCNTs.

5.6 Drug Release

Abdullahi et al. successfully created a hydrogel that was loaded with fluoxamine using ultrasound, demonstrating the medication’s-controlled release in a body-like environment. They contend that a number of variables, including the hydrogel's composition, geometric structure, preparation technique, drug type, and environmental conditions throughout the release period, affect the mechanism of drug release from the hydrogel, with pH being one of the most crucial variables [54]. Ganji et al. used injectable hydrogel, which is sensitive to chitosan temperature in vitro, to successfully release pyridostigmine bromide gradually in another experiment. They think that a darker solution than the unsalted chitosan solution was seen after adding glycerol phosphate salt. Over time, turbidity variations were observed in chitosan and chitosan / glycerol phosphate solutions with 8% salt content by weight/volume. Turbidity in chitosan solution did not vary appreciably over time at 37°C. Chitosan solution without glycerol phosphate salt is not temperature sensitive and can remain stable at 37°C for a long time. However, at 37° C, the chitosan / glycerol phosphate solution showed a small rise in turbidity over time. After 9 minutes, the darkness suddenly increased. The point of gel formation is when an abrupt increase in opacity is seen. This experiment demonstrated that adding glycerol phosphate salt to the chitosan solution decreased its stability at 37° C and quickly altered the phase from the solution state to the gel state [55].

Bakhshashi et al. successfully created a Baghdadit-vancomycin nanostructured scaffold with drug release, antibacterial activity, and biocompatibility. The drug release was tested on a net basis. The study used vancomycin as a drug model in the Baghdad scaffold to observe drug release behavior [56]. Drug release from the scaffold was detected in both an explosive and regulated way after immersion in phosphate-buffered saline solution (PBS). During the first 6 hours of explosive release, the release remained stable. However, after 36 hours, 45-75% of the medication was released from the scaffold for all compounds. Vancomycin effectively killed S. aureus and prevented replication on the scaffold. The study found that 35% of the antibiotic persisted in the scaffold, suggesting that the Baghdadit/vancomycin scaffold can effectively suppress bacterial infection in early phases of bone infection [56-58].

6. Conclusion

This review article discusses the various forms of hydrogels, their production mechanisms, and medical applications. Hydrogels are formed using several crosslinking processes. Hydrogels with physical networks have been employed for tissue engineering in medicinal applications such as releasing bioactive chemicals and limiting cell proliferation. These networks provide the advantage of not requiring organic solvents. Hydrogels release medicinal ingredients by structural swelling. Recently, there has been a growing interest in stimulus-responsive hydrogels. Temperature and pH influence the release of medicinal drugs from the hydrogel network. Hydrogels are a popular biomaterial for medical applications due to their ability to absorb water, their soft structure, biocompatibility, and similarity to the extracellular matrix.

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