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Review Article

A Comprehensive Review on Hydrogel

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ABSTRACT

Hydrogels are three-dimensional, hydrophilic polymeric networks capable of absorbing and retaining large amounts of water or biological fluids while maintaining their structural integrity. Due to their unique physicochemical properties, biocompatibility, flexibility, and similarity to natural tissues, hydrogels have gained significant attention in pharmaceutical, biomedical, agricultural, and environmental applications. This review provides a comprehensive overview of hydrogels, including their introduction, definition, types, classification, methods of preparation, and polymers used in their fabrication. Hydrogels can be categorized based on source, ionic charge, cross-linking mechanism, responsiveness to stimuli, and structural characteristics. They may be prepared through physical or chemical cross-linking techniques such as freeze-thaw cycling, ionic interactions, radiation polymerization, graft polymerization, and covalent cross-linking. Various natural polymers, including alginate, chitosan, gelatin, collagen, and hyaluronic acid, as well as synthetic polymers such as poly(vinyl alcohol), poly(ethylene glycol), polyacrylamide, and poly(N-isopropylacrylamide), are widely employed in hydrogel development. The selection of polymers and preparation methods significantly influences the mechanical strength, swelling behavior, biodegradability, and functionality of hydrogels. This review highlights the fundamental aspects of hydrogel technology and emphasizes their growing importance in advanced drug delivery systems, tissue engineering, wound healing, and other emerging biomedical applications.

INTRODUCTION

Hydrogels are the hydrophilic polyelectrolyte polymeric system of three dimensional which are physically or chemically cross-linked structures that absorb considerable quantity of water with

tunable biocompatibility, acute environmental sensing, biodegradability, and mechanical properties [1] [2] [3] [4] [5] [6] [7]. Hydrogels can be created by physically or covalently cross-linking natural or synthetic polymers. Hydrogels are sometimes referred to as hydrophilic gels

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because they can greatly expand in water media when biological fluid or water is used as the dispersion medium. Among the most important properties of hydrogels include swelling, softness, elasticity, flexibility, absorbent nature, and water storage capacity. This polymer can be produced naturally or synthetically [5] [6]. Water can be absorbed by the hydrogels for approximately ten to twenty times its molecular weight [6]. Hydrogels' ability to absorb water is attributed to the hydrophilic functional group attached to the polymeric support, and their resistance to dissolution is provided by the crosslinks between the network chains [7]. Another title for dry hydrogels is "Xerogels," even though the term "hydrogels" implies that the material has already swelled in water. This dehydrated hydrogel, known as aero gel, is incredibly light and has a porosity of up to 98% once water is removed without upsetting the polymeric network, either by lyophilization or extraction with organic solvents. During the drying process, surface tension causes the gel body to break down. Hydrogels are frequently utilized as drug carriers because of their ease of production and self-application. The creation of a large and continuous surface area is one of their primary benefits for widespread use in scientific and clinical applications. A number of polymer combinations are created into hydrogel

formulations in order to investigate their potential as a drug delivery method. The complimentary properties of natural and synthetic polymers may lead to mechanical stability and biological acceptability when combined. It is found that the hydrogels are stable and robust [8] [9] [10]. Because the acid groups are ionized in water, the polymer picks up several negative charges along its length. There are two possible outcomes. First, because the negative charges repel one another, the polymer is forced to expand. Second, polar water molecules are drawn to the negative charges. The resultant combination is more viscous because the polymer chain now takes up more space and prevents solvent molecules from moving around it. Although the polymer and the surrounding water are in equilibrium, this equilibrium could be disrupted in a number of ways. When the ionic concentration of the solution is increased, for as by adding salt, positive ions bind to the negative sites on the polymer. The charges are effectively neutralized by this. The polymer thus folds in on itself once more. When alkali is added, the point of equilibrium moves to the right; when acid is added, it moves to the left, which has the opposite effect. Hydrogels are available in various distinct types, and they all expand and contract at different pH levels, temperatures, and ionic concentrations [7].

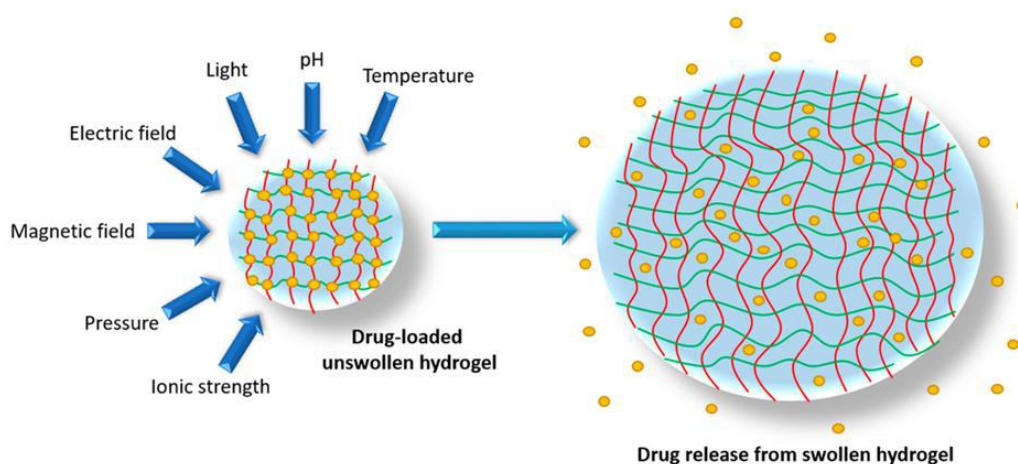


Figure 1 Swelling carried on by various chemical and physical events in a drug delivery hydrogel. The interwoven matrix structure of a hydrogel is represented by red and yellow lines, with yellow spots denoting drug molecules [11].s

there are two types of hydrogels on the market: chemical and physical hydrogels. Since this would lead to the creation of ionically connected domains and molecular entanglement clusters, physical hydrogels are not uniform. Free chain ends or chain loops indicate transient network defects in physical gels. Physical hydrogels known as ionotropic hydrogels, such as calcium alginate hydrogels, are produced when a polyelectrolyte is combined with a multivalent ion that has the opposite charge. In certain situations, complex coacervates or poly-ions are created when polyelectrolytes with different charges are mixed together. These interactions can be affected by variations in temperature, pH, ionic strength, stress applications, or the addition of the solute that competes with polymeric ligands for the protein's affinity site. Cross-linking water-soluble polymers or converting hydrophobic polymers into hydrophilic polymers and then cross-linking to create a network are two methods used to create chemical hydrogels. Hydrogels that are covalently cross-linked into networks are referred to as "permanent" or "chemical" gels. Additionally, chemical hydrogels are not homogeneous. They usually have "clusters," or regions with high crosslink density and low water swelling, dispersed throughout regions with high swelling and low crosslink density [12]. Hydrogels are the artificial biomaterials that most closely resemble genuine living tissue. Their delicate, tissue-like texture and high water content are the reasons behind this. Hydrogels' unique physical properties have drawn a lot of interest in their usage in medication delivery applications [3]. High water content materials also improve their biocompatibility. Hydrogels are therefore utilized in medication delivery systems, biosensor

membranes, prosthetic skin, contact lenses, and artificial heart linings. Hydrogels are also used as carriers that can interact with the mucosa lining of the GI tract, colon, vagina, nose, and other parts of the body because they can remain longer at the delivery location. The interaction between these carriers and the glycoproteins in the mucosa is thought to be mostly dependent on hydrogen bonding. Therefore, it appears that materials having a high density of hydroxy and carboxyl groups are appealing for this kind of use. The most common monomers used in the synthesis of mucoadhesive polymers (MAA) are acrylic and methacrylic acid [8].

Advantages

1. Hydrogel is more elastic and stronger.
2. Hydrogel possess good transparent properties and easy to modification.
3. Due to their significant water content they possess a degree of flexibility very similar to natural tissue.
4. They are biocompatible, biodegradable and can be injected.
5. Hydrogel have ability to sense change pH, temperature, or the concentration of metabolite and release their load as result of such a change.
6. Release of Medicines or nutrients timely.

Disadvantages

1. High cost.
2. Non-adherent and may need to be secured by secondary dressing and also cause sensation felt by movement of the maggot.
3. Difficult to sterilize.



4. In contact lens less deposition hypoxia, dehydration and red eye reactions.

translucent hydrogel is produced by adding epichlorohydrin as a cross-linker.

Classification of hydrogel

On the basis of origin

Natural polymers forming hydrogels

These hydrogels are biodegradable, biocompatible, and nontoxic. However, their mechanical quality is low, and batch variation may lead to poor reproducibility [41, 24].

Synthetic hydrogels

Synthetic hydrogels are cross-linked polymers made by ring-opening polymerization or the addition reaction under carefully regulated circumstances. In the production of synthetic hydrogels, polyacrylic acid and its derivatives, polyvinyl alcohol polyethylene glycol and its copolymers [17], and polyvinylpyrrolidone, among others, are frequently used as skeletons.

On the basis of composition

Hydrogels are categorised into four types based on their polymer composition: (1) homopolymeric hydrogels, (2) copolymeric hydrogels, (3) semi interpenetrating networks (semi-IPNs), and (4) IPNs [2].

Homopolymeric hydrogel

Polymer networks created from a single monomer species are known as homo-polymers. In every polymer network, it acts as the basic structural component (Lizawa et al., 2007). One example of a homopolymeric hydrogel made from natural polymers is cellulose hydrogel, which is made by dissolving cellulose in a urea/NaOH solution during a one-step polymerization process. A

Copolymeric hydrogel

One of the two types of monomers that make up co-polymeric hydrogels is hydrophilic. [16] developed a biodegradable triblock poly (ethylene glycol)-poly (caprolactone)-poly (ethylene glycol) (PECE) co-polymeric hydrogel for use in drug delivery systems.

On the basis of charge

Hydrogels are classified into five categories based on the sort of charges present on the polymer network.

1. Nonionic (neutral) substances such as dextran, agarose, and pullulan;
2. Anionic substances like carrageenan
3. Cationic substances, such as chitosan;
4. Amphoteric electrolyte collagen
5. Polybetaines with zwitterionic properties, such as polyanionic xanthan and polycationic chitosan (2, 46)

On the basis of cross linking

Physical hydrogels

Noncovalent interactions (secondary bonds) between linear molecules that produce physical cross-linking joints, such as electrostatic contact, hydrogen bonding, chain entanglement, and hydrophobic interaction, result in three-dimensional networks known as physical hydrogels [13]. Because it takes relatively little energy to break the physical bonds between the molecules, physical hydrogels often show reversible solgel conversion [14]. They can be used in biological applications because they don't require a chemical reaction during production and the conditions are usually mild.



Chemical hydrogels

Chemical cross-linking between molecules forms chemical hydrogels, and this cross-linking is irreversible. Chemical hydrogels often feature stable qualities, adjustable architectures, excellent mechanical properties, and so forth [15].

On the basis of physical state

Solid hydrogels

Solid hydrogels can swell in aqueous environments including water, buffer solutions, and biological fluids, but they are typically chemically cross-linked and solid at room temperature. They can be used to create hydrogels for biomedical, environmental, and ecological applications since they can mimic the physical, chemical, electrical, and biological properties of the majority of biological tissues. The mechanical properties of the polymer matrix are enhanced by the addition of nanoparticles. For instance, gelatin-collagen enhanced with bioactive glass nanoparticles for cardiac tissue engineering and methacrylate gelatin reinforced with multiwalled COOH-functionalized carbon nanotubes (CNTs) are two examples [17].

Semisolid hydrogels

The adhesive interactions of semisolid hydrogels with soft-tissue networks and interfacial forces (van der Waals, hydrogen bonds, and electrostatic forces) set them apart. These hydrogels are also referred to as bioadhesive or mucoadhesive hydrogels due to their bioadhesive characteristic. They are used in the biomedical industry for efficient dosage and prolonged drug administration [17]. Hydrogels derived from the naturally occurring polysaccharides sterculia gum and poly (vinylpyrrolidone), both of which are biological in nature, fall under this category.

Recently, a starch nanocrystal-based hydrogel was developed for transdermal use [17].

Liquid hydrogels

liquid hydrogels are in a liquid phase at room temperature, but at a specific temperature they have an elastic phase that resembles soft tissue and is functional. There are numerous biomedical uses for these hydrogels, which are injectable. High mannuronic alginate hydrogels that are employed as wound dressings in cutaneous wound healing fall under this group. Microbial TG and human-like collagen might be combined to create a smart injectable hydrogel that could be utilized as a soft substance to create skin tissue. One option for a dressing material is keratin silica hydrogel [17].

Characterization Of Hydrogels[18]

Generally hydrogels are characterized for their morphology, swelling property and elasticity. Morphology is indicative of their porous structure. Swelling determines the release mechanism of the drug from the swollen polymeric mass while elasticity affects the mechanical strength of the network and determines the stability of these drug carriers. Some of the important features for characterization of hydrogels are as follows

Morphological characterization

Hydrogels are characterized for morphology which is analyzed by equipment like stereomicroscope. Also the texture of these biomaterials is analyzed by SEM to ensure that hydrogels, especially of starch, retain their granular structures

***In-vitro* release study for drugs**

Release studies are conducted to comprehend the process of release over a period of application



because hydrogels are swollen polymeric networks that contain drug molecules.

FTIR (Fourier Transform Infrared Spectroscopy)

Hydrogels' IR absorption spectra are altered by any morphological changes because of stretching and O-H vibration. The emergence of bands near 1648 cm indicates the formation of a coil or helix, which is indicative of cross linking.

Swelling behavior

To determine the swellability of these polymeric networks, the hydrogels are submerged in an aqueous media or a liquid with a particular pH. These polymers exhibit swelling-related increases in dimensions.

Rheology

Hydrogels are evaluated for viscosity under constant temperature of usually 4°C by using Cone Plate type viscometer

Methods of Hydrogels Synthesis

Depending on the desired structure and use, hydrogels can be generated by establishing

cross-linking in various ways. Grafting polymerization, physical cross-linking,

chemical cross-linking, and radiation cross-linking are among the different preparation

procedures used.

Physical Cross-Linking

Hydrophobic groups, hydrogen bonds, and electrostatic attraction interact to form self-assembling hydrogels, such as physically cross-linked hydrogels, which are then followed by non-

self-assembling macromolecules. Physically cross-linked hydrogels cannot dissolve due to physical interactions between different polymer chains [19].

Because the physically cross-linked hydrogels can revert to their polymer chain, these connections are disrupted by the application of stress or changes in the physical environment. Due to their ease of synthesis and lack of a cross-linker, physically cross-linked hydrogels are the newest materials. The dependability of the materials to be imprisoned (such as cells, proteins, etc.) is affected by these cross-linking agents

Ionic-Interaction

Ionic polymers are cross-linked using this method. For example, ionic polymers can be cross-linked by inserting divalent or trivalent counter-ions [20]. The idea behind this method is to gel a polyelectrolyte solution (like Na-alginate) using multi-valent ions of opposing charge (like $\text{Ca}^{2+} + 2\text{Cl}^-$).

Complex Coacervation

When a poly-anion and a poly-cation polymer interact, complex coacervate gels can be created. This process creates soluble and insoluble complexes by the opposing polymer charges adhering to one another. The concentration and pH of the solution determine these complexes [21]. One such instance is the coacervation of polycationic chitosan with polyanionic xanthan. Polyion complex hydrogels are created when positively charged proteins below their isoelectric point bond to anionic hydrocolloids [22].

Hydrogen bonding

Hydrogel synthesis can be regulated by temperature, solvent type, solution concentrations, and degree of polymer functionalities. Increasing



the polymer content causes hydrogels to become more stable and organized due to increased entanglements and hydrogen (H-) bonding interactions [23]. However, diluting the solution causes the network H-bonding to weaken and become disrupted in a few of hours. For example, during the formation of the PVA and adipic acid dihydrazide hydrogel, a hydrogen bond is formed between the –OH of PVA and the –NH₂ group of adipic acid dihydrazide [24].

Hydrogels can be prepared by repeating the freezing and thawing processes, i.e., they are frozen and then melted at room temperature for several cycles. The creation of structures including microcrystals is part of this process [25]. PVA is the most representative and most researched polymer that can be freeze-thawed into a hydrogel, however this process also depends on the creation of H-bonds between the polymeric chains [24].

Heat-Induced Aggregation

This process creates the hydrogel by applying heat, which causes the polymers to aggregate. Although the majority of gum Arabic (acacia gums) is made up of carbohydrates, it also contains 2-3% protein as an inherent component. Gum Arabic is composed of glycoprotein (GP) and arabinogalactan protein (AGP). Heat-induced proteinaceous component aggregation increases molecular weight, improving the produced hydrogels' mechanical properties and water-holding capacity [26].

Hydrophobic Interactions

An amphiphilic copolymers can be used to make the hydrogels by using an equilibrium part of hydrophilic and hydrophobic polymers [27]. Many hydrogels can be formed by the polymers cross-linking method via the hydrophobic interactions.

When the polymer is heated and dehydrated, the interactions of hydrophobic units occurred with each other result in the formation of hydrophobic linkages. This linkage is facile because the heating process of the polymer solution accelerates the gelation process. These hydrophobic associations are supposed to cross-link the hydrogels. Diblock, triblock, and multiblock copolymers are examples of amphiphilic block copolymers. The nature of the polymer and the solution concentration of the polymer greatly influenced the characteristics of the amphiphilic block copolymer, because hydrogel is formed at an optimum concentration of the polymer solution [28,29].

Chemical Cross-Linking

To create permanent cross-linking in the hydrogels, the chemical cross-linking approach is preferred, which employs covalent interaction between polymer chains. Chemical cross linking is the use of cross-linking agents or monomer grafting on the polymer backbone. Functional groups such as amino, carboxyl, and alcoholic groups containing synthetic and natural polymers that can be cross-linked by using the interaction between polymer functional groups and cross-linkers such as aldehyde groups. (e.g., glutaraldehyde, adipic acid dihydrazide, etc.). Enzyme catalyzed processes, polymer-polymer conjugation, and photosensitive agents accomplished the production of cross-linking [30]. Chemical cross-linking is carried out by the following methods.

Chemical cross-linkers

Hydrogels with both natural and synthetic polymers were created using cross-linking agents such glutaraldehyde and epichlorohydrin. This method involves adding additional molecules to the polymeric chains to create cross-linked chains. The use of 2-acrylamido-2-methylpropanesulfonic



acid to cross-link acrylic acid and β -carrageenan to create biodegradable hydrogels has also been documented in literature. Additionally, the business uses carrageenan hydrogels to immobilize enzymes. Heating and freezing methods can be utilized to create hydrogels from cellulose using epichlorohydrin as a cross-linker [31].

Grafting

Grafting is the process of polymerizing a monomer on a polymer backbone that has already been produced. Grafting is classified into two categories: radiation grafting and chemical grafting. Chemical grafting, such as the use of N-vinyl-2-pyrrolidone to graft starch with acrylic acid, entails the activation of polymer chains by chemical reagents. Electron beam radiation was used by Said et al. to create CMC hydrogel [31].

Radiation Cross-linking

Cross-linking the polymers is another method for creating these systems. Using this technique, the polymer is exposed to a high energy source after producing free radicals. Because it doesn't require any chemical additions, it is a beneficial procedure. Additionally, it is an economical method of modifying biopolymers for use in biomedical applications [31].

Polymers used in hydrogel preparation

Natural polymers

Natural polymers usually have the advantage of biocompatibility and biodegradability, but because of the distress of purification, their most common limitation is batch to batch variation which causes differences in final formulation (32). Some of the most important natural polymers which are used in hydrogel preparation are listed in the following section

Chitosan

Chitosan is a well-known natural polymer and has many advantages in drug delivery systems such as mucoadhesive and bioadhesive properties, absorption-enhancing properties and controlled drug release (33). Chitosan is a water soluble (in light acidic pH) and cationic (positive charge) polymer. The most important advantages of this polymer are its biocompatibility and low toxicity potential. Chitosan also has some limitations such as small specific surface area and void fraction that should be overcome (33).

Hyaluronic acid

Hyaluronic acid is a mucopolysaccharide which is naturally existing in cartilage and connective tissue. Hyaluronic acid has a poly-anionic nature and so it could be cross-linked to cationic polymers. Hyaluronic acid could be degraded by hyaluronidase (34). Hyaluronic acid is a suitable choice in local delivery because of its biocompatible and biodegradable nature and visco-elastic and unique rheological properties. It could induce cell mobility and cell proliferation and also cause wound healing. Hyaluronic acid is used in hydro-gel preparation and has the advantage of delayed drug release and prolonged duration of action (35)

Carrageenan

Carrageenan is a natural polysaccharide and because of its gelation capability has the advantage of controlled drug release. Carrageenan is an anionic, biocompatible and low toxic polymer. It seems that κ -carrageenan is the most suitable form of carrageenan for drug delivery and tissue

engineering purposes (36)

Alginate acid



Alginic acid is an anionic polysaccharide which is highly water soluble and has the gelation capability. It has the advantage of biocompatibility and low toxicity. Alginic acid could instantly form a gel in combination with calcium ions (37) and could be used as drug carrier.

Collagen

Collagen is a biocompatible and biodegradable polymer which is widely used in pharmaceutical industries (38). It is a very suitable choice for implant preparation. Collagen could control drug release from hydrogels and also could induce cell growth (39). Collagen would be widely used as a scaffold for tissue engineering purposes.

Synthetic polymers

Poly ethylene glycol (PEG)

Polyethylene glycol is a synthetic polymer with biological functions. The benefits of PEG include its biocompatibility and flexible physical characteristics. PEG can undergo regulated gelation in response to a photo-initiator or when combined with cross-linkers (40). PEG hydrogels have hydrolyzable (degradable) components in their architecture

Poly lactic acid (PLA)

Poly lactic acid is a synthetic polymer that is hydrophobic, biodegradable, and biocompatible. The inability of PLA to disperse hydrophilic compounds in its polymeric structures and unconventional controlled the pattern of medication release. By cross-linking PLA to hydrophilic PEG polymers, which are highly hydrated and able to maintain and extend drug release, these issues could be resolved. Hydrophilic and biodegradable polymers are two benefits of PEG/PLA copolymerization. When PEG is grafted onto PLA, the polymer becomes

hydrophilic, and the PLA portion of the molecule becomes biodegradable ().

Poly lactic co-glycolic acid (PLGA)

PLGA is a biocompatible, biodegradable polymer with suitable mechanical characteristics. PLGA could be used in drug delivery systems because of the advantage of sustained and prolonged drug release(39). Although PLGA is a biodegradable polymer, but micro environmental acidity following polymer degradation, could induce irritation at the site of formulation application and could also damage peptide/protein drugs which are loaded to hydrogels

Poly vinyl alcohol (PVA)

PVA is a synthetic polymer that is hydrophilic, biodegradable, and biocompatible. In tissue engineering, PVA would be extensively utilized to restore damaged organs or tissues. A appropriate hydrogel that could mimic the characteristics of natural tissues could be created by using the ideal PVA to water ratio (40).

Poly caprolactone (PCL)

PCL is a synthetic polyester that is hydrophobic, biocompatible, and biodegradable. It is widely employed in tissue engineering and drug delivery (28). Because PCL may easily and potentially copolymerize with other polymers, it has the advantage of being adaptable to physical, mechanical, and chemical changes. PCL can be used with both synthetic and natural polymers. Lipophilic medications may be uniformly distributed in a polymeric matrix due to their hydrophobic character, whereas hydrophilic drugs primarily migrate to the surface of PCL polymers (29). Semisynthetic polymers (Cellulose derivatives)

Carboxymethyl cellulose (CMC)



CMC is a biodegradable and biocompatible polymer that finds extensive application in biomedicine. CMC, a water-soluble cellulose derivative (ether of cellulose), is currently reasonably priced and very pure (30). Hydrogels might be readily created by cross-linking CMC.

Hydroxyethyl cellulose (HEC)

HEC is a cellulose derivative that is biocompatible and soluble in water. Grafting polymerization with vinyl groups allows for the modification and improvement of HEC, which contains a large number of OH groups in its structure (31). When cross-linked with CMC, HEC produces new hydrogels with superior swelling characteristics. By modifying the appropriate HEC/CMC ratio and quantity of cross-linkers utilized in hydrogel synthesis, these hydrogels' capacity to absorb water and expand could be maximized (32).

Methyl cellulose (MC)

MC is a semi-flexible linear derivative of cellulose that has methoxy groups in place of certain hydroxyl groups. The water solubility of this polymer may be determined by the ratio of hydroxyl to methoxy groups. The nucleation and growth mechanism may cause MC to gel at the sol-gel transition temperature (T_g) (41).

Hydroxypropyl cellulose (HPC)

HPC is a cellulose derivative that is very soluble in water. HPC hydrogels are typically employed in aqueous solution dye removal. The most significant drawback of HPC hydrogels is their poor water absorption capacity, which might be overcome by adding nanofillers with a high specific surface area to these hydrogels (42).

Hydroxypropyl methyl cellulose (HPMC)

A common component of controlled drug delivery systems is HPMC, a water-soluble derivative of cellulose. The most significant feature of HPMC is its high swelling capacity, which may regulate the release of the active pharmaceutical ingredient. The molecular weight, methoxy substitute content, and hydroxypropoxy substitute content of HPMC polymers can be used to characterize their physicochemical qualities.

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