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REVIEW

An Overview on Starch-Based Sustainable Hydrogels: Potential Applications and Aspects

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Abstract

Hydrogels are a kind of three dimensional polymeric network system which has a significant amount of water imbibing capacity despite being soluble in it. Because of the potential applications of hydrogels in different fields such as biomedical, pharmaceutical, personal care products, biosensors, and cosmetics, it has become a very popular area of research in recent decades. Hydrogels, prepared from synthetic polymers and petrochemicals are not ecofriendly. For preparing biodegradable hydrogels, most available plant polysaccharides like starch are utilized. In its structure, starch has a large number of hydroxyl groups that aid in hydrogel networking. For their easy availability and applications, starch-based hydrogels (SHs) have gained huge attention. Moreover, SHs are non-toxic, biocompatible, and cheap. For these reasons, SHs can be an alternative to synthetic hydrogels. The main focus of this review is to provide a comprehensive summary of the structure and characteristics of starch, preparation, and characterization of SHs. This review also addresses several potential multidimensional applications of SHs and shows some future aspects in accordance.

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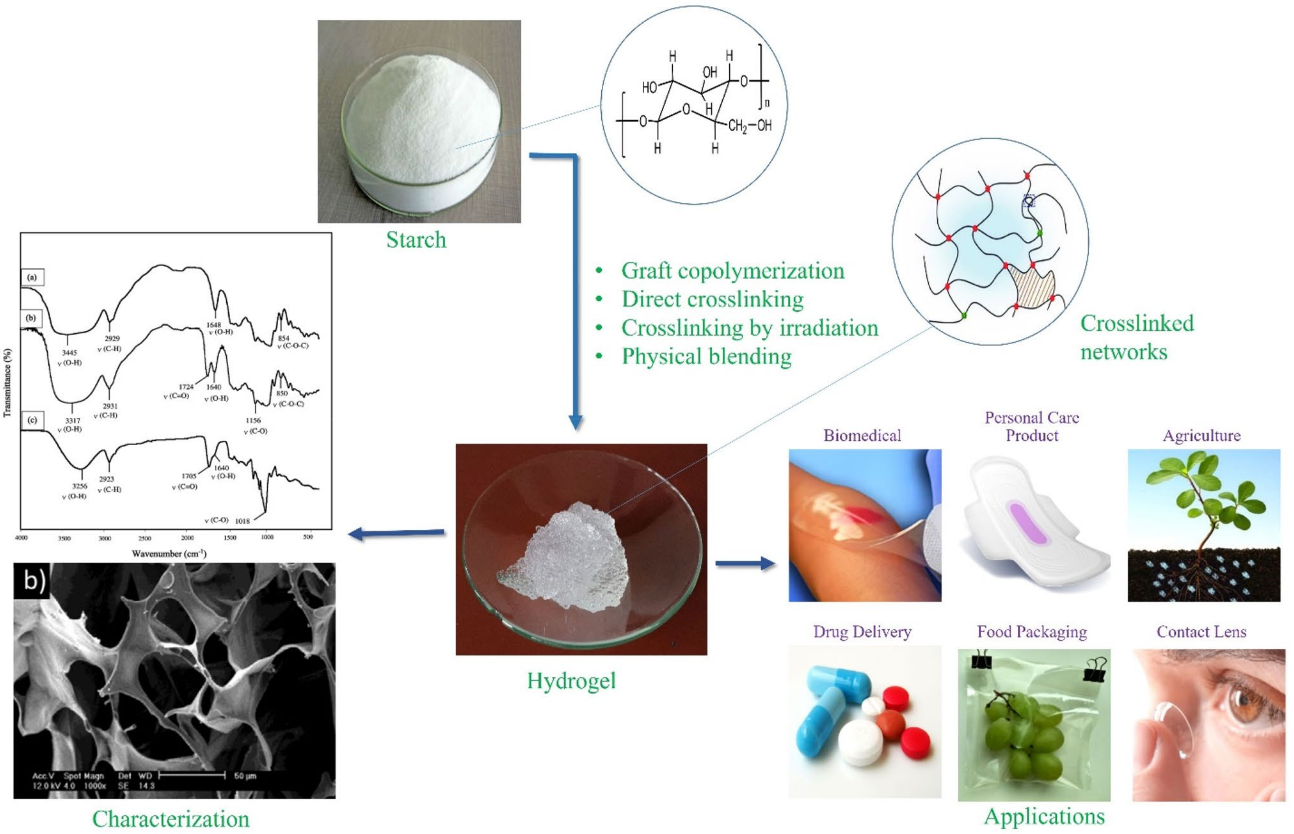
Graphic abstract



Keywords Hydrogel · Polymeric-network · Starch · Biodegradable-hydrogel · Preparation · Application

Abbreviations

SHs	Starch based hydrogels
PLA	Poly lactic acid
PGA	Poly glycolic acid
AA	Acrylic acid
KC	Kappa-carrageenan
PVA	Polyvinyl alcohol
AM	Acrylamide
MBA	*N*,*N′*-Methylenebisacrylamide
DMDAAC	Dimethyl diallyl ammonium chloride
EG	Ethylene glycol
MAA	Methacrylic acid
PCL	Polycaprolactone
AMPS	2-Acrylamido-2-methylpropane-1-sulphonic acid
DMAEMA	Dimethylaminoethyl methacrylate
AFM	Atomic force microscopy
FTIR	Fourier transform infrared spectroscopy
XRD	X-ray diffraction
TGA	Thermogravimetric analysis
DTA	Differential thermal analysis
DTG	Differential thermogravimetry
DSC	Differential scanning calorimetry
GMA	Glycidyl methacrylate
LDPE	Low-density polyethylene
APS	Ammonium persulphate
TEMED	*N*,*N*,*N*,*N*-Tetramethylethylene diamine
LRD	Laponite RD
HEMA	2-Hydroxyethyl methacrylate
HES	Hydroxyethyl starch
CB	Coomassie brilliant
MB	Methylene blue
MV	Methyl violet
MO	Methyl orange
AMPS	2-Acrylamido-2-methylpropanesulfoacid

Introduction

Hydrogels are called hydrophilic polymeric networks with huge water absorption capability (up to thousand times their dry weight) [1–3]. Hydrogels can be prepared from both synthetic and natural materials. The key feature of hydrogels is their large amount of solvent (e.g., water, biological fluids) retaining ability under different conditions. Like living tissue, hydrogels are soft and flexible which makes them a very suitable material for various potential applications [4]. In recent decades, hydrogels have been used in various important areas such as in tissue engineering [5–7], in delivery of drugs [8, 9], wound treatment [10, 11], for the purification of water [12–14], in biomedical sectors [15, 16], in production of personal care items [17–19] and agriculture [20, 21]. Another significant character of hydrogels is their responding ability to different environmental conditions such as the change of temperature, pH, variation in magnetic fields, and some electric stimuli. Due to having the ability to quickly respond to the environment, they are called smart polymers (e.g., stimuli-responsive hydrogels). Hydrogels are also termed as novel biomaterials of this era because of some unique properties like (I) ability of biodegradation, (II) stability in chemicals and bio-fluids, (III) stable shape, (IV) biocompatible and ecofriendly, (V) large absorption capability of metabolites and nutrients which are soluble in water [22]. The use of hydrogels as bio-material is also well-known because of their permeability of small molecules, soft consistency, low interfacial tension like the properties of living tissues [23]. The use of hydrogels is growing steadily because of their unique properties [24–26].

In the polymeric structure of hydrogel networks, covalent bonds are involved due to the reaction between the co-monomers and hydrogen bonds and the interaction of van der Waals force between the chains and physical crosslinking can be associated [17]. The cross-linked structure of hydrogels prohibits them from disintegration at the time of water-retaining. Generally, crosslinking agents are used along with the polymers for the initiation of chemical crosslinking in hydrogel networks. In hydrogels, there is the presence of different hydrophilic groups like –COOH, –SO_3H, –OH, and –NH_2 which increase the hydrophilic property of the networks [27]. The water retention ability of hydrogels depends on two main factors—interactions of polymeric chains with water and temperature. A number of steps are involved in the water retention process of hydrogels—(I) hydrophilic groups present in hydrogels are hydrated, (II) interaction of water with the hydrophobic groups in hydrogels, (III) dilution of the hydrogels are stopped by crosslinks present in hydrogel networks, so the additional water can be absorbed [28].

Synthetic polymers are heavily used to manufacture hydrogels that are currently available on the market because they show some outstanding mechanical, physical, and chemical properties. They also have some drawbacks too. They are not ecofriendly, production is not cost-effective, nonrenewable and bio-degradation is difficult [8]. For these reasons hydrogels produced from naturally produced polysaccharides like starch and cellulose are getting more attention from researchers [29–34]. In starch, hydrophilic groups (e.g., –OH) are distributed in the molecular chains [35, 36]. These groups increase the hydrophilic character and biodegradability of the resulting hydrogels [37]. An abundant amount of starch is naturally produced in plants by the photosynthesis process by using CO_2 and water [38–40]. Plants are an abundant source of starch polysaccharides in which chloroplasts of the green leaves and amyloplasts of the tubers and seeds contain a large amount of starch as granules [41–43]. Potatoes, wheat, corn, and rice are the main production source of commercial starch [44]. Mainly by three steps of hydration–plasticization starch, turns into gelatin. Hydrophilic starch granules adsorb water and swell in the first step. The granule structure destructed when the starch is dissolved in water and form gelatin in the second step. The final step consists of starch hydrogel network formation by cooling and aging and the structure of polysaccharide get reorganized. This step is also called the step of retrogradation. Hydrogel formation primarily depends on the amount of amylose present in starch and the temperature of gelatin formation [45, 46]. For these characteristics and easy availability, hydrogel formation by starch has become popular from the 1970s [47, 48]. In addition, starch-based hydrogel products have gained attention for reducing the impact on the environment and more potential applicability [49–51].

To prepare SHs, various chemical methods can be utilized such as etherification and grafting method of starch. In etherified starches, various ether groups like carboxymethyl starch substitute the –OH groups from the starch structure. On the contrary, hydrogels are produced by grafting different vinyl monomers on starch in the grafted starch method. The use of SHs in different fields has both merits and demerits. Some merits of the use of SHs are—(I) ample in nature, (II) available all over the world, (III) cheap and attractive, (IV) preparation methods are comparatively easier, (V) eco-friendly, (VI) solvent absorption capacity is higher, (VII) usable in different process [52]. Some drawbacks of SHs are—(I) surface area is low, (II) for enhancing the sorption capacity chemical derivatization is needed, (III) low durability, etc. [53, 54]. Although being completely biodegradable, the use of SHs is quite a few in different fields like drug delivery systems [55]. But other polymers like poly (lactic acid) (PLA), poly (glycolic acid) (PGA) are being used in this field [56–59]. Hence more study should be continued on SHs.

Henceforth, this review aims to provide a brief overview of the characteristics and structure of starch and SHs with an emphasis on their recent developments. In addition, properties and different potential applications of SHs have been discussed. Specifically, this review provides a summary of several synthesis pathways of SHs, and finally, some of the gaps future research directions have also been suggested.

Characteristics and Structure of Starch

Starches are polysaccharides. Monosaccharides (glucose units) are bonded together by α-D-(1–4) and α-D-(1–6) linkages in starch structure [60–62]. In the starch structure, there are primarily two types of polysaccharides, which are amylose and amylopectin representing about 98–99% dry weight of starch [63, 64]. Amylose (molecular weight approximately 1×10^5–1×10^6) and amylopectin (molecular weight approximately 1×10^7–1×10^9) have different structures [44, 65–68]. Amylose has a linear structure whereas amylopectin is composed of branched linkage [61, 63]. The structure of starch is shown in Fig. 1. The amount of amylose and amylopectin present in starch mainly depends on two factors which are—(I) source of the starch (potato, wheat, corn) and (II) polysaccharide crystallinity [69]. The proportion of the amorphous and crystalline mass in starch is about 70% and 30% respectively [70].

The main component of the amorphous region in starch is amylose. Although a very small amount of amylopectin is available in the amorphous part, the crystalline region mainly consists of amylopectin [71]. In water, amylose solubility is high and a helical structure is formed [72]. Starches occur as a form of granular structure in the plant's body and amylopectin can form a helical structure and crystallized. The hydrogen bonds occurred by the –OH groups present in the starch surface are the main reason for exhibiting hydrophilic property and strong inter-molecular attraction by starch granules [30, 64]. Starches contain a huge amount of –OH groups in their structure. In each glucose unit of starch, there are two types of –OH groups present. One is secondary –OH group (at carbon 2 and 3 positions) and another is primary –OH group (carbon 6 position when it is not linked). The reactivity of starch is highly dependent on these –OH groups. Starch can be oxidized or reduced and form hydrogen bonds, esters, and ethers [73].

As starch is hydrophilic in nature, water molecules can change the internal interaction and morphological

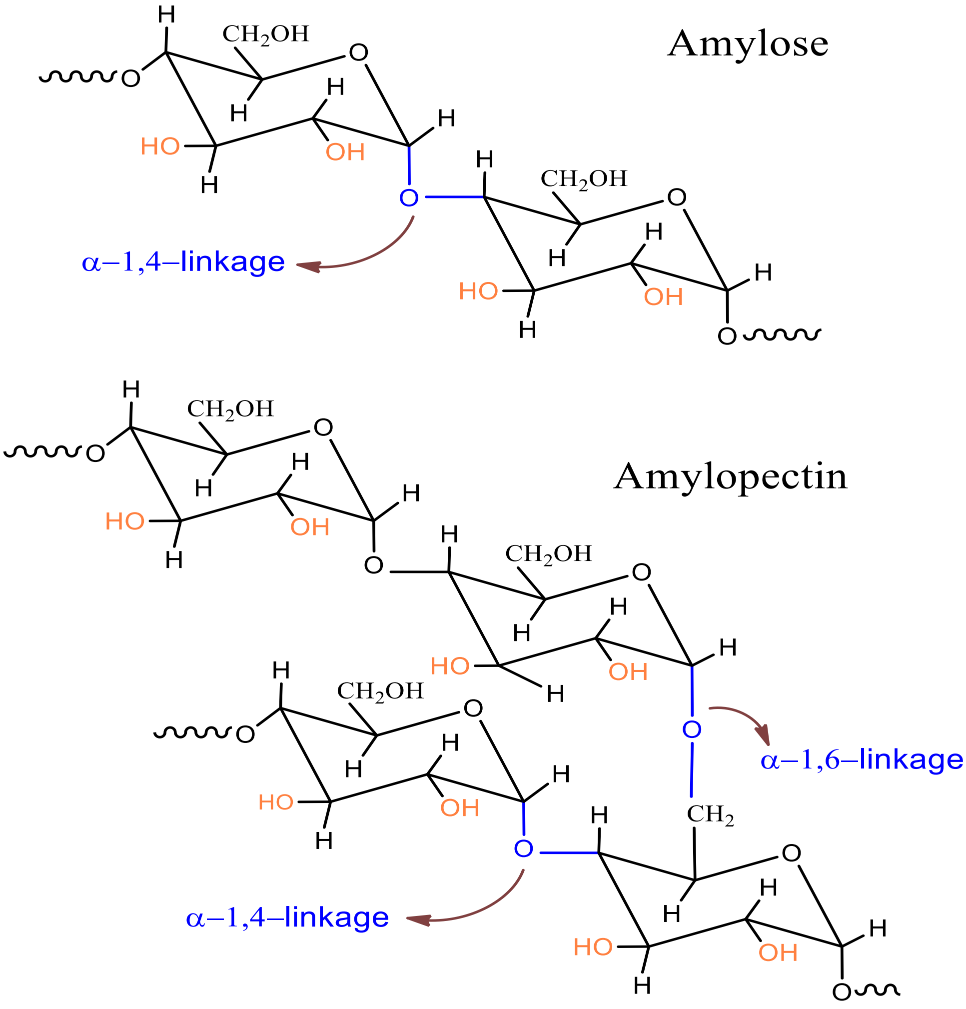


Fig. 1 Structure of amylose and amylopectin and in starch [30]

arrangements of starch. For this reason various properties of starch such as physical and mechanical properties and glass transition temperature (Tg) dependent on water content [74]. One of the important properties of starch is complete biodegradability in different environments. Using microorganism or enzymatic reaction, glucose can be found from starch by hydrolysis process. Further, it can be metabolized into CO_2 and water [75]. Starch is biocompatible, non-toxic, cheap, shows good mechanical properties, and easily available [76, 77]. Although starches are found abundantly in nature, it has some drawbacks such as dimensional stability is low in native starch, comparatively poor gel content, mechanical properties, and processability are also poor for the end products [78–80]. For this reason, using native starch is not feasible. Thus, by using chemical or/and physical interaction, starches need to be modified [81, 82]. In this regard, SHs can be good starch based product for various potential applications.

Classifications of Hydrogels

There are various types of hydrogel depending on different properties, such as (I) swelling property, (II) physical characteristics, (III) preparation technique, (IV) source, (V) biodegradation property, and (VI) crosslinking nature [4, 8, 83–85]. Different types of hydrogels are shown in Fig. 2. Physical crosslinking techniques are utilized in the physical gels (reversible for conformational changes) achieved by chain aggregation, the complexion of polymer chain, and hydrogen bonding. In chemical hydrogels process like covalent crosslinking is used. But chemical hydrogels are irreversible in nature for their configurational changes. On the other hand, both the chemical and physical hydrogels are combined in dual-network hydrogels. Dual-network hydrogels are more advantageous than the physical and chemical hydrogels because they

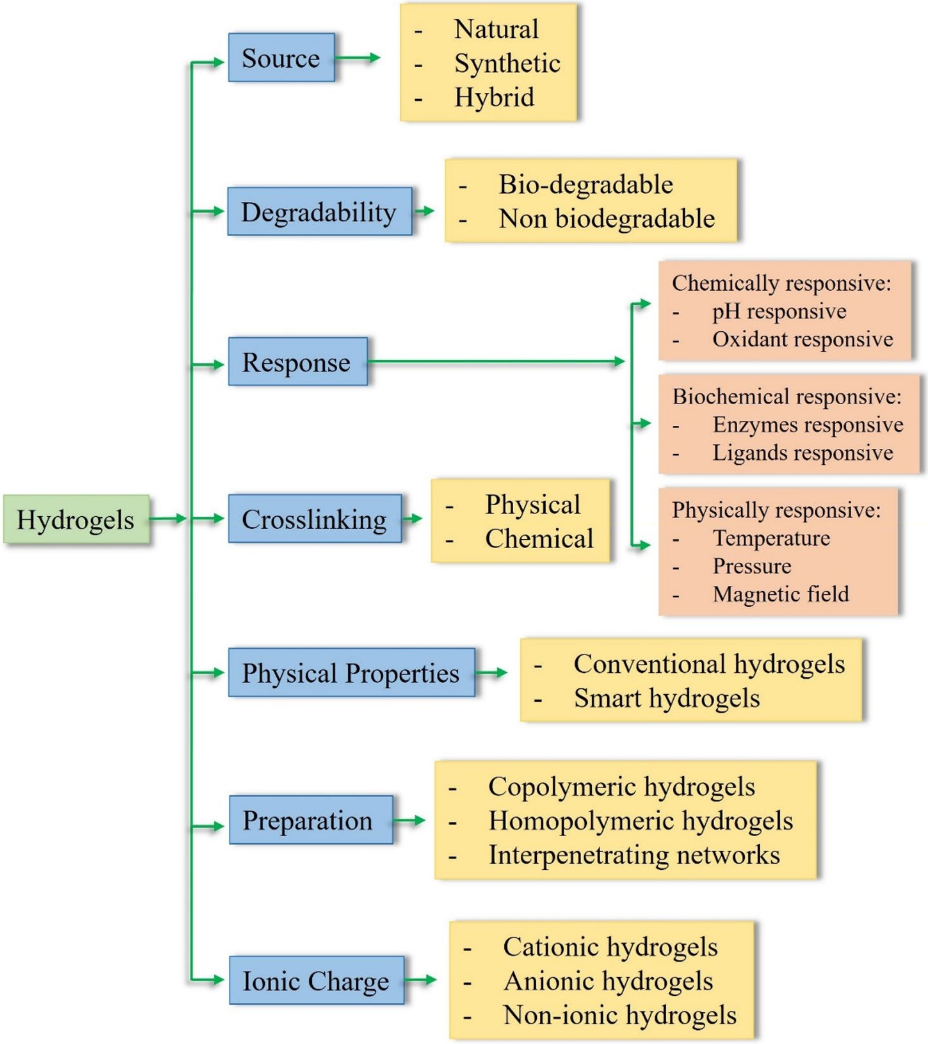


Fig. 2 Classifications of hydrogels according to different properties [4]

can retain a comparatively large amount of liquid and their sensitivity in different pH range is higher than the chemical or physical hydrogels alone. Recently, Cong et al. [86] and Yalpani [87] introduced a dual-network hydrogel with comparatively higher mechanical characteristics by combining graphene–polymer composites. Hydrogels can be either natural or synthetic depending on their origins [88]. Depending on the monomers used in the preparation of hydrogels, they can be homopolymeric or copolymeric hydrogels. In the hemolytic hydrogels, a single type of monomer is used [89]. On the other hand, two or more types of monomers are utilized for the preparation of copolymeric hydrogels [90]. They are arranged in different ways in the polymeric chain such as random, block, or alternating configurations. Another important class of hydrogel is interpenetrating polymeric hydrogel (IPN). Both the cross-linked and non-cross-linked polymers are present in the semi IPN hydrogels [91, 92].

Responsive hydrogels are also an important type of hydrogel because they are sensitive to different environmental conditions like temperature, pressure, pH, etc. [93]. Stimuli-responsive hydrogels are smart hydrogels that are sensitive to environmental stimuli [94–97]. Molecular interactions change due to the change in chemical (pH, ionic factors, chemical agents) and physical (light, pressure, temperature, electric fields, magnetic fields, mechanical stress) stimuli change [4]. In biomedical and pharmaceutical and biomedical sectors stimuli-responsive hydrogels are very popular [98].

With the alteration of surrounding temperature, swelling and shrinkage of the hydrogels change in thermo-responsive hydrogels [99]. Dai et al. [100] prepared thermo-responsive hydrogel by the copolymerization of polyacrylamide and poly(acrylic acid) for controlled drug delivery system. They can be both positive and negative temperature-responsive hydrogels [101].

pH responsive hydrogels are another important type of hydrogels that can response to the change of the pH of the environment and for this property, this type of hydrogels are widely used in drug delivery systems [102–106]. The degree of ionization in pH sensitive hydrogels change instantly for the change in medium pH [107]. pH responsive hydrogels can be anionic (contains carboxylic or sulfonic acid pendent groups) and cationic type (amine pendant groups) [108–111]. Han et al. (2020) prepared dual pH-responsive hydrogel actuator and successfully used in lipophilic drug delivery system [112].

Hydrogel Preparation Techniques

Hydrogels are hydrophilic polymer networks. According to the different application aspects, hydrogels are prepared by different methods using both hydrophilic and hydrophobic monomers. Either synthetic polymers or natural polymers are used in hydrogel preparation. Although hydrogels prepared from synthetic polymers are mechanically much stronger than the natural ones, they are less biodegradable [113, 114]. The main task during the hydrogel production is creating crosslinking between the monomers using different types of crosslinkers. Copolymerization/cross-linking free-radical polymerizations are commonly used methods for hydrogel production. Mainly hydrogels are prepared from the monomers considering the bond forming principles below [92, 115]:

a. Physical interaction between the monomers,
b. Linking polymer chains via chemical reaction,
c. Using ionizing radiation.

Monomer, initiator, and cross-linker are three major parts of hydrogel production. Diluents are used for controlling the hydrogel properties. Different polymerization techniques for hydrogel preparation are described below.

Bulk Polymerization

Bulk polymerization is one of the simplest methods of hydrogel preparation. This method mainly consists of different monomers, initiators, and a suitable amount of crosslinking agents. Vinyl monomers are highly used in this technique. A wide variety of monomers are used for altering the physical characteristics of final hydrogel products [92]. Chemical catalysts and radiation is used for initiating the reaction. Initiators are selected according to the monomers and solvents. The degree of polymerization and polymerization rate are high in this method. Rods, particles, films and membranes, as well as emulsions, are all possible forms for the bulk polymerized hydrogels [115].

Solution Polymerization

In this technique, copolymerization occurs between the monomers with the help of crosslinkers. UV-irradiation or redox initiator initiates the reactions. The solvents used in the polymerization work as a heating bath for the reaction which is the main advantage of this method compared to the bulk polymerization. This process is much preferred because of cost-effectiveness, easy preparation and heat control. At the beginning of this method concentration of monomers are kept high and low concentration of initiators and crosslinkers are utilized [116]. For removing the impurities and unreacted monomers the prepared hydrogels are washed with distilled water. Different types of solvents are used in this polymerization method such as water, ethanol, and benzyl alcohol, etc. [115]. Hydroxyethylcellulose (HEC) and carboxymethylcellulose sodium salt hydrogels were prepared by

Sannino et al. [117–120] using this polymerization method. Keeping water as a common solvent they prepared hydrogels altering different crosslinkers and catalysts.

Free Radical Polymerization

Free radical polymerization is a common chain-growth polymerization technique [121]. The main advantages of this technique are—it is a fast technique, can be carried out as bulk, solution (also in water), emulsion, and suspension polymerization. Acrylates, vinyl lactams, and amides monomers are highly used in this process. Free radical polymerization technique include different steps such as propagation, chain transfer, initiation, and termination [115]. This process starts with the decay of initiators and free radicals generate which attack the monomers to make them active [122]. Polymerization stops during the termination stage with the deactivation of the active center.

Suspension Polymerization

Suspension polymerization is a technique in which monomers and initiators are dispersed in the organic phase and create a homogeneous mixture. The reacting molecules are dispersed as minute droplets in the continuous phase. The heat is transferred from the droplets to the water, which has a large heat capacity and a low viscosity. Preparation of hydrogels by this method is dependent on different conditions such as monomer viscosity, speed of the agitation, and rotor design [123–125]. One advantage of this method is that additional grinding is not necessary for the prepared hydrogel product because hydrogels are obtained in the powder or beads form. A low hydrophilic–lipophilic-balance (HLB) suspending agent is required to be added in the dispersion because of the thermal instability of the dispersion [92].

Physical Cross-linking

Physical cross-linking is one of the easiest ways of hydrogel formation by crosslinking the polymers. This process is gaining popularity because of its ease of production and an additional cross-linking agent is not required. Physical hydrogels are also called self-assembled hydrogels. Different types of interactions occurred between the macromolecules such as hydrophobic, polyelectrolyte complexation, electrostatic, and H-bonding [126]. These physical interactions prevent the dilution of the product hydrogels. These interactions are reversible and can be altered by changing physical conditions [127]. As toxic cross-linkers are not used in this method, it is much usable in the biomedical applications like drug delivery systems. In preparation of hydrogels by physical crosslinking, different methods like heating/cooling of polymer solution, complex coacervation, ionic interaction, hydrogen bonding are utilized [115].

Chemical Cross-linking

For the production of mechanically stable hydrogels, the chemical cross-linking method is highly used. For grafting the monomers and cross-link the polymers, different types of cross-linkers are used. Cross-linking is produced by the reaction between the functional groups (e.g., –OH, –COOH, and $–NH_2$) of the monomers and cross-linking agents (e.g., -glutaraldehyde, adipic acid dihydrazide) [128]. Mainly covalent bonds are developed between the polymer chains [129]. Sometimes toxic cross-linking agents are used which need to be extracted before using the gel for different purposes [126]. For instance, there are huge number of hydroxyl groups present in starch and PVA structures. PVA–starch based hydrogels can be prepared using glutaraldehyde as cross-linking agent where glutaraldehyde replaces the –OH groups and intermolecular bridges are formed [2].

Hydrophilic Nature of Hydrogel

Depending on the techniques of absorbing water, hygroscopic materials are mainly two types—(I) Chemical absorbers and (II) Physical absorbers. Chemical reaction is occurred when chemical absorbers (e.g. metal hydrides) absorb water and the properties of the absorbers are completely altered. On the other hand, different mechanisms work behind the absorption of water in physical absorber which are—(I) reversible changes in crystal structure, (II) physical water entanglement in the porous framework, (III) combining with second mechanism hydration of functional groups and (IV) thermodynamic expansion and dissolution of polymeric chains are prohibited by crosslinking with combining second and third mechanism [130]. Two types of water can be present in hydrogels which are free water and bound water. Most of the polar and hydrophilic groups present in the hydrogel are hydrated during the absorption of water molecules by dry hydrogel which results in the "primary bound water". Further, "secondary bound water or hydrophobically-bound water" results when the swelling of the polymeric network starts and hydrophobic groups are exposed interacting with the water molecule. The combination of the primary and secondary bound water is termed as "total bound water" [17]. The hydrogel networks start to gain more water molecules when the bound water is interacted with the hydrophilic and hydrophobic parts of the networks because of the osmotic driving forces for complete dilution. Physical and covalent cross-linkages present in the hydrogel network restrict the additional swelling which results in an elastic polymeric network. For this reason, infinite dilution cannot be occurred in hydrogel and reaches an equilibrium swelling. Various groups

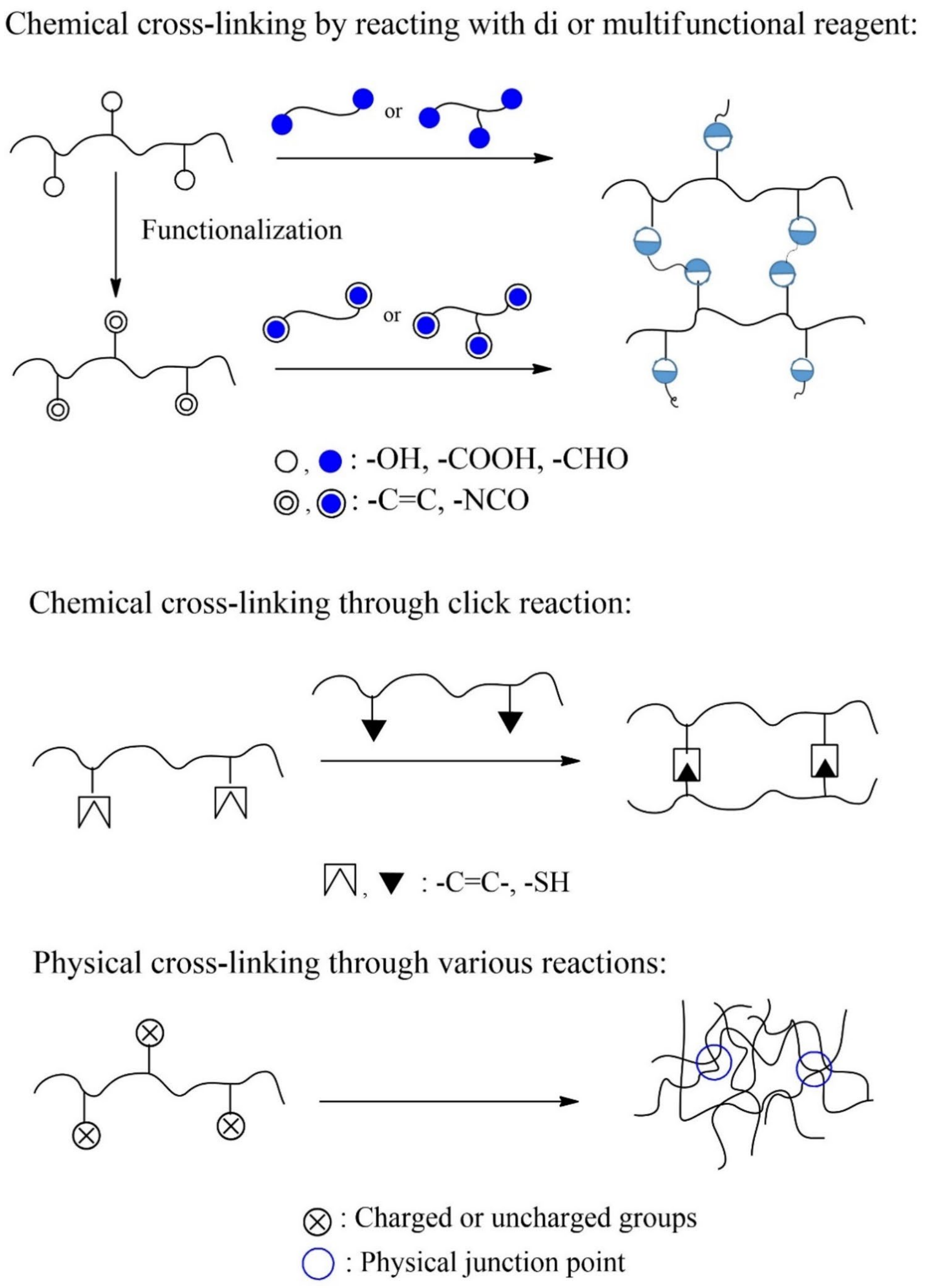


Fig. 3 Formation of starch-based hydrogels. Reused with permission from [143]. Copyright 2012, John Wiley and Sons

such as polar groups, ionic groups, and hydrophobic groups absorb the additional water and are saturated with bound water which is termed as "free water". Big pores and voids along with the gaps between the polymeric chains of hydrogels can be filled by free water [3].

Different Crosslinking Strategies of Starch-Based Hydrogels

Hydrogels possess three dimensional networks in their structure. The formation of crosslinking between the chains of macromolecules is the main aim during the synthesis of hydrogel which finally develops three dimensional networks [131]. At the primary stage of the network structure development, branched polymers are formed first. The size of these branched polymers gradually develops and finally forms the structure of gel networks. This gel network formation process is called gelation or sol–gel transition. Different processes can be utilized for the development of these crosslinking networks. Properties of hydrogels including swelling and several physical characteristics mainly depend on the degree of crosslinking [132, 133]. This crosslinking either can be physical [134] or chemical [131, 135] (Fig. 3). Researchers have utilized different synthesis routes for the development of polysaccharide-based hydrogels (e.g., cellulose, starch, chitin) [22, 136]. The stability of the hydrogels prepared by physical blending is not very good although they are insoluble in aqueous media. These types of hydrogels are reversible in nature [137, 138]. Oppositely, chemically crosslinked hydrogels are more stable forming covalent crosslinking [139].

Chemical cross-linking in hydrogels can be achieved in different ways such as—(I) different reagents (bi or multifunctional) can be reacted with polymers, (II) polymerize end functional macromonomers, and (III) click reactions between different polymeric groups [140–142].

By functionalizing, reactive groups can be added with polymers or they can exist in polymer precursors. Different crosslinking agents (glutaraldehyde, formaldehyde, epoxy compounds, dialdehyde) are used which are low or high molecular weight [144–147]. In physical crosslinking, hydrogen bonding or interaction between the opposite charge macromolecules are incorporated [148]. Although crosslinking structure and composition are more perfect in chemical crosslinking, sometimes crosslinking agents used in this process are toxic. But in physical crosslinking, comparatively mild conditions are utilized. For this reason, they could be utilized in drugs or other biomaterials for encapsulation [3]. In situ techniques are utilized in quick crosslinking (both physical and chemical crosslinking). Different parameters govern this process like-macromolecular structure (amount of crystalline or amorphous region, presence of functional groups) and environmental parameters (temperature, pH) [143].

Physical Blend

Although chemical cross-linking is more advantageous than other mechanisms due to its wide versatility, it has some drawbacks too. The reagents used for chemical crosslinking in this process are usually toxic and high temperature is needed for the reaction which is not very suitable for the bio-active materials [149]. In this regard, the physical cross-linking process has advantages for using in life science (e.g., tissue engineering) and bio-medical sectors (e.g., drug delivery) [143].

In physically blended hydrogel, different forces (hydrophobic interaction, hydrogen bonding, and ionic interaction) act for the cross-linking of the networks, and cross-linking agents are not necessary. For SHs production, different techniques have been used such as heat-induced aggregation, freeze-drying, and complex coacervation [22]. Hydrogel is formed along with the formation of helix and junction jones when the hot solution of polysaccharides is cooled. For the development of carrageen hydrogels from the hot solution, the cooling process was used [150]. Produced hydrogels become more stable if K^+, Na^+ like salts are present. A new technology for the formation of physically blended hydrogel is stereocomplex in which lactic acid and methyl methacrylate oligomers are used. Coupling is formed between polymer chains and oligomers. De Jong et al. 152] synthesized self-assembled hydrogels by stereocomplex by grafting enantiomeric lactic oligomers in dextran.

Gelation of native starch is easy [151]. By thermal treatment, hydration starts in starch resulting in gelation [152, 153]. For starch aerogel formation, gelating behavior is combined with emulsion [153]. By retrogradation, heating the starch solution and finally drying starch aerogel is formed [154]. Different characteristics (e.g., swelling and release properties) of starch hydrogels are changeable by modifying the non-ionic components [155, 156]. By utilizing the freezing/thawing process repeatedly, physical PVA based ecofriendly hydrogel can be easily developed [157, 158]. Using the graft copolymerization process PVA–starch based hydrogels are synthesized by radical copolymerization and alcoholysis [159, 160]. Starch-based networks are highly improved blending with PVA [161]. By utilizing the freezing and thawing method starch-*g*-PVA hydrogel is prepared [162, 163]. A nano-precipitation method is utilized for preparing SHs which are pH-sensitive [164].

Blend with Synthetic Degradable Polymers

Previously, starch carbon–carbon backbone was utilized for making vinyl polymers biodegradable [165]. Due to the use of starch, the total surface area increases which enhance the attack of microorganisms. By this process, the biodegradability increased slightly but was not totally eco-friendly [30]. Biodegradable polymers can be utilized to prepare completely biodegradable hydrogel blending with starch. Aliphatic polyesters, polyvinyl alcohol (PVA), and biopolymers are highly utilized for blending with starch [166–168]. Enhancing biodegradability along with maintaining cost-effectiveness and other characteristics are the main aim of this blending [169, 170]. In respect of biodegradability, PLA (polyacetic acid) is a good polyester and has wide use in biomedical sectors. Some of the excellent features of PLA are high biocompatibility, high strength, and modulus. PLA has some drawbacks too—comparatively costly, shows physical aging when applied (brittle under load) [3, 171]. For increasing their compatibility poly (hydroxyester ether) and poly(vinyl alcohol) are used [172, 173]. Researches have been attempted to produce hydrogel by blending starch polysaccharides and PLA [171, 174]. Starch granules are hydrophilic in nature and PLA is hydrophobic. For this reason, the interfacial interaction between them is low resulting in poor mechanical properties of the blend [175]. To overcome this drawback, different plasticizers have been utilized such as glycerol, sorbitol, and formamide, etc. [176] Suitable compatibilizers can also be used for enhancing the compatibility between starch and PLA. Gelatinization is another technique that increases the interfacial affinity. This method helps in easy dissolve of the starch granules and dispersion is also good [177, 178]. Polycaprolactone (PCL) is an eco-friendly and biodegradable polymer that was also used for blending with starch. PCL/starch blends were formed by reinforcing with nano-clay and fiber etc. which showed improved biodegradability, hydrolytic stability [179, 180].

Polyvinyl alcohol (PVA) is a water-soluble synthetic polymer with good biodegradability. It is compatible with starch and shows good mechanical characteristics. For this reason, PVA is most widely used for blending with starch [36]. In different microbial conditions, both the starch and PVA are degradable which resulting in a completely biodegradable PVS/starch blend polymer [171, 181]. Isolated microorganisms from landfill and municipal sewage line were used for the biodegradability test which shows the complete biodegradable nature of the blend [182]. Processability and mechanical properties are also excellent because of the bonding between the –OH groups of PVA and the chain of starch [183, 184]. With rising PVA concentration, tensile strength also improved [182]. For increasing the compatibility of other starch-based blends such as starch/PLA, PVA is utilized. A continuous phase is developed between the starch and PVA during blending because both of them are polyols [173]. The interfacial interaction of starch and PLA is poor where starch works as a filler and is loaded in PLA matrix without PVA.

Blend with Biopolymers

Due to biodegradability and other characteristics of different polysaccharides like cellulose and chitosan, they are investigated for blending with starch. Another reason is the easy availability of starch and chitosan polysaccharides in nature, they are inexpensive, non-toxic, and renewable as well [185]. Starch has a huge amount of –OH groups and chitosan has –OH and –NH_3 groups in their structure. The main mechanism behind this blending is the formation of hydrogen bonds between the hydroxyl and amino groups. The blended hydrogels show excellent mechanical and film formation properties. Depending on the ratio of chitosan and starch, properties of hydrogels like mechanical properties and biodegradability varies [168]. Several authors have studied starch-based biodegradable films and chitosan blend with biopolymers [186–190]. Different plasticizers (e.g., glycerol) are also utilized for edible film production [191]. Strength rupture of hydrogel is enhanced with the increase of cellulose amount. On the other hand vapor permeability decrease with the increase of cellulose content in SHs. Derivatives of cellulose like carboxymethylcellulose form thermodynamically compatible hydrogel with starch and exhibit biodegradable property [192]. Nano-cellulose with plasticized starch is cast for SHs composite film production which shows improved mechanical properties. Large changes in mechanical properties like tensile strength are experienced improvement in nanocomposite hydrogel. Tensile strength increases from 11.9 MPa in the unreinforced hydrogel to 498.2 in the nanocomposite hydrogel [193].

Chemical Blend

Covalent bonds are present in the hydrogels which are produced by chemical crosslinking. The equilibrium swelling capacity of these hydrogels depends on the density of crosslinking and water polymer interaction. Different approaches can be utilized for chemical crosslinking such as—(I) polymerization using cross-linkers, (II) polymer–polymer cross-linking, (III) chain-growth polymerization, (IV) addition and condensation polymerization, and (V) gamma and electron beam polymerization etc. [22, 194, 195]. There are some drawbacks to starch–polymer blend hydrogel production. Most of the different polymers are immiscible with starch resulting in unstable physical characteristics of produced hydrogels. For this reason, the chemical crosslinking technique is widely used. A large number of –OH groups are present in the starch structure. Typically, these –OH groups react (substituted with other groups or chains) during the chemical functionalization of the starch molecules [196, 197]. The characteristics of native starch and modified starch are different. But modified starch is biodegradable as well as the parent starch. Chemical crosslinking can be achieved in different ways in SHs.

Graft Copolymerization Method

At the beginning of the graft copolymerization method, reaction between an initiator and polysaccharides (starch) starts. This reaction can proceed in two methods. In the first method, a complex (redox pair) is formed when the hydroxyl group present in the starch monomer reacts with the initiator (e.g., Ce^{4+}). Free radicals are formed on the polymer backbone due to the dissociation of the evolved complex. During the dissociation, C–C bonds experience homogeneous cleavage.

Next, the graft polymerization of vinyl monomers and cross-linker starts on the chain by the produced free radicals. Athawale and Lele [199] prepared SHs by the first method of graft copolymerization of acrylic acid on maize starch in which the initiator was Ce^{4+}. Dragan and Apopei [198] proposed graft polymerization of acrylamide on starch with the same initiator. The mechanism is represented in Fig. 4. By this method properties of the starch can be easily modified and produce SHs [200–202].

Another method consists of the formation of free radicals on the polymer backbone by the removal of hydrogen radicals from the –OH groups with $S_2O_8^{2-}$ initiator. This method is more temperature dependent than the first one because a thermal initiator has been used here. Pourjavadi et al. [203] used *N*,*N′*-methylene-bis-acrylamide (MBA) as a cross-linker for grafting acrylic acid (AA) on kappa-carrageenan (KC) during the production of the hydrogel. Here ammonium persulfate was used as the initiator. This mechanism is shown in Fig. 5. First of all, sulfate anion-radical is produced by the decomposition of the $S_2O_8^{2-}$ initiator, and hydrogen is removed from –OH groups of KC by this radical to produce alkoxy radical. Then the grafting of acrylic acid started because of the presence of active centers in the persulfate–saccharide redox system. Graft copolymerization can be utilized in natural or synthetic polymers and the combination of both [133, 204, 205].

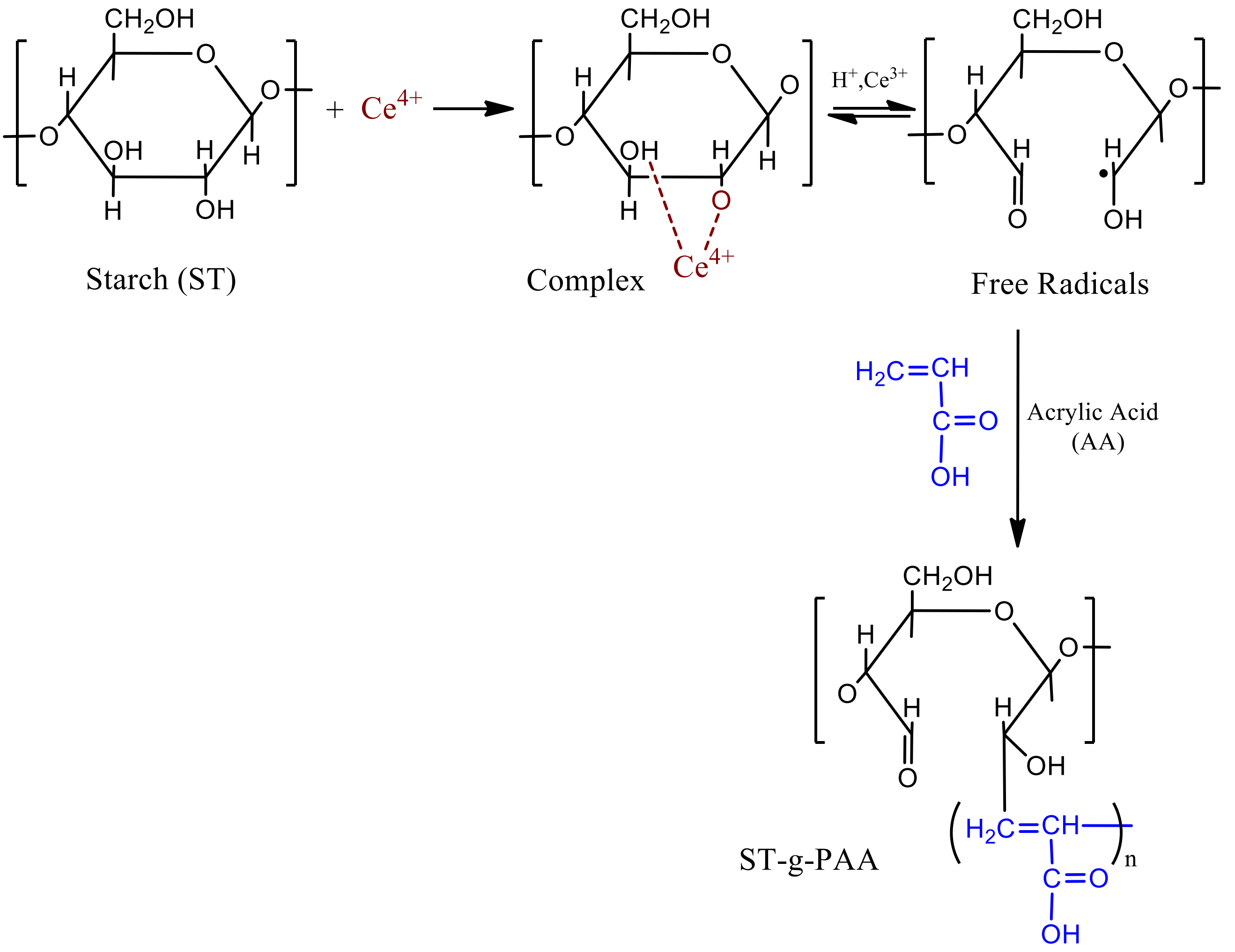


Fig. 4 Grafting mechanism of acrylic acid on starch with Ce^{4+} as free-radical initiator. Reused with permission from [198]. Copyright 2011, Elsevier

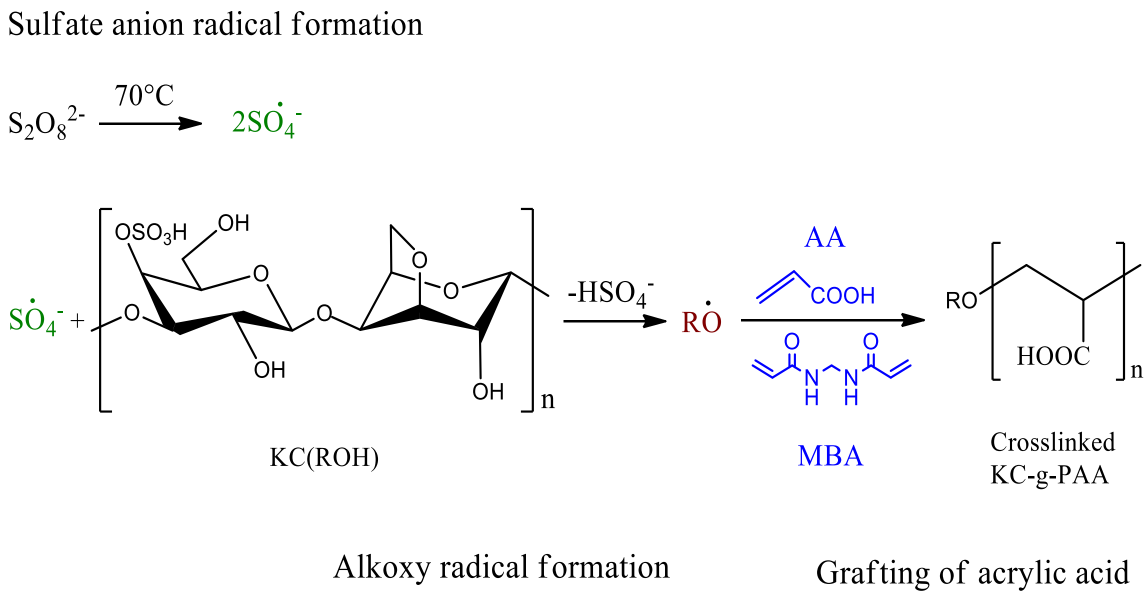


Fig. 5 Mechanism for graft copolymerization of acrylic acid (AA) onto kappa-carrageenan (KC) [17]

Direct Crosslinking Method

Polyfunctional compounds like glycerol, glyoxal, or polyvinyl compounds such as polyvinyl alcohol (PVA), polylactic acid (PLA), and divinyl sulphone are generally utilized in the direct crosslinking method of polymerization of hydrogel [130]. As crosslinking agents, glutaraldehyde, citric acid, glyceraldehyde are commonly used [124, 206, 207]. Hydrogel membrane was prepared by Pal et al. [208] by direct crosslinking of polyvinyl alcohol

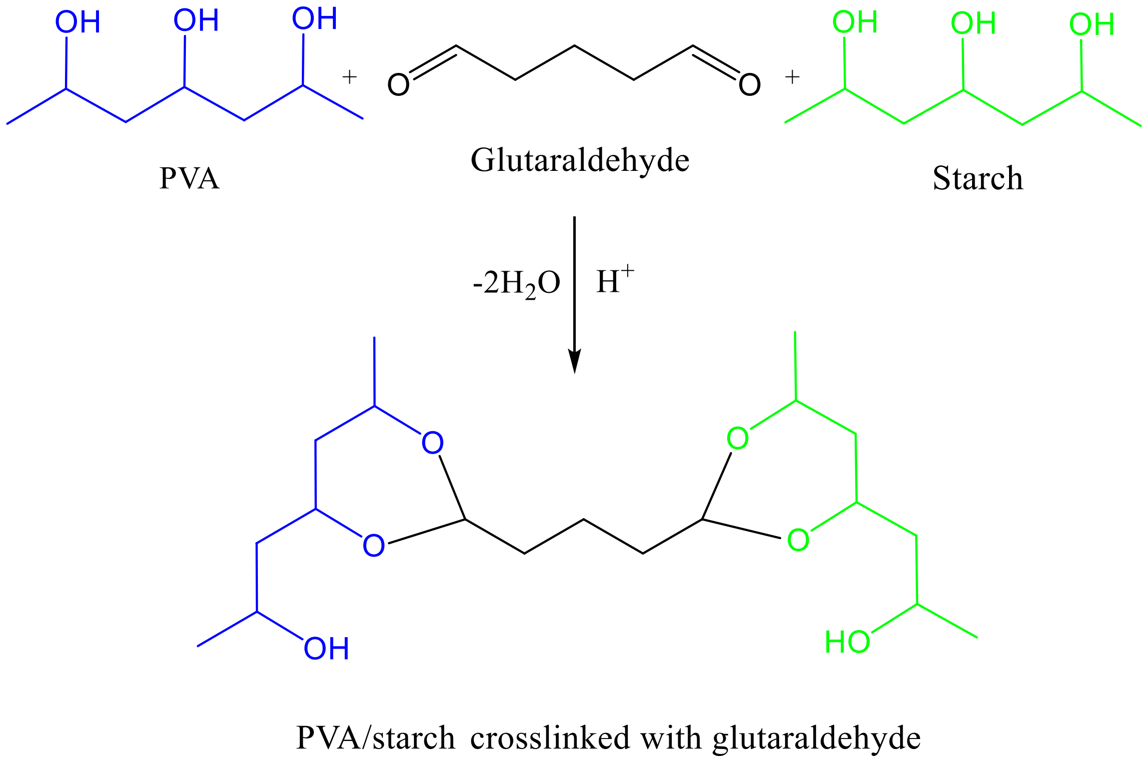


Fig. 6 Crosslinking mechanism of PVA–starch with glutaraldehyde [211]

(PVA) with starch using glutaraldehyde crosslinker. The mechanism is represented in Fig. 6. Acetyl bonds are formed when the glutaraldehyde reacts with the hydroxyl groups present in polyvinyl alcohol [209, 210]. Glutaraldehyde is a reagent which provides multifunctional property. Glutaraldehyde crosslinks starch with PVA by reacting with the hydroxyl groups present in starch. As a result, an intermolecular bridge between PVA and starch chains are formed.

In chemical functionalizing, starches are transformed into anionic polysaccharides [212]. For example, by esterifying starch with maleic anhydride, maleic starch half-ester acid is produced which is an anionic polyelectrolyte [143]. Sangseethong et al. [213] showed the mechanism of crosslinking between starch and citric acid during the preparation of rice starch-based superabsorbent hydrogel. This mechanism of the reaction is based on the anhydride intermediate formation of citric acid [214, 215]. A reactive cyclic anhydride is formed when the citric acid is dehydrated under controlled heating. An ester linkage is formed when the reactive anhydride reacts with starch by the esterification reaction. By this mechanism further crosslinking is achieved.

Radiation Induced Crosslinking Method

To fabricate SHs by chemical crosslinking method, sometimes different types of high energy radiations such as gamma radiation and electron beam irradiation are used [216–219]. The efficiency of this method is high and further contamination is not occurred [220]. Homolytic scission of C–H bonds of starch is occurred during this process. Hydroxyl radicals are generated from the water molecules due to irradiation. Starch radicals are formed when these hydroxyl radicals remove hydrogen from the starch chain [221, 222]. Final crosslinked network structures are developed by the covalent bonding of starch radicals and different chains. Starch radicals quickly react with oxygen. For this reason, an inert environment is required for crosslinking reaction [62]. Lv et al. [223] grafted acrylamide (AM) and dimethyl diallyl ammonium chloride (DMDAAC) onto corn starch using the gamma irradiation method (Fig. 7).

The development of SHs by the process of high energy irradiation has been studied by numerous authors. For instance, Zhai et al. [218] synthesized PVA/starch grafted hydrogel by inducing gamma and electron beam radiation at room temperature. ^{60}Co-gamma radiation-induced graft polymerization method was used by Geresh et al. [224]

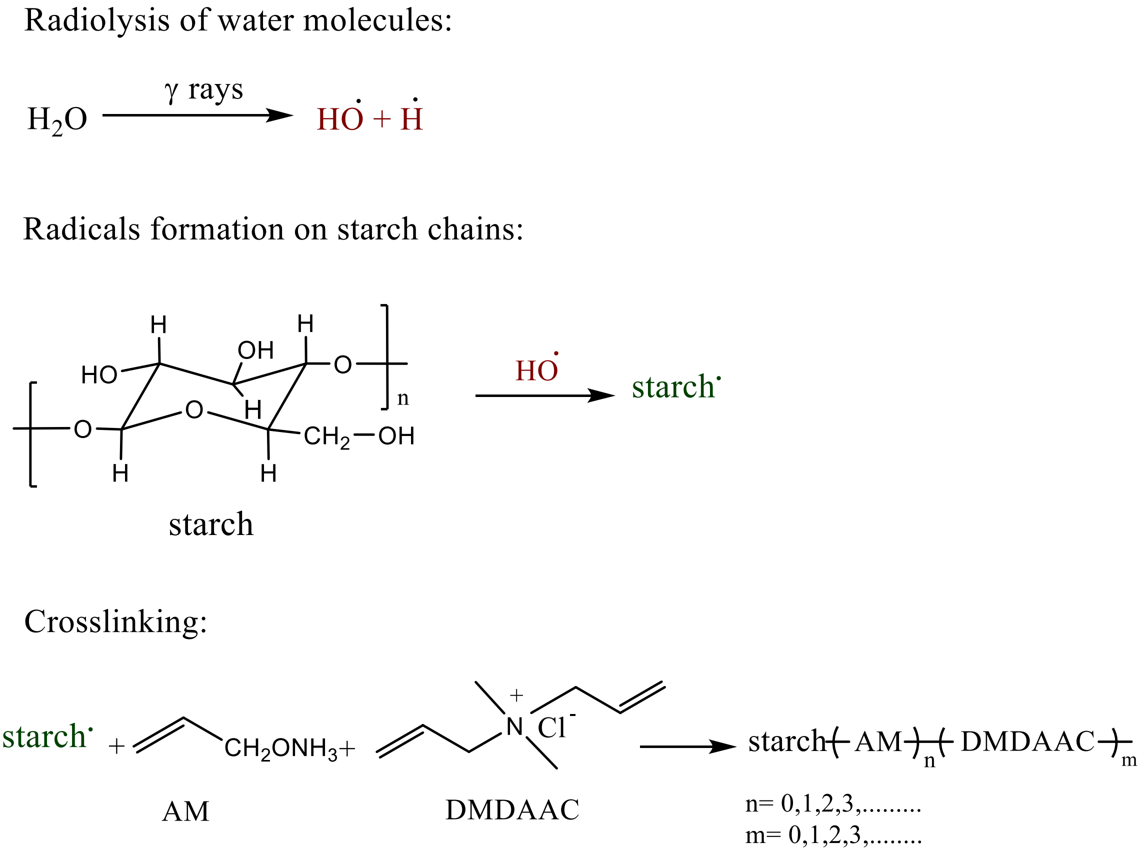


Fig. 7 Gamma radiation graft copolymerization mechanism of AM and DMDAAC onto corn starch [223]

to develop bioadhesive grafted starch. Acrylic monomers (acrylonitrile, acrylic acid) were used for grafting on multiple kinds of starch (potato, corn, and rice starches). The crosslinking formation was indicated by the insoluble nature of prepared polymer in water and NaOH solution of different pH range.

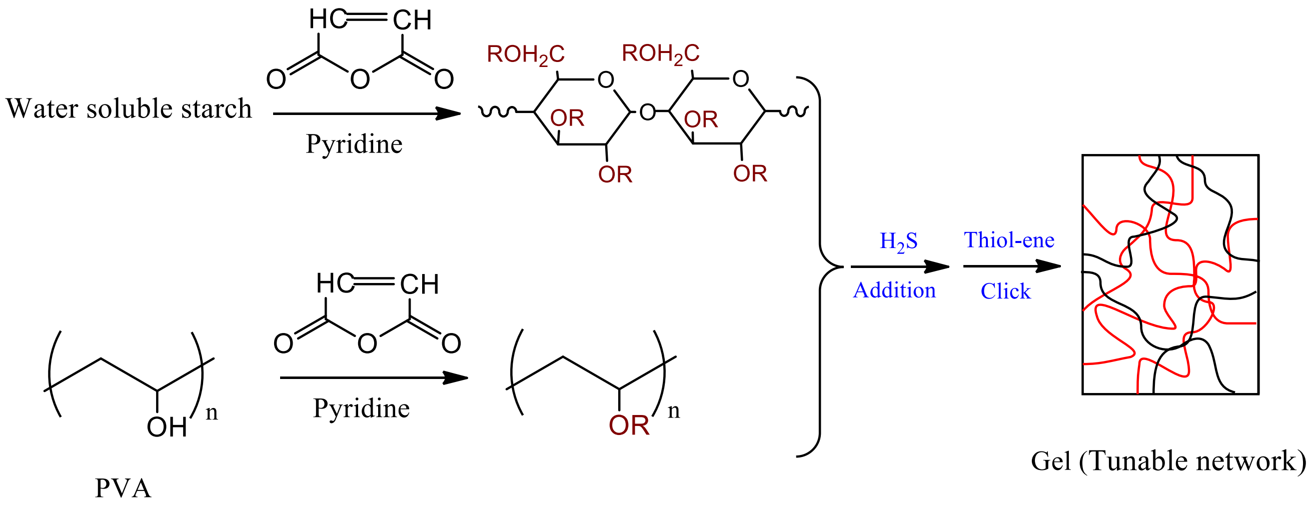
Abd El-Mohdy et al. [225] synthesized SHs (EG-*co*-MAA) by the copolymerization of ethylene glycol (EG) with methacrylic acid (MAA) which grafted on starch gamma initiated radiation. It was found that with increasing MAA content, temperature, and pH, swelling of the hydrogel is increased. The nature of crosslinking is affected by starch concentration and radiation. The degree of swelling reduces with increasing starch content and dose of irradiation.

A Combined Strategy

In a recent study, tunable SHs are proposed by Huang and Xiao [226] combining five different techniques—(I) addition–fragmentation chain transfer polymerization, (II) saponification, (III) esterification, (IV) addition, and (V) click reaction. First, both starch and PVA are utilized to prepare macromonomers. Potassium persulfate was used as an initiator. After that two reaction mechanisms (concurrent electrophilic addition and thiol-ene reactions) are utilized to synthesize SHs. Thiol-ene is a very rapid and efficient reaction mechanism for generating polymer networks [227, 228]. Here toxic H_2S is used for SHs preparation (Fig. 8) [226].

From Fig. 8, it is clear that SHs are formed by different reactions among the functional groups. In this regard old traditional techniques, new techniques, or the combining effort can be utilized for the development of crosslinking between the chains for SHs formation.

Comparing to the other chemical methods, the direct cross-linking method, drastic conditions have to be maintained like low pH, high temperature, and addition of quencher [229, 230]. The cross-linking agents used in this method are sometimes toxic (e.g., low concentration of glutaraldehyde is toxic showing cell growth inhibition) [231]. Hence they can't be directly used in biomedical applications [232, 233]. These hydrogels have good mechanical properties also and the characteristics can be modified by altering the concentration of the dissolved polymer and cross-linking agents. In the radiation-induced cross-linking technique, high energy irradiation (gamma and electron beam) is used for hydrogel production. Free radicals are formed on the polymer chain due to the radiation [234]. Different properties of prepared hydrogels such as the swelling property is dependent on radiation dose and polymer concentration. This process is advantageous than the direct cross-linking method because mild conditions (room temperature and physiological pH) can be used here. On the other hand addition of toxic cross-linkers are not necessary. For this reason, prepared hydrogels can be quickly used in drug delivery systems [131]. The thermal stability of hydrogel is dependent on the types of monomer being used and methods of crosslinking. Literature studies showed that thermal degradation of chemically crosslinked hydrogels starts at



R: H, CO-CH=CH-COOH

R': H, CO-CH=CH-COOH, $COCH_2CH(SH)$-COOH or COCH(SH)-CH_2COOH

Fig. 8 A combined method of SHs (tunable) formation. Reused with permission from [226]. Copyright 2012, John Wiley and Sons

180 °C. For increasing the thermal stability of hydrogels, cross-linking strategy plays a vital role [235–237].

Starch-Based Hydrogel Characterization Techniques

To know about various properties of hydrogels such as mechanical, structural, morphological, chemical, and physicochemical properties, characterization of hydrogels are necessary. Different characterization methods are employed according to the application of the hydrogels. There are various established characterization methods are available to study biodegradability, antimicrobial property, water uptake ability, mechanical properties, etc. [238].

Generally, to investigate the characteristics of SHs at micro and nanoscale, microscopic studies are utilized. For instance, the surface morphology of SHs can be examined and compared by using SEM (scanning electron microscope). For instance, Sangseethong et al. [213] synthesized superabsorbent carboxymethyl starch hydrogel using citric acid crosslinker. The internal morphology of native starch and prepared SHs were determined using scanning electron microscope (SEM, Quanta 450, FEI, Oregon, USA). It is found from the SEM image that three-dimensional interconnected porous structures are present in the prepared hydrogel (Fig. 9a, b). Chemical modification by graft copolymerization of the hydrogel surface can be observed from SEM images. In another study, Farag et al. [239] prepared bifunctional hydrogel by grafting AMPS (2-acrylamido-2-methylpropane-1-sulphonic acid) and DMAEMA (dimethyl aminoethyl methacrylate) on starch. Figure 9c shows the SEM image of starch and Fig. 9d shows the SEM of grafted SHs. The difference in the morphology of the SHs surface after the crosslinking can be marked from the SEM images. Homogeneous surface can be seen in the native starch whereas rough surface is observed in the crosslinked hydrogel.

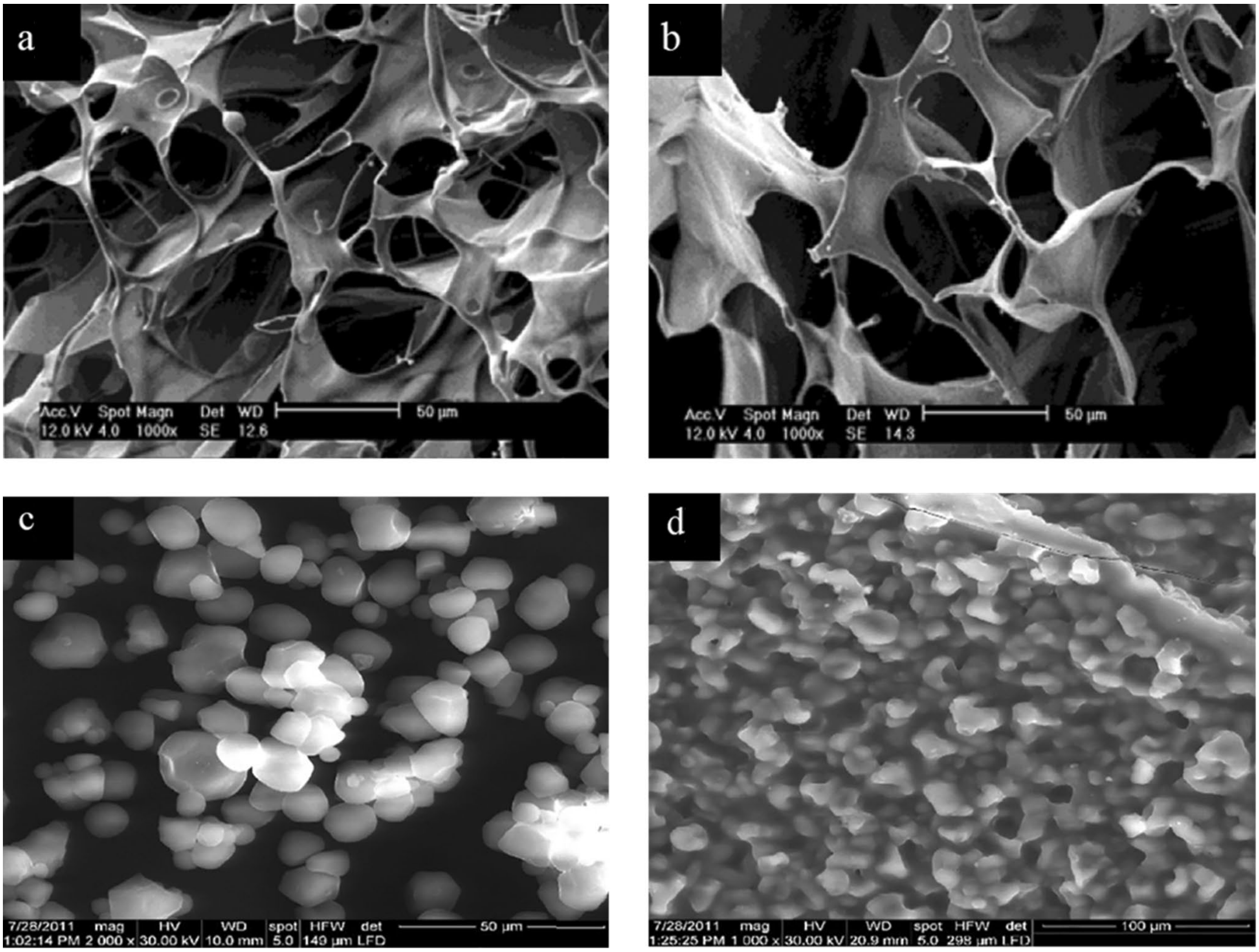


Fig. 9 SEM images for SHs characterization. **a** and **b** Reused with permission from [213]. Copyright 2018, John Wiley and Sons; **c** and **d** reused with permission from [239]. Copyright 2018, Elsevier

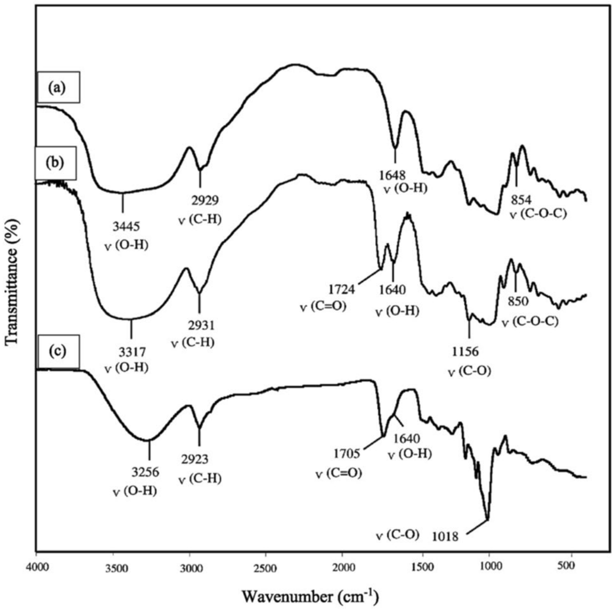


Fig. 10 FTIR images of (a) starch, (b) starch-citrate, and (c) starch-citrate hydrogel. Reused with permission from [242]. Copyright 2019, Elsevier

AFM (atomic force microscopy) is an important technique for hydrogel characterization. Quantitative information about the topography of the hydrogel and smooth gel surface with nanometer sized protrusions can be studied with AFM. Information regarding the mechanical and elastic properties of hydrogel can also be studied with AFM [240].

For observing the modification nature of hydrogels and identifying interactions between polymeric chains, Fourier transform infrared spectroscopy (FTIR) is utilized [241]. Hydroxyl group's presence in a hydrogel can be detected by FTIR analysis. For example, Chin et al. [242] prepared SHs by crosslinking of starch citrate, poly(vinyl alcohol) (PVA), polyethylene glycol (PEG) and studied the FTIR spectra of native starch (Fig. 10a), starch-citrate (Fig. 10b) and starch-citrate hydrogel (Fig. 10c). From the FTIR spectrum of starch-citrate, peaks at 1724 cm^{-1} and 1156 cm^{-1} indicate the ester carbonyl (C=O) group and C–O–C stretching of the ester group respectively. Peaks of 854 cm^{-1}, 917 cm^{-1} and 1018 cm^{-1} attributed to C–O–C glycosidic bond in starch, starch-citrate, and hydrogels respectively. OH-stretching peak at 1668 cm^{-1} in starch was decreased to 1640 cm^{-1} in the hydrogel. The absorption peak at 3317 cm^{-1} and 3256 cm^{-1} indicates the presence of –OH groups in starch-citrate and prepared hydrogel. It is known that the water retention capacity of the hydrogels mainly relies on the availability of free –OH groups [3, 243]. Hence from the FTIR spectrum, the water holding ability of the hydrogel can also be predicted.

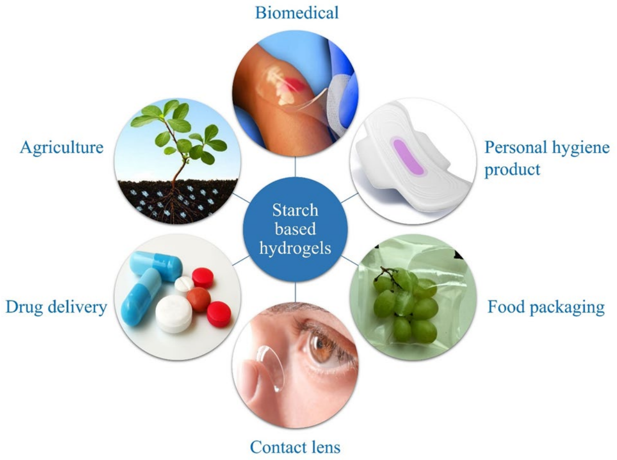


Fig. 11 Potential applications of starch-based hydrogels

The amount of crystallinity in SHs can be determined by XRD [208, 244]. Different characterization techniques such as TGA, DTA, DTG, and DSC are utilized for studying thermal characteristics and stability of hydrogels. The weight loss of the hydrogels with increasing temperature is analyzed by these thermogravimetric methods [245]. One of the significant properties of hydrogels is their mechanical properties which can be measured by tensile strength study [208]. Different types of tensile strength testers are available for this study [5, 246].

Water retention capability is the most important feature of SHs. Swelling behavior can be measure against different solutions such as water, sodium chloride (NaCl), and magnesium chloride ($MgCl_2$) [247]. Generally, cut pieces of the hydrogel are immersed in the solution and kept for 24 h, and swelled hydrogel is weighted. Finally, the swelling ratio can be determined by the following equation [2, 248]:

$$\text{Swelling ratio }(\%) = (-\text{Ws}-\text{Wd})/\text{Wd} \times 100.$$

Here, Ws is the weight of swelled membrane and Wd is the weight of dry membrane.

For evaluating the antimicrobial properties of hydrogels, anti-bacterial activity measurement is important. Disk diffusion technique is a popular method for antimicrobial test of starch-based hydrogels [243, 249]. Different types of gram-negative (e.g., *Escherichia coli*) and gram-positive (e.g., *Staphylococcus aureus*) bacteria are used in this method. From the zone of inhibition in the agar plate, antibacterial activity can be evaluated [250, 251].

Biocompatibility is an important property of starch-based hydrogels. Hence hydrogels need to undergo biodegradability test. Two highly used degradation tests are—(I) direct microbial studies, and (II) soil burial test. Hydrogel is buried in soil for a period of time. Then the sample is washed and weighted. Weight loss percentage is the result of degradation. The direct microbial study is a similar process to the soil burial test. Additionally, cultured microorganisms are used here for the degradation of SHs [252, 253].

Applications of Starch-Based Hydrogel

The popularity of SHs is increasing day by day because of their characteristics such as biodegradability and dynamic mechanical properties. SHs have different applications in biomedical sectors (e.g., drug delivery system, contact lenses), agricultural sectors (e.g., controlled pesticide release, water storage), food industries (e.g., food packaging), and so on. SHs have various industrial applications. In the cosmetic industry, electrophoresis, and food industries, SHs are widely used [22, 254, 255]. Different applications of SHs are represented in Fig. 11.

Biomedical Applications

In the biomedical sectors, the use of SHs have several advantages due to having some unique properties like the biological systems [256–262] which are—(I) biocompatible nature, (II) higher biodegradability, (III) good mechanical characteristics, (IV) hydrophobic property, (V) permeability and so on. For giving the patients quick structural support nowadays bone cement is utilized which is starch-based and gradually degrade from the placement site. In addition, different bio-active materials are used with starch-based bone cement resulting in the growth of bone at the cement–bone interface and unnecessary portion degrade naturally [263]. Along with water absorbing capacity SHs show good mechanical properties which enhances their applicability in biomedical sectors. Hassan et al. [2] developed PVA/starch-based antibacterial hydrogel membrane for wound dressing. For the first time, they used turmeric as an antimicrobial agent in SHs. Batool et al. [243] synthesized starch-based antimicrobial nanocomposite hydrogels for wound dressing by incorporating Ag nanoparticles. Artificial skin had been fabricated from corn starch-based hydrogels by Pal et al. [264].

Tissue Engineering

SHs are soft and flexible like living tissues and can retain a huge amount of water. For this reason, in tissue engineering, the use of SHs are becoming popular [265]. Starch-based scaffolds are an excellent innovation in tissue engineering. They represent similar characteristics to the body matrix of the human tissue which are mainly different amino acids and sugar-based macromolecules. SHs can mimic the properties of body matrices being completely biocompatible [266]. The ability of starch to create pores in ceramics and membranes can be utilized in scaffold bone tissue engineering [77, 267, 268].

Several types of SHs such as chitosan-starch hydrogels are utilized in tissue engineering applications [59, 265]. Salgado et al. [269] synthesized novel scaffolds based on starch particularly for bone tissue engineering. Prepared scaffolds are completely nontoxic, have good mechanical properties (compressive modulus and compressive strengths are 117.50 ± 3.7 MPa and 20.8 ± 2.4 respectively), don’t inhibit cell growth, and increase the growth of human osteoblast like cells. Kamoun [270] prepared SHs based on dialdehyde starch (DAS) and *N*-succinyl chitosan (SCS) without using any traditional chemical crosslinking agents. The synthesized hydrogel is injectable, non-toxic, biodegradable, biocompatible, and showed good mechanical properties. The use of prepared hydrogel in tissue engineering and cartilage repairing is proposed.

Another important characteristic of starch-based materials is their different configurations. Available configurations of starch-based materials are fibers, films, and beads etc. [271–274]. Different authors have used several techniques to prepare starch-based scaffolds such as—(I) Fiber bonding method [275–277], (II) Particulate leaching methods [275, 278], (III) Phase separation method [279–281], (IV) Microwave backing method, etc. [282]. But utilization of SHs in tissue engineering is studied by few authors. For instance, Gomes et al. [278] prepared starch-based polymeric scaffolds and studied their configurations, mechanical properties, morphology, and degradation behavior. Tunable SHs were fabricated by Noè et al. [283] from maize starch by photocuring process. The prepared SHs were light processable and showed good mechanical properties along with biocompatibility. As the hydrogel showed good human cell viability it can be utilized in human tissue engineering and cell carriers. Thermosensitive SHs have been prepared by blending starch and chitosan by Viyoch and colleagues [284]. They have potential applications in cartilage tissue engineering and chondrocyte delivery through injection. The pore size and hydrophobicity of the fabricated hydrogel are enhanced because of using starch which is more advantageous for tissue engineering [285].

Drug Delivery Systems

The use of SHs in drug delivery devices is one of the successful biomedical applications of SHs and they are already used at the industrial level. Delivering the site-specific drug in the body with the highest efficiency and minimal side-effects are the main purpose of creating a drug carrier system. Different hydrogels based on polysaccharides (e.g., dextran, chitosan, pectin) is used for site-specific drug carrier of the colon [286–288]. These types of site-specific drugs show higher efficiency because the drug molecules are out of contact with other tissues and organs of the body [289]. SHs possess some unique and modifiable physical characteristics which are very much useful for using them in the drug delivery system. The surface of SHs are porous resulting in great affinity for liquid environment and swollen. The porosity of SHs is controllable by altering the density of crosslinking. Porosity is an important property because it assists in the quick and easy loading of drug molecules into hydrogels and helps in releasing the drug in a specific area [148]. Some studies on SHs in drug delivery systems showed very good results. For instance, Ketoprofen drug loaded SHs based on starch-methacrylic acid (MAAc) copolymer was prepared by Ali et al. [288]. The study showed very good results in retaining the drug at a very low pH (pH 1) and released the loaded drug at pH 7. Prepared drug loaded SHs showed excellent pH sensitivity and they can also be utilized in the site-specific drug of the colon. For the treatment of colon disease like Crohn's disease, SHs prepared from starch (starch-M hydrogel) with GMA (Glycidyl methacrylate) was studied by Reis et al. [290]. This SH showed excellent behavior in preserving and transporting corticoids which is an acid-responsive drug. For controlled released drug delivery system, xanthan gum hydrogels can be a good alternative because it shows selective permeability on ionizable drug loading. Shalviri et al. [291] studied SH (starch–xanthan gum hydrogels) for drug delivery system. Their study suggests that prepared novel SHs can be used in drug delivery systems for different types of drugs because of their selective permeability property.

Agricultural Applications

Agriculture is one of the most important industries. Good agriculture mostly relies on an adequate quantity of water and nutrients (enhanced by fertilizers). Plants can absorb about 30–60% of the N_2 from fertilizers and the rest is leached into groundwater and river water causing extreme environmental pollutions [292–295]. For reducing this problem controlled-release fertilizers can be a good choice in which fertilizers will be released slowly preventing the loss of the valuable amount of fertilizers [296, 297]. Currently, scarcity of water is a global issue. An enormous amount of water is needed for irrigation. In summer seasons, a shortage of water is seen and in some desert areas, rainfall is not available. In this regard, hydrogels with high water absorbing capacity can be used. Hydrogels can retain water for extended periods and release water slowly. Hence hydrogel-based agriculture can be widely used [298]. Advantages of using hydrogels in farmlands are many, such as—(I) less amount of water is required for irrigation, (II) fertilizer is retained in the soil for a longer period, (III) decrease plant death, (IV) enhance plant growth, and (V) decrease environment pollution [299].

SHs can be utilized in this sector for water storage and fertilizer retention [22]. Starch-based polymers are utilized not only in water and fertilizer storage but also in the greenhouse hood and mulch film [300]. SHs can prevent the fertilizers from early leaching and surface runoff [292, 299, 301, 302]. It is found that the use of starch-based superabsorbent hydrogels enhances bacterial growth in the farmland. By utilizing this property, weedy plants and harmful microorganisms can be controlled from the land. Hence controlled pesticide formulations are another use of SHs in agriculture [83]. Along with water and fertilizers, proper use of pesticides is also important for increased production of foods and preventing environmental problems [303, 304]. Hence controlled released pesticides are required to reduce environmental leaching and dermal toxicity [305–307]. For example, SHs were prepared by Singh et al. [308] for the controlled release of thiram fungicide. The SH was prepared from starch and poly (methacrylic acid) adding initiator (ammonium persulphate) and crosslinker (*N*,*N*′-methylene-bis-acrylamide). It showed a very good result in controlled thiram release [309]. Starch–alginate beads (adsorbents—bentonite and kaolin) were used for controlling the release of thiram by Singh et al. [310].

For the slow release of thiophanate methyl, fluometuron, and trifluralin, Abd El-Mohdy et al. [311] prepared starch/(EG-*co*-MAA) hydrogels. SH with the copolymers acrylic acid and acrylamide was used as the membrane of urea by Liu et al. [299] for slow release purposes. The observation result showed that 61% N_2 was released on the 13th day from the hydrogel and its water intake capacity was 80%. Fang et al. [312] used the graft copolymerization technique to prepare SHs. Acrylamide and acrylic acid was grafted onto starch and ethylene glycol was the crosslinker. These SHs were used to observe the corn seed germination. Germination energy was comparatively higher than normal germination when SHs were used.

Food Industry

The use of starch-based biodegradable polymers for the packaging of food is getting attention nowadays. The use of petroleum-based plastic can be minimized by using natural biodegradable polymers [313]. In the food industry, starch-based biopolymers are primarily used for food packaging [314]. For using starch-based polymers in food packaging some requirements should be maintained such as—(I) packaging materials should be cheap, (II) protection from chemical, biological, and physical influences, and (III) keep the food fresh [315]. Traditionally food packaging made from LDPE (low-density polyethylene) is not ecofriendly and takes a long time to be disposed of [316]. Starch-based biodegradable polymers can be a good alternative to this packaging material [188]. The main limitation in this respect is the retrogradation of starch in presence of water. To overcome this issue, thermoplastic starch can be prepared by adding different types of plasticizers [315]. Different methods like thermo-processing or casting techniques can be used for fabricating starch films [317–319]. Avella et al. [320] prepared starch/clay nanocomposite-based biodegradable packaging materials for food. Good mechanical properties (modulus and tensile strength) had been shown by the packaging. Different forms of starch like modified, soluble, or pre-gelatinized starch are used to prepare starch films [321, 322]. Some advantages of using starch film are—these films are non-toxic, biodegradable, odorless, transparent, and low oxygen permeability at less humidity [323, 324]. Furthermore, foamed materials can be prepared from starch which can be the replacement of polystyrene foam [325]. For food packaging, chitosan-starch based hydrogels can be used [326]. Jyothi et al. [327] crosslinked (cassava starch with epichlorohydrin) polymer for food packaging with enhanced physicochemical, thermal, and retrogradation properties. Food packaging film coated by starch-graft-acrylic acid was prepared by Tsuji et al. [17]. For controlling humidity (for fruits and vegetable preservation), CO_2 and O_2 concentration urethane was used as binder. Hence, in the food packaging industry, the starch-based biodegradable polymers can bring revolutionary change by implementing eco-friendly packaging.

Electrical Applications

Nowadays superabsorbent hydrogels are used in numerous kinds of electrical sectors such as in electronic water-resistant cable coatings, corrosion-resistant coatings etc. SHs are utilized in telecommunication cable production. When the outer layer of the electric cables are damaged, water enters the inner side and damages the cable. Cables can be protected from unwanted water by using hydrogels in the cable. They can also be utilized in the water blocking tape for electronics by dispersing on the fabric [328].

Water resisting cable was prepared by Schoeck et al. [329]. For enhancing the water blocking capacity of the cables, starch-based super absorbent hydrogels coated filaments were utilized. Starch grafted sodium polyacrylates, and partial sodium salts of polypropenoic acid were used in the filament. Alcohol soluble yarn binders were used to attach the hydrogel to the surface [17, 328, 330].

Starch/graphene hydrogels were prepared by González et al. [331] by Diels–Alder cross-linking reactions between starch (furan modified) and bismaleimide (BMI) compound. It was hypothesized that enhanced antimicrobial and electrical properties could be represented by the prepared SHs. Electrically conductive SHs (polyaniline impregnated polyacrylate-starch hydrogel) were prepared by Prabhakar and Kumar [332] and conductivity was studied with a varying crosslinker, initiator, and monomer concentration.

Personal Hygiene Applications

Superabsorbents are mainly used in diapers, sanitary napkins, adult incontinence products, feminine hygiene products as an absorbent for water. These personal care products are very important for keeping individuals dry and hygienic especially during the period time [333–335]. It has taken the place of cloth, cotton, cellulose fiber etc. which were traditionally used as absorbent. Because they have limitations like low water absorption capacity, recyclability [336]. There are mainly two parts in diapers—core and chassis. Superabsorbent polymers are mainly used in the core part to absorb body fluids [337]. Hydrogels can absorb huge amounts of body wastes, fluids, and retain them under pressure. All these features have made hydrogels one of the most suitable materials for using in personal hygiene products. By reviewing the literature, it is seen that lots of studies have been done on the cellulose based superabsorbent hydrogel and its use in hygiene applications [338–344]. But a few works have been found on starch-based superabsorbent polymers used on this site. A starch-based film was prepared by Weisman et al. [17] from starch-graft-acrylic acid. This film could be used for the storage of fluid in diapers. Nowadays cellulose and acrylic-based hydrogels have got huge attention. Hence further research on the use of SHs in this sector should be continued.

Water Treatment

Water pollution by heavy metals (Ni, Pb, Cd, Ag, Hg) is one of the main environmental issues all over the world. If the number of heavy metals in water crosses the permissible limit then it may cause serious environmental and human health issues [345–350]. Traditionally different types of water recycling methods (ion exchange, solvent extraction, and precipitation) are used for the purification of water from heavy metals. But these methods are expensive and a large quantity of solid wastes are produced [17]. Using different adsorbents for heavy metal ion removal from water is a very effective method. Nowadays activated carbon is highly used for the heavy metals (Cd, Ni, Cr, Cu) [351–354] removal from aqueous media [355, 356]. Although it has several advantages (adsorption capacity is higher, large surface area), it is not a very cost-effective approach for water recycling. Hydrogels prepared from polysaccharides can be a great alternative for high-cost adsorbents for water recycling because they are inexpensive and non-toxic [357, 358]. Several authors have studied the use of inexpensive adsorbents like starch biopolymers and SHs to extract toxic heavy metals from wastewater [359–363].

Cassava starch-based hydrogels were prepared by Ekebafe et al. [362] by grafting acrylonitrile onto native cassava starch. Ceric ammonium ion was used as an initiator. Under equilibrium conditions, sorption mechanisms were studied for Ni^{2+}, Cu^{2+}, Pb^{2+}. The sorption data were a good fit for the Freundlich model represented the sorption capacity for grafted hydrogel: 72 mg Pb/g, 76.6 mg Cu/g, and 86.5 mg Ni/g. With increased hydrolysis, heavy metal affinity also increased.

Abdel-Aal et al. [364] synthesized maize starch/acrylic acid hydrogels by radiation-induced graft copolymerization technique to remove heavy metal ions such as Fe^{3+}, Cr^{3+}, Pb^{2+}, and Cd^{2+} from wastewater. The sequence of metal uptake capacity for the hydrogel was $Fe^{3+} > Cr^{3+} > Pb^{2+} > Cd^{2+}$ which increases with increased pH.

For the effective removal of Cu^{2+} ions from the wastewater, Dai et al. [365] prepared thermoresponsive composite SHs. The novel SH was synthesized from sodium alginate and 2-hydroxy-3-isopropoxypropyl starch and its response to the pH was investigated. It was observed that inside the hydrogel structure there are sufficient Cu^{2+} ions binding sites. The maximum adsorption capacity for Cu^{2+} was 25.81 mg/g.

Chauhan et al. [366] synthesized functional SHs (polycarboxylated) by the free radical initiation technique for sorption of Cu^{2+} ions where *N*,*N*′-methylene-bis-acrylamide was used as crosslinker. At pH 7, maximum adsorption capacity for the ions was investigated 128.26 mg/g^{-1} (2 h at 40 °C). From the FTIR data it was seen that ions were adsorbed by chelation mechanism between the $–CO_2$ and –C=O or –OH groups of the hydrogel with Cu^{2+} ions.

Yu et al. [367] proposed starch/PVA hydrogels crosslinking with Laponite RD (LRD) by freezing/thawing technique for cadmium ion absorption. It was investigated that with raising LRD concentration, Cd^{2+} ion absorption was increased.

For the efficient sorption (exothermic and spontaneous) of Fe^{2+} ions from the solution, a range of hydrogels were prepared. As initiator accelerator ammonium persulphate (APS) and *N*,*N*,*N*,*N*-tetramethyl ethylene diamine (TEMED) were utilized. The investigation showed that metals ion adsorption capacity is strongly relied on the environment (temperature and pH). It was clear from the characterization techniques that metal ions intake capability of *N*,*N*′- methylene-bis-acrylamide crosslinked hydrogels were less than the hydrogels crosslinked with ethylene glycol dimethacrylate [368].

Dye Sorption

Water pollution is a dangerous environmental issue [369]. Different industrial effluents carry a variety of toxic chemicals, dyes, and heavy metals. Plastic, leather, paint, textile, paper, and cosmetic industries use toxic dyes to color their products and release a huge amount of toxic dyes in rivers and groundwater without appropriate treatments [370, 371]. Textile dying industries are the major contributor to this dye pollutions [372, 373]. The World Bank reported that 17% to 20% of the polluted water comes from the dyeing and finishing sectors of the textile industries [374–376]. These water containing colorants and dyes are very harmful to the ecosystem and public health [377–383]. Different types of methods have been utilized to remove industrial dyes from wastewater such as—adsorption, anaerobic degradation, coagulation, flotation, biological treatment, chemical treatment, and so on [384, 385]. Among these methods adsorption method is becoming popular day by day because of its high efficiency, low cost, and easy handling [386–389]. The sorption process is subjected to different factors like pH, temperature, the surface area of solvent, bonding between the dye, sorbent, and particle size, etc. [17, 385].

To remove dyes from wastewater different types of adsorbent materials are utilized such as—different types of clays [390], synthetic hydrogels [391–393], and biopolymers [394–396]. Different adsorbents based on natural polysaccharides (chitosan, starch-based hydrogels) have got much attention these days [53, 364, 397–399].

Mahmoud et al. [400] prepared starch, acrylic acid, and 2-hydroxyethyl methacrylate (HEMA) based hydrogels by gamma radiation for the adsorption of acid-fast red dye from industrial wastewater. With increased gamma irradiation gel content of the prepared hydrogel also increased. The effect of temperature, pH, and concentration of the dye medium during adsorption was also investigated. With the decrease of pH, the adsorption capacity of the hydrogels increased whereas capacity decreased with lower initial dye concentration and temperature.

IIgin et al. [401] proposed the preparation of hydroxyethyl starch (HES) based hydrogels, hydroxyethyl starch/p(sodium acrylate), hydroxyethyl starch/p(3 (acrylamidopropyl) trimethyl ammonium chloride), and hydroxyethyl starch/p(acrylamide) to adsorb cationic dyes [e.g., methyl violet (MV) and methyl orange (MO) from water]. The hydrogels were prepared by redox polymerization technique. The result showed that the organic dye adsorption efficiency of the hydrogels was over 90% (maximum adsorption of MO and MV was 238.1 mg/g and 185.2 mg/g).

Starch/poly(alginic acid-g-acrylamide) nano-hydrogel (ST/PL(AA-g-AAm) NHG) was synthesized by Sharma et al. [402] by copolymerization technique. The prepared hydrogels were utilized for the adsorption of Coomassie brilliant (CB) blue R-250 dye from wastewater. The adsorption isotherm was investigated from the regression coefficient value of Langmuir (0.99), Freundlich (0.93), and Tempkin (0.98) isotherm which shows that Langmuir isotherm fits the adsorption process. The dependency of adsorption on temperature, time, pH, and dye concentration was also studied in detail.

For the removal of different dyes such as anionic dyes [direct brown 2 (D. Brown 2), direct blue 21 (D. Blue 21)], and a cationic dye (methylene blue), Hashem et al. [403] prepared bi-functional starch composite hydrogels by grafting acrylic acid (AA) and acrylamide (AAm) onto maize starch. Gamma irradiation induced grafting mechanism was utilized for the hydrogel preparation. The adsorption of D. Brown 2 and D. Blue 21 onto a prepared composite followed the Freundlich isotherm.

Different studies had been done on dye adsorption by SHs such as—Dragan et al. [198] synthesized semi-IPN composite hydrogels from potato starch for the adsorption of Methylene Blue dye. Bhattacharyya et al. [388] prepared SHs based on acrylamide, hydroxyethyl methacrylate, and starch for effective removal of low (2.5–40) and high concentration (200–500 mg/L) cationic dyes from aqueous solution. Superabsorbent chitosan-starch (ChS) hydrogels were prepared by Ngwabebhoh et al. [371] for the adsorption of Direct Red 80 (DR80) from water. Moharrami and Motamedi (2020) synthesized starch-based hydrogel nanocomposites from agricultural waste for the adsorption of two cationic dyes—crystal violet (CV) and methylene blue (MB). Malachite green and methyl violet are two widely used dyes in industries for being cheap and easily available [404, 405]. Al-Aidy and Amdeha [406] proposed the preparation of polyacrylic acid and acrylamide grafted starch hydrogels for the enhanced adsorption of malachite green dye from water.

Miscellaneous Applications

The use of SHs are gaining huge attention in numerous areas and industrial sectors such as petroleum industries, paper industries, plastic and ion exchange resin manufacturing, and so on [407, 408]. The use of SHs for making biodegradable plastic film can be a good alternative to synthetic polymers. Thermoplastics can be fabricated by grafting starch with acrylate esters, methacrylate esters, and styrene. Starch-g-poly(methyl methacrylate-*co*-acrylonitrile) polymers can be highly used in making bio-plastics because of their good toughness. The starch grafted copolymers are easily attacked by soil microorganisms and degrade rapidly [409, 410]. Biodegradable polyethylene was developed by Maharana and Singh [411] by graft copolymerization of starch.

For reducing the filtrate loss in the oilfield drilling muds, starch graft copolymers are frequently used. Acrylamide and vinyl alcohol monomers grafted on starch are utilized in drilling mud [412–414]. Song et al. [415] prepared copolymer of starch with acrylamide (AM) and 2-acrylamido-2-methylpropanesulfoacid (AMPS) by grafting mechanism and used the copolymer as flooding agent in oil recovery. The study showed that with a higher grafting level oil recovery rate also increased. Acrylonitrile-graft-starch and quick lime particles were utilized by Kobayashi et al. [416] for the development of demolition agents for demolishing the rigid structures. SHs are potentially used in other sectors such as sizing agent of textiles (e.g., starch-g-poly(vinyl acetate-sizing agent of cotton)) [417], dye thickening agents etc.[418].

Research Gaps and Future Aspects

Research on starch-based hydrogel is flourishing due to its distinctive characteristics and potential applications. Synthesis methods of SHs are developing. The combination of both physical and chemical cross-linking is getting much attention. Different studies have been conducted on starch-based smart hydrogels such as responsive hydrogels (e.g., pH sensitive and stimuli responsive SHs) [198, 237, 419–421], dual and multi-responsive SHs [422, 423], hybrid nanothermochromic SHs [424], starch-based tunable hydrogels [425], magnetic SHs [426], clickable SHs for 3d encapsulation [427] etc. Further studies and investigations are necessary for developing hydrogels with smart properties. In this regard nanostructured, hybrid and bioinspired SHs can be the field of further interest. Hydrogel's characteristics depend heavily on its method of preparation, composition, and structure. So, various kinds of starch-based hydrogels with numerous unique properties can be developed by closely customizing the structure of the macromolecules based on starch. Sometimes the properties of native starch-based hydrogels are not as good as synthetic polymers such as having poor mechanical properties. Hence different types of synthetic polymers (e.g., PVA, PLA) are utilized for the modification of starch hydrogel properties. To catch the commercial fields, the performance (e.g., mechanical properties, antimicrobial properties, swelling behavior) of starch-based hydrogels need to be improved. Although different studies are going on for applying SHs in biomedical and other fields, commercialization of these products are not satisfactory. For the development of more commercial products with more advanced and unique properties, further research on SHs should be continued.

Conclusion

There are many beneficial properties of starch-based hydrogels. They are hydrophilic, biodegradable, biocompatible, pH-responsive, and non-toxic in nature. In addition, they are cheap and their high accessibility from natural sources makes them more appealing, intelligent polymer materials. Due to their environment-friendly nature, starch-based sustainable hydrogels can be the alternative to petroleum-based polymeric materials. People will be no longer highly reliable on synthetic polymers. Preparation and characterization methods of SHs are comparatively easier. Although, SHs have some drawbacks such as lower sustainability and poor mechanical properties, promising application opportunities of SHs are widening day by day. They are occupying the place of synthetic hydrogels in the field of agriculture, biomedical (e.g., tissue engineering, drug delivery), water recycling, and food industries. More studies on starch-based sustainable hydrogels are required. In this review, a brief summarization of preparation methods, characterization techniques, and several potential applications of SHs are given. This review might be helpful for the researchers studying in this field.

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