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A review of polymerization techniques for preparing hydrogels

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ABSTRACT

The present study reviews articles on polymerization techniques for preparing hydrogels. Hydrogels are three-dimensional networks of polymer chains that are cross-linked through a simple reaction of one or more types of monomers and have a high ability to absorb and retain water. The ability of hydrogels to absorb water comes from hydrophilic functional groups such as COONa, OH, NH_2, COOH, SO_3H that are attached to the base chain. While their insolubility in water is due to cross-links between the main chains. Hydrogels are three-dimensional, hydrophilic polymer networks capable of absorbing large amounts of water or biological fluids. Due to their unique physicochemical properties, biocompatibility, and tunable characteristics, hydrogels have found widespread applications in biomedical engineering, drug delivery, wound healing, and environmental science. This review provides a comprehensive overview of various polymerization techniques used to synthesize hydrogels, including physical and chemical crosslinking methods. Key polymerization strategies such as free radical polymerization, photopolymerization, click chemistry, radiation polymerization, and enzyme-mediated polymerization are discussed in detail, along with their advantages, limitations, and typical applications.

Introduction

When the chemical potential of water in a hydrogel is higher than the environment, water penetration from the hydrogel to the surrounding environment occurs, or in other words, swelling occurs and water is transferred to the environment [1]. This phenomenon is accompanied by shrinkage of the hydrogel. This property has led to the use of these materials to maintain soil moisture. Hydrogels can be of natural or synthetic origin. Natural hydrogels can include polysaccharide groups (chitosan, alginate, hyaluronic acid, and dextran), protein-based hydrogels (fibrin, collagen, and gelatin), and DNA-based hydrogels. Natural hydrogels are of interest due to their biocompatibility, cheapness, versatility, and wide availability [2]. Over time, synthetic hydrogels have replaced natural hydrogels due to their higher water absorption capacity, long life, and wide range of chemical sources. Natural polymers are fully hydrated in less than two hours, while synthetic polymers take about six hours or more to fully hydrate.

Different types of nanoparticles such as ceramic mineral nanoparticles such as hydroxyapatite, silica, silicate, calcium phosphate, metal or metal oxide based nanoparticles such as gold, silver and iron oxide, carbon based nanoparticles such as carbon nanotubes and graphene and polymer nanoparticles such as dendrimers, liposomes and micelles are combined with polymer network to produce nanocomposite hydrogels [3].

The placement of these nanoparticle in the hydrogel structure is done by various interactions. Based on the thermodynamic equilibrium in hydrogels, when the chemical potential of water in the environment is greater than that of the hydrogel, water will penetrate from the environment into these materials, and this water absorption will cause these polymers to swell to several times their initial volume. Based on the preparation method, hydrogels can be classified into homopolymeric, copolymeric, and multipolymeric categories. In homopolymeric hydrogels, the polymer network is formed from only one type of monomer. Copolymeric hydrogels are

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composed of two or more different types of monomers and are arranged randomly, block wise, or alternately in the polymer network. Multipliers or interwoven polymer hydrogels have two natural or synthetic polymer components. Hydrogels are classified according to their physical structure (Configuration) as amorphous, semi-crystalline, and crystalline. The type of cross-linking between monomers can be physical or chemical. Physical connections between polymers can be made using non-covalent interactions such as hydrophobic interactions, ionic interactions, hydrogen bonding, or a combination of these [4]. Chemical connections are made through covalent bonds between polymer chains, and the resulting hydrogel network is usually more resistant to mechanical stress than physically connected networks (Table 1).

Table 1. Comparative Summary of Techniques

Technique	Advantages	Limitations	Applications
Free Radical	Fast, broad monomer range	Toxic residues, uncontrolled kinetics	Drug delivery, hydrogels
Photopolymerization	Spatial/temporal control	UV damage, depth limitation	Tissue engineering
Click Chemistry	High specificity, bio-orthogonality	Requires functionalized precursors	Cell encapsulation, bio-printing
Radiation Polymerization	Sterile, no initiator	Expensive, specialized equipment	Medical hydrogels
Enzymatic Polymerization	Mild conditions	Slow, enzyme cost	In situ biomedical applications

These interactions include: covalent, hydrogen and electrostatic interactions. The resulting nanocomposites have physical properties more suitable for drug release through stimulating agents. The type of nanoparticles used in the hydrogel structure determines the type of stimuli that can control the drug release under desired conditions. The addition of nanoparticles to the hydrogel nanocomposite and their interaction with the hydrogel matrix occurs in three ways.

- ✓ Micro- or Nano-sized hydrogel particles stabilizing inorganic or polymeric nanoparticles.
- ✓ Nanoparticles immobilized in the hydrogel with non-covalent bonds [5].
- ✓ Nanoparticles immobilized in the hydrogel with covalent bonds.

Studies

Yu and colleagues fabricated 3D ternary nanostructured electrodes made of PPy-Si-CNTs that had a reversible capacity of 1600 mAhg-1 and a storage capacity of more than 85% after 1000 cycles. The use of carbon nanotubes greatly enhances the electron transport and conduction capabilities of the PPy framework by improving both the physical and electromagnetic coupling between the silicon nanoparticles and the conductive polymer gel framework.

Bao and colleagues developed a ternary system consisting of PEDOT: PSS hydrogel, carbon nanotubes, and active nanoparticles such as TiO_2 and Si. The TiO_2-based electrodes achieved a capacity of 76 mAhg-1, while the silicon nanoparticle-based electrodes had a capacity of about 2.25 mAhcm-2 [6]. Bao and colleagues prepared phytic acid crosslinked PANI hydrogels by thermal annealing and used these frameworks to fabricate flexible electrodes for micro patterned super capacitors. The relatively small size of the carbon particles provides features such as scalability and high processing flexibility [7]. The connected fibers provide mechanical flexibility, which leads to a large mass increase.

Researchers have developed a new type of hydrogel that can be used in biosensors, high-performance energy storage devices and electrodes. Researchers have developed a new type of hydrogel that can be used in biosensors, high-performance energy storage devices and electrodes. According to ISNA Technology Service, for this work, Stanford University researchers developed a conductive polymer based on porous nanostructures that has suitable electronic and electrochemical properties. Lijia Pen and Guihao Yu, researchers of the project, say: "The material we have developed can combine the advantages of hydrogels with organic conductive materials. This will achieve good electrochemical and conductive properties [8].

This will allow us to produce high-performance electrochemical devices such as super capacitors and highly sensitive biosensors." Hydrogels are three-dimensional polymer networks that hold large amounts of water. These materials have a structure very similar to living tissues. Most hydrogels are made of non-conductive polymer materials. Therefore, their applications in the electronics industry are very limited. In this project, phytic acid was used, which is a very good ionic conductor. Using this material, it is possible to create communication bridges in polyaniline, which in a way strengthens the polymer and overcomes the problem of non-conductivity. To make this material, two solutions are combined. The first solution starts the polymerization process, and the second solution contains aniline monomers and the phytic acid reinforce [9].

The gelation process takes three minutes, which is due to the presence of six phosphorus groups in each phytic acid molecule. These groups react with multiple polymer chains at once. "We have shown that we can mass-produce conductive polymer hydrogels and then pattern them using a printer and spray," says Penn. "This means we may be able to mass-produce electronic components such as biosensors and microsupercapacitors." The hydrogel has a specific capacitance of about 480 F/g and a high charge/discharge rate. This means that this material could be used for data storage and electric vehicles. To better understand the high specific capacitance of this hydrogel, it is worth remembering that the specific capacitance of carbon is 100 F/g. The researchers used this hydrogel to make glucose sensors. The results showed that the response time of this sensor is 0.3 seconds, while in conventional glucose sensors this time is 20 seconds [10].

He and his colleagues synthesized a 3D carbon network of a PANI gel by modifying and completely sealing PANI inside NaCl using crystallization. This modified method allows the formation of porous structures with various pore sizes from micro to meso and large scale. These pores provide a high density of ORR active sites located along a mass transfer path, which results in high conductivity, high ORR activity and high stability in acidic electrolytes.

Dai and his colleagues synthesized a N, P doped carbon framework of a PANI hydrogel with phytic acid linkers and its catalytic compounds and demonstrated dual ORR and OER catalytic activity. The minimum and maximum potentials of N- and P-doped carbon frameworks for ORR and OER were reported to be V 0.44 and V 0.39, respectively, which are lower than their theoretical values. Yu and co-workers fabricated ternary 3D nanostructured electrodes made of PPy-Si-CNTs with a reversible capacity of 1600 mAhg-1 and a storage capacity of more than 85% after 1000 cycles [11].

The use of carbon nanotubes greatly enhances the electron transport/conductivity capability of the PPy framework by improving both the physical and electromagnetic coupling between the silicon nanoparticles and the conductive polymer gel framework. The experimental results showed good electrochemical performance of these frameworks, with high capacities of 225 and 162 Fg-1 at current densities of 0.5 and 50 Ag-1, and 95% of the capacitance was retained after 10,000 cycles at 5 Ag-1. The group also used porous graphitic carbon for lithium-sulfur batteries. In these experiments, a high specific capacity was achieved, which was attributed to the hierarchical pore structure and N and O doping, which led to strong LixS interactions, through which it could control the formation of lithium sulfide species while preventing excessive material consumption. In addition to lithium-ion batteries, Dai and his colleagues fabricated zinc-air batteries using porous graphitic carbon as the air cathode. The initial two-electrode zinc-air batteries showed an open circuit potential of about 1.48 V, a current density of about 70 mAcm-2, and a peak power density of about 55 mWcm-2, indicating good catalytic activity of the electro catalyst [12].

Kim and colleagues prepared a magnetite/polyorganophosphazene temperature-sensitive nanocomposite hydrogel and studied its potential as a contrast agent in magnetic resonance imaging. To achieve the contrast agent, magnetic cobalt ferrite ($CoFe_2O_4$) nanoparticles with a hydrophobic surface were attached to the initial hydrogel through hydrophobic interactions between the hydrophobic surface of the nanoparticle and the L-isoleucine ethyl ester groups of the polymer. The prepared magnetic hydrogel, while maintaining the properties of the pure initial hydrogel such as temperature sensitivity, biocompatibility, biodegradability, injection into the body, and cell-gel phase transition within the body temperature range, exhibited two important properties for use as a contrast agent, namely very low toxicity to the body and sufficient magnetization for use as a contrast agent, magnetic imaging. The synthesized magnetic hydrogel was injected into the mouse brain and showed potential for use as a contrast agent for about 4-5 weeks.

In another study, Ang and colleagues prepared a magnetic hydrogel based on magnetite nanoparticles and N-isopropyl acrylamide and studied its potential for use in the process of thermotherapy as well as drug release. In this study, the study was carried out at a fixed frequency of 375 kHz, but the strength of the external magnetic field was varied in the range of 1.7-2.5 kA/m. The experimental results showed that the highest heat was obtained in a system with a high percentage of magnetite and for the highest strength of the external magnetic field [13].

Sakhvatpour and colleagues prepared a porous magnetic hydrogel based on starch grafted to polyvinyl imidazole/polyvinyl alcohol and used it to remove crystal violet and Congo red dyes as well as the divalent metals lead, copper and cadmium. The results showed that the designed adsorbent has a high adsorption capacity for the removal of the aforementioned metals and dyes. The dye removal process for this carrier followed second-order kinetics and the results were consistent with the Langmuir isotherm. The reusability of the adsorbents after the adsorption process is one of the most important criteria for the desirability of the adsorbent for environmental and economic reasons. Therefore, the adsorption/desorption cycle was investigated to investigate the potential of the prepared granular hydrogel. In general, the higher the number of adsorption/desorption cycles of the adsorbent, the greater the possibility of reusing the adsorbent, which is better from an environmental and economic perspective. The adsorbent proposed in the work of Sakhvatpour and his colleagues showed its efficiency for four consecutive adsorption/desorption cycles [14].

Changes in soil chemical properties

Silberbusch, Adar and Demalach (1992) used Agrosec hydrogel in field corn cultivation and stated that this material causes the release of sodium into the soil. Experiments have shown that plants that are resistant to salt respond better to these materials. They attributed the release of sodium by these materials to the presence of this element in the hydrogel structure [14]. This is because the polymerization of this material occurs at high acidity and in the presence of a strong acid. Therefore, it must be neutralized with a neutralizing agent, which is used to achieve this purpose. It is recommended to use potash instead of sodium. Over time, the amount of sodium released by this substance is reduced.

In a study by Bowman and Owens in 1991, the application of calcium in the form of calcium nitrate significantly reduced the amount of water that polyacrylamide hydrogels could retain. The hydrophobicity of this hydrogel mixed with calcium nitrate was reduced by the successive addition of potassium. Hydrogels have a number of k-coo+ groups that may behave as salts and increase their water absorption. It is possible that the multivalent cations actively displace and replace water molecules at charged sites around and within the polymer. In another study, all three types of hydrogels mentioned showed a decrease in water absorption, but SCP was less affected by the addition of fertilizer compounds. However, SCPs could not absorb as much water as the polyacrylamide hydrogel group. Of the three hydrogel groups, the PAM group was found to have the greatest buffering capacity in soil. Therefore, they are significantly abler to retain water under different conditions. In some cases, an element such as nitrogen was retained by one formulation of the hydrogel but not by another. When using these materials in tomato culture, it was observed that ammonium CNH^{4+} retained in the soil surface increased compared to the control, but when nitrate was added to the hydrogel, the amount of nitrate leached was the same in all treatments. The difference in response to the two formulations of nitrogen could be related to the difference in charge of the ion, and thus the difference in solubility. Peterson (2002) [15] cited Henderson and Hensley as stating that polymers retain more cations when dehydrated and less water when rehydrated. Most hydrogels have a high ph. High pH reduces the growth of some plants. The retention of nitrate and ammonium in tomato culture media increases with increasing hydrogel application. Nitrogen concentration also increases in tomato leaves compared to the control. The use of hydrogels in coarse sandy soils will give the best results compared to other soils, which could be due to the lower cation holding capacity of these soils compared to other soils. In general, PAM hydrogel has the greatest potential for wide application and has a high ability to play its role in saline conditions [16].

Henderson and Hensley (1987) observed that the germination of acacia and coffee tree seeds with hydrogel coating showed a negative response. The reduction in germination was related to the lack of oxygen transfer between the soil and the seed. In faba bean, when the seeds were covered with hydrogel, the germination rate decreased. A certain amount of hydrogel increases the germination rate, while increasing the amount of hydrogel stops germination [17].

Woodhouse and Johnson (1991) used moisture-absorbing materials in sandy soils and limited irrigation conditions in lettuce and barley, and observed that it increased the interval between field capacity and wilting, and reduced the frequency of irrigation and the total amount of water used [18].

Hydrogels are commonly used for the establishment of moisture-loving plants and are most beneficial under drought conditions (Patterson, 2002). Mixtures of hydrogels with sand in tomato, lettuce, radish and wheat increased dry weight and time to wilting. When hydrogel was added to the soil in two plants, Azalia and Scaklete, it increased plant growth and survival. Drought-sensitive annuals such as Petunia responded well to drought conditions, increasing the number of flowers and their dry weight. The response of moisture-loving plants to hydrogels has sometimes been negative in the field or in containers [19]. The addition of hydrogels to the medium of European birch reduced plant mass and available water. After investigating the lack of available water, it was attributed to the amount of dissolved salts in the medium, which was explained by the fact that cations were substituted for water molecules in the polymer [20]. Austin and Bandari (1992) observed that after 11 years of research, hydrogels did not show any increase in blueberry growth and in some cases inhibited the growth of young plants, which was explained by the presence of high salt content in the soil. Adding hydrogel to the soil of drought-tolerant perennials such as Vinca did not show any good response to increased soil water. Hydrogels in soil reduce the effects of salts. Increased salt concentrations can cause poor growth and malformed leaves in horticultural plants. In one study, adding hydrogel to a culture medium with different salt concentrations increased the viability of plants [21]. The hydrophilic polymer may reduce the temperature of soil aggregates. The use of hydrogels can keep roots cooler during hot summer days. Often, the amounts recommended by commercial companies are not sufficient for the plant to respond and more of this substance is needed [22].

In a study conducted by Silberbusch, Adar, and Demalach in 1993, they found that the use of hydrogels increased corn yield and that the increase in plant growth was directly related to the increase in the amount of hydrogel used. They stated that when using hydrogels, the type of irrigation system should also be considered. They used a drip irrigation system for corn cultivation and observed that more water was stored around the outlets of the irrigation system and that points further away from the outlets benefited little from water intake. As a result, plant growth at these points was lower than at the points where the outlets were located. This problem can be solved by changing the drip irrigation system to a sprinkler irrigation system [23].

Mohammadinejad et al. (2018) successfully produced a superabsorbent hydrogel composite containing poly (acrylamide-co-itaconic acid) and multi-walled carbon nanotubes (MWCNT). The stability of the hydrogel increased with increasing proportion of multi-walled carbon nanotubes, which may be attributed to the hydrophobic nature of multi-walled carbon nanotubes as well as the increase in cross-linking density [24]. The water retention capacity of the hydrogel also increased in the presence of multi-walled carbon nanotubes (10 wt%). In this study, the effects of ultrasound were evaluated in terms of uniform distribution of carbon nanotubes on the surface of the superior polymer. The multi-walled carbon nanotubes were intact without any disruption in the polymer structure. In addition, the strength of the obtained nanocomposite was increased and its water retention capacity and adsorption of other soluble materials such as lead(II) were increased [25]. Ultrasound dispersed multi-walled carbon nanotubes as an excellent filler in the polymer chains under elevated temperature. The researchers conclude that these reaction conditions cannot be achieved by conventional methods and homogeneity and good dispersion of particles in the host cannot be achieved. In addition, ultra-sonication dissociates nanoparticles into single particles, while stirring cannot do this. Another mechanism of size reduction is the effect of strong acoustic waves on secondary bonds such as hydrogen bonds, which breaks the H-bonds of the particles, subsequently separating the aggregated particles and increasing the number of free radical groups such as -OH and Thus, this important event makes ultra-sonication a superior method to other methods such as magnetic stirring used in the literature [26].

Researchers have produced a new type of hydrogel that can be used in biosensors, high-performance energy storage devices and electrodes. To do this, researchers at Stanford University have developed a conductive polymer based on porous nanostructures that has suitable electronic and electrochemical properties [27].

Another study conducted at Kansas State University in 1997 examined the effects of environmental conditions on polyacrylamides. The final results showed that the properties of this type of polymer can lead to the release of acrylamide under certain conditions.

In the field of polymer products, researchers at the end of the 20th century realized that synthetic-natural and modified natural hybrid polymer materials, which are biocompatible, are gradually replacing synthetic types. In this regard, superabsorbents based on natural polymers such as cellulose and starch or based on modified natural polymers such as carboxymethyl cellulose, etc. were prepared. In many studies conducted by researchers in the field of preparing hybrid hydrogels, polymer synthesis and grafting operations were carried out simultaneously. Although the obtained hydrogels have acceptable properties, this method has some disadvantages:

- ✓ First, the homopolymerization reaction of the added monomers can be carried out in competition with the copolymerization reaction and causes the production of a significant amount of by-products (Table 1).
- ✓ Second, natural polymers in water condense the system, which can cause process limitations [28].

Table 2. Comparative Table: Research Background on Polymerization Techniques for Hydrogels

Polymerization Technique	Materials Used	Key Findings	Limitations / Notes
Free-radical polymerization	PEG, PVA	Basic mechanisms and biomedical applications	Lacks details on crosslinking diversity
Photo polymerization	Acrylates, PEGDA	High control over structure and shape	UV sensitivity may harm biomolecules
Michael addition	PEG-SH, PEG-VS	Biocompatible hydrogels for tissue engineering	Limited mechanical strength

Enzymatic crosslinking	Gelatin, HRP, H2O2	Mild conditions suitable for cell encapsulation	Slower gelation compared to UV methods
Ionic gelation	Alginate, $CaCl_2$	Environmentally friendly; no need for toxic initiators	Limited mechanical stability
Click chemistry (azide-alkyne)	PEG, HA derivatives	Bio-orthogonal and efficient crosslinking	Requires synthetic modification
Photopolymerization	PEGDA, ECM proteins	Enables patterning and tunable stiffness	Needs photo initiators and light exposure
Self-assembly	Peptides, amphiphilic molecules	Forms Nano fibrous hydrogel scaffolds	Less mechanically robust than covalent gels
Thiol-ene polymerization	PEG-norbornene, thiols	Cytocompatible and orthogonal chemistry	Requires specific monomers
Reversible Diels-Alder	Furan-maleimide systems	Thermo-reversible hydrogels with recyclability	Limited to certain temp. ranges
RAFT polymerization	Acrylamide-based polymers	Controlled molecular weight and structure	Requires RAFT agents and initiators
Dynamic covalent polymerization	Hydrazone, boronate esters	Adaptable, stimuli-responsive gels	Slower equilibrium kinetics
Non-covalent host-guest polymerization	Cyclodextrin, adamantane	Shear-thinning and injectable hydrogels	Sensitive to environment (pH, salt)
Shear-thinning hydrogel (self-healing)	Alginate, synthetic polymers	Used for 3D bioprinting, rapid recovery	Lower long-term stability
Dual-crosslinked systems	Chitosan, genipin, aldehydes	Improved toughness and tunable degradation	Complex synthesis route

In addition to this simultaneous system, we can use an asynchronous process, so that the polymerization reaction is carried out separately and then the grafting operation is performed on the natural polymer. In the innovative method used in this invention, unlike previous methods, the polymerization reaction between the modifying monomers is carried out in the absence of the natural polymer and through inverse emulsion polymerization as a method capable of producing polymers with high molecular weight and at high polymerization rates. By performing surface crosslinking and creating higher network density at the level of mechanical hardness of the outer layers of the particle, it is improved. Since the surface layer is formed separately on each particle, the hydrogel particles after preparation must be crushed and cut to the desired size, and then any two or more agent material that has the ability to react with polymer chains can be used as a crosslinking agent [29]. In this study, the crosslinker {3-(2,3-epoxypropoxy) propyl} trimethoxysilane (GPS) was used as the surface crosslinking agent. The properties of the hydrogel after production were investigated by tests such as swelling measurements, rheological measurements, infrared spectroscopy, electron microscopy and thermogravimetric analysis. All these tests confirm the production of hybrid hydrogels with 70% natural component and 30% synthetic component and the improvement of swelling properties under load from 6.5 g/g in the unmodified hybrid hydrogel sample to 17 g/g in the modified hybrid hydrogel sample. Haas et al. presented a comprehensive review of the electrochemical processes and material design of conducting polymers for use as electrodes. However, conducting polymers in reduced states usually have poor stability and conductivity, and these weaknesses seriously hinder their wider application. In addition to their use as electrode materials in lithium-ion batteries, conductive polymers can also be used as binder materials. To maximize energy utilization, each active particle in the battery must have a specific shape and size and be connected to a current collector and a solid or liquid electrolyte. However, the traditional binder synthesis system is a binary combination in which the conductive phase is randomly distributed and narrow paths or weak contacts prevent effective access to some parts of the battery. Unlike the conventional binder system, conductive polymers can have dual functionality due to the synergistic properties of conventional polymers and organic conductors. As a result, the possible accumulation of conductive additives and separation of the polymer layer during cycling are avoided. Most importantly, the chemical structure and physical/chemical properties of conductive polymers can be tuned by modifying the polymer chains [30].

Cieddo and colleagues prepared polyacrylamide/magnetite hydrogels by in situ precipitation and then used them to investigate their adsorption capacity of bovine serum albumin as a model protein. In this work, iron II and III ions are attached to the acrylamide hydrogel through electrostatic interactions due to the porous structure of the hydrogel and the presence of amide groups in the main chain of the hydrogel. Subsequently, magnetite-acrylamide hydrogel nanocomposites were prepared by oxidation induced by ammonium solution. This method has several advantages:

First: the introduction of a large number of mineral particles into the hydrogel network confirms that colloidal-sized particles are well dispersed in the polymer network [31].

Second: the preparation process is simple and inexpensive. However, this method is only suitable for hydrogels with a stable network. Otherwise, the hydrogel network may be destroyed by alkaline solution during the preparation process of magnetic hydrogel. In addition, the use of alkaline solution limits the application of magnetic hydrogel synthesized by this method to surround cells.

Third: Another important point is that the efficiency of Fe_3O_4 magnetic nanoparticles in hydrogels that have negatively charged ligands may be low due to the complex formation reaction of these ligands with Fe^{+2} and Fe^{+3} present in the salt solution [32].

Sattarkar and his colleagues prepared a temperature-sensitive magnetic hydrogel based on N-isopropyl acrylamide and magnetite nanoparticles and studied its ability to release model drugs methylene blue and vitamin B12 under the influence of a high-frequency alternating magnetic field. The drug release results showed that the prepared system has the potential to be used as a drug delivery agent. N-Isopropyl acrylamide is a temperature-sensitive monomer with a low critical solution temperature of 30-35$^{\circ C}$.

Samantha and colleagues prepared biocompatible polyethylene glycol and N-isopropyl acrylamide/magnetite nanocomposite hydrogels at different magnetite percentages and studied their swelling power and ability to generate heat as a result of applying an alternating magnetic field. The results of the swelling study showed that the prepared nanocomposite hydrogel has a temperature-sensitive swelling behavior, such that it shows a water-swelling behavior at high temperatures. Also, the temperature-sensitive swelling behavior of the prepared nanocomposite can be controlled by changing the strength of the external alternating magnetic field. Their results also showed that the magnetic field strength required for thermotherapy depends on the percentage of magnetite nanoparticles used to produce the nanocomposite hydrogel, and with increasing the percentage of magnetite in the magnetic hydrogel structure, a weaker magnetic field is required to generate heat during the thermotherapy process [33].

Kass et al. (2010) prepared several acrylic hydrogels by ultrasonic polymerization of water-soluble monomers and macro monomers. Ultra sonication was used to generate initiator radicals in viscous aqueous monomer solutions using glycerol, sorbitol, or glucose additives in an open system at $37^{\circ C}$. Water-soluble additives were essential for hydrogel production, and glycerol was the most effective. Hydrogels were prepared from the monomers 2-hydroxyethyl methacrylate, poly (ethylene glycol) methacrylate, dextran methacrylate, acrylic acid/ethylene glycol methacrylate, and acrylamide/bisacrylamide. The application of ultrasound using a probe ultrasonicator was an effective method for the polymerization of water-soluble vinyl monomers and the subsequent preparation of hydrogels. Ultrasonic polymerization occurs rapidly in the absence of chemical initiators [34].

Conclusion

Silk hydrogel is a type of hydrogel that can be produced by both fibroin and sericin. Sericin can be easily separated from silk in vitro. When sericin is separated from fibroin, a softer, whiter, and shinier structure is obtained. So, in general, it can be said that sericin is a useless protein. However, studies have shown that gel can also be obtained from this protein. Fibroin can be extracted as a clear and uniform solution for various applications. Fibroin solution can be converted into gel using substances such as acids, various ions, and other additives. There are various factors such as temperature and pH that are involved in the gelation process of fibroin solution. When the concentration of silk solution and also temperature increase and the pH value decreases, the gelation time decreases. The size of the gel pores can be adjusted by changing the concentration of the fibroin solution, ambient temperature, pH, and calcium ion concentration. During the gelation process, the non-crystalline regions of the protein are converted into beta sheets. What causes gel formation in silk fibroin solution is the formation of intramolecular and extra molecular hydrogen bonds. To increase the properties of the fibroin solution hydrogel, other biopolymers such as chitosan and gelatin can be added to it. Such composite scaffolds have appropriate intramolecular connections and permeability. Porous hydrogel polymers are known as new advanced materials due to their unique properties in various scientific and industrial fields. These materials are formed as three-dimensional polymer networks that are capable of absorbing and retaining water or other liquids. Generally, due to their porous structure, these hydrogels can absorb large volumes of liquids and are used as a suitable environment for cell culture, drug delivery, and medical applications such as wound dressings and tissue prostheses. In addition, these materials are widely used in different industries such as food, cosmetics, and pharmaceuticals due to their improved mechanical properties and the ability to be designed to meet specific needs. These unique properties of this group of materials have attracted the attention of many researchers around the world. In the latest scientific projects registered by the researchers, a porous hydrogel copolymer of N-vinylpyrrolidone (NVP) and 2-hydroxyethyl methacrylate (HEMA) has been prepared by the high internal phase emulsion method. The research group used methylene bisacrylamide as a crosslinking agent and ammonium persulfate and tetra methylene diamine as initiators to synthesize this copolymer. In order to optimize the process, the water absorption of the synthesized copolymers was measured and the best value was recorded at each stage. The highest water absorption of the porous hydrogel synthesized in this method was 8.9 g/1 g of hydrogel with an average pore size of 5.2 μm, while the hydrogel with the same amounts of reactants and without the aforementioned method was 1.2 g/1 g of hydrogel. This research showed that using High internal phase Emulsion polymerization, copolymers can be prepared that are very suitable for increasing water absorption and can be a suitable alternative to super absorbents.

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Authors' Contributions

All authors contributed to data analysis, drafting, and revising of the paper and agreed to be responsible for all the aspects of this work.

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