



Review

Potential of mucilage-based hydrogel for passive cooling technology: Mucilage extraction techniques and elucidation of thermal, mechanical and physiochemical properties of mucilage-based hydrogel

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Abstract: Current air-conditioning and refrigeration systems utilize active cooling technology, which consumes a lot of energy from fossil fuels, thereby increasing global warming and depletion of the ozone layer. Passive cooling is considered an alternative to active cooling because it is effective and less expensive and does not require the use of electricity, so cooling can be achieved in locations where there is no electricity. Hydrogels are flexible and soft 3-dimensional networks with high water content and evaporative and radiative cooling properties that make them suitable for use in passive cooling technology. Natural hydrogels are considered alternatives to synthetic hydrogels because they are biodegradable, biocompatible, sensitive to external environments and mostly sourced from plant-based sources. There are limited studies on the application of mucilage-based hydrogel for passive cooling, despite its excellent thermal, mechanical and physiochemical properties. Therefore, this study evaluates the properties of mucilage-based hydrogel as a plausible alternative to synthetic hydrogel for passive cooling. The possibility of using mucilage-based hydrogel in passive cooling technology depends on the mucilage biomass feedstock, mucilage extraction techniques, polymerization techniques and additives introduced into the hydrogel matrix. Different mucilage extraction techniques; mucilage percentage yield; the effects of crosslinkers, polymers and nanoparticle additives on the properties of mucilage-based hydrogel; and the potential of using mucilage-based hydrogel for passive cooling technology are examined in this review.

Keywords: mucilage; natural hydrogel; extraction technique; properties; passive cooling

1. Introduction

Current air-conditioning and refrigeration systems utilize active cooling technology, which consumes a lot of energy from fossil fuels, thereby increasing the amount of greenhouse gases (GHG) emitted into the environment. At the Montreal Protocol conference, the hydrochlorofluorocarbons (HCFCs) and chlorofluorocarbons (CFCs) refrigerants commonly used in these cooling systems were phased out due to their impact on ozone layer depletion [1]. Some locations in developing countries do not have access to the national electricity grid, while others experience load shedding. With these challenges, drugs, blood, vaccines and food are difficult to preserve using active cooling and temperature-controlled storage space. Electronic components dissipate heat when under load. Conventional active cooling technologies (water cooling and forced air circulation) are currently used for the removal of heat and cooling of these components. The high energy consumption, considerable weight, complicated structure and high carbon footprint of these cooling technologies limit their application in wearable and portable devices. Therefore, the need for alternative cooling systems (technology) that address the high energy consumption, depletion of the ozone layer, weight of appliances, comfort of humans and preservation of food and drugs becomes eminent. Passive cooling is considered an alternative to active cooling, as shown in Figure 1, because it is effective and less expensive, and it does not require the use of electricity or a refrigeration system. Insulated boxes have been used over the years to maintain the cold temperature of farm produce, food and drugs during transportation, but the cold temperature cannot be maintained at high temperatures over a long distance. Evaporative and radiative cooling can be adopted in passive cooling technology to reduce reliance on active cooling technologies. Radiative cooling involves the dissipation of heat through infrared radiation into cold space, while evaporative cooling is water-based and involves the dissipation of a high quantity of heat caused by the high enthalpy of vaporization of water. The use of hydrogel in passive cooling technology has been proposed as an alternative to active cooling technology to address energy challenges in developing countries.

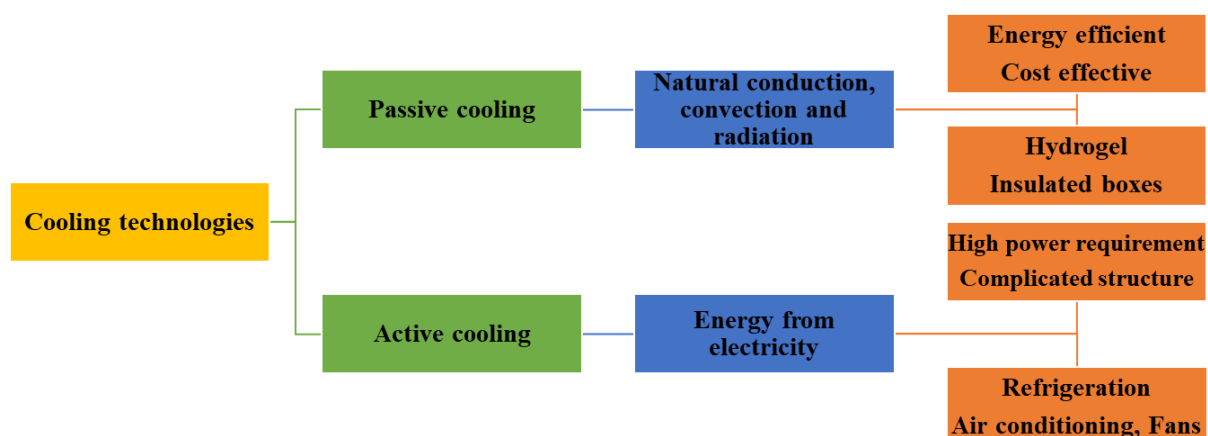


Figure 1. Cooling technologies for energy systems and electrical components.

Hydrogels are flexible and soft 3-dimensional networks of insoluble polymer chains that are cross-linked. They have high water content and usually freeze and dry at low and high temperatures, respectively [2]. Hydrogels are formed by chemical and physical crosslinking of natural and synthetic

polymers, and they have different properties due to their source, chemical composition, hydrophobicity and crosslinking density. Hydrogels are commonly prepared from polar monomers, and they are classified as synthetic, natural or a combination of the two classes (hybrid). Synthetic hydrogels are usually a 3-dimensional network of hydrophilic homopolymer or copolymer cross-linked polymer, while natural hydrogels are from natural sources, which gives them excellent biodegradability and biocompatibility properties. Hydrogels can absorb water because of the presence of $-OH$, $-COOH$, $-NH_2$ hydrophilic functional groups and other polar groups in the polymer, while their inability to be soluble in water is attributed to the cross-linking network structure [3]. The hydrogel water content is governed by the density of networking and the polymer's properties [4]. The unique properties of hydrogels make them useful in biomedical, cooling, electronic, tissue engineering and sensing applications.

Synthetic hydrogel is chemically stronger; has higher gel strength, water absorption capacity and service life; and is hydrophobic in nature when compared with natural hydrogel. However, natural hydrogel is gradually replacing synthetic hydrogel because it is biodegradable, biocompatible and sustainable. Several production techniques have been developed to improve the polymeric properties of natural hydrogel, thereby making them acceptable, sustainable and competitive with synthetic hydrogel. Nanotechnology is an emerging technology with various applications in material science, energy systems, medicine, drug delivery and biology. This technology has been used to enhance the properties of natural hydrogels through the incorporation of nano- and micro-sized particles into the matrix of the hydrogel, thereby improving the mechanical properties and potentiating the desired solar reflectance needed to enhance radiative cooling.

Plant-based biopolymers like glucans, gums, cellulose and mucilage are commonly used to synthesize hydrogels used for drug delivery, medicine and tissue engineering; but the application of mucilage-based hydrogels for passive cooling, despite their excellent thermal, mechanical and physiochemical properties, has not been fully exploited by researchers. The various techniques used in the mucilage extraction from various plants' parts, the mucilage percentage yield and the effects of crosslinkers, polymers and nanoparticle additives on the thermal, mechanical and physiochemical properties of mucilage-based hydrogel, with the aim of using mucilage-based hydrogel as a plausible candidate for passive cooling, are reviewed in this paper. The performance of synthetic hydrogel in different applications was also considered to obtain a clear understanding of hydrogel performance in passive cooling technology, since there are limited studies on the direct application of natural hydrogel in passive cooling technology.

2. Classification of natural polymers

Recently, plant-based natural polymers have gained the attention of researchers because of their acceptable biocompatibility and chemical and mechanical properties. These hydrogels can be classified as conventional or smart hydrogels. Smart hydrogels are sensitive to external stimuli like temperature, enzymes, sound, pressure, antigens, solvent composition, ionic strength and pH. Comparing natural hydrogel with synthetic hydrogel, natural hydrogels are less expensive, biodegradable, biocompatible and sensitive to the external environment, while synthetic hydrogels are chemically stronger, possess controllable properties, have a high water absorption capacity and have a longer service life. The use of agricultural feedstock in the production of hydrogel will increase the use of sustainable and environmentally friendly (green) materials that are not hazardous to food, humans and the environment.

Research on natural polymers is growing rapidly to develop advanced materials such as hydrogels, membranes, films and coatings that are alternatives to their synthetic polymer counterparts and have the potential to neutralize the negative effects caused by the use of synthetic polymers on the environment. Natural hydrogels are mostly synthesized from natural polymers obtained from agricultural sources such as polysaccharides, proteins, animals and microorganisms. Natural polymers from polysaccharides can be classified as alginate, guar gum, inulin, lignin, starch (amylopectin and amylose) and cellulose, while those from proteins are classified as collagen and gelatin. Hyaluronic acid, xanthan gums, chitosan and chitin are the different categories of polymers from animals [5], as represented in Figure 2.

Chitosan is a natural linear polysaccharide found in the walls of certain fungi and animal shells (chitin), which are exoskeletons of crustaceans such as crabs, lobsters and shrimp. Chitosan is readily available and abundant in nature. It is mostly obtained through the deacetylation of chitins with enzymes [6]. Liu et al. [7] prepared hydrogel from chitin through chemical transformation. The hydrogel was prepared through the acetylation of the chitosan precursor with the assistance of agarose. The hydrogel formed contained over 95% water, but it suffers from some drawbacks like low stability in an acidic medium, low mechanical strength and low thermal stability.

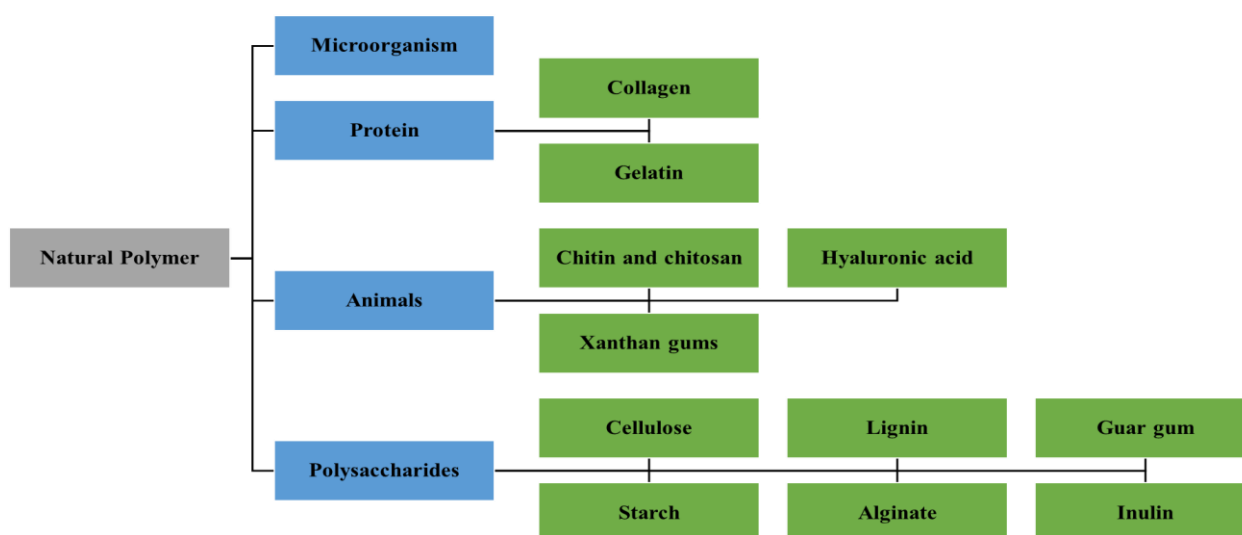


Figure 2. Classification of natural polymers.

Protein-based hydrogels consist of amino acids that are connected to each other through peptide bonds using the condensation reaction. Gelatin is a natural polymer that is obtained when collagen from animal sources such as fish, bovine and porcine skin is processed. The physical and mechanical properties of the gelatin polymer are affected by the source of the animal and the collagen treatment process used in the polymer preparation [8]. Protein-based hydrogels are usually categorized as smart hydrogels because of the solution-gel transition that occurs as the environment changes. Polysaccharides are widely distributed in nature, and they can be found in plants and animals. Mucilage is a type of polysaccharide that is found in different parts of the plant, and it is commonly used in different food industries due to its broad-spectrum application. The various monosaccharide units that

make up the mucilage give it the ability to retain water and release it when it is needed by the plant parts. This property makes mucilage a viable feedstock for the synthesis of natural hydrogel.

3. Polysaccharide-based hydrogel from mucilage

Mucilages are complex polysaccharides that contain a molecular structure of about 30,000 monosaccharides and dietary fiber and are soluble in water [9]. They are found in the roots, stems, fruits, leaves and bark of plants. Some of the plants that contain mucilage are chai seed, flaxseed, basil, psyllium, camelina seeds, cactus, quine seed, aloe vera and others. The polysaccharide comprises uronic acid and monosaccharides linked with bioactives, glycoproteins, and glycosidic bonds that usually contain l-arabinose, methyl glucuronic acid, d-xylose, galacturonic acid, d-mannose, d-glucose and glucuronic acid residues [10,11]. The presence of mucilage in leaves assists in retaining water and food during water deficiencies in the soil. The gel-forming capacity and water affinity of mucilage are determined by the protein and fiber contents, while the lipid content determines the oil-holding capacity [12]. The mucilage feedstocks influence the conformation of the polysaccharide chain, type of glycosidic bond and monosaccharide units [13]. Mucilage has various functional properties, like high film-forming ability, viscosity-modifying properties, emulsifying gelling and high water absorption capacity [14,15]. These mucilage properties make them suitable for hydrogel production, additives in food, bioplastics for packages, edible coatings and film [12,16,17]. The plant species and parts, climatic and environmental conditions, genetic modification and extraction process affect the chemical composition of mucilage [18,19]. The purity of the mucilage is determined by the quantity of protein in the mucilaginous extract and extreme processing conditions decrease the purity of the mucilage [20,21].

3.1. Mucilage extraction techniques

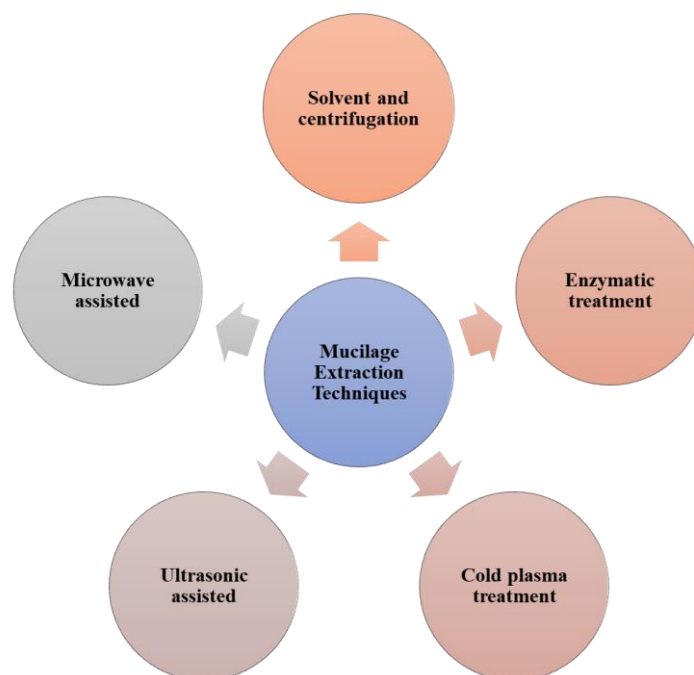


Figure 3. Mucilage extraction techniques.

The extraction techniques used in the production of mucilage affect the percentage yield, properties and quality of the mucilage. The states of the extraction processes, such as extraction time, temperature, solvent to seed ratio and pH, are essential to the optimization of the mucilage yield. Figure 3 gives a summary of the different mucilage extraction techniques discussed in this paper, while Table 1 gives a summary of the different mucilage extraction techniques used for different plants and extraction variables for optimum percentage yield.

3.1.1. Conventional (solvent) and centrifugation method

Due to the water-soluble properties of plants' seeds, stems and roots, mucilage can be extracted using distilled water and alcohol. Mechanical crushing or hulling is required if the presence of the seed's pod makes the mucilage extraction difficult. In the solvent method, the plant's parts are soaked in distilled water and stirred continuously. The solution is then filtered, and alcohol is added to the filtrate to precipitate the mucilage [22]. Wild leaves of *Talinum triangulare* (Jacq.), *Amaranthus spinosus* Linn. and *Abelmoschus manihot* Linn. were used to produce mucilage by Ang and Raman [23]. Purified water was used to wash the leaves, which were then air-dried and ground. The powdered leaves were soaked in distilled water for 24 h at 60 °C with continuous stirring. Using the solvent method, ethanol was added to the supernatant to precipitate the mucilage. The mucilage was washed with acetone and dried. The percentage yields obtained were 5.30%, 5.85% and 8.76% for *Talinum triangulare* (Jacq.), *Amaranthus spinosus* Linn. and *Abelmoschus manihot* Linn. Leaves, respectively. *Dioscorea opposita* Thunb. (tuber) was used in the preparation of mucilage using centrifugation and solvent methods [24]. The paste was centrifuged for 5 min at 4000 rpm, and ethanol of 75% concentration was added to the supernatant to precipitate the mucilage. The yield of the mucilage was approximately 5.71%, which was lower than the expected yield of 8.08%. Nazir et. al [25] used the surface response method to optimize the mucilage yield from basil seed. The temperature, extraction time and ratio of water to seed varied in the optimization process. The mucilage yield was observed to increase as the temperature increased. The optimum yield of 20.5 g/100 g was obtained at a temperature of 56.7 °C, a water to seed ratio of 66.84:1 and an extraction time of 156 min. Figure 4 shows the schematic method used in the extraction of mucilage from taro using low-temperature methods such as cold-water extraction, ethanol precipitation and centrifugation [26]. Estévez et al. [27] investigated the influence of solvent pH value on the mucilage yield from mesquite seed gum using alkaline and acid solvents of 0.2 and 0.4 w/v, respectively, and a temperature range of 10–60 °C. Mucilage yields of 24.9% and 17.7% were obtained for alkaline and acid extraction, respectively. The protein content in the acid extraction was low due to hydrolysis. The pH value of the solvent did not affect the viscosities of the mucilage dispersions. The difference between the mucilage yields of dehydrated and fresh *O. ficus-indica* cladodes was studied by Quintero-García et al. [28] using the extraction and centrifugation methods. The percentage yield of the dehydrated cladodes (15.69% ± 0.04 g/100 g) was higher than the fresh ones (13.06% ± 0.19 g/100 g). The cladodes' maturity stage was reported to affect the percentage yield. The young cladodes produce more mucilage because of their higher soluble content fiber, and as the cladodes mature, the insoluble fiber in them increases, thereby reducing the mucilage yield. Rodríguez-González et al. [29] adopted different methods for the extraction of mucilage from six (6) wild *Opuntia* species with the aim of preserving the structure of the mucilage. The fresh cladodes were blended with varying proportions of water (1:8 and 1:2 w/v). The suspensions were centrifuged at 3500 rpm for 15 min, after which ethanol was added to them, and the mucilage

mass was separated using the centrifugation technique. The other method involves mixing the fresh cladodes with ethanol in a ratio of 1:1 without using water. The suspension was maintained at 22 °C and centrifuged at 3500 rpm for 15 min to extract the mucilage. The chemical compositions of mucilage and the percentage yields varied in all species. The quantity and quality of mucilage extracted were influenced by the solvent (type, concentration) and extraction conditions (process time, temperature, pH) [30,31]. Ethanol is the preferred solvent used in the extraction of mucilage because it is sustainable, and its danger to humans and the environment is relatively low [32]. The addition of heat is not required in the solvent method, but it is expensive (high cost of solvent) and time-consuming, while the mucilage yield is usually low. The centrifugation method allows the separation of impurities in the mucilage [33,34].

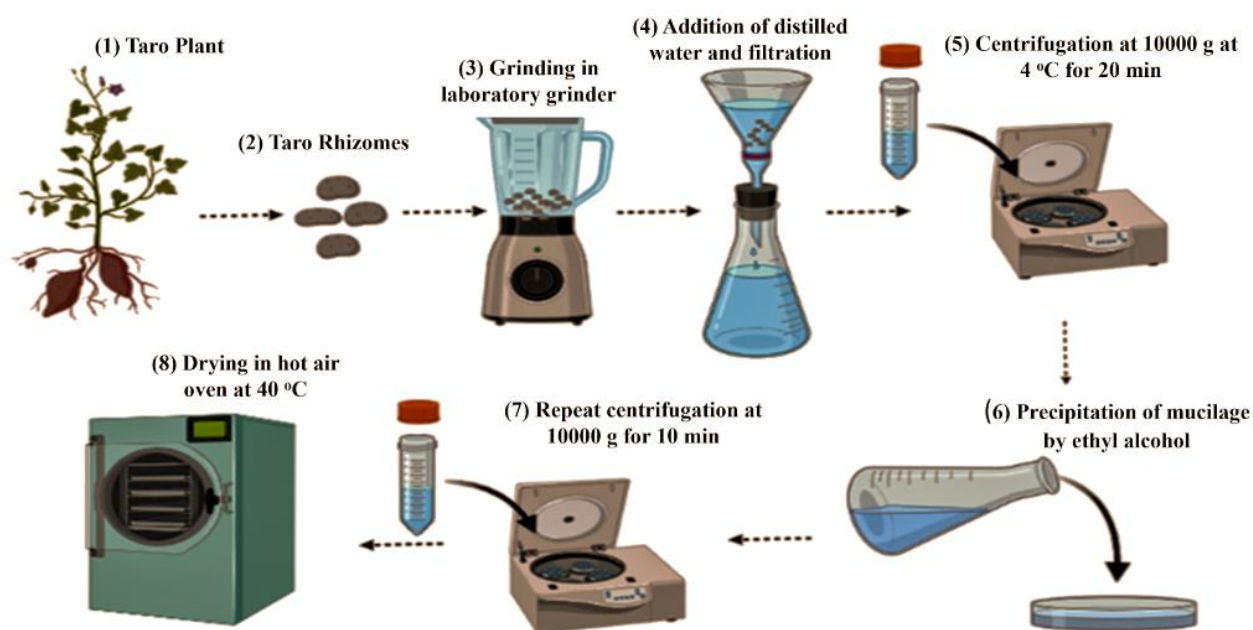


Figure 4. Schematic representation diagram of the extraction of taro mucilage at low temperature with ethanol precipitation (Reproduced from Ref. [26] with permission).

3.1.2. Microwave-assisted extraction method

The microwave-assisted extraction method (Figure 5) is a modern extraction method that is used to overcome some of the challenges faced by the conventional extraction technique [35]. In this method, the energy from the microwave penetrates the plant part, reaching the inner vascular system, trichomes and glandular system, thereby causing the wall of the plant seed to relax and swell. The sudden rise in temperature in the plant tissues makes them open extensively, therefore increasing the contact of the solvent with the plant material rapidly. The quantity of solvent that penetrates the plant matrix accelerates the vaporization of volatile materials [36]. The microwave-assisted extraction method is known as a green technology because of its low solvent usage and extraction time. The mucilage produced from this technique has improved quality and quantity [37].

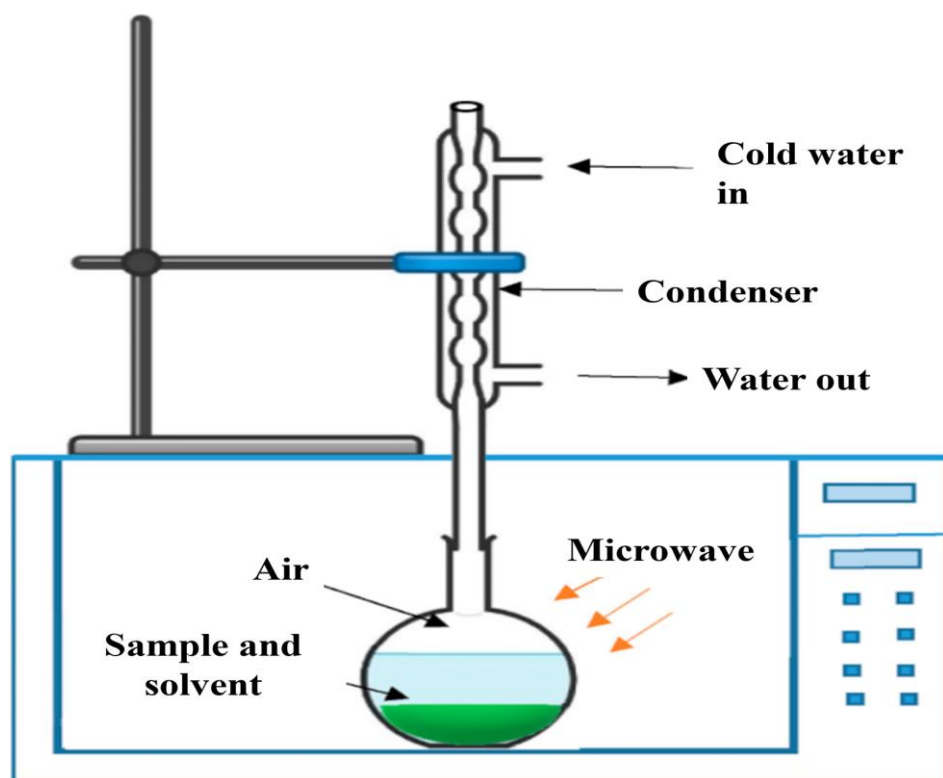


Figure 5. Microwave-assisted extraction system for lemon myrtle leaves (Reproduced from Ref. [35] with permission).

The percentages of mucilage yield using conventional and microwave-assisted extraction techniques with *Ocimum basilicum* var. *album* (L.) seed were compared by Shiehnezhad et al. [38]. The influences of ratio of sample to solvent, microwave power and extraction time on the percentage yield showed that the optimum yield of $17.00\% \pm 0.14\%$ was obtained at a microwave power of 570.32 W, an extraction time of 4.33 min and solution sample ratio of 40:1 mLg, while the conventional extraction method recorded a mucilage yield of $14.53\% \pm 0.25\%$. The mucilage yield increased between 360 and 560 W of microwave power. At higher microwave power, above 560 W, the mucilage yield is reduced due to the thermal degradation and denaturation of the polysaccharide [39]. Han et al. [40] extracted mucilage from the peel of *Opuntia dillenii* Haw. fruits and optimized the process using the response surface method. An optimum mucilage yield of $15.62\% \pm 0.37\%$ was obtained at a microwave power of 367 W, an extraction time of 150 s and a ratio of water to raw materials of 37 mLg when compared with the hot water extraction technique, which has an optimum value of $13.36\% \pm 0.71\%$. The viscosity of hot water-extracted mucilage was observed to be lower than that of microwave-assisted extraction. The influences of pH, ratio of raw material to water, microwave extraction power and time on the mucilage recovery of *Opuntia ficus-indica* cladodes was investigated by Felkai-Haddache et al. [41]. Higher microwave irradiation power enhanced the penetration of the solvent into the plant matrix, thereby increasing the mucilage extraction, but excessive exposure to high irradiation degrades the molecules of the polysaccharides. It was reported that the mucilage recovery increased rapidly with the increase in extraction time but decreased as the pH of the solvent increased. The higher water to raw material ratio increased the diffusivity of polysaccharide into the solvent, thereby increasing the mucilage recovery, while the pH value caused the dissociation of the acidic group in the polysaccharides

and repulsion among the negative charges. However, Zhao et al. [42] explained that increasing the water ratio decreased the viscosity and density of the extraction solution, thereby facilitating the dilution of the polysaccharide in the water. The optimum mucilage yield of 25.6 g was observed at a pH of 11, a water to raw material ratio of 4.83, an extraction time of 5.15 min and a microwave power of 700 W. Comparing the mucilage yield obtained from [28], it is observed that the yield from the microwave-assisted extraction method is higher than that of the conventional extraction method. Wei et al. [43] also compared the percentage yields of mucilage using the conventional method and microwave-assisted extraction from sea buckthorn berries. The water to material ratio, microwave power, extraction temperature and time were varied in the ranges of 5:1–25:1 mL/g, 500–700 W, 75–95 °C and 4–12 min, respectively. The maximum mucilage yield of $0.264\% \pm 0.005\%$ was obtained from the heat reflux extraction using distilled water at 85 °C and 6 min, while $0.207\% \pm 0.006\%$ was obtained at extraction time and temperature of 6 min and 85 °C, respectively, with a microwave power of 600 W and a liquid to material ratio of 10:1 mL/g for the microwave-assisted technique. The authors also reported that the increase in microwave power over a long period caused partial degradation of the polysaccharide, thereby reducing the yield, while the high liquid content in the mixture caused lower viscosity and density, which facilitated the dilution of polysaccharide in the water. The microwave-assisted extraction method consumes less energy and less organic solvent and has a low extraction time with improved mucilage quality and quantity when compared to the conventional extraction method [44]. A non-homogenous structure that caused low reproductivity was observed in the mucilage produced using microwave-assisted extraction [45].

3.1.3. Ultrasonication-assisted extraction

The ultrasonic method (Figure 6) is an efficient physical technique used in the rapid extraction of mucilage from plants, especially seeds. Acoustic waves are used to improve the rate of diffusion by creating a high temperature of about 4000 °C, uneven pressure and high shear stress through the collapse of cavitation bubbles [46]. The cavitation breaks down the cell wall of the seeds, thereby enhancing the interaction between the substrate and the solvent [47,48]. The ultrasonic-assisted extraction method gradually removes the seed mucilage from its inner and outer layers until the layers of the mucilage are removed from the core of the seed. The ultrasonic-assisted extraction technique decreases the extraction time with a high extraction yield. High ultrasound intensity was used in the extraction of mucilage from mutamba [49]. The ultrasonic process time and power varied. The mucilage was completely recovered at a process time of 7 min without modifying the primary structure of the mutamba seed mucilage. Tian et al. [33] evaluated the influences of water/raw material ratio, ultrasonic extraction time and power and ultrasound radiation on the yield of polysaccharide in *Angelica sinensis* using experiments and the response surface method. An optimum yield of $21.89\% \pm 0.21\%$ was reported at an ultrasonic power of 396.83 W, an ultrasonic time of 28.06 min and a water to raw material ratio of 43.31 mL/g. In the study conducted by Zhao et al. [50], mucilage from *Arabidopsis* seed was completely extracted after 20 s when the ultrasound technique was used, while it took an hour of vigorous agitation to extract a small amount of mucilage using water in the conventional method. The effects of extraction power, temperature and time on mucilage yield from barhang seeds was studied by Niknam et al. [21]. The yield increased with increasing sonication time (15 to 30 min) and temperature (20 to 40 °C). At lower ultrasonic power, the mucilage from the seed was not released, but as the power increased from 200 to 400 W, the mucilage yield increased.

However, the mucilage yield was reduced by the ultrasonic power exceeding 400 W. The authors concluded that the sonication power was more significant than the time and temperature in the extraction process. Akhtar et al. [51] considered the influences of amplitude level, water to seed ratio, extraction time and temperature and pH on the mucilage yield from flaxseed gum. The variation of the values of the extraction ultrasonic parameters affected the mucilage yield, and the optimum mucilage yield of 11.04% was obtained at 70% amplitude and 90 °C while the other parameters were at their mean values. The authors reported that the amplitude (%) and the temperature had a more predominant influence on the percentage yield of the mucilage, but in the study conducted by Souza et al. [52], the pH values and extraction time were reported to have the most significant effect on mucilage yield from psyllium. The optimum mucilage yield was obtained at a pH of 10 and an extraction time of 3 h. The percentage of mucilage yield from chia seed using the ultrasonic method was lower than that of the conventional method [53], but Zhu et al. [54] reported that the ultrasonic-assisted extraction method caused maximum transfer of mucilage to solvent. Yeh and Lai [55] used ultrasonic and enzymatic-assisted techniques to extract mucilage from *Citrus grandis* Osbeck seed and reported that the performance and structural properties of *Citrus grandis* Osbeck seed mucilage are influenced by the extraction techniques adopted.

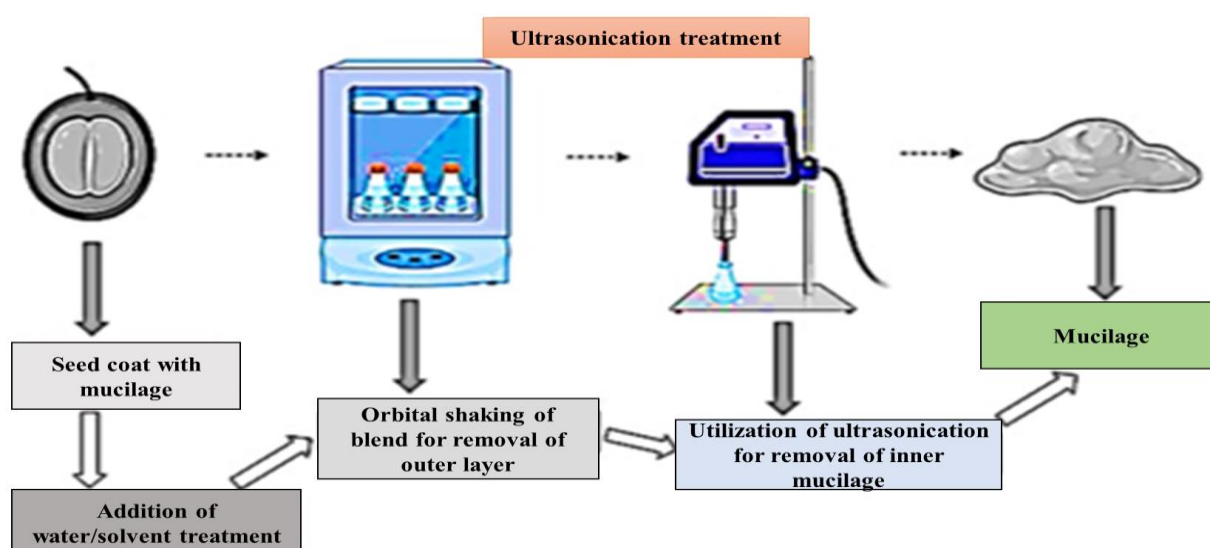


Figure 6. Ultrasonication-assisted technique (Reproduced from Ref. [9] with permission).

3.1.4. Enzymatic treatment

The efficiency of mucilage extraction from cactus cladodes was enhanced with degrading enzymes such as Viscozyme L, Ultraflo L, Rapidase TF, Pectinex 5XL, Optidex L-400, Econase CE, Spezyme Prime, Cytolase PCL5 and Celluclast 1.5 L [56]. The use of enzymes enhanced the degradation of the walls of the plant cell, thereby increasing the mucilage yield. The influences of extraction temperature (30–60 °C), time (2 to 6 h) and enzyme concentration to raw material ratio (30–60 U/g) on the mucilage yield of tender young fronds of *A. australasicum* using endo-1,4- β -xylanase and β -glucanase as enzymes were investigated by Chiang and Lai [57]. The result obtained in this study was compared with the mucilage yield from water-assisted extraction. The

optimum extraction yields of 3.64%, 6.04% and 6.77% were obtained for the water-, xylanase- and glucanase-assisted extractions at a temperature of 50 °C and a concentration of 50 U/g. The enzyme-assisted extraction significantly increased the extraction efficiency. The combination of 20 U/g xylanase and 30 U/g glucanase gave a yield of 6.86%, while the 50 U/g xylanase and 50 U/g glucanase combination gave a yield of 8.62%. The mucilage from glucanase- and xylanase-assisted extraction is composed primarily of high-molecular-weight polysaccharides with approximately 85% neutral sugar and 15% uronic acid. Zeng and Lai [58] used endo-1,4-b-xylanase and β -glucanase enzymes to extract mucilage from *Asplenium australasicum* leaves, and high mucilage yield with excellent physicochemical properties was reported.

3.1.5. Cold plasma treatment

The application of heat during the extraction of mucilage affects its properties. Mucilage produced using heat exhibits a less uniform microstructure and is brittle, while that produced through cold extraction and freeze-drying has better rheological and solubility properties along with higher porosity [59]. The properties of the mucilage produced through the cold extraction technique are enhanced because the mucilage structure is not altered at low temperatures [60]. The cold plasma technique is characterised by active particles like free radicals, excited and neutral atoms and molecules, free electrons, negative and positive ions and ultraviolet (UV) photons, and it involves the production of ionized gas through the application of energy to gas or a gas mixture [61,62]. This technique efficiently breaks down the plant's cell wall to release valuable compounds [63]. The structure and properties of the polysaccharide are altered when they interact with the plasma [64]. The cold plasma treatment consumes less energy when compared with other extraction technologies. Zielinska et al. [65] used cold plasma treatment, a non-thermal treatment, to extract pectin from okra pods. In this treatment, the structure and properties of the polysaccharides are changed through their interaction with the plasma. In the cold plasma treatment, the time was varied (5, 15 and 30 s), and this technique significantly influenced the extraction yield, rheological properties and structure of the pectin derived from the okra pods. Pectin extraction was optimal for a longer cold plasma treatment. Limited studies are available on the extraction of mucilage from plant cell walls using this technology.

Table 1. Mucilage extraction techniques and percentage yields.

Extraction method	Plant	Optimum percentage yield Extraction variables	Reference
Conventional	Basil seed	20.49/100 g Temperature—56.7 °C Extraction time—1.6 h Water/seed ratio—66.84:1	[25]
Conventional	Mesquite seed gum	24.9%—Alkaline solvent 17.7%—acidic solvent	[27]
Hot extraction	Chia seed	6.97% Temperature—80 °C Seed/water ratio—1:40	[66]
Microwave assisted	Sea buckthorn berries	0.264 ± 0.005% Microwave power—600W Extraction time—6 min Liquid/Material—10:1 mL/g Temperature—85 °C	[43]
Microwave assisted	<i>Ocimum basilicum</i> seed	17.00 ± 0.14% Microwave power—570.32 W Extraction time—4.33 min Solution/sample ratio—40:1 mL/g	[38]
Conventional		14.53 ± 0.25%	
Ultrasonic assisted	Flaxseed	11.04% Amplitude—70% Temperature—90 °C	[51]
Ultrasonic assisted method	Chia seed	10.52%	[67]
Hot extraction		1.86% Temperature—80 °C	
Enzymatic treatment	Tender young fronds of <i>A. australasicum</i>	6.04%—xylanase 6.77%—glucanase 3.64%—water	[57]

3.2. Hydrogel synthesis from mucilage

The properties of the hydrogel matrix are significantly determined by the type of physical or chemical crosslinking technique. Examples of different types of physical and chemical crosslinking techniques are shown in Figure 7. Crosslinkers are used in the synthesis of hydrogel from mucilage to prevent the solubilization of the mucilage and to increase the tensile strength. The physical process involves physical crosslinking interaction, which comprises temperature and ionic interaction, forming bonds between the polymer chains through weak interaction, while the chemical crosslinking involves the creation of new covalent bonds within the polymer chain of the hydrogel. These two techniques can also be used in the synthesis of hydrogels from plant-based polymers. The chemical crosslinking

technique produced hydrogels that have adjustable degradation behavior, desirable mechanical properties and better stability under physiological conditions when compared to hydrogels formed through physical crosslinking techniques [68]. This is attributed to the permanent covalent bonds formed in the matrix of the hydrogel [69]. The difference between physical and chemical hydrogels [70] is shown in Table 2.

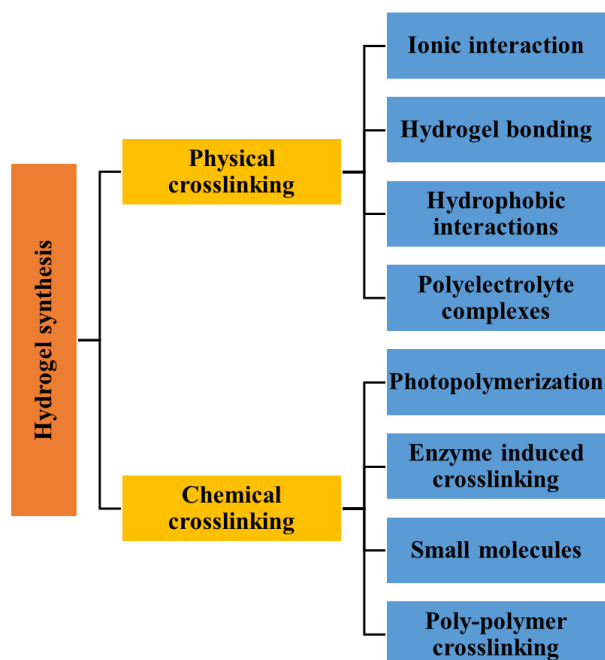


Figure 7. Physical and chemical crosslinking techniques used in hydrogel synthesis.

Table 2. Difference between physical and chemical hydrogels.

	Physical hydrogel	Chemical hydrogel
Formation	Secondary forces and/or molecular entanglement	Covalent crosslinking
Bond	Weak and the hydrogel are considered as reversible gel	Strong covalent bond that are irreversible
Degradability	Less stable against degradation	Very stable against degradation
Homogeneity	Homogenous	Non-homogenous
Mechanical properties	Poor mechanical properties due to reversible physical interaction	Enhanced mechanical properties
Applications	Bioactive substance	Pharmaceutical, agriculture, food industry and cosmetics

The synthesis of natural hydrogels requires the use of non-toxic and friendly components that are less harmful to humans and the environment. To regulate the heat of polymerization during the polymerization of natural hydrogel, the mucilage is often dissolved in diluents such as aqueous solutions and water. Impurities such as unreacted monomers, cross-linkers, initiators and by-products in the hydrogel are removed by washing it. Polymerization techniques like graft polymerization, polymer hydration, radiation polymerization, cross-linking reactions, physical mixtures and bulk polymerization are used in the synthesis of hydrogel [71]. The major components in the preparation of

hydrogel are crosslinkers, initiators and monomers. Hydrogel-like structures can be obtained from plant mucilage, such as from basil and chia seeds, without the use of external crosslinkers. These hydrogels are characterized by an extensive network of nanoscale fiber protruded from the seed's surface into the aqueous bulk [72]. Sacco et al. [17] prepared chia hydrogel by dispersing the powdered chia mucilage in distilled water at 50 °C with continuous stirring until the macroscopic aggregates disappeared. Various crosslinkers and biopolymers are used in hydrogel formulation, and mucilage is sometimes used as a crosslinker or primary biopolymer component [73]. Hydrogel was prepared in various weight concentrations from *Mimosa pudica* L. mucilage using water as the solvent, 10% v/w of glycerol as the co-solvent and 0.2% w/w methyl paraben as the preservative [74]. The mucilaginous polysaccharide was mixed in water, after which glycerol and methyl paraben were added and mixed to form a homogenous mixture using a magnetic stirrer. The mixtures were then heated in a water bath at a temperature of 70–80 °C, after which they were allowed to cool to room temperature. Sharma et al. [75] used the free radical co-polymerization technique to prepare hydrogel from little millet, proso millet and adzuki bean mucilage. The crosslinker used was N,N'-methylenebisacrylamide, while potassium persulfate was used as the initiator. The mucilages were dissolved in 10 mL of deionized hot water, after which the crosslinker and initiator were added and stirred continuously. The mixture was sonicated to remove air bubbles and then transferred to a tube and put in a water bath for an hour. The hydrogel was air-dried and later dried in an oven at 60 °C. To prepare graft copolymeric hydrogel using linseed mucilage, [76] used free radical polymerization. In this process, the powdered mucilage was mixed in purified water at 70 °C, and potassium persulfate was added to the mucilage solution. Methylene bis-acrylamide (crosslinker) and acrylamide (monomer) solutions were added to the polymer initiator solution. Insufficient hydrolytic stability in the linseed mucilage was enhanced through the introduction of acrylamide with groups like hydroxyl alkyl and alkyl. The obtained mixture was put in a water bath (50 °C) for 1 h and later increased to a temperature of 80 °C until hydrogel was formed. The hydrogel was then washed in a water/ethanol solution to remove any unreacted content before drying in an oven (50 °C) for 24 h. The free radical polymerization was also used in the preparation of hydrogel from okra while varying the ratio of the mucilage to monomers (1:5, 1:2, 1:1) and using ethylene glycol dimethyl acrylate (crosslinker) and potassium persulfate (initiator). This simple production process produced natural hydrogel with properties that are valuable when compared to synthetic hydrogel [77]. The polymeric matrix of composite hydrogel, as shown in Figure 8, was developed by polymerizing acrylic acid using N,N'-methylenebisacrylamide and ammonium persulfate as the crosslinker and initiator, respectively, in the presence of linseed mucilage and chitosan [78]. The influence of pH on water absorption and water swelling percentages was investigated in this study. The linseed mucilage of 0.1 g was dissolved in 20 mL of water at a temperature of 70 °C, and 0.1 g of chitosan was mixed with 2 g of acrylic acid to form a homogeneous solution, after which 0.08 g of N,N'-methylenebisacrylamide was added. Ammonium persulfate was added to the solution to begin the acrylic polymerization. The mixture was then mixed for 2 h to form the hydrogel.

In recent times, mucilage-coated metal nanoparticles and nanostructure hydrogels have been utilized for different hydrophobic and hydrophilic applications [79]. Hydrogel can also be synthesized into smaller sizes, such as microgel and nanogel, which have the combined exclusive properties of nanoparticles and hydrogel with enhanced efficiency, possess a higher exchange rate per unit volume and respond faster to stimuli [80,81]. The synthesis of nanohydrogel from plant-based polymers is illustrated in Figure 9. The diameter of the nanohydrogel is usually less than 100 nm [9]. Intramolecular crosslinking is predominately involved in nanogels, while hydrogel involve intermolecular

crosslinking [82]. Sabzevar et al. [83] produced natural hydrogel with enhanced magnetic properties from basil seeds, azobisisobutyronitrile (radical initiator), EGDMA (crosslinker), vinyl-modified BSG, methacrylic acid (monomer) and $\text{Fe}_3\text{O}_4/\text{SiO}_2$ nanoparticles for drug delivery. Nanohydrogel produced with mucilage has enhanced stability when compared with alternative plant-based biopolymers [84]. Sen et al. [85] produced a nanofiber mat from *Hibiscus rosa-sinensis* leaves for wound healing scaffolds. For effective release of mucilage from the leaves, the leaves were sliced into small pieces and then soaked in distilled water for 12 h at room temperature. The solution was then sieved to separate the mucilage from the leaves in the water, and acetone was added to the filtrate to precipitate the mucilage. In the preparation of polymeric blends, 10% w/v PVA, 2% and 5% w/v pectin and 1% w/v dried mucilage were used to form a homogenous solution, which was electrospun to obtain the nanofiber. The nanofiber was cross-linked with 25% glutaraldehyde and 0.5 M hydrochloric acid vapor to enhance its durability and stability. The properties of the non-crosslinked and crosslinked nanofibers were evaluated in this study. Nanocomposites, nanofibers and nanoparticles (metallic and non-metallic) are introduced into the hydrogel to act as nanocarriers in different medical and industrial applications.

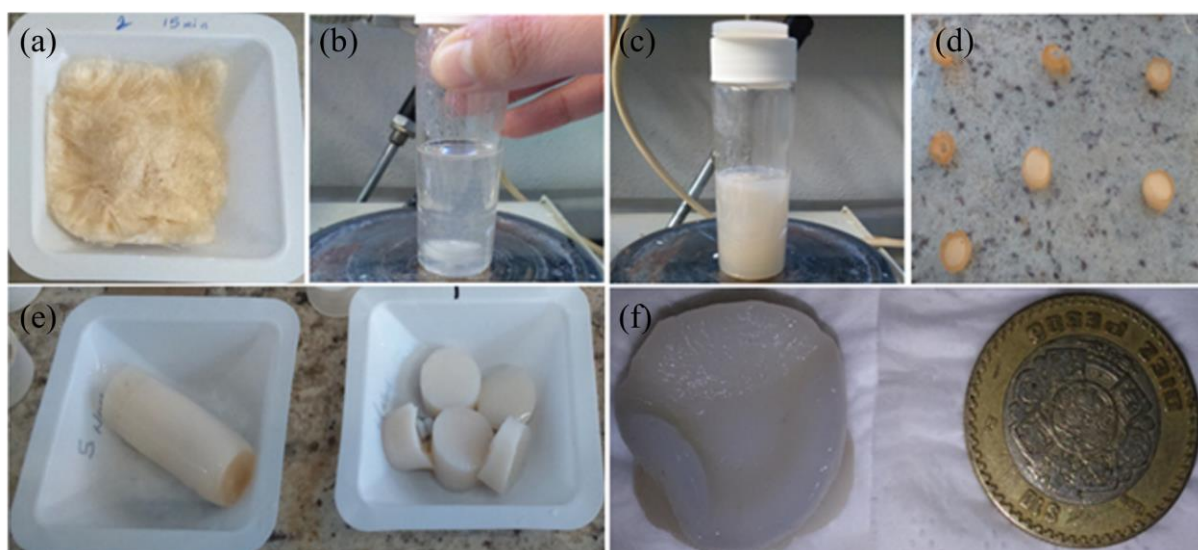


Figure 8. Physical aspect of (a) purified and dried linseed mucilage, (b) reaction mixture consisting of dissolved linseed and chitosan in acrylic acid, (c) hydrogel after 2 h of polymerization, (d) dried hydrogels (xerogels), (e) hydrogel after polymerization and slicing, (f) sample of swollen hydrogel at pH = 9.0 and its comparison with a coin of 1 inch diameter (Reproduced from Ref. [78] with permission).

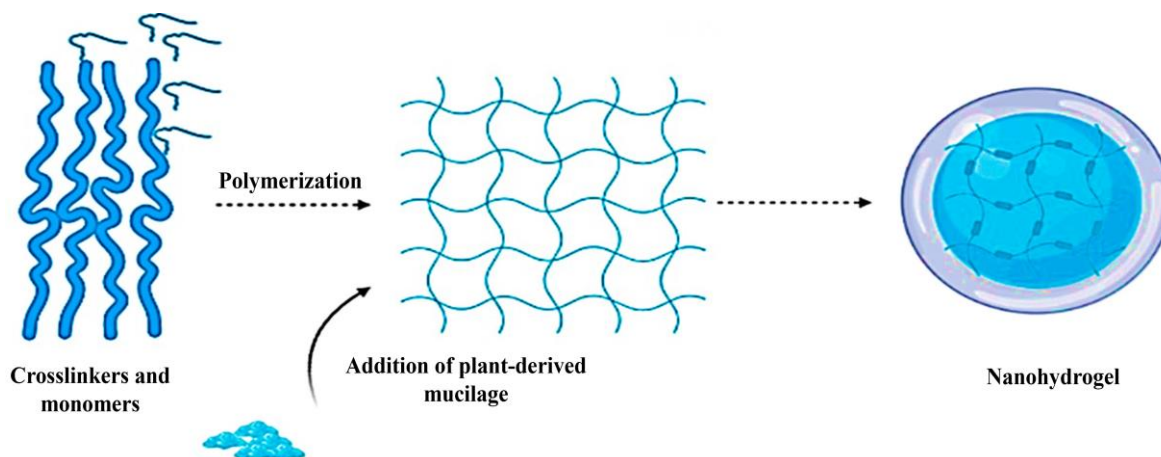


Figure 9. Formation of nanohydrogel using plant-based mucilage (Reproduced from Ref. [9] with permission).

4. Properties of mucilage-based hydrogel for passive cooling

Hydrogels are susceptible to temperature and environmental changes because of the large amount of water they contain. The properties of natural hydrogels from mucilage should be evaluated to determine the possibility of using them in passive cooling technology.

4.1. Physiochemical properties

4.1.1. Solubility of hydrogel

The solubility of mucilage in chloroform, methanol, ethanol, acetone and water was investigated by [75]. The mucilage was insoluble in chloroform, methanol and acetone, soluble in hot water and partially soluble in cold water. The solubility of flaxseed mucilage at different temperatures was evaluated by Emadzadeh et al. [86] while considering the effects of extraction techniques and different species of flaxseed. The solubility of the mucilage increased with an increase in temperature. The exposure of the hydroxyl groups in the mucilage to water molecules causes the breakdown of the hydrogen bonds that exist within the polysaccharide chains, which leads to an increase in solubility. The extraction method was observed to affect the solubility of the mucilage, and they produced mucilages with different molecular weight, which affected the solubility for each technique. The structure and molecular weight of the polysaccharide affect the solubility of mucilage. The solubility increased with a decrease in molecular weight [87]. The ultrasonic method produced mucilage with a lower molecular weight due to the structural destruction and irregularity in the molecules of the polysaccharides caused by the cavitation phenomenon. Hydrogel from cellulose has low solubility in most organic solvents and water because of the presence of a hydrogen-bonded structure.

4.1.2. Swelling properties

The swelling property of hydrogels is a unique characteristic that determines their potential use in several applications. Hydrogel can be considered a heat sink that absorbs water when it comes into

contact with moisture to realize evaporative cooling, which provides a more effective cooling effect than radiative cooling. The deswelling and swelling properties of the hydrogel cause reversible changes in the volume of the hydrogel, thereby affecting its structure, surface morphologies and mechanical properties [88]. In passive cooling, the high content of water in the hydrogel will cause it to freeze often at low temperatures and dry out easily at a higher temperature. The swelling ratio of hydrogel is dependent on its chemical structure. Hydrogels with a higher hydrophilic group content will swell more than those with a higher hydrophobic group content. The hydrophobic group breaks down in the presence of water, thereby reducing its interaction with water molecules [89]. The elasticity of the polysaccharide network, crosslinking density and the presence of hydrophilic and hydrophobic groups determine the swelling of natural polysaccharides. The influences of varying concentrations of monomer, crosslinker and linseed mucilage on the swelling properties of linseed mucilage were studied by Mahmood et al [76]. The increases in monomer and crosslinker concentrations caused decreases in the swelling ratio. The reduction in total concentration of the monomer caused an increase in the degree of dilution of the matrix at constant gel, which caused an increase in the equilibrium water content of the gel, while a lower concentration of monomer increased the swelling [90]. A high concentration of crosslinker gave rise to a higher crosslink density, thereby reducing the water absorbency of the hydrogel. Considering the mucilage concentration, an increase in the mucilage content increased the swelling of the mucilage from linseed. This increase was attributed to the increase in hydrophilic group content and the reduction in crosslinking density [91]. The effect of polymer and the concentration of crosslinkers on the swelling property of quince mucilage-based hydrogel was studied by Sumaira et al. [92] using acrylamide (monomer) and methylene bis-acrylamide (crosslinker). The swelling ratio of the quince hydrogel decreased with an increase in polymer concentration, and this was attributed to the reduced crosslinking density. The increase in polymer concentration was also observed to enhance the porosity of the hydrogel. The production of repulsive forces and ionization of the ionizable groups in the hydrogel were responsible for the hydration of the hydrogel as the polymer concentration increased. Considering the crosslinking density of the polymeric network in this study, the hydrogel swelling ability was strongly determined by the degree of crosslinking. Choudhary et al. [77] examined the swelling capacity of okra hydrogel using glucose, saline and water solutions. The rate of swelling in the hydrogel increased with time, and the highest swelling was reported for water, while the saline solution had the least, due to the presence of sodium ions around the hydrogel. The effect of pH on the swelling capacity was also considered in this study. It was observed that the swelling was lower in an acidic condition but higher in an environment of pH between 7 and 8. The $-\text{COOH}$ group dissociated at a pH value above 7, causing a slow swelling rate and reducing the osmotic pressure while increasing the mobile ions. The swelling property of hydrogel can be altered by altering the pH or concentration of salt in the solution.

Using nanoparticles, the swelling property of nanocomposite hydrogel decreased as the concentration of the nanoparticles increased when immersed in deionized water for 24 h [93]. In this study, the swelling rate of hydrogel without the nanocomposites was 2.5 times that with nanocomposites. The addition of nanocomposite reduced the porosity of the hydrogel, thereby reducing the swelling rate, enhancing the hydrophobic property and improving the crosslinking degree of the hydrogel initiated by the non-covalent interactions.

4.2. Mechanical properties

4.2.1. Tensile strength and flexibility

Hydrogel displays excellent self-healing, flexibility and elasticity properties, making it an appropriate polymeric material for numerous applications such as environmental remediation, energy storage, biomedical engineering and cooling of food and drugs. The mechanical strength of hydrogels is essential because they experience a reduction in mechanical strength when they absorb water. Excellent mechanical properties and flexibility are essential for evaporative and radiative cooling. The mechanical strength of mucilage-based hydrogel is dependent on the combination of cellulose and pectin due to their poroelasticity effect [94]. Hydrogels have the tendency to lose their mechanical toughness and become brittle and hard at low temperatures when the water freezes and at high temperatures with water loss due to their high water content. The freezing and dehydration of water in the hydrogel make it lose its elasticity and conductivity. To enhance the mechanical properties of hydrogel, Xu et al. [93] improved the flexibility of the hydrogel by dispersing nanoparticles in the matrix of the hydrogel. In this study, natural deep eutectic solvent was used as a plasticizer. The nanocomposite hydrogel had the ability to withstand twisting and folding when compared with the hydrogel without plasticizer. The excellent flexibility and tensile properties are attributed to the enhancement of the hydrogen bonding forces provided by the plasticizer system, but the tensile stress decreased sharply during water absorption due to the typical swelling-weakening effect. The addition of nanoparticles in this study enhanced the tensile strength of the nanocomposite hydrogel but significantly reduced the elongation. Nanocellulose can improve the dimensional and mechanical stability of natural hydrogels [94]. Zhou et al. [95] used cellulose nanofiber as reinforcement to enhance the mechanical properties of hydrogel. The elongation of the hydrogel increased with an increase in nanofiber content, but the continuous addition of nanofiber causes the elongation to decrease. This trend was attributed to the production of stress concentrations caused by denser networks. The elastic module and elasticity of the hydrogel increased with an increase in the nanofiber content.

4.2.2. Tensile strength and flexibility

The rheological properties of *M. pudica* mucilage hydrogel were evaluated by Deore and Mahajan [74], varying the concentration of the mucilage content in the hydrogel matrix. The hydrogel showed excellent hydrogel properties at low concentrations; and at higher concentrations, the hydrogel was colloidal in nature and showed shear-thinning flow behavior. The viscosity decreased with an increase in the shear rate over time. The viscosity of mucilage from *Opuntia ficus-indica* showed less viscosity at a lower mucilage content [96]. The effect of temperature on the Newtonian viscosities at steady-state conditions was determined using creep experiments [17]. At a polymer concentration of 2% w/w, chia hydrogel behaved as a viscoelastic material, but mild stiffening was observed as the temperature increased. At a low chia concentration of 0.2%–1% w/w, there was a decrease in viscosity as the temperature increased. At large deformations, the chia hydrogel exhibited strain-softening behavior. The authors reported that the polymer network of the different polymers in the chia hydrogel is influenced by the inter-chain interactions. These interactions influenced the different behaviors towards network slipping under constant applied stress. Considering the effect of temperature on the viscosity of mucilage, the viscosity of *Pereskia aculeata* leaf mucilage was observed to decrease with

an increase in temperature. The increase in temperature enhanced the molecular motion, which influenced the intermolecular interaction and the hydrogen bonding [97]. The effect of temperature on the viscosity of *Diospyros melanoxylon* Roxb. seed mucilage solution was investigated by Singh et al. [98], and it was reported that the viscosity decreased with an increase in temperature. The pectin in the mucilage was identified as the major component responsible for the viscosity of the mucilage. Mucilage with a high content of pectin has been observed to possess non-Newtonian behavior, while low content results in Newtonian behavior [99]. The viscoelastic matrix of the hydrogel gives it self-healing properties. The self-healing property is the ability of the hydrogel to heal itself when it is damaged. This property improves the durability and reliability of the hydrogel while reducing the waste of resources and the cost of repair.

4.3. Thermal properties

4.3.1. Thermal stability

The thermal stability of hydrogel needs to be enhanced to improve its adaptability to extreme temperatures and workability at different temperatures. The thermal stability of okra mucilage was evaluated by Glaue et al. [100], and it was reported that there was no considerable weight loss in the sample at temperatures below 240 °C. However, in the study conducted by Santos et al. [101], okra mucilage had three different mass loss stages at 30 to 150 °C, 180 to 400 °C and 400 to 600 °C, with mass losses of 12.36%, 49.42% and 10.23%, respectively. The mucilage was observed not to be thermally stable after the first loss. Choudhary et al. [77] explained that the first stage of decomposition observed was the evaporation of water, while the second stage involved the decomposition of small volatile molecules, and the third stage is the complete degradation. The authors attributed the high thermal degradation rate to the significant amount of pectin in the mucilage. The thermal stability of mucilage-based hydrogel is an essential property to consider in the selection of a suitable material, especially when the material will operate in a high-temperature environment. Mucilage has the potential to be used in the production of hydrogel for passive cooling because it is thermally stable and can maintain its structure and properties at varying levels of heat and high temperatures. The thermal stability of natural hydrogels can be enhanced by introducing nanomaterials, ions and organic solvents that modify the polymer chains, thereby enhancing the thermal stability of the hydrogel. Under different working temperatures, the thermal stability of the hydrogel is essential for it to adapt to extreme environments.

4.3.2. Anti-freezing and anti-drying properties

The mechanical stability of hydrogels over a wide temperature range is affected by the cyclic drying of hydrogels in an arid condition and freezing below the freezing point of water in a low-temperature environment. Anti-freezing and anti-drying properties of mucilage-based hydrogel are critical in its application in passive cooling because it becomes brittle and hard when the water dries up, losing its mechanical flexibility and electrical conductivity [95]. The addition of soluble ions such as sodium chloride, potassium chloride and calcium chloride to hydrogel reduces the freezing point by inhibiting the formation of ice crystals while increasing the thermal conductivity [102]. The addition of organic solvents such as glycerol and ethylene glycol to the hydrogel assists in preventing

dehydration, decreasing the freezing temperature and maintaining the properties of the hydrogel at high temperatures [95]. At high temperatures, the water in hydrogel evaporates in an open space, making it brittle and hard. The addition of alcohol improves the anti-drying properties of the hydrogel, while the addition of nanoparticles improves durability and bond water content by absorbing water and hydrated ions through hydrogen bonding and dipole interaction [103].

4.3.3. Thermoresponsiveness

Natural hydrogels are very sensitive to the environment and temperature changes. Natural hydrogels experience changes in physical properties and discontinuous changes in shape, size and appearance over a small change in temperature. They exhibit phase transitions in response to external conditions like electric currents, ionic strength, pH and temperature [104]. This property makes hydrogel suitable for use as a soft and smart material in aqueous solutions. At higher temperatures, hydrogels form semi-solid gels, while some are liquid at low temperatures. The polymer chains in the hydrogel change from hydrophilic to hydrophobic above the low critical solution temperature due to the water content of the hydrogel at a slight change in temperature [105].

5. Hydrogel for passive cooling technology

Mucilage-based hydrogels are usually used in medicine and drug delivery, but there are limited studies in the literature on the application of mucilage-based hydrogels for passive cooling technology, despite their excellent thermal, mechanical and physiochemical properties. Most of the literature available is on the use of synthetic hydrogel for passive cooling. Passive cooling technology involves the use of natural ventilation, microclimate and evaporative and radiative cooling to reduce the temperature of a system without consuming electricity [106].

Passive cooling technology has been used in various applications, such as the storage and distribution of pharmaceutical products and food, the thermal management of human beings and buildings, semiconductor devices and solar panels. Evaporative and radiative cooling are used to dissipate heat to cold surfaces. Evaporative cooling technology is established on the property of water absorbing heat to change its form from a liquid to a vapor state, and a large amount of heat is dissipated due to the high enthalpy of the water, resulting in a reduction in the temperature of the system. The evaporative cooling technology consists of air-mediated and complementary medium cooling [107]. A water or air volume is required for the evaporative cooling process to occur, and it serves as the cooling medium, which is initially chilled and then later used to cool the object of interest. The natural hydrogel is charged, and during the process of discharging, the air surrounding the hydrogel is first cooled due to the evaporation of water from the hydrogel, and air at a low temperature is consequently used as a medium to deliver the cooling effect on the object to be cooled. The cooling by evaporation phenomenon is used in the cooling of solar panels during the day using hydrogel. The hydrogel absorbs water at night and is used to cool the solar panels when the water evaporates during the day. Evaporative cooling is an environmentally friendly, energy-efficient and cost-effective process since it does not require the use of refrigerant or energy from fossil fuels.

Over the years, several studies have been conducted to investigate the application of radiative cooling technology in photovoltaic cooling, passive power generation, textile cooling and energy-saving buildings [108–110]. Radiative cooling deals with the dissipation of heat to a cold surface using

infrared radiation. Materials such as ultra-thin paints, porous materials, metamaterials and photonic materials are commonly used in radiative cooling technology because of their high thermal emissivity and solar reflection [111]. The high manufacturing cost and complex fabrication process limit the application of multilayer photonic materials [112]. The cooling performance indicators for radiative cooling are the maximum cooling power and the maximal temperature reduction, but the maximal temperature reduction is the well-known indicator for sub-ambient radiative cooling. It deals with the variance between the ambient temperature and the radiative cooler. The larger the maximal reduction of the radiative cooler is, the better the radiative cooler [113].

Feng et al. [114] showed that hygroscopic hydrogel has the potential to be used for radiative and evaporative cooling simultaneously, and the difference in humidity between the ambient and the hydrogel surface causes the flow of vapor, which allows the transfer of energy to the surroundings. This flow phenomenon occurs once the temperature of the hydrogel is lower than that of the surroundings. The mass and heat transfer phenomena of evaporation–insulation bilayer cooling designs using aerogel and hydrogel were investigated by Lu et al. [115]. This study reported that the bilayer containing the transparent aerogel lengthens the cooling time by 400% when compared with the conventional single-layer design of the same thickness. The introduction of the porous insulation layer enabled fine-tuning between the cooling time and the temperature drop. Xu et al. [116] evaluated the effect of evaporative and radiative cooling on fruit preservation under solar radiation using the one-pot synthesis method to produce a natural eutectic solvent-based polyacrylamide/poly(vinyl alcohol) hydrogel with nanoparticles. The addition of zirconium dioxide nanoparticles (ZrO_2 NPs, 400 nm) and poly-(tetrafluoroethylene) nanoparticles (PTFE NPs, 400 nm) at high concentrations enhanced the mechanical properties, atmospheric window emittance and solar reflectance. The addition of the natural deep eutectic solvent improved the moisture sorption capacity and stretchability of the nanohydrogel. The nanohydrogel showed an excellent passive cooling effect by associating evaporative and radiative cooling. The result obtained showed that the fruit treated with nanohydrogel has the maximum average temperature reduction and the most similar quality to the fresh ones after exposure to the sun. Xu et al. [93] developed polyacrylamide/polyvinyl alcohol hydrogel with nanoparticles and natural deep eutectic solvent (NPs/NADES@PAAm/PVA) for passive cooling employing evaporative and radiative cooling technology. During the daytime, the nanocomposite hydrogel had an atmospheric emissivity and a solar reflectance of approximately 0.92 and 0.95, respectively. After water absorption, the nanocomposite hydrogel concurrently achieved evaporative and radiative cooling. Under the sun, the cooling effects and cooling powers of the NPs/NADES@PAAm/PVA, hygroscopic NPs/NADES@PAAm/PVA and swollen NPs/NADES@PAAm/PVA were examined, and average temperature drops of 2.9, 3.6 and 6.2 °C were obtained, respectively. Through the combination of evaporation and radiative cooling, the swollen and hygroscopic NPs/NADES@PAAm/PVA nanocomposite hydrogels possessed better cooling performances after absorbing water, but the evaporative cooling effect reduced as the water content reduced, while the radiative cooling effect remained until the next rehydration. The efficiency of the regeneration of the hydrogel during the night does not match the evaporation of vapor during the day. This limits the cooling time, especially in arid regions. The use of hygroscopic salt can effectively extend the cooling span by assisting hydrogel regeneration [117].

Recently, passive cooling has been adopted in the cooling of electronics components to reduce energy costs. In the study conducted by Pollard [118], a thin layer of hydrogel was applied to the fin of a heatsink. The heat generated caused the water in the hydrogel to evaporate, thereby causing a

cooling effect under load. During the idle state of the electronics, the hydrogel quickly absorbs atmospheric water when it is cool, preparing for the next workload. Mu et al. [119] developed a hydrogel for rapid evaporative cooling of electronic components using polyacrylamide, water molecules and ions. The cobalt/lithium polyacrylamide hydrogel enhanced the photoelectric conversion efficiency of commercial polycrystalline silicon solar cells by 1.26% under solar lighting of 1 kW/m^2 . The temperature of solar panel cells was reduced by 9 and $9.6 \text{ }^\circ\text{C}$ at 600 and 1000 W/m^2 when hydrogels were used for passive cooling [120].

Amith and Joshi [121] used acacia gum to prepare an edible natural hydrogel used as fluid in roof ponds for building cooling. The result showed a substantial reduction in the temperature of the air in the room, and the acacia gum and water mixture were reused for several days without any noticeable degradation of the hydrogel. The authors concluded that acacia gum hydrogel is effective and economical in cooling buildings, and the rate of evaporation of water in the hydrogel was reduced. Currently, the practical application of mucilage-based hydrogel for passive cooling is still not prevalent, and further investigation needs to be conducted for further quantitative analysis to evaluate its energy-saving potential alongside the costs of production, installation, maintenance and disposal during its life cycle.

6. Conclusions, outlook and challenges

Global hydrogel value is estimated to be 6.7 billion dollars by 2023, and this exponential growth is driven by the increasing applications of hydrogel in different industries [122]. Hydrogels are usually used in the pharmaceutical and medical industries for drug delivery, contact lenses and wound dressing. Due to their ability to absorb and retain water, they are also used in the agricultural industry to enhance water management by decreasing water loss through evaporation and making water available to plants during dry seasons and droughts. Synthetic hydrogels derived from fossil fuel sources are commonly used, and their continuous use is harmful to humans and the environment due to their non-degradability and incompatibility with the human system during wound dressing and drug delivery. As a result of these limitations, polymers from plants, animals and microorganism have gained huge demand from the medical and food industries due to their unique properties and diverse applications in the production of film coatings, drug delivery, gelling agents and binders. Mucilage, a type of gelatinous polysaccharide, is found in almost all plants, animals and microorganisms. The major components of mucilage, such as galacturonic acid, L-rhamnose, D-galactose, D-xylose, L-arabinose and glycoproteins, give it exceptional properties that allow for its broad-spectrum applications, such as hydrogel production. The technique used in the extraction of mucilage from the plant's parts affects the mucilage yield and properties. The presence of functional groups such as carboxylic, hydroxyl, amide and sulfate groups in the mucilage-based hydrogel increases its water holding and swelling capacity and influences the microscopic pores and elasticity of the hydrogel. The development of natural hydrogels has evolved from traditional hydrogels to intelligent or smart hydrogels. An intelligent hydrogel responds to external stimuli such as light, mechanical forces, molecules, solvent, temperature and pH, and it has the potential to be used for passive cooling of space, electronics, agricultural produce and medical products.

The massive production of natural hydrogel from plants' mucilage for passive cooling is still evolving, and its utilization has the potential to reduce the overall energy consumption required for electronic and space cooling; provide cooling of food, drugs, vaccines and blood in locations that do not have access to an electricity grid; address the challenges faced with the disposal and degradation

of synthetic hydrogel; and reduce the overall negative effects of the use of fossil fuels on the environment. Currently, most of the feedstock used in the production of mucilage and its synthesis into hydrogel is from first-generation feedstock, such as okra, linseed, flaxseed, basil seed and starch, which are edible. The continuous use of this edible agricultural produce, especially for large-scale production of natural hydrogel, will affect the food supply chain, thereby affecting the cost and availability of these foods for human and animal consumption. Therefore, it is essential that researchers source alternative feedstocks from second- and third-generation biomass feedstocks, as shown in Figure 10, that are abundant and non-toxic when the natural hydrogel comes into contact with food or humans during the passive cooling process. To promote the development and use of natural hydrogel and save our climate from further ozone layer depletion and climatic change, legislative policies should be developed, and financial incentives should be given to companies, individuals and research and development institutions that are dedicated to manufacturing, trading and advocating the use of natural hydrogel from mucilage.

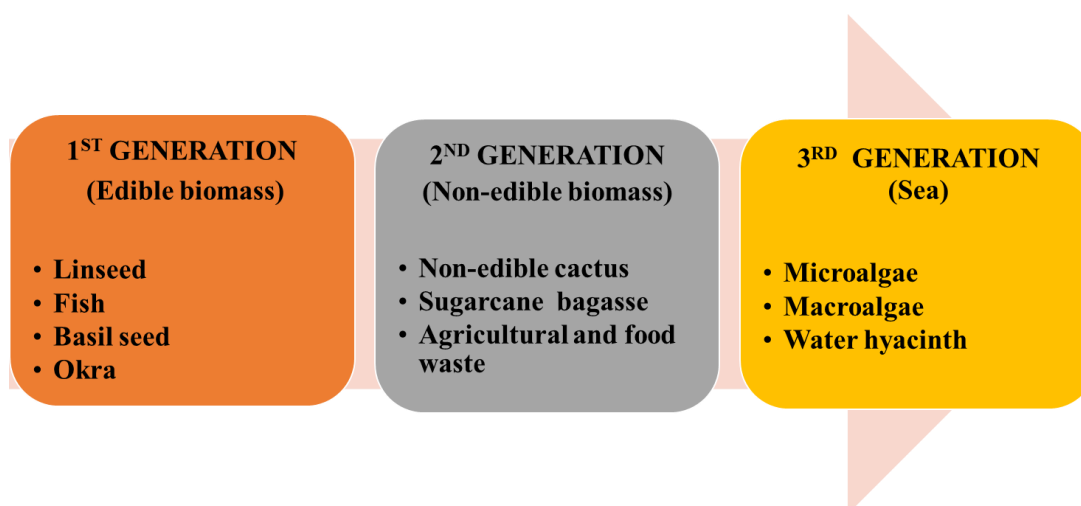


Figure 10. Feedstocks for natural hydrogel production.

Mucilage-based hydrogel is regarded as a potential material for passive cooling considering its mechanical, thermal and physiochemical properties, but synthetic hydrogel is chemically stronger and hydrophobic in nature when compared with mucilage-based hydrogel. Further studies need to be conducted to enhance the rigidity, hydrophobic nature, mechanical properties, thermal stability, pH dependence, physiochemical properties and characteristics of mucilage-based hydrogel through modification of the extraction technique, polymerization components and techniques and introduction of nanoparticles and nanofiber into the matrix of the natural hydrogel. Studies have reported that mucilage contains about 10% water, and the quality of mucilage-based hydrogel tends to change as storage conditions vary. These changes may pose the risk of microbial contamination during production and when used as food preservatives and cooling. Further investigation needs to be conducted to investigate microbial activity in mucilage-based hydrogel, ensure proper control of the various handling processes to enhance the quality and reduce the rate of degradation at different stages of the storage and supply chains.

Use of AI tools declaration

The authors declare they have not used artificial intelligence (AI) tools in the creation of this article.

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Conflict of interest

The authors declare no conflict of interest.

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