GSC Advanced Research and Reviews

eISSN: 2582-4597 CODEN (USA): GARRC2

Cross Ref DOI: 10.30574/gscarr

Journal homepage: https://gsconlinepress.com/journals/gscarr/

(REVIEW ARTICLE)

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Leaves, cuticles and green stems as untapped feedstocks for natural hydrogels: Extraction, functionalization and applications

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GSC Advanced Research and Reviews, 2026, 26(01), 014-022

Publication history: Received on 19 November 2025; revised on 28 December 2025; accepted on 30 December 2025

Article DOI: https://doi.org/10.30574/gscarr.2026.26.1.0395

Abstract

Leaves, green stems and cuticles are underutilized. They can be useful to make natural hydrogels for a sustainable bioeconomy. They are different parts of plants like fruit peels or the sticky stuff from seeds. Leaves, green stems and cuticles have their special mix of chemicals like cellulose, hemicellulose, pectin, cutin and mucilage. These chemicals help leaves, green stems and cuticles to hold water. Leaf cuticles have an ability to control moisture. They also have phenolics and secondary metabolites. The article explains how to extract and separate the hydrogel from leaves, green stems, etc. All the methods in detail including the extraction and fractionation strategies are explained. Overall, valorization of leaves, cuticles, and green stems offers a promising pathway toward circular bioeconomy-driven hydrogel technologies.

Keywords: Natural Hydrogels; Leaf Biomass; Green Stems; Plant Cuticles; Biopolymer Extraction; Sustainable Materials

1. Introduction

Recent studies show that the leaves, cuticle and green stems can act as a water-retaining hydrogel additive, indicating a good potential but no comprehensive synthesis exists yet. Leaves, cuticles or stems differ chemically from typical feedstocks, which suggests a unique extraction workflow. A distinct related framework is needed beyond the general agro-waste reviews. Leaves, green stems, and cuticles should be given more importance in natural hydrogel as they are rich source of natural polymers that is, leaves, stems, and cuticles are rich in biopolymers such as cellulose, lignin, pectin, hemicellulose, etc. which are the key components for the hydrogel formation. These polymers have a good water holding capacity, biodegradability, and biocompatibility which make an eco-friendly hydrogel synthesis. For instance, leaf cuticles are designed naturally to retain moisture which is a desirable property in a hydrogel [1].

Leaves and stems are highly renewable and abundantly available, plant by-products or agricultural waste. Unlike fruit peels or seeds, they are often discarded after the harvest, leading to the biomass waste, which can be utilized for the hydrogel production and thereby it promotes the zero waste and supports the bioeconomy [2]. Leaf cuticles and epidermal layers naturally regulate water absorption, retention and transpiration in the plants. This makes the polymer well suited for moisture management [3]. Leaves and green stems often contain antimicrobial or antioxidant phenolic, and also secondary metabolites, which can be co-extracted or retained in the hydrogels to add the functional properties such as in medical dressing, cosmetics [4]. Valorizing leaves, green stems, and cuticles refers to finding valuable uses for these plant components, especially which are often considered as agricultural by-products or waste. Their availability, seasonal abundance, and integration into the circular bioeconomy make them important resources for sustainable development and agricultural innovation [5]. Leaves, stems, and cuticles are often available in large quantities due to the regular pruning, harvesting, and natural plant life cycles in agriculture. Leaves and stems are non-

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edible biomass, reducing competition with food supply chains. Agricultural residues stems represent 30-60% of total plant biomass after harvest. By this, agro-waste accumulation and open burning can be minimized, which adds economic value to underused biomass streams. Plant leaves and green stems follow predictable seasonal growth patterns, which enables the strategic collection. Seasonal peaks in leaf or stem biomass allows the batch-scale hydrogel or biopolymer production [5-7]. Using these wastes, supports sustainable rural economies, reduces environmental pollution, and adds value to agricultural systems. Leaf and cuticle valorization also reduces landfill pressure and promotes eco-innovation in biopolymer development. So, by converting these agro-waste into sustainable hydrogels which reduces the landfill. It also emphasizes conversion of biological waste into high-value materials. which reduces the rancid waste and fossil dependence. Existing reviews on natural hydrogels derived from leaves highlight their properties such as best biocompatibility, biodegradability, water absorption, swelling ability, and potential in pharmaceutical and also in agricultural and dermatological applications. Current research has characterized hydrogels from various leaf sources like bay leaves focusing on physiochemical properties like antioxidant, anti-inflammatory, and also antimicrobial activities as well as applications like wound healing, dermatitis treatment, and soil moisture retention for increased plant resilience.

2. Biochemistry of leaves and green stems relevant to gelation

The biochemistry of leaves and green stems, focusing on the major polymers relevant to gelation, structure, water relations, barriers etc. These include cellulose, hemicellulose, pectin, cutin, suberin, epicuticular waxes, and mucilage — their chemistry, distribution, roles,

2.1. Cellulose

It is a linear β-(1→4) D-glucose polymer forming microfibrils with crystalline and amorphous regions. Cellulose is the major structural component of primary and secondary cell walls in leaves and stems. Cellulose itself does not act as gel but serves as a rigid scaffold embedding gel forming polysaccharide like pectin and hemicellulose. It is synthesized by cellulose synthase complexes (CESA)in the plasma membrane [8].

2.2. Hemicellulose

It is a branched polysaccharide such as xyloglucans, xylans, and mannans with β-(1→4)-linked backbones. It is embedded in the cell wall matrix between cellulose microfibers. Function in gelation is, it modulates wall flexibility and hydration, cross-linking cellulose microfibers to pectin. It influences gel-like consistency [9].

2.3. Pectin

Complex polysaccharide rich in α-(1→4)-linked D-galacturonic acid. It is located at middle lamella and primary cell wall matrix. Its function in gelation is main gel forming polymer. It forms hydrated gels through, Ca^{2+}crosslinking, and hydrogen bonding. It controls the cell adhesion, wall porosity, and also water retention. It is synthesized in Golgi apparatus and secreted in the wall [10].

2.4. Mucilage

It is a heterogeneous hydrophilic polysaccharide rich in galactose, xylose, rhamnose, and uronic acid. It is found in epidermal cells, seed coats, and glandular tissue, in green stems or leaves of some species which are adapted to arid conditions. Its function in gelation is it is highly gel-forming. It can absorb and retain water, forming viscous gels. It helps in hydration protection and desiccation tolerance [11].

The compositional differences between leaves/green stems and fruit peels/seeds matter a lot for making hydrogels as fruit peels are rich in pectin which is often concentrated in the outer tissues and middle lamella. Pectin from peels tends to be relatively soluble and have high galacturonic acid content and readily gellable under food-style conditions. Its high pectin content makes strong, easily formed hydrogels at lower polymer concentrations. Leaves and green stems contain pectin too, but generally have more cellulose and hemicellulose compared to peel pectin. Their cell walls are optimized for mechanical strength, not for high soluble pectin yield from fruit peel. The pectin in leaves may be more tightly associated with cellulose or hemicellulose, altering its extractability and gelling behavior. The lower extractable pectin and higher insoluble matrix make weaker or more difficult to extract hydrogels unless pre-treated [12,13].

Seeds mucilage forms highly soluble and low concentration gels. It contains mucilage, which is highly hydrophilic, contain soluble polysaccharides, that swell and form viscous gels at very low concentrations. Seed mucilage forms high-water-retaining hydrogels at low polymer loading, which is excellent for thickening wound dressing, edible films, and injectable gels. This shear-thinning hydration kinetics differ from pectin gels [14].

3. Extraction and fractionation strategies

3.1. Gentle aqueous mucilage extraction (petioles, pith)

Gentle aqueous mucilage extraction mechanically shaves the tissue to increase surface area. Soaked in deionized water at a mild temperature of 50 degree Centigrade with slow stirring. The pH was adjusted to neutral to slightly alkaline of pH 7 to 9 to improve solubilization for some mucilage. Mild physical aids such as low-power ultrasonication or heat-assisted extraction were used if yield is low. Clarified by filtration or centrifugation. The polysaccharide was precipitated if needed with ethanol which minimizes depolymerization and native rheology associated with the bio actives was preserved [15].

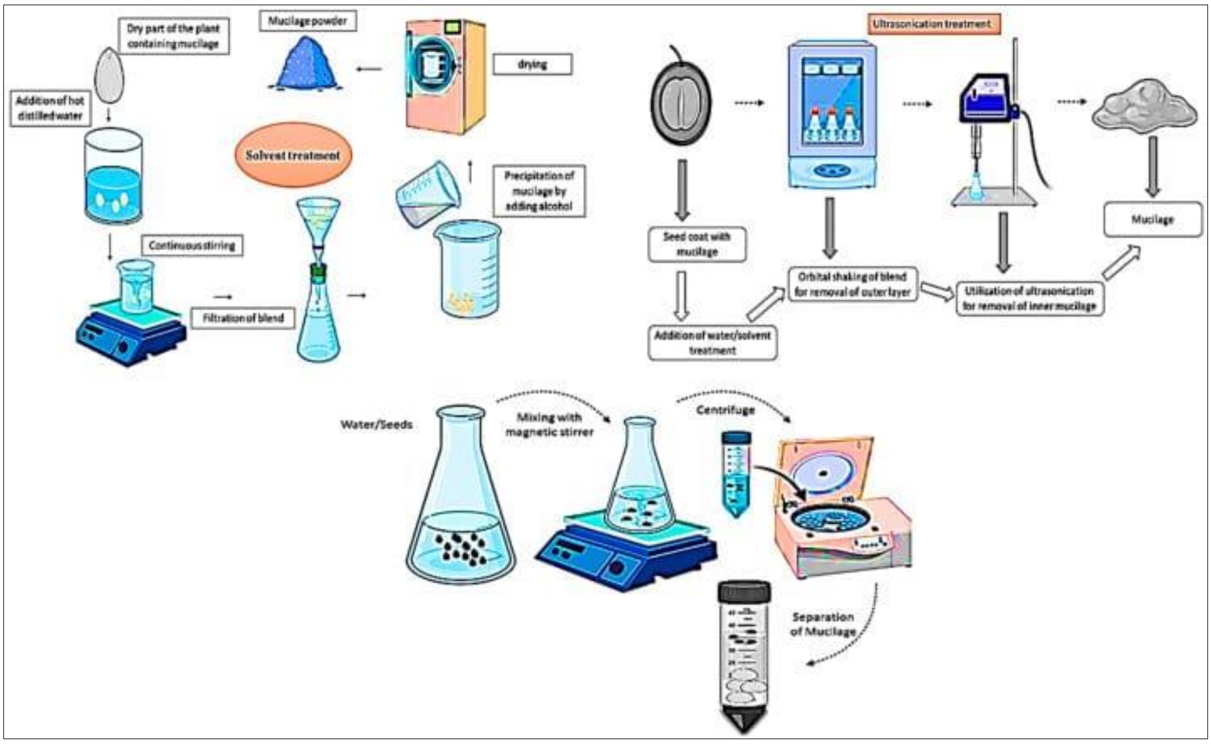

Figure 1 Gentle aqueous mucilage extraction [15]

3.2. Delignification/bleaching for fiber/cellulose recovery

For chemical delignification, Kraft, Soda, Organosolv are the chemicals used. Based on the feedstock and desired purity, the chemicals were chosen. Kraft or Soda are robust for industrial fiber separation and Organosolv gives cleaner lignin and easier cellulose recovery. Oxidative or peroxidase bleaching or chlorine-free sequence are used to remove residual lignin and brightens the fibre, and peroxidase bleaching is milder and more environment-friendly. Mild pre-treatments can enhance delignification efficiency for denser tissues. Chemical delignification yields high cellulose purity but may produce undesired effluents unless controlled [16].

3.3. Cuticle isolation (solvent dewaxing, enzymatic peeling) and preparation of cutin/cutin-nanofibers

Dewaxing (solvent extraction) or Soxhlet/accelerated solvent extraction to remove free and bound waxes is performed. In enzymatic peeling/enzymatic isolation cell-wall degrading enzymes (pectinase, cellulose, hemicellulose) are used under controlled conditions to remove the underlying cell wall. This preserves cutin network better than harsh hydrolysis. Chemical depolymerization for cutin monomers or nanofibers-controlled transesterification or mild alkaline hydrolysis can solubilize cutin into oligomers/monomers; tune severity to avoid complete decomposition [17].

3.4. Chemical modifications useful for gelation (carboxymethylation, TEMPO oxidation, graft polymerization)

Carboxymethyl groups ($–CH_2–COO^−$) can be introduced onto hydroxyls (cellulose, mucilage polysaccharides) to increase hydrophilicity, charge density and gellability. At typical conditions, alkali activation, then monochloroacetic acid at controlled temperature degree of substitution is adjusted by reagent ratios and times. As it promotes water swelling, ion-mediated gelation and better colloidal stability and also useful as a pre-treatment to reduce mechanical energy for fibrillation [18]. TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl radical) - mediated oxidation introduces negative charges that give electrostatic repulsion but also allows ionic crosslinking (Ca^{2+}, Al^{3+}) to form ionically crosslinked gels; TEMPO-oxidized nanofibers form strong shear-thinning hydrogels at low solids [19]. Graft polymerization from polysaccharide backbone to introduce synthetic side-chains that increase network formation and tunable responsiveness such as pH, ionic strength [20].

4. Gelation mechanisms and crosslinking chemistries

4.1. Ionic (Ca^{2+}) gelation of pectin/mucilage from petioles

Ionic gelation mainly happens due to electrostatic interaction between the divalent ions and negatively charged groups of pectin or mucilage polysaccharides. It results in 3D networks. This process is often done via external gelation where polymer solution droplets are dropped into a calcium solution. The calcium ions get diffused into the polymer, cross-linking primarily at the surface first and then gradually inside hydrogel beads are formed. This technique is effective for pectin hydrogels from various plant sources [21].

4.2. Physical interpenetrating polymer networks (IPNs) with polyvinyl alcohol (PVA), gelatin, and leaf-derived polysaccharides

Physical IPNs combine polymer networks connected without covalent bonds, relying on physical entanglements. For instance, PVA and gelatin can form physical IPNs with polysaccharides which are extracted from leaves by freezing or physical gelation. These gels demonstrate enhanced mechanical properties and biocompatibility useful in bio application [22].

4.3. Chemical crosslinking with citric acid, genipin, and EDC

Citric acid acts as a non-toxic, multi-carboxylic acid crosslinker by forming ester bonds with hydroxyl groups of polysaccharides, which therefore improves network stability. Genipin, a crosslinker reacting primarily with amine groups through covalent bonds for stable gels. EDC (1-ethyl-3-(3-dimethylaminopropyl) carbodiamide) facilitates zero-length crosslinking by activating carboxyl groups to form amide bonds with amino groups in biopolymers. These chemical crosslinkers create stronger and permanent gels than ionic or physical gels [23].

The principles and applications of various gelation mechanisms are given in Table 1.

Table 1 Gelation mechanisms

Method	Principle	Type of network formed	Application
Ionic gelation	Electrostatic interaction between divalent Ca^{2+} ions and negatively charged carboxyl groups of polysaccharides	Ionic, reversible 3D network	Simple and mild process; external gelation forms beads; suitable for plant-derived pectin hydrogels
Physical IPNs	Physical entanglement and hydrogen bonding without covalent bonds	Physically crosslinked IPN	Improved mechanical strength; biocompatible; formed by freeze–thaw or physical gelation; useful in biomedical applications
Chemical crosslinking	Covalent bond formation between functional groups of polymers	Covalent network	Stronger and stable gels; citric acid forms ester bonds, genipin reacts with amines, EDC enables amide bond formation

5. Properties and Characterization

Commonly, hydrogels or superabsorbent polymers can absorb fluid, which can be expressed as swelling ratio or percentage. This property is especially relevant in biomedical, environmental, and packaging applications where water uptake impacts performance and function [24]. Rheology involves measures a material's viscoelastic characteristics.

The storage modulus (G') measures elasticity (solid-like behavior) while the loss modulus (G'') reflects viscosity (liquid-like behavior). These values are determined through oscillatory rheometry. Understanding rheology is an important aspect for polymers, gels, and soft matter systems in engineering and scientific research [25]. Porosity is assessed using scanning electron microscopy. It provides visualization and quantification of a material's pore structure i.e. size, shape, and distribution. This technique is used for characterizing materials in fields like filtration, tissue engineering [26]. Thermogravimetric analysis measures the mass of a sample as it is heated. It indicates the decomposition, temperatures, and stability. TCA is vital for polymers, composites, and natural products to predict their thermal limits and lifespans [27]. Monitors heat flow into or out of the material as it is heated or cooled. It detects transitions like melting glass transitions or crystalline points. Differential scanning calorimetry (DSC) yields heat capacity and transition enthalpies which are useful for research and quality control.

6. Applications and case studies

Hydrogel-based materials and biopolymers have become an important across multiple applied sectors, from agricultural and environmental engineering to biomedical and food packaging systems, agriculture.

6.1. Agricultural applications

Hydrogels and biopolymer composites improve soil water retention and serve in seed coating and fertilizer control. Seaweed-derived polysaccharides such as alginate, agar, and carrageenan have been used to prepare biodegradable hydrogels, which improves drought resistance and nutrient efficiency. These materials act as slow-release carriers for urea, herbicides, and pesticides, reduces groundwater contamination and improves the yield [28]. Case studies include seaweed hydrogel granules functioning as micro-reservoirs for water and nutrients under drought conditions [28]. Chitosan-based seed coatings improving germination rates and early root development due to the polymer's bioadhesive and antifungal properties [29].

6.2. Environmental applications

Hydrogel composites are widely researched as absorbents for dye and heavy metal removal for some wastewater. Modified chitosan, cellulose, and alginate hydrogel shows high capacity for Pb^{2+}, Cr^{2+}. Their biodegradability provides environmental advantages over traditional polymeric sorbents [28,29]. Case study includes carboxylated chitosan hydrogel films demonstrated over 90% removal efficiency for methylene blue dye and 95% sorption for lead ions in simulated wastewater studies [29].

6.3. Biomedical applications

Hydrogels are optimized for wound dressings, mucosal systems, and tissue engineering matrices due to their ability to reduce the extracellular matrix. Quadrant hydrogels dressings exhibit hemostatic antibacterial self-healing mechanism [30]. Case studies include self-healing hydrogel adhesives made from catechol Fe^{3+}, cross-linked polymers, outperformed medical glues and sutures in closing incisions and preventing infection [30]. Hyaluronic acid hydrogels, cross-linked with benzaldehyde-functionalized PEGs, promote regeneration and demonstration of dual antibacterial capability against *E. coli* and Methicillin-resistant *Staphylococcus aureus* (MRSA) [31].

6.4. Packaging and food applications

Hydrogels and biodegradable films in food sectors acts as edible coating or active packaging materials for perishable goods. They provide controlled release of antimicrobial or anti-oxidants to extend the shelf life [32]. Case studies include antimicrobial edible coatings containing chitosan or essential oils reduce microbial decay in minimally processed fruits and sausages [32].

7. Challenges and technical gaps

Heterogeneity of raw materials for composition and microstructure vary by species, tissues, and processing history. Consequences are inconsistent material properties like mechanical, rheological, and barrier, and also poor reproducibility between batches. The suggested actions are standardized source selection and use fraction or characterization work of monosaccharide profile, degree of branching, and report the batch reports [33]. Seasonal and environmental variability in yield and bioactive composition. Consequences are seasonal changes, polysaccharide content, sugar or ash, and antioxidant levels, and functional performance of mucilage or cutin derived fractions. Suggested actions are seasonal sampling studies and also developed for harvest windows. Blending or post-harvest condition should be implemented to homogenize input [15]. Residual phytochemical traces for toxins and bioactive components complicate purification and biocompatibility. Consequences are unwanted phenolics, alkaloids, and also pesticide residues can alter geragination, trigger cytotoxicity, and immunogenic responses in downstream medical and food uses. Suggested actions are targeted removal processes like absorption, solvent washes, dialysis, and also validated impurity assays and bioactivity or toxicity screening early in process development [34].

Scale-up of cuticle or cuticle isolation and downstream purification is technically difficult and poorly optimized. Consequences are lab methods like enzymatic peeling, solvent de-waxing, and delignification are often low through throughput and results. Solvent-intensive or give-heterogeneous fractions are creating bottlenecks for kilogram-plus productions. Suggested actions are developing continuous or flow extraction, green solvents or enzymatic cascades, and couple with validated analytics. Examples for validated analytics like spectroscopic and chromatographic fingerprints to control product specification [35,36]. Regulatory, safety and standardization gaps for plant-derived biomaterials. Consequences are lack of clear product specifications, biocompatibility standards, and regulatory person's slow translation to medical and food applications. Suggested actions like early regulatory mapping by ISO-ASTMA-FDA guidance, standardization in vitro and in vivo biocompatibility panels, and interlaboratory rink trails to accept criteria [37].

8. Research roadmap and opportunities

8.1. Standardized extraction protocols

To produce reproducible, scalable, low-impact extraction protocols for polysaccharide-rich fractions from leaves, green stems, and peels, the standard workflow should be followed. The first step is raw material handling and reporting (record species, part, age, pretreatment, particle size, and moisture content). Then followed by sequential fraction baseline i.e aqueous extraction for soluble mucilage, enzyme-assisted delignification or mild alkali for hemicellulose, solvent de-waxing for cuticle isolation can be done from mild to strong. Green intensification options such as ultrasound-assisted, microwave-assisted improve yield reducing harsh chemicals. The next step includes purification and drying by ethanol precipitation, dialysis for molecular weight fractionation. At last yield, energy use, solvent mass and waste streams are recorded [15].

8.2. Comparative screening of leaf vs peel vs stem

Systematically feedstocks are compared using the same extraction and characterization to identify best sources for target applications in agricultural, biomedical, and food packaging. Firstly, select representative species and then use identical pre-treatment. Followed by characterization panel i.e, chemical composition, monosaccharide profile and functional traits such as swelling ratio, equilibrium, water content, rheology, tensile properties. Evidence from comparative studies shows peel-derived pectin often give high water holding and gelation but leaves and stems can provide cellulose or hemicellulose fraction with higher mechanical strength in composition.[38].

8.3. Green modification pathways

To replace toxic crosslinkers or solvents with natural while retaining function, promising green pathways like physical crosslinking by thermal gelation, freeze-thaw cycles to avoid covalent reagents are used. Enzymatic crosslinking, laccase or transglutaminase to form network under mild conditions. Bio-based crosslinkers and click-like green chemistries, citric acid esterification and enzyme coupling are used. Physical nano-structuring, mechanical fibrillation, templating to make interpenetrating networks without synthetic crosslinkers. Reviews on green routes for cellulose and surface modification summarized many of the highlights which reduce environmental footprints when replacing strong acids or base and toxic crosslinkers [23,39].

8.4. Pilot scale demonstration path to commercialization

Pilot milestones are pilot design and feedstock sourcing (secure a stabilized feedstock supply, map seasonal composition changes). Process intensification for scale (implement solvent recycling, continuous extraction and energy recovery). Quality control and standardization. Regulatory and safety: for food biomedical. Use setup GMP compatible lines. Pilot preliminary economic assessment (PEA) or life cycle assessment (LCA) update i.e; update model with pilot data and identify bottlenecks to use this, determine commercial feasibility and investment needs. Case studies of pilot LCA, PEA and pilot plant setup in relation to bioproduct fields show the necessity of refining process modelling and proactive waste valorization to improve economics [40].

9. Conclusion

Leaves and green stems represent promising underutilized biomass feedstocks for natural hydrogel production due to the rich content of bioactive compounds such as polysaccharides, flavonoids, which provides anti-oxidative, antimicrobial, and wound healing properties. Natural hydrogels formulated with leaf extracts such as bay leaves have demonstrated efficacy in wound healing, anti-inflammatory effects, and sustained drug release with desirable physical and rheological properties. Green extraction methods and biopolymer-based hydrogel formulations provides eco-friendly, biocompatible, and effective platforms for exploiting plant biomass, supporting the sustainability goals and a circular bioeconomy. There is a need for systematic standardized protocols for extraction and comparative screening of leaves versus stems versus other parts to optimize better yield bioactivity and hydrogel formulation characteristics.

Compliance with ethical standards

Disclosure of conflict of interest

The authors declare no conflict of interest.

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