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Blue biorefinery: obtaining advanced lignocellulosic materials using marine biotechnology as a tool.

Biorrefinería azul: obtención de materiales lignocelulósicos avanzados mediante la biotecnología marina como herramienta.

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Abstract

Marine biomass from macroalgae, microalgae, and seagrasses is emerging as a fundamental pillar for next-generation biorefineries. This review focuses on the structural fundamentals, experimental methods, and emerging applications of these biomaterials, highlighting the sustainable use of marine resources. The first section of the state of the art covers marine cellulose, or "blue cellulose," as a precursor to advanced materials such as microcellulose and marine nanocellulose. These nanomaterials, characterized by their high crystallinity, mechanical strength, and large surface area, are essential in high value-added biomaterial applications. The second section considers marine lignin as a valuable source of sustainable aromatic compounds. Lignin nanoparticles are crucial as natural antioxidants, UV filters, emulsion stabilizers, and precursors for advanced biocomposites. The exploitation of these marine biopolymers accelerates the transition to a circular and sustainable economy.

Keywords: *Blue cellulose, marine cellulose, marine nanocellulose, marine lignin, marine biotechnology, biomaterials*

Resumen

La biomasa marina procedente de macroalgas, microalgas y praderas marinas se está consolidando como un pilar fundamental para las biorrefinerías de nueva generación. Esta revisión se centra en los fundamentos estructurales, los métodos experimentales y las aplicaciones emergentes de estos biomateriales, haciendo hincapié en el uso sostenible de los recursos marinos. La primera sección, que presenta el estado del arte, abarca la celulosa marina, o "celulosa azul", como precursora de materiales avanzados como la microcelulosa y la nanocelulosa marina. Estos nanomateriales, caracterizados por su alta cristalinidad, resistencia mecánica y gran superficie, son esenciales en aplicaciones de biomateriales de alto valor añadido. La segunda sección considera la lignina marina como una valiosa fuente de compuestos aromáticos sostenibles. Las nanopartículas de lignina son cruciales como antioxidantes naturales, filtros UV, estabilizadores de emulsiones y precursores de biocompuestos avanzados. La explotación de estos biopolímeros marinos acelera la transición hacia una economía circular y sostenible.

Palabras clave*: Celulosa azul, celulosa marina, nanocelulosa marina, lignina marina, biotecnología marina, biomateriales.*

1 Introducción

Cellulose and lignin are the most abundant structural biopolymers on the planet. They are the essential pillars of biomass, the bioeconomy, and the materials industry, as well as sustainable and sustainable energy. Cellulose is a linear polysaccharide, formed by β-D-glucose units linked by β (1→4) bonds, and is the main component of plant cell walls. Lignin, on the other hand, is a three-dimensional aromatic polymer that surrounds cellulose to provide rigidity, structural support, and impermeability. Both materials have gained relevance in the market for their applications as bioplastics, fibers, biosurfactants, bioadhesives, and biofuels (Habibi et al., 2010; Eichhorn et al., 2010).

The extraction of cellulose is an important process due to the diversity of sources from which this biomaterial can be extracted, notably agroforestry species, terrestrial and marine angiosperms, and phycophora. The traditional uses of this material can range from paper and packaging manufacturing to textiles and applications in the cellulose derivatives industry. In contrast, lignin is seen as a by-product in the processing of paper pulp and bioethanol, with only 2% of lignin estimated to be used in the production of high value-added materials (Habibi et al., 2010).

These lignocellulosic materials are being promoted in the polymer industry due to their natural aromatic origin (Habibi et al., 2010; Eichhorn et al., 2010), in response to the need to reduce dependence on petrochemical polymers and their carbon footprint. Biomaterials from marine sources have notable advantages over synthetic ones (Trivedi et al., 2016; Omran et al., 2021; Jose et al., 2025), because they are biodegradable, biocompatible, renewable, environmentally friendly, and also offer attractive production yields. Marine biotechnology has emerged as a strategic field that promotes the valorization of macroalgae, microalgae, cyanobacteria, and marine phanerogams for the use of functional lignocellulosic biopolymers.

Rapid algal growth, high productivity per surface area, and low freshwater use for the production of macroalgae and microalgae make them an attractive raw material compared to agricultural and forestry products (Geng et al., 2025; Iqbel et al., 2025). However, the limitations associated with taxonomic variability, the presence of salts, and effluent treatment act as an economic counterweight, hindering scaling up (Zaimes and Khanna, 2013).

Technological advances have made it possible to combine green processes (enzymatic hydrolysis, organosolv extraction, ultrasound, among others) to obtain lignocellulosic materials. These biomaterials generate industrial-scale production routes that favour their profitability and promote the circular economy (Geng et al., 2025).

There is growing interest in materials engineering in relation to cellulose and lignin, mainly in the pharmaceutical, medical, food, and cosmetics industries, as well as in interfacial phenomena, among others. Algal cellulose, for example, is used for its high crystallinity, which is inversely proportional to particle size (French et al., 2017; Mihranyan, 2011), improving this property as its dimensional scale decreases (Ioelovich, 2022). In the case of algal lignin, it is a reserve of versatile aromatic compounds (Karlsson et al., 2023; Melikoglu, 2025), used in the synthesis of resins and advanced carbonaceous materials (nanolignin, graphenes, and graphene oxides).

However, the engineering challenges involved in exploiting these resources remain to be solved (Sadaan et el., 2024). The presence of salts, biological variability, changing environmental conditions, and waste treatment affect the economic viability of industrial scaling (Klap et al., 2007). Nevertheless, advances in separation, purification, and molecular characterization processes have provided an important basis for the exploitation of these biomaterials, positioning them as high value-added products derived from clean technologies that contribute to the circular economy (Mateo et al., 2025; Zanchetta et al., 2025).

This work aims to provide a state-of-the-art review of marine lignocellulosic biomaterials from the perspective of their attractive properties for industry, the production process, and emerging applications for the development of sustainable biomaterials with high added value.

2. Results and Discussion

2.1 Marine cellulose

Cellulose is the most industrially produced polysaccharide in the world, with an estimated annual production of 7.5E10 tons. It has a linear structure composed of β-D-glucose monomers linked by β (1→4) bonds.

An alternative source of cellulose extraction is macroalgae and marine phanerogams, which have the particularity of forming cellulose with a highly crystalline system that is insoluble in water and has significant mechanical and thermal resistance (Habibi et al., 2010). The formula associated with this macromolecule $(C_6H_{10}O_5)_n$ is organized into microfibrils with larger alternating crystalline regions and smaller disordered or amorphous regions (French et al., 2017).

Green algae of the genus Ulva sp and Cladophora sp are the species most commonly used for the production of marine cellulose. Algae cellulose is characterized by its high degree of polymerization and crystallinity greater than 90% (Mihranyan, 2011).

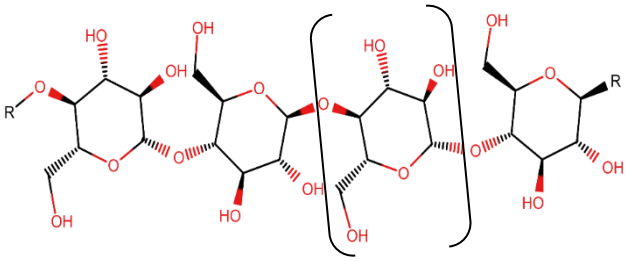

Fig 1. Polymeric configuration (Fisher projection) of cellulose, formed by D-glucose units linked by β-(1→4) bonds, unlike starch, which is α-(1→4). O: oxygen, H: hydrogen. FTIR spectra of non-activated carbons obtained from coconut shells (CC2) and peach pits (CD1).

The most common protocols for algal cellulose production begin with washing and drying the macroalgae, followed by an alkaline treatment stage with sodium hydroxide between 2 and 5% w/v at 80-90 °C for 2 to 4 hours to remove proteins, lipids, and other soluble polysaccharides, lignin, and pigments (Gomaa et al., 2021). This is followed by oxidative bleaching using 2% w/v sodium hypochlorite or 3% hydrogen peroxide at room temperature for 1 to 2 hours (Moral et al., 2019).

Other authors recommend using acetic or hydrochloric acid (1M) for demineralization if calcareous or siliceous remains are known to be present (Díaz-Vásquez et al., 2015). Continue with centrifugation (5000 rpm for 10-15 min) and separate the effluents. Finally, wash with distilled water until

a neutral pH is reached and dry by heat or freeze-drying (Svagan et al., 2023).

From the same cellulose, it is possible to obtain marine nanocellulose (nanofibrillar or nanocrystalline) by mechanical processes such as homogenization, ultrasound, or by chemical techniques of acid hydrolysis that generate fiber breaks in diameters of different scales (Wahlstrom et al., 2020). This biomaterial is used in tissue engineering, drug encapsulation, biosensors, cosmetics, biodegradable films, and food, taking advantage of its ability to form hydrogels (Patrichi et al., 2023).

2.2 Marine microcellulose

Also known as marine microcrystalline cellulose, it consists of D-glucose molecules linked together by β (1→4) glycosidic bonds (Tarchoun et al., 2019), rearranging themselves into a crystalline, three-dimensional system. This system is stabilized by intramolecular hydrogen bonds, which generate high rigidity, insolubility in water, and resistance to organic solvents (Poudel et al. 2025).

The most common sources of microcellulose are red macroalgae (Gracilaria, Gelidium), green algae (Ulva sp.), and especially marine microorganisms such as cellulolytic bacteria (Vibrio sp, Pseudoalteromonas sp) and microalgae of the genera Chlorella sp, Cladophora sp, Valonia sp, and Boergesenia sp (Madadi et al., 2021). Other references point to the extraction of microcellulose from crustacean and marine sponge waste rich in cellulosic and chitinous matrices (Pamungkas and Budhiyandi, 2021).

For the production of microcellulose, it is necessary to remove any lipid and mineral impurities from the source. This can be achieved by washing with 80% ethanol and 1% hydrochloric acid at room temperature for 2 hours, followed by a final wash with distilled water (Trivedi et al., 2016). The material is then subjected to basic hydrolysis (liquid:solid ratio 1:10 with 2-4% m/v sodium hydroxide) at a maximum temperature of 90°C for 2 to 4 hours, removing proteins, lignin, and hemicellulose from the effluent. The resulting solid is subjected to a bleaching stage with 2.5% sodium hypochlorite at 70°C for one hour to remove lignin and pigment residues (He et al., 2018).

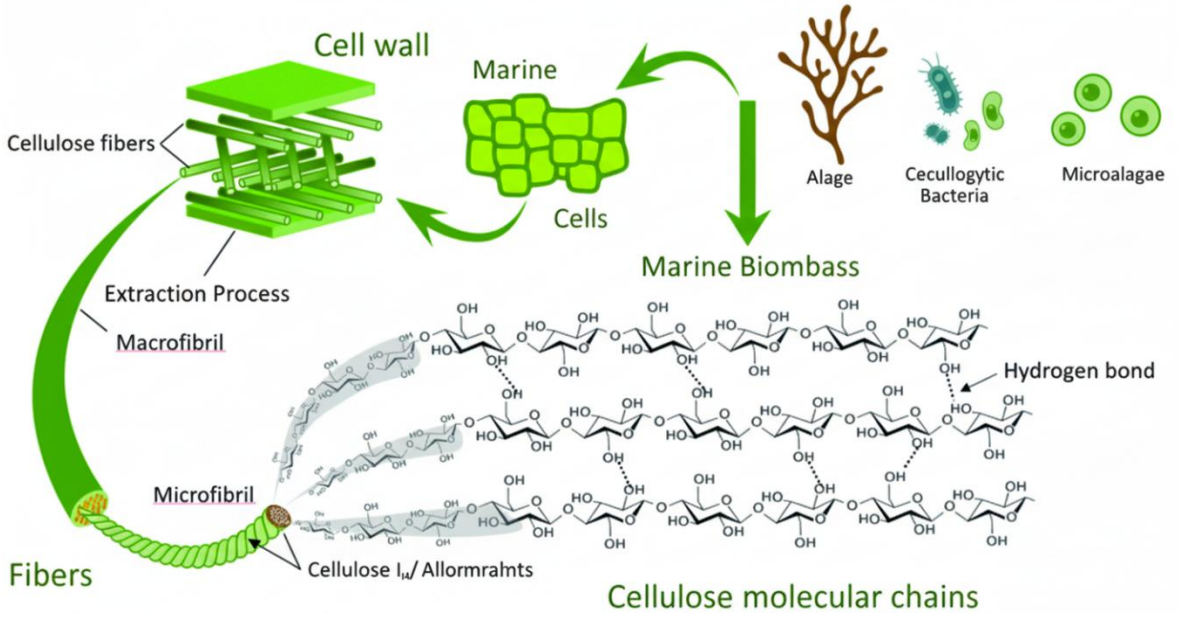

Fig 2. Composition of microcellulose as microfibrils; when hydrolysis is applied, they are released from the structure and generate microcrystalline regions. Inspired by Han. 29.

In order to increase the crystallinity of marine microcellulose, hydrochloric acid is used at a concentration between 2.5 and 3 N at 45°C for 1 hour (Revol et al., 1992), breaking down the non-crystalline areas. It is then washed with distilled water to remove excess acid until a neutral pH is reached, and then centrifuged (10,000 rpm for 10 min) until the phases are completely separated. Finally, sonication (20 kHz, 750 W, 15 min) is applied to disperse the cellulose microfibers (Dong et al., 1998).

Similar to other materials, microcellulose has applications in the field of biomedicine for tissue regeneration, controlled drug release, and scaffolds for tissue engineering, due to its biocompatibility and porosity (Trache et al., 2016). In cosmetics, it is used as a natural thickener, in

foods as stabilizers, surfactants, and biodegradable packaging (Kowalka et al., 2025). Its application is expanding due to the boom in the sugar chemical industry.

2.3 Marine nanocellulose

In recent years, marine nanocellulose has become popular as a biomaterial of interest in research and development. This is due to its high-performance physicochemical properties, such as its crystallinity, specific surface area, high mechanical strength, and ability to form stable three-dimensional networks (Yuan et al., 2021). This type of cellulose mainly occurs in three forms: cellulose nanofibers (CNF), cellulose nanocrystals (CNC), and bacterial cellulose (BC). Its structure is composed of linear chains of β-(1→4)-D-glucopyranose, with alternating crystalline and amorphous regions (Eichhorm et al., 2010, Ren et al. 2014).

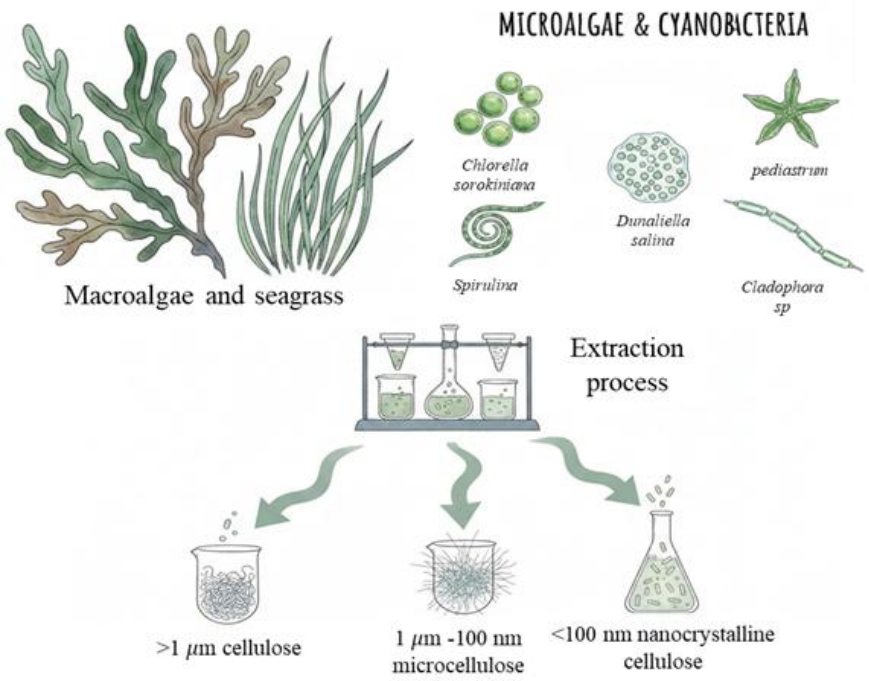

Fig 3. Types of marine cellulose from macroalgae, seagrasses, microalgae, and cyanobacteria.

Among marine sources of nanocellulose, macroalgae, microalgae, and cyanobacteria such as Chlorella sorokiniana, Spirulina, Dunaliella salina, and Cladophora sp. stand out (Plianwong and Sirirak, 2024). The structures of their cellulose have a higher crystallinity than terrestrial sources due to their denser organization in nanofibrils (Stromme et al., 2002).

The methods for extracting marine nanocellulose consist of a series of separation and purification processes. In the case of Cladophora glomerata, it is recommended to start with dry material and subject it to an alkaline treatment with 2% (m/v) caustic soda at 80°C for 4 hours to remove proteins and hemicelluloses. The bleaching process is carried out with 1.5% hydrogen peroxide at 60°C for 2 hours to remove pigments and other residual compounds (Mihhels et al., 2023).

The cellulose nanocrystals are then isolated by hydrolysis with 64% sulfuric acid (v/v) at 45°C for 45 min with constant stirring. This process breaks down amorphous regions, releasing crystals with lengths of 100 to 300 nm and diameters of 5 to 10 nm (Xu et al.,2022). The crystals are then washed with distilled water and dialysis is applied to remove excess acid. Finally, molecular centrifugation is applied at 10,000 rpm for 15 minutes to obtain the CNC (Ren et al., 2014).

When using green macroalgae, high-pressure homogenization (1500 bar) should be used for 10 cycles to release CNF, as an alternative technique to acid hydrolysis (Berglund et al., 2020). Finally, the material obtained is freeze-dried for proper storage.

The food industry uses nanocellulose as a biodegradable film for food due to its antimicrobial properties. It has also been used for the production of packaging, taking advantage of its transparency and mechanical strength (Casalini and

Giacinti, 2022). In the field of medicine, it is used in wound dressings, controlled drug delivery systems, and tissue regeneration support due to its porous structure (Machado et al., 2024). Nanocellulose has had an impact on colloidal chemistry due to the replacement of industrial surfactants with nanocellulose surfactants.

Marine-derived cellulose, microcellulose, and nanocellulose are compared in Table 1 below:

Table 1. Comparison of the characteristics of marine-derived celluloses.

Property	Cellulose	Microcellulose	Nanocellulose (CNC/NFC)	References
Scale	Micrometric scale	1–10 µm	3–100 nm (CNC)	(Habibi et al., 2010; Eichhorm et al., 2010; Mihranyan, 2011; Tarchoun et al., 2019; Madadi et al., 2021; Zanchetta et al., 2025; Berglund et al.,2020; Moon et al., 2011).
Morphology	Thick fibers	Short, amorphous fibers	Rods or fibrils	(Habibi et al., 2010; Eichhorm et al., 2010; Pamunkas and Budhiyandi, 2021; He et al., 2018; Plianwong and Sirirak, 2024; Stromme et al., 2002; Mihhels et al., 2023).
Percentage of crystallinity	40-80% (estimated)	60-80%	70-95%	(Eichhorm et al., 2010; French et al., 2017; Ioelovich, 2022; Revol et al., 1992; Dong et al.,1998; Stromme et al., 2002; Xu et al., 2022; Ren et al., 2014; Moon et al., 2011).
Applications	Support, paper, cardboard	Pharmaceutical excipients, additives	Biomedicine, packaging, etc.	(Zanchetta et al., 2025; Moral et al., 2019; Patrichi et al., 2023; Trache et al., 2016; Kowalka et al., 2025; Yuan et al., 2021; Casalini and Giacinti, 2022; Machado et al., 2024; Moon et al., 2011).

2.4 Marine lignin

Lignin is a complex, amorphous aromatic polymer that provides rigidity, with high thermal stability and relative resistance to biological degradation. It is located in the cell walls of vascular plants and forms three-dimensional structures of phenylpropane units (p-hydroxyphenyl, coniferyl, and sinapyl) that interact through ether bonds (β-O-4, α-O-4, 4-O-5) and carbon-carbon (β–1, β–β, β–5, 5–5) bonds (Karlsson et al., 2023). These characteristics determine its reactivity and value as a raw material for aromatic compounds.

Broadly speaking, marine lignin sources can be classified into two categories: phanerogams and marine macroalgae. The former are vascular sources such as angiosperms that contain lignin in their structure in a similar way to terrestrial sources. Seagrass beds of Zostera sp, Posidonia sp, and Thalassea sp are exploitable sources of cellulose and lignin. On the other hand, macroalgae biosynthesize lignin in smaller proportions than marine phanerogams. There is scientific evidence identifying

complex aromatic groups of the macrophyte-assembly type or functional groups of macroalgae (Sargassum sp) that suggest taxonomic and environmental variations with the presence of high levels of lignin-type aromatic compounds (Li et al., 2012; Alzate-Gaviria et al., 2021).

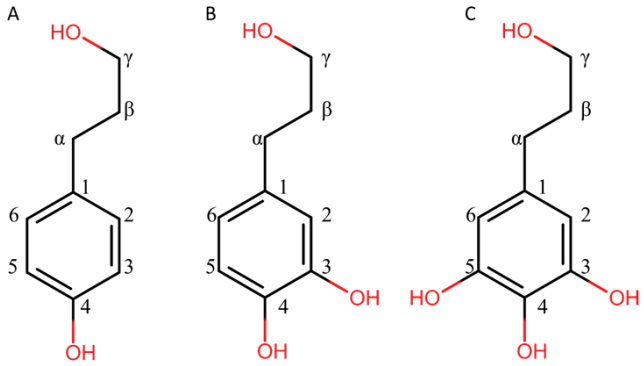

Fig 4. Lignin monomers in Fisher projection, (A) p-hydroxyphenyl, (B) coniferyl, (C) sinapyl. O: oxygen, H: hydrogen.

Marine lignin is extracted in a similar way to terrestrial lignin, but with the application of additional methods for salt treatment. First, it is dried and desalinated with fresh water, and then the material is crushed. The biomass is hydrolyzed in an alkaline medium of NaOH or KOH at a ratio of 1:10 to 1:20 of dry material to alkali solution with a concentration of 1 M. The process takes 1 to 4 hours at a temperature of 80 °C. Dissolved lignin and part of the hemicellulose and cellulose are obtained and removed by centrifugation. The resulting liquid phase is acidified to a pH of 2 with 1 M hydrochloric acid to precipitate the lignin, which is separated by filtration, and the material is then washed and dried (Ley et al., 2023; Li et al., 2012).

Another method used is organosolv, which guarantees the absence of sulfur (in the paper industry), a lower degree of condensation, and a higher content of β-O-4 bonds. The material is pretreated in the same way as in the previous method. Ethanol and water are used in a 60:40 ratio, for a solid:liquid ratio of 1:5-1:10. Hydrochloric acid at 0.1-0.5 M is added at a ratio of 1.5:50 acid:dry solid at a working temperature of 140-200 °C for 20-120 min and a pressure of 10-20 bar. It is then centrifuged to obtain the cellulose and water is added to the liquid phase to precipitate the lignin. It is centrifuged again to obtain the wet lignin ready for the drying process (Melikoglu, 2025; Saadan et al., 2024).

Lignin has many applications, including as a binding agent and a component of resins and panels when mixed with other polymers. It has also been used in chemical routes to obtain monomers such as phenols used in polymer synthesis. Activated carbon has been manufactured from lignin for use in water treatment and electrodes. Due to its high bioactivity, this biomaterial exhibits antioxidant and UV-screening properties and has pharmacological benefits (Yu and Kim, 2020; Liu et al., 2024; Centeno-Bordones, 2022). On the other hand, Gómez et al (Gómez et al., 2025). have studied oxidative processes of lignin to use it in the formulation of animal feed in order to take advantage of its metabolic energy in ruminants.

The marine lignin production industry faces challenges due to the variability of the raw material. Different taxonomies and climatic effects restrict yield and the efficiency of production processes. Brine management often involves treatment processes that affect economic profitability. In this regard, industrial scaling using the organosolv method requires the management of solvents that need to be reused to reduce costs (Saadan et al., 2024; Alam et al., 2024).

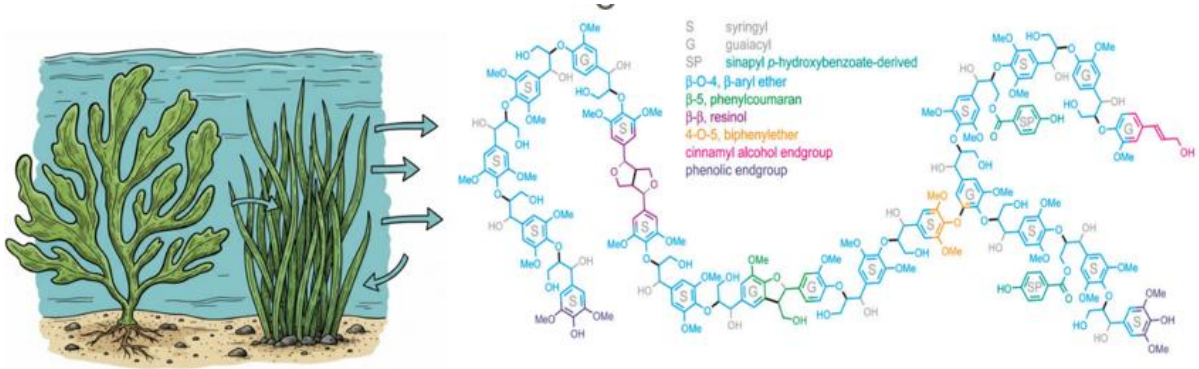

Fig 5. Reference molecule of marine lignin from macroalgae and seagrasses (Vanholme et al., 2010; Stewart et al., 2009).

2.5 Marine lignin

The reduction in particle size in lignin results in an increase in specific surface area, meaning that the reactivity and availability of functional groups (hydroxyls) are more reactive for their applications. This allows interactions through hydrogen bonds, Π-Π stacking, ionic chelation, and coupling reactions, facilitating integration with

biodegradable polymers (such as cellulose, chitosan, or algae polysaccharides). In addition to being an intrinsic antioxidant, it has thermal stability and the ability to absorb UV A, B, and C radiation (Camargos et al., 2025; Centeno-Bordones, 2022).

The traditional sources of extraction are the same as those outlined in the previous section. However, the key to obtaining this nanoproduct lies in its chemical processing methods. Nanolignin is obtained through top-down or bottom-up routes. The top-down route is based on size reduction through mechanical grinding, ultrasound, high-pressure homogenization, and steam explosion (Grappa et al., 2024). The literature has shown the effectiveness of obtaining spherical nanolignin with diameters between 50-300 nm through the top-down route. While the bottom-up route induces controlled precipitation, lignin dissolved in an organic solvent (acetone, ethanol, methanol, tetrahydrofuran) precipitates on contact with water, losing solvation and self-assembling to form nanoparticles. This process allows for easy scaling, but results in purification processes for solvent recovery. However, there are other methods such as: application of templating agents/assisted polymerization, hydrothermal processes, emulsification, and pH-directed self-assembly (Grappa et al., 2024).

A top-down method for obtaining lignin nanoparticles is the solvent exchange and ultrasound method applied by Centeno-Bordones, using black liquor effluent from cellulose extraction. Once the lignin has been precipitated in an acidic medium and dried, it is placed in contact with a water:alcohol (methanol or ethanol) mixture (6:4), keeping it under agitation for 30 minutes. This mixture is then subjected to continuous pulse sonication at 42,000 Hz and 135 W for 30 to 45 minutes. After this time, the solution is vacuum filtered through a 0.45 μm filter. Next, 200 mL of distilled water is added drop by drop and stirred at 400 rpm for 20 minutes. Finally, it is centrifuged for 30 minutes at 3,000 rpm. This methodology yields lignin nanoparticles between 15 and 65 nanometers in size (Centeno-Bordones, 2022).

Cellulose, nanocellulose, or chitosan composites increase their mechanical strength, thermal stability, and UV blocking capacity by incorporating nanolignin into their system. On the other hand, biomedicine is investigating nanolignin as an antioxidant agent, hydrophobic drug carrier, and component of bioactive hydrogels. Nanolignin's ability to interact with metals makes it useful in adsorption processes for water treatment and soil remediation. Positive effects have been demonstrated as a coating for antimicrobial agents, sustainable cosmetic formulations, and emulsion stabilizers (Bueno-Suescun et al., 2025).

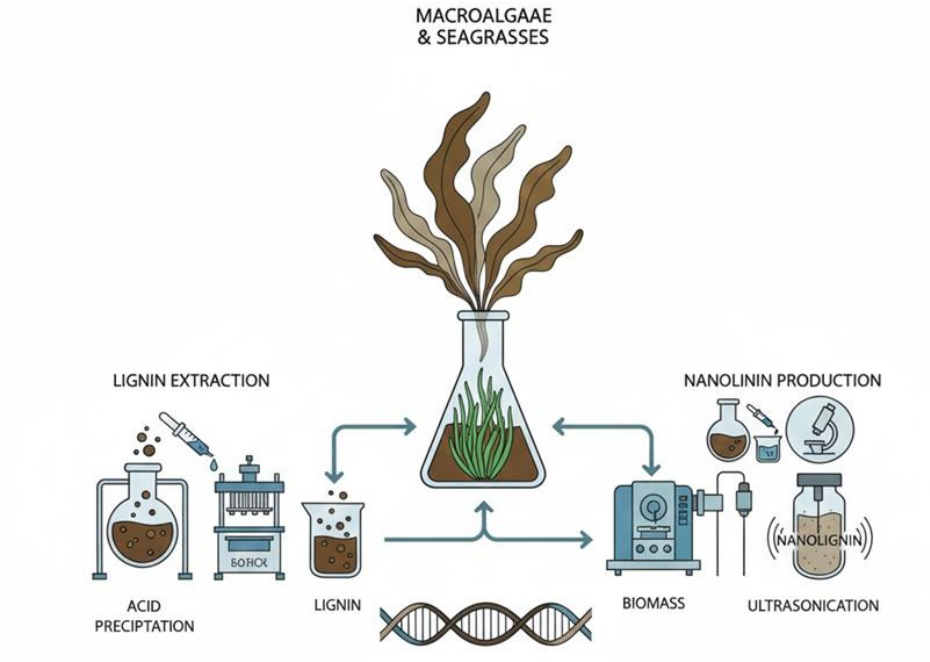

Fig. 6. Processes for obtaining lignin and marine nanolignin from macroalgae and seagrass.

Conclusions

Blue cellulose, obtained from marine biomass, represents a paradigm shift in the bioeconomy. This unconventional marine source competes with terrestrial crops, offering a sustainable and abundant way to obtain biomaterials. Its importance lies in the fact that it allows the

production of microcellulose and nanocellulose, materials with superior properties of high strength and surface area. Thus, blue cellulose and its nanostructured derivatives are key to decarbonizing industry and moving towards a circular economy based on renewable ocean resources.

Blue cellulose and its different scales stand out for their physicochemical properties, derived from the structural adaptability of marine organisms. Marine cellulose has a high degree of crystallinity, enabling the development of biodegradable and biocompatible materials, while micro- and nanocellulose (extractable from microalgae and cyanobacteria) expand the capabilities for contributions to tissue engineering, controlled drug delivery, sustainable packaging, and the development of the advanced materials industry.

Marine lignin is an emerging frontier in blue biotechnology. Complex aromatic structures offer opportunities in the development of biopolymers, natural antioxidants, and advanced functional materials. However, it should be noted that its use faces technical challenges due to biological variability, brine treatments, and industrial scaling costs.

Marine biomaterials are renewable resources with high added value, favoring the transition to a circular economy and enhancing the value of marine resources. Optimizing their extraction, purification, and modification processes will expand their impact on the development of green technologies, turning marine ecosystems into strategic pillars of technological innovation and sustainable development.

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