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Abstract: Polymeric materials such as hydrogels have become necessary in our daily lives. Hydrogels are three-dimensionally cross-linked polymeric networks that can absorb large amounts of water. Thanks to their properties, hydrogels are very useful in multiple application fields. However, they have usually been obtained from petroleum-derived polymers, contributing to plastic pollution and global warming. In this context, biomass has emerged as an appropriate alternative for the production of chemicals, building blocks and biopolymers. Lignin is the second most ample and disposable biopolymer in the world. This biopolymer has been underutilised until recent decades, but as it has demonstrated to provide composite materials such as hydrogels with excellent features, its valorization would mean a step forward in sustainability and circular economy. Hence, this review focuses on the state-of-art of hydrogels and lignin-hydrogels, as well as on their current applications.

Keywords: biopolymers, hydrogels, bioeconomy, sustainability, biomass

Synthesis of advanced bio-based green materials from renewable biopolymers

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Abstract

Polymeric materials such as hydrogels have become necessary in our daily lives. Hydrogels are three-dimensionally cross-linked polymeric networks that can absorb large amounts of water. Thanks to their properties, hydrogels are very useful in multiple application fields. However, they have usually been obtained from petroleum-derived polymers, contributing to plastic pollution and global warming. In this context, biomass has emerged as an appropriate alternative for the production of chemicals, building blocks and biopolymers. Lignin is the second most ample and disposable biopolymer in the world. This biopolymer has been underutilised until recent decades, but as it has demonstrated to provide composite materials such as hydrogels with excellent features, its valorization would mean a step forward in sustainability and circular economy. Hence, this review focuses on the state-of-art of hydrogels and lignin-hydrogels, as well as on their current applications.

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1. Introduction

Currently, synthetic polymer-based materials such as hydrogels have become indispensable for our everyday lives. These materials are usually obtained by polymerizing low molecular weight organic compounds which derive from fossil resources. Hydrogels are considered three-dimensionally cross-linked polymeric networks that contain many hydrophilic groups which allow huge absorption of water molecules within their porous structure without being dissolved [1]. Apart from their excellent swelling properties, these materials stand out for their morphology as well as for their resistance to compression [2]. Different properties (control diffusion process, response to different factors -ionic strength changes, pH and/or temperature- and capacity to trap chemical species) [2] have made that hydrogels grow in popularity in the last decades. Due to their outstanding features and the wide variety of substrates and forms they can be presented in, hydrogels have been applied in multiple fields such as personal hygiene, agriculture (water retention), environmental remediation (CO_2 capture) and biomedicine (drug delivery, among others) [1]. In fact, their global market represented in 2016 a total of $15.6 billion and the market should total $22.3 billion in 2022 [3]. However, the high cost related to the complex production process of hydrogels, coupled with environmental hazards from disposable synthetic hydrogel products, are some of the major drawbacks restraining the growth of the market globally.

Since the middle years of the 20th century, the petroleum-derived chemistry, refinery and engineering processes have been the basis of the polymer industry [4]. The first decades of the 21st century have led to a big economic growth together with an irrecoverable environmental concern in which plastic pollution has been remarked as a global crisis. The latest involves plastic's whole life-cycle, i.e. from their production to their disposal and incineration. In this context, the development of biodegradable polymers has become one of the most suitable alternatives in this industry to face ecological problems [4], contributing at the same time both to sustainability and circular economy [5]. Therefore, the urgent transition from a petroleum-based to a bio-based economy has marked the beginning of the current century, shifting the type of carbon-resources from fossil to natural and renewable ones [6].

In the search of an appropriate solution to the exposed problem, biomass has resulted to be an alternative to petrochemicals for the production of chemicals, sugars and biopolymers [6]. Biopolymers are classified as natural polymers produced by plants, animals and microorganisms, which present biodegradability [4] as their main advantage. More specifically, lignocellulosic feedstock has emerged as a promising, renewable and vast source for chemicals. This biomass is mainly constituted by carbohydrate polymers (cellulose and hemicelluloses) as well as by aromatic polymers (lignin and tannins), which can be isolated and employed to form hydrogels without competing with other biopolymers with food applications such as starch. Although biopolymer-based hydrogels can sometimes exhibit poorer mechanical properties than synthetic ones, many strategies have recently been explored to overcome this drawback [7] and, for this reason, renewable polymers are showing the tremendous potential to replace traditional polymers due to their capacity for reducing energy consumption and pollution.

Among the above mentioned biopolymers, lignin, just after cellulose, is the second most plenty and fascinating terrestrial biopolymer [8]. In fact, about 150 billion tons of lignin are annually synthesized by Earth plants [9]. This intricate aromatic macromolecule is mostly constituted by three different types of phenyl propane monomers (p-coumaryl, coniferyl and sinapyl alcohols) which are cross-linked through different stable bondages types [8]. Lignin can be obtained by several isolation methods using different raw materials such as agricultural residues, dedicated crops or wood [8]. Due to this, it is widely available as an important by-product from different industries, such as pulp and paper industry, among others, and it can be specifically produced in treatments of the new biorefinery schemes.

In the last ten years, the development of biorefinery approaches for biomass revalorization has increased the academic and industrial research about lignins. In this sense, renewable lignin as a new feedstock for bio-materials' synthesis is being investigated for its positive effect on the thermal behaviour, hydrophilicity, biocompatibility and biodegradability of these systems [10]. Furthermore, its three-dimensional structure makes lignin suitable as a new potential crosslinking component for the development of hydrogels. Thus, the formulation of these materials has experimented a rapid shift from synthetic polymers to biopolymers [11].

Since less than 2% of the total amount of lignin generated by year in pulp and paper industry has an added-value [8], the development of new green materials from lignin has great attraction from an environmental and economic point of view. Limited oil reserves, the environmental problems caused by the toxic compounds from the degradation of this type of materials together with the expanding hydrogel market generate an urgent need for the development of environmentally friendly and safe alternative hydrogels from renewable materials, which would also expand the life cycle opportunities for reuse and recycling of lignocellulosic biomass and contribute to circular bioeconomy (see **Figure 1**). Hence, the novelty and potentiality of this review are focused on the use of this biopolymer for the synthesis of hydrogels and their applications in several fields to promote the sustainable development.

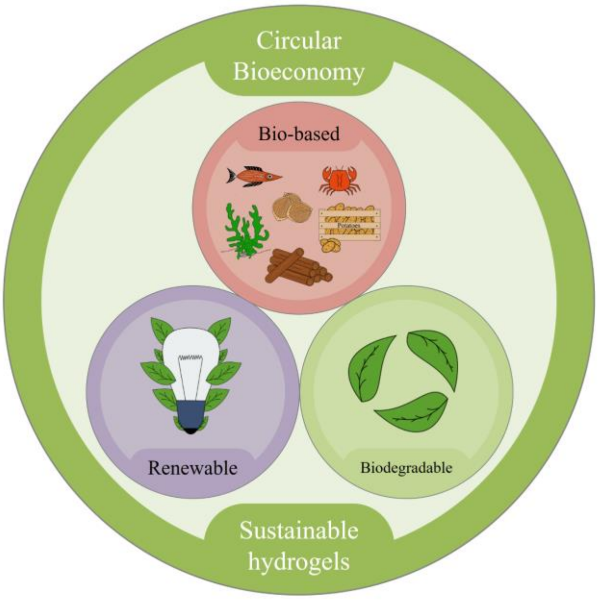

Figure 1. Graphical definition of sustainable hydrogels.

2. Hydrogels and Lignin

Over the last few years, studies on biopolymer-based hydrogels have become very popular due to their applicability in the fields of food, cosmetics, pharmaceuticals, and biomedical implants [12]. Besides, the integration of biopolymers together with other molecules can improve the functionality of the resultant hydrogels, making possible to formulate hydrogels capable of imitating the interrelated nature of human tissue [13]. Among these biopolymers, lignin is the only aromatic one, which makes it interesting for its application on functional hydrogels [12]. Although there is not a defined structure for lignin [2], many studies have recently demonstrated that it can be successfully employed in for the formulation of lignin-derived composites used for tissue scaffolding and drug release or as UV absorbents, for instance [14]. Thus, if adequate routes for improving the handling of lignin and the development of technologies to process it were further investigated, this natural polymer would undoubtedly play an important role in the synthesis of new materials [15]. A clear increasing trend of the publications about lignin and lignin applications in the last years has been reported [9,15,16].

The lignin extraction method determines the type and features of the resultant lignin, which can be more or less reactive depending on the used technique. The typical types of lignin employed for hydrogel synthesis are Kraft, lignosulphonate, alkaline and organosolv [2,12]. In any case, there are usually two pathways to valorize this biopolymer: the first one that uses the lignin as a macro-polymer (to obtain valuable materials) whereas the second one implicates the lignin depolymerization (to obtain low-molecular weight monomers) [16]. In the case of hydrogels, lignin is usually combined with other bio- or synthetic polymers due to the lack of self-gel-forming properties. For this reason, it has been included in different polymeric networks through physical interactions or chemical reactions which promote hydrogels structure [2].

3. Applications of lignin-hydrogels

A big part of the synthesized lignin-based hydrogels has been used to remove heavy metal and cationic compounds presented in aqueous solutions [17]. Nonetheless, Mohammadinejad et al. (2019), also report their use on fields such as biomedicine for drug delivery and tissue engineering. Some hydrogel applications studied till date are described below.

The controlled release of water is a commonly used term in agriculture. In climates where the rain is scarce, soils need to be irrigated frequently. Nevertheless, equipping soils with small and smart hydrogels, such as lignin-hydrogels, have proven to absorb significant amounts of water and retain it until the crops require it [18]. The water content of hydrogels can vary according to their degree of cross-linking, which can be controlled during their synthesis. Mazloom *et al.* (2019, 2020), for instance, synthesized a lignin-based hydrogel (lignin alkali polymers-poly(ethylene glycol) diglycidyl ether) hydrogels and then tested them as controlled water releasers in agricultural soils, which resulted in promising findings [19, 20]. Meng *et al.* (2019) also prepared some lignin-hydrogels from the copolymerization of acrylic acid and the spent sulphite cooking solution (or red liquor) directly obtained from the paper industry [21]. The materials developed by Meng *et al.* exhibited super-swelling and slow release behaviours in water. Song *et al.* (2020) combined lignin, sodium alginate and konjaku flour for the synthesis of hydrogels with excellent soil conditioning properties tested with tobacco plants [22].

Lignin has functional groups (phenolic hydroxyl, alcohol hydroxyl, methoxy and carboxyl groups) that can act as adsorption sites for dye molecules and heavy metals [23]. According to Meng *et al.* (2019), these structures are therefore capable of attracting positive molecules through the negatively charged functional groups and aromatic organics via π-π reactions [18]. For this reason, they have been used for the absorption of both inorganic ions and organic compounds in soils and water [18]. Table 1 summarises some works in this field:

Table 1. Literature review of hydrogels as pollutant removers.

Lignin type in hydrogels	Pollutant	Medium	Removal	Reference
Sodium lignosulfonate	Cadmium ions (Cd^{2+})	Soil	< 61.77 ± 1.09 mg/g	[24]
Lignin	Methylene blue (MB) and lead ions (Pb^{2+})	Aqueous	201.7 mg/g MB and 753.5 mg/g Pb^{2+}	[25]
Alkali, Kraft and organosolv lignins	Toluene	Aqueous	164 –170 mg/g	[26]
Aminated alkali lignin	Heavy metal cations (Pb^{2+}, Hg^{2+} and Ni^{2+}) and dyes (Methylene blue, methyl, orange and malachite green)	Aqueous	2.1–55 mg/g for heavy metal cations and 2–155 mg/g for dyes	[27]
Alkali, Kraft and enzymatic hydrolysis lignin	Rhodamine 6G, crystal violet, methylene blue and methyl orange	Aqueous	10–196 mg/g	[23]
Soda, Kraft and organosolv lignins	Methylene blue	Aqueous	69 to 629 mg/g	[28]
Pine Kraft lignin Indulin AT	Prednisolone drug and 3,4-dichloroaniline	Aqueous	1.35 mg/g for Prednisolone and 4 mg/g for 3,4-dichoroaniline	[29]
Alkali lignin	Hexavalent chromium Cr(VI)	Aqueous	599.9 mg/g	[30]
Acid-pretreated alkali lignin	Pb^{2+}, Cu^{2+} and Cd^{2+} ions	Aqueous	1.076 mmol/g for Pb^{2+}, 0.3233 mmol/g for Cu^{2+} and 0.059 mmol/g for Cd^{2+}	[31]

Thus, according to Andrade Batista *et al.* (2019), hydrogels would be an attractive alternative for their application in food packaging systems as absorbent pads. Apart from water absorption properties, these materials must increase the stored food products shelf-life and avoid microbial growth on food surface [32]. Therefore, the antimicrobial activity of the polymers constituting the hydrogel network is crucial. In this context, lignin and its derivates, and lignin-hydrogel coatings have demonstrated to present antimicrobial behaviour [33–35]. For this reason, Yang *et al.* (2018) proposed that their lignin-hydrogels with antioxidant and antimicrobial properties could be employed in food packaging.

Antimicrobial, antioxidant and low cytotoxicity features are also important for the synthesis of biocompatible materials. In this sense, hydrogels can provide effective removal of undesirable metabolites from wounds due to their high absorption capacity, and the addition of lignin may help to protect the wound from future problems such as further injury or contamination thanks to its good mechanical properties [16]. Hence, lignin has also gained interest in wound dressing [36] and tissue engineering [37].

Lignin-hydrogels have also demonstrated to be useful in another important part of the biomedical field: the controlled release of both hydrophobic [34] or hydrophilic drugs. Stimuli-responsive hydrogels can be created by combining other (bio)polymers and lignin. These hydrogels are of great interest in drug delivery since they are able to alter their volume in response to environmental stimuli [38] such as pH, temperature, or light [2]. Some authors, for instance, have studied the release kinetics of curcumin loaded hydrogels at simulated human body conditions [34]. They concluded that as the lignin content was higher, the hydrogels were loaded with higher amounts of the drug, promoting a higher release of it [34].

Finally, the use of lignin-based materials applied to energy devices has proven to be promising [39]. Among other applications, these materials have gained attention as electrode materials for supercapacitors as a result of their high surface areas and chemical stability, and superior electrical conductivity [39]. Liu *et al.*, for instance, created hybrid double-cross-linked lignin-hydrogels by combining corncob lignin media with poly (ethylene glycol) diglycidyl ether in alkaline media and immersing them in sulphuric acid afterwards. These authors reported not only excellent electrochemical performance for their supercapacitors but also improved mechanical properties [40].

Conclusions

Taking all the above mentioned into account, it can be concluded that, although lignin has been underused in the past, it presents multiple advantages not only from the economical (e.g. high availability and low cost) perspective but also from the environmental (i.e. renewable and biodegradable) and technological (i.e. excellent physicochemical features) points of view, making them greatly potential for many applications. Although lignin-based materials are still at early stages in the global market, many spin-offs, start-ups and companies have placed their bet on them, producing lignin aerogels (Aerogelex UG., Hamburg–Germany) for instance. A deeper understanding of the chemical structure and physicochemical properties of lignin would make it even more interesting and useful for the synthesis of globally used commodities in the near future. Thus, the standardization of lignin-manufacturing processes would lead to the synthesis of bountiful sustainable products such as thermoplastic and elastomeric materials for automotive or building applications, for instance, as well as to the production of materials for packaging and electronic applications. Therefore, it is important that research continues in this field to create green and sustainable materials for the substitution of conventional ones.

Declaration of interest:

None.

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