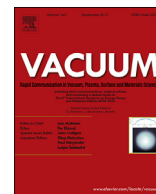




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Progress in lignin hydrogels and nanocomposites for water purification: Future perspectives

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ABSTRACT

Lignin is one of the most abundant natural polymers after cellulose and has attracted great attention in academia as well as in industry for miscellaneous applications. Lignin also exhibits very high potential as a renewable sustainable resource for a galaxy of high value biochemicals. Due to the typical structure of lignin, it can be converted into different types of useful products. From a ligno-cellulose bio-refinery prospective point of view, it can be regarded as an economic bio product and can also be used as filler in several polymers to form blends or composites. Recently lignin-based hydrogels have shown excellent performance for removal of various pollutants from water. The adsorption properties of lignin based hydrogels can further be improved by using a combination of nanomaterials and lignin that results in promising hydrogel nanocomposites. In nature, the most abundant structures are formed by the combination of lignin, cellulose and hemicelluloses. In this article, we have attempted to comprehensively review the research work carried out in the direction of usage of lignin-based hydrogel for removal of toxic pollutants including metal ions and dyes.

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1. Introduction

Hydrogels belong to a category of soft matter comprising a polymeric cross-linked three-dimensional structure [1,2]. The characteristics of hydrogels such as flexibility, elasticity and permeability are attributed to their high water absorption ability [3]. They show both half liquid-like and half solid-like properties [4,5]. The three-dimensional structure of hydrogel is generally organized by (a) chemical cross-linking and (b) physical cross-linking [6,7]. Coordination bonds, electrostatic interaction, hydrogen bonding and hydrophobic interaction are responsible for the formation of physically cross-linked hydrogels. The aqueous

solution of amphiphilic organic molecules [8] or inorganic nanoparticles [9] can produce hydrogel through supramolecular chemistry. Physically cross-linked hydrogels have a sol–gel phase transition [10] on exposure to external stimuli [11,12]. On the other hand, a change in external conditions such as pH [13], electric field [14], temperature [15,16] and ionic strength [5] leads to the volume phase transitions [17] in chemically cross-linked hydrogels. These hydrogels exhibit permanent three-dimensional networks. Because of these features, hydrogels have potential use in various applications such as superabsorbents, bio-actuators, microfluidic devices, packaging materials catalyst supports and biomedical materials [18–26]. The super absorbent characteristics of hydrogels are attributed to their ability to absorb huge quantity of water, about 10–20 times as compared to their original molecular weight, and, after absorbing water, the hydrogel achieves a swollen state [27,28]. On the other hand, Xerogels (an important class of gels) may be defined as hydrogels without water, i.e. dried hydrogels, where water is removed from the hydrogels network, whereas aerogels exhibit the removal of water molecules from a hydrogel network with no deformation of structure [29]. The diffusion process of

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hydrogels depends on their structure and the thermodynamic behavior of components used in the formation of the hydrogel network [7]. The swelling capability of a hydrogel is linked to the porosity of the network and porosity generally depends on (a) cross-linking density and (b) attraction between hydrogel and aqueous solution. The porosity of structure also permits the adsorption of pollutants from wastewater or water [30,31]. Water exists in different forms in the network of the hydrogel. The presence of water in hydrogel can be divided into four types [32]: (a) free water present in the outer part that can be removed easily or easily extractable bulk water from the hydrogel structure, (b) physically entrapped water, existing in interstices of the network, named interstitial water, (c) chemically attached bound water through functional groups and linked directly to the network, it cannot be removed under normal conditions from the hydrogel network, (d) semi-bound water, neither free nor bound water, but existing in between these two types of water. A Differential Scanning Calorimeter (DSC) thermogram can be used to identify and characterize the water present in hydrogel [33]. The development of the network of hydrogel depends on the kind and quantity of cross-linker used in the formation of the hydrogel. Different types of cross-linkers are required for different applications [32]. Cross-linkers can be classified as (a) physical and (b) chemical cross-linkers. Physical cross-linking includes hydrogen bonding, freeze thawing, thermo gelation and charge interactions [34–39]. Chemical cross linking in hydrogels may be due to the radical polymerization, high energy irradiation, enzymes and reaction of functional groups [40–44]. Insolubility of hydrogels is because of the presence of chemical and physical cross-links. At an equilibrium swelling state, dispersion and cohesive forces of hydrogel become equal [32,45]. Dipole–dipole, electrostatic and hydrophobic forces can contribute to cohesive forces, a covalent bond is usually responsible for cohesive forces. Biopolymer-based materials such as hydrogels and fibres are a currently highlighted topic and represent the most promising bio-renewable materials for various applications [46–49]. Tunable and degradation characteristics are advantages of bio-polymer based hydrogels. The use of lignin for different applications has been reported earlier and well-reviewed due to the inherent advantages of lignin [48,50–56]. In this article, we will primarily focus on the recent progress carried out in lignin based hydrogel material for water treatment.

2. Lignin structure

In 1838, the structure of wood was first elucidated by Anselme Payen, a French scientist [57]. He obtained a material by reacting wood with nitric acid which looks like a thread called cellulose. There was a substance enriched in carbon even after the extraction of cellulose. Anselme Payen used a better extraction process and successfully obtained a new encrusting substance named lignin [56]. It was found in 1890 that lignin did not belong to polysaccharides and cellulose and lignin were differing by its methoxy groups [56]. Lignocellulosic biomass has attracted great attention because of its accessibility and lack of contest with supply of feed [58–63]. Lignocellulosic biomass is made up of three components [64]: (a) cellulose, (b) hemicellulose and (c) lignin (Fig. 1) [65–68]. Study regarding lignin is still going on because of its specific characteristics.

Lignin is found in plants and most of the plant biomass can consist of 30% of lignin [69]. In the earth, after cellulose, lignin is most abundant aromatic natural complex polymer [52,70,71]. Strength, integrity and inflexibility [72] of plant cell are provided by binding of lignin with cellulose and hemicelluloses [73–75] as shown in Fig. 2a. Several pre-treatment processes are used for the extraction of lignin from different lignocellulosic resources. Most of

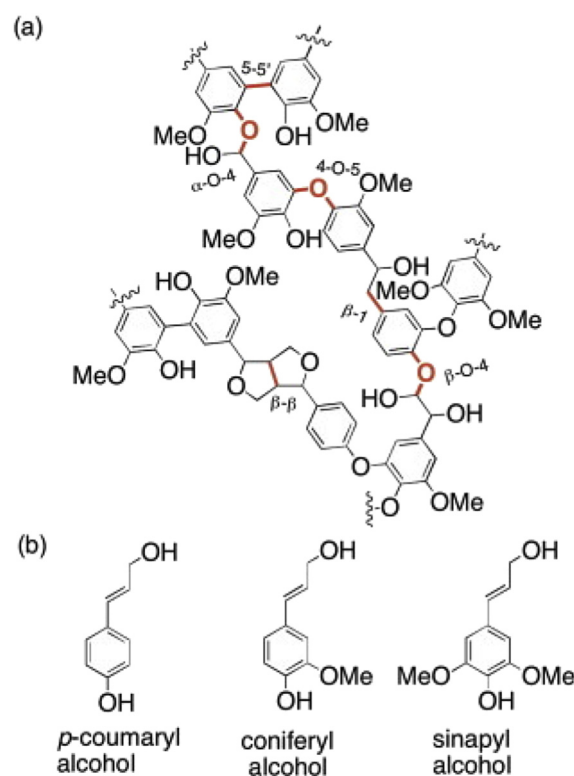


Fig. 1. (a) Proposed lignin structure with several linkages; (b) three primary alcohol structures in lignin [55]. Reprinted with permission [55]. Copyright 2016 Elsevier.

the currently used pre-treatment processes have been found to generate toxic compounds and inhibitors during the lignin processing (Fig. 2b).

Amorphous lignin polymer has significant use in fluid flow, pathogen resistance and response to stress [76]. Such a lignin-polysaccharides network helps in bio-refining by cell walls. Monolignols—p-coumaryl, sinapyl and coniferyl alcohols [77,78] are the three phenylpropanoids from which lignin is derived giving p-hydroxyphenyl (H), syringyl (S) and guaiacyl (G) units, respectively. Generally, the linkages β-O-4', β-5', α-O-4', 4-O-5', β-β' exist in between the sub-units, β-1' and 5-5' linkages also exist but in small amount. 0–5/95–100/0, 0–8/25–50/46–75 and 5–33/33–80/20–54 are the H/G/S ratio in softwood, hardwood and grasses respectively [79]. The structure of lignin can be best depicted by a common empirical chemical formula. The common empirical chemical formula is based on the C₉ formulae which represent the nine carbon atom molar ratio with respect to other elements. The formula shows common repeating units which are formed by monolignols. C₉H_{7.92}O_{2.40}(OCH₃)_{0.92} and C₉H_{7.49}O_{2.53}(OCH₃)_{1.39} [80] are the C₉ formula of softwood and hardwood lignin, respectively. The difference between softwood and hardwood is due to their different monolignols ratio.

Freudenberg [81] (1965), Adler [82] (1977), Brunow et al. [83] (1998) and Gellerstedt [84] (2007) respectively developed structure for spruce (softwood) lignin and a beech (hardwood) lignin structure was produced by Nimz [85] (1974). According to Adler [82] and Gellerstedt [84], models for structure of lignin can be represented in different ways. The structure of type 1 and type 2 lignin existing in plants was given by Gellerstedt et al [84,86]. The weight/weight percentage (w/w %) of spruce lignin is 48 in type 1 whereas it is 40 in type 2 [84,86]. Glucmannan-lignin-xylan (w/w = 9:9:1) and xylan-lignin-glucmannan (w/w = 2:3:1) are the complexes containing type 1 lignin and type 2 lignin, respectively.

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