



# Effective purification of oily wastewater using lignocellulosic biomass: A review

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## ABSTRACT

Due to the frequent occurrence of oil spills and the large-scale production of oily wastewater, the treatment of oily sewage has become an important issue for sustainable development. Recently, materials prepared from lignocellulosic biomass (LCB) for oil-water separation have been found to be effective due to their high separation efficiency, good recyclability, and superior sustainability. However, few reviews have focused on the advantages and limitations of LCB for sewage treatment. This review summarizes the performance of modified LCB in oily wastewater treatment, in terms of the advanced modification methods applied and the structural dimensions of LCB materials according to the principle of superwetting oil-water separation. Research on the preparation technologies, separation mechanisms, and treatment efficiency of different LCB materials are briefly summarized, along with the characteristics of different LCB material types for oily wastewater treatment. Finally, the future prospects and challenges faced in the development of LCB materials are discussed.

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## 1. Introduction

The increasing global demand for energy, along with rapid social and economic development, has resulted in a continual increase in the discharge of oily industrial wastewater and domestic sewage in recent years [1,2]. Human survival and the health of most habitats worldwide are dependent in some manner on ocean ecosystems and therefore, oil spills or the release of untreated oily wastewater is a critical issue for marine ecosystem [3,4]. Oily wastewater and oil spills introduce toxic compounds into terrestrial and marine environments, threatening species from every level of the trophic system from the bottom (e.g., algae) to the top (e.g., mammal), including humans. The improper handling of oil-water mixtures causes significant harm to both the waterbody and the surrounding ecological environment [5–8]. Therefore, the materials and technologies applied to oily wastewater treatment are receiving increased research attention and development.

Oily wastewater can be categorized as a simple oil-water mixture (oil-water stratification), a highly dispersed (uniform) mixture, or a miscible (homogeneous phase) mixture, according to its macroscopic physical state [9,10]. The primary oil pollutants in do-

mestic and industrial sewage are the lipids formed from biological fats and petroleum, respectively, including fatty acids, fats, waxes, and other similar substances. Crude oil is a mixture containing water, which is mainly composed of alkanes, cycloalkanes, aromatics, and unsaturated hydrocarbons [11,12]. In addition to the pollution caused by oil itself, various pollutants dissolved in oil can also induce serious toxic effects, such as organic pesticides, polycyclic aromatic hydrocarbons, aromatic hydrocarbons, polychlorinated biphenyls, and dyes [13,14]. The traditional treatment technologies used for oily wastewater treatment generally adopt physical methods such as gravity sedimentation and coarse-grained oil removal. More sophisticated treatment methods utilize physical (air flotation and filtration), chemical (flocculation and chemical oxidation), and biological (active sludge and biofilms) methods [15,16]. However, these traditional methods are time-consuming, labor-intensive, and incur high operational costs. Furthermore, the additional use of fillers such as flocculants, produces secondary pollution and results in the need for stringent control mechanisms [17].

Degradable lignocellulosic biomass (LCB) material has exhibited good potential for practical application for the purification of oily wastewater [18,19]. It is attractive to use LCB as a promising candidate material for the purification of oily wastewater [20,21]. The different components and structures of LCB are powerful modification platforms for technological development, which can be

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selectively modified according to specific requirements, allowing an advanced level of oily wastewater treatment that achieves effective separation and purification. A wide variety of functional materials have been manufactured for oily wastewater treatment; however, few review articles are available on the application of LCB in this field. In this review, we summarize the latest research progress in the preparation of materials from LCB for oily wastewater treatment. The mechanism of action and the wettability of LCB materials which are beneficial for oily wastewater treatment are discussed initially, followed by the advantages and disadvantages of their application for oily wastewater treatment, in view of the structures and compositions of LCB. Based on this, new methods of LCB material modification for oily wastewater treatment were reviewed, summarizing the oil-water separation efficiency and recycling performance of LCB materials with 0-dimensional (0D) powder, 2-dimensional (2D) membrane, and 3-dimensional (3D) structures. In particular, the emerging materials, derived from LCB and exhibiting special wettability performance for oily wastewater treatment are discussed, demonstrating the relationship between the material structure and oil-water separation efficiency. Finally, the potential for large-scale application of LCB materials is summarized. By assessing the existing methods of LCB material preparation, this review is intended to provide clear information about the latest technological developments on the use of LCB biomass materials for oily wastewater treatment, and provide reference material for scientists investigating how to achieve optimized, low-cost biomass capable of high efficiency oil-water separation.

## 2. The mechanism of superwettability for oily wastewater treatment

Adjusting the wettability of materials is an effective method to improve oil absorption capability [22,23]. Materials with superhydrophobic and superlipophilic properties can simultaneously repel the water phase and make it easier for the oil phase to diffuse, absorb and penetrate substances, achieving oil-water separation [24]. The wettability of materials is affected by the surface roughness and chemical composition. Fig. 1a presents a schematic of the effect of contact angle (CA). The wetting system can theoretically be divided into three-phase gas/liquid/solid and liquid/liquid/solid systems according to the different media in contact with the func-

tional surface, according to Young's equation (Eq. 1) [25]:

$$\gamma_{SV} = \gamma_{SL} + \gamma_{LV} \cos\theta \quad (1)$$

where,  $\gamma_{SV}$ ,  $\gamma_{SL}$  and  $\gamma_{LV}$  represent the surface tensions of solid-gas, solid-liquid, and gas-liquid interfaces, respectively. Young's equation is only applicable for smooth, non-deformed, and ideal isotropic surfaces, while for the non-ideal solid surfaces, the Wenzel equation must be applied (Eq. 2) [26]:

$$\cos\theta_r = r \cos\theta \quad (2)$$

where,  $\theta_r$ ,  $\theta$  and  $r$  represent the apparent contact angle, balanced contact angle, and roughness, respectively. Wenzel model is shown in Fig. 1b. Considering the non-ideality of solid surfaces, Eq. 2 was optimized, assuming that the material surface consists of two substances (1 and 2) as shown in Fig. 1c, with the intrinsic contact angles of substance 1 and 2 being  $\theta_1$  and  $\theta_2$ , respectively. Therefore, the Cassie-Baxter equation was proposed (Eq. 3) [27]:

$$f_1 \cos\theta_1 + f_2 \cos\theta_2 = \cos\theta_r \quad (3)$$

where  $f_1$  and  $f_2$  represent the surface area fractions of two substances on the solid surface ( $f_1 + f_2 = 1$ ). When the solid surface possesses strong hydrophobicity, the droplets would have non-wetting contact with the solid surface (Fig. 1d), with  $f_1$  and  $f_2$  defined as the surface area fractions of liquid-solid and droplet-air pore contacts ( $f_1 + f_2 = 1$ ), respectively. The CA between droplet and the air is  $180^\circ$  and therefore, Eq. 3 can be developed to form Eq. 4 as follows:

$$\cos\theta_r = f_1 \cos\theta_1 - f_2 \quad (4)$$

According to Eq. 4, the hydrophobicity of the material is enhanced with an increase in  $f_2$ . The wettability of the material surface, therefore, can be reasonably designed according to the Cassie-Baxter equation (Eq. 3) in order to endow materials with the superwettability required for effective oil-water separation [28,29].

For the liquid/liquid/solid three-phase wetting system,  $L_1$ ,  $L_2$ ,  $S$ , and  $V$  are expressed as the liquid phase 1, liquid phase 2, ideal solid surface, and gas phase, respectively. Schematics of the CA in different three-phase systems are shown in Fig. 1e (liquid 1 in air), Fig. 1f (liquid 2 in air), and Fig. 1g (liquid 1 in liquid 2). Introducing the Young's equation (Eq. 1) into this three-phase system generates Eqs. 5–7 as follow:

$$\gamma_{SV} = \gamma_{SL1} + \gamma_{L1V} \cos\theta_1 \quad (5)$$

$$\gamma_{SV} = \gamma_{SL2} + \gamma_{L2V} \cos\theta_2 \quad (6)$$

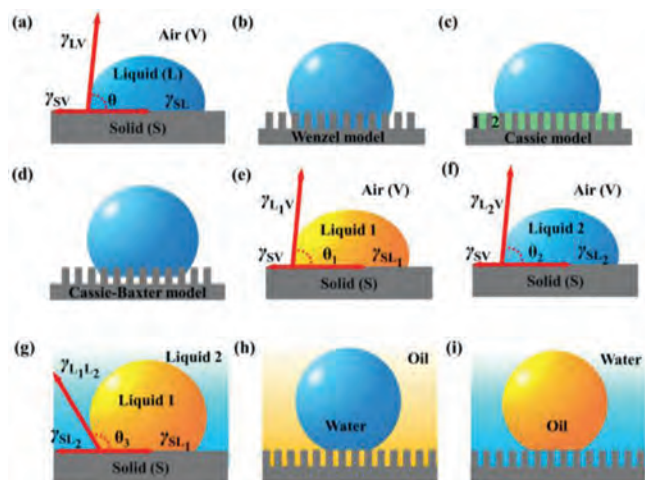
$$\gamma_{SL2} = \gamma_{SL1} + \gamma_{L1L2} \cos\theta_3 \quad (7)$$

Combining Eqs. 5–7 results in Eq. 8 as follows:

$$\cos\theta_3 = \frac{y_{L1V} \cos\theta_1 - y_{L2V} \cos\theta_2}{y_{L1L2}} \quad (8)$$

where, the  $\theta_1$ ,  $\theta_2$  and  $\theta_3$  individually represent the CA of liquid 1 to the solid surface in air, the CA of liquid 2 to the solid surface in air, and the CA of liquid 1 to the ideal solid surface in a liquid/liquid/solid three-phase system. Eq. 8 can describe the wetting behavior of smooth, non-deformed, and isotropic ideal surfaces in a liquid/liquid/solid three-phase system, with  $\theta_3$  calculated from  $\theta_1$  and  $\theta_2$ , as the liquid phases are known [30,31].

For the wettability model of liquid/liquid/solid three-phase systems with a rough surface, the Cassie-Baxter equation (Eq. 3) should be introduced into the system for derivation. The state of rough solid surfaces in the liquid phase can be regarded as a binary composite interface formed between one liquid and the solid surface, resulting in the contact state between the other liquid and the solid surface being referred to as the Cassie state. In



**Fig. 1.** Schematic diagram of three-phase wetting system. (a) Schematic of the contact angle. (b) Wenzel model. (c) Cassie model. (d) The transitional state between Wenzel's and Cassie's states. (e) Schematic of the contact angles of liquid 1 in air. (f) Schematic of the contact angles of liquid 2 in air. (g) Three-phase system of liquid 1 in liquid 2. (h) Underoil superhydrophobic surface. (i) Underwater superoleophobic surface.

this case,  $f_1$  is defined as the surface area fraction of contact between one liquid and the solid, while  $f_2$  is defined as the surface area fraction of contact between the other liquid and the solid ( $f_1 + f_2 = 1$ ), with the CA of the droplet to another liquid phase considered to be  $180^\circ$ . Thus, the wettability equation for the liquid/liquid/solid three-phase system with a rough surface can be obtained, as shown in Eq. (9):

$$\cos \theta'_3 = f_1 \cos \theta_3 + f_1 - 1 \quad (9)$$

where,  $\theta'_3$  represents the CA of liquid 1 to the rough solid surface in the system [32,33].

The above equations illustrate the theoretical wettability of three-phase systems. Materials selectively absorb water or oil on their surface as they possess higher hydrophilicity and oleophobicity or lipophilicity and hydrophobicity, respectively, resulting in oil-water separation. In liquid/liquid/solid three-phase systems, the underoil superhydrophobic (Fig. 1h) and the underwater superoleophobic surfaces (Fig. 1i) are the two surfaces with mutually contradictory wettability characteristics, which depend on an exact match of the materials surface geometry and chemistry [34–36]. Generally, the oil-water separation effect can only be achieved when the material surfaces show the opposite wettability for water/oil. The superhydrophobic/superlipophilic surfaces are conducive for oil filtration and absorption, while the underoil superhydrophobic and the underwater superoleophobic surfaces possess more comprehensive oil-water separation ability, the former and the latter are mainly used to separate water-in-oil and oil-in-water mixtures, respectively. In the next section, we will elaborate on various new modification methods for LCB, so that the readers can quickly understand the methods based on reasonable adjustment of the wettability of LCB for the oily wastewater treatment.

### 3. The use of LCB as an applied material for oily wastewater treatment

#### 3.1. LCB components and pretreatment methods

##### 3.1.1. LCB components

LCB is a main component of plant resources and its effective utilization is an important way of developing environmentally sustainable industrial practices, as the biomass resource LCB usually has surplus availability [37]. LCB is composed of cellulose, hemicellulose, and lignin as its three major components, with the proportion of cellulose being the highest (35%–50%). Cellulose is a polymer (up to 10,000 units) that is connected linearly via  $\beta$ -1,4 glycosidic linkages [38]. Glucose molecules contain a large number of intramolecular and intermolecular hydrogen bonds, which result in the cellulose compound being difficult to degrade. Meanwhile, the large number of hydroxyl groups (–OH) endow the LCB surface with high activity and surface energy. Hemicellulose is an amorphous polysaccharide compound that contains various unit compositions including xylose, glucose, mannose, galactose, and arabinose, along with other types of structural units. The number of sugar units in hemicellulose ranges from 100 to 200, resulting in its molecular weight being far lower than that of cellulose [39]. The hemicellulose component acts as a binder between cellulose and lignin, improving the rigidity of the biomass overall [40]. In contrast, lignin has a 3D-grid polymeric structure without a fixed shape, in which the basic unit is composed of methoxylated phenylpropanoid units including sinaphyl, coniferyl, and *p*-coumaryl alcohols [41,42]. Cellulose and hemicellulose can tightly combine with lignin to form lignocellulose. These components provide plants with the strength and hardness required to resist external forces, while also forming a protective layer to prevent external microbial degradation of polysaccharides [43].

##### 3.1.2. A brief description about pretreatment of LCB

LCB must be pretreated before functionalization. Generally, the pretreatment includes physical, chemical, physicochemical, and biological methods. The particle size and degree of polymerization of the LCB material largely depend on the pretreatment methods applied. [44–46]. Physical pretreatment methods are generally used to reduce the size of biomass, increase the specific surface area and pore size, or reduce the crystallinity and polymerization degree of cellulose. Chemical treatment is mostly performed using acids, alkalis, organic solvents, or co-solvents, which are capable of selectively removing the main structural components [47,48]. Physicochemical approaches include steam explosion, ammonia fiber explosion (AFEX), and liquid hot water (LHW) methods [49,50]. Biological pretreatment allows lignin to be specifically degraded and obtains the corresponding products [51].

#### 3.2. Advantages of LCB application for oily wastewater treatment

The recent rapid development of biomass energy technologies and bionics has provided new ideas for the invention of low-cost materials capable of high efficiency oily wastewater treatment [20,52]. The complete separation of oil-water mixtures using LCB materials without the consumption of any external energy, has become a key research focus.

The linear molecular structure of cellulose ( $\beta$ -D-glucopyranose,  $(C_6H_{10}O_5)_n$ ), is the main component of LCB and is, therefore, difficult to dissolve using common polar and non-polar solvents as the chemical bonds are not easily broken. The strong hydrogen bonding is attributed to the large number of hydroxyl functional groups (–OH) on the surface of cellulose, which make LCB inherently hydrophilic, enabling it to chemically react with different functional groups. The main –OH functional group reactions of LCB include oxidation to aldehyde groups (–CHO), oxidation to carboxyl groups (–COOH), hydroxyl group substitution reactions, alcoholysis with alkoxy groups to form new alkoxy bonds, condensation reactions with phenolic and melamine resins, hydrogen bonding, coordination, electrostatic, and van der Waals interactions. The interconnected lignin, cellulose, and hemicellulose compounds in LCB form a lignin-carbohydrate complex, providing mechanical strength and forming the internal structure of LCB materials. Furthermore, the lignin in LCB contains active groups such as aromatic, phenolic hydroxyl, alcoholic hydroxyl, carbonyl, methoxy, carboxyl groups, and conjugated double bonds, capable of selective functional group chemical reactions [53]. Therefore, suitable reactions can be selected to facilitate the addition or removal of specific functional groups on LCB, supporting further functionalization. For example, the use of different long-chain silanes for hydrolysis and grafting onto the surface of LCB, allows precise control over the wettability of LCB, endowing it with the characteristics required for oily wastewater treatment [54,55]. In addition, the 3D pore structure of the original LCB material makes it suitable as a natural filter and filler material [56,57]. Due to the 3D network structure of LCB, there are an abundance of controllable factors (pretreatment modification, pore shape, length, diameter, etc.), allowing the 3D channels of LCB to be filled using functional particles and surface modifications [58].

#### 4. Advanced LCB modification techniques for oily wastewater treatment

LCB has the advantages of excellent sustainability, degradability, and an abundance of natural resources, resulting in it becoming a sustainable development research hotspot. The different potential wettability characteristics and multilayer structures of LCB materials are the main factors contributing to the oil-water separation

capability. In order to improve the efficiency of oil-water separation, the selection of suitable LCB material modification methods has been found to be the key factor.

#### 4.1. Dip coating

Dip coating is an effective method for superwetting oily wastewater treatment, in which the pretreated LCB substrate is fully immersed into a solution containing micro/nano particles, a low surface energy substance, or a mixture of both, forming an LCB structure with low surface energy and achieving superwettability. The micro/nano particles commonly used to construct micro-nano surface structures include  $\text{SiO}_2$ ,  $\text{TiO}_2$ , ZnO, carbon materials, and metal nanoparticles [59]. Generally, the low surface energy substances used include fluorine-containing silanes, fluorine-containing resins, silicone resins, and fluorine-free long-chain silanes. Wang *et al.* immersed wood in a mixed solution of zinc acetate and triethylamine, modifying the wood with stearic acid to obtain a stable superhydrophobic surface with a water contact angle (WCA) of up to  $151^\circ$  [60]. Furthermore, modifying wood using potassium methyl silicate (PMS) and controlling the dipping conditions, resulted in the formation of superhydrophobic wood with a WCA of  $153^\circ$  [61]. Rahman *et al.* constructed micro-nano structures on a fabric surface by enzymatic hydrolysis and then treated it using the polydimethylsiloxane (PDMS) dip coating method [62]. Superhydrophobic cellulosic biomass could be obtained by optimizing the treatment conditions. In practice, the effectiveness of the dip coating method is limited due to the fragile adhesion between the micro-nano structure and the LCB substrate surface, affecting the stability and recyclability of the modified LCB material, although this limitation can be overcome using other methods and techniques. Overall, the dip coating technique is simple to implement and provides a convenient method for achieving superwettability of LCB and other materials [63,64].

#### 4.2. Spray coating

The spray coating technique atomizes raw materials and applies them to the substrate surface through a spray gun or an atomizer, avoiding the limitations of the substrate and efficiently accomplishing coating on a large-scale. Hydrophobic silica nanoparticles dispersed in a solution can be sprayed on tape to prepare a multifunctional superhydrophobic surface, using a simple and practical method [65]. Xie *et al.* first utilized polydopamine (PDA) coating to generate a double-sided superhydrophilic surface on a regenerated cellulose (RC) membrane, then spraying superhydrophobic attapulgite (SOATP) onto one of the surfaces to finally obtain a membrane with opposing characteristics on each side [66]. This modified RC membrane achieved efficient oil-water separation of up to 99%. Shang *et al.* produced a superhydrophobic cotton fabric by spraying it with a solution composed of the oligomer and hydrophobic  $\text{SiO}_2$  nanoparticles [67]. Due to its excellent superhydrophobicity and superlipophilicity, the functional cotton fabric effectively separated a variety of oil-water mixtures and emulsions with ultrahigh efficiency and reusability, with the separation efficiency remaining above 99.990% after 30 separation cycles. The spray coating method can effectively construct micro-nano structures on the surface of various complex substrates, although these structures can easily be damaged. Another key limitation is that spray coating cannot be applied to the modification of 3D micro/mesoporous internal structures [68].

#### 4.3. Chemical vapor deposition (CVD)

In the chemical vapor deposition (CVD) method, precursors are reacted and the products are deposited on the substrate to form

multifunctional nanomaterial coatings. CVD coating can be used to efficiently modify LCB and other substrates for oily wastewater treatment. Ultra-light cellulose aerogel can be modified by CVD to become hydrophobic, resulting in a capability for oil absorption, with high surface and internal hydrophobicity [69]. Zhang *et al.* conducted a comprehensive study on the CVD of  $\text{SiO}_2$  on soot-coated copper mesh and prepared a network of  $\text{SiO}_2$  nanofibers with superhydrophobicity and superlipophilicity, offering a solution for the treatment of oily wastewater [70]. Similarly, silk fabric was enzymatically etched using the CVD process, achieving stable oil-water separation performance [71]. Furthermore, an oil-water separation aerogel with thermal management function was fabricated via CVD modification by Zhu *et al.* using a top-down strategy to combine balsa wood, PDMS, and carbon nanotubes (CNTs) [72]. The photothermal effect of CNTs promotes rapid volatilization of the absorbed oil and due to the automatically continuous output path, oil absorption by the superhydrophobic CNTs/wood aerogel does not become saturated, providing a novel system for the development of functional materials capable of oily wastewater treatment.

#### 4.4. Sol-gel technique

The sol-gel method incorporates precursors into solution and gel states, then processes them to form a composite solid [73]. Briefly, the preparation process hydrolyzes the precursor to form a solution and then forms a compound with the substrate to generate the functional material. For example, a hydrophobic cellulose membrane was modified with hexadecyltrimethoxysilane (HDTMS) by Medina-Sandoval *et al.*, using a  $\text{SiO}_2$  sol-gel method, achieving the separation of immiscible oil-water mixtures at an efficiency of over 99% and remaining effective (separating a water/oil (1:1) emulsion) after 18 cycles of reuse [74]. Through a one-step sol-gel strategy, Xie *et al.* utilized sol-gel strategy for the hydrolysis and polycondensation of tetraethylorthosilicate (TEOS) and hexadecyltrimethoxysilane (HDTMS), preparing a superhydrophobic cellulose membrane (SOCM) with nano-micro structures and a low surface energy, achieving a separation efficiency of over 98% for various oil-water mixtures and exhibiting good stability and recyclability [75]. Using a simple vapor-liquid sol-gel method, underoil superhydrophobic and underwater superoleophobic fabrics with excellent durability were prepared by Chen *et al.*, effectively separating heavy oil/water, light oil/water, water-in-oil (W/O), oil-in-water (O/W) emulsions, and immiscible organic solvents [76]. Overall, the sol-gel technique provides a foundation for the preparation of multifunctional materials for use in oil-water separation.

#### 4.5. Other advanced modification techniques

Due to recent technological progress, novel LCB modification methods and LCB smart materials are likely to be applied to the oily wastewater treatment, especially techniques forming smart materials with self-healing properties, catalytic activity, photothermal performance, and multi-functionality.

Cao *et al.* developed a carbon black membrane (CBM) with a layered structure and suitable chemical composition, exhibiting excellent multiphase emulsion separation performance due to its underwater superoleophobicity and underoil superhydrophobicity [77]. Using a process based on dual-function aqueous suspensions, a superhydrophobic nanocellulose fiber (SHNCF) aerogel was developed by Wang *et al.*, achieving high capacity oil absorption (13.03–32.95 g/g), with efficiently continuous oil-water separation [78]. Although materials with special wettability have been proven capable of oil-water separation, they can easily be polluted by oil during practical application. The preparation of materials

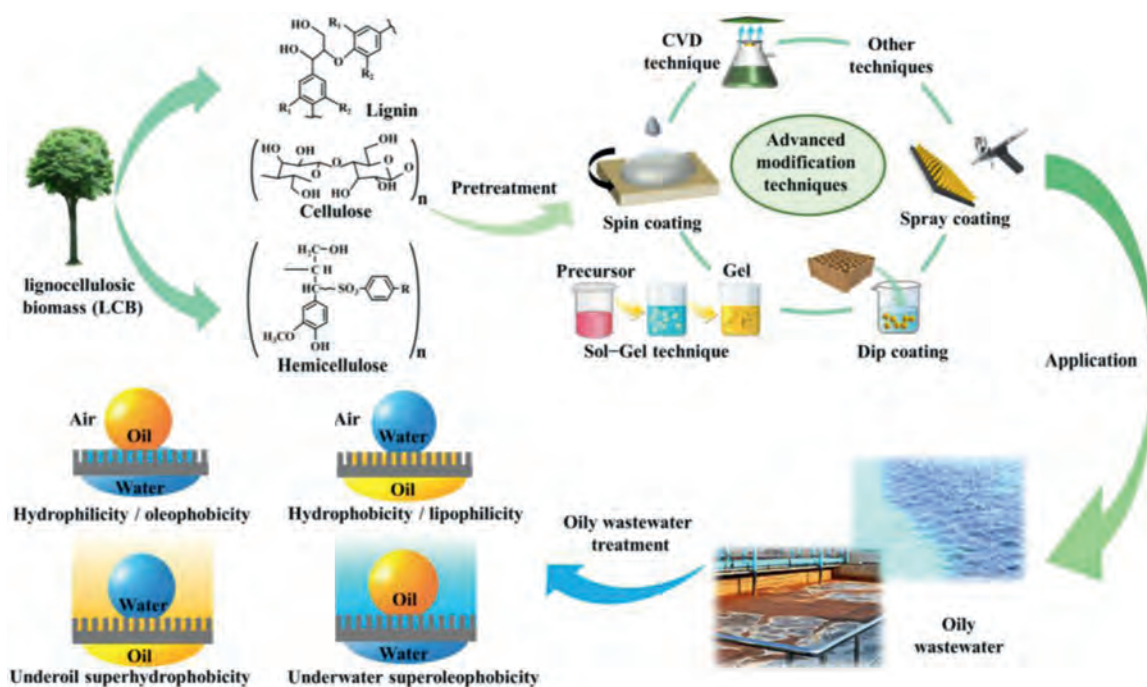


Fig. 2. Schematic diagram of the process of LCB used in oily wastewater treatment after refining and advanced modification techniques.

capable of catalytic degradation or intelligent wettability conversion is a promising solution to overcome this limitation. Yang *et al.* proposed a fluorine-free strategy for the modification of superhydrophobic cotton with  $\text{TiO}_2$  nanoparticles, generating a cotton fabric capable of self-cleaning via superhydrophobicity and photocatalytic degradation, with an oil-water separation efficiency of >99.0% maintained after 7 cycles of reuse [79]. Chen *et al.* fabricated a smart fabric coating based on pH-responsive and UV-curable polyurethane, which endowed the fabric with wettability characteristics switching from superhydrophobic to underwater superoleophobic with changing pH conditions [80]. The smart Janus membrane has shown much promise for application in oily wastewater treatment, exhibiting different wettability characteristics on both sides and effectively separating various emulsions and oil-water mixtures. Lehtinen *et al.* prepared a Janus filter by spraying polydimethylsiloxane (PDMS) on one side of cotton fabric and treating the other side with dimethylaminoethyl methacrylate (DMAEMA) [81]. Due to the demulsification, the Janus fabric can achieve a high rate of oil-water separation ( $4.9 \times 10^{-4}$  mL/s). Despite significant progress in the development of intelligent materials for oily wastewater treatment, their use in large-scale applications remains a significant challenge.

These various techniques of advanced modification applied to LCB for oily wastewater treatment are summarized in Fig. 2. The different modification methods and corresponding key findings are summarized in Table 1. By comparing the effect of different modification methods, this review summarizes key references for the practical application of modified LCB materials in oily wastewater treatment.

## 5. Construction of 0D, 2D and 3D-LCB materials for oily wastewater treatment

Different types of LCB materials exhibit specific characteristics for oily wastewater treatment due to varying structures and wettability behaviors. As a basis for representative comparison, the dimensions of materials are used as the discussion standard: 0D-

LCB modified powders, 2D-LCB membranes, and 3D-LCB structures with continuous channels for oily wastewater treatment.

### 5.1. 0D-LCB materials for oily wastewater treatment

The mature LCB powder materials used in oily wastewater treatment include superhydrophobic-superlipophilic (water-removing) and superhydrophilic-underwater superoleophobic (oil-removing) powders. Using sawdust as a raw material, a superhydrophobic and superoleophilic oil sorbent LCB was designed by Zang *et al.*, combining sawdust fibers with  $\text{SiO}_2$  particles and self-assembly octadecyltrichlorosilane (OTS) monomers [82]. The WCA and oil contact angle (OCA) of sawdust oil absorbents were  $153^\circ$  and  $\sim 0^\circ$ , respectively, exhibiting an excellent oil absorption capacity of 14.4 g/g. In order to improve the oil absorption and recycling capacity of LCB powders, hollow spherical ZnO particles and HDTMS were deposited on the surface of corn straw powders via chemical hydrophobic modification, effectively improving surface roughness and reducing the surface free energy of corn straw fibers [83]. The WCA and OCA of the corn straw oil absorbent were  $155^\circ$  and  $\sim 0^\circ$ , respectively, achieving an absorption capacity of 20.4 g/g, resulting in high efficiency oil removal (99%–100%). Similarly, Xu *et al.* fabricated a novel superhydrophobic/superoleophilic corn straw fiber using the impregnation method, achieving excellent superhydrophobicity (WCA  $\sim 152^\circ$ ) and superlipophilicity (OCA  $\sim 0^\circ$ ), with the modified corn straw fiber exhibiting an oil absorption capacity of up to 27.8 g/g [84]. Furthermore, the WCA and OCA remained unchanged after 150 days of storage under ambient temperature and humidity conditions. Overall, the corn straw oil absorbent has high potential for use in the treatment of oily wastewater, due to its good environmental stability and high efficiency oil-water separation capability. Hydrophobic wood powders modified with ZnO nanoparticles can effectively treat oily wastewater, with modified wood flour shown to possess excellent stability with a WCA and OCA of  $156^\circ$  and  $0^\circ$ , respectively [85]. The oil absorption capacity of modified wood flour was 20.81 g/g, with an oil absorption efficiency ranging from 98% to 100%. A key advantage is

**Table 1**  
Comparison of different modification methods and corresponding key findings.

Methods	Material	Wettability	Key findings	Ref.
Dip coating	Poplar lumber, zinc acetate, triethylamine	WCA = 151°	Superhydrophobic wood surface showed that the surface roughness and low-surface energy are needed to form superhydrophobicity.	[60]
	Poplar wood, PMS	WCA = 153°	Using the waterproof reagent PMS as the reactant in a solution-immersion method, the superhydrophobic surface was assembled on the wood surface.	[61]
	Cellulase from <i>Aspergillus niger</i> , polydimethylsiloxane (PDMS)	WCA = 162°	Using the biological enzyme dipping method, by changing the conditions to find the enzyme value with the largest superhydrophobicity.	[62]
Spray coating	1H,1H,2H,2H-Perfluorodecyltrimethoxysilane, SiO <sub>2</sub> , Ausbond 92 conformal coating	WCA = 156.1°	Excellent scratch and abrasion resistance, strong acid and alkali resistance.	[65]
	Cellulose, attapulgite (ATP), HDTMS	WCA = 150.4°, OCA = 152.6° (underwater)	A Janus membrane possessed broad development potential in the design of special wettability oil-water separation membranes.	[66]
Chemical vapor deposition	Octavinyl polyhedron oligomeric sesquioxane, SiO <sub>2</sub> , fabric	WCA = 159.6° ± 1.8°	Modified fabric separated oil-water mixture multiple times, stable, low-cost, and environmentally friendly.	[67]
	Cotton linter, NaOH, urea	WCA = 128.4° (inside)	Aerogel possessed high hydrophobicity with oil absorption capacity of 59.32 g/g.	[69]
	SiO <sub>2</sub> , soot, copper mesh	WCA = 165°, OCA ≈ 0°	A kinetic model was proposed and a device of tubular separator to purify the oily wastewater is demonstrated.	[70]
	Cellulose, cotton fabrics, papain, alkaline protease	WCA = 156.7°	Enzymes were used to form structures on the surface of silk fabrics.	[71]
Sol-gel	Balsa wood, carbon nanotubes, PDMS	WCA ≈ 155°	A condensation/reflux platform married to CNT/wood aerogel system was set up for oil condensation/reflux.	[72]
	Cellulose filter paper, SiO <sub>2</sub> , TEOS, HDTMS	WCA ≈ 121°	Oil and water can be filtered under gravity by the modified membrane, which is an attractive method for treating oily wastewater.	[74]
	Cellulose membrane, HDTMS	WCA = 164.4°	The separation efficiency was > 98% on harsh conditions and could be reused > 10 times.	[75]
Other techniques	Cotton fabric, TEOS	WCA = 162° (under oil), OCA = 162° (under water)	Modified cotton fabric was used for separation of oil and water (99.99%), and had excellent durability.	[76]
	Carbon black, polyurethane, PVDF	WCA = 155° (under oil), OCA = 152° (under water)	The membrane overcoming the problem of liquid layer clogging during continuous filtration of multiphase emulsions.	[77]
	Nano cellulose fiber, SiO <sub>2</sub> , HDTMS, aminopropyltriethoxysilane (AS)	WCA = 160°	Innovatively proposed a water-based superhydrophobic suspension and prepared a super-hydrophobic aerogel with a maximum oil absorption of 32.95 g/g.	[78]
	Cotton fabric, titanium tetraisopropoxide, 3-mercaptopropyltriethoxysilane, 2-hydroxy-2-methylpropionophenone	WCA = 157.6°	Modified cotton with oil-water separation and photocatalytic degradation ability, fluorine-free and environmentally friendly.	[79]
	Cotton fabrics, silane coupling agent, etc.	OCA = 155° (under water)	Modified cotton fabric with self-cleaning and pH-controllable oil/water separating ability.	[80]
	Cotton fabric, PDMS, HEMA, MMA	WCA = 146° ± 4° (one side)	Janus membrane with oil-water separation and antibacterial functions.	[81]

that modified wood flour can be recycled after removal of the absorbed oil. Therefore, the superior oil-water separation efficiency and environmental durability of the modified wood flour greatly enhance its commercial applicability and feasibility. The composite modification of LCB with a variety of nanoparticles is an important method for improving the capability for oily wastewater treatment. Similarly, through the composite modification of LCB with ZnO/SiO<sub>2</sub> and octyltriethoxysilane (OTES), a superhydrophobic-superoleophilic corn straw material was fabricated for the separation of oil from liquid mixtures, achieving absorption capacities for crude and bean oils of 20.05 and 22.50 g/g, respectively [86]. Although LCB powdered materials possess high oil absorption and oil removal efficiencies, they remain limited by their capacity for recycling and reuse. Di *et al.* designed Fe<sub>3</sub>O<sub>4</sub>/sawdust composites (HFSCs) capable of high efficiency oil-water separation (up to 99%) [87]. It is of note, that the oil-absorbing composites of LCB were easily separated magnetically, allowing the oil-absorbed Fe<sub>3</sub>O<sub>4</sub>/sawdust to be collected using a magnet and reused after treatment. Recyclability, cost-effectiveness, and environmental impact are the important evaluation criteria for the selection of materials for use in oily wastewater treatment. Overall, OD-LCB materials have been shown to exhibit great potential for application in the treatment of oily wastewater.

## 5.2. 2D-LCB materials for oily wastewater treatment

Oil-water mixtures can be passed through 2D-LCB membranes modified by superwetable materials to achieve complete oily wastewater treatment. The separation efficiency, flux, stability, cost, and sustainability of LCB membranes are the key parameters for assessing whether they can be applied in practice. Therefore, establishing simple preparation processes, low-cost raw materials, excellent separation efficiency, and high membrane strength, requires the further exploration of materials such as LCB composites for use in filter membranes. Yu *et al.* prepared a corn straw powder-nylon 6,6 membrane (CSPNM) using the phase-inversion method, achieving superhydrophilicity and underwater superoleophobicity [88]. After 20 separation cycles, the oil rejection rate remained > 99.50%, with a flux of 1561.09 L m<sup>-2</sup> h<sup>-1</sup>, indicating that the CSPNM possessed good reuse capability and environmental stability. A similar nylon membrane coated with a superhydrophilic/underwater superoleophobic cellulose-starch-silica (CSS) composite was designed by Zhang *et al.*, exhibiting a WCA and underwater OCA of 0° and 159.5°, respectively [89]. The membrane possessed a mixture flux of 31,847 L<sup>-1</sup> m<sup>-2</sup> h<sup>-1</sup> bar<sup>-1</sup> and after 100 cycles, the separation efficiency remained above 97%, with the membrane able to be maintained for at least 24 h un-

der extreme environmental conditions (pH 4–10), exhibiting high removal flux, good stability, and reusability. The separation of two-phase oil-water mixtures under laboratory conditions is idealized, while three-phase or more complex light oil/water/heavy oil mixtures are more representative of real wastewater conditions. Using the strategy of continuous gravity separation of three-phase oil-water mixtures, a dually prewetted LCB membrane with underwater superoleophobicity and underoil superhydrophobicity, has been shown to possess superior oil-water separation capabilities [90]. A superamphiphilic waste corn straw powder (CSP)-coated fabric (CSPF) was designed by spraying CSP and polyurethane (PU) solutions onto cotton fabrics. The CSPF was dually prewetted using water and oil (DCSPF) to form a water-containing region (WCR) and an oil-containing region (OCR), respectively, in which the oil and water were selectively passed through the WCR or OCR to separate the three-phase oil-water mixtures. The average fluxes of water, light oil, and heavy oil in the DCSPF were  $\sim 3.8$ ,  $\sim 8.9$  and  $\sim 13.3 \text{ L m}^{-2} \text{ s}^{-1}$ , respectively, with a three-phase mixture separation efficiency of over 97% maintained after 50 separation cycles. The Janus membrane with asymmetric surface properties was also found to perform very well in oily wastewater treatment. Utilizing cellulose and Ag nanoparticles, a double-sided Janus composite membrane (JCM) was fabricated by Lv *et al.*, with both superhydrophilic and superhydrophobic properties [91]. The separation fluxes of oil-in-water and water-in-oil emulsions in the JCM were  $640 \text{ L m}^{-2} \text{ h}^{-1}$  and  $323.04 \text{ L m}^{-2} \text{ h}^{-1}$ , respectively, with the separation efficiency exceeding 96%. At present, the trend in development of 2D-LCB materials for oily wastewater treatment has shifted from single- to multi-functional modifications and from single emulsion to multiphase emulsion treatments, supporting the development of low-cost, readily available, environmentally friendly, and sustainable materials. Therefore, further studies are required in order to transition from laboratory research to industrial applications.

### 5.3. 3D-LCB materials for the oily wastewater treatment

The high porosity, suitable pore sizes and compressibility of 3D-LCB porous oil-absorbing materials make them suitable for application in oily wastewater treatment. Due to the complexity and diversity of oily wastewater, there is an urgent need to develop low-energy and high-efficiency 3D-LCB materials for oily wastewater treatment. Yang *et al.* performed controlled dissolution treatment using raw cotton, freeze-drying the product to develop a cellulose sponge with excellent underwater superoleophobicity [92]. The oil-water separation efficiency and flux of the 3D cellulose sponge were 99.2% and up to  $485 \text{ L m}^{-2} \text{ h}^{-1}$ , respectively. Sawdust waste can also be used for oily wastewater treatment [93]. A 3D liquefied-larch-based polymer foam (LLB-PF) with a honeycomb interconnected structure and its carbonized product, LLB-CF, were prepared using larch sawdust waste as the raw material. The unique 3D structure endowed LLB-PF/CF with a high abundance of pores, hydrophobicity, and superoleophilicity. The absorption of tetrachloromethane and epoxidized soybean oil by LLB-PF and LLB-CF reached 88-fold and 153-fold greater than their own weight, respectively. A balsa wood sponge with the fluoroalkyl silane modified reduced graphene oxide (F-rGO@WS) was produced by Huang *et al.* for oily wastewater treatment, achieving an oil-water separation efficiency of 99.0% due to its hydrophobicity (WCA =  $145^\circ$ ) and longitudinal channels [94]. In addition, F-rGO@WS possesses electric-heating performance, resulting in a 10-fold higher heavy oil and water separation rate under a voltage of 20 V, significantly enhancing its capability for the treatment of complex oily wastewater. Similarly, innovative 3D-LCB aerogels have shown great potential for oily wastewater treatment, with cellulose aerogels (CEA) and aerogels coated with Cu nanoparticles (Cu/CEA) possessing high oil

absorption capabilities (67.8–164.5 g/g) [95]. Cu/CEA can quickly separate oil-water mixtures with a high separation efficiency of >97%, while also exhibiting good recyclability (>10 times) making it a promising material for practical application. Meng *et al.* constructed a lignin-based carbon aerogel enhanced by graphene oxide (LCAGO), resulting in synergistic superhydrophobicity and good mechanical properties for oily wastewater treatment [96]. The oil absorption capacity of LCAGO was 32–34 g/g, making it suitable for use in environmental remediation. The combination of wood pulp cellulose nanofibers (CNF) and three silane modifiers, resulted in the fabrication of a novel hydrophilic and oleophobic composite aerogel, which achieved a good absorption capacity of 11.5 g/g. After 5 absorption-regeneration cycles, the oil absorption capability of the modified aerogels remained at 10 g/g, with its rugged compression performance and excellent water resistance being beneficial for practical industrial applications [97]. Yuan *et al.* utilized natural sisal cellulose as a raw material, using carbonization and  $\text{MnO}_2$  self-assembly to prepare a hierarchical biomass carbon@ $\text{SiO}_2$ @ $\text{MnO}_2$  (HBCSM) aerogel with a high specific surface area and good mechanical flexibility [98]. The WCA of the HBCSM aerogel was  $155^\circ$ , resulting in a large absorption capacity for different oils and organic solvents (60–120 g/g). Furthermore, the HBCSM aerogel remained effective for more than 9 cycles of reuse, making it a promising candidate material for oily wastewater treatment. LCB porous oil-absorbing materials are a promising development in the field of oily wastewater treatment due to their unique 3D structures, with the research focus now aimed at developing environmentally friendly, economical, and low-energy consuming 3D-LCB materials for oily wastewater treatment.

Factors such as separation efficiency, separation quality, and recyclability determine the potential for practical application of LCB materials. A comparison of the parameters of 0D-, 2D- and 3D-LCB materials for oily wastewater treatment is shown in Table 2. Overall, selecting LCB materials with an appropriate structure for oily wastewater treatment according to the actual treatment conditions is the key to achieving optimal treatment performance.

## 6. Current challenges and future prospects

The diversity and biocompatibility of the natural components of LCB, provide biomass materials with inherent advantages and potentially transformative characteristics. Taking LCB biomass as raw materials possesses the advantages of being green and sustainable. They are nontoxic after degradation and capable of avoiding secondary pollution effectively. In addition, LCB biomass is economically feasible, and most of them are waste resources such as wood flour and corn stalk. Therefore, the use of LCB to prepare oil-water separation materials not only meets the needs of environmental protection but also the requirements of low cost. However, despite the achievements made in the preparation, microscopic morphology, chemical structure, and composition of LCB materials, there remain various limitations and deficiencies, as shown in Fig. 3. The challenges include single functions, poor stability and circulation, low efficiency oil-water separation, and high environmental-impact modification processes. Therefore, developing strategies for the conversion of LCB components into the materials capable of high efficiency oil-water separation is essential, particularly simple strategies to adjust the morphology of LCB materials from 0D fibers, 2D membranes to 3D structures for oily wastewater treatment.

The recycling and regeneration capabilities of materials for oily wastewater treatment are of great significance to their practical application. Due to the high recycling cost and complex recycling processes, most 0D-LCB powder absorbents are difficult to use repeatedly. In contrast, 2D-LCB membranes rarely involve recovery problems, but they are often used in the later stages of water treat-

**Table 2**  
Comparison of the oily wastewater treatment parameters of 0D-, 2D-, 3D-LCB materials.

Structure state	Material	Wettability	Separation efficiency/absorptive capacity	Recyclable performance	Ref.
0D-LCB oily wastewater treatment materials	Sawdust	WCA = 153°, OCA ≈ 0°	14.4 g/g	/	[82]
	Corn straw powder	WCA = 155°, OCA ≈ 0°	99%–100%/20.4 g/g	77%~89% (3 cycles)	[83]
	Corn straw fiber	WCA ≈ 152°, OCA ≈ 0°	27.8 g/g	/	[84]
	Wood	WCA = 156°, OCA ≈ 0°	98%–100%/20.81 g/g	77% (3 cycles)	[85]
	Corn straw	WCA = 152°, OCA = 0°	22.50 g/g	/	[86]
	Sawdust	WCA > 150°	99%	> 10 cycles	[87]
2D-LCB oily wastewater treatment materials	Corn straw powder-nylon	WCA = 0°, OCA = 157° (under water)	oil rejection > 99.60% flux > 660.00 L m <sup>-2</sup> h <sup>-1</sup>	20 cycles	[88]
	6,6 membrane	WCA = 0°, OCA = 159.5°	31,847 L <sup>-1</sup> m <sup>-2</sup> h <sup>-1</sup> bar <sup>-1</sup>	100 cycles (97%)	[89]
	Cellulose-starch-silica nylon membrane	WCA = 152° (under oil), OCA = 155° (under water)	~13.3 L m <sup>-2</sup> s <sup>-1</sup> (heavy oil)	50 cycles (97%)	[90]
	Waste corn straw	WCA = 150.3° (hydrophobic side), OCA = 153° (under water)	640 L m <sup>-2</sup> h <sup>-1</sup> (oil-in-water) 323.04 L m <sup>-2</sup> h <sup>-1</sup> (water-in-oil)	10 cycles (94%)	[91]
3D-LCB oily wastewater treatment materials	Cotton	WCA = 141 ± 1.6°, OCA = 160 ± 1.3° (underwater)	485 L m <sup>-2</sup> h <sup>-1</sup>	> 10 cycles	[92]
	Larch sawdust	WCA = 144.6°	55–153 g/g	> 5 cycles	[93]
	Balsa wood	WCA = 145°	99.0%/16.73 g/g (Chloroform)	30 cycles (95%)	[94]
	biomass cellulose	WCA = 150.3°	> 97%/67.8 ~ 164.5 g/g	> 10 cycles	[95]
	Alkali lignin	WCA > 150°	32–34 g/g	/	[96]
	pulp cellulose nanofibers	WCA = 0°, OCA = 106.5°	11.5 g/g	> 5 cycles	[97]
	natural sisal cellulose	WCA = 155°	60–120 g/g	> 9 cycles	[98]



**Fig. 3.** The advantages and disadvantages of LCB in wastewater treatment applications.

ment processes, as membrane filtration requires high water quality and large impurities may damage the membrane. In particular, in filtration membranes for the separation of oil-water emulsions, if the emulsion enters the 2D membrane together with a large amount of un-emulsified water or oil, a serious decline in flux will occur in the membrane [99]. 3D-LCB materials are suitable for oily wastewater treatment due to their abundance of interconnected pores or channels [100]. Whether LCB materials can be used industrially for oily wastewater treatment is a main factor in the evaluation of their practical value. It is essential to develop sustainable LCB materials with catalytic properties such as visible light catalysis, electrocatalysis, and advanced oxidation for oily wastewater treatment, overcoming the limitations of single-performance separation materials.

A comprehensive technical and economic sustainability analysis should be conducted to determine the optimal LCB pretreatment methods and modification routes, to achieve high-efficiency

oily wastewater treatment. It is of note, that most of the chemicals used in wettability modification are not environmentally friendly and in particular, fluorine-free modifiers should be selected for use in the modification process [101–103]. Separation efficiency, recyclability, and cost are the preferred evaluation criteria for LCB materials usable in industrial and domestic oily wastewater treatment. The ideal intelligent sewage separation systems are easy to repair itself after being damaged and capable of treating composite pollutants. The characteristics of rapid degradation and self-decontamination of intelligent systems will enable the treated wastewater to meet the qualified standards, which possess the advantages of low cost, high separation efficiency, easy self-cleaning, and low labor cost. The cost optimization of the intelligent system should be operated in accordance with the process and technical requirements. Controlling the cost of raw materials and optimizing the design and structure of the LCB treating medium can improve separation efficiency and quality. A cost-benefit calculation



Fig. 4. The development prospect of LCB in wastewater treatment applications.

system should be established to optimize the treatment process to improve the economic benefits of the intelligent system. More efforts should be made to analyze the mechanism of biomass materials for removal of complex pollutants in sewage on the basis of ensuring the effect of oil-water separation [104,105]. Therefore, there is a strong need to develop LCB materials with different dimensions and structures, in order to meet the actual requirements of different field applications.

At present, most of the materials used in oily wastewater treatment reported in the literature have been non-renewable materials [106]. Fig. 4 highlights the development prospects of LCB for wastewater treatment applications. From the perspective of sustainable development, some important aspects need to be considered during the deployment of LCB materials for oily wastewater treatment:

- (1) In the utilization of a certain component of LCB, it is necessary to develop efficient separation processes to achieve environmentally friendly and low-cost separation.
- (2) Further research is needed to explore the internal relationship between the mechanical properties of LCB-2D/3D porous materials and their structure. LCB materials should be optimized to allow the sustainable recycling of oil and water resources.
- (3) LCB materials are susceptible to damage after repeated use and mechanical abrasion, resulting in a reduction or even loss of separation efficiency. The construction of sustainable and recyclable superwetting LCB materials for oily wastewater treatment materials remains a key problem that needs to be solved.
- (4) The design of LCB materials capable of simultaneously treating oily wastewater and pollutants on a large scale remains a challenge, especially the large-scale treatment of organic dyes, heavy metal ions, pesticides, antibiotics and other pollutants.
- (5) The construction and development of multifunctional, intelligent sewage separation systems using LCB are essential, with self-healing capabilities and multiple response mechanisms (such as temperature, light, electric, pH, and magnetic responses).

## 7. Conclusions

The application of LCB materials has become an attractive solution for oily wastewater treatment, although there are still some

shortcomings that must be overcome. In order to effectively separate and purify oily wastewater, it is necessary to develop sustainable and multifunctional LCB materials. Multi-dimensional wastewater treatment and purification system utilizing LCB is a key trend in sustainable development, requiring thorough evaluation for effective large-scale application. Considering the feasibility of taking LCB as raw materials to build the sewage treatment system with cost optimization, treatment efficiency, and recyclability, which is conducive to the development of a high-efficiency intelligent sewage treatment system. Ultimately, the current research indicates that these materials are promising for sustainable oily wastewater treatment and that further development of LCB materials will overcome the present challenges and limitations.

## Declaration of competing interest

The authors declare no conflict of interest.

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