Recent Developments in Water Treatment by Cellulose Aerogels from Agricultural Waste

Phuong X T Nguyen\textsuperscript{1,2,3}, Kim H Ho\textsuperscript{1,2}, Chi T X Nguyen\textsuperscript{1,2}, Nga H N Do\textsuperscript{1,2}, Anh P N Pham\textsuperscript{3}, Tai C Do\textsuperscript{3}, Kien A Le\textsuperscript{4}, Phung K Le\textsuperscript{1,2,*}

\textsuperscript{1}Faculty of Chemical Engineering, Ho Chi Minh City University of Technology (HCMUT), 268 Ly Thuong Kiet Street, District 10, Ho Chi Minh City, Vietnam
\textsuperscript{2}Vietnam National University Ho Chi Minh City, Linh Trung Ward, Thu Duc District, Ho Chi Minh City, Vietnam
\textsuperscript{3}Hong Bang International University (HIU), 215 Dien Bien Phu Street, Ward 15, Binh Thanh District, Ho Chi Minh City, Vietnam
\textsuperscript{4}Institute for Tropicalization and Environment, 57A Truong Quoc Dung Street, Phu Nhuan District, Ho Chi Minh City, Vietnam
\textsuperscript{*}Corresponding author’s e-mail: phungle@hcmut.edu.vn

Abstract. Water pollution caused by inorganic and organic compounds affects seriously health risks and the destruction of the water balance of the ecosystem. Water remediation is one of the greatest challenges of the modern age, which is faced by several countries in over the world. Many advanced technologies have been developed in recent years to manage this alarming problem. One of the recent research directions is to utilize agricultural waste, which is an abundant, low-cost, eco-friendly source, to manufacture cellulose aerogels for the removal of heavy metals, dyes, and oils from contaminated water. This review paper focuses on the latest green fabrication of bio-aerogels from various cellulose-based plant waste and studies on the kinetics and influence of factors such as contact time, pH solution, initial concentration, and temperature on the adsorption process to better understand the mechanisms of water treatment by the cellulose aerogels. Based on the discussion and summary, the potential of agro-waste sources in their value-added chain by developing efficient water treatment cellulose aerogels is comprehensively overviewed.

1. Introduction
Biomass is a heterogeneous combination formed by organic compounds and a small number of inorganic substances, in which solar energy is accumulated as chemical connections. Raw biomass comprises 30–60% carbon, 30–40% oxygen, and 5–6% hydrogen by weight, and 1% other elements. Natural photosynthesis produces $1.7 \times 10^{11}$ tons of biomass per annum [1]. Lignocellulosic biomass plays an important role as a potential sustainable power and contributes approx. 25% of global energy to replace fossil fuels [2]. The remaining can be utilized to fabricate the renewable materials as novel adsorbents [3, 4], pharmaceutical carriers [5, 6], electrode materials [7, 8], functional composites [9], catalysis/photocatalytic [10, 11], or discharge into the soil as fertilizer slow-release, nutrient controlled-release [12, 13]. Lignocellulosic compositions from biomass resources vary include lignin, hemicellulose, and cellulose, in which the application for cellulose is significantly dominated.
Cellulose was discovered in 1839 by Anselme Payen (French) for the first time and has been researching and developing in numerous modern fields [14]. It has the chemical formula of \((C_6H_{10}O_5)_n\), which is one of the most ubiquitous biopolymers on the earth and the crucial component of biomass. It is a polysaccharide encompassing a linear chain connected of several thousands of \(\beta(1\rightarrow4)\) linked anhydrous-D-glucose units, which is a major component of the plant cell wall. Together with Vander Waals forces, intramolecular hydrogen bonds keep the structure parallel stacking of cellulose microfibrils into crystalline cellulose [15]. During the New Material Revolution, eco-friendly and recyclable properties are critical aspects to study and develop products from natural resources. With the abundant quantity from agricultural waste, for example, over worldwide yearly, more than 7.64×10^6 tons of sugarcane bagasse [16], approximately 7.64×10^7 tons of pineapple leaves by-product [17], it is the fact that cellulose aerogel based on agro-waste is a prospect of the synthesized high-performance materials. Cellulose-based materials in terms of fibers [18], nano [19], films [20], hydrogels [21], aerogels [16], and composites [22] have been successfully synthesized. Among that applications, natural cellulosic aerogels have excellent potentiality in various applications for adsorption [23], heat and sound insulation [24], biomedical materials [25]. It was known that aerogel is a 3D-porous structure material in terms of low density (0.001–0.500 g/cm^3), high porosity (≥ 90 %) with pore size in 2–50 nm, large specific surface area (100–1600 m^2/g), good mechanical strength (5.20 kPa–16.67 MPa), high thermal conductivity (0.03–0.04 W/m.K). Aerogels were firstly fabricated by Kistler from a silica-based source by subjecting a wet gel precursor and supercritical drying [26].

The rapid development of industry in terms of the production activities, exploitation of fossil fuel, textile, metallurgical, manufacturing of pesticides and fertilizers results in intensifying environmental pollutions, particularly contaminated wastewater. Known as a “ubiquitous solvent” water can dissolve more substances than other liquids, thus, water is uniquely vulnerable to contamination. According to Gallup’s yearly Environment survey, the contamination of both drinking water and rivers, lakes, reservoirs remain most worried. Almost a quarter of the world’s population has a problem with using water safety, around 4 billion humans will perambulate extreme water deficiency, data from the United Nations [27]. The benefits of the Industrial Revolution leading to economic development are not denized but chemical toxic from industrial activity deliberately or unintentionally released into the water system. Heavy metals are transported by drainage from the factory to the rivers and oceans. Finally, these metals are accumulated in the soil and sediments [4, 28]. From Qiaqiao investigation in 2017, the contents of toxic metals in the global river and lake water were higher in Africa, Asia, South America, but lower in Europe and North America [29]. In recent years, dyes are extensively applied in textile, leather, paper, photography, paints, printing inks, plastics, and electronics. Approximately 100,000 various pigments and dyes are synthesized with a production of over 7×10^7 tons. Textile manufacture consumes higher than 70 % of the total produced dyes over the world. However, about 1–2% of dyes stuff are lost during manufacturing processes and 10–15 % of dyes are removed as wastewater [30, 31]. Oil spills occupy around 12 % of the oil that enters marine environments, besides discharging from navigation travel, drains, and dumping [32].

To protect human health and sustain ecological balance, successfully removing the toxic chemical from wastewater and restricting agricultural waste are still significant and challenging issues. Regenerated cellulose aerogels having distinctive physicochemical attributes likely high porosity, selectivity, and specific surface area are mostly studied to clean the environment. Considering the importance and prospect of biomass utilization for aerogel products, as well as the speedy study operation in this area, for the first time, this review article is a comprehensive overview of the recent advances in the synthesis processing of natural cellulosic aerogels from agro-waste and the applications of environmental Remediations such as heavy metal ion, dye, and oil contaminants. The impact of the adsorption process related to contact time, pH solution, initial concentration, and temperature is mentioned. To better clarify the mechanisms of polluted water treatment by the cellulose aerogels, the investigation on the kinetics and isotherm adsorption is also introduced. Finally, the challenges and potentialities of cellulose aerogel from agro-waste will be encountered in the future.
2. **Fabrication of cellulose aerogels from lignocellulosic biomass**

The preparation of cellulose aerogels primarily includes four steps, (i) cellulose extraction from different biomass sources, (ii) dissolution by suitable cross-linker solvents (iii) Gelations or regenerations, and (iv) drying processes. These synthesis pathways have important influences on the structure formation and characterization of bio-aerogels.

2.1. **Cellulose extraction from different biomass sources**

The biomass sources are principally distributed into agro-waste (coir, pineapple, sugarcane bagasse, bamboo, rice/wheat husk/straw) in figure 1 – left branch) and industrial waste (wood pulp, paper, cotton, Kapok fiber) in figure 1 – right branch). The generality of plant cell walls consists of cellulose, hemicelluloses, and lignin. Lignin is a linker between cellulose and hemicellulose, which essentially provides stiffness and stability to the cell. Hence, the preliminary step is the pretreatment of lignocellulosic to separate the hemicelluloses and lignin.

![Figure 1. Categorization of biomass](image)

A variety of methods to purify natural-origin cellulose materials includes biological, physical, and chemical technicalities [33]. Biological techniques are taken part in the enzyme but a high cost and slow process rate. Chemical pretreatment is more efficient and less energy than physical methods (ball milling, homogenization, high-pressure homogenizer). Two chemical methodologies are commonly applied acid and alkaline hydrolysis to break the strong structure with their various mechanisms. While acid hydrolysis using hydronium ions attacks the lignocellulosic links to stretch out the internal fibers, the alkaline solutions can destroy the lignin structure by the destruction of glycosidic ester or ether links [34].

A sulfur-free method to remove lignin from coconut fibers is proposed by Fauziyah et al.. A combination of mechanical and chemical treatments with 6% NaOH was effectively used, which results obtained in the highest α-cellulose content (92.5 wt%) and the Kappa number of the pulp equaling 14.8 [23]. In the alkaline pretreatment stage for cellulose purification from apple pomace, three elements such as the NaOH content (6–12%), extracting time (30–240 min), and heating (30–90 °C) were investigated, which followed the aims as highest yield, maximum α-cellulose content, and whiteness index (WI). the yield equaled 27.96±0.78%, 85.31±0.91% of α-cellulose purity, and 47.79±0.65% of WI when the experimental conditions were done as 10.23% NaOH, 69.82 °C, and 161.54 min [35]. Sugarcane bagasse [16], bamboo microfibers [36] were also treated by NaOH solution for isolate cellulose. While an alkali process removes the amorphous region of hemicelluloses and lignin from lignocellulosic, the elimination of most of the lignin is done by acid-chlorite and bleaching [37]. Alana et al. isolated cellulose nanocrystal structures from the paper residue by an
aqueous acid solution of 92% CH₃COOH and 1.4% H₂SO₄ at 80 °C for 3 h [38]. In summary, the alkaline method is regularly chosen in the pretreatment step due to its efficiency.

2.2. Dissolution by suitable cross-linker solvents
Cellulosic chains are difficult to dissolve in water and several organic and inorganic solvents, because of the determined structure, and robust inter- and intramolecular hydrogen bonding causing in the hydroxyl groups. The original challenge to dissolve cellulose is the solvent having the accessible aptitude into their chains and directly attacking its crystalline and amorphous network. Over the last period, the scientific groups have made positive contributions to cellulose dissolution [39]. Generally, cellulose solvents are classified into two major types, such as derivatizing, and non-derivatizing systems. The term “derivatizing” group comprehends all the methods in which cellulose dissolution takes place through covalent transformation developing an “unstable” ester, ether, or acetal derivative. The “non-derivatizing” solvents are alkaline and related systems, chemical cross-linkers systems, transition metal complexing agents, non-aqueous systems, ionic liquids [40]. Those systems are capable of dissolving their chains by disrupting the physical interactions which hold them without any chemical modification. In recent years, polyvinyl alcohol (PVA) solvents and cross-linking of chemicals get special attention, which is based on the purpose of producing materials with high-value performances, sustainability, and eco-friendliness (table 1).

| Raw material          | Type solvents  | Cross-linker solvents | Drying methods | Density (g/cm³) | Porosity (%) | BET surface area (m²/g) | BJH pore size (nm) | Ref. |
|-----------------------|----------------|-----------------------|----------------|----------------|--------------|-------------------------|--------------------|------|
| Sugarcane bagasse     | non-derivatizing| PVA                   | freeze drying  | 0.016–0.122   | 91.9–98.9    | 309.3–368.3          | 4.4–11.3           | [16] |
| Sugarcane bagasse     | non-derivatizing| PVA/Glyoxal           | freeze drying  | 0.012–0.108   | 92.9–99.2    | 368.3–4.4            |                   | [41] |
| Pineapple              | non-derivatizing| DETA/CMC/PVA         | freeze drying  | 0.063–0.093   | 91.1–94.4    | -                      | -                  | [17] |
| Pineapple              | non-derivatizing| PVA                   | freeze drying  | 0.063–0.093   | 99           | -                      | -                  | [24] |
| Pineapple              | non-derivatizing| PVA                   | freeze drying  | 0.127–0.326   | 96.9–98.8    | 5.6                    | 1.3–2.2            | [42] |
| Pineapple and cotton   | non-derivatizing| PVA                   | freeze drying  | 0.019–0.046   | 96           | -                      | -                  | [43] |
| Rice straw             | non-derivatizing| PVA                   | freeze drying  | 0.05–0.06     | 97           | -                      | -                  | [44] |
| Rice straw             | derivatizing    | Polyamidene           | freeze drying  | 0.002–0.024   | 98.4–99.8    | 178.8                  | 12–50              | [45] |
| Lupin hull             | non-derivatizing| Water/ethanol         | scCO₂ drying   | 0.019         | 98.8         | 93                     | 11.7               | [46] |
| Lupin hull             | non-derivatizing| water                 | freeze drying  | 0.030         | 98.1         | 18                     | 10.9               | [46] |
| Sorghum stem           | non-derivatizing| NaOH/urea/water       | freeze drying  | 0.146–0.167   | 90           | -                      | 50                 | [47] |
| White Bamboo           | non-derivatizing| NaOH/urea/water       | freeze drying  | 0.085–0.144   | 90.58–94.46  | 8.155–13.419          | -                  | [48] |
2.3. Gelations or regenerations

Synthesis of cellulose aerogel includes many steps where the most important stage depends on the sol-gel or the generation process to create a 3D-porous structure. The cross-links created within the polymeric chains are categorized into two types in terms of the physical and chemical bond. Physical gels are cross-linked by weak forces as hydrogen bonds, Van der Waals interactions, hydrophobic or electronic associations. Chemical gel processes are replaced from a liquid into another liquid phase, typically water (polar) and a non-polar organic solvent. For the similar cellulose content, if replacing water with methanol, coagulating aerogels by methanol resulted in lower density than water-coagulated materials [49]. The enhancement in the physicochemical properties of aerogels and the decrease gelation time is directly related to adding a monovalent salt (LiCl, NaCl, and KCl) and divalent salts (CaCl$_2$ and MgCl$_2$) [50]. Sandeep A et al. studied that the mechanical technicalities via high-speed homogenization or high-intensity ultrasonication treatment influenced the stability of cellulose fibers dispersion in dilute suspensions, as well as cylindrical and dimensional shapes of rice straw cellulose aerogels after freeze-drying. While the micron-sized disintegration by high shear homogenizer was effortlessly precipitated at the bottom bottle, the nano-dimensionality of fibers with the emergence of stable aqueous suspensions was utilized ultrasonic treatment in combination with high-speed shearing [45]. Besides, the size alteration from micro to nano-fibrillated was due to the cavitation of ultrasonic energy, which caused the erosion of the surface fibers and breakdown of the intermolecular hydrogen bonds in microfibers. In the same way, Mandana et al. also presented the similar morphology and structure of freeze-dried cellulose micro/nanofibers (0.1 wt%) from rice straw [51]. Hence, the foundation of more nano-sized cellulose fibers led to a drastic increase in the available surface area and supported favorable conditions for adsorption processes.

2.4. Drying processes

The characterization in connection with morphology via SEM images, density, the porosity of cellulose aerogels strongly depends on the drying techniques. The current methods of cellulose aerogel preparation are usually utilized the vacuum freeze-drying and supercritical drying [52]. Freeze drying is the most regular technique used to remove the solvent in the gel system and manipulate the network inside the aerogel to prevent collapse structure (table 1) [53]. Supercritical drying relates to the renewal of the solvent in the cellulose sol-gel with supercritical fluids establish on their special solubility and lower surface tension. The befitting pressure and temperature are directed to permit the solvent to come to its critical point, then slowly dropped the pressure and cooling down. The solvent is transformed from liquid phases to gas phases and obtained a three-dimensional 3D-porous aerogel. Carbon dioxide (CO$_2$) is more frequently employed because of the ideal supercritical conditions (31.3 °C and 138 bar) and their safety [9, 54]. Reporting to Ciftci et al., while CO$_2$ supercritical drying developed in Lupin hull cellulose aerogel without main fibers aggregation, the freeze-drying evolved in a sheet cellulose network including large pores with several micrometers in diameter due to frozen water [46].

3. Applications of biomass-derived aerogels to treat contaminated wastewater

Enhancing the cleanup of water pollutants urges scientists to develop novel bio-adsorbents. Cellulose aerogels giving porous structure, large specific surface area, and light density are highly adsorptive for heavy metals, dyes, and oils.

3.1. Heavy metals

The presence of heavy metals in the form of hydroxides, oxides, salts... such as lead (Pb), cadmium (Cd), arsenic (As), mercury (Hg), nickel (Ni), chromium (Cr) in the ecosystem cause many adverse sequels [4]. Contaminations of heavy metal ions in the water are difficult to biodegradability and result in a negative impact on human health and the eco-systems. These influences may be decreased activities of organs (liver cirrhosis, chronic anemia, or stomach and intestine irritation), disorganized
of the nervous system (dizziness, fatigue, depression, loss of memory, and lethargy), caused in cancer, life-threatening if acute toxicity cases. Heavy metals may be rigid precipitation and accumulation in bone, muscle, fat, or joint, causing many complications in the body. Consequently, the purification and elimination of heavy metals before releasing them to water resources is compulsive in environmental troubles. The efficiency of removing pollutant metals is a challenge required for cellulose aerogels absorption of researchers in recent years.

The present-day articles published that the mechanisms of heavy metals adsorption relate to the static electricity interaction or binding between functional groups on material and metal ions. The surface of bio-materials containing rich in hydroxyl groups (−OH) has a stronger adsorption capacity for metal ions. In recent studies, it has been a trend that modification of natural cellulosic aerogel by various crosslinkers containing carboxylic (−COOH) [55], amine (−NH₂) groups [17], or magnetic-controlled [56] are effective policies to improve its contaminated water removal (table 2). The chelation of complexation, ion exchange, or coordination of −OH, −NH₂, and −COOH functional groups attract and connect contaminant ions to the active sites for adsorption.

### Table 2. Heavy metals adsorption capacity of various cellulose aerogels

| Adsorbents                      | Adsorbates | q_{max} (mg/g) | The kinetic model | The isotherm model | pH | t (°C) | ΔG (kJ) | ΔH (kJ/mol) | ΔS (J/mol) | Ref. |
|---------------------------------|------------|----------------|-------------------|--------------------|----|--------|--------|-------------|-----------|------|
| DETA pineapple leaf fibers aerogel | Ni^{2+}    | 48.4           | 2^{nd}            | Langmuir-Freundlich| 2–6| -      | -      | -           | -         | [17] |
| Carboxylated MnFe_{2}O_{4}/cellulose aerogel | Cu^{2+}    | 73.7           | 2^{nd}            | Langmuir           | 2–9| 35–55 | -      | -           | -         | [55] |
| MnFe_{2}O_{4}/cellulose aerogel   | Cu^{2+}    | 63.3           | 2^{nd}            | Langmuir           | -  | -      | -      | -           | -         | [56] |
| Fe_{3}O_{4}@bagasse@chitosan     | Cr^{6+}    | 41.0           | 2^{nd}            | Freundlich         | 65 | -      | -22.01 | 0.028       | -12.46    | [57] |
|                                | Cu^{2+}    | 137.6          |                   |                    |    | -      | -13.44 | (-12.79)    | -19.7     | -0.0213 |
|                                | Cd^{2+}    | 89.6           | 2^{nd}            | Langmuir           | 1–7| 25–55 | -12.3  | (-11.58)    | -15.1     | -0.0103 |
|                                | Pb^{2+}    | 655.2          |                   |                    |    | -      | -13.96 | (-11.45)    | -32.7     | -0.062  |
|                                | Cu^{2+}    | 99.0           |                   |                    |    | -      | -8.83  | (-7.66)     | 3.965     | 3.9      |
|                                | Cd^{2+}    | 124.5          | 2^{nd}            | Langmuir           | 3–7| 25–55 | -5.11  | (-4.18)     | 5.06      | 3.1      |
|                                | Hg^{2+}    | 242.1          |                   |                    |    | -      | -4.43  | (-2.27)     | 19.28     | 7.2      |

DETA-PF aerogel from pineapple leaf fibers demonstrated Ni^{2+} ion adsorption from aqueous solutions, with maximum adsorption of 48.4 mg/g. The amine groups are commonly known as the highly effective functional groups for heavy metal ion removal via adsorption that the main mechanism is formed by metal-ligand coordination linking with only central metal ion/atom [17]. Carboxylated MnFe_{2}O_{4}/cellulose aerogel [55] and MnFe_{2}O_{4}/cellulose aerogel [56] performed rapid removal efficiency and excellent adsorption ability to eliminate Cu^{2+} ions, which were higher than traditional cellulose aerogel (compared 73.70 and 63.30 mg/g with 6.40 mg/g). Hui et al. were synthesized a magnetic double-base aerogel Fe_{3}O_{4}@bagasse@chitosan from waste bagasse fiber,
which removed Cr\(^{6+}\) reaching 41 mg/g at 65 °C. The removal mechanism of Cr\(^{6+}\) ions by FBC aerogel was the redox reaction between the active component magnetic nanoparticles-chitosan and metals ions, as well as the number of amino -NH\(_2\) and carboxyl -COOH groups on the surface of the chitosan via electrostatic interaction [57]. Moreover, the modified adsorbents are better reusability in addition to their effective adsorption capacity [56].

3.2. **Dyes**

Adsorption of dyes is based on dye characteristics namely molecular structure, existing types, the number of functional groups, and site of the substituent in the dye molecules. Dyes are organic compounds and categorizing into cationic and anionic. Cationic dyes (basic dyes) depend on substituted aromatic groups containing the mono-azo, diazo, and azine compound, which is water-soluble and yield colored positive ions in solution. Cationic dyes have been widely used in dye adsorption experiments such as methylene blue (MB), crystal violet (CV), malachite green (MG), safranin. Anionic dyes (acidic dyes) are exhibited different characteristics of their structure in terms of salts of sulphuric, carboxylic, anthraquinone, triphenylmethane, nitro, or phenolic groups. Acid dyes are good water solubility dividing negative ion charges and can be partly/totally connected to the cationic substances such as fabric, silk, nylon, or modified acrylic fibers by covalent bonds. Methyl orange (MO), Congo red (CR), methyl red (MR), indigo carmine, rhodamine B (RhB), eriochrome black-T (EBT) are the most noteworthy sources of industrial contaminations arising from the textile, leather, food, pharmaceutical-cosmetics, paint and varnish, and pulp/paper manufacturing. Dyes are an important concern for scientists because of their complex structures and non-biodegradable nature, which makes them poison for aquatic ecosystems due to restricting sunlight penetration [30, 31]. Hence, the adsorption technique by three-dimensional macroscopic cellulose aerogels has gained favor procedure among all the physicochemical procedures and introduced in table 3. The contribution of both physical-chemical attractiveness and π-π interactions (principally for aromatic adsorbates) are also increased the efficiency of the adsorbent [60]. The attractive force is related to the electrostatic interplays and hydrogen bonding via hydroxyl functional groups existing on the surface of the material [61].

### Table 3. The dyes adsorption capacity of different cellulose aerogels

| Adsorbents       | Adsorbates | q\(_{max}\) (mg/g) | The kinetic model | The isotherm model | pH  | t (°C) | ΔG (kJ) | ΔH (kJ/mol) | ΔS (J/mol) | Ref. |
|------------------|------------|--------------------|-------------------|-------------------|-----|--------|---------|-------------|------------|------|
| Wheat straw      | CR         | 120                | 2\(^{nd}\)        | Langmuir          | 2–11| 8–40   | -9.05–(-0.95)| 29.44        | 33.84      | [62] |
| Coir fibers      | MB         | 62.01 (g/g)       | -                 | Thomas            | -   | -      | -       | -           | -          | [23] |
| Sago pith        | MB         | 222.20             | 2\(^{nd}\)        | Langmuir          | 7   | 20     | -       | -           | -          | [63] |
| Rice straw       | MG         | 212.70             | 2\(^{nd}\)        | Langmuir          | -   | -      | -       | -           | -          | [64] |
| Sorghum stem     | MB         | 11.55              | -                 | -                 | -   | -      | -       | -           | -          | [47] |
| Fe\(_3\)O\(_4\)/MCC | MB      | 57.36              | 1\(^{st}\)        | Freundlich        | -   | -      | -       | -           | -          | [65] |
| Cellulose-MXene  | MB         | 168.93             | 2\(^{nd}\)        | Langmuir          | 3–11| -      | -       | -           | -          | [61] |
| Graphene         | MB         | 111.20             | 2\(^{nd}\)        | Langmuir          | -   | -      | -       | -           | -          | [66] |
| oxide/spruce     | MB         |                    |                   |                   | -   | -      | -       | -           | -          |      |
| kraft pulp       | EBT        | 2297               | 2\(^{nd}\)        | Langmuir          | 3–10| 27–67 | -       | -           | -          | [67] |
Following the Thomas model, the cellulose aerogel from coconut fibers had the rate constant of MB adsorption of 0.00159 mL/mg.min and the equilibrium adsorption capacity of 62 g/g [23]. The modified wheat straw cellulose aerogel via poly(propylene glycol adipate) as a cross-linker had not only stability of network structure but also improved CR adsorption ability (reaching 120 mg/g) [62]. As well as from wheat straw, the fabricated cellulose aerogel by eco-friendly NaOH/PEG solvent system was efficiently absorptive capacities of approximately 16.8, 17.3, and 17.4 times by weight for MO, RhB, and indigo blue [71]. Jeng et al. were fabricated successfully sago pith waste cellulose nanofibrils aerogel, which was efficient in MB eliminating with the highest uptake of 222.2 mg/g at 20 °C [63].

In sub-summary, recently, the improvements in dyes adsorbents by cellulose aerogel have been widely published, but it is mainly functionalized with high-purity sources in terms of nanofibrils [60, 67], carboxylated cellulose nanofibers [70], and microcrystalline cellulose [72], or industrial waste as cotton [68, 69], new-paper [73]. The potentialities of modified-cellulose aerogel by agricultural by-products such as coir, pineapple leaves, rice straw/husk, and bamboo would be extensively researched.

### 3.3. Oils

Some investigation can be informed that the viscosity and density of the oil especially have related to oil-water separation efficiency. Lower viscosity oils are easier to irrupt into the porous microstructure adsorbent, which is resulted in reaching the absorption saturation in a shorter time and more effective adsorption [45, 74]. Therefore, three-dimensional (3D) porous networks of cellulose aerogel convenience for separation are recognized as very potential materials for oil pollutants treatment. However, a challenge for bio-based absorbent materials is their hydrophilicity due to a great deal of -OH groups (i.e., poverty-stricken of oil/water selectivity) and subordinate mechanical handleability (i.e., inferior shape recovery from deformation). As can be seen from table 4, various strategies have been utilized to advance the hydrophobicity and oleophobicity of cellulose aerogels via chemical vapor deposition technique (CVD) using alkoxy groups and silane coupling agents in methyltrimethoxysilane (MTMS), triethoxy(octyl)silane (OTES), trimethylchlorosilane (TMCS), methyltrichlorosilane (MTCS), hexamethyldisilazane (HMDS), or silicon ceramic coating powder (SCCP).
Table 4. The oils adsorption capacity of different modified-cellulose aerogels

| Source              | Characterization materials | Density (mg/cm³) | Porosity (%) | WCA (◦) | Oil Capable uptake (g/g) | Reusability (times) | Ref. |
|---------------------|---------------------------|------------------|--------------|---------|-------------------------|---------------------|------|
| Sugarcane bagasse   | MTMS                      | 16–22            | 91.9–98.9    | 142–147 | crude                   | 25                  | -    | [16] |
| Sugarcane bagasse   | SCCP                      | 12–108           | 92.8–99.2    | 140.1   | crude                   | 23                  | 10   | [41] |
| Sugarcane bagasse   | Alkyl ketene dimer (AKD)  | 47.3             | 95.9         | 148     | remarkable              | 31.65               | 10   | [75] |
| Pineapple fiber     | MTMS                      | 13–33            | 96.98–98.85  | 140     | motor oil 5w30 diesel, pump, corn, mineral, motor diesel, pump, engine, corn | 37.9               | -    | [76] |
| Moso bamboo         | MTMS                      | 102              | 99.4         | 140     | lubricant               | 32–96               | 35   | [77] |
| Bamboo              | GO + TMCS                 | 17.95            | 98.8         | 142     | crude, pump, motor three various oils | 112–130           | 5    | [74] |
| Coir fibers         | TMCS and HMDS             | -                | -            | 110–148 | crude                   | 10                  | -    | [78] |
| Rice straw          | MTMS                      | 2–24             | 98.4–99.8    | 151±7   | crude, pump, motor      | 112–130             | 5    | [45] |
| Rice straw          | MTMS                      | 50–60            | 97           | 141–150 | three various oils      | 13                  | 3    | [44] |
| Canola straw        | HDTMS                     | 11–17.5          | 98.8–99.3    | 121–139 | motor, cooking oil      | 78.8–162.4          | 20   | [79] |
| Posidonia oceanica leaves | Poly (lactic acid)       | 110–230          | -            | 95–130  | sunflower               | 34                  | 34   | [80] |

Crosslinking by thermodynamics occurred the chemical interaction between the lactone rings of AKD emulsion and -OH groups of cellulose bagasse fibrils to form β-ketone esters. The alkyl chains of AKD stretched outward to the fibrils resulting in the hydrophobic structure of AKD-modified aerogels which exhibited an ultrahigh oil spill-cleaning (31.65 g/g) [75]. To modify the hydrophobicity of aerogels, MTMS was firstly acid-hydrolyzed in water to yield silanol groups (Si-OH). When the silanol sol was then dropped by drop into PVA/CNF suspension, two chemical reactions parallelly took place in the mixture: (i) the Si-OH radicals could be directly substituted with the -OH groups of the cellulosic chains thereby constructing Si-O-C bonds, (ii) the Si-OH groups could also condense themselves on the surface, leading to form polysiloxanes with steady Si-O-Si bonds. The silylated sol mixture was continuously frozen by a custom-built freeze-caster. In the final step, polysiloxanes formed stiff layers on the aerogel via dehydration counteractions after the freeze-drying stage [16, 76, 77]. Functionalized-nanofibers aerogel from Canola straw was silylated by stretching vibration of the
Si-O or Si-C bonds in HDTMS, which was quickly absorbed the motor and cooking oil in the first 300 and 42 s, respectively [79].

The water contact angle (WCA) on the aerogel surface characterized the hydrophobicity of the materials, the higher the contact angle of a water droplet, the more non-hydrophilic silane-modified aerogels [81]. Table 4 can be informed that the WCA is generally higher than 120°. Effect of the heating temperature and concentration of TMCS and HMDS on the WCA was researched by Fauziyah et al. The best experimental method of hydrophobicity was established at 50% TMCS and 40% HMDS dissolving in n-hexane with a temperature of 60 and 120 °C [78].

4. Effect of different parameters on adsorption capacity

4.1. Effect of equilibrium time

The primary adsorption occurs rapidly at initial times until the taken time to reach equilibrium. The primary adsorption occurs rapidly at initial times until the taken time to reach maximum equilibrium capacity values. The sorption operations by a large type of bio-sorbents are a mass-transfer controlled process conforming to two sequential steps. The first step is rapidly occurred because of the plentiful and available activities of sites on the surface, in which the adsorbed capability is decreased due to the little-by-little possession of these active linking sites with lasted time. Secondly, the equilibrium step is mostly based on the absolute saturation of the aerogel-sorption with the contaminant molecules [58]. In the beginning period at 0–20 min, the adsorbed rate of Cu^{2+} ion is reached higher than 50%. At this stage, the different concentration of Cu(II) between the solution and adsorbent surface causes the diffusion of the phenomenon. The adsorption process slows down as the reaction progresses and finally reaches equilibrium after 150 min [55]. The removal of basic MG dye onto CNF aerogels quickly happens in the first 40 min, then virtually non-changes beyond 80 min at all initial concentrations (10–400 mg/L) [64]. The difference in contact time may be related to the pore volume, specific surface area, several functional groups of bio-materials, or molecule size of contaminants. The bulky structure or ionic radius can affect of mobility of dyes or ion metals in contaminated solution to move to the adsorbent and cause in extending equilibrium time.

4.2. Effect of pH

Solution pH has been known to be a very significant aspect of adsorption due to it affects the chemical property of not only the adsorbate but also the adsorbent. The magnitude of electrostatic forces between heavy ions and adsorbent materials decides capable of uptake. The pH value increasing from 3 to 7, the amino groups commonly endured in the neutral form, which dopped the electrostatic repulsion between the ions on surface materials and Cu^{2+}, Cd^{2+}, and Hg^{2+} ions, hence increased adsorption capacity [59]. Following Lim et al., the lower pH values medium contained a relatively higher concentration of H^+ protons, which could compete with the Ni^{2+} ions over the amino groups of the tridentate ligands. Furthermore, strong electrostatic repulsions of both the protonated amino groups and Ni^{2+} cations may be disadvantaged for nickel ions to contact with the PF-DETA aerogel to take place adsorption [17]. As the pH adjusting from 6 to 11, a large number of -OH radicals can precipitate with Cr^{6+} ions, causing a decreased adsorption capacity [57]. The pH value (2 to 11) of the solution affected the existence of advantageous radicals on the propylene glycol adipate-modified wheat straw aerogel (PCA) and then influenced the adsorption. Under acidic aqueous, PCA was protonated with positive point charges and strongly adsorbed CR. Under basic conditions, the PCA contained a large amount of -OH on the surface, which generated repulsive electrostatic force with the negative CR dye, resulting in a speedy reduction in adsorption capacity [62]. The highest removal rate of MB of robust cellulose sponge was attained 99.92 % at pH solution of 11, but the removal rate of CV was lower (92.74%). This difference was due to the large hindrance of bulky CV size, which reduced the adsorption capacity [69]. In summary, the contaminated adsorptions by cellulose aerogel may be performed over a wide pH range.
4.3. **Effect of initial concentration**

The change tendency in the initial concentration of the adsorbate and the shape of isothermal adsorption linear can estimate the maximum adsorption capacity of a bio-adsorbent and be applied to describe the interactivity between contaminants and cellulose materials. The adsorption capacity generally decreases with an increasing the initial concentration because of the saturation of active sites on the adsorbent aerogels [64]. The uptake for MB had a rapid upward trend from 162.3 to 421.4 mg/g when the initial MB concentration was changed from 50 to 300 mg/g. Thereafter, the effective adsorption of HKUST-1/CCSA was remarkably smaller with the increasing initial concentration. The adsorption capacity at 600 mg/g was almost similarly kept in as those at 500 mg/g and was becoming less obvious of the rate of uptake [68]. The total MG adsorbed capacity increased from 44 to 206 mg/g at 10 to 400 mg/L original concentrations, with 76 to 99 % in the first 40 min. This rapid adsorption capacity is attributed to the hierarchical meso-/macroporous construction of the rice straw aerogels [64].

4.4. **Effect of temperature**

The mobility of heavy ions or dye molecules is related to the temperature, hence, the thermodynamic parameters in terms of changing in the standard Gibbs free energy (ΔG), enthalpy (ΔH), and entropy (ΔS) are generally investigated and calculated as equation (1) and (2). Almost the results negative ΔG indicates that the uptake process is spontaneous because adsorption is principally dominated by a gradient concentration. Therefore, Gibbs of free energy is more advantageous in concluding that the adsorption is favored at higher or lower temperatures [82]. The ΔH change in the ranges of 20–40 kJ/mol and 80–400 kJ/mol represents physisorption and chemisorption. The positive ΔS performs an enhancing disorder at the solid/liquid interface during the adsorption process [62, 72].

\[
\Delta G = -RT\ln K_d 
\]

\[
\ln K_d = \frac{\Delta S}{R} - \frac{\Delta H}{RT} 
\]

The representation in tables 2 and 3, the elimination of polluted water is the commonly endothermic process and the general randomness of the reaction. The equilibrium adsorption amount increased with the increase of lower temperature 30 ℃ and started to decrease with the increased temperature. It clearly clarifies that the hydrogen clinkers interior the adsorbent is broke down due to high temperature, and the pyretic movement inside the material enhances, lots of dye molecules are desorbed, thus the adsorption ability significantly declines. ΔH = 129.441 kJ/mol indicated that the adsorption process of PCA on CR was an endothermic reaction [62]. According to Geng et al., the values of ΔG at 25–55 ℃ equaled -7.657, -8.047, -8.437, and -8.827 kJ/mol, implying the spontaneous nature of Cu²⁺ ion adsorption by bleached kraft bamboo pulp aerogel. A positive value of ΔH = 3.965 kJ/mol quoted that Cu²⁺ ion elimination on APTMS-modified TO-NFC aerogel was endothermic reaction [59]. At various temperatures, |ΔH| > |TΔS| presents that the adsorption process is controlled by enthalpic rather than the entropic changes [57, 58].

5. **Adsorption models**

5.1. **Adsorption kinetics**

The effect of contact time is utilized to predict adsorption kinetic models through pseudo-first-order (described by equation (3)) and pseudo-second-order (equation (4)) [83].

\[
\ln(q_e - q_t) = \ln q_e - k_1 t 
\]

\[
t = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} 
\]
The pseudo-first-order dynamics presumes that the adsorption speed is proportional to the discrepancy between the adsorbate concentration and the available plots for solid-liquid systems suggesting by Lagergren. The pseudo-second-order equation simulates that the rate-limiting step dominates by the chemisorption and supposes a second-order proportionality between the sorption rate and saturated concentrations. The correlation coefficient $R^2$ value and the comparison of the calculated and experimental adsorption capacity ($q_{cal}$ and $q_{exp}$) are almost utilized standard to validate data fitting. Following these kinetics models, the adsorption process is concerned by physisorption or chemisorption [82, 83].

The data statistic from tables 2 and 3 illustrate the best fitting with pseudo-second-order model that may be principally controlled by chemisorption. For all amino-silylated nano-cellulose aerogels (TO-NFC) to remove Cu$^{2+}$ ion, the conjectured $R^2$ values from the 2nd kinetic model reached more than 0.99, while the $R^2$ ones from the 1st kinetic were 0.94 and 0.91 for the TO-NFC-Si-NH$_2$ (1:0.5) and TO-NFC-Si-NH$_2$ (1:0.8). In addition, the $q_{cal}$ values determining from the pseudo-second-order model are approximated to be 17.1, 26.2, 31.0, and 31.3 mg/g, respectively, which were consistent with the $q_{exp}$ ones (17.3, 25.9, 30.9 and 31.1 mg/g) [59]. The correlation coefficient values $R^2$ deducing by pseudo-second-order model are almost greater than that fitted by pseudo-first-order model, demonstrating that the redox effect of Fe$_3$O$_4$ beads was dominant in the Cr(VI) removal process. The adsorption also plays out with chemical connections due to the formed complexes by the carboxyl and amino groups on the chitosan linker [57].

5.2. Adsorption isotherm

The adsorption process based on the initial concentration can carry out more in-depth research to clarify the mechanism via disparate adsorption isotherms models, such as Langmuir, Freundlich, Tempkin, Thomas, Yoon-Nelson..., in where Langmuir and Freundlich linearization is the most popular studied. While the Langmuir model (equation (5)) assumes identical monolayer adsorption the Freundlich equation (equation (6)) describes a heterogeneous or multilayer adsorption of adsorbent [82, 83].

$$q_e = \frac{K_L q_{max} C_e}{1 + K_L C_e}$$  \hspace{1cm} (5)

$$q_e = K_F C_e^{1/n}$$  \hspace{1cm} (6)

In general, according to tables 2 and 3, the absorption processes usually follow the Langmuir model. The well-fitting Langmuir isotherm showed that MG was adsorbed as a homogeneous monolayer on CNF-rice straw aerogel surfaces at a maximum capable adsorption of 212.7 mg/g [64]. A coefficient $K_L = 0.0103$ (smaller than 1) deducing from the Langmuir model indicated that PCA easily adsorbed CR dye. In addition, $n = 3.125$ is greater than 1 calculating by the Freundlich model, which also performs that PCA easily removes CR solution. However, the linear correlation coefficient of the Langmuir model ($R^2 = 0.9984$) is remarkably greater than that of the Freundlich model (R-square = 0.8444). It clearly represents that PCA has a similar surface of adsorption capacity at different positions and follows a well-fit Langmuir isothermal model [62].

6. Conclusion and Prospects

In recent years, environmental issues related to the uncontrolled discharge the heavy metal ions, dye compounds, or oil spill is a top concern. The elimination of various contaminants of wastewater from industrial areas and the polluted large oceans is an important problem to consider by researchers. The novel adsorbents by three-dimensional 3D-porous aerogels containing the potentiality in removing various contaminants are introduced in the current review paper. The original subjects can be concluded:
Cellulose aerogels with high porosity, high specific surface area, ultra-low density are synthesized from various and abundant agro-biomass. Many green cross-linkings such as PVA, alkaline-urea combinations, amine-functionalized polymers are utilized to replace the toxic derivatizing solvents in the dissolution of cellulose. The functionalization of the cellulose chains in aerogel by amine/carboxyl/magnetic groups can be significantly improved in the efficient adsorption of dyes or heavy metals. For oil-spill cleanup applications, the silylation via chemical vapor deposition methodology by MTMS, HDMS, TMCS to enhance the hydrophobicity is regularly chosen. Removability of contaminants by novel aerogel crucially depend on the contact time, pH solution, initial concentration, and temperature. The adsorption is a spontaneous and endothermic process. Adsorption capacity of polluted chemistry can be presumed by fitting the experimental data with pseudo-second-order kinetics and Langmuir isothermal model.

Although many improvements have been performed in the fabrication, modification, and application of cellulose aerogel in water treatment, a simplified synthesis process is still necessary to develop in industrial manufacture. More in-depth research in removing real water samples is essentially needed.

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