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Review

Bio-Based Polymeric Flocculants and Adsorbents for Wastewater Treatment

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Abstract: With the growing demand for clean and safe water, there is a pressing need to explore novel materials for water treatment applications. In this regard, bio-based polymeric materials have emerged as a promising solution for water purification. This article highlights the numerous advantages offered by bio-based polymeric materials, including their biodegradability, low cost, and renewable nature. Moreover, it discusses in depth the two primary applications of these materials in water treatment, namely flocculation and adsorption, showcasing their effectiveness in removing contaminants. Furthermore, this review addresses the future prospects and challenges associated with the development of bio-based polymeric materials for water treatment applications. This review provides valuable insights for researchers in the field, driving further advancements in the utilization of bio-based polymeric materials to ensure clean and sustainable water resources.

Keywords: flocculation; adsorption; biopolymers; polysaccharide; regenerations; biodegradation; wastewater treatment; sustainability

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1. Introduction

Water is an essential resource for life, and access to clean and safe water is crucial for the well-being of individuals and communities [1]. However, rapid population growth, urbanization, and industrialization have led to global water scarcity and pollution issues. Shockingly, more than 2 billion people lack access to safe drinking water, and around 3 billion people experience water scarcity for at least one month annually, as reported by the World Health Organization [2–12].

Water treatment is the key to removing contaminants and impurities from water, rendering it safe for consumption and various purposes [13–15]. Recently, there has been a surge of interest in utilizing bio-based polymeric materials for water treatment applications [16–21]. Biopolymers offer a cost-effective and practical solution, as they possess numerous active sites that can coordinate with pollutants and integrate with synthetic polymers [22].

Among biopolymers, polysaccharides have gained significant prominence in water treatment [22]. Comprising repeating units of sugar molecules, polysaccharides can be sourced from various natural origins, such as plants, algae, and bacteria [23]. Remarkably, polysaccharides exhibit exceptional water affinity and demonstrate the ability to absorb and eliminate diverse contaminants, such as heavy metals, dyes, and organic pollutants. Notably, chitosan, derived from chitin, stands out as a widely employed polysaccharide in water treatment, offering a remarkable adsorption capacity and antibacterial properties [24–26]. Although there are concerns regarding the water-insolubility and limited biodegradability of cross-linked polysaccharides, such as chitin, chitosan, starch, and cyclodextrin, these materials have found utility in effluent treatment [27].

Synthetic polymers such as polyacrylamide, polyacrylic acid, and polystyrene sulfonic acid, which are soluble in water, have been employed as effective flocculants and adsorbents. The main responsible sites for coordination are $-CONH_2$ groups, $-COO^-$ and $-COOH$

groups, and $-SO_3H$ and $-SO_3^-$ groups, which are present in polyacrylamide, polyacrylic acid, and polystyrene sulfonic acid, respectively [28]. For example, poly(glutamic acid) (PGA) stands out as a biodegradable, edible, and non-toxic anionic polymer widely used in commercial applications and water purification [29,30]. Biopolymers derived from okra (*Hibiscus/Abelmoschus esculentus*) have demonstrated promising results in solid suspended particle removal, comparable to commercial polyacrylamide flocculants [31]. An added benefit of this bio-based flocculant is its ability to be utilized without pH adjustment or the addition of a coagulant.

Acrylamide-based synthetic polymeric adsorbents dominate the commercial market, captivating researchers for decades due to their high reactivity, excellent water solubility, and affordability [32,33]. These water-soluble synthetic polymers have found applications in various water treatment processes, including flocculation, sedimentation, and membrane separation. They effectively eliminate contaminants such as suspended solids, bacteria, and organic pollutants. However, their use is hindered by drawbacks such as toxicity, and non-biodegradability. Consequently, scientists are dedicated to developing polymeric materials that are less toxic, biodegradable, cost-effective, and possess extraordinary contaminant removal capabilities [32,33].

Significant efforts have been undertaken to mitigate the harmful effects of synthetic polymers by employing grafting methods with natural polymer backbones, particularly focusing on the development of acrylamide-free polymeric materials [34]. The emergence of non-acrylamide-based polymers offers a compelling alternative that fulfils the requirements of cost, performance, and biodegradability, without the concerns of potentially carcinogenic residual acrylamide monomers or toxic degradation by-products. These non-acrylamide-based polymer flocculants can be derived from alternative synthetic monomers or, more notably, from bio-based materials. The recent literature demonstrates an encouraging trend toward the synthesis of sustainable and environmentally friendly flocculants and adsorbents [32,35–38].

This review offers valuable insights into the recent advancements in utilizing biodegradable or bio-based polymeric materials for water treatment applications. It sheds light on the numerous benefits associated with the use of bio-based polymeric materials and focus on flocculation and adsorption in water treatment. Furthermore, the review delves into the existing challenges and presents an outlook on the future prospects of utilizing bio-based polymeric materials in the field of water treatment.

2. Methods of Water Purifications

Water purification aims to eliminate harmful chemical compounds, organic and inorganic elements, and biological pollutants from water sources. Various parameters, including pH, odor, color, and taste, are examined to assess water quality and contamination levels. Additionally, assessments encompass factors such as radioactive materials, microbial pathogens, dissolved and suspended solids, as well as organic and inorganic chemicals, such as chloride, copper, manganese, sulfates, and zinc. Home-water purification systems, including boiling, slow sand filtration, and chlorination, are commonly employed. Municipal water treatment systems typically involve slow sand filtering, chlorination, storage sedimentation, and up-flow roughing filtration. While methods such as boiling and activated carbon filtration can eliminate some water pollutants, concerns have arisen in recent years regarding the infiltration of pesticides, fertilizers, and other contaminants into wells from surface sources.

Industrial wastewater treatment encompasses diverse methods for purification, including coagulation [39,40], flocculation [41,42], adsorption [43,44], membrane filtration [45,46], froth floatation [47,48], advanced oxidation techniques [49,50], solvent extraction [51,52], ion exchange [53,54], anaerobic treatment [55,56], microbial treatment [57,58], acoustic cavitation [59], electrolysis [60,61], and activated sludge [62]. Typically, pre-treatment is carried out before subjecting industrial wastewater to these purification processes. The wastewater is initially pumped from its source or transported via pipelines into storage

tanks, after which it is delivered to a water treatment plant for purification. Pre-treatment involves the removal of chemicals, biological pollutants, and other substances such as large particles and debris. Further insights into these processes are provided in Table 1.

Table 1. Details about the water treatment methods.

Water Purification Method	Meaning	Advantages	Limitations
Boiling water	Boiled water in a clean and covered pot.	• Cheapest. • Easily kills microorganisms.	• Requires fuel. • Pesticides cannot be removed.
Sedimentation	Heavy particles settle down at the bottom.	• Reduces water treatment cost. • Reduces chemicals need for water treatment.	• Suspended particles or smaller particles do not settle down. • Microbes do not settle.
Decantation	Separation of immiscible liquid–liquid and liquid–solid mixtures.	• Reduces the turbidity. • Reduces settable solids.	• Unable to reduce pathogens. • Laborious process.
Chlorination	Mixing chlorine in the water.	• Chlorine is readily available. • Kills germs. • Can be used in emergency cases. • Oxidizes organic matter. • Low-cost method.	• Chlorine is a potent biocide. • Chlorinated water is not safe for pregnant women. • It cannot kill Cryptosporidium and Giardia.
Filtration	Contaminated water passes through the sands and activated carbon cartridge.	• Facile method. • Removes toxins from water. • Improves taste and odor. • Removes excess chlorine.	• Cannot completely remove germs. • Filter needs to be cleaned thoroughly. • Cartridge needs to be carefully disposed of.
Coagulation	Inorganic chemicals are applied to remove suspended particles. For electro coagulation, there is no need for chemicals.	• Small particles clump together and settle down. • Toxic metal cannot be removed. • Cannot remove pathogens and pesticides.	• Operation costs are expensive. • High dose needed. • High sludge volume. • Costly disposal. • Required to maintain pH, temperature, and dose.
Flocculation	Adding organic chemicals or polymers to separate colloidal particles or suspended particles.	• The particles' charges are neutralized by coagulation. • Neutralized particles bind together and become larger by flocculation. • Easy floc formation by a polymer.	• Produces a large amount of toxic sludge. • Mixing speed and time affects the removal efficiency. • It cannot make crystal clear water. • Tiny floc makes noticeable turbidity.
Adsorption	Adding adsorbents to the contaminated water and attaching contaminates to the surface of the adsorbent.	• Wide pH range. • High performance. • Easy operation. • Low cost. • Selectivity.	• Adsorbent saturation. • Adsorbent disposal. • Variable performance. • Difficult separation of adsorbents from adsorbates.
Membrane filtration	Physically separates contaminates using pressure difference between the two sides of the membrane. Ultrafiltration, nanofiltration, and reverse osmosis are different membrane filtration processes.	• No requirement for chemicals. • Can remove pathogens. • Energy efficient process. • Greater degree of purity.	• Limited lifetime. • Fouling of membrane. • Expensive method. • Ruptures easily.
Froth floatation	Selectively separates hydrophobic contaminates from hydrophilic contaminates.	• Easy separation of microplastics. • Widely used in minerals separation. • Highest separation capacity.	• Maintenance of pH. • Creates environmental pollution. • High reagent consumption. • High processing cost.
Advanced oxidation techniques	Separation of organic and inorganic contaminates from water using •OH radicals.	• Fast reaction rate. • Reduces toxicity of organic pollutants. • Mineralization of organics matter. • Removes pathogens • No sludge production.	• Expensive process. • Dose depends on the process and is very calculative. • Complex chemistry.

Table 1. *Cont.*

Water Purification Method	Meaning	Advantages	Limitations
Solvent extraction	Separation of aromatic compounds from water, transforming solubility from one solvent to another solvent.	• Separation of sparingly soluble components from water. • Separation of azeotropic mixtures. • High selectivity of separation.	• Highly polar molecules are incompatible. • High operational costs.
Ion exchange	Ionic pollutants separation by exchange with other ions passing through resin matrix.	• Used to neutralize highly acidic or basic water. • Converts hard water into soft water. • Removes toxic contaminates. • Low-cost method. • Ecofriendly.	• Fouling. • Bacterial contamination. • Chlorine contamination. • Organic contamination from resin. • Inadequate regeneration. • Resin degradation.
Anaerobic treatment	Transform organic matters to biogas in the absence of oxygen.	• Low-energy process. • Environmentally friendly. • Less expensive. • Reduces pathogens.	• Cannot remove inorganic components. • Respiration generates lactic acids.
Microbial treatment	Removing pollutants from water using bacteria.	• Environmentally friendly. • Widely used to remove oil from water. • Phenols, nitrates, and food particles become separated.	• Short life span. • Costly process. • Fouling. • Low production rate. • Instability.
Acoustic cavitation	Separation of contaminates using high-power ultrasounds.	• Improves anaerobic digestion. • Pathogen reduction. • Odor removal.	• Damages the impeller. • High energy consumption. • Expensive.
Electrolysis	A sacrificial metal anode and cathode generate electrically active coagulants and tiny bubbles of hydrogen and oxygen in the water.	• Excellent pretreatment process. • Generates fuel energy. • Reduces the total number of suspended solids. • It can be operated by solar energy.	• Regular cleaning of electrodes is needed. • Requires more labor. • Short life span of the electrode. • Several factors affect electrolysis, such as pH, conductivity, gaps between two electrodes, current density, and size of particles.
Catalytic process	Adding catalyst improved the rate of removal of contaminates' selectively. It includes a catalytic oxidation and reduction process.	• Less energy used. • Reduces production cost. • Reusable. • A small amount is needed. • Photocatalysis is an attractive process.	• No sludge or negligible sludge. • May generates toxic products. • Expensive.
Activated sludge treatment	Used microorganisms to treat wastewater.	• Removes organic materials from water. • Low-cost method for installation.	• Not suitable for industrial wastewater treatment. • Sludge needs to be removed and disposed of. • Activated sludge loses activity with time.
Disinfection	It is the final step of the water treatment. Adding disinfectant chemicals, ultraviolet radiation treatment, or ultrafiltration	• UV radiation is a nonchemical, energy efficient, and low-cost treatment. • Removes most bacteria and viruses.	• Chemical disinfectant is limited in its use due to its harmfulness. • UV radiation cannot remove any inorganic metals.

While advanced oxidation techniques and membrane treatment technologies have proven effective in eliminating organic and inorganic pollutants, as well as pathogens, their widespread practical implementation is hindered by high energy consumption and the associated overhead costs. In contrast, flocculation and adsorption methods offer practical advantages due to their simplicity and affordability. Consequently, conducting a comprehensive review that focuses on the role of biopolymers in water treatment, specifically in the context of flocculation and adsorption, is of paramount importance. Such a review would provide valuable insights into these practical and cost-effective approaches for water purification.

3. Flocculation

Flocculation is a common term applied to aggregate formation when soluble polymers are added to the aqueous medium of colloidal particles. Usually, the term flocculation is synonymously used with coagulation, but the techniques are entirely different. Coagulation occurs through the accumulation of charges with the addition of inorganic chemicals. However, organic polymers are used as additives in flocculation techniques. Alternatively, coagulation is formed by aggregation and is an electrostatic phenomenon [63,64]. The aggregates formed are compact because there is little loose space between the constituent particles. A schematic of the flocculation method is shown in Figure 1.

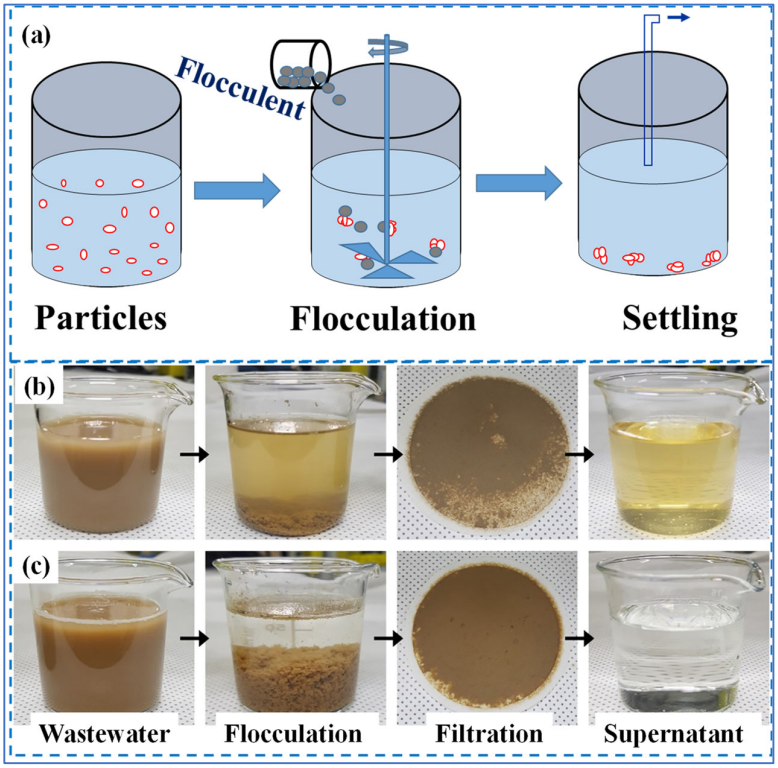


Figure 1. (**a**) A schematic of bench test of flocculation; (**b**) practical use of untreated wastewater; and (**c**) treatment with polyacrylamide [65]. Copyright 2022; reproduced with permission from Elsevier Ltd. (Amsterdam, The Netherlands).

In contrast, flocculation mainly involves the simultaneous adsorption of high molar mass polymeric materials onto the surface of the particle mixture in aqueous medium and form flocs (Figure 2a) [66]. The main driving parameter of suspended particles in water is the van der Waals force, which induces relative stability (Figure 2b) [67,68]. Colloidal particulate suspensions are characterized according to their liquid (water)–solid interface. The first one creates a more distinct interface between the solid and liquid among the hydrophobic and hydrophilic constituent parts. However, the hydrophilic particle does not influence the phase boundary. The mechanism controlling constituent particle's steadiness is mainly static repulsion [69]. The cations and anions in hydrophobic surfaces are accumulated at the boundary, producing electric potential energy that can resist the colloidal particles of the same surface charge. The dissociation of organic acid salts mainly generates electrical potential in the hydrophilic surfaces or boundaries. In addition to the electrostatic repulsion, particulates may also be rationally stable due to adsorbed H_2O molecules that offer a liquid barrier to successful particulate collision. The surface charge of the colloidal particles increases at the adsorption layer. It has two significant effects: (a) repulsion force responsible for the precipitation of particles, and (b) attraction of counter ions to the particle surroundings. These two opposite forces create a diffuse cloud of ions surrounding the

particulate matter. The co-existence of the original surface charge and the neutralizing excess of opposite ions over co-ions dispersed in diffuse mode is the electrical double layer or the ion cloud (Figure 2c) [70]. The electrical double layer around the colloidal particles determines the minimum distance between the two charged particles corresponding to their stable equilibrium. The size of such an ion cloud depends on several factors: (a) the magnitude of the surface charge depending on the adsorbing ion concentration and (b) the electrolyte concentration in the solution. A higher concentration corresponds to the greater approachability of two close particles.

The ion cloud close to the solid surface forms the stern and diffuse layers, referred to as the Gouy–Chapman layers [71]. The oppositely charged ions are attracted to the surface of the solid and form the stern layer.

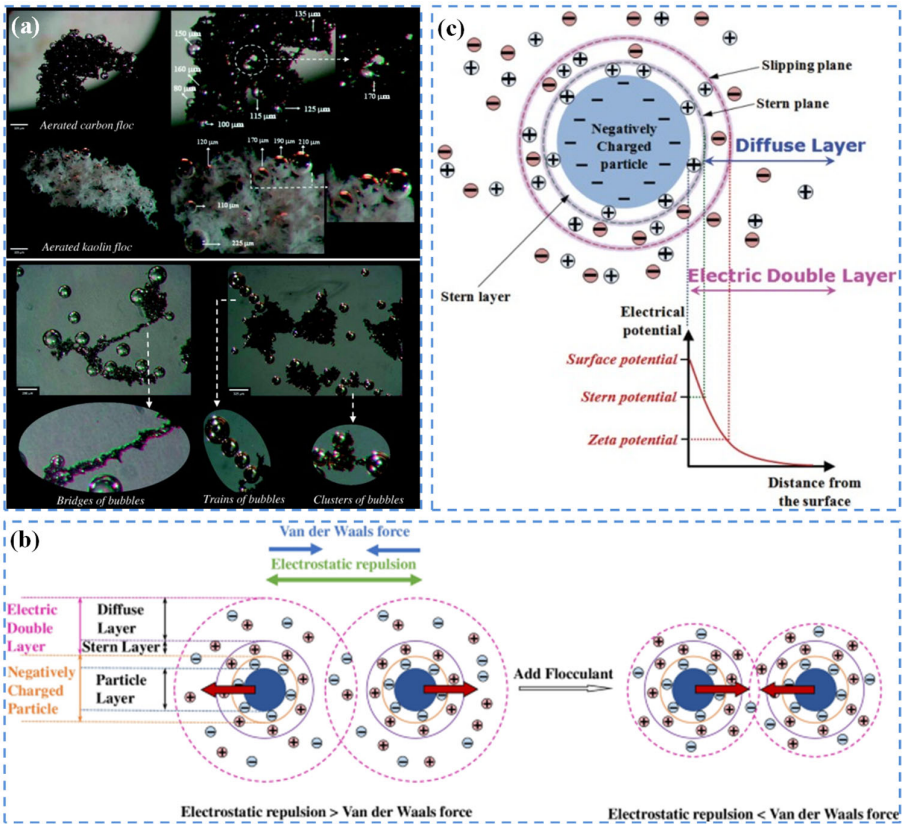


Figure 2. (**a**) Aerated carbon and kaolin flocs by polymer [66]. Copyright 2012; reproduced with permission from Elsevier Ltd. (**b**) Van der Waals force interaction [68]. Copyright 2022; reproduced with permission from Saudi Society for Geosciences. (**c**) Diagram of electric double layer [70]. Copyright 2011; reproduced with permission from Elsevier Ltd.

The oppositely charged solid's surface also attracts the same charged ions of the layer, and simultaneous repulsion is governed by the exact charges in the stern layer. The ionic attraction and repulsion equilibrium form a diffuse layer [72]. A double layer is formed through the Gouy–Chapman layer. The inner layer and the diffuse double layer significantly impact the actions of particulate surfaces in an aqueous medium [73]. The perspective of the stern layer considerably affects the stability of suspended particles.

The colloidal particles may come closer or may remain as a stable suspension, which is well-known as the DLVO theory [74]. This theory is fundamental to the steadiness of colloidal suspension balancing between electrostatic repulsion and the van der Waals force of attraction. This idea additionally states that an energy barrier ensuing from the repulsion force prevents approaching these two particles and adhering together.

Flocculants

A diverse variety of coagulants and flocculants are available for wastewater treatment. Commonly, these are classified into two categories: inorganic compounds and organic compounds. Inorganic coagulants (such as iron chloride, aluminum sulphate, aluminum chloride, polyaluminum chloride, aluminum chlorohydrate, and iron sulphates) and organic flocculants (such as polymeric materials or polyelectrolytes) are usually used. Extensive tonnage usage of inorganic coagulants generates much sludge. Environmental and ecological concerns justify using biodegradable flocculants in wastewater and industrial effluent treatments. Polymeric flocculants (both synthetic and natural) are favored owing to their low dose, facile, less sludge generation, and varied tailorability.

Polymer flocculants can be categorized based on their source as natural and synthetic. Synthetic polymers are classified into four categories based on surface charge: cationic, anionic, non-ionic, and amphoteric. Cationic flocculants are more active than anionic flocculants in treating powerfully and negatively charged particle suspensions and vice versa. Some synthetic polymeric flocculants that are commercially used for effluent treatment are shown in Table 2.

Table 2. Commercial polymeric flocculants with their charge and uses in various effluents.

Sl. No.	Commercial Polymeric Flocculants	Charge	Type of Effluent Used	Optimum Results (TSS %)	Ref.
1.	Polyacrylamide	Anionic	Slaughterhouse	94.0	[75]
2.	Polyacrylamide	Non-ionic	Beverage and food units	97.0	[76]
3.	Organopol 5415	Cationic	Paper mills	99.5	[77]
4.	Chemfloc 430A	Anionic	Pulp industries	99.5	[77]
5.	KP 1200B	Cationic	Ceramic (Boron)	80.0	[78]
6.	AP 825C	Anionic	Ceramic plants (Boron)	40.0	[78]
7.	Actipol A-401	Anionic	Sauce manufacturing units	72.0	[79]
8.	Magnafloc 155	Anionic	Polymer effluents	91.0	[80]
9.	Polyacrylamide	Amphoteric	Paper mills	95.0	[81]
10.	Flocan 23	Anionic	Olive mills	~100	[82]
11.	Magnafloc LT 7991 Magnafloc LT 7992 Magnafloc LT 7995 Magnafloc LT 22S Magnafloc E 38	Cationic	Aquaculture	89.0 84.0 84.0 91.0 45.0	[83]
12.	Hyperfloc CE 854 Hyperfloc CE 834 Hyperfloc CE 1950	Cationic	Aquaculture	98.0 87.0 94.0	[83]
13.	Organopol 5415	Anionic	Pulp and paper mills	97.0	[84]
14.	Polydiallyldimethylammonium chloride	Cationic	Textile dye	90.0	[85]
15.	Polyamine	Cationic	Dye effluents	96.0	[86]
16.	Polydiallyldimethylammonium chloride	Cationic	Wastewater of paper mills	~100	[87]
17.	Poly etheleneoxide	Non-ionic	Tale, graphite, chalcopyrite, and covellite dispersions	98.0, 99.5, 82.0 and 93.0	[88]

Table 2 shows that acrylamide-based synthetic polymeric materials are effectively used in practical wastewater treatment. Controlling molecular weight and chemical structures can enhance the efficiency of these polymers. However, synesthetic polymers are not shear resistant.

Recently, bio-based flocculants have attracted considerable interest due to their advantages over typical synthetic polymers or inorganic agents. Biopolymers, mostly polysaccharides, are shear resistant, biodegradable, non-toxic, and easily obtained from agricultural or forest resources. Biopolymer-based flocculants have a variety of excellent advantages, such as a wide pH range, adaptability, ease of modification, being eco-friendly, their mini-

mum dosage requirements, and high flocculation efficiency [89]. Important and naturally available water-soluble biopolymers primarily used in the field of water remediation are discussed in Table 3.

Table 3. List of readily available biopolymers.

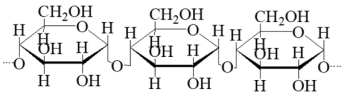
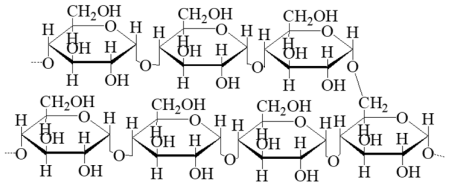
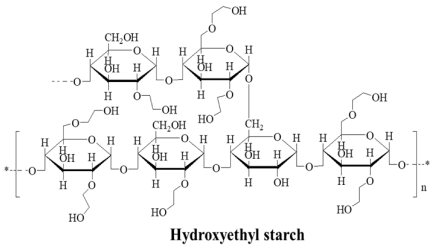
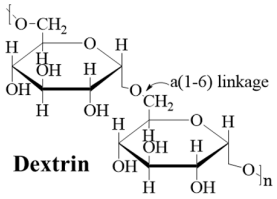
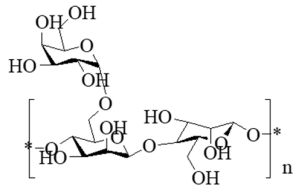
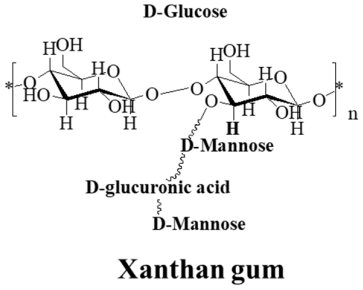
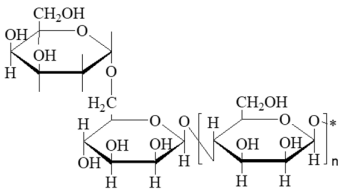
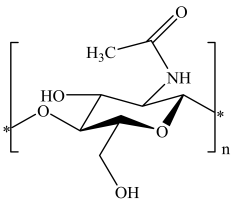
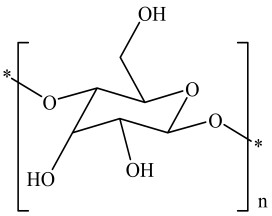
Name	Properties	Chemical Structures	Ref.
Starch	Sources: seeds and stem crops, wheat, potato, corn, sago, sweet potato, arrowroot, cassava, and rice. Chain of D-glucose-α-glycosidic linkages mixtures with amylose [(1-4)-α-D-glucan)] and amylopectin [(1-4)-α-D-glucan and (1-6)-α-D linkages].	 Amylose  Amylopectin	[90]
Hydroxyethyl starch	It is chemically modified starch and dextrin. Primarily used in the medical specialty pharmaceutical field and industrial effluent treatment.	 Hydroxyethyl starch	[90]
Dextrin and cyclodextrin	Potato, corn, wheat, rice, and tapioca all contain cyclic oligosaccharides.	 Dextrin	[91]
Guar gum	Plant's polysaccharide; linear chain of mannose and galactose.	 Guar gum	[92]
Xanthan gum	Natural polysaccharides: Chain β-(1→4)-linked-d-glucopyranose glucan and side chain β-(1→3)-α-linked d-mannopyranose-(1→2)-β-d-glucuronic acid-(1→4) β-d- mannopyranose.	 Xanthan gum	[93]

Table 3. *Cont.*

Name	Properties	Chemical Structures	Ref.
Locust bean gum	Source is the seeds of carob trees: linkage of (1-4)-β-D mannose and the side chain of (1-6)-α-D galactose.	 **Locust bean gum**	[94]
Chitosan	Source is the shells of shrimps, fish scales, crabs, and lobsters. Linkage of β-1,4-D-glucosamine.		[95]
Cellulose	Main source of cellulose is wood, cotton, sugarcane bagasse, grass, and banana peel. Covalently linked β-D-anhydroglucopyranose units.		[96]

However, the quick biodegradation of biopolymers reduces their shelf life and needs careful modification. The biopolymer modification could increase the flocculation efficiency of biopolymer-based flocculants or obtain multifunctionality of flocculants [97,98]. Some chemical modification processes include graft copolymerization, etherification, acetylation, esterification, acylation, oxidation, cross-linking, and the Mannich reaction [99]. Compared to others, the grafting process is more straightforward and cheap. The biopolymer-based graft copolymers for flocculants and adsorbents are described in below.

Recently reported graft copolymers and their capacity to remove various pollutants from water at pH are shown in Table 4.

Table 4. Recently reported biopolymer-based graft copolymer flocculants for water treatment.

Sl. No.	Flocculent	Pollutant	Results (%)	Refs.
1.	AP-g-PAM	Iron ore Kaolin Coal Silica Paper mill wastewater Municipal wastewater	89.3%, pH 6.5 86.8%, pH 7.6 90.5%, pH 7.5 43.4%, pH 6.5 70.9%, pH 6.5 88.3%, pH 6.8	[100,101]
2	AP-g-PDMA	Iron ore Kaolin Coal Silica Paper mill wastewater Municipal wastewater	93.5%, pH 6.5 93.3%, pH 7.6 90.9%, pH 7.5 85.8%, pH 6.5 72.5%, pH 6.5 90%, pH 6.8	[100]
3	AP-g-PAA	Mining industries' wastewater	87.31%	[102]

Table 4. *Cont.*

Sl. No.	Flocculent	Pollutant	Results (%)	Refs.
4	AP-g-PAA	Paper mill effluent Mine process water Textile wastewater	82.1% 81.8% 98.0%, pH 8.2	[103]
5	AP-g-poly (AM-co-ATMAC)	Kaolin	5.4 NTU	[104]
6	Dxt-g-PAM	Kaolin suspension	Settling velocity 0.0370 cm/s	[105]
7	HES-g-PAM	Iron ore Kaolin Coal Silica Paper mill wastewater Municipal wastewater	91.8%, pH 6.5 87.2%, pH 7.6 91.3%, pH 7.5 51.5%, pH 6.5 71.8%, pH 6.5 88.9%, pH 6.8	[100,106]
8	HES-g-PDMA	Iron ore Kaolin Coal Silica Paper mill wastewater Municipal wastewater	95.5%, pH 6.5 93.8%, pH 7.6 92.17%, pH 7.5 87.4%, pH 6.5 74.0%, pH 6.5 90.5%, pH 6.8	[100,106]
9	HES-g-Poly (DMA-co-AA)	Iron ore Kaolin Coal Silica Paper mill wastewater Municipal wastewater	89.5%, pH 6.5 94.5%, pH 7.6 90.1%, pH 7.5 51.0%, pH 6.5 76.4%, pH 6.5 77.7%, pH 6.8	[100]
10	St-g-PAM	Iron ore Kaolin Coal Silica Paper mill wastewater Municipal wastewater	86.4%, pH 6.5 83.9%, pH 7.6 88.2%, pH 7.5 42.2%, pH 6.5 69.6%, pH 6.5 79.3%, pH 6.8	[90,100]
11	St-g-PDMA	Iron ore Kaolin Coal Silica Paper mill wastewater Municipal wastewater	92.42%, pH 6.5 85.9%, pH 7.6 89.1%, pH 7.5 83.0%, pH 6.5 70.2%, pH 6.5 88.2%, pH 6.8	[100]
12	St-g-poly(AM-co-2-methacryloyloxy ethyl trimethyl ammonium chloride)	Blast furnace effluent	19.7 NTU	[107]
13	St-g-poly (2-methacryloyloxyethyl) trimethyl ammonium chloride)	Kaolin suspensions	25 NTU, pH 4.0	[108]
14	St-g-poly(2-methacryloyloxyethyl trimethyl ammonium chloride)	Kaolin Escherichia coli suspensions Kaolin and Escherichia coli suspensions	98.7%, pH 11.0 95.5%, pH 11.0 98.7%, pH 11.0	[109]

Table 4. *Cont.*

Sl. No.	Flocculent	Pollutant	Results (%)	Refs.
15	St-g-poly(AM-co-sodium xanthate)	Kaolin and $CuSO_4$	~100%, pH 5.0	[110]
16	St-g-poly(AM-co-AA)	Kaolin suspensions	~100% transparent	[111]
17	Barly-g-PMMA	Coal fine Iron ore Kaolin Municipal wastewater	86.0% 75.0% 82.0% 67.61%, pH 7.2	[112]
18	CMC-g-p(DADMAC)	Kaolin suspension Coal suspension Iron ore	80.9% 83.6% 79.5%	[113]
19	Moringa gum-based	Coal River water Kaolin MWCNTs	90% 91% 92% 98%	[114]
20	Chitosan-g-acrylamidopropyltrimethylammonium chloride	Kaolin	90%	[115]
21	Kraft lignin-based polymers	Kaolin	62.91%	[116]
22	Carboxymethylcellulose-AM-4-vinylpyridine	Kaolin Bentonite	75% 89%	[117]
23	Starch-Based Flocculants	Kaolin Hematite	97.7%, pH 11 98.6%, pH 7	[118]

The flocculation performance of the grafted polymers from the starch family, such as starch-grafted-poly(acrylamide), starch-grafted-poly(dimethyl acrylamide), starch-grafted-poly(dimethyl acrylamide-co-acrylic acid), amylopectin-grafted-poly(acrylamide), amylopectin-grafted- poly(dimethyl acrylamide), amylopectin-grafted-poly(dimethyl acrylamide-co-acrylic acid), hydroxyethyl starch-grafted-poly(acrylamide), hydroxyethyl starch-grafted-poly(dimethyl acrylamide), and hydroxyethyl starch-grafted-poly(dimethyl acrylamide-co- acrylic acid), were assessed in four synthetic suspensions of iron ore slime (0.25 wt.%), kaolin clay (1.0 wt.%), coal (1.0 wt.%), and silica (1.0 wt.%) [90,100]. The results showed that the polysaccharide-based PDMA graft copolymers exhibited higher flocculation properties than the polysaccharide-based PAM polymers in coal, silica, and municipal wastewater. However, polysaccharide-based poly(DMA-co-AA) showed better performance than the PAM- and PDMA-based polymers in iron ore slime and kaolin clay suspension. This is due to the fact that grafting polyacrylamide branches onto polysaccharides, which results in dangling grafted chains that are incredibly approachable to pollutants [119]. It has recently been found that the PAM-grafted biopolymer backbone is stiffer due to intramolecular hydrogen bonding between the >C=O and $–NH_2$ groups of PAM molecules, restricting the approachability of the grafted polyacrylamide chains to suspended particles compared to PDMA molecules. The absence of hydrogen bonding in PDMA chains increases the flexibility and approachability of PDMA chain-grafted polysaccharides to contaminants [90,100]. Binary groups such as $–CONMe_2$ and –COOH in the biopolymers' backbone could be beneficial for the adsorption compared to the flocculation performance. The static repulsion decreased the flexibility of the grafted chains, decreasing the flocculation characteristics. The binary monomers and polysaccharide chain types in graft copolymers affect their water treatment performance. For example, hydroxyethyl starch-g-PDMA shows better flocculation performance than starch and amylopectin-based graft copolymers. This may be

due to the branching chains in hydroxyethyl starch that are more flexible and approachable toward pollutants than linear chains in starch and amylopectin [100]. In addition, the hydroxyethyl starch-g-PDMA polymer has a more remarkable ability to remove suspended particles (iron ore slime, silica, and coal suspension) from wastewater than industrially available flocculants such as magnafloc 1011, telfloc 2230, and percol 181, as shown in Figure 3a–f.

This might be due to the linear chains in commercial flocculants such as PAM. On the other hand, acrylic acid (AA) moieties in the graft copolymers showed improved removal of suspended particles from industrial effluents, e.g., kaolin clay [100,102,120]. This is due to the electrostatic interaction between –COOH groups in hydroxyethyl starch-g-poly (DMA-co-AA) chains and aluminum silicate in kaolin clay suspension. Sarkar et al., reported that the AP-g-PAA polymer was used to treat wastewater from mining industries and found that the turbidity decreased from 510 NTU to 64.7 NTU [102]. Recently, a lignocellulose-acrylamide-carboxymethyl cellulose copolymer was synthesized using microwave-assisted copolymerization, which removed 92% of the cationic dye Methylene Blue. This can potentially reduce treatment costs by 32% compared to commercially available anionic poly acrylamide [121].

Furthermore, a carboxymethyl cellulose-acrylamide-dimethyl diallyl ammonium chloride cationic polymer was synthesized using the γ-ray co-irradiation grafting approach, which removed 86% of the kaolin suspension from wastewater [122]. On the other hand, grafting copolymerization was used to develop a new, environmentally friendly natural polymer bagasse cellulose-based flocculant that significantly improves the removal (91.6%) of humic acid from lake water [123]. Furthermore, by grafting methacrylic acid 2-(benzyldimethylamino) ethyl chloride and 2-(methacryloyloxy)-N,N,Trimethylethanium chloride, cationic starch-based flocculants with variable hydrophobicity and charge densities were synthesized, named CS-DMLs and CS-DMCs, respectively. These polymers provided a new and effective method for designing graft natural polymeric flocculants that were used effectively to condition sludge at a cheap cost and with excellent performance. More cationic groups in the St-based flocculants improved their capacity to neutralize charge for more efficient agglomerations of negatively charged sludge particles.

Additionally, the polymeric flocculants with a greater charge density increased the length of the chain conformation in the solution, which enhanced the bridging flocculation effects. Therefore, with the charge density increase, the dewatering performance of CS-DMCs and CS-DMLs was enhanced (Figure 3g–h) [124].

From the above discussion, it has been found that there are three main mechanisms in flocculation: surface charge neutralization (Figure 4a), polymer bridging, and electrostatic patch. Surface charge neutralization completely neutralizes charged particles by opposite charges on the adsorbent surface. In contrast, the electrostatic patch mechanism involves the adsorption of charged particles onto localized charge patches on the adsorbent surface. Both mechanisms rely on electrostatic forces, but the degree of neutralization differs [125,126].

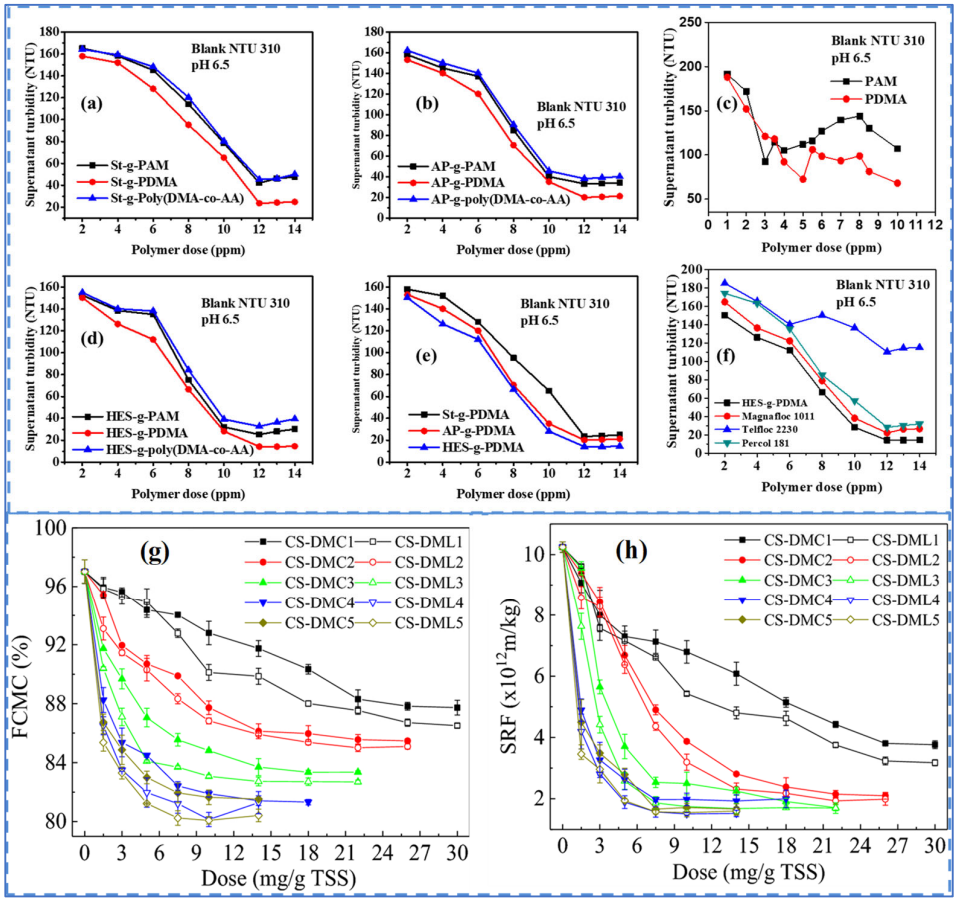


Figure 3. The flocculation characteristics in an iron ore slime colloidal suspension of PAM-, PDMA-, and poly (DMA-co-AA)-grafted (**a**) St and (**b**) AP, (**c**) monomers, (**d**) HES, (**e**) comparison of best performance, and (**f**) evaluation of best performance with three commercial flocculants [100]. Copyright 2016; reproduced with permission from Springer Nature. Copyright 2012; reproduced with permission from Wiley. Dose effects of CS-DMCs and CS-DMLs on sludge dewatering performance: (**g**) filter cake moisture content and (**h**) specific resistance of filtration [124]. Copyright 2021; reproduced with permission from Springer Nature.

In the polymer bridging mechanism (Figure 4b), polymer chains attach to the surface of colloidal particles, creating loops and tails in the surrounding solution. These loops and tails can then bind to nearby particles, forming larger aggregates or flocs. The adsorption of polymers can occur through various forces such as van der Waals forces, hydrogen bonding, and chemical reactions. This bridging mechanism is more effective with high molecular weight polymers (>106 Da) that have the same charge as the colloidal particles. The degree of branching, chemical composition, and molecular weight of the polymers affect the efficiency of this mechanism [99]. Flocs formed through polymer bridging are generally larger, stronger, and more stable, even under high shear rates, compared to those formed through charge neutralization, electrostatic patch mechanisms, or in the presence of metal salts. The flexibility of polymeric chains allows for stretching before the flocs break, contributing to their stability. The formation of flocs reaches an equilibrium or steady state, depending on the applied shear rate or stirring speed [127].

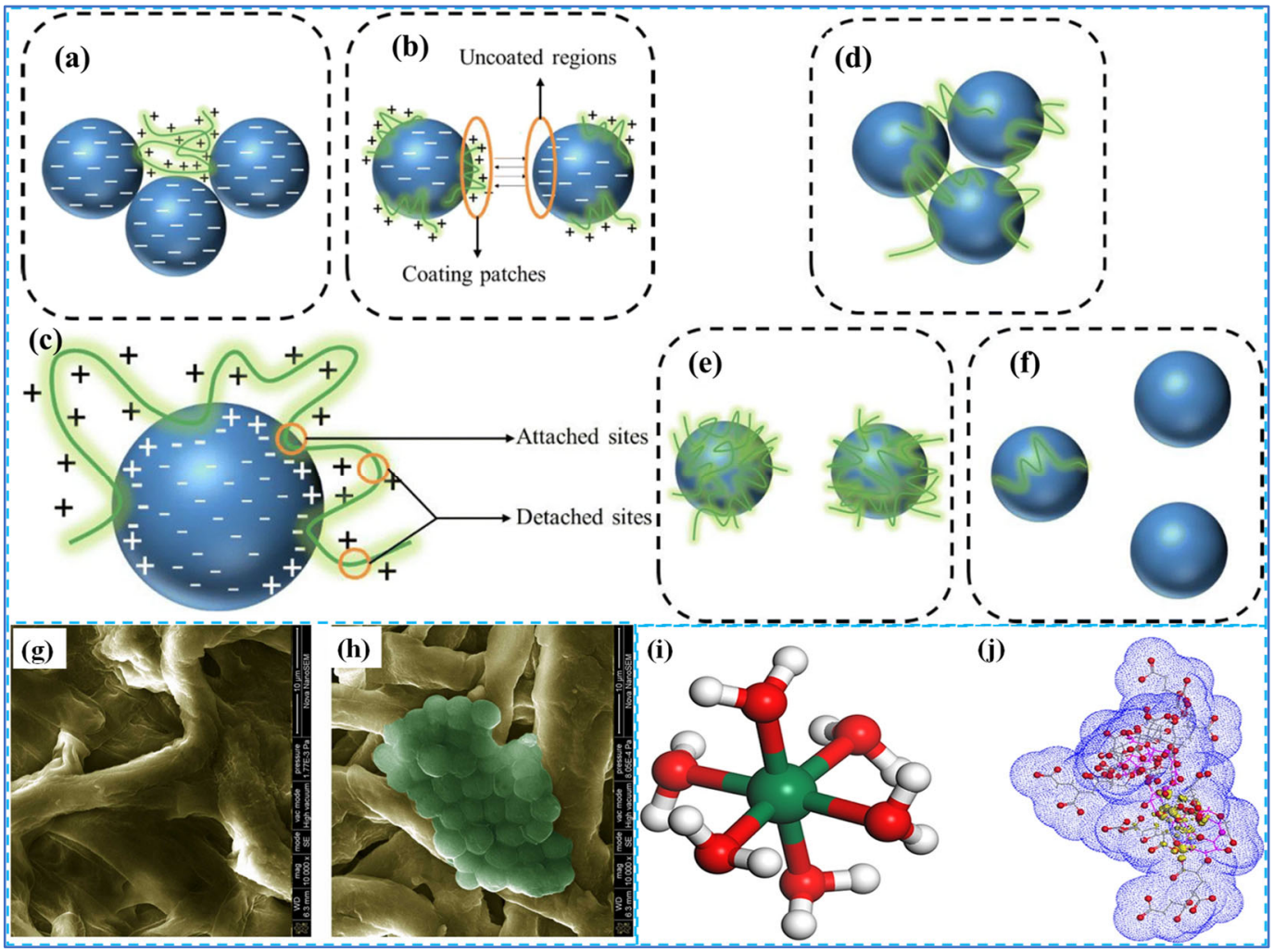


Figure 4. Flocculation mechanism of surface charge neutralization, (**a**) ordinary, (**b**) patching mechanism, or (**c**) adsorption mechanism; polymer bridging mechanism (**d**) at an optimum dose of flocculants, (**e**) excessive dose, and (**f**) inadequate dosage [99]. Copyright 2021; reproduced with permission from Springer Nature. (**g**) SEM images of fungi and (**h**) fungi-microalgae composite structures [128]. Copyright 2022; reproduced with permission from Elsevier B.V. DFT (**i**) Cr $(OH)_6$ and (**j**) Cr (III) adsorption by polymer chain bridging [129]. Copyright 2016; reproduced with permission from Society of Plastics Engineers.

Furthermore, the chains involved in polymer bridging are more resistant to breaking under higher shear levels. While floc breakage may be irreversible, they may not reform, even at lower shear levels [130]. Long-chain polymers with a high molecular weight are cost-effective, non-ionic, and suitable for bridging. The charge density of polyelectrolytes plays a crucial role in the efficiency of bridging, as the repulsion between charged fragments affects the expansion and straightening of polymer chains, making them more accessible to contaminant particles [131]. However, an excessively high charge density can hinder the absorption of particles with similar charges by the polymeric segments due to the loss of flexibility and rigidity of the polymer chains [132]. For instance, Figure 4g–j depicts optical microscopy images illustrating the absorption of Microcystis aeruginosa cells by *A. oryzae* fungal-microalgae using flocculation treatment [128]. Notably, the structural integrity of the microalgal cells remained intact throughout the flocculation process, which effectively minimized the release of extracellular secretions and the need for further processing.

In the case of the electrostatic patch mechanism, polymers with low molar mass and high charge density are moderately effective in flocculating anionic colloidal suspensions.

In the presence of highly cationic polymers and anionic suspensions, the high interaction energy leads to a flattened adsorbed alignment, individually reducing the formation of loops and tails for bridging particles. Instead, the ionic charges on the polymer segments form opposing charges with patch-like regions. Particles near the polymeric segments with patch formation are influenced by the opposite surface charges and are attracted electrostatically. The flocs formed through this mechanism are weaker than those formed through bridging, but stronger than those formed through simple charge neutralization or the presence of metal salts.

Furthermore, a higher ionic strength can accelerate flocculation through the polymer bridging mechanism, whereas lower ionic strength favors the electrostatic patch mechanism [133]. Additionally, several factors impact the performance of flocculation, including the type of colloidal solution and flocculants (their charge), the dosage of flocculants, solution pH, ionic strength, mixing speed, mixing rate, temperature, and the specific mechanism involved. Optimization of these factors is necessary to achieve efficient flocculation [134].

Moreover, the size of the flocs formed is a critical parameter in the flocculation process. Although the most commonly employed method for water treatment worldwide involves aggregation combined with gravity separation or sedimentation (as shown in Figure 5a), this approach incurs high financial and environmental costs due to the production of approximately 8 million tons of metal-based sludge waste annually from the chemicals used, such as coagulants. To address the challenges of process sustainability, cost, and efficiency, researchers have developed re-engineered materials from virgin or waste fibers that function as super-bridging agents, adsorbents, and ballast media. Forming a giant flock could reduce the settling time and size of the settling tank. The formation of larger flocs can reduce the settling time and the size of the settling tank. A recent study by Lapointe et al., demonstrated the enhancement of floc size using super-bridging fibrous materials in industrial water treatment [135]. These fibrous materials were synthesized using cellulosic fibers grafted with silicon (Si). Both pure fibers and fibers derived from recycled paper were utilized to create Si-fibers and porous Si-microspheres. In conjunction with a coagulant, synthetic flocculant, and a bioflocculant extracted from potato residue, these Si-grafted materials, along with other waste fibers such as polyester and cotton, were employed to maximize floc size, density, and ultimately enhance the efficiency of the contaminant removal. The fibrous materials exhibited the formation of supersized flocs, which were ten times larger than those achieved through traditional treatment methods. This approach improved settling with minimal doses of coagulants and flocculants, resulting in reduced capital and process costs, as well as an eco-friendly footprint [135], as depicted in Figure 5b–f. Here, the researchers focused on flock removal using a screening process instead of settling. One significant advantage of screening, as illustrated in Figure 5g, is that the removal of flocs is primarily determined by their size rather than their density and settling velocity. The study demonstrated that using reusable grafted fibers, fiber-based microspheres, and fiber-based flakes in conjunction with screening can greatly reduce the chemical consumption in water treatment and decrease the size of water treatment facilities. The formation of Si-grafted cellulosic fiber materials and the mechanistic approach for separating contaminants from water were explored.

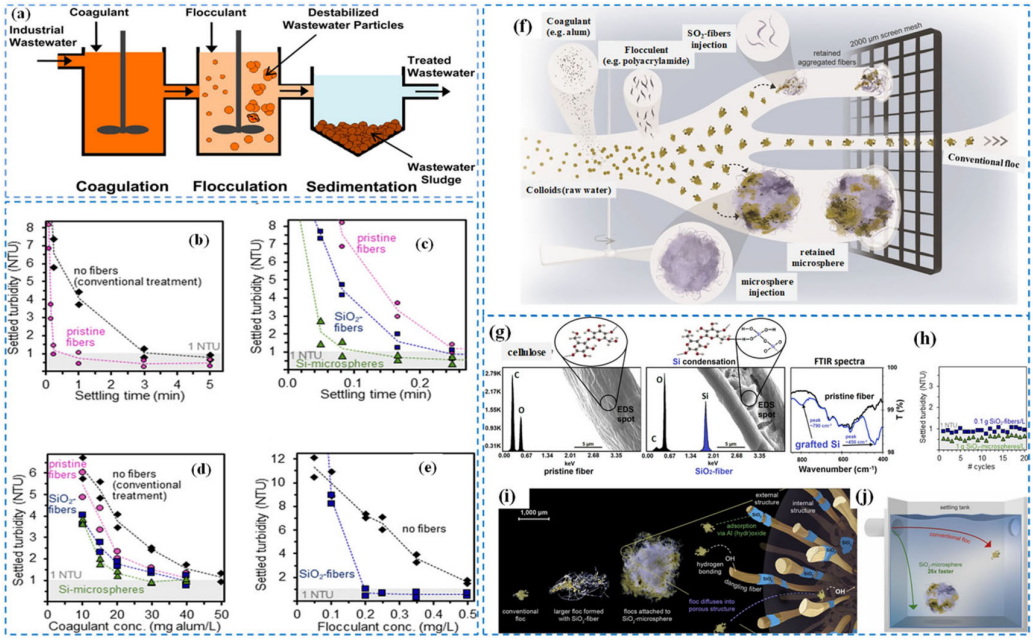


Figure 5. Provides several illustrations and information related to the topic. In (**a**), it shows the sedimentation process [136]. Copyright 2016; reproduced with permission from American Chemical Society. Figures (**b**,**c**) depict the impact of different fibers (pristine cellulosic fibers, Si-fibers, and Si-microspheres) on turbidity removal rates compared to conventional treatment (no fibers). (**d**,**e**) shows the effect of fiber materials on the minimum concentration of the coagulant (alum) and the flocculant (polyacrylamide). The conditions for these experiments involved a flocculant consisting of 50% starch and 50% polyacrylamide, a coagulant concentration of 30 mg/L (alum), and a pH of 6.5 +/− 0.2. Dashed lines connect the average results from independent studies. The shaded area after treatment represents the industry standard for turbidity (1 NTU). Figure (**f**) shows screening methods for water treatment, while (**g**) presents the characterization of Si-fibers using SEM-EDS and FTIR analysis. (**h**) displays the settling cycle for turbidity removal; (**i**) compares floc formation with conventional treatment; and (**j**) provides a schematic of floc size in a settling tank [135]. Copyright 2022; reproduced with permission from Springer Nature.

Furthermore, iron oxide-grafted cellulosic fibers were found to form large flocs that could be easily separated using settling techniques and innovative screening methods. These fibrous materials showed the potential to reduce the demand for coagulants by approximately 60% in removing natural organic matter during multiple water treatment cycles [137]. The fibers also demonstrated the ability to remove the targeted pollutant phosphorus from synthetic wastewater using either a lower dose of coagulant or no coagulant at all. Jar tests, which simulated the water treatment process, were conducted to evaluate the bridging capacity of the Fe-grafted fibers. Extensive characterization techniques were employed to analyze micrographs of the iron-grafted fibers, and the results are presented in Figure 6a–d. A comparison was made between the fiber-bridged flocs, the conventional flocs formed solely with alum, and the flocculant polyacrylamide (PAM), as depicted in Figure 6e–k.

The flocculation process discussed above presents important advancements in water treatment. The use of high molecular weight polymers in the polymer bridging mechanism leads to the formation of larger and stronger flocs compared to other mechanisms. Optimizing parameters such as flocculant type, dose, pH, and mixing speed are crucial for efficient flocculation. The development of super-bridging fibrous materials, such as Si-grafted cellulosic fibers, offers a sustainable and cost-effective solution, reducing the settling time and chemical consumption. Screening-based floc removal, focusing on floc size rather than density, improves efficiency and reduces the size of treatment facilities. Characterization techniques provide valuable insights into fiber properties and functional-

ity. These advancements contribute to enhanced contaminant removal, reduced chemical consumption, and improved process sustainability in water treatment.

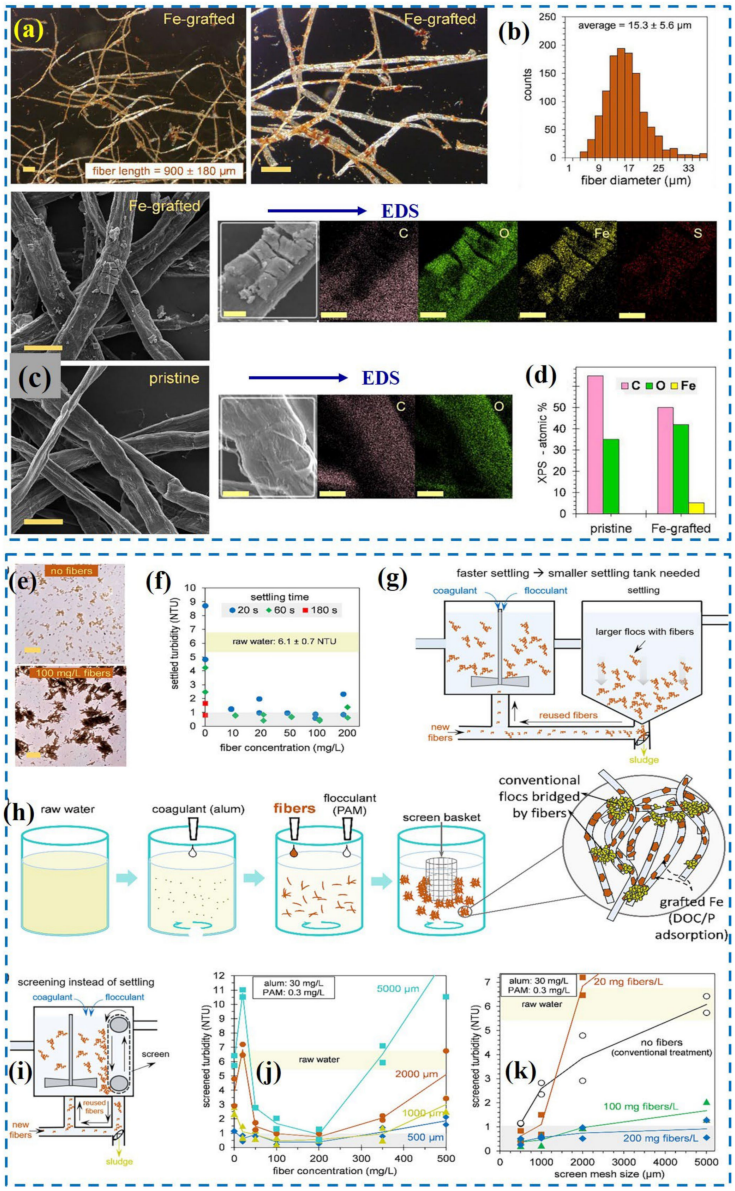


Figure 6. Characterization and experimental results of the Fe-grafted cellulose fibers in water treatment [137]. Copyright 2022; reproduced with permission from Elsevier. The characterization of cellulose fibers involves several steps, as described below: (**a**) Optical images of Fe-fibers showing the average fiber length (scale bars = 100 µm). (**b**) Distribution of fiber diameter after surface functionalization. (**c**) Morphological analysis (SEM, left panels, scale bar = 20 µm) and elemental analysis (EDS, right panels, scale bar = 5 µm) of Fe-grafted and pristine cellulose fibers. (**d**) Surface chemical analysis of Fe-grafted cellulosic fibers in water treatment. (**e**) Microscope images of flocs after jar tests, comparing samples without fibers and with 100 mg/L of fibers (scale bar = 5000 µm). (**f**) Settled turbidity at different time intervals. (**g**) Illustration depicting the traditional treatment process, which can utilize a smaller settling tank due to the formation of larger flocs with the presence of fibers. (**h**) Illustrations demonstrating the screening-based jar tests and the resulting post-treatment flocs. (**i**) Schematic illustrating how screening is integrated into the flocculation tank instead of settling, enabling the separation of flocs. (**j**) Turbidity levels with different fiber concentrations using screens with various aperture sizes (500, 1000, 2000, and 5000 µm). (**k**) Screened turbidity as a function of screen aperture size for specific fiber concentrations.

4. Adsorption

Adsorption is a widely used process worldwide for the removal of chemical contaminants from water. It is favored among various water treatment methods due to its simplicity and excellent ability to remove metal ions [138,139] and dye molecules in water [140,141]. Unlike other processes, adsorption is sludge-less and can be effective for both high and low concentrations of pollutants. The presence of numerous active sites on the surface of the adsorbent is crucial for effective adsorption. Activated carbon is commonly used as an adsorbent in wastewater treatment; however, the cost associated with its processing and regeneration increases due to the required energy consumption. Hence, there is a growing emphasis on developing inexpensive alternative adsorbents using industrial and agricultural waste materials.

4.1. Adsorbents

Adsorbents play a crucial role in the process of adsorption and are responsible for the separation of mass. The effectiveness of adsorption largely depends on the quality of the adsorbent, which should possess high adsorption activity. Fixed-bed adsorber columns are commonly employed for adsorption, where separation occurs through continuous interaction and equilibration between the fluid and sorbent phases. A single column can undergo hundreds to thousands of equilibrium phases, enabling efficient separation. In industrial wastewater treatment, only a few types of adsorbents are commonly used, including activated carbon, silica gel, zeolites, and alumina. Commercially, a limited number of adsorbents find application in this field. Adsorbents utilized for pollutant sorption in wastewater treatment should fulfill certain requirements, such as cost-effectiveness, high sorption capacity, favorable chemical and physical properties, ability for economic regeneration, and tolerance for significant waste parameters.

In recent decades, there has been significant progress in the development of novel nanoporous materials, allowing for customization of their porosity and surface chemistry. This includes oxide molecular sieves, various forms of carbon (such as carbonaceous molecular sieves, activated carbon, activated carbon fiber, and graphite nanofibers), and zeolites. However, the full potential of these materials in terms of their adsorption capabilities for future applications is yet to be fully exploited. Activated carbon, a widely used sorbent with hydrophobic characteristics, has been extensively researched since World War I for air pollution control. Other desiccants such as activated alumina and silica gel are employed for specific purification requirements. Moreover, polymeric materials have gained attention for industrial wastewater treatment due to their defined pore structures and high specific surface area, which enhance pollutant adsorption. The pore size affects surface area, where smaller pores contribute to a larger surface area, but they should also allow molecules to access the adsorbing surface within the polymer matrix. The interaction between adsorbents and adsorbates is influenced by factors such as pH, temperature, reaction time, and solvents. Various interactions, including van der Waals forces, hydrogen bonding, ionic interactions, π-π bonds, and hydrophobic interactions, play a role in the adsorption process [142]. The selection of polymeric adsorbents should consider both their ionic or non-ionic properties and the surface chemistry (hydrophobic, ionic, and hydrogen bonding), in relation to the properties of the adsorbates, such as solvent polarity and molecular structure.

Currently, the majority of commercially available synthetic polymeric adsorbents are derived from acrylamide (as shown in Table 5). Acrylamide has been a subject of interest for many years due to its high reactivity, water solubility, and low cost as a monomer. However, the use of acrylamide in water purification has raised concerns regarding its carcinogenic properties, leading to ecological and health risks [143]. With increasing regulations on air and water pollution, there is a growing demand for cleaner environments. To address these concerns, improved sorbents are required, but they are not yet commercially available. The development of sorbents has traditionally been empirical, but to overcome the current challenges, tailor-made sorbents need to be designed based on fundamental principles.

Table 5. Synthetic polymeric adsorbents, with their charge and uses in various effluents.

Sl. No.	Commercial Polymeric Adsorbents	Charge	Type of Effluent Used	Maximum Adsorption Capacity (mg/g)/Removal (%)	Ref.
1.	Poly(quaternary ammonium salt)	Cationic	Acid Blue 25 and Acid Red 18 dye removal from single and binary systems	2000 and 1667	[144]
2.	Epicholorohydrin-dimethylamine polyamine	Cationic	Bentonite clay	94.9% at 323 K	[145]
3.	Jalshakti®	Anionic	Cationic dye such as Safranine T, Methylene Blue, Malachite Green, Brilliant Green, Rhodamine B, Crystal Violet, and Basic Fuchsine	181.8, 172.4, 34.2, 17.6, 15.4, 12.9, and 11.7	[146]
4.	Polyacrylamide	Cationic	Kaolinite	46.9% at 298 K	[147]
5.	2-acrylamide-2-methylpropanedimethyl ammonium chloride	Cationic	Activated sludge	Not available	[148]
6.	Polyamidoamine	Cationic	Pb (II) ions	694.4 at 293 K	[149]
7.	Poly (ethyleneimine)	Cationic	Cu (II), Ni (II) and Co (II) ions	Not available	[150]
8.	Poly (ethyleneimine)	Cationic	Methyl orange	275.74 at 323 K	[151]
9.	Poly (4-vinyl pyridine)	Cationic	Cr (VI) ions	72.2% at 298 K	[152]
10.	Poly acryl acid	Anionic	Cu (II) removal	303 at 298 K	[153]
11.	Polyacrylamide	Non-ionic	Methylviolet	1136 at 298 K	[154]

Furthermore, the use of various adsorbents has been reported for the removal of heavy metals. However, these adsorbents often suffer from limitations such as low adsorption capacity, weak interactions with metal ions, and difficulties in extraction and regeneration from water [155]. As a result, there has been a growing interest in the use of carbohydrate biopolymers. Researchers are actively working to develop polymeric adsorbents that are less toxic, biodegradable, cost-effective, and possess a high capacity for contaminant removal and reusability. Numerous research articles, reviews, and book chapters have been published on the use of biopolymer-based adsorbents for adsorbing pollutants, including dyes and metal ions, from wastewater [156,157].

4.2. Biopolymer-Based Adsorbents

Biopolymers derived from renewable natural resources are abundant, non-toxic, and biodegradable, making them highly suitable as adsorbents. These biopolymers possess unique structures and physicochemical characteristics, including chemical reactive groups such as hydroxyl, acetamide, or amino functions in their polymer chains, which contribute to their chemical stability, high sensitivity, and selectivity. Carbohydrate-based biopolymers such as chitosan, lignin, carboxymethyl cellulose, and alginate have gained significant attention as adsorbents due to their affordability, effectiveness, and biocompatibility. These biopolymers exhibit various functional groups, such as amine, hydroxide, phenolic hydroxyl, methoxyl carboxyl, and high hydrophilicity, which enhance the removal efficiency of heavy metals from water by chelating metal ions and forming complexes. Chitosan, in particular, with its abundant amino and hydroxyl groups, offers excellent adsorption capacity and biodegradability [158]. However, its limited functionality, solubility in acidic media, mechanical properties, swelling ratio, and inadequate specific surface area hinder its widespread use as an adsorbent. To overcome these limitations, chitosan can be physically, chemically, or biologically modified to create various forms such as nanofibers, nanopar-

ticles, microspheres, membranes, and scaffolds. Chemical modification can be achieved through cross-linking and blending. It involves esterification, etherification, N-alkylation, and graft copolymerization [159]. Surface modification of chitosan is also an effective strategy to enhance its adsorption capacity and other characteristics, which can be achieved through cross-linking, grafting, or functionalization techniques (Figure 7a) [160]. Porous carbon materials derived from biomass have also emerged as cost-effective and renewable adsorbents for water purification and the recovery of metal ions and organic dyes. These materials offer porous structures and accessibility derived from biomass, making them promising alternatives for clean water production (Figure 7b,c) [158,161,162].

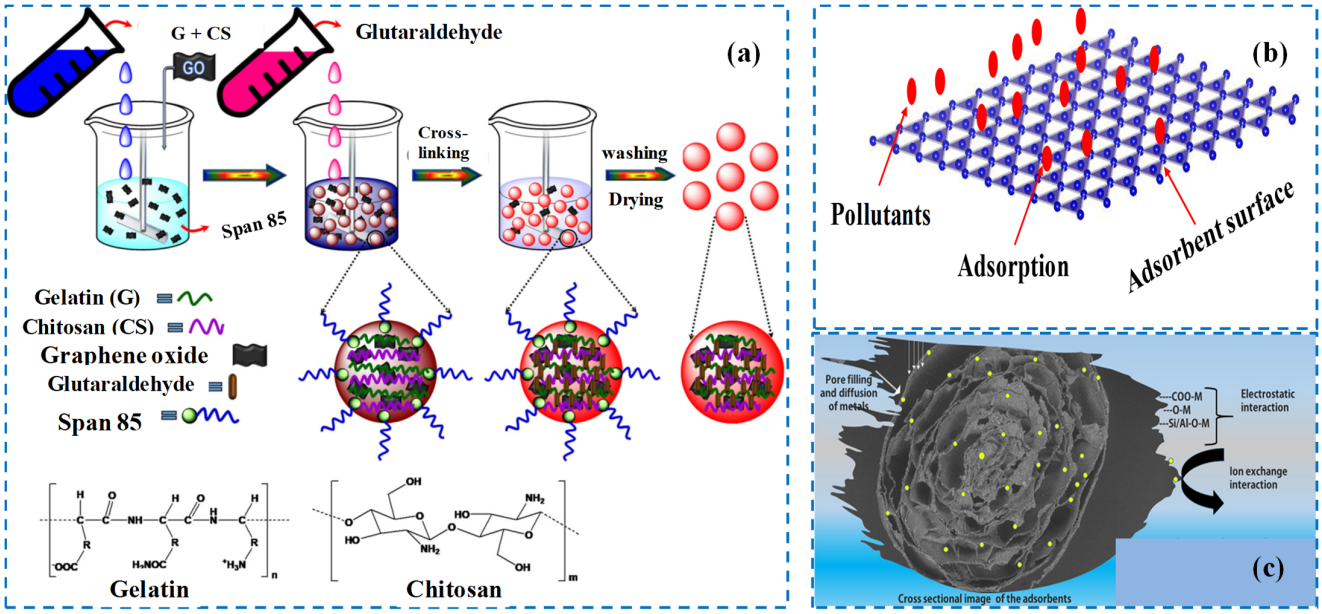


Figure 7. (**a**) Schematic preparation of cross-link polymer [160]. Copyright 2020; reproduced with permission from Springer Science Business Media, LLC, part of Springer Nature. (**b**) Schematic surface interaction and adsorption sites for pollutant adsorption. (**c**) Cross-sectional SEM image of adsorbent with pollutants (metal ions) [162]. Copyright 2021; reproduced with permission from American chemical society.

Lignin, a common aromatic biopolymer derived from renewable plants and a waste product of pulping industries, has emerged as an affordable carbon precursor. Lignin-derived porous carbon (LPC) has shown significant progress in producing clean water from organic dye effluents [163–167]. However, simultaneously recycling organic dyes has proven to be challenging. The preparation of LPC often requires large amounts of activating agents or pore-forming compounds, leading to increased costs and resource loss. Additionally, LPC's performance in producing clean water and recycling organic dyes is hindered by its specific surface area and microstructure. To overcome these limitations, the design of LPC with a remarkable porosity microstructure and minimal chemical usage is crucial [168]. Recently, a cost-effective LPC with a nanoporous hierarchical structure and high specific surface area was synthesized, demonstrating a high adsorption capacity for organic dyes (979.3 mg/g) [168]. Figure 8a illustrates the synthetic process and surface morphology of the material. The characterization results, shown in Figure 8b–g, provide additional insights. To assess the adsorption capabilities of LPC-0 and LPC-3, two cationic dyes (BG and CV) and two anionic dyes (AF and O-II) were used at a high concentration of 700 mg/L in adsorption studies (Figure 8h). LPC-3 exhibited superior adsorption performance compared to LPC-0 for all four organic dyes. The UV–Vis adsorption spectra indicated the complete disappearance of the peak at 485 nm for O-II after two minutes (Figure 8i). During the first minute of adsorption, a slight reduction in color was observed

(Figure 8j). However, LPC-3 successfully removed O-II molecules within 5 min, enabling the collection of clean water during subsequent filtration (Figure 8k). These findings highlight the efficient adsorption capabilities of LPC-3 and its potential for effective water purification processes.

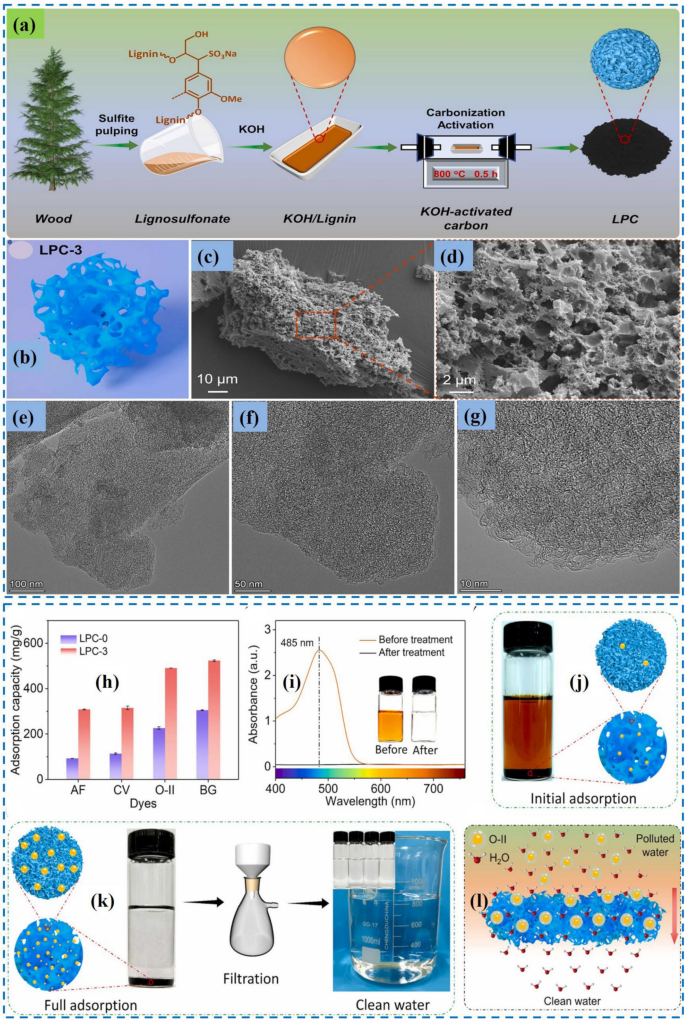


Figure 8. (**a**) Schematic of LPC preparation. (**b**–**d**) SEM and enlarged SEM micrographs of the LPC. (**e**–**g**) TEM images of LPC. (**h**) The AF, CV, O-II, and BG adsorption capacity of LPC-0 and LPC-3 in 10 min. (**i**) UV–Vis adsorption spectra and color change photos of the O-II solution before and after 2 min of LPC-3 treatment. (**j**) The initial adsorption status of O-II and LPC-3. (**k**) Complete O-II adsorption on LPC-3 and formation of pure water. (**l**) Schematic depiction of the LPC-3 filter treatment intercept of clean water production. [168]. Copyright 2022; reproduced with permission from Elsevier.

Recently, there has been an increasing focus on the development and performance of bio-based polymeric adsorbents for water treatment. Table 6 presents a compilation of recent reports highlighting various biopolymer-based adsorbents. These adsorbents utilize biopolymers that are derived from renewable sources and exhibit promising adsorption properties for the removal of contaminants from water. The table provides valuable insights into the types of biopolymers used, the target pollutants, and the reported adsorption capacities.

Table 6. Reported bio-based polymeric adsorbents for water purification.

Sl. No.	Adsorbent	Pollutant	Results % or Q_{max} mg/g	Ref.
1.	AP-g-poly(DMA-co-AA)	Cu (II), Ni (II), and Zn (II)	11.23 mg/g, 8.02 mg/g, 7.77 mg/g	[139]
2.	AP-g-poly(AM-co-AA)	Pb (II)	58.8 mg/g	[169]
3.	AP-g-Poly(NMA-co-AA)	MG	99.1%	[101]
4.	AP-g-poly(MA-co-sodium acrylate)	Cd(II)	5.742 mg/g	[170]
5.	AP-g-AM (sulphated) AP-g-PDMA (sulphated) AP (sulphated)	CR CR CR	66.97 mg/g 51.41 mg/g 37.18 mg/g	[141]
6.	AP-g-poly (AM-co-ATMAC)	MB Kaolin	97.74%, pH 7 5.4 NTU	[104]
7.	Cross-linked starch polymer	Acetophenone 1-phenylethanol	1.29 mg/g 0.70 mg/g	[171]
8.	Guar gum/bentonite	Pb Crystal Violet	72.5 mg/g 48.40 mg/g	[172]
9.	HES-g-Poly (DMA-co-AA)	MG	93.53%, pH 5.5	[120]
10.	Polyaluminum-$FeCl_3$-St-g-poly (AM-co-Dimethyl diallyl ammonium chloride)	Dye blue KN-R Green KE-4B Textile wastewater	89.7%, ~100%	[173]
11.	St-g-poly(AM-co-dimethyl diallyl ammonium chloride)	Disperse Yellow E-3G Dye blue KN-R Reactive Green KE-4B Disperse blue 2BLN	96.0% 89.8% 96.0% 96.0%	[174]
12.	St-phosphate	Pb (II)	99.99%, pH 5.5	[175]
13.	St-g-poly(N,N-Diethylaminoethyl methacrylate) hydrogel	Direct Red 81	95.65%, pH 1.0	[176]
14.	St-g-3-chloro-2-hydroxypropyl trimethylammonium chloride	Sludge dewatering S-EPS LB-EPS TB-EPS	76.40% 50.90% 48.30%	[177]
15.	$FeCl_3$-St-3-chloro-2-hydroxypropyl trimethylammonium chloride	Sludge dewatering of extracellular polymeric substances (EPS) Soluble EPS Loosely bound EPS Tightly bound EPS	93.5% 55.4% 53.9%	[177]
16.	St-g-poly(AM-co-AA)	MB	2276 mg/g	[178]
17.	St-g-DADMAC	Kaolinite and minerals from drilling fluids	98%	[179]
18.	Cross-linked dibenzo-18-crown-6-St	Cd (II) Zn (II) Ni (II) Cu (II)	45 mg/g 42 mg/g 50 mg/g 84 mg/g	[180]
19.	St-g-poly(DMA-co-AA)	Cr (VI)	6.70 mg/g	[181]
20.	St-g-poly(NMA-co-AA)	Hg (II)	11.0 mg/g	[182]
21.	Chitosan on alkali-activated inorganic material	Zn (II) and Cu (II)	98–100%	[183]
22.	St-g-poly(2-methacryloyloxyethyl trimethyl ammonium chloride)	Kaolin Escherichia coli suspensions Kaolin and Escherichia coli suspensions	98.7%, pH 11.0 95.5%, pH 11.0 98.7%, pH 11.0	[109]

Table 6. *Cont.*

Sl. No.	Adsorbent	Pollutant	Results % or Q_{max} mg/g)	Ref.
23.	St-g-dimethylaminoethyl methacrylate	Cu (II) Pb (II)	2.12 mg/g 2.09 mg/g	[184]
24.	St-g-poly(AM-co-sodium xanthate)	Kaolin and $CuSO_4$	~100%, pH 5.0	[110]
25.	St-g-(AA–chitosan)	Acid Blue 113	99.7% pH 4–10.0	[185]
26.	St-g-poly (AM-co- DMDAAC)	Oil—sludge suspension	93.6% transmittance, pH 6.0	[186]
27.	Dxt-g-poly(hydroxyethyl methacrylate)	Methylene Blue Red rose Bengal Fe (III) Cu (II)	91.6% 88.6% 92.4% 81.9%	[187]
28.	St-g-poly(AM-co-AA)	Kaolin suspensions	~100% transparent	[111]
29.	St-g-PAA	Crystal Violet	0.8 mg/g, pH 12.0	[188]
30.	St-g- 3-chloro-2-hydroxypropyltrimethyl ammonium chloride	Red 195 Golden yellow SNE	101.73 mg/g 120.14 mg/g	[189]
31.	Chitosan-g-AM-fulvic acid	Acid blue 113 MO Reactive Black 5	97.0% 91.6% 38.2%	[190]
32.	Carboxylated cellulose nanocrystals	MB Cu (II)	95.6% 82.3%	[191]
33.	Opuntia stricta	TSS Color	80% pH 10.6 77.8%, pH 10.6	[192]
34.	AA-g- xanthan gum hydrogel	Rhodamine B	2777.77 mg/g, 323 K	[193]
35.	Arabic gum-g-PAA/PAM	MB	2300 mg/g, pH 7.0	[194]
36.	Peach gum	MB and MV	98%, pH 6–10	[195]
37.	Modified chitosan beads	Hg (II)	2.3 mmol/g, pH 7	[196]
38.	Leuconostoc citreum B-2 extracellular polysaccharides	Pb (II) Zn (II)	269.54 mg/g, pH 5.5, 298 K 49.88 mg/g, pH 5.5, 318 K	[197]
39.	Alginate-hydrogel	Pb (II) Cd (II) Ni (II) Cu (II)	534.25 mg/g 258.6 mg/g 187.0 mg/g 224.5 mg/g	[198]
40.	Polyacrylic acid/starch graft copolymer	Cd (II)	588 mg/g	[199]
41.	Modified HEMP	Cd (II), Co (III), Ni (II), Cu (II), Mn (II), and Zn (II)	80–100%	[200]
42.	Corn silk/sepiolite	Pb (II) Zn (II)	93.13%, pH 5.5 89.04%, pH 5.5	[201]
43.	Algae residue CCLR	Cu (II), Pb (II), Cd (II), and Mn (II)	78.1, 108.8, 87.3, 57.8 mg/g	[202]
44.	Activated carbon–chitosan beads	Diclofenac	99.29 mg/g	[203]
45.	Lignin-based superhydrophobic melamine resin sponges	Oil/water	98.6%	[204]
46.	Lignin-derived hierarchical porous carbon	Organic dye	1980.63 mg/g	[205]
47.	Macroporous cellulose	Methylene Blue	454 mg/g	[206]
48.	Oxidized cellulose	Pb (II) and Mn (II)	272.5 and 52.9 mg/g	[207]
49.	Carboxymethyl Cellulose Nanofibrils	Cu (II)	380.03 ± 23 mg/g	[208]
50	Pyridine-2,6-dicarboxylic acid cross-linked chitosan	Cu (II)	2186 mg/g, pH 7.5	[209]

The adsorption of dyes by an adsorbent increase at a high pH (>5.5) due to the enhanced ionic interaction between the color particles and the adsorbent molecules. However, for metal ion adsorption, pH 5.5 is considered optimal, as higher pH levels can lead to the precipitation of metal hydroxides, resulting in lower metal ion adsorption by the polymer. Table 6 highlights the recent literature featuring superior polymers for the removal of dye molecules and metal ions from wastewater, with reported removal efficiencies ranging from 90% to 100%. While the focus has predominantly been on removing organic color molecules, rather than metal ions, it is worth noting that most investigations have been carried out using synthetic solutions rather than real industrial effluents. However, Sarkar et al., demonstrated the efficacy of naturally abundant polymer-based flocculants/adsorbents for treating wastewater from mining industries [102]. The AP-g-PAA polymer exhibited a remarkable 99% removal of Malachite Green dye (352.11 mg/g) and metal ions such as Mn (II), Fe (III), Ca (II), and Mg (II) from paper mill effluents [103]. Additionally, Zhou et al., recently reported that natural peach gum could efficiently remove a large amount of cationic dye (98%) from water within a short period of 5 min. Even after undergoing five cycles of desorption–adsorption, the regenerated peach gum retained a high adsorption capacity [195]. Furthermore, adsorbents derived from renewable materials such as rice husk, peach gum, castor seed shell, banana peel, wheat shell, and peanut hull have been explored for color removal from aqueous solutions [195].

5. Commercial Applications

Bio-based polymeric materials have gained significant commercial interest and application in water treatment for industrial wastewater. One notable example is poly glutamic acid, an anionic polymer that is non-toxic and biodegradable. It has emerged as a promising candidate for industrial wastewater treatment. Recent research reports have highlighted its effectiveness in removing metal ions and color molecules from water, including both synthetic and industrial effluents, with removal rates ranging from 90% to 100%. In addition, bio-based polymeric materials, such as chitosan and its derivatives, have demonstrated excellent adsorption capabilities for heavy metals and pharmaceutical contaminates. Chitosan, derived from the shells of crustaceans, possesses amino and hydroxyl functional groups that can complex with metal ions. Studies have shown the effective adsorption of metals such as lead, cadmium, and copper using chitosan-based adsorbents. Activated carbon derived from bio-based sources has adsorbed organic contaminants such as phenols, dyes, pesticides, and pharmaceutical compounds effectively. The commercial use of biopolymeric materials in water treatment offers several advantages. Firstly, these materials possess higher porosity, surface area, and pollutant removal effectiveness compared to traditional acrylamide-based polymeric materials. This enhanced performance is attributed to the unique properties of biopolymers, such as activated multifunctionality and a larger surface area, which make them suitable as adsorbent materials. The trend in recent research is focused on synthesizing more sustainable and green flocculants, adsorbents, and catalysts for the treatment of industrial effluents. By copolymerizing acrylamide with biopolymers or other monomers, the toxicity of polyacrylamide can be minimized while improving its biodegradability. This approach allows for the development of alternative materials that are less toxic and more environmentally friendly.

6. Regeneration of Polymeric Adsorbents

Adsorption is a widely used non-destructive approach for removing pollutants from water. However, one of its primary drawbacks is that adsorbents retain impurities without degrading the pollutants. To address this issue, regeneration techniques are employed to restore the adsorbents' adsorption capacity and enable their reusability. Regeneration is crucial for cost-effectiveness, resource conservation, waste reduction, and recovering the adsorbate. Figure 9a presents a schematic of basic regeneration techniques [210]. Desorption is a common method used to regenerate adsorbents by removing the adsorbed pollutants. The effectiveness of the recovery, decontamination, and regeneration of used adsorbents

determines their reusability. A suitable sorbent can be reused and recovered for commercial and industrial applications, significantly reducing the cost associated with synthesizing new adsorbents. However, regenerated adsorbents generally exhibit a lower adsorption capacity compared to fresh adsorbents [210]. Choosing the appropriate regeneration process is essential for enhancing the pollutant desorption performance. Several factors need to be considered, including the type of adsorbent, eco-friendliness, cost-effectiveness, and energy requirements. Various regeneration strategies have been investigated, but none have been implemented on a large scale [211]. Some of the crucial regeneration approaches for pollutant-loaded adsorbents include microwave-assisted regeneration (Figure 9b) [212], electrochemical regeneration (Figure 9c) [213], ultrasound regeneration, microbial regeneration, thermal regeneration techniques, and chemical regeneration [210,211]. These techniques have been extensively discussed in the referenced articles. The electrochemical regeneration method, in particular, shows promise, as it can effectively regenerate adsorbents that are loaded with both organic and inorganic species. It offers higher regeneration efficiency and porosity recovery compared to thermal or chemical regeneration methods. However, the overall regeneration rate is slow, and the fouling of electrodes can increase the regeneration cost. It is worth noting that there is no single regeneration approach that can effectively regenerate the wide range of adsorbents available today. The optimal regeneration strategy depends on the specific adsorbent, regeneration time, overall cost of the adsorption–regeneration process, and environmental considerations [211]. Disposing of used adsorbents and recovering adsorbate pollutants are significant practical challenges.

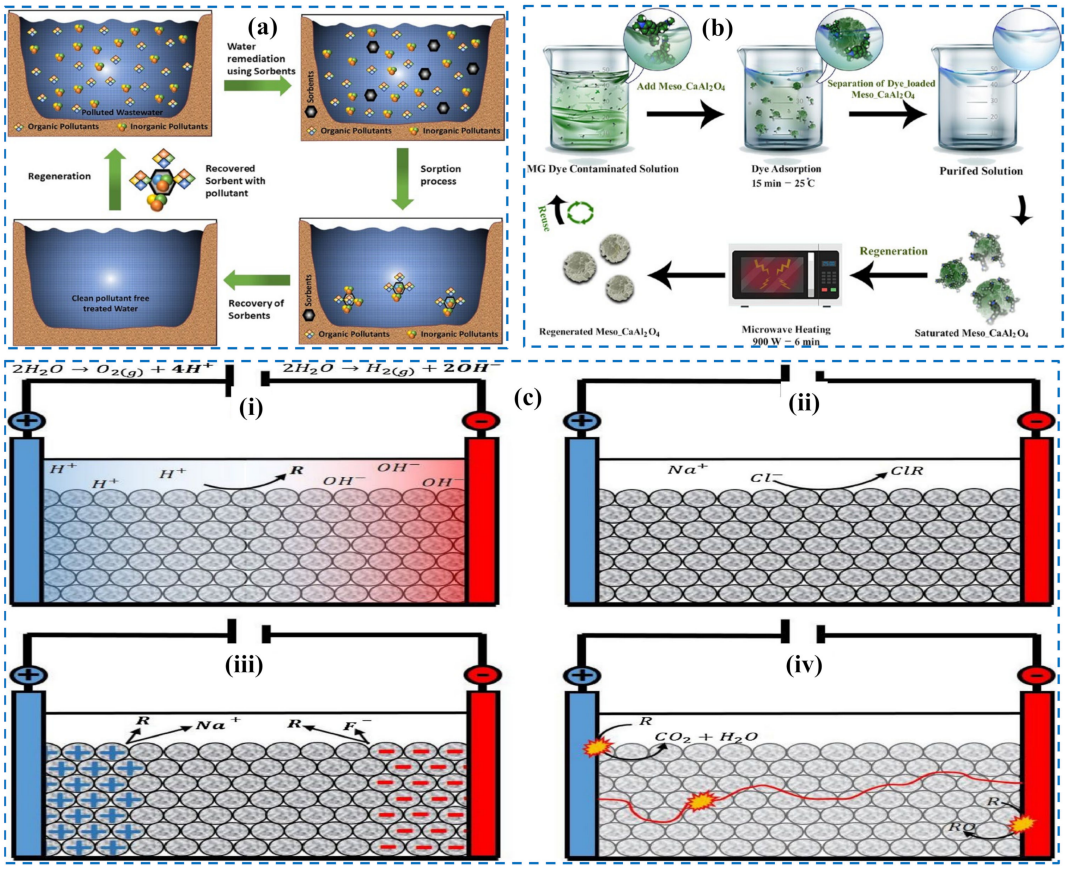


Figure 9. (**a**) Schematic of basic regeneration techniques [210]. Copyright 2022; reproduced with permission from Elsevier B.V. (**b**) MG dye adsorption by meso-CaAl2O4 and microwave-assisted regeneration [212]. Copyright 2021; reproduced with permission from Springer Nature. (**c**) Electrochemical regeneration and enhancement of desorption. (**i**) pH change; (**ii**) salinity; (**iii**) electrode sorption; (**iv**) oxidation/degradation reaction of granular activated carbon reactions [213]. Copyright 2018; reproduced with permission from Elsevier B.V.

Researchers and companies will need to continue exploring and developing ideal regeneration strategies, taking into account the specific characteristics of the adsorbents, regeneration time, cost implications, and environmental impacts. Finding sustainable solutions for regeneration is crucial for ensuring the long-term viability of adsorbent-based water treatment processes.

7. Biodegradation of Polymeric Materials

Earlier studies have investigated the regeneration of polymeric materials from loaded pollutants, with a particular focus on the biodegradability of these polymers. Biodegradation refers to the degradation of materials through the metabolic activities of microorganisms [214]. Fungi, bacteria, and algae play important roles in the biodegradation of chemical components. Biopolymers or natural polymers can undergo degradation in the environment, leading to the formation of biomass, CO_2, and CH_4 [215]. Several factors influence the biodegradation of polymeric materials, including the stability of functional groups, hydrophilicity, swelling properties, reactivity, molecular weight, and surface morphology.

In a study by Kolya et al., the enzymatic hydrolysis of polysaccharide-based graft copolymers and virgin polysaccharides using α-amylase was observed [120,139,181,182,216]. *α-amylase* is an enzyme capable of cleaving the glycosidic linkage (1,4-α-D-glycosidic) in starch chains, releasing glucose residues. The concentration of glucose can be determined by measuring the optical density (OD) using a photoelectric colorimeter after adding DNS (2,4-dinitrosalicylic acid) at pH 6.7 in a phosphate-buffered solution (PBS) [216]. The results of the study are summarized in Figure 10a [120,139,181,216]. The polysaccharides exhibited a more rapid degradation compared to the graft copolymers. This can be attributed to the modification of the polysaccharide backbones through the grafting technique. Consequently, the polysaccharide contained a higher number of glucose moieties compared to the graft copolymers. As a result, the release of glucose was relatively lower for the graft copolymers in the solution treated with α-amylase. Additionally, chemically modified starch (HES) exhibited a slower degradation compared to starch and amylopectin. Overall, polysaccharide-based graft copolymers demonstrate biodegradability.

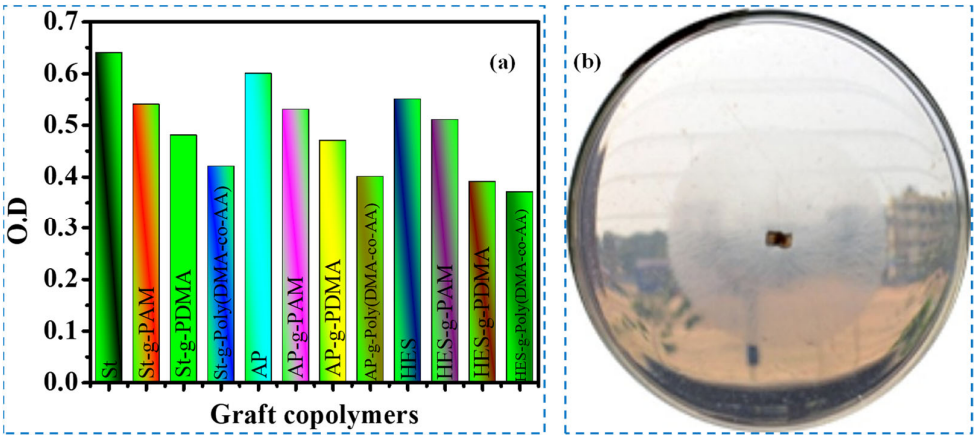


Figure 10. (**a**) Biodegradation studies by α-amylase enzyme and (**b**) growth of *Fusarium sp.* on a partially hydrolyzed AP-g-PAM after 7 days [170]. Copyright 2016; reproduced with permission from Elsevier B.V.

Furthermore, a study by Sasmal et al., demonstrated the potential of *Fusarium sp.* for degrading partially hydrolyzed AP-g-PAM. The growth of *Fusarium sp.* on the partially hydrolyzed AP-g-PAM was observed in a sterilized modified Czapek–Dox medium, as depicted in Figure 10b [170]. This finding suggests that *Fusarium sp.* can play a role in the degradation of the polymer. Moreover, the enzyme *lysozyme* has been found to be effective in degrading polysaccharide-based nanocomposite hydrogels (referred to as NHA), resulting in improved removal efficiency of MG dye. In a PBS medium, lysozyme

was able to degrade 55% of the NHA within a span of 15 days [217]. Based on these studies, it can be concluded that polysaccharide-based polymers exhibit superior biodegradability compared to synthetic polymers. This characteristic makes them suitable candidates for applications in wastewater purification processes.

8. Limitations, Challenges, and Opportunities

The use of commercially applied polymers has shown satisfactory performance in wastewater treatment. These polymers are derived from inexpensive monomers, such as acrylamide, acrylic acid, and methyl methacrylate. However, a major limitation of these polymers is their non-biodegradability or limited biodegradability, making their reuse difficult and posing environmental concerns. As a solution, bio-based polymers have emerged as a potential alternative to reduce environmental toxicity. However, using biopolymers alone may not meet the specific requirements for commercial applications, necessitating chemical modifications to enhance their properties.

Polysaccharide-based polymers, including starch, hydroxyethyl starch, dextrin, cellulose, chitosan, and guar gum, have garnered significant attention due to their natural abundance and low manufacturing cost. Chitosan, in particular, has been extensively studied for polymer, composite, and nanocomposite production, due to its eco-friendliness and the presence of amino groups ($–NH_2$). However, challenges related to chitosan's solubility and cost implications in adjusting reaction conditions and polymer solubility hinder its direct implementation. Consequently, chemical modifications are required to improve chitosan's water solubility and charge density while maintaining its efficacy and cost-effectiveness in water treatment.

While many researchers have reported their approaches as economical and environmentally beneficial, few studies account for the operational costs and biodegradability. Additionally, most investigations focus more on removing organic color molecules rather than metal ions or their mixtures, often employing synthetic solutions instead of real industrial effluents. Future research in this field should emphasize the development of environmentally friendly polymeric materials that are user-friendly, multifunctional, resistant to interference, efficient, cost-effective, and accompanied by comprehensive research reports to promote their adoption in the industry.

To overcome these limitations and challenges, it is essential for researchers in the field to collaborate with the industry to develop bio-based polymers for practical applications. This collaboration should aim to address the need for low-cost regeneration methods, consider the regeneration cost of adsorbents and flocculants, and provide a holistic understanding of the material's performance and potential in real-world scenarios. By focusing on these opportunities, the development of bio-based materials can contribute to more sustainable and effective wastewater treatment processes.

9. Conclusions

In conclusion, the development of bio-based polymeric materials for wastewater treatment presents an opportunity to address the limitations and challenges associated with commercially applied polymers. While the currently used polymers demonstrate satisfactory performance, their non-biodegradability and limited reusability raise environmental concerns. Biopolymers, such as polysaccharide-based materials, offer a promising alternative due to their natural availability and low manufacturing costs. However, chemical modifications are often necessary to enhance their properties for commercial applications.

Chitosan, a widely studied biopolymer, exemplifies the need for solubility improvements and charge density enhancements. By overcoming these challenges, biopolymers can offer cost-effective and environmentally friendly solutions for water treatment. Collaborations between researchers and industry are crucial to developing practical applications and ensuring that the materials meet the requirements of real-world scenarios.

Future research should focus on developing bio-based polymeric materials that are user-friendly, multifunctional, and resistant to interference. Emphasis should also be placed

on comprehensive research reports that consider the operational costs, biodegradability, and regeneration methods of adsorbents and flocculants. By addressing these factors, the industry can promote the adoption of bio-based polymers for efficient and sustainable wastewater treatment.

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Abbreviations

AA	Acrylic acid		
AM	Acrylamide	NTU	Nephelometric turbidity unit
AML	Amylose	OD	Optical density
AN	Acrylonitrile	PAA	Poly(acrylic acid)
AP	Amylopectin	PAM	Polyacrylamide
ATMAC	(3-acrylamidopropyl) trimethylammonium chloride	PDMA	Poly(N,N-dimethyl acrylamide)
CR	Congo red	PEG	Polyethylene glycol
DADMAC	Dimethyl diallyl ammonium chloride	PMETAC	Poly (2-methacryloyloxy ethyl trimethyl ammonium chloride)
Dextrin	Dt	ppb	Parts per billion
DMA	N,N-dimethyl acrylamide	ppm	Parts per million
DNS	2,4-dinitrosalicylic acid	PPS	Potassium peroxydisulfate
-g-	Grafting	PVA	Polyvinyl alcohol
HES	Hydroxyethyl starch	St	Starch
MB	Methylene Blue	UV	Ultraviolet
MF	Microfiltration	UV–VIS	Ultraviolet–Visible
MG	Malachite Green		
MMA	Methyl methacrylate		

References

1. Hussain, D.; Khan, S.A.; Alam Khan, T.; Alharthi, S.S. Efficient liquid phase confiscation of nile blue using a novel hybrid nanocomposite synthesized from guar gum-polyacrylamide and erbium oxide. *Sci. Rep.* **2022**, *12*, 14656. [CrossRef] [PubMed]
2. Khan, I.S.; Ali, N.; Hamid, R.; Ganie, S.A. Genotoxic effect of two commonly used food dyes metanil yellow and carmoisine using *Allium cepa* L. as indicator. *Toxicol. Rep.* **2020**, *7*, 370–375. [CrossRef] [PubMed]
3. Dutta, S.; Gupta, B.; Srivastava, S.K.; Gupta, A.K. Recent advances on the removal of dyes from wastewater using various adsorbents: A critical review. *Mater. Adv.* **2021**, *2*, 4497–4531. [CrossRef]
4. Kaur, J.; Bhat, S.A.; Singh, N.; Bhatti, S.S.; Kaur, V.; Katnoria, J.K. Assessment of the Heavy Metal Contamination of Roadside Soils Alongside Buddha Nullah, Ludhiana, (Punjab) India. *Int. J. Environ. Res. Public Health* **2022**, *19*, 1596. [CrossRef]
5. Pasdaran, A.; Azarpira, N.; Heidari, R.; Nourinejad, S.; Zare, M.; Hamedi, A. Effects of some cosmetic dyes and pigments on the proliferation of human foreskin fibroblasts and cellular oxidative stress; potential cytotoxicity of chlorophyllin and indigo carmine on fibroblasts. *J. Cosmet. Dermatol.* **2022**, *21*, 3979–3985. [CrossRef]
6. Emenike, P.C.; Neris, J.B.; Tenebe, I.T.; Nnaji, C.C.; Jarvis, P. Estimation of some trace metal pollutants in River Atuwara southwestern Nigeria and spatio-temporal human health risks assessment. *Chemosphere* **2020**, *239*, 124770. [CrossRef]
7. Cao, X.; Lu, Y.; Wang, C.; Zhang, M.; Yuan, J.; Zhang, A.; Song, S.; Baninla, Y.; Khan, K.; Wang, Y. Hydrogeochemistry and quality of surface water and groundwater in the drinking water source area of an urbanizing region. *Ecotoxicol. Environ. Saf.* **2019**, *186*, 109628. [CrossRef]
8. Hossain, S.; Anik, A.H.; Khan, R.; Ahmed, F.T.; Siddique, M.A.B.; Khan, A.H.A.N.; Saha, N.; Idris, A.M.; Alam, M. Public Health Vulnerability Due to the Exposure of Dissolved Metal(oid)s in Tap Water from a Mega City (Dhaka, Bangladesh): Source and Quality Appraisals. *Expo. Heal.* **2022**, *14*, 713–732. [CrossRef]

9. Muhammad, S.; Usman, Q.A. Heavy metal contamination in water of Indus River and its tributaries, Northern Pakistan: Evaluation for potential risk and source apportionment. *Toxin Rev.* **2022**, *41*, 380–388. [CrossRef]
10. Rizk, R.; Juzsakova, T.; Ben Ali, M.; Rawash, M.A.; Domokos, E.; Hedfi, A.; Almalki, M.; Boufahja, F.; Shafik, H.M.; Rédey, Á. Comprehensive environmental assessment of heavy metal contamination of surface water, sediments and Nile Tilapia in Lake Nasser, Egypt. *J. King Saud Univ. Sci.* **2022**, *34*, 101748. [CrossRef]
11. Rahman, M.S.; Ahmed, Z.; Seefat, S.M.; Alam, R.; Islam, A.R.M.T.; Choudhury, T.R.; Begum, B.A.; Idris, A.M. Assessment of heavy metal contamination in sediment at the newly established tannery industrial Estate in Bangladesh: A case study. *Environ. Chem. Ecotoxicol.* **2022**, *4*, 1–12. [CrossRef]
12. Küçüksümbül, A.; Akar, A.T.; Tarcan, G. Source, degree and potential health risk of metal(loid)s contamination on the water and soil in the Söke Basin, Western Anatolia, Turkey. *Environ. Monit. Assess.* **2021**, *194*, 6. [CrossRef]
13. Kang, C.-W.; Kolya, H. Green Synthesis of Ag-Au Bimetallic Nanocomposites Using Waste Tea Leaves Extract for Degradation Congo Red and 4-Nitrophenol. *Sustainability* **2021**, *13*, 3318. [CrossRef]
14. Kolya, H.; Kang, C.-W. Biogenic Synthesis of Silver-Iron Oxide Nanoparticles Using Kulekhara Leaves Extract for Removing Crystal Violet and Malachite Green Dyes from Water. *Sustainability* **2022**, *14*, 15800. [CrossRef]
15. Kolya, H.; Maiti, P.; Pandey, A.; Tripathy, T. Green synthesis of silver nanoparticles with antimicrobial and azo dye (Congo red) degradation properties using Amaranthus gangeticus Linn leaf extract. *J. Anal. Sci. Technol.* **2015**, *6*, 33. [CrossRef]
16. Kundu, R.; Mahada, P.; Chhirang, B.; Das, B. Cellulose hydrogels: Green and sustainable soft biomaterials. *Curr. Res. Green Sustain. Chem.* **2022**, *5*, 100252. [CrossRef]
17. Lustenberger, S.; Castro-Muñoz, R. Advanced biomaterials and alternatives tailored as membranes for water treatment and the latest innovative European water remediation projects: A review. *Case Stud. Chem. Environ. Eng.* **2022**, *5*, 100205. [CrossRef]
18. Zaman, A.; Ali, M.S.; Orasugh, J.T.; Banerjee, P.; Chattopadhyay, D. *Biopolymer-Based Nanocomposites for Removal of Hazardous Dyes from Water Bodies BT—Innovations in Environmental Biotechnology*; Arora, S., Kumar, A., Ogita, S., Yau, Y.-Y., Eds.; Springer Nature: Singapore, 2022; pp. 759–783. ISBN 9789811644450.
19. Alias, N.H.; Abdullah, N.; Othman, N.H.; Marpani, F.; Zainol, M.M.; Shayuti, M.S.M. *Sustainability Challenges and Future Perspectives of Biopolymer BT—Biopolymers: Recent Updates, Challenges and Opportunities*; Nadda, A.K., Sharma, S., Bhat, R., Eds.; Springer International Publishing: Cham, Switzerland, 2022; pp. 373–389. ISBN 9783030983925.
20. Kour, G.; Majhi, P.K.; Bharti, A.; Kothari, R.; Jain, A.; Singh, A.; Tyagi, V.V.; Pathania, D. Biopolymer-Based Nanocomposites and Water Treatment: A Global Outlook. In *Biorenewable Nanocomposite Materials, Vol. 2: Desalination and Wastewater Remediation*; ACS Symposium Series; American Chemical Society: Washington, DC, USA, 2022; Volume 1411, pp. 2–25. ISBN 9780841297807.
21. Patel, M.; Patel, J. Improving Sustainable Environment of Biopolymers Using Nanotechnology. In *Sustainable Nanotechnology: Strategies, Products, and Applications*; Wiley: Hoboken, NJ, USA, 2022; pp. 71–87. [CrossRef]
22. Zhao, C.; Liu, G.; Tan, Q.; Gao, M.; Chen, G.; Huang, X.; Xu, X.; Li, L.; Wang, J.; Zhang, Y.; et al. Polysaccharide-based biopolymer hydrogels for heavy metal detection and adsorption. *J. Adv. Res.* **2023**, *44*, 53–70. [CrossRef]
23. Torres, F.G.; Troncoso, O.P.; Pisani, A.; Gatto, F.; Bardi, G. Natural Polysaccharide Nanomaterials: An Overview of Their Immunological Properties. *Int. J. Mol. Sci.* **2019**, *20*, 5092. [CrossRef]
24. Gough, C.R.; Callaway, K.; Spencer, E.; Leisy, K.; Jiang, G.; Yang, S.; Hu, X. Biopolymer-Based Filtration Materials. *ACS Omega* **2021**, *6*, 11804–11812. [CrossRef]
25. Mamba, F.B.; Mbuli, B.S.; Ramontja, J. Recent Advances in Biopolymeric Membranes towards the Removal of Emerging Organic Pollutants from Water. *Membranes* **2021**, *11*, 798. [CrossRef] [PubMed]
26. Picos-Corrales, L.A.; Morales-Burgos, A.M.; Ruelas-Leyva, J.P.; Crini, G.; García-Armenta, E.; Jimenez-Lam, S.A.; Ayón-Reyna, L.E.; Rocha-Alonzo, F.; Calderón-Zamora, L.; Osuna-Martínez, U.; et al. Chitosan as an Outstanding Polysaccharide Improving Health-Commodities of Humans and Environmental Protection. *Polymers* **2023**, *15*, 526. [CrossRef] [PubMed]
27. Langer, R. Polymer Systems for Controlled Release of Macromolecules, Immobilized Enzyme Medical Bioreactors, and Tissue Engineering. In *Advances in Chemical Engineering*; Elsevier: Amsterdam, The Netherlands, 1994; Volume 19, pp. 1–50. [CrossRef]
28. Kolya, H.; Tripathy, T. Preparation, investigation of metal ion removal and flocculation performances of grafted hydroxyethyl starch. *Int. J. Biol. Macromol.* **2013**, *62*, 557–564. [CrossRef] [PubMed]
29. Bhattacharyya, D.; Hestekin, J.A.; Brushaber, P.; Cullen, L.; Bachas, L.G.; Sikdar, S.K. Novel poly-glutamic acid functionalized microfiltration membranes for sorption of heavy metals at high capacity. *J. Membr. Sci.* **1998**, *141*, 121–135. [CrossRef]
30. Bajaj, I.; Singhal, R. Poly (glutamic acid)—An emerging biopolymer of commercial interest. *Bioresour. Technol.* **2011**, *102*, 5551–5561. [CrossRef]
31. Lee, C.S.; Chong, M.F.; Robinson, J.; Binner, E. Optimisation of extraction and sludge dewatering efficiencies of bio-flocculants extracted from *Abelmoschus esculentus* (okra). *J. Environ. Manag.* **2015**, *157*, 320–325. [CrossRef]
32. Vandermeulen, G.W.M.; Boarino, A.; Klok, H.-A. Biodegradation of water-soluble and water-dispersible polymers for agricultural, consumer, and industrial applications—Challenges and opportunities for sustainable materials solutions. *J. Polym. Sci.* **2022**, *60*, 1797–1813. [CrossRef]
33. Aravamudhan, A.; Ramos, D.M.; Nada, A.A.; Kumbar, S.G. Chapter 4—Natural Polymers: Polysaccharides and Their Derivatives for Biomedical Applications. In *Natural and Synthetic Biomedical Polymers*; Elsevier: Oxford, UK, 2014; pp. 67–89. ISBN 9780123969835.

34. Phalle, S.P.; Choudhari, P.B.; Choudhari, S.P.; Bhagwat, D.A.; Kadam, A.M.; Gaikwad, V.L. Microwave-assisted grafting of acrylamide on a natural xylan gum for controlled drug delivery. *Polym. Bull.* **2023**, 1–18. [CrossRef]
35. Kong, W.; Li, Q.; Liu, J.; Li, X.; Zhao, L.; Su, Y.; Yue, Q.; Gao, B. Adsorption behavior and mechanism of heavy metal ions by chicken feather protein-based semi-interpenetrating polymer networks super absorbent resin. *RSC Adv.* **2016**, *6*, 83234–83243. [CrossRef]
36. Rajabi, M.; Keihankhadiv, S.; Suhas; Tyagi, I.; Karri, R.R.; Chaudhary, M.; Mubarak, N.M.; Chaudhary, S.; Kumar, P.; Singh, P. Comparison and interpretation of isotherm models for the adsorption of dyes, proteins, antibiotics, pesticides and heavy metal ions on different nanomaterials and non-nano materials—A comprehensive review. *J. Nanostructure Chem.* **2023**, *13*, 43–65. [CrossRef]
37. Liu, J.; Su, D.; Yao, J.; Huang, Y.; Shao, Z.; Chen, X. Soy protein-based polyethylenimine hydrogel and its high selectivity for copper ion removal in wastewater treatment. *J. Mater. Chem. A* **2017**, *5*, 4163–4171. [CrossRef]
38. Ekrayem, N.A.; Alhwaige, A.A.; Elhrari, W.; Amer, M. Removal of lead (II) ions from water using chitosan/polyester crosslinked spheres derived from chitosan and glycerol-based polyester. *J. Environ. Chem. Eng.* **2021**, *9*, 106628. [CrossRef]
39. Lin, S.H.; Lin, C.M. Treatment of textile waste effluents by ozonation and chemical coagulation. *Water Res.* **1993**, *27*, 1743–1748. [CrossRef]
40. El-Gohary, F.; Tawfik, A. Decolorization and COD reduction of disperse and reactive dyes wastewater using chemical-coagulation followed by sequential batch reactor (SBR) process. *Desalination* **2009**, *249*, 1159–1164. [CrossRef]
41. Leentvaar, J.; Rebhun, M. Effect of magnesium and calcium precipitation on coagulation-flocculation with lime. *Water Res.* **1982**, *16*, 655–662. [CrossRef]
42. Semerjian, L.; Ayoub, G. High-pH–magnesium coagulation–flocculation in wastewater treatment. *Adv. Environ. Res.* **2003**, *7*, 389–403. [CrossRef]
43. Ruthven, D.M. *Principles of Adsorption and Adsorption Processes*; John Wiley & Sons: New York, NY, USA, 1984; ISBN 0471866067.
44. Cooney, D.O. *Adsorption Design for Wastewater Treatment*; CRC Press: Boca Raton, FL, USA, 1998; ISBN 1566703336.
45. Blöcher, C.; Dorda, J.; Mavrov, V.; Chmiel, H.; Lazaridis, N.K.; Matis, K.A. Hybrid flotation—Membrane filtration process for the removal of heavy metal ions from wastewater. *Water Res.* **2003**, *37*, 4018–4026. [CrossRef]
46. Kolya, H.; Singh, V.K.; Kang, C.-W. *Polymeric Membranes and Hybrid Techniques for Water Purification Applications BT—Polymer-Based Advanced Functional Materials for Energy and Environmental Applications*; Subramani, N.K., Nataraj, S.K., Patel, C., Shivanna, S., Eds.; Springer: Singapore, 2022; pp. 75–91. ISBN 9789811687556.
47. Mavros, P.; Daniilidou, A.C.; Lazaridis, N.K.; Stergiou, L. Colour removal from aqueous solutions. Part I. Flotation. *Environ. Technol.* **1994**, *15*, 601–616. [CrossRef]
48. Zouboulis, A.I.; Lazaridis, N.K.; Grohmann, A. Toxic metals removal from waste waters by upflow filtration with floating filter medium. I. The case of zinc. *Sep. Sci. Technol.* **2002**, *37*, 403–416. [CrossRef]
49. Al-Kdasi, A.; Idris, A.; Saed, K.; Guan, C.T. Treatment of textile wastewater by advanced oxidation processes– A review. *Glob. Nest J.* **2004**, *6*, 222–230. [CrossRef]
50. Oturan, M.A.; Aaron, J.-J. Advanced Oxidation Processes in Water/Wastewater Treatment: Principles and Applications. A Review. *Crit. Rev. Environ. Sci. Technol.* **2014**, *44*, 2577–2641. [CrossRef]
51. Mo, K.B. Membrane-based solvent extraction for selective removal and recovery of metals. *J. Membr. Sci.* **1984**, *21*, 5–19. [CrossRef]
52. Yang, C.; Qian, Y.; Zhang, L.; Feng, J. Solvent extraction process development and on-site trial-plant for phenol removal from industrial coal-gasification wastewater. *Chem. Eng. J.* **2006**, *117*, 179–185. [CrossRef]
53. Tran, D.; Weidhaas, J. Ion exchange for effective separation of 3-nitro-1,2,4-triazol-5-one (NTO) from wastewater. *J. Hazard. Mater.* **2022**, *436*, 129215. [CrossRef] [PubMed]
54. Edgar, M.; Boyer, T.H. Nitrate adsorption and desorption during biological ion exchange. *Sep. Purif. Technol.* **2022**, *285*, 120363. [CrossRef]
55. Yulistyorini, A.; Sunaryo, K.; Mujiyono; Camargo-Valero, M.A. *Hybrid Anaerobic Baffled Reactor and Upflow Anaerobic Filter for Domestic Wastewater Purification BT—Environmental Degradation: Challenges and Strategies for Mitigation*; Singh, V.P., Yadav, S., Yadav, K.K., Yadava, R.N., Eds.; Springer International Publishing: Cham, Switzerland, 2022; pp. 149–164. ISBN 9783030955427.
56. Kuroda, K.; Narihiro, T.; Shinshima, F.; Yoshida, M.; Yamaguchi, H.; Kurashita, H.; Nakahara, N.; Nobu, M.K.; Noguchi, T.Q.; Yamauchi, M.; et al. High-rate cotreatment of purified terephthalate and dimethyl terephthalate manufacturing wastewater by a mesophilic upflow anaerobic sludge blanket reactor and the microbial ecology relevant to aromatic compound degradation. *Water Res.* **2022**, *219*, 118581. [CrossRef] [PubMed]
57. Bora, A.; Mohanrasu, K.; Swetha, T.A.; Ananthi, V.; Kumar, P.; Govarthanan, M.; Arun, A. Chapter 21—Microbes incorporated nanomaterials for water purification. In *Handbook of Microbial Nanotechnology*; Hussain, C.M., Ed.; Academic Press: Cambridge, MA, USA, 2021; pp. 439–459. ISBN 9780128234266.
58. Zhao, Z.; Cheng, M.; Li, Y.; Song, X.; Wang, Y.; Zhang, Y. A Novel Constructed Wetland Combined with Microbial Desalination Cells and its Application. *Microb. Ecol.* **2021**, *83*, 340–352. [CrossRef]
59. Alfonso-Muniozguren, P.; Bohari, M.H.; Sicilia, A.; Avignone-Rossa, C.; Bussemaker, M.; Saroj, D.; Lee, J. Tertiary treatment of real abattoir wastewater using combined acoustic cavitation and ozonation. *Ultrason. Sonochemistry* **2020**, *64*, 104986. [CrossRef]
60. Gatidou, G.; Samanides, C.G.; Fountoulakis, M.S.; Vyrides, I. Microbial electrolysis cell coupled with anaerobic granular sludge: A novel technology for real bilge water treatment. *Chemosphere* **2022**, *296*, 133988. [CrossRef]

61. Heebner, A.; Abbassi, B. Electrolysis catalyzed ozonation for advanced wastewater treatment. *J. Water Process. Eng.* **2022**, *46*, 102638. [CrossRef]
62. Kamali, M.; Aminabhavi, T.M.; Tarelho, L.A.; Hellemans, R.; Cuypers, J.; Capela, I.; Costa, M.E.V.; Dewil, R.; Appels, L. Acclimatized activated sludge for enhanced phenolic wastewater treatment using pinewood biochar. *Chem. Eng. J.* **2022**, *427*, 131708. [CrossRef]
63. Lin, R.; Li, Y.; Yong, T.; Cao, W.; Wu, J.; Shen, Y. Synergistic effects of oxidation, coagulation and adsorption in the integrated fenton-based process for wastewater treatment: A review. *J. Environ. Manag.* **2022**, *306*, 114460. [CrossRef]
64. Sibiya, N.P.; Rathilal, S.; Tetteh, E.K. Coagulation Treatment of Wastewater: Kinetics and Natural Coagulant Evaluation. *Molecules* **2021**, *26*, 698. [CrossRef]
65. Lee, H.-K.; Chang, S.; Park, W.; Kim, T.-J.; Park, S.; Jeon, H. Effective treatment of uranium-contaminated soil-washing effluent using precipitation/flocculation process for water reuse and solid waste disposal. *J. Water Process. Eng.* **2022**, *48*, 102890. [CrossRef]
66. Oliveira, C.; Rubio, J. A short overview of the formation of aerated flocs and their applications in solid/liquid separation by flotation. *Miner. Eng.* **2012**, *39*, 124–132. [CrossRef]
67. Zhao, J.; Li, B.; Wang, A.; Ge, W.; Li, W. Floc formation and growth mechanism during magnesium hydroxide and polyacrylamide coagulation process for reactive orange removal. *Environ. Technol.* **2022**, *43*, 424–430. [CrossRef]
68. Wu, Y.; Tran, Q.C.; Zhang, X.; Lu, Y.; Xu, J.; Zhang, H. Experimental investigation on the treatment of different typical marine dredged sludges by flocculant and vacuum preloading. *Arab. J. Geosci.* **2022**, *15*, 642. [CrossRef]
69. Na, S.-H.; Kim, M.-J.; Kim, J.-T.; Jeong, S.; Lee, S.; Chung, J.; Kim, E.-J. Microplastic removal in conventional drinking water treatment processes: Performance, mechanism, and potential risk. *Water Res.* **2021**, *202*, 117417. [CrossRef]
70. Park, S.-J.; Seo, M.-K. Chapter 1—Intermolecular Force. In *Interface Science and Composites*; Park, S.-J., Seo, M.-K., Eds.; Elsevier: Amsterdam, The Netherlands, 2011; Volume 18, pp. 1–57. ISBN 1573-4285.
71. Cheng, Y.; Shi, J.; Zhang, Q.; Fang, C.; Chen, J.; Li, F. Recent Progresses in Adsorption Mechanism, Architectures, Electrode Materials and Applications for Advanced Electrosorption System: A Review. *Polymers* **2022**, *14*, 2985. [CrossRef]
72. Bedzyk, M.J.; Bommarito, G.M.; Caffrey, M.; Penner, T.L. Diffuse-Double Layer at a Membrane-Aqueous Interface Measured with X-Ray Standing Waves. *Science* **1990**, *248*, 52–56. [CrossRef]
73. Grahame, D.C. Diffuse Double Layer Theory for Electrolytes of Unsymmetrical Valence Types. *J. Chem. Phys.* **1953**, *21*, 1054–1060. [CrossRef]
74. Ninham, B.W. On progress in forces since the DLVO theory. *Adv. Colloid Interface Sci.* **1999**, *83*, 1–17. [CrossRef]
75. Amuda, O.S.; Alade, A. Coagulation/flocculation process in the treatment of abattoir wastewater. *Desalination* **2006**, *196*, 22–31. [CrossRef]
76. Amuda, O.S.; Amoo, I.A. Coagulation/flocculation process and sludge conditioning in beverage industrial wastewater treatment. *J. Hazard. Mater.* **2007**, *141*, 778–783. [CrossRef]
77. Ahmad, A.L.; Wong, S.S.; Teng, T.T.; Zuhairi, A. Improvement of alum and PACl coagulation by polyacrylamides (PAMs) for the treatment of pulp and paper mill wastewater. *Chem. Eng. J.* **2008**, *137*, 510–517. [CrossRef]
78. Chong, M.F.; Lee, K.P.; Chieng, H.J.; Ramli, I.I.S.B. Removal of boron from ceramic industry wastewater by adsorption–flocculation mechanism using palm oil mill boiler (POMB) bottom ash and polymer. *Water Res.* **2009**, *43*, 3326–3334. [CrossRef]
79. Martin, M.A.; González, I.; Berrios, M.; Siles, J.A.; Martin, A. Optimization of coagulation–flocculation process for wastewater derived from sauce manufacturing using factorial design of experiments. *Chem. Eng. J.* **2011**, *172*, 771–782. [CrossRef]
80. Sher, F.; Malik, A.; Liu, H. Industrial polymer effluent treatment by chemical coagulation and flocculation. *J. Environ. Chem. Eng.* **2013**, *1*, 684–689. [CrossRef]
81. Irfan, M.; Butt, T.; Imtiaz, N.; Abbas, N.; Khan, R.A.; Shafique, A. The removal of COD, TSS and colour of black liquor by coagulation–flocculation process at optimized pH, settling and dosing rate. *Arab. J. Chem.* **2017**, *10*, S2307–S2318. [CrossRef]
82. Sarika, R.; Kalogerakis, N.; Mantzavinos, D. Treatment of olive mill effluents: Part II. Complete removal of solids by direct flocculation with poly-electrolytes. *Environ. Int.* **2005**, *31*, 297–304. [CrossRef]
83. Ebeling, J.M.; Rishel, K.L.; Sibrell, P.L. Screening and evaluation of polymers as flocculation aids for the treatment of aquacultural effluents. *Aquac. Eng.* **2005**, *33*, 235–249. [CrossRef]
84. Wong, S.S.; Teng, T.T.; Ahmad, A.L.; Zuhairi, A.; Najafpour, G. Treatment of pulp and paper mill wastewater by polyacrylamide (PAM) in polymer induced flocculation. *J. Hazard. Mater.* **2006**, *135*, 378–388. [CrossRef]
85. Kang, Q.; Gao, B.; Yue, Q.; Zhou, W.; Shen, D. Residual color profiles of reactive dyes mixture during a chemical flocculation process. *Colloids Surfaces A: Physicochem. Eng. Asp.* **2007**, *299*, 45–53. [CrossRef]
86. Yue, Q.Y.; Gao, B.Y.; Wang, Y.; Zhang, H.; Sun, X.; Wang, S.G.; Gu, R.R. Synthesis of polyamine flocculants and their potential use in treating dye wastewater. *J. Hazard. Mater.* **2008**, *152*, 221–227. [CrossRef]
87. Razali, M.A.A.; Ahmad, Z.; Ahmad, M.S.B.; Ariffin, A. Treatment of pulp and paper mill wastewater with various molecular weight of polyDADMAC induced flocculation. *Chem. Eng. J.* **2011**, *166*, 529–535. [CrossRef]
88. Rubio, J. The flocculation properties of poly (ethylene oxide). *Colloids Surfaces* **1981**, *3*, 79–95. [CrossRef]
89. Liu, Y.; Zheng, H.; Sun, Y.; Ren, J.; Zheng, X.; Sun, Q.; Jiang, S.; Ding, W. Synthesis of novel chitosan-based flocculants with amphiphilic structure and its application in sludge dewatering: Role of hydrophobic groups. *J. Clean. Prod.* **2020**, *249*, 119350. [CrossRef]

90. Kolya, H.; Tripathy, T. Grafted polysaccharides based on acrylamide and N,N-dimethylacrylamide: Preparation and investi-gation of their flocculation performances. *J. Appl. Polym. Sci.* **2013**, *127*, 2786–2795. [CrossRef]
91. Zare, E.N.; Lakouraj, M.M.; Kasirian, N. Development of effective nano-biosorbent based on poly m-phenylenediamine grafted dextrin for removal of Pb (II) and methylene blue from water. *Carbohydr. Polym.* **2018**, *201*, 539–548. [CrossRef]
92. Saya, L.; Malik, V.; Singh, A.; Singh, S.; Gambhir, G.; Singh, W.R.; Chandra, R.; Hooda, S. Guar gum based nanocomposites: Role in water purification through efficient removal of dyes and metal ions. *Carbohydr. Polym.* **2021**, *261*, 117851. [CrossRef]
93. Maji, B.; Maiti, S. Chemical modification of xanthan gum through graft copolymerization: Tailored properties and potential applications in drug delivery and wastewater treatment. *Carbohydr. Polym.* **2021**, *251*, 117095. [CrossRef]
94. Pandey, S.; Do, J.Y.; Kim, J.; Kang, M. Fast and highly efficient removal of dye from aqueous solution using natural locust bean gum based hydrogels as adsorbent. *Int. J. Biol. Macromol.* **2020**, *143*, 60–75. [CrossRef] [PubMed]
95. Kou, S.G.; Peters, L.M.; Mucalo, M.R. Chitosan: A review of sources and preparation methods. *Int. J. Biol. Macromol.* **2021**, *169*, 85–94. [CrossRef] [PubMed]
96. Akhlaq, M.; Maqsood, H.; Uroos, M.; Iqbal, A. A Comparative Study of Different Methods for Cellulose Extraction from Lignocellulosic Wastes and Conversion into Carboxymethyl Cellulose. *Chemistryselect* **2022**, *7*, e202201533. [CrossRef]
97. Du, Q.; Wang, Y.; Li, A.; Yang, H. Scale-inhibition and flocculation dual-functionality of poly(acrylic acid) grafted starch. *J. Environ. Manag.* **2018**, *210*, 273–279. [CrossRef]
98. Yang, R.; Li, H.; Huang, M.; Yang, H.; Li, A. A review on chitosan-based flocculants and their applications in water treatment. *Water Res.* **2016**, *95*, 59–89. [CrossRef]
99. Jiang, X.; Li, Y.; Tang, X.; Jiang, J.; He, Q.; Xiong, Z.; Zheng, H. Biopolymer-based flocculants: A review of recent technologies. *Environ. Sci. Pollut. Res. Int.* **2021**, *28*, 46934–46963. [CrossRef]
100. Kolya, H.; Sasmal, D.; Tripathy, T. Novel Biodegradable Flocculating Agents Based on Grafted Starch Family for the Industrial Effluent Treatment. *J. Polym. Environ.* **2017**, *25*, 408–418. [CrossRef]
101. Sasmal, D.; Singh, R.P.; Tripathy, T. Synthesis and flocculation characteristics of a novel biodegradable flocculating agent amylopectin-g-poly(acrylamide-co-N-methylacrylamide). *Colloids Surfaces A Physicochem. Eng. Asp.* **2015**, *482*, 575–584. [CrossRef]
102. Sarkar, A.K.; Mandre, N.R.; Panda, A.B.; Pal, S. Amylopectin grafted with poly (acrylic acid): Development and application of a high performance flocculant. *Carbohydr. Polym.* **2013**, *95*, 753–759. [CrossRef]
103. Sarkar, A.K.; Ghorai, S.; Patra, A.S.; Mishra, B.K.; Mandre, N.R.; Pal, S. Modified amylopectin based flocculant for the treatment of synthetic effluent and industrial wastewaters. *Int. J. Biol. Macromol.* **2015**, *72*, 356–363. [CrossRef]
104. Kumar, K.; Adhikary, P.; Karmakar, N.C.; Gupta, S.; Singh, R.P.; Krishnamoorthi, S. Synthesis, characterization and application of novel cationic and amphoteric flocculants based on amylopectin. *Carbohydr. Polym.* **2015**, *127*, 275–281. [CrossRef]
105. Pal, S.; Nasim, T.; Patra, A.; Ghosh, S.; Panda, A.B. Microwave assisted synthesis of polyacrylamide grafted dextrin (Dxt-g-PAM): Development and application of a novel polymeric flocculant. *Int. J. Biol. Macromol.* **2010**, *47*, 623–631. [CrossRef]
106. Kolya, H.; Tripathy, T. Polyacrylamide and Poly N, N-Dimethylacrylamide Grafted Hydroxyethyl Starch: Efficient Biode-gradable Flocculants. *J. Colloid Sci. Biotechnol.* **2014**, *3*, 150–159. [CrossRef]
107. Das, S.; Patra, P.; Singha, K.; Biswas, P.; Sarkar, S.; Pal, S. Graft copolymeric flocculant using functionalized starch towards treatment of blast furnace effluent. *Int. J. Biol. Macromol.* **2019**, *125*, 35–40. [CrossRef]
108. Wang, J.-P.; Yuan, S.-J.; Wang, Y.; Yu, H.-Q. Synthesis, characterization and application of a novel starch-based flocculant with high flocculation and dewatering properties. *Water Res.* **2013**, *47*, 2643–2648. [CrossRef]
109. Huang, M.; Liu, Z.; Li, A.; Yang, H. Dual functionality of a graft starch flocculant: Flocculation and antibacterial performance. *J. Environ. Manag.* **2017**, *196*, 63–71. [CrossRef]
110. Chang, Q.; Hao, X.; Duan, L. Synthesis of crosslinked starch-graft-polyacrylamide-co-sodium xanthate and its performances in wastewater treatment. *J. Hazard. Mater.* **2008**, *159*, 548–553. [CrossRef]
111. Yang, Z.; Wu, H.; Yuan, B.; Huang, M.; Yang, H.; Li, A.; Bai, J.; Cheng, R. Synthesis of amphoteric starch-based grafting flocculants for flocculation of both positively and negatively charged colloidal contaminants from water. *Chem. Eng. J.* **2014**, *244*, 209–217. [CrossRef]
112. Dey, K.P.; Mishra, S.; Sen, G. Synthesis and characterization of polymethylmethacrylate grafted barley for treatment of industrial and municipal wastewater. *J. Water Process. Eng.* **2017**, *18*, 113–125. [CrossRef]
113. Mehta, A.; Aryan, A.; Pandey, J.P.; Sen, G. Synthesis of a Novel Water-Soluble Graft Copolymer for Mineral Ore Beneficiation and for River Water Treatment towards Drinking Water Augmentation. *Chemistryselect* **2022**, *7*, e202103289. [CrossRef]
114. Kumari, N.; Mishra, S. Synthesis, characterization and flocculation efficiency of grafted Moringa gum based derivatives. *Carbohydr. Polym.* **2022**, *281*, 119079. [CrossRef] [PubMed]
115. Suresha, P.; Badiger, M.V. Cationic chitosan graft flocculants: Synthesis, characterization and applications in kaolin separation. *Mater. Today Proc.* **2022**, in press. [CrossRef]
116. Hasan, A.; Fatehi, P. Flocculation of kaolin particles with cationic lignin polymers. *Sci. Rep.* **2019**, *9*, 2672. [CrossRef] [PubMed]
117. Belaid, A.; Bouras, B.; Hocine, T.; Tennouga, L. Flocculation of Clay Suspensions Using Copolymers Based on Acrylamide and Biopolymer. *Phys. Chem. Res.* **2023**, *11*, 221–230.
118. Li, H.; Cai, T.; Yuan, B.; Li, R.; Yang, H.; Li, A. Flocculation of Both Kaolin and Hematite Suspensions Using the Starch-Based Flocculants and Their Floc Properties. *Ind. Eng. Chem. Res.* **2015**, *54*, 59–67. [CrossRef]
119. Jyothi, A.N. Starch Graft Copolymers: Novel Applications in Industry. *Compos. Interfaces* **2010**, *17*, 165–174. [CrossRef]

120. Kolya, H.; Tripathy, T. Hydroxyethyl Starch-g-Poly-(N,N-dimethylacrylamide-co-acrylic acid): An efficient dye removing agent. *Eur. Polym. J.* **2013**, *49*, 4265–4275. [CrossRef]
121. Guan, G.; Gao, T.; Lou, T.; Wang, X. Lignocellulose–acrylamide–carboxymethyl cellulose copolymer as a cost-effective anionic flocculant. *Iran. Polym. J.* **2022**, *31*, 587–594. [CrossRef]
122. Zhou, P.; Ru, X.; Yang, W.; Dai, Z.; Ofori, M.A.; Chen, J.; Hou, J.; Zhong, Z.; Jin, H. Study on preparation of cationic flocculants by grafting binary monomer on cellulose substrate by γ-ray co-irradiation. *J. Environ. Chem. Eng.* **2022**, *10*, 107138. [CrossRef]
123. Han, Z.; Huo, J.; Zhang, X.; Ngo, H.H.; Guo, W.; Du, Q.; Zhang, Y.; Li, C.; Zhang, D. Characterization and flocculation performance of a newly green flocculant derived from natural bagasse cellulose. *Chemosphere* **2022**, *301*, 134615. [CrossRef]
124. Hu, P.; Shen, S.; Yang, H. Evaluation of hydrophobically associating cationic starch-based flocculants in sludge dewatering. *Sci. Rep.* **2021**, *11*, 11819. [CrossRef]
125. Tripathy, T.; De, B.R. Flocculation: A new way to treat the waste water. *J. Phys. Sci.* **2006**, *10*, 93–127.
126. Lü, T.; Luo, C.; Qi, D.; Zhang, D.; Zhao, H. Efficient treatment of emulsified oily wastewater by using amphipathic chitosan-based flocculant. *React. Funct. Polym.* **2019**, *139*, 133–141. [CrossRef]
127. Mühle, K. Floc Stability in Laminar and Turbulent Flow. In *Surfactant Science Series*; Marcel Dekker AG: New York, NY, USA, 1993; p. 355.
128. Nie, Y.; Wang, Z.; Wang, W.; Zhou, Z.; Kong, Y.; Ma, J. Bio-flocculation of Microcystis aeruginosa by using fungal pellets of *Aspergillus oryzae*: Performance and mechanism. *J. Hazard. Mater.* **2022**, *439*, 129606. [CrossRef]
129. Ma, K.; Zhao, L.; Jiang, Z.; Huang, Y.; Sun, X. Adsorption characteristics of Cr (III) onto starch-graft-poly(acrylic acid)/organo-modifed zeolite 4A composite: A novel path to the adsorption mechanisms. *Polym. Compos.* **2018**, *39*, 1223–1233. [CrossRef]
130. Korhonen, M.H.J.; Rojas, O.J.; Laine, J. Effect of charge balance and dosage of polyelectrolyte complexes on the shear resistance of mineral floc strength and reversibility. *J. Colloid Interface Sci.* **2015**, *448*, 73–78. [CrossRef]
131. Bratby, J. *Coagulation and Flocculation in Water and Wastewater Treatment*; IWA Publishing: London, UK, 2006; ISBN 1843391066.
132. Singh, R.P.; Tripathy, T.; Karmakar, G.P.; Rath, S.K.; Karmakar, N.C.; Pandey, S.R.; Kannan, K.; Jain, S.K.; Lan, N.T. Novel biodegradable flocculants based on polysaccharides. *Curr. Sci.* **2000**, *78*, 798–803.
133. Pal, S.; Das, R.; Ghorai, S. Hydroxypropyl Methyl Cellulose Grafted with Poly (Acrylamide): Application as Novel Polymeric Material as Flocculant as well as Adsorbent. In *Cellulose-Based Graft Copolymers*; CRC Press: Boca Raton, FL, USA, 2015; pp. 320–353.
134. Maćczak, P.; Kaczmarek, H.; Ziegler-Borowska, M. Recent Achievements in Polymer Bio-Based Flocculants for Water Treat-ment. *Materials* **2020**, *13*, 3951. [CrossRef]
135. Lapointe, M.; Jahandideh, H.; Farner, J.M.; Tufenkji, N. Super-bridging fibrous materials for water treatment. *NPJ Clean Water* **2022**, *5*, 11. [CrossRef]
136. Teh, C.Y.; Budiman, P.M.; Shak, K.P.Y.; Wu, T.Y. Recent Advancement of Coagulation–Flocculation and Its Application in Wastewater Treatment. *Ind. Eng. Chem. Res.* **2016**, *55*, 4363–4389. [CrossRef]
137. Kurusu, R.S.; Lapointe, M.; Tufenkji, N. Sustainable iron-grafted cellulose fibers enable coagulant recycling and improve contaminant removal in water treatment. *Chem. Eng. J.* **2022**, *430*, 132927. [CrossRef]
138. Ngah, W.W.; Hanafiah, M.A.K.M. Removal of heavy metal ions from wastewater by chemically modified plant wastes as adsorbents: A review. *Bioresour. Technol.* **2008**, *99*, 3935–3948. [CrossRef] [PubMed]
139. Kolya, H.; Tripathy, T. Metal complexation studies of amylopectin-*graft* -poly[(*N*,*N* -dimethylacrylamide)-*co* -(acrylic acid)]: A biodegradable synthetic graft copolymer. *Polym. Int.* **2015**, *64*, 1336–1351. [CrossRef]
140. Jana, S.; Ray, J.; Mondal, B.; Pradhan, S.S.; Tripathy, T. pH responsive adsorption/desorption studies of organic dyes from their aqueous solutions by katira gum-cl-poly(acrylic acid-co-N-vinyl imidazole) hydrogel. *Colloids Surfaces A: Physicochem. Eng. Asp.* **2018**, *553*, 472–486. [CrossRef]
141. Sasmal, D.; Maity, J.; Kolya, H.; Tripathy, T. Study of congo red dye removal from its aqueous solution using sulfated acrylamide and N, N- dimethyl acrylamide grafted amylopectin. *J. Water Process. Eng.* **2017**, *18*, 7–19. [CrossRef]
142. Tan, K.B.; Vakili, M.; Horri, B.A.; Poh, P.E.; Abdullah, A.Z.; Salamatinia, B. Adsorption of dyes by nanomaterials: Recent developments and adsorption mechanisms. *Sep. Purif. Technol.* **2015**, *150*, 229–242. [CrossRef]
143. Lu, L.; Pan, Z.; Hao, N.; Peng, W. A novel acrylamide-free flocculant and its application for sludge dewatering. *Water Res.* **2014**, *57*, 304–312. [CrossRef]
144. Mahmoodi, N.M.; Sadeghi, U.; Maleki, A.; Hayati, B.; Najafi, F. Synthesis of cationic polymeric adsorbent and dye removal isotherm, kinetic and thermodynamic. *J. Ind. Eng. Chem.* **2014**, *20*, 2745–2753. [CrossRef]
145. Yue, Q.-Y.; Li, Q.; Gao, B.-Y.; Yuan, A.-J.; Wang, Y. Formation and characteristics of cationic-polymer/bentonite complexes as adsorbents for dyes. *Appl. Clay Sci.* **2007**, *35*, 268–275. [CrossRef]
146. Dhodapkar, R.; Rao, N.N.; Pande, S.P.; Nandy, T.; Devotta, S. Adsorption of cationic dyes on Jalshakti®, super absorbent polymer and photocatalytic regeneration of the adsorbent. *React. Funct. Polym.* **2007**, *67*, 540–548. [CrossRef]
147. Tekin, N.; Demirbaş, O.; Alkan, M. Adsorption of cationic polyacrylamide onto kaolinite. *Microporous Mesoporous Mater.* **2005**, *85*, 340–350. [CrossRef]
148. Tanaka, H. Copolymerization of cationic monomers with acrylamide in an aqueous solution. *J. Polym. Sci. Part A Polym. Chem.* **1986**, *24*, 29–36. [CrossRef]

149. Zhou, S.; Xue, A.; Zhang, Y.; Li, M.; Li, K.; Zhao, Y.; Xing, W. Novel polyamidoamine dendrimer-functionalized palygorskite adsorbents with high adsorption capacity for Pb^{2+} and reactive dyes. *Appl. Clay Sci.* **2015**, *107*, 220–229. [CrossRef]
150. Kislenko, V.N.; Oliynyk, L.P. Complex formation of polyethyleneimine with copper(II), nickel(II), and cobalt(II) ions. *J. Polym. Sci. Part A: Polym. Chem.* **2002**, *40*, 914–922. [CrossRef]
151. Li, X.; Wang, Z.; Ning, J.; Gao, M.; Jiang, W.; Zhou, Z.; Li, G. Preparation and characterization of a novel polyethyleneimine cation-modified persimmon tannin bioadsorbent for anionic dye adsorption. *J. Environ. Manag.* **2018**, *217*, 305–314. [CrossRef]
152. Briones, O.X.; Tapia, R.A.; Campodónico, P.R.; Urzúa, M.; Leiva, A.; Contreras, R.; González-Navarrete, J. Synthesis and characterization of poly (ionic liquid) derivatives of N-alkyl quaternized poly(4-vinylpyridine). *React. Funct. Polym.* **2018**, *124*, 64–71. [CrossRef]
153. Lv, Q.; Hu, X.; Zhang, X.; Huang, L.; Liu, Z.; Sun, G. Highly efficient removal of trace metal ions by using poly(acrylic acid) hydrogel adsorbent. *Mater. Des.* **2019**, *181*, 107934. [CrossRef]
154. Rahchamani, J.; Mousavi, H.Z.; Behzad, M. Adsorption of methyl violet from aqueous solution by polyacrylamide as an adsorbent: Isotherm and kinetic studies. *Desalination* **2011**, *267*, 256–260. [CrossRef]
155. Fouda-Mbanga, B.; Prabakaran, E.; Pillay, K. Carbohydrate biopolymers, lignin based adsorbents for removal of heavy metals (Cd^{2+}, Pb^{2+}, Zn^{2+}) from wastewater, regeneration and reuse for spent adsorbents including latent fingerprint detection: A review. *Biotechnol. Rep.* **2021**, *30*, e00609. [CrossRef]
156. Sayyed, A.J.; Pinjari, D.V.; Sonawane, S.H.; Bhanvase, B.A.; Sheikh, J.; Sillanpää, M. Cellulose-based nanomaterials for water and wastewater treatments: A review. *J. Environ. Chem. Eng.* **2021**, *9*, 106626. [CrossRef]
157. Musarurwa, H.; Tavengwa, N.T. Advances in the application of chitosan-based metal organic frameworks as adsorbents for environmental remediation. *Carbohydr. Polym.* **2022**, *283*, 119153. [CrossRef] [PubMed]
158. Mallik, A.K.; Kabir, S.F.; Rahman, F.B.A.; Sakib, M.N.; Efty, S.S.; Rahman, M.M. Cu(II) removal from wastewater using chitosan-based adsorbents: A review. *J. Environ. Chem. Eng.* **2022**, *10*, 108048. [CrossRef]
159. Wang, J.; Zhuang, S. Chitosan-based materials: Preparation, modification and application. *J. Clean. Prod.* **2022**, *355*, 131825. [CrossRef]
160. Perumal, S.; Atchudan, R.; Yoon, D.H.; Joo, J.; Cheong, I.W. Graphene oxide-embedded chitosan/gelatin hydrogel particles for the adsorptions of multiple heavy metal ions. *J. Mater. Sci.* **2020**, *55*, 9354–9363. [CrossRef]
161. Sheoran, K.; Kaur, H.; Siwal, S.S.; Saini, A.K.; Vo, D.-V.N.; Thakur, V.K. Recent advances of carbon-based nanomaterials (CBNMs) for wastewater treatment: Synthesis and application. *Chemosphere* **2022**, *299*, 134364. [CrossRef]
162. Hassan, M.; Liu, Y.; Naidu, R.; Du, J.; Qi, F.; Donne, S.W.; Islam, M.M. Mesoporous Biopolymer Architecture Enhanced the Adsorption and Selectivity of Aqueous Heavy-Metal Ions. *ACS Omega* **2021**, *6*, 15316–15331. [CrossRef]
163. Liu, S.; Wei, W.; Wu, S.; Zhang, F. Preparation of hierarchical porous activated carbons from different industrial lignin for highly efficient adsorption performance. *J. Porous Mater.* **2020**, *27*, 1523–1533. [CrossRef]
164. Tan, Y.; Wang, X.; Xiong, F.; Ding, J.; Qing, Y.; Wu, Y. Preparation of lignin-based porous carbon as an efficient absorbent for the removal of methylene blue. *Ind. Crop. Prod.* **2021**, *171*, 113980. [CrossRef]
165. Zhu, S.; Xu, J.; Kuang, Y.; Cheng, Z.; Wu, Q.; Xie, J.; Wang, B.; Gao, W.; Zeng, J.; Li, J.; et al. Lignin-derived sulfonated porous carbon from cornstalk for efficient and selective removal of cationic dyes. *Ind. Crop. Prod.* **2021**, *159*, 113071. [CrossRef]
166. Sun, Y.; Wang, T.; Sun, X.; Bai, L.; Han, C.; Zhang, P. The potential of biochar and lignin-based adsorbents for wastewater treatment: Comparison, mechanism, and application—A review. *Ind. Crop. Prod.* **2021**, *166*, 113473. [CrossRef]
167. Jiang, M.; Chen, L.; Niu, N. Enhanced adsorption for malachite green by functionalized lignin magnetic composites: Optimization, performance and adsorption mechanism. *J. Mol. Struct.* **2022**, *1260*, 132842. [CrossRef]
168. Zhu, S.; Xu, J.; Yu, W.; Kuang, Y.; Wang, B.; Ying, G.; Li, J.; Cheng, Z.; Li, J.; Chen, K. Simultaneous production of clean water and organic dye from dyeing wastewater by reusable lignin-derived porous carbon. *Ind. Crop. Prod.* **2022**, *187*, 115314. [CrossRef]
169. Sasmal, D.; Maity, J.; Kolya, H.; Tripathy, T. Selective adsorption of Pb (II) ions by amylopectin-g-poly (acrylamide-co-acrylic acid): A bio-degradable graft copolymer. *Int. J. Biol. Macromol.* **2017**, *97*, 585–597. [CrossRef]
170. Sasmal, D.; Kolya, H.; Tripathy, T. Amylopectin-g-poly(methylacrylate-co-sodium acrylate): An efficient Cd(II) binder. *Int. J. Biol. Macromol.* **2016**, *91*, 934–945. [CrossRef]
171. Chai, K.; Lu, K.; Xu, Z.; Tong, Z.; Ji, H. Rapid and selective recovery of acetophenone from petrochemical effluents by crosslinked starch polymer. *J. Hazard. Mater.* **2018**, *348*, 20–28. [CrossRef]
172. Ahmad, R.; Mirza, A. Synthesis of Guar gum/bentonite a novel bionanocomposite: Isotherms, kinetics and thermodynamic studies for the removal of Pb (II) and crystal violet dye. *J. Mol. Liq.* **2018**, *249*, 805–814. [CrossRef]
173. Zhou, L.S.; Zhou, H.; Yang, X. Preparation and performance of a novel starch-based inorganic/organic composite coagulant for textile wastewater treatment. *Sep. Purif. Technol.* **2019**, *210*, 93–99. [CrossRef]
174. Zhou, H.; Zhou, L.; Yang, X. Optimization of preparing a high yield and high cationic degree starch graft copolymer as environmentally friendly flocculant: Through response surface methodology. *Int. J. Biol. Macromol.* **2018**, *118*, 1431–1437. [CrossRef]
175. Bahrami, M.; Amiri, M.J.; Bagheri, F. Optimization of the lead removal from aqueous solution using two starch based adsorbents: Design of experiments using response surface methodology (RSM). *J. Environ. Chem. Eng.* **2018**, *7*, 102793. [CrossRef]
176. Abdel-Halim, E. Preparation of starch/poly(N,N-Diethylaminoethyl methacrylate) hydrogel and its use in dye removal from aqueous solutions. *React. Funct. Polym.* **2013**, *73*, 1531–1536. [CrossRef]

177. Wei, H.; Ren, J.; Li, A.; Yang, H. Sludge dewaterability of a starch-based flocculant and its combined usage with ferric chloride. *Chem. Eng. J.* **2018**, *349*, 737–747. [CrossRef]
178. Mahmoodi-Babolan, N.; Nematollahzadeh, A.; Heydari, A.; Merikhy, A. Bioinspired catecholamine/starch composites as superadsorbent for the environmental remediation. *Int. J. Biol. Macromol.* **2019**, *125*, 690–699. [CrossRef] [PubMed]
179. Jiang, G.; Peng, S.; Li, X.; Yang, L.; Soares, J.B.P.; Li, G. Preparation of amphoteric starch-based flocculants by reactive extrusion for removing useless solids from water-based drilling fluids. *Colloids Surfaces A Physicochem. Eng. Asp.* **2019**, *558*, 343–350. [CrossRef]
180. Ibrahim, B.; Fakhre, N. Crown ether modification of starch for adsorption of heavy metals from synthetic wastewater. *Int. J. Biol. Macromol.* **2019**, *123*, 70–80. [CrossRef]
181. Kolya, H.; Roy, A.; Tripathy, T. Starch-g-Poly-(N, N-dimethyl acrylamide-co-acrylic acid): An efficient Cr (VI) ion binder. *Int. J. Biol. Macromol.* **2015**, *72*, 560–568. [CrossRef]
182. Kolya, H.; Das, S.; Tripathy, T. Synthesis of Starch-g-Poly-(N-methylacrylamide-co-acrylic acid) and its application for the removal of mercury (II) from aqueous solution by adsorption. *Eur. Polym. J.* **2014**, *58*, 1–10. [CrossRef]
183. Mondal, S.K.; Wu, C.; Nwadire, F.C.; Rownaghi, A.; Kumar, A.; Adewuyi, Y.; Okoronkwo, M.U. Examining the Effect of a Chitosan Biopolymer on Alkali-Activated Inorganic Material for Aqueous Pb(II) and Zn(II) Sorption. *Langmuir* **2022**, *38*, 903–913. [CrossRef]
184. Zhang, L.-M.; Chen, D.-Q. An investigation of adsorption of lead(II) and copper(II) ions by water-insoluble starch graft copolymers. *Colloids Surfaces A Physicochem. Eng. Asp.* **2002**, *205*, 231–236. [CrossRef]
185. Xun, J.; Lou, T.; Xing, J.; Zhang, W.; Xu, Q.; Peng, J.; Wang, X. Synthesis of a starch-acrylic acid-chitosan copolymer as flocculant for dye removal. *J. Appl. Polym. Sci.* **2019**, *136*, 47437. [CrossRef]
186. Ying, C.; Gongwei, T.; Yuning, L.; Qi, Z.; Jing, L.; Ji, L. Ammonium persulfate-initiated polymerization of cationic starch-grafted-cationic polyacrylamide flocculant for the enhanced flocculation of oil sludge suspension. *J. Dispers. Sci. Technol.* **2018**, *40*, 1246–1255. [CrossRef]
187. Amini-Fazl, M.S.; Ahmari, A. Dextran-graft-poly(hydroxyethyl methacrylate) biosorbents for removal of dyes and metal cations. *Mater. Res. Express* **2019**, *6*, 055324. [CrossRef]
188. Wang, X.-M.; Zhang, C.-N.; Dai, W.-Y.; Xu, Q. Study on the adsorption of crystal violet by starch-g-poly (acrylic acid). In *Materials Engineering and Environmental Science*; World Scientific: Singapore, 2015; pp. 21–29. [CrossRef]
189. Guo, J.; Wang, J.; Zheng, G. Synthesis of cross-linking cationic starch and its adsorption properties for reactive dyes. *IOP Conf. Series: Earth Environ. Sci.* **2019**, *227*, 062034. [CrossRef]
190. Lou, T.; Wang, X.; Song, G.; Cui, G. Synthesis and flocculation performance of a chitosan-acrylamide-fulvic acid ternary copolymer. *Carbohydr. Polym.* **2017**, *170*, 182–189. [CrossRef]
191. Fan, X.-M.; Yu, H.-Y.; Wang, D.-C.; Mao, Z.-H.; Yao, J.; Tam, K.M.C. Facile and Green Synthesis of Carboxylated Cellulose Nanocrystals as Efficient Adsorbents in Wastewater Treatments. *ACS Sustain. Chem. Eng.* **2019**, *7*, 18067–18075. [CrossRef]
192. Hussain, G.; Haydar, S. Textile Effluent Treatment Using Natural Coagulant *Opuntia stricta*in Comparison with Alum. *CLEAN –Soil, Air, Water* **2021**, *49*, 2000342. [CrossRef]
193. Du, J.; Yang, X.; Xiong, H.; Dong, Z.; Wang, Z.; Chen, Z.; Zhao, L. Ultrahigh Adsorption Capacity of Acrylic Acid-Grafted Xanthan Gum Hydrogels for Rhodamine B from Aqueous Solution. *J. Chem. Eng. Data* **2021**, *66*, 1264–1272. [CrossRef]
194. Elbedwehy, A.M.; Atta, A.M. Novel Superadsorbent Highly Porous Hydrogel Based on Arabic Gum and Acrylamide Grafts for Fast and Efficient Methylene Blue Removal. *Polymers* **2020**, *12*, 338. [CrossRef]
195. Zhou, L.; Huang, J.; He, B.; Zhang, F.; Li, H. Peach gum for efficient removal of methylene blue and methyl violet dyes from aqueous solution. *Carbohydr. Polym.* **2014**, *101*, 574–581. [CrossRef]
196. Jeon, C.; Höll, W.H. Chemical modification of chitosan and equilibrium study for mercury ion removal. *Water Res.* **2003**, *37*, 4770–4780. [CrossRef]
197. Xu, X.; Zhang, L.; Han, Y.; Zhou, Z. Biosorption of Pb2+ and Zn2+ by Ca-alginate immobilized and free extracellular polysaccharides produced by *Leuconostoc citreum* B-2. *Int. J. Biol. Macromol.* **2021**, *193*, 2365–2373. [CrossRef] [PubMed]
198. Mohammad, N.; Atassi, Y.; Tally, M. Synthesis and swelling behavior of metal-chelating superabsorbent hydrogels based on sodium alginate-g-poly(AMPS-co-AA-co-AM) obtained under microwave irradiation. *Polym. Bull.* **2017**, *74*, 4453–4481. [CrossRef]
199. Abdel-Halim, E.S.; Al-Deyab, S.S. Preparation of poly(acrylic acid)/starch hydrogel and its application for cadmium ion removal from aqueous solutions. *React. Funct. Polym.* **2014**, *75*, 1–8. [CrossRef]
200. Morin-Crini, N.; Staelens, J.-N.; LoIacono, S.; Martel, B.; Chanet, G.; Crini, G. Simultaneous removal of Cd, Co, Cu, Mn, Ni, and Zn from synthetic solutions on a hemp-based felt. III. Real discharge waters. *J. Appl. Polym. Sci.* **2020**, *137*, 48823. [CrossRef]
201. Moradi, M.; Sabzevari, M.H.; Marahel, F.; Shameli, A. Response surface methodology for the high efficiency removal of lead and zinc from effluents using natural sepiolite particles on the corn silk. *Int. J. Environ. Anal. Chem.* **2022**, 1–18. [CrossRef]
202. Li, R.; Zhang, T.; Zhong, H.; Song, W.; Zhou, Y.; Yin, X. Bioadsorbents from algae residues for heavy metal ions adsorption: Chemical modification, adsorption behaviour and mechanism. *Environ. Technol.* **2021**, *42*, 3132–3143. [CrossRef]
203. Kumar, A.; Patra, C.; Rajendran, H.K.; Narayanasamy, S. Activated carbon-chitosan based adsorbent for the efficient removal of the emerging contaminant diclofenac: Synthesis, characterization and phytotoxicity studies. *Chemosphere* **2022**, *307*, 135806. [CrossRef]
204. Sun, H.; Liu, Z.; Liu, K.; Gibril, M.E.; Kong, F.; Wang, S. Lignin-based superhydrophobic melamine resin sponges and their application in oil/water separation. *Ind. Crop. Prod.* **2021**, *170*, 113798. [CrossRef]

205. Zhu, S.; Xu, J.; Wang, B.; Xie, J.; Ying, G.; Li, J.; Cheng, Z.; Li, J.; Chen, K. Highly efficient and rapid purification of organic dye wastewater using lignin-derived hierarchical porous carbon. *J. Colloid Interface Sci.* **2022**, *625*, 158–168. [CrossRef]
206. Salim, M.H.; Kassab, Z.; Ablouh, E.-H.; Sehaqui, H.; Aboulkas, A.; Bouhfid, R.; Qaiss, A.E.K.; El Achaby, M. Manufacturing of macroporous cellulose monolith from green macroalgae and its application for wastewater treatment. *Int. J. Biol. Macromol.* **2022**, *200*, 182–192. [CrossRef]
207. He, Y.; Dietrich, A.M.; Jin, Q.; Lin, T.; Yu, D.; Huang, H. Cellulose adsorbent produced from the processing waste of brewer's spent grain for efficient removal of Mn and Pb from contaminated water. *Food Bioprod. Process.* **2022**, *135*, 227–237. [CrossRef]
208. Si, R.; Chen, Y.; Wang, D.; Yu, D.; Ding, Q.; Li, R.; Wu, C. Nanoarchitectonics for High Adsorption Capacity Carboxymethyl Cellulose Nanofibrils-Based Adsorbents for Efficient Cu^{2+} Removal. *Nanomaterials* **2022**, *12*, 160. [CrossRef]
209. Bisiriyu, I.O.; Meijboom, R. Adsorption of Cu(II) ions from aqueous solution using pyridine-2,6-dicarboxylic acid crosslinked chitosan as a green biopolymer adsorbent. *Int. J. Biol. Macromol.* **2020**, *165*, 2484–2493. [CrossRef]
210. Baskar, A.V.; Bolan, N.; Hoang, S.A.; Sooriyakumar, P.; Kumar, M.; Singh, L.; Jasemizad, T.; Padhye, L.P.; Singh, G.; Vinu, A.; et al. Recovery, regeneration and sustainable management of spent adsorbents from wastewater treatment streams: A review. *Sci. Total. Environ.* **2022**, *822*, 153555. [CrossRef]
211. Omorogie, M.O.; Babalola, J.O.; Unuabonah, E.I. Regeneration strategies for spent solid matrices used in adsorption of organic pollutants from surface water: A critical review. *Desalination Water Treat.* **2014**, *57*, 518–544. [CrossRef]
212. Arabkhani, P.; Javadian, H.; Asfaram, A.; Hosseini, S.N. A reusable mesoporous adsorbent for efficient treatment of hazardous triphenylmethane dye wastewater: RSM-CCD optimization and rapid microwave-assisted regeneration. *Sci. Rep.* **2021**, *11*, 2275. [CrossRef]
213. McQuillan, R.V.; Stevens, G.W.; Mumford, K.A. The electrochemical regeneration of granular activated carbons: A review. *J. Hazard. Mater.* **2018**, *355*, 34–49. [CrossRef]
214. Gautam, R.; Bassi, A.S.; Yanful, E.K. A review of biodegradation of synthetic plastic and foams. *Appl. Biochem. Biotechnol.* **2007**, *141*, 85–108. [CrossRef]
215. Kyrikou, I.; Briassoulis, D. Biodegradation of Agricultural Plastic Films: A Critical Review. *J. Polym. Environ.* **2007**, *15*, 125–150. [CrossRef]
216. Kolya, H.; Tripathy, T. Biodegradable flocculants based on polyacrylamide and poly(N,N-dimethylacrylamide) grafted amylopectin. *Int. J. Biol. Macromol.* **2014**, *70*, 26–36. [CrossRef]
217. Hosseinzadeh, H.; Ramin, S. Fabrication of starch-graft-poly(acrylamide)/graphene oxide/hydroxyapatite nanocomposite hydrogel adsorbent for removal of malachite green dye from aqueous solution. *Int. J. Biol. Macromol.* **2018**, *106*, 101–115. [CrossRef] [PubMed]