Biocoagulant/bioflocculant application for drinking water

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Contributor: Setyo Budi Kurniawan

The utilization of metal-based conventional coagulants/flocculants to remove suspended solids from drinking water and wastewater is currently leading to new concerns. Alarming issues related to the prolonged effects on human health and further pollution to aquatic environments from the generated nonbiodegradable sludge are becoming trending topics. The utilization of biocoagulants/bioflocculants does not produce chemical residue in the effluent and creates nonharmful, biodegradable sludge.

Keywords: alum ; coagulation ; environment ; green technology ; biocoagulant ; bioflocculant ; flocculant ; coagulant ; flocculation

1. Introduction

Water is part of our life and a basic necessity for humans. It is one of the main life supports for humans. Humans need water for use as a source of body fluids and for several activities, such as bathing, washing, and using latrines. Some of these activities later cause the generation of wastewater. Treatment processing is needed to maintain the stability and continuity of the water supply. Particularly, drinking water and wastewater treatment are an important part of the water cycle in human life.

Several treatment technologies are used to process raw water sources into drinking water and transform wastewater into treated effluent before it is discharged to water bodies, and these treatments include conventional and advanced technologies [1][2]. Most of the treatment processes, whether for water or wastewater, cannot be separated from coagulation and flocculation stages, as part of the treatment processes. Coagulation and flocculation are parts of a water treatment system that have the main function of separating suspended particles in water to produce clear and suspension-free effluent [3]. The step of the coagulation–flocculation process is normally in the primary treatment of a water or wastewater treatment system [4][5].

The processes of coagulation and flocculation require the addition of compounds known as coagulants and flocculants [6]. The main types of coagulants and flocculants used in the treatment of drinking water and wastewater are divalent positively charged chemical compounds. Negatively charged polymers are also largely used in water treatment, notably as high molecular weight flocculants [7]. The chemical compounds commonly used as coagulants/flocculants include iron salts ($FeCl_3$ or $Fe_2(SO_4)_3$) [8], aluminum salts ($Al_2(SO_4)_3$) [8][9], hydrated lime [8], magnesium carbonate [8], and polymers (aluminum chlorohydrate, polyaluminum chloride (PAC), polyaluminum sulfate chloride, and polyferric sulfate) [10]. Some of the mentioned compounds have been shown to be effective in reducing suspended solid concentrations in water.

The application of these compounds is not necessarily free from impacts [11]. Several environmental problems due to the chronic toxicity of coagulants/flocculants are currently being discussed, specifically for environmental observers worldwide [12][13][14]. In-depth analysis has been conducted in relation to the impact that can be caused by the use of chemical compounds as coagulants and flocculants [15][16][17][18]. The environmental impacts include increasing the corrosion rate of metallic utilities [19], changing the pH, limiting root elongation, and inhibiting seed germination [20][21]. Water and wastewater treatment involving conventional coagulants/flocculants also generates excessive chemical sludge in addition to the suspended solids to be removed; thus, the handling of chemical sludge becomes another issue to resolve. Aside from these impacts on the environment, concerns related to human health arise. Metallic-based coagulants/flocculants are nondegradable or nonbiodegradable, and their residuals in drinking water can induce a direct impact on human health when consumed and can be accumulated in body cells [22][23]. The residuals of chemical coagulants/flocculants, when used in wastewater treatment, in treated effluent discharged to the environment may be trapped in food chains [22][23]. Some indications regarding the impacts of chemical coagulants/flocculants on human health, including central nervous system failure, dementia, Alzheimer’s disease, and severe trembling, have been reported [24][25][26][27].

Biocoagulants/bioflocculants can be an alternative solution to minimize the environmental pollution and health risks caused by the use of chemical coagulants/flocculants. Biocoagulants and bioflocculants come from living things or their parts and are totally organic and biodegradable; therefore, they are environmentally friendly and have minimal impacts on human health [28]. Research related to biocoagulants and bioflocculants has undergone many stages until their application to treatment processing units [29][30]. Some biocoagulants and bioflocculants obtained from various sources have already been analyzed and been proven efficient for application to treatment processes as a substitution for the currently widely used chemical coagulants and flocculants [30][31][32][33][34].

2. Characterization of Biocoagulants and Bioflocculants

2.1. Origin of Biocoagulants and Bioflocculants

During ancient times, people were not well exposed to proper water treatment. For normal household usage, water was just boiled and filtered to acquire consumable water. This scenario is still present in certain regions with limited access to proper water sources and technologies. As time evolved, people found a method to clarify unclean water by adding some plant powder, which is termed biocoagulant, to turbid water to settle down the dirt. In the 19th century, metal coagulants were introduced and started to gain global attention. China was the first country to use alum for wastewater treatment [35]. Chemical coagulants were continuously improved afterward to achieve the highest efficiency and suitability with the greatest operating conditions; hence, enhanced coagulants were produced, and their relevance developed over time.

2.2. Chemical Characteristics

Biocoagulants can be extracted from plants, animals, or microorganisms [36][37]. The important characteristics of these sources that enable them to be used as biocoagulants are the contents of polysaccharides [38], protein polymers [36], and some functional groups [35], such as hydroxyl and carboxyl groups. Polysaccharides, protein, and some functional groups promote the mechanisms of adsorption, polymer bridging, and charge neutralization (Section 4.3). Several major compounds that could perform as biocoagulants/bioflocculants are summarized in Table 1.

Table 1. Characterization of chemical contents in biocoagulants/bioflocculants.

No	Type	Species	Chemical Compounds/Functional Groups	Source
1	Animal-based	Shellfish	Chitin and polysaccharides	[33]
2	Animal-based	Shrimp shell	Chitosan and carboxy methyl cellulose	[39]
3	Animal-based	Periwinkle shell	Alcohol, phenol, secondary amide group, amine group, alkyne group, and polysaccharides	[40]
4	Animal-based	Crab shell	Chitosan	[41]
5	Microorganism-based (bacteria)	*Bacillus agaradhaerens* C9	Carboxyl, hydroxyl, amino, and glycoprotein groups	[42]
6	Microorganism-based (bacteria)	*Bacillus mucilaginosus*	Uronic acid, neutral sugar, amino sugar, carboxyl group, and hydroxyl group	[43]
7	Microorganism-based (bacteria)	*Bacillus salmalaya* 139SI-7	Carboxyl group, hydroxyl group, amino group, polysaccharides, and proteins	[44]
8	Microorganism-based (bacteria)	*Paenibacillus polymyxa*	Polysaccharides and proteins	[45]
9	Microorganism-based (bacteria)	*Bacillus licheniformis* strain W7	Polysaccharides, protein, hydroxyl group, carboxyl group, and amino group	[46]
10	Microorganism-based (bacteria)	*Bacillus velezensis*	Xylose and glucose	[47]
11	Plant-based	Rice starch	Cellulose, lignin, aldehydes, ketones, esters, and carboxylic acids	[48]
12	Plant-based	*Lens culinaris*	Hydroxyl and carboxyl groups	[49]

No	Type	Species	Chemical Compounds/Functional Groups	Source
13	Plant-based	Cassava	Amino acids, carboxyl group, and hydroxyl group	[50]
14	Plant-based	*Dillenia indica*	Polysaccharides	[38]
15	Plant-based	Potato starch	Branched-structure polymers	[51]
16	Plant-based	*Moringa oleifera* seed	Cationic protein, starch, glucose, fatty acids, and phenolic compounds	[52]
17	Plant-based	*Moringa oleifera* seed	Alcoholic compound, polysaccharides, and amides	[53]
18	Plant-based	*Moringa oleifera* seed	Amines, carboxylate groups, and alcoholic compounds	[54]

2.3. Working Mechanism of Natural Coagulants/Flocculants

The mechanisms of natural coagulation are mainly adsorption, charge neutralization, polymer bridging, precipitative coagulation, and electrostatic patching. The first three are the main mechanisms of biocoagulation, as described below.

- Adsorption: Natural polymers provide a free surface to adsorb colloid particles and form larger particles that are easier to settle down [55].
- Polymer bridging: Colloid particles will attach to a part of a long-chain polymer, while the other free part of the chain will form a loop and a tail. The molecules will continue to form a larger molecule when the free tail attaches with another free colloid, increasing the particle size. The correct dosage of coagulants to provide a free surface for the process is important [55].
- Charge neutralization: Colloid particles are normally negative in charge and cannot form a larger particle because they repel one another. Thus, the addition of cationic biocoagulants will produce carboxylate and H^+ ions to neutralize the suspension near to zero zeta potential and make the formation of a large floc possible. A low dosage of coagulants will be needed for the treatment if they have a high charge density.

Natural coagulants produce a five times lower volume of sludge compared with inorganic salts. Dorea [111] stated that this condition occurs because alum requires as many as three molecules of water hydration to fulfil its covalent bond, thus resulting in an increment in sludge volume. The sludge produced in biocoagulation is biodegradable, with high nutritional value, and it is safe and suitable for land usage (biofertilizer) [56][57].

In addition to being a clarification agent, biocoagulants have also been reported to have antimicrobial and heavy metal removal properties, which are effective in high-turbidity water [58]. Choy et al. mentioned that aside from starch, phytochemicals, such as tannins and alkaloids, help in antimicrobial activities. On the contrary, natural coagulants will increase the organic matter concentration in the water, thus leading to undesired microbial activities because the antimicrobial efficiency of biocoagulants is normally low. Organic matter will also affect the color, odor, and taste of water. Accordingly, Gunaratna et al. [58] suggested removing the content in natural coagulants through simple purification/filtration.

2.4. Ethical Utilization and Toxicity

In coagulation treatment, traces of coagulants may remain in treated water. Hence, the usage of natural coagulants is safe, and no serious problems regarding pipe corrosion will occur due to their noncorrosive properties. The application of alum to water treatment has been reported to lead to health problems, such as Alzheimer's disease [58]. Thus, the substitution of chemical coagulants with green coagulants, which are safer, eco-friendly, and low-cost, is recommended. Natural coagulants have effectiveness comparable with that of chemical coagulants for treating wastewater but have not been successfully commercialized yet due to the lack of scientific proof of their working mechanism and efficiency.

3. Application of Biocoagulants/Bioflocculants to Drinking Water and Wastewater Treatment

The utilization of biocoagulants/bioflocculants shows reliable performance in treating drinking water and wastewater. Most of the parameters of pollutants in drinking water and wastewater can be removed via the utilization of biocoagulants/bioflocculants. Those parameters include the total suspended solids (TSSs), biological oxygen demand

(BOD), chemical oxygen demand (COD), color, and nutrients. A summary of the performance of biocoagulants/bioflocculants in removing pollutants in drinking water and wastewater is presented in Table 2.

To summarize the compilation of data in Table 2, the performance of biocoagulants/bioflocculants in removing pollutants (e.g., TSSs, COD, BOD, algae, and color) is undeniably great compared with that of conventional metal-based coagulants/flocculants. Biocoagulants/bioflocculants can achieve similar or even higher pollutant removal efficiency than the conventional flocculants. Most of the countries involved in the research into biocoagulants/bioflocculants are tropical and developing countries. This phenomenon can be ascribed to the abundance and diversity of potential resources (whether from waste or by-products), especially plants and crustaceans, to be utilized as biocoagulants/bioflocculants due to the tropical climate [59][60].

Plant-based biocoagulants/bioflocculants are still being specialized in this research topic. Most research has already implemented the utilization of local resources (native plants) or isolation from the indigenous environment (for microorganisms). However, research that utilizes waste or by-products to seek their potential as biocoagulants/bioflocculants is still limited； further study on this particular theme could be a future direction. Most of the animal-based biocoagulants/bioflocculants come from crustacean studies [61] because the composition of the chitosan of crustaceans is beneficial for coagulation/flocculation. Additional study on another phylum might be interesting to provide alternative technologies. Extensive studies on fungus- and alga-based biocoagulants/bioflocculants will contribute to this topic, considering that research from these sources is currently still scarce [62][63].

Table 2. Performance of biocoagulants/bioflocculants in treating drinking water and wastewater.

No.	Name	Type	Function	Treated Water	Summary	Country
1	*Achatinoidea* shell	Animal-based	Biocoagulant	Paint industry wastewater	*Achatinoidea* shell could reduce total dissolved solid (TDS) by up to 13% for 35 min of settling time with a dosage of 4 g/L at pH 7.9. Optimum performance of 99.22% was obtained at pH 4, 4 g/L dosage, and 45 °C.	Texas
2	Crab shell	Animal-based	Biocoagulant	Lake water	The crab shell could aid alum as a biocoagulant to enhance turbidity removal (97%) with 0.2 mg/L dosage after 45 min of settling time. Crab shell could be used as a natural aid coagulant for drinking water treatment with the lowest risks of organic release.	Algeria

3	Crab shells	Animal-based	Biocoagulant	Drinking water	Combining crab shell as biocoagulant and alum could reduce turbidity of low-, medium-, and high-turbidity water by up to 74.8%, 96.7%, and 98.2%, respectively. This removal was higher than that using only alum as coagulant. This biocoagulant could reduce the alum dose by up to 75%, and the sludge by-product is readily biodegradable. The optimum pH and biocoagulant dose for removing turbidity were 7 and 1.5 mg/L, respectively.	India
4	Periwinkle shell	Animal-based	Biocoagulant	Petroleum wastewater	Varying the dosage of periwinkle shell and pH had a significant effect on the coagulation–flocculation efficiency. The optimum conditions were pH 4 and a 100 mg/L periwinkle shell dosage. The removal of particles was up to 83.57%.	Texas
5	Shrimp shells	Animal-based	Biocoagulant	Wastewater containing oil	The chitosan from shrimp shell as a biocoagulant could reduce oil by up to 96.35% at pH 4 over 60 min of contact time. The removal of oil by using chitosan was increased after adding carboxy methyl cellulose (CMC), with percentage efficiency of 99% at (90% chitosan and 10% CMC) with 30–60 min of contact time.	Egypt

6	Snail shell	Animal-based	Biocoagulant	Wastewater containing dye	The snail shell alone as biocoagulant could reduce malachite green (MG) dye by up to 60% with a dosage of 100 mg/L. The combination of snail shell and alum could enhance the removal of MG dye. The optimum pH for MG dye removal was found to range between 4 and 5. The optimum flocculation time was 30 min with an alum–snail shell dosage of 20–100 mg/L. The sludge produced from the alum–snail shell combination had better settling characteristics than the sludge obtained from the use of snail shell alone.	Nigeria
7	Alginate	Microorganism-based (algae)	Biocoagulant	Drinking water	Algal alginate has a high polysaccharide content that could perform as a biocoagulant. Alginate removed up to 98% of suspended solids from high-turbidity water. A low dosage of the coagulant (as low as 0.02 mg/L) still achieved high turbidity removal.	Turkey
8	*Achromobacter xylosoxidans* strain TERI L1	Microorganism-based (bacteria)	Bioflocculant	Wastewater containing heavy metals	*Achromobacter xylosoxidans* strain TERI L1 could produce exopolysaccharide as a bioflocculant. The bioflocculant contained 75% total sugar, with 72.9% neutral sugar and 11.5% protein. *Achromobacter xylosoxidans* strain TERI L1 could flocculate Zn, Pb, Ni, Cd, and Cu by up to 90%.	India

9	*Bacillus agaradhaerens* C9	Microorganism-based (bacteria)	Bioflocculant	Wastewater containing microalgae	A bioflocculant was extracted from *Bacillus agaradhaerens* C9 and contained 65.42% polysaccharides, 4.70% proteins, and 1.65% nucleic acids. The optimum conditions for producing bioflocculant from *Bacillus agaradhaerens* C9 were 10 g/L of glucose, 10 g/L of yeast extract, and an initial pH of 10.2. The flocculation rate for kaolin suspension was 95.29%, with optimum dosage, pH, and temperature of 1.5 mg/L, 6.53, and 29 °C, respectively. The bioflocculant had the potential to treat alkaline wastewater.	China
10	*Bacillus licheniformis* strain W7	Microorganism-based (bacteria)	Bioflocculant	Synthetic wastewater containing kaolin and river water	A bioflocculant (MBF-W7) was produced using *Bacillus licheniformis* strain W7. The optimum conditions for flocculant production were a 5% (*v/v*) inoculum size with maltose and NH_4NO_3 as carbon and nitrogen sources. The pH and cultivation time were 6 and 72 h, respectively. The flocculation rate for kaolin clay suspension was 85.8%, observed at pH 3, and MBF-W7 of 0.2 mg/mL. MBF-W7 could remove turbidity and chemical oxygen demand (COD) by up to 86.9% and 75.3%, respectively, in Tyume River.	South Afri

11	*Bacillus mucilaginosus*	Microorganism-based (bacteria)	Bioflocculant	Starch wastewater	A bioflocculant (MBFA9) was produced from *Bacillus mucilaginosus*. The major component was a polysaccharide that contained uronic acid (19.1%), neutral sugar (47.4%), and amino sugar (2.7%). The flocculation rate for kaolin suspension was 99.6% with a 0.1 mL/L MBFA9 dosage. MBFA9 could reduce total suspended solid (TSS) and COD by up to 85.5% and 68.5%, respectively.	Singapore
12	*Bacillus salmalaya* 139SI-7	Microorganism-based (bacteria)	Bioflocculant	Organic-rich wastewater	A bioflocculant (QZ-7) was synthesized using *Bacillus salmalaya* strain 139SI with flocculation activity of 83.3%. The optimum temperature, pH, and incubation time conditions for flocculant production were 35.5 °C, 7, and 72 h, respectively, with inoculum size of 5% (*v*/*v*), sucrose as carbon source, and yeast extract as nitrogen source. Bioflocculant QZ-7 could remove COD and BOD by 93% and 92.4%, respectively.	Malaysia

13	*Bacillus velezensis*	Microorganism-based (bacteria)	Bioflocculant	Lake water	This study investigated the effects of incubation time and temperature on the production of bioflocculants by using *Bacillus velezensis* grown in sago mill effluent (SME) and palm oil mill effluent (POME) as a fermentation feedstock. The highest bioflocculant yield (2.03 g/L) at a temperature of 40 °C was achieved in POME medium. The bioflocculant produced from a fermented SME medium (BioF-SME) showed the highest activity. Bioflocculants from POME and SME had performance comparable with alum's in removing color and turbidity from lake water.	Malaysia
14	*Chromobacterium violaceum* and *Citrobacter koseri*	Microorganism-based (bacteria)	Bioflocculant	Tapioca wastewater	*Chromobacterium violaceum* and *Citrobacter koseri* were isolated from tapioca wastewater and had high flocculation activities of 68.92% and 71.38%, respectively. The optimum pH and temperature for *Chromobacterium violaceum* and *Citrobacter koseri* were 2–4 and 6–8 and 40 °C and 30 °C, respectively.	Indonesia

15	*Paenibacillus polymyxa*	Microorganism-based (bacteria)	Bioflocculant	Formaldehyde wastewater	A novel bioflocculant-producing bacterium (MBF-79) was isolated from formaldehyde wastewater sludge. The optimum inoculum size, pH, and formaldehyde concentration for bioflocculant production were 7.0%, 6, and 350 mg/L, respectively. The major components of MBF-79 were polysaccharide (71.2%) and protein (27.9%). The optimum MBF-79, pH, contact time for the removal of arsenate and arsenite by using MBF-79 were 120 mg/L, 7, and 60 min, respectively, with removal efficiencies of 98.9% and 84.6%, respectively.	China
16	*Aspergillus niger*	Microorganism-based (fungi)	Bioflocculant	Aquaculture wastewater	*Aspergillus niger* was applied to flocculate microalgae from aquaculture wastewater. More than 90% harvesting efficiency was obtained at pH 3.0 to 9.0 and a mixing rate of 100–150 rpm.	Malaysia
17	*Aspergillus niger*	Microorganism-based (fungi)	Bioflocculant	Potato starch wastewater	Two milliliters of the bioflocculant produced using *A. niger* was able to remove up to 91.15% of COD and 60.22% of turbidity within 20 min of treatment. Compared with the conventional coagulants (alum- and iron-based), this bioflocculant showed nearly identical performance with a lower material cost and a smaller yield of sludge.	Hong Kon
18	*Penicillium sp.* and *Trichoderma sp.*	Microorganism-based (fungi)	Biocoagulant	Domestic wastewater	Suspension of fungal spores was proven to reduce 84% (relative to alum efficiency) of turbidity from sewage at pH 7.8 with 60 min of treatment.	Iraq

19	*Abelmoschus esculentus*	Plant-based	Biocoagulant	Industrial textile wastewater	*Abelmoschus esculentus* as biocoagulant is more efficient for treating textile wastewater than chloride ferric. *Abelmoschus esculentus* can remove turbidity, COD, and color by up to 97.25%, 85.69%, and 93.57%, respectively, with optimum pH and concentration of biocoagulant of 6 and 3.2 mg/L, respectively.	Brazil
20	Dragon fruit foliage	Plant-based	Biocoagulant	Concentrated latex wastewater	Dragon fruit foliage as biocoagulant could reduce COD, SS, and turbidity from latex effluent by up to 94.7%, 88.9%, and 99.7%, respectively, at pH 10. The biocoagulant dosage range of 200–800 mg/L showed consistent removal of pollutants. The removal percentage for pollutants using ferric sulfate was higher than that using dragon fruit foliage.	Malaysia
21	*Moringa oleifera*	Plant-based	Biocoagulant	Synthetic turbid wastewater	Raw *Moringa oleifera* seed contains high amounts of oil, which can reduce the potential for coagulation activity. Oil extraction significantly increased the coagulation activity of *Moringa oleifera* seed. The utilization of this biocoagulant showed 82.43% oil and grease removal from water.	Brazil
22	*Moringa oleifera*	Plant-based	Biocoagulant	Hospital wastewater	*Moringa oleifera* extract contains dimeric protein. Utilization of this biocoagulant showed 65% removal of turbidity, 38% of COD, and up to 90% removal of *Pseudomonas aeruginosa.*	Benin

23	*Moringa oleifera*	Plant-based	Biocoagulant	Drinking water	Integrating seed powder of *Moringa oleifera* into solar water disinfection could reduce turbidity by up to 85% in 24 h and remove *Escherichia coli* in 6 h.	Ireland

References

1. Madaki, Y.S.; Seng, L. Palm Oil Mill Effluent (Pome) from Malaysia Palm Oil Mills: Waste or Resource. Int. J. Sci. Environ. Technol. 2013, 2, 1138–1155.
2. Rajasulochana, P.; Preethy, V. Comparison on efficiency of various techniques in treatment of waste and sewage water – A comprehensive review. Resour. Technol. 2016, doi:10.1016/j.reffit.2016.09.004.
3. Duan, J.; Gregory, J. Coagulation by hydrolysing metal salts. Adv. Colloid Interface Sci. 2003, doi:10.1016/S0001-8686(02)00067-2.
4. Edzwald, J.K. Coagulation in drinking water treatment: Particles, organics and coagulants. Water Sci. Technol. 1993, 27, 21–35.
5. Ebeling, J.M.; Sibrell, P.L.; Ogden, S.R.; Summerfelt, S.T. Evaluation of chemical coagulation-flocculation aids for the removal of suspended solids and phosphorus from intensive recirculating aquaculture effluent discharge. Aquac. Eng. 2003, 29, 23–42, doi:10.1016/S0144-8609(03)00029-3.
6. Hosseini, M.; Koohestanian, A.; Abbasian, Z. The Separation Method for Removing of Colloidal Particles from Raw Water. J. Agric. Environ. Sci. 2008, 4, 266–273
7. Lapointe, M.; Barbeau, B. Understanding the roles and characterizing the intrinsic properties of synthetic vs. natural polymers to improve clarification through interparticle Bridging: A review. Sep. Purif. Technol. 2020, 231, doi:10.1016/j.seppur.2019.115893.
8. Moran, S. Engineering science of water treatment unit operations. In An Applied Guide to Water and Effluent Treatment Plant Design; Elsevier: Amsterdam, The Netherlands, 2018; pp. 39–51.
9. Aragonés-Beltrán, P.; Mendoza-Roca, J.A.; Bes-Piá, A.; García-Melón, M.; Parra-Ruiz, E. Application of multicriteria decision analysis to jar-test results for chemicals selection in the physical–chemical treatment of textile wastewater. J. Hazard. Mater. 2009, 164, 288–295, doi:10.1016/j.jhazmat.2008.08.046.
10. Weydts, D.; De Smet, D.; Vanneste, M. Processes for reducing the environmental impact of fabric finishing. In Sustainable Apparel; Elsevier: Amsterdam, The Netherlands, 2015; pp. 35–48.
11. Barakwan, R.A.; Trihadiningrum, Y.; Bagastyo, A.Y. Characterization of alum sludge from Surabaya Water Treatment Plant, Indonesia. J. Ecol. Eng. 2019, 20, 7–13, doi:10.12911/22998993/104619.
12. Imron, M.F.; Kurniawan, S.B.; Ismail, N. ‘Izzati; Abdullah, S.R.S. Future challenges in diesel biodegradation by bacteria isolates: A review. J. Clean. Prod. 2020, 251, 119716, doi:10.1016/j.jclepro.2019.119716.
13. Cardwell, A.S.; Adams, W.J.; Gensemer, R.W.; Nordheim, E.; Santore, R.C.; Ryan, A.C.; Stubblefield, W.A. Chronic toxicity of aluminum, at a pH of 6, to freshwater organisms: Empirical data for the development of international regulatory standards/criteria. Environ. Toxicol. Chem. 2018, 37, 36–48, doi:10.1002/etc.3901.
14. Tomljenovic, L. Aluminum and Alzheimer’s disease: After a century of controversy, is there a plausible link? J. Alzheimer’s Dis. 2011, 23, 567–598, doi:10.3233/JAD-2010-101494.
15. Tetteh, E.K.; Rathilal, S. Application of Organic Coagulants in Water and Wastewater Treatment. Org. Polym. 2019, doi:10.5772/intechopen.84556.
16. Fort, D.J.; Stover, E.L. Impact of toxicities and potential interactions of flocculants and coagulant aids on whole effluent toxicity testing. Water Environ. Res. 1995, 67, 921–925, doi:10.2175/106143095x133149.
17. Rahmani, R.; Shahmoradi, B.; Maleki, A. Bioassay testing the toxicity of nano-structure polymer (PAMAM G2) as coagulant aid in water treatment. Res. J. Environ. Toxicol. 2015, 9, 261–267, doi:10.3923/rjet.2015.261.267.
18. Al-Mutairi, N.Z. Coagulant toxicity and effectiveness in a slaughterhouse wastewater treatment plant. Ecotoxicol. Environ. Saf. 2006, 65, 74–83, doi:10.1016/j.ecoenv.2005.05.013.
19. Niquette, P.; Monette, F.; Azzouz, A.; Hausler, R. Impacts of substituting aluminum-based coagulants in drinking water treatment. Water Qual. Res. J. Canada 2004, 39, 303–310, doi:10.2166/wqrj.2004.041.
20. Zhang, K.; Zhou, Q. Toxic effects of Al-based coagulants on Brassica chinensis and Raphanus sativus growing in acid and neutral conditions. Environ. Toxicol. 2005, 20, 179–187, doi:10.1002/tox.20093.
21. Ismail, N. ‘Izzati; Abdullah, S.R.S.; Idris, M.; Kurniawan, S.B.; Effendi Halmi, M.I.; AL Sbani, N.H.; Jehawi, O.H.; Hasan, H.A. Applying rhizobacteria consortium for the enhancement of Scirpus grossus growth and phytoaccumulation of Fe

and Al in pilot constructed wetlands. J. Environ. Manage. 2020, 267, doi:10.1016/j.jenvman.2020.110643.

22. Kluczka, J.; Zołotajkin, M.; Ciba, J.; Staroń, M. Assessment of aluminum bioavailability in alum sludge for agricultural utilization. Environ. Monit. Assess. 2017, 189, 422, doi:10.1007/s10661-017-6133-x.

23. Mortula, M.; Bard, S.M.; Walsh, M.E.; Gagnon, G.A. Aluminum toxicity and ecological risk assessment of dried alum residual into surface water disposal. Can. J. Civ. Eng. 2009, 36, 127–136, doi:10.1139/S08-042.

24. Barakwan, R.A.; Hardina, T.T.; Trihadiningrum, Y.; Bagastyo, A.Y. Recovery of alum from Surabaya water treatment sludge using electrolysis with carbon-silver electrodes. J. Ecol. Eng. 2019, 20, 126–133, doi:10.12911/22998993/109861.

25. Ahmad, T.; Ahmad, K.; Alam, M. Sustainable management of water treatment sludge through 3′R' concept. J. Clean. Prod. 2016, 124, 1–13, doi:10.1016/j.jclepro.2016.02.073.

26. Exley, C. Aluminum Should Now Be Considered a Primary Etiological Factor in Alzheimer's Disease. J. Alzheimer's Dis. Rep. 2017, 1, 23–25, doi:10.3233/ADR-170010.

27. Mirza, A.; King, A.; Troakes, C.; Exley, C. Aluminium in brain tissue in familial Alzheimer's disease. J. Trace Elem. Med. Biol. 2017, 40, 30–36, doi:10.1016/j.jtemb.2016.12.001.

28. Adnan, O.; Abidin, Z.Z.; Idris, A.; Kamarudin, S.; Al-Qubaisi, M.S. A novel biocoagulant agent from mushroom chitosan as water and wastewater therapy. Environ. Sci. Pollut. Res. 2017, 24, 20104–20112, doi:10.1007/s11356-017-9560-x.

29. Oladoja, N.A. Headway on natural polymeric coagulants in water and wastewater treatment operations. J. Water Process Eng. 2015, 6, 174–192, doi:10.1016/j.jwpe.2015.04.004.

30. Chethana, M.; Sorokhaibam, L.G.; Bhandari, V.M.; Raja, S.; Ranade, V.V. Green Approach to Dye Wastewater Treatment Using Biocoagulants. ACS Sustain. Chem. Eng. 2016, doi:10.1021/acssuschemeng.5b01553.

31. Braga, W.L.M.; Roberto, J.A.; Vaz, C.; Samanamud, G.R.L.; Loures, C.C.A.; França, A.B.; Lofrano, R.C.Z.; Naves, L.L.R.; José Henrique De Freitas Gomes, J.H.; Naves, F.L. Extraction and optimization of tannin from the flower of Musa sp. applied to the treatment of iron ore dump. J. Environ. Chem. Eng. 2018, doi:10.1016/j.jece.2018.05.058.

32. Yongabi, K.A; Lewis, D.M.; Harris, P.L. A Moringa Oleifera Disinfectant-Sand Filter Integration : A Review of an Alternative Sustainable Technology for Household Water Treatment. Environ. Sci. Eng. 2011, 5, 1100–1108.

33. Lichtfouse, E.; Morin-Crini, N.; Fourmentin, M.; Zemmouri, H.; do Carmo Nascimento, I.O.; Queiroz, L.M.; Tadza, M.Y.M.; Picos-Corrales, L.A.; Pei, H.; Wilson, L.D.; et al. Chitosan for direct bioflocculation of wastewater. Environ. Chem. Lett. 2019, 17, 1603–1621, doi:10.1007/s10311-019-00900-1.

34. Ogunlade, A.O.; Oyetayo, V.O.; Ojokoh, A.O. Effect of different biocoagulants on the microbial quality and mineral composition of west African cheese produced from sheep milk. Food Res. 2019, doi:10.26656/fr.2017.3(3).238.

35. Choy, S.Y.; Prasad, K.M.N.; Wu, T.Y.; Raghunandan, M.E.; Ramanan, R.N. Utilization of plant-based natural coagulants as future alternatives towards sustainable water clarification. J. Environ. Sci. 2014, 26, 2178–2189, doi:10.1016/j.jes.2014.09.024.

36. Amran, A.H.; Syamimi Zaidi, N.; Muda, K.; Wai Loan, L. Effectiveness of Natural Coagulant in Coagulation Process: A Review. Int. J. Eng. Technol. 2018, 7, 34, doi:10.14419/ijet.v7i3.9.15269.

37. Muruganandam, L.; Kumar, M.P.S.; Jena, A.; Gulla, S.; Godhwani, B. Treatment of waste water by coagulation and flocculation using biomaterials. IOP Conf. Ser. Mater. Sci. Eng. 2017, 263, doi:10.1088/1757-899X/263/3/032006.

38. Manholer, D.D.; De Souza, M.T.F.; Ambrosio, E.; De Souza Freitas, T.K.F.; Geraldino, H.C.L.; Garcia, J.C. Coagulation/flocculation of textile effluent using a natural coagulant extracted from Dillenia indica. Water Sci. Technol. 2019, 80, 979–988, doi:10.2166/wst.2019.342.

39. Hosny, R.; Fathy, M.; Ramzi, M.; Abdel Moghny, T.; Desouky, S.E.M.; Shama, S.A. Treatment of the oily produced water (OPW) using coagulant mixtures. Egypt. J. Pet. 2016, 25, 391–396, doi:10.1016/j.ejpe.2015.09.006.

40. Menkiti, M.C.; Ndaji, C.R.; Ezemagu, I.G.; Uddameri, V. Application of Periwinkle Shell Coagulant (PSC) for the Remediation of Petroleum Produced Water (PPW) by Coag-Flocculation. J. Dispers. Sci. Technol. 2016, 37, 760–774, doi:10.1080/01932691.2015.1060488.

41. Zemmouri, H.; Drouiche, M.; Sayeh, A.; Lounici, H.; Mameri, N. Coagulation flocculation test of Keddara's water dam using chitosan and sulfate aluminium. Procedia Eng. 2012, 33, 254–260, doi:10.1016/j.proeng.2012.01.1202.

42. Subudhi, S.; Bisht, V.; Batta, N.; Pathak, M.; Devi, A.; Lal, B. Purification and characterization of exopolysaccharide bioflocculant produced by heavy metal resistant Achromobacter xylosoxidans. Carbohydr. Polym. 2016, 137, 441–451, doi:10.1016/j.carbpol.2015.10.066.

43. Deng, S.B.; Bai, R.B.; Hu, X.M.; Luo, Q. Characteristics of a bioflocculant produced by Bacillus mucilaginosus and its use in starch wastewater treatment. Appl. Microbiol. Biotechnol. 2003, 60, 588–593, doi:10.1007/s00253-002-1159-5.

44. Tawila, Z.M.A.; Ismail, S.; Dadrasnia, A.; Usman, M.M. Production and characterization of a bioflocculant produced by bacillus salmalaya 139si-7 and its applications in wastewater treatment. Molecules 2018, 23, 2689, doi:10.3390/molecules23102689.

45. Zhao, H.; Zhong, C.; Chen, H.; Yao, J.; Tan, L.; Zhang, Y.; Zhou, J. Production of bioflocculants prepared from formaldehyde wastewater for the potential removal of arsenic. J. Environ. Manag. 2016, 172, 71–76,

doi:10.1016/j.jenvman.2016.02.024.

46. Okaiyeto, K.; Nwodo, U.U.; Mabinya, L.V.; Okoli, A.S.; Okoh, A.I. Evaluation of flocculating performance of a thermostable bioflocculant produced by marine Bacillus sp. Environ. Technol. 2016, 37, 1829–1842, doi:10.1080/09593330.2015.1133717.
47. Hasan, H.A.; Ezril Hafiz, R.; Muhamad, M.H.; Sheikh Abdullah, S.R.; Hasan, H.A.; Ezril Hafiz, R.; Muhamad, M.H.; Sheikh Abdullah, S.R.; Hassimi, A.H.; Ezril Hafiz, R.; et al. Bioflocculant production using palm oil mill and sago mill effluent as a fermentation feedstock: Characterization and mechanism of flocculation. J. Environ. Manag. 2020, 260, doi:10.1016/j.jenvman.2019.110046.
48. Teh, C.Y.; Wu, T.Y.; Juan, J.C. Potential use of rice starch in coagulation-flocculation process of agro-industrial wastewater: Treatment performance and flocs characterization. Ecol. Eng. 2014, 71, 509–519, doi:10.1016/j.ecoleng.2014.07.005.
49. Chua, S.C.; Malek, M.A.; Chong, F.K.; Sujarwo, W.; Ho, Y.C. Red lentil (Lens culinaris) extract as a novel natural coagulant for turbidity reduction: An evaluation, characterization and performance optimization study. Water 2019, 11, 1686, doi:10.3390/w11081686.
50. Othman, N.; Abd-Rahim, N.S.; Tuan-Besar, S.N.F.; Mohd-Asharuddin, S.; Kumar, V. A Potential Agriculture Waste Material as Coagulant Aid: Cassava Peel. IOP Conf. Ser. Mater. Sci. Eng. 2018, 311, doi:10.1088/1757-899X/311/1/012022.
51. Lapointe, M.; Barbeau, B. Dual starch–polyacrylamide polymer system for improved flocculation. Water Res. 2017, 124, 202–209, doi:10.1016/j.watres.2017.07.044.
52. Magalhães, E.R.B.; Fonseca de Menezes, N.N.; Silva, F.L.; Alves Garrido, J.W.; Angélica dos Santos Bezerra Sousa, M.; Santos, E.S. dos Effect of oil extraction on the composition, structure, and coagulant effect of Moringa oleifera seeds. J. Clean. Prod. 2021, 279, 123902, doi:10.1016/j.jclepro.2020.123902.
53. Nonfodji, O.M.; Fatombi, J.K.; Ahoyo, T.A.; Osseni, S.A.; Aminou, T. Performance of Moringa oleifera seeds protein and Moringa oleifera seeds protein-polyaluminum chloride composite coagulant in removing organic matter and antibiotic resistant bacteria from hospital wastewater. J. Water Process Eng. 2020, 33, doi:10.1016/j.jwpe.2019.101103.
54. Vigneshwaran, S.; Karthikeyan, P.; Sirajudheen, P.; Meenakshi, S. Optimization of sustainable chitosan/Moringa. oleifera as coagulant aid for the treatment of synthetic turbid water—A systemic study. Environ. Chem. Ecotoxicol. 2020, 2, 132–140, doi:10.1016/j.enceco.2020.08.002.
55. Bolto, B.; Gregory, J. Organic polyelectrolytes in water treatment. Water Res. 2007, 41, 2301–2324, doi:10.1016/j.watres.2007.03.012.
56. Dorea, C.C. Use of Moringa spp. seeds for coagulation: A review of a sustainable option. Water Sci. Technol. Water Supply 2006, 6, 219–227, doi:10.2166/ws.2006.027.
57. Jadhav, M.V.; Mahajan, Y.S. Investigation of the performance of chitosan as a coagulant for flocculation of local clay suspensions of different turbidities. KSCE J. Civ. Eng. 2013, 17, 328–334, doi:10.1007/s12205-013-2021-2.
58. Gunaratna, K.R.; Garcia, B.; Andersson, S.; Dalhammar, G. Screening and evaluation of natural coagulants for water treatment. Water Sci. Technol. Water Supply 2007, 7, 19–25, doi:10.2166/ws.2007.147.
59. Antov, M.G.; Šćiban, M.B.; Prodanović, J.M.; Kukić, D.V.; Vasić, V.M.; Đorđević, T.R.; Milošević, M.M. Common oak (Quercus robur) acorn as a source of natural coagulants for water turbidity removal. Ind. Crops Prod. 2018, 117, 340–346, doi:10.1016/j.indcrop.2018.03.022.
60. Idris, J.; Som, A.M.; Musa, M.; Ku Hamid, K.H.; Husen, R.; Muhd Rodhi, M.N. Dragon fruit foliage plant-based coagulant for treatment of concentrated latex effluent: Comparison of treatment with ferric sulfate. J. Chem. 2013, 2013, doi:10.1155/2013/230860.
61. Bakshi, P.S.; Selvakumar, D.; Kadirvelu, K.; Kumar, N.S. Chitosan as an environment friendly biomaterial—A review on recent modifications and applications. Int. J. Biol. Macromol. 2019, doi:10.1016/j.ijbiomac.2019.10.113.
62. Hassan, K.F.; Obeid, S.H. Efficiency of fungi suspension spores as Biocoagulants for suspended solid sedimentation in wastewater. Int. J. Sci. Eng. Res. 2016, 7, 578–585.
63. Pu, S.; Ma, H.; Deng, D.; Xue, S.; Zhu, R.; Zhou, Y.; Xiong, X. Isolation, identification, and characterization of an Aspergillus niger bioflocculant-producing strain using potato starch wastewater as nutrilite and its application. PLoS ONE 2018, 13, e0190236, doi:10.1371/journal.pone.0190236.
64. Nasir, N.M.; Mohd Yunos, F.H.; Wan Jusoh, H.H.; Mohammad, A.; Lam, S.S.; Jusoh, A. Advances in water and wastewater treatment harvesting of Chlorella sp. microalgae using Aspergillus niger as bio-flocculant for aquaculture wastewater treatment. J. Environ. Manag. 2019, 249, doi:10.1016/j.jenvman.2019.109373.
65. Menkiti, M.C.; Ejimofor, M.I. Experimental and artificial neural network application on the optimization of paint effluent (PE) coagulation using novel Achatinoidea shell extract (ASE). J. Water Process Eng. 2016, 10, 172–187, doi:10.1016/j.jwpe.2015.09.010.
66. Oladoja, N.A.; Aliu, Y.D. Snail shell as coagulant aid in the alum precipitation of malachite green from aqua system. J. Hazard. Mater. 2009, 164, 1496–1502, doi:10.1016/j.jhazmat.2008.09.114.

67. Devrimci, H.A.; Yuksel, A.M.; Sanin, F.D. Algal alginate: A potential coagulant for drinking water treatment. Desalination 2012, 299, 16–21, doi:10.1016/j.desal.2012.05.004.
68. Liu, C.; Wang, K.; Jiang, J.H.; Liu, W.J.; Wang, J.Y. A novel bioflocculant produced by a salt-tolerant, alkaliphilic and biofilm-forming strain Bacillus agaradhaerens C9 and its application in harvesting Chlorella minutissima UTEX2341. Biochem. Eng. J. 2015, 93, 166–172, doi:10.1016/j.bej.2014.10.006.
69. Suryani; Ambarsari, L.; Artika, I.; Susanti, H.E. Characterization of Bioflocculant Producing-Bacteria Isolated from Tapioca Waste Water. HAYATI J. Biosci. 2011, 18, 193–196, doi:10.4308/hjb.18.4.193.
70. Keogh, M.B.; Elmusharaf, K.; Borde, P.; Mc Guigan, K.G. Evaluation of the natural coagulant Moringa oleifera as a pretreatment for SODIS in contaminated turbid water. Sol. Energy 2017, 158, 448–454, doi:10.1016/j.solener.2017.10.010.
71. Rasool, M.A.; Tavakoli, B.; Chaibakhsh, N.; Pendashteh, A.R.; Mirroshandel, A.S. Use of a plant-based coagulant in coagulation-ozonation combined treatment of leachate from a waste dumping site. Ecol. Eng. 2016, 90, 431–437, doi:10.1016/j.ecoleng.2016.01.057.