Current Pollution Reports (2021) 7:379–391
https://doi.org/10.1007/s40726-021-00191-7

BIOLOGY AND POLLUTION (R BOOPATHY AND Y HONG, SECTION EDITORS)

Recent Advances on Coagulation-Based Treatment of Wastewater: Transition from Chemical to Natural Coagulant

Muhammad Burhanuddin Bahrodin[1] · **Nur Syamimi Zaidi**[1,2] · **Norelyza Hussein**[1] · **Mika Sillanpää**[3] · **Dedy Dwi Prasetyo**[4] · **Achmad Syafiuddin**[5]

Accepted: 13 May 2021

Abstract

Purpose of Review The use of conventional chemical coagulant in treatment of wastewater is gaining great attention. Drawbacks related to the prolonged effects on human health and environment due to the generation of by-product non-biodegradable sludge are becoming the latest topics. Transition from chemical to natural coagulant can be a good strategy to reduce the aforementioned drawbacks. Therefore, this review aims to provide critical discussions on the use of natural coagulant along with the comparative evaluation over the chemical coagulant.

Recent Findings Treatment performances by chemical and natural coagulant have been reviewed on various types of wastewater with different success rates. Based on this review, a transition from the use of chemical to natural coagulant is highly suggested as the performance of the natural coagulant is comparable to that of the chemical coagulant and in some cases even better. The comparative advantages and disadvantages also convinced that the natural coagulant stands a great chance to be used as an alternative over the chemical coagulant.

Summary Though the current utilization of natural coagulant is encouraging, three main aspects were overlooked by researchers: active coagulant agent, extraction, and optimization due to different wastewater characteristics. Furthermore, delving into these aspects could clarify the uncertainties on the natural coagulant. Hence, it makes this transition a prospect of green technology with sustainable application towards wastewater treatment.

Keywords Coagulation · Chemical coagulant · Natural coagulant · Wastewater treatment

Introduction

The world's freshwater resources are decreasing at an alarming rate. Acquiring access to clean and good water quality is difficult as some under-developed countries have limited or no access, yet good water quality is taken for granted by many others. More than 2 billion people in the world live in under-developed countries with little or no access to clean water, while 4 billion people in developing and developed countries experience extreme water scarcity at least 1 month per year [1]. Since 1980, it was recorded that water use for all human activities worldwide, not limited to industrialization, has been rising by about 1% per year. This trend was predicted to continue at a similar rate until 2050, which is equivalent to a 20 to 30% increment of water use exceeding the current water level due to rising demand in the industrial and domestic sectors [2].

Several treatment technologies are used to process raw water sources into drinking water and transform wastewater into

---

This article is part of the Topical Collection on *Biology and Pollution*

✉ Nur Syamimi Zaidi
nursyamimi@utm.my

✉ Achmad Syafiuddin
achmadsyafiuddin@unusa.ac.id

[1] School of Civil Engineering, Universiti Teknologi Malaysia, 81310 Johor Bahru, Johor, Malaysia

[2] Centre for Environmental Sustainability and Water Security (IPASA), Universiti Teknologi Malaysia, 81310 Johor Bahru, Johor, Malaysia

[3] Institute of Research and Development, Duy Tan University, Da Nang 550000, Vietnam

[4] Department of Statistics, Institut Teknologi Sepuluh Nopember, Surabaya 60111, Indonesia

[5] Department of Public Health, Universitas Nahdlatul Ulama Surabaya, 60237 Surabaya, East Java, Indonesia

treated effluent before it is discharged to water bodies [1, 3]. Most of the treatment processes, whether for water or wastewater, cannot be separated from coagulation and flocculation stages, as part of the treatment processes. The coagulation-flocculation process is a prevalent method in water and wastewater treatment due to its effectiveness in removing organic matter, suspended solids, turbidity, and color [4, 5]. The conventional coagulation process involved the addition of divalent positively charged chemical compounds such as aluminum sulfate and ferric chloride which are known to have various drawbacks from both health and environmental perspectives. In-depth analysis has been conducted concerning the impact that can be caused by the use of chemical compounds as coagulants including high levels of chemical residuals, toxic sludge, and health diseases upon prolonged consumption [6–9].

The transition from chemical to natural coagulants can be an alternative solution to minimize the environmental pollution and health risks caused by the use of chemical coagulants [9, 10]. The use of natural coagulants in water treatment has become imperative due to its biodegradable and environment-friendly nature. Studies related to natural coagulants have undergone many stages until the application to treatment processing units [11, 12]. In the past few years, there have been many reports on natural coagulants obtained from various plant species such as *Moringa oleifera* [13, 14], *Jatropha curcas* [4, 15], banana peels [16–18], and bagasse [14]. These findings achieved the same agreement that natural coagulants pose promising treatment performance, thus standing a great chance in replacing the conventional chemical coagulant.

Therefore, this paper aims to provide critical discussions on the use of various types of natural coagulants, especially in terms of optimum operating conditions and their respective treatment performances, along with comparative evaluation over the chemical coagulant. Besides, this paper provides detailed elaboration on fundamental mechanisms, factors affecting the coagulation process, and drawbacks of chemical coagulants with regard to the health and environmental aspects, which supports the need for transition from chemical to natural coagulant. A full description of the origin, potential sources, and characterization of the natural coagulant, including its unique aspect of being active coagulant agents, is also extensively highlighted. Towards the end, future outlooks for the utilization of natural coagulants in wastewater treatment are summarized as a basis for researchers to further explore research gaps.

Fundamentals of Coagulation Process

Coagulation can be defined as a process that converts stable, unsettled, or slow-settling fine-sized particles into larger sizes by the addition of a coagulant that increases the effective size (flocs) and settling velocities of the particles or the destabilization process [19, 20]. Initially, the colloidal particles in water/wastewater are in a stable phase. With the addition of divalent positive-charged chemical compounds, the colloidal particles are destabilized, thus creating a neutral form of combined compounds. Further addition of coagulant will cause the colloidal particles to restabilize, hence inhibiting agglomeration of the particles. Different coagulants react differently with colloidal particles, with the aim of destabilizing the colloidal particles [21]. The destabilization of colloidal particles can then be categorized into four (4) mechanisms, which are double-layer compression, sweep flocculation, charge neutralization, and interparticle bridging.

Double-Layer Compression

Double-layer compression can be defined as a mechanism that uses ions with counter charge of the colloids to penetrate the double layer surrounding the colloids. The counter ions will alter the properties of the double layer thinner and smaller in volume [22]. Continuous compression from the electrolyte will reduce the repulsive electrostatic repulsion and increase the van der Waals forces which will encourage binding of two destabilized colloids [23, 24]. The flocs formed are bigger due to a higher rate of aggregation, but they have a low degree of sedimentation due to unnecessary friction force between largely formed flocs [25]. In addition, the strength of the flocs is dependent on the ionic charge of the coagulant. Monovalent ions that are weakly charged will produce large but loose flocs that require a longer time to settle. At weakly charged ions, the double layer still strongly charges with a strong repulsive force that reduces the chance of agglomeration [26].

Sweep Flocculation

Sweep flocculation can be identified as a mechanism that removes colloids by enmeshment of colloids in a net-like structure. The net-like structure is made up of precipitation of amorphous metal hydroxide through the hydrolysis process [22, 27]. Analysis such as flocculation index (FI), initial floc aggregation (IFA), and relative settling factor (RSF) showed that flocs formed by sweep flocculation are smaller and have good settling ability but possess a slower rate of floc formation [27]. Floc formed shows a high fractal dimension indicating that the flocs formed are complex [28]. Theoretically, the high fractal dimension will result in stronger flocs and resistance to breakage, but flocs of sweep flocculation are only big and have a higher rate of floc formation and are prone to breakage [22, 29]. It is due to the repulsion force that is still present between flocs. Sweep flocculation only enmeshes colloids in the net-like structure but did not neutralize the repulsion force between colloids which result in formation of weak flocs [28].

Charge Neutralization

Charge neutralization occurs through adsorption between oppositely charged coagulants and the surface of the colloids [24]. Chemical coagulants will undergo a hydrolysis process and produce various cationic species before reacting with colloids [28]. Charge neutralization occurs on the surface of the colloids through a patch-wise manner known as electrostatic patch mechanism. Various cationic species will patch on the surface of the colloids resulting in particle surfaces with positive and negative charges. Colloids' surface with mix charged will reduce the repulsive forces and increase van der Waals forces between particles [30]. Flocs formed by charge neutralization mechanism are stronger compared to those by sweep flocculation but weaker compared to interparticle bridging despite smaller sizes with spherical shapes [27, 31]. In addition, flocs formed also have high fractal dimension; but unlike flocs formed by sweep flocculation, flocs formed by charge neutralization are more compact [28]. This statement was supported by Huang et al. [29] who prove the flocs formed by charge neutralization are stronger based on lower floc strength constant which indicates that flocs formed can resist shear force and not easily broken. It is admitted that flocs by charge neutralization are strong but not too strong due to their reliance on the physical bond which is weaker than chemical bonds [28, 32].

Interparticle Bridging

Interparticle bridging relies on a polymeric chain of polymers that are a long, highly reactive group dangling in the wastewater. One part of the polymeric chain will attach itself to colloids, while the unattached parts of the polymeric chain get attached to other colloidal particles and form a complex structure of colloid-polymer-colloid in which the polymer served for bridging [28]. Several colloid-polymer-colloid structures may become enmeshed and forming readily settleable flocs [20]. A study by Lek et al. [30] shows that the flocs formed as flaky with irregular void space between the network structure as the flocs formed are by polymeric chain attaching to colloids. Meanwhile, the fractal dimension of the floc formed is the lowest which indicates it is not as complex as flocs formed by other mechanisms [28]. Theoretically, a low fractal dimension will result in weak flocs that are prone to breakage, but they are very strong and not easily broken into smaller clusters due to polymers that serve as bridges that are strong enough and have formed enough chemical bonds among flocs [32]. A study carried out by Choy et al. [11] shows that the use of natural coagulant with interparticle bridging mechanism enhances floc growth by at least three times compared to the use of a chemical coagulant due to the ability of polymeric chains to stretch and attach to as many colloids as possible.

Factors Affecting Coagulation Process

Identifying the optimum condition of a coagulation process is crucial as it fully utilizes the added coagulant for the removal of most pollutants. Different coagulants have different optimum conditions. Understanding the interaction between coagulant and pollutant is important in identifying the maximum efficiency of the coagulant besides minimizing operational cost and sludge volume. Several factors affecting the coagulation processes in water and wastewater treatment include the types of coagulants, the coagulant dosage, the mixing processes, and the characteristics of the water/wastewater to be treated.

Coagulant Dosage

The effect of coagulant dosage against the removal of pollutants can be analyzed through three different conditions, namely underdosage, optimum dosage, and overdosage. Underdosage can be defined as insufficient coagulant dosage to adhere to the existing pollutant in the wastewater and require additional coagulants to achieve optimum condition [23]. Addition of more coagulants to the wastewater will provide more active coagulant sites to attract and absorb pollutants.

In addition, coagulants added to the wastewater will further neutralize the electrical load until it reaches zero zeta potential, which allows attraction and absorption of pollutants to coagulants [33]. However, adding more coagulants exceeding the optimum dosage will only pollute the wastewater. The excess coagulant will saturate the surface of colloids. The saturated coagulant will cause particle restabilization, which eventually creates the repulsive force among pollutants and thus hinder floc formation.

pH

Chemical coagulants added into water will go through the hydrolysis process where they will be broken down into various hydrolysis products. These hydrolysis products are responsible for successful pollutant removal. Different pH will produce different hydrolysis products, and different products are effective for a different mechanism. For example, alum in acidic conditions will be dissolved to form an aqueous aluminum ion (Al^{3+}) that is effective in double-layer compression and charge neutralization. Meanwhile, alum in alkali conditions will be dissolved into aluminate or amorphous hydroxide $(Al(OH)_4)^-$ that forms into a net-like structure that is effective in sweep flocculation [34].

Meanwhile, the effect of pH in natural coagulants depends on the active coagulant agent and its mechanism. If the active coagulant agent is protein such as *Moringa oleifera* seed and its mechanism is charge neutralization, the effect of pH is more prominent. Proteins are made up of a chain of amino acids. Amino acids can be classified as polar and non-polar. Amino acids such as lysine and arginine are positively charged, while an amino acid such as aspartic acid is negatively charged [35]. Different pH and isoelectric points (pI) will decide which amino acids will dominate. Optimum pH less than the recorded pI resulted in domination of positively charged amino acids, while pH higher than pI resulted in domination of negatively charged amino acids [4].

Initial Turbidity

The coagulation process relies on the reaction among pollutants with a coagulant for floc formation. Coagulants will collide with pollutants and agglomerate to form flocs. High initial turbidity will have more pollutant molecules. It will enhance the number of collisions between a coagulant and a pollutant. More collisions will produce larger and sturdier flocs which will result in faster settling [12, 36]. Meanwhile, in low initial turbidity, fewer collisions between coagulant and pollutant occur due to an insufficient number of pollutants. Smaller floc formation will slow down the sedimentation rate, thus increasing the sedimentation time [37].

In addition, low initial turbidity in the coagulation-flocculation process will form a flake-like structure with low sedimentation capability. The flake-like structure will cause the sedimentation time to increase and disturb the coagulation-flocculation process. The optimum sedimentation time will be disturbed by the flake-like structure [38].

Previous studies have proved the relationship between different initial turbidities with the coagulation efficiency. A study conducted by Rajput et al. [39] shows turbidity removal is better at higher initial turbidity. They found that for low (20 NTU), medium (45 NTU), and high (100 NTU) initial turbidities, the turbidity removals are 87%, 67%, and 58%, respectively. However, a study by Abidin et al. [35] shows that increasing the initial turbidity to higher than optimum initial turbidity will reduce coagulation performance. Optimum initial turbidity (500 NTU) shows turbidity removal of 99.5%, but high initial turbidity (8000 NTU) reduces turbidity removal to 98.4%.

Rapid Mixing

Rapid mixing is one of the important steps in the coagulation process. The main objective of rapid mixing is to evenly distribute coagulants in the wastewater. However, identifying the optimum time and speed is important to ensure homogeneous dispersion of coagulants and prevention of floc shear and tear [30]. Studies conducted by Ramphal and Sibiya [40] and Ding et al. [41] showed the optimum speed of rapid mix to be in a range of 40 to 200 rpm, with the most desirable mixing speed of 120 rpm. A too slow mixing speed (below 80 rpm) fails to homogeneously disperse coagulants in the wastewater. In addition, flocs formed from the coagulation-flocculation process are fragile and easily broken. Broken flocs will reduce the removal efficiency and increase the pollutant concentration in the wastewater. Meanwhile, a too fast mixing speed (above 160 rpm) is also not desirable as the action increases the shearing and tearing of flocs formed. Tear and shear of flocs will restabilize the pollutant and increase the pollutant concentration in the wastewater [42]. Besides that, the flocs formed from the too fast rapid mix will produce smaller, denser, and less porous flocs [40]. Although the flocs formed are small, they are dense and thus do not allow more pollutant adsorption, with the result that flocs take a longer time to settle. In addition, the production of smaller flocs is caused by increasing zeta potential as the mixing speed increases, causing lesser agglomeration between pollutants and coagulants [40, 41].

According to Kan et al. [43], for a successful coagulation process, rapid mixing time should be kept between times required for destabilizing pollutants and identification of optimum coagulant dosage to reduce the dosage but cannot exceed the time required for floc formation. This indicates that the extended rapid mixing time is not favorable. During the rapid mix, the treated wastewater was exposed to fluid shear rates. Increasing the rapid mixing time will expose the treated wastewater to fluid shear rates for a longer time, which will lead to floc breakage. In addition, flocs formed will collide with other flocs, and thus the rate of floc breakup will increase besides stunting the process of floc growth [40].

Temperature

Temperature is not a crucial parameter that will affect the coagulation efficiency as much as other factors. According to Katayon et al. [44], there was no change in pollutant removal, whether the wastewater sample was kept in the refrigerator or left at room temperature. However, this finding was contradicted by studies by Betran-Heredia et al. [45] and Mataka et al. [46]. Based on these studies, there were changes in the removal upon the change in temperature of wastewater due to the exothermic reaction between active sites and pollutants.

Generally, it is more preferable for the coagulation process to be carried out in a warmer environment. The coagulation process involves agglomeration between coagulants and pollutants. In warmer conditions, the particles will move faster and collisions between the coagulant and pollutant will be more frequent for bigger and sturdier floc formation [47]. Chilled water is not preferable for the coagulation process because it will decrease the coagulant solubility, increase water viscosity, and retard the kinetic energy for particle flocculation. Therefore, the coagulation process will require a higher coagulant dosage and extend flocculation time to achieve optimum pollutant removal. In return, the originally lower cost treatment process will incur more cost and will increase the total treatment cost [48]. In addition, an increase in water viscosity will require a coagulation process with higher shear stress (increase mixing speed). Higher shear stress will exert more shearing and tearing force to the originally formed flocs causing pollutants to redisperse into the wastewater [49].

Factors such as coagulant dosage, pH, and mixing speed were studied in many pieces of research [50]. However, a lack of literature has focused on external factors such as temperature. Some researchers consider that the effect of temperature is less significant, but based on previous studies, there were improvements in pollutant removal with increasing or decreasing the temperature [51, 52]. Different studies have reached different conclusions on the importance of temperature to the coagulation process. Therefore, more studies are required to identify factors that affect this outcome.

Types of Coagulants

Conventional coagulation process relies on the use of chemical coagulants. These include metal salt coagulants, activated silica, synthetic polymer, and natural coagulant. From these coagulants, metal salt coagulants are commonly used in water and wastewater treatment plants. The advantages of metal salt coagulants over other coagulants are that they have been proven to be very efficient in the removal of various pollutants such as heavy metals, turbidity, chemical oxygen demand (COD), and biological oxygen demand (BOD). Metal salt coagulants are also available commercially and are capable of inactivating bacteria [53].

Metal salt coagulants are widely used due to their availability and reasonable price compared to other chemical coagulants. Among the commonly used metal salt coagulants are aluminum-based and iron-based coagulants. Aluminum sulfate (alum) and ferric chloride are the most conventional chemical coagulants used in treating water and wastewater. Upon addition of the coagulant into the water, it will react through the hydrolysis process and its products are responsible for pollutant's removal. By comparing iron-based coagulants with aluminum-based coagulants, both coagulants show good removal of pollutants. The merit of using an iron-based coagulant over aluminum-based coagulant is that an iron-based coagulant is cheaper, has a broader pH range, and is less sensitive to overdosage. Correspondingly, the flocs formed by using iron-based coagulants are tougher and denser which make them prone to breakage [54].

Transition from Chemical Coagulant to Natural Coagulant

Transition from chemical to natural coagulant can be an alternative solution to minimize the environmental pollution and health risks, at the same time promoting green technology in water and wastewater treatment application. Natural coagulants can be extracted from natural sources from either plants or animals. A natural coagulant from plant sources is not new as it was first discovered over decades ago.

A natural coagulant was first applied for water treatment way before chemical coagulants were discovered. Over the years, the use of natural coagulants was recorded in various countries, while chemical coagulants were continuously being researched. In the twentieth century, the development of chemical coagulants was at its peak as researchers had finally identified the mechanisms that were responsible for better coagulation process. Despite its greatest interest in the treatment process, various disadvantages of the chemical coagulants were also discovered and evidenced. Alternatively, the interest to explore the feasibility of natural coagulants as a viable application to replace chemical coagulants in treating water and wastewater gradually increased until today.

Natural coagulants can be obtained from natural sources from either plant or animal waste. A lot of studies have been carried out on various sources that can potentially be used as comprehensive natural coagulants. Waste from plant-based sources is mostly studied as natural coagulants. Every part of plants from stem to leaf has been tested for suitability in becoming a potential natural coagulant. Among others, *Moringa oleifera* is the most established natural coagulant. Its utilization has been proven in treating various types of polluted water [55–57] and wastewater such as dairy wastewater [58]. Without prior extraction and purification, most of the natural coagulants derived from plant-based waste were still capable of obtaining about 60–98% turbidity removal, 55–89% organic content removal, and 80–91% fecal coliform removal [55, 59, 60]. Additionally, the use of natural coagulant as a coagulant aid to alum was able to reduce the optimum dosage of alum itself by 40% [61].

Natural coagulants from animal waste such as shells and bones were least studied. Out of many advantages, one of the problems with regard to the natural coagulant is its ability to constantly being produced for large-scale treatment processes. Natural coagulants from animal sources are least studied due to their ability of providing continuous supply as abundant sources [62]. In the year 2018, the global production of fruits was around 868 million metric tons and has steadily increased since the year 1960 [63]. Based on this pattern, it is expected that there will be more production of various fruits in the future which eventually will increase the production of fruit waste. Therefore, the utilization of plant-based waste from various fruits can be potentially used as a suitable source of natural coagulant with continuous supply in the future.

The active coagulant agents are compounds that play a vital role in successful coagulation activity by natural coagulants. Different sources of natural coagulant either from plant-based or from animal-based waste comprise different active coagulant agents. According to Jagaba et al. [54], the active coagulant agent can be classified into protein polymers, polysaccharides, and some functional groups such as hydroxyl and carboxyl groups. In addition, a study by Camacho et al. [13] reported otherwise, with the main active coagulant agent in natural coagulants being phytochemicals such as phenolics and phytic acids. Main active coagulant agents that were commonly extracted from the natural coagulants are observed to be protein polymer, carbohydrate, polysaccharides, and phenolic compounds [64–67]. With regard to the mechanism of coagulation by the act of natural coagulants, proteins, polysaccharides, and some functional groups promote the mechanisms of adsorption, polymer bridging, and charge neutralization [68].

Comparative Evaluation on Performance of Chemical Coagulant and Natural Coagulant in Wastewater Treatment

A chemical coagulant as a conventional coagulant poses many disadvantages that affect both environment and health upon prolonged aftereffects. Substituting chemical coagulants with natural coagulants as safer and environment-friendly alternatives attracted the attention of researchers for the past few years. *Moringa oleifera* is among the natural coagulants that have been studied by many researchers. Its seeds, leaves, and stems can be processed into natural coagulants, with its seeds showing similar or better performance compared to chemical coagulants [69]. A natural coagulant relies on the polymeric chain that is affected by molecular weight and charge density for better removal. Sources with higher molecular weight will have a longer polymeric chain that will attach to the pollutants, while higher charge density will encourage extension of the polymeric chain [11, 70]. Based on previous studies, natural coagulants can remove pollutants as good as chemical coagulants. Table 1 shows the comparison of water and wastewater treatment using chemical and natural coagulants.

Based on Table 1, in most studies, natural coagulants managed to achieve removal efficiency as good as the chemical coagulant. However, most of the findings indicated slightly higher removal efficiency performed by chemical coagulants compared to that by the natural coagulant. It is worthy to note that the performance by natural coagulants can be further increased by optimizing the extraction and purification to attain the respective active coagulant agent. Natural coagulants such as *Moringa oleifera* seeds rely on protein polymers, while *Jatropha curcas* relies on polysaccharide compounds as the active coagulant agent [36, 38]. With an appropriate extraction process, *Moringa oleifera* seeds and *Jatropha curcas* can perform way better in the coagulation process, thus resulting in higher removal efficiency. Still, without/lack of optimized or appropriate extraction, *Moringa oleifera* seeds and *Jatropha curcas* managed to obtain more than 85% and 70% removal efficiency, respectively, mainly for the parameters turbidity and COD [71, 81].

A significant comparison was also observed for the removal of bacteria in polluted river water. Using the *Moringa oleifera* seeds, the coliform bacteria and mesophilic bacteria were removed by averages of 81.7% and 96.0%, respectively, higher compared to the 40.0% and 32.9%, by addition of alum [12]. This observation suggested that the natural coagulant shows potential in having antibacterial properties that are capable of disinfecting the water upon treatment. Besides, comparative observations as summarized in Table 1 also suggest that the natural coagulant is feasible to be applied along with chemical coagulant as an aid. The use of alum and banana peels independently resulted in turbidity removals of 73.1% and 65.6%, respectively. Both types of coagulants, either chemical or natural coagulants, failed to achieve more than 80% turbidity removal. However, the combination of both, where banana peels acted as coagulant aid along with alum, had successfully resulted in higher turbidity removal of 94.1% [80].

In terms of sludge production, it was evident that the natural coagulant is capable of producing less sludge compared to the chemical coagulant. A study by Tuddao and Gonzales [86] indicated that the use of *Jatropha curcas* seeds generated about 40 mL/L sludge volume compared to 58 mL/L when using alum. Additionally, the sedimentation time was shorter when using *Jatropha curcas* seeds compared to the alum. This observation highlights the primary advantages of a natural coagulant which is biodegradable.

Other than that, the advantages of natural coagulant over chemical coagulant can also be seen in the form of treatment capacity and environmental friendliness. Jagaba et al. [54] had evaluated the performance of three coagulants, namely alum as the representative of the chemical coagulant, with *Moringa oleifera* and chitosan as the representatives of natural coagulants. With regard to the treatment capacity, chitosan showed the highest removal performance, while *Moringa oleifera* and alum showed comparable removal performance. Chitosan managed to remove 95% color, 95% oil and grease, 98% TSS, and 91% NH_3-N at the optimum coagulant dosage of 500 mg/L. *Moringa oleifera* managed to remove 93% color, 87% oil and grease, 95% TSS, and 90% NH_3-N at optimum coagulant dosage of 2000 mg/L, while alum removed 93% color, 95% oil and grease, 97% TSS, and 95% NH_3-N at optimum coagulant dosage of 4000 mg/L. Although all coagulants showed remarkable high treatment performance, the amount of dosage used plays a crucial role. It is evident that low dosage is required when using the natural coagulant compared to the chemical coagulant. As for *Moringa oleifera*, the dosage used can still be reduced when the active agent is extracted. Meanwhile, with regard to environmental friendliness, toxicity becomes an important benchmark. Awolola

Table 1 Comparison performance of natural coagulant and chemical coagulant in wastewater treatment

Type of wastewater	Chemical coagulant	Removal performance	Natural coagulant	Removal performance	Ref.
Paper mill industry	Alum	Turbidity: 97.1% COD: 92.7%	*Moringa oleifera* seed	Turbidity: 96.0% COD: 97.3%	Boulaadjoul et al. [71]
Concrete plant	Alum and ferric chloride	Turbidity: 99.9%		Turbidity: 99.9%	De Paula et al. [72]
River water	Alum	Turbidity: 75% Coliform bacteria: 40% Mesophilic bacteria: 32.9%		Turbidity: 62.5% Coliform bacteria: 70.0–93.3% Mesophilic bacteria: 93.7–98.3 %	Alo et al. [73]
POME	Alum	TSS: 99.7% Turbidity: 98.7% NH_4-N: 98.5% Color: 94% Oil and grease: 95%		TSS: 95.4% Turbidity: 88.3% NH_4-N: 89.8% Color: 90.2% Oil and grease: 87.1%	Jagaba et al. [54]
	Ferric chloride	TSS: 99.1% Turbidity: 95.8% NH_4-N: 93.7% Color: 66.4% Oil and grease: 95.7%			
Paint industry	Ferric chloride	Color: 89.4% COD: 83.4% Turbidity: 88.5%	Cactus	Color: 88.4% COD: 78.2% Turbidity: 82.6%	Vishali and Karthikeyan [74]
Confectionary	PAM	TSS: 93.5% COD: 95.9%		TSS: 92.2% COD: 95.6%	Sellami et al. [75]
Glue	PAM	TSS: 90.7% COD: 86.3%		TSS: 90.3% COD: 82.1%	
Paper and mill	PEI and HE	Color: 80% TOC: 30%	Chitosan	Color: 90% TOC: 70%	Ganjidoust et al. [76]
	Alum	Color: 80% TOC: 40%			
POME	Alum	TSS: 99.7% Turbidity: 98.7% NH_4-N: 98.5% COD: 75% Color: 94% Oil and grease: 95%	Chitosan	TSS: 99% Turbidity: 98.4% NH_4-N: 95.6% COD: 68.3% Color: 96% Oil and grease: 94.9%	Jagaba et al. [54]
POME	Alum	Turbidity: 82.2% COD: 49.1%	Rice starch	Turbidity: 92.5% COD: 30.9%	Teh et al. [77]
Leachate	PACl	TSS: 78% Turbidity: 76% Color: 70% COD: 65% NH_4-N: 25%	*Diplazium esculentum* leaf as coagulant aid	TSS: 88% Turbidity: 87% Color: 76% COD: 68% NH_4-N: 34%	Zainol et al. [78]
Leachate	Alum	Color: 69% Iron: 60% TSS: 45% Turbidity: 36% NH_4-N: 25%	*Hibiscus rosa* leaf as coagulant aid	Color: 61% Iron: 100% TSS: 72% Turbidity: 60% NH_4-N: 54%	Awang and Aziz [79]
Synthetic turbid water	Alum	Turbidity: 73.1%	Banana peel Banana peel as coagulant aid	Turbidity: 65.6% Turbidity: 94.1%	Kian-Hen and Peck-Loo [80]
POME	Alum	COD: 59% BOD: 61% TSS: 71%	*Jatropha curcas* seed	COD: 70% BOD: 65% TSS: 88%	Abidin et al. [81]
Latex effluent	Ferric sulfate	COD: 98% TSS: 98% Turbidity: 89%	Dragon fruit foliage	COD: 94.7% TSS: 85.9% Turbidity: 99.7%	Idris et al. [82]
Artificial turbid water	Alum	Turbidity: 97.9%	Mango seed	Turbidity: 92%	Seghosime et al. [61]
Dam water	Alum	Turbidity: 98.5%	Watermelon seed	Turbidity: 89.3%	Muhammad et al. [83]

Table 1 (continued)

Type of wastewater	Chemical coagulant	Removal performance	Natural coagulant	Removal performance	Ref.
		Color: 98.5%		Color: 93.9%	
			Watermelon seed as coagulant aid	Turbidity: 99.3% Color: 100%	
Cassava effluent	Alum	Color: 49% Turbidity: 56%	*Acacia negara*	Color: 81.5% Turbidity: 91%	Dos Santos et al. [84]
Synthetic wastewater	Alum	Turbidity: 99.6% Sedimentation time: 40 min Sludge volume: 58 mL/L	*Jatropha curcas* seed	Turbidity: 99.6% Sedimentation time: 20 min Sludge volume: 40 mL/L	Abidin et al. [35]
Paint effluent	Ferric chloride	COD: 82% Color: 89% Sludge volume: 172 mL/L	Commercial NC based on tannin	COD: 87% Color: 99% Sludge volume: 116 mL/L	Aboulhassan et al. [85]
	Alum	COD: 81% Color: 89% Sludge volume: 220 mL/L			

et al. [87] had tested the toxicity (LC_{50}) on *Carica papaya* seeds (natural coagulant) and $CaSO_4$ (chemical coagulant). A high value of LC_{50} indicates that the coagulant is less toxic. The results indicated that the *Carica papaya* seed is significantly less toxic with LC_{50} of 196.49 μg/mL compared to the $CaSO_4$ with LC_{50} of 7.64 μg/mL. The findings also reported that the use of chemical coagulants as such is extremely toxic not only to the generated sludge but also to the effluent.

Nonetheless, the comparative observations also show that the removal performances by natural coagulants are quite inconsistent. These inconsistencies might be due to the excess organic or inorganic content retained in the natural coagulant. For that, the purification process plays a greater role to ensure the residues are removed, thus allowing efficient extraction of active coagulant agents to take place.

Comparative Evaluation on Drawbacks Between Chemical Coagulant and Natural Coagulant

Undeniably, both coagulants show good removal of various pollutants for water and wastewater treatment with almost similar removal efficiency. However, like any other treatment alternatives, the disadvantages are unavoidable. Figure 1 provides a comparative evaluation of the drawbacks between chemical and natural coagulants [51, 88–90].

Based on Fig. 1, the drawbacks of a chemical coagulant are more serious and long-lasting as it mostly affects the environment in terms of high and toxic sludge generation, and affects consumers' health due to the prolonged consumption of water containing chemical residuals that can potentially cause neurodegenerative disease. On the other hand, the drawbacks that arise upon the use of natural coagulants rely mostly on the coagulant itself such as the impact of seasons and storage duration, which in turn can affect the production and continuous supply of the natural coagulant. However, many pieces of research to date have been conducted which aim to reduce the impacts from drawbacks of the natural coagulant. Current research that was conducted on a mixture of plant waste such as banana and pomegranate peels proved suitability of mixing different raw material sources, thus providing insight on exploring other possible sources in little quantity to be used as natural coagulants [91].

Other drawbacks of natural coagulants that may affect the environment are due to the release of organic content into the treated effluent, which in turn increased the COD, BOD, and TOC concentrations. However, unlike the environmental drawbacks of chemical coagulants, the increased organic content upon the use of natural coagulants can be reduced by associating extraction and purification processes such as acid and alkali extraction.

Future Outlooks

The transition applications of chemical substances towards natural resources have drawn much attention from researchers worldwide [92–94]. Researchers started to have a deep interest in natural resources due to their immense advantages; for example, natural resources are relatively cheaper compared to chemical substances and produce less sludge volume [95–99]. Worldwide, countries are now exploring various raw material sources either from plants or from animals to be developed as a

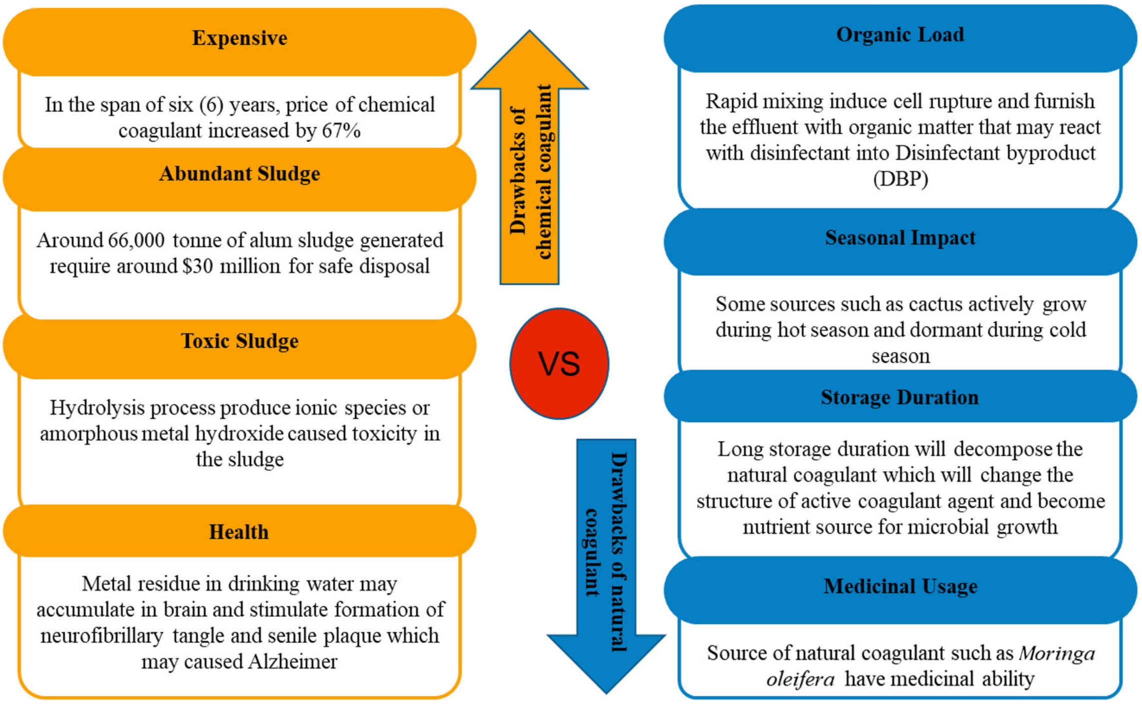

Fig. 1 Comparison on drawbacks between chemical and natural coagulants

natural coagulant. Countries such as Malaysia, Brazil, and India documented a lot of researches related to the natural coagulant. However, the findings are mainly focused on the commonly known and established plants such as *Moringa oleifera*, *Jatropha curcas*, and *Opuntia ficus indica*. Although these sources have proven their ability in removing various pollutants, it is crucial to explore the possibility and feasibility of other local raw material sources as well. Other local resources such as waste from coconut, cassava peels, bagasse (sugarcane), papaya for both peels and seeds, and banana remain unexplored. According to Zakari and Abdul Rahim [100], in Malaysia alone, the supplies of local fruits such as coconut and banana in the year 2011 are around 627,104 MT and 306,923 MT, respectively. These trees, in fact, produce among the largest percentage of fruit waste—80% (from banana tress) and 50% (from coconut tree) —which eventually adds to more waste production in landfill [101, 102]. Therefore, exploring the use of waste from varieties of other local resources may contribute towards improvement in water and wastewater treatment by means of extending the list of natural resources that are locally accessible [103–108].

The extraction and purification process can improve the treatment performance of natural coagulants by extracting the active coagulant agent compounds. Solvents such as salt, alcohol, acid, and base can be used for the extraction process, and methods such as precipitation and lyophilization can be used for the purification process. However, selection on a suitable solvent and method is important as a different active coagulant agent reacts differently. In addition, the purification process is not preferable as it will increase the total treatment cost which did not suit the purpose of using the natural coagulant. Therefore, providing a simple and reliable method for extracting the active coagulant agent from raw materials of natural coagulants could be the solution to overcome the complex extraction of natural coagulants. Many pathways of research into minimizing and simplifying the extraction processes can be carried out depending on the chosen raw materials (plant-based or animal-based). Optimization of operating coagulation conditions is crucial as it can maximize the efficiency of natural coagulants. To date, optimization findings with regard to the potential local sources of natural coagulants are lacking. Therefore, determining the optimum conditions for coagulation processes using the natural coagulant will be relevant to conduct in the future. Additionally, a thorough analysis of the utilization of bacteria-based coagulants and their impact on the treated water is also needed to be conducted. This is due to the many observations reported that the plant-based natural coagulant can remove coliform bacteria successfully. Apparently, research on this matter is still limited. Hence, further research on the use of bacteria-based coagulants is needed.

Lastly, an analysis on the suitability of natural coagulants to treat certain characteristics of water and wastewater will be useful in minimizing trial-and-error processes, given the many characteristics of wastewater. To date, natural coagulants have been used on various wastewater such as textile, industrial, domestic, leachate, river water for drinking water purposes, and groundwater. However, other types of wastewater such as pharmaceuticals and palm oil mill effluent (POME) are less likely to be explored. Compatibility of natural coagulants with these unexplored wastewaters can serve a better understanding on the suitability and ability of the natural coagulant to remove high-strength pollutants.

Conclusion

Several studies related to the application of natural coagulants are currently being conducted at the laboratory scale. The utilization of natural coagulant is a promising technology in water and wastewater treatment due to its environmentally friendly and its reliable treatment performance, which is comparable with that of the chemical coagulant. Even so, several factors have to be considered upon utilization of natural coagulants mainly due to their complex extraction and purification process, limited availability of the raw materials for continuous supply, and various characteristics of water and wastewater to be treated. The limitations of utilizing the natural coagulant open up new challenges for future study. Future researches including simplification on the feasible extraction and purification methods, characterization on the potential local resources (plant-based and/or animal-based) to be used as a natural coagulant, optimization of the operating conditions as well as extraction process, and evaluation on other types of wastewater with different characteristics are then worthy to be explored. To sum up, by considering of all the advantages, the development and utilization of natural coagulants have good prospects as a green technology with a viable sustainable application as water pollution control.

Acknowledgements The authors thank the Universitas Nahdlatul Ulama Surabaya in realizing the current work.

Funding The authors received financial support from the Ministry of Higher Education (MOHE) Malaysia (FRGS/1/2019/TK01/UTM/02/11) and Universiti Teknologi Malaysia (UTM) (Q.J130000.2651.16J76).

Compliance with Ethical Standards

Conflict of Interest The authors declare that they have no conflict of interest.

Human and Animal Rights and Informed Consent This article does not contain any studies with human or animal subjects performed by any of the authors.

References

1. UNESCO. The United Nations World Water Development Report 2019: leaving no one behind. In: UNESCO Digital Library. 2019.
2. UNESCO. The United Nation World Water Development Report 2018: transforming our world: the 2030 agenda for sustainable development. In: UNESCO Digital Library. 2018.
3. Kumar V, Othman N, Asharuddin S. Applications of natural coagulants to treat wastewater − a review. MATEC Web Conf. 2017;103:06016.
4. Abidin ZZ, Mohd Shamsudin NS, Madehi N, Sobri S. Optimisation of a method to extract the active coagulant agent from Jatropha curcas seeds for use in turbidity removal. Ind Crop Prod. 2013;41(1):319–23.
5. Kakoi B, Kaluli JW, Ndiba P, Thiong'o G. Banana pith as a natural coagulant for polluted river water. Ecol Eng. 2016;95:699–705.
6. Vatvani C. The toxic waste that enters Indonesia's Citarum River, one of the world's most polluted. In: Channel news Asia. 2018. https://www.channelnewsasia.com/news/asia/indonesia-citarum-river-worlds-most-polluted-toxic-waste-.
7. EPA. The Passaic's River, Polluted Past. United States Environmental Protection Agency. In: www.ourpassaic.org. 2014.
8. De Pippo T, Donadio C, Guida M, Petrosino C. The case of Sarno River (Southern Italy): effects of geomorphology on the environmental impacts. Environ Sci Pollut Res. 2006;13(3):184–91.
9. Montuori P, Lama P, Aurino S, Naviglio D, Triassi M. Metals loads into the Mediterranean Sea: estimate of Sarno River inputs and ecological risk. Ecotoxicology. 2013;22(2):295–307.
10. Septiono MA, Roosmini D, Salami IRS, Ariesyadi HD, Lufiandi. Industrial activities and its effects to river water quality (case study Citarum, Bengawan Solo and Brantas), an evaluation for Java Island as an economic corridor in master plan of acceleration and expansion of Indonesia economic development (Mp3Ei) 2011. 12th Int Symp Southeast Asian Water Environ. 2016;(November).
11. Choy SY, Prasad KN, Wu TY, Raghunandan ME, Ramanan RN. Performance of conventional starches as natural coagulants for turbidity removal. Ecol Eng. 2016;94:352–64.
12. Amran AH, Zaidi NS, Muda K, Loan WL. Effectiveness of natural coagulant in coagulation process: a review. Int J Eng Technol. 2018;7(3.9):34.
13. Camacho FP, Sousa VS, Bergamasco R, Ribau TM. The use of Moringa oleifera as a natural coagulant in surface water treatment. Chem Eng J. 2017;313:226–37.
14. Chitra D, Muruganandam L. Performance of natural coagulants on greywater treatment. Recent Innov Chem Eng. (Formerly Recent Patents Chem. Eng.). 2019;13:81–92.
15. Sibartie S, Ismail N. Potential of Hibiscus sabdariffa and Jatropha curcas as natural coagulants in the treatment of pharmaceutical wastewater. MATEC Web Conf. 2018;152.
16. Maurya DP, Singla A, Negi S. An overview of key pretreatment processes for biological conversion of lignocellulosic biomass to bioethanol. 3 Biotech. 2015;5(5):597–609.
17. Zaidi NS, Muda K, Loan LW, Sgawi MS, Abdul Rahman MA. Potential of fruit peels in becoming natural coagulant for water treatment. Int J Integr Eng. 2019a;11:140–50.
18. Zaidi NS, Muda K, Abdul Rahman MA, Sgawi MS, Amran AH. Effectiveness of local waste materials as organic-based coagulant in treating water. IOP Conf Ser Mater Sci Eng. 2019;636(1).
19. Choy SY, Prasad KMN, Wu TY, Raghunandan ME, Ramanan RN. Utilization of plant-based natural coagulants as future alternatives towards sustainable water clarification. J Environ Sci (China). 2014;26(11):2178–89.
20. Peavy HS, Rowe DR, Tchobanoglous G. Environmental engineering. 1985.
21. Delphos PJ, Wesner GM. Mixing, coagulation, and flocculation (ch. 6), Water treatment plant design. 4th ed. McGraw-Hill; 2005.
22. Ghernaout D, Ghernaout B. Sweep flocculation as a second form of charge neutralisation-a review. Desalin Water Treat. 2012;44(1–3):15–28.
23. Choy SY, Prasad KMN, Wu TY, Ramanan RN. A review on common vegetables and legumes as promising plant-based natural coagulants in water clarification. Int J Environ Sci Technol. 2015;12(1):367–90.

24. Miller SM, Fugate EJ, Craver VO, Smith JA, Zimmerman JB. Towards understanding the efficacy and mechanism of Opuntia spp. as a natural coagulant for potential application in water treatment. Environ Sci Technol. 2008;42:4274–9.
25. Szilagyi I, Polomska A, Citherlet D, Sadeghpour A, Borkovec M. Charging and aggregation of negatively charged colloidal latex particles in the presence of multivalent oligoamine cations. J Colloid Interface Sci. 2013;392(1):34–41.
26. Xia X, Lan S, Li X, Xie Y, Liang Y, Yan P, et al. Characterization and coagulation-flocculation performance of a composite flocculant in high-turbidity drinking water treatment. Chemosphere. 2018;206:701–8.
27. Jiao R, Fabris R, Chow CWK, Drikas M, van Leeuwen J, Wang D, et al. Influence of coagulation mechanisms and floc formation on filterability. J Environ Sci (China). 2017;57:338–45.
28. Li T, Zhu Z, Wang D, Yao C, Tang H. Characterization of floc size, strength and structure under various coagulation mechanisms. Powder Technol. 2006;168(2):104–10.
29. Huang C, Lin JL, Lee WS, Pan JR, Zhao B. Effect of coagulation mechanism on membrane permeability in coagulation-assisted microfiltration for spent filter backwash water recycling. Colloids Surf A Physicochem Eng Asp. 2011;378(1–3):72–8.
30. Choong Lek BL, Peter AP, Qi Chong KH, Ragu P, Sethu V, Selvarajoo A, et al. Treatment of palm oil mill effluent (POME) using chickpea (Cicer arietinum) as a natural coagulant and flocculant: evaluation, process optimization and characterization of chickpea powder. J Environ Chem Eng. 2018;6(5):6243–55.
31. Ma C, Hu W, Pei H, Xu H, Pei R. Enhancing integrated removal of Microcystis aeruginosa and adsorption of microcystins using chitosan-aluminum chloride combined coagulants: effect of chemical dosing orders and coagulation mechanisms. Colloids Surf A Physicochem Eng Asp. 2016;490:258–67.
32. Wang B, Shui Y, He M, Liu P. Comparison of flocs characteristics using before and after composite coagulants under different coagulation mechanisms. Biochem Eng J. 2017;121:107–17.
33. Momeni MM, Kahforoushan D, Abbasi F, Ghanbarian S. Using Chitosan/CHPATC as coagulant to remove color and turbidity of industrial wastewater: optimization through RSM design. J Environ Manag. 2018;211:347–55.
34. Metcalf W, Eddy C. Metcalf and eddy wastewater engineering: treatment and reuse. New York: Wastewater Eng Treat Reuse McGraw Hill; 2003.
35. Abidin ZZ, Ismail N, Yunus R, Ahamad IS, Idris A. A preliminary study on Jatropha curcas as coagulant in wastewater treatment. Environ Technol. 2011;32(9):971–7.
36. Choudhary M, Ray MB, Neogi S. Evaluation of the potential application of cactus (Opuntia ficus-indica) as a bio-coagulant for pre-treatment of oil sands process-affected water. Sep Purif Technol. 2019;209(July 2018):714–24.
37. Odiyo JO, Bassey OJ, Ochieng A, Chimuka L. Coagulation efficiency of Dicerocaryum eriocarpum (DE) plant. Water SA. 2017;43(1):1–6.
38. Dotto J, Fagundes-Klen MR, Veit MT, Palácio SM, Bergamasco R. Performance of different coagulants in the coagulation/flocculation process of textile wastewater. J Clean Prod. 2019;208:656–65.
39. Rajput SK, Bapat KN, Choubey S. Bioremediation - natural way for water treatment. An Int J Life Sci Chem. 2012;29(2):88–99.
40. Ramphal S, Muzi SS. Optimization of time requirement for rapid mixing during coagulation using a photometric dispersion analyzer. Procedia Eng. 2014;70:1401–10.
41. Ding Y, Zhao J, Wei L, Li W, Chi Y. Effects of mixing conditions on floc properties in magnesium hydroxide continuous coagulation process. Appl Sci. 2019;9(5).
42. Ernest E, Onyeka O, David N, Blessing O. Effects of pH, dosage, temperature and mixing speed on the efficiency of water melon seed in removing the turbidity and colour of Atabong River, Awka-Ibom State, Nigeria. Int J Adv Eng Manag Sci. 2017;3(5):427–34.
43. Kan C, Huang C, Pan JR. Time requirement for rapid-mixing in coagulation. Colloids Surf A Physicochem Eng Asp. 2002;203(1–3):1–9.
44. Katayon S, Noor MJMM, Asma M, Ghani LAA, Thamer AM, Azni I, et al. Effects of storage conditions of Moringa oleifera seeds on its performance in coagulation. Bioresour Technol. 2006;97(13):1455–60.
45. Beltrán-Heredia J, Sánchez-Martín J, Delgado-Regalado A. Removal of carmine indigo dye with moringa oleifera seed extract. Ind Eng Chem Res. 2009;48(14):6512–20.
46. Mataka LM, Sajidu SMI, Masamba WRL, Mwatseteza JF. Cadmium sorption by Moringa stenopetala and Moringa oleifera seed powders : batch , time , temperature, pH and adsorption isotherm studies. Int J Water Resour Environ Eng. 2010;2(3):50–9.
47. Guan D, Zhang Z, Li X, Liu H. Effect of pH and temperature on coagulation efficiency in a North-China water treatment plant. Adv Mater Res. 2011;243–249:4835–8.
48. Sahu O, Chaudhari P. Review on chemical treatment of industrial waste water. J Appl Sci Environ Manag. 2013;17(2).
49. Joudah RA. Effect of temperature on floc formation process efficiency and subsequent removal in sedimentation process. J Eng Dev. 2014;18(4):1813–7822.
50. He J, Liu F, Ouyang L, Xu K. Optimim operating conditions confirmation and effectiveness analysis based on research of the coagulation and precipitation integrated process. Procedia Environ Sci. 2011;10:541–8.
51. Warrier RR, Sing B, Balaji C, Priyadarshini P. Storage duration and temperature effects of Strychnos potatorum stock solutions on its coagulation efficiency. J Trop For Environ. 2018;4(2):45–56.
52. de Souza MTF, de Almeida CA, Ambrosio E, Santos LB, Freitas TKF d S, Manholer DD, et al. Extraction and use of Cereus peruvianus cactus mucilage in the treatment of textile effluents. J Taiwan Inst Chem Eng. 2016;67:174–83.
53. Crini G, Lichtfouse E. Advantages and disadvantages of techniques used for wastewater treatment. Environ Chem Lett. 2018;17(1):145–55.
54. Jagaba AH, Kutty SRM, Hayder G, Latiff AAA, Aziz NAA, Umaru I, et al. Sustainable use of natural and chemical coagulants for contaminants removal from palm oil mill effluent: a comparative analysis. Ain Shams Eng J. 2020;11:951–60.
55. Arafat MG, Mohamed SO. Preliminary study on efficacy of leaves, seeds and bark extracts of Moringa oleifera in reducing bacterial load in water. Int J Adv Res. 2013;1(October):124–30.
56. Rodiño-Arguello JP, Feria-Diaz JJ, de Jesús Paternina-Uribe R, Marrugo-Negrete JL. Sinú River raw water treatment by natural coagulants. Rev Fac Ing. 2015;76:90–8.
57. Balamurugan P, Shunmugapriya K. Treatment of urinal waste water using natural coagulants. Int J Recent Technol Eng. 2019;8(2):355–62.
58. Pallavi N, Mahesh S. Feasibility study of Moringa oleifera as a natural coagulant for the treatment of dairy wastewater. Int J Eng Res. 2013;2(3):200–2.
59. Gopika GL, Kani KM. Accessing the suitability of using banana pith juice as a natural coagulant for textile wastewater treatment. Int J Sci Eng Res. 2016;7(4):260–4.
60. Alwi H, Idris J, Musa M, Ku Hamid KH. A preliminary study of banana stem juice as a plant-based coagulant for treatment of spent coolant wastewater. J Chem. 2013;(February).
61. Seghosime A, Awudza JAM, Buamah R, Ebeigbe AB. Effect of locally available fruit waste on treatment of water turbidity. Civ Environ Res. 2017;9(7):7–15.

62. Lagade VM, Taware SS, Muley DV. Seasonal variations in meat yield and body indices of three estuarine clam species (Bivalvia: Veneridae). Indian J Geo-Marine Sci. 2014;43(8):1586–93.
63. Shahbandeh M. Statista: global production of fresh fruit from 1960 to 2018. 2020. https://www.statista.com/statistics/262266/global-production-of-fresh-fruit/#statisticContainer. Accessed 17 Nov 2020.
64. Fahey JW. Moringa oleifera: a review of the medical evidence for its nutritional, therapeutic, and prophylactic properties. Part 1. Trees for life J. 2005;1(5):1–5.
65. Garnayak DK, Pradhan RC, Naik SN, Bhatnagar N. Moisture-dependent physical properties of jatropha seed (Jatropha curcas L.). Ind Crop Prod. 2008;27(1):123–9.
66. Gomez-Flores R, Calderon CL, Scheibel LW, Tamez-Guerra P, Rodriguez-Padilla C, Tamez-Guerra R, et al. Immunoenhancing properties of Plantago major leaf extract. Phytother Res. 2000;14(8):617–22.
67. Díaz MD, de la Rosa AP, Héliès-Toussaint C, Guéraud F, Nègre-Salvayre A. Opuntia spp.: characterization and benefits in chronic diseases. Oxid Med and Cell Longev.2017.
68. Kurniawan SB, Abdullah SRS, Imron MF, Said NSM, Ismail N. 'Izzati, Hasan HA, et al. Challenges and opportunities of biocoagulant/bioflocculant application for drinking water and wastewater treatment and its potential for sludge recovery. Int J Environ Res Public Health. 2020;17(24):1–33.
69. Jassim N, AlAmeri M. Single objective optimization of surface water coagulation process using inorganic/organic aid formulation by Taguchi method. Period Eng Nat Sci. 2020;1924-34.
70. Bratby J. Coagulation and flocculation in water and wastewater treatment. Water 21. 2006.
71. Boulaadjoul S, Zemmouri H, Bendjama Z, Drouiche N. A novel use of Moringa oleifera seed powder in enhancing the primary treatment of paper mill effluent. Chemosphere. 2018.
72. de Paula HM, de Oliveira Ilha MS, Sarmento AP, Andrade LS. Dosage optimization of Moringa oleifera seed and traditional chemical coagulants solutions for concrete plant wastewater treatment. J Clean Prod. 2018;174:123–32.
73. Alo MN, Anyim C, Elom M. Coagulation and antimicrobial activities of Moringa oleifera seed storage at 3 °C temperature in turbid water. Appl Sci Res. 2012;3(2):887–94.
74. Vishali S, Karthikeyan R. Cactus opuntia (ficus-indica): an eco-friendly alternative coagulant in the treatment of paint effluent. Desalin Water Treat. 2015;56:1489–97.
75. Sellami M, Zarai Z, Khadhraoui M, Jdidi N, Leduc R, Ben RF. Cactus juice as bioflocculant in the coagulation-flocculation process for industrial wastewater treatment: a comparative study with polyacrylamide. Water Sci Technol. 2014.
76. Ganjidollst H, Tatsumi K, Yamagishi T, Gholian RN. Effect of synthetic and natural coagulant on lignin removal from pulp and paper wastewater. Water Sci Technol. 1997;35(2–3):291–6.
77. Teh CY, Wu TY, Juan JC. Optimization of agro-industrial wastewater treatment using unmodified rice starch as a natural coagulant. Ind Crop Prod. 2014;56:17–26.
78. Zainol NA, Aziz HA, Lutpi NA. Diplazium esculentum leaf extract as coagulant aid in leachate treatment. AIP Conf Proc. 2017;1835(April).
79. Awang NA, Aziz HA. Hibiscus rosa-sinensis leaf extract as coagulant aid in leachate treatment. Appl Water Sci. 2012;2(4):293–8.
80. Kian-Hen C, Peck-Loo K. Potential of banana peels as bioflocculant for water clarification. Prog Energy Environ. 2017: 47–56.
81. Abidin ZZ, Madehi N, Yunus R. Coagulative behaviour of Jatropha curcas and its performance in wastewater treatment. 2017;00(00):1–10.
82. Idris J, Som AM, Musa M, Ku Hamid KH, Husen R, Muhd Rodhi MN. Dragon fruit foliage plant-based coagulant for treatment of concentrated latex effluent: comparison of treatment with ferric sulfate. J Chemother. 2013;2013:1–7.
83. Muhammad IM, Abdulsalam S, Abdulkarim A, Bello AA. Water melon seed as a potential coagulant for water treatment. Glob J Res Eng C Chem Eng. 2015;15(1):17–24.
84. Ang WL, Mohammad AW. State of the art and sustainability of natural coagulants in water and wastewater treatment. J Clean Prod. 2020;262:121267.
85. Aboulhassan MA, Souabi S, Yaacoubi A, Baudu M. Coagulation efficacy of a tannin coagulant agent compared to metal salts for paint manufacturing wastewater treatment. Desalin Water Treat. 2015:1–7.
86. Tuddao VB, Gonzales E. Updates on water environment management in the Philippines. Philippines: Dept of Environ Manag Bureau; 2016.
87. Awolola GV, Oluwaniyi OO, Solanke A, Dosumu OO, Shuiab AO. Toxicity assessment of natural and chemical coagulants using brine shrimp (*Artemia salina*) Bioassay. Int J Biol Chem Sci. 2010;4(3):633–41.
88. Gunten A Von, Ebbing K, Imhof A, Giannakopoulos P. Brain aging in the oldest-old. 2010.
89. Dassanayake KB, Jayasinghe GY, Surapaneni A, Hetherington C. A review on alum sludge reuse with special reference to agricultural applications and future challenges. Waste Manag. 2015;38(1):321–35.
90. Zhang Q, Zhang F, Ni Y, Kokot S. Effects of aluminum on amyloid-beta aggregation in the context of Alzheimer ' s disease. Arab J Chem. 2019;12(8):2897–904.
91. Mohana R. Utilization of natural coagulant in turbidity removal and oxygen demand reduction. J Environ Eng Stud. 2019;4(2):18–24.
92. Kueh ABH. Spent ground coffee–awaking the sustainability prospects. Environ Toxicol Manage. 2021;1(1):1–6.
93. Al Farraj DA, Elshikh MS, Al Khulaifi MM, Hadibarata T, Yuniarto A, Syafiuddin A. Biotransformation and detoxification of antraquione dye green 3 using halophilic Hortaea sp. Int Biodeterior Biodegradation. 2019;140:72–7.
94. Al Farraj DA, Hadibarata T, Yuniarto A, Alkufeidy RM, Alshammari MK, Syafiuddin A. Exploring the potential of halotolerant bacteria for biodegradation of polycyclic aromatic hydrocarbon. Bioprocess Biosyst Eng. 2020;43(12):2305–14.
95. Al Farraj DA, Hadibarata T, Yuniarto A, Syafiuddin A, Surtikanti HK, Elshikh MS, et al. Characterization of pyrene and chrysene degradation by halophilic Hortaea sp. B15. Bioprocess Biosyst Eng. 2019;42(6):963–9.
96. Hadibarata T, Syafiuddin A, Al-Dhabaan FA, Elshikh MS. Rubiyatno. Biodegradation of Mordant orange-1 using newly isolated strain Trichoderma harzianum RY44 and its metabolite appraisal. Bioprocess Biosyst Eng. 2018;41(5):621–32.
97. Syafiuddin A, Boopathy R, Hadibarata T. Challenges and solutions for sustainable groundwater usage: pollution control and integrated management. Curr Pollut Rep. 2020;6(4):310–27.
98. Syafiuddin A, Fulazzaky MA. Decolorization kinetics and mass transfer mechanisms of Remazol Brilliant Blue R dye mediated by different fungi. Biotechnol Rep. 2021;29:e00573.
99. Mahmud KN, Wen TH, Zakaria ZA. Activated carbon and biochar from pineapple waste biomass for the removal of methylene blue. Environ Toxicol Manage. 2021;1(1):30–6.
100. Zakaria NA, Abdul Rahim AR. An overview of fruit supply chain in Malaysia. J Mech. 2014;37:36–46.
101. Padam BS, Tin HS, Chye FY, Abdullah MI. Banana by-products: an under utilized renewable food biomass with great potential. J Food Sci Technol. 2014;51(12):3527–45.
102. FAOSTAT. Understanding coconut as a biomass fuel. In: Food and Agriculture Organization of the United Nations. 2014.

103. Syafiuddin A, Fulazzaky MA, Salmiati S, Kueh ABH, Fulazzaky M, Salim MR. Silver nanoparticles adsorption by the synthetic and natural adsorbent materials: an exclusive review. Nano Env Engg. 2020;5(1):1–18.
104. Syafiuddin A, Salmiati S, Hadibarata T, Kueh ABH, Salim MR. Novel weed-extracted silver nanoparticles and their antibacterial appraisal against a rare bacterium from river and sewage treatment plan. Nanomaterials. 2018;8(1):1–17.
105. Syafiuddin A, Salmiati S, Hadibarata T, Kueh ABH, Salim MR, Zaini MAA. Silver nanoparticles in the water environment in Malaysia: inspection, characterization, removal, modeling, and future perspective. Sci Rep. 2018;8(1):1–15.
106. Syafiuddin A, Salmiati S, Hadibarata T, Salim MR, Kueh ABH, Sari AA. A purely green synthesis of silver nanoparticles using *Carica papaya*, *Manihot esculenta*, and *Morinda citrifolia*: synthesis and antibacterial evaluations. Bioprocess Biosyst Eng. 2017;40(9):1349–61.
107. Syafiuddin A, Salmiati S, Hadibarata T, Salim MR, Kueh ABH, Suhartono S. Removal of silver nanoparticles from water environment: experimental, mathematical formulation, and cost analysis. Water Air Soil Pollut. 2019;230(5):102–17.
108. Syafiuddin A, Salmiati S, Jonbi J, Fulazzaky MA. Application of the kinetic and isotherm models for better understanding of the behaviors of silver nanoparticles adsorption onto different adsorbents. J Environ Manag. 2018;218:59–70.