



The potential and constraints of replacing conventional chemical coagulants with natural plant extracts in water and wastewater treatment

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ABSTRACT

Coagulation is a vital stage in treating contaminated water using coagulants that can be either synthetic or natural. Currently, water is being treated by water treatment plants that use aluminum- or iron-based salts and synthetic polymers for coagulation. However, these synthetic coagulants have flaws, including the production of large amounts of chemical sludge, considerable effects on the pH of the purified water, relatively expensive to use, and potentially toxic effects on the environment. The potential and constraints of replacing chemical coagulants with natural plant extracts for water treatment are reviewed in this paper. Various natural coagulants previously investigated for the removal of heavy metals, turbidity, pathogens, and other contaminants from surface and synthetic water were analyzed based on dosage rates, active ingredients responsible for coagulation, and extraction methods among other criteria. A comparative analysis was performed in studies in which both conventional chemical coagulants and natural plant extracts were used for water treatment. Various studies have shown that it is possible to replace chemical coagulants with plant-based extracts. It has been demonstrated that it is possible to replace chemical coagulants with natural plant-based extracts because seed extracts such as those of fenugreek can achieve turbidity removal efficiencies as high as 98% as compared to 85% for alum. The use of natural coagulants may be beneficial for water treatment plants because they may produce less sludge than chemical coagulants; this increases environmental sustainability while decreasing the cost of handling sludge. They are less toxic than chemical coagulants and do not pose any adverse threats to the environment; therefore, they are a safer alternative. However, studies are needed on how these plant-based coagulants can be commercialized in the same way that chemical coagulants are readily available.

1. Introduction

Poor access to adequate, clean, and safe water is one of the greatest problems in global development (Pandey *et al.*, 2021). In 2020, approximately 122 million people were getting water directly from surface water sources (UNICEF/WHO, 2021). In 2021, about 2.2 billion people did not have safely managed drinking water services (World Bank, 2021).

Water can be contaminated by natural or anthropogenic sources of pollutants (Ojha, 2020). Many anthropogenic activities occurring in the environment have increased the contamination of available water resources (Fernando *et al.*, 2018). Moreover, an increase in population has accelerated urbanization, economic growth, and efforts to lessen poverty, resulting in intensified food production, resource mining, and industrial development. The increase in population density in urbanized

areas has escalated environmental pressure from agricultural and industrial practices and waste disposal habits (Boelee *et al.*, 2019). When groundwater flows through the pores of sedimentary rocks and various soils and infiltrates them, it can contaminate the water through natural processes by mixing it with various natural substances, including iron, nitrate, arsenic, and fluoride. Consequently, high concentrations or undesirable quantities of naturally occurring compounds can contaminate groundwater, the consumption of which can cause severe health risks (Ali & Ahmad, 2020).

In both developed and developing countries, the challenges of water contamination are mainly due to chemical exposure, which includes, but is not limited to agricultural sources (Sharma & Bhattacharya, 2017). Contaminants such as insecticides, herbicides, pesticides, chemical fertilizers, heavy metals (e.g., mercury and lead), and hazardous chemicals mainly emanate from agricultural and industrial activities (Bekele &

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Yegrem, 2021; Ashbolt, 2004; Fawell et al., 2001). Disposal of batteries, diesel fuel, and household chemical wastes, such as oils, medicines, antiseptics, and paint solvents, can also result in groundwater contamination and severe health challenges (Neksumi et al., 2022; Kass et al., 2005; Anwar & Yaqout, 2003). Furthermore, the release of chemicals from wastewater can cause groundwater pollution. Riverbeds and wetlands are also at risk because of rising sediment impoundments and the presence of persistent and hazardous contaminants. Such pollution can persist even after the sources have stopped producing pollutants (Edokpayi et al. 2017).

Globally, at least two billion people use a drinking water source with fecal contamination, causing approximately 485,000 diarrheal deaths each year (WHO, 2019). The pollution of drinking water can result in high mortality and morbidity due to waterborne diseases such as typhoid, cholera, diarrhea, dysentery, hepatitis, and many protozoan and helminth infections (Pal et al., 2018; Aryal et al., 2012). Consumption of water that contains pathogens or elements that could be toxic can result in health impacts that range from discomfort to death (UNEP, 2016).

Raw water often requires treatment to make it potable. If the water originates from ground sources via mountain springs, the water quality is usually good, and in most cases, only disinfection is sufficient (Dursun & Sarcan, 2022). However, suspended particles are of great significance if the water is from a surface water supply such as a river, lake, or dam. Various techniques to remove suspended particles include the introduction of coagulants and the use of membranes (Owodunni et al., 2023).

Owing to potential disturbances with downstream purification processes and adverse impacts on consumer acceptance, effective turbidity reduction is one of the main objectives in the treatment of drinking water. Turbidity removal during water treatment has two purposes. The removal of turbidity results in the removal of some microbes, while reducing the levels of organic matter and other particles, which increases the effectiveness of downstream treatment processes (Soros et al., 2019). The removal of heavy metals is also of significant importance because they can be harmful and dangerous to consumer health, as excessive quantities of copper can be toxic and carcinogenic (Kahlson & Dixon, 2022). Bacterial removal is an important process in water treatment because infectious diseases caused by bacteriological agents are the most common and widespread health risks associated with drinking water (WHO, 2017; Mcallister, 2005).

Water turbidity can be reduced and removed using several standard techniques, including precipitation, adsorption, flotation, ion exchange, membrane filtration, and biological and electrolytic processes (Rajoria et al., 2022). The process of turbidity elimination in the form of suspended and colloidal debris and dissolved matter to generate drinkable water from the majority of raw water sources requires the use of a coagulant during the coagulation-flocculation stage of water treatment operations. Coagulants and flocculants exist in a variety of forms and are used in conventional water treatment processes. These can be grouped into inorganic coagulants, such as aluminum and ferric salts; synthetic organic polymers, such as polyacrylamide derivatives and polyethylene imine; and natural coagulants, which can be obtained from microorganisms, plants, and animal tissues (Halder, 2021; Vijayaraghavan et al., 2011).

In recent years, there has been significant interest in the use of natural coagulants (Amran et al., 2022; Alenazi et al., 2020; Mumbi et al., 2018; Unnisa and Bi, 2018). Natural coagulants are biodegradable, making them a potentially sustainable option for water treatment (Agarwal & Saini, 2022). According to Makarfi et al. (2022), the utilization of plant-derived natural materials of plant origin to treat turbid raw water is not a new concept. Natural organic polymers have been used as coagulants and coagulant aids for water with high turbidity in China, India, and Africa for more than 2,000 years (Freitas et al., 2018). Various plants such as *Moringa oleifera* seeds have been identified as natural coagulants (Nhut et al., 2021; Pandey et al., 2020; Hussian et al.,

2012; Saulawa et al., 2011; Okuda et al., 1999), Nirmali seeds (Alenazi et al., 2020), and Cactus (Choudhary et al., 2018).

The advantages of using natural coagulants for water treatment include low coagulant cost, biodegradability, production of lower sludge volume compared to chemical coagulants, and that they are easily obtainable (Kristianto et al., 2018). In a study carried out by Hadadi et al. (2022), natural coagulants *Moringa oleifera* (MO) seeds, *Pinus halepensis* seeds (PHS), *Opuntia ficus indica* (OFI), and Algerian *Aloe vera* (AV) produced sludge volumes of 25 ± 1 mL/L, 30 ± 1 mL/L, 26 ± 1 mL/L, and 22 ± 1 mL/L, respectively, which were less than the sludge volumes produced by chemical coagulants ferric chloride and alum of 52 ± 1 mL/L and 62 ± 1 mL/L, respectively. These coagulants are derived from plant materials; therefore, the byproducts are organic and biodegradable, making their use environmentally friendly (Hadadi et al., 2022). In terms of cost, a study conducted in India by Thirugnanasambandham & Karri (2021) revealed that natural coagulant *Azadirachta indica* A. Juss had lower operating costs (\$USD 0.56/m³) than alum (\$USD 1.73/m³) for treating urban wastewater. Despite these advantages, the level of acceptance of natural coagulants for water and wastewater treatment plants has hitherto remained low (Ang & Mohammad, 2020). This is mainly because of a lack of industrial confidence in adopting natural coagulants owing to concerns about the efficiency and consistency of these coagulants when they are used for water treatment on a large scale. Therefore, this study aimed to investigate the potential and constraints of replacing conventional chemical coagulants with natural plant extracts for water treatment. These issues will be discussed in the following sections: coagulation and flocculation processes in water treatment, classification of coagulants, classification of natural plant extracts according to the removal of contaminants, criteria for analysis of natural coagulants, coagulation performance of conventional chemical coagulants compared to coagulants derived from plant extracts, potential constraints of using natural plant extracts for water treatment, and opportunities and drawbacks for the commercialization of natural coagulants. These sections will aid in providing an overall understanding of how water is treated so that it is potable and the role of coagulants in the purification of water in removing various contaminants. The criteria for analyzing coagulants are important as they help in selecting coagulants that are appropriate for a specific water treatment setup and for water with different concentrations of contaminants.

2. Coagulation and flocculation processes in water treatment

Coagulation and flocculation processes are regularly used in the treatment of drinking water (Maćczak et al., 2022) and industrial effluents (Teh et al., 2016). In an aqueous medium, the majority of solids or suspended particles are smaller and have negative charges. Consequently, the particles must group together to create larger flocs to speed up the sedimentation process. Electrostatic repulsive forces, which prevent negatively charged particles from collecting on the substance, pose a challenge to this process. As a result, it takes longer to settle. To fix this issue, the particles have to be destabilized with a coagulant (El-taweel et al., 2023). Therefore, coagulation happens when coagulants are added during relatively intense mixing to destabilize naturally occurring particles and macromolecules (Al-Risheq et al., 2021).

The process of coagulation alone does not significantly help, it needs the process of flocculation to perform effectively (Mohd-Salleh et al., 2019). Therefore, flocculation which allows slow mixing is a crucial step in water treatment to achieve optimum performance. The purpose of the flocculation stage is to detect the collision and enhance the aggregation of the particles as a result (El-taweel et al., 2023).

2.1. Mechanisms of coagulation

In general, the coagulation process consists of four steps that are grouped according to the primary mechanism that causes aggregates to

develop. These four steps include double-layer compression, charge neutralization, the formation of electrostatic patches, and the formation of inter-particle bridges (Gheraout et al., 2020; Mortadi et al., 2017; Bordini et al., 2004). Furthermore, flocculation takes place as a result of two basic phenomena, namely; orthokinetic which is a result of fluid motion, and perikinetic which is caused by Brownian motion (Beljon, 2021). Fig. 1 shows the overall process of coagulation-flocculation in water treatment. The mechanisms for the coagulation process are described in the following subsections.

2.1.1. Charge neutralization

The colloidal particle's surface, which is typically negatively charged, is coated by an oppositely charged coagulant as part of the charge neutralization mechanism, which lowers and neutralizes the colloidal particle and brings the zeta potential down to almost zero net charge (Choi et al., 2015). Reduced zeta potential, which arises from reducing and neutralizing the surface charge, aids in lowering or removing the energy barrier. As a result, colloidal particles are destabilized and begin to gather together and settle down (Alnawajha et al., 2022).

2.1.2. Formation of inter-particle bridges

When long-chain polymers or polyelectrolytes stretch into a solution and form threads or fibers, they bridge the gap between numerous colloidal particles. A crucial component of bridging flocculation is the molecular weight of polymeric flocculants (Mohd-Salleh et al., 2019; Wei et al., 2018). Polymers that possess a low degree of cationic charge and a high molecular weight should mainly work through the bridging mechanism since the polymers with large molecular weights have long-chained structures which significantly increase the number of vacant adsorption sites. (Alnawajha et al., 2022).

2.1.3. The formation of electrostatic patches

Charged polymers or colloids bind to an oppositely charged particle in this process. Through areas of opposite charges, particles are drawn to one another, thereby coagulating the suspension (Agunbiade et al., 2016). For the charge neutralization procedure, a patch-wise medium which is referred to as an electrostatic patch mechanism is used. The surface of the colloids will patch with different cationic species, resulting in particle surfaces with positive and negative charges. Surfaces of colloids with mixed charges will have fewer repelling forces and more van der Waals forces between the particles (Chong et al., 2021).

2.1.4. Double-layer compression

The negatively charged colloidal particles draw ions of opposite

charge to form a dense layer adjacent to the particle which is known as the Stern layer. The Stern layer's repulsive force and the dynamic equilibrium between the excess positive ions drawn in by the negatively charged core colloids cause the diffuse layer to form. The double-layer is a term used to describe such two layers in the colloid particle interfacial region (Zhao, 2020). The double-layer will be squeezed as soon as a coagulant (positively charged) is added to a colloidal solution due to electrostatic interaction between the ions and colloids. Double-layer compression is a crucial destabilizing mechanism in naturally occurring aquatic systems, even though it does not control the colloid destabilization process in water treatment (Gheraout et al., 2020).

3. Classification of coagulants

Different types of coagulants can be used for treating water (Tetteh et al., 2017). These coagulants can be chemical, non-chemical, synthetic, or natural coagulants. Each type of coagulant possesses its unique properties with some of the coagulants having ions that will be positively charged and these will entrap the negative charge of the organic matter in the water that causes turbidity (Tetteh & Rathilal, 2020). An overview of synthetic organic polymers, natural coagulants, and inorganic coagulants and coagulant aids is provided in this section.

3.1. Inorganic coagulants and coagulant aids

Conventional chemicals which are utilized for coagulation are mainly aluminum or iron-based salts. Metal-based coagulants are most widely utilized because of their advantages which include the fact that they are low-cost, widely available, and can achieve removal efficiencies as much as 98.8%, 99.7%, and 92.3 % for removal of turbidity, total suspended solids and COD respectively (Al-Wasify et al., 2023; Kurniawan et al., 2023). The use of these metal-based coagulants for the purification of water is not without drawbacks, though. They may change the pH of the water since they deplete alkalinity; thereby resulting in lime or soda ash being added to maintain the desired pH levels (Iwuozor, 2019). Additionally, the use of metal-based coagulants causes the production of large amounts of sludge as well as the presence of residue metals in the sludge (Freitas et al., 2018; de Souza et al., 2016). The benefits and drawbacks of some inorganic coagulants for water treatment are listed in Table 1.

The settling properties of flocs produced by coagulants are improved by coagulant aids. A coagulant aid that has previously been utilized is activated silica. Activated silica was utilized as a coagulant aid together with Poly Aluminium Chloride (PAC) for enhanced coagulation of raw water from Yellow River Water (YW) and South-to-North Water

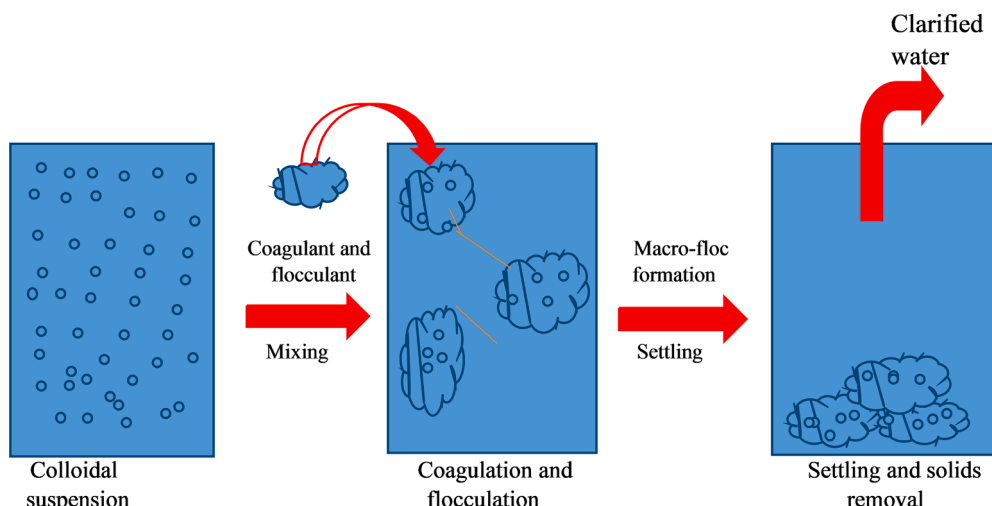


Fig. 1. Overview of the coagulation-flocculation process, adapted from Wilson (2014).

Table 1Benefits and drawbacks of inorganic coagulants. Adapted from [Thakur et al. \(2021\)](#).

Coagulant	Benefits	Drawbacks	References
Aluminum sulfate (Alum) $\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$	Low sludge volume index	- Limited pH effective range - Introduces dissolved aluminum salts residues to the water	Thakur et al., 2021 ; Neukirchen & Ries, 2020 ; Zand and Hoveidi, 2015
Sodium aluminate $\text{Na}_2\text{Al}_2\text{O}_4$	- Efficient in hard waters - Mostly, low concentrations are needed.	- High cost - Ineffective for the treatment of soft water	Krupinska, 2020 ; Tripathy et al., 2019
Polyaluminium chloride (PAC) $\text{Al}_{13}(\text{OH})_{20}(\text{SO}_4)_4\text{Cl}_{15}$	- Rapid aggregation velocity - Bigger and heavier flocs than conventional Al(III) salts - Less dosage is required than conventional Al(III) salts - The pH of the treated water is unaffected.	It has considerably lower molecular weight and size than organic polymers, and its stability to withstand subsequent hydrolysis is also significantly lower than for organic polymers flocculants	Abdo et al., 2020 ; Gao et al., 2002 ;
Ferric sulfate $\text{Fe}_2(\text{SO}_4)_3$	Less sensitive to overdosage than aluminum-based coagulants	- Very caustic - It causes rust-colored stains that are visible and related to chemical spills and leaks. - Depletes alkalinity	Bahroodin et al., 2021 ; Sadeghi et al., 2017 ; Hassan, 2005
Ferric chloride $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$	Effective in a broad range of pH between 4 and 11	- Salts are added as dissolved solids to the water. - Doubles the amount of alkalinity that alum consumes	Bouchareb et al., 2021 ; Bouchareb et al., 2020
Ferrous sulfate $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$	It is less sensitive to pH than lime.	- Salts are added as dissolved solids to the water. - Normally, alkalinity must be added.	Marey, 2019
Lime $\text{Ca}(\text{OH})_2$	Does not add dissolved salts to the water	- pH-dependent - Produces vast quantities of sludge	Thakur et al., 2021 ; Kumar et al., 2017

Diversion (SNW) in Zhengzhou, China, with turbidity removal efficiencies of up to 96.5% obtained at PAC dosage of 35mg/L and activated silica dosage of 7 mg/L for source water YW and SNW blended in the ratio of 2:1 ([Lu et al., 2018](#)). Other aids used at the time were sodium alginates and some soluble starch products which are still being utilized. Soluble starch products such as sago and tapioca were used for the removal of turbidity in river water achieving turbidity removal efficiencies of 93.7% at a dosage of 1000 mg/L and 96.4% at an optimum dosage of 2000 mg/L for tapioca and sago starch respectively ([Zainol et al., 2020](#)). The fact that these compounds were well-known and had previously been employed in the food business gave them the benefit of being accepted as safe for water purification. Polyelectrolytes were later utilized and were more effective. They now consist of many synthetic products, particularly long-chain organic chemicals which can either be cationic, anionic, or non-ionic ([Johnson & Elphinston, 2017](#)).

3.2. Synthetic organic polymers

Synthetic organic polymers have been utilized for decades for the treatment of water and wastewater as flocculants and also as chemical aids for sludge dewatering ([Heiderscheidt et al., 2016](#)). These high molecular weight polymers which are commonly referred to as polyelectrolytes are synthetic compounds that tend to strongly adsorb the particles on the surface in an aqueous suspension ([Koshani et al., 2020](#)). Utilizing organic polymers in the process of treating drinking water can increase the settling rate, reduce costs and improve the water quality ([Iwuozor, 2019](#)).

Some of the advantages of using organic polymers are that they sometimes require lower dosages than metal salt coagulants and that they produce less sludge with better dewatering characteristics ([Dayarathne et al., 2021](#)). It has been observed that floc re-growth after breakage caused by initial turbulent mixing is almost fully reversible with cationic polymers while mostly irreversible when metal salt coagulants are used ([Zhu & Dou, 2021](#)). Moreover, the introduction of organic coagulants does not consume alkalinity; therefore there will be no need for any pH adjustments of the treated water ([Naceradska et al., 2019](#)). This was demonstrated by [Sellami et al. \(2014\)](#) in a study in which there was a slight change in pH from 4.94 to 4.56 when synthetic organic polymer polyacrylamide was used as a flocculant for the treatment of wastewater from the food industry. According to [Bradney et al. \(2019\)](#), there have been some concerns concerning the toxicity of synthetic polymers and their impacts on human health and also on the

aquatic ecosystem which is a disadvantage of using synthetic organic polymers for water treatment. However, the toxicity of these products has mostly been linked to residual concentrations of non-reacted monomers and other contaminants which might be present in the treated water and are strictly limited by legislation worldwide ([Heiderscheidt et al., 2016](#)).

3.3. Natural polymers

Natural polymers are readily available and environmentally friendly hydrophilic compounds that are also non-toxic to people ([Tetteh & Rathilal, 2020](#)). Many naturally occurring polymers possess intrinsic cationic properties which can be modified to give a cationic polyelectrolyte to be used for solid-liquid separations as flocculants ([Sillanpää et al., 2018](#)). Examples of natural polymers are chitosan, starch, and cellulose derivatives such as carboxymethyl cellulose (CMC), methylcellulose (MC), and chitin ([Benabid & Zouai, 2016](#)). Like most natural polymers, chitosan is non-toxic, making it generally accepted on a health basis ([Dibyangana et al., 2015](#)).

Plants are a primary source of natural polymers. Examples of plant coagulants are Nirmali seeds, *Moringa oleifera* seeds, *Strychnos potatorum* seeds, *Jatropha curcas* seeds, *Cicer arietinum*, and Cactus ([Nisa & Kishor, 2021](#); [Patchaiyappan & Devipriya, 2021](#)). The main merits of utilizing natural plant-based coagulants as point-of-use water treatment mechanisms are that they are cost-effective, unlikely to produce treated water that has extreme pH(outside the WHO recommended range of 6.5 to 8.5) and are highly biodegradable ([Patchaiyappan et al., 2020](#)). These benefits are particularly enhanced if the plant from which the coagulant is produced is naturally located nearby in a rural area.

Natural coagulants have the disadvantage of being unable to handle huge pH variations. Most coagulants work effectively within a specified pH range, but their efficacy is greatly affected outside of this range ([Gautam & Saini, 2020](#)). Another disadvantage of employing natural coagulants is that they sometimes require higher dosages than synthetic coagulants ([Aliyu et al., 2015](#)). The application of natural coagulants such as *Moringa oleifera* seeds in water treatment contributes to the surge of organic matter in treated water. This increase of organic matter in treated water is a disadvantage of using *Moringa oleifera* seeds in water treatment because organic matter contributes to the development of carcinogenic trihalomethanes when water is disinfected using chlorine ([Camacho et al., 2017](#)).

4. Characterization of natural plant extracts according to the removal of key contaminants

The findings of investigations on the removal of turbidity, pathogens, and heavy metals from the water will be presented in this section.

4.1. Turbidity

To lower the turbidity of synthetic water using a standard jar test apparatus, Asrafuzzaman et al. (2011) evaluated the seeds of *Moringa oleifera*, *Cicer arietinum*, and *Dolichos lablab* for usage as locally accessible natural coagulants in Bangladesh. After introducing water-soluble extracts to the water, turbidity removal efficiencies obtained were 94.1%, 95.89%, and 88.9%, respectively. Seeds of *Cicer arietinum* treated the water to a final turbidity of 3.9 NTU which met WHO standards for turbidity in water as it was less than 5NTU whilst seeds of *Moringa oleifera* and *Dolichos lablab* achieved a final turbidity of 5.9 and 11.1 NTU and thus did not meet WHO standards for turbidity in drinking water. The natural coagulants in the study worked better with high turbidity water (90–120 NTU) in comparison to medium (40–50 NTU) or low turbidity water (25–35 NTU). These classifications of turbidity are according to Asrafuzzaman et al., (2011) and may differ from one study to another. Fig. 2 shows the seeds of *Moringa oleifera*, *Cicer arietinum*, and *Dolichos lablab* which can be utilized in water treatment.

The turbidity reduction efficiencies obtained by Asrafuzzaman et al. (2011) were higher than those obtained in another study which was done in Malaysia using *Azadirachta indica*, *Stenochlaena palustris*, *Dicranopteris linearis*, *Syzygium polyanthum*, *Manihot esculenta*, *Piper sarmentosum*, and *Melastoma malabathricum*. The optimum dosage for *Piper sarmentosum* and *Manihot esculenta* was 5 g/L, which removed 24.2% and 22.2% of the turbidity, respectively (Ahmad et al., 2022). *Azadirachta indica* removed 26.9% of the turbidity at 10 g/L, followed by *Stenochlaena palustris*, 24.9%, *Syzygium polyanthum*, and 17.5% of *Dicranopteris linearis*. *Melastoma malabathricum* finally achieved the removal of 12.2% with a dosage of 0.1 g/L, with the final turbidities ranging from 353.0 NTU to 412.5 NTU from an initial of 500 ± 50 NTU (Ahmad et al., 2022).

The results obtained by Asrafuzzaman et al. (2011) are comparable to the turbidity removal efficiency used when calcium alginate was utilized as a natural coagulant to treat synthetic turbid water in a study by Saranya et al. (2022). The coagulation-flocculation process produced turbidity values of less than 5 NTU, which are acceptable for drinking water while utilizing alginate concentrations of 6–10 mg/L and calcium concentrations larger than 50 mg/L. A turbidity removal of greater than 93% was achieved and maximum removal of 99.3% was obtained (Saranya et al., 2022).

Another study by Ali et al. (2010) produced similar results to what Asrafuzzaman et al., (2011) obtained in their study. *Moringa oleifera* was used as a natural coagulant by Ali et al. (2010) to treat synthetic turbid water in a study carried out in Malaysia. A dosage of 0.4 mg/L resulted in final turbidity of 1.28 to 1.36 NTU, which is under the advised WHO

guideline value of 5 NTU, and a turbidity removal effectiveness of 96.23%.

4.2. Pathogens

In a study conducted by Amran et al. (2021), *Carica papaya* seeds were used in Malaysia at a dosage of 196 mg/L and a pH of 4 to treat river water. Removal efficiencies of 57.6% and 62.1% were achieved for total coliform and *Escherichia coli* respectively. However, higher removal efficiencies of total coliform (99.61%) and *Escherichia coli* (99.74%) when *Moringa oleifera* seeds with a dosage of 600 mg/L were used to treat domestic wastewater in Brazil (Andrade et al., 2021). In a separate study, *Magnifera indica* seeds with a dosage of 0.5 g/L were also used to remove 100% of both total coliform and fecal coliform in stream water (Okunola et al., 2020).

Asrafuzzaman et al. (2011) also carried out a study in which they used seeds from the plants *Moringa oleifera*, *Cicer arietinum*, and *Dolichos lablab* to successfully reduce the total coliform in turbid water by 89–96%. Total coliform in water was reduced by *Moringa oleifera* seeds from 1.35×10^3 cfu/100mL to 5.4×10^1 cfu /100mL. Additionally, the total coliform was reduced to 1.0×10^2 cfu/100mL and 1.1×10^2 cfu/100mL by *Cicer arietinum* and *Dolichos lablab*, respectively from 1.05×10^3 cfu/100mL. The water samples which were treated using the three natural coagulants did not meet the WHO drinking water quality standards of 0 cfu/100mL for total coliform in water and will therefore require further disinfection. Furthermore, a study by Ramavandi (2014) which was conducted in Iran also showed that fecal streptococcus was removed to almost 0 cfu/100mL from turbid water using a coagulant extracted from *Plantago ovata*. However, the fecal coliform was not reduced to 0 cfu/100mL and did not meet WHO drinking water quality standards.

Tubers of *Maerua subcordata* and seeds of *Moringa stenopetala*, which are shown in Fig. 3 below were used in a study in which coagulation and microbial reduction experiments were done on surface river waters in Ethiopia (Megersa et al., 2016). Both plants displayed excellent performances in the reduction of microbial load (total coliform, *Escherichia coli*, and heterotrophic bacteria). The five rivers that were sampled contained *Escherichia coli* in the range of 153 to 166 cfu/100mL. After being treated with the natural coagulants, four of these water samples had 0 cfu/100mL, which complies with World Health Organization (WHO) drinking water quality requirements. However, one sample did not meet the WHO water quality standards as the final *Escherichia coli* load after treatment with *Maerua subcordata* and *Moringa stenopetala* was 2 and 1 cfu/100mL, respectively. Therefore, in this case, residual disinfection is very important to obtain zero colony-forming units in treated water (Pritchard et al., 2009). A greater percentage of total coliform removal (100%) was noticed for the lowest turbidity value of 20 NTU than (98.4%) for the highest turbidity level of 195 NTU which was used in the study by Megersa et al. (2016). This higher percentage of microbial load removal in lower turbidity water than in higher turbidity water may be due to the increase in particles suspended in the

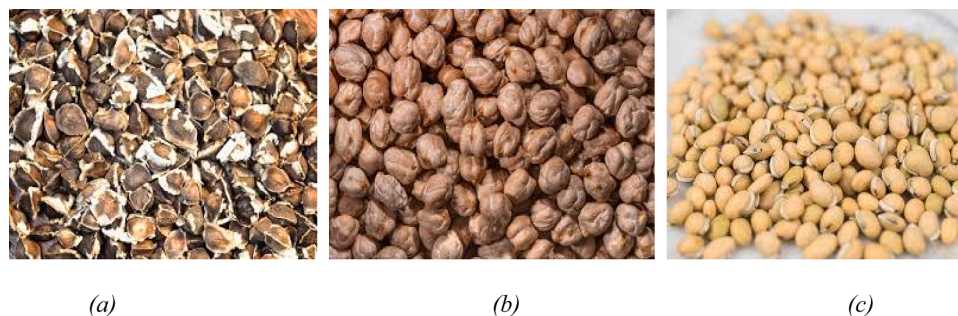


Fig. 2. Seeds of *Moringa oleifera* (a) (Paisley Farm & Crafts, 2022), *Cicer arietinum* (b) (Sergeev, 2015) and *Dolichos lablab* (c) (Exporters India, 2022).

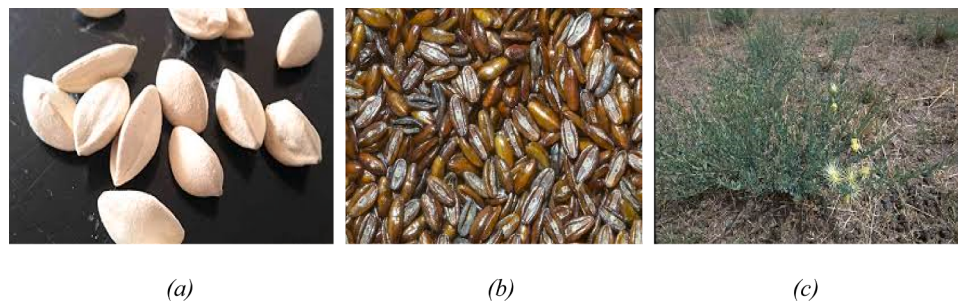


Fig. 3. Seeds of *Moringa stenopetala* (a) (Moringa Matters, 2022), *Plantago ovata* (b) (Botany Today, 2018) and tubers of *Maerua subcordata* (c) (Gerald and Buff, 2002).

high-turbidity water that protects the bacteria from the action of the natural coagulants (Megersa *et al.*, 2016). Sharma & Bhattacharya (2017) suggest that the presence of clay, silts or sand, or organic, algae, and leaf particles causes turbidity which may shield bacteria, thus preventing disinfection chemicals such as chlorine from invading and destroying cells.

The antimicrobial effect of plant coagulants may be accredited to both flocculation (Nwaiwu & Lingmu, 2011) and bactericidal action (Oluduro *et al.*, 2010). The presence of alkaloids and tannins in plant species could contribute to antimicrobial activities (Shojaemehr *et al.*, 2020). Bacterial re-growth was raised as a huge issue of concern with natural coagulants and can be made better by purification of the active agent (Ahizi *et al.*, 2023).

4.3. Heavy metals

The seed cake of *Moringa oleifera* acts as a natural adsorbent for removing heavy metals from water samples (Ndibewu *et al.*, 2011). In a study which was done in Fiji, 60% of cadmium was eliminated by using *Moringa oleifera* seed cake (Nand *et al.*, 2012). This result concurs with the findings of Meneghel *et al.* (2013) in which 57.21% of cadmium was removed using *Moringa oleifera* seeds as a biosorbent in a study conducted in Brazil.

However, in a study done in Malaysia by Shan *et al.* (2017), more than 90% of cadmium was removed from the water samples collected from “The Sungai baluk” river when the seeds of *Moringa oleifera* were

used as a natural coagulant for removal of heavy metals. The initial concentrations of heavy metals which included copper (1.401 mg/L), cadmium (1.887 mg/L), iron (1.414 mg/L), and lead (1.854 mg/L) were high. Results from this study as shown in Table 2 indicate that increasing *Moringa oleifera* seed cake concentration resulted in high removal efficiency of heavy metals from the “Sungai baluk” river samples. (Shan *et al.*, 2017).

In a study done in Malawi by Sajidu *et al.* (2005), 92.14% of iron was removed from synthetic contaminated water using *Moringa oleifera* seeds. This result is comparable with what was concluded in another study carried out by Shan *et al.* (2017), in which iron was reduced to zero in the water samples treated with *Moringa oleifera* seed cake. Moreover, the iron removal efficiency (92.14%) obtained in the study by Sajidu *et al.* (2005) is almost the same as the result obtained in Finland in the study by Righetto *et al.* (2021) in which 92.00% of iron was removed in landfill leachate using a tannin based product named HTH.

The percentage removal of lead using *Moringa oleifera* seeds in synthetic contaminated water was 89% in a study which was done by Sajidu *et al.* (2005) and 80% in ground and surface water in the study done by Subramaniam *et al.* (2011). These values differed from that of 71% which was obtained by Shan *et al.* (2017) when *Moringa oleifera* seeds were used to treat water from “Sungai baluk” river water. Concerning the level of lead in the treated water, the result which was obtained by Shan *et al.* (2017), did not meet the drinking water standards of WHO (2006). The level of lead was lowered to 0.537 mg/L. This amount of lead was above the allowable limit, which is less than 0.05 mg/L.

Table 2
Removal of heavy metals in water using natural coagulants.

Natural coagulant	Metal removed	Final Concentration (mg/L)	Level of removal (%)	WHO standards(mg/L)	Complied with WHO standards	Reference
Neem wood powder	Copper	0.5	95	2	Yes	Arshad & Imran, 2020
	Zinc	0.4	96	3-5	Yes	
	Manganese	0.6	94	<0.1	No	
	Lead	2.8	72	0.01	No	
	Chromium	2.5	75	0.05	No	
Pine Cone powder	Arsenic	0.75	25	0.01	No	Seetha <i>et al.</i> , 2012
	Copper	0.1	99	2	Yes	
	Zinc	0.3	97	3-5	Yes	
	Manganese	0.4	96	<0.1	No	
	Lead	1.2	88	0.01	No	
Carica papaya leaf powder	Chromium	1.4	86	0.05	No	Mane <i>et al.</i> , 2011
	Arsenic	0.75	25	0.01	No	
	Lead	10.2	89.8	0.01	No	
	Chromium	3.34	66.6	0.05	No	
	Nickel	1.16	88.4	0.02	No	
Moringa oleifera	Copper	0.001	99.9	2.00	Yes	Shan <i>et al.</i> , 2017
	Cadmium	0.029	98.5	0.003	No	
	Iron	0	100.0	No health-based guideline	Yes	
	Lead	0.537	71.0	0.01	No	
Dolichos lablab	Iron	<0.05	97.17	No health-based guideline	Yes	Kabir, <i>et al.</i> , 2019

In a study which was conducted in Fiji, Tonga, and Kiribati by Subramaniam et al. (2011), *Moringa oleifera* seed cake was able to remove up to 90.00% of copper in water. This percentage removal is lesser than the one obtained in the study done by Shan et al. (2017), in which more than 99.90% of copper was removed from a water sample using *Moringa oleifera* seed cake. However, the copper removal efficiency of 90.00% obtained by Subramaniam et al. (2011) is slightly greater than the copper removal efficiency of 87.02% which was obtained in a study by Skotta et al. (2023) in which *Lepidium sativum* seeds were used for removing copper in laboratory synthetic samples.

Copper, zinc, and iron were treated to meet WHO water quality standards among the heavy metals examined in this section, but manganese, chromium, lead, arsenic, and cadmium did not after treatment with various natural coagulants like neem wood, pine cones, and *Moringa oleifera* seeds. Manganese, chromium, lead, arsenic, and cadmium would therefore require additional treatment to comply with WHO water quality standards.

5. Criteria for analysis of natural coagulants

Natural coagulants that are derived from plants were analyzed by using environmental and technical criteria which include dosage, active ingredients, extraction method, sludge produced, toxicity, technical complexity, availability, environmental impact, and costs as suggested by Omran et al. (2021).

5.1. Dosage

In a study by Rayudu et al. (2022), dragon fruit foliage was employed as a natural coagulant to remove turbidity. The turbid water was prepared in the laboratory with an initial turbidity of 55 NTU. The optimum coagulant dosage was found to be 250 mg/L with final turbidity of 9 NTU and will therefore require further treatment to achieve the desired WHO standard of 5 NTU in drinking water (WHO, 2017). The results from the study by Sciban et al. (2005) showed that seeds of common bean *Phaseolus vulgaris* and indigobush *Amorpha fruticosa* at a dose of 5 mg/L were best used for treating water with a turbidity of 17.5 NTU, which was the lowest turbidity in the study. The extracts of the seeds of black locust *Robinia pseudoacacia* had good coagulation activity (greater than 50%) in water with a turbidity of 70 NTU in a dose of 30 mg/L and for lesser turbidity, lower dosages have a better performance. The optimum coagulant dosage obtained by Taiwo et al. (2020) when treating water with turbidity less than 50 NTU using *Moringa oleifera* seeds was 60 mg/L. The turbidity of the water dropped significantly ($p < 0.05$) at this dosage, from 41.19 ± 1.5 NTU to 23.51 ± 1.03 NTU.

Seven plant species that are native to Malaysia were investigated for their potential to treat the turbidity of water. The plants are *Azadirachta indica*, *Stenochlaena palustris*, *Dicranopteris linearis*, *Syzygium polyanthum*, *Manihot esculenta*, *Piper sarmentosum*, and *Melastoma malabathricum* (Ahmad et al., 2022). The optimum coagulant dosage for *Melastoma malabathricum* was 0.1g/L with turbidity removal of 12.2%. As for *Azadirachta indica*, *Stenochlaena palustris*, *Syzygium polyanthum*, and *Dicranopteris linearis*, the optimum coagulant dose was 10 000 mg/L with the removal efficiency being 26.9%; 24.9%; 24.9% and 17.5% respectively. *Piper sarmentosum* and *Manihot esculenta* achieved optimum coagulant dosages of 5000 mg/L with removal efficiencies of 24.2% and 22.0% respectively. The low turbidity removal efficiencies are due to the fact that the water extraction method was used therefore the ingredients which are responsible for coagulation were not adequately extracted (Ahmad et al., 2022).

Moringa oleifera leaves extracts were used in a study by Pandey et al. (2020) for the removal of turbidity, fluoride, and *E. coli* in water. The initial values of turbidity, fluoride, and *E. coli* in water were 15.6 NTU, 2.3 mg/L, and 315 cfu/100mL respectively. After treatment at a dosage of 150 mg/L, *Moringa oleifera* leaves reduced the turbidity, fluoride, and *E. coli* to 6.1 NTU, 1.1 mg/L, and 41 cfu/100mL respectively. Turbidity

and *Escherichia coli* did not meet WHO standards of 5 NTU and 0 cfu/100mL respectively whilst fluoride met the standard of 1.5 mg/L fluorides in drinking water (WHO, 2017).

5.2. Active ingredients

The active ingredients of *Moringa oleifera* seeds are soluble cationic proteins with a molecular weight of about 13 kDa and isoelectric pH values of 10 and 11 (Ndabigengesere et al., 1995). Legumes store proteins in seed organelles to provide nutrition for seed growth and development (Duranti & Gius, 1997). These proteins, including albumins, globulins (soluble in dilute salt solutions), prolamins (soluble in alcohol/water mixtures), and glutens (soluble in dilute acid or alkali), are categorized as storage proteins based on their solubility characteristics (Shewry et al., 1995). Globulins, such as arachin and coarachin in peanut (*Arachis hypogaea*) seeds, are the predominant proteins in legumes and play a crucial role when used as natural coagulants (Kristianto, 2017). *Moringa oleifera* seeds, known for their coagulation activity, contain a water-soluble protein called lectin (Ferreira et al., 2011). The active ingredients in *Moringa oleifera* seeds are reported to be stable dimeric proteins that are completely soluble in water (Sapana et al., 2012). Water-soluble proteins, ranging from 18.6% to 37.2%, are the main components found in *Moringa oleifera* seeds after lipids (Kumar et al., 2022; Kristianto, 2017; Ferreira et al., 2011; Duranti and Gius, 1997; Shewry et al., 1995).

In a study conducted by Hussain et al. (2019), pine cone extract was investigated as a natural coagulant for purifying turbid water. The study found that pine cone extracts contain both cationic and anionic coagulating components. The anionic components are responsible for reducing turbidity at lower pH levels, while the cationic protein components are activated at higher pH levels. Other studies have shown that rice hull ash, which is rich in silica, can be used as a coagulant aid to enhance the effectiveness of aluminum or iron coagulants by forming larger and denser flocs (Adams & Mulaba-Bafubandi, 2014; Abo-El-Enen et al., 2011). Additionally, starch can be utilized as a natural coagulant due to its properties as a low-cationic polyelectrolyte (Oladaja, 2015). Table 3 shows the active ingredients from various coagulants which are responsible for the coagulation process.

Banana pith powder has been successfully used as a natural coagulant for treating river water, effectively removing turbidity, suspended solids, and some heavy metals (Kakoi et al., 2016). The functional groups present in the banana pith, such as hydroxyl, carboxylic and ketone, aldehyde, and lactone groups, contribute to its coagulation capabilities. Inulin, found in banana pith juice, acts as an active coagulant agent by forming bridges of flocs, particularly in the treatment of spent coolant wastewater (Alwi et al., 2013). Papaya seeds are known for their antimicrobial properties due to their various phytochemical contents (Peter et al., 2014). Additionally, approximately 10% of papaya seed protein belongs to the albumin type, which can also serve as a coagulant agent (Kristianto, 2017). According to Jones & Bridgeman (2016), when Okra seed is used as a coagulant, it relies on proteins for turbidity removal. White mustard seed proteins have recently been employed to remove anionic dye direct black 19 from textile wastewater, achieving removal efficiencies of up to 90.9% (Tie et al., 2019).

The active ingredients for most natural coagulants which have been studied for water treatment in several studies are mainly proteins (Choudhary & Neogi, 2017; Kristianto, 2017; Ramavandi, 2014; Ferreira et al., 2011; Ndabigengesere et al., 1995).

5.3. Extraction method

Okuda et al. (1999) developed a method for extracting the active ingredient for coagulation from *Moringa oleifera* seeds using a solution of sodium chloride and other salts. This method showed better coagulation activity for removing kaolin turbidity compared to the conventional water extraction method. The salt extraction method achieved over 95%

Table 3

Active ingredients from various coagulants responsible for coagulation in water treatment.

Coagulant	Active ingredient	Coagulation mechanism	References
<i>Moringa oleifera</i>	• Cationic proteins • Water-soluble proteins • Protein lectin	• Charge neutralization and adsorption of the colloidal particles	Choudhary and Neogi, 2017 Ferreira et al., 2011 Ndabigengesere et al., 1995
Nirmali Rice husk ash	• Anionic polyelectrolytes • Silica	• Interparticle bridging • Enmeshment in a precipitate	Sanghi et al., 2002 Adams and Mulaba-Bafubandi, 2014 Abo-El-Enein et al., 2011
Papaya seed	• Protein- albumin type	• Electrostatic interactions	Melinda et al., 2022 Kristianto, 2017
Banana pith powder; banana pith juice	• O-H and C-H groups • The carboxylic (COO-double bond) and asymmetrical ionic carboxylic (-COOH) groups, • Other function groups such as the C-O group of ketones, aldehydes, and lactones, • C-O-C and O-H groups of polysaccharides and the C-N groups • Inulin	• Charge neutralization combined with low bridging	Kakoi et al., 2016 Alwi et al., 2013
Okra seed	• Okra seed protein	• Charge neutralization	Jones & Bridgeman, 2016
Mustard seed	• Mustard seed protein	• Adsorption and charge neutralization	Tie et al., 2019

Table 4

Extraction methods used when utilizing natural coagulants for water treatment.

Coagulant	Extraction method	References
<i>Moringa oleifera</i> seeds	• Salt (NaCl, KNO₃, KCl, NaNO₃) • Water • Hexane • Acetone • Ethanol	Bouchareb et al., 2021 Okuda et al., 1999 Camacho et al., 2017 García-Fayos et al., 2016
Peanut seeds	• Water • Salt (NaCl, KNO₃, KCl, NH₄Cl and NaNO₃) • Salt (NaCl) • Water • Salt (FeCl₃)	Birima et al., 2013 Birima et al., 2015
<i>Plantago ovata</i> seeds	• Water • Salt (NaCl) • Ammonium acetate • Ultrasound-assisted extraction (UAE) • Water • Water	Ramavandi et al., 2015 Ramavandi, 2014 Dhivya et al., 2017
<i>Opuntia ficus-indica</i>	• Ultrasound-assisted extraction (UAE) • Water	Adjeroud- Abdellatif et al., 2020 Gomes et al., 2016
Pine cone	• Water	Hussain et al., 2019

coagulation of the initial 50 NTU kaolin turbidity using a significantly lower dose (4 mL/L) compared to the water extraction method, which required 32 mL of coagulant to remove about 78% of the same turbidity. The improved efficiency of coagulation with sodium chloride is attributed to its salting-in mechanism, increasing protein solubility as the salt's ionic strength rises. Another study by Bouchareb et al. (2021) found that salt extraction with sodium chloride resulted in a higher amount of dissolved coagulant agent compared to water extraction.

In a study by Birima et al. (2015), a de-oiled extract from peanut seeds was utilized for treating palm oil mill effluent. The active coagulation component was extracted using three different concentrations of sodium chloride: 0 mol/L (distilled water), 1 mol/L, and 2 mol/L. The highest removal of total suspended solids (TSS) and chemical oxygen demand (COD) was achieved with the 2 mol/L sodium chloride extract, resulting in 94.7% and 73.5% removal, respectively. In comparison, using distilled water for extraction yielded lower removal efficiencies of 90% for TSS and 66% for COD. Table 4 shows different extraction methods which are utilized when using natural coagulants for water treatment.

In a study by Ramavandi et al. (2015), salt extraction using ferric chloride was used for the extraction of proteins from the seeds of *Plantago ovata*. Adjeroud-Abdellatif et al. (2020) made use of ultrasound-assisted extraction to extract proteins from the natural coagulant *Opuntia ficus-indica* by subjecting the coagulant to ultrasound waves by a sonicator probe.

Many methods have been used to extract active ingredients from

natural coagulants. These include water and salt extraction, ultrasound-assisted extraction, and extraction using hexane, acetone, and ethanol. The most common extraction methods in several studies include water and salt extraction (Bouchareb et al., 2021; Hussain et al., 2019; Birima et al., 2015; Ramavandi et al., 2015; Okuda et al., 1999). Salt extraction method produced better results than water extraction methods in several studies in which both of the methods were used (Behera & Balasubramanian, 2019; Benalia et al., 2018; Birima et al., 2015; Vishali & Karthikeyan, 2015; Okuda et al., 1999).

5.4. Sludge produced and how safe it is for reuse

Ndabigengesere et al. (1995) conducted a study to compare the effectiveness of *Moringa oleifera* seeds and alum as coagulants in water treatment. At a dosage of 500 mg/L, the volume ratio of sludge produced by alum, shelled *Moringa oleifera* seeds, and non-shelled *Moringa oleifera* seeds was 5:2.5:1 respectively. The type and amount of coagulated materials influenced the sludge generated when using *Moringa oleifera* seeds. However, the advantage of using *Moringa oleifera* seeds is that all the by-products are organic and biodegradable. Additionally, the sludge produced by *Moringa oleifera* coagulation is non-toxic and significantly smaller in volume compared to the chemical sludge produced by alum coagulation, which contains aluminum hydroxide precipitates. The differences in sludge volume can be attributed to the sweep coagulation process. In the case of *Moringa oleifera*, the sludge is mainly dependent on the dosage and suspended particles (Vega et al., 2021).

In another study, the efficiency of two natural coagulants, namely calcium lactate and tannic acid was assessed and compared to conventional coagulants; namely, polyaluminium chloride and ferric chloride (Kaji et al., 2020). An initial turbidity of 900 NTU was used to test the four coagulants to compare the sludge produced. After 30 minutes, it was observed that the two natural coagulants created less deposited sludge than the other two chemical coagulants. It was therefore noted that the utilization of natural coagulants results in less sludge produced which is also biodegradable.

5.5. Toxicity

Toxicity is an important factor in evaluating natural coagulants. Genotoxicity refers to the damaging impact on the genetic material of a cell, potentially leading to cancer, mutations, or birth defects (Cinthura et al., 2017). Cytotoxicity, another type of toxicity, refers to the harmful effects on cells caused by toxic agents, which may ultimately result in cell death. (Salih et al., 2019). Studying the cytotoxic, genotoxic, and mutagenic effects of plant compounds can help minimize the potential health risks associated with these substances

According to Igboke et al. (2018), an acute toxicity test demonstrated that *Moringa oleifera* seeds are safe for consumption, with an LD50 value of 14 g/kg. Rolim et al. (2011) conducted a study on the genotoxicity of an aqueous seed extract of *Moringa oleifera*, using various assay systems. The results indicated that the seed extract was not genotoxic and did not pose a health risk. Araújo et al. (2013) evaluated the cytotoxicity of an aqueous extract of *Moringa oleifera* seeds and found no signs of systemic toxicity or mortality in mice. Additionally, Arnoldsson et al. (2008) observed that *Moringa oleifera* seed cake is a non-toxic agent used for water treatment. Furthermore, chitosan, as highlighted by Aider (2010) is a natural, abundant, and non-toxic biodegradable linear copolymer.

Sanmugapriya & Venkataraman (2006) conducted acute and chronic toxicity studies on the aqueous extract and seed powder of *Strychnos potatorum* Linn seeds, commonly used for water treatment. The study involved Wistar albino mice and rats, and no toxic effects were observed in the animals even at a dose of 2,000 mg/kg administered orally. Similarly, Doppalapudi (2019) administered high doses (up to 2000 mg/kg) of seed extracts of *Cicer arietinum* to normal rats orally. No mortality was observed in the rats, indicating the absence of toxic effects from the seed extracts of *Cicer arietinum* even at high doses.

From the studies which were considered, it is evident that various natural coagulants such as *Moringa oleifera*, *Strychnos potatorum* Linn, and *Cicer arietinum* are non-toxic and can therefore be safely used for water treatment.

5.6. Technical complexity

Varkey (2020) described a household technique for reducing the turbidity of surface water from rivers. The method involved adding 0.25g of seed powder to 1 liter of water in a container and vigorously swirling it for 20 to 25 seconds. After allowing the water to settle for an hour, the clear supernatant was poured off and filtered using a fine mesh cotton cloth. According to Babu & Chaudhuri (2005), seeds of *Strychnos potatorum* and *Moringa oleifera* can be easily implemented at the household level, since the implementation procedure does not require extensive technical expertise.

An uncomplicated setup for treating water at the household level would be to dose 10 L of contaminated water with *Strychnos potatorum* seed or *Moringa oleifera* seed as a coagulant (2% suspension prepared with distilled water). This is then followed by mixing via hand-stirring for a period of 2 to 3 minutes and filtering the water directly via a sand filter. The sand filter can be composed of 1,200 mm long and 150 mm internal diameter PVC pipe packed up to a depth of 500 mm using fine river sand, with 20-30 mm of pea-sized gravel at the bottom and arrangement for acquiring an initial flow rate of 3 L/minute. About 2 or

3 filter runs can be done every day. The sand in the filter must then be removed, thoroughly cleaned with water, and put back in the filter when the rate of filtration has significantly decreased (Babu & Chaudhuri, 2005).

According to Price (2007), *Moringa oleifera* seeds can be easily utilized for household water treatment using a simple process that involves removing the wings from the dry seeds and grinding them into a powder. The powder is then mixed with water, agitated for approximately five minutes, and filtered through a piece of woven fabric after an hour, resulting in treated water. Another technique involves immersing a cloth with seed powder in water, typically overnight, to allow contaminants to coagulate. The purified water is then decanted, leaving the coagulated particles at the bottom, and the cloth holding the seeds is removed. Foidl et al. (2001) reported that this method can eliminate up to about 99% of colloidal material, with one seed per liter recommended for slightly polluted water and two seeds for heavily polluted water.

In a study conducted by Burgos-vergara et al. (2020) in Colombian villages, the use of *Moringa oleifera* and *Cassia Fistula* was explored. The seeds of both plants were peeled and ground separately using laboratory and special seed grinders. The resulting fine powders were stored in Ziploc bags to prevent moisture absorption. 10 g of each powder was added to 1 liter of raw filtered river water in two 2-liter plastic bottles. The bottles were vigorously shaken for 1 and 2 minutes respectively (fast mixing), followed by an additional 5 minutes of slower shaking. To ensure complete dissolution of the natural compounds, the water and coagulant were manually mixed using consistent top-to-bottom and bottom-to-top movements. The water was then left undisturbed for 2 hours to allow for sedimentation to occur.

In terms of the practical application of natural coagulants, the studies which were considered show that the coagulants require minimal technical expertise to implement and can therefore be easily implemented even in low-income communities.

5.7. Availability

Previous studies have demonstrated the availability and extraction of natural coagulants from a variety of sources, including waste biomass, natural products, and even microbes (Saleem & Bachmann, 2019; Lee & Chang, 2018; Kanmani et al., 2017). Thus, it can be inferred that some naturally occurring coagulants have dependable and readily accessible sources. Since plants like *Moringa oleifera* only produce mature, dried seeds twice a year, maintaining a consistent supply of raw materials that meet the minimum quality requirement is an inconvenience when considering using natural coagulants for large-scale water treatment facilities (Saleem & Bachmann, 2019).

5.8. Environmental Impact

Water utilities may be interested in using biodegradable natural coagulants that can reduce sludge production while also not adding to the sludge's toxicity (Dorca et al., 2018). If the organic sludge produced by natural coagulants could be used for other tasks, such as wastewater treatment, in the construction industry for producing building materials such as cement, concrete, and mortar, for agricultural use as soil conditioners, and land-based uses, it might provide environmental sustainability (Ahmad et al., 2016).

Rarely is the secondary waste produced by the chemical-based extraction, purification, or synthesis of natural coagulants highlighted. Natural coagulant residues are regarded as organic materials that may interact with disinfectants which would have been produced by a subsequent disinfection process to produce carcinogenic disinfection byproducts. Therefore, natural coagulants should be used cautiously if a downstream disinfection process is available (Ang & Mohammad, 2020).

5.9. Costs

According to cost estimates, the cost of obtaining the natural coagulant from *Moringa oleifera* seeds includes the cost of the raw materials, chemical compounds (n-hexane), the electrochemically activated solution, energy consumption, labor costs, and cost for storage of the substance (Hoa & Hue, 2018). Income can be gained from the sale of oil extracted from *Moringa oleifera* seeds and the fatty de-cake for reuse in composting. The unit prices of the natural coagulants alginate and chitosan are \$12/kg and \$19/kg (Saranya et al., 2014). In Ethiopia, *Moringa stenopetala* seeds are sold on the market and a kilo costs about 40 Ethiopian Birr (USD\$1.40) with the cost of treating water per m³ using the same coagulant being USD\$0.042 (Megersa et al., 2019). On the other hand, the price for alum is about \$0.30–\$0.50/kg (Saranya et al., 2014). The cost of treating water using alum differs, for instance, Keeley et al. (2012) state that it is USD\$0.05 per m³, Phillip (2013) reports that it is USD\$1.50 per m³ whilst Hamawand et al. (2017) state that it is USD\$0.10 per m³.

In the study by Kaji et al. (2020) both natural coagulants (calcium lactate and tannic acid) had higher water treatment costs than chemical coagulants polyaluminium chloride and ferric chloride as shown in Table 5. The cost was calculated based on the coagulant optimum dosage and the cost per kilo of each of the coagulants for 1 m³ of the treated water. According to Kangama et al. (2018), the estimated cost per m³ of treating water using chitosan is USD\$0.025. Table 5 gives the costs associated with using different natural and chemical coagulants.

In the studies examined in this paper, the cost of water treatment using natural coagulants was found to be higher than that of chemical coagulants in certain cases. However, there were also instances where natural coagulants had lower water treatment costs. These findings indicate that, in certain situations, natural coagulants can be economically viable for large-scale use.

6. Coagulation performance of conventional chemical coagulants as compared to coagulants derived from plant extracts

In a comparative study by Gandiwa et al. (2020), the effectiveness of blending extracts of *Cactus Opuntia* pads and *Moringa oleifera* seeds with a chemical coagulant in raw water treatment was examined. The results showed that alum achieved the highest turbidity removal efficiency, reducing turbidity from 28.4 to 2.4 NTU (91.5% removal) at an optimum dosage of 45 mg/L. *Moringa oleifera* achieved a turbidity reduction of 88.7% to 3.2 NTU at a dosage of 50 mg/L, while *Cactus Opuntia* reduced turbidity by 89.4% to 3 NTU. All coagulants achieved turbidities below the WHO guidelines of 5 NTU (WHO, 2017).

Cactus Opuntia extracts were successful in removing turbidity from water through adsorption and interparticle bridging mechanisms (Gandiwa et al., 2020; Nharingo et al., 2015). *Moringa oleifera* seed protein also exhibits coagulating properties based on adsorption (Nouhi et al., 2019). In a study by Tsamo et al. (2021), alum demonstrated the highest turbidity removal efficiency in raw water and slaughterhouse wastewater (64.61%), surpassing rice husk, cypress leaves, and eucalyptus leaves with removal efficiencies of 58.53%, 54.16%, and 28.10%, respectively. However, in a study conducted by Elsayed et al. (2020), it was found that Fenugreek seed extracts achieved a high turbidity removal efficiency of 98% in synthetic turbid water, outperforming alum with a lower efficiency of 85%. The flocculation process using Fenugreek extract likely involved an adhesion mechanism, where the proteins adhered directly to the surface and bridged the kaolin particles, facilitating their flocculation (Elsayed et al., 2020). On the other hand, the coagulation-flocculation process using rice husk was attributed to charge neutralization and particle bridging mechanisms (Huzir et al., 2019).

In a comparative study conducted by Beyene et al. (2016), cactus powder, alum, and their combination were analyzed for their impact on various physiochemical parameters using the jar test. The study found that increasing the dosage of cactus powder from 1000 mg/L to 7000 mg/L resulted in a turbidity reduction ranging from 23.90% to 54.00%. Similarly, increasing the alum dosage from 1000 mg/L to 7000 mg/L led to a turbidity reduction ranging from 28.46% to 58.20%. The use of cactus powder had a minimal effect on the water's pH, with the final pH values remaining relatively unchanged, while alum caused a significant drop in pH to 4.18 (acidic) at a dosage of 7000 mg/L, compared to the initial pH of 7.81. The decrease in pH caused by alum usage in water treatment can deplete alkalinity, requiring additional measures and costs to maintain a neutral pH (Hashim et al., 2013; Napacho & Manyele, 2010). In a study by Beyene et al. (2016), using cactus powder and alum as coagulants resulted in increased total dissolved solids (TDS), conductivity, and salinity of the water sample. Cactus powder increased these parameters by 93.8%, 68.7%, and 97.1%, respectively, while alum increased them by 167.8%, 68.7%, and 180.0%, respectively. This indicates that cactus powder performed relatively better in maintaining TDS, conductivity, and salinity. In another study using alginate as a natural coagulant (Saranya et al., 2022), negligible changes were observed in the pH and conductivity of the water sample.

In the study by Gandiwa et al. (2020), the initial pH of the untreated water was 7.05. With the introduction of alum at a dosage of 60 mg/L, the pH decreased to 5.00. When *Moringa oleifera* and *Cactus Opuntia* were used at the same dosage, they slightly reduced the pH to 6.4 and 6.5, respectively. These pH values obtained with *Cactus Opuntia* fell within the WHO's pH range of 6.5–8.5 for potable water (WHO, 2017).

Table 5
Costs for treating water using natural and chemical coagulants.

Coagulant	Cost of treating water using coagulant per m ³ (USD)	Source of raw water	Country of study	Reference
Calcium lactate	0.009	Zayandehrood River	Iran	Kaji et al., 2020
Tannic acid	0.004			
Poly aluminium chloride	0.0017	Bisalpur dam	India	Solanki et al., 2022
Ferric chloride	0.0024			
Synthesized inorganic polymeric coagulant-medium basicity	0.076	Laboratory-prepared turbid water samples	Turkey	Çoruh, 2005
Inorganic polymeric coagulant-ultrahigh basicity	0.076			
Calcium chloride	0.064	Inlet point of Ankara Ivedik Drinking Water Treatment Plant	Ethiopia	Megersa et al., 2019
Alum	0.008			
<i>Moringa stenopetala</i>	0.042	Synthetic turbid water	South Korea	Kangama et al., 2018
Chitosan	0.025	Raw water from the Han River	India	Thirugnanasambandham & Karri, 2021
<i>Azadirachta indica</i> A. Juss	0.56	Urban waste water		
Alum	1.73			

However, when *Moringa oleifera* was used, the final pH was slightly outside the recommended range. The conductivity of the water increased by 51% with a dose of 60 mg/L of alum. Similarly, the conductivity increased by 38% for *Moringa oleifera* and 36% for *Cactus Opuntia* when the dosage was increased from 0 to 60 mg/L. The increase in conductivity is attributed to the dissociation of the coagulant, generating ions that enhance electrical conductivity. *Moringa oleifera* and *Cactus Opuntia* have a lower dissociation capacity in water compared to alum, resulting in a lesser increase in conductivity for these coagulants (Marobhe et al., 2007).

In a separate study in which both natural and chemical coagulants were used for the treatment of palm oil mill effluent, it was observed that alum had the greatest dose requirement of 4000 mg/L as the optimum whilst *Moringa oleifera* had 2000 mg/L, chitosan 400 mg/L and zeolite 1000 mg/L (Jagaba et al., 2020). The study demonstrated that more alum coagulant was required to treat the sample water as compared to natural coagulants which include *Moringa oleifera*.

From the studies which were analyzed, it was observed that alum had generally higher turbidity removal efficiencies than natural coagulants such as cactus, rice husk, and eucalyptus leaves. However, treatment with some of the natural coagulants results in final turbidity which met WHO guidelines for drinking water quality. Natural coagulants were also generally better than alum in maintaining the physicochemical parameters of the water such as pH, TDS, conductivity, and salinity.

7. Potential for using natural plant extracts for water treatment

Various natural coagulants such as *Moringa oleifera*, *Cicer arietinum*, *Plantago Ovata*, *Maerua subcordata*, and *Carica papaya* have been investigated in various studies and have proved to be able to remove impurities such as chromium, nickel, cadmium, *Escherichia coli*, and turbidity (Saranya et al., 2022; Arshad & Imran, 2020; Megersa et al., 2016). The successes of these studies at lab scale call for further upscaling of use of these coagulants at water and waste water treatment plants. Moreover, an intriguing idea that might improve the coagulation process is the idea of multifunctionality. Coagulants that can remove both dissolved and undissolved pollutants can speed up the treatment process and provide several advantages, including cost savings and reduced environmental footprint. There is also potential to come up with hybrid coagulants by combining natural coagulants with other already existing coagulants to achieve favorable levels of removal efficiency for different contaminants. Furthermore, since these coagulants are obtained from natural sources, the coagulants can be easily accessed (Ang & Mohammad, 2020).

8. Constraints of using natural plant extracts for water treatment

Plant-based coagulants have to go through some processes before they can be utilized for treatment unlike conventional chemical coagulants which are readily available for purchase and use (Kurniawan et al., 2020). The extraction of natural coagulants from plants depends on the characteristics of the plant in question but generally, the processes involve crushing, sieving, dialysis, washing, delipidation, removal of insoluble matter, filtration, and ion exchange (Ali et al., 2010; Okuda et al., 2001). Therefore, this processing which is required every time they are used is a challenge since the conventional chemical coagulants which are being currently used are readily available to use. To make the utilization of bio coagulants/bio flocculants practical and accessible, it is necessary to simplify these complex extraction processes. Future research should focus on preserving the longevity of active chemicals and converting them into powdered form for easier handling and storage (Kurniawan et al., 2020). Moreover, resources that are used as natural coagulant have to compete with other needs for which they have to be used such as basic foods.

Furthermore, the use of these resources as coagulants has to be

sustainable to avoid encountering scarcity challenges in the long-term application (Kurniawan et al., 2020). Therefore, considering the huge amounts of water being currently treated using conventional coagulants, it implies that a vast quantity of the plant extracts has to be exploited in the long-term application and this may not be sustainable in the long run. Plants like *Moringa oleifera* also require processing and storage, increasing expenses and making it difficult to, meet the demand for natural coagulants as a replacement for chemical coagulants (Saleem & Bachmann, 2019). However, to deal with the issue of vast quantities of plant extracts being exploited, large plantations of the plants can be developed such that the plants are produced on a commercial scale and continuously to ensure viability.

Furthermore, another constraint of using natural coagulants for water treatment is that in some cases, these natural coagulants have been proven to be good for high turbidity but limited efficiency has been attained for low turbidity water (Asrafuzzaman et al., 2011). Another challenge which is being faced in the use of natural plant extracts as coagulants is the lack of industrial confidence to adopt natural coagulants for water treatment due to concerns about the cost-effectiveness and performance consistency of the coagulants for large-scale water treatment (Ang & Mohammad, 2020). In addition, most of the performance results of natural coagulants which have been reported have been limited to lab-scale studies and the move to real industrial applications has been hard to come by.

The potential of natural coagulants to perform well under real application challenges such as fluctuating feed water quality, steady supply of natural coagulants with consistent quality, and storage and handling of the coagulants has not yet been established. Natural coagulants are also vulnerable to degradation where the active compounds such as proteins and polysaccharides may age during storage or be altered because of microbial attacks and external environmental factors during extraction and purification of the coagulants (Abidin et al., 2019; Ho et al., 2014). This will create a problem for the long-term storage of natural coagulants as the degradation of active compounds will affect the coagulation efficiency. Consequently, this has critical impacts on the commercialization potential of natural coagulants. This is because the performance of the coagulants would have been affected due to transportation and storage problems for the coagulants (Ang & Mohammad, 2020). This can be addressed by coming up with ways to process the extracts and also adding preservatives which will ensure that the extracts can be stored for a long time without being altered or degraded.

9. Opportunities and drawbacks for commercialization of natural coagulants

The successful commercialization of natural coagulants relies on their ability to maintain treatment performance at a comparable or lower cost than traditional chemical coagulants. However, making direct comparisons of coagulant types, processing stages, and costs across different geographic regions is challenging due to factors such as currency rates, inflation, and cost value accuracies (Freitas et al., 2018).

The commercialization of natural coagulants often faces obstacles due to financial dependence and investor scepticism toward new products. To enhance the development of natural coagulants, it is important to understand the associated risks and weaknesses. However, most published results are limited to laboratory-scale studies, which might not be sufficient for real-time applications (Bahrodin et al., 2021). A breakthrough in this sector can be achieved by emphasizing the combination of natural and chemical coagulants. Approval from regional governing organizations is also necessary for successful commercialization, but the stringent screening and documentation requirements can make obtaining approval challenging. To incentivize water treatment operators to adopt natural coagulants, tax refunds, and subsidy programs could be introduced (Choy et al., 2014).

There is also a need for synergies between the research institutions researching these natural coagulants with the industries and

municipalities which treat water and wastewater so that pilot studies can be done to determine how the natural coagulants perform in a large-scale water or wastewater treatment plant and the operating conditions under which they work.

In addition, providing loans to individuals interested in cultivating plants like *Moringa oleifera* and *Elephantorrhiza goetzei* for natural coagulant production on large areas of land can be considered. The main challenge in implementing plant-based natural coagulants in terms of sustainability lies in the large-scale cultivation of plants to meet industrial demands. Initially, there may be increased industrial costs due to technological adjustments, the development of new tools and machinery, and the establishment of new professional requirements. However, over time, there will be a significant reduction in the process costs and associated pollutants. Moreover, this approach can contribute to job creation since extensive land cultivation will be required for growing these plants (Freitas et al., 2018).

There is potential for these natural coagulants to be commercialized and made available on the market in the same way in which chemical coagulants are readily available on the market. For this potential to be realized, investigations should be done on how to process the natural coagulants so that they are used in powder form and also find ways to make sure that there is a constant supply of the natural coagulants on the market to ensure industrial confidence in the products.

10. Discussion

Various studies have demonstrated that plant-based extracts have been successful in the removal of contaminants such as heavy metals, turbidity, and pathogens in water (Hussain et al., 2019; Shan et al., 2017; Sharma & Bhattacharya, 2017; Megersa et al., 2016;). Turbidity removal efficiencies higher than 90% can be achieved using natural coagulants (Saranya et al., 2022; Asrafuzzaman et al., 2011). However, there is a challenge of lower efficiencies with water of lower turbidity. In the study by Asrafuzzaman et al. (2011), the removal efficiency increased from 86.9% to 94.1% for water with 100 NTU as the coagulant dosage increased from 50 mg/L to 100 mg/L as compared to an increase of removal efficiency from 65.6% to 69.4% as the coagulant dosage increased from 50 mg/L to 100 mg/L for lower turbidity of 48 NTU. Therefore, there is a need for further study on how best water with low turbidity can be treated with natural coagulants. A possible solution would be to spike the water with kaolin clay to increase the raw water turbidity before the natural coagulant is introduced. Pathogens can also be successfully removed using natural coagulants such as *Plantago ovata*, where removal efficiencies of approximately 96.1%, 98.1%, and 98.3% were achieved for fecal coliforms, total coliforms, and fecal streptococcus respectively (Ramavandi, 2014).

To achieve the best pathogen removal results for some of the coagulants, the water has to be of low turbidity. In the study by Yimer and Dame (2021), salt crude extract of papaya seed achieved a higher total coliform removal (88.97%) at a lower initial turbidity of 94.20 NTU than the 87.65% removal efficiency achieved at a higher initial turbidity of 392.70 NTU. Moreover, Marobhe & Sabai (2021) achieved 51.3% removal of total coliforms at initial turbidity of 1000 NTU using *Moringa oleifera* seed whilst Vunain et al. (2019) obtained a higher microbial load (96.65%) at lower initial turbidity of 278 NTU using *Moringa oleifera* seed. Therefore, if the water has higher turbidity, it can be treated in two stages. For the first stage, the natural coagulant is first introduced to reduce turbidity, and further dosing of the coagulant is done in a separate stage where the natural extract will now be working as a disinfectant. Heavy metals can also be successfully removed in water (Arshad & Imran, 2020). Heavy metal removal of up to 89.80% has been obtained for dangerous heavy metals such as lead in water using natural coagulants such as *Carica papaya* leaves (Seetha et al., 2012). Natural extracts can be used in conjunction with conventional methods of removing heavy metals in water such as the ion exchange process and adsorption-based separation. This approach is useful when natural

coagulants cannot reduce the concentration of the heavy metals to acceptable WHO standards.

Natural coagulants can be analyzed based on the dosage required for optimum removal of contaminants to determine whether it will be viable to use the coagulant, especially on a large scale. The active ingredient in the coagulant is also another important aspect of analyzing natural coagulants. Different components within natural coagulants can be responsible for the coagulation process. These include proteins, silica, organic polyelectrolytes, and starch (Oladoja, 2015; Ramavandi, 2014; Sanghi et al., 2002). It is therefore important to be aware of the active ingredient responsible for coagulation as that information can be exploited to enhance the coagulation capabilities of the natural coagulant. The extraction method is also another critical component when analyzing natural coagulants. This is because different extraction methods such as water and salt extraction result in differing coagulation activity and this can be used to design multi-stage experiments (Bouchareb et al., 2021). There are also several other factors to consider when analyzing natural coagulants. These include the nature of the biomass produced and determining whether it is safe to use as a soil enhancer based on its components and toxicity since any residual toxic substances will pose a danger to the health of humans and the environment at large. The practical application of natural coagulants in communities has to be relatively easy so that even people in marginalized areas can easily adopt the technology.

11. Conclusion

The information gathered in this study shows that plant-based extracts can potentially replace conventional chemical coagulants if the challenges which are associated with using the extracts are addressed. Various plant extracts from plants such as *Moringa oleifera*, *Cactus Opuntia*, *Maerua subcordata*, *Cicer Arietinum*, *Dolichos Lablab*, and pine cone have been successfully used to treat raw water samples. *Moringa oleifera* and *Cactus Opuntia* have reduced turbidity to less than 5 NTU which is the WHO standard for turbidity in drinking water. Various natural coagulants have also been used to remove *Escherichia coli*, total coliform, and heavy metals such as copper, zinc cadmium, and lead in water. Reduction efficiencies of up to 90% for heavy metals and up to 99.9% for microbial load have been achieved using natural coagulants. Copper and zinc have successfully been treated to WHO drinking water standards using natural coagulants whilst the standards could not be met for manganese, lead, cadmium, chromium, arsenic, and nickel. These heavy metals for which the WHO drinking water standards were not met would require further treatment such as adsorption-based separation for further removal of the contaminants. In some cases, natural coagulants managed to remove microbial load up to 0 cfu/100mL thus achieving WHO drinking water standards. However, in some cases, the natural coagulants could not achieve total removal of microbial load and will therefore need further disinfection for the water to meet WHO standards for water quality.

Natural coagulants can maintain the pH within the range for portable water which is 6.5 to 8.5, thereby reducing costs for pH correcting which may be needed when using alum for treating water. However, when some of the plant extracts are used at the household level in conventional systems, the crude extract is used, which is not processed. In the crude extract form, the natural coagulants release organic matter and nutrients into the water which will result in the water that would have been treated decreasing its quality in terms of organic matter content. Moreover, many of the studies which have been done are lab-scale studies, with large-scale implementation at water treatment plants and commercialization being scarce.

12. Future directions

More studies are needed to address the issue of the higher levels of organic matter in the treated water when the crude form of natural

coagulants is utilized for household water treatment. This is because the increased organic matter in the treated water can result in color, taste, and odor problems in the treated water and it also facilitates the development of microorganisms in the water when it is stored. Further studies are also required to come up with a way of processing and packaging coagulants from plant extracts so that they can be made available for sale as a packaged product and also to ensure that they can be stored for longer times. It is also important to investigate other plant-based coagulants which have not yet been explored such as *Elephantorrhiza goetzei* and to study the sludge produced from natural coagulants to determine whether the nitrogen and phosphorous content are enough for the sludge to be efficiently used as a soil conditioner. Furthermore, options must be explored on whether the natural coagulants can be commercially cultivated so that there will be a continuous supply of the coagulants to ensure that they can be effectively used on a large scale at water treatment plants.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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