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Assessment and Optimization of Coagulation Process in Water Treatment Plant: A Review

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A B S T R A C T

The rapid growth of the human population, industrialization, and urbanization has threatened the global demand for safe drinking water. Water treatment plant plays a vital role in purifying the raw water for consumer use. The typical water treatment process are coagulation-flocculation, sedimentation, and filtration. Among them, the coagulation-flocculation process is the primary stage in the treatment process. This paper reviews and discusses the optimization strategies of a coagulation-flocculation process to enhance overall treatment efficiency. The working principle of the coagulation-flocculation process is first discussed to understand the treatment process better. Next, the importance of aluminum-based coagulants is addressed as chemical coagulants are one of the key factors that can improve the process. The removals of natural organic matter (NOM) by the coagulation-flocculation process were reviewed as NOM normally contributes to the discoloration of water. The optimization of coagulant dosage was also discussed to depict the consequence of uncontrolled dosage. Finally, dosing control strategies in real-time were discussed, namely direct and indirect dosing control.

1. INTRODUCTION

Surface water is the principal source of drinking water distribution system in the majority of the cases. For the treatment of surface water, the issues are mostly related to turbidity removal, color removal, and disinfection of bacteria (Herman, 1983). Chemical and physical treatments such as coagulation-flocculation, sedimentation, and filtration are used to treat the raw water in a water treatment plant. Among them, being the primary treatment stage, coagulation-flocculation plays a vital role in the control of drinking water quality, specifically particulates, microorganisms, natural organic matter (NOM), synthetic organic carbon, disinfection byproducts (DBPs) precursors, and some inorganic ions and metals (Jiang, 2001). With proper understanding and operational control, this process can improve particulate species separation in downstream processes such as sedimentation and filtration, improving the overall effectiveness of the treatment process.

Coagulation in water treatment is defined as the process of adding a chemical coagulant or coagulants to suspended, colloidal, and dissolved matter for subsequent processing by flocculation or to produce conditions that will allow the particulate and dissolved matter to be removed later. Generally, this process involves three reaction stages: (1) destabilization of small suspended and colloidal particulate matter, (2) adsorption of the colloidal and dissolved NOM to particles, and (3) forming of flocculant precipitate that sweep through the water, entangling tiny suspended, colloidal, and dissolved particles as they settle. Furthermore, during coagulation, factors such as raw water parameter, coagulant type, coagulation pH, and dose of coagulation have been studied to influence the coagulation performance. The optimum combination of these parameters can produce a high-efficiency treatment process (Trinh & Kang, 2011).

Flocculation, on the other hand, is the aggregation of destabilized particles (a lowered electrical surface charge) and precipitation products created by adding coagulants to bigger particles known as flocculant particles or "floc". It is often regarded as one process with coagulation, hence, forming the term coagulation-flocculation. With that, the phases throughout the coagulation-flocculation process can be visualized in **Figure 1** (Kurniawan *et al.*, 2020).
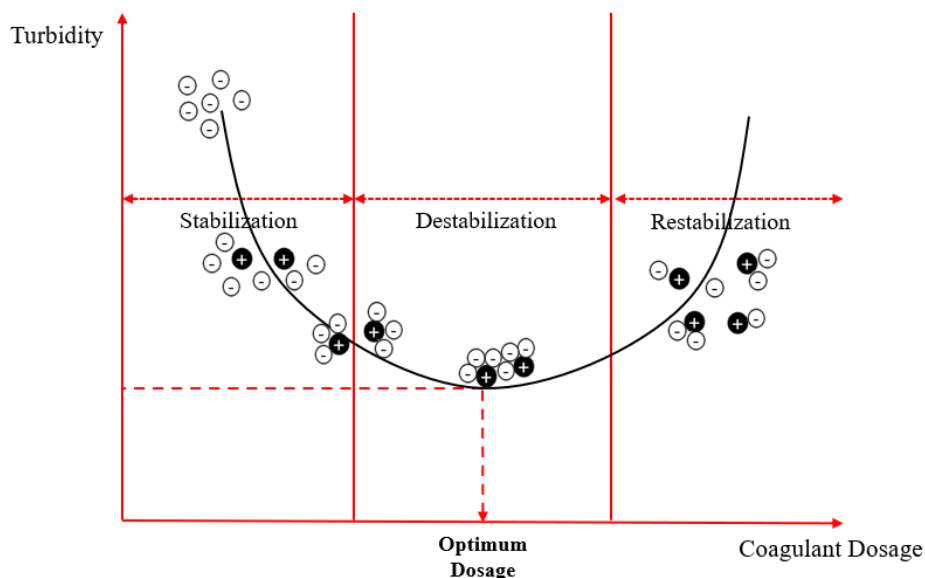

Figure 1. Stages in the process of coagulation-flocculation [6].

In addition, the flocculation process can occur by natural reaction using flocculants and physical processes. Two types of flocculation can be formed through these process mediums: micro and macro flocculation. The former is also known as perikinetic flocculation, where particle aggregation is caused by fluid molecules' naturally random thermal motion. The latter arises when induced velocity gradients cause particle aggregation in the liquid, also known as orthokinetic flocculation. Regardless of its types, the purpose of flocculation is to form particles that can be removed by the next separation techniques such as gravity sedimentation and filtration.

Ultimately, implementing high efficiency and suitable coagulation-flocculation process in a water treatment plant can enhance the overall production of clean water. Numerous methods have been studied and reported in the past decade to improve the removal of contaminants by cost-effective coagulation-flocculation. **Table 1** presents a literature summary on the method to increase the effectiveness of the coagulation-flocculation process.

Table 1. Overview summary method to enhance the coagulation-flocculation process.

Study	Description	Results	Reference
Combining Al and Fe coagulants	Process of blending Al-based and Fe-based coagulants for water purification	- The removal efficiency of DOC and turbidity is 7.3-8.4% and 2.6-14% higher than single coagulants - Improved floc size, floc strength, and dewaterability - Less affected by pH, temperature, and mixing intensity	(Lee *et al.*, 2008)
Rice starch as a natural coagulant	Improve removal efficiency by using environmental-friendly non-autoclaved rice starch	- Turbidity removal efficiency can reach up to 94.8% (at pH = 2.37) - Turbidity removal efficiency is 70.17% when pH = 5	(Usefi & Asadi, 2019)
Use of algal alginate as a coagulant	Employing calcium alginate in treating turbid water	- Highly dependent on the initial turbidity and calcium concentration - Turbidity removal, over 98%, at high initial turbidity water - Use of greater viscosity alginate and longer rapid mixing can improve the performance for low turbidity water	(Devrimci *et al.*, 2012)
Use of composite polyaluminum chloride (HPAC)	Uses a novel composite coagulant for high alkalinity and micro-polluted water	- 30% more efficiency than alum in removing dissolved organic carbon (DOC) and very effective in turbidity removal - Low residual aluminum is produced - It can be used as an alternative to acidify raw water and apply increased doses of hydrolyzing coagulants to improve NOM removal	(Barrows *et al.*, 2018)
Anionic polyacrylamide as coagulant aid	Application of polyelectrolyte to improve the coagulation-flocculation process	- Increased flocculation efficiency and settling speed - Reduced sludge volume by 42% - Reduced amount of coagulant required and reduced cost of coagulation-flocculation process	(Aguilar *et al.*, 2005)

Table 1 (continue). Overview summary method to enhance the coagulation-flocculation process.

Study	Description	Results	Reference
Ballast reaction nuclei-enhanced coagulation	Uses ballasted nuclei, such as silica sand, magnetite sand, anthracite, recycled glass to improve the floc formation	- Potential to treat surface water - Increased the floc density and sedimentation speed of flocs - Improved removal of turbidity in humic-rich raw water	(Cui *et al.,* 2020)
Pump diffusion mixer (PDM)	Uses a different type of rapid mixing technique for the removal of NOM in water treatment	- Uses lower coagulant dosage to remove NOM and turbidity efficiently - Unnecessary for additional chemicals for pH control - Better performance in noise, energy waste, and maintenance cost	(Cui *et al.,* 2020)
Chemical pre-oxidation enhanced coagulation	Process of employing pre-oxidants such as ozone, permanganate, and ferrate	- Favorable in source waters that contain a high level of NOM, algae cells - Ability to destroy the organic coating on the particle surface to change the zeta-potential - Improved formation of flocs	(Xie *et al.,* 2016)
Coupling Ultrasound/Ozone with coagulation	Process of combining US/O_3 with coagulation (either before or after) for water treatment	- Able to increase the removal efficiency of NOM and turbidity - Uses less amount of coagulants - Less production of sludge	(Setareh *et al.,* 2021)
Combination of electrocoagulation (EC) and dead-end micro-filtration (MF)	Pretreatment with iron- and aluminum-based EC to improve NOM removal	- 20% increase in NOM removal - The hybrid process of EC-MF is superior to ultrafiltration - Can mitigate NOM fouling and reduce the energy consumption of filtration	(Setareh *et al.,* 2021)

2. METHODS

In this chemical process, destabilization of colloidal particles is achieved after the addition of hydrolyzing electrolytes such as metal salts, or in other terms, the metallic chemical coagulant and synthetic organic polymer. Several mechanisms will arise within the coagulation-flocculation process with this chemical coagulant present in the water. This section will introduce the principal coagulation mechanism in charge of particle destabilization and removal.

There are four mechanisms involved in this phenomenon: (1) double layer compression, (2) adsorption and charge neutralization, (3) adsorption and interparticle bridging, and (4) enmeshment in a precipitate, or "sweep floc.".

2.1. Double-Layer Compression

Suspended particles present in the surface water are mostly negatively charged ions. With only the same charges in the water, the repulsive energy barrier between particles causes stability. To destabilize the particles, introducing simple electrolytes or metal coagulants, which carries positively charged ions, into the stabilized colloidal water concentration will penetrate the electrical double layer surrounding the particles. In theory, this process, coupled with the micro-flocculation and macro-flocculation in the flocculation process

mentioned earlier, can destabilize the particles by compressing the double-layer. **Figure 2** shows the occurrence between negatively charged particles on the positive ions (frequently called counter ions).
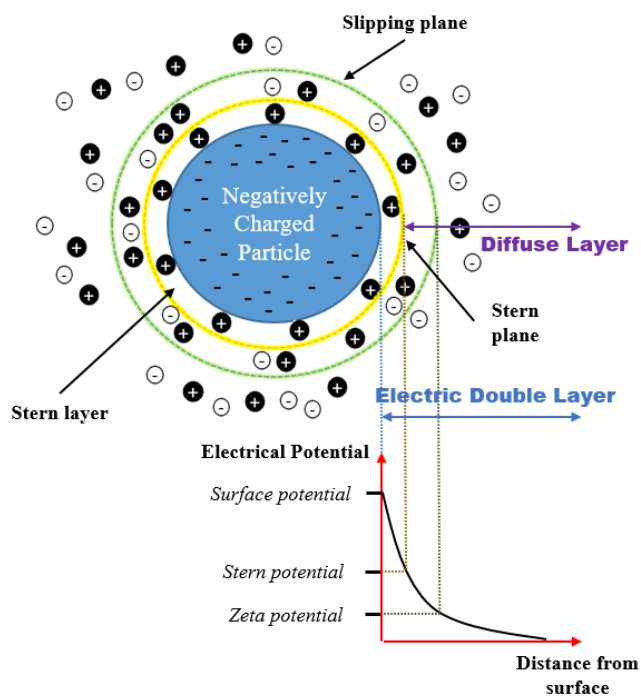

Figure 2. Structure of electrical double layer.

From the diagram, the negative colloid first attracts some of the positive ions, causing them to form a firmly bonded later around the colloid's surface; such layer is known as the stern layer. When more counter ions are attracted to the negative particles, some of them will be repelled by the stern layer or by other counter ions approaching the particles; this results in forming a diffuse layer of counter ions. Consequently, the double layer comprises attached counter ions in the stern layer and the charged atmosphere in the diffuse layer. Moreover, the zeta potential measures the electrical charge of particles suspended in the liquid. Here, bringing the zeta potential between -30mV to +30mV is a good rule of thumb for aggregating particles.

Additionally, particles electroneutrality can be achieved at a shorter distance if there are more ions in solution or if the ions have a higher charge (divalent or trivalent instead of monovalent). With increasing ionic concentration, the double layer compression will also increase and weakens the repulsive forces that separate the particles and allow for coagulation due to van der Waals forces. However, promoting the ionic strength to reduce the thickness of the double layer is not realistic for destabilizing particles in water because the required ionic strength is greater than those regarded as acceptable in drinkable water. Therefore, coagulating chemicals are required to destabilize the particles.

2.2. Adsorption and charge neutralization

The ability of a chemical agent to destabilize and coagulate suspending particles is, for all intents and purposes, the outcome of a mixture of mechanisms. In the adsorption and charge neutralization route, the charge on the particles is neutralized, and electrostatic repulsion is reduced or removed, resulting in particle destabilization and, as a result, agglomeration

happens (**Figure 3**). If the particle surface has no net charge, there will be no electrical double layer, and the van der Waal forces will cause particles to stick together.

Figure 3. Schematic diagram of charge neutralization.

Following the same aspects, the foundation of charge neutralization occurs in a 'patch-wise' fashion. The adsorption between high charge density electrolytes (coagulant) and low charge density colloidal particles occurs (**Figure 4**). This is known as the electrostatic patch mechanism, which refers to patches of positive and negative regions on the particle's surface (Amran *et al.,* 2018). Based on this mechanism, a lower coagulant dosage is required for water treatment with a higher charge density (Kurniawan *et al.,* 2020). It is also important to note that overdosing of coagulant can cause the colloid's charge to be reversed and redistributed as a positive colloid, or in other words, re-stabilization occurs.
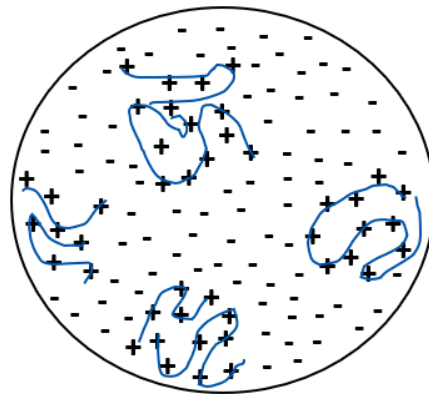
Figure 4. Model of an electrostatic patch in charge neutralization mechanism.

2.3. Adsorption and Interparticle Bridging

In this mechanism, the inter-particle bridging instability occurs when polyelectrolytes with highly active surfaces and linear or branching structures are used as a coagulant aid to increase the aggregation of micro-flocs during the flocculation process (Ghernaout *et al.,* 2020). Technically, polymer chain adsorb on the surface of particles at one or more locations is due to (1) coulombic (charge-charge) interactions, (2) dipole relation, (3) hydrogen bonding, and (4) van der Waals forced of attraction. The remainder of the polymer chain extends away into the water. It gets absorbed on another available particle surface, creating a "bridge" binding them together and forming a larger particle that can subside more effortlessly, as shown in **Figure 5**.

Figure 5. Schematic diagram of bridging mechanism.

Furthermore, the presence of enough unoccupied surface on a particle for binding polymer chains segment adsorbed on other particles is crucial for polymer bridging (Bolto & Gregory, 2007). Suppose no other particle is accessible or there is an excessive polymer. In that case, the original particle will be extensively covered by the free extended parts of the polymer molecule, thus re-stabilizing the colloid.

Other than the aforementioned conditions, factors such as molecular weight (MW) of polymer, charge density (CD) of polyelectrolytes, and ionic strength will also influence the process of polymer bridging. In sum, a study has stated that (i) the most effective polymers are linear polymers with a high MW, (ii) only a small amount of adsorbed polymer is required, and excessive quantities can cause re-stabilization, (iii) an optimum CD is desirable for polyelectrolytes, (iv) the presence of specific metal ions, as well as ionic strength, may play a role, and (v) the bridging process creates very strong flocs; however, damaged flocs may be irreversible (Bolto & Gregory, 2007).

2.4. Enmeshment of particles in the precipitate, or "sweep floc"

When high enough dosage of coagulants such as alum $[Al_2(SO_4)_3]$ or ferric chloride $(FeCl_3)$ are added, an insoluble precipitate will form, and raw water colloid becomes enmeshed in the amorphous precipitates. This is one of the most common methods to destabilize a turbid suspension and is usually called sweep coagulation, as shown in **Figure 6**.
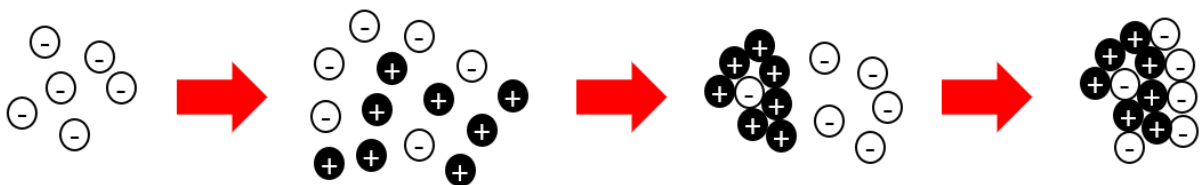

Figure 6. Schematic diagram of sweep coagulation mechanism (Lambert *et al*., 1999).

Theoretically, sweep floc is a non-selective process that aggregates colloidal size particles (Ghernaout & Ghernout., 2012). With this characteristic, the addition of a large number of precipitate particles can improve the likelihood of colloidal interaction with raw water colloid and promotes floc formation. This is particularly advantageous in low turbidity water (Lambert *et al*., 1999). Moreover, this type of mechanism is most prevalent in water treatment applications where the pH is kept between 6 and 8 (neutral), and the coagulant salts (Al or Fe) are utilized at concentrations concerning the produced amorphous metal hydroxide solid, typically higher than adsorption. This shows that this mechanism has a higher coagulant demand, which inevitably raises chemical costs and results in thick sludges that are more difficult to dewater (Lambert *et al*., 1999).

On the other hand, sweep coagulation has a clear advantage in process control compared to destabilization by adsorption and charge neutralization. It is more insensitive to the deviations from the optimal dose conditions, especially for overdosing. According to a study, this mechanism has three influencing factors: oversaturation, anions, and colloids concentration. For oversaturation, a higher concentration of insoluble precipitate is required to obtain fast precipitation and effective sweep coagulation. Subsequently, different anions in water, particularly sulfate ions, will improve the precipitation rate. Finally, the higher the concentration of colloids, the better the precipitation rate as the colloids could serve as nuclei for precipitate formation.

3. RESULTS AND DISCUSSION
3.1. Aluminum-Based Coagulants

The coagulant types used are also one of the determining factors in the overall effectiveness of the coagulation-flocculation process, other than coagulant dosage and pH control. In the context of this research paper, aluminum sulfate will be the main focus as it is the standard coagulant used in the studied WTP. Other aluminum-based will also be discussed for exploratory purposes.

Aluminum sulfate ($Al_2(SO_4)_3$), often known as alum, is the most commonly used aluminum-based coagulant in drinking water production as it can improve the coagulation process for the removal of particulate, colloidal, and dissolved substances. In addition, alum comes in a variety of solid grades, including block, kibbled, and ground, as well as in solution form. Other similar inorganic aluminum-based coagulant includes aluminum chloride ($AlCl_3$) and sodium aluminate.

When alum is added into the water solution, they react and produce aluminum hydroxide ($Al(OH)_3$) and sulfuric acid as by-products. Then, the created sulfuric acid reacts with the alkalinity in the raw water to produce carbon dioxide, thus lowering the pH. Consequently, it is frequently necessary to add alkalinity to the raw water. Moreover, it has been reported that increasing the alum dose only increases the removal of contaminants such as NOM to a certain extent. The use of alum can leave relatively large aluminum residuals in the final treated water, especially during low pH levels or low temperature, which might pose a health risk or produce other problems in the distribution system, such as spontaneous flocculation (Cui *et al*., 2020; Bolto & Gregory, 2007).

Although surface water treatment using alum has been practiced worldwide for over a century, the procedure of removing residual aluminum in the treated water is not being emphasized enough. Thus, using aluminum salts as a flocculant to filter drinking water has long been criticized (Chao *et al*., 2020). According to the Environmental Protection Agency (EPA), a Secondary Maximum Contaminant Level (SCML) of 0.05-0.2 mg/dm3 for aluminum in drinking water has been recommended (Krupinska., 2020). The World Health Organization (WHO) has also set a limit of 0.2mg/dm3 of residual aluminum in drinking water. In the circumstances where the residual aluminum concentration exceeds these limits, health risks such as presenile dementia and Alzheimer's disease may arise (Krupinska., 2020). Not only that, but a high concentration of residual aluminum also increases turbidity, reduces disinfection efficiency, and decreases the volume capacity of the water distribution system (Driscoll & Letterman, 1995).

On account of this, several alternatives of aluminum-based coagulants have been proposed to treat water while producing less residual aluminum. One of the options is the use of inorganic-organic composite coagulants, PACPE, which is the combination of a cationic polyelectrolyte (p-DADMAC) and an inorganic pre-polymerized coagulant (Tzoupanos & Zouboulis, 2010). Another study states that pre-hydrolyzed aluminum coagulants are more effective than non-prehydrolyzed coagulants such as aluminum sulfate or sodium aluminate in treating water intended for human consumption (Krupinska, 2020; Nowacka *et al*., 2014).

Pre-hydrolyzed aluminum coagulants, for example, polyaluminum chloride (PACl) and polyaluminum sulfate (PAS), have been produced and investigated in recent years [31]. PACl, where its uses are steadily increasing, is made by partially neutralizing AlCl3 to various basicity ratios. Additionally, pre-hydrolyzing AlCl3 increases the amount of Al13 in the coagulation process, which has been identified as the most effective Al-species for contaminant removal because of their bigger size and higher positive charges (Matilainen *et al*., 2010; Hu *et al*.,

2006). **Table 2** presents the comparison between inorganic salts (Aluminum sulfate), pre-hydrolyzed coagulant (PACl), and inorganic-organic composite coagulants (PACPE).

Table 2. Overview comparison between three aluminum-based coagulants (Adapted from (Matilainen *et al.*, 2010)).

Aluminum-based coagulants	Pros	Cons
Aluminum sulfate or Alum ($Al_2(SO_4)_3$)	• It is stable, simple to handle, and quickly dissolving • In many circumstances, turbidity removal is superior to ferric salts. • In low dosages, it may be more effective than ferric. • Better color efficiency can be achieved.	• In some circumstances, the finished water contains relatively high coagulant (aluminum) residuals. • It carries potential health risks such as Alzheimer's disease. • Consumption of alkalinity is high. • Corrosivity is increased by sulfate in finished water.
Polyaluminum Chloride (PACl)	• It depends less on temperature and pH when compared to alum salts. • Alkalinity consumption is not high. • In many circumstances, it has a higher capacity for removing NOM than alum. • Less sludge produced and a lower dose requirement. • Less residual aluminum in treated water.	• Coagulant hydrolysis species have a major impact on coagulant efficacy. • During coagulation, pre-hydrolyzed Al-species are stable and cannot be further hydrolyzed.
PACPE	• Charge neutralization is improved, resulting in increased dissolved organic carbon (DOC), (SUVA), and turbidity removal capability. • Less residual aluminum in treated water.	• It is mostly still at the development stage. • They are not being utilized widely yet.

The table shows that alternatives for aluminum-based coagulants are available other than the conventional alum. PACl has been extensively studied in removing humic acid (HA), a kind of NOM (Wu *et al.*, 2020; Saxena *et al.*, 2019; Musteret *et al.,* 2021). Despite all that, raw water characteristics still acquire a significant contributing factor in choosing a proper coagulant (Matilainen *et al.*, 2010). This section serves as an introductory purpose to a water treatment plant that is in pursuit of different coagulants to improve the plant operation.

3.2. Removal of Natural Organic Matter (Nom) By Coagulation-Flocculation

To deal with NOM in the coagulation-flocculation process, it is imperative to acquire knowledge about their characteristics, content, and type beforehand. According to *Matilainen et al.* (2021), Musteret *et al.* (2021), Sillanpaa *et al.* (2017), the definition of NOM is generally similar. In this regard, this paper defined NOM as a complex matrix of heterogeneous hydrophobic and hydrophilic substances that naturally form in water sources due to various hydrological, biological, and geological interactions. NOM has been found in most water sources, including lakes and rivers (Hussein *et al.*, 2018). As a result, the amount,

type, and characteristics of NOM vary greatly in waters of various origins can be influenced by the biogeochemical cycles of the surrounding habitats (Matilainen *et al.,* 2010).

The characteristics of NOM are mainly categorized based on hydrophobicity or hydrophilicity, molecular weight (MW), and charge of the functional groups. This characterization can be made by resin adsorption, size exclusion chromatography (SEC), nuclear magnetic response (NMR) spectroscopy, and fluorescence spectroscopy (Loganathan *et al.,* 2020). In particular, by using fractionation techniques, the mixture of organic compounds of NOM can be divided into hydrophobic fractions (humic substances) and hydrophilic fractions (non-humic substances) (Alfaro *et al.,* 2016). Among the two fractions, biopolymers such as proteins-peptides, nucleic acid residues, carbohydrates, and lipids are considered to be non-humic substances (Chow *et al.,* 2009). In contrast, humic and fulvic acid, high in aromatic carbon, phenolic structures, and conjugated double bonds, make up the majority of hydrophobic NOM (Matilainen *et al.,* 2010). Moreover, humic and fulvic acids are the most abundant in NOM as they account for 53-68% of dissolved organic carbon (DOC) in natural water bodies (Wu *et al.,* 2020).

Although NOM itself is not poisonous, its presence in drinking water sources is extremely harmful because NOM alters the organoleptic properties (taste, odor, and color) of potable water and causes public health concerns with the formation of disinfection by-products (DBP) (Varjani., 2017; Hussein *et al.,* 2018). Owing to this, the removal of NOM in drinking water treatment is of great importance and cannot be overlooked. With that said, throughout the history of water treatment, coagulation-flocculation is the most prevalent and economically practical conventional technique for removing NOM (including particle matter), followed by sedimentation/flotation and sand filtration (Musteret *et al.,* 2021).

From a scientific viewpoint, the presence of NOM has impacted significantly on coagulation chemistry. Conventionally, coagulation has been used in water treatment to reduce turbidity, improve color conditions, and remove pathogens (Varjani *et al.,* 2017). However, it should be noted that optimal turbidity or color removal does not necessarily occur under the same condition as NOM removal. To address this, one of the most feasible and cost-effective ways in NOM removal is the utilization of enhanced coagulation at existing coagulation plants. Here, enhanced coagulation refers to increasing the amount of coagulant dosage or adjusting the coagulation pH to maximize the removal of NOM from drinking water sources (Alfaro *et al.,* 2016; Hussain *et al.,* 2019).

Furthermore, coagulant dosage is more dependent on the presence of NOM in water than turbidity (Pernitsky & Edzwald., 2006). Naturally, this shows that NOM will affect the amount of coagulant dose to a great extent. When a coagulant is added into the water, it removes dissolved-phase NOM through complexation reactions followed by a phase change. The NOM is removed from the solution by becoming a solid or absorbed onto a solid (Pernitsky & Edzwald., 2006). That being said, the ideal coagulant dose for water containing high-molecular-weight NOM, such as humic substances (hydrophobic), is likely to be low as the removal mechanism is mainly charge neutralization.

On the contrary, the optimal coagulant dose is substantially larger if the water comprises low-molecular-weight NOM such as non-humic substances (hydrophilic) with the removal mechanism involving adsorption onto metal hydroxide surface. It is also reported that the hydrophobic fraction of NOM can be removed more efficiently than the hydrophilic fraction by coagulation [50]. In addition, regarding the coagulation mechanism for NOM removal, charge neutralization mechanism and sweep mechanism are two frequently described coagulation processes, among other mechanisms. Between these two, the sweep mechanism

was reported to be the best coagulation mechanism for NOM removal due to the charge density of the humic substances (Alfaro *et al.*, 2016).

Apart from the mechanism, the efficiency of coagulation-flocculation to remove NOM from water in the real-world application is primarily determined by the coagulant type and dosage, pH value, mixing conditions, water temperature, and flocculant type and dosage, on the one hand, and the properties of NOM, on the other (Musteret *et al.*, 2021). Besides, a few general parameters, such as dissolved organic carbon (DOC), UV/Vis absorbance, and specific UV absorbance (SUVA), may be utilized to analyze the type of NOM better and to monitor these compounds during technological processes effectively (Musteret *et al.*, 2021).

3.3. Optimization of Coagulant Dosage

Coagulation, being the primary step in conventional treatment, must be properly and effectively controlled to produce decent quality water. One of the most significant factors to consider when determining the best conditions for the coagulation-flocculation process is the coagulation dosage (Saritha *et al.*, 2017). The optimal coagulant dosage is the least amount of coagulants necessary to achieve the desired treated water quality.

Over the years, as water quality standards become stricter, optimizing coagulant dosage in the water treatment process becomes more important. Naturally, improving water quality is a key priority. However, other objectives could include the ability to intensify water production while maintaining water quality, lower chemical costs and other operating costs such as backwashing or sludge handling costs, improve sludge properties or reduced sludge volume to ease the management of solids, reduce operator attention and hence, allow them to conduct other tasks, or complete automation of plant operation during certain hours (Dentel, 1991).

Theoretically, the control of coagulant dosage depends on several parameters. For instance, raw water quality includes colloidal concentration, pH, alkalinity, hardness, temperature, NOM concentration, and ionic strength. Furthermore, the treated water quality is often being tested to determine the suitability of added coagulant dosage (Kim *et al.*, 2006). For these reasons, controlling coagulant dosage in the large-scale treatment plant is usually faced with various challenges that hinder the overall treatment efficiency.

In general, the coagulant dosage control is somewhat straightforward when the raw water quality is stable. However, this is often not the case, especially for water sources from the river. The coagulant dosing is associated with raw water's chemical and physical features. Thus, the diurnal, seasonal, and storm-related changes in raw water quality pose a considerable difficulty in controlling the dosage (Dentel., 1991; dos Santos *et al.*, 2017). Apart from the irregular variation in the raw water quality, other challenges include chemical reagent stock depletion, system failures, and errors from plant operators (Bella *et al.*, 2014).

If the dosage is not controlled or cannot be controlled, not only will the coagulation-flocculation process be affected, but its subsequent process (sedimentation, filtration, and chlorination) may also suffer undesirable consequences. For this aspect, more information will be discussed in the next section. Typically, the uncontrolled dosage can be classified into two scenarios: underdosing and overdosing. The former indicates that there is insufficient coagulant dosage, while the latter suggests that there is an excessive amount of coagulant dosage in the treatment process. By measuring the degree of removing particles that causes turbidity in water, a report has studied its relationship with the amount of coagulant dosage. According to Figure 7, four zones have been identified with increasing coagulant dosage from zone 1 to 4.
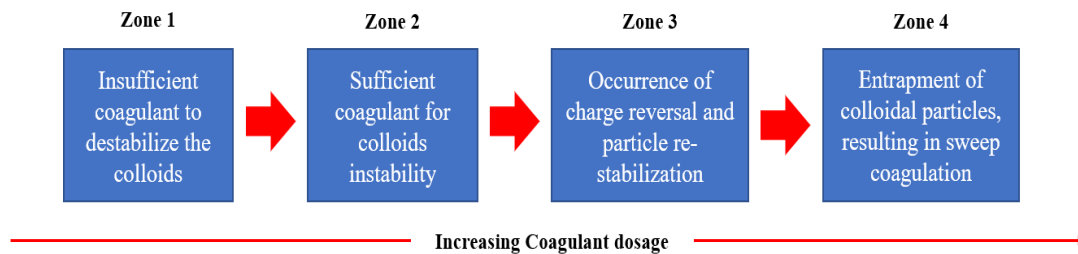
Zone 1		Zone 2		Zone 3		Zone 4
Insufficient coagulant to destabilize the colloids	➡	Sufficient coagulant for colloids instability	➡	Occurrence of charge reversal and particle re-stabilization	➡	Entrapment of colloidal particles, resulting in sweep coagulation

Increasing Coagulant dosage ⟶

Figure 7. Reactions with different amounts of coagulant.

Zone 1 presents the reactions during underdosing, resulting in the inability to reach the water quality target (Farhoui & Derraz., 2016) and less efficient operation of the WTP (Gagnon *et al.*, 1997). Conversely, Zone 3 and 4 both demonstrate the reactions related to overdosing. Due to these reactions, the removal efficiency of turbidity decreases by increasing the coagulant dosage (Saritha *et al.*, 2017).

Aside from affecting the process reaction, the uncontrolled dosage can also influence the WTP operational cost and raises public health concerns (Gagnon *et al.*, 1997). Concisely, chemical costs have accounted for up to 20% of total operational costs, and some reports suggest that operational costs are roughly equivalent to coagulant costs. An example is that overdosing in the coagulation-flocculation process could produce unnecessarily large amounts of sludge, resulting in additional sludge treatment costs. Moreover, overdosing could also decrease the pH in treated water, which increases the risk of corrosion in the water transport system. Therefore, to tackle this issue, more state-of-the-art approaches in dosing control should be studied and reviewed thoroughly to enhance the coagulation-flocculation process.

3.4. Approaches for Coagulant Dosage Control in Real-Time

Considering the complex physical dynamics and relationship between control parameters and dosage in the coagulation-flocculation process, no comprehensive or universally recognized mathematical process has been developed so far. Despite that, more process concepts for dosage control based on online-measurable water quality parameters are emerging (Ratnaweera & Fettig., 2015). In this section, two approaches that use online parameters for dosing control have been identified, and they are: direct dosing control and indirect dosing control.

In direct dosing control, physical sensors are utilized to control the dosage concerning the influence of water quality. Generally, there are three direct control approaches: (1) Feed-forward control based on raw water quality, (2) Feed-backward control based on dosed and treated water quality, and (3) Feedforward-feedback control approach.

3.4.1. Feed-Forward Control Based on Raw Water Quality

This control strategy involves altering the number of chemical coagulants introduced to a process stream in response to the sensory data obtained from the raw water parameter [61]. This type of control is also known as an open-loop system (Liu & Ratnaweera., 2016). Conventionally, settled/treated water quality changes, among other parameters, were mainly used to set the coagulant dosages. This type of dose management is reactive, and it took hours to recover performance if the settling water turbidity began to rise. On the contrary, utilizing the feed-forward (FF) coagulant dose prediction would shift the control strategy from reactive to proactive, preventing any turbidity from rising in settling water (Fabris *et al.*, 2013). **Figure 8a** illustrates the basis of feed-forward control.

In theory, an FF control approach can react quickly to any measured disturbing variables (raw water qualities) via manipulated variables (coagulant dose), with controlled variables (treated water qualities) responding accordingly (Liu & Ratnaweera., 2016). Therefore, a practical relationship between raw water characteristics and the ideal coagulant dose for each location is required to produce an effective FF control approach. Data from jar tests, pilot-plant runs, or technical-scale operations must be accessible for this purpose. They must include all parameters fluctuations that may occur for at least a few months, or ideally a year (Ratnaweera & Fettig., 2015). In addition, since the NOM concentration and characteristics have a significant impact on coagulant dosage and coagulant efficiency, the FF dosing control can be established on online measurements of UV absorbance spectrophotometers or fluorescence spectroscopy, and TOC in raw water (Ratnaweera & Fettig., 2015).

3.4.2. Feed-Backward Control Based on Dosed and Treated Water Quality

In a feedback (FB) control system, the output is determined by the generated feedback signal. More specifically, the feedback control system processes feedback signals, which are then used as inputs by the system. For the drinking water treatment plant, the changing raw water quality and water production rate fluctuations depending on consumer demand patterns in a 24-hour operating cycle and rising chemical costs have encouraged the need for a feedback control system for continuous coagulant dose control (Sibiya., 2014).

The application of the feedback control approach generally involves using sensors such as streaming current detectors (SCD) to measure the residual charge on colloidal, color, and turbidity of particles in the water, comparing the process values to the setpoint and adjusting the coagulant dose pump accordingly to rectify any divergence from the intended results (Sibiya., 2014; Bello *et al.,* 2014). The SCD, which can be used as automated coagulant control (Sibiya., 2014), consists of a sensor and a signal processor. The effectiveness of SCD depends on the charge neutralization mechanism. As discussed in section 2.1.1, this mechanism forms neutralized colloidal particles when cationic coagulant is added into the raw water, which typically contains negatively charged colloids. Since the neutralized colloidal particles do not contribute to the ion charge, the overall net charge is more positive. Correspondingly, SCD controls this net charge at a set point that has been proved to deliver close to optimum coagulation under various raw water conditions in jar testing (Sibiya., 2014).

A feed-backwards (FB) control strategy can be proposed by employing SCD as a direct method to monitor dosed water quality. A feed-backwards (FB) control strategy can be proposed, as shown in Figure 8b. Nonetheless, one of the most important aspects of current streaming monitoring is that the signal fluctuates with pH and that abrupt changes in flow or poor chemical mixing can result in unstable signals. Decoupling control strategy based on this statement, which decreases the interaction between pH and coagulant dosage loops. It follows that this strategy was found to be less susceptible to disturbances.

Another feed-backward control approach is based on the treated water quality. **Figure 8c** shows the principle of this type of control approach. In practice, the treated water characteristics are rarely used for real-time control, despite being one of the most important criteria for optimal coagulation (Ratnaweera & Fettig., 2015). This can be attributed to the system delay or dead time between dosing and effluent from the separation stage. Consequently, this control approach may not function properly, resulting in under- or over-dosing chemical coagulants, especially during seasons when raw water quality often varies widely (Bello *et al.,* 2014).

3.4.3. Feedforward-Feedback Control Approach

According to a study, the results indicate that the feed-forward control system cannot respond to unpredicted treated water qualities during heavy rain. In addition to that, the FF control system is less competent in handling circumstances involving unmeasured disturbance, resulting in unexpected quality of treated water. On the other hand, the FB control system can correct the unmeasured disturbance and incorrect dosage [68]. It can alter a measured error between the setpoint and controlled variables (Liu & Ratnaweera., 2016). As a result, it is critical to combine these two control strategies' benefits to improve coagulation performance. **Figure 8d** shows the principle of combining FF and FB control systems.

With the combination of the two control systems, the dosing control approach has performed better than those based on the FF control system only. The test results by Liu & Ratnaweera, (2016) demonstrate that the dosage adjustment of the FF-FB control system improves the system capacity, ranging from 66% to 197% compared to FF control. With this flexibility, the coagulant consumption can be further decreased in the range of 3/7% to 15.5%, producing a more stable treated water quality while avoiding the possibility of overdosing.
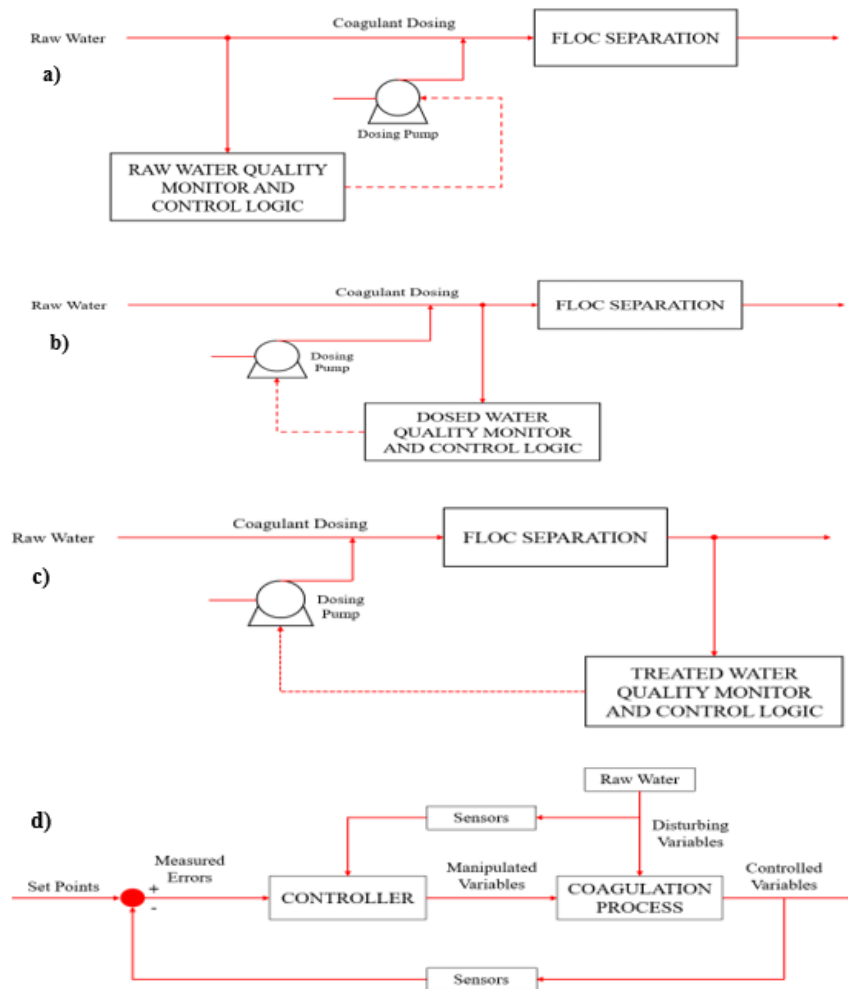

Figure 8. Schematic diagram of (a) feed-forward control approach based on raw water quality, (b) feed-backward control approach based on dosed water quality, (c) feed-backward control approach based on treated water quality, and (d) Feedforward-Feedback control approach.

Furthermore, the system performance can be enhanced by data with more precise dosages because the empirical model used in this system can significantly construct a relationship between variables from historical data Liu and Ratnaweera, (2016). However, due to the long hydraulic retention time of conventional sedimentation tanks, the treated water sensor is always several hours late in delivering feedback information, thus limiting the use of the FF-FB control approach. Besides, the FF-FB system's reliability is strongly dependent on the operational condition of online instruments, which can malfunction and go out of service. Despite that, a silver lining that still encourages applying the FF-FB control approach is that, in most situations, the inlet (raw water) quality variations are measured in hours and days rather than minutes Liu and Ratnaweera, (2016). In summary, combining feed-forward and feed-backward control systems has proved its potential to be a superior dosing control strategy. However, a more innovative approach should nullify its existing limitations.

3.4.4. Approach Control

The other control approach is indirect dosing control. Over the years, advanced mathematical methods have been used to assess data from jar testing, pilot-plant tests, and full-scale operation in a variety of studies to determine the connections between many input variables and one or more output parameters. The resulting correlations are sometimes called models, although they are based on a mathematical analysis of current data rather than a physical knowledge of the process (Ratnaweera & Fettig., 2015). In a nutshell, there are three primary approaches to consider: (1) Artificial Neural Network models (ANN), (2) Multivariate Regression (MVR) analysis, and (3) Fuzzy Logic models.

Long water quality data and operational parameters should be accessible for model calibration and verification to utilize the above models. However, the model's practical application is still limited due to the need for large data based on water and process-related factors for data analysis.

3.4.4.1. Artificial Neural Network Models (ANN)

The artificial neural network (ANN) is a good estimator of the nonlinear relationship between the numerical data input and output. ANN is, essentially, a network made up of artificial neurons that are interconnected and attempt to replicate the human brain's problem-solving abilities (Joo *et al*., 2000; Baxter *et al*., 2001). Three types of models can be distinguished based on their architecture: multilayer perceptron (MLP), time-delay neural network (TDNN), and radial basis function (RBF) neural network (Ratnaweera & Fettig., 2015).

The ANN model is divided into three networks: calibration, validation, and simulation (Leon-Luque *et al*., 2016). Taking turbidity as a parameter, the calibration network developed a pattern of coagulant dose behavior for each level of initial turbidity that tends to rise as the value of initial turbidity increases. The validation network compares the jar test's coagulant dose values to the Validation Network simulated values. If it demonstrates that both variables contain the same behavior, the network has already trained and adapted the coagulant dose pattern for each turbidity level. Lastly, the simulation network revealed the coagulant dose to apply to the random initial turbidity entered into the network previously. If the difference between the coagulant dose and the dosage acquired with the jar test is minor, it proves the ANN model to be an effective control strategy (Leon-Luque *et al*., 2016).

Since the ANN model is data-driven, it has significant advantages over traditional modeling methods when employed to the drinking water treatment plants (DTWPs) (Maier *et al*., 2004). Moreover, ANN is failure-tolerant as it can efficiently adjust to data discontinuities, variable levels of data precision, noise, and scatter of data (Joo *et al*., 2000 & Baxter *et al*., 20010. For

these reasons, ANN has been widely used to model the coagulant dose prediction approach and to ease process control and automation in WTPs (Baxter *et al.,* 2001). An example study by van Leeuwen et al. (1999) presents that ANN has been developed to predict the optimal coagulant (alum) doses based on jar tests on surface water collected in southern Australia. Another study by Zhang and Stanley (1999) and Baxter et al. (2001) shows that a process control system based on the ANN model to remove NOM is a possible dosing control technology and can aid in water treatment cost savings.

Concerning the shortcomings of the ANN model, there is very little information about the model's applicability to data outside the domain in which they were trained. In addition, there is no standard process for generating ANN models; therefore, each model may use different modeling strategies Baxter et al. (2001). More importantly, owing to the high dependency on data, ANN is only well suited to process that contains a large amount of data. More research efforts are required to address the existing limitations of the ANN model to encourage usage in the industry.

3.4.4.2. Multivariate Regression (MVR) Analysis

Multivariate regression (MVR) analysis or multivariate statistical analysis is a control technique that deals with data containing several measurements of variables or objects. This type of control technique is progressively gaining popularity in studying complex data. It can provide analysis when there are multiple dependent or independent variables, all with different degrees of relationship with one another. Additionally, the MVR approach generally includes multiple linear regression (MLR), a powerful chemometric method such as principal components analysis (PCA), and projection to latent structures like partial least squares (PLS).

Several studies have proposed the utilization of MVR analysis. Set of regression models for predicting coagulant (alum) dosages in coagulation (turbidity and color removal) and enhanced coagulation (NOM removal) processes. This model was further validated by (Staaks et al.,2011) when they used it to estimate starting coagulant dosages in a coagulation analysis and optimization research. In another study to optimize the coagulation process in WTPs, Trinh and Kang [80] suggested quadratic regression models based on response surface methodology (RSM). The dependent variables in the model were turbidity and total organic carbon (TOC), while the independent variables were coagulant (alum) dose and coagulation pH. The constructed model was evaluated using statistical indices, and the model was found to be adequate, with the expected response being extremely near the experimental results. Further, a slightly different approach was adopted by Joo et al. (2000), where MVR analysis was used to confirm that the ANN's prediction capability has improved. Regardless, these statistical methods do not reflect the system's dynamic reaction and, thus, in some cases, may not be suitable for autonomous coagulation control in WTPs (Bello *et al.,* 2014).

3.4.4.3. Fuzzy Logic Model

Fuzzy controllers or modeling have found various applications in the engineering process over the last 30 years. They work by taking real-time data as input and utilizing a series of logic rules (if-then) to generate an output signal that keeps the parameters as close to the specified setpoint as possible. More specifically, the input variables are fuzzified using a membership function, which is a curve that translates input variables to membership grades between the range of 0 and 1 (Bello *et al.,* 2014), where 0 means the statement is completely false, and 1 means the statement is completely true.

These conditional statements of fuzzy modeling effectively describe the indistinct method of human thinking required to make decisions in uncertain and imprecise situations.

Essentially, Mamdani and Takagi-Sugeno are the two popular techniques for fuzzy conditional statements (Bello *et al.*, 2014). For Mamdani fuzzy rules, both the antecedent (if-part) and consequent (then-part) are stated in terms of fuzzy sets. An example is: "if the coagulant flow rate is low, then the surface charge is high". Here, the membership functions' language values are low and high, and surface charge and coagulant flow rate are linguistic variables. Conversely, the fuzzy rule of Takagi-Sugeno contains only fuzzy sets in the antecedent section, while the consequent part is written as a constant, linear, or nonlinear input variables equation (Bello *et al.*, 2014).

With a deeper understanding of these conditional statements, they are the building components of the fuzzy inference system. Accordingly, as a fundamental part of fuzzy models, this system produces the final results. **Table 3** shows the step used in the fuzzy inference system.

Table 3. Overview Process of Fuzzy Inference System.

Steps	Description
1	Compare the input variables with the membership function to determine the membership grade of each linguistic value on the antecedent section
2	Establish the firing strength or weight of each fuzzy rule by combining the membership grades on the antecedent section
3	Calculate each fuzzy rule's qualified consequent as a function of firing strength
4	Combine the qualified consequent to create a single-valued output

It has been observed that the use of the fuzzy model is often coupled with other control strategies such as feedforward, feedback, and ANN model. For example, in a DWTP in Taiwan, Chen and Hou (Chen & Hou., 2006) designed a feed-forward control system with a fuzzy feed-backward component. The fuzzy control rules were established using full-scale data collected over four years, and pH and turbidity in treated water were used to create the models. By utilizing this method, it was found that the fuzzy control method can help minimize field operator errors in water purification operations. One other study by Wu and Lo (2008) combines a neural network with fuzzy logic for coagulant control, where promising results were obtained.

In sum, water purification, particularly the coagulation-flocculation process, is a complex time-varying system that raises concerns about inconsistency, urgency, and safety. To overcome this problem, both direct dosing control (uses physical sensors) and indirect dosing control (uses software sensors) in an appropriate control system have presented their ability to enhance the stability of the coagulation process. Regarding this study, the selection of methods will be conducted based on literature and, more importantly, the availability of technologies.

4. CONCLUSION

The coagulation process in a water treatment plant is undoubtedly one of the determining factors in producing good quality drinking water. When employed in a water treatment process, it is essential to study the operating principle to optimize the coagulation process. The literature discussed in this paper has proposed or reviewed different chemical and physical changes that enhance process efficiency. These changes should be assessed based on the existing factors that affect the coagulation process in the water treatment plant. One of the crucial factors that will affect the coagulation process is the control of coagulant dosage. Uncontrolled dosage will corrupt the coagulation process's outcome and may also

influence the subsequent treatment process. Direct and indirect dosing control, which uses physical equipment and software sensors, respectively, are viable options to control the dosage in real-time according to the raw water condition. Depending on the available resources and issues, the water treatment plant can implement the optimization method to improve the coagulation process, resulting in better overall treatment efficiency.

5. AUTHORS' NOTE

The authors declare that there is no conflict of interest regarding the publication of this article. Authors confirmed that the paper was free of plagiarism.

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