


# Adsorption and coagulation in wastewater treatment – Review

HADID SUKMANA<sup>1\*</sup> , NAOUFAL BELLAHSEN<sup>1</sup>,  
FERNANDA PANTOJA<sup>1</sup> and CECILIA HODUR<sup>2</sup>

<sup>1</sup> Doctoral School of Environmental Sciences, University of Szeged, H-6720 Szeged, Hungary

<sup>2</sup> Department of Biosystems Engineering, University of Szeged, H-6725 Szeged, Hungary

## REVIEW PAPER

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## ABSTRACT

Wastewater issues became a complex challenge in the world. There are several methods in wastewater treatment, such as chemical, physical, biological, and the combination of each method. However, each process has advantages and disadvantages. The physicochemical methods are common methods used in wastewater treatment, such as adsorption and coagulation. Adsorption and coagulation are excellent methods to remove pollutants. The adsorption process is greatly influenced by pH, adsorbent dose, temperature, and contact time. Coagulant dose, settling time, and pH are the main factors in the coagulation process. Chemical material as an adsorbent and coagulant has been studied in previous research, but recently, to substitution chemical materials is a challenging subject. Natural substances are potential new materials in wastewater treatment and became popular due to their efficiency and environment friendly characteristics. This review investigated the role of adsorption and coagulation in wastewater treatment and the utilization of natural materials as adsorbents and coagulants.

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## KEYWORDS

adsorption, coagulation, natural adsorbents, natural coagulants, wastewater treatment

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\* Corresponding author. E-mail: sukmanahadid@gmail.com

## INTRODUCTION

Water is an essential factor for all life and human survival and has an important role in the drinking water supply and economic sectors (Tiri et al., 2018). The need for water in household and industrial activities is still increasing every year. Nowadays, the world faces a water crisis due to industrial globalization, increasing residential and commercial areas, and agricultural lands that lead to enormous wastewater production (Abdelbasir and Shalan, 2019).

According to global trends, in high-income countries, municipal and industrial wastewater treatment is about 70%, the percentages drop to 38 and 28% in upper-middle-income and lower-middle-income countries, respectively. Besides, in low-income countries, only 8% undergoes treatment of any kind, and over 80% of all wastewater is discharged without treatment (WWAP (United Nations World Water Assessment Programme), 2017). Wastewater treatment is an essential factor in industrial processes because wastewater containing hazardous pollutants has negatively affected all environmental elements, such as air, soil, and water (Mohd Udaiyappan et al., 2017).

The presence of non-biodegradable organic compounds in wastewater, even in small amounts, causes severe public health problems due to endocrine-disrupting, toxic to living organisms, and potential carcinogens (Malik et al., 2020). For example, the industrial process, such as palm oil, olive oil, sunflower oil production, etc., generates many oily contents to wastewater, forming pollution. Oily content is a harmful waste that is discharged into the environment. The concentration and physical nature of the oily content and the droplet sizes affect the removal of oily content in wastewater (Coca-Prados and Gutiérrez-Cervelló, 2010).

In general, the wastewater treatment methods are biological, chemical, physical, and the combination of each method (Kontos et al., 2014). The essential process of wastewater treatment are divided into five steps such as 1) pre-treatment using physicochemical and mechanical methods; 2) primary treatment using physicochemical and chemical methods; 3) secondary treatment using physicochemical and biological methods; 4) tertiary treatment or final treatment using physical and chemical methods; and finally, 5) sludge treatment (Crini and Lichtfouse, 2019). For instance, wastewater treatment technologies include biological techniques, photocatalysis, membrane technology, ultrasonication, adsorption, and coagulation/flocculation. The advantages and disadvantages of technologies of wastewater treatment are shown in Table 1.

Efficient wastewater treatment is the basic need of the present society. The application of wastewater treatment could be a solution for protecting the environment. Adsorption and coagulation are wastewater effective treatment methods with a simple design and lower processing cost used by industry (Cai et al., 2019; dos Santos et al., 2018).

Today, research about adsorption and coagulation for wastewater treatment is still under development. In addition to using chemical adsorbents and coagulants, researchers continue to develop alternative raw materials appropriate for use in the future. Hence, one of the most exciting research objectives is to find out new materials for wastewater treatment. The development of the world that leads to environmentally friendly materials has also encouraged increased research of natural adsorbents and coagulants.

Natural materials could be a potential resource for wastewater treatment that is low-cost, efficient, and environmentally friendly. Therefore, natural adsorbents and coagulants would be alternative solutions for the mitigation of pollution. This review presents and discusses recent information concerning the role of adsorption and coagulation methods in wastewater



Table 1. Technologies of wastewater treatment

Type of treatment	Advantages	Disadvantages	References
Biological	potential for removing metals	technology is still under development	(Ahmaruzzaman, 2009)
Photocatalysis	high degradation rate	potential to harmful due to exposure to carcinogenic UV light	(Cheng et al., 2019)
Membrane technology	membrane properties could be adjusted small occupation area high processing efficiency	membrane fouling	(Saleh and Gupta, 2016; Zhang et al., 2016)
Ultrasonication	compact environmentally friendly	require high energy	(Budiman and Wu, 2016; Mahvi, 2009)
Adsorption	simple design cost-effectiveness excellent approach for removing organic pollutants	require regeneration of the adsorbent	(Ariffin et al., 2017; Bazrafshan et al., 2016; Cai et al., 2019; Diaz de Tuesta et al., 2018)
Coagulation/ flocculation	simple process good for reclamation or removed pollutants	require high dosage produce massive sludge and large particles	(Ang and Mohammad, 2020; Ariffin et al., 2017; Chethana et al., 2016)

treatment. This review also compared the removal effectiveness of different natural adsorbents and coagulants as potential materials in the future.

## ADSORPTION

Adsorption is a change in concentration of a given substance at the interface compared with the neighboring phases (Dąbrowski, 2001). Adsorption can occur in the following systems: solid-liquid, solid-gas, liquid-gas, and liquid-liquid.

Nowadays, adsorption is one of the most common methods used in industrial wastewater treatment. For instance, adsorption could be used to remove and recover heavy metals in wastewater, even at a low concentration. Therefore, adsorption is a practical and simple process to apply in wastewater treatment compared with other methods. Adsorption methods have several parameters to determine an effective technique, such as pH, adsorbent dose, temperature, and contact time.

The solution pH plays an essential role in the adsorption process; determining the initial pH condition is vital for increasing pollutants' removal effectivity. pH condition in the solution affects the ability between hydrogen ions in the functional group such as hydroxyl (OH), carboxyl (COOH), amine (NH), and metal ions on the adsorbent surface (Huang and Liu, 2013).

The adsorbent dose is one of the critical parameters in the adsorption method due to the adsorbent capacity's effect on the adsorbate initial concentration and plays an important role in determining the optimum process (Afroze et al., 2016). The increasing adsorbent dose could



increase removal capacity due to greater surface area and more active adsorption sites (Kumar et al., 2016).

The temperature is one of the parameters that impact the adsorption process due to affecting physicochemical reactions. In endothermic reactions, the increasing temperature would increase the reaction rate, whereas, in exothermic reactions, the increasing temperature would decrease the reaction rate (Xu et al., 2018).

Adsorption capacity could be described by contact time. Therefore, the adsorption process requires a specific time to reach equilibrium, which is the time when the adsorption is completed (Ariffin et al., 2017). In the application of contact time, there is time variation to achieve maximum process, depending on adsorbent material and adsorbate.

### Mechanisms of adsorption

The adsorption method is widely used in wastewater treatment due to effective and efficient factors. During adsorption, several steps occur until the process finishes. The adsorption mechanism is shown in Fig. 1.

The adsorption mechanism follows four steps, i.e. (Sotelo et al., 2013):

1. The solution transfers the contaminant molecules/solute to the boundary layer of the adsorbent.
2. Diffusion occurs from the boundary layer to external surface of the adsorbent.
3. Transport from the external surface to active sites of pores.
4. The adsorption of the sorbate to the solid phase.

Adsorption processes are divided into two types, shows in Fig. 2. Physical adsorption (physisorption) and chemical adsorption (chemisorption) have different processes. The physisorption is responsible for attractive forces in molecules and generally occurs with a relatively low degree of specificity, whereas chemisorption is responsible for the structure of chemical compounds due to the involved electrons exchanges (Rouquerol et al., 2013; Tan and Hameed, 2017).

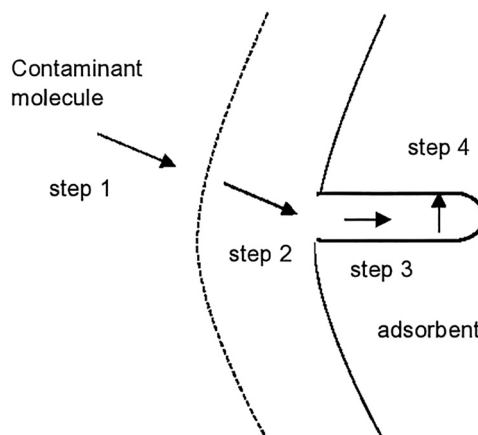


Fig. 1. Adsorption mechanism (Bellahsen et al., 2018)



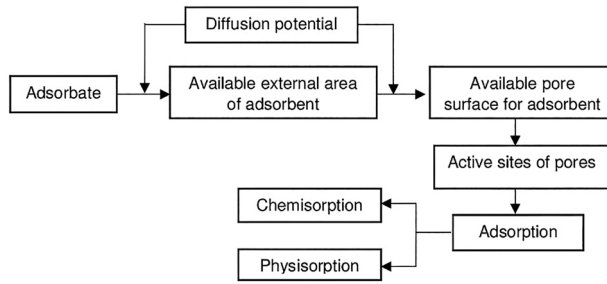


Fig. 2. Pathways of adsorption process (Singh et al., 2018)

## Isotherm and kinetics

The common method for evaluating the adsorption mechanism is adsorption isotherm and kinetics. The adsorption isotherm is described as equilibrium concentration between the solute concentration in the liquid and that on the surface of the adsorbent (Foo and Hameed, 2010). In contrast, adsorption kinetics is described as diffusion behavior and the adsorption rate of solute (Das, 2020). The linear forms of adsorption isotherm and kinetic models are shown in Table 2.

According to Table 2, there are several models in isotherm adsorption. Langmuir isotherm model has two key points in the adsorption process. First, homogeneous adsorption occurs in the adsorbent. Second, adsorbed molecules form a saturated layer on the adsorbent surface so that maximum and monolayer adsorption occurs. The Freundlich isotherm model assumes adsorption capacity has relation to the ion concentration at equilibrium, and adsorption occurs in the heterogeneous surface, also not suitable for low sorbate concentration (Alves et al., 2017; Sadeek et al., 2015).

Dubinin-Redushkevich isotherm model describes the adsorption with a Gaussian energy distribution on a heterogeneous surface. This model could be used to determine the difference in the physical or chemical adsorption process (Üner et al., 2016). Furthermore, the standard model in the kinetic models is pseudo-first-order and pseudo-second-order.

The kinetic model could explain the adsorption process and possible rate-controlling steps, such as chemical reaction processes or mass transport (Anastopoulos et al., 2017). Generally, when adsorption occurs through diffusion through the interface, the kinetic model follows pseudo-first-order. However, pseudo-second-order has a benefit where equilibrium adsorption capacity could be calculated from the model and there is no need to calculate adsorption equilibrium capacity from the experiment (Sarma et al., 2019).

## Adsorbent

The adsorbent is an essential factor in the adsorption process. Adsorbents can capture pollutant substances onto itself, has porosity and is also insoluble in water (Abdi et al., 2017). The utilization of adsorbent usually considers several aspects, such as cost and adsorbent characterization. However, the cost and characteristics of these compounds can influence maximum removal in wastewater.

Firstly, the adsorbent key factor is adsorption capacity, where the adsorbent could adsorb the adsorbate onto its surface. Secondly, excellent adsorbent criteria are short adsorption periods in



Table 2. The linear forms of adsorption isotherm and kinetic models (Lima et al., 2015; Varala et al., 2019)

Isotherm models	Equation form	Parameters
Langmuir	$\frac{1}{q_e} = \frac{1}{Q_0} + \frac{1}{Q_0 K_L C_e}$	$C_e$ : equilibrium concentration of adsorbate $q_e$ & $Q_e$ : amount of sorbed per gram of adsorbent $Q_0$ : maximum monolayer coverage capacity of sorbent $K_L$ : Langmuir isotherm constant related to the energy of adsorption
Freundlich	$\log Q_e = \log K_f + \frac{1}{n} \log C_e$	$K_f$ : Freundlich isotherm constant, an approximate indicator of adsorption capacity $n$ : adsorption intensity/heterogeneity parameter
Dubinin-Redushkevich (D-R)	$\ln q_e = \ln q_s - (K_{ad} \varepsilon^2)$ where $\varepsilon = RT \ln [1 + \frac{1}{C_e}]$ and $E = \frac{1}{(2K_{ad})^{0.5}}$	$K_{ad}$ : D-R isotherm constant $q_s$ : theoretical isotherm saturation capacity
Sips	$q_e = \frac{Q_0 \cdot K_s \cdot C_e^{1/n_s}}{1 + K_s \cdot C_e^{1/n_s}}$ where $0 < \frac{1}{n_s} \leq 1$	$K_s$ : Sips isotherm constant $n_s$ : Sips exponent
Liu	$q_e = \frac{Q_0 \cdot (K_g \cdot C_e)^{n_L}}{1 + (K_g \cdot C_e)^{n_L}}$	$K_g$ : Liu isotherm constant; $n_L$ : Liu exponent
Redlich-Peterson	$q_e = \frac{K_{RP} \cdot C_e}{1 + a_{RP} \cdot C_e^g}$ where $0 < g \leq 1$	$K_{RP}$ and $a_{RP}$ : Redlich-Peterson isotherm constants $g$ : Redlich-Peterson exponent (dimensionless) whose value should be $\leq 1$
Hill	$q_e = \frac{Q_0 \cdot C_e^{nH}}{KH + C_e^{nH}}$	$KH$ : Hill isotherm constant $nH$ : Hill exponent (dimensionless)
Khan	$q_e = \frac{Q_0 \cdot K_k \cdot C_e}{(1 + K_k \cdot C_e)^{n_k}}$	$K_k$ : Khan isotherm constant $n_k$ : Khan exponent (dimensionless)
Radke-Prausnitz	$q_e = \frac{Q_0 \cdot K_{RAP} \cdot C_e}{(1 + K_{RAP} \cdot C_e)^{1/n_{RP}}}$	$K_{RAP}$ : Radke-Prausnitz isotherm constant $n_{RP}$ : Radke-Prausnitz exponent (dimensionless)
Toth	$q_e = \frac{Q_0 \cdot K_T \cdot C_e}{[1 + (K_T \cdot C_e)^{1/n_T}]^{n_T}}$	$K_T$ : Toth isotherm constant $n_T$ : Toth exponent (dimensionless)
Kinetic models	Equation form	Parameters
Pseudo-first-order	$\log(q_e - q_t) = \log q_{e,m} - \frac{k_1}{2.303} t$	$q_t$ : amount of metal sorbed at time $t$ $k_1$ : the first-order rate constant
Pseudo-second-order	$\frac{t}{q_t} = \frac{1}{k_2 q_{e,m}^2} + \frac{1}{q_{e,m}} t$	$k_2$ : the second-order rate constant
Elovich	$q_t = \frac{1}{\beta} \ln(\alpha \cdot \beta) + \frac{1}{\beta} \ln(t)$	$\alpha$ : initial adsorption $\beta$ : related to the extent of surface

(continued)



Table 2. Continued

Kinetic models	Equation form	Parameters
Avrami	$q_t = q_e \cdot \{1 - \exp[-(k_{AV} \cdot t)]^n\}$	coverage and the activation energy involved in chemisorption to: $1/\alpha\beta$ . $k_{AV}$ : Avrami kinetic constant $n$ : fractional adsorption order

the adsorption process (Katheresan et al., 2018). Thirdly, adsorbent with high porosity has a higher surface area with high adsorption capacity (Ballav et al., 2018).

Adsorbents could be classified into two types that are conventional and non-conventional adsorbents. Conventional adsorbents consist of activated carbons, ion-exchange resins (polymeric organic resins), and inorganic materials such as activated alumina, silica gel, zeolites, and molecular sieves. Non-conventional adsorbents consist of industrial/agriculture by-products such as sawdust, bark, solid waste, red mud, etc., and biological by-product such as biomasses, peat, chitosan, other polysaccharides (Crini et al., 2019).

### Conventional adsorbent/inorganic adsorbent

Conventional adsorbents are widely used as commercial adsorbent in the industrial sector or laboratory practice such as activated carbons, zeolites, commercial activated alumina, silica gels, ion-exchange resins. Activated carbons are excellent adsorbents to remove organic pollutants in secondary or tertiary treatment. These materials' benefits are that they do not generate by-products and could be combined with other techniques such as an ultrafiltration membrane. However, activated carbons are expensive and nonselective. On the other hand, ion-exchange resins could reduce water pollution in the environment. The benefits of this material are no loss of adsorbent on regeneration and reclamation of solvent. Like activated carbons, this material is expensive (Crini et al., 2019).

Several adsorbents from inorganic materials have been studied for removed pollutants in wastewater. The research conducted by Feng et al. (2012) used superparamagnetic ascorbic acid-coated  $\text{Fe}_3\text{O}_4$  nanoparticles to remove arsenic in wastewater. The result shows maximum adsorption capacity for arsenic (V) and arsenic (III) are  $16.56 \text{ mg g}^{-1}$  and  $46.09 \text{ mg g}^{-1}$ , respectively. The Langmuir model was the best model for this study.

The study was carried out by Mahmoud et al. (2016) and used three MgO nanomaterials ( $\text{MgO}_S$ ,  $\text{MgO}_N$ , and  $\text{MgO}_U$ ) to remove Remazol Red RB-133 in aqueous solution. The result shows the removal efficiency of Remazol Red RB-133 on  $\text{MgO}_S$ ,  $\text{MgO}_N$ , and  $\text{MgO}_U$  was achieved 100, 92, and 76.5% at 11 min, respectively. Simultaneously, the removal was completed on  $\text{MgO}_N$  at 40 min and  $\text{MgO}_U$  at 60 min. The Langmuir and pseudo-first-order models were the best models for this study.

Wang et al. (2019) observed the use of Polyaniline/ $\text{TiO}_2$  ( $\text{PANI}/\text{TiO}_2$ ) as an adsorbent to remove methylene blue (MB). The result shows that the maximum adsorption capacity was  $458.10 \text{ mg g}^{-1}$  with pH application (pH 3–11). The regeneration of adsorbent was studied with used HCl solution ( $0.1 \text{ mol L}^{-1}$ ) as a stripping agent and, after ten cycles, adsorption efficiency could be maintained around 99%.



## Non-conventional/natural adsorbent

Natural adsorbent origin is from natural materials or industrial waste/by-products such as agriculture, food, etc. It has a low cost and could be directly used or after minor treatment (Gupta and Suhas, 2009). For instance, agriculture waste is one of the foremost abundant renewable resources globally and available in a considerable amount (Adegoke and Bello, 2015), becoming a source of porous materials rich in active functional groups (da Rocha et al., 2020). Therefore, agriculture waste is a potential material that could be utilized in wastewater treatment.

Currently, the research is focused on developing a natural adsorbent as an alternative for substituting a chemical adsorbent (Martini et al., 2020). The main advantages of natural adsorbents are increased efficiency economically due to low cost and a high removal rate for the highly toxic wastewater (Mathurasa and Damrongsiri, 2018; Mo et al., 2018). However, natural adsorbents also have disadvantages such as the adsorption process running slow and pH parameter as the main factor to influence the adsorption effect (Mo et al., 2018).

Several natural adsorbents have been studied for pollutants removal from wastewater. For instance, Bellahsen et al. (2021) applied natural adsorbents from the banana peel, compost, bark, wheat husk, wheat bran, sugar beet pulp, and pomegranate peel for the adsorption of ammonium. The result shows pomegranate peel powder (PPP) could achieve 97% removal, however, other materials showed a negative and low adsorption ability.

Furthermore, Baby et al. (2019) reported the use of agricultural waste palm kernel shell as an adsorbent to remove heavy metals-contaminated water. In this study, heavy metals such as  $\text{Pb}^{2+}$ ,  $\text{Cr}^{6+}$ ,  $\text{Cd}^{2+}$ , and  $\text{Zn}^{2+}$  could be removed effectively from water. The optimum condition of contact time was 60 min for  $\text{Pb}^{2+}$  and  $\text{Cr}^{6+}$  and 90 and 120 min for  $\text{Cd}^{2+}$  and  $\text{Zn}^{2+}$ , respectively. The percentage of removal obtained was 99% for  $\text{Pb}^{2+}$  and  $\text{Cr}^{6+}$ , and  $\text{Zn}^{2+}$  and  $\text{Cd}^{2+}$  was 83%. The adsorption capacity of  $\text{Pb}^{2+}$  and  $\text{Cr}^{6+}$  was achieved  $49.64 \text{ mg g}^{-1}$  and  $49.55 \text{ mg g}^{-1}$ , respectively, whereas  $\text{Cd}^{2+}$  and  $\text{Zn}^{2+}$  were achieved  $43.12 \text{ mg g}^{-1}$  and  $41.72 \text{ mg g}^{-1}$ , respectively.

Other than that, Zulkania et al. (2020) found the application of activated carbon and bio-sorbent produced from palm fibre wastes (PFW) and activated by phosphoric acid. The adsorbent was investigated to remove MB with the effect of adsorbent type and phosphoric acid concentration. The contact time was found to be 90 min. The result shows optimum adsorption was obtained using activated carbon with 10% (v/v) bio-sorbent activating agent concentration and using bio-sorbent with 30% (v/v) activating agent concentration. The adsorption capacity and percentage of removal from activated carbon were  $9.850 \text{ mg g}^{-1}$  and 98.50%, respectively, while bio-sorbent was achieved  $9.984 \text{ mg g}^{-1}$  and 99.84%, respectively.

The modification of rice husk (RH) or rice husk ash (RHA) was investigated by Phan et al. (2019). It used Triamine-activated rice husk ash (TRI-ARHA) as a potential natural adsorbent for nitrate and other anions removal. The TRI-ARHA shows a nitrate adsorption capacity ( $>160 \text{ mgNO}_3^-/\text{g}$ ) compared to the anion exchange resin akulite A420 ( $\sim 80 \text{ mgNO}_3^-/\text{g}$ ), with 10 cycles of adsorption-desorption. Similar results were obtained by Mor et al. (2016) using the activated rice husk ash (ARHA) to remove phosphate in wastewater and water. The ARHA would enhance the adsorption capacity for phosphate with maximum removal of 89% with pH 7,  $2 \text{ g L}^{-1}$  doses, and a time of 120 min.





Furthermore, the study by [Thuy et al. \(2020\)](#) applied magnesium chloride modified carbonized rice hull (MCRH) to remove ammonium from synthetic and domestic wastewaters. During the 27 h long treatment, MCRH with  $1.8 \text{ g L}^{-1}$  concentration could remove 90.7% ammonium (capacity of  $41.0 \text{ mg g}^{-1}$ ) for  $81.3 \text{ mg L}^{-1}$  synthetic wastewater and 86.8% for real domestic wastewater. The Langmuir model was the best model for this study, and based on the Dubinin-Radushkevich model, ammonium was physically adsorbed on MCRH.

[Quansah et al. \(2020\)](#) used rice husk utilization for removing methylene blue (MB) and crystal violet (CV). The rice husk used varying dosage in the range of 0.05–1.0 g with the initial concentration of  $300 \text{ mg L}^{-1}$  (MB) and  $400 \text{ mg L}^{-1}$  (CV). The result shows the optimum temperature at  $75^\circ\text{C}$  with removal percentage of MB and CV from 53.74–97.74% and 57.40–98.19%, respectively. [Mitra et al. \(2019\)](#) showed that the addition of rice husk as an adsorbent in continuous column mode for the removal of Pb (II) and Cr (VI) ions with pH 2–6. The result shows increasing influent concentration ( $10\text{--}30 \text{ mg L}^{-1}$ ) and indicates the increased capacity of adsorbent for Pb (II) and Cr (VI) from 5.72 to  $22.99 \text{ mg L}^{-1}$  and  $2.798\text{--}10.18 \text{ mg L}^{-1}$ , respectively. According to the result, statistical analysis showed the Thomas model and the Yoon–Nelson model.

## COAGULATION

The coagulation process has an essential function in wastewater treatment for reclamation or removal of pollutants ([Ang and Mohammad, 2020](#)). The coagulation-flocculation process is the major physicochemical treatment method used in industrial wastewater treatment to reduce colloidal turbidity and suspended solids ([Gautam and Saini, 2020](#)). According to previous studies, 40% of organic materials and nitrogen could be removed from wastewater using the coagulation method ([Loloei et al., 2014](#)).

Coagulation is a process where the pollutants, suspended particles lead to the sediment through collision with opposite particles and obtain agglomerate to form an insoluble agglomerate complex ([Syam Babu et al., 2020](#)). In several types of research, a coagulant dosage, settling time, and pH are the main factors determining the removal of the contaminant in wastewater. These factors will lead to the optimum conditions in the JAR test. The factor affecting coagulation shows in [Table 3](#).

### Mechanisms of coagulation

In general, there are four steps in the coagulation/flocculation mechanism, including charge neutralization, sweep coagulation, bridging, and patch flocculation ([Amran et al., 2018](#)). The coagulation/flocculation mechanism is shown in [Fig. 3](#).

There are colloidal particles in the coagulation process that are negatively charged, which causes repulsion to occur. The coagulant is added to stabilize the colloid particles so that there is no repulsive force. Usually, the added coagulant is a poly-electrolyte that causes the zeta potential of the colloid close to zero points. Therefore, this process is called the charge neutralization mechanism ([Amran et al., 2018](#)). Furthermore, a high concentration of metal salts is added to the water causing the precipitation of amorphous metal hydroxides, and gradually large lumps are formed; this process is called sweep coagulation ([Li et al., 2006](#); [Zhao et al., 2011](#)).



Table 3. Factors affecting coagulation (Kumar et al., 2017)

Coagulation application	Effective characteristics	Properties of natural water
a) Extraction of coagulants	a) Settling time	a) pH
b) Solubility of coagulants	b) Turbulence	b) Alkalinity
c) Dosage of coagulants	1. Rapid Mixing	c) Availability of bacteria
d) Charge of particles	2. Slow Mixing	d) Dissolved Oxygen
e) Basicity of a coagulant	c) Coagulant adds quantity	e) Temperature
	d) Particle's type	f) The element contains (Cl, Na, Mn, Si, Fl, NH <sub>3</sub> , Fe)
		g) Suspended solids
		h) Turbidity
		i) Total dissolved solids

The bridging flocculation process could produce a very large floc due to the adsorption of high molecular weight linear-chain compounds commonly based on polyacrylamide. This process occurs at the same time in several particles (Hogg, 2013). Meanwhile, the patch flocculation process occurs because of the adsorption of the polymer onto the particles, thereby a local charge reversal is effected. The result of this process is an attraction force between each particle (Hjorth and Jørgensen, 2012).

## Coagulant

Coagulants could be classified into two types, i.e. chemical and natural coagulants. Both coagulants aim to remove pollutants in the chemical (BOD & COD) or physical (suspended solids & turbidity) forms (Kumar et al., 2017). Chemical coagulant includes hydrolyzing metallic salts such as ferric chloride, ferric sulphate, magnesium chloride, and alum; pre-hydrolyzing metallic salts such as poly aluminum chloride (PAC), poly ferric chloride (PFC), poly ferrous sulphate (PFS), poly aluminum ferric chloride, and synthetic cationic polymers such as aminomethyl polyacrylamide, polyalkylene, polyethylenimine, polyamine. Natural coagulant contains microorganism substance such as bacterial, microalgae, fungal; animal-based such as chitosan and isinglass and plant-based such as seed and plant extracts, starch and fruit waste (Al-Sahari et al., 2020; Pandey et al., 2020).

Generally, coagulants are polymers or polyelectrolytes that are synthetic organic compounds with a high molecular weight that make flocs stronger, bigger, and easily settling. Good coagulants could form multi-charged polynuclear complexes in solution with enhanced adsorption characteristics (Iwuozor, 2019).

Several coagulants from chemical materials have been studied for removing pollutants in wastewater. Dhanjal et al. (2018) analyzed the use of alum, potash alum, and poly aluminum chloride for pre-treatment of municipal wastewater. The result shows the most effective coagulant is poly aluminum chloride. The optimum dosage of 500 mg L<sup>-1</sup> to remove pH, EC, TDS, TSS, and turbidity were 7.16, 700.3 μS cm<sup>-1</sup>, 973 mg L<sup>-1</sup>, 715 mg L<sup>-1</sup>, and 4.5 NTU, respectively.



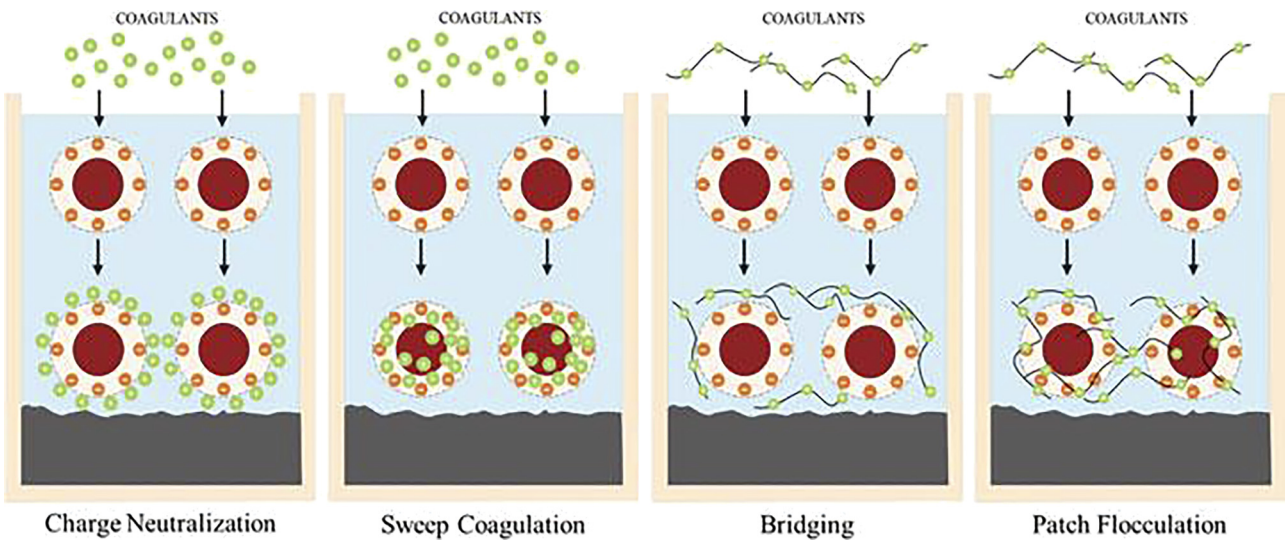


Fig. 3. Coagulation/flocculation mechanisms at the treatment process (Al-Sahari et al., 2020)



The study was investigated by [Zahrim et al. \(2017\)](#), which used several chemical coagulants such as calcium lactate, ferric chloride, magnesium hydroxide, poly diallyl dimethyl ammonium chloride (polyDADMAC), and aluminum chlorohydrate to remove color in palm oil mill biogas plant wastewater (POMBPW). The result shows ferric chloride is the best coagulant to remove color. Ferric chloride could remove color up to 80% with an optimum dosage of  $8,000 \text{ mg L}^{-1}$  and pH 10.

Furthermore, the research was carried out by [Li et al. \(2016\)](#) used magnesium hydroxide as a chemical coagulant for dyes wastewater treatment. The different dosages and initial turbidity used to determine the optimum condition in coagulation. The result shows optimum dosage, initial turbidity and pH was achieved  $144 \text{ mg L}^{-1}$ , 45 NTU, and 12, respectively. Besides, coagulant dosage increases have an effect of decreasing final turbidity.

### Natural coagulant

Natural coagulant is produced from other natural sources, which have a functional group. For instance, agriculture waste has properties that provide excellent coagulation ability due to the presence of carboxyl and silanol groups ([Ahmaruzzaman and Gupta, 2011](#)). Natural coagulant is an environmentally friendly green product, unlike chemical coagulants that could affect human health and the environment ([Othmani et al., 2020](#)).

The utilization of natural coagulants in the researches shows an essential improvement to nature and the ecosystem due to biodegradable components. Besides, the utilization of natural coagulants is growing rapidly over the years due to the low impact on pH and fast reaction. In contrast, a disadvantage of natural coagulants from plant-based is the price so that their use on an industrial scale compared to synthetic coagulants is ineffective. The utilization of these coagulants more effectively removes low-medium turbidity in wastewater treatment ([Litu et al., 2019](#); [Nath et al., 2020](#)).

Besides, natural coagulants have been studied in some previous research, such as rice husk ash, *Moringa oleifera*, *Trigonella foenum-graecum*, and *Hibiscus esculentus*, *Cassia obtusifolia*, and papaya seed powder.

### Rice husk ash

The previous research on RH and RHA for natural coagulants is still limited. For instance, [Huzir et al. \(2019\)](#) investigated the application of RHA as a potential natural coagulant for palm oil mill effluent (POME) treatment with optimum conditions was pH 3.6, 6.0 g with a time of 57 min. The results show RHA could remove the total suspended solids (TSS) and chemical oxygen demand (COD) up to 83.88 and 52.38%, respectively.

### Moringa oleifera

[Jagaba et al. \(2020\)](#) observed that the use of *M. oleifera* as a natural coagulant to compare with a chemical coagulant. The result shows the optimum dosage of *M. oleifera* obtained  $2,000 \text{ mg L}^{-1}$  for removing turbidity, oil & grease, TSS, COD, Color, and  $\text{NH}_3\text{-N}$ . [Ahmad et al. \(2015\)](#) reported the pre-treatment for POME using *M. oleifera* and chitosan as a natural coagulant combined with the integrated membrane. The result shows the percentage of recovery for turbidity value 20 NTU at 78%. Furthermore, a similar result was obtained by [Nikam et al. \(2012\)](#) shows the effectivity of *M. oleifera* as a natural coagulant for reducing turbidity in



wastewater. The removal percentage of turbidity obtained at 32.52% with an optimum dosage of  $10 \text{ mg L}^{-1}$ .

Adelodun et al. (2020) used *M. oleifera* to remove turbidity, COD, and biochemical oxygen demand (BOD) in municipal wastewater. The result shows the percentage of removal turbidity, COD and BOD were 94.44, 57.61, and 68.72%, respectively, with the optimum dosage of  $150 \text{ mg L}^{-1}$  and settling period of 250 min. Also, Muruganandam et al. (2017) reported the pre-treatment for tannery effluent using *M. oleifera*, Cactus, and Aloe vera as a natural coagulant. The result shows the percentage of recovery for turbidity with *M. oleifera*, Cactus, and Aloe vera achieved 59.43, 51.50, 46.76%, respectively then for COD achieved 37.82, 59.35, 52.60%, respectively. The optimum dosage and pH condition for *M. oleifera*, are  $15 \text{ g m}^{-1}$  with pH 6, for Cactus  $40 \text{ g mL}^{-1}$  with pH 7 and for Aloe vera 5% v/v with pH 5.

### ***Trigonella foenum-graecum* and Okra**

Sui Kim et al. (2020) applied *T. foenum-graecum* (fenugreek) as a natural coagulant and *Hibiscus esculentus* (okra) as a natural flocculant for the treatment of POME. The optimum conditions of removal percentages for TSS, COD, and turbidity were found in a dosage of fenugreek is  $4.6 \text{ g L}^{-1}$ , a dosage of okra is  $40 \text{ mL}/500 \text{ mL}$  POME, pH 4, and speed of mixing is 155 rpm. The results from fourier transform infrared (FTIR) spectroscopy show fenugreek and okra have properties that provide excellent coagulation-flocculation ability due to the presence of polysaccharides groups.

In addition, Freitas et al. (2015) studied the combination and inorganic coagulant ( $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ ) and Okra mucilage as natural coagulant removal of turbidity, COD, and color. The result shows it used an inorganic dosage of  $88.0 \text{ mg L}^{-1} \text{ Fe}^{3+}$  and okra mucilage dosage  $3.20 \text{ mg L}^{-1}$  with pH 6 could remove turbidity, COD, and the color were 97.24, 85.69, and 93.57%, respectively.

### ***Cassia obtusifolia* and papaya seed powder**

Shak and Wu (2015) analyzed the addition of *C. obtusifolia* seed gum as a natural coagulant with alum application for treatment of POME. The optimum conditions using response surface methodology (RSM) were found in a dosage of alum of  $1.15 \text{ g L}^{-1}$ , a dosage of *C. obtusifolia* seed gum is  $2.47 \text{ g L}^{-1}$ , and a settling time of 35.16 min. The removal percentages for COD and TSS were up to 48.22 and 81.58%, respectively. Kristianto et al. (2018) studied the use of papaya seed powder to remove color in synthetic textile wastewater. The result shows the percentage of removal of 84.77% with optimum conditions were found in coagulant dosage is  $0.57 \text{ g L}^{-1}$ , and pH 1.97.

## **CONCLUSIONS**

There are several methods to remove pollutants in wastewater treatment. Adsorption and coagulation are the most common methods used; they are simple to use and have more advantages, such as low cost, practice, and efficiency. Therefore, these methods are effective in removing pollutants and obtaining the quality of wastewater according to standards. On the other side, the development of these methods also was followed by new researches about



materials in adsorption and coagulation from natural resources. One of the sources which was used as a material for adsorbent and coagulant is agriculture waste. Natural materials became a potential opportunity due to their availability in a considerable amount. The utilization of natural adsorbents and coagulants, which are environmentally friendly, could reduce the negative effect on the environment and human health. Previous studies showed that natural adsorbents and coagulants are useful for removing pollutants in wastewater. Therefore, modification of natural adsorbents and coagulants can further be explored for enhanced stability and effectiveness in all wastewater types. Besides, recovery and reuse of natural adsorbents and coagulants have to be continuously developed to ensure environmental sustainability. Finally, applying natural adsorbents and coagulants on the industrial scale requires further investigation as a strategy that will fulfill the goal of the circular economy.

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