International Journal of Engineering & Technology, 7 (3.23) (2018) 56-60

International Journal of Engineering & Technology

Website: www.sciencepubco.com/index.php/IJET

Research paper

A Review on Performance of Chemical, Natural and Composite Coagulant

Mohamad Azizan Bin Omar[1*], Nur Shaylinda Binti Mohd Zin[1], Siti Nor Aishah binti Mohd Salleh[1]

[1]*Department of Water and Environmental, University Tun Hussein Onn Malaysia, 86000 Johor, Malaysia.*
Corresponding author E-mail: zizangojan92@gmail.com

Abstract

Coagulation and flocculation process are commonly used in the treatment of water and wastewater. Normally the chemical coagulant (inorganics coagulants) is widely used as primary coagulant due to its availability, show good efficiency, cheaper and ease to handle. However, from the previous study, the application of conventional coagulant causes environmental effect and consumed large dosage. Many studies were carried out to overcome this problem such as the development of new chemical coagulant, utilization of natural coagulant, application of dual coagulant and composite coagulant. Development of new chemical coagulant such as pre-hydrolysing coagulant (combination of two or more chemical coagulant) and synthetic cationic polymers that richer with positive charge ion are seen able to enhance the removal efficiency at a lower dosage. However, this coagulant still is made from chemical substances that probably contribute to the toxicological issues. To date, natural coagulant is widely explored due to its capability, besides free from physical, and chemical changes in the treated water. However, most of the natural coagulant as single coagulant is not effective in removal compared to the chemical coagulant. Thus, to improve the efficiency while combining the best properties of both coagulants, the development of composite coagulant made from natural and chemical coagulant is necessary. The objective of this paper is to gives an overview of the performance of single, dual, and composite coagulants in order to develop the novel composite coagulant.

Keywords: *Natural coagulant, composite coagulant*

1. Introduction

Nowadays, there are many industrial activities are carried out especially in the developing countries. This indirectly contribute to the production of wastewater and leachate liquid that contains small suspended solids, inorganic and organic substances, heavy metals, toxicity substances and other impurities [1, 2]. Due to the stable surface charge from small particles that keep away from each other, the mission is to neutralize the charge and make the particles added in size since it is crucial for settling process [3, 4]. There are many technologies have been explored to treat water, such as membrane filtration, ion exchange, precipitation, solvent extraction, adsorption, flotation, coagulation and flocculation, biological and reverses osmosis methods [5].

Coagulation and flocculation is one of simple technique used for the removal of suspended solid and dangerous substances present in wastewater and leachate liquid. In this method, after the addition of coagulants, small particles are stickled together to form macro flocs that easily settle, which directly purify the water. Alum and ferric chloride are the most common chemical coagulant used as a single or primary coagulant due to its effectiveness in removing pollutant substances, besides cheaper and easily available. However, many researchers found the chemical coagulant promote secondary pollution, unfriendly to the environment, non-biodegradability, produced toxic sludge which directly lead to its complex and costly treatment [6, 7]. Nowadays, due to the negative drawbacks from chemical coagulant, the natural coagulants are widely explored and used as coagulant aids.

Natural coagulants are normally originated from the animal base, plant base, microorganisms, and bacteria. It is widely used to enhance the formation of micro flocs to macro flocs [8]. However, natural coagulants as single coagulant is not effective in removing pollutants compared with single chemical coagulant. In order to combined the good characteristic of chemical and natural coagulant, the dual coagulant or composite coagulant is suggested. The use of natural coagulants as coagulant aids in dual coagulant method had improved the efficiency of coagulation process, reduced the dosage of chemical coagulants, indirectly reduced the pollutions and cost to treat the sludge [9, 10]. However, the dosing procedure of dual coagulant method are complex which are done separately at rapid and slow mixing stages. Hence, to simplify the procedure, the composite coagulant method is suggested.

Composite coagulant can be described as the combination of two or more types of coagulants either organic or inorganic as one coagulant. The preparation of composite coagulant is by mixturing the coagulant in specific condition. Moreover, by using the composite coagulant method the dosing procedure will be more convenient as it simplify the processes and reduce the cost [11]. Currently, little attention has been made in combining natural coagulant with chemical coagulant as a composite coagulant. Most study on composite coagulant combines two chemical coagulants or chemical coagulant and synthetic polymer. This paper elaborated the types of coagulants, performance of the chemical, natural and composite coagulants.

2. Coagulation and Flacculation

Coagulation is a process to gather the suspended solids in water by adding chemical or natural coagulants [12]. The suspended solids or colloid particles have negative charge (stable particles) which cause them to keep away from one another. By charge neutralization the positive charged ions will neutralize the negative charges particles. When the charge is neutralized, the colloid particles are closed and stick together to form micro flocs, which are not noticeable by naked eyes. While in flocculation process, the gentle mixing is required to allow the high and effective collision between micro flocs to form macro flocs (aggregates) that are heavier and easily removed by settlement. [13].

Usually, the flocs formed in coagulation process are dense and weakly bound, while flocs formed in flocculation process are denser, bigger in size and tightly bound [14, 15, 16]. There are several magnetism and repulsion forces that influence the stability and instability of colloid particles such as Van der Waals forces, Brownian notion, Electrostatic forces and Universal forces [17]. The effectiveness of coagulation and flocculation process depends on few main factors which is type of coagulant, dose of coagulant, pH of water settling time, rapid and slow mixing energy.

2.1. Coagulant

The common coagulants used in water treatment can be classified into inorganic coagulants, synthetic organic polymers or natural coagulants [18]. Chemical or inorganic coagulants can be characterized into three parts as shown in Table 1. Inorganic coagulants which are hydrolyzing metallic salts (aluminum sulphate, ferric chloride, and ferric sulphate) are most commonly used in coagulation and flocculation process due to the lowest price, effective and able to entrap bacteria as it settles [19, 10]. Pre-hydrolyzing metallic salts (combination of chemical with chemical coagulant) found to be more effective than hydrolyzing metallic salts due to better removal at low dosage, produce less volume and strength sludge [20].

In the recent years, polyaluminum chloride (PAC) is widely applied as coagulants for wastewater treatment [21, 22]. Although the cost of PAC is expensive than alum salts and iron salts, but PAC require lower dosage, has high efficiency, do not have requirement for any neutralizing agent, shorter flocculation time, smaller amount of sludge, reduced number of backwashing steps and is effective in the adsorption of micropollutants, toxic organic substances and heavy metals [23, 24]. PAC has the ability to form a large and strong floc with shorter flocculation time compared with alum [25]. Apart from that, PAC has higher ability to neutralize the negative charge of colloidal particles and form a chain of the colloidal particles [26].

Organic or natural coagulant are getting widely explored and used as coagulant aids due to its best properties such as high molecular weight and long chained structures [28]. Natural coagulants have the significant advantages over the commercial ones as they are available in abundance, cheaper and environmental friendly [29]. Moreover, in term of floc characteristics, natural coagulants show better result in settling time, size and strength of flocs, and produced small volume of sludge. The natural coagulants show that unaffected to pH change when compared with chemical coagulant [30]. Table 2 shown the categories of natural coagulant comes from plant, animal and micro-organism based.

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Table 1: Categorization of chemical coagulant [27]

Categories of chemical coagulant	Coagulant
Hydrolyzing metallic salts	Aluminium sulphate, Magnesium chloride, Ferric chloride and Ferric sulphate.
Pre-hydrolizing metallic salts	Polyaluminum chloride, Polyferric chloride, Polyferrous sulphate, Polyaluminum ferric chloride and Polyaluminum sulphate.
Synthetic polymers	Aminomethyl polyacrylamide, Polydiallylmethyl ammonium chloride (poly- DADMAC), Polyethylenimine, Polyalkylene, Polyamine.

Table 2: Categorization of natural coagulants [31]

Categories of natural coagulant	Coagulant
Plant base	Moringa, cactus, tannin, tamarind, nirmali, okra, sago, green pea, mung bean, nuts, tapioca, potato, rice tubers, oat, wheat and corn.
Animal base	Chitosan
Micro-organism based	Xanthan gum

2.2 Composite Coagulant

In the recent year, the high demand for effective coagulants in wastewater and leachate treatment has brought the development of composite coagulant. This new material poses great potential in treating polluted water due to its unique characteristic with high performance compared to the conventional chemical coagulant [32]. There are many composite coagulants such as polymethylmethacrylate psyllium (Psy- PMMA), polyacrylamide starch (St-PAM), polyacrylamide carboxyethyl guar gum (CMG-PAM) and hydroxypropyl methyl cellulose polyacrylamide (HPMC-PAM) have been synthesized and investigated, while its properties were tested on synthetic wastewater. There are several composite coagulants that have been developed to treat various wastewaters as shown in Table 3.

Table 3: Composite coagulants

Composite coagulant	Type of wastewater	Reference
- Poly(2-methacryloyloxethyl) trimethyl ammonium chloride + Chitosan	Paper recycling wastewater	[33]
-(2-methacryloyloxyethyl) trimethyl ammonium chloride + Chitosan.	Pulp mill wastewater	[34]
-Polyacrylamide + Carboxymethyl guar gum	Municipal sewage wastewater	[35]
-Polyacrylamide + Oatmeal	Municipal sewage wastewater	[36]
-Polyacrylamide + Carboxymethyl chitosan	Dye solution	[37]
-Polyaluminum chloride (PAC) + rhizobiumradiobacter F2 + bacillus sphaericus F6, microorganism in soil	Kaolin solution	[38]
-Polyaluminum chloride (PAC) + Chitosan	Synthetic water and reservoir water	[39]
-Polyaluminum chloride + cationic polymer + anionic polymer	Leachate	[21]
-Polyaluminum chloride + Polyacrylamide	Leachate	[40]
-Prehydrolyzed iron + Tapioca starch	Leachate	[11]

3. Performance of Single, Dual and Composite Coagulants on Various Wastewater

Table 4 shown the result of percentage removals by coagulation and flocculation processes for the treatment of various wastewater by single, dual and composite coagulants. The natural coagulants from plant based are widely used in wastewater treatment because of the ability to treat water and environmental-friendly. Studied by Teng [41], the natural coagulant which is corn starch, sago and rice flour were able to remove COD and turbidity from synthetic wastewater with the optimum dose of all coagulant at 20 mg/L, by removing 58% and 78% for corn, 84% and 79% for sago respectively, while 73% and 83% for rice flour respectively. According to Garcia and Ali [42, 43], the used of moringa at dose 300 mg/L was able to remove turbidity over 99%. While, Omar [41], found that tapioca starch as natural coagulant from plant based with 100 mg/L dose, was able to treat semiconductor wastewater with 87% and 99% percentage removals of COD and turbidity respectively. These showed the natural coagulants are able to treat wastewater but do not effective compared to chemical coagulants.

Table 4: Percentage removal for various wastewater treatments

Sample	Coagulants (mg/L)	Percentage removal	Author
Synthetic wastewater (pH 6)	PAC (4.32)	80 (dissolved organic carbon, DOC)	[38]
	composite - PAC (0.86) + chitosan (3.46)	87 (DOC)	
Reservoir water (pH 6)	PAC (4.32)	35 (DOC)	
	composite - PAC (0.86) + chitosan (3.46)	40 (DOC)	
Kaolin (pH 6)	PAC (4.6)	96 (flocculation rate)	[39]
	dual – PAC (1.15) + (CBF) (3.45)	97 (fr)	
	composite - PAC (1.15) + (CBF) (3.45)	99 (fr)	
	composite - aluminum chloride (1.15) + (CBF) (3.45)	99 (fr)	
Kaolin (pH 7)	dual - ferric chloride (10) + chitosan (0.1)	92 (turbidity)	[44]
wastewater (semiconductor) (pH 12)	tapioca starch (100)	87 (COD), 99 (turbidity)	[41]
wastewater (semiconductor) (pH 12)	corn starch (20) sago (20) rice flour (20)	78 (turbidity), 58 (COD) 84 (turbidity), 79 (COD) 83 (turbidity), 73 (COD)	[41]
Wastewater (synthetic) (pH 7)	moringa (300)	97 (turbidity)	[42]
River water (pH 7)	moringa (300)	99 (turbidity)	[43]

Normally, the dose of natural coagulant as primary coagulant is higher due to its less efficiency compared to chemical coagulants. Proved by Ng [38], showed the percentage removal of dissolved organic carbon (DOC) at optimum dose of single PAC at 4.32 mg/L was 80% for synthetic wastewater and 35% for reservoir water. While, in dual coagulant consist of PAC and chitosan, the removal of DOC for synthetic wastewater and reservoir water was increased to 87% and 40% at optimum dose of PAC 0.86 mg/L and chitosan 3.46 mg/L respectively. These showed the reduction of chemical coagulants dose in dual coagulant method. According to Shahriari [44], the dual coagulant by ferric chloride at dose 10 mg/L and chitosan as coagulant aid at dose 0.1 mg/L showed the good removal of turbidity of kaolin solution where the percentage is 92%. These showed, the application of natural coagulants such as chitosan as coagulant aids can increase the removal and reduced the dosage of chemical coagulants. It seem able to increase the removal and reduced the dosage of both coagulant in application of composite coagulant method.

Ni [39], investigated the efficiency of flocculation rate of kaolin solution by coagulation and flocculation process by using single coagulant, dual coagulant and composite coagulant technique. The dose of PAC as single coagulant at 4.6mg/L shown the percentage of flocculation rate was 96% and for dual coagulant the percentage was 97% at a dose of PAC at 1.15 mg/L and dose of CBF at 3.45 mg/L. While for composite coagulant (made from PAC and bioflocculants (CBF, a mixture of rhizobium radio acter F2 + bacillus sphaericus F6, microorganism in soil) with same dose of dual coagulant, the flocculation rate increases to 99%. These showed the composite coagulant is more efficient than dual coagulant in treated wastewater.

4. Performance of Single, Dual and Composite Coagulants on Leachate

In leachate treatment, the dose of coagulant used is higher than another wastewater treatment. Table 5 shows the result of percentage removals by chemical, natural coagulant and composite coagulants for leachate treatment. In single coagulants, alum and iron sulphate were used by Ilhan [45], at optimum dose for both coagulant at 2000 mg/L with 31% and 22% percentage removals for COD respectively. As investigated by Daud [46], the study showed that the ferric chloride at optimum dose of 950 mg/L was more effective than alum at optimum dose of 3000 mg/L with the percentage removal of 37%, 26%, 84% and 96% for COD, ammonia, colour and suspended solid respectively for ferric chloride and 33%, 14%, 78%, and 68% by using alum.

According to Aziz [47], the best chemical coagulant used in leachate treatment for removal of suspended solid and colour was ferric chloride at the optimum dose of 800 mg/L with percentage removals of 99% and 96%, followed by ferrous sulphate at 1000 mg/L dose with percentage removals of 81% and 63%, while for alum at dose of 2200 mg/L with percentage removals of 74% and 60% respectively. While Ghafari [48], found out the optimum dose of alum at 9400 mg/L was able to remove 85% of COD, 92% of colour, and 95% of suspended solid. While, for PAC at optimum dose of 1900 mg/L the percentage removal was of 57% of COD, 97% of colour, and 99% of suspended solid. These had proved that the application of PAC required low dosage compared to alum and ferric chloride. However, PAC is pre-hydrolyzing metallic salts contains chemical substances that may contributed to the toxic sludge. Thus, to reduce the usage of chemical coagulants, the application of composite coagulant by utilizing natural coagulant is suggested.

The natural coagulant is not yet widely used and investigate in leachate treatment due to low efficiency compared to the chemical coagulant. However, the natural coagulants have several significant advantages over chemical coagulants such as green chemistry and environmental friendly. As investigated by Faiz [49], the durian seed starch at dose of 4000 mg/L as a primary coagulant was able to remove COD and turbidity in leachate with percentage removal of 36% and 34%. Zin [50], recorded the percentage removals of ammonia, colour and suspended solid by tapioca starch (TS) with 13%, 55% and 12% respectively. These show the natural coagulants are able to remove impurities in leachate but still cannot be the best compared with chemical coagulants. Thus, to increase the efficiency removal, the combination of both properties of chemical and natural coagulants, the application of dual coagulant or composite coagulant method is suggested.

Table 5: Percentage removal for leachate treatments

Sample	Coagulants (mg/L)	Percentage removal	Author
Leachate (8.2)	alum (2000) iron sulphate (2000)	31 (COD) 22 (COD)	[45]
Leachate (pH 7)	alum (3000) ferric chloride (950)	33 (COD), 14 (ammonia), 78 (colour), 68 (turbidity) 37 (COD), 26 (ammonia), 84 (colour), 96 (turbidity)	[46]
Leachate (pH 4)	aluminum sulphate (2200)	60 (colour), 74 (ss)	[47]
	ferric chloride (800)	96 (colour), 99 (ss)	
	ferrous sulphate (1000)	63 (colour), 81 (ss)	
Leachate (pH 7)	alum (9400)	85 (COD), 92 (colour), 95 (ss), 95 (turbidity)	[48]
	PAC (1900)	57 (COD), 97 (colour), 99 (ss), 99 (turbidity)	
Leachate (pH 6)	PAC (7200)	55 (COD), 80 (colour), 95 (ss)	[51]
	alum (11000)	58 (COD), 79 (colour), 78 (ss)	
	dual - PAC (7200) + psyllium husk (400)	64 (COD), 90 (colour), 96 (ss)	
Leachate (pH 6)	durian seed starch (4000)	34 (colour), 37 (turbidity)	[49]
Leachate (pH 4)	tapioca starch (2500)	13 (ammonia), 55 (colour), 12 (SS)	[50]
Leachate (pH 5)	composite - prehydrolyzed iron (60) + tapioca starch (140)	60 (COD), 11 (ammonia), 96 (colour), 98 (ss)	[11]
Leachate (pH 7)	composite - PAC (750) + polyacrylamide (15)	28 (COD), 66 (turbidity)	[40]
	PAC (2000)	49 (COD), 29 (ammonia)	[21]
	composite – PAC (2000) + cationic polymer (10)	59 (COD), 49 (ammonia)	
	composite – PAC (2000) + cationic polymer (10)	56 (COD), 46 (ammonia)	

The addition of polymer or natural coagulant in dual coagulant or composite coagulant method with PAC is able to increased coagulant and flocculation efficiency. Regarding to Yusof [51], dual coagulants by PAC at the optimum dose of 750 mg/L and psyllium husk at dose of 15 mg/L were able to remove COD, colour and suspended solid with percentage removals at 64%, 90% and 96% compared to single PAC which were 55%, 80% and 95%. Meanwhile, the experiment run by Zin [11], showed the leachate treatment using composite coagulant that was made from prehydrolyzed iron (PHI) at dose of 60 mg/L and tapioca starch (TS) at dose of 140 mg/L, the percentage removal of COD, ammonia, colour and suspended solid were 60%, 11%, 96% and 98% respectively.

While Rui [21], the optimum dose of PAC to treat leachate was 2000 mg/L where the percentage removal was 49% of COD and 29% of ammonia. The author also found out that the usage of PAC at dose of 2000 mg/L and cationic polymer at dose 10 mg/L as composite coagulant were obtained with the percentage removals 59% COD, and 46% ammonia, while for PAC and ionic polymer with the same dose the percentage removals were 56% and 45% respectively. These showed when composite coagulant is made from PAC and cationic polymer the percentage removal is higher compared with PAC alone and PAC with anionic polymer. Li [40], used the composite coagulant made from PAC and polyacrylamide in treatment leachate. The optimum dose of PAC was 750 mg/L and polyacrylamide was 15mg/L with percentage removals of COD and turbidity at 28% and 66%. These showed the composite coagulant is more efficient compared to single coagulant and dual coagulant by achieving the highest percentage removals. In order to develop and optimize the composite coagulant processes, it is very important to recognize and understand the best combination of chemical and natural coagulant. However, the usage of composite coagulant in leachate treatment with natural and composite coagulants is still lacking and need particular attention.

5. Conclusion

The application of chemical, natural, and composite coagulants in wastewater treatment including leachate has been analyzed. Although all coagulant have showed satisfies results in removing polluted substances, the coagulation performance still need to improve. The ways to improve the coagulation and flocculation efficiency is by monitoring the range of molecular weight and charge density carried out by each coagulant. Different charge density and molecular weight generated different neutralization or bridging system. For further research, the optimization of this factor could improve the treatment efficiency and initiate lowest cost for used chemicals.

The chemical coagulants showed the highest percentage removal compared to the natural coagulants. Since the chemical coagulant contributed to the environmental pollution, the usage of natural coagulant is able to reduce the dosage of chemical coagulant and indirectly reduced the drawback. The natural coagulant is an environmental-friendly product that able to treat water but not efficient as a chemical coagulant. It is also the inexpensive alternatives, which easily available for the reduction of the dosage of the chemical coagulant. In order for development of novel composite coagulant, the selection of high efficient coagulant is important for the successful treatment process. It is necessary to develop further research for the development of best coagulant that able to remove pollutants at wider pH, environmental friendly, has simple and economically standard process, and shows good result without much environmental issues.

Acknowledgement

The authors are grateful to Geran Penyelidikan Pascasiswazah (GPPS), VOT Project (U705), Universiti Tun Hussein Onn Malaysia and Fundamental Research Grant Scheme (FRGS), VOT Project (1570), Kementerian Pendidikan Tinggi for the financial support for this research.

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