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A Review of Wastewater Treatment using Natural Material and Its Potential as Aid and Composite Coagulant

(Suatu Penilaian Perawatan Sisa Air menggunakan Bahan Semula Jadi dan Potensinya sebagai Pembantu dan Komposit Koagulan)

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ABSTRACT

The introduction of natural materials into the coagulation-flocculation process need to be practiced as common as possible. The previous literature indicated that utilization of natural material is actually worth to be developed and if possible, into the commercial one. However, the application of natural coagulants itself as the primary treatment is not sufficient, due to the rise in constraints which limited its performance. Emerging technologies and depth studies are actually helping these limited conditions to grow them as good as the performance of chemical coagulants. Alternatively, the natural-based coagulants are commonly used as coagulant aids alongside chemical coagulants, which have created a highlight in water research. This review covers the type of coagulants used in the coagulation-flocculation of wastewater treatment especially in the usage of natural-based coagulants. This review paper also outlines the future prospects of natural materials as aids and its potential as sustainable composite coagulants.

Keywords: Chemical coagulant; composite coagulant; natural material; wastewater

ABSTRAK

Pengenalan bahan-bahan semula jadi ke dalam proses pemberbukuan-pembekuan perlu diamalkan sedapat mungkin. Kepustakaan terdahulu menunjukkan bahawa penggunaan bahan semula jadi sebenarnya berbaloi untuk dibangunkan dan jika boleh secara komersial. Walau bagaimanapun, penggunaan koagulan semula jadi sendiri sebagai rawatan utama tidak mencukupi, disebabkan oleh peningkatan kekangan yang membatasi prestasinya. Teknologi yang berkembang dan kajian mendalam sebenarnya membantu keadaan terhad ini untuk membangunkannya menjadi sebaik prestasi koagulan kimia. Sebagai alternatif, koagulan berasaskan semula jadi biasanya digunakan sebagai alat koagulan di samping koagulan kimia, yang telah menarik perhatian dalam penyelidikan air. Kajian ini merangkumi jenis-jenis koagulan yang digunakan dalam pemberbukuan-pembekuan pembuangan air sisa terutamanya dalam penggunaan koagulan berasaskan semula jadi. Kertas ini juga menggariskan prospek masa depan bahan semula jadi sebagai bantuan dan potensinya sebagai koagulan komposit yang mampan.

Kata kunci: Air sisa; bahan semula jadi; koagulan kimia; komposit koagulan

INTRODUCTION

Water is such the greatest gift of nature for the sustainability of ecological system and human beings. The cycle of water happen on Earth ensure the continuous supply for all forms of living organisms, from the mountaintops down into the oceans and to the smallest rivers. It is generally known that 71% of the Earth's surface is covered by water, but only 2.5% of the Earth's water is fresh water. The used fresh water is returned into the environment as wastewater, which not in the same sort of conditions when it was withdrawn anymore. Humans utilize and divert fresh waters a lot in many ways to drive significant economic, agricultural and support countless livelihood activities, thus unfortunately give tense pressure to the natural water bodies (Gani et al. 2017; Kalpana et al. 2016; Omotosho 2016). According to Corcoran et al. (2010), immense urbanization will lead to the raising of global populations and is expected to surpass nine billion by 2050, with 1 to 1.4 billion escalations in just a decade come from people living in slums alone. The growing population will cause more consumption of water thus generate larger volume of wastewater. The deteriorate quality of water resources cause more duty and pressure to the self-purification of streams and river bodies itself as well (Rajasulochana & Preethy 2016).

WASTEWATER AND ITS IMPACT

Wastewater can be interpreted in many variations of definitions. As being seen from a broad prospective, it can be interpreted as amalgamation of more than one of domestic effluent (black water and grey water), agricultural effluent, water from commercial institutions and manufacturing industries (Corcoran et al. 2010; De Souza et al. 2014; Jones et al. 2017; Raschid-Sally & Jayakody 2008). The developing countries suffer the most when people need to face the sanitation constraints and health risks due to the consumption of polluted water. Certain nitrogenous compounds including ammonia, nitrates, and nitrites have been notified to be in drinking water and various types of wastewater (Omotosho 2016). Today's emerging technologies coupled with proper management, wastewater can be redefined as beneficial sources. For instance, most of wastewater from Mexico City is utilized for irrigation districts, especially the Tula Valley (Corcoran et al. 2010). Wastewater reiteration in agriculture can perk the farmers in many ways such as conserving fresh water resources and balancing economic efficiency (Maryam & Büyükgüngör 2017).

Innumerable of divergent components from different sources can contaminate wastewater such as pathogens, organic matter and heavy metals. Sometimes their concentrations exceed the permissible discharge levels, thus build significant hazards (Simate & Ndlovu 2015). They are either soluble or emerge as particulate matter in the solution, which threatening the security of human, ecosystem and food production. Thus, considerable amounts of investment need to be consummated in order to have successful overcoming drawbacks of wastewater. Both urban and rural areas have to be addressed with their suitable integrated wastewater management to face the different context of challenges. In contrary of being the origin of problems, sound wastewater management will contribute a lot of helpful additives to the environment, health and economy.

Significant impact can be identified from improper management of wastewater. The crisis that the world faced today can affect the exploiting water demands in future, unless the wastewater management is reconstructed with sophisticated goals. The thriving problems that across the globe can emerge catastrophic influence on environmental and ecological sustainability, human health, food supply chain, and economic performance and the well-being of all forms of living organisms (Jones et al. 2017) in short and long term impacts such as eutrophication and loss of wildlife. The problems exasperate in the remote regions and undeveloped countries (Jadhav & Mahajan 2013), where the reliance on the untreated wastewater is high due to the lacking supply of hygiene water and sanitation service. The most abhorrent issues when it is involving mortality, which human need to deal with life insecure threats. The water borne diseases remain the greatest cause to halt the synergy between water and human worldwide. (Rajasulochana & Preethy 2016). It was reported that in 2012, about more than 800,000 deaths came from poor countries involving mostly children due to inadequate sanitation and contaminated drinking water (WWAP 2016).

Waterborne diseases that spread diseased and parasitic microorganisms into water bodies are the consequences from resources and knowledge lacking to construct and sustain a good sanitation system (Jadhav & Mahajan 2013). The high cases of Malaria disease is an instance showing worsen the further impact of wastewater. According to WHO (2017), an estimated 429,000 malaria deaths worldwide recorded that Africa region has the most portion of it. An estimated 70% of deaths occurred in children under the age of five in 2015. Besides, sanitation approached inadequacy trigger the bacterial illness such as cholera epidemics to spread as well. This worsens especially after natural disaster and civil disturbance, when the community services and facilities breakdown and cause clean water supply deterioration. Cholera outbreak in Haiti is a recent example of terrible consequences due to the lack of clean fresh water access, as thousands of people face the possibility to infect themselves from any water sources (Lewis 2017). Hence it has become a crucial need to protect water from getting polluted or to develop cost effective remedial method for its protection.

COAGULATION-FLOCCULATION TREATMENT METHOD

The emerging technologies nowadays lead to the introducing of more green conventional methods of waste water treatment. As the population grow, there is always an exciting need to keep developing the remedial action according to present and future problems. The grant funded by government give the opportunity to have on-going collaboration research between higher education, academic institutes and professional sectors to come out with implementable solutions (Maryam & Büyükgüngör 2017). According to the United Nations' prediction as stated by Goh and Ismail (2018), particularly half of the countries around the globe in future will confront with water stress and shortages. Water reuse and desalination are among the most favoured application to address the issues by offering safe water for daily consumption in many arid, remote and coastal regions. Conjointly, limited land available cause for wastewater treatment insufficiency. Thus urge the creation of under-control-advanced remediation by accelerating the forces of nature that compatible in smaller areas (Rajasulochana & Preethy 2016). Nature has such infinity occasion yet to be developed by present technologies.

Various domestic, industrial and agricultural activities involving water have caused the introducing of organic and inorganic substances into the environment. Some of them are easily treated such particulate matters that can be done through filtration method. However, according to Rajasulochana and Preethy (2016), to treat most of the compounds in a single step is not yet available. Several established conventional techniques to treat wastewater effluent such as ion exchange, solvent extraction, reverse osmosis, sedimentation and filtration require high maintenance and operational costs but less efficient in reducing the pollutants such as heavy metals (Simate & Ndlovu 2015). Historically, the application of conventional methods for treatment of waste water have been existed and cited since archaic times back in 19th century specifically coagulation-flocculation method (Choy et al. 2014; Rajasulochana & Preethy 2016; Saravanan et al. 2017). The primary goals of this method are to get rid the colloidal impurities as well as turbidity present in water. Coagulant is such a portent chemical developed by researchers to stabilize the colloidal particles by withdrawing forces that cause them to hang down (Saravanan et al. 2017). According to Zazouli and Yousefi (2008), coagulation is also the finest way to remove heavy metals. As if wastewater is treated through biological process, the non-biodegradable characteristics of heavy metal can pass through it thus makes it inefficient. Nonetheless, the reduction of chemical oxygen demand (COD) is often recorded low in aerobic and anaerobic processes of biological treatment due to the existence those heavy metals (Rasool et al. 2016).

Meanwhile, the technology of coagulation-flocculation has been used widely in diverse disciplines of water treatment (Shak & Wu 2015). The properties of used coagulants are the crucial factor in determining the coagulation performance, as it can boost or hinder the treatment efficiency (Sun et al. 2017; Verma & Kumar 2016). Coagulation and flocculation is also preferred in water engineering, resulted from a conducted survey over 20 treatment plants (Rui et al. 2012a). The simple operating conditions is said to be one of the factors for the treatment to be chosen (Rui et al. 2012b). Other than type of coagulants, several factors have been pointed out in order to help the efficiency of coagulation, namely volume dosage of coagulant, pH, mixing speed and its duration, temperature and settling time (Rui et al. 2012b). These parameters need to be controlled to come out with the satisfying and optimized impurities/contaminant reduction. Coagulation needs flocculation process to work efficiently. Coagulation by itself does not help much when the addition of coagulant may increase the insoluble compounds in treated sample (Al-Sameraiy 2012). Thus the subsequent flocculation step is needed, by allowing slow mixing as a key to the optimum performance. The second step concedes the collision, thus promote the agglomeration between particles.

In the laboratory scale, coagulation-flocculation technique is carried out by using jar test equipment. The experiment is conducted with two stages of mixing speed; rapid and slow mixing. The adding of sole coagulants usually is done during the rapid mixing, to blend the coagulants with water samples, before the slow mixing speed is applied in their respective time. Sufficient mixing time for both stages are important so that the agglomeration of particles can grow during sedimentation process (Al-Sameraiy 2012), which separate the solid or sludge from treated water and wastewater sample. Alongside cationic coagulants such as aluminium or iron salts as primary coagulants, the treatment also can be coupled with another polymer flocculants that help in faster accumulation of the time-taking settlings of small flocs (Verma & Kumar 2016). However, the overuse of chemical coagulants also lead to the additional problem (Abidin et al. 2011) such as big volume of sludge generation, that shows an urge to use natural-based coagulant as coagulating agents, which will be reviewed in detail in later section.

PERSPECTIVE ON USING COAGULANT IN WASTEWATER TREATMENT THROUGH THE YEARS

The particular impurities in wastewater are almost impossible to be removed without the adding of coagulant agents. The colloidal or suspended particles are negative loads that demand destabilisation to flocculate in the bunch as stability cause them to not sink in the bottom. The repellence between the colloidal particles will be reduced after the annexation of coagulant, resulted in small floccules. Assisted continuation of serene stirring will allow the floccules grow and further clustered together, settled down at bottom and separated from the water suspension itself (Saravanan et al. 2017; Zainol et al. 2011). The understandable mechanisms of it makes coagulation as an uncomplicated way for water purification since the 19th century (Choy et al. 2014; Jiang 2001). A lot of debates arise from researchers before regarding the mechanisms, proposing of what it actually takes to work in the best conditions. According to Jiang (2001), the scientific and depth study about coagulation starts approximately 100 years ago, after chemical coagulants such as aluminium sulphate and ferric chloride have been used for water treatment such in a big scale extensively. It is documented that the first proposed mechanism to explain coagulation was from Schulze-Hardy rule, while the theory of particle collision function was developed back in early 1917 (Jiang 2001).

Early in 1940s, further studies were done towards colloidal interaction, best known as van der Waals attraction but it was in 1960s when the introduction of micro-electrophoresis started, for the next encyclopedic coagulation theory. This led to the farther significant study of colloidal destabilization, by quantifying the zeta potential on the colloidal particles itself. It is also recorded that the stoichiometric relationship study between required coagulant dose to neutralize colloids and colloidal particles concentration in coagulation treatment bloomed around this decade (Jiang 2001). The usage of coagulants in wastewater's physical-chemical treatment has even been recorded since the earliest times, but using natural-based as the prime material. It was way foretime the natural-based coagulants especially the one that extracted from plants were used as coagulant for water clarification (Choy et al. 2014). For instance, Debora et al. (2013) stated that natural-based coagulants were used since 4000 years ago with utilization of by nirmali (*Strychnos potatorum)* in the countries such as India and Myanmar, while almonds and soaked beans were practiced in Sudan and Egypt since 16th century as water purifying agents to enhance water quality.

The globalization era starts to take over the consumption of old and aged treatment methods as sign of urbanization world, with massive technology and advancing knowledge, left the traditional ways behind. Over the years, obvious gaps are growing that differentiate the employment of natural and chemical coagulants. Both chemical and natural-based coagulants have been effectively acknowledged in various ancient records and past studies (Choy et al. 2014). However, it is sadly to be accepted that natural-based coagulants are no longer the main priority with the invasion growth of more advanced synthetic material, except the poor countries that have limitation in purchasing these goods. Nonetheless, there is arising concerns regarding chemical coagulants thus make researchers nowadays start to develop a more environmental friendly water and wastewater clarification agent, likely from non-chemical based. Chemical coagulant agents such as polyaluminium chloride and aluminium sulphate are among the most well-known and used in wastewater treatment. They are used in coagulation which is primarily occupied to diminish the content of impurities and organic matters that are devoting to water quality index (Zonoozi et al. 2011).

CHEMICAL AND NATURAL COAGULANTS

An efficient and easy operation is the ultimate reason for the wide usage and practice of inorganic coagulants such instance alum (aluminium sulphate), ferric chloride, and polyaluminium chloride in wastewater treatment (Ghafari et al. 2010; Ni et al. 2012). It is cited that metal coagulants had been used since 18th century in United States, which is also the third wold-biggest country, followed by China. While China had been using alum as an effective coagulant in their reign for water clarification and become one of the first pioneers ever since (Choy et al. 2014; Shen & Dempsey 1998). Apart from wastewater treatment, these synthetic coagulants also had been used in drinking water industry. Conventional metal coagulants, such as Fe (III) and Al (III), are the most widely used coagulants have bigger advantages but still cannot escape the reality of some drawbacks, generally known as lacking of green chemistry (Choy et. al. 2014). Despite the superiority of common and familiar used of these chemical-based, past literature have recorded few list of their disadvantages that cannot be ignored. The sustainability of treatment process slightly have negative view when it is linking to the neurological disease such as Alzheimer, especially when the aluminium residual penetrate into the human body and get accumulate in the brain (Amagloh & Benang 2009; Birima et al. 2013; Debora et al. 2013; Saravanan et al. 2017; Subramonian et al. 2014; Teixeira et al. 2017) although only few evidences are convincingly stated to clear the doubts (Amagloh & Benang 2009).

Another hazards on human health and living organisms in environmental surrounding also have been mentioned, particularly as intestinal constipation, convulsions, abdominal colic's, and loss of energy (Rasool et al. 2016; Saravanan et al. 2017; Subramonian et al. 2014). These synthetic metals and inorganic salts also have gloomy history associated in physical properties. Even though the cost of raw material to advent the products are claimed to be relatively efficient, the price will be blatantly higher when it enter the commercial market (Debora et al. 2013; Hesami et. al. 2014; Rasool et al. 2016) thus affecting the social factor in the rural communities. The post-treatment after coagulation also resulted in generation of large volume of toxic sludge that complicating the disposal process (Rasool et al. 2016; Teixeira et al. 2017) has become a source of debate. The toxic sludge is usually due to unreacted chemical residuals and reaction of by-products of polymers in treated water (Hesami et al. 2014; Subramonian et al. 2014). The generating sludge cannot be avoided as during coagulation process, since larger flocs are formed by smaller ones approaching to each other (Rong et al. 2013). The produced sludge need to have another stage of treatment process since coagulation is set to be as pre and primary treatment only. Furthermore, the pH of wastewater also needs to be altered first because of the efficient coagulation exclusively work only in certain pH and conditions. They are also inefficiently worked in low temperatures (Rong et al. 2013; Teixeira et al. 2017). The review of these hydrolysable cations' drawbacks in recent years, resulted in more breakthrough of natural polymers and organic coagulants. The search for eco-friendly and sustainable treatment processes are always encouraged as it gives lot of benefit to others. As stated by Debora et al. (2013), the reduction or replacement of these contentious coagulating agents will add the value to the sources of origin, while avoiding the chemical and physical post-effects in treated medium.

A great attention has been shifted to the sustainable water management, where the challenge to develop efficient processes especially in wastewater industry. Starting from natural ingredients to the laboratory-produced synthetic chemical based, on-going researches always look forward to natural materials again for the successful approaches of wastewater treatment. Natural coagulants especially from plants extracts always available in abundance and well-known as toxic free (Awang & Aziz 2012; Teixeira et al. 2017). Several studies have been done on utilization of natural coagulants or coagulant aids, either produced or extracted from plants, animal and agricultural wastes such as *Moringa oleifera* (MO) seeds, chitosan and cassava peel (Choy et al. 2014; Mohd-Asharuddin et al. 2017). Over the years, these organic coagulants such as (MO) seed and chitosan have been the most cited in the previous literature (Camacho et al. 2017; Debora et al. 2013; Ghebremichael et al. 2005; Rusdizal et al. 2015; Sánchez-Martín et al. 2012; Vieira et al. 2012; Zoonozi et al. 2011). The trending application of these coagulants are generally speaking up for the environmental-friendly technology, since they are renewable resources and achievable especially in the remote areas (Yin 2010). Biodegradable characteristics of natural coagulants that are assumed safe for ecological and living organisms, essentially exist way before the advent of chemical salts to clarify wastewater (Awang & Aziz 2012; Vieira et al. 2012). Originally, the lessen molecular weight of natural polymers are found to be the main excuse for the less efficient of coagulation (Altaher et al. 2016; Qudsieh et al. 2008). Thus it is usual to be applied as coagulant aid and not as the first preference. As eloquently stated by Altaher et al. (2016), natural polymer helps the formation of chemical bridge with the suspended solids during coagulation process when used as coagulant aid.

The sole reason for the drawback performance reported due to the increasing presents of organic matter from coagulants which may cause steric difficulty between polymer molecules at high concentration. At the same time, this also encourage for more depth researches regarding the substitution of natural materials in the commercial coagulants.

The emerging interests towards the usage of natural materials lead to more depth studies regarding their characteristics and behaviour in the coagulation process and for instance, the wide used of MO seed coagulant. It come from a multi-benefit tropical tree as highlighted by Food and Agricultural Organization of the United Nations (FAO), which abundantly found especially in the sub-Himalayan valleys, Asia, Africa and Latin America (Camacho et al. 2017; Debora et al. 2013; Marañón et al. 2010). Previous literature has stated that not only coagulant can be generated from the seed, the extracted edible oils also have been used in health and beauty products due to its nutritious source, while the remaining residual rind used as fertilizer, animal feed as well as activated carbon (Ghebremichael et al. 2005; Sánchez-Martín et al. 2012; Vieira et al. 2012). It is also reported to produce less volume sludge and showed comparable acts as alum in coagulation of high turbidity water (Ghebremichael et al. 2005). It has been identified that the MO seed extract contain cationic protein of polymerization which help in the low cost clarification procedures, has been widely used in the rural community areas to clear the cloudy water (Debora et al. 2013; Marañón et al. 2010). Another example, a natural polyelectrolyte of chitosan also has drawn much attention due to its high molecular weight and charged density along with its biodegradable polymer origin (Mohammadi et al. 2014; Pontius 2016; Zonoozi et al. 2011). Chitosan that is derived from deacetylation of chitin, are extensively found on natural sources and marine creatures such as arthropods, insects, crustaceans and fungi (Chi & Cheng 2006; Vieira et al. 2012; Zonoozi et al. 2011) while being the second most abundant biopolymer on Earth after cellulose (Jadhav & Mahajan 2013). Upon react with acidic solution, chitosan emits positive charged that work interactively with the anionic suspended colloidal, make it much favoured when is used as coagulant. The positive charge of chitosan's chain structure is due to the natural existence of amine functional groups (Jadhav & Mahajan 2013; Mohammadi et al. 2014; Zonoozi et al. 2011).

The natural ingredient of coagulants also could be potentially derived from natural waste, such as agricultural waste. Agricultural wastes are among the most type of waste found that contribute to its largest proportions in the landfills, as they lack in economic value and have commercialization constraints (Bories et al. 2009). For instance, the available waste-derived coagulant is cassava peel. The utilization of cassava peel has great concerns since the industries related to cassava processing produce huge amount of wastes, due to only its cassava starch is being used for further commercialization (Heuzé et al. 2016; Howeler et al. 2001). Statically, there are about 47 tons of fresh by-products, produced by 100 tons of tubers per day from a cassava starch processing unit that can cause negative impacts to ecological surrounding if being disposed carelessly (Heuzé et al. 2016). Cassava peels contain large amounts of cynogenic glucosides, besides they also can emit a strong offensive smell and carbon dioxide from the rotten waste heaps. A study carried out by Zayadi et al. (2016) stated that that cassava peel has potential to be selected as waste-based coagulant due to its ability to remove nutrient and heavy metals, supported by the presence of Si, Ca, Mg, Fe, abundant functional groups and surface porosity of media as the main features. This natural-polymer coagulant shows a promising choice for water coagulants due to their biopolymers composed of proteins and polysaccharides (Mohd-Asharuddin et al. 2017; Zayadi et al. 2016). Summarize in a Table 1 are various natural coagulants applied on wastewaters with its contaminants' removal percentages to portray its efficiency.

FUTURE POTENTIAL AND CONSTRAINT OF NATURAL MATERIALS AS COMMERCIAL COAGULANT

A satisfying wastewater management will give benefits to many parties, with environment and society as the first preference. As stated by Teixeira et al. (2017), the real challenge arise since there is a need to find eco-friendly and economical viable actions toward efficient water treatment. Besides removing hazardous contents, the adding of nutritional value (Teixeira et al. 2017) is also the extra points in regulating wastewater through various researches that might be convenient in future prospects. In the perspective of natural coagulants, it is not impossible to develop them as commercial ones based on the bright future potentials. Essentially, past studies have recorded that natural material-based coagulants produce lesser volume of sludge (Altaher et al. 2016; Rasool et al. 2016), thus, convince the utilization of these alternative and possible choice of coagulants, since it cut the cost of sludge treatment. The naturally occurring materials also highly biodegradable (Awang & Aziz 2012; Birima et al. 2013; Rasool et al. 2016; Subramonian et al. 2014), non-hazardous and toxic free (Rasool et al 2016; Rusdizal et al. 2015; Teixeira et al. 2017), differ than chemical-based that insecure the human health and cause environmental hazards.

Coagulants derived from natural materials can also be found in abundance especially when it is plant-based and agro waste. The plants such as *Moringa oleifera* and rice starch can be locally grown, where there is a lot of land space available (Rasool et al. 2016; Teixeira et al. 2017). The thoughts in commercialization of natural materials as coagulants might also boost the social factor of the locals that rely on these agricultural economy (Pondja et al. 2017; Yin 2010), also probably as solution towards wastewater and agro waste pollution. The renewable sources of them also mean the continuous supply of raw materials in development of coagulants. Therefore, the cheaper cost of coagulating agents could be obtained in future in attempt to

TABLE 1. Research summary of natural coagulants used in wastewater treatment

No.	Coagulant	Type of wastewater	Removal parameter (%)	Reference
1	*Moringa oleifera* seed	Landfill leachate	Heavy metals; Fe (100%), Cu & Cd (98%), Pb (78.1%), respectively	Shan et al. (2017)
		Textile industry	Heavy metals; Cd, Cr, Mn (100%), respectively Turbidity (98.6%), biochemical oxygen demand, BOD (11.7%)	Hendrawati et al. (2016)
		Palm oil mill effluent (POME)	Suspended solids (99.2%), chemical oxygen demand, COD (52.5%)	Othman et al. (2008)
2	Chitosan	Oily produced water	Oil (96.35%)	Hosny et al. (2016)
		Acid dyes	Chitosan as aid with polyaluminium chloride (PAC); dye AB292 (52%)	Zonoozi et al. (2011)
		Wastewater from milk processing plant	Turbidity (94.3%)	Chi & Cheng (2006)
3	*Ocimum basilicum* (basil)	Landfill leachate	Basil as aid with alum; COD (64.4%), Colour (77.8%)	Rasool et al. (2016)
		Textile wastewater	Basil as aid with alum; COD (76.4%), Colour (80.4%)	Shamsnejati et al. (2015)
4	Banana pith	Polluted river water	Turbidity (98.5%), COD (54.3%), Suspended solids (96.03%), sulphates (98.9%), nitrates (88.7%), Cu (100%), Cr (100%), Fe (92%), Zn (81%), Pb (100%), Mn (60%)	Kakoi et al. (2016)
5	*Corchorus olitorius* L.	Humic acids wastewater	Turbidity (95.38%) Total organic carbon, TOC (100%)	Altaher et al. (2016)
6	*Opuntia ficus-indica*	Textile effluents	Jeans laundry effluent; COD (64.77%), turbidity (91.26%) Fabric dyeing mill effluent; COD (87.19%), turbidity (93.62%)	De Souza et al. (2014)
7	*Jatropha curcas* seed	Kaolin synthetic wastewater	Turbidity (96%)	Abidin et al. (2011)
8	*Cassia obtusifolia* seed gum	POME	Aid with alum; total suspended solid, TSS (81.58%), COD (48.22%)	Shak & Wu (2015)
		Raw pulp and paper mill effluent	TSS (86.9%), COD (36.2%)	Subramonian et al. (2014)
9	Nirmali seed	Laundry waste surfactant	TSS (75.5%), turbidity (95.5%)	Mohan (2014)
10	Pectin of orange peel pith		Dual coagulants with nirmali seed; turbidity (89.5%), TSS (81.5%)	
11	Lateritic soil	Landfill leachate	COD (65.7%), colour (81.8%), ammonia (41.2%)	Syafalni et al. (2012)
12	Jackfruit seed starch (JSS)	Landfill leachate	JSS as aid with PAC; COD (26.85%)	Yusoff & Mohamad Zuki (2015)
13	Rice starch	POME	TSS (84.1%)	Teh et al. (2014)
14	*Hibiscus sabdariffa* Roselle Seed	Synthetic wastewater	Turbidity (93.13%)	Saharudin & Nithyanandam (2014)
		Industrial wastewater	Turbidity (87.18%)	
15	*Hibiscus rosa-sinensis* leaf extract	Textile wastewater	Hibiscus leaf as aid with PAC; COD (77.8%), suspended solids (99.4%), colour (78.4%)	Aziz et al. (2012)
16	Tapioca starch (TS)	Partially stabilized leachate	Suspended solids (12%), colour (54.7%), ammonia (13.2%)	Mohd-Zin et al. (2016)
		Semiconductor wastewater	Turbidity (99%) COD (87%)	Fatehah et al. (2013)

replace the hazards of using inorganic metal salts (Altaher et al. 2016; Yin 2010). Fundamentally, the utilization of plant-based coagulants during early years also initiates further researches regarding the potential natural materials to be developed as coagulants (Awang & Aziz 2012; Birima et al. 2013; Subramonian et al. 2014).

However, a few constraints also could be seen in commercializing the coagulants derived from these bio-based. A study by Choy et al. (2014) stated few distinct hinder factors that have the tendency in failing the growth of natural materials into profit-making products, which are financial source, research and development, and market awareness. Basically, the potential investors are likely having doubts in investing in new products, lead to the limited financial supply. The depth study of new knowledge is also the key for further development in researches, so its certify factors such as strength and weakness can be identified. Awareness regarding natural materials also needs to move simultaneously to seek for demands in replacing the synthetic-inorganic coagulants. Besides, the working conditions of it to earn optimal achievements might be questioned. Next, the accurate target market must be aimed to the achieve practicability of both suppliers and consumers.

While Altaher et al. (2016) and Choy et al. (2014) mentioned that natural polymers could emerge the poor performance when it is used as sole coagulants in remediating turbid wastewater. The after treatment might resulted in immense concentrations of organic matter, thus worsen the turbidity level. The identification of this problem can be tackled by accentuating the merge of natural and chemical coagulants, either in the same or different treatment. The convincing coagulation performance of combined coagulants will likely to improve treatment performance and reduce the ratio of chemical coagulant as well (Rasool et al. 2016). The combined coagulants can work either as coagulant aid or composite coagulant (Shak & Wu 2015).

UTILIZATION OF NATURAL MATERIALS AS COAGULANT AID AND COMPOSITE COAGULANT

The pressure on wastewater treatment intensifies, leading to tension, conflict among users and excessive pressure on the environment (Mohan 2014). The goal for addition of aids during coagulation is to further optimize the conditions together with primary coagulant. By doing so, faster and stronger formation of flocs, lower coagulant dosage volume, and higher reduction of impurities are expectantly done (Hesami et al. 2014). In recent years, natural coagulants have been researched as a more environmentally friendly approach to substitute the commercial coagulants in coagulation flocculation treatment. Natural-based coagulants have been applied on wastewater as either a primary coagulant or as a coagulant aid for a chemical coagulant (Rusdizal et al. 2015). However, the effectiveness study of natural coagulants is still at the initial state. Further analysis is important since the combine mechanisms of composed mixture help to understand the coagulation performance (Kumar et al. 2017).

An urgency to develop affordable and green coagulant that benefit others should not be lagged. Previously, several successful application of natural materials as sole coagulant and coagulant aid has been mentioned; such as using lateric soil (Syafalni et al. 2012), natural seed gum (Shak & Wu 2015), jackfruit seed starch (Yusoff & Zuki 2015), MO seed (Choy et al. 2014), cassava peel (Mohd-Asharuddin et al. 2017) and chitosan (Vieira et al. 2012). Undeniably, dual coagulant is able to reduce the dose of chemical, thus minimize the toxicological effect. Substitution of chemical coagulant with natural material can be applied to reduce the negative sides, simplify the process and tackle the other drawbacks (Kumar et al. 2017; Mohd-Zin et al. 2014; Syafalni et al. 2012). As eloquently stated by Mohd-Zin et al. (2014), composite coagulant is made by combining two types of coagulant as one reagent by premixing the coagulant, through certain working conditions. The operation using composite coagulant will be more convenient as it abridges the treatment practice and effective in financial perspective. The association of both materials can be reviewed by past studies; many researches have interests in synthesizing of inorganic-inorganic (Moussas & Zouboulis 2009; Tzoupanos & Zouboulis 2011) and inorganic-organic materials (Lee et al. 2012). A study by Moussas and Zouboulis (2009) stated that the new combination of materials has increase the polymer species concentration and promote good properties of coagulant. Increasing the quantity of polymeric species in the original structure or producing new composite coagulants to insert other components are techniques that can improve the coagulation-flocculation efficiency (Moussas & Zouboulis 2009). They also exhibit good stability better than primary single coagulant (Huang et al. 2014), as the increase molecular size enhance the aggregating ability. It is assumed to improve the formation capability of bridge among colloidal, high molecular weight coagulants are more effectual (Wang et al. 2017a), due to an intrinsic unity of relation among the floc properties under various coagulation mechanisms. The polymer bridging is likely to occur when the polymer and particle inside have similar charge, lead to the strengthen flocs and size.

The application of composite coagulant also said to be able of overcoming the arise problems such as generated small flocs during slow mixing speed of sole coagulants (Zhu et al. 2012), as a positive result from modifiers that comprise of high cationic charge coagulating species. Furthermore, study by Wang et al. (2017b) indicated that the aggregated flocs induced by the composite coagulant could be more resistant to shear and more recoverable than those induced by the non-composite coagulant. Alongside with less operation time and stable aggregating ability, composite coagulant also conceivably produces low sludge amount. The fast formation and lesser volume of floc and sludge after settling are preferred because it means low-cost sludge treatment (Mohd-Zin et al. 2015). The lower residual of metal salts that remains in the treated sample is one of the main advantage of composite coagulant as well (Tzoupanos & Zouboulis 2011). The substitution of natural polymer into the commercial coagulant has given a significant role to assist the better performance of coagulation in research and development of wastewater treatment.

Conclusion

Natural material is relatively environmental-friendly, thus become an appealing substantial in wastewater treatment process. The utilization of natural coagulants can be further commercialized by tackling its limitation factors via progressing them as aids or composite ones. Despite the disadvantages of sole chemical coagulants and the limited ability of natural materials to work at their finest, the formation of coagulants from both materials might be the next big-key solution. The rise curiosity in exploring water and wastewater treatment industries, especially via coagulation-flocculation method shows the bright future of natural coagulants indeed.

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