



REVIEW PAPER

## Use of natural coagulants for industrial wastewater treatment

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### ABSTRACT

Industrial effluents are a menace to the environment and the fact that their characteristics vary from industry-to-industry only adds to the complex challenge they offer to the engineers and scientists. Resource-efficient and environment-friendly solutions to this hazard are a call of the hour. Coagulation, by synthetic chemicals, has been used as a cost-effective and efficient method for managing the effluents generated by a large number of industries. However, the synthetic chemicals themselves are a cause of concern due to their non-native nature, non-degradability, and health conditions associated with their left-over residues. Natural coagulants offer a cost-effective, environment-friendly, and sustainable alternative to the application of synthetic chemicals. Such natural coagulants, despite their demonstrated effectiveness in treating the industrial wastewaters, have their own limitations and are yet to be investigated for large-scale applications. The current work presents a state-of-the-art review of the natural coagulants' application in treating industrial wastewaters and their relative advantages and disadvantages as compared to the chemical coagulants. Future research areas have also been identified that may ultimately lead to the large-scale commercial application of natural coagulants and will result in an environment-friendly and sustainable solution to the problems created by industrial effluents and synthetic chemical coagulants.

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## INTRODUCTION

Rapid industrialization has posed many threats to the environment due to the wastewater generation through various industrial processes. Indiscriminate disposal of this wastewater (with or without an appropriate level of treatment) can cause water pollution and land pollution. This has a profound effect on the health of living beings apart from the impacts on the abiotic components such as soil (Chhonkar *et al.*, 2000) and water (Pal *et al.*, 2010). Some key sources of industrial wastewater are palm oil mills (Bhatia *et al.*, 2007; Jagaba *et al.*, 2020), paper and pulp industry (Ashrafi *et al.*, 2015; Wang *et al.*, 2011; Chaudhari *et al.*, 2010), brewery and winery (Brito *et al.*, 2004), cosmetic industry (Naumczyk *et al.*, 2014), tannery industry (Rameshraj and Suresh, 2011), slaughterhouses (Bustillo-Lecompte *et al.*, 2015), paint industry (Aboulhassan *et al.*, 2014), dairy industry (Triques *et al.*, 2020), etc. A huge quantity of industrial wastewater is generated worldwide daily. Saha *et al.* (2005) claimed that a single unit of Indian distillery uses 1133.5 KLD of water, and, after processing, around 668 KLD of wastewater is generated. In another attempt to quantify the wastewater generation in India, Majumder *et al.* (2014) estimated that every day around 13,468 MLD of wastewater is generated, out of which, more than 50% is discharged into the environment untreated. The quantity and quality of industrial effluent are functions of the ongoing industrial process, raw material utilized, and products manufactured in each industrial unit, and therefore the composition and constitution of wastewater generated are different for different industries. For instance, the textile dyeing processes in the textile industry generate colored wastewater rich in chemical dyes (Ariffin *et al.*, 2009). On the other hand, the effluent from paper and pulp industry contains large quantities of sodium salts of organic acids, lignin, etc. (Wang *et al.*, 2011). Wastewater from the cosmetic industry has significant concentrations of COD, oils, detergents, fats, and suspended solids (Tobajas *et al.*, 2014). Muralimohan *et al.* (2014) studied the various processes contributing to the wastewater generation in the textile industry. The study revealed that processes like cleaning, de-sizing, etc. are responsible for toxic wastewater generation. The main high-risk constituents of this wastewater are chemicals, dyes, bleaching agents, oils, acids, bases, etc. Sometimes,

wastewater contains heavy metals which are a potential cause of concern for the environment and human health (Ahemd *et al.*, 2020). Prasad and Rao 2016 studied the impact of the cadmium pollution, caused due to the discharges from the mining activities. Elevated levels of cadmium can cause diseases like Itai-Itai; whereas, at low levels, it may cause problems like kidney damage, sterility among males, flu disorders, and high blood pressure. The characteristics of wastewater have been studied in terms of key parameters like BOD (Sehar *et al.*, 2013), TDS, COD (Rathi and Puranik, 2002), and color. These parameters help in gauging the level of potential hazard the effluent can pose to the environment and human health. Due to the potential toxicity, industrial effluents may require prior treatment before their release into the environment. Improper disposal of the wastewater can cost liability in addition to their environmental impacts (Elsheikh and Al-Hemaidi, 2013). Sahu and Chaudhari 2013 suggested that industrial wastewater management is necessary to eliminate the health and socio-economic concerns. Wastewater treatment technologies can be physical, chemical, or biological (Elsheikh and Al-Hemaidi, 2013). Among the physicochemical processes, coagulation-flocculation has been frequently used. In this process, the charge of the colloidal particles is destabilized with the help of coagulants (typically aluminium or iron salts) which results in floc formation due to collision of destabilized particles and their aggregation, which ultimately gets separated from the liquid phase. An alternative to the conventional coagulation-flocculation is the electrocoagulation process. Here, coagulants are formed due to electro-dissolution of the anode which causes hydrolysis products that result in the destabilization of the particles (Verma and Kumar, 2018). Electrocoagulation has been successfully used for the treatment of textile wastewater using iron and aluminium electrodes (Kobyta *et al.*, 2003), synthetic textile wastewater (Merzouk *et al.*, 2011), sewage (Carballa *et al.*, 2005), vegetable oil refinery wastewater (Tezcan *et al.*, 2009), paint industry wastewater (Akyol, 2012), olive oil mill wastewater (Salameh, 2015), petroleum refinery wastewater (El-Naas *et al.*, 2009), etc. This process has also been used to successfully remove harmful substances, such as Diclofenac (responsible for vulture population decline) (Oaks *et al.*, 2004; Green *et al.*, 2004), and

a carcinogenic dye, acid red 14 (Ahangarnokolaie *et al.*, 2017). Electrocoagulation requires very small settling time due to the rapid settling of the flocs and results in lesser sludge production (Ahangarnokolaie *et al.*, 2017). However, complicated experimental setup with the electricity or current input is required for the treatment which limits its commercial application by increasing the operational cost of the treatment unit (Perren *et al.*, 2018). Therefore, electrocoagulation cannot be seen as an economical option for a large-scale industrial wastewater treatment. Coagulation, on the other hand, is an important chemical treatment method and is very effective for wastewater treatment (Carballa *et al.*, 2005). Wastewater streams obtained from industries like tannery (Ariffin *et al.*, 2009), hospital (Wang *et al.*, 2011), paper and pulp mill (Wang *et al.*, 2011), etc. have been treated by coagulation using synthetic chemicals. The use of chemical coagulants for coagulation of wastewater has various implications such as their potential to cause diseases (WHO Guidelines, 2010), the possibility of groundwater contamination or surface runoff of treated water containing high residual aluminium concentration and therefore their use for the wastewater treatment is not an eco-friendly option. On the other hand, the use of natural substances for coagulation in place of chemicals is a promising alternative for the treatment of industrial wastewater. Natural coagulants are safe for consumption (owing to their plant-origins) and are biodegradable in the environment (Feria-Díaz *et al.*, 2016; Nath, *et al.*, 2020). Plant components, which are otherwise considered as waste are an economical option, as compared to the synthetic

chemicals. Natural coagulation has been used for treating wastewater from various industries like textile (Muralimohan *et al.*, 2014; Shankar *et al.*, 2014; Dotto *et al.*, 2019; Prabhakaran *et al.*, 2020), dairy (Sivakumar *et al.*, 2014; Sivakumar, 2015; Sivakumar *et al.*, 2016; Triques *et al.*, 2020), tannery (Ahmed *et al.*, 2020), etc. A compilation of the various chemical and natural coagulants used for the treatment of industrial wastewater, from literature studies, has been provided in Table 1.

In comparison, some of the synthetic coagulants used for the treatment of domestic sewage include alum (Sarparastzadeh *et al.*, 2007), ferric chloride (Sarparastzadeh *et al.*, 2007), poly-aluminium chloride (El Samrani *et al.*, 2008) and aluminium sulphate (Fabres *et al.*, 2017). Several research studies have shown the potential of natural coagulants in treating the wastewaters originating from different industries and have shown them to be equivalent to chemical coagulants in terms of treatment efficiencies for various parameters of interest. Coagulation using natural coagulants has also been effectively used for the water treatment. However, the current work is solely focused on industrial wastewater. A brief introduction of the coagulation-flocculation process and factors affecting this process is presented, followed by a review of the applications of chemical coagulants and their advantages and disadvantages. The need for reducing the residual aluminium in water is emphasized by a brief review of the relevant literature. Natural coagulants are discussed as an attractive and environment-friendly alternative to the chemical coagulants and some of the more commonly studied natural coagulants are

Table 1: Chemical and natural coagulants investigated for the coagulation of industrial wastewaters.

Chemical coagulants	Natural coagulants	
Potassium Aluminium Sulphate (KAl(SO <sub>4</sub> ) <sub>2</sub> ·12H <sub>2</sub> O)	Chitosan	<i>Opuntia mucilage</i>
Aluminium sulphate Al <sub>2</sub> (SO <sub>4</sub> ) <sub>3</sub>	<i>Moringa oleifera</i>	<i>Psyllium husk</i>
Ferric chloride (FeCl <sub>3</sub> )	<i>Abelmoschus esculentus</i> (okra)	<i>Plantago major L.</i>
Iron(III) chloride hexahydrate	<i>Malva sylvestris</i> (mallow)	<i>Hibiscus Rosa sinensis</i>
Poly aluminium chloride (PAC)	<i>Ocimum Basilicum</i>	<i>Trigonella foenum-graecum</i>
Lime (CaO)	<i>Sechium edule</i> (Chayote)	<i>Dolichos lablab</i>
Magnesium chloride (MgCl <sub>2</sub> )	<i>Plantago ovata</i>	<i>Cicer arietinum</i>
Ferric sulphate	<i>Zea mays</i> (maize)	Roselle seeds
Poly ferric acetate	Cactus	Banana juice
Poly ferric sulphate	<i>Tamarind Indica</i>	<i>Strychnos Potatorum</i>
Poly aluminium ferric chloride (PAFC)	Bitter gourd seed	Castor oil cake
Poly aluminium silicate chloride (PASic)	Cotton seed oil cake	Surjana seed powder
	Maize seed powder	

described in detail. The advantages offered by these plant-based substances and the issues impeding their commercial-scale application are also discussed. The authors have also identified several constraints that prevent the commercial adoption of the natural coagulants for wastewater treatment. To overcome these constraints, some areas of future research have been identified and are likely to promote the large-scale adoption of natural coagulants. Recommendations have also been made for usage of natural coagulants over the synthetic chemicals. Natural and environment-friendly options for managing industrial wastewaters are not only cost-effective and sustainable; but may also ensure that industrial effluents are treated (before disposal) in the far-flung areas in developing and the under-developed world, where the technical expertise is absent and chemical coagulants are not easily available or are too expensive to be considered a viable option. This study is aimed at providing state-of-the-art and comprehensive review of the application of natural coagulants for treating industrial wastewaters. This work was done in Greater Noida, Uttar Pradesh, India during 2018-20.

#### Chemical coagulation

Chemical coagulation is the process of destabilizing the colloidal impurities in water or wastewater by using chemically produced substances. The process of formation of flocs on charge neutralization is known as flocculation (Hamawand, 2015). Aluminum sulfate (Quintero-Jaramillo *et al.*, 2016), potassium aluminium sulfate, Iron (III) chloride hexahydrate, ferric sulfate (Karamany, 2010), etc. can be used as coagulants for water and wastewater treatment. Alum is the most commonly used chemical coagulant. Alum contains potassium and aluminum, and is chemically represented as  $KAl(SO_4)_2 \cdot 12H_2O$ . Alum or potassium aluminium sulfate is very frequently used for water and wastewater treatment. Alum is easily available and is an inexpensive alternative for the wastewater treatment. Madhavi *et al.* 2014 used alum for treating wastewater from the metal fabrication industry. Alum, at a concentration of 450mg/L, removed 99% of color from the wastewater at a pH of 8.0 (Madhavi *et al.*, 2014). The effectiveness of alum can be increased by using it with other coagulants. In a study, alum, when used with polyacrylamide (PAA) and poly ferric sulfate (PFS), resulted in an increase

in the efficiency of COD removal to 82% from 68% (Loloei *et al.*, 2014). Jiang and Llyod 2002 reviewed the potential of ferrate salts for the treatment of wastewater and reported their potential application for wastewater treatment. Iron (III) chloride, also called as ferric chloride (represented as  $FeCl_3$ ), can also be used as a coagulant for wastewater treatment. Bogacki *et al.* 2011 used ferric chloride for the treatment of cosmetic industry wastewater with the aim of COD reduction. Using ferric chloride, up to 63.9% reduction of COD was achieved at a pH of 6.0. Poly aluminium ferric chloride can also be used as a coagulant (Ebrahimi *et al.*, 2014). Liang *et al.* 2009 used ferric chloride for treating the molasses wastewater. At optimum conditions, 96% color and 86% COD could be removed from the wastewater (Liang *et al.*, 2009). Ebeling *et al.* (2003) used ferric chloride to remove 93% orthophosphate from aquaculture discharge. Iron (II) sulfate, also called ferrous sulfate (represented as  $FeSO_4 \cdot xH_2O$ ), can act as coagulating agent for wastewater treatment. In a comparative study using coagulants: lime, alum, ferric chloride ( $FeCl_3$ ), ferrous sulfate ( $FeSO_4$ ), and magnesium chloride ( $MgCl_2$ ) for the treatment of textile industry wastewater, Bidhendi *et al.* 2007 found that ferrous sulfate was the most effective and could remove color at low coagulant dose, had minimum settled sludge volume and maximum decolorization of wastewater. Loloei *et al.* 2014 did a comparative study of alum and ferric sulfate for treating the wastewater from the dairy industry. Ferric sulfate could remove up to 95% turbidity and 62% COD (Loloei *et al.*, 2014). Aluminium sulphate is an aluminium salt that can be used as a coagulant for wastewater treatment. Poly aluminium chloride (PAC) can be used as a coagulant for wastewater treatment. Fattah *et al.*, 2013 used poly aluminium chloride for automotive wastewater treatment and reported that it could remove 98% iron, 83% zinc and 63% nickel. Chemical coagulants, after immobilization, were found to be more effective in reduction of TDS, phenolphthalein, total phenolphthalein, COD, and chromium as compared to their native forms (Imran *et al.*, 2012 ; Aravind *et al.*, 2016). Immobilized ammonium aluminium sulphate was found to be more effective for the chromium removal from tannery industry wastewater (Imran *et al.*, 2012). Gregory and Duan, 2001 discussed the charge neutralization by hydrolyzing metal coagulants and the effects associated with precipitated metal

hydroxide. The study also reviewed the basic principles of colloid stability and metal ion hydrolysis. Some of the hydrolyzing salts that can be used as coagulants to aid coagulation and flocculation process are PAC (Pal *et al.*, 2010; Alavijeh *et al.*, 2017), Poly ferric sulphate, poly aluminium ferric chloride, and. They have superior color removal efficiency even at a small dosage and are effective for treating wastewater over a wide pH range (Muralimohan *et al.*, 2014). Wei *et al.*, 2017 used poly ferric acetate to remove phosphorus from synthetic wastewater prepared using 0.2 g/L kaolin solution. It was found that, under optimum pH range of 7.0-9.0, the removal percentage of phosphorus was 96.1 % using poly ferric acetate, proving it to be a promising alternative for phosphorus removal from wastewater. Irfan *et al.*, 2017 used chemical coagulants such as aluminium chloride, poly-aluminium chloride, and anionic PAM for treating black liquor wastewater. The combination could remove 88% color, 95% total suspended solids and 81% COD. Response surface methodology (RSM) is a new approach in the wastewater treatment. This technique uses a combination of mathematical and statistical models to determine the efficiency of the coagulants in the coagulation and flocculation process. Thirugnanasambandham *et al.*, 2014 used this technique for the bagasse wastewater treatment with the help of biopolymer. Wang *et al.*, 2011 used aluminium chloride as a chemical coagulant for paper mill wastewater treatment. In addition, a modified natural polymer was used as a flocculant. The models were developed to obtain the efficiency of chemical coagulants in terms of turbidity and lignin removals. For the experiments, the dose of coagulant used was 871 mg/L and to aid the process, a flocculant dose of 22.3 mg/L at pH 8.35 was selected. Experiments confirmed that combination of the Uniform Design and RSM is an efficient approach for the treatment of paper and pulp mill wastewater. Ghafari *et al.*, 2009 investigated the efficiency of poly aluminium chloride and alum for leachate treatment. Using Central composite design (CCD) and response surface method (RSM), quadratic models were developed. Using 2 g/L PAC (at a pH of 7.5) and 9.5 g/L alum, the efficiency of the coagulants was determined. The predicted efficiency of the coagulants matched with the experimental data. The COD, turbidity, color, and TSS removal efficiencies for PAC were reported to be 43.1%, 94.0%, 90.7%, and 92.2%, respectively.

The comparable efficiencies for alum were found to be 62.8%, 88.4%, 86.4%, and 90.1%, respectively. Chemical coagulants have several advantages. The easy availability and cheap prices are some of the reasons which outweigh the issues and concerns associated with the large-scale application of these coagulants on the commercial level. The efficiency of chemical coagulants is not affected by the pH variation as they are effective over a wide pH range. The optimum conditions are obtained at very low dosages. The easy availability of chemicals, ease of storage, no efficiency loss with long-term storage, reliability of the system and better efficiency are some of the key advantages of the chemical coagulants (Semerjian and Ayoub, 2003). The efficiency of the chemical coagulant gets influenced by several different factors. Next section will comprehensively detail the factors affecting the efficiency of chemical coagulants.

#### *Factors affecting the efficiency of chemical coagulants*

The factors affecting the efficiency of coagulation process are characteristics of wastewater, the type and the quantity of coagulant used (Azhar *et al.*, 2016). Temperature also has a very significant effect on the flocculation process. Joudah 2014 studied the effect of temperature on the flocculation efficiency of coagulants. The study claimed that, at lower temperatures, the flocculation efficiency is reduced as the bigger flocs get broken (Joudah, 2014). Warmer temperatures are therefore, preferred to ensure better flocculation (Fitzpatrick *et al.*, 2004). The effectiveness of coagulants in the wastewater treatment depends on the wastewater pH. Altaher and Alghamdi 2011 comprehensively studied the effects of pH, temperature and stirring rate on the coagulation and flocculation efficiency of four coagulants: alum, ferrous sulfate, ferric chloride, and commercial synthetic cationic polymer. The study reported that the wastewater pH significantly affected the turbidity reduction. The highest removal efficiency was obtained at higher pH values. Variation in the stirring rate did not affect coagulation efficiency. The process of coagulation has been identified as an effective method for treatment of the wastewater having variations in temperature, dosage, and concentration. Zhang *et al.* 2018 used the combination of poly aluminium chloride and chitosan for the treatment of low temperature and low

turbidity water. This coagulant combination yielded removal efficiencies of turbidity, DOC and UV254 of approximately 87%, 63%, and 82%, respectively. In another study, [Dhivya et al. 2017](#) noted that the natural coagulant *Plantago ovata* (*P. ovata*) worked best at the room temperature and when tested for the varying turbidity values removal efficiencies in range 98.2% to 80.2% were obtained. However, PO-NaCl was found to be most effective in alkaline conditions. [Saritha et al. 2020](#) used various combinations of natural and synthetic coagulants for water treatment and turbidity removal of 99.29 % was attained at all the pH variations and dosages. Thus, it is necessary to consider the effect of parameters such as temperature, dosage and concentration of the source water on the coagulant dosage and efficiency.

#### *Addition of flocculants*

The efficiency of the chemical coagulants can be further improved by the addition of flocculants. Flocculants are the substances which link destabilized particles thereby causing settling of the colloidal impurities in the form of flocs. [De Godos et al. 2011](#) used  $\text{Fe}_2(\text{SO}_4)_3$ ,  $\text{FeCl}_3$  and five commercial polymeric flocculants (Chitosan, Floccudex CS/5000, Drewfloc 447, Flocusol CM/78, and Chemifloc CV/300) to evaluate their efficiency of algal biomass removal from the piggery wastewater. Biomass removals of up to 98% were achieved using the combination of 150–250 mg/L ferric salts and 25–50 mg/L flocculants. [Vanerkar et al. 2013](#) treated food industry wastewater using coagulation and flocculation process and discussed the behavior of coagulant supplemented with polyelectrolytes. Different coagulants and flocculants were tried to attain the best results and lime was selected as the optimum coagulant based on the cost factor. Lime was used for the coagulation and resulted in 53.59% COD and 57.19% BOD reductions respectively, at the dosage of 200 mg/L. Only, 25 mL/L of sludge was developed at this dose of the coagulant. Alum dosage was not very effective for the treatment. 0.3 mg/L of Magnafloc E-207 in combination with 200 mg/L of lime effectively reduced 67.61%, 71.01% and 81.53% of COD, BOD and SS, respectively ([Vanerkar et al., 2013](#)). [Wong et al. 2006](#) used anionic and cationic polyacrylamides to treat paper and pulp mill wastewater. [Aguilar et al. 2005](#) also used anionic polyacrylamide for improving coagulation-flocculation process for treatment

of slaughterhouse effluent. The use of anionic polyacrylamide helped in increasing the coagulation efficiency of the coagulants, ultimately reducing the cost of treatment. Chemical coagulants have also been used in conjunction with other processes to get zero discharge. [El-Awady et al. 2019](#) studied the possibility of reusing treated water from a paper recycling mill industry to obtain zero discharge from the unit. The wastewater treatment was carried out using synthetic coagulants like alum, ferric chloride and cationic polymer. Removal efficiency up to 98.9% was obtained for TSS and up to 79.4% was obtained for COD. It was noted that the treated water was fit to be reused at the unit for the industrial activities. [Mohammadtabar et al. 2019](#) tried five chemical membrane hybrid processes, in conjunction with coagulation, for the treatment of boiler blowdown water from oil and sand stream. This experimental study showed that the direct treatment of blowdown water by nano-filtration resulted in up to 97% of TDS removal. A flux recovery of 97% was obtained by simple hydraulic washing which necessitated the need of the pre-treatment with ion exchanger regeneration wastewater and soda ash solutions. These two processes combined can help attain Zero liquid discharge and calcium sulfate extracted from the sludge has direct applications in cement manufacturing, food factories, and water treatment, whereas the nano filter soda ash can be reused for generation of ion exchanger. [Semblante et al. 2018](#) tried to find better management options for brine and suggested pre-treatment by coagulation and flocculation process followed by thermal treatment to obtain a Zero liquid discharge.

A summary of the various chemical coagulants, the industrial wastewaters studied, and key results is presented in [Table 2](#). From this listing, it is clear that alum, poly aluminium chloride (PAC) and ferric chloride hexahydrate are the most efficient chemical coagulants and have been used to treat wastewaters generated by different industries such as tannery, paper and pulp, textile, beverage, etc.

#### *Reducing the residual aluminium in the supernatant*

The residual aluminium varies based on the type of the coagulant used for the treatment. There are a number of health effects associated with high levels of residual aluminium in the water (discussed in more detail in the following section) and hence it needs

Table 2: A summary of the various chemical coagulants and their effectiveness for the treatment of industrial wastewaters

S.N.	Coagulant	Industry	Form of coagulant	Optimum dosage	Parameters studied	Removal efficiency	Reference
1	Alum and cationic polymer (C-492)	Tannery wastewater	Dry	100 and 5 mg/L	Turbidity	97%	Haydar and Aziz, 2009
					TSS	93.50%	
					Total COD	36.20%	
					Chromium	98.40%	
2	Alum and cationic polymer (Cytec)	Textile dyes wastewater	Dry	200 and 1.0 mg/L	Color	94.00%	El-Gohary and Tawfik, 2009
					COD	44.00%	
					Turbidity	99.70%	
					TSS	96.30%	
3	Alum and Anionic polymer	Tannery wastewater	Dry	160 and 5 mg/L	Total COD	48.30%	Haydar and Aziz, 2009
					Chromium	99.70%	
					Turbidity	99.70%	
					TSS	96.40%	
4	Alum+ organopol	Paper and pulp mill wastewater	Dry	1000 mg/L	Total COD	53.30%	Haydar and Aziz, 2009
					Chromium	98.90%	
					COD	58.00%	
					Color	79.00%	
5	Alum	Tannery wastewater	Dry	240 mg/L	TSS	96.40%	Haydar and Aziz, 2009
					Total COD	53.30%	
					Chromium	98.90%	
					COD	58.00%	
6	Alum	Leachate treatment	Dry	11 g/L	Color	79.00%	Al-Hamadani et al., 2011
					TSS	78.00%	
					Turbidity	99.80%	
					TSS	99.40%	
7	Alum	Paper and pulp mill wastewater	Dry	1000 mg/L	COD	91%	Ahmad et al., 2008
					COD	52%	
					Turbidity	92%	
					TSS	82%	
8	Alum	Oil refinery wastewater	Powder	40 mg/L	Turbidity	96%	Dehghani and Alizadeh, 2016.
					TSS	82%	
					Turbidity	96%	
					COD	71%	
9	PAC	Wastewater	powder	30 mg/L	Turbidity	95%	Awad et al., 2013
					COD	63%	
					Turbidity	96%	
					COD	61%	
10	PAC and Shendi	Wastewater	powder	30 mg/L and 400mg/L	Turbidity	96%	Awad et al., 2013
					COD	63%	
					Turbidity	96%	
					COD	61%	
11	PAC and Singa	Wastewater	powder	30 mg/L and 400 mg/L	Turbidity	96%	Awad et al., 2013
					COD	61%	
					Turbidity	96%	
					COD	61%	
12	PAFS	sewage wastewater	aqueous solution	45 mg/L	COD	83%	Zhu et al., 2012
					Turbidity	98%	
					Turbidity	82%	
					Cr	93%	
13	Ferric chloride	Coal washery wastewater	solution	400 ppm	Turbidity	98%	Babarao and Verma, 2015
					Turbidity	82%	
					Cr	93%	
					TDS	71%	
14	Ferric chloride	Tannery wastewater	powder	150 mg/L	TSS	95%	Ahmed et al., 2016
					Turbidity	72%	
					TSS	95%	
					BOD	81%	

*Natural coagulant for industrial wastewater*

Table 2: A summary of the various chemical coagulants and their effectiveness for the treatment of industrial wastewaters

S.N.	Coagulant	Industry	Form of coagulant	Optimum dosage	Parameters studied	Removal efficiency	Reference
15	Ferric Chloride and Aluminium Sulfate	Hospital wastewater	Dry		COD	85%	<a href="#">Suarez et al., 2009</a>
					TSS	92%	
					Diclofenac (DCF)	46%	
					Naproxen (NPX)	42%	
16	Ferric chloride and lime	Coal washery wastewater	Solution	400 ppm	Ibuprofen (IBP)	23%	<a href="#">Babarao and Verma, 2015</a>
17	Iron III chloride Hexahydrate	Beverage industry wastewater	Dry	300 mg/L	Turbidity	100%	<a href="#">Amuda and Amoo, 2007</a>
					COD	73%	
					TP	95%	
18	Iron III chloride Hexahydrate	Textile wastewater	Stock solution	0.37 mol/L	TSS	97%	<a href="#">Wang et al., 2011</a>
					Turbidity	94%	
19	Iron III chloride Hexahydrate	Cosmetic industrial wastewater	Powder	500 mg/L	COD	50%	<a href="#">Carpinteyro et al., 2012</a>
					Turbidity	70%	
					O&G	87%	
20	Iron III chloride Hexahydrate and polyelectrolyte	Beverage industry treatment	Dry	100 and 25 mg/L	COD	48%	<a href="#">Amuda and Amoo, 2007</a>
					COD	91%	
					TP	99%	
21	PAC	Paper and pulp mill wastewater	Dry	500 mg/L	TSS	97%	<a href="#">Ahmad et al., 2008</a>
					Turbidity	99.90%	
					TSS	99.50%	
22	PAC	Leachate wastewater	Dry	7.5 g/L	COD	91.30%	<a href="#">Al-Hamadani et al., 2011</a>
					COD	55.00%	
					Color	80.00%	
24	Lime	Textile dyes wastewater	Dry	600 mg/L	TSS	95.00%	<a href="#">El-Gohary and Tawfik, 2009</a>
					Color	100%	
25	Magnesium chloride	Textile dyes wastewater	Dry	120 mg/L	COD	50%	<a href="#">El-Gohary and Tawfik, 2009</a>
					Color	100%	
26	Lime+ Magnesium chloride	Textile dyes wastewater	Dry		COD	40%	<a href="#">El-Gohary and Tawfik, 2009</a>
					Color	97-100%	
					COD	40-50%	
28	Ferrous sulfate with lime	Coal washery wastewater	solution	400 ppm	Turbidity	56.67%	<a href="#">Babarao and Verma, 2015</a>
29	Ferric sulfate trihydrate	Beverage industry	Dry	500mg/L	Turbidity	81.81%	<a href="#">Amuda and Amoo, 2006</a>
					COD	78%	
					TSS	74%	
30	Ferric sulfate trihydrate and polyelectrolyte	Beverage industry	Dry	500 mg/L and 25 mg/L	TP	75%	<a href="#">Amuda and Amoo, 2006</a>
					COD	93%	
					TSS	94%	
					TP	96%	

to be taken care of. [Zouboulis and Tzoupanos 2009](#) realized the issues that may arise due to the high residual aluminum content in the treated water by the use of aluminum-based coagulant and therefore

suggested the use of silica-based coagulants instead. Silica-based coagulants have low residual aluminium content in the treated water ([Zouboulis and Tzoupanos, 2009](#)). Presence of residual aluminium



and its concentration in the supernatant is a function of the coagulant type. The use of the PASiC was suggested due to smaller residual aluminium content in the treated water. It was found that PASiC is resilient to pH variation as compared to the other coagulants (Zouboulis and Tzoupanos, 2009). A comparative study to see the flocs formation efficiency of silica-based and aluminium based coagulants showed that though silica-based coagulants form bigger flocs, if the results are compared to Poly aluminium chloride (PAC) and alum, there was not a very substantial difference in phosphate removal efficiencies of both types of coagulants (Zouboulis and Tzoupanos, 2009). Zouboulis and Traskas 2005 conducted a comparative study to understand the effect of basicity on the efficiency of poly aluminium chloride. The study reported that the PAC with intermediate basicity produced better coagulation results as compared to PAC with higher basicity and alum. The PAC has lesser residual aluminium concentration than alum. Poly aluminium chloride had better efficiency in removal of suspended particulates. Furthermore, the study concluded that the residual aluminium content in the treated water is also affected by the pH (Zouboulis and Traskas, 2005). Tolkou and Zouboulis 2014 tried different coagulants for water and wastewater treatment and suggest that PsiFAC (Poly-aluminium-ferric-silicate-chloride) is the most effective coagulant for water and wastewater treatment and has minimum residual aluminium concentration (140 µg Al/L). The coagulant could remove up to 99% turbidity using the coagulant dose of 100 mg/L.

#### *Disadvantages of chemical coagulants: Health and environmental impacts*

Chemical coagulation is carried out using synthetic chemical coagulants. This practice has the potential of leaving a bad impact on the environment and the public health. Chemical coagulants are non-biodegradable and remain in the water even after the coagulation process is completed. There is a possibility that the treated supernatant contains the traces of metals present in the chemical coagulants due to the presence of residual aluminium in the supernatant (Zouboulis and Tzoupanos, 2009). Use of chemical coagulants can cause neurological diseases like Alzheimer's disease (WHO Guidelines, 2010; Niquette et al., 2004), Encephalopathy (Srinivasan et al., 1999) leading to dementia, Down's syndrome

and staining of Hippocampal neurons (Walton, 2006). The treated water containing the high concentration of residual aluminium may either get seeped into the groundwater or may have a surface runoff (WHO Guidelines, 2010). Chemical coagulants are non-biodegradable (Muralimohan et al., 2014) and remain in the water because of which, synthetic chemicals are said to generate a sludge that may not be amenable to safe environmental disposal and may result in contamination of our water and land. Parmar et al. 2012 in his study suggested that supernatant obtained from dairy industry using alum and ferrous sulfate was not suitable for discharge into the municipal drains due to the high values of various parameters like BOD and COD in the supernatant. Major issues with the use of aluminium-based coagulants are that they lead to increased concentration of residual aluminium in the supernatant (WHO Guidelines, 2010). This aluminium may either seep into the groundwater or may have a surface runoff (WHO Guidelines, 2010). Conventional, water and wastewater treatment plants do not remove aluminium and water with elevated aluminium content (Walton, 2006) is supplied to the end consumers (Srinivasan et al., 1999). If aluminium entered the public distribution system, it could lead to precipitation of hydrous aluminium in the water, which is to be supplied to the consumers (Srinivasan et al., 1999). Residual aluminium in the treated water is found to negatively impact the health of consumers. Exposure to aluminium is linked to Alzheimer's disease (WHO Guidelines, 2010; Niquette et al., 2004) as it stains the Hippocampal neurons (Watson, 2006). Aluminium is neurotoxic and is responsible for disorders like Parkinson's disease and Down's syndrome (Watson, 2006). Its accumulation in the bloodstream for the long term can result in severe Encephalopathy (Srinivasan et al., 1999), and consequently contributing to dementia (Oaks et al., 2004).

#### *Natural coagulants for wastewater treatment*

The potential environmental and human health hazards associated with the use of chemical coagulants has necessitated the need for the use of natural coagulants for industrial wastewater treatment. Natural coagulants are gaining a lot of attention these days as they are an effective alternative to the chemical coagulants (Yin, 2010). Natural coagulants are a sustainable approach

(Folkard *et al.*, 1995). Plant-based materials have been investigated for treating industrial effluents from different industries. Plant-based substances like *Moringa Oleifera* (Chonde and Raut, 2017; Sivakumar, 2013), chitosan and chitin, *Abelmoschus esculentus*, *Opuntia ficus-indica*, *Synchorous Potatorum*, *Prosopis laevigata* Seed Gum, *Hibiscus rosa-sinensis* (Awang and Aziz, 2012), *Acacia mearnsii* (Beltrán-Heredia *et al.*, 2011), etc. can be used as coagulants. Generally, the natural coagulants are directly used as a powder or a stock solution. In some cases, the deoiled powder is also used, after extraction of oil from the coagulant. The plant-based products (such as seeds, etc.) are first extracted from the plant, cleaned to remove any impurities that may interfere with coagulation, and then dried. The powder is then formed (with or without the oil extraction, as per need) by grinding (Miller *et al.*, 2008). This powder may be directly used, or a stock solution can be prepared from it. In some cases, proteins may be extracted from the specific plant parts and used as a coagulant. This may require extensive extraction and purification steps (Kansal and Kumari, 2014). Kumar *et al.* 2017 and Tariq *et al.*, 2015 reported that the microbial polysaccharides, starches, gelatin galactomannans, cellulose derivatives, chitosan, glues, and alginate can be used for wastewater treatment. Natural coagulants can also be used with synthetic coagulants to assist the coagulation process for wastewater treatment. Mohamed *et al.* 2014 used *Moringa oleifera* and *Strychnos potatorum* seeds as a natural coagulant for car wash wastewater. The turbidity and COD reduction efficiency of coagulants was studied. Using *Moringa oleifera*, 94% turbidity, 60% COD, 81% phosphorus removal were obtained, whereas using *Strychnos potatorum*, 97% turbidity, 54% COD, and 82% phosphorus removal were obtained. These results were compared with synthetic coagulants, and natural coagulants were suggested for coagulation process as they provide better treatment, are cost-effective and are safe for environment. Sundaresan and Anu 2016 investigated the feasibility of natural coagulant for the treatment of dairy wastewater. *Artocarpus heterophyllus* (jack fruit) and *Phaseolus vulgaris* (Common Beans) were used to remove turbidity from wastewater. *Artocarpus heterophyllus* (jack fruit) seeds attained 94% turbidity removal efficiency and *Phaseolus vulgaris* (common beans) seeds gave up to 99% of

turbidity removal (Sundaresan and Anu, 2016). Jeon *et al.* (2009) used the grape seed for removal of cationic dyes and confirmed that grape seed-derived coagulants induced decolorization of cationic dyes. Nharingo and Moyo 2016 used derivatives of *Opuntia ficus-indica* for treatment of the wastewater. The study demonstrated coagulant as a very promising alternative to remove dyes, metallic species, heavy metals, turbidity and COD. Gandhi *et al.* 2013 used plant-based polyelectrolytes as coagulants (derived from the fruits of *Opuntia ficus indica*, fruits of *Jatropha gossypifolia* and *Borassus flabellifer*) to remove chromium. It was concluded that there is a significant improvement in the physicochemical characteristics of wastewater and heavy metal chromium was successfully controlled by natural coagulants. These polyelectrolytes can be effectively used for removal of chromium as they destabilize and reduced the repulsive forces between the molecules. Prodanovic *et al.* 2015 used the common bean for distillery wastewater treatment and it was concluded that pH value of stillage influenced activity of the natural coagulant. The optimum pH for the treatment was reported to be 8.5. Saharudin and Nithyanandam 2014 assessed possibility of using natural coagulants as an alternative to the aluminium sulphate and to optimize the parameters related to the working for treatment of synthetic wastewater samples. It was concluded that roselle seeds are a viable commercial alternative to aluminium sulfate. The highest removal efficiency with roselle seeds powder was within the range 81.2% to 93.13% for synthetic wastewater at a pH 4.0. However, the highest removal efficiency for industrial wastewater was within 76.8% to 87.18% at a pH value of 10.0. Kani *et al.* 2016 used banana pith juice for textile wastewater treatment. At pH 4, 97.5% turbidity and 50.1% total solids were removed from the wastewater. There was a significant improvement in the electrical conductivity. The results confirmed that banana stem juice has an enormous potential for turbidity removal from the textile wastewater. Wu *et al.* 2005 used white-rot fungi for degradation of lignin from the paper mill wastewater. The lignin and COD removal efficiencies were reported to be 71% and 48%, respectively. Optimum pH range for lignin removal by *Chrysosporium*, *Pleurotus ostreatus* and s22 was observed to be 9.0-11.0. Gaurang and Punita 2012 used fruit mucilages of *Coccinia indica* and *Abelmoschus esculentus*, and the dry seed powder

of *Moringa* for treatment of dairy wastewater. Using *Abelmoschus esculentus*, 60.33% of turbidity could be removed whereas *C. indica* gave highest turbidity removal of 77.67%. [Nicholas et al. 2018](#) used *Hibiscus sabdariffa* seeds for removing dyes from the synthetic wastewater. The study involved preparation of models and ANOVA (analysis of variance). Response surface methodology showed 98.68% dyes removal from the wastewater. Oyster mushroom has been tested for the reduction of parameters like turbidity and TSS (total suspended solids) from domestic wastewater. The basic coagulation mechanism of natural coagulants differs from that of the synthetic coagulants due to their inability in forming the hydroxide precipitates in the water. Synthetic coagulants generally work on the mechanism of charge destabilization and sweep flocculation. In comparison, the polymeric nature of plant-based coagulants and presence of functional groups suggest that polymer bridging, and charge neutralization are the dominant mechanism for the natural coagulants ([Ang and Mohammad, 2020](#); [Saleem and Bachmann, 2019](#)). For some of the natural coagulants, adsorption in conjunction with either charge neutralization or polymer bridging has been cited as the dominant coagulation mechanism ([Kansal and Kumari, 2014](#); [Daverey et al., 2019](#)).

#### Commonly studied natural coagulants

A large number of natural coagulants, mostly plant-based, have been studied for their potential in treating industrial effluents. A brief description of some of the more commonly studied coagulants is provided here.

*Moringa Oleifera* is a small or medium-sized tree grown in northwest India, several parts of Asia, Africa, and South America. It is a multipurpose plant ([Muthuraman and Sasikala., 2014](#)) and is cultivated in tropical, sub-tropical or semi-arid regions at an altitude between 0-2000 m. Cultivation of *Moringa* requires an annual rainfall between 250-3000 mm. It belongs to the *Moringaceae* family ([Okuda et al., 2001](#)). *Moringa* can grow on the low altitude tropical belt on the less humid soil. *Moringa Oleifera* is an organic polymer. *Moringa* grows rapidly from the seed or cutting and does well even in poor soils ([Adebayo et al., 2017](#)). *Moringa oleifera* is one of the most effective and certainly the most investigated natural coagulant for wastewater treatment ([Bhuptawat et al., 2007](#)). Typically, the grounded seed powder of

*Moringa oleifera* is used as a coagulant. [Sulaiman et al. 2017](#) studied the potential of *Moringa* and reported excellent results for wastewater treatment. The seeds of moringa do not further deteriorate the environment and are amenable to biodegradation and thus are environment friendly. [Hemapriya et al. 2015](#) used *Moringa* for treating the textile mill effluent. *Moringa oleifera* was reported to cause significant COD reduction for textile mill wastewater ([Dotto et al., 2019](#)). [Parmar et al. 2012](#) used grounded seed powder of *Moringa Oleifera* for dairy wastewater treatment. The *Moringa oleifera* seeds left the water clear and reduced the turbidity by almost 100 percent and 99.50-100 percent removal of fecal coliforms was observed. [Ashmawy et al. 2012](#) used *Moringa Oleifera* for treating the laundry wastewater and obtained up to 83.63% of turbidity removal.

*Abelmoschus Esculentus* belongs to *A. esculentus* species of plants and *Malvaceae* family ([Kumar et al., 2010](#)). It is commonly known as ladyfinger or Gumbo Okra. Okra is a vegetable crop cultivated in Asia and Africa ([Kumar et al., 2010](#)). [Ameena et al. 2010](#) studied the binding effects of the *Hibiscus rosasinensis* Linn and Okra (*Abelmoschus esculentus* Linn) mucilages, both of which were found to be amorphous (after drying). It was reported that instead of starch, lower concentrations levels of okra can be used. [Anastasakis et al. 2009](#) suggested that Okra has better efficiency than *Malva sylvestris* (mallow) in removing turbidity, even at much lower doses.

*Opuntia ficus indica*, a species of cactus, is grown in different parts of the world with different habitats with different climates. *Opuntia ficus* mucilage has enormous potential to treat industrial wastewater and is safe for the environment. It can serve as a potential replacement of aluminium and iron salts ([Bogacki et al., 2011](#)). Different species of cactus have been tried to ascertain the potential of cactus in coagulation and flocculation process. Cactus is used in different forms such as powder, mucilage for wastewater treatment. [De Souza et al. 2014](#) used *Opuntia ficus-indica* (OFI) for textile effluents treatment and reported 95% turbidity removal. The coagulant was stored for 4 days which did not affect the efficiency of the coagulation mechanism. [Rebah and Siddeeg 2017](#) reviewed the potential of cactus for wastewater treatment. Renewability, abundance, adaptability, and biodegradability of cactus plants make them a suitable alternative to the chemical

coagulants. It has a potential to be used as coagulant/flocculant, as biosorbent and as packed material for bio-filter. The presence of cactus enzymatic system can transform toxic textile dyes when used for textile wastewater treatment. However, limitation of using cactus is that there is variation in the efficiency on basis of wastewater characteristics.

*Chitosan* is a cellulose-like biopolymer and is widely distributed in nature. It is yellow colored in powder form and completely biodegradable (Renault *et al.*, 2009). The major sources of chitin and chitosan in the environment are shells of crabs, shrimp (Feria-Diaz *et al.*, 2018), lobsters (Kumar *et al.*, 2010), fungi, mollusks, diatoms, and marine and freshwater sponges (Malerba and Cerana, 2018). Chitosan can be used for the process of coagulation (Rao, 2015). Renault *et al.* 2009 reviewed the potential of chitosan for coagulation, and flocculation process and confirmed that chitosan has potential for the wastewater treatment. Kumar *et al.* 2000 suggested the different applications of chitin and chitosan for use in a variety of industries due to the minimum toxicity, which makes them safe for discharge into the environment. Wan Ngah *et al.* 2011 suggested using chitosan and its derivatives for dyes and heavy metals removal from the wastewater. Anteneh and Sahu 2014 used chitosan for treating soap and detergent wastewater. Chitosan reduced 83% of COD as well as 90 % of color from wastewater. The authors concluded that easy availability and unique properties of chitosan as a coagulant make it a promising alternative to chemical coagulants. Sarkar *et al.* 2006 investigated the possibility of treating dairy wastewater using chitosan. They found that chitosan worked efficiently even at very low dosages (10 mg/L). The study suggested that PAC treatment of supernatant following chitosan treatment completely removed color and odor from wastewater before membrane processing (Sarkar *et al.*, 2006). Savant and Torres 2000 used chitosan complexes for treating cheddar cheese whey. Turbidity reductions of 40-43% and 65-72% were obtained and up to 70% protein recovery from cheese whey was obtained. Feria-Diaz *et al.* 2018 used chitosan for turbidity removal from slaughterhouse wastewater. The study claimed that as much as 90% of turbidity could be removed from the wastewater using chitosan as a coagulant.

*Tannins* are aromatic compounds and have polycyclic structures (Thakur and Choubey,

2014). Beltran-Heredia 2011 showed that tannin-derived coagulant is a novel product for dye (azo, anthraquinonic, indigoid, and triphenylmethane ones) removal.

*Cicer arietinum* is grown in arid and semi-arid zones (Hiremath *et al.*, 2014). It is grown in areas experiencing good rainfall where the soil has good moisture content. Jaseela and Chadaga 2015 used *cicer arietinum* for COD and turbidity removal from the dairy industry wastewater. Using the coagulant in the powder form, 86.29% of turbidity removal was achieved.

*Ocimum basilicum*, commonly known as basil, is traditionally used in medicines and studies have indicated that it can be used as a coagulant. With 1.6 mg/L of coagulant in 50 mg/L solution of pigment, up to 61.6% of BOD, and 68.5% of color removal was attained from textile wastewater. With a dosage of 9.6 mg/L *Ocimum basilicum* solution, 50.9% color, and 56.6% COD removal efficiencies can be achieved (Shamsnejati *et al.*, 2015). Mosaddeghi *et al.* 2020 reported 85% color removal and 82% total suspended solids removal with *O. basilicum* for paper recycling wastewater.

*Cassia seed gums*-based coagulants like *Cassia javahikai*, *Cassia angustifolia*, *Cassia fistula*, *Cassia obtusifolia*, and *Cassia tora* can be used for textile wastewater treatment (Karoliny *et al.*, 2018). Ngan *et al.* 2017 used *Cassia fistula* seed gum with PAC for the treatment of fish processing wastewater.

Table 3 shows the details of the natural coagulants used for the effluents obtained from various industries. The details of the effective doses and treatment efficiencies are also listed. From this listing, it can be seen that *Moringa oleifera* and chitosan are the most efficient natural coagulants and have shown potency for wastewaters generated from different industries.

#### Advantages of natural coagulants

Wastewater treatment using natural coagulants is an eco-friendly option. Natural coagulants are non-toxic, biodegradable, and environment friendly (Verma *et al.*, 2012; Muralimohan *et al.*, 2014). Unlike synthetic coagulants, treated water contains no residual aluminium. Prodanović *et al.* 2013 used common bean extract for the treatment of distillery wastewater treatment. The study claimed that anaerobic sludge contained no aluminium salts. This

ensures that aluminium-related health issues (such as Parkinson's disease, Alzheimer's disease, and other neurological diseases), as stated earlier, would be absent when natural coagulants are used for the

coagulation-flocculation of the wastewater. The supernatant and sludge produced on treatment using natural coagulants can be used for other purposes, unlike those obtained from chemical coagulation.

Table 3: Summary of the key literature findings of industrial wastewater treatment by using natural coagulants

S.N.	Coagulant	Wastewater source	Original form of coagulant prepared	Form of coagulant used	Optimum dosage	Parameters	Removal efficiency	Reference
1	<i>Moringa oleifera</i>	Palm oil mill effluent	Deoiled powder		6000 mg/L	Suspended Solids	95%	Thirugnanasambandham <i>et al.</i> , 2014
2	<i>Moringa</i> (425 µm)	Dairy wastewater	Deoiled	direct powder	300 mg/L	Turbidity	90%	Pallavi and Mahesh, 2013
3	<i>Moringa</i> (212 µm)	Dairy wastewater	Deoiled	direct powder	500 mg/L	Turbidity	93.50%	Pallavi and Mahesh, 2013
4	<i>Moringa</i> (150 µm)	Dairy wastewater	Deoiled	direct powder	500 mg/L	Turbidity	96.80%	Pallavi and Mahesh, 2013
5	<i>Moringa oleifera</i>	Secondary oxidation pond effluent	Powder	stock solution	100mg/L	COD Alkalinity BOD(5-day) Turbidity	60% 99% 75% 99%	Katayon <i>et al.</i> , 2007
6	<i>Moringa oleifera</i>	Dairy wastewater	Powder	solution	1g/L	COLOUR COD color	95% 55% 71%	Vieira <i>et al.</i> , 2010
7	<i>Moringa oleifera</i>	Domestic wastewater	Powder	solution	250 mg/L	turbidity total coliform <i>Escherichia coli</i>	85% 96% 83%	Silva <i>et al.</i> , 2013
8	<i>Moringa oleifera</i>	Dairy waste water	Powder	powder	0.2 g/L	turbidity COD	61.60% 65.00%	Patil and Hugar, 2015
9	<i>Moringa oleifera</i>	Tannery wastewater	Powder	powder	0.6 g/L	turbidity COD	82% 83%	Kazi <i>et al.</i> , 2013
10	<i>Moringa oleifera</i>	Dairy waste water	Powder	suspension	60 mg/L	BOD (5-day) COD turbidity	55% 60% 78%	Bhutada <i>et al.</i> , 2006
11	<i>Moringa oleifera</i>	Dairy wastewater	Powder	powder	0.3 mg/L	BOD(3-day) COD TDS TSS turbidity	80% 86% 9% 95% 85%-94%	Neethu <i>et al.</i> , 2017
12	<i>Moringa oleifera</i>	Leachate treatment	Deoiled seed powder	stock solution	-	Cu and Cd Pb	98% 78%	Shan <i>et al.</i> , 2017
13	<i>Moringa oleifera</i>	Batik effluent	Powder	powder	4000 mg/L 5000 mg/L	Turbidity TSS	96% 88%	Effendi <i>et al.</i> , 2015
14	<i>Moringa oleifera</i>	Oil refinery wastewater	Solution	powder	70 mg/L	TSS Turbidity	62% 64%	Dehghani and Alizadeh, 2016
15	<i>Moringa oleifera</i>	Textile effluent	Powder	stock solution	40 mL	TDS BOD COD	71% 72% 79%	Muralimohan <i>et al.</i> , 2014
16	<i>Moringa oleifera</i> +NALCO 7751	Palm oil mill effluent	Deoiled powder	-	4000+7000 mg/L	Suspended solids COD	99.30% 52.50%	Bhatia <i>et al.</i> , 2007
17	<i>Moringa Oleifera</i> and fuller earth	Dairy wastewater	Powder	suspension solution	60 mg/L+ 15mg/L	BOD(5-day) COD	60.00% 66.00%	Bhutada <i>et al.</i> , 2006

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S.N.	Coagulant	Wastewater source	Original form of coagulant prepared	Form of coagulant used	Optimum dosage	Parameters	Removal efficiency	Reference
18	<i>Moringa</i> and Alum	Oil refinery wastewater	Powder	solution	70 and 80 mg/L	COD turbidity TSS	50.41% 86.14% 81.52%	Dehghani and Alizadeh, 2016
20	<i>Moringa</i> and Alum	Textile industry effluent	Powder	stock solution	50:50:00	turbidity BOD COD	61.60% 80.67% 66.73%	Muralimohan <i>et al.</i> , 2014
21	Chitosan	Olive mill wastewater	Powder	solution 5g/L	400 mg/L	TSS	81%	Rizzo <i>et al.</i> , 2008
22	Chitosan	Paper and pulp wastewater	Powder	-	1.8 g/L	turbidity BOD COD	84% 90% 93%	Thirugnanasambandham <i>et al.</i> , 2014
23	Chitosan	Brewery wastewater	-	stock solution	120 mg/L	Turbidity COD	95% 50%	Maya <i>et al.</i> , 2014
24	Chitosan	Textile wastewater	Powder	stock solution	30 mg/L	COD Turbidity	73% 95%	Ariffin <i>et al.</i> , 2009
25	Chitosan	Textile wastewater	Powder	powder	30 g/L 25 g/L and 30 g/L	COD Color	65% 43%	Patel and Vashi, 2013
26	<i>Okra mucilage</i>	Textile wastewater	Mucilage	-	3.20 mg/L	Color Turbidity COD	93.57% 97.24% 85.69%	Wang <i>et al.</i> , 2011
27	<i>Okra</i>	Coal washery wastewater treatment	Powder	stock solution	400 ppm	Turbidity	61.03%	Babarao and Verma, 2015
28	<i>Okra</i> + Iron (III) chloride Hexahydrate	Textile wastewater	-	stock solution	3.20 mg/L + 88 mg/L	turbidity COD color	97.24% 85.69% 93.57%	Wang <i>et al.</i> , 2011
29	<i>M. sylvestris</i> (mallow) + aluminium Sulfate	Synthetic wastewater Biologically treated effluent	-	stock solution	12 mg/L 62.5 mg/L	turbidity turbidity	97.40% 66%	Anastasakis <i>et al.</i> , 2009
30	<i>Okra mucilage</i> + aluminium sulphate	Synthetic wastewater Biologically treated effluent	-	stock solution	5 mg/L 2.5 mg/L	turbidity turbidity	97.70% 74%	
31	<i>Psyllium husk</i> + PAC	Leachate treatment	Powder	-	0.4 g/L + 7.2 g/L	COD Color TSS	64% 90% 96%	Al-Hamadani <i>et al.</i> , 2011
32	Guar	Cosmetic wastewater	Powder	-	300mg/L	turbidity	67.82%	Carpinteyro <i>et al.</i> , 2012
33	Locust bean gum	Cosmetic wastewater	Powder	-	300mg/L	turbidity	67.82%	Carpinteyro <i>et al.</i> , 2012
34	<i>Opuntia mucilage</i>	Cosmetic wastewater	Powder	-	150mg/L	turbidity	49.56%	Carpinteyro <i>et al.</i> , 2012
35	<i>Ocimum basilicum</i>	Textile wastewater	Powder	stock solution	1.6 mg/L	color COD	68.50% 61.60%	Shamsnejati <i>et al.</i> , 2015

Table 3: Summary of the key literature findings of industrial wastewater treatment by using natural coagulants

S.N.	Coagulant	Wastewater source	Original form of coagulant prepared	Form of coagulant used	Optimum dosage	Parameters	Removal efficiency	Reference
36	<i>Plantago major</i> L.	Dye wastewater	Powder	stock solution	297.6 mg/L	color COD	92.40% 81.60%	<a href="#">Chaibakhsh et al., 2014</a>
37	<i>Plantago psyllium</i>	Textile wastewater	Mucilage solution	-	10 mg/L	Golden dye reactive black	71.40% 35%	<a href="#">Mishra and Bajpai, 2014</a>
38	<i>Chyote</i> + Iron trichloride	Textile wastewater	Powder	-	15 mg/L + 47.40 mg/L	turbidity COD	97.95% 83.84%	<a href="#">Almeida et al., 2017</a>
39	<i>Hibiscus Rosa sinensis</i> + Alum	Leachate wastewater	Powder	-	500 mg/L + 4000 mg/L	Suspended solids turbidity Iron (III)	72% 60% 100%	<a href="#">Awang and Aziz, 2012</a>
40	<i>Trigonella foenum-graecus</i>	Dairy wastewater	Powder	powder	0.1 g/L	turbidity COD	58% 63%	<a href="#">Patil and Hugar, 2015</a>
41	<i>Dolicus lablab</i>	Dairy wastewater	Powder	powder	0.2 g/L	turbidity COD	71.74% 75%	<a href="#">Patil and Hugar, 2015</a>
42	<i>Cicer arietinum</i>	Dairy wastewater	Powder	powder	0.1 g/L	turbidity COD	78.33% 83%	<a href="#">Patil and Hugar, 2015</a>
43	<i>Cicer arietinum</i>	Tannery wastewater	-	powder	0.2 g/L	turbidity COD	81% 90%	<a href="#">Kazi et al., 2013</a>
44	<i>Cicer arietinum</i>	Slaughter house wastewater	Powder	powder	0.5 g	Turbidity	68%	<a href="#">Sheikh et al., 2016</a>
					2 g	TDS	82%	
					2 g	BOD	83%	
					1.5 g	COD	84%	
45	Roselle seeds	Glove manufacturing	Powder	powder	60 mg/L	Turbidity	87.18%	<a href="#">Saharudin and Nithyanandam, 2014</a>
46	Cactus	Tannery wastewater	Powder	powder	0.4 g/L	Turbidity	78.54%	<a href="#">Kazi et al., 2013</a>
						COD	75%	
47	Tanfloc SG	Dairy wastewater	Powder	stock solution	20 mg / L	turbidity	71.20%	<a href="#">Wolf et al., 2015</a>
						COD	77.28%	
						Total solids	0.48%	
						Total coliform	100.00%	
48	Tanfloc SH	Dairy wastewater	Powder	stock solution	20 mg/L	Turbidity	65.60%	<a href="#">Ayangunna et al., 2016</a>
						COD	44.14%	
49	Tamarind seed powder	Detergent wastewater	Powder	stock solution	400 mg/L	Total solids	0.43%	<a href="#">Ayangunna et al., 2016</a>
						Total coliform	69.00%	
50	Tamarind Indica	Textile effluent	Powder	stock solution	60 mL	TDS	41.50%	<a href="#">Babarao and Verma, 2015</a>
51	Bitter gourd seed	Coal washery wastewater treatment	Powder	solution	400 ppm	Turbidity	61.03%	<a href="#">Babarao and Verma, 2015</a>

Table 3: Summary of the key literature findings of industrial wastewater treatment by using natural coagulants

S.N.	Coagulant	Wastewater source	Original form of coagulant prepared	Form of coagulant used	Optimum dosage	Parameters	Removal efficiency	Reference
52	Cotton seed oil cake	Sago effluent	Oil	oil	30 mL	TSS	66.27%	<a href="#">Narmatha et al., 2017.</a>
53	<i>S. Potatorum</i>	Textile effluent	Powder	stock solution	60 mL	TDS BOD COD Turbidity	52.30% 65.23% 72.71% 75.20%	<a href="#">Dehghani and Alizadeh, 2016</a>
54	<i>S. Potatorum</i> and Alum	Textile wastewater	Powder	Stock solution	40: 60 40: 60	TDS TSS BOD COD	67.91% 79.23% 69.43% 74.93%	<a href="#">Muralimohan et al., 2015</a>
55	Castor oil cake	Sago effluent	Oil	oil	40 mL	TSS	66.67%	<a href="#">Sheikh et al., 2016</a>
56	Surjana seed	Textile wastewater	Powder	powder	25 g/L 30 g/L	COD Color	74.11% 62.82%	<a href="#">Patel and Vashi, 2013</a>
57	Maize seed	Textile wastewater	Powder	powder	30 g/L	COD Color	68.82% 47.03%	<a href="#">Patel and Vashi, 2013</a>
58	Chayote and FeCl <sub>3</sub>	Textile wastewater	Powder		15 mg/L and 47 mg/L	Turbidity COD	97.95% 83.84%	<a href="#">Almeida et al., 2017</a>
59	Banana juice	Spent coolant wastewater		juice	90 mL	COD SS Turbidity	80.10% 88.60% 98.50%	<a href="#">Alwi et al., 2013</a>
60	<i>P. ovata</i>	Textile wastewater	De-oiled seed powder	solution	1.5 mg/L	COD	89.30%	<a href="#">Ramavandi and Farjadfard, 2016</a>

The sludge obtained after the treatment with natural coagulants is non-toxic and biodegradable and can be used for soil improvement. Paula *et al.* 2016 suggested the use of treated water for washing vehicles and flushing toilets. In the study, a portion of Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> was used with *Moringa Oleifera* seed extract to make the concrete wastewater eco-friendly. For the treatment, MO and Al<sub>2</sub>(SO<sub>4</sub>)<sub>3</sub> were used in the ratio of 20:80. 97.5% of turbidity was removed in 60 min using this arrangement. Feria-díaz *et al.* 2016 recommended the use of sludge produced, after treatment of water samples collected from Sinu River, Monteria, Cordoba using *Moringa oleifera*, for soil improvement. The study evaluated the agricultural application of the coagulation sludge from raw water treated with moringa. The properties of sludge were found to be suitable for application to soils (Feria-díaz *et al.*, 2016). Wei *et al.* 2018 suggested dewatering of the sludge and converting them into sludge cakes. Natural coagulants are cost-effective. Mohamed *et al.* 2014 after confirmatory experiments on commercial and natural coagulants for car wash wastewater treatment, recommended

the use of natural coagulants as they are eco-friendly and involve low cost as compared to the chemical coagulants. Vijayaraghavan *et al.* 2011 in their work stated that the natural coagulants form stronger flocs via bridging effect as compared to alum. Even at high shear levels, these linkages are resistant to breakage (Karoliny *et al.*, 2018). Therefore, it can be said that natural coagulants form better flocs. Cost comparison between the synthetic and natural coagulants is interesting due to the consideration of a large number of parameters. The natural coagulants are locally sourced, are available in large amounts, produce less sludge, generate biodegradable sludge (which can be reused) and may be based on waste materials (such as banana peels, etc.). On the other hand, in comparison to synthetic coagulants, the natural coagulants generally require a higher dosage, need processing and storage, may not be available year-round, and may have alternate economically attractive applications. Due to the absence of detailed studies and field-scale data on some of these parameters, it is difficult to do a detailed cost comparison between the natural and



synthetic coagulants (Ang and Mohammad, 2020). However, a number of recent studies have reported that natural coagulants are an economically attractive alternative to the synthetic coagulants (Behera and Balasubramanian, 2019; Daverey *et al.*, 2019; Meraz *et al.*, 2016).

#### Disadvantages of natural coagulants

A major drawback of using natural coagulants is their inability to manage large pH variation. Most of the coagulants work well under certain pH, beyond this range, their efficiency is severely affected. Vieira *et al.* 2010 used *Moringa Oleifera* for dairy wastewater treatment. The results demonstrated that the adsorption power of *Moringa Oleifera* seeds was best in the pH range of 5 - 8. Ferrari *et al.* 2016 used natural coagulants for the treatment of the food industry effluent in place of ferric chloride. Although the supernatant contained no traces of metal, Chitosan was found to be less efficient as its dilution occurs best at acidic pH. This may increase the cost of wastewater treatment using chitosan (Ferrari *et al.*, 2016). Natural coagulants contain organic matter and therefore the efficiency of these coagulants may decrease over time. Proper arrangements are required to store these coagulants in stock. This will increase the overall cost of the treatment. Another drawback of using natural coagulants for wastewater treatment is the requirement of high dosages as compared to the synthetic coagulants (Aliyu *et al.*, 2015). Aliyu *et al.*, (2015) used moringa and *Abelmoschus esculentus* for the treatment of the water from River Yamuna, India. Results showed that for treating the water from the same source, 150 mg/L to 200 mg/L of moringa and *Abelmoschus esculentus* were required, whereas better results were obtained using alum at a much lower dose of 30 mg/L under the same experimental conditions. In a comparative study between alum and natural coagulant *Moringa* for treatment of dairy industry wastewater, it was found that alum had better efficiency as compared to *Moringa oleifera* (Bangar *et al.*, 2017).

#### Issues in the large-scale implementation of natural coagulants

Natural coagulants are an economical and environment-friendly alternative to synthetic chemicals for wastewater treatment. Despite the

enormous potential, the treatment using natural coagulants faces challenges, and this has limited the application of natural coagulants to the laboratory-scale studies so far. Some of the issues preventing the large-scale application of natural coagulants for wastewater treatment are:

#### 1. Competitiveness

Many natural coagulants have varied applications and their use for wastewater treatment will affect the availability of these coagulants for other purposes. *Moringa oleifera* is used for the treatment of various diseases including asthma, syphilis, asthma, etc. (Somchit *et al.*, 2012). The use of *okra* for wastewater treatment also has various implications because of the multipurpose uses of the *Okra* plant. For instance, *okra* is consumed as a vegetable in Western Africa and Southeast Asia (Lamont 1999). Dried *okra* stems are used as a fuel as it burns considerably and produces considerable heat (Lamont 1999). *Okra* mucilage is used for manufacturing paper in Malaysia (Lamont 1999). Kumar Dutta *et al.* 2004 investigated the chemistry, properties, and applications of chitin and chitosan. Chitin and chitosan have also been used in various other fields like agriculture, cosmetics, photography, chromatographic separations, solid-state batteries, biomedical applications, burn treatment, wound healing/ wound dressing, artificial skin, Ophthalmology, drug delivery system, and in LEDs (light-emitting devices) applications (Kumar Dutta *et al.*, 2004). To ensure the large-scale application of natural coagulant, a non-competitive coagulant needs to be found. Besides, plant-based materials that are otherwise considered waste may also be investigated for their coagulation potential. *Benincasa hispida*, commonly known as ash gourd, is a commonly used vegetable and source of Petha candy (a common sweet popular in Indian sub-continent). Its seed is usually discarded. The seed powder of *Benincasa Hispida* has recently been studied for its coagulation potential for the treatment of river water (Saini, 2017).

#### 2. The absence of source for mass availability

Not all the natural coagulants discussed in this work are available universally and it raises concerns over the sustained mass-scale supply that would be needed for commercial applications. Due to the non-uniform distribution of plants, the same

natural coagulant may be costlier as compared to the synthetic coagulants in some parts of the world (Kansal and Kumari, 2014). Chemical coagulants, on the other hand, can be produced in industries practically anywhere and supplied as needed. A potential solution is the application of locally sourced natural coagulants.

### 3. Potency losses

Natural coagulants contain organic matter and may decay over time. This decay may affect the efficiency of coagulants in treating wastewater. Studies on potency losses are non-existent and are required to determine the possibility of potency losses of natural coagulants. De Souza *et al.* 2014 studied the possibility of potency losses with the use of cactus. They studied the effects of the storage time and temperature on the coagulation efficiency of *Opuntia ficus-indica*. The study indicated that there was no effect on the efficiency for up to 4 days (De Souza *et al.*, 2014). However, no other research is available that discussed potency beyond 4 days or ways to improve the shelf life of natural coagulants. More research is needed in determining and enhancing the shelf life of natural coagulants.

### 4. High quantity requirement

For treating the same quantity of the wastewater with the same values of the various parameters, the optimum dosage is very high in the case of natural coagulants as compared to the chemical coagulants. This implies that the wastewater can be treated with a very small quantity of chemical coagulants as compared to the natural coagulants. Omar *et al.* 2008 used sago and potato flour for COD reduction from semiconductor wastewater. Poly aluminium chloride and aluminium sulfate coagulants were also used. The researchers found that sago helped in COD and turbidity reduction at a low dose of 1.5 g L<sup>-1</sup>. On the other hand, considerable reductions in COD and turbidity were seen using poly aluminium chloride and ammonium sulphate at much lower concentrations (0.02–1.0 g L<sup>-1</sup>), although much longer settling time of 30 to 60 min was required (Omar *et al.*, 2008).

### 5. High inventory and processing cost

Natural coagulants are likely to have high inventory and processing costs as compared to chemical

coagulants due to their biodegradable nature. Also, storing the natural coagulants in stock for later use will require some arrangements.

### 6. Lack of awareness, market interest, and guidelines

There is currently a lack of awareness in general public and utility boards regarding the adverse effects of synthetic coagulants and the availability of natural coagulants as an eco-friendly alternative to synthetic coagulants. Due to economic uncertainties, utility services and plant operators are less likely to opt for a new technology esp. when there are more aspects of the natural coagulants that need detailed research. Standard guidelines related to the production, storage, and use of natural coagulants are not available. All these need to be taken care of, to promote the commercial adoption of the natural coagulants.

## CONCLUSION

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Synthetic chemicals have been used as chemical coagulants for the treatment of industrial wastewater for a long time. Aluminium salts are the most commonly used coagulants. Despite their high efficiency of treatment, chemical coagulants have inherent disadvantages. These include the production of nondegradable sludges, high cost, and (most importantly) medical conditions due to leftover aluminium in the water. Many studies have been conducted to assess the potential of using natural coagulants as an alternative to synthetic chemicals. Many of these plant-based coagulants (especially, *Moringa oleifera*) have shown significant promise in treating the wastewater emanating from different types of industries. This review comprehensively presented the advantages and disadvantages of both the natural and synthetic coagulants. Despite being the most economical and environment-friendly alternative, natural coagulants are not preferred over chemical coagulants for the wastewater treatment, esp. on a large scale. Competitiveness, high inventory, and operating cost, lack of source for mass availability are some of the hurdles in their large-scale application. There is a significant lack of studies to utilize the potential of natural coagulants for wastewater treatment. Studies to discover the shelf life of these coagulants will help in determining the possibility of the potency losses with these coagulants. This will be a

major contribution in ascertaining a reduction in the efficiency of these coagulants over a given period of time. In addition, further studies can be conducted to determine the techniques to improve the shelf life of natural coagulants. There is a scarcity of literature to understand the nature of sludge produced. This may also help in encouraging the treatment using natural coagulants. The use of natural coagulants should be tested for large-scale wastewater treatment, to verify results obtained in the laboratory. Hopefully, the review presented here will stimulate the research in the identified directions and motivate the researchers in finding sustainable solutions to industrial wastewater treatment.

### RECOMMENDATIONS

Given the inherent disadvantages of synthetic coagulants, it is recommended that the natural coagulants be used as an alternative for the treatment of industrial effluents. To support this transition, several areas of research have been identified, including, improvements in the processing, efficiency loss due to storage, use of waste plant-based products as coagulants, development of standard guidelines, etc. It is suggested that the natural coagulants are first used at small or medium-scale effluent treatment plants and the data and experience thus generated is used for large-scale implementation.

### AUTHOR CONTRIBUTIONS

S. Gautam wrote the first draft of the manuscript. G. Saini developed the concept, supervised the manuscript preparation and edited it.

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### CONFLICT OF INTEREST

The authors declare that there is no conflict of interest regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/or falsification, double publication and/or submission, and redundancy have been completely observed by the authors.

### ABBREVIATIONS

ANOVA	Analysis of variance
BOD	biological oxygen Demand
COD	Chemical oxygen demand
Cd	Cadmium
Cr	Chromium
Cu	Copper
FeCl <sub>3</sub>	Ferric chloride
FeSO <sub>4</sub>	Ferrous sulfate
g/L	grams per litres
KAl(SO <sub>4</sub> ) <sub>2</sub> ·12H <sub>2</sub> O	Potassium Aluminium Sulphate
KLD	Kilo litres per day
MgCl <sub>2</sub>	Magnesium chloride
mg/L	Milligram per litre
mL/L	Milliliter per litre
MLD	Million litre per Day
Mg	Microgram
O&G	Oil and grease
PAA	Polyacrylamide
PAC	Poly aluminium chloride
PAFC	Poly aluminium ferric chloride
PASiC	Poly-aluminium-silicate-chloride
PFS	Polyferric sulfate
<i>P. ovata</i>	<i>Plantago ovata</i>
PsiFAC	Poly-aluminium-ferric-silicate-chloride
TDS	Total Dissolved Solids
TP	Total phosphorus
TSS	Total suspended solids
WHO	World Health Organization

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