AKAApplications of Coagulants in Leachate Treatment – A critical review

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***Abstract:** Among the various applications of coagulants, leachate treatment is considered as one of the most significant domains. Sanitary landfill is the most common way to eliminate solid waste. In any case, sanitary landfill creates expansive amount of leachate. Leachate can be characterized as a fluid that percolates through a landfill and leached out dissolved and suspended matter. The nearness of vast amount of contaminants in leachate is unsafe to human and natural condition. This can prompt well-being issues, including gastrointestinal ailment, contraceptive issues, and neurological issue. There are several significant techniques have been made to overcome the problem of leachate pollution including photo catalytic oxidation and bioremediation etc. However, the applications have been restricted by many factors, such as processing efficiency, operational method, energy requirements, and economic benefit. The main aim of this review paper is to throw insight on the overview of research studies concerned with the coagulants, coagulants aid and their potential applications in leachate treatment.*

***Keywords:** Leachate, Coagulation, Landfill, Flocculation, Treatment*

Introduction:

Disposal of solid waste in the landfill is one of the common and economically viable waste management approach implemented across the world. In general, most of the metropolitan cities normally perform the landfilling techniques. Thus 90% of the solid wastes in developing countries like India are disposed in an unempirical approach. The accessibility of land for discarding the generated waste is decreasing day by day **(Mor *et al.*, 2006; Siddiqui *et al.*, 2006; Sharholy *et al.*, 2005).** In course of time the solid waste disposed in landfill produces a slimy liquid called "**Leachate**". The production of leachate and managing the produced leachate is one of the environmental concerns as the produced leachate causes damage to soil and water (both underground and surface water). Unless precautionary procedures are taken, this will lead to a great menace to environment **(Baccini *et al.*, 1989; Lopez 2004).** Increased concentration of leachate makes the underground water to contaminate very easily **(Petruzzelli *et al.*, 2007; Sarvako, 2007, Yang, 2006).**

Coagulation-flocculation is widely used for wastewater treatment. This treatment is efficient to operate. It have many factors can influence the efficiency, such as the type and dosage of coagulant/flocculants, pH, mixing speed and time and retention time. The optimization of these factors may influence the process efficiency **(Ozkan *et al*, 2004).** Coagulation process followed by flocculation process was seen to be more effective in organic pollutant removal **(Zhang et al, 2004).** A polymer or polyelectrolyte acts as a bridging agent in promoting the formation of greater flocs that settle more efficiently, ultimately resulting in higher removal rates **(Zouboulis *et al*, 2004).**

Coagulation followed by flocculation process is an effective way for removing high concentration of organic pollutants **(Wang *et al*, 2002).** Coagulation-flocculation is destabilizing the colloidal suspension of the particles with coagulants and then causing the particles to agglomerate with flocculants. After that, it will accelerate separation and thereby clarifying the effluents **(Gnandi *et al*, 2005).**

Leachate Characteristics:

COD, BOD, BOD/COD, pH, Suspended solids (SS), Ammonium nitrogen (NH3-N), Total Kjeldahl nitrogen (TKN) and heavy metals are the basic parameters for determining the quality of a leachate. The leachate as reported in different literature demonstrates a wide variation in their composition.

Table 1: Typical composition of Landfill Leachate (Source: Vesilind and Rimer, 1981)

Parameters	Typical Value (mg/L)	Range (mg/L)
BOD_5	10,000	200-40,000
COD	30,000	300-90,000
TOC	6,000	1,500-20,000
TSS	500	200-1,000

pH	6	4.2-7.8
Zn	50	25-250
Pb	2	0.2-10
Fe	60	25-2,500
Hardness as $CaCO_3$	3,500	300-10,000
TKN	1,000	0.2-13,000
NH_3-N	766	>400

Table 2: Standard for Limit values for the discharge of treated leachate according to German standards (Rahmen *et al,* 1996)

Parameters	Limiting Concentration (mg/L)
COD	200
BOD_5	20
TKN	70
NH_3-N	2
Pb	0.5
Zn	2
TOC	70
TSS	100

Review of Coagulation-Flocculation process of Landfill Leachate Treatment:

Keenan *et. al,* 1983 [36] analyzed the results of full scale physico-chemical treatment of sanitary landfill leachate , the treatment scheme consists of equalization, lime precipitation, sedimentation and air stripping of NH_3 and the results demonstrated that the overall percent removal efficiencies of $BOD_{5,}$ N, Cd, Cu, Fe, Pb, Hg, Ni, Zn, TSS was found to be 69%, 53%, 59%, 38%, 98%, 66%, 52%, 54%, 95%, 95% respectively and it was also observed that equalization markedly reduces operating costs.

Slater *et. al,* 1983 [19] conducted a study and examined the efficiency of physic-chemical process using lime and he found that there was substantial reduction in dispersed oil, turbidity and some heavy metal species and the recabronation was suggested to adjust the pH desirable for successive treatment processes.

Otal *et. al,* 2002 [35] studied the application of combination of physicochemical treatment along with the zeolite treatment in which commercial zeolite and synthetic zeolite prepared from fly ash was used and the results showed that this treatment achieved an important improvement in COD and Total kjeldahl nitrogen (TKN) reduction. No differences were observed in COD reduction between the two zeolites tested, while regarding nitrogen removal, the zeolitised fly ash showed a better behavior.

Ahn *et. al,* 2002 [6] carried out a systematic enquiry on the effect of pre-removal of ammonia from leachate using zeolite on the efficiency of biological treatment and it was inferred that 87% NH_3-N was removed effectively and remarkably improved COD removal efficiency (83%), since high concentration of NH_3-N is toxic to the microorganisms in biological treatment and result in the sludge rising.

Zouboulis *et. al,* 2004 [5] examined the efficiency of bioflocculant along with the coagulant and the bioflocculant was produced by the bacterium *Rhizomonas sp*. and the test was performed using conventional jar-test coagulation experiments and it was found that the optimal pH for the removal of humic acid was 7-7.5 while a dosage of 20 mg/L was sufficient in removal of 85% humic and the results also indicated that almost 45% COD removal could be achieved using either the bioflocculant or coagulant. However, the use of bioflocculant was more advantageous as the bioflocculant produced similar results as the use of alum did, but with substantially lower dosage.

Sang *et. al,* 2005 [17] comprehensively studied the flocculation treatment of organic substances present in leachate using a novel inorganic polymer flocculants Poly-ferric(III)-magnesium(II) aluminum(III)-sulfate (PFMAS) and it indicate that the removals of color, COD, and BOD_5 by coagulation with PFAMS can reach 89%, 68%, and 58% respectively which greatly reduces the extent of pollution and improves its appearance. Particularly, the species with no benzene ring were removed greatly (about 60-100%), and some were removed completely, while species with a benzene ring were partially removed (about 25-70%).

Aziz *et. al*, 2006 [22] investigated the efficiency of coagulation and flocculation processes for removing color from a semi-aerobic landfill leachate. In this study 4 coagulants were used and their effects were studied using standard jar apparatus test and the results indicated that ferric chloride was superior to the other coagulants and removed 94% of color at an optimum dose of 800 mg/L at pH 4.

Luna *et. al*, 2007 [14] conducted a pilot plant study using zeolitised coal fly ash as adsorbent and alum and polyelectrolyte as coagulant and the results showed that it is possible to remove 43%, 53% and 82% of COD, NH_3–N, and TSS respectively. Therefore, this method may be an alternative for ammonical nitrogen removal.

Zoupanos *et. al*, 2008 [20] evaluates the efficacy of applying polyaluminium silicate chloride as coagulant aid in treating old leachate that is relatively stable and are difficult to treat further and the results suggested that the silica based composite coagulants exhibit better coagulation performance, than the relevant conventional coagulants.

The main objectives of the research of **Yousefi *et. al*, 2008 [23]** was to determine the removal efficiency of heavy metals (Ni, Cd, Cr, Zn and Cu) and COD present in raw leachate using coagulation-flocculation process and the results indicated that the removal efficiencies of heavy metals and COD was 80% and 25% respectively and the results illustrated that the physico-chemical process may be used as a useful pretreatment step especially for fresh leachates.

Moreover, **Zazouli *et. al*, 2009 [24]** conducted a research on Composting plant Leachate treatment using $Al_2\ (SO_4)_3.18H_2O$ (Alum) and $FeCl_3.6H_2O$ as coagulants and the experimental results show that 18% removal of COD and 90% removal of heavy metals can be attained at pH 6.5 (optimum for alum) with the addition of 1400 mg/L of alum and 28% removal of COD and 86% removal of heavy metals can be attained at pH 10 (optimum for ferric chloride) with the addition of 2000 mg/L of ferric chloride.

Ghafari *et. al*, 2009 [12] carried out his experiment using Poly Aluminum chloride (PAC), Alum and cationic polymer (CP) as coagulants and demonstrated that PAC was very effective in removal of physical parameters such as color, turbidity and SS but less effective in removal of COD compare to alum. Optimum dose of PAC and CP used in the experiments were 1000 mg/L and 8mg/L respectively at pH 7 by optimizing different experimental conditions. Results showed that 52.66% removal of COD, 97.16% removal of SS and 96.44% removal of color were achieved under optimum condition.

Zhang *et. al*, 2009 [13] observed that xenobiotic organic substances (XOC's) such as phthalic acid esters can be sorbed to dissolved organic matter (DOM), removal of DOM from leachate results into overall removal of phthalic acid esters.

Rui *et. al*, 2010 [14] further used Modified Micro sand (MMS) in the above experiment of Ghafari *et. al*, 2009 to enhance the settling down of sludge particle and to increase the rate of flocculation, Modified Micro sand was prepared by prepared by dissolving the sodium aluminum and sodium hydroxide in distilled water with micro sand and results showed that with 300mg/L of MMS, the settling time was reduced to 1 minute using MMS as compared to 45 minutes without using MMS.

Samadi *et. al*, 2010 [15] conducted a research on treatment of landfill leachate of Hamadan Landfill in Iran using coagulation- flocculation process using PAC, Alum and $FeSO_4$ and the results showed that the efficiency for COD removal by PAC at pH 12 and 2500 mg/L of dose, by alum at pH 12 and 1000 mg/L of dose and by $FeSO_4$ at pH 12 and 1500 mg/L of dose were 60%, 62.33% and 70.66%, respectively. Also results showed that, the efficiency for TSS removal by PAC that was obtained at pH 12 and 2500mg/L concentration, by alum at pH 2 and 1500 mg/L concentration and by $FeSO_4$ at pH 7 and 2500mg/L of $FeSO_4$, were 39.14% , 58.37% and 35.58%, respectively.

Zhan-meng *et. al*, 2010 [12] introduced the Pre-treatment of sanitary landfill leachate using a novel coagulant i.e., Poly-ferric (III)-magnesium (II)-sulfate (PFMS) and the results indicated that using PFMS was far better than other traditional coagulants as COD and color removal rate can reach 67% and 88% respectively. Furthermore, the biodegradability of the leachate was enhanced from 0.46 to 0.68. He also studied the decolourization performance and observed that the coagulation efficiency of PMAS for the treatment of a secondary effluent was generally better than the traditional coagulants with the decolourization efficiency greater than 90%.

Maranon *et. al*, 2010 [33] conducted a research on treatment of leachate by coagulation-flocculation process using different coagulants and polyelectrolyte and the findings were such that the Higher pollutant removal (73% COD, 98% color and 100% turbidity) efficiencies were obtained using ferric chloride at pH 5.0–5.5 and dosage of 0.6 g/L and the volume of sludge generated after centrifugation represents about 4.0–4.6% when ferric chloride is used an and 10% when employing aluminum polychloride.

Weng *et. al*, 2010 [18] carried out three stage physic-chemical and biological treatment of domestic landfill leachate that concentrates complex organic substances and this 8 month pilot operation was divided in to three stages in which UASB reactor and SBR were used along with the coagulation tank and the results indicate that stage 1 removes most pollutants, and stages 2 and 3 remove recalcitrant pollutants. The combined effect of the three stages ensured that the effluent characteristic meets the discharge standards.

Comstock *et. al*, 2010 [9] compared three types of coagulants which focused on removal of dissolved organic matter (DOM) from leachate. The presence of DOM was measured using specific ultraviolet absorbance at 254 nm (SUVA254) and fluorescence excitation–emission matrices. The performance of the metals salts was in the order of:- ferric sulfate > aluminum sulfate > ferric chloride and DOM removal followed the trend of color > UV254 > dissolved organic carbon > COD.

Ling *et. al*, 2011 [20] used organically modified Bentonite (OMB) to treat and dispose of old leachate (>10 years) and after treatment using OMB, the COD of the sample decreases from 2400 mg/L to 245 mg/L which is attributed to 90% reduction and the Gas chromatography-Mass spectrometry results indicated that most of the organic compounds were removed during the process. The results have provided a new cost efficient method for treatment of landfill leachate.

A combined process was employed in the study of **Zhang *et. al,* 2011 [21]** to treat old phosphorus slag leachate and it was inferred that the dispersed aeration reduced yellow phosphorus to 1 ppm and the addition of sodium hypochlorite accelerated the reaction. Beside this, the coagulation process reduced turbidity as well as the concentration of Fluorides and Fe^{2+} and the final process of the combined membranes technology (UF + NF + RO) efficiently removed total phosphorus, COD and $Fe2^{+}$ to acceptable limits and it was clearly detected that the effluent did not contains yellow phosphorus.

Hamadani *et. al,* 2011 [7] tests the potential of psyllium husk as coagulant aid with PAC and alum for treatment of semi-aerobic leachate and it was observed that additional 9, 10 and 2% removal efficiencies were recorded for COD, color and TSS respectively when psyllium husk was used as coagulant aid along with PAC and it was also concluded that psyllium husk was more effective as coagulant aid when used with PAC as compared to alum (5, 4 and 2% respectively).

Awang *et. al,* 2012 [29] carried out an experiment using ***Hibiscus rosa-sinensis*** leaf extract as natural coagulant aid in treating landfill leachate objective of this study is to examine the efficiency of coagulation–flocculation processes for the removal of color, iron (Fe^{3+}), suspended solids, turbidity and ammonia nitrogen (NH_3–N), from landfill leachate using 4,000 mg/L alum in conjunction with H. rosa-sinensis leaf extract (HBaqs) and the result showed that The $Fe3^{3+}$ removal rate using 4,000 mg/L alum as sole coagulant was approximately 60 %, and increased to 100 % when 4,000 mg/L alum was mixed with 500 mg/L HBaqs. By mixing, 4,000 mg/L alum with 100–500 mg/L HBaqs, 72 % of SS was removed as compared with only 45 % reduction using 4,000 mg/L alum as sole coagulant. The research has shown a good potential of HBaqs to be used as natural coagulant aid in leachate treatment as alternative to the commonly use polymeric commercial coagulant aid.

Syafalni *et. al,* 2012 [24] investigated the capability of lateritic soil as coagulant in treating stabilized landfill leachate and the evaluation was done using conventional jar test and the inference that was drawn from the experiment showed that The optimal pH was 2.0, while 14 g/L of lateritic soil coagulant was sufficient in removing 65.7% COD, 81.8% color and 41.2% NH_3-N. Conversely, the optimal pH and coagulant dosage for the alum were pH 4.8 and 10 g/L respectively, where 85.4% COD, 96.4% color and 47.6% NH_3-N were removed from the same leachate sample. Additionally, the Sludge Volume Index (SVI) ratio of alum and lateritic soil coagulant was 53:1, which indicated that less sludge was produced and was an environmentally friendly product. Hence, it can be concluded that the lateritic soil coagulant can be considered a viable alternative in the treatment of landfill leachate.

Liu *et. al,* 2012 [13] reported that the recalcitrant compounds such as humic acids can also be removed by coagulation-flocculation process. Humic acid removal efficiencies of 80, 53, 70 % when ferric chloride 6-hydrate, ferric sulphate 7-hydrate, and poly ferric sulphate were used, respectively.

The main goal of the study of **Raghab *et. al,* 2013 [27]** was to utilize a natural low cost material ''as an accelerator additive to enhance the chemical treatment process using Alum coagulant and the accelerator substances were Perlite and Bentonite. The performance of the chemical treatment was enhanced using the accelerator substances with 90 mg/L Alum as a constant dose. Perlite gave better performance than the Bentonite effluent. The removal ratio for conductivity, turbidity, BOD and COD for Perlite was 86.7%, 87.4%, 89.9% and 92.8% respectively, and for Bentonite was 83.5%, 85.0%, 86.5% and 85.0% respectively at the same concentration of 40 mg/L for each.

Gandhimathi *et. al,* 2013 [10] studied the use of combined coagulation(alum and $FeCl_3$)-adsorption(fly ash) process for pre-treatment of leachate and was found that overall COD removal efficiency of 82% was obtained by coagulation using alum and adsorption using fly ash for stabilized leachate.

Guneş *et. al,* 2014 [16] aims at seasonal characterization of the leachate of Corlu which was heavily industrialized and also examined the efficiency of coagulation-adsorption process of treatment using granular activated carbon (GAC0 and waste metal hydroxide (WMH) as adsorbent, alum and lime as coagulant and at optimum conditions the results clearly indicates that by coagulation/flocculation/GAC adsorption and coagulation/flocculation/WMH adsorption the COD removal efficiencies were respectively 56-63% and 45-48% and color removal efficiencies were 77-91% and 81-92%, respectively. Hence, the adsorption study showed that these adsorbents could be used for only color removal for this landfill leachate the process was more efficient during summer and spring season.

Pushplata *et. al,* 2015 [11] utilizes Titanium chloride as an effective coagulant in her study for treatment of landfill leachate and it was found that $TiCl_4$ was much more efficient at low dosage than other coagulants used in the study.

Sobri *et. al,* 2015 [8] examined the efficacy of starch based coagulant aid from sago trunk for semi aerobic leachate treatment and found that dosage of PAC was reduced up to 35% when sago was used as coagulant aid and the removal performance for color, SS and turbidity were 94.7, 99.2, and 98.9 %, respectively.

Zamri *et. al,* 2015 [2] applied the coagulation-flocculation for semi-aerobic landfill leachate using mechanically treated starch from oil palm trunk waste as coagulant and the jar test clearly showed that this botanical coagulant was much more effective in COD removal than PAC, however it was less effective in removing suspended solids.

Rusdizal *et. al,* 2015 [32] conducted a study to treat stabilized leachate by applying PAC and tobacco leaf extract as a coagulant and coagulant aid and the morphology of tobacco leaf extract was determined using Scanning electron microscopy (SEM) and it indicated that extract was positively charged and further, it was concluded that the removal rate of the COD using 1500 mg/L PAC as a sole coagulant was approximately 63% and increased to 91% when 1000 mg/L PAC was mixed with 1000 mg/L of tobacco leaf extract. Additionally, 1500 mg/L PAC with 250-1000 mg/L tobacco leaf helps in achieving 54% NH_3-N removal efficiency compared with only 46% using 1500 mg/L of PAC. Therefore, this study reveals that PAC is more efficient in improving physical characteristics of leachate, but a combination of PAC and tobacco leaf extract better removes COD and NH3-N.

Chakrabarty *et. al,* 2016 [31] in his study determined the most appropriate type of coagulant and optimum dosages for removing of pollutants in leachate. The coagulation process was applied with H_2O_2 and $FeSO_4$ at different dosages and with definite pH of 2, 4, 6, 8 and 10. Finally, the experimental results demonstrated that $FeSO_4$ had the best performance to reduce TSS while H_2O_2 effectively reduced color, turbidity and EC.

Zin *et. al,* 2016 [34] investigated the performance of a natural coagulant synthesized using tapioca starch for treatment of partially stabilized leachate and the results suggested that tapioca starch might not be the best coagulant compared to the chemical ones because of its low removal efficiency but combining tapioca starch as a coagulant aid in future might improve the coagulation-flocculation performance of it.

Yusoff *et. al,* 2016 [5] in his attempt used Jackfruit seed starch as a coagulant aid along with PAC and it was demonstrated that the addition of seed reduced the dosage of PAC up to 33% and the removal efficiencies were 33.6%, 93.6%, 13.1 %, 94%, 92% respectively for COD, Color, NH_3-N, Turbidity and TSS.

Zengnian *et. al,* 2016 [35] illustrate the treatment of composting leachate by combination of coagulation and nanofiltration process using Poly ferric sulphate as coagulant and PMIA (Poly m-phenylene isophthalamide) as nanofiltration membrane and its cross-sections were analyzed using SEM and the final leachate effluent concentration of COD, NH_3-N, TOC and TSS were 92 mg/L, 21 mg/L, 73 mg/L and 23 mg/L respectively which were under acceptable limit.

Poveda *et. al,* 2016 [**30]** experimentally compared the effectiveness of four different leachate pre-treatment options viz. air stripping, chemical coagulation, electro-coagulation and advanced oxidation in terms of COD and ammonia removal and it was demonstrated that combination of air stripping and chemical coagulation removed up to 50% COD and 85% of ammonia and the order in which the methods were applied did not affect the removal efficiencies.

Recently, **Zouaghi *et. al,* 2017 [25]** carried out investigation on treatment of leachate from urban waste using Alumina sulphate (10g/L) as coagulant and anionic polyelectrolyte (1g/L) as flocculent and by using different optimization techniques the physicochemical parameters gave a pH of 8.46, an electrical conductivity of 18.24mS/cm, an orthophosphate concentration of 0.35mg/L, an oxidability of 125mg O_2/L and turbidity of 252 FAU.
Furthermore, in the above experiment adsorbents were also used mainly Palm leaves (4 gm), Alep pine bark (2 gm) and Activated carbon (6 gm) and the adsorption with 2gm of Alep pine bark/100ml of leachate gave the best results of turbidity of 93 FAU at pH 9.14 with stirring time of 120 minutes.

Zin *et. al,* 2017 [1] conducted an experiment using dual coagulant (Alum + Barley) for removing color from landfill leachate and it can be concluded that barley as coagulant aid able to reduce 33% usage of alum for achieving 98% removal efficiency of color. Thus, the dual coagulant consisting alum and barley has the potential to be applied as a coagulant for leachate treatment.

Zainal *et. al,* 2017 [3] examined the effectiveness of $SnCl_4$ (Tin chloride) for treatment of stabilized leachate and the best conditions were obtained at pH 8 and with optimum dosage of 15g/L, the removal efficiencies were found to be 98.40 % for color, 99.54 % for SS, and 71.53 % for COD.

Chouki *et. al,* 2017 [4] aimed to investigate the feasibility of coagulation-flocculation process to treat MSW landfill leachate of Casablanca city of Morocco using lime, alum and $FeCl_3$ and it was concluded that heavy metals were removed most effectively using $FeCl_3$ and it was accepted 1.4-1.6 times more than lime and alum.

Bazrafshan *et. al,* 2017 [32] conducted an experimental study to illustrate the effect of combined coagulation and adsorption on leachate using tea waste as a low cost adsorbent and it was observed that the combined process was sufficient for treatment of this leachate and hence modified tea waste should be applied as an adsorbent.

Simonic *et. al,* 2017 [26] predominantly focused on proper pre-treatment method in order to reduce the fouling index of polysulfone nano-membrane and to assess its filtration capacity and it was cleary observed that using PAC as coagulant helps in removing 75% colloids and it was absolutely necessary for removing foulants like humic acid and improving the flux performance and filtration index of nano-membrane.

Thaldiri *et.al,* 2017 [28] observed the effect of modified micro-sand, poly-aluminum chloride and cationic polymer on coagulation-flocculation process of semi-aerobic landfill leachate treatment and the final outcome inferred 52.66 % removal of COD, 97.16% removal of SS and 96.44% removal of under optimum condition. The durations of settling time to settle down the flocs were recorded as 1 minute (modified sand), 20 minutes (raw micro sand) and 45 minutes (without micro sand).

Conclusion:

The physico-chemical process may be used as a useful pretreatment step especially for fresh leachate prior to biological treatment or as a post-treatment step for partially stabilized leachate. Despite having recent advancement of landfill leachate treatment technology, it is well perceived that coagulation and flocculation is still predominant for the treatment of landfill leachate is widely practiced by the many landfill operators. In fact, Ferrous sulfate can also considered as a better coagulant over other metallic salts. Emerging of starch-based coagulants in leachate treatment is seen as promising approach which can reduce dependent of pure coagulant which can effectively improve sludge production during flocculation process.

Furthermore, optimization of this process can be done by applying Central composite design (CCD) and Response surface methodology (RSM) to optimize the operating variables i.e., coagulant dosage and pH.Quadratic models developed for the responses are studied to demonstrate the optimum condition.

Acknowledgement

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