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# Coagulation-Flocculation Process in Landfill Leachate Treatment: Focus on Coagulants and Coagulants Aid

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**Abstract.** In physico-chemical treatment, the separation of suspended particles from the liquid phase is usually accomplished by coagulation, flocculation and sedimentation. Coagulation-flocculation processes have been widely used as alternative treatment to remove leachate pollutants such as BOD, COD, TSS, heavy metals, colour, and nitrogen compounds prior to other treatment methods. It is often coupled with treatment methods like biological process, chemical oxidation, adsorption or filtration to achieve desirable effluent quality. In spite of being economical, the dewatering and disposal of the precipitated sludge could be laborious and time-consuming. In this manuscript, brief discussions on coagulant and coagulants aid in landfill leachate treatment is discussed with respect to their mechanism.

## 1. Introduction

Coagulation is generally defined as destabilization of a colloidal suspension or solution. On the other hand, flocculation is the process whereby the destabilized particles are propagated to gather and agglomerate into larger flocs. Although coagulation and flocculation are two distinct processes, they have been used interchangeably due to the confusion resulted from water treatment industry itself [1].

The same goes for the terms ‘coagulant’ and ‘flocculant’. However, it is pertinent to assert that ‘coagulant’ is often used to describe any substances that can induce both coagulation and flocculation [2].

### 1.1. Colloids in Landfill Leachate

Dispersed solids in landfill leachate comprise non-sedimentary suspending materials with negligible sedimentation velocity; and colloids are the major constituents of these non-sedimentary particles. Colloids are generally defined as solid particles with a size between 1 nm and 1 mm [3,4,5]. Some



studies had even revealed that particles with a size up to 10  $\mu\text{m}$  might act as colloids as well [6].

Matura and co-researchers had noted that the colloids in landfill leachate were mostly inorganic and mainly consisted of carbonates and clays. In addition, colloids can be classified as hydrophobic (resistant to water bonding) or hydrophilic (affinity for water bonding). Owing to the attraction of hydrophilic colloids for water, they are difficult to be removed from dispersion. However, this classification does not work for colloids in wastewater as it is hard to differentiate between the two [1].

### 1.2. Colloid and Contaminant Mobilization

Colloids are important in the context of speciation, fate, transport, biogeochemistry, bioavailability and toxicity of contaminants in the environment [7]. For landfill leachate particularly, several researchers had asserted similar conclusions [8,9]. Heavy metals are bound to colloids by adsorption, precipitation (and surface precipitation) and ion exchange [11]. Owing to the great affinity of colloids for heavy metals, around 70 to 90% of metals in landfill leachate are in colloidal or organic complexes (Baun & Christensen, 2004). Excluding highly mobile elements like arsenic, selenium and rubidium, most trace elements bind at least 20% individually to colloids and some metals like aluminium, iron and lead may go up to nearly 100% as shown in table 1.

**Table 1.** Binding of elements to colloidal particles in landfill leachate.

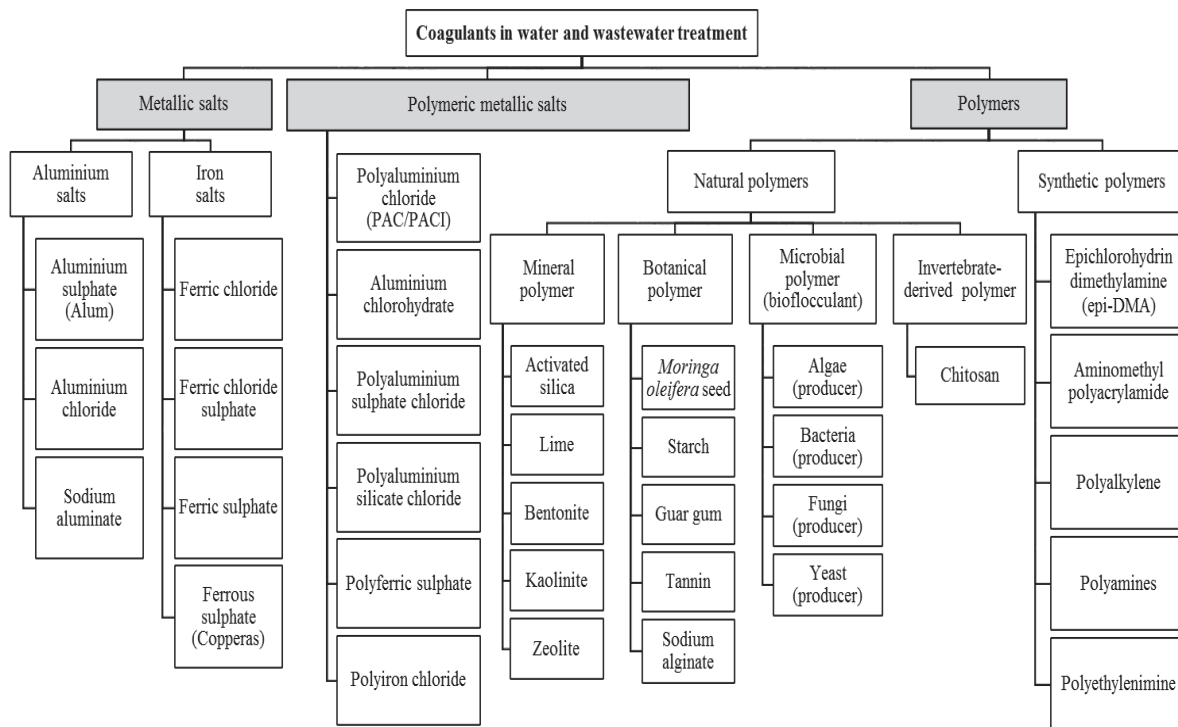
Element	Binding to colloids (%)
As, Se and Rb	0
Ca, Co, Cr, Ni, Cu, Zn, Sr, Ba, V and Sb	$\geq 20$
Al, Fe and Pb	$\sim 100$

## 2. Coagulants and Coagulant Aids

### 2.1. Coagulants

Coagulants are chemicals added into water or wastewater to perform coagulation. They can be classified broadly into three types i.e. metallic salts (or hydrolysing metallic salts), polymeric metallic salts (or pre-hydrolysed/pre-polymerised metallic salts) and polymers. Polymeric coagulants encompass both natural and synthetic polymers. Detailed classification of coagulants is depicted in Figure 1. Amongst them, microbial flocculants or bioflocculants are not produced or applied in industrial scale, albeit literatures regarding their bench scale coagulation efficiencies are scanty in recent years. This is due to their low yield and high production cost [12].

Moreover, their coagulation mechanisms are inherently complicated. Therefore, bioflocculants are not reviewed in depth in this work. The same goes for other less applicable coagulants. They are excluded since they are less favoured in water treatment industry. In addition, general requirements for coagulants in water treatment are: (1) trivalent metallic cations or polymers that have been proven efficient as coagulants; (2) nontoxic and do not cause any detrimental physiological effects on human health; and (3) insoluble or low solubility in the pH ranges common in water treatment to minimise chemical residual in effluent [13]. Owing to that, coagulant for particular wastewater treatment has to be selected carefully based on their feasibilities. Advantages and drawbacks of commonly used coagulants are shortlisted in table 2.



**Figure 1.** Classification of coagulants in leachate treatment.

**Table 2.** Degree of colloid stability as a function of zeta potential.

Zeta potential (mV)	Colloid stability
from 0 to $\pm 5$	Rapid coagulation or flocculation
from $\pm 10$ to $\pm 30$	Incipient instability
from $\pm 30$ to $\pm 40$	Moderate stability
from $\pm 40$ to $\pm 60$	Good stability
more than $\pm 61$	Excellent stability

## 2.2. Coagulant Aids

Coagulant aids are applied in tandem with primary coagulants (mostly metallic coagulants) to optimize coagulation process. They accelerate clarification, strengthen the flocs, improve sludge settleability, reduce coagulant dosage and treatment cost, provide a wider effective pH range and elevate contaminant removal efficiency [14]. Shammas [11] had postulated that there are four types of typical coagulant aids i.e. alkalinity additives, pH adjusters, particulate additives and polymers.

Despite most literatures only consider particulate additives and polymers as coagulant aids (by means of post-orthokinetic agglomeration), alkalinity additives and pH adjusters are also included in this review because they provide conducive environments for optimum coagulation-flocculation (by means of orthokinetic agglomeration). Indeed, coagulant aids are also commonly classified into two types i.e. particle binders and nucleating agents based on their action mechanisms. For particle binders, they promote flocculation like primary coagulants while the nucleating agents act as nucleation sites to facilitate floc formation.

Most of the commonly used coagulant aids are polyelectrolytes, especially those synthetic polymers like polyacrylamides, polydiallyldimethyl ammonium chloride (poly-DADMAC) and polystyrene sulfonate. Polyelectrolytes are polymers with ionisable functional groups such as

carboxyl, amino or sulphonic groups. They can be cationic (with positive functional groups), anionic (with negative functional groups) or amphoteric (with both positive and negative functional groups). Polymers without any ionisable functional group are nonionic. Comparison of these typical coagulant aids is summarised in Table 3.

### 2.3. Polyaluminium Chloride (PAC)

Polyaluminium chloride (PAC) is a highly effective polymerised inorganic coagulant. It is the dominant variety of polymeric inorganic salts or so-called the New Age coagulants in wastewater treatment. Indeed, PAC is a generic name for various salts with a general formula,  $Al_nCl_{(3n-m)}(OH)_m$  that can be formed by polymerising aluminium chloride under different proprietary reaction conditions. They include aluminium chlorohydrate, basic aluminium chloride, polybasic aluminium chloride, aluminium hydroxide and aluminium oxychloride. They are sold in both aqueous and dry powder forms. Among them, aluminium chlorohydrate (ACH),  $Al_2Cl(OH)_5$  is the most common PAC in the market. Comparison between these two PACs is shown in **Error! Reference source not found.**

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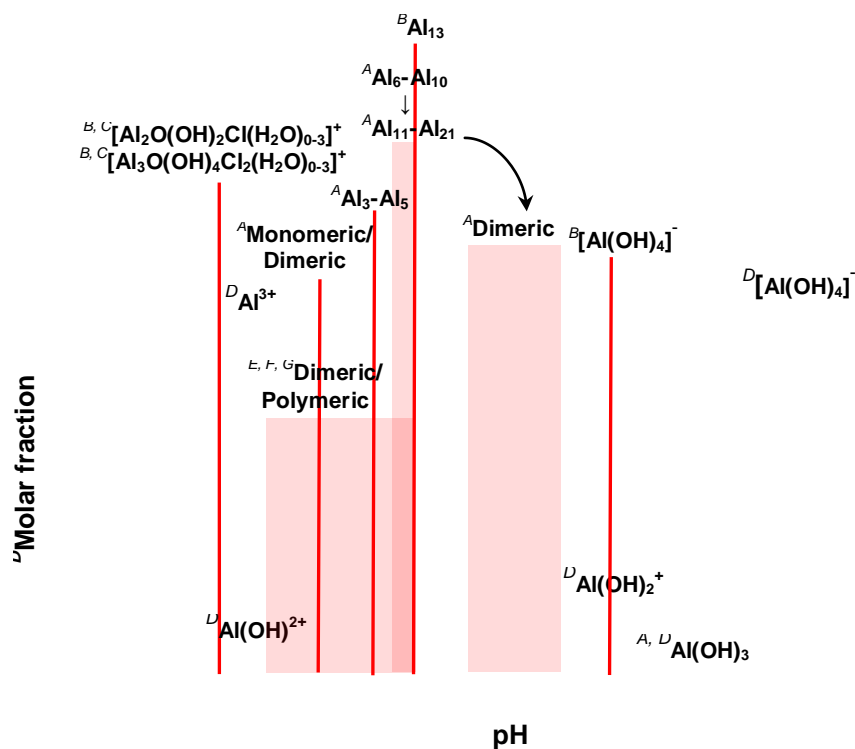
Knowledge on Al speciation is crucial for PAC coagulation-flocculation optimisation. When PAC is introduced into water, aluminium (Al) hydrolyses into diverse monomeric and polymeric forms, depending upon pH [16,17,18,19], counter-ions, Al salt concentration [20], temperature [21] and hardness [22]. Though Al hydrolysis reactions at different pH values is not yet ascertained, several studies in the past have shed some light on their predominant species in different pH ranges (Figure 3).  $Al^{3+}$  [ $Al(H_2O)_6^{3+}$ ],  $Al_{13}$  [ $AlO_4Al_{12}(OH)_{24}(H_2O)_{12}^{7+}$ ] and aluminate ions [ $Al(OH)_4^-$ ] seem to be the dominant species at pH <4,  $\approx 5$  and >7, respectively. In the presence of counter-ions like humic acid, fulvic acid, sulphates, phosphates and fluorides, speciation and precipitation of Al will vary. In light of space limitation, other less significant alluded factors are left out of this discussion.

Amongst all hydrolysed Al species, metastable pre-hydrolysed polynuclear  $Al_{13}$  species (a.k.a. tridecameric Al or Keggin  $Al_{13}$ ) is believed to be the main active species for coagulation. Its superiority over other Al species especially  $Al^{3+}$  (key Al species from alum and  $AlCl_3$  hydrolysis) is supported by the fact that it has the greatest charge neutralisation ability, higher structural stability and nanosized molecular diameter. However, this theory is now challenged by the discovery of  $Al_{30}$  [ $\{(AlO_4)_2Al_{28}(OH)_{56}(H_2O)_{26}\}^{18+}$ ] and its characterisation in recent years [4,9,11,12]. Its preponderance over  $Al_{13}$  has been postulated since it is bigger (1 nm diameter and 2 nm length) more positively charged, more thermal resistant and stable in hydrolysis process especially at low pH. Despite the coagulation mechanism of PAC is still under debate, it is suggested that adsorption, charge neutralisation, sweep flocculation and bridging do occur during coagulation. [8] had revealed that precipitate formation of PAC increased coagulation performance substantially; contrariwise, the charge neutralisation was insignificant after certain PAC dosage. Studies had proven that it is effective in removing suspended matters, dissolved organic matter, turbidity, COD, colour and humic acid.

**Table 3.** Comparison of coagulant aids in water and wastewater treatment.

Type	Chemical	Advantage	Disadvantage
Synthetic cationic polymers	Polydiallyldimethyl ammonium chloride (poly-DADMAC), polydimethyl aminomethyl polyacrylamide, polyvinylbenzyl & trimethyl ammonium chloride	Produces denser shear-resistant sludge; For the use with metallic coagulants.	Increases complexity of coagulation-flocculation stage but improves process performance and economy when used properly.
			<u>Remarks</u> Application of polyacrylamides may

Synthetic neutral polymers	Polyacrylamides & polyethylene oxide		release residual acrylamide into the environment. It is possibly genotoxic and neurotoxic carcinogenic.
Synthetic anionic polymer	Anionic polymer A110 (Kemira), hydrolysed polyacrylamides, polystyrene sulfonate, polyacrylic acid & polyacrylates	Produces larger shear-resistant floccs by promoting bridging.	Its biodegradation could take up to a few weeks' time. Therefore, polyacrylamide usage is banned or limited in several countries.
Natural polymers	Sodium alginate	Particularly suited for the use with ferric salts, but if used properly can be effective when used with alum	Less expensive, but less efficient than synthetic polymers
	<i>Moringa oleifera</i> seed extract, starch, chitosan, guar gum, tannin	Cheaper additives for increasing settling velocity and reducing coagulant dosage.	
Inorganic coagulant aids	Aluminium chloride	For the use with organic polymer coagulants	Increases complexity of coagulation-flocculation, but improves process performance and economy when used properly



**Figure 2.** Predominance of Al species at different pH values.

PAC was developed in Japan in 1960s and has been commercially applied in water and wastewater treatment in Japan, the former USSR and Europe since the '70s, followed by America in the 1980s, rivalling against traditional metallic coagulants like alum and ferric chloride. In recent years, utilisation of PAC has surged and substitutes traditional metallic coagulants gradually due to its high coagulation efficiency, rapid precipitation, less dosage, less effect on pH, lower alkalinity consumption, lesser sludge production, low cost and wider availability compared to traditional coagulants. Now, it is extensively utilised in water and wastewater treatment across numerous countries, namely Japan, France, Germany and North America.

Despite of its prominence in landfill leachate treatment, the application of PAC may introduce toxic Al residuals into the aquatic environment. This had been verified by [18] using optimum PAC-treated synthetic and surface water samples. Residuals like very toxic  $Al_{13}$  and toxic  $Al^{3+}$  are of utmost concern. In soil environment,  $Al_{13}$  can be formed at pH 3.6-5.7 with peak dominance at pH 4.1-4.8, and subsequently persist over wide pH range (pH 3.6-10.6). Other organic Al-complexes and monomeric Al species like sulphate, phosphate and silicate Al species are far less toxic. Prolonged exposure of Al may induce bioaccumulation in human brain, bones and liver [22]. In fact, low molecular weight Al species can even penetrate biological membrane, threatening any living organism.

**3. Conclusions**

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 operator. Accordingly, it is assumed that pre-hydrolysed coagulants such as PACl, PFCl, PFS and PAFCl can be considered as the superior coagulants due to better colour removal and high pH affinity towards leachate. In fact, Ferrous sulfate can also considered as a better coagulant over other metallic salts. Having said that, 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. However, until now, the applicability of the discussed coagulants are yet to be documented and extensively discussed. Further studies required to evaluate their application for landfill leachate treatment with respect to broad range of its parameters that need to be observed in following to Department of Environment permissible discharge limit.

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