Treatment for Landfill Leachate via Physicochemical Approaches: An Overview

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Leachate waste consists of various mixtures of organic, inorganic, and heavy metal contaminants, which are responsible for groundwater and surface water contamination. Landfills apply physical, chemical, and biological processes for the treatment of leachate. Most studies on leachate treatment by coagulation and flocculation are based on the selection and performance of natural based biopolymers in comparison with various inorganic metal salts and grafted polymers used for the removal of contaminants. In addition, adsorption processes utilizing non-conventional activated carbons as absorbents are the current emerging focus of the researchers in leachate treatment. These adsorbents are low-in-cost, efficient, and renewable compared to conventional adsorbents. The present paper aimed to evaluate and review the technology utilising various greener approaches in coagulation, flocculation, and adsorption as the physicochemical approaches to leachate treatment. The challenges and future work regarding the development of these green products in the commercial markets were comprehensively evaluated.

Keywords:
landfill leachate, leachate treatment, coagulation, flocculation, adsorption

Introduction

According to Kreith *et al.*, information on solid waste generation is fundamental to all aspects of solid waste management.[1] Such information will help to strategize a sustainable solution to solid waste management in the long run. Sanitary landfill is a common solid waste management method chosen to protect the environment from being polluted, and to create a hygienic living environment for the human being. Apart from the selection of a well-engineered design for a sanitary landfill, which is capable of managing the increasing amounts of solid waste, another key element for the success of the sanitary landfill is its leachate waste management. With the known quantities and types of solid waste generation in a sanitary landfill, an ultimate solution should be planned to ensure the current leachate treatment technology is sustainable. The adaptability of the new emerging technology should be viable for taking advantage of the current mature conventional methods to be further optimised in an innovative way.

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Table 1 – *Comparison of waste generation rate among Asia Pacific countries from 2009 to 2025 (Projection)*

Waste generation in South East Asia	Year 2009	Year 2011	Year 2025 (Projection)
	(kg/capita/day)		
Philippines	0.52	1.56	0.8
Myanmar	0.45	0.44	0.85
Indonesia	0.76	0.88	1.0
Cambodia	0.52	0	1.1
Laos	0.55	0.7	1.1
Brunei	0.66	0.87	1.3
Singapore	1.1	1.49	1.8
Vietnam	0.67	1.46	1.8
Malaysia	1.3	1.5	1.9
Thailand	0.64	1.76	1.95
India	0.34	0.5	0.7
Nepal	0.4	0.5	0.7
Bangladesh	0.25	0.43	0.75
Mongolia	0	0.66	0.95
Sri Lanka	0.9	0.73	1.0
Republic of Korea	1.0	1.24	1.4
China	0.8	1.02	1.7
Japan	1.1	1.7	1.7

Main components of solid waste are food, paper, and plastics, which comprise 80 % of the total weight.[2] Out of the total weight, 60 % of the solid waste is organic waste.[3] This composition suggests that high organic content will be dissolved into liquid phase as leachate, due to contact with rainwater. Household waste comprises two main categories, which are: i) recyclable items, and ii) non-recyclable items. If both categories are not segregated properly while disposing to the local licensed concessionaires, the complexity of the domestic waste categories in the treatment facilities will increase substantially. Based on the statistics obtained for the projected waste generation rate in the Asia Pacific Region by the year 2025, Malaysia is ranked second top waste generator after Thailand, whereby the waste generation rate will increase from 1.3 kg/capita/day in year 2009 to 1.9 kg/capita/day in year 2025, (projection) as shown in Table 1.[4–6]

Looking at the perspective of the overall trend, the generation of solid waste is closely related to the population not only in Malaysia,[6] but also in the other countries of the Asia Pacific Region. The rate is rapidly growing due to the increase in population growth. Waste generation rates are also influenced by the standard of living and urbanisation, degree of industrialisation, public habits, and local climate of the individual country. For instance, Philippine's waste generation rate is projected to decrease from 1.52 kg/capita/day in year 2011 to 0.8 kg/capita/day in year 2025, with almost 50 % reduction due to the socioeconomic conditions whereby the people reuse waste for a living.

In Malaysia, the government has set the 3Rs strategy (i.e., Reduce, Reuse, and Recycle) as part of the currently implemented Solid Waste and Public Cleansing Act 2007, in order to achieve a recycling rate of 22 % by year 2020, which is equivalent to a 25 % CO_2 reduction in total greenhouse gases (GHGs) release. Without segregation of waste at the source under the act enforced in September 2015, Malaysia will be foreseen as the top-ranking country in the Asia Pacific Region in 2025 with respect to its waste generation rate.[7] This was also seen in 2009, when Malaysia was compared with highly populated countries like China and Japan.[8] Prior to this mandatory enforcement, a comprehensive survey on solid waste composition, characteristics, and existing practice of solid waste recycling in Malaysia was undertaken from September 2011 to September 2012 tasked by the Ministry of Housing and Local Government (KPKT). Fig. 1 shows the statistics revealing that the waste generation rate in Malaysia, combining households (rural), commercial (urban), industrial, and institutions, amounts to 1.17 kg/capita/day on average, with the total generation of 33,130 MT/day. Generally, due to differences in the standard of living, the residents from urban areas tend to generate more waste (e.g., 1.24 kg/capita/day) than rural residents (e.g., 1.01 kg/capita/day).

The quantitative survey did not cover construction and demolition debris, sewage sludge, automobile bodies, municipal sludge, combustion ash, wastes from imports or exports, or production waste from industries being disposed of at landfills in Malaysia. Table 2 comprises the hazardous waste components. The maximum waste generation rate in 2011 was seen on par with the rate in 2009, as compared with 1.5 kg/capita/day as reported by Samsudin *et al.*[8] This reflected that the introduction of the new act brought about a positive impact on the reduction of overall waste generation rates. Therefore, the projected waste generation rate in 2025 could be improved. Subsequently, it will help the government to contribute some potential savings of 38.76 US$ per year on leachate treatment costs.[9] The operating cost of the leachate treatment system increases with high contaminant loading of the waste input. The effluent water quality might also exceed the environmental compliance discharge limits due to the constraints of the current technology, which mostly depends on the conventional biological
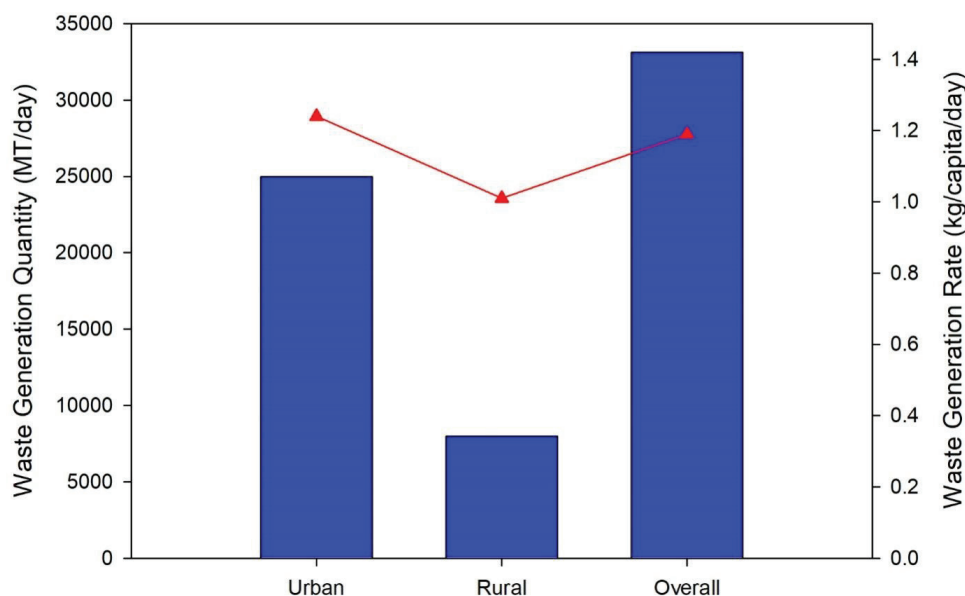
F i g . 1 – *Overview of waste generation from urban, rural, and overall areas in Malaysia (September 2011 to September 2012)*

treatment of the leachate waste coming from a vast variety of waste components contributed especially by hazardous wastes (e.g., 1.2 – 1.3 %).

Leachate waste composition consists of organic matter and heavy metals. Therefore, it is required to undergo both physicochemical and biological treatment. The selection of technology for leachate treatment depends on the leachate characteristics, which vary from one site to another. The higher complexity leachate waste requires a complicated and multi-processed technology for the leachate treatment.[10] The effluent discharge hardly complies with the effluent discharge limits set by the governing authority, with a single process designed for leach-

ate treatment.[11] This is particularly true for old and mature leachate, which contains a higher concentration of refractory compounds. Thus, a multi-stage treatment process is required, which combines physical, chemical, and biological treatment. The advantages of individual processes with their own ability to treat certain components of the leachate could thus be synergised.[11,12]

The present review aims to evaluate previous studies on the physicochemical approaches used in leachate treatment technology (i.e., coagulation, flocculation, and adsorption processes). The selection of the best available technology (BAT) option should be based on the aspects that target both the process and performance. For the process target, each element of the leachate characteristics requires different types of treatment methods. The chemical treatment was proven to be more effective in eliminating non-biodegradable organic matter and colloidal particles.[11] For the performance target, the selection of suitable materials for achieving better process performance is vital. Suitable materials for coagulation and flocculation processes for leachate treatment can be synthesized from metal salts, synthetic polymers, biopolymer or grafted polymers. The performance target is also applicable for the adsorption process, where adsorption can be done by conventional or non-conventional adsorbents.

Characteristics of leachate waste

Leachate is an aqueous material generated from the percolation of rainwater through the waste, owing to the inherent moisture, decomposition of waste, and biochemical reactions in the landfill.[13,14]

T a b l e 2 – *Components of disposed hazardous waste*

Hazardous waste	Description
Aerosol cans	Parasites repellent sprays, detergents, household chemical containers and cans
Batteries	Ni-Cd/Li-ion rechargeable batteries, small lead batteries
E-waste	Electronic components like printed circuit boards, computer parts, radio, CD/DVD players and parts
Fluorescent tubes/bulbs	Mostly spoilt and cracked bulbs
Medical waste	Cough syrup bottles, swabs, syringes (with and without needles), expired pills
Paint containers	Paint spray cans and containers
Motor service waste	Oil and air filters, plugs, oil and grease containers
Others	Asbestos sheets, fire extinguishers

Table 3 – *Properties and chemicals present in leachate as per their age (young leachate, intermediate leachate, and old leachate)*

Type of leachate	Young leachate		Intermediate leachate	Old leachate	Ref.
Period	Transition	Acid formation	Methane fermentation	Final maturation	26
Stability	Unstable		Moderately stable	Stable	27
Age	0 – 5 years	5 – 10 years	10 – 20 years	> 20 years	26,27
Phase	Acidogenic		Methanogenic	Stabilisation / Methanogenic	23,27
Colour appearance	Light yellow		–	Dark black-green	13,28
Total suspended solids	200 – 2000 mg L^{-1} (landfill age < 2 years)		100 – 400 mg L^{-1} (landfill age > 10 years)	$\geq$ 72.33 mg L^{-1}	29
Total dissolved solids	4000 – 55000 mg L^{-1}		1100 – 6400 mg L^{-1}	1460 – 4640 mg L^{-1}	26
Chemicals present in the leachate					
Sulphate (mg L^{-1})	50 – 1000 (landfill age < 2 years)		20 – 50 (landfill age > 10 years)		29,30
	70 – 1750 (landfill age < 2 years)		10 – 420 (landfill age > 10 years)		30
Chloride (mg L^{-1})	200 – 3000 mg L^{-1} (landfill age < 2 years)		100 – 400 mg L^{-1} (landfill age > 10 years)		29,31,32
Calcium (mg L^{-1})	10 – 2500 (landfill age < 2 years)		20 – 600 (landfill age > 10 years)		30
	200 – 3000 (landfill age < 2 years)		100 – 400 (landfill age > 10 years)		29
Iron (mg L^{-1})	20 – 2100 (landfill age < 2 years)		3 – 280 (landfill age > 10 years)		30
	50 – 1200 (landfill age < 2 years)		20 – 200 (landfill age > 10 years)		29
Magnesium (mg L^{-1})	50 – 1150 (landfill age < 2 years)		40 – 350 (landfill age > 10 years)		30
	50 – 1500 (landfill age < 2 years)		50 – 200 (landfill age > 10 years)		29
Manganese (mg L^{-1})	0.3 – 65 mg L^{-1}		0.03 – 45 mg L^{-1}		30
Zinc	0.1 – 120 mg L^{-1}		0.03 – 4 mg L^{-1}		
Potassium	200 – 1000 mg L^{-1} (landfill age < 2 years)		50 – 400 mg L^{-1} (landfill age > 10 years)		29
Sodium	200 – 2500 mg L^{-1} (landfill age < 2 years)		100 – 200 mg L^{-1} (landfill age > 10 years)		
Heavy metal (overall)	Low to medium		Low	Low	21,23,27,33

Untreated leachate can permeate the soil to reach groundwater or it can mix with surface water, which contribute to soil pollution, groundwater, and surface water pollution.[15] Therefore, leachate management has become a major environmental issue.[16] Leachate waste is a complex mixture of various high-strength organic and inorganic contaminants including heavy metals.[17–20] It is a mixture of different fractions of molecular weight that can be lower than 2,000 Dalton (Da), and even higher than 100,000 Da.[11] High amount of heavy metals can contaminate groundwater and surface water.

Leachate waste can be categorised as (a) dissolved organic matter; (b) inorganic macro-components; (c) heavy metals; (d) xenobiotic organic compounds.[21–24] A comparative study was carried out between a non-sanitary landfill and sanitary landfill in Malaysia, and the results revealed that the hydrogeological profile of the groundwater was very much affected by the leachate waste from the non-sanitary landfill as compared with the well-engineered sanitary landfill.[25] As the number of years of landfill operation increases, the leachate characteristics will also vary in terms of the content level of dissolved organic matter, inorganic macro-components, heavy metals, and xenobiotic organic compounds.[26] These characteristics were identified as young leachate, intermediate leachate, and old leachate, as shown in Table 3.

Leachates have moderately high strength of recalcitrant compounds and range between (medium:low) BOD (biochemical oxygen demand) to COD (chemical oxygen demand) ratio depending on the age of landfill.[14] This was categorised into four different periods, namely transition period (0 – 5 years), acid formation period (5 – 10 years), methane fermentation (10 – 20 years), and final maturation (> 20 years).[26] The content of the fatty acids in the acidogenic phase could reach as much as 80 to 95 % of the organic content, which can cause the pH of the leachate to decrease to between 4.5 and 7.5.[23,34,35] As the pH of leachate decreases, the solubility of many compounds, including heavy metals, increases.

During the methanogenic phase, the accumulated fatty acids will be consumed by methanogenic bacteria and released as methane gas and carbon dioxide.[36] This causes an increase in pH due to the consumption of fatty acids. Therefore, the pH for the intermediate leachate rises from 6.6 to 9.0.[34,37] In the stable and mature methanogenic phase for old leachate (>20 years), pH will continue to increase from 7.5 to 9.0.[27,33,38,39] However, the BOD_5 to COD ratio will decrease throughout the landfill age to <0.1 due to the consumption of volatile fatty acids.[14,27]

The colour of the old leachate is dark black-green caused by the presence of humic substances and the oxidation of ferrous iron,[14,40,41] as compared with the yellowish colour of the young leachate, which has a higher content of biodegradable material.[14] The humic substances are recalcitrant compounds and are poorly biodegradable.[14,24,27]

Heavy metals found in the leachate waste come from the disposed hazardous waste, which includes batteries, fluorescent tubes, bulbs and electronic waste, commonly disposed by the household and commercial sectors, as well as industrial and institution sectors. The hazardous waste comprises 1.2 % to 1.3 % of the total municipal solid waste.[28] Metal components (e.g., ferrous metal, aluminium, and other non-ferrous metals) are also disposed in the landfill, and may release heavy metals into the leachate. Typically, the heavy metal content is higher in young leachate than in old stabilised leachate, due to the metal solubility in the acidic environment.[27,28] As the landfill increases in age, the metal solubility decreases with the increase in pH values.[27] Heavy metals undergo oxidation-reduction reactions at alkaline conditions, reducing the dissolution of heavy metals.

Organic pollutants (OPs) in the leachate may be classified into dissolved and colloid-bound groups.[42] The various types of compounds: petroleum hydrocarbons (PHCs), alkylphenols (APs) including 4-nonylphenol (4-NP), 4-*t*-octylphenol (4-*t*-OP) and 4-*t*-butylphenol (4-*t*-BP), bis-phenol A (BPA), phthalates and polycyclic aromatic hydrocarbons (PAHs) and their degraded products, such as oxygenated PAHs (oxy-PAHs), have all been reported in the landfill leachate.[42–44] Oxy-PAHs are likely to form by the action of sunlight at high nitrogen and iron oxide concentrations under prevailing aerobic conditions.[45,46] Hydroxyl radicals, which may be generated by photochemical reactions in the pond (including photolysis of nitrate and nitrite, photo-Fenton chemistry, ligand-to-metal charge-transfer reactions or direct photolysis of coloured dissolved organic matter), could degrade PAHs.[45,47] Petroleum hydrocarbons could be degraded through physical processes, which are described in the next section.[47]

Treatment systems for leachate waste

Physicochemical treatment

Physicochemical treatment (i.e., air stripping, chemical oxidation, coagulation, flocculation, adsorption by activated carbon) are coupled mutually or combined with biological treatment.[14] Biological treatment process is more effective in treatment of young leachate in comparison to old leachate. This is due to the presence of rich organic matter in young leachate. Physicochemical treatment of old leachate is more favourable biological treatment.[24,48] Oil separator and sedimentation pond is not sufficient enough for landfill leachate; adsorption and filtration will be required for the removal of organic pollutants in the form of dissolved and colloidal material.[49,50] Therefore, tertiary treatment is an important option to further improve the quality of treated water after physicochemical and biological treatment.

Selection for the treatment process depends on the strength of the leachate.[14] Colloidal particles, dissolved solids, and organic matter are all present in the landfill leachate. These contaminants are physically tiny having a similar surface layer charge in the aqueous medium. Thus, this makes it more challenging to bring the particles closer and subsequently to form a heavier mass for settling to obtain water clarity.[51,52] The most common traditional physicochemical treatment for solid-liquid separation is coagulation and flocculation.[15,51] However, the mechanism of the removal process should be known beforehand in order to obtain a better selection for the technology in the later stage.

Characteristics and mechanism of coagulation process via chemical coagulants

Chemical coagulants can be categorised into two groups: pre-hydrolysing metallic salts, and hydrolysing metallic salts.[53] Pre-hydrolysing metallic salts are polyaluminum chloride, polyferric chloride, polyferrous sulphate, polyaluminum ferric chloride, and polyaluminum sulphate. These are pre-neutralised coagulants as compared with the hydrolysing metallic salts, which need higher dosage of pH correction chemicals. Among the hydrolysing metallic salts are aluminium sulphate, ferric chloride, ferrous sulphate, and magnesium chloride.

Generally, small dispersed particles will agglomerate together to form microflocs, and if the coagulant is added, the flocs will settle to the bottom.[51,54] In most cases, the colloidal and suspended particles carry negative charges in the aqueous medium. Once the inorganic coagulant is added, the hydrolysis process takes place, and the metal salts will rapidly hydrolyse to form cation at the isoelec-

tric point. The negative surface charge of the colloidal particles will be reduced simultaneously and neutralised to form microflocs.[51,55] The microflocs formed are fragile and easily breakable when subjected to physical forces accidently. The sedimentation of microflocs is slow. Subsequently, the clarity of water at the effluent discharge will also be affected as the microflocs might be carried off with their low strength of agglomeration.[56]

Characteristics and mechanism of flocculation process via organic synthetic polymers

Flocculants may be derived from organic synthetic polymers. Originally, organic flocculants are linear soluble polymers extracted from oil-based and non-renewable materials with acrylamide and acrylic acid based repeated monomer units.[51,55] Since polymers vary in molecular weight, charge density, type of charge, composition, and structural framework (linear or branched), organic polymer-based flocculants can be classified into groups known as cationic, anionic, amphoteric (contain both cationic and anionic charge), and non-ionic (neutral). Flocculants act as coagulation promoters that could facilitate the agglomeration process to bring the microflocs together into larger and denser flocs. There are three functions of flocculants, which are: charge neutralisation, polymer bridging, and electrostatic patch function. A high molecular weight cationic or anionic polymer with medium charge density can neutralise the opposite charges of the colloidal and suspended solids. In most practical cases, the colloidal particles are negatively charged. Therefore, the cationic polymers are preferable in combination with inorganic coagulants (e.g., metal salts) for reducing the surface charge in terms of zeta potential to values close to zero (i.e., to the isoelectric point). As a result, the electrical repulsion forces between the colloidal particles are weakened. This will allow the van der Waals forces of attraction to aggregate the colloidal particles and other suspended materials to form microflocs.[51] However, a reverse effect will happen if the polymer is overdosed. This causes the microflocs to be dispersed with positive charge instead of negative charge.[57]

Apart from the surface charge neutralisation, the flocculants can also bridge the agglomerated and charge-destabilised colloidal particles together to form bigger and denser flocs.[58] The macroflocs are more resistant to breakage under elevated shear impacts. Flocculants use the mechanism of loops and tails to bridge the colloidal particles into a dense and closely packed form.[59] This is necessary in case of fragile and loose microflocs which have underwent charge neutralisation only, causing the slow settling of microflocs and affecting the water

clarity at the final discharge.[57] Therefore, long-chain polymers with high or medium molecular weight (up to 1,000,000 Da) are used to extend their loops and tails beyond the electrical double layer of the destabilised colloidal particles. In this case, the charge density is not determined as an important factor in affecting the polymer bridging.[60] The examples of the low charge density polymers with high and medium molecular weight are cationic polyacrylamide (C-PAM), and cationic copolymers of acrylamide and diallyldimethylammonium chloride, respectively. If the polymer chains are shorter in terms of low molecular weight, the loops and tails of the polymer could not reach out from one colloidal particle surface to another particle.[51,61,62] The polymer bridging effect will be reduced.[51,63] The polymer may also function as an electrostatic patch. In some cases, when the colloidal particles with low negative charge are adsorbed by the high charge density cationic polymers with low molecular weight, the surface charge of the colloidal particles is not fully neutralised.[61] Cationic patches are formed on the particles, which still have negatively charged surface sites. In this case, flocculation occurs when the cationic patched particles are attracted and adsorbed by the particles with the uncoated negatively charged area. Though the strength of the macroflocs formed in this manner is not as high as the macroflocs formed via polymer bridging, the tendency of the flocs breakage is much lower than with the flocs formed by simple charge neutralisation and coagulation in the presence of metal salts. However, the charge density of the polymers should be high enough for the electrostatic patches to act efficiently. Otherwise, polymer bridging would be a better way to form macroflocs.[64]

Biopolymers, their characteristics, and mechanism of the flocculation process

Biopolymers are environmentally friendly flocculants that have recently emerged as an alternative solution in treating water and wastewater. Wastewater treatment by natural polymers or biopolymers has increased in recent years.[65] Generally, biopolymers are grouped into three categories: plant-based biopolymers, animal-based biopolymers and microorganism-based biopolymers.[53] Researchers have studied many types of biopolymers, such as chitin and chitosan, vegetable tannin, *Cassia javahikai* and *Ipomoea dasysperma* seed gum, *Strychnos potatorum, Moringa oilifera* seed extract, okra gum, xanthan gum (microorganism based), and guar gum (used in potable water treatment and in food processing industries).[66–70] Guar gum is also a sizing additive in the paper industry.[64]

Cellulose exists naturally as an abundant polysaccharide. Recently, it has become the subject of

studies and undergone physical and chemical structure modification to enhance its ability to act as a biopolymer.[71] Anionic sodium carboxymethylcellulose (CMCNa) and anionised dicarboxylic acid nanocellulose (DCC) are among the modified starches tested in drinking water treatment and municipal wastewater treatment, respectively.[55,72]

Tannins are biodegradable anionic polymers, secondary plant metabolites that can be extracted from vegetal tissues like bark, fruits, and leaves.[73,74] Various studies have shown that tannins could be used as coagulation promoters to agglomerate the destabilised colloidal particles into macroflocs through polymer bridging.[74–76] Due to its anionic nature, which has restricted its application for charge neutralisation, the tannin extracted from *Acacia mearnsii* bark was modified through a physicochemical process by adding hydrocolloid gums and soluble salts to convert the tannin into cationic form. This is due to the inclusion of quaternary nitrogen into the modified tannin to be used as a flocculant.[74]

Chitosan is an animal-based biopolymer obtained by the alkaline deacetylation of chitin found in shellfish. It is of cationic nature due to the linear and rigid structure of hydrophilic amino polysaccharides containing glucosamine and acetylglucosamine units. It is insoluble in water and organic solvents, but soluble in the presence of dilute organic acids such as acetic acid and inorganic acids, with the exception of sulphuric acid. The free amino groups in the hydrophilic part of chitosan molecules are protonated and become soluble cationic biopolymers with high charge density at acidic pH below 5.0.[77–79] This facilitates the charge neutralisation via protonated amino groups, and polymer bridging via long polymer chains with high molecular weight. Therefore, chitosan has two molecular functionalities to coagulate and flocculate the negatively charged colloidal particles into bigger flocs to be settled.

Xanthan gum is a non-toxic, biodegradable, and widely available polysaccharide secreted from bacterium *Xanthomonas campestris*.[64] The benefits of this biopolymer are its wide availability, strongly hydrophilic nature, and production from a renewable resource.[79] The sludge produced after the coagulation process can be degraded efficiently by microorganisms.[52,76] Therefore, secondary pollution due to the excess of biopolymer could be avoided.[52,63]

Anionic biopolymers such as cellulose, tannin, and sodium alginate, have carboxyl and hydroxyl groups in their macromolecular structures. These functional groups could interact with the colloidal particles by adsorbing counter ions to neutralise the surface charge of the particles. Eventually, biopolymers reduce the zeta potential by decreasing the thickness of the diffuse part of the electrical double layer and destabilising the colloidal particles. However, they are unable to neutralise negatively charged colloidal particles or anionic contaminants. Therefore, the addition of metal salts or polymers with cationic charge is required prior to the promotion of polymer bridging by the anionic biopolymer (carboxyl and hydroxyl groups).[55,69,72,76] Unlike anionic biopolymers, chitosan is a high molecular weight polymer with cationic behaviour due to its amino and hydroxyl groups. It could be applied in the charge neutralisation as well as polymer bridging. The mechanism was also examined by Guibal and Rousy, where the charge neutralisation was performed by the protonated amine groups from chitosan, and the flocculation was enhanced by the long chain bridging mechanism to agglomerate and settle down the charge-destabilised colloidal particles.[65] The above described phenomena were confirmed by Szygula *et al.*; the flocculation mechanism of chitosan involves hydrophobic interactions and chain association through its hydrogen bridges.[78]

Characteristics and mechanism of flocculation process via grafted polymers

Grafted polymers are modified polymers obtained by grafting synthetic polymers onto the backbone of natural polymers. This grafting mechanism combines the best attributes of both polymers for further enhancement of the active component and cationic charge of the polymers.[80] Many grafted polymers have been synthesized, such as poly(methyl methacrylate) grafted psyllium (Psy-*g*-PMMA), poly(acrylamide) grafted starch (St-*g*-PAM), poly(acrylamide) grafted carboxymethyl guar gum (CMG-*g*-PAM), hydroxypropyl methyl cellulose grafted with poly(acrylamide) (HPMC-*g*-PAM) and poly(2-hydroxyethyl methacrylate) grafted agar.[81,82] It was found that high molecular weight grafted polymers have branched structures with fewer and longer dangling polymer chains.[52,63,83] Due to alteration of the regular structure of natural biopolymers with the insertion of organic synthetic polymers, the biodegradability of the grafted polymers reduces substantially, and the polymer becomes more stable with respect to shear induced degradation.[52,83] Furthermore, grafted polymers could be "tailor-made" by adjusting the properties of polysaccharides to treat the different types of surface charge of the colloidal particles. In principle, cationic grafted polymers deal effectively with negatively charged colloidal particles. Similarly, amphoteric grafted polymers would be preferable for colloidal particles containing both cationic and anionic charges.[84–86]

Charge neutralisation and polymer bridging are the main flocculation mechanisms for grafted polymers and copolymers.[82,84,86–88] Firstly, the surface

charge of the colloidal particles is neutralised rapidly to produce insoluble complexes. The insoluble complexes are then aggregated by the flexible polymeric graft chains through the bridging effect. Finally, the closely packed macroflocs form and settle down rapidly. All grafted polymers have a similar mechanism regardless of their (amphoteric, cationic or anionic) nature. This is due to their extended configuration with respect to the linear polymers. The longer chains and higher radii of gyration of graft polymers enable adsorption of their segments onto the surfaces of different particles. Therefore, interaction with more than one particle is possible prior to the formation of bridges.

Factors affecting coagulation and flocculation

Coagulants are pH dependent and the precipitation of metal hydroxides is only obtained at the desired pH after the addition of metal salts. The destabilising effect is stronger for counter ions of higher valence, and therefore the dosage of coagulants should be reduced. Unlike coagulants, cationic or anionic polymers can work in a wide range of pH values, from acidic, neutral, to alkaline media.

Zeta potential is another key factor determining the application of coagulants and flocculants.[53] If the zeta potential is below the isoelectric point of the colloidal particles, cationic polymers will be preferred, and adsorption of these polymer chains onto the negatively charged colloidal surface will occur for charge neutralisation as well as bridge formation. If the zeta potential is above the isoelectric point for colloidal particles, anionic polymers will work better and adsorption of these polymer chains onto the positively charged colloidal surface will occur for charge neutralisation as well as bridge formation. For polymer bridging, the effectiveness of agglomeration of the colloidal particles to form macroflocs depends on the dosage of the polymers. The dosage should not be too high, as it could cause charge reversal. The particle surfaces may become overly coated with polymers, which would reduce the bridging efficiency as well, as there would be less available sites on the particle surface to bridge with other particles.[89,90] In general, molecular weight and charge density are two important factors to influence and determine the acting mechanism.

Sludge volume index (SVI) is a parameter used to study the settling characteristics of the flocs. It helps to establish the relationship between the settling velocity and amount of suspended solids. The removal of suspended particulate matter depends on the physicochemical reaction between active groups of the flocculants and the waste particles. Other parameters to be studied are dosage, pH, and mixing speed to obtain effective flocculation.[64]

Applications of coagulation and flocculation

Coagulation and flocculation processes utilise the principle of charge neutralisation and polymer bridging to remove the colloidal and dissolved particles in the leachate. However, not all particles can be removed effectively by coagulation and flocculation alone. Ferric chloride is mostly chosen as a coagulant due to its good performance in treating leachate containing humic substances, especially stabilised leachate.[90] When ferric chloride is used, it can form complexes with organic matter. Optimum pH for coagulation by ferric chloride is around 4.5 for the concentration of 7.0 mmol Fe^{3+} L^{-1}.[91,92]

Upon increasing the dosage of coagulants, the removal rate for the dissolved organic carbon, COD, and total suspended solids increases as well. For the sake of comparison, ferric chloride was tested both on young leachate and on old leachate. Dissolved organic carbon and COD removal efficiencies of 10 % to 25 %, and 50 % to 60 % for young and old leachate were obtained, respectively.[92,93]

Studies have shown that the dissolved organic matter in leachate consists of organic compounds ranging widely in their molecular weight and including volatile fatty acids, humic acids, and fulvic acids in high concentrations. Upon 60 % removal of dissolved organic carbon, the colour of leachate changed from dark-brown to brownish yellow, which is the characteristic of fulvic acids.[10,92] High molecular weight humic acids were likely precipitated and eliminated during the coagulation process. This was also in agreement with the study carried out by Wu *et al.*, where the removal efficiencies of humic acids, fulvic acids, and hydrophilic fraction of organics of 71 %, 53 %, and 37 % were achieved, respectively, through the coagulation step.[94] This is related to the charge neutralisation of the functional groups of the humic acids reducing their solubility and followed by the adsorption and precipitation of the destabilised negatively charged humic acids.[91,92] However, the disadvantage of the dosage increase is the generation of higher amounts of acid sludge that need to be properly disposed. This is caused by the coagulation process that only promotes the agglomeration of organic matters from the dissolved state to the particulate state.

In the case of coagulation and flocculation using ferrous sulphate as metal salt and polyacrylamide as organic synthetic polymer, the dissolved organic matter with molecular weight higher than 4,000 Da, has a higher removal rate than that with molecular weight lower than 4,000 Da.[11] Another study conducted by Trebouet *et al.*, reported that the majority of COD can be attributed to components with molecular weight lower than 500 Da after coagulation by ferric chloride as metal salt.[95] This

confirms the findings of Yoon *et al.*, that components with molecular weight higher than 500 Da were much easier to remove from leachate than components with molecular weight lower than 500 Da.[96] In general, larger molecular weight fraction of leachate is more easily removed via coagulation and flocculation process.

UV-Vis spectra and FTIR have been widely used to identify the structure of dissolved organic matter qualitatively and quantitatively.[11,15] From the analysis of UV-Vis spectra of dissolved organic matter, the absorption by double bonds and polycyclic aromatic structures characteristic for humic acids and fulvic acids, decreased after the coagulation and flocculation process. It showed that the refractory components could be partly removed through charge neutralisation and polymer bridging process.[11]

Based on the zeta potential of leachate, most of the colloidal particles are negatively charged. The presence of colloidal particles causes the turbid appearance of leachate. Application of cationic metal salts or cationic polymers will form electrostatic attraction with the surface charge of the colloidal particles to form flocs. Most of the published papers reported turbidity reduction in the range of 70–99 %.[95–98] With the removal of organic matter from the leachate, the turbidity also improved by 87 %, as shown in Table 4(a)(b).

Regarding the anions, a high percentage of NO_2^-, NO_3^- and SO_4^{2-} ions removal of more than 90 % was obtained by using ferric chloride. The results show that it is suitable to combine coagulation with biological treatment to achieve denitrification. Diverse results were obtained with the application of ferric chloride for the removal of heavy metals, depending on their types. More than 90 % of Ag, Bi, Cr, Fe, P, Pt and Sn was removed, while the removal of Al, As, Cd, Si, Ti, V, W, and Zn was in the range of 40 %–85 %.[93]

Ammoniacal nitrogen, (NH_3-N) with relatively high concentrations in the stabilised or old leachate was not removed significantly by the coagulation and flocculation process. Coagulation with ferric chloride does not remove NH_3-N[93] directly, which results in its low removal (11.5 %), as reported by Liu *et al.*[97] However, in another study carried out by Syafalni *et al.*, the NH_3-N removal using alum and lateritic soil was higher, within the range of 40–50 %.[103] This could be explained by the predominance of ammonium ion (NH_4^+) under acidic conditions. Ammonium ion could be predominantly protonated under optimal pH of 5 – 7 to adsorb electrostatically onto the colloidal particles with negative surface charge, which will then settle down.

Magnesium ammonium phosphate (MAP) is commonly applied as the final step in the removal of NH_4^+ after the biological process both for medium-age and young-age landfill leachate. Magnesium chloride and sodium hydrogen phosphate are two chemicals commonly used in MAP process.[111,112] MAP treatment mechanism involves the adsorption of organics on the surface of the crystal, and is subject to the physical properties of the crystals formed. MAP can eliminate the ammonium ion effectively.[111,112] The advantage of MAP sludge is in the fact that it can be used as a fertilizer due to the high content of soluble magnesium, phosphorus, and ammonium.[110,111,113] The disadvantage is the cost of the chemicals used for the treatment, although this could be compensated with the value of the generated sludge. In the application of MAP, it is important to assess pH and the molar ratio of magnesium, ammonium, and phosphate ions. Upon addition of the chemicals, crystals form rapidly and settle to the bottom of the container accompanied with a drop in pH from 9.2 to 5.9. Around 50 % of NH_4^+ can be removed without pH adjustment. With pH adjusted to 8–10, the crystals are much more visible due to the insolubility of MAP under alkaline conditions. Optimal pH for maximum ammonium ion removal at molar ratio of $Mg:NH_4:PO_4 = 1:1:1$ is shown in Table 5. In studies of Akkaya *et al.* and Ozturk *et al.*, 50 % removal of COD for the anaerobically treated leachate was achieved.[111,114] A maximum removal of NH_4^+ of 96.6 % was obtained at pH = 9.0. The results of the above studies are summarised in Table 6.

Three types of organic biopolymer coagulants (i.e., guar gum, locust bean gum, and xanthan gum) have been studied for their performance in the leachate treatment.[64] A comparison was made among the mentioned organic coagulants, as well as with alum as the selected inorganic coagulant for the effectiveness of the effluent treatment. The advantage of the application of organic coagulants was production of denser flocs. SVI increased with the increase in biopolymer dose. More compact sludge is easier to handle for disposal.[64]

In terms of floc size, guar gum has the largest floc size within 500 – 2,000 μm followed by alum, locust bean gum, and xanthan gum. As compared with guar gum, floc appearance for alum is less compact and its distribution is not uniform. This is due to the influence of pH. At high pH, zeta-potential of the alum-treated effluent is –19.3 mV, which causes re-stabilisation of the colloid and consequently less floc formation. At lower pH values, (pH 5.0 and 7.0), zeta potential is 0.557 mV and –0.627 mV, respectively. This indicates that the destabilisation and neutralisation of colloid took place for improved floc formation. Unlike alum, guar gum has a wide range of working pH, which does not affect its efficiency. Floc formation using xan-

Table 4(a) – *Removal percentages for leachate using coagulation and flocculation process*

pH	Coagulant	Dosage (mg L^{-1})	Flocculant	Dosage (mg L^{-1})	Removal percentage (%)	Reference
			COD removal			
3.8	FeCl$_3$	400	–	–	28.1	[98]
4.0	FeCl$_3$	5,000	polyacrylamide	70	82	[99]
4.2	FeCl$_3$	240	–	–	58	[92]
4.5	FeCl$_3$	223	–	–	54	[91,92]
5.0	FeCl$_3$	970	polyacrylamide grafted gum ghatti	100	79	[100]
5.5	FeCl$_3$	600	–	–	68	[101]
6.0	FeCl$_3$	1,400	–	–	32.5	[99]
7.9	FeCl$_3$	1,400	–	–	66	[102]
7.94	FeCl$_3$	1,000	–	–	65.7	[93]
4.8	Al$_2$(SO$_4$)$_3$·16H$_2$O	10,000	–	–	85.4	[103]
6.0	Al$_2$(SO$_4$)$_3$·17H$_2$O	800	–	–	27.1	[98]
6.0	Al$_2$(SO$_4$)$_3$	600	–	–	53	[101]
6.5	Al$_2$(SO$_4$)$_3$·18H$_2$O	11,000	–	–	58	[104]
6.5	Al$_2$(SO$_4$)$_3$·18H$_2$O Psyllium husk	10,000 400	–	–	63	[104]
7.0	Alum	250	–	–	36	[105]
7.0	Alum *O. basilicum*	5,000 (Ratio 1:1)	–	–	64.4	[106]
5.0	Fe$_2$(SO$_4$)$_3$	800	–	–	36	[107]
–	Fe$_2$(SO$_4$)$_3$ KMnO$_4$	3,000 1,000	polyacrylamide	10	60.2	[99]
–	Fe$_2$(SO$_4$)$_3$ Ca(OH)$_2$	3,000 10,000	polyacrylamide	10	60.2	[99]
12.8	Ca(OH)$_2$	18,000	polyacrylamide	1,000	41	[99]
5.0	poly ferric sulphate	1,200	–	–	65.3	[9]
5.5	poly ferric sulphate	300	–	–	70	[101]
6.0	poly ferric sulphate	1,200	–	–	62.8	[108]
5.5	polyaluminum chloride	600	–	–	61	[101]
7.5	polyaluminum chloride	7,200	–	–	55	[104]
7.5	polyaluminum chloride *Psyllium husk*	7,200 400	–	–	64 17	[104]
7.9	polyaluminum chloride	1,400	–	–	44	[102]
8.0	polyaluminum chloride	6,000	–	–	36.8	[98]
2.0	Lateritic soil	14,000	–	–	65.7	[103]
			Organic matter			
4.2	FeCl$_3$	240	–	–	60	[92]
7.94	FeCl$_3$	1,000	–	–	86.6	[93]
			Ammoniacal nitrogen			
7.94	FeCl$_3$	1,000	–	–	15.2	[93]
4.8	Al$_2$(SO$_4$)$_3$·16H$_2$O	10,000	–	–	47.6	[103]
6.0	Al$_2$(SO$_4$)$_3$·18H$_2$O	4,000	–	–	25	[109]
6.0	Al$_2$(SO$_4$)$_3$·18H$_2$O *Hibiscus rosa-sinensis*	4,000 500	–	–	28	[109]
5.0	poly ferric sulphate	1,200	–	–	26.9	[9]
2.0	Lateritic soil	14,000	–	–	41.2	[103]
10.0	*Hibiscus rosa-sinensis*	2,000	–	–	54	[109]
			Turbidity			
3.8	FeCl$_3$	400	–	–	90.2	[98]

T a b l e 4 (b) – *Cont.*

pH	Coagulant	Dosage (mg L^{-1})	Flocculant	Dosage (mg L^{-1})	Removal percentage (%)	Reference
5.0	FeCl$_3$	970	polyacrylamide grafted gum ghatti	100	93	[100]
5.5	FeCl$_3$	600	–	–	97	[101]
7.9	FeCl$_3$	500	–	–	90.3	[102]
7.94	FeCl$_3$	1,000	–	–	87	[93]
6.0	Al$_2$(SO$_4$)$_3$·17H$_2$O	800	–	–	93.2	[98]
6.0	Al$_2$(SO$_4$)$_3$	600	–	–	95	[101]
6.0	Al$_2$(SO$_4$)$_3$·18H$_2$O	4,000	–	–	36	[109]
6.0	Al$_2$(SO$_4$)$_3$·18H$_2$O *Hibiscus rosa-sinensis*	4,000 500	–	–	60	[109]
5.5	poly ferric sulphate	300	–	–	96	[101]
6.0	poly ferric sulphate	1,200	–	–	62.8	[110]
5.5	polyaluminum chloride	600	–	–	99	[101]
7.9	polyaluminum chloride	4,000	–	–	97.7	[102]
8.0	polyaluminum chloride	4,000	–	–	97.7	[98]
			Colour			
3.8	FeCl$_3$	400	–	–	78.4	[98]
5.0	FeCl$_3$	970	polyacrylamide grafted gum ghatti	100	60	[100]
4.8	Al$_2$(SO$_4$)$_3$·16H$_2$O	10,000	–	–	96.4	[103]
6.0	Al$_2$(SO$_4$)$_3$·17H$_2$O	800	–	–	84.3	[98]
6.0	Al$_2$(SO$_4$)$_3$·18H$_2$O	7,000	–	–	69	[109]
6.0	Al$_2$(SO$_4$)$_3$·18H$_2$O *Hibiscus rosa-sinensis*	4,000 500	–	–	61	[109]
6.5	Al$_2$(SO$_4$)$_3$·18H$_2$O	11,000	–	–	79	[104]
6.5	Al$_2$(SO$_4$)$_3$·18H$_2$O *Psyllium husk*	10,000 400	–	–	82	[104]
7.0	Alum *O. basilicum*	5,000 (Ratio 1:1)	–	–	77.8	[106]
7.5	polyaluminum chloride	7,200	–	–	80	[104]
8.0	polyaluminum chloride	4,000	–	–	91.2	[98]
2.0	Lateritic soil	14,000	–	–	81.8	[103]
–	*Psyllium husk*	400	–	–	27	[104]
			Total suspended solids			
7.5	polyaluminum chloride *Psyllium husk*	7,200 400	–	–	90	[104]
6.0	Al$_2$(SO$_4$)$_3$	600	–	–	87	[101]
6.0	Al$_2$(SO$_4$)$_3$·18H$_2$O	4,000	–	–	45	[109]
6.0	Al$_2$(SO$_4$)$_3$·18H$_2$O *Hibiscus rosa-sinensis*	4,000 500	–	–	72	[109]
6.5	Al$_2$(SO$_4$)$_3$·18H$_2$O	11,000	–	–	78	[104]
6.5	Al$_2$(SO$_4$)$_3$·18H$_2$O *Psyllium husk*	10,000 400	–	–	81	[104]
7.0	Alum	250	–	–	46	[105]
5.5	poly ferric sulphate	300	–	–	93	[101]
5.5	polyaluminum chloride	600	–	–	91	[101]
7.5	polyaluminum chloride	7,200	–	–	91	[104]
7.5	polyaluminum chloride *Psyllium husk*	7,200 400	–	–	96	[104]
–	*Psyllium husk*	400	–	–	41	[104]
			Anions			
7.94	FeCl$_3$	1,000	–	–	>90 (NO$_2^-$, NO$_3^-$, SO$_4^{2-}$)	[93]
7.94	FeCl$_3$	1,000	–	–	68.2 (Br$^-$)	[93]
7.94	FeCl$_3$	1,000	–	–	64.7 (Cl$^-$)	[93]
7.94	FeCl$_3$	1,000	–	–	4.17 (F$^-$)	[93]
			Heavy metals			
6.0	Al$_2$(SO$_4$)$_3$·18H$_2$O	4,000	–	–	60 (Fe^{3+})	[109]
6.0	Al$_2$(SO$_4$)$_3$·18H$_2$O *Hibiscus rosa-sinensis*	4,000 500	–	–	100 (Fe^{3+})	[109]
7.94	FeCl$_3$	1,000	–	–	>90 (Ag, Bi, Cr, Fe, P, Pt and Sn)	[93]
7.94	FeCl$_3$	1,000	–	–	40 % – 85 % (Al, As, Cd, Si, Ti, V, W and Zn)	[93]

Table 5 – *Optimal pH for maximum ammonium ion removal at molar ratio of Mg:NH$_4$:PO$_4$ (1:1:1)*

pH range	pH at maximum NH$_4^+$ removal	Reference
8.5 – 11.0	9.5	[1]
–	8.5	[101,111,114–116]
–	9.2	[2]
–	8.5–9.0	[3]

than gum occurs effectively at pH 7.0 and pH 12.0. On the other hand, locust bean performs best at lower pH, and the efficiency decreases in alkaline effluents. Therefore, locust bean is more suitable for acidic effluents. Unlike inorganic coagulants, which destabilise the colloidal particles by charge neutralisation to form flocs, organic coagulants do not affect the charge but act by polymer bridging.[117] If the dosage applied to the effluent is above the optimal concentration, repulsive interactions can be developed between the coagulants and suspended particulate matter (SPM), which results in the re-dispersion of aggregated particles.[64] Therefore, it is important to determine the optimum concentration for the floc settling. Based on the experimental results obtained by Mukherjee *et al.*, the amount of guar gum applied to achieve high turbidity removal (>90 %) is much lower than that of inorganic coagulant.[64] Among the biopolymers, guar gum and locust bean gum showed the highest removal rate, followed by xanthan gum. Nevertheless, alum has the highest fraction as compared with biopolymers. In terms of settling velocity, alum is ranked first, followed by guar gum, xanthan gum, and locust bean. Nitrogenous groups could be removed from the effluent and detected in the flocs of guar gum, as proven by the presence of C=N stretches in FTIR

spectra. The spectra reveal the removal of halide groups as well.[117]

Lateritic soil is a natural coagulant rich in aluminium and iron ions. Both cations can destabilise the negatively charged colloidal particles in the leachate achieving COD removal of 65.7 %.[117] Another natural coagulant extracted from the leaf of *Hibiscus rosa-sinensis* was also studied; it is a high molecular weight anionic polymer. Better removal efficiency was achieved when combining it with the inorganic coagulant.[109]

Leachate treatment by adsorption

Conventional and non-conventional adsorbents used in the treatment processes

By definition, adsorption is a process of attaching components from gas or liquid mixtures to the surface of a solid absorbent via physical or chemical bonds.[118] The criteria for selecting adsorbents or filters are their affinity for the targeted pollutant, adsorption capacity, and hydraulic properties.[119] Sand filter is inefficient to improve water quality to meet the environmental water standards. Sand filter provides pre-treatment and removal of precipitated iron that would otherwise lower the removal efficiency for PAHs and PHCs in peat and granular activated carbon (GAC) filters.[119] *Sphagnum* peat is a low-cost adsorbent. Oxygen-containing functional groups in the peat such as carboxyl, hydroxyl, and carbonyl groups provide adsorption function for metal and polar compounds, while non-polar groups such as waxes and methyl groups adsorb the hydrophobic organic molecules.[119] Previous studies have utilised the peat for removal of heavy metals, oils, pesticides, phosphorous, and nitrogen.[120–122]

Table 6 – *Removal rate of the effluent parameters for MAP application*

MAP application	Removal rate (%) 4.	Removal rate (%)		
		Molar ratio of Mg:NH$_4$:PO$_4$		
5.	6.	[1:1:1]	[1:1.2:1.2]	[1:1.4:1.4]
COD	50	–	–	25.7
Turbidity	–	–	–	36.4
Colour	–	18.5	–	22.5
Sludge volume generation	–	15.6	–	38.2
Ammonium ion	–	50 % (with no adjustment pH dropped to 5.9)	–	–
		86.6 % (with pH adjustment to 8.0)	–	–
		93.9 % (with pH adjustment to 9.0)	96.6 % (with pH adjustment to 9.0)	–
Ref.	[64,111]		[111]	

Table 7 – *List of potential sources for non-conventional activated carbon adsorbents*

Agricultural waste	Industrial by-products	Food waste
Corncob		Bone meal
Oil palm fibre		Banana frond
Palm shell	Iron fines	Coffee ground
Sugarcane bagasse	Rattan sawdust	Durian peel
Rice husk carbon composite		Tamarind fruit seed
		Orange peel

GAC has shown a good capacity for adsorption of dissolved and colloidal compounds. This was due to the presence of a large and non-polar surface. GAC is efficient for non-polar pollutant adsorption. In the presence of carbon-oxygen surface groups, polar compounds and metal cations can be adsorbed.[119] Recently, non-conventional adsorbents have been developed for the leachate treatment because large volumes of landfill waste are generated. Some of the non-conventional adsorbents are activated carbons produced from bamboo dust, bark husk, chitin, coir pith, lignite, maize cob, palm shell, palm fibre, palm stone, peat, pinewood, rice husk, sago waste, sawdust, sugarcane bagasse, and tea leaves.[123–125] Table 7 and Table 8 provide a list of some of non-conventional adsorbents and their characteristics. These materials can be chemically modified and are easily sourced at lower cost than conventional activated carbon.[126]

Mechanism of the adsorption process

In the adsorption process, the most crucial properties for good performance of an adsorbent are the surface area and porosity. Those features could be developed during the activation process and determined by the impregnation ratio.[126] An increase in impregnation ratio could widen the pore size of the adsorbents from micropores to mesopores via catalytic oxidation. The common types of catalysts used are potassium hydroxide, zinc chloride, and phosphoric acid.[131–135] Absorbents with high surface area are suitable for adsorbing small molecules.[123] This vital property corresponds to its adsorptive capacity.[128]

The participation of the functional groups for pollutant binding in the leachate could be determined bz FTIR analysis. Important functional groups which should be present on the surface of absorbents are carboxyl, carbonyl, hydroxyl, and

Table 8 – *Characteristics of the non-conventional activated carbons*

| Characteristics	Before activation		After activation (Activated carbon)				
	[1]Char	[2]Char	Tamarind fruit seed	Palm shell	Banana frond	Sugarcane bagasse	[3]Sugarcane bagasse
BET surface area ($m^2\ g^{-1}$)	28.92	6.388	1,090.01	–	847.66	1,620.69	99.949
Micropore surface area ($m^2\ g^{-1}$)	6.43	0.00158	511.78	–	313.91	659.25	0.0231
External surface area ($m^2\ g^{-1}$)	22.49	10.0164	578.23	–	533.75	961.44	43.514
Langmuir surface area ($m^2\ g^{-1}$)	44.61	7.688	1,616.84	–	1297.40	2,430.16	111.786
Surface area ($m^2\ g^{-1}$)	–	–	–	595	–	–	–
Pore size (nm)	–	–	1.5 – 3.5	–	2 – 4.5	2 – 4	–
Average pore size (nm)	2.606	–	2.178	–	3.417	2.417	–
Total pore volume ($cm^3\ g^{-1}$)	0.020	–	0.594	0.36	0.726	0.979	–
Micropore volume ($cm^3\ g^{-1}$)	0.004	–	0.281	–	0.169	0.515	–
Mesopore volume ($cm^3\ g^{-1}$)	0.016	–	0.313	–	0.557	0.464	–
Fourier Transform Infrared Spectrometer (FTIR)	–	–	–OH (3,842–3,568 cm^{-1}) –COOH (2,287 cm^{-1}) –CO (1,054 cm^{-1}) 7.	–	–OH (3,234 cm^{-1}) –COOH (2,364–2,348 cm^{-1}) –CO (1,053 cm^{-1})	–COOH (2,038 cm^{-1}) –CO (1,053 cm^{-1}) 8.	–OH (hydroxyl) 3,434 cm^{-1} 9.
Reference	[127]	[128]	[129]	[130]	[127]	[127]	[128]

Remarks:

[1] 700 °C carbonisation temperature under purified (N_2) flow,

[2] 700 °C carbonisation temperature under purified nitrogen (N_2) flow at 150 $cm^3\ min^{-1}$ with ramping temperature at 10 °C min^{-1},

[3] 687 °C activation temperature, 2 h activation time, and 2.59 KOH/char impregnation ratio

lactonised carboxyl.[123] The adsorption occurring on the surface of the adsorbent is often attributed to two proposed mechanisms, namely, sorption via ion exchange according to Reaction 1 or complex formation with negatively charged sites of the absorbents according to Reaction 2, especially for the activated carbon.[118,136,137]

$$xCOH + M^{y+} \rightarrow (CO)_x M^{(y-x)+} + xH^+ \qquad (1)$$

$$xCO^- + M^{y+} \rightarrow (CO)_x M^{(y-x)+} \qquad (2)$$
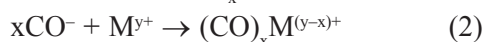
Based on Reaction 1, ion exchange occurs between the activated carbon and any types of contaminants. The release of ion H^+ causes a decrease in pH and the sorption will cease with the depletion of ion H^+.[118] Another dominating mechanism is shown by Reaction 2, where the specific chemical absorption energy prevails over the electrostatic energy.[138] However, there are other possible mechanisms not pertaining to those proposed above.

Although not important for the granular activated carbon, the mechanism of ion exchange is still important for other types of absorbents, such as bone meal and iron fines. Ion exchange of metals in leachate with Ca^{2+} ion on the poorly crystalline hydroxyapatite, $(Ca_{10}(PO_4)_6(OH)_2)$ found in the inorganic constituent of bone meal, is the main mechanism responsible for metal removal.[139] It occurs through the dissolution of Ca^{2+} ion from hydroxyapatite and its replacement by metal ions form leachate. The sorption of heavy metal composed of phosphate minerals occurs due to the formation of complexes with the organic matter present in the bone meal.[139] This shows that bone meal is a positively charged absorbent, and produces no electrostatic repulsion with phosphate minerals, as found in the negatively charged absorbents.[139] The mechanism of iron fines has yet to be understood clearly regarding its ion exchange with metal ions in the effluent. Ion exchange with the iron fines only occurs when the iron corrosion takes part aerobically or anaerobically, as shown in Reaction 3 and Reaction 4, respectively.

$$Fe\ (s) + (1/2)\ O_2 + H_2O \rightarrow Fe^{2+} + 2OH^- \qquad (3)$$

$$Fe\ (s) + 2H_2O \rightarrow Fe^{2+} + H_2 + 2OH^- \qquad (4)$$
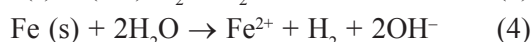
Chloride ions could also take part in the anion exchange with the iron fines to release OH^- ion.[118] The released hydroxide ion will further react with the metal ions besides controlling the pH.[22,140] However, most of the metal ions in the leachate carry a positive charge, so the ion exchange with OH^- could not be the dominating mechanism. There is another possible mechanism with the formation of iron hydroxide due to the reaction of Fe^{2+} ion with water aerobically, as shown in Reaction 5.

$$4Fe^{2+}\ (s) + O_2 + 10H_2O \rightarrow 4Fe(OH)_3\ (s) + 8H^+ \quad (5)$$
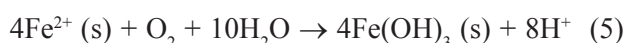
As the precipitation of iron minerals occurs with no significant decrease in pH, the mechanism of co-precipitation of metal cation with iron hydroxide is more likely to be the possible mechanism.[22,140]

Adsorption treatment application

Adsorption treatment is regarded as one of the most effective tertiary treatments for removal of high molecular weight compounds of leachate, especially for stabilised leachate.[141] Activated carbon was used as a medium to treat the complexity of the leachate either in a single stage or in combination with other best available technologies, including physicochemical and biological processes. Based on the reports, the conventional activated carbon was usually studied as an adsorbent to test its performance in treating different categories of leachate, ranging from young leachate, intermediate leachate to stabilised leachate. The results of the studies are presented in Table 9. Different types of activated carbon are available commercially. Granular activated carbon and powdered activated carbon are two most common products applied in the leachate treatment process. Some suppliers use their own brand name for the activated carbon. Therefore, it is essential to carry out the experiments for comparison of treatment capabilities and suitability of the products for the identified leachate. The main removal parameters measured include COD, NH_3-N, organic matter, color, phosphate, and heavy metals.

With reference to COD parameters shown in Table 9, the activated carbon has higher maximum adsorption capacity (4,300 mg g^{-1}) for stabilised leachate in comparison with intermediate leachate (0.258 mg g^{-1} only). This is in agreement with Aziz *et al.*, who have recognised activated carbon as a reliable means to treat non-biodegradable, refractory organic compounds found in stabilised leachate.[153] Generally, the COD removal rate for stabilised leachate, intermediate leachate, and young leachate is 59 %, 54 % and 49 %, respectively.

NH_3-N is another parameter whose adsorption onto the activated carbon was measured. The removal rate of 90 % was achieved in treating stabilised leachate due to the presence of functional acidic groups on the surface of the activated carbon. These functional acidic groups are hydroxyl group, carboxyl group, and carbonyl group, which may impart a polar character to the activated carbon surface.[129] Moreover, the dissolved organic carbon and hydrophobic organic chemicals in the leachate can also be removed via activated carbon. Organic matter, which forms complexes with the heavy metals, can also be removed with the activated carbon. Inorganic macro-compound-like phosphate, which

Table 9 – *Leachate removal percentage for conventional activated carbons*

Activated carbon type/ precursor	Maximum adsorption capacity (mg g^{-1})	Removal percentage (%)			Reference
		Stabilised leachate	Intermediate leachate	Young leachate	
COD					
Commercial PAC	4,300	38	–	–	[142]
DARCO	–	38	–	–	[143]
GAC (type PHO 8/35 LBD)	165.46	60	–	–	[144]
Calgon Filtrasorb 400	564	70	–	–	[145]
Norit 0.8	88.8	90	–	–	[146]
Commercial PAC	–	–	24.6	–	[147]
Norit SA 4	–	–	38	–	[148]
Picacarb 1240	0.148	–	48	–	[141]
Commercial PAC	–	–	49	–	[149]
Chemviron AQ40	0.258	–	55	–	[141]
Norit 0.8	0.253	–	68	–	[141]
Commercial PAC	–	–	75	–	[150]
Carbotech	0.250	–	75	–	[151]
Commercial PAC	–	–	**10.**	49	[152]
Ammoniacal nitrogen					
GAC (type PHO 8/35 LBD)	53.58	95	–	–	[144]
Commercial PAC	–	–	16	–	[149]
Commercial PAC	–	–	44	–	[150]
Commercial PAC	–	–	–	78	[152]
Organic matter					
DARCO	–	40 [Dissolved organic carbon]	**11.**	**12.**	[143]
Commercial PAC	–	–	89.2 [Hydrophobic organic chemicals]	–	[147]
Colour					
PAC	–	–	–	50	[152]
Phosphate					
PAC	–	–	44	–	[150]
Heavy metals					
Commercial GAC	–	90 [Cobalt (Co), Chromium (Cr), Iron (Fe), Manganese (Mn) and Nickel (Ni)]	–	–	[118]

was negatively charged, was also removed in portion, and the mechanism of the process has already been discussed in the earlier section. Non-conventional activated carbons (NACs) have also received attention for their potential in removing contaminants from leachate. Basically, many of them could be sourced locally and originated from food wastes, agricultural wastes, and industrial by-products. Similar parameters were monitored and analysed for the performance of the selected non-conventional activated carbons, like for the conventional activated carbon, as discussed earlier (Table 10).

Table 10 – *Leachate removal percentage for non-conventional activated carbons*

Activated carbon type/precursor	Maximum adsorption capacity (mg g^{-1})	Removal percentage (%)	Reference
COD			
Rice husk carbon composite	3.11	27.61	[154]
Durian peel	61.72	41.98	[155]
Palm shell	1,460	50	[130]
Rice husk	–	70	[156]
Sugarcane baggase	–	77.8	[128]
Tamarind fruit seed	64.93	79.93	[129]
Sugarcane baggase	126.58	83.61	[126]
13. Ammoniacal nitrogen			
14. Sugarcane baggase	15. –	16. 41.05	17. [128]
Sugarcane baggase	14.62	46.65	
Rice husk carbon composite	12.9	51.0	[154]
Sugarcane baggase	138.46	79.63	[127]
Organic matter			
Colour			
Durian peel	100	39.68	[155]
Rice husk	–	60	[156]
Sugarcane baggase	–	87.3	[128]
Tamarind fruit seed	168.57	91.23	[129]
Sugarcane baggase	555.56	94.74	[126]
18. Phosphate			
19. Coffee ground	20. –	21. 84	22. [123]
Sugarcane baggase	–	85.06	[157]
Metals			
Banana frond	26.15	95.14 [Iron (Fe)]	[127]

For COD removal, the sugarcane baggase NAC achieved the highest percentage, which was 83.61 %, as reported by Azmi *et al.*, though the maximum adsorption capacity was obtained with the palm shell NAC, which was 1,460 mg g^{-1}.[128] Nevertheless, the minimum of 27.61 % removal could still be achieved by using the rice husk carbon composite NAC. An average of 61.6 % removal in overall performance showed that the non-conventional activated carbons could be competitive or even better than the conventional activated carbons, which could only achieve 59 % removal rate in average for COD reduction in leachate.

Sugarcane baggase and rice husk carbon composite NACs were analysed for the removal of NH$_3$-N from leachate. Both adsorbents showed almost similar performance in terms of maximum adsorption capacity and removal percentage.[126,128,156]

Rice husk carbon composite NAC still achieved a slightly better performance than sugarcane baggase.[157] However, another study carried out by Foo *et al.*, showed that sugarcane bagasse NAC managed to remove 79.63 % of NH$_3$-N, which was higher than the rice husk carbon composite NAC (51.0 %).[157] On average, 54.58 % removal was achieved by using the non-conventional activated carbons as compared with 68 % achieved by the conventional activated carbon.

Orange peel based non-conventional activated carbon was the only one investigated for the removal of total organic carbon (TOC), and 59.7 % removal was achieved based on the study carried out by Xie *et al.*[158] Colour of the effluent discharge is also another parameter of environmental concern, due to the presence of humic acids and fulvic acids, which requires the solution for its treatment. Sugar-

cane bagasse and tamarind fruit seed NACs were found to remove more than 85 % of colour in the leachate, and the highest removal percentage of 94.74 % was obtained using sugarcane bagasse NAC. The lowest percentage was found with durian peel NAC, which was only 39.68 %.

Metal ions present in leachate represent a threat to river ecosystems if they are not properly treated before discharge. Therefore, non-conventional activated carbons have been studied for their adsorption potential in treating metal ions. Based on the analysis of metals in leachate, banana frond and coffee ground NACs, as well as bone meal and iron fines have shown impressive removal rates above 75 % and >90 % of some of the metals, including boron (B), iron (Fe), strontium (Sr), manganese (Mn), chromium (Cr), mercury (Hg), and calcium (Ca).[127,129,139]

A comparison has also been made of the treatment performances of sand filter, peat filter, and granular activated carbon (GAC) filter, with respect to removal of some refractory organic compounds, based on the study carried out by Kalmykova *et al.*[119] GAC filter and peat filter have good adsorption capability for bisphenol-A, and 4-*N*-nonylphenol 80 % of bisphenol-A was removed by the peat filtration. Polycyclic aromatic hydrocarbons (PAHs) were removed 63 % and 50 % by the peat filter and GAC filter, respectively. However, GAC filter had higher removal percentage than peat filter in adsorbing 4-*tert*-octylphenol and 4-*tert*-octylphenol monoethoxylate (OP1EO). Petroleum hydrocarbons (PHCs), containing mostly C_{16}-C_{35} alkane fraction and less abundantly C_{35}-C_{40} alkane fraction, were also tested for the efficiency of removal in GAC filter and peat filter. GAC filter and peat filter achieved 50 % and 35 % removal percentage, respectively.

GAC filter is capable of removing colloidal and dissolved pollutants. Low molecular weight PAHs were more effectively removed in GAC filter as compared with the peat filter. This is attributed to the capability of GAC filter to adsorb volatile hydrocarbons rather than to the degradation of the PAHs. Microbial degradation is the route to degrading PAHs.[159,160] GAC filter has excellent removal rate for 4-*tert*-butylphenol (dissolved phase) and 4-*tert*-pentylphenol as compared with peat filter and sand filter. Sand filter could only remove 4-*tert*-butylphenol though being in dissolved phase. Furthermore, GAC filter could effectively remove humic and fulvic acids (belonging to the class of organic colloids) as well as alkylphenols.[121] However, GAC filter was not effective in removing high molecular weight PAHs as well as low molecular weight petroleum hydrocarbons belonging to C_{10}-C_{12} alkane fraction. Peat filter has better removal efficiency than GAC filter in removing phthalates. According to Jonsson *et al.*, phthalates could be degraded in the peat filter.[161] However, phthalates can be degraded to mono-esters and phthalic acid both under biotic and abiotic conditions, and their degradation was observed in landfills, where the abiotic degradation route can be neglected. The degradation of phthalates may occur even during the sample storage, due to improper storage conditions.[119] Additionally, peat filter can remove oxygenated PAHs (oxy-PAHs) more effectively than GAC filter. However, as compared with GAC filter, the peat filter could not absorb humic acid colloids and attached compounds due to the repulsion of similar negative charges. Humic and fulvic acids are the degradation products of peat, and constantly emitted in trace amounts from the peat filters.[162] Peat filters were also ineffective in removing alkylphenols, which could leach out after the saturation of the filter bed. Based on the study by Kalmykova *et al.*, the removal percentage of the peat filter decreases in the order of bisphenol-A > 4-nonylphenol > 4-*tert*-octylphenol > 4-*tert*-butylphenol > 4-*tert*-pentylphenol, due to the increase in colloidal phase content.[163] Organic colloids originating from humic and fulvic acids were removed completely. The proposed treatment order was (i) sand filter (auto backwashed to remove the particles), (ii) peat filter (removes high molecular weight PHCs, di-(2-ethylhexyl) phthalate, oxy-PAHs, and traps small particles escaped from the sand filter), (iii) GAC filter (removes most volatile PHCs and PAHs). Filters based on non-conventional absorbents could be potentially applied as an alternative to GAC filter.

Challenges and future works

Determination of the optimum dosage of coagulants and flocculants is required to enable the precipitation of humic acids, and to avoid excessive sludge generation at the end of the process. Another challenge is to manage the generated chemical sludge during the coagulation/flocculation process. Metal-containing sludge could be accumulated in huge quantities due to high amount of leachate volume generated from the landfill, especially in tropical climates where it is hot and humid throughout the year. Improper management and disposal of the sludge is a cause for environmental concern. Therefore, additional metal contribution from the inorganic coagulants could be avoided if natural coagulants and biopolymers could be fully understood in terms of their biochemical reactions and molecular structure. This will also help to better understand the removal characteristics of the organic matter in leachate.

One of the main problems with adsorption treatment is the potential of leaching out or desorption of the xenobiotic organic compounds from the filter bed.[119] Further exploration of the characteristics of xenobiotic organic compounds and their adsorption interaction with the surface of the absorbent could be another future perspective in this field. In addition, non-conventional absorbents are still under lab-scale experimental investigation, and have yet to be commercialised in the market. The performance of non-conventional absorbents has not been fully tested in the pilot scale or industrial scale, which opens the possibility for future research.

After physicochemical treatment, there remain recalcitrant compounds like humic substances and fulvic fractions with low molecular weight commonly found in old and stabilised leachate. Those compounds require attention in order to select proper tertiary treatment methods. Unlike other heavy metals, which are easily coagulated and precipitated under acidic conditions, the concentration of manganese increases many times during treatment, as manganese easily dissolves in the leachate. Therefore, a separate pre-treatment or post-treatment method may be required to remove this particular metal.[91]

Conclusion

In this review, the physicochemical treatment methods for landfill leachate, especially coagulation and flocculation as well as adsorption, have been discussed. Research papers reporting an environmentally friendly approach aimed to reduce the dosage of inorganic coagulant and generation of acid sludge have been discussed. The renewable source of non-conventional absorbents is an alternative option to conventional activated carbon. In chemical processes, coagulation and flocculation have shown potential in removing pollutants from the leachate with average removal of >75 % for COD, organic matter, turbidity, total suspended solids, anions, and heavy metals, with the only exception of ammoniacal nitrogen. The combination of inorganic coagulants with grafted polymers will be a better option as compared to organic synthetic polymers, since the usage of inorganic coagulants may be reduced substantially. However, the optimal working pH for coagulation and flocculation is around 4 – 6.5. This indicates that pH adjustment of the leachate is necessary. The results have also proved that higher removal is obtained if pH adjustment is performed in the early stage of the process, in comparison to the process without pH adjustment. The remaining fraction of low molecular weight humic acids and fulvic acids, depending on the leachate characteristics, could be further removed by adsorption treatment or by the biological processes. Non-conventional activated carbons are the focus in leachate treatment, as they are renewable and more environmentally friendly compared to conventional activated carbons. In terms of performance, both types could achieve almost similar treatment efficiency.

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