Landfill leachate treatment technology via electrocoagulation: a review of operating parameters, intensification, and modelling

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ABSTRACT

Electrocoagulation has been emerging as a promising future and offers various advantages in dealing with highly recalcitrant organic wastewater. For the past years, electrocoagulation managed to deal with different types of water and wastewater pollutant removal efficiently. Compared to previous conventional method of coagulation/flocculation, electrocoagulation is highly rated and improves the removal efficiency, especially for a multitude of recalcitrant pollutants. Landfill leachate is considered as one of the highly recalcitrant organic wastewater that results from the degradation of complex organic and inorganic material, which requires serious attention for treatment before being released back to the environment. In this review, a comprehensive discussion was carried out on the theory, operating parameters, intensification, and modelling of electrocoagulation, especially in treating landfill leachate. The recent development of landfill leachate studies is explored in this paper, including the theory and fundamentals of solid waste management. This review provides an extensive cover to the theory, factors, modelling, intensification, and advanced electrocoagulation treatment for landfill leachate treatment.

Keywords: Electrocoagulation; Landfill leachate; Municipal solid waste; Advanced oxidation process (AOP); Modelling

1. Introduction

1.1. Fundamental of solid waste management

The growth of the industrial revolution movement worldwide, particularly in the developing country leads to the opening of new industrial zones and migration of workers into these hotspot areas, thereby increasing the generation of solid waste produced. The higher production of municipal waste leads to high waste disposal spending, particularly in a country that relies heavily on landfills. According to a report by the World Bank Group, the projection for solid waste is expected to grow up to 3.40 billion tonnes globally by 2050, as shown in Fig. 1 [1]. The rapid generation of solid waste drives researchers worldwide to explore possible alternatives to reduce the abundance of solid waste and maximise pollutants removal via treatment technologies. This review
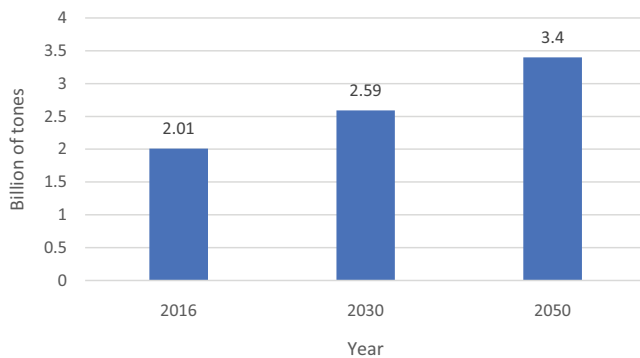
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Fig. 1. Projected global waste generation [4].

will discuss the current state of solid waste management in landfills and its derivative, such as leachate in detail. Solid waste management differs from one country to another based on the approaches, policies, and cost-efficiency. In many countries, municipal solid waste is a major problem that causes environmental issues. The global issues on waste generation cause land scarcity, as the waste requires a longer time to degrade and produce other consequences effects. The global municipal solid waste is contributed by various sources, including industrial, commercial, and agricultural by-products that continuously increases from time to time [2]. According to a report by Kaza et al. [1], the world generates waste of 0.74 kg/cap/d, which varies from 0.11 to 4.54 kg/ca/d depending on the native country. The report also suggested that the key factors that will influence the rapid acceleration in waste generation are economic and population growth. The expenses allocated for the cycle of solid waste management starting from the collection, transportation, until the disposal and recovery process involve a high-value amount of money, which is repeated every single year to combat the abundance of waste produced. The trend of solid waste management is diverting from the conventional method to a modern and systematic approach. Numerous technologies are proposed by engineers and researchers worldwide aiming

to provide alternatives to counter the vast amount of waste generated and its derivatives. In the hierarchy of waste management, landfilling is considered the least preferred method for waste disposal, even though various precautionary steps are taken to reduce the drawback and their consequences. A proper plan for landfill operation needs to be taken into account as the landfill will involve a large-scale area and only last until full capacity before being closed down and recovered. The ultimate and safest landfill is known as sanitary landfill, which applied the principle of engineering and modern constructed facilities with continuous monitoring of leachate and gas emissions. Fig. 2 illustrates the design of a modern constructed landfill [3]. The main concern on the landfilling process is regarding the uncontrolled generation of leachate and gas emission, which deteriorated the environment [4]. Due to various consequences of leachate and gas emission, the landfill needs to fulfil the requirement of local authorities with regard to the management and discharge limits to prevent contamination in the chemosphere. The municipal waste undergoes degradation and decomposition via aerobic, anaerobic, or semi-aerobic processes. The decomposition process will be heavily influenced by several factors, such as solid waste structure, moisture content, potential hydrogen (pH), landfill age, and landfill operation [5].

1.2. Leachate generation and its properties

The landfill leachate is a derivate from the decomposition of heterogeneous waste by physicochemical and biological means, resulting in the production of highly concentrated liquid containing organic and inorganic compounds. Leachate poses a serious problem towards the flora and fauna causing detrimental effects on the environment. Furthermore, it could either flow into the water stream due to runoff and illegal discharge, or dissolved through the soil matrix and contaminated groundwater source. The requirement of a modern facilitated landfill in level three sanitary landfill that is equipped with a proper liner at the bottom of each cell and daily cover, drainage for leachate flow as well as methane gas collection would
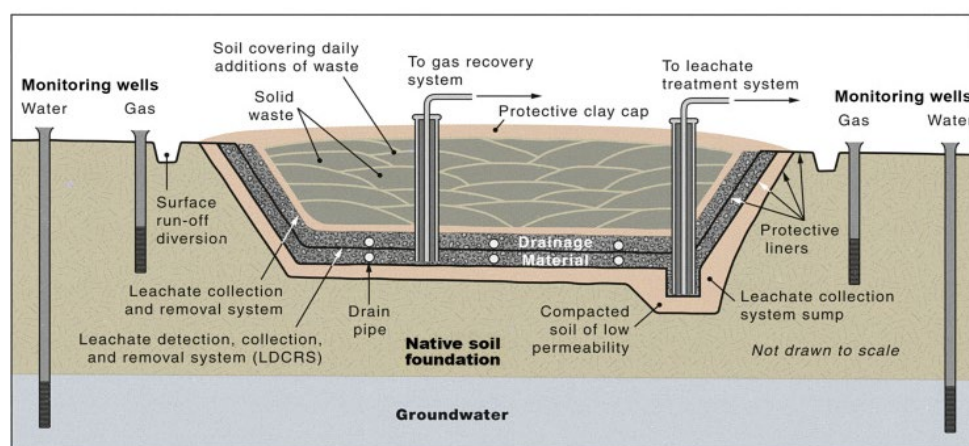

Fig. 2. Design of a modern constructed landfill [3].

Table 1
Composition of constituents in a different state of leachate [7]

Types of leachate	Young	Intermediate	Stabilised
Age (years)	<5	5–10	>10
pH	<6.5	6.5–7.5	>7.5
Biodegradability	Important	Medium	Low
Kjeldahl nitrogen (g/L)	0.1–0.2	–	–
Ammonia nitrogen (mg/L)	<400	–	>400
TOC/COD	<0.3	0.3–0.5	>0.5
Heavy metals (mg/L)	Low to medium	Low	Low
BOD_5/COD	0.5–1.0	0.1–0.5	<0.1
COD (mg/L)	>10,000	4,000–10,000	<4,000

reduce the possibility of pollution. However, a complete and modern sanitary landfill requires massive investment and recent technologies that cannot be fully afforded for all landfills worldwide, especially in developing countries. In these countries, the commonly applied landfill involves open dumping and controlled tipping with only a few landfills that are considered proper sanitary landfills for stages one and two, and even fewer for stage three landfill. The impact of a low number of sanitary landfills equipped with waste recycling facilities will increase the burden on the landfill and reduce the lifespan. The common physicochemical parameters studied in leachate included biochemical oxygen demand (BOD), chemical oxygen demand (COD), alkalinity, pH, ammonia nitrogen, heavy metals, odour, colours, total dissolved solids, and organic compounds [6]. Information on the landfill's age and pH, combined with leachate composition parameters are crucial to provide the actual phase of landfill leachate, which provides guidance for suitable selection of landfill leachate treatment. The range of pollutant composition in landfill leachate with different classifications is summarised in Table 1.

The quality of landfill leachate produced depends on the degradation phase, meanwhile the amount of leachate produced is related to the solid waste composition, particle size, degree of compaction, hydrology of the site, landfill age, moisture and temperature condition, and available oxygen [8]. In the early stage of the landfill leachate (acidogenesis phase), the presence of volatile organic compound influences the low pH of the wastewater [9]. The concentration of acid in the leachate would eventually decline (methanogenic phase), resulting in an increase in pH and leaving an acute volume of organic compound left behind [10]. The average pH of landfill leachate in this phase varies between 7.6 and 8.2 [11]. The degradation process of landfill leachate will eventually change the wastewater pH and other constituents. Therefore, different type of treatment system needs to be employed for different level of landfill leachate. As stated by Shadi et al. [12], the landfill leachate approximately undergoes five different stages for stabilisation process, which is illustrated in Table 2. The climatic conditions change from aerobic to anaerobic before return to aerobic state once the refuses degraded by Adam et al. [13].

1.3. Leachate treatment technologies

The priority of handling and treating landfill leachate turns the researchers' attention from conventional methods to current advanced technologies that are more promising and effective. The technologies vary from biological, chemical to physicochemical treatment. The variation in treatment technologies depends on the target pollutants, and the type of stages of the leachate phase. The knowledge regarding the landfill leachate is crucial to deploy suitable treatment methods.

The biological treatment utilises microbes to carry out the degradation process of organic compounds. Common unit operation in biological treatment technologies involves the presence of dissolved oxygen that influenced the treatment process. This treatment is known to be reliable, simple, and highly cost-effective to tackle problems associated with organic and nutrient in wastewater, especially BOD [14]. The microbes grow in the attached or suspended system. According to Kamaruddin et al. [2] the efficiency of biodegradation by microbes showed a decline performance due to the formation of refractory compounds, such as humic and fulvic acid that have low biodegradability. The structure of humic-like substrates made of aromatic rings and aliphatic chains is the reason for the low degradation of organic compounds [15]. The humic substance can be classified into three types, which are humin, humic acid, and fulvic acid [16]. The solubility of each type of humic substance is different, whereby humic acid requires an alkaline condition to become soluble, fulvic acid in aqueous solution, while humin is insoluble at any pH condition. The biological treatment process is frequently employed for treatment of young leachate, which is rich in high molecular organics and less consumable COD. Treatment of mature landfill leachate is known to be less effective unless biological treatment is combined with other non-biological treatment technology [17].

The chemical treatment method is known through the addition of chemicals to react with the desired pollutant before eliminating it. Neutralisation is usually employed in this type of treatment, whereby charge neutralisation occurs with the addition of acid or base. Moussa et al. [18] stated that the addition of chemicals will increase the total dissolved solids (TDS), thus making this treatment method undesirable for leachate treatment. The treated leachate

Table 2
Stages for stabilisation process of landfill leachate [12]

Stages of landfill leachate	Description
Initial adjustment phase	- Microbial decomposition of biodegradable organic matter takes place in aerobic conditions due to the presence of air.
	- Still a low formation of landfill leachate.
Transition phase	- Covering of waste cells will cause a cut in oxygen supply to the cell and limits the aerobic microbial decomposition.
	- Increase in heat temperature and magnify the leachate production.
Acidogenic phase	- Anaerobic state of the landfill.
	- High organic content and strong acidic pH level due to the presence of CO_2.
	- Increase in ammonia and metal concentrations.
	- The phase occurs for four months, while stabilisation of landfill gas generation level lasts for 1–2 years.
Methane fermentation phase	- Establishment of leachate in a neutral or slightly alkaline state, which takes months or years.
	- Composition of methane and CO_2 ranges between 55%–60% and 40%–45% once the phase has been stabilised.
	- Bacterial activities at high temperatures, such as mesophilic bacteria and thermophilic bacteria, which consumed CO_2 and acetate.
Maturation phase	- Re-establishment of aerobic states with new aerobic microorganisms' growth.
	- Biodegradable refuse has transformed into CO_2 and methane.

would require further treatment before discharge, as the net dissolved content increases and need to comply with the standard discharge limits. According to Chawaloesphosiya et al. [19], the flocs derived from chemical coagulation were bulky and tend to break up when exposed to minimum agitation making the separation of flocs difficult. Production of high sludge volume causes usage of chemical unfavourable [20].

The inefficiency of physical, biological, or chemical treatment to stand alone in the treatment process leads to the combination of two or more treatment processes, which is known as the physicochemical process. Conventional physicochemical proposed for landfill leachate treatment involves adsorption, chemical oxidation, electrochemical technology, advanced oxidation processes (AOPs), and membrane filtration technology. AOPs that utilise strong oxidants combined with various irradiation manage to provide high removal and degradation pollutants. However, the possibility for chlorine oxidation, resulting in the creation of chlorine or hypochlorite, and the major drawback of poor economic acceptability for large-scale procedures reduce the treatability using AOPs [21]. Membrane filtration technology, which commonly involves ultrafiltration, nanofiltration, microfiltration, and reverse osmosis are undoubtedly efficient to eliminate colloids and suspend materials at high removal efficiency. In contrast, pressure-driven processes are vulnerable to fouling by a wide range of constituents (which necessitates significant pretreatment, or chemical cleaning of the membranes, resulting in shorter membrane lifetime and lower productivity process), and a large volume of concentrate formation (which is unusable and need further treatments) [22]. The role of electrocoagulation in wastewater treatment can be considered significant and able to provide an efficient treatment. Worldwide, various researchers'

reviews have recognised the contribution of electrocoagulation in wastewater treatment. Verma and Kumar [23] compared the conventional coagulation and electrical conductivity (EC) as a method of pretreatment, while Mousazadeh et al. [24] covered the application of electrical conductivity/elemental chlorine free (EC/ECF) process under different operating conditions. Didar-Ul Islam [25] provided the application of electrocoagulation in various fields of wastewater. Moussa et al. [18] provided the potential and challenges of EC for water treatment. Ghernaout [26] described the electric field role contribution in EC treatment. Hakizimana et al. [27] reviewed and explained the electrocoagulation modelling approaches in water and wastewater treatment. The major aim of this review is to understand the theoretical aspects behind electrocoagulation and compare the current intensification and advanced electrocoagulation treatment focusing on landfill leachate. The author also listed the available tools and modelling to integrate with electrocoagulation.

2. Fundamental

2.1. Theory of coagulation and flocculation

Coagulation and flocculation had been applied for centuries, estimated around as early 2000 BCE, commonly for water clarification and potabilisation [28]. In the 21st century, coagulation and flocculation remain relevant and important for water and wastewater treatment technology. Coagulation and flocculation refer to two different processes that complement each other. Coagulation involves the destabilisation of particles via the reduction of repulsive forces between particles, or through entrapment in precipitates. Hogg [29] and Harif et al. [30] indicated that insoluble particles will undergo inter-particle repulsion caused by electrical double layer interaction. The author

also suggested that the addition of soluble ionic species might disrupt and change the surface potential difference of the colloids, either via adsorption to the particle surface, or by double-layer compression. The destabilisation process is heavily linked with the concentration of metal ions and pH. Typically, the pollutants that are present in water and wastewater involve colloidal particles that are known to be very difficult to remove as a result of land surface runoff, the decay of vegetation, and domestic and industrial effluent discharge. According to Moussa et al. [18], the colloid is a stable microscopic particle, with a size ranging from 1 nm to 2 µm, consisting of a total surface area larger than their respective mass and size. Colloidal particle stability is often referred to as repulsive force exerted on the surface of particles that prevent agglomeration [31]. According to the infamous Derjaguin-Landua-Verwey-Overbeek (DLVO) theory, colloidal particle stability is influenced by the effects of van der Waals attractive force, and repulsive electrostatic forces exerted by particle surfaces double layer. Due to the nature of both different forces, the net charge remained at zero to balance in the colloidal system. However, to promote agglomeration, the addition of coagulating agent is essential to reduce the distance of the electrical double layer, which reduces the electrostatic repulsion, narrowing the surface potential, and changing the net forces, thus allowing aggregation [32,33]. The principle of destabilisation of the colloidal system follows any of these four mechanisms (compression of the electrical double layer, charge neutralisation, adsorption, and bridging between particles, entrapment of particles in the precipitate). This is influenced by several factors, such as chemical and physical properties of wastewater, coagulation/flocculation, type of wastewater and pollutant Moussa et al. [18] and Matilainen et al. [34] described that this process reduces the repulsive potential double layer of colloids by turning it into microparticles, which will collide with each other to form larger flocs. Similar to coagulation and flocculation, electrocoagulation possesses an identical mechanism for colloidal destabilisation and removal by overcoming the repulsive force and initiates agglomeration.

2.2. Theory of electrocoagulation

The development of electrocoagulation (EC) treatment technologies started in late 1880s in the European countries for treating and disinfecting sewage [35]. A few years later, EC technologies were adopted for plant-scale treatment of sewage and canal water. The genuine interest in EC remains high until now to mitigate various kinds of wastewater. EC is classified as one of the emerging electrochemical technologies available and is important in the field of electrochemistry. Electrochemistry is dubbed as one of the clean technology due to the utilisation of electrons as the main precursor. Numerous research-based on electrochemistry are developed worldwide to cope with pollutants in gases, liquids, or soils. According to Feng et al. [36], the field of electrochemistry via electrochemical technology provided various advantages, such as acceptable cost, high-efficiency energy, versatility, and low resource consumption. The author also recognised electrochemical technology for pretreatment, or as an advanced treatment method, and listed the available technologies under electrochemical technology, as illustrated in Fig. 3 [36]. Another researcher found
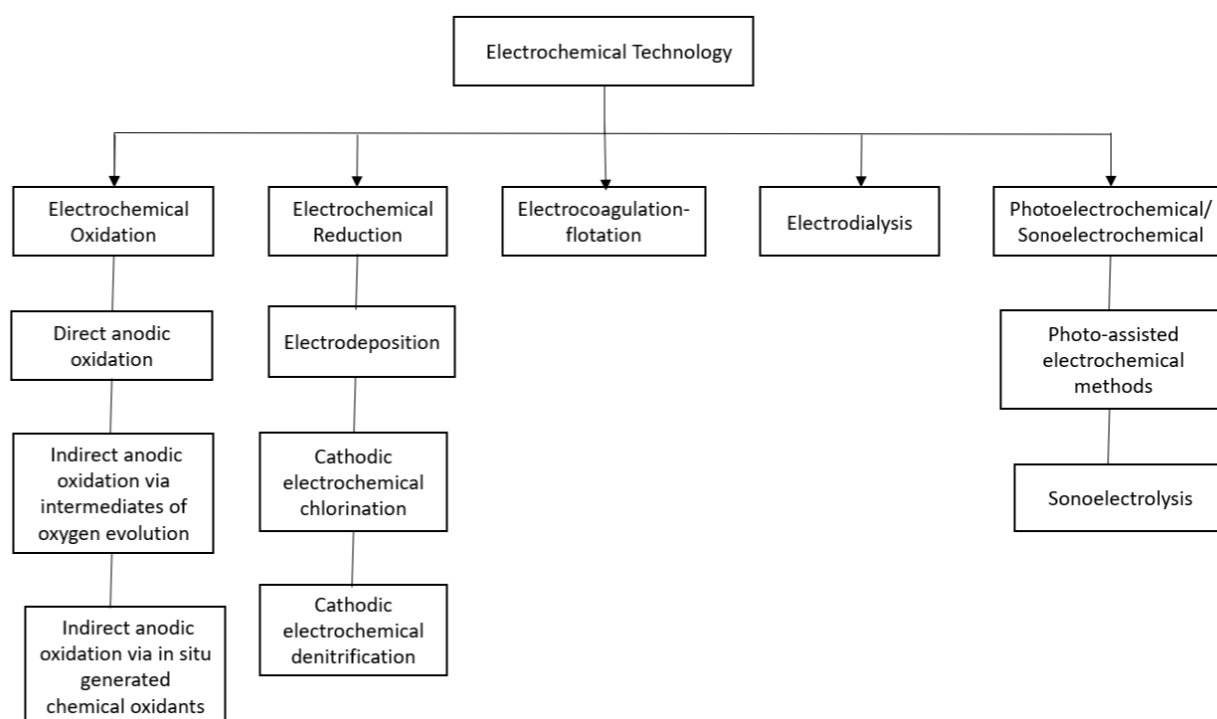

Fig. 3. Recently available technologies under electrochemical technology [36].
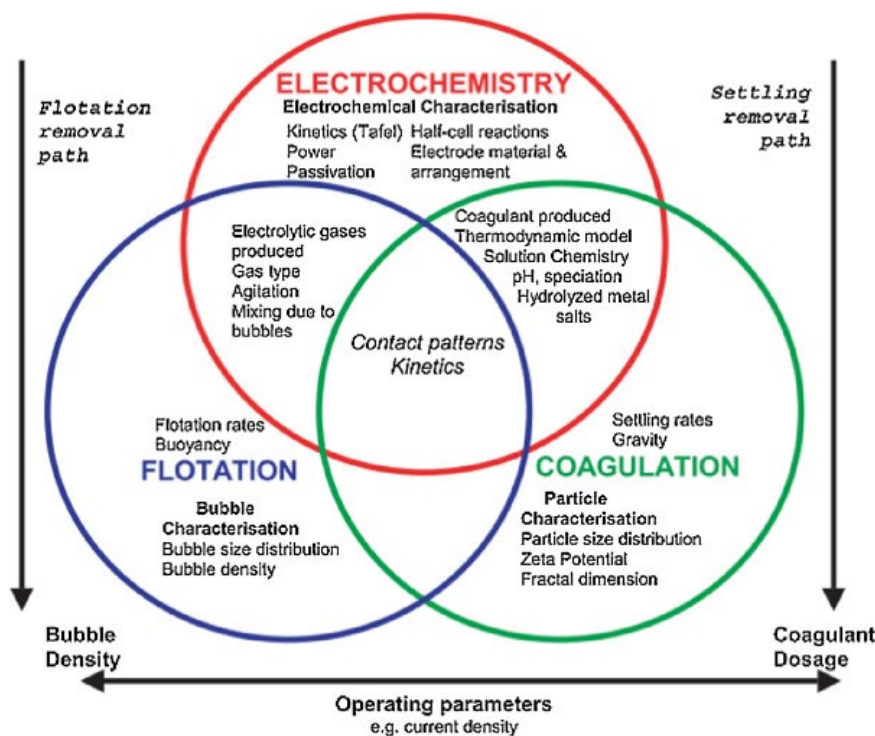
Fig. 4. Conceptual framework for electrocoagulation as a synthesis technology [38].

that electrochemical technology provided an environmental-friendly option with no chemical additives, low sludge generation, and minimal footprint that provided efficient treatment results [18].

Amongst the listed technologies, EC emerged as one of the current alternatives in treating wastewater, including landfill leachate. The shift from traditional coagulation-flocculation process to EC is a major upgrade for the leachate treatment process, as many advantages are achieved through this replacement. Researchers worldwide suggested that EC could provide an efficient treatment system. EC is considered as an AOP, a process under the generation of electricity [37]. EC combines three fundamentals of knowledge into one technology, which is electrochemistry, coagulation, and flotation. As with all of the technologies applied in EC, it is important to understand the key interactions that occur within it. Holt et al. [38] in their works came out with a conceptual Venn diagram (Fig. 4) to highlight and relate the interaction between all the knowledge. The phenomena that described EC as a whole, involved contact patterns and kinetics. The former explained how the different species (coagulant, pollutant particles, bubbles) travel and interact with one another, while the latter indicated the pace at which interactions between the various species occur. As a result, these characteristics are positioned at the junction of all three lobes. The physicochemical process within EC forced the removal and separation into two ways, settling and flotation. Both mechanisms are influenced by the combination of reactor configuration and operating parameters. The complicated interconnections between the three foundation technologies have resulted in an inescapable relationship between design and operating factors.

According to Fryda et al. [39], the ability of electrochemical advanced oxidation processes to treat a wide range of COD makes it a preferred method to treat landfill leachate compared to other treatment technologies. The finding indicated that the significant advantage for the implementation of EC in electrochemical advanced oxidation process was a wide range of COD concentrations. As stated by Shahedi et al. [40], EC treatment covered a wide range of particle sizes from an ionic range up to macro particle range, which is commonly attributed to particles, such as metal ions, suspended solids, humic, tannic, folic acids, colloids, and few other particulate materials. The EC treatment system includes a high removal of contaminants, which allows the treated wastewater to further be reused for some cases of wastewater. Padmaja et al. [41] and Sirés et al. [42] stated that EC able to provides various range of COD treatment which reflect the flexibility of this treatment method. Other benefits gained from this treatment system are short reaction time, in situ coagulant formation, low sludge formation, no chemical addition, and less selective.

Generally, EC is a process of dissolution of sacrificial anode with simultaneous hydroxyl ion and hydrogen gas formation at cathode through an induced chemical reaction in water by electric current [30]. EC combines several mechanisms, involving electrochemical, chemical, and physical that occur either sequential and/or parallel [27,43]. The basic EC reactor consists of direct current (DC) power supply connected to the anode and cathode electrode that is immersed in the wastewater reactor. The EC technique involves an in situ dissolution of the sacrificial anode as the ionic coagulant into the wastewater,

depending on the applied current, pH, and conductivity of wastewater [44]. A significant difference between traditional coagulation/flocculation and EC involves the release of metal hydroxides, which are directly added in the EC process. Meanwhile, multivalent cation salt is released in the conventional coagulation/flocculation process, requiring a neutralization process afterwards, and thus making the previous treatment unsuitable [45]. The tendency for coagulation is much higher in electric field potential compared to the addition of chemicals [46]. The metallic ions dissolution obeys the Faraday's law theory.

During the EC process, several reactions took place at both the anode and cathode as well as in the bulk solution. A cationic coagulant is simultaneously produced in situ to assist the sludge production. Different for coagulation and flocculation, counter-ions should be added during the dissolved phase and need to be neutralised using a chemical, such as lime $Ca_2(OH)_2$ to prevent the pH drop due to the hydrolysis of metal salts [32]. Fig. 5 illustrates the general process and reaction that occurred in an EC batch reactor [18]. The following is a summary of the general mechanism and equation of reaction that occurred [17,24,27,47,48]:

- The metal anode dissociated the metal ions when DC was passed through the cell. The dissolution of metal into ionic coagulant depended on the properties of anode material as well as the properties of wastewater.
- In the bulk solution, water was oxidised into hydroxyl ions and interacted with the released metal ions forming amorphous metal hydroxide flocs. The hydroxide flocs are known for having large surface areas that provide good adsorption and trapping of colloids (sweep coagulation). However, a high anode potential could lead to a secondary reaction to occur. The presence of chloride ions and alkaline pH could influence the water to be oxidised as hydronium cation and oxygen, while chloride anion, Cl^- converted into Cl_2. The presence of Cl_2 as a strong oxidant would enhance the

oxidation of dissolved organic compounds, or leads to the formation of ClOH that would act as an oxidiser.
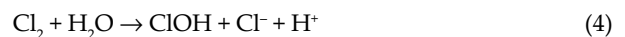- Contrary to an insoluble contaminant, dissolved contaminants, including the recalcitrant organic compound that could be obtained from landfill leachate would be eliminated using different mechanisms, such as precipitation, co-precipitation, adsorption, or complexation depending on the contaminants species. Colloids and emulsion, both considered insoluble contaminant favours removal via compression of double layer, charge neutralisation, adsorption, or bridging between particles, entrapment of particles in the precipitate (sweep coagulation).
- On the other side of the electrode, water was reduced into hydrogen gas and hydroxyl ions, as a result of side reactions. The presence of hydroxyl ions would drift the pH solution towards a more alkaline condition.
- The flocs would be further removed by sedimentation, or electro-flotation via attachment at hydrogen (H_2) bubbles from the cathode or oxygen (O_2) bubbles from the anode. If the flocs are not attached to the bubbles, they will settle due to their high density relative to the solution.
- Flow and mixing are desirable to improve the interaction between contaminants, coagulated particles, and gas bubbles.

General equation:
At anode:

$$M_{(s)} \rightarrow M^{n+}_{(aq)} + ne^- \tag{1}$$

$$2H_2O_{(l)} \rightarrow 4H^+_{(aq)} + O_{2(g)} + 4e^- \tag{2}$$

In case the presence of Cl^-:

$$2Cl^- \rightarrow Cl_2 + 2e^- \tag{3}$$

$$Cl_2 + H_2O \rightarrow ClOH + Cl^- + H^+ \tag{4}$$

At cathode:

$$M_{(aq)}n^+ + ne^- \rightarrow M_{(s)} \tag{5}$$

$$2H_2O + 2e^- \rightarrow 2OH^- + H_2 \tag{6}$$

In the bulk solution:

$$M^{n+}_{(aq)} + nOH^- \rightarrow M(OH)_{n(s)} \tag{7}$$

where $M_{(s)}$ = metal, $M^{n+}_{(aq)}$ = metallic ion.

Application of EC has been widely used in various treatments of EC. The variety of pollutants removal by EC increases the efficiency applicability in industrial and commercial use. Several industries, such as synthetic fibre, printing, drilling, pharmaceutical, and food industries widely applied EC in their treatment system, either as a pretreatment, primary treatment, or even as an advanced treatment system. Table 3 summarises several treatments of wastewater utilising EC in recent years. Landfill leachate is not excluded from utilising EC for treating highly recalcitrant organic matter and other contaminants. Table
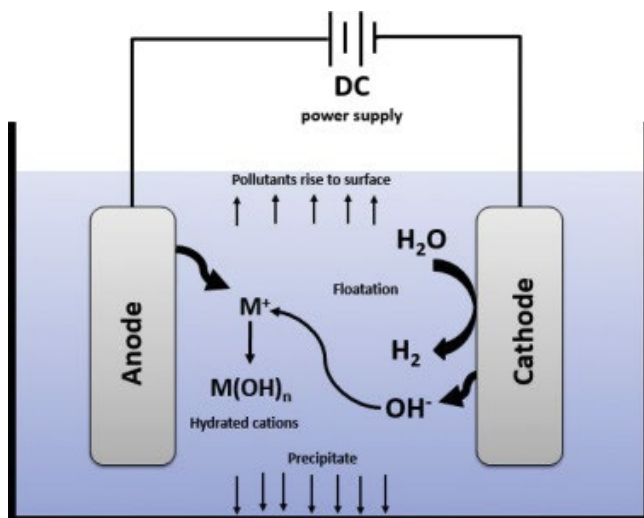

Fig. 5. General process and mechanism that occurs within an electrocoagulation reactor [18].

Table 3
Treatments of various wastewater utilising electrocoagulation

Effluent	Stage of EC	Electrodes	Arrangement	Parameter removal	Properties of wastewater	References
Wet-spun acrylic fibre (WAF) wastewater	Pretreatment	Fe–Fe	MP-P	TOC: 44% BOD/COD: 0.35%	- Effluent from WAF produces a large amount of wastewater rich in organic, inorganic, polymers, and colloid that are toxic and harmful. - High chemical oxygen demand and less biodegradability is the main problem of these types of wastewater.	[50]
Drilling fluid wastewater	Primary	Al–Fe	MP-P	COD: 89%	- Removal of petroleum organic matter from the wastewater of oil drilling process is necessary to prevent harm to the environment.	[51]
Printing wastewater	Pretreatment	Zn-Steel	MP-P	COD: 41% TDS: 19% OG: 14%	- Printing wastewater contains non-biodegradable compounds, high concentrations of COD, trace amount of toxic metals, intense colours, adhesives, and pigments that need to be addressed before being released or reused.	[52]
		Ti-Steel	MP-P	COD: 47% TDS: 5% OG: 19%		
Pharmaceutical effluent	Primary	Fe–Al	MP-P	COD: 92.3% TDS: 91.5% Chloride: 90.4% SS: 86.3%	- Water is commonly used in this industry for the process, such as a solvent or heat transfer agent. - Wastewater from this industry is very hazardous and contains high contents of BOD, COD, TDS, and SS. - High content of BOD could cause depletion of dissolved oxygen and renders the water unsuitable for further use.	[41]
Paper recycling wastewater	Primary	Fe–Al	MP-P	COD: 79.5% TSS: 83.4% Colour: 98.5% Ammonia: 85.3%	- Pulp and paper industry consumes high water usage and produces large volume of wastewater that is a complex mixture of toxic and recalcitrant substance. - This type of wastewater commonly employed to a high amount of COD, BOD, wood debris, and other contaminants.	[53]
Tanning wastewater	Primary	Al–Al	MP-P	COD: 72% TOC: 57% Turbidity: 98% Cr: 99%	- Tanning industry consumes and produces a high amount of water and wastewater. - Common problems related to tannery wastewater is related to a high amount of chromium and organic matter.	[54]
		Fe–Fe	MP-P	COD: 69% TOC: 60% Turbidity: 99% Cr: 99%		
Pulp and paper wastewater	Primary	Fe–Al	MP-P	COD: 85% Lignin: 78.5% Colour: 100%	- Pulp and paper industry is considered the sixth-largest polluter of wastewater associated with high organic matter0, blackish colour, COD, BOD, and SS.	[55]
Vinegar industry wastewater	Primary	Al–Al Fe–Fe	MP-P MP-P	COD: 90.91% COD: 93.60%	- Effluent from vinegar wastewater contains high organic pollutant loads.	[56]

Table 4
Previous works using electrocoagulation on landfill leachates

Types of leachate	Stage for EC	Electrodes	Arrangement	Current density	Reaction time	Parameter removal	References
Raw leachate	Primary	Cu/Al–Al	MP-P	20 mA/cm^2	6 h	TN: 32.66% TOC: 34.01%	[57]
		Cu/Al–MS	MP-P	20 mA/cm^2	6 h	TN: 19.54% TOC: 44.36%	
Diluted raw leachate (1:4)	Primary	Cu/Al–Al	MP-P	50 mA/cm^2	6 h	TN: 38.25% TOC: 24.74% NH$_3$–N: 27.18% NO$_3$–N: 97.53%	
				20 mA/cm^2	6 h	TN: 88.63% TOC: 32.78% NH$_3$–N: 87.42% NO$_3$–N: 100%	
				20 mA/cm^2	12 h	TN: 88.11% TOC: 36.51% NH$_3$–N: 86.89% NO$_3$–N: 100%	
Raw landfill leachate	Pretreatment (Combined with anodic oxidation)	Fe–Fe	MP-P	2.5 A	3 h	BOD$_5$/COD: 0.9% COD: 90%	[44]
Raw landfill leachate	Sequential treatment (EC, EO, PMS/UV/CuFe$_2$O$_4$	Fe–Fe	MP-P	20 mA/cm^2	50 min	COD: 60% TSS: 98% TOC: 40% Ammonia: 30% Colour: 85%	[58]
		Al–Al				COD: 50% TSS: 89% TOC: 36% Ammonia: 40% Colour: 80%	
Bio-filtrated landfill leachate	Tertiary	Stainless steel-Al	Recirculation mode	8 mA/cm^2	20 min	COD: 70% TOC HA: 100% FA: 57% Hyl: 60%	[17]
		Stainless steel-Fe		10 mA/cm^2		COD: 65% TOC HA: 100% FA: 37% Hyl: 46%	

4 listed the research for EC on a different phase of landfill leachates. According to Deng et al. [49], EC is considered an electrochemical standalone process that can treat water and wastewater efficiently. In recent years, researchers integrated EC with various treatments to increase pollutant removal and increases the capability of EC utilisation.

Nanayakkara et al. [57] in their studies introduced a copper layer on the surface of aluminium electrodes for removing nitrogenous compounds from the landfill leachate. The combination Cu/Al anode combined with aluminium and mild steel cathode, however, provides low removal of total nitrogen and total organic carbon for removal of raw leachate below 45% for both parameters. The low removal of parameters, especially ammonium-nitrogen (N) is possibly due to the low oxidation ability of aluminium. The author applied a similar treatment for diluted raw leachate, which showed a significant improvement for all parameters related to ammonium-nitrogen except for total organic carbon. The presence of chloride ions as an active chlorine is an important factor to enhance the removal of ammonium compound, however, it also reduces the current efficiency, low oxidation of Al anode, and limit mass transfer for total organic carbon removal. The author also suggested that the raw leachate is pretreated and EC is used as a secondary or tertiary step to increase the efficiency of removal.

An integrated treatment of electrochemical treatments, involving EC is another key idea to improve the removal of organic compounds in wastewater. Ghanbari et al. [58] introduced an electro-oxidation and PMS/UV/$CuFe_2O_4$ process along with EC to treat highly recalcitrant landfill leachate. The sequence of this integrated system started with EC, electro-oxidation and finished by PMS/UV/$CuFe_2O_4$ process. As EC is the starting point for the treatment, EC managed to remove pollutants ranging from 80%–90% COD, 89%–98% total suspended solids (TSS), 36%–40% total organic carbon (TOC), 30%–40% ammonia, and 80%–85% colour using both types of electrodes. EC simply reduces the burden of pollutants in wastewater and other treatments. The combination treatments that resulted in the overall removal of 95.6% COD, 90.5% TOC, and 99.8% ammonia could be considered highly efficient. Fernandes et al. [44] utilised EC as a pretreatment before being treated using anodic oxidation. The pretreatment seemed to reduce the organic load and enhanced the biodegradability index via COD/BOD_5 ratios.

The mixture of organic, inorganic, and microbial pollution substances remains burdensome for wastewater treatment, including its presence in landfill leachate. The properties of the landfill leachate that changes from time to time require a proper treatment technology. The biological treatment is not flexible for different range of landfill leachate ages, which see only young leachate works effectively using this kind of treatment. Biological treatment offers the removal of biodegradable organic compounds and inorganic pollutants simultaneously and effectively. Even though the high removal of pollutants could be achieved, recalcitrant organic compound, such as humic substances could remain in a large amount and potentially form as a precursor for undesirable organochlorine by-products once released into the water stream. Besides relying solely on biological treatment, a hybrid treatment combining

biological and non-biological treatment could provide an extensive and wider range of pollutants removal. Dia et al. [17] implemented the combination of biological treatment with EC for landfill leachate treatment. Raw landfill leachate was treated using an aerated trickling bio-filter composed of peat and wood chips before being handled by EC. The purpose of a biological treatment is to remove nitrogen, biodegradable organic compounds, and turbidity simultaneously, while EC aims to remove colloids, suspended solids, and other high-molecular-weight compounds. The hybrid treatment can remove high COD as well as TOC of humic substances using a combination of stainless steel with aluminium and iron as electrodes. The removal of humic substances is directly influenced by the presence of humic substances, especially humic and fulvic acid which are abundantly available in wastewater.

As stated by Ghernaout et al. [31] and Huang et al. [59] during the electrochemical process, such as electroflotation, electro chlorination, and electrodisinfection, the production of chlorine species might result in the formation of harmful disinfection by-products (DBPs). Oxidation of available chloride ions into chlorine gas and formation of active chlorine species (free chlorine and combined chlorine), which co-exist with organic compounds will form carcinogenic by-products (CBPs) [60]. There are five classes of DBPs involving halocetic acid (HAA), trihalomethanes (THM), haloacetonitriles (HAN), haloketones (HK), and halonitromethanes (HNM). Xu et al. [61] conducted a study on DBP formation and contribution of individual DBP towards the toxicity of wastewater by integrating EC and granular activated carbon using two different sources of landfill leachates (A and B). The EC phase could provide removal of 60%–70% of COD depending on the electrodes pairing and various amount of organic compounds. The initial composition of landfill leachate TOC and COD consists of a combination of humic and fulvic acid at almost 90%. Organic constituents are measured to determine the degree of aromaticity for each fraction. Humic acid dominates the leachate and contains more aromatic rings compared to fulvic acid and hydrophilic compounds. EC effectively removes the organic compound using any pair of electrodes. All three organic compounds have different molecular weights and surface charges, which influence the removal selection. The higher molecular weight of humic acid results in a higher ability to interact with metallic hydroxides and co-precipitate compared to others.

2.3. Disadvantages of electrocoagulation

Even though EC had been successfully applied to various kinds of wastewater, several issues regarding the efficiencies of EC treatment need to be addressed. A common problem associated with EC treatment is related to the passivation of the electrode that reduces the efficiency of EC treatment. Passivation is related to the formation of an oxide layer on the surface of the electrode that inhibits the anode dissolution from anode and electron transfer from the cathode [62,63]. The formation of this layer will increase the resistance and indirectly increase the power consumption as well as the overall cost. Due to this problem, several approaches and modifications have been

carried out to overcome this problem, such as ultrasonic waves, alternating pulsed current, electrode polarity reversal, chloride ions addition as well as modification of the electrode [62]. Cleaning of the electrodes may provide a reduction in electrode passivation. Other factors include improper and unsystematic reactor of EC cells. Various reactor shapes and configurations have been suggested in various research, but unfortunately, none are ideally suitable to all types of wastewater, and require modification based on research interest. As the electrodes are being used repeatedly, periodical change of the electrodes is required, as the thickness of electrodes reduces, and the efficiency decrease. In some cases, the presence of chloride ions in wastewater causes production of toxic chlorinated organics [64]. Landfill leachate, which contains a high amount of humic substances of humic and fulvic acid could eventually produce trihalomethanes. The wastewater suspension should be very conductive. The overpotential generated by the suspension resistance can be reduced by increasing the conductivity of the suspension using a supporting electrolyte. Chloride ions, as an example, are advantageous because they can break the passivating layers at the anode via the pitting corrosion mechanism [65]. Above all the limiting factors, the cost of EC treatment is the main focus and those limiting factors will directly increase the operating cost, which is essential especially for scale-up purposes.

2.4. Factors affecting electrocoagulation

The EC process is influenced by various factors that need to be tuned into optimal conditions. Some of the factors depend on each other, and thus makes EC a complex system. The optimal performance for EC varies for different types of wastewater, according to their composition and parameter of subjected removal. Common factors for EC involve electrode material, pH, interelectrode distance, temperature, electrode arrangement, and design of the reactor. Current density and electrolysis time is considered as the controller for coagulant production.

2.4.1. Effect of electrode material

The selection of material for sacrificial anode is crucial for any EC process. The anode used will determine the type of ionic metallic species released in the treatment process. According to Garcia-Segura et al. [32], the higher charger valence metal-ionic coagulants were normally used as they posed higher electrical double-layer compression that promoted colloids aggregation. Table 5 listed several types of anode material and their advantages, as described by researchers worldwide.

Anode dissolution, pollutant removal efficiency, and coagulant quantity are all significant characteristics in terms of EC effectiveness. They have a direct link to the metal species that have been discharged. As a result, metal ion coagulants with a larger charge valence are favoured in this situation because their electric double-layer compression effect is stronger, improving pollutant coagulation efficiency. Higher oxidation potential can be used as a guidance for electrodes selection, especially to achieve higher oxidation and coagulant rate. Generally, researchers choose Fe

and Al anodes over other electrode materials because they can use the flocculation characteristics of the generated multivalent metal ions. Iron and aluminium are two commonly used electrodes due to low purchasing cost and can be easily found commercially, especially for large-scale purposes. Both electrodes pose easy maintenance and are widely accepted in various kinds of wastewater. However, that does not mean that other metal electrodes cannot pose similar removal efficiency as the removal efficiency will depend on the type of wastewater and targeted pollutants removal. The electrode material chosen has a significant impact on the electrochemical reactor's performance. The likes, such as graphite and lead (IV) dioxide (PbO_2) are cost-effective and readily available, however, both showed low durability and create high oxygen evolution over its potential. PbO_2 also produces high toxic Pb^{2+}, which leads to the generation of secondary pollution [47]. The electrodes IrOx and Ti/IrOx–Ta_2O_5 are highly costly. Although Ti/PbO_2 and Ti/SiO_2 anodes can produce identical average Faradic yields, the latter is favoured because of its superior capacity to oxidise hazardous chemicals. Metal dissociation is related to the pH of wastewater. Electrode, such as iron electrodes may dissociate into various monomeric or polymeric hydroxo complexes, and adsorb the pollutants. In landfill leachate cases, various electrodes have been used to determine the removal efficiency. Malinovic et al. [70] compared the effect of iron and aluminium anode on the removal of total dissolved solids (TDS) and turbidity. The verdict of the research showed that iron electrodes achieved higher removal of turbidity and could provide better removal of TDS at low current densities. However, the researcher also tried replacing the iron cathode with stainless steel, and interestingly the removal efficiency increased. The combination of electrodes could be considered as an alternative to improve the removal efficiency. In other research, Mahmad et al. [71] found that aluminium and stainless steel managed to provide efficient removal of total chromium, colour, and turbidity of landfill leachate. Aluminium showed 99% removal of colour and turbidity, while stainless steel managed to remove 88% of chromium. The material of electrode should be suitable for the targeted removal of pollutants for EC.

2.4.2. Effect of pH

The pH hugely influenced performance of an EC reactor. The efficiency of pollutant removal will rely on the pH value, as different metallic ions favour different conditions for optimal removal. The tendency for ions to form metal hydroxo-complexes and polymeric species hugely depended on the initial pH of wastewater [53]. The variation of metallic ions presents different interactions with pollutants in wastewater. As reported by Xu et al. [72], the electro-dissolution of electrodes would increase the presence of hydroxide ions, and thus increased the pH of the treated wastewater. During the EC process of an initially low pH of wastewater, the hydrogen evolution at the cathodes would create a disturbance and enhanced the release of CO_2 from the wastewater. In alkaline wastewater, fewer changes that occur cause only a small drop of pH. This scenario could be described that the EC system could act as a pH buffer due to the balance provided between the

Table 5
Electrode materials and descriptions

Materials	Descriptions	References
Iron	- Coagulating properties of multivalent ions; - Widely accepted in the water treatment process; - Easily available; - Low-cost maintenance; - High electrode dissolution rates; - Involved in two complex interactions due to two ions caused by Fe^{2+} and Fe^{3+} (divalent or trivalent); - Low toxicity; - Requires a lower dosage than aluminium; - Forms heavier flocs; - Wide range of pH for operation.	[32,66]
Aluminium	- Widely accepted in the water treatment process; - Forms trivalent ion; - pH neutraliser.	[18]
Stainless steel	- Commonly used, besides iron and aluminium; - Highly effective; - Cheap and readily available.	[67]
Magnesium	- Highly effective for nutrient recovery, especially phosphorus; - Improved electrocoagulation stabilisation.	[68,69]
Inert	- Handling with wastewater that has a concentrated amount of calcium or magnesium ions; - Great chemical resistance.	[67]
Dimensionally stabilised anode (DSA)	- Great chemical resistance; - High efficiency; - Cost-effective and easily available.	[47]

generation and consumption of hydroxide ions. According to Moussa et al. [18], three keys areas were influenced by pH, which were the solution's conductivity, zeta potential, and electrode dissolution. Garcia-Segura et al. [32] found that neutral and alkaline conditions were more preferred for the coagulation process. However, studies from different researchers proved vice versa, which showed EC was more efficient in acidic conditions. Sahu et al. [47] stated that the formation of effective coagulant species ranged from acidic, neutral, and until slightly alkaline pH, depending on the electrode material. The optimum conditions for EC are still debatable with large gaps that still need to be filled. Asaithambi et al. [73] reported that the performance of EC dropped when the pH shifted towards acidic or alkaline. The author suggested that the precipitation of hydroxides at the cathode presumably contributed to the high removal efficiency. In the cases of landfill leachate, the leachate ages will reflect the pH and the organic constituents within it. The volatile organic molecules dominate the leachate throughout its early life (acidogenesis), keeping the pH low, generally less than four. However, due to the actions of the methanogens, the pH of the landfill shifted towards alkaline (above seven) as it matures. The effect of the initial pH is very crucial for the treatment of landfill leachate using EC to ensure optimum removal of pollutants.

2.4.3. Effect of current density and voltage

Another important parameter in controlling EC process involves the current density and voltage applied in the process. Current density is defined as the current flowing through an electrode per unit active surface. It is directly related to the rate of coagulant and bubble formation, the size and development of flocs as well as the mixing and mass transfer of solution. Click or tap here to enter text [47]. The amount of metal ions dissociates should be proportional to the current density applied. The current density will directly influence the reaction rate in EC by limiting the ion released from the electrode and coagulation rate that also reflects the performance and operational cost of the system. The removal rate of pollutants depends on the available binding site of metal hydroxides, and the number of metal hydroxides produced [74]. The formation of metal hydroxide species subsequently lowers the inter-particle repulsion charge of suspended particles and allows van der Waals forces to promote agglomeration. Simultaneously, the rate of bubble generation is directly influenced by applied current density, and thus affects the mixing and mass transfer in the reactor [36]. Another unique feature posed by the current density is the sedimentation, or flotation of flocs that depends heavily on the weightage of current applied. According to Orkun and Kuleyin [75], the increase in current density applied

would cause a simultaneous increase in dissolution of the anode to increase the formation of the floc. The formation of hydroxides, oxyhydroxides, and metal oxides provided an adsorption surface for pollutants before being removed via electroflotation [52]. The author also stated that the high amount of current density generated high hydrogen gas and smaller bubble size, which provided larger surface areas and increased the flotation rate of flocs formed. However, at higher current density, the turbulence in the electrogenerated gases as well as excess flocs in the reactor, could cause flocs to rupture [11]. The usage of current density beyond optimum level did not produce any improvised result, and created excess flocs. The excessive usage of current might cause the passivation of the electrode, and simultaneously increase the EC voltage. The poor performance of the EC system will directly influence the Faradaic efficiency of the electrodes and the EC system. The Faradaic efficiency, which is the ratio of empirically measured coagulant mass to the theoretical mass predicted by the Faraday's Law, is frequently stated when evaluating EC's performance [76]. It is desirable to have a Faradaic efficiency of 100% or 1.0 computed since it indicates effective current consumption. Faradaic efficiency can be calculated using the equations below:

$$\text{Theoretical amount of coagulant metal dissolved} = \frac{ItM}{zF} \quad (8)$$

$$\text{FE}(\%) = \frac{\text{Observed coagulant}}{\text{Theoretical coagulant}} = \frac{zFm}{ItM} \times 100\% \quad (9)$$

where F (Faraday constant) = 96,485 c/mol; g = mass; I = current density (Å); t = electrolysis time (s); M = molar mass of metal (g/mol); z = number of electron transfer.

In certain circumstances, readily presence of chloride ions in wastewater may enhance the pitting of the electrodes. The corrosion of the electrode's surface may rapidly increase the rate of metallic ions. This scenario will increase the Faradaic efficiency to a value exceeding 100% or 1.0. This phenomenon is called super-faradaic dissolution [Eq. (10)]. The current density usage level at the optimum level should be considered efficient. Usage below or above the optimum level can cause longer treatment time or higher consumption of current. A large supply of current density beyond the consumption may lead to a high ohmic drop between electrodes with an increase in voltage and temperature of the treated wastewater. Excess O_2 productions can be observed at a high stake of current density, which eventually prevents maximum removal of pollutants. However, the current density does not solely influence the performance of EC. Therefore, other factors such as pH, temperature, water flow, reaction time, wastewater, and types of wastewater should also be considered.

$$\text{FE} = \frac{(ZmF)}{(MIt)} \times 100\% \quad (10)$$

where Z = number of electron transfer; m = mass loss of anode (g); F, Faraday constant = 96,485 C/mol; M, molar mass = mass of electrode g/mol; I = applied current (Å); t = duration of EC (s).

The flow of current can either used direct current (DC) or alternating current (AC). DC has been widely applied in various EC treatments, involving different kinds of wastewater. The electrode passivation is identified as one of the common problems, especially when being conducted in DC electrocoagulation system. Electrodes passivation formed an oxide layer at the outer of the electrode preventing the current flow between the electrodes, and increasing the power consumption and operating costs. Passivation reduces the EC efficiency and lowers the Faradic efficiency, which may reflect ineffective treatment results. The drawbacks due to passivation in DC electrocoagulation requires frequent maintenance and periodical electrodes replacement, which leads to higher operating cost. Some research on AC have found that using AC could improve the performance of EC by reducing power and electrodes consumption, while providing greater removal efficiency. Asaithambi et al. [73] found that the implementation of AC in electrocoagulation treatment of distillery wastewater provides a higher removal of colour and COD percentages with a lesser electrical consumption compared to direct EC. The AC resulted in 100% colour removal, 95% COD with only 3.20 kWh/m³ electrical energy consumption, while DC obtained 90.57% colour removal, 86.54% COD removal with 3.50 kWh/m³ electrical consumption. The sludge production and formation of impermeable oxide were lower in the AC system compared to the DC system. The influence of the flow of the current could be observed on the surface structure of the electrodes. As observed by Kamaraj et al. [74] in their study, when the AC was applied, less disordered pores and a smooth microstructure of magnesium were detected, indicating that the magnesium electrodes dissolved uniformly during the electrolysis. The electrode surface on DC-fed electrodes, on the other hand, was observed to be rough, with a lot of dents. These dents arose around the nucleus of the active sites, whereby metal hydroxides were produced as a result of electrode dissolution. The anode material consumption at active areas due to the creation of oxygen at its surface might be responsible for the formation of a large number of dents [77].

2.4.4. Effect of electrolysis time

According to the Faraday's law, the amount of ions generated are highly influenced by electrolysis time, which eventually contributed to the efficiency of the EC process. Different electrode materials will produce different monomeric and polymeric ions for the respective period. The variety and amount of ions are decisive factors that require sufficient electrolysis time to achieve optimum removal of pollutants [55]. The metal hydroxide generation is directly proportional to the electrolysis time. With a constant current density, the amount of metal hydroxide produced increases as the electrolysis time increased. The production of flocs increases as the electrolysis duration increased, resulting in a rise in pollutant removal efficiency [36]. Once the removal efficiency achieved maximum removal, a constant rate pattern could be seen and there was no significant improvement [75]. This could be caused by the limit achieved via sweep coagulation and co-precipitation [24]. The prolonging electrolysis time will

indirectly change other factors, such as pH. The results showed that depending on the anode and cathode activities, pH progressively increased due to the cathode's dominating activities.

2.4.5. Interelectrode gap distance

Another element that impacts EC performance is the electrode distance, which can alter the electrostatic field, reactor size, flow regime, mass transfer inside the electrodes, and energy consumption. The distance between the anode and cathode, which are known as the interelectrode gap is related to the ohmic drop of the reactor due to the solution resistance. Ohmic drop is a major concern to the selection of interelectrode gap distance. According to Sahu et al. [47], the gap between the electrodes was partially filled with gas that evolved in the reaction process. The presence of gas will decrease the gap distance between electrodes and leads to electrolyte resistance Asselin et al. [78] and Wang et al. [79] reported that the ohmic drop potential is directly proportional to the interelectrode distance, and thus influence the cost and energy consumption of the operation. The distance between the electrodes is found to influence the redox reaction rate. According to Bhagawan et al. [80], the decrease in electrode gaps showed high removal rates due to faster anion discharge and an improved oxidation process. The small interelectrode distance allowed high electrical conductivity and better electrostatic field with high metal dissolution for particle neutralisation and flocs formation. However, a too close interelectrode gap could be a disadvantage. The accumulation of bubbles and floc in between both electrodes could prevent the electron transfer between the anode, cathode, and the bulk solution. If the interelectrode spacing is too near, solid and fluid transmission is impeded, resulting in an increase of electrical resistance. Pore sizes for an electrode in close distance electrodes are understandably bigger. The pore size diminished as one move far away. Voltage is commonly related to the interelectrode distance [81]. The voltage drop along the current route in the electrolyte may have caused the pore size reduction. The ohmic resistance between the two electrodes increased when the anode was pushed further from the cathode, raising the system's total voltage [82]. The resistance posed by the solution could be estimated using Eq. (11). From Eq. (11), it is expected that the IR drop, also known as ohmic potential drop, is directly proportional to the interelectrode distance. In real study cases, the applied current should be increased to achieve a constant current input, which was caused by the rising of ohmic loss (IR drop), resulting in inhibition of the anodic oxidation process. When working with low cell voltages that favour the formation of strong, well-settled flocs, the interelectrode spacing should be kept to a minimum of 10 mm; otherwise, the EC might require frequent polarity reversal due to frequent floc deposits caused by the lower level of turbulence between the electrodes [78]. When designing highly active electrode surface area to volume ratio reactors, the interelectrode gap should be sufficient to allow turbulence between the electrodes, hence initiating mass transfer within the EC [47].

$$V = IR \tag{11}$$

$$R = \frac{d}{(Sk)} \tag{12}$$

$$IR = \frac{Id}{(Sk)} \tag{13}$$

where R = ohmic resistance of electrolyte (Ω); I = current (Å); d = electrode distance (m); S = electrode surface (m²); k = conductivity of the solution (mS/m).

2.4.6. Electrode arrangement

The electrode arrangement is directly involved in cost calculation and removal efficiency. Generally, there are three types of electrode arrangements, which are monopolar electrodes in parallel connection (MP-P), monopolar electrodes in series connection (MP-S), and bipolar electrodes in serial connection (BP-S). The preference of electrodes arrangement depends on the operating parameter and also the nature of wastewater. Hakizimana et al. [27] described the characteristics of each electrode arrangement, which is summarised in Table 6.

Table 6
Electrode arrangement in electrocoagulation treatment [27]

Electrode arrangements	Descriptions
Monopolar electrodes in parallel connection (MP-P)	- Electrode arrangement placed at the same anodic or cathodic potential; - Voltage is the same in each pair; - Current of each electrolytic cell is additive.
Monopolar electrodes in series connection (MP-S)	- Internally connected and no connection with outer electrodes; - Electricity passing through is the same; - Total voltage is the sum of individual voltage.
Bipolar electrodes in serial connection (BP-S)	- Two electrodes are connected to the power supply, while the other electrode is arranged in-between the outer electrode; - Outer electrodes are monopolar, while in-between are bipolar; - Bipolar electrodes are not interconnected with each other, and each side acts differently at different charges.

Monopolar parallel modes setting connect the anodes and the cathodes in their connection. The connection ensures that the voltage is evenly distributed amongst the electrodes, and thus reduced the potential difference.

The outermost electrodes of a monopolar series system are linked to a power source, and current flows through the other electrodes, polarising them. Due to electrodes linked in series have a greater resistance, a higher voltage is frequently required for a given current. The electrodes may pose different voltages and the total voltage of the system is the sum of the individual voltage. The internal electrode is considered as 'sacrificial electrodes' due to its function to prevent the passivation of the cathode and reduce the anode consumption [18].

In a bipolar parallel connection, two parallel electrodes are linked to an electric power source with no power connection to the sacrificial electrodes, and system maintenance is relatively straightforward owing to the simplified setup. When current flows through the main electrode, the opposite sides of the internal electrode become polarised and transport opposing charges to surrounding electrodes. As a result, the two outermost electrodes are unipolar, whereas the two innermost electrodes are bipolar. This implies that each internal electrode serves as both an anode on one side and a cathode on the other side.

Generally, no exact arrangement has been identified the best, as the results of the arrangement mostly depended on the wastewater constituent and its operating parameters. Several authors have compared the result of their works using different electrode configurations. Ding et al. [83] identified that the bipolar system performed better than the monopolar system by providing 65%, 100%, and 91% removal for organic pollutants, ammonia, and phosphorus, respectively. In another research, Nasrullah et al. [84] found that monopolar series electrode indicated better removal efficiency compared to others by providing 74%, 77%, and 66% removal for COD, BOD, and suspended solids (SS), respectively. However, the findings also showed that the MP-S arrangement had a higher operating cost compared to others.

2.4.7. Shape of the reactor and electrode

Reactor design and configuration are important for the overall performance of EC as well as forecasting for scale-up purposes. The performance of EC reactor involves operating parameters, such as flow regime, flocs formation, removal yield, and flotation/settling characteristics [47]. The common type of reactor used in research involves plate electrodes that is used in an open batch cell with additional mechanical stirring to allow mixing within the reactor. Various shapes have been used, including box-shaped, rectangular, and cylindrical reactors to provide high removal efficiency. Rectangular cells with the use of rectangular electrodes have been used widely for lab-scale study. The cell normally has an opening on top of the reactor with electrodes dipped into the reactor, which allows easy maintenance and set-up. The system within the EC reactor often involves a continuous or batch system. A continuous system is known for having a continuous flow feed of wastewater within a reactor, while a batch system involves a fixed amount of wastewater within a reactor. EC reactor design

affects EC through the reactor working volume, which intervenes to define electrode area/volume ratio (A/V), and through EC geometry, in addition to electrode arrangement and spacing. When utilising electrode plates, the electrode A/V ratio is the sole key scale-up characteristic in plant design that allows constructing EC full-scale equipment from laboratory tests, while maintaining the same interelectrode spacing. Hakizimana et al. [27] suggested an average of 15 and 45 m^2/m^3 for the range of electrode A/V ratio. When the A/V ratio is increased, both the treatment time and the optimum current density are reduced. The Reynolds number (fluid flow regime) and Froude number were chosen as dimensionless scale-up parameters to assure proper reactor sizing and pro-portioning (buoyancy). Weber's surface tension, gas saturation, and reactor geometry were maintained [47]. Comparing the reactors' performances is difficult because all the reactors have their advantages and disadvantages [25].

Different reactors and electrodes have been implemented in EC. Naje et al. [15] in their study, have been working on a novel rotating electrode reactor. The majority of EC studies used a traditional reactor design with a static electrode and magnetic stirrer to agitate the solution. Due to poor molecular diffusion, inefficient solution mixing limits mass transfer during the process, resulting in the creation of a passive film on the electrode surface. The author found that at 50 rpm, the removal efficiency of COD, TSS, and TDS achieved 94.5%, 95.5%, and 93%, respectively, at 40 min neutral pH state. Proper shape of an electrode can lead to improvement in energy consumption and reduction of operating costs. Khandegar and Saroha [85] demonstrated a study on the effect of electrode shape on the performance of EC. Bhagawati et al. [86] used two-dimensional concentric aluminium tube electrodes (CATE) for treating wastewater using EC. EC manages to remove pollutants at optimum factors that involved pH 7, 30 min treatment time, 12 voltage current with a surface area of 446 cm^2. The electrode was modified by punched holes with different numbers, diameters, and geometry. The result showed that the performance of punched electrode was better compared to the normal plane electrode. The optimum number and diameter of holes also benefitted the EC treatment and could be considered as an option. Novel perforated zinc electrode provides good removal of Pb(II) by 99.9% in wastewater treatment within 10 min of treatment time at 1.13 mA/cm^2 current density using solar-photovoltaic EC [87]. Another research Ibrahim et al. [62] showed that perforated electrodes managed to reduce energy consumption by about 70% compared to typical plate electrodes. As the EC cell had been optimised, organic contaminants, such as TOC, TPH, and oil and grease (O&G) managed to reduce by 97%, 98%, and 95%, respectively. The research works mentioned proved that modification of electrode shape and morphology were able to increase the removal efficiency of EC. However, there is still a gap of knowledge that needs to be addressed to understand the effect of electrode shape as well as the EC reactor.

2.4.8. Effect of temperature

Several studies suggested that temperature affects pollutant removal in EC, either as advantages or disadvantages.

The high temperature exceeded the optimum temperature to reduce the metal hydroxide formation, while low temperature caused a slow anodic dissolution rate in the bulk solution. In general, the temperature shows an increasing trend due to the electrolytic reactions influenced by the contact time, electrode type, and applied current. According to Naje et al. [15], increased in the solution temperature until optimum temperature helped to improve the treatment of landfill leachate using EC. The author described that the solution temperature increased the dissolution of the anode, whereby in this study is the dissolution and hydrolysis of Al^{3+} to $Al(OH)_3$ as well as improved Al^{3+} diffusivity from the anode surface to the bulk of solution according to Einstein's standard equation. The result was supported by El-Ashtoukhy et al. [88], who obtained an improvement of electrode dissolution in bulk solution for phenolic removal of wastewater. However, too high temperature would cause shrinkage of large pores for metal hydroxide gel that would eventually form denser flocs that deposit on the surface electrode [89]. The increase in temperature helps to improve the adsorption capacity of hydroxide flocs for removing contaminants. Adsorption processes, such as physisorption and chemisorption, act differently with the increase in temperature, as stated by Le Chateliers's principle. Theoretically, the weak forces between the adsorbate and adsorbent caused the physisorption to decrease with the increase in temperature, while the chemisorption increased in the process with the increase in temperature. Elnakar and Buchanan [90] identified that the chemisorption process showed a high adsorption capacity for insoluble hydroxide flocs, such as $Fe(OH)_3$, as they require activation energy for chemical reactions to occur and decrease reflecting the desorption effect. The removal of phosphate in a study by Bharath et al. [91] showed an increasing trend from 20°C to 50°C, but beyond 50°C is not preferable as it causes a reduction in the rate of reaction, removal efficiency, and is non-economical. The Joule heating law identified that the temperature will simultaneously increase as a result of voltage increment von Meier [92] and Rafiee et al. [93] stated that the changes in interelectrode distance will directly affect the voltage. The low voltage can also be explained by the selection of electrode material using the standard electrode potential as a reference.

2.5. Mechanism of flocs formation

The EC process involves various multiple reactions taking place simultaneously. According to Mollah et al. [64], the mechanism of EC could be divided into three phases, which were (i) formation of coagulant by electrolytic oxidation of sacrificial anode, (ii) destabilisation of the contaminants, particulate suspension, and breaking of emulsions, and (iii) aggregation of the destabilised phases to form flocs. The formation of coagulants is influenced by factors, such as the applied current and pH. Coagulation reduces the electrostatic forces repulsion between the colloids, while flocculation introduces the chemical bonds interaction between the particles, which later form a large floc network. Commonly, the dissolution of metal ions that have a +3 charge or more tends to hydrate and produce hydronium cation [94]. Hydroxyl ions that are produced

act as a connector between the metal hydroxides and most probably pose a positive charge that can donate another hydrogen ion to hydroxyl anions as well as bond with metals for the polymerisation process [94,95]. The authors also mentioned that the capability and specificity of pollutants aggregation could vary depending on the wastewater constituents. According to Safwat [52], EC could provide an efficient floc formation in a short period, even in various pH states. Electrical charge, porosity, and forms of bonding produced in the hydroxide, or with the contaminants are all important aspects of the generated gelatinous hydroxides [95]. The flocs structure formed is normally fragile and porous, which requires a series of compaction and structuring [52,96] focused in detail on formations of particles or flocs in wastewater via EC treatment, as there are five phases for flocs growth. The series of phases involved are arranged in ascending sequence as follows: (i) aggregation, (ii) steady-state, (iii) breakage, (iv) re-growth, and (v) second steady state. Based on the sequence, the flocs breakeven after a steady-state condition is obtained. The first steady-state is achieved when the flocs achieved a steady-state, whereby it can no longer agglomerate and the charge has been neutralised. However, as the treatment continued, the flocs became unbalance, cracked into smaller flocs, and thus made the flocs unstable. The continuous generation of coagulant stabilised the smaller flocs by joining the unstable flocs together, therefore forming larger flocs.

2.6. Bubble formation and electro-flotation

Parallel with the EC process, another process that occurs is the electroflotation process. The pollutant will be floating because of natural buoyancy due to the formation of bubbles gas that evolves at both the anode and cathode [97]. The bubbles gas produced could be varied due to several factors, such as current density, voltage, pH, electrodes material, position, and structure of electrode [98]. The evolution of gases (typically O_2 and H_2), as a result of water electrolysis, occurred at both electrodes at sufficiently high overpotentials to allow some coagulated impurities to float to the surface. The presence of impurities, such as chloride will lead to the formation of chlorine gas bubbles to be released [99]. The mechanism of bubble captures relies on selective surface hydrophobicity and hydrophilicity. According to Prakash et al. [100], the generation of an electric field between the anode and cathode, as a result of liquid conductance, led to the production of tiny gas bubbles. Furthermore, the authors stated that electroflotation technique generated bubbles that were tiny and uniform in sizes, which could not be obtained by conventional method.

3. Intensification and advanced electrocoagulation process

EC is still an emerging technology, whereby researchers worldwide are trying to bring out the best from this process. There are various novel studies on EC technology, coupling with other conventional and advanced technologies to combat arising issues on water securities globally. Therefore, this review article discussed a few novel studies that could treat wastewater, especially landfill leachate.

3.1. Intensification of electrode/reactor

A novel study by Muhammad Niza et al. [101] introduced vibration-induced electrode plates into an EC system to enhance optimum ionic transfer between the electrode. The internal resistance due to the arising and evolving bubbles gases at the electrode causes ineffective ionic transfer and uneven dispersion of bubbles throughout the water matrix. A previous study applied physical-mechanical mean of mixing, such as agitation to ensure a total mixing in the reactor. However, an optimum agitation speed is very much needed as the speed below, or above the optimum speed could decrease the flocs' formation due to low ion mobility and collisions of bubbles [102]. The author managed to remove 90% of colour and 35% of COD by applying this concept at optimum conditions, considering a few factors that affected the EC process, including the vibration intensity. According to Ivanov and Ksenofontov [103], the method of intensification is divided into two classes, chemical, and physical intensification. Vibration is considered under physical class, which mechanically promotes the intensification process along with ultrasound and sound. As reported by Nicol et al. [104], an acoustic field superimposed over a fine-particle flotation process might assist to improve recovery in the low size range (less than 20 μm). Vibration may result in the splitting of bubbles into tiny microbubbles, which contribute to higher collision and improves the mixing. A study by Ivanov and Ksenofontov [103] showed an improvement in terms of efficiency up to three times compared to traditional pneumatic flotation.

Alkhatib et al. [105] introduced the novel cylindrical electrode configuration for inducing dielectrophoretic forces in EC and utilising unsymmetrical electrodes to improve the pollutants removal efficiency. The use of unsymmetrical electrodes is expected to generate extra forces, known as dielectrophoresis (DEP) force. DEP force can be defined as the motion of free particles induced by dielectric polarisation in an inhomogeneous electric field. The DEP forces aim to redistribute the charge inside any suspended particle and medium at their interface. The main advantage of DEP is that the force can affect any suspended particles without being influenced by their electrical charge. The DC and AC, which were adopted for this research indicated that the AC topping removal for TP and COD was at 30 min and 4.3 mA/cm^2, respectively. The increase of electrolysis time helps to improve the removal percentages. DC power supply faces cathode passivation as the treatment time increase, and thus affects the performance of EC. DEP is found to have low effectiveness at low current density. At high current density, the DEP forces will push particles further from the inner electrode contributing to more agglomeration as well as preventing the deposition of flocs on the electrode surface. The use of AC-DEP in EC also manages to reduce the weight loss for both electrodes, allowing a longer lifespan and reduces the overall costs. The AC-DEP module achieved optimum removal efficiency for 88.3% of TP, 82.4% of COD, and only 0.02 mg of electrode weight loss. The team also worked on the removal of TOC in primary treated municipal wastewater [106]. In this research, the performance of EC using the DEP effect was compared to a sole EC treatment process. The TOC was found to have an increasing trend with the electrolysis time. However, the EC-DEP was found to have lower TOC removal compared to normal EC at all electrolysis time. The author suggested that the DEP force was found to be minimal, and thus did not affect the TOC removal. Interelectrode distance was found to affect the TOC removal efficiency. Greater distance between the electrodes caused a reduction in metal ions dissociation. The maximum TOC removal was obtained at a distance of 0.5 cm. The EC and EC-DEP obtained 80.5% and 87.7% removal, respectively. The effect of DEP forces is undoubtedly efficient to improve the removal of pollutants in EC and found to be economical in treating landfill leachate.

3.2. Sono-electrocoagulation

The ultrasound irradiation in the EC process, known as sono-electrocoagulation is an AOP that brings promising advantages for wastewater treatment, especially leachate. Sonification utilises the energy of sound to simulate particles in the leachate. The ultrasound produced will create a high-energy microenvironment that is influenced by the frequency applied. An intense mixing is created, and thus enhanced the collision and flocculation. Ultrasonic irradiation can be observed via hydrodynamic cavitation that involves the growth, formation, and motion of micro-bubbles. At a certain point, a certain condition of bubbles leads to a forceful micro-jet liquid that increases the mixing as well as mass transfer. The ultrasound irradiation could lead to two types of bubbles, caused by the radial motion of the bubble that produced stable cavitation (small-amplitude oscillatory) type, or transient cavitation (large-amplitude non-linear type, depending on static pressure in the medium, frequency, and pressure amplitude) [107,108] worked on sono-electrocoagulation of fresh leachate from municipal solid waste for COD and TSS removal. The sono-electrocoagulation was compared to ultrasonic waves and EC treatment and performed the best amongst the three. The optimum removal for COD and TSS was achieved for 98% and 68% at 30 V in 60 min. The effect of operating parameters, such as reaction time and voltage notably boosted the effectiveness of electrode dissolution. An excessive amount of energy produced due to high turbulent pressure waves, however, broke the flocs and decelerated the sedimentation process. The electrical conductivity of the leachate showed a decreasing trend contributed by many bubbles as a result of cavitation. Another research that involved sonification and EC was reported by Asaithambi et al. [109]. The author combined sono(ultrasound)-ozone(O_3)-electrocoagulation in treating landfill leachate, which was described by the colour and COD removal as well as power consumption. The hybrid technology successfully removed colour and COD by 100% and 97.50%, respectively. The influence of various operating parameters, such as initial effluent pH, COD concentration, O_3 production, sonication power, electrolyte concentration, and current density were studied to understand the effect of each parameter on the performance of the hybrid technology. The introduction of O_3 increases the removal of colour and COD. The increase in

the concentration of O_3 allows better mass transfer, and thus increases the efficiency. Cavitational activity was found to increase with an increase in power density due to the higher production of hydroxyl radicals. The situation leads to improvement in colour and COD removal. A further understanding of the concept and mechanism of ultrasound irradiation was described by Torkashvand et al. [107] and Nazimudheen et al. [110]. Besides cavitation, another mechanism induced by ultrasound involves micro-streaming and microturbulence. Micro-streaming is a result of the high-velocity oscillatory motion of fluid elements, which further creates a micro-mixing within the leachate. The third mechanism in ultrasound is known as microturbulence. This mechanism resulted in transient cavitation caused by the generation of acoustic waves of high amplitude pressure, and oscillatory liquid motion in the close vicinity of the cavitation bubble. Overall, the mechanisms stated helped to improve pollutants removal and increase efficiency [110].

3.3. Peroxi-coagulation and electrocoagulation-electro-Fenton-like process

The potential of an AOP to degrade recalcitrant pollutants is taking the research world by storm. The promising potential of oxidants to break down organic pollutants provides an alternative to treat polluted wastewater, especially landfill leachate. EC alone cannot deal with all types of pollutants. Therefore, several researchers suggested a combination of EC and advanced oxidation process [80]. The common oxidants used in the advanced oxidation process were ozone, hydrogen peroxide, and sodium persulfate of Fenton's reagent [103]. Different oxidant poses different oxidation potential for degradation organics pollutant in wastewater. The most used oxidant in treating wastewater worldwide is hydrogen peroxide in a process called the Fenton's process. The application of hydrogen peroxide in an induced electrochemical condition is known as the Electro-Fenton process. According to Alkhatib et al. [105], the combined advantages of the electrochemical and the Fenton treatment method provided a powerful tool to handle concentrated wastewater. The oxidant requires some sort of activation to form highly reactive species from the raw oxidant. Hawari et al. [106] stated three activation methods of oxidant via chemical activation, activation by light irradiation, and activation by ultrasound (US) irradiation. A study carried out by Torkashvand et al. [107] found that the addition of oxidant hydrogen peroxide in an EC process could increase the removal of organic pollutants and reduce energy consumption compared to typical EC. However, the author also acknowledged that there is still a huge gap in understanding the role of hydrogen peroxide in EC, and therefore more research needs to be carried out to optimise this combination. Other studies conducted, such as Afsharnia et al. [108] also acknowledged the combination of the treatment for evaluating the COD removal from the landfill leachate. The author coupled the EC with the Fenton process that is referred to as electro-Fenton (EF) process. The combination managed to obtain a rapid treatment in only 15 min until completion, which took a total of 60 min. The treatment process could remove 74.21% COD at 5,000 ppm hydrogen peroxide and 60 min treatment,

which was considered as optimum operating condition. The author also reported that the EF treatment process is far more superior compared to the conventional Fenton not only in removal efficiency terms, but also cost-wise. The electrocoagulation-Fenton (persulfate) process combination is also used to treat landfill leachate that is pretreated using continuous flow sequencing bed biofilm reactor (CF-SBBR) bio-digested [109]. The trend involved an increased in pollutant removal efficiency with the increase of persulfate concentration. However, beyond the optimal addition of persulfate, the removal efficiency will decline, and treatment undergoes a negative impact. The condition also applies to the current density of the treatment process. Although the combination of treatment could improve the removal of pollutants and its efficiency, there is still a concern about the massive amount of sludge generated by this process that might increase the overall cost of the treatment. The combination between EC and EF-like process also draws attention from researchers worldwide. As reported by Nazimudheen et al. [110], EC coupled with the EF-like process is used to concentrate leachate from incineration facility using a dual anode system. At optimum conditions (7 Ah/L, 7 V, and pH 7), the removal of pollutants achieved 57% removal of organics and 60% removal of ammonia. The removal of organics had been justified by the high removal of suspended solids at smaller interelectrode gap conditions. The organics fraction was represented by the presence of humic acid and fulvic acid. The combination between EC and EF-like process showed a better removal of TOC compared to individual process, either EC or EF. The organic removal according to this study followed the order: EC/EF-like process > EF-like process > EC process.

3.4. Electrocoagulation with activated carbon

The combination between EC and various activated carbon was carried out to increase the efficiency of treating landfill leachate. Hamid et al. [111] proposed a combination of EC and zeolite (clinoptilolite) augmentation for landfill leachate treatment. According to the author, the natural zeolite species, such as clinoptilolite type provides high strength cation exchange and adsorption, which potentially offers great treatment efficiency. The research suggested that 70% ammonia and 88% colour were removed at optimum conditions. The biodegradability ratio (BOD_5/COD) of the leachate also managed to improvise from 0.05 to 0.27. The combination was applied to another research, involving a sequencing batch reactor (SBR) [112]. The reduction of ammonia and colour was determined without any addition of auxiliary electrolytes. The optimum removal of ammonia and colour was achieved at 83% and 95% using a constant 5 h system. Other operating parameters were obtained for the optimum removal, such as 3,750 mL treated effluent, 95 min for reaction and settling, and 110 g/750 mL clinoptilolite loading. Ammonia and colour removal adsorption kinetics fit the pseudo-second-order kinetic model. The author described that the hybrid technology only requires low operating costs with the possibility of multiple cycles.

3.5. Electrocoagulation coupled with biofiltration

The biofiltration process is one of the treatment methods that has been conventionally applied for the treatment

of wastewater, including landfill leachate. Often considered as an efficient technology, especially due to its ability to remove biodegradable organic matter and nitrogen, the advantages of this treatment method are also due to its simplicity and low treatment cost [113]. Biofiltration utilises the immobilisation of bacteria on media, which provides a high surface area for the reaction between the pollutants and microbes. The biological reaction takes place, which eventually degrades the pollutant in the landfill leachate. The performance of biofiltration declines especially for treating mature landfill leachate due to the presence of bio-refractory compounds. Recently, biofiltration has been coupled with EC to improve the efficiency of treatment for the organic and inorganic treatment of landfill leachate. Dia et al. [17] proposed the merging of EC and biofiltration (EC-BF) treatment of landfill leachate by utilising the mixture of peat and wood chips for the biofiltration unit. The findings reported that the EC process alone managed to remove 37% of the initial total COD and 15.1% of TOC. The landfill leachate was then fractionated into three groups based on the polarity properties (HA, FA, and Hyl). It was noted that the humic substances of COD and TOC were slightly different from each other. These results could be explained by the differences of molecular structure between the organic fractions, such as HA poses a limited amount of oxygen and several aromatic rings [114]. Another reason could be described due to the chemical oxidants needed to oxidise these molecules, which resulted in a difference in COD removal. The properties of Hyl fraction that compromised only a small fraction, eventually caused the compounds to easily oxidise. The performance of other pollutants, such as turbidity, true colour, zinc, and phosphorus were effectively removed (82%, 60%, 95%, and 82%, respectively). The following treatment using the biofiltration method managed to reduce the COD and BOD by an average of 42% and 97%, respectively. The author estimated the operating cost of this hybrid process to be around USD1.23M, lower than most alternative processes employed for landfill leachate treatment. Another study by Oumar et al. [115] showed the effectiveness of EC-BR coupling using magnesium-based anode on the removal of COD, ammonia nitrogen, BOD, turbidity, and phosphorus. Biofiltration showed inconsistent and low removal of COD due to the leaching of humic substances from the media. However, in the following treatment using EC, the removal of COD increases rapidly, which is attributed to the mechanisms, such as complexation, co-precipitation, and adsorption of organic matters. The COD was removed at 53% and true colour at 85% at 10 mA/cm^2 current density for 30 min treatment time. The pH showed an increasing trend, but requires at least pH 10 for magnesium ions to start precipitating. As observed, the hybrid combination between EC and biofiltration managed to promote an increase in removal efficiency of pollutants in landfill leachate. However, more research is still needed to fill the knowledge gaps for this combination.

4. Modelling and optimisation

In general, the EC process involves various phenomena that occur simultaneously, which is affected by various factors. Assessment for one single factor at a time is not efficient enough to understand the overall process that occurs. Therefore, EC modelling is an important tool to provide better insight into the EC concept and optimisation. As far as this study is concerned, the modelling that involved EC could be divided into two principal categories, which were statistical modelling and modelling funded on the knowledge. Both modelling has been widely applied in various treatments of wastewater, including EC. Statistical modelling is a tool that is typically used to find optimal operating settings that can improve EC performance. Modelling based on knowledge includes models that used EC as one way as well as others to characterise a physical or chemical process that occurs throughout the technique. The emerging computational fluid dynamics (CFD) modelling studied the fluid flow and current density inside the reactor, and could forecast complicated restrictions and limitations within the process [116]. The author summarised the key modelling, advantages, and limitations for all users in EC, which were adopted from Hakizimana et al. [27] and other research, as shown in Table 7.

4.1. Statistical modelling

The chemical and physical processes that occurred within the EC process as a result of different factors' contributions, commonly are difficult to understand. The theoretical and experimental work could differ due to a few considerations that need to be addressed. Commonly, the works for EC can be carried out by optimisation for each factor, which is considered as an excessive experimental work. The old-fashioned optimisation process, however needs several experimental works for verification and could lead to weak optimisation, such as underestimation or overestimation due to neglecting the mutual contribution amongst the variables [42,75]. The most common statistical model analysis used is the response surface methodology (RSM). The RSM is a tool used to optimise the parameters chosen for the EC process and their interactions [117]. RSM is a regression study that predicts the value of the dependent variable based on the controlled values of the independent variables. In a short amount of time, a large number of experiment combinations may be created, allowing researchers to determine if the investigated parameter has a substantial influence on the study effort. There are various forms of RSM widely used in research, such as partial factorial design (FD), central composite design (CCD), D-optimal design (DOP), and Box–Behnken design (BDD). As explained by Gong et al. [118], FD is commonly used to determine the principal effects of the independent variable as well as their interactions. Other forms of RSM, such as CCD, DOP and BBD are employed for the determination of optimum operating conditions by generating the empirical model, which is represented by a second-order polynomial regression model. The operating conditions required for the optimisation process should include the highest and lowest limit to obtain minimisation and maximisation. The response of the optimisation process can be achieved for a single factor removal or multiple response optimisation. Therefore, the optimisation using RSM should reduce operating costs, while covering adequately the operating parameters.

Table 7
Variation of models, advantages, and limitations [27]

Models	Advantages	Limitations
RSM	- RSM is simple to combine with a techno-economic or environmental study; - Excellent for process optimisation.	- Insufficient for scaling up; - There is no physical backdrop; - Process control is inefficient.
Phenomenological	- Simple to utilise for scaling up; - Likened to kinetically regulated chemical reactions.	- Poor compatibility with complicated pollutants; - Pseudo-kinetic constants are frequently included; - Assumes excellent mixing, settling, and flotation.
Electrochemical	- Estimation of the power input and the amount of coagulant released; - Takes into account electro-oxidation/reduction.	- Prediction of a zeroth-order mechanism in the absence of any additional limiting step (usually occurs only for a short period); - Perfect mixing and settling/flotation are assumed.
Adsorption	- Simple and adaptable (isotherm); - Comparison with a classical adsorption.	- Perfect settling/flotation is assumed; - Only applies if adsorption is the controlling factor; - It is assumed that equilibrium exists.
Complexation	- Simple and adaptable (sequential and parallel responses); - Chemical reactions that are thermodynamically regulated are analogous.	- Perfect settling/flotation is assumed; - It is assumed that equilibrium exists; - The complicated mechanisms postulated may not always correspond to the real mechanism.
VOK	- Model that is dynamic; - It is versatile since it can account for both equilibrium and kinetic limiting stages.	- Assumption of complete flotation/settling; - A limiting step should be selected.
Flocculation/Settling		- The description of hydrodynamics is required for orthokinetic flocculation.
CFD	- Based on basic principles; - Mixing and mass transfer are taken into account as well as electrochemistry; - Adapted for the aim of scaling up; - For techno-economic analysis, correlations can be derived.	- Simulations might take a long time to complete; - The impact of microbubbles and particles is frequently overlooked; - Models of flotation and settling should still be linked with hydrodynamics; - When compared to simpler models, coupling with cost estimation is more difficult.

4.2. Modelling based on knowledge

4.2.1. Phenomenological models

The phenomenological model involves the EC kinetics, which is used in various works for simulation of EC system according to the classical kinetic law [Eq. (14)]. The removal kinetics for pollutants may vary from one to another, which could be recognised as an *n*-order kinetic model.

$$\frac{dc}{dt} = KCn \qquad (14)$$

where C = initial concentration of the pollutant; K = reaction rate constant; n = order of the reaction.

4.2.2. Modelling detailed mechanisms

4.2.2.1. Electrochemical phenomena

Electrochemistry is the fundamental science behind the complex EC process along with charge transport, electrochemical kinetics, knowledge of electrodes interface, and thermodynamics [118]. The EC system remains in equilibrium unless there is applied potential across the electrodes. However, the applied current should overcome the equilibrium potential difference, anode and cathode overpotential, and ohmic potential drop of the solution [119]. The reaction rate should depend on the electrical charge transfer on the electrode interface.

4.2.3. Adsorption and VOK model

Adsorption has been widely studied in various treatments of wastewater, including EC. Terms, such as adsorption isotherm and adsorption kinetics models have been focused to provide sufficient knowledge on the EC mechanism and aid for modelling. It is necessary to obtain sufficient knowledge on the adsorption mechanism, which can be used for pollutant abatement steps. Langmuir, Freundlich, and Langmuir-Freundlich models are familiar adsorption isotherms that are used based on the basis of thermodynamic control. According to Gong et al. [118], Langmuir isotherm can be assumed as a monolayer deposition of the adsorbate on the homogenous adsorbent surface,

while the Freundlich isotherm is not for the reversible adsorption, which is followed by the multilayer formation. Adsorption kinetics models are another way around to determine pollutant removal. The adsorption is known as pseudo-first-order and pseudo-second-order kinetic models. The pseudo-first-order kinetic model suggests that the rate of adsorption site occupancy is proportional to the number of available active sites, whereas the pseudo-second-order kinetic model explains adsorption equilibrium.

4.2.4. Flocculation modelling

The flocculation modelling rises from the acts of two colliding particles adjoining together forming a floc network. The flocculation process involves two stages, which are motion leading to collision and the particle attachment to become flocs. The first stages of flocculation are normally best described by either Brownian motion of the particles (perikinetics flocculation), fluid motion (orthokinetic flocculation), and differential settling velocities due to gravity (differential sedimentation), while the attachment floc is normally an act due to interparticle forces. Flocculation can occur after particle-cluster and/or cluster-cluster aggregation. In the case of cluster-cluster and irreversible perikinetic aggregation, two limiting regimes have been identified: diffusion-limited cluster aggregation (DLCD) and reaction-limited cluster aggregation (RLCD) [120]. The former happens when the repulsive barrier between particles is low, resulting in looser, more tenuous structures; the latter happens when the repulsive barrier between particles is high, resulting in a more compact and stronger structures [30,121]. However, the time consumed for the particles or clusters to reach each other limits the DLCD aggregation. The Fractal dimension has been successfully used to study the flocs growth by analysing using the scattering, image analysis, and sedimentation velocity. The dimension has also been used to determine the flocs geometrical characteristics, and define the aggregation regime that regulates flocculation.

4.2.5. Flotation and settling

During EC, the flocs formed may be removed either by electroflotation or sedimentation. Electroflotation occurs due to the gas that evolved at the electrodes produced micro-bubbles formation, which adsorbs and rise to the surface of the wastewater. The formation of microbubbles ranging from 20 to 50 µm is affected by several factors. Current density is normally dubbed as the main contributor to the efficiency and characteristics of microbubbles, but the contribution of other factors should still be considered. Even though electroflotation has been able to remove a major fraction of flocs, the gravitational settling of flocs still can occur and none of the two machines can avoid each other.

4.2.6. Complexation

The complexation of metal hydroxide with suspended material is a result of adsorption equilibrium that occurs during the EC process. The complexation model is a new and promising phenomenological model used for wastewater treatment, including EC. Two models were proposed for the complexation process, the 'overall model' and 'contribution model'. The former model was applied with the assumption that the waste is mixed in the treatment process regardless of the origin of the affluent, while the latter model considers the separated contribution of the species.

4.2.7. Modelling utilising computational fluid dynamic

The flow and mixing are important for the design of an EC reactor. Computational fluid dynamics turn out to be a promising tool to understand the hydrodynamic and residence time distribution. Both data are important to identify the flow pattern and forecast the features of EC, such as velocity profile, reaction rate distribution at the electrodes, and the cell voltage. The CFD model simulates the process using various numerical solutions of the equation of motion, electrical potential, energy, and current distribution in the flow geometry. Another set of equations defined the transport mechanics (diffusion and migration), including the effects of electric field and reactions of chemical species. Laminar and turbulent flow are two common flow patterns that govern the EC process. According to Azargoshasb et al. [122], the distribution of current and potential could be divided into three, which were primary, secondary, and tertiary. Due to the low overpotential and concentration gradient, the primary current/potential distribution did not account for charge transfer on the electrode. The overpotential of activation, whereby charge transfer occurs on the electrode surface, is considered in secondary current/potential distribution. Since the current density is still low in contrast to the limiting current specified by diffusional mass transfer in the electrodes' mass transfer boundary layer, the overpotential of concentration is ignored [123]. Inside the EC cells, several hydrodynamic phenomena, such as channelling, internal recirculation, and dead zones were identified. Cell geometry is responsible for this complicated behaviour. CFD models and secondary- and tertiary-current distribution calculations may be used to investigate mass transport in the electrolyte and reaction kinetics at the electrodes as well as a starting point for experiment development. CFD can potentially handle multiphase flows, such as those combining a dispersed solid and a dispersed gas phase as well as particle and bubble size distributions using a population balance method [122]. However, several limitations had been identified, especially the need for an expert for various simulated considerations. Due to the overall CPU and memory limitations of multiphase flow simulations, CFD uses imprecise assumptions on the physics of the process, ignoring the influence of hydrogen microbubbles and solid particles on the flow.

5. Conclusion

EC poses similar concept, such as coagulation/flocculation with an improvement through the in situ electrochemical dissolution. EC provides an improvement to conventional treatment with several advantages. However, extensive research is required to explore the potential and optimised the output of EC, especially for landfill leachate treatment. Future research should focus more on optimisation,

intensification and the advanced of EC. Besides, establishing a proper systematic approach, such as modelling would benefit future research. By having this type of model will accommodate the industry with future prediction for operational scale-up and pollutants removal, especially when dealing with high recalcitrant organic wastewater, such as landfill leachate. Economic feasibility of an EC treatment, whether as a standalone or combined with other treatment method should be estimated. This is a major criterion for adoption of technology by the industry. As for landfill leachate treatment via EC, the majority of research focused on the small-scale batch reactor. Therefore, future research should demand more on the continuous flow mode, which are more relevant towards industry.

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