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#### REVIEW

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# A critical review of state-of-the-art technologies for electroplating wastewater treatment

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#### ABSTRACT

The discharge of untreated electroplating wastewater could lead to the formation of toxic metal-organic complexes that is harmful for the environment. There are several processes reported to treat electroplating wastewater such as chemical precipitation, adsorption, coagulation and flocculation, ion exchange, electrocoagulation, membrane filtration, advanced oxidation process and biorecovery. These methods provide the possibility to capture different toxic metals and remove organic pollutants. The combination of different processes together has shown higher efficiencies in removing a wide range of contaminants than single treatment systems. The main challenge is identifying the highest treatment performance and most cost-efficient process. This review provides an overview of treatment technologies for electroplating wastewater and describes the strengths and drawbacks among them. The efficiency of different systems and kinds of pollutants removed, along with a comparison between these systems have been analysed. When comparing the treatment technologies, it is hard to identify the most efficient one as each method has been successful in removing certain pollutants that has not been recovered from the other methods. Therefore, making the decision on identifying the best treatment technology depends on the objective of the treatment and the targeted pollutants. Furthermore, the cost of different technologies has been evaluated based on literature to have a brief look at cost differences between the technologies. When targeting removal of heavy metals, the highest cost of treatment has been reported at around 4.45 \$/m<sup>3</sup> for the adsorption method employing nano zero valent iron (nZVI). While the lowest cost of treatment has been reported at about 0.35 USD/m<sup>3</sup> employing an adsorption method followed by electrodeposition.

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# 1. Introduction and background

Electroplating refers to the process of applying a metal coating onto a metal by hydrolysis to prevent corrosion of the metal or for decorative purposes[1]. The electroplating industry is pivotal to today's community and our modern way of life. The industry plays a crucial role in supporting key industries that play a large part in shaping the economy. Electroplating is used to produce a variety of components for numerous industries including electronic, automotive, aerospace and petrochemical industries [2]. Electroplating also enables manufacturers to reduce production costs by reducing the required amount of expensive raw materials where the expensive metal is only required for coating the less expensive material (metal of plastic) [3]. The plating is normally applied on barrels or racks to ensure an efficient process, especially when plating many products. The anode is the desired metal coating so it could be either an inert anode (carbon or platinum) or a sacrificial anode, and the cathode is normally the piece to be plated [4]. Both the anode and the cathode are placed in an electrolyte solution such as cyanide solution or hydrochloric acid solution [5]. Lamentably, the electroplating industry is a contributor to some of the main environmental issues facing the community; water guality deterioration and the future scarcity of natural resources.

Typically, electroplating industries produce a large amount of wastewater while washing the electroplated parts [6]. This wastewater is highly toxic since it contains high concentrations of metal ions [7]. The exact composition of the wastewater depends on the materials used in the electroplating process. However, it usually includes hazardous metal ions such as copper (Cu), nickel (Ni), chromium (Cr) and lead (Pb) [8]. These ions are classified as toxins and carcinogens; a characteristic which is amplified by the fact that they do not degrade by natural processes [9]. It is estimated that about 2–20% of the chemicals and metals used in the process are washed away and end up in the generated wastewater [10]. Improper treatment and disposal of electroplating wastewater may lead to direct contamination of water bodies resulting in a plethora of health and environmental issues.

Human exposure to toxic metals is perilous where even low concentrations of metals have been shown to cause adverse impacts on the human health [11]. Although the health impacts vary based on the quantity and duration of exposure, they include immunosuppression, nerve damage, organ damage and in some cases, death [12]. The ingestion of toxic metals by living organisms also gives rise to health issues and inevitable damage to the organism including deformities, reproduction issues and death [13]. The persistence of the metals in the ecosystem also lead to the accession of their concentrations through the food chain, also known as bioaccumulation, which leads to further exaggeration of the impact of heavy metal exposure, especially in larger animals [14]. Thus, the wastewater must be sufficiently treated to remove toxic metals before it is released to the environment [7].

Additionally, and aside from the deterioration of water quality caused by the generation of electroplating wastewater, the industry also has an effect on the global natural reserves [15]. The industry is a heavy consumer of natural metals and ores and is a contributor to the diminishing reserves of high-grade ore. One example of diminishing resources is the reduction of copper concentration in natural copper ore from 4% in the early 20<sup>th</sup> century to 1% currently due to excessive mining [16]. Therefore, nowadays, a growing body of research is investigating the feasibility of incorporating resource recovery measures in the treatment of electroplating wastewater. Successful metal recovery from wastewater would allow the utilisation of industrial waste water as an artificial ore, which, in turn, would reduce reliance on natural resources.

The current conventional methods used for electroplating wastewater treatment include chemical precipitation, adsorption, coagulation and flocculation, ion exchange, electrochemical treatment, and membrane filtration [4]. However, these methods have been criticised for their relatively high capital and operational costs, excessive sludge production and the difficulty of metal recovery after treatment [10]. Advanced oxidation processes (AOPs) are novel methods that can be used for treating electroplating wastewater since they accelerate the degradation and the oxidation of a wide range of contaminants that are resistant to conventional treatment methods [17]. Metal bioreduction is a green method proposed by many researchers in recent years to overcome the main drawbacks of the conventional and AOPs treatment methods including the high capital and operational costs, generation of sludge that requires further treatment, slow recovery process, and low selectivity of precious metal [4].

The treatment of industrial wastewater, generally, and electroplating wastewater, specifically, is crucial for achieving the Sustainable Development Goals (SDGs). It has a direct contribution to SDG 6 on Clean Water and Sanitation and specifically on target 6.3 on water quality and wastewater. Efforts to achieve SDG target 6.3 also contribute to other targets such as those on drinking water, sanitation, water-related ecosystems, and sound chemical management (SDG targets 6.1, 6.2, 6.6, and 12.4) [18–20]. Wastewater treatment also indirectly contributes to most of the SDGs as improving water quality would result in the promotion of the overall wellbeing of the community and enable them to target the remaining SDGs. This paper aims at providing an overview of electroplating wastewater treatment through exploring potential challenges to treatment and critically reviewing the available technologies to highlight their main advantages and disadvantages. The cost of each treatment method is also elaborated on in this study. Finally, the study concludes by establishing how the different treatment technologies differ and the criteria upon which the appropriate technology can be selected.

#### 2. Electroplating industry wastewater: challenges

Wastewater generated from electroplating industries is highly toxic inhibiting microbial activity for any biological treatment to be performed [21]. The treatment of electroplating wastewater may generate large quantities of metal sludge, a highly toxic and hazardous waste [22] that could be a source of secondary pollution to the environment [23]. Furthermore, untreated electroplating wastewater could lead to the formation of more toxic metal-organic complexes [24]. The quantity and quality of electroplating wastewater is complex and uncertain that results in incomplete removal of toxic metals during treatment process [25]. Moreover, the treated wastewater cannot be reused again in the electroplating process [26]. The removal of toxic metals at low concentrations, below 10 mg/L, is a complex and difficult process [27]. Rapid development in the plating process introduces other waste types, which requires continuous consideration of more efficient treatment alternatives [28]. Toxic metals in alkaline media, where pH is greater than 12.5, do not precipitate and usually requires the addition of expensive precipitators and resins

[29]. The discharge of both acidic and alkaline plating wastewater is responsible for corrosion of concrete structure and clogging of sewer systems due to the suspension of impurities [30].

# 3. Treatment methods

Different methods are used for electroplating wastewater treatment. These methods include physical treatment, chemical treatment, bio-recovery, and a combination between different mechanisms (Figure 1).

#### 3.1. Physical treatment

#### 3.1.1. Electrocoagulation

Electrocoagulation (EC) is the process of producing metal ions such as iron (Fe) and aluminium (AI) (as indicated in the Table 1) in the water by applying DC voltage to electrodes that allow the neutralisation of electric charge and thus remove pollutants (emulsified, suspended, and dissolved contaminants) by precipitation or by flotation [31]. The coagulant that is responsible for the neutralisation process is normally



Figure 1. Treatment methods for electroplating wastewater.

	Iron (Fe)	Aluminium (Al)
Anode	$Fe_{(s)} \rightarrow Fe_{(aq)}^{2+} + 2e^{1-}Fe_{(aq)}^{2+} + 2OH_{(aq)}^{1-} \rightarrow Fe(OH)_{2_{(s)}}$	$AI_{(s)} \rightarrow AI_{(aq)}^{3+} + 3.e^{1-}AI_{(aq)}^{3+} + 3OH_{(aq)}^{1-} \rightarrow AI(OH)_{3_{(s)}}$
Cathode	$2H_2O_{(l)} + 2e^{1-} \rightarrow H_{2(gas)} + 2OH_{(aq)}^{1-}$	$2H_2O_{(l)} + 2e^{1-} \rightarrow H_{2(gas)} + 2OH_{(aq)}^{1-}$
Overall	$Fe_{(s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_{2_{(s)}} + H_{2(gas)}$	$AI_{(aq)}^{3+} + 3H_2O_{(I)} \rightarrow AI(OH)_{3_{(s)}} + 3H^{1+}$

Table 1. Reactions in electrocoagulation proc	cess [49,164,165].
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generated in-situ by the formation of hydroxyl ions at the cathode with simultaneous dissolution of metal from the anode [32]. The cathode also produces hydrogen gas that can be stored. The EC process is characterised by ease of operation, no chemicals or pH control required, time-efficiency, effectiveness, and low sludge production [33]. Additional advantages of electrocoagulation include the fact that it combines oxidation, coagulation and precipitation processes allowing it to target a variety of metals, and the limiting of the presence of competitive anions since the addition of metallic salts (ions) is not required [34]. The main drawback of applying this technology in a large scale is the higher power cost, but, the technology has been improved in recent years to reduce electrical power consumption [35]. Recently, several authors [36] and [37] suggest that the EC is a low-cost technology for water and wastewater treatment compared to other technologies. Nonetheless, EC, like any other process, faces some disadvantages [34,38]. Among the most notable of which is that the sacrificial nodes need to be regularly replaces as they are consumed during the process and that metal deposition or passivation could decrease the efficiency of the process or even inhibit it. Moreover, while EC produces less sludge than traditional coagulation, the sludge produced from EC contains high concentrations of toxic metals which is considered an environmental hazard if not treated and disposed of properly.

The performance of the EC process depends on several factors: electrode material, gap between electrodes, electrode arrangement, current density, electrolysis time, pH, temperature, and reactor design [32]. Several electrode materials can be used in EC process, such as aluminium, iron, silver, calcium, arsenic, cadmium, barium, chromium, caesium, magnesium, silicon, sodium, strontium, and zinc [39]. Other materials that can be used include nickel, graphite, lead(IV) oxide (PbO<sub>2</sub>), tin(IV) oxide (SnO<sub>2</sub>), and boron-doped diamond (BDD) [32]. Aluminium and iron electrodes are the most commonly to be used for wastewater treatment due to their low cost, availability, proven effective, and nontoxic [40,41]. Iron electrodes are more preferable than the aluminium one because of its low cost [32]. The gap between two electrodes affects the reactor size, energy consumption, and the total cost of the treatment [42]. Wide gaps increase cell voltage and power consumption, while narrow gaps enhance mass transfer and consume less energy [43]. An electrode arrangement has a great effect on the efficiency of the treatment. Water flow between the electrodes can follow horizontal or vertical direction [32]. Electrodes can be bipolar or monopolar [32]. In bipolar systems, a power source connects the outermost electrodes, and the electrical current passes through the other electrodes. In monopolar systems, all cathodes are connected to each other, and similarly, all anodes are also connected to each other. The monopolar system has been reported to be better than bipolar systems in the treatment of laundry wastewater, oily water, and textile mill wastewater [44-46], while the bipolar system was better in the treatment of slaughterhouse and dairy wastewaters [45,47]. Current density has an effect on bubble generation rates and coagulant dosage, and also has a great influence on mass transfer and solution mixing at the electrodes [32]. At high current densities, the removal of chemical oxygen demand (COD) and colour increase due to increasing the hydroxo-cationic complexes in the liquid. However, a trade-off needs to be performed between operational costs and other parameters, such as flow rate, pH, and temperature, to investigate optimal current density [48]. The pH of the solution has an effect on dissolution of the electrodes, zeta potential of colloidal particles, conductivity of the solution, and speciation of hydroxides

[31]. It has been reported that the optimal pH for electrocoagulation treatment is 3.0 [49]. The temperature change may have positive or negative effects on the removal efficiency depending on the type of wastewater treated [31,50,51].

Djaenudin et al., 2018 investigated the removal of nickel from electroplating wastewater by applying electrocoagulation processes; copper was used as the cathode and aluminium was used as the anode [52]. At pH 7, a treatment time of 90 min, and supply voltage of 5 VDC, a 14.8% reduction was reported for Ni<sup>2+</sup>. The treatment process has shown effectiveness towards the removal of dissolved nickel but not of soluble nickel. Moreover, the optimum removal efficiency was not obtained.

Akbal et al., 2011 studied the removal of Cu, Cr, and Ni from electroplating wastewater by applying the electrocoagulation process [53]. Four different combinations of iron and aluminium electrolytes were tested: Fe-Fe, Al-Al, Fe-Al, and Al-Fe. All the experiments were conducted at a constant conductivity of 4 mS/cm, pH 3.0, and current of 2.0 A. At an initial Cu concentration of 335 mg/L, the removal efficiencies of Cu reached 100% after 30 mins using Fe-Fe electrodes, after 40 min using Al-Al electrodes, after 40 mins using Fe-Al electrodes, and after 60 mins using Al-Fe electrodes. At an initial Cr concentration of 193 mg/L, the removal efficiencies of Cr reached 100% after 15 min using Fe-Fe electrodes, after 20 min using Al-Al electrodes, after 20 min using Fe-Al electrodes, and after 40 min using Al-Fe electrodes. At an initial Ni concentration of 526 mg/L, the removal efficiencies of Ni reached 100% after 60 min using Fe-Fe and Fe-Al electrodes. The removal efficiency was higher than 98% for Al-Al and Al-Fe electrodes after 60 min of contact time. It could be noted that Fe-Fe and Fe-Al electrodes were more effective at removing Ni than Cu-Al electrodes employed by [52].

Adhoum et al., 2004 investigated the removal of COD and metal ions  $(Zn^{2+}, Cu^{2+}, and Cr^{4+})$  from electroplating wastewater by applying electrocoagulation process with aluminium electrodes [7]. At a current density of 4.8 A/dm<sup>2</sup> and an anode surface of 50 cm<sup>2</sup>, the removal efficiency of COD reached 64% after 25 min. At the same conditions, the removal efficiency of  $Zn^{2+}$ ,  $Cu^{2+}$ , and  $Cr^{4+}$  reached 97, 96, and 76%, respectively. The electricity consumptions and electrode were found to be 32 A.h/L and 1 g/L, respectively. The study has shown that EC processes are effective for removing toxic metals from wastewater in a considerable small amount of time. These results are about similar to the results of [53] for removing Cu, whom also employed Al-Al electrodes in their study. However, they showed different results for removing Cr.

#### 3.1.2. Membrane filtration

Membrane filtration is a pressure driven process, where separation efficiency depends on the membrane material's selectivity and permeability [54]. Membrane technology is a simple and flexible process, that requires low operational energy, could be easily integrated with other technologies and has high selectivity and separation efficiency [55]. However, a major disadvantage of membrane technologies is membrane fouling due to deposition of particles within the membrane pores, where antifouling strategies have been employed [56]. There are a number of membrane filtration technologies, which include microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO), where each reject contaminants based on particle size [57–59]. For industrial applications, UF is most commonly used [60]. The performance of the membrane depends on its permeability, surface porosity and membrane's pore structure, hydrophilicity, and antifouling nature [61]. Ultrafiltration requires low operation pressure reducing capital and operational costs and thus considered to be a promising membrane process [62,63]. Nanofiltration membranes are recognised for their versatility, high permeation flux, high removal ratio and low energy demand [64].

Lu et al. 2020 used ion-imprinted membranes (IIMs) to selectively capture palladium  $Pd^{2+}$  in electroplating wastewater [65]. Selective adsorption of  $Pd^{2+}$  was explored by IIMs in the presence of other competitive ions, which included  $Cu^{2+}$ ,  $Cd^{2+}$ ,  $Co^{2+}$  and  $Ni^{2+}$ . The adsorption capacity for  $Pd^{2+}$  was 1.2106 mg/g, while  $Co^{2+}$ ,  $Cu^{2+}$ ,  $Cd^{2+}$  and  $Ni^{2+}$  showed an adsorption capacity around 0.5 mg/g within a contact time of 180 min and  $Pd^{2+}$  concentration of 25 mg/L in a 10 mL solution. The IIMs were tested against ultrasonic treatment for 3.0 h to analyse their stability performance. The membranes were hardly damaged by ultrasonic and showed an insignificant decline in their rebinding capacity. A solution of 1.0 M HCL within a contact time of 180 min was used to regenerate the IIMs. The membranes have shown a high regeneration performance reaching up to 92% after five adsorption and desorption cycles. Moreover, the membranes, after being overused, displayed their capability to treat up to 99.87% of methylene blue indicating the potential for multiple treatments.

Aloulou et al. 2020 employed ceramic ultrafiltration (UF) membranes to remove toxic metals present in the effluent discharged from electroplating industries [66]. The membranes were fabricated by applying the layer-by-layer method and supported by smectite (Sm) nanoparticles and natural zeolite. Four different membranes with four different coated layers, ranging from 4 to 7 layers, were developed (Sm/Z4, Sm/Z5, Sm/Z6 and Sm/Z7), where the letter Z refers to the number of layers. The membrane resistance for each membrane was studied, showing that the membrane with six layers (Sm/Z6) has the highest resistance with no cracks or pinholes found in the membrane. Furthermore, Sm/ Z6 exhibited high textural properties of all the other membranes. Sm/Z6 was studied in two different water samples (EF1 and EF2) with different pollutants composition, where EF1 had a pH of 6.2 and EF2 had a pH of 2.9 at room temperature and different applied pressures ranging between 3 bar and 7 bar. A pressure of 3 bar achieved the highest rejection of pollutants. Sm/Z6 has achieved COD removal of 100% and  $Cr^{2+}$  and  $Co^{2+}$ removal of 80% and 30% respectively, when tested in EF1. While being in EF2, Sm/Z6 achieved COD removal of 96% and Cr and Co<sup>2+</sup> removal of 89% and 60% respectively. Deionised water was used to regenerate the membranes achieving total regeneration for two cycles and then about 10% performance loss. The ceramic membranes has the potential to eliminate almost all COD and Cr<sup>2+</sup> and remove moderate amount of Co<sup>2+</sup> at relatively low operating pressure, even after numerous times of membrane regeneration.

Ma et al. 2020 prepared poly(4-vinylpyridine)-b-polysulphones-b-poly (4-vinylpyridine)/polysulphones blend membranes to uptake  $Pd^{2+}$  found in electroplating wastewater [67]. The membranes were fabricated through phase separation, surface segregation and block polymer self-assembly processes, while using Amphiphilic molecules as the membrane base material. Firstly, the membranes were tested in a solution containing only  $Pd^{2+}$  without the presence of any other ions. A solution with  $Pd^{2+}$  concentration of 60 mg/L was prepared, where 0.05 g of membranes were added at 25°C, pH range between 0.5–4.0, and a contact time of 24 hr. The membranes showed a considerable adsorption capacity of around 103.1 mg/g towards  $Pd^{2+}$  at pH 2.

Moreover, the performance and selectivity of the membranes towards  $Pd^{2+}$  were investigated in the presence of different metal cations and anions. The cations have shown almost negligible rejection rates, while the cations have obtained relatively higher rejection rates. Still,  $Pd^{2+}$  showed the highest rejection rate, which was around 96.8%, indicating that the membranes are highly efficient to selectively remove  $Pd^{2+}$  from electroplating wastewater. A solution containing 1.0 M HCl and 1.0 wt% thiourea was sufficient to desorb  $Pd^{2+}$  and regenerate the membranes. The regeneration process was evaluated after seven cycles showing negligible loss in adsorption capacity and excellent chemical stability of the membranes. The blend membranes had excellent selectivity and removal of  $Pd^{2+}$ , even with the coexistence of different metal ions. The study has also concluded its applicability to perform in wastewater with low concentration of  $Pd^{2+}$ .

Hosseini et al. 2017 fabricated poly (acrylonitrile) nanofiltration membranes to extract  $Ni^{2+}$  and  $Cr^{2+}$  from electroplating wastewater [68]. The membranes achieved the highest rejection rate at about 87% and 83% for Ni<sup>2+</sup> and Cr<sup>2+</sup>, respectively. The study showed that rejection rates of the membranes were enhanced by decreasing porosity and mean size pore through optimisation of the membrane structure. Moreover, the addition of  $TiO_2$ nanoparticles and polyacrylonitrile polymer on the structure of the membrane increasing the rejection rates towards Ni<sup>2+</sup> and Cr<sup>2+</sup>. Noah et al. 2018 has also investigated the capture of Cr<sup>2+</sup> by preparing emulsion liquid membranes (ELM), where wastewater samples were obtained from an electroplating industry [69]. The experiment was performed at an agitation speed of 250 rpm and contact time of 3 min. The extraction process has reached equilibrium at the carrier concentration of 0.04 M and any further increase in the concentration did not effect the removal process. Almost 100% removal of chromium was achieved, while a 0.05 M NaOH solution was 100% efficient at desorbing Cr<sup>2+</sup> from the membranes for regeneration. An increase in the solution concentration has not shown any effect towards the desorption process. ELM has shown nearly similar removal of Cr<sup>2+</sup> as the ceramic (UF) membranes, while both were more efficient than poly (acrylonitrile) nanofiltration membranes.

Nanofiltration (NF) membranes have been employed in three different studies. Wei et al. 2013 investigated the removal of Cr<sup>2+</sup>, Cu<sup>2+</sup> and Ni<sup>2+</sup> using NF hollow-fibre membranes [70]. At an operating pressure of 0.4 MPa, the membrane has rejected 95.76% Cr<sup>2+</sup>, 95.33% Cu<sup>2+</sup> and 94.99% Ni<sup>2+</sup>. Increasing the feed temperature had negligible effect on the membrane rejection rates, while the membrane permeation flux increased. Highest membrane stability was obtained at pH of 2.31. Moreover, Boricha and Murthy 2009 prepared two NF membranes to evaluate their treatment performance on electroplating wastewater [71]. Maximum rejection rates of 94% for Zn<sup>2+</sup>, 93% for Fe<sup>3+</sup> and over 99.4% for SS and TDS have been achieved. It was also recognised that increasing pressure and feed flow rate increased the rejection rates of heavy metal ions. The feed pH had negligible effect on the rejection rates; however, an increase in pH from 2 to 8 has significantly decreased the membrane permeation flux. Wang et al. 2007 considered three NF membranes to remove  $Cu^{2+}$  and  $Cr^{2+}$  from electroplating wastewater [72]. Maximum rejection rates of 96.6% for  $Cr^{2+}$  and 90.0% for  $Cu^{2+}$  were obtained at pH range of 4 to 5. An increase in feed temperature increased the the membrane permeation flux, while it had no influence on the rejection of the metal ions.

#### 3.2. Chemical treatment

#### 3.2.1. Chemical precipitation

Chemical precipitation (pH adjustment) is the process of adding reagent, such as hydroxides or sulphides to react with dissolved metal ions found in the wastewater to form insoluble metal precipitates [73]. Hydroxide precipitation is more preferable than the sulphide precipitation due to the availability and low-cost of its precipitant agents like limestone and lime. However, sulphide precipitation is not amphoteric and can achieve high metal removal [74]. The metal precipitates are normally recovered using coagulation and/or filtration or sedimentation [75]. The Chemical extraction process is normally used for precious metals purification [76]. The main advantages of this method is the low cost and simple operation, while the disadvantages are slow metal precipitation, aggregation of metal precipitates, generation of excessive amount of sludge, and poor settling [73–75,77].

Zainuddin et al., 2019 investigated the removal of  $Cu^{2+}$ ,  $Ni^{2+}$ , and  $Zn^{2+}$  from electroplating wastewater using hydroxide and sulphide precipitation [78]. The optimum pH for hydroxide precipitation was from 8.5 to 9.5 for  $Cu^{2+}$ , from 10 to 10.5 for  $Ni^{2+}$ , and from 9.0 to 9.5 for  $Zn^{2+}$ , while for sulphide precipitation, the optimum pH was 8 for copper and from 10 to 10.5 for nickel and zinc. Sulphide precipitation was more efficient than hydroxide precipitation in the removal of toxic metals. For nickel rinse sample, 68.8% of  $Zn^{2+}$  was removed using hydroxide precipitation, while 93.75% % was removed using sulphide precipitation. For the acid rinse sample, 76.66% of  $Ni^{2+}$  was removed using hydroxide precipitation, while 95.32% was removed using sulphide precipitation. For  $Ni^{2+}$ and acid rinse samples, a 100% removal efficiency of copper was achieved for both sulphide and hydroxide precipitation.

#### 3.2.2. Adsorption

Adsorption is a separation process defined as the mass transfer between pollutant molecules present in water and the adsorbent surface [79–83]. The adsorbate, which is the compound being adsorbed, moves across the adsorbent diffusing into its pores and adhering onto the active sites [84]. Adsorption depends on either physical or chemical interaction between the molecules and the adsorbent surface [85–89]. Electrostatic attraction and Van der Waals forces are the basis for physical adsorption, while chemical adsorption is achieved due to the generation of new bonds between the molecules and the adsorbent [90]. Adsorption isotherm and kinetics could be studied to evaluate the adsorption performance of a specific adsorbent and provide information on adsorption mechanisms [67,91].

Adsorption is a low cost method, rapid, easily operated and has been considered as one of the most efficient process to remove heavy metal ions and is more preferable at low concentrations, while minimising the generation of secondary waste [92]. Moreover, adsorption has the potential to regenerate the adsorbent and restore nearly all of its adsorption capacity [93]. Adsorption efficiency depends on the adsorbent's porosity, surface area, adsorption capacity, water stability, recyclability and selectivity for toxic metals [94]. Manyangadze et al. 2020 noted that the adsorbent should be chemically and hydrothermally stable to be able to withstand adverse environmental conditions [95]. The extent of adsorption also depends on the adsorbent dosage, pollutant characteristics and concentration, pH, temperature, and contact time [96,97].

Wang et al. 2020 addressed the possibility of simultaneously removing heavy metal cations and oxyanions present in electroplating wastewater [98]. They employed pinewood sawdust derived engineered biochar (BC) fabricated with MgAI layered double hydroxide (MgAI-LDH) nanosheets to examine its performance for the removal of Pb<sup>2+</sup> and Cr<sup>6+</sup> from solution. The physical blend between BC and MgAl-LDH considerably improved adsorption capacity for both ions. The maximum adsorption capacity of  $Pb^{2+}$ occurred at pH 7.0 as the electronegativity of the adsorbent's surface had increased. In the case of Cr<sup>6+</sup> removal, maximum adsorption capacity was achieved at pH 2.0. Still, at pH 7.0, a considerable amount of  $Cr^{6+}$  was captured due to the synergy between BC and MgAl-LDH rather than using each material separately. The maximum removal efficiency occurred at a dosage of 1.0 g/L removing 99.8% of Pb<sup>2+</sup> and 72.4% of Cr<sup>6+</sup>. On the other side, adsorption capacity decreased with increased dosage indicating that increased competition in the solution leads to this outcome. Adding to that, the adsorption capacity has increased with increased pollutant concentration. According to the kinetics study, adsorption equilibrium for Pb<sup>2+</sup> was reached after 400 min, while for Cr<sup>6+</sup> it required 1200 min. The study has shown that MgAI-LDH nanosheets were efficient at simultaneously removing  $Pb^{2+}$  and  $Cr^{6+}$ , while being feasible to be applied on industrial wastewater.

Qu et al. 2020 investigated the removal of  $Pb^{2+}$ ,  $Cd^{2+}$  and  $Ni^{2+}$  using an adsorbent named RH<sub>MW</sub>-X, which is a microwave-functionalised cellulose derived from rice husk [99,100]. The experiments were performed on an industrial electroplating industry, which contained Zn<sup>2+</sup>, Cd<sup>2+</sup>, Mn<sup>2+</sup>, Ni<sup>2+</sup>, Cu<sup>2+</sup>, Pb<sup>2+</sup>, Aq<sup>1+</sup>, Na<sup>1+</sup>, K<sup>1+</sup>, Ca<sup>2+</sup> and Mq<sup>2+</sup>. RH<sub>MW</sub>-X achieved substantial removal efficiencies of 99.2, 99.8 and 96.38% for Pb<sup>2+</sup>, Cd<sup>2+</sup> and Ni<sup>2+</sup> respectively. In the case of adsorption capacity, the results were high reaching 295.20, 151.51 and 72.80 mg/g for Pb<sup>2+</sup>, Cd<sup>2+</sup> and Ni<sup>2+</sup> respectively. It was also noted that the adsorbent has successfully reduced the other metal ions below the required standards achieving a removal efficiency of 99.76, 99.84, 98.39 and 99.35% for Ag<sup>+</sup>, Cu<sup>2+</sup>, Zn<sup>2+</sup> and Mn<sup>2+</sup> respectively. In comparison to Wang et al. 2020 with respect to Pb<sup>2+</sup> removal, they both achieved high removal efficiencies. However, RH<sub>MW</sub>-X proved to have higher adsorption capacity than MgAI-LDH/BC. The adsorption equilibrium time was 30 min and the adsorbent worked effectively over a wide range of pH, while the uptake process was analysed to be exothermic. A solution of 0.5 M HNO<sub>3</sub> was successful to regenerate 100% of RH<sub>MW</sub>-X and achieved more than 96% of regeneration after five cycles of treatment. The results obtained from this study show that the adsorbent RH<sub>MW</sub>-X has the potential to effectively eliminate heavy metal ions from industrial wastewater at low cost.

Peng et al. 2019 utilizedraw and calcined electroplating sludge (RES and CES) to uptake nickel from electroplating wastewater [27]. Electroplating sludge (ES) is concentrated with metal hydroxide, which was formed due to the precipitation of heavy metal ions in the electroplating wastewater. The removal efficiency of Ni was dependent on the composition, characteristics and methods of preparation of ES, which included sample pH and pyrolysis temperature. RES and CES both achieved considerable high adsorption capacities of 210.9 and 163.6 mg/g respectively. This proved to be much higher than RH<sub>MW</sub>-X investigated by Qu et al. 2020. In terms of applicability, RES proved to be more economical and is more energy efficient than CES. On the other hand, CES had a higher mechanical strength making it more suitable for developing an adsorption column. Liu et al. 2020 have also recycled ES to prepare erdite-bearing nanorods.

A dosage of 0.3 g/L achieved a removal efficiency 99.7, 99.4, 37.9 and 53.3% for  $Zn^{2+}$ ,  $Cu^{2+}$ ,  $Ni^{2+}$ , and  $Co^{2+}$ , respectively. They indicated that the nanorods were more efficient than activated carbon, polyaluminum chloride, polyferric sulphate and  $Na_2S$ . These nanorods achieved similar removal efficiencies for  $Zn^{2+}$  and  $Cu^{2+}$  as  $RH_{MW}$ -X, but failed to obtain results for  $Ni^{2+}$ .

Hamdy et al. 2019 studied the efficiency of zero-valent iron nanoparticles (nZVI) for capturing a wide range of heavy metal ions, Cr<sup>6+</sup>, Pb<sup>2+</sup>, Ag<sup>+</sup>, Cu<sup>2+</sup>, Ni<sup>2+</sup>, Mn<sup>2+</sup>, Zn<sup>2+</sup>, Fe<sup>3+</sup>, Al<sup>3+</sup>, and Co<sup>2+</sup>, in real electroplating wastewater, obtaining removal efficiencies of 66.7, 91.5, 83.3, 80.8, 17.4, 47.1, 54.6, 94.7, 100, and 42.1%, respectively [101]. Moreover, the treatment process achieved 91.3, 68.3, 94.2, and 98.5% removal of total suspended solids (TSS), chemical oxygen demand (COD), nitrogen (N), and phosphorus (P), respectively. Moreover, selective capturing of  $Cu^{2+}$  was investigated in synthetic wastewater achieving almost complete removal of  $Cu^{2+}$  and an adsorption capacity of 81.3 mg/g using a dosage of 1.6 g/L at pH 7.3, temperature 30 C, contact time of 36 min, and agitation speed of 180 rpm. Huang et al. 2018 has also tested the removal of Cu<sup>2+</sup> using diethylenetriaminepentaacetic acid (DTPA) functionalised chitosan micro-gel (DTCS) attaining a removal efficiency of more than 80% at pH 3.0–5.0, 28 C and agitation speed of 150 rpm [102]. An adsorption capacity of 106 mg/g and adsorption equilibrium time of 1 min were achieved. The study revealed that nZVI has the potential to nearly eliminate TSS, N, P, Fe<sup>3+</sup>, Al<sup>3+</sup>, and Pb<sup>2+</sup> and remove Ag<sup>+</sup>, and Cu<sup>2+</sup> at high efficiency. However, highly concentrated sludge with toxic metals was produced.

Suksabye and Thiravetyan 2012, Kong et al. 2011 and Álvarez-Ayuso et al. 2007 investigated the removal of  $Cr^{6+}$  from electroplating wastewater using different adsorbents [103–105]. Suksabye and Thiravetyan 2012 used chemically modified coir path by grafting with acrylic acid [105]. An adsorption capacity of 196.0 mg/g and complete removal of  $Cr^{6+}$  was achieved at pH 2.0, temperature 30 C, and a contact time of 22h. Kong et al. 2011 tested an adsorbent provided by a Chinese company (CHS-1 resin) at low  $Cr^{6+}$  concentrations [104]. The adsorption capacity obtained was 347.22 mg/g at 25 C and pH 2.0–3.0. They also concluded that 5% NaOH-5% NaCl could recover up to 98.02% of the adsorbent without decreasing its adsorption capacity. Álvarez-Ayuso et al. 2007 studied the possibility of capturing  $Cr^{6+}$  at different concentrations using amorphous aluminium oxide [103]. An adsorption capacity of 78.1 mg/g and a removal efficiency of 90% of  $Cr^{6+}$  was achieved, where it took up to 24 h to capture high  $Cr^{6+}$  concentrations and about 30 min for low concentrations. Dosages between 1–5 mg/l of aluminium oxide were required to attain the highest removal efficiency depending on  $Cr^{6+}$  concentration.

#### 3.2.3. Coagulation and flocculation

Coagulation-flocculation (CF) technology is widely used for the treatment of domestic and industrial wastewater due to its effectiveness in removing suspended solids [106– 108]. After the addition of specific volume of coagulation reagents, insoluble and dissolved particles combine together forming larger aggregates. During the flocculation process, slow stirring and agitation take place to enable the formed flocs to settle out and become sediment [109]. The global market for coagulants and flocculants has been estimated to reach around USD 6.01 billion by 2022, indicating the importance and strong demand towards this treatment technology. There is also a growing interest in natural coagulants to replace inorganic and synthetic ones for achieving sustainable

development goals [110]. A main drawback of CF process is its inability to completely remove soluble toxic metals present in wastewater and may require a follow up by another treatment system [111].

Yang et al. 2019 employed O-xanthogenated chitosan Schiff base (XCTS) and Fe<sup>3+</sup> ions (Fe<sup>3+</sup>/XCTS) to capture Cd<sup>2+</sup> from wastewater [112]. The compound showed to achieve maximum treatment efficiency in acidic media, where pH range between 3.0 and 5.0. At Cd<sup>2+</sup> initial concentration of 5, 15, 25 and 50 mg/L, removal efficiency reached up to more than 99.5%. Shen et al. 2013 prepared four different composite coagulants by mixing polyferric sulphate (PFS) with cationic polyelectrolyte (CP) coagulants of different weight percentages (0, 0.5, 1.0 and 5.0%) [113]. The coagulants were investigated against the removal of cyanide present in wastewater. The compound with 5.0% CP has achieved the highest removal of COD and total cyanide (TCN). The results obtained showed between 95% and 97% removal of TCN and between 50% and 55% removal of COD at a pH of 7.5 and a coagulant dosage of 500 mg/L. Furthermore, the flocs formed from that compound had the highest growth rate, strength factor, and structural density, while having the lowest recovery factor. Chen et al. 2007 investigated the precipitation of Cr using zero valent iron (ZVI) [114]. At ZVI dosage of 41.0 g/L, pH 1.5 and hydraulic retention time of 5.6 min, 99.9% of chromium was precipitated. Fe  $^{3+}$ /XCTS and ZVI were very efficient working in highly acidic media. In conclusion, the three coagulants previously mentioned were highly effective in removing different contaminants from wastewater showing that CF process is promising to treat electroplating wastewater.

#### 3.2.4. Ion exchange

lon exchange processes are known for their low cost, eco-friendliness and resistance to high temperatures [115]. Ion exchangers are preferred due to their high uptake efficiency of toxic metals, high treatment capacity and rapid kinetics [116]. It is also possible to recover and reuse ion exchange resins through a regeneration process [117]. An Ion exchanger could be made using organic polymers or in organic polymers [118]. Furthermore, ion exchangers could be distinguished based on material of composition, which can be natural or synthetic, and functional groups, which can be cationic, anionic, or chelating [119].

Inorganic resins are considered to be more stable at higher temperatures than organic ones and exhibit higher affinity for metal ions [120]. On the other hand, they posses low chemical and mechanical strength and cannot be naturally replenished [121]. Organic ion exchangers are prepared by repeatedly joining together organic monomers, a process called polymerisation, through chemical bond formation to form larger molecules [122]. There are efforts to couple organic and inorganic ion exchangers in order to modify its structure and increase overall stability and selectivity towards specific heavy metal ions [123].

Yan et al. 2020 employed a hybrid system to remove both  $Cu^{2+}$  and  $Cr^{6+}$  from wastewater [124]. Synthetic wastewater was prepared containing 20 mg/L of  $Cu^{2+}$  and 20 mg/L of  $Cr^{6+}$ , while adjusting to a pH of 3.0 using 0.1 M HNO<sub>3</sub>. A TiO<sub>2</sub>-ZrO<sub>2</sub> solid solution was established to remove  $Cr^{6+}$ , reaching a removal efficiency of up to 95%. A commercial cation exchange resin (TP207) was employed to selectively capturphoto-catalysis process using e  $Cu^{2+}$ , where the results showed no change in  $Cr^{6+}$ . A column setup was used to determine the breakthrough curve of TP207. A breakthrough point

was obtained at  $Cu^{2+}$  concentration of 2.0 mg/L. An adsorbent dosage of 1.0 g/L was sufficient to completely remove  $Cu^{2+}$  in 4 h. It was observed that coupling the removal processes together had no effect on the removal efficiency of  $Cu^{2+}$ . However, the adsorption rate decreased due to competition among the different ions. The most optimum combination of the two removal processes with the highest efficiency was observed to be ion exchange followed by photocatalysis with 99.17%  $Cu^{2+}$  and 96.29%  $Cr^{6+}$  removal. 10 wt% of nitric acid was adequate to recover 94.58% of  $Cu^{2+}$ ; but only 41.06% recovery of  $Cr^{6+}$ . The hybrid system presented in this study provides a promising and effective solution for the selective capturing of heavy metal ions present in acidic electroplating wastewater.

Ye et al. 2019 prepared a silica-supported pyridine resin (SiPyR-N4) through in-situ polymerisation to capture and recover Cr<sup>6+</sup> from electroplating wastewater [125]. The influence of time, solution pH, Cr<sup>6+</sup> concentration and coexisting anions on the removal efficiency were examined. Sorption equilibrium was achieved within 5 min of the treatment process, removing 99.3% of Cr<sup>6+</sup>. These results were obtained at a Cr<sup>6+</sup> initial concentration of 100 mg/L, when increasing the concentration up to 200 mg/L, the observed equilibrium time was 10 min. It was concluded that the resin's large specific surface area is causing its high adsorption rate. It was observed that the optimum pH ranged between 2.0 and 6.0, where the maximum removal efficiency was obtained at pH 4.0. The presence of sulphate, nitrate, and chloride ions is common in wastewater, thus, their effect on Cr<sup>6+</sup> removal efficiency was also investigated. A solution of 1.0 M HCL and 0.1 M NaHSO<sub>3</sub> achieved a desorption efficiency of 93% after 5 min of contact time. A salt concentration of 5 mmol/L had little impact on the treatment process, where the resin maintained a removal efficiency exceeding 90%. However, when increasing the salt concentration up to 100 mmol/L, a decline in the removal efficiency resutled with the nitrate ions having the greatest influence. SiPyR-N4 proved to be very selective and effective to remove Cr<sup>6+</sup> at very high rate in acidic media. Yan et al. 2020 also showed nearly similar results for removing Cr<sup>6+</sup> under acidic conditions but at a much lower rate [124].

Dai et al. 2015 investigated the removal of  $Cr^{6+}$  from electroplating wastewater using fibrous weak anion exchanger (FFA-1) [126]. The fibrous ion exchanger is recognised for its rapid adsorption and desorption rate, high adsorption capacity and exceptional osmotic capacity. A column system was arranged with the anion exchanger and facilitated at lab and pilot scale to evaluate its Cr<sup>6+</sup> removal efficiency. The system was tested against synthetic wastewater, prepared by dissolving different concentrations of potassium dichromate (K<sub>2</sub>Cr<sub>2</sub>O<sub>7</sub>) in deionised water, and real wastewater obtained from an electroplating factory. The anion exchanger showed an increase in operation capacity with increasing initial concentration of  $Cr^{6+}$  until reaching a maximum of 426.3 mg/g at initial concentration of 149.9 mg/L for the synthetic solution. A breakthrough adsorption test was performed to evaluate the system's maximum adsorption capacity when the effluent's concentration exceeds 0.5 mg/L of  $Cr^{6+}$ . The results ranged from 237.1–291.2 mg/g at Cr<sup>6+</sup> initial concentrations ranging from 58.0–149.9 mg/L. The experiments performed on real electroplating wastewater showed an adsorption capacity ranging from 160-230 mg/g. The influence of coexisting  $SO_4^{2-}$  and  $CI^-$  ions was also investigated, showing the fibre selectivity towards Cr<sup>6+</sup>. Moreover, the fibre was evaluated for its regeneration

capacity and stability. The fibre could withstand up to 80 operational-elution cycles, losing only about 20% of it adsorption capacity. A solution of 2.5 M NaOH was sufficient for carrying out the desorption process of the fibre in 15 min.

Ismail et al. 2014 used a strong acid cation exchange resin to capture Ni from electroplating wastewater [127]. Moreover, applying an electric current to the removal process was investigated to enhance treatment efficiency. An adsorption capacity of 91 mg/g was obtained and the removal efficiency exceeded 80%. The effect of applied electric current was studied at two different flow rates, 240 mL/h and 500 mL/h, enhancing the removal process by 12.7% and 2.5% respectively. A desorption process was performed using a solution of 2.0 M HCL at a flow rate of 500 mL/hr. There wasn't a very noticeable difference between the cases of absence and presence of electric current. Juang et al. 2006 have also investigated the removal of Ni<sup>2+</sup> from electroplting wastewater using a strong acid cation exchange resin [128]. A washing process of NaOH, HCL and n-hexane was applied to the resin to remove any impurities. It was observed that adsorption capacity increased with increasing pH, reaching a plateau after pH of 2.5. The highest adsorption capacity was obtained at a Ni<sup>2+</sup> concentration of 5.1 mol/m<sup>3</sup>, reaching up to 1.6 mol/kg at different temperatures ranging from 15 to 45 C. The two studies showed that a strong acid cation exchange resin is promising to effectively capture Ni found in wastewater under strong acidic conditions. No regeneration experiments experiments were performed by Juang et al. 2006, while Ismail et al. 2014, showed the possibility for regenerating a strong acid cation exchange resin [127,128].

Cavaco et al. 2007 considered adopting ion exchange processes to remove  $Cr^{3+}$  from wastewater evaluating the performance of a chelating exchange resin (Diaion CR11) and a weak cationic resin (Amberlite IRC86) [129]. A solution of 2.0 M HCl and 2.0 M NaOH was used to clean the resins and remove any impurities from the preparation process. A column setup was prepared using Amberlite IRC86 achieving complete saturation within 135 min. The column was also tested for the removal of  $Cr^{2+}$  in the presence of  $Cu^{2+}$  showing low selectivity towards  $Cr^{2+}$ , where the adsorption rate was almost the same. A solution of 1.0 M NaOH and 0.15 M H<sub>2</sub>O<sub>2</sub> was capable to desorb  $Cr^{3+}$  from the resins with a contact time of 24 h. Isotherm studies were performed on the resins, showing an increase in the Diaion CR11 adsorption capacity at a temperature of 25° than Diaion CR11. At low pH, the sorption capacity for  $Cr^{2+}$  decreases due to excess hydrogen ions with competing  $Cr^{2+}$  for the active sites. Sodium and calcium ions could also reduce the resin uptake of  $Cr^{2+}$ , requiring a pretreatment to maintain adequate removal efficiency.

#### 3.2.5. Advanced oxidation processes (AOPs)

Advanced oxidation processes (AOPs) involve treatment techniques that use hydroxyl radical reactions to remove contaminants from wastewater [17]. AOPs are expensive, complex and normally applied as pre-treatment prior to conventional methods as they aid in the partial degradation of non-biodegradable organics, transforming them into biodegradable intermediates which enhances the efficiency of biological degradation [130]. Therefore, AOPs are useful in the removal of contaminants that are resistant to other treatment methods (i.e. biological treatment) through chemically transforming the contaminant and achieving complete mineralisation [131]. Moreover, AOPs can treat

contaminants at low concentrations and are ideal for decreasing the concentration of compounds formed due to pretreatment methods, thus, eliminating the effects of disinfectants and oxidants on human health and improving the organoleptic properties of water [132,133]. Similar to other treatment technologies, AOPs have some disadvantages in addition to being costly when compared to biological processes. For example, the byproducts formed by AOP reactions could be as toxic as the original compound being treated [133]. This causes the efficiency of downstream processes to be unaffected by using AOP, and thus, be a waste of resources. Generally, AOPs can be divided into two categories [17]: (1) Non-Photochemical Methods: Ferrate, Fenton systems, Peroxone, Ozone, and Catalytic Ozone, and (2) Photochemical Methods: Ultrasound/sonication, Photo Fenton/Fenton-like Systems, Photocatalytic ozonation, Ultraviolet radiation (UV), Ozone – Hydrogen Peroxide – UV Radiation, Photocatalytic Oxidation (UV/ titanium dioxide(TiO<sub>2</sub>)), Ozone-UV Radiation, and Electron Beam Irradiation.

**3.2.5.1.**  $O_3$ ,  $O_3/H_2O_2$ ,  $O_3/UV$  and  $O_3/H_2O_2/UV$ . Wang et al., 2019 evaluated the removal of copper ions (Cu<sup>2+</sup>) from electroplating wastewater by ozone-based AOP [134]. They conducted a comparison study between  $O_3$ ,  $O_3$ /hydrogen peroxide(H<sub>2</sub>O<sub>2</sub>),  $O_3/UV$  and  $O_3/H_2O_2/UV$ . All the experiments were conducted at an initial concentration of ethylenediaminetetraacetic acid (EDTA) of 100 mg/L and an initial pH of the EDTA–Cu solution of 4.7–5.0. By applying  $O_3$  only at a concentration of 40 mg/L, the Cu<sup>2+</sup> removal efficiency reached 49% after 60 min. By applying  $O_3$  at a concentration of 40 mg/L and H<sub>2</sub>O<sub>2</sub> at a molar ratio of 1 H<sub>2</sub>O<sub>2</sub>: 4 O<sub>3</sub>, the Cu<sup>2+</sup> removal efficiency reached 43% after 60 mins. By applying  $O_3$  at a concentration of 40 mg/L, H<sub>2</sub>Cu<sup>2+</sup> removal efficiency reached 65% after 60 mins. By applying  $O_3$  at a concentration of 40 mg/L, H<sub>2</sub>Cu<sup>2+</sup> removal efficiency reached 58% after 60 mins. By applying  $O_3$  at a concentration of 40 mg/L, H<sub>2</sub>Cu<sup>2+</sup> removal efficiency reached 58% after 60 mins. The results obtained from this study indicate that ozone-based treatment processes were not very effective at removing Cu<sup>2+</sup>, where the highest removal efficiency achieved was 65%.

**3.2.5.2.** *UV/TiO*<sup>2</sup> *photocatalysis*. Hudaya et al., 2018 investigated the reduction of cyanide and cadmium from electroplating wastewater by UV/TiO<sub>2</sub> Photocatalysis [135]. A 64-Watt low-pressure UV amalgam lamp (intensity of approximately 20 Watt/L at wavelength equal 254 nm) was used in this study. The efficiency of cyanide removal exceeded 95% at pH 13 by applying 1.0 g/L TiO<sub>2</sub>. The removal efficiency of cadmium exceeded 95% at pH 13 by applying 2.0 g/L TiO<sub>2</sub>. UV/TiO<sub>2</sub> Photocatalysis is very efficient at removing cyanide and cadmium under alkaline conditions. However, the process is time consuming that requires more than 90 min.

#### 3.2.5.3. $H_2O_2$ and $H_2O_2$

**/FeSO**<sub>4</sub>. Husain et al., 2014 studied the removal of Ni<sup>2+</sup>, Cu<sup>2+</sup>, and Cr<sup>6+</sup> from electroplating wastewater using hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) combined with ferrous sulphate (FeSO<sub>4</sub>) (Fenton's reagent) [136]. Their results showed that the H<sub>2</sub>O<sub>2</sub> alone was not effective in removing toxic metals from electroplating wastewater. However, a combination of H<sub>2</sub>O<sub>2</sub> and FeSO<sub>4</sub>.7H<sub>2</sub>O<sub>2</sub> has proven to be effective for toxic metals removal, especially at pH 4. By applying H<sub>2</sub>O<sub>2</sub> at a dosage of 20 ml and FeSO<sub>4</sub>.7H<sub>2</sub>O<sub>2</sub> at a dosage of 1.0 gram, while keeping the other parameters constant (pH = 4 and reaction time = 24 hr), the removal

			Initial	Dose of	Fenton's reagent	Reaction	Removal
Parameter	Source	System	concentration	$H_2O_2$	FeSO <sub>4</sub> .7H <sub>2</sub> O <sub>2</sub>	time	efficiency
Unit	real electroplating	Multimetal	mg/L	ml	g	hour	%
Ni	wastewater	system	5.82	20	1.0	24	49
Cu			12.94	20	1.0	24	60
Cr			0.190	20	1.0	24	60

Table 2. Ni, Cu, and Cr removal from electroplating wastewater after applying Fenton's reagent [136].

efficiency of Ni<sup>2+</sup>, Cu<sup>2+</sup>, and Cr<sup>6+</sup> reached 49, 60, and 60%, respectively (see Table 2). There is a strong correlation between the three metals removal efficiencies and the doses of hydrogen peroxide. In general. The removal process employed in this study did not achieve a high treatment efficiency, in addition, the process is very time consuming.

3.2.5.4. Mixed-Fe<sub>3</sub> $O_4@SiO_2/metal$  oxide magnetite photocatalysts. Huang et al., 2014 investigated the recovery of heavy metal from electroplating wastewater for preparation of four magnetite photocatalysts: M-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>/CuO, M-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>/NiO, M-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>/ZnO, and M-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>/Fe<sub>2</sub>O<sub>3</sub> [137]. The efficiency of these photocatalysts were tested for the degradation of methyl orange (MO). Their results showed that M-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>/ZnO has the best photocatalytic performance, with a MO removal efficiency of 91.5% in 150 min. This was mainly due to the high •OH content compared to the other magnetite photocatalysts. Using M-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>/ZnO has also reduced the total organic carbon (TOC) concentration by about 27.9%. The MO removal rate was lower for other magnetite photocatalysts, where the removal efficiency did not exceed 37.4%, 19.0%, and 17.6% for M-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>/NiO, M-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>/Fe<sub>2</sub>O<sub>3</sub>, and M-Fe<sub>3</sub>O<sub>4</sub> @SiO<sub>2</sub>/CuO, respectively. However, M-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>/NiO was more efficient than the other two photocatalysts due to stronger •OH generation ability, higher absorption intensity of light in the UV band, and higher Brunauer-Emmett-Teller (BET) specific surface area of 177 m<sup>2</sup>/g, compared to 16 m<sup>2</sup>/g for M-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>/Fe<sub>2</sub>O<sub>3</sub> and 18 m<sup>2</sup>/g for M-Fe<sub>3</sub>O<sub>4</sub>@SiO<sub>2</sub>/CuO.

#### 3.3. Bio-recovery

Biorecovery is the introduction of biomaterials such as bacteria, algae, fungi and plants into aqueous media to remove contaminants [138]. The process is ecofriendly, sustainable and economically feasible [139]. Moreover, the biomaterials are abundant in nature and has high removal efficiency of metals from wastewater [140]. Biorecovery could follow a number of mechanisms such as biosorption, bioaccumulation, bioreduction and biomineralization [141]. Algae have proved to be strong binders to metal ions due to the presence of surplus electronegative charge on their cell wall. In addition, many microorganisms have shown to be highly tolerant to several metals ions with high growth rates [142]. The selection of a specific bacterial strain is also critical to achieve highest recovery efficiency [143]. The performance of biorecovery mainly depends on three growth parameters, which are temperature, pH, and dissolved oxygen level [144]. Wang et al. 2020 employed Taihu blue algae, which are considered as an abundant and hazardous biomass waste, to develop a biochar composite for removing Ni from electroplating wastewater [145]. KOH was used to activate the algal biochar with high specific area and enhance its porous structure. Then, iron was loaded into the biochar, by mixing it with a solution of ferric nitrate, to increase the adsorption capacity and improve catalytic performance. The wastewater samples were obtained from an electroplating industry with a Ni<sup>2+</sup> initial concentration of 2.82 mg/L and pH 11.7. Varying concentrations of the biochar, between 0.5 and 5.0 g per 200 mL of wastewater, were added. The pH was adjusted to a range of 2 to 8 and, then, the samples were stirred for 20 min at 200 rpm to achieve an adsorption balance. The biochar achieved a removal efficiency of 99.14% and adsorption capacity of 2.099 mg/g for the biochar with 20% mass fractions. The stability and recyclability of the biochar was investigated by washing it with alkaline and distilled water and then drying it in a vacuum. The biochar still exhibited high removal rates after four recycles reaching up to 93.26% removal.

Hu et al. 2020 assessed the performance of sulphate-reducing bacteria (SRB) for simultaneously removing sulphate and Ni from electroplating wastewater [146]. It has been noted that SRB has the capability to reduce sulphate to  $S^{2-}$ ,  $HS^{-}$  and  $H_2S$ , which in turn initiates the precipitation of toxic metals present in wastewater. This study has investigated the influence of different parameters on the sulphate and Ni removal efficiency by SRB. These parameters include pH, COS/S molar ratio, presence of a carbon source and the coexistence of valent iron elements. A maximum removal efficiency of 95.8% for Ni and 20.1% for sulphate were achieved at pH 7 after 3 days. It has also been indicated that the maximum activity for SRB is achieved at pH 6.0 to 7.5 and that activity is lost when pH is lower than 5 and higher than 9. Different types of carbon sources; sodium lactate, sodium succinate, sucrose and glucose, were used to study their effect on SRB treatment performance. After the addition of sodium lactate, SRB practically removed all the present Ni and removed 50.1% of the sulphate, while sucrose and glucose decreased SRB activity removing only around 32% of Ni and 12.5% of sulphate. COD/ Sulphate molar ratio also had an effect on SRB growth and removal efficiencies; when the molar ratios increased from 2 to 10, the removal of Ni<sup>2+</sup> and sulphate increased from 75.29% and 17.49% to 99.29% and 62%, respectively. Furthermore, the addition of  $Fe^{2+}$ and Fe<sup>3+</sup> enhanced the removal efficiency of Ni<sup>2+</sup> and sulphate, where Ni<sup>2+</sup> was completely removed and between 61.6% and 82.7% of sulphate was removed. SRB showed to be very effective to remove high amounts of Ni<sup>2+</sup> and moderate amounts of sulphate. SRB also had the efficiency as Taihu blue algae in removing Ni<sup>2+</sup> from wastewater.

Barquilha et al. 2019 used a brown algae named *Sargassum sp.* to prepare a biosorbent for the removal of Ni<sup>2+</sup> and Cu<sup>2+</sup> [147]. The experiments were first performed in synthetic solutions at metal ion concentrations of 0.5–3.0 mmol/L, pH of 4.5 and flow rate of 4.5 L/min into a bed column with a height of 30.6 cm. Maximum biosorption capacities of 1.404 and 1.656 mmol/L were obtained for Ni<sup>2+</sup> and Cu<sup>2+</sup>, respectively. A 0.5 M solution of acidified calcium chloride at pH 3.0 was used to desorb the metal ions from the biosorbent, where the biosorption process was performed at metals concentration of 1.0 mmol/L. The results showed that the desorption process has decreased the biosorption capacities by 25% after the first cycle. The study has also presented data from [148] showing that a solution of MgSO<sub>4</sub> had only a 15% decrease in

the biosorption capacity after the third cycle. The biosorbent was then tested in real electroplating wastewater at the same operating conditions used with the synthetic solutions. The wastewater first passed through a chemical precipitation process and cyanide oxidation process for pretreatment. The biosorption capacities for Ni<sup>2+</sup> and Cu<sup>2+</sup> decreased by about 29% and 26%, respectively, due to the high competition with other co-existing metals.

Wen et al. 2018 isolated five different microbial strains from electroplating wastewater [149]. The influence of exposure time, temperature, and pH, on the treatment process to remove Cu<sup>2+</sup> was investigated. Control experiments without any bacterial addition were also prepared. The equilibrium time was determined by adding bacterial isolates to  $Cu^{2+}$ solutions with concentration of 10 mg/L, and then incubated for 3 h at 28°C and an agitation speed of 150 rpm. The lowest equilibrium time achieved was 30 min and the highest equilibrium time was 120 min. At equilibrium, the highest adsorption capacity for  $Cu^{2+}$  obtained was 8.72 mg/g and the lowest adsorption capacity was 1.84 mg/g. The results obtained from isothermal experiments showed that the highest adsorption capacity was 45.68 mg/g and the lowest adsorption capacity was 18.20 mg/g. Temperatures between 22 and 32 C had negligible influence on  $Cu^{2+}$  removal, while 32 C reduced removal efficiency of all strains. The optimum pH achieving the highest removal efficiency was at pH 6. Furthermore, the highest COD removal efficiency was 71.6% and the lowest COD removal efficiency was 61.5%. The three most efficient bacterial strains were identified and inoculated for microbial bioaugmentation in a H/A/O-MBR system. Bioaugmentation with repeated inoculation has shown a considerable improvement in the removal of COD and Cu<sup>2+</sup>. Bioaugmentation treatment is a promising process for the application in electroplating wastewater to remove COD and Cu<sup>2+</sup> as indicated by the study.

## 3.4. Combined systems

#### 3.4.1. Adsorption and UV

Vyas and Kulkarni, 2011 evaluated the reduction of COD, biological oxygen demand (BOD), and total dissolved solids (TDS) concentrations in electroplating wastewater using adsorption and ultraviolet (UV) processes [150]. Figure 2(a) shows the flowsheet of the treatment processes. The adsorbent material used in this study is called 'Dashmool' and it is a combination of ten Ayurvedic plants, such as Uraria picta, Aegle Marmelos, Tribulus Terrestris, Desmodium Gangeticum, Solanum Xanthocarpum, Solanum Indicum, Gmelina Arborea, Stereospermum Suaveolens, Oroxylum Indicum, and Premna Serratifolia. The raw electroplating wastewater was loaded at a rate of 5 mL/min to a 30 cm Dashmool column. The adsorption process has reduced the BOD, COD, and TDS by about 32%, 44%, and 14%, respectively. Following the adsorption process, the partially treated wastewater samples were irradiated for 5 hours with a 24 Watts UV lamp. The combined processes reduced the BOD, COD, and TDS by about 92%, 86%, and 63%, respectively. Supplementary Table 1 also summarises the removal efficiency of the studied parameters for different treatment steps. Dashmool adsorbent was found to be very strong in removing BOD, COD, and TDS from electroplating industrial effluent. It is also important to notice that the UV reaction to degrade several contaminants is strongly affected by the pH conditions.



**Figure 2.** Flowsheet of the combined: a) adsorption and UV processes, b) resin adsorption and electrodeposition processes, c) Coagulation and  $H_2O_2/UV$ , d) coagulation + PAC +  $H_2O_2/UV$  processes, e) chemical precipitation and electrodialysis processes, f) electrocoagulation using an aluminium electrodes with simultaneous ozonation plus ozonation processes, g)  $H_2O_2$  oxidation with simultaneous anodic Fenton or electrocoagulation process (Adapted from [150–155]).

#### 3.4.2. Adsorption and subsequent electrodeposition

Li et al., 2019 have developed a pilot-plant which consisted of two treatment steps: adsorption through a fixed-bed resin to recover Ni<sup>2+</sup> ions from electroplating wastewater, and electrodeposition to produce nickel sheet through reduction of the concentrated Ni<sup>2+</sup> ions [151]. The adsorption process is comprised of three main units: 1) feed and pre-treatment system to adjust the pH to 5-7 and remove impurities and other toxic metals through settling for 2 h; 2) ion-exchange fixed-bed resin columns (three columns contain a weak acid cation resin and one column contains a chelating resin) in which supernatant was fed at 5 bed volumes (BV)/h in the down flow mode, and 3) a regeneration system in which 4% (w/w) hydrochloric acid (HCI) is passed in a downflow mode through the column bed at a flow rate of 1 BV/h to allow the desorption of nickel ions from the resin. The adsorption capacity of the resin was reported as 63 mg/g for  $Ni^{2+}$  ions and the concentrations of  $Ni^{2+}$  ions in the effluent of the adsorption process were less than 0.1 mg/L. Additionally, about 1.5 BV (more than 30 g/L) of concentrated neutral nickel solution was obtained during the regeneration step to be used as input in the electrodeposition process. In the electrodeposition process, a reactor was used which was divided into two independent compartments that are separated by a filter press cloth. A titanium plate was used as the cathode, while pure crystalline ruthenium-iridium oxide ( $RuO_2/IrO_2$ ) plate was used as an anode. Figure 2(b) shows the flowsheet of the treatment processes. The experiment results showed that about 95.6% of Ni<sup>2+</sup> ions could be recovered as an elemental nickel (Ni) on the cathode under the optimum electrodeposition conditions: initial Ni<sup>2+</sup> concentration of 28.5  $\alpha/L$ , temperature of 45°C-55°C, current density of 120 A/m<sup>2</sup>, pH of 3.5-4.1, and additives (saccharin 1.5 g/L, Boric acid (H<sub>3</sub>BO<sub>3</sub> 30 g/L)). The combined process was efficient to recover Ni from wastewater at a very low cost. Moreover, the process is promising to recover heavy metal ions from industrial wastewater rather than removing them as waste.

## 3.4.3. Adsorption and $H_2O_2/UV$ oxidation

Yen et al., 2015 used a batch photo-reactor to investigate the reduction of toxic metals and organic compounds from electroplating wastewater through a combination of the following treatment processes: (1) coagulation process using aluminium or iron salts (rapid mixing: 100 rpm for 10 min, slow mixing: 40 rpm for 30 min, and 30 min for settling), (2) adsorption process using 5.0 g/L powder activated carbon (PAC) (mixing for 120 min at 40 rpm), and (3) oxidation process using hydrogen peroxide  $(H_2O_2)/UV$  (oxidation time = 90 min,  $H_2O_2$  dosage of 100 mg/L, and 16 UV lamps each had 8 watts and emitting 254 nm wavelength) [152]. Figures 2(c,d) shows the flowsheet of the treatment processes. The combined processes in Figure 2(d) reduced the COD, dissolved organic carbon (DOC),  $Fe^{3+}$ ,  $Zn^{2+}$ , and Ni<sup>2+</sup> from electroplating wastewater by about 99.3%, 98.2%, 100.0%, 99.8%, and 98.4%, respectively. Supplementary Table 2 also summarises the removal efficiency of the studied parameters for different treatment processes. The results indicated that applying either coagulation followed by  $H_2O_2/UV$  or coagulation followed by PAC will reduce heavy metals and organics, but without satisfying the reuse criteria. In order to meet the reuse criteria, the combined process of coagulation, PAC, and  $H_2O_2/UV$  must be applied.

#### 3.4.4. Chemical precipitation and electrodialysis

Peng et al., 2004 investigated the removal of hexavalent chromium (Cr<sup>6+</sup>), as well as other ions, such as sodium (Na<sup>+</sup>), chloride (Cl<sup>-</sup>), magnesium ion (Mg<sup>2+</sup>), calcium ion (Ca<sup>2+</sup>), phosphate (PO $_{4}^{3-}$ ), and sulphate (SO $_{4}^{2-}$ ) from electroplating wastewater using chemical precipitation and electrodialysis processes [153]. Figure 2(e) shows the flowsheet of the treatment processes. First, the raw electroplating wastewater passes through two chemical precipitation processes which was sodium sulphide (Na<sub>2</sub>S) and ferrous chloride (FeCl<sub>2</sub>) to reduce Cr<sup>6+</sup> to chromium(III) (Cr<sup>3+</sup>). Then, the wastewater passes through another precipitation processes to adjust the pH from 8 to 10 using sodium hydroxide (NaOH), which allows the precipitation of  $Cr^{3+}$  and ferric cation (Fe<sup>3+</sup>) in the form of hydroxides. After the chemical precipitation process, the sludge is separated from solution. The solution is then passed through multiple electrodialysis treatment processes; a two-step rougher and a one-step scavenger. The electrodialysis unit consists of anionic exchange membrane (AEM) and cationic exchange membrane (CEM) with an effective area of 150 cm<sup>2</sup> and the distance between them is 0.1 cm. From the electrodialysis process, one concentrated water and one diluted water stream are produced. The flow rates are controlled at 2.0 L/h for concentrated water and 5.0 L/h for diluted water. The temperature was maintained at  $30 \pm 3^{\circ}$ C throughout the experiment. The combined chemical precipitation and electrodialysis treatment have reduced the Cr<sup>6+</sup>, Na<sup>+</sup>, Cl<sup>-</sup>, Mg<sup>2+</sup>, Ca<sup>2+</sup>, PO<sub>4</sub><sup>3-</sup>, and SO<sub>4</sub><sup>2-</sup> by about 95%, 94.9%, 96.2%, 93.8%, 95.1%, 100%, and 94.0%, respectively.

#### 3.4.5. Electrochemical and ozonation

Orescanin et al., 2013 developed a pilot-plant to treat electroplating wastewater which consists of four treatment steps: (1) 15 min electroreduction using iron electrode plates (distance between plates is 10 mm; total surface area is  $0.6552 \text{ m}^2$ ; I = 70 A; U = 12 V); (2) 15 min electrocoagulation using an aluminium electrode set (distance between plates is 10 mm; total surface area is  $0.5688 \text{ m}^2$ ; I = 65 A; U = 12 V) with simultaneous ozonation (constant flow rate of 2.5 mL/min); and (3) 30 min ozonation [154]. The sludge is allowed to settle for 45 min. Figure 2f shows the flowsheet of the treatment processes. The combined treatment reduced the COD, total organic carbon (TOC), lead (Pb), nickel (Ni), Fe<sup>3+</sup>, Cu<sup>2+</sup>, Zn<sup>2+</sup>, and Cr<sup>6+</sup> by about 93.43%, 93.24%, 96.81%, 95.86%, 100.00%, 98.66%, 99.97%, and 99.94%, respectively. The treatment process was very efficient at almost eliminating COD, TOC, and six different heavy metal ions. This study has shown that the combination of these two different processes have certainly greater performance than relying on one treatment system.

#### 3.4.6. $H_2O_2$ oxidation and anodic Fenton process or electrocoagulation

Zhao et al., 2013 investigated the removal of organic contaminants, toxic metals, and cyanide from an electroplating wastewater using  $H_2O_2$  oxidation followed by anodic Fenton process [155,156]. At all pH values, the cyanide concentration in the wastewater is expected to decrease by about 60% within 10 min when adding  $H_2O_2$  with a dosage of 3.0 mg/L. The effluent is then further treated by either anodic Fenton or electrocoagulation process. Figure 2(g) shows the flowsheet of the treatment processes. In the case of applying the Fenton process, a 30 Mm  $H_2O_2$  was added to an electrochemical system while keeping constant pH of 4 and current density of 5 mA/cm<sup>2</sup>. The removal efficiency

of cyanide was 100% after 15 min, COD reached 73.5% after 30 min,  $Cu^{2+}$  removal reached 100% after 5 min, and Ni<sup>2+</sup> removal reached 94% after 5 min. In case of applying electrocoagulation process at a current density of 20 mA/cm<sup>2</sup> and after 30 min of reaction, the removal efficiency of COD reached 18.4%, and removals of  $Cu^{2+}$  and Ni<sup>2+</sup> exceeded 99%. Moreover, the removal efficiency of cyanide reached 97.5% after 30 min of reaction at a current density of 5 mA/cm<sup>2</sup>. The results show that the combined system was able to effectively remove cyanide,  $Cu^{2+}$ , and Ni<sup>2+</sup> in a considerable short amount of time.

## 4. Comparison of different treatment methods

Different treatment technologies could be applied for the treatment of electroplating wastewater, which includes chemical precipitation, adsorption, coagulation and flocculation, ion exchange, electrocoagulation, membrane filtration, advanced oxidation process and biorecovery. These methods provide the possibility to capture different toxic metals and remove organic pollutants. For example, biochar was used as an adsorbent and managed to almost remove  $Pb^{2+}$  completely, while a cation exchange resin removed  $Cu^2$ <sup>+</sup>. Hydroxide and sulphide precipitation managed to remove Cu ions completely, followed by adsorption using electroplating sludge. The main advantage of treatment using sludgedriven adsorbent is the possibility to reduce and recycle waste. Even though chemical precipitation was successful at removing  $Cu^{2+}$ , it was not very efficient at removing  $Zn^{2+}$ . The adsorption method employing electroplating sludge and the electrocoagulation methods employing Al electrodes were both very efficient at removing Zn<sup>2+</sup>. The diasdvantage of electrocoagulation over adsorption is its requirement for power consumption. Still, Electrocoagulation was very efficient at removing Cu<sup>2+</sup>, Cr<sup>2+</sup>, Ni<sup>2+</sup>, Zn<sup>2+</sup> and Cu<sup>2+</sup>, while utilising different metal electrodes, with almost complete removal. Zero-valent iron nanoparticles was successful at removing multiple toxic metals at the same time, which is an advantage not reported by many treatment methods. One of the drawbacks of Zero-valent iron nanoparticles is its toxicity to the organic life if released into the environment [157]. Various methods of membrane filtration have reported high electroplating wastewater treatment efficiency removing toxic metals such as Ni<sup>2+</sup>, Zn<sup>2+</sup>, Fe<sup>3+</sup>, Cu<sup>2+</sup>, Cr<sup>2+</sup> and Pd<sup>2+</sup>. Still, of the main drawbacks of membrane filtration is energy demand and the production of permeate that requires further processing. Moreover, The combination of different treatment methods have reported very high removal of toxic metals. The main strength of processes combination is limiting the drawbacks of each treatment method and increasing treatment efficiency. The combined systems also succeeded to remove multiple pollutants and at very high efficiency, unlike other treatment methods that fail to effectively remove a large number of pollutants within the same process. Table 3 provides an overview of all the treatment processes discussed in this review and their removal efficiencies for specific pollutants.

#### 5. Cost of electroplating wastewater treatment

Evaluating the cost of wastewater treatment should be covering the whole process starting from freshwater withdrawal till the final discharge of effluent to the environment [158]. From economic point of view, the aim is to enhance cost efficiency by identifying the highest treatment performance and determining the cost reduction

Table 3. Efficiency	of removing heavy metals from real electroplating w	astewater using v	/arious tr	eatment t	echniques	
Treatment Process		Source	Pollutant	Removal Efficiency	References	Details about Methodology
Chemical precipitation	Hydroxide precipitation	Real electroplating wastewater	Zn <sup>2+</sup>	68.8%	78	Multimetal system
			$Ni^{2+}$	93.75%		
			Cu <sup>2+</sup>	100%		
	Sulfide precipitation		Zn <sup>2+</sup>	76.6%		
			Ni <sup>2+</sup>	95.32%		
			Cu <sup>2+</sup>	100%		
Adsorption						
	Biochar		$Pb^{2+}$	99.8%	98	
			Cr <sup>6+</sup>	72.4%		
	Microwave-functionalized cellulose derived from rice husk		$Pb^{2+}$	99.2%	66	pH = 5.5, time = 60 minutes, temperature= 20 °C, multimetal system
			Cd <sup>2+</sup>	99.8%		
			Ni <sup>2+</sup>	96.35%		
	Electroplating sludge		Zn <sup>2+</sup>	99.7%	27	Multimetal system
			Cu <sup>2+</sup>	99.4%		
			Ni <sup>2+</sup>	37.9%		
			Co <sup>2+</sup>	53.3%		
	zero-valent iron nanoparticles		Cr <sup>6+</sup>	66.7%	101	
			$Pb^{2+}$	91.5%		
			$Ag^+$	83.3%		
			Cu <sup>2+</sup>	80.8%		pH = 7.3, time = 36 minutes, temperature= 30 °C monomeral system
			Ni <sup>2+</sup>	17.4%		Multimetal system
			$Mn^{2+}$	47.1%		×
			Zn <sup>2+</sup>	54.6%		
			Fe <sup>3+</sup>	94.7%		
			Al <sup>3+</sup>	100%		
			Co <sup>2+</sup>	42.1%		

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(Continued)

Table 3. (Continue	d).					
Treatment Process		Source	Pollutant	Removal Efficiency	References	Details about Methodology
	Amorphous aluminium oxide		Cr <sup>4+</sup>	%06	103	pH = 3-9,
Coagulation and flocculation	O-xanthogenated chitosan Schiff base (XCTS) and $Fe^{3+}$ ions		Cd <sup>2+</sup>	99.5%	112	Multimetal system
	polyferric sulfate (PFS) mixed with cationic polyelectrolyte (CP) coagulants					
	Zero-valent iron		Cr <sup>6+</sup>	<b>%6.</b> 66	114	
lon Exchange	cation exchange resin followed byphotocatalysis process using TiO <sub>2-</sub> ZrO <sub>2</sub> solid solution		Cu <sup>2+</sup>	99.1%	124	
			Cr <sup>6+</sup>	96.29%		
	silica-supported pyridine resin		Cr <sup>6+</sup>	99.3%	125	
	fibrous weak anion exchanger		Cr <sup>6+</sup>	ı	126	
	strong acid cation exchange resin		Ni <sup>2+</sup>	> 80%	127	
	Chelating exchange resin and a weak cationic resin		Cr <sup>6+</sup>	,	129	
Electrocoagulation	Cu-Al electrodes		Ni <sup>2+</sup>	ı	52	Constant Current
	Fe-Fe, Al-Al, Fe-Al, and Al-Fe electrodes		Cu <sup>2+</sup>	100%	53	Constant Current
			Cr <sup>2+</sup>	100%		
			Ni <sup>2+</sup>	100%		
	Al electrodes		Zn <sup>2+</sup>	97%	7	Constant Current
			Cu <sup>2+</sup>	%96		
			Cr <sup>6+</sup>	76%		
Membrane filtration	ion-imprinted membranes		Pd <sup>2+</sup>	1.2106	65	Multimetal system
				mg/g		
	Ceramic UF		Cr <sup>2+</sup>	89%	99	
			C0 <sup>2+</sup>	60%		
	blend membranes		Pd <sup>2+</sup>	96.8%	67	
	poly (acrilonitryle) nanofiltration membranes		Ni <sup>2+</sup>	87%	68	
			Cr <sup>2+</sup>	83%		
	emulsion liquid membranes		Cr <sup>2+</sup>	≈100%	69	
	hollow-fiber membranes		Cr <sup>2+</sup>	95.76%	70	
						(Continued)

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Treatment Process		Source	Pollutant	Removal Efficiency	References	Details about Methodology
			Cu <sup>2+</sup>	95.33%		
			Ni <sup>2+</sup>	94.99%		
	nanofiltration membranes		Zn <sup>2+</sup>	94%	71	
			Fe <sup>3+</sup>	93%		
	nanofiltration membranes		Cu <sup>2+</sup>	96.6%	72	
			Cr <sup>2+</sup>	90.0%		
Advanced Oxidation Processes	ozone-based and ultraviolet		Cu <sup>2+</sup>	65%	134	
	$H_2O_2$ combined with FeSO <sub>4</sub>		Ni <sup>2+</sup>	49%	136	
			Cu <sup>2+</sup>	60%		
			Cr <sup>2+</sup>	60%		
Bio-recovery	Taihu blue algae		Ni <sup>2+</sup>	99.14%	145	
	sulfate-reducing bacteria		Ni <sup>2+</sup>	100%	146	
	microbial strains		Cu <sup>2+</sup>		149	
<b>Combined Systems</b>						
	Adsorption and subsequent electrodeposition		Ni <sup>2+</sup>	95.6%	151	
	Adsorption and H2O2/UV oxidation				152	
			Fe <sup>3+</sup>	100.0%		
			Zn <sup>2+</sup>	99.8%		
			Ni <sup>2+</sup>	98.4%		
	Chemical precipitation and electrodialysis		Cr <sup>6+</sup>	95%	153	
			$Na^+$	94.9%		
			CI^	96.2%		
			Mg <sup>2+</sup>	93.8%		
			Ca <sup>2+</sup>	95.1%		
	Electrochemical and ozonation				154	
			$Pb^{2+}$	96.81%		
			Ni <sup>2+</sup>	95.86%		
			Fe <sup>3+</sup>	100.00%		

(Continued)

		s Details about Methodology									
		Reference				155					
	Removal	Efficiency	98.66%	99.97%	99.94%		100%	94%		> 99%	> 99%
		Pollutant	Cu <sup>2+</sup>	Zn <sup>2+</sup>	Cr <sup>6+</sup>		Cu <sup>2+</sup>	Ni <sup>2+</sup>		Cu <sup>2+</sup>	Ni <sup>2+</sup>
		Source									
Table 3. (Continued).	Treatment	Process				H <sub>2</sub> O <sub>2</sub> oxidation and anodic Fenton process			$\rm H_2O_2$ oxidation and electrocoagulation		

		Operation	
Process	Removal	Cost	Reference
Electrocoagulation	Cr <sup>6+</sup> , Cu <sup>2+</sup> , Zn <sup>2+</sup> Fe <sup>3+</sup> , Ni <sup>2+</sup> , Cu <sup>2+</sup> , Zn <sup>2+</sup> ,	1.7 \$/m <sup>3</sup>	[39]
Chemical Coagulation	$Pb^{2+}, Cd^{2+}$	3.5 \$/m <sup>3</sup>	
fixed-bed resin adsorption and subsequent	Ni <sup>2+</sup>	0.35 \$/m <sup>3</sup>	[151]
electrodeposition		2	
lon exchange and photocatalytic process	Cr <sup>6+</sup>	3.34 \$/m <sup>2</sup>	[166]
Sequential reduction – precipitation – settling process		2.10 \$/m <sup>2</sup>	
Nanoparticles of zero-valent iron (nZVI)	Cr <sup>6+</sup> , Pb <sup>2+</sup> , Ag+, Cu <sup>2+</sup> , Ni <sup>2+</sup> , Mn <sup>2+</sup> , Zn <sup>2+</sup> ,	4.45 \$/m <sup>3</sup>	[167]
	$Fe^{3+}$ , $Al^{3+}$ , $Co^{2+}$		

Table 4. Cost of electroplating wastewater treatment by targeting heavy metals.

potential of the identified process [159]. Achieving that would require an extensive experimentation process that is costly. Therefore, computer models could be introduced to simulate and optimise the most efficient treatment technology [160]. The cost of treatment normally includes capital costs and operation costs [161], where operational costs include chemical additives and energy requirements [162]. As illegal discharge of industrial wastewater poses significant environmental damage, effective management and treatment options must be implemented. Therefore, there is a demand to identify cost effective technologies, reduce operational costs and improve product guality [163]. The optimisation of energy consumption is also very critical for reducing negative environmental impact due to greenhouse gas emission [159]. In this section, the cost for electroplating wastewater treatment has been adopted from several literature to analyse the cost differences between different treatment technologies. Table 4 presents the possible operating costs for different treatment technologies of electroplating wastewater. As shown in the table, chemical precipitation recorded highest operational cost followed by adsorption by nZVI and then chemical coagulation. The combined system of adsorption followed by electrodeposition has been successful achieving the lowest operational costs among the other mentioned treatment technologies. Average operational costs have been achieved by electrocoagulation and two combined systems employing lon exchange with photocatalytic process and Sequential reduction - precipitation - settling process.

#### 6. Conclusions

This review has presented different treatment technologies, such as adsorption, chemical precipitation, coagulation flocculation, ion exchange, electrocoagulation, membrane filtration, advanced oxidation processes, biorecovery, and combined systems, to remove or reduce different contaminants from electroplating wastewater. Various studies have been investigated outlining the adopted materials, applied parameters, observed conditions and the treatment performance achieved. The selection of a specific technology mainly depends on its effectiveness and applicability to work in wastewater with specific characteristics to reach a desired quality. In addition, economic factors play a major role in selecting a suitable treatment system. Investigating selective recovery of specific metal ions has been the objective for most of the studies, where the influence of coexistence of other ions was considered.

The combination of several treatment systems has also shown capability of removing a wide range of contaminants at higher efficiencies than particular single systems. Choosing the most efficient treatment technology depends on the objective of the treatment and the pollutants required to be removed. This is because each treatment method has been successful in removing specific pollutants and none of the methods were able to remove all kinds of pollutants present in the wastewater. The construction of an economical, environmentally friendly and highly efficient treatment system is the optimum goal to effectively treat electroplating wastewater. In this review, chemical precipitation has reported the highest cost of treatment at 13.35 \$/m<sup>3</sup>, while combined process of adsorption and electrodeposition reported the lowest cost at 0.35 USD/ton.

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#### **Data availability**

The article includes all data generated or analysed during the investigation.

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