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Removal of xenobiotics from wastewater by electrocoagulation: A mini-review

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Xenobiotics are man-made, non-biodegradable compounds present in the environment at higher concentrations than usual. Phenols, dyes, surfactants, pesticides, pharmaceuticals are some of the xenobiotics. These pollutants have an adverse effect on the environment because of their toxicity, complex structure and carcinogenic nature and are not efficiently removed by conventional wastewater treatment technologies. Researchers round the globe have demonstrated various physical and chemical methods for the treatment of xenobiotics. This mini-review focuses on the application of electrocoagulation for the removal of xenobiotics. Different operating parameters that affect the removal efficiency, the types of electrodes used and the treatment cost of xenobiotics are elaborated in this article.

Keywords: Electrocoagulation, pesticides, pharmaceuticals, surfactants, wastewater treatment.

Introduction

Industrial effluents and municipal sewage contain pollutants that are detrimental to the environment. Hence, these pollutants need to be removed before releasing the effluent into natural water bodies. Some of these pollutants are xenobiotic, i.e. not found naturally in the environment and are non-biodegradable compounds, which are present at a higher concentration than usual in the environment[1]. Phenols, dyes, surfactants, pesticides and pharmaceuticals are some of the commonly used xenobiotics. Phenols and surfactants are used in a variety of household applications such as washing, cleaning and are also used in institutions, hospitals and other public places. Furthermore, pesticides are used in public parks, playing grounds and gardening[2,3]. Pharmaceuticals consumed by animals and humans are discharged into the environment either directly or are indirectly excreted from their body along with faeces and urine[4]. Thus, following these pathways the xenobiotics end up in municipal sewage.

The occurrence of these pollutants in the surface water bodies is a significant concern because of their detrimental effect on the ecosystem. Though, the concentration of these pollutants is very low in municipal sewage, however they can enter the food chain through various pathways and can have an adverse effect on human and animal health[5]. Xenobiotics present in the environment even at lower concentrations have various toxicological impacts on both plants and animals. Xenobiotics can alter the physiological and biochemical structures of aquatic organisms[6]. Xenobiotics are also endocrine-disrupting compounds and can have an adverse effect on the reproductive, immune and neurological system of humans and other organisms[7]. These compounds have a complex structure and forms an aromatic or polycyclic ring structure with compounds of high molecular mass. Hence, they are insoluble or partially soluble in water[8].

The conventional wastewater treatment plants are originally engineered for the removal of organic matter and suspended solids. Thus, xenobiotic compounds are not efficiently removed in conventional wastewater treatment plants[9]. Different technologies such as adsorption, nanofiltration, reverse osmosis, photo-catalytic degradation, biological degradation, etc. have been used for the removal of these xenobiotics. However, each technology has some drawbacks, such as reverse osmosis is expensive[10]. Electrocoagulation (EC) is an efficient technique used for the treatment of wastewater over the years, because of its short treatment time, no chemical addition, smooth operation, and minimal footprint. Furthermore, even the smallest colloidal particles can be removed using EC.

The theory behind EC is same as coagulation and flocculation, where the colloidal particles are removed by desta-

bilizing with the addition of coagulants, thus making the force of attraction dominant and then forming flocs to settle the particle down[11]. This mini-review focuses on the application of EC for the removal of various xenobiotic compounds like dyes, surfactants etc. Also, the various parameters affecting the efficiency of EC have also been discussed. Thus, this mini-review is intended to help the researcher in this subject to take this technology forward for the removal of xenobiotic compounds from contaminated water and thus to produce treated water safe for various reuse purposes.

Electrocoagulation

Electrocoagulation follows the basic principle of electrolysis. The process occurs on the action of transfer of ions between two electrodes, i.e. anode and cathode, in an electrolyte. Due to the application of current between the electrodes, cations move towards cathode and anions towards the anode, respectively[12]. A basic EC setup consists of an anode and cathode electrodes connected to an external power source either in monopolar or in bipolar configuration and immersed into the solution to be treated (Fig. 1). The metals ions generated from the sacrificial anode are destabilizing agents that neutralize the electric charge present on the surface of the pollutants, thus removing the contaminants[13]. At the cathode, hydrogen gas is liberated and helps to buoyant the flocculating particles onto the water surface[14].

The reactions occurring at the anode and cathode are given below from eq. (1) to (4).

At anode:

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

$$2H_2O(l) \rightarrow 4H^+(aq) + O_2(g) + 4e^- \quad (2)$$

At cathode:

$$M^{n+}(aq) + ne^- \rightarrow M(s) \quad (3)$$

$$2H_2O(l) + 2e^- \rightarrow H_2(g) + 2OH^- \quad (4)$$

where, M can be any metal like iron or aluminum. The mass of metal ions generated from the anode is given by Faraday's law, following eq. (5).

$$m = ItM_w/zF \quad (5)$$

where, m is the mass of metal ions generated (g), I is the current supplied (A), t is the time of operation (s), M_w is the molecular weight of the metal (g/mol), F is Faraday's constant (96,485 C/mol), z is the number of electrons involved in the reaction. However in some cases, the mass of metal ions generated calculated from Faraday's law has deviated from the actual mass of ions generated because of the occurrence of other reactions on the anode. When the pH of the solution is alkaline, the evolution of oxygen at anode takes place following eq. (2). It might be the reason for the deviation in the actual mass to the theoretical value for the metal ions[11].

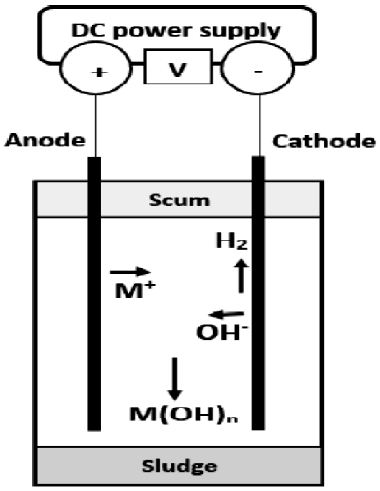

Fig. 1. Electrocoagulation reactor.

Factors affecting electrocoagulation

The efficiency of EC to remove pollutants from the wastewater is influenced by various operating parameters such as current density, pH of the electrolyte, initial concentration of the contaminant, time of electrolysis, the distance between the electrodes, electrode configuration, etc.[11]. This section provides an overview of some important factors affecting the performance of EC.

(*A) Current density:*

Current density (CD) is defined as the current applied per unit area of the electrode[11]. The amount of metal ions released from the electrodes is determined by CD. The anodic metal dissolution rate increases with the increase in CD. Hence, a higher number of metal hydroxide flocs are formed for higher CD; as a result, the pollutant removal efficiency increases. However, an increase in the CD after the critical limit of CD will not show any further improvement in the removal efficiency of the pollutant as sufficient flocs would have already been formed in the EC setup for the settling of the contaminants[15]. Electrocoagulation with aluminium elec-

trodes was used for the removal of endosulphan pesticide, which demonstrated that with the increase in CD from 2.5 to 12 mA/cm^2, the removal efficiency increased from 74.6% to 92.6%[16]. Dyes have also been successfully removed by EC from wastewater and effect of CD on removal efficiency was investigated, and the result obtained shows that increasing CD from 13.9 to 41.7 mA/cm^2, the dye removal efficiency increased from 35.9 to 73.6%[17].

(*B) Solution pH:*

Solution pH is another vital parameter for EC, as the electrolyte pH affects the solution conductivity, zeta potential and the rate of metal dissolution into the solution. During EC, the pH of wastewater changes and it is difficult to establish a relationship between pH and efficiency of the process. Hence, the initial pH of the electrolyte is considered as the solution pH for EC[11]. The formation of complex species during the dissolution of metals depends on the solution pH as a result, the overall mechanism of EC changes with the change in solution pH[18]. Effect of solution pH was noted for the removal of phenolic compounds using EC, and it was observed that the change in solution pH from 1 to 7 demonstrated an increase in removal efficiency of phenolic compounds from 18% to 88%. However above pH of 7, the efficiency of EC was decreased because at very high pH aluminium complex formed was soluble and will not absorb the pollutant, whereas at very low pH aluminium hydroxide has amphoteric nature and will not precipitate[19].

(*C) Initial concentration of pollutant:*

The initial concentration of pollutants also affects the efficacy of EC. The removal efficiency is inversely proportional to the initial concentration of a pollutant at a constant CD. At higher pollutant concentration, insufficient numbers of metals flocs are available for the adsorption of the pollutants. Hence, settling is less for wastewater with a higher initial concentration of the pollutant[15]. Methyl orange dye was treated using EC with iron electrodes, and the effect of initial concentration was noted. The result obtained showed that the removal efficiency decreased from 71.11% to 60% with the increase in initial concentration of methyl orange dye from 25 to 55 mg/L by keeping the other parameters constant[20].

(*D) Electrolysis time:*

The pollutant removal efficiency and the reaction rate depends on the time of electrolysis. The increase in electrolysis time will increase the removal efficiency. This is because a higher number of flocs will be formed but up to a critical time after which the removal efficiency remains constant and does not increase with further increase in electrolysis time[21]. When electrolysis time was increased from 15 to 35 min, the removal efficiency of diazinon pesticide increased from 57.7% to 89%[22]. Removal efficiency of propiconazole pesticide also increased from 40% to 70% when electrolysis time was increased from 10 to 60 min[23].

(E) Distance between the electrodes:

The amount of electrical energy imposed into the system for the generation of electric field and to induce motion into ions depends on the distance between the electrodes. The removal efficiency is inversely proportional to the distance between the electrodes. The energy consumption and removal efficiency decreases and increases, respectively, with the decrease in electrode gap. This is because, less electrical energy is required for the motion of ions with smaller electrode gap due to the shorter travel path[24]. Colour removal from the textile industry effluent decreased from 98.15% to 72.18%, when the electrode gap was increased from 1.5 to 4.5 cm, respectively[25]. Methyl orange removal efficiency also decreased from 86% to 80%, when the electrode gap was increased from 0.8 to 3.1 cm[20].

(*F) Electrode configuration:*

Electrodes connected in monopolar or bipolar mode affect the cost of the treatment of EC, as the consumption of energy is influenced by the polarity of the electrode. Monopolar parallel arrangement is cost-effective as compared to monopolar series and bipolar series arrangement. The parallel arrangement requires a lower potential difference, whereas a series arrangement requires a higher potential difference. Hence, energy consumption is less in the case of a parallel arrangement for the same degree of pollutant removal[26]. In a demonstration, the minimum energy consumed for the removal of malathion pesticide in bipolar and monopolar connection was found to be 1.5 and 0.223 kWh/kg, respectively[10].

Applications of electrocoagulation

Electrocoagulation process has been widely used since the last two decades for the treatment of different types of wastewater generated either from households or industries[27]. Resource recovery has also been achieved from the sludge produced during the process of EC[28,29]. Electrocoagulation

is found to be a feasible solution for the removal of xenobiotics from wastewater[30]. In this section, an overview on the removal of some xenobiotics employing EC is presented.

(A) Phenols:

Phenolic compounds are used in various household applications such as emulsifiers, detergents, dispersants, and wetting agents. Agricultural and industrial uses of these compounds are the major sources that lead to their occurrence in surface water bodies[31]. Phenol removal using EC from paper mill effluent was investigated with aluminium and iron electrodes. The influence of various operating parameters was also explored, and it was observed that with an applied voltage of 12 V, electrolysis time of 2 min and at a current intensity of 10 mA/cm^2, 98% and 93% of phenol was removed with aluminium and iron electrodes, respectively[10]. The reason for the difference in removal efficiency for two different metal electrodes was that these electrodes would produce different compounds at different pH, during the process of EC[10,11].

In another exploration, EC was used for the removal of phenol and organic matter from synthetic wastewater. With zinc anode and stainless steel cathode pair, a CD of 25 mA/cm^2, 1.5 g/L of NaCl as an electrolyte, the removal efficiency of phenol and chemical oxygen demand (COD) was observed to be 84.2% and 40.3%, respectively[34]. It was also observed that without the addition of electrolyte NaCl, the phenol and COD removal efficiencies were 72.3% and 20.9%, respectively. The difference between the removal efficiencies may be explained by the fact that the addition of NaCl in electrolyte improves the conductivity. Moreover, zinc dissociation into the effluent forms chemical species that are potent oxidizing agents, which decompose the organic matter present in wastewater[34].

In another demonstration, the effect of presence of different chemical species on phenol removal using the process EC was inspected. Iron electrodes were used both as anode and cathode to examine the reduction in phenol concentration with a CD of 45 mA/cm^2 and NaCl concentration of 6 g/L. The effect of the addition of $NaHCO_3$, $CaSO_4$, $MgSO_4$, and $MgCl_2$ on removal efficiency of phenol was also noted[35]. The result obtained showed that the addition of $CaSO_4$ into the solution increases the time required for the complete removal of phenol. Addition of $MgSO_4$ demonstrated similar results for the removal of phenol except that the time needed for complete removal of phenol was 12 min which was 20 min for $CaSO_4$. Furthermore, the addition of $NaHCO_3$ took 30 min for the complete removal of phenol from wastewater, and it is quite evidently more than the time required for the complete removal of phenol with the addition of $CaSO_4$ or $MgSO_4$.

Addition of $MgCl_2$ had shown that after 5 min of electrolysis time, complete removal of phenol could be achieved, which is very less when compared to the addition of $MgSO_4$, $CaSO_4$, or $NaHCO_3$ into the electrolyte. The reason for this reduced time of electrolysis is because of lesser cathode passivation with the addition of $MgCl_2$ as compared to the other three chemical species. Moreover, the destruction of the passive oxide film, which forms on the anode is supported with higher chloride ion concentration and as a result, short treatment time is required for the removal of phenol when $MgCl_2$ is added in the electrolyte[13,14].

(*B) Dyes:*

Textile industries are the major source of release of dye in wastewater because of their usage of dyes as a colouring agent for textile fibres[36]. Worldwide estimation for the release of reactive dyes is approximately 8000 tons per year in the receiving water bodies[37]. Brilliant green dye was removed using iron electrodes in EC with an initial dye concentration of 100 mg/L at a CD of 4.17 mA/cm^2 and initial pH of 4.0 during the time of electrolysis of 30 min. It was observed that with the above-mentioned operating parameters, 99.59% of the dye was removed. Moreover, the optimum pH range was found to be 4–10 to obtain the highest dye removal efficiency. With initial dye concentration of more than 100 mg/L, the removal efficiency of dye decreased because, at the higher concentration of dye, the flocs produced by ion dissociation with iron anode were insufficient to absorb all molecules of dye present in the solution[17].

In another exploration pertaining to EC, aluminium, steel and bronze electrodes produced by metallurgical filing waste was employed and their efficiency of degradation of carmine dye was compared with commercial electrodes[38]. When the commercial electrodes were used with the treatment time of 80 min, applied current of 10 mA and at a voltage of 5 V, dye discoloration of 84% was achieved for aluminium electrode, 90% for steel electrode and 96% for bronze electrode. On the other hand, when electrodes produced from metal waste

were used, dye discoloration of 72% was achieved for aluminium electrode, 92% for steel electrode and 90% for bronze electrode, with the same duration of treatment. The application of electrodes produced from waste in the process of EC emphasized that nearly similar dye removal efficiency can be achieved with these materials as compared to the commercial electrodes[38].

Electrocoagulation process was also used for the removal of indigo carmine dye from synthetic solution with high purity magnesium and magnesium-aluminium-zinc alloy as anodes[39]. With an initial concentration of dye being 100 mg/L and CD of 5 mA/cm^2, magnesium-aluminium-zinc alloy as anode, the dye discoloration of 96% was achieved at room temperature. When pure magnesium was used as anode, 75% of dye discoloration was achieved with the similar initial concentration of dye and CD. Use of magnesium alloy demonstrated better results as compared to pure magnesium, warranting its use for the treatment of wastewater[39].

(*C) Surfactants:*

Surfactants are used in a wide variety of household cleaning products such as soap and detergents. They are also used in different industrial applications like paint, agrochemicals, paper coatings, pharmaceuticals, etc.[2]. Removal of the surfactant sodium dodecyl sulphate by the process of EC using iron electrodes was investigated and it was observed that with the initial sodium dodecyl sulphate concentration of 100 mg/L, a CD of 1 mA/cm^2 and time of electrolysis of 10 min, 99% of sodium dodecyl sulphate was removed[40].

In another examination, titanium electrodes were used in the process of EC for the treatment of carwash water. With the pH of wastewater being in the range of 4 to 8, a CD of 0.25–3 mA/cm^2 and the stirring speed of 150–350 rpm, the removal efficiency of anionic surfactants and COD was found to be 99.3% and 84%, respectively. This result showed that the removal of surfactant is feasible with EC using titanium electrodes, however the cost of the process increases due to the use of costly titanium electrodes. Hence, titanium electrodes can be hybridized with low-cost materials such as iron or aluminium to minimize the fabrication cost of EC setups[18]. Aluminium electrodes were also employed for the process of EC, to determine the removal of nonylphenolpolyethoxylate surfactant from hospital wastewater. The result obtained showed that 95% removal of nonylphenolpolyethoxylate could be achieved with 30 min of electrolysis time and current of 1.5 A[41].

(D) Pesticides:

Agricultural applications are the major source of pesticides released in the environment. However, their presence in urban wastewater treatment plants is because of their non-agricultural uses, such as their application in golf courses, public parks, institutions, etc.[42]. Electrocoagulation process for the degradation of different types of pesticides was investigated by various researchers. In one demonstration, aluminium electrodes were used for the removal of dicofol pesticide from synthetic wastewater. With the time of electrolysis being 120 min, an applied voltage of 15 V, solution pH of 5, stirring at 750 rpm and electrode spacing of 8 cm, 95.52% dicofol removal efficiency was achieved[43].

In another demonstration, malathion pesticide removal from wastewater through EC was determined with aluminium electrodes. Over 90% of malathion pesticide was removed with the treatment time of 10 min and with initial pesticide concentration of 40 mg/L, initial pH of 6, a CD of 10 mA/cm^2, electrolyte concentration of 2.5 g/L and electrode distance of 2 cm[10]. Malathion pesticide was also removed using iron electrodes and it was observed that 98% of malathion pesticide was removed from wastewater with 10 min of treatment time, initial pH of 6–7 and CD of 1 mA/cm^2. Moreover, the result obtained also showed that the degradation of pesticide follows the pseudo-second order reaction kinetics[44]. Therefore, both aluminium and iron electrodes have ability to remove the malathion pesticide with iron showing better removal efficiency as compared to aluminium.

Propiconazole removal with aluminium electrodes employing EC was also investigated. It was observed that 79.83% of propiconazole pesticide was removed from wastewater with initial concentration of 25.93 mg/L, 36.44 min of treatment time and a CD of 10.95 mA/cm^2. Moreover, it was also observed that pH is not an essential parameter for the removal of propiconazole, however neutral pH was found to be ideal for attaining higher removal efficiency[23].

(E) Pharmaceuticals:

One of the major sources from which pharmaceuticals are released into the environment is domestic sewage as because both humans and animals are dosed with drugs or medicines. Untreated hospital effluent is also one of the sources of pharmaceuticals getting released in the natural

water bodies[9]. In several investigations, EC was used for the removal of pharmaceuticals from wastewater. Aluminium electrodes were used for the removal of ciprofloxacin from hospital wastewater. The optimal operating conditions were observed to be 20 min of treatment time, pH of 7.7 and a CD of 12.5 mA/cm^2. For initial ciprofloxacin concentration of 32.5 mg/L, 88.57% removal efficiency of ciprofloxacin was achieved. Moreover, the consumption of electrode material and energy were found to be 66.80 g/m^3 and 0.613 kWh/m^3, respectively for the process[45].

In another investigation, iron electrodes were used for the removal of metronidazole by EC from the hospital wastewater. Under optimal operating conditions of 20 min electrolysis time, CD of 6 mA/cm^2 and initial metronidazole concentration of 21.6 mg/L, 100% of the pollutant was removed (Table 1). It was also observed that the kinetics of the EC process for the removal of metronidazole followed the pseudo-second order reaction model[46].

Electrocoagulation process with aluminium electrodes was also used for the removal of dexamethasone from wastewater. Only 38% of the dexamethasone removal was achieved under optimized operating conditions. When real wastewater was used, almost similar results were obtained with 45 min of treatment time. Moreover, it was also observed that when the pH of influent was 8.5, the residual aluminium ion concentration was lower than 10 mg/L and when pH was brought down to 6.5, the concentration of aluminium obtained was 0.30 mg/L. Hence, pH adjustment is essential when aluminium is used as electrode material; otherwise, it will have an adverse effect on the environment as more residue of aluminium could be found in the treated effluent[47].

Table 1. Removal efficiency (η%) of some of the major xenobiotics

Xenobiotics	Electrodes	Current density (mA/cm^2)	Time (min)	η (%)	Ref.
Phenols	Al	75	25	91.0	48
	Al	25	60	91.0	49
	Fe	70	30	85.0	50
Dyes	Fe	12.78	06	83.0	51
	Al	15	05	98.0	52
	Fe	7.50	05	98.0	52
Surfactants	Al	5.26	05	90.0	53
	Fe	0.50	10	81.6	54
	Fe	3.12	60	94.9	55
Pesticides	Fe	1	10	97.0	44
	Al	6.2	60	91.2	16
	Al	10.6	70	95.0	56
Pharmaceuticals	Al	19.44	210	72.8	57
	Al	5.39	80	90	58
	Fe	6	20	100	46

Cost analysis of electrocoagulation

The economics of any treatment system plays an essential role in the desired degree of pollutant removal from the wastewater. Hence, it is vital to discuss the cost of treatment of the EC process. The cost involved in EC includes the cost of energy consumption, electrode material and the cost of addition of any external chemicals for changing the parameters, thus improving the removal efficiency of EC process[15]. Cost analysis of textile dye wastewater treatment demonstrated that when iron and aluminium electrodes were used, the operating cost was 0.1 and 0.3 US$/kg COD, respectively. Consumption cost of the electrodes was 80% of the total cost of aluminium electrodes and 50% of the total cost of iron electrodes[59]. The operating cost for 95% removal of malathion pesticide was found to be 0.76 US$/m^3 with a treatment time of 60 min and applied current of 1 A[10]. Electrocoagulation was also successfully used for the removal of cypermethrin. It was observed that 96.2% removal of cypermethrin was achieved with the operational cost of 22.4 US$/m^3 [60].

Conclusion and outlook

Electrocoagulation technology is feasible for the removal of xenobiotic compounds from the wastewater and it has the potential of offering tertiary treatment to the wastewater. Aluminium and iron have been used as electrodes in most of the investigations. Xenobiotics like phenolic compounds, dyes, surfactants, pesticides and pharmaceuticals have been reported to be removed with efficiencies higher than 95% employing metal electrodes within shorter reaction time. However, limited research has been focussed on the treatment of pharmaceuticals and emerging pollutants using EC. Thus, more investigations should be directed on the kinetics of the process for the removal of xenobiotic in order to understand the mechanism of pollutant removal. However, sludge generation, cathode passivation and high operating cost are the limitations of EC. Integrating EC with existing technology such as the combination of stacked microbial fuel

cell with EC can help in minimizing the electricity consumption and operating cost. Therefore, researchers should work on these bottlenecks, which presently limit the application of EC for the treatment of wastewater.

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