Electrochemical Treatment of Hazardous Organic Pollutants – A Status Review

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Abstract

The rapid industrial development, while giving a significant boost to the quality of human life, has also taken a heavy toll on our environment. One of the manifest effects is the proliferation of hazardous organic pollutants in the environment. These pollutants have many environmental and health implications like persistence and biomagnification, mutagenic and carcinogenic nature, endocrine disruption, and ecotoxicity to name a few.

Being mostly non-biodegradable or biorecalcitrant, they cannot be effectively treated with biological wastewater treatment methods. Physical separation methods only shift the pollutants from one stream to the other pending a final disposal. Advanced treatment techniques are, therefore, needed to deal with such pollutants. Against this backdrop, the electrochemical treatment methods are significant and relevant.

Electrocoagulation utilizes a sacrificial anode, often iron or aluminium, for the in-situ generation of Fe(III) or Al(III) ions as the coagulationg agent. This method has been found to be partially successful in the remediation of wastewater containing hazardous organic pollutants. But the most important of the electrochemical treatment methods is fast emerging technique of Electrochemical Advanced Oxidation, also known as Electrochemical Incineration. This aims at mineralization of the target pollutant by generating highly reactive hydroxyl radicals at the anode through dissociation of the water molecule. Certain pollutants also undergo direct oxidation at the anode through electrochemical charge transfer. The anode material, the inter-electrode potential, current density, and the supporting electrolyte have all been found to affect the process. Hydrogen is generated at the cathode which further adds to the attractiveness of the process.

Keywords: Hazardous organic pollutants, electrochemical incineration, electrocoagulation, anode material

1. Introduction

Today synthetic organic chemicals are ubiquitous and make their presence felt in almost every walk of life. While these chemicals have several advantageous characteristics one cannot wish away their darker side too. They often pose potential threat to the environment after their immediate intended use. These substances can be toxic and persistent. Many of them show bioaccumulation through the food web. Quite a few organic substances have been found to be mutagens and carcinogens and some are endocrine disruptors.

In recognition of the environmental implications of these substances the International conventions and government regulatory authorities of most of the countries have included them in their priority lists. The objective of the Stockholm Convention on Persistent Organic Pollutants is to 'protect human health and the environment from persistent organic pollutants' (Stockholm Convention, 2001). It lists 26 regulated organic chemicals and 5 more chemicals/chemical classes are under review for possible inclusion. Concise International Chemical Assessment Documents (CICADs) from International Programme on Chemical Safety lists about 60 odd organic chemicals with associated environmental issues (CICADS, IPCS). The United States Environmental Protection Agency (USEPA) under its National Waste Minimization Program provides a list of 31 priority chemicals, 28 of which are organic chemicals (USEPA, Priority Chemicals). The listed organic chemicals are persistent, bioaccumulative, and toxic (PBT). Environment Canada has provided two Priority Substances Lists containing 34 and 16 organic chemicals, respectively, many of which are conclusivey proven to be toxic (Environment Canada, PSL1 1989, PSL2 1995). In 1993, the Council of the European Communities adopted Council Regulation 793/93/EEC, thereby introducing a comprehensive framework for the evaluation and control of 'existing' chemical substances (EC, 1993). All but two of the 41 substances listed in the Priority Lists published from time to time are organic chemicals. The Manufacture, Storage and Import of Hazardous Chemical (Amendment) Rules, 2000, of the Ministry of Environment and Forests, Government of India, regulate the use of 94 toxic organic chemicals (MoEF, GOI, 2000). The priority chemicals list of 28 substances promulgated by The Environmental Management Bureau of Phillipines includes 18 organic chemicals

(Environmental Management Bureau, Phillipines, 1998). An illustrative (not exhaustive) list of some hazardous organic chemicals is presented in Table 1.

Pollutant	Class	Environmental impact
Nitrobenzene	Aromatic nitro compound	carcinogenic
Chlorobenzenes	Chlorinated organic	Persistent, volatile organic compound; bioaccumulate; toxic; carcinogenic
Chlorinated naphthalenes	Chlorinated organic	Highly bioaccumulative; toxic
Heptachlor	Biocide	Persistent; buioaccumulative; neurotoxic; potentially carcinogenic
Chlordane	Biocide	Persistent; buioaccumulative; neurotoxic; potentially carcinogenic
Polychlorinated biphenyls	Chlorinated organic	Persistent; bioaccumulative; Adverse effects on childbirth; carcinogenic
Polychlorinated dibenzo-para-dioxins	Chlorinated organic	Persistent, bioaccumulative, toxic, carcinogenic
Polychlorinated dibenzofurans	Chlorinated organic	Persistent, bioaccumulative, toxic
CI Acid Red 114	Azo dye	carcinogenic
CI Direct Blue 15	Azo dye	Carcinogenic; toxic
Phenobarbital	Pharmaceutical	carcinogenic

Table 1. Some biorecalcitrant organics and their environmental impacts

Most of the above mentioned organic substances cannot be adequately treated through conventional biological treatment methods. Physical separation methods like adsorption and membrane separation are also limited in their scope as they are unable to destroy the pollutants. Thus a final disposal problem is always imminent. Recently, advanced oxidation processes (AOPs) have been found to be effective in dealing with such pollutants. AOPs not only remove the pollutant but also achieve very high degree of mineralization in many cases due to the generation of highly reactive radical species, mainly hydroxyl radicals (Andreozzi et al., 1999; Hernandez et al., 2002). Electrochemical treatment is one of the very significant developments in AOPs that provide several benefits.

2. Electrocoagulation

The use of polyvalent metal ions for chemical precipitation of pollutants from effluent streams is well known. In these techniques a coagulant is added as an external source of the polyvalent metal ion. The examples are alum and ferric chloride with Al(III) and Fe(III) being the corresponding metal ions, respectively. In electrocoagulation the desired metal ions are generated in situ by using a sacrificial anode dipped in the effluent and applying a DC electrical bias potential to it. It consists of creating a floc of metal hydroxides by electrodissolution of the soluble sacrificial anode (Sengil and Ozacar, 2009).

2.1 Electrocoagulation process description

Aluminum and iron are the two most widely used anode materials used in electrocoagulation. In the case of aluminum the reactions are believed to be (Daneshvar et al., 2006; Kobya et al., 2003):

At anode:

$$Al \leftrightarrow Al^{3+} + 3\,e^- \quad (1)$$

At cathode:

$$H_2O + e^- \leftrightarrow H_2 + OH^- \quad (2)$$

The Al(III) ions and the hydroxyl ions generated in the above reactions combine to produce several monomeric and polymeric hydroxylated species like $Al(OH)^{2+}$, $Al(OH)_2^+$, $Al(OH)_4^-$, $Al_6(OH)_{15}^{3+}$, $Al_7(OH)_{17}^{4+}$, $Al_8(OH)_{20}^{4+}$, and $Al_{13}(OH)_{34}^{5+}$ which finally precipitate as $Al(OH)_3$ (Daneshvar et al., 2006; Mollah et al., 2004; Ugurlu et al., 2008). These freshly formed $Al(OH)_3$ aggregates, owing to their large specific surface area, allow rapid adsorption of organic pollutants onto them. Colloidal impurities are also trapped inside. The flocks so formed either settle to the bottom or float with the generated Hydrogen bubbles (Kobya et al., 2003; Mollah et al., 2004). For iron anode different mechanisms have been proposed based on the solution pH (Daneshvar et al., 2006;

Sengil and Ozacar, 2009). The reactions involved are:

Acidic pH; at anode:

$$\mathrm{Fe} \leftrightarrow \mathrm{Fe}^{2+} + 2\,e^- \qquad (3)$$

$$2\,H_2O \leftrightarrow O_2 + 4\,H^+ + 4\,e^- \qquad (4)$$

$$4\,\mathrm{Fe}^{2+} + 10\,H_2O + O_2 \leftrightarrow 4\,\mathrm{Fe(OH)}_3 + 8\,H^+ \qquad (5)$$

At cathode:

$$2\,H^+ + 2\,e^- \leftrightarrow H_2 \qquad (6)$$

Alkaline pH; at anode:

$$\mathrm{Fe} \leftrightarrow \mathrm{Fe}^{2+} + 2\,e^- \qquad (3)$$

$$\mathrm{Fe}^{2+} + 2\,OH^- \leftrightarrow \mathrm{Fe(OH)}_2 \qquad (7)$$

At cathode:

$$2\,H_2O + 2\,e^- \leftrightarrow H_2 + 2\,OH^- \qquad (8)$$

Various monomeric and polymeric hydroxy complexes are then formed such as $\mathrm{Fe(H_2O)_6}^{3+}$, $\mathrm{Fe(H_2O)_5(OH)}^{2+}$, $\mathrm{Fe(H_2O)_4(OH)_2}^{+}$, $\mathrm{Fe_2(H_2O)_8(OH)_2}^{4+}$, and $\mathrm{Fe_2(H_2O)_6(OH)_4}^{4+}$ which act as adsorbent for the organic pollutants (Mahesh et al., 2006; Mollah et al., 2004; Ugurlu et al., 2008; Sengil and Ozacar, 2009). In some cases they can also form complexes with the organics. In a variation of the process called the peroxy electrocoagulation process externally added hydrogen peroxide generates Fenton reactive system and synergistically works with electrocoagulation (Qiang et al., 2003; Yuksel et al., 2009).

2.2 Wastewater treatment using electrocoagulation

Electrocoagulation has been investigated as a potential remediation method for several pollutants and wastewater. It has been effective in the removal of colour, COD and TOC from effluents containing dyestuffs like those from textile industries or pulp and paper industries. To name a few, reactive blue, disperse red, CI acid red, bomaplex red, CI reactive black have been treated by electrocoagulation (Aleboyeh et al., 2008; Sengil and Ozacar, 2009; Yildiz, 2008; Zidane et al., 2008). Electrocoagulation was found to be effective in the treatment of effluents from pharmaceuticals industries (Boroski et al., 2009). In one study sodium dodecyl sulfate was effectively treated with peroxy electrocoagulation (Yuksel et al., 2009). Black liquor as well as effluents from paper industry has been treated with electrocoagulation (Mahesh et al., 2006; Zaied and Bellakhal, 2009). Several studies have suggested the efficacy of electrocoagulation for the treatment of wastewaters containing oil emulsions (Bensadok et al., 2008; Canizares et al., 2007). Electrocoagulation is also suited for the removal of heavy metals from aqueous systems. Removal of Zn(II), Cu(II), Ni(II), Ag(I) and Cr(VI) have been attempted (Heidmann and Calmano, 2008). Other wastewaters from diverse sources that have been treated with electrocoagulation are tannery wastewater (Feng et al., 2007), landfill leachate (Li et al., 2011), petroleum refinery wastewater (El-Naas et al., 2009), and laundry wastewater (Wang et al., 2009).

3. Electrochemical incineration

This is a process where hydroxyl radicals are generated at the anode by the oxidation of water or hydroxyl ions, depending on system pH. The reactions involved are (Dhaouadi and Adhoum, 2009; Ghatak et al., 2008):

Acidic pH; at anode:

$$H_2O \leftrightarrow OH\bullet + e^- + H^+ \qquad (9)$$

Alkaline pH; at anode:

$$OH^- \leftrightarrow OH\bullet + e^- \qquad (10)$$

The hydroxyl radicals so generated react with the organic pollutants due to their very high oxidizing power, ultimately leading to mineralization with the production of CO_2, H_2O and other inorganic ions. The reactions at the cathode are the same as in the case of electrocoagulation and represented by reactions (6), and (8) above.

3.1 The choice of electrode material

Besides reacting with the organics, the hydroxyl radicals generated at the anode can undergo a second anodic charge transfer to produce nascent oxygen as (Scialdone et al., 2008):

$$OH\bullet \leftrightarrow \bullet O\bullet + e^- + H^+ \qquad (11)$$

The nascent oxygen can react with the organic pollutants but its reactivity is much less than that of hydroxyl radicals. Instead they combine together to produce molecular oxygen through the reaction

$$2\ \bullet O\bullet \rightarrow O_2 \qquad (12)$$

These side reactions, (11), and (12), effectively waste the hydroxyl radicals and reduce the current efficiency of the process for mineralization. To minimize this anodes with high oxygen evolution overpotential are preferred (Marselli et al., 2003). In addition the material should be able to withstand the extreme oxidizing environment prevailing. With the current state of the art Boron Doped Diamond (BDD) electrodes best fulfil these criteria. They have high oxygen evolution overpotential and high anodic stability (Scialdone et al., 2008). This is attributed to the inactivity of BDD to chemisorption of hydroxyl radicals due to the low adsorption enthalpy (Kapalka et al., 2009; Pereira et al., 2012), thus minimizing the progress of the reaction (11). The hydroxyl radicals are, therefore, much more available in the bulk of the liquid for the destruction of the pollutants (Bejan et al., 2012). Other anodes with high oxygen evolution overpotential are βPbO_2, and Ti/SnO_2. They are known as “non-active” anodes, because they do not bind the oxygen atom of the hydroxyl covalently (Bejan et al., 2012). PbO_2 has the disadvantage of possible lead contamination while SnO_2 loses its activity over time. The electrical conductivity of SnO_2 can be significantly improved by doping with antimony, ruthenium, iridium and rare earth elements (Gomez et al., 2006; Hu et al., 2007). The electrocatalytic activity of PbO_2 can also be improved by incorporating other metal oxides such as Bi, and Ce (Ai et al., 2004; Mohd and Pletcher, 2006). Noble metal and noble metal oxide anode have also been tried for electrochemical incineration applications. Materials like Pt, Ti/RuO_2, and Ti/IrO_2-Ta_2O_5 have exhibited low overpotential for oxygen evolution as they chemisorb the hydroxyl radicals with formation of covalent bond between the metal and oxygen. Thus, they have less ability to provide hydroxyl radicals for pollutant mineralization (Panizza and Cerisola, 2009). Such “active anodes are, therefore, less suitable for electrochemical incineration.

3.2 Wastewater treatment using electrochemical incineration

Nitrophenols were electrochemically incinerated on Ce-Ru-SnO_2 anodes with 60-80% COD reduction in 300 minutes (Liu et al., 2012). In related studies o-nitrophenol was mineralized with electrochemical incineration on Ti/Bi-PbO_2 anode (Liu and Liu, 2008). Several chlorophenols were electrochemically incinerated on BDD anode and the kinetics and mechanism were examined (Canizares et al., 2003). Other investigators treated chlorophenols with glassy carbon electrode and observed the formation of a polymeric film (Berrios et al., 2008). Trichloroethylene could be degraded and mineralized on the ceramic anode ebonex which is a nonstoichiometric titanium oxide comprised of Magneli phase titanium oxides Ti_4O_7 and Ti_5O_9 (Chen et al., 1999). Similarly, several chloroethanes and carboxylic acids were effectively treated with BDD anode (Scialdone et al., 2012). Aqueous phenol wastes have been subjected to combined process of electrochemical incineration and electrocoagulation with stainless steel anode (Canizares et al., 2002). 90% COD abatement of bisphenol A was achieved by electrochemical incineration over BDD anode in the presence of NaCl as supporting electrolyte (Pereira et al., 2012). The pharmaceutical sulfamethoxazole was mineralized to a high degree with high current efficiency on BDD anode (Li et al., 2008). Azoic dyes Reactive Red 141, Direct Black 22, and Disperse Orange 29 and anthraquinonic dye Acid Blue 62 were completely decolourized and highly mineralized with BDD anode electrochemical incineration in the presence of different supporting electrolytes (Aquino et al., 2012). Similarly, the azoic dyes Eriochrome black T, methyl orange, and congo red were degraded with significant reduction of COD and TOC with BDD anode (Faouzi et al., 2006azoic dyes). Several pollutants, namely phenol, ibuprofen, acid orange 7, and diclofenac were mineralized between 70 – 90% by electrochemical incineration on Ti/SnO_2–Sb_2O_4 (Ciriaco et al., 2011).

4. Conclusions

Hazardous organic pollutants pose a serious threat to the environment and cannot be adequately managed using conventional biological or physico-chemical treatment methods. Electrochemical methods have shown promise in removal of such pollutants. Compared to other chemical precipitation methods electrocoagulation is cleaner and compact as it does not require the addition of an external agent. Electrochemical incineration has the potential to not only remove the target pollutants but also to mineralize them. Among the various anode materials investigated BDD has shown the highest efficiency in mineralizing the pollutants.

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