J. Bangladesh Acad. Sci., Vol. 39, No. 2, 125-134, 2015

REMOVAL OF LEAD FROM BATTERY EFFLUENT BY ELECTRO-COAGULATION

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ABSTRACT

The efficiency of iron electrode based electrocoagulation (EC) technique at laboratory scale to remove lead (Pb) from battery industrial effluent in Bangladesh is investigated. Different combinations of voltage (15, 30 and 45), effluent pH (1, 3, 5, 7 and 9) and electrolysis time (15, 30, 45 and 60 minutes) at the EC reactor was examined for searching the ideal operating conditions of maximum lead removal. Initial battery effluent pH of 3, electro coagulating at 30 V for 15 minutes would be the optimum conditions for treatment where 99.9% Pb removal was achieved. Treated effluent quality was compared with national environmental standard to discharge into surface water bodies and found physico-chemical parameters (TDS, TSS, DO and pH) were within prescribed limit except electrical conductivity.

Key words: Removal of lead, Electrocoagulation, Battery effluent

INTRODUCTION

One of the unwanted concerns of industrial booming in recent decades is the increasing of metal concentrations in natural water bodies, caused by the large output of industrial effluents contaminated with heavy metals (Rahman *et al.* 2012a). Heavy metal, such as lead (Pb) pollution has become one of the most serious environmental nuisances recently. Pb, often presents and discharges by number of industrial processes to the aquatic ecosystem, is hazardous even in low concentration (Martins *et al.* 2004). High levels of Pb (II) can be traced at industrial discharges from variety of sources, such as batteries, paints, pigments and ammunition, petrol, cables, alloys and steels, plastics, ceramic and glass industries (Ghaedi *et al.* 2007). Pb contamination is also due to emission from vehicular traffic and the mixing of roadside run-offs (Sari and Tuzen 2009). The presence of Pb (II) in drinking water is known to cause various types of health

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problems (Ahmedna *et al.* 2004). Although the inorganic form of Pb is a general metabolic poison and enzyme inhibitor, organic forms are even more poisonous (Lo *et al.* 1999). Lead poisoning in human causes severe damage to kidney, nervous system, circulatory system, reproductive system, cardiovascular system, liver and brain (Naiyaa *et al.* 2009). In adults, symptoms of neurological effects include dullness, irritability, poor attention span, headaches, muscular tremors, loss of memory, and hallucinations. The condition may then be worsen, sometimes abruptly, to delirium, convulsions, paralysis, coma and death.

Therefore, the removal of Pb from water and wastewater is important for the protection of public health and environment. The most widely practiced methods for removing heavy metals from wastewater include ion-exchange, chemical precipitation, filtration, reverse osmosis, evaporation, membrane filtration, pre-concentration, bio-sorption, adsorption, biological treatment etc. (Ghaedi *et al.* 2007). Most of these methods suffer from some drawbacks, such as high capital and operational cost, generation of secondary pollutants, disposal issue of the residual metallic sludge and are not suitable for small-scale industries (Kobya *et al.* 2005).

Electrocoagulation (EC) process has attracted a great attention in treating industrial wastewaters because of its versatility and environmental compatibility. This method is characterized by easy and safe operation with simple equipment set-up, relatively cheap, energy efficient, a shortened reactive retention period, and importantly not suffers from overburden of sludge production (Islam *et al.* 2011a, Mollah *et al.* 2004, Rahman *et al.* 2012b). Moreover, during EC, the salt content of the liquid does not increase appreciably, as in the case of chemical treatment. EC is a proved efficient, effective and reliable technology used for treating various process effluents (Islam *et al.* 2011a, b, Rahman *et al.* 2011b, 2012b). But, there is no literature to treat Pb from battery industry effluent by EC. Thus the goal of present research was to assess the performance of EC using sacrificial iron electrodes, on the removal of Pb from battery effluent of Rahimafrooz Accumulators Ltd. (RAL) industry, Bangladesh. To do so this work ultimately explored the effects of various process parameters in EC technique such as voltage, pH and reaction time on overall Pb removal.

MATERIALS AND METHODS

Untreated battery effluent samples were collected from effluent treatment plant (ETP) of RAL located in Zirani (23°59'45.70"N, 90°15'20.15"E), Gazipur district of Bangladesh. RAL produces and markets a wide range of industrial batteries which are used in telecommunication, power station, railways, electric vehicles, forklifts, ships, buoy lighting, ISP, UPS, inverter and solar power systems. Before discharging the effluent into the environment, RAL usually treats it chemically and thus the ETP requires high amount of chemicals. For this current experiment glass bottles of 500 ml capacity were used for effluent sampling from RAL and prior to sampling these containers were washed with 10% (V/V) HNO_3 solution and then rinsed thoroughly with deionized water. The containers were completely filled with sample effluent and sealed carefully. Samples were labeled with specific code, stored in an ice box maintaining ~5°C temperature and then immediately transferred to the laboratory for EC treatment. Physico-chemical parameter of samples such as total suspended solids (TSS), total dissolved solids (TDS), dissolved oxygen (DO), pH and conductivity were determined using gravimetric method, TDS meter (HANNA H18734), DO meter (Cyber Scan DO 110) and pH meter (EcoScan Ion 6), respectively.

EC is a complicated process involving many chemical and physical phenomena that use consumable electrodes to supply ions into the wastewater stream. In an EC process the coagulating ions are produced *in situ* and it involves three successive stages: (i) formation of coagulants by electrolytic oxidation of the ‘sacrificial electrode’, (ii) destabilization of the contaminants, particulate suspension, and breaking of emulsions and (iii) aggregation of the destabilized phases to form flocs (Mollah *et al.* 2004). The destabilization mechanisms of the contaminants, particulate suspension, and breaking of emulsions have been described in broad steps and may be summarized as follows: (a) compression of the diffuse double layer around the charged species by the interactions of ions generated by oxidation of the sacrificial anode, (b) charge neutralization of the ionic species present in wastewater by counter ions produced by the electrochemical dissolution of the sacrificial anode. These counter ions reduce the electrostatic inter-particle repulsion to the extent that the van der Waals attraction predominates, thus causing coagulation. A zero net charge results in the process and (c) floc formation as a result of coagulation creates a sludge blanket that entraps and bridges colloidal particles still remaining in the aqueous medium (Mollah *et al.* 2004). The solid oxides, hydroxides and oxy-hydroxides provide active surfaces for the adsorption of the polluting species.

The experiments were performed at a laboratory bench scale (Fig. 1) by employing effluent from RAL battery industry. A detail description of the EC experimental set up is available (Islam *et al.* 2011a and b, Rahman *et al.* 2012b). Low-cost two iron plates as the sacrificial electrodes have been deployed with 4 cm gap in a reactor which was 250 ml capacity Pyrex glass beaker. Within this reactor roughly 4 A constant current was passing through two iron electrodes of approximately 30 cm^2 surface area in combination of both anode and cathode and thus the overall current density of this electrochemical process was nearly 0.1 A/cm^2. The initial pH of the raw water was 1 and hence post adjustment was required using NaOH to get the desire pH of the solution. The experiment was carried out at different pH (1, 3, 5, 7 and 9), voltage (15, 30 and 45) and electrolysis time (15, 30, 45 and 60 minutes). The purity of the iron electrodes was about 99.8%. Before use, the electrodes were cleaned manually by abrading with sand papers and then washed with dilute acid to remove impurities on the metal surface. At the end of each run, the solution was filtered and then the filtrate was analyzed, the electrodes were washed thoroughly with water to remove any solid residues on the surfaces, dried and reweighted.

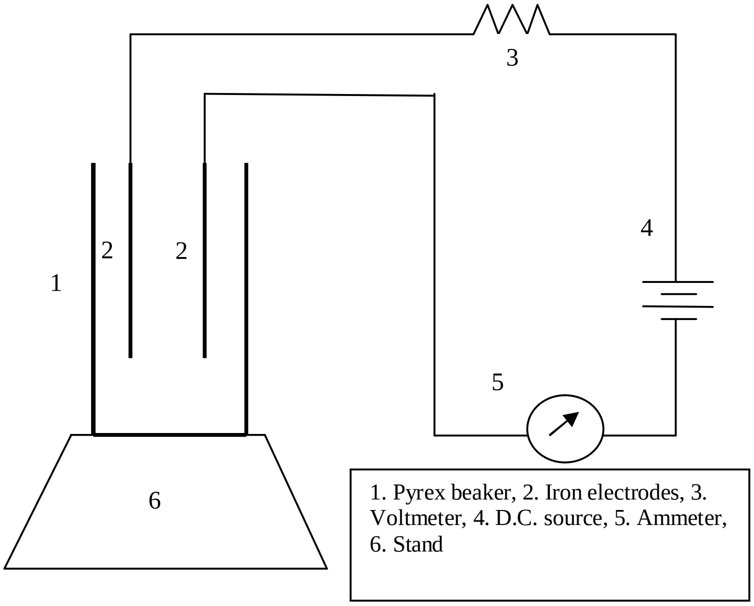

Fig. 1. Schematic diagram of the EC cell (1. Pyrex beaker, 2. Iron electrodes, 3. Voltmeter, 4. D.C. Source, 5. Ammeter and 6. Stand).

Pb concentration of before and after treatment was measured by ICPMS (Inductively Coupled Plasma Mass Spectrometry) (Varian, Australia), F-AAS (Flame-Atomic Absorption Spectrometry) (Varian, AA 240 FS) and GF-AAS (Graphite Furnace-Atomic Absorption Spectrometry (Varian, AA 240Z). This advanced instrumental analysis was carried out from the Analytical Research Division, Bangladesh Council of Scientific and Industrial Research (BCSIR), Dhaka, Bangladesh. Pb removal efficiency (% RE) by EC was calculated following the equation 1.

$$\%RE = (C_0 - C_f)/C_0 \times 100 \qquad (1)$$

where, C_o and C_f are the concentrations of Pb before and after the experiment, respectively.

RESULTS AND DISCUSSION

Characteristics of raw battery effluent were shown in Table 1. Effluent was strongly acidic in nature with suspended materials. Several operating parameters, such as initial pH, voltage and electrolysis time were investigated in order to evaluate the efficiency of EC for Pb removal.

Table 1. Physico-chemical characteristics of battery effluent.

Parameter	Unit	Effluent value	DoE standard (DoE, 2003)
pH		1	6-9
TSS	mg/l	36	150-500
TDS	”	7480	2100
DO	”	7.80	4.5-8
EC	µs/cm	14023	1200
Pb	µg/l	4600	100

With the increased electrolysis time and voltage, pH value of the solution reached maximum 5.28 from initial pH 1 (Fig. 2). But this was lower than the standard value to discharge in the environment. Therefore, pre-adjustment of pH was required and was adjusted to 3, 5, 7 and 9 using NaOH of untreated effluents. The new solutions of different pH were kept to see the effect of operating time with pH.

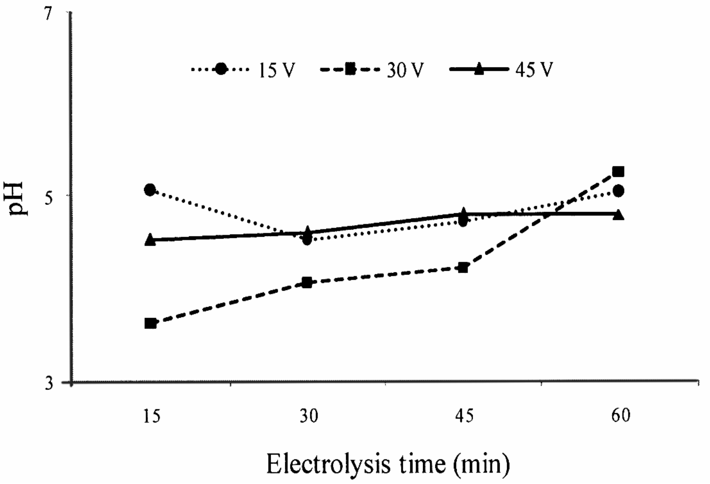

Fig. 2. Effects of voltage and electrolysis time on initial wastewater pH (at pH = 1).

The final pH also increased with the increased operating time (Fig. 3), which was due to the OH^- ion accumulation in aqueous solution during the process. Increase of pH at acidic condition was attributed to hydrogen evolution at cathodes (Nabil *et al.* 1998).

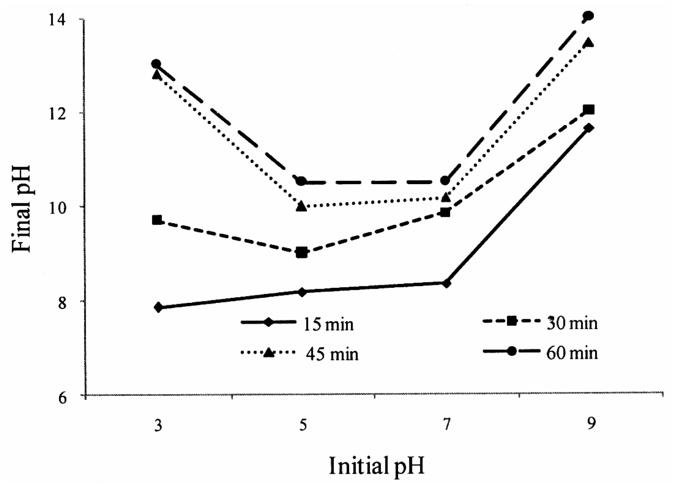

Fig. 3. Variation of pH at various times on electrolysis at constant voltage 30.

When EC proceeds in aqueous solution, the pH of the medium has a considerable effect on its performance as well as on the solubility of the analyte (Escobar *et al.* 2005). To improve the efficiency of Pb removal, the effect of the initial wastewater pH was also explored in the range of 1 to 9 at various electrolysis times. With the increased pH at various electrolysis times, the removal efficiency of Pb also increased (Fig. 4). Subsequently,

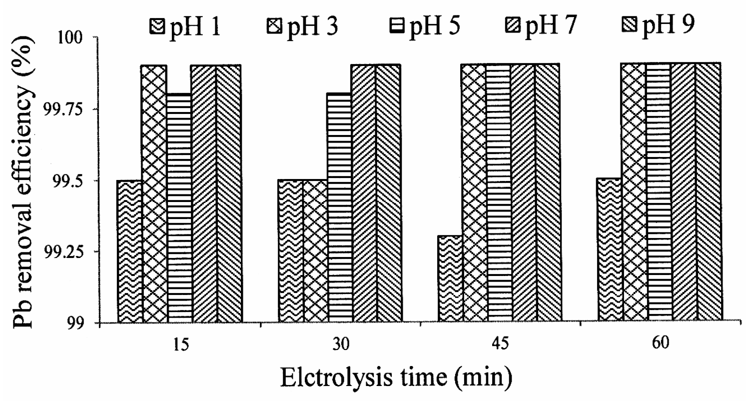

Fig. 4. Removal of Pb as a function of time at different pH at 30V.

when EC was applied to the wastewater with a high initial pH, the degree of removal of metals increased rapidly to their maximum because, at high initial pH, the structures of ferrous ions in the system were in the monomeric ($Fe(OH)_3$) form and polyhydroxyl iron (III) complexes such as $Fe(OH)^{2+,}$ $Fe(OH)_2^+$, $Fe(H_2O)_5OH^{2+}$ and $Fe(H_2O)_4(OH)_2^+$ which can combine with the metal ion molecules causing the coagulation and the reduction of heavy metals occurred.

Pb removal efficiency was fluctuated with increased voltage at different electrolysis time (Fig. 5). In high voltages, size and growth rate of produced flocs increase and affects the efficiency of the process (Zhu *et al.* 2005). At constant time of 45 and 60 minutes, the removal efficiency increased with increased voltage and reached above 99%.

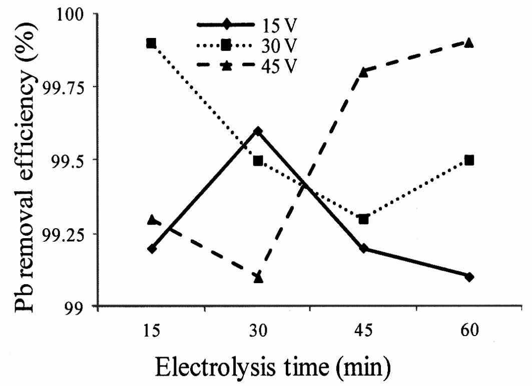

Fig. 5. Removal of Pb as a function of time at various voltages at constant pH 1.

Increase in retention time in the case of constant voltage and pH, increases the efficiency of lead removal due to precipitation of metal from effluent by forming flocs. The removal efficiency increased with increasing electrolysis time with change of pH 1, 3, 5, 7 and 9 and voltage 15, 30 and 45 V (Fig. 6). This is expected since a high applied current over a long electrolysis time will generate a large amount of coagulant according to Faraday's law, leading to lower amount of heavy metals in the treated wastewater (Chaloempan *et al.* 2009). With the formation of coagulants, during the treatment process, hydrogen gas was released from the cathode which can help to float the flocculated particles out of the water, a process which is sometimes called electro-flocculation (Chen 2004). The main reactions were as equations 2 - 7:

Anode reaction (oxidation):

$$Fe \leftrightarrow Fe^{2+} + 2e^{-} \quad (2)$$

$$Pb^{2+} + Fe^{2+} \leftrightarrow Fe + Pb^{4+} \quad (3)$$

Cathode reaction (reduction):

$$2H_2O + 2e^{-} \leftrightarrow H_2 + 2OH^{-} \quad (4)$$

Co-precipitation:

$$Pb^{2+} + 2OH^{-} \leftrightarrow Pb\,(OH)_2\downarrow \quad (5)$$

$$Pe^{3+} + 30H^{-} \leftrightarrow Fe\,(OH)_3 \quad (6)$$

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

In the presence of iron electrodes, the ferric ions generated by the electrochemical oxidation of the iron electrode, according to equation 2, may form mono-meric species, such as $Fe(OH)^{2+}$, $Fe(OH)_2^{+}$, $Fe(H_2O)_5OH^{2+}$, $Fe(H_2O)_4OH_2^{+}$, $Fe(OH)_3$, $Fe(OH)_4$, and polymeric species, such as $Fe_2(H_2O)_8OH_2^{4+}$ and $Fe_2(H_2O)_6OH_4^{2+}$, depending on the pH of the aqueous medium in the EC process (Gomes *et al.* 2007, Kobya *et al.* 2008). These complex molecules can act as adsorbents and /or traps for Pb ions and so eliminate them from wastewater.

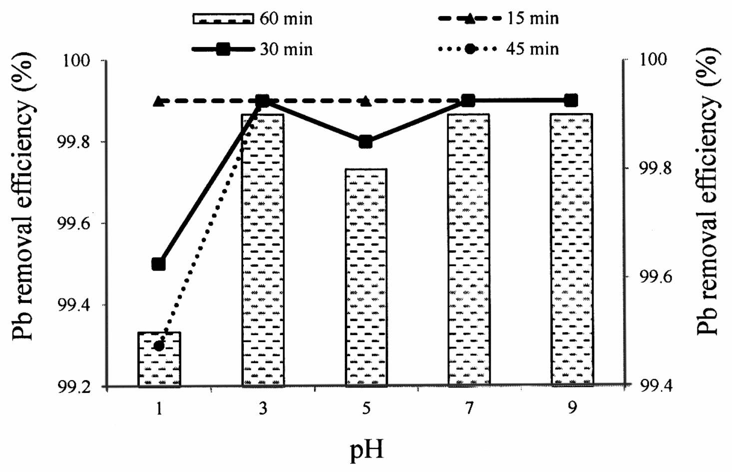

Fig. 6. Removal percentage of Pb as a function of pH with different electrolysis time at constant voltage 30 (secondary Y axis is for 60 min experiments only).

The treated effluent quality was compared with wastewater reuse quality standards for inland surface water (Table 2). The comparison allowed to judge the effectiveness of EC process for the treatment of Pb from battery effluent of RAL. After EC treatment all parameters of effluent except electrical conductivity were within standard of Department of Environment, Bangladesh (DoE 2003).

Table 2. Comparison of treated wastewater quality with various wastewater quality standards.

Parameter	Unit	After treatment			DoE standard (DoE, 2003)
		Minimum	Maximum	Mean	
TDS	mg/l	470	1450	640	2100
TSS	mg/l	2.74	97.4	50.7	150
pH	-	3.62	14	8.81	6 - 9
EC	µs/cm	960	3220	2090	1200
DO	mg/l	5.10	8.48	6.79	4.5 - 8
Pb	µg/l	0.03	37.9	19	100

The final pH of treated effluent was varied from 3.62 to 14. At pH 3, 5 and 7 with 15 minutes electrolysis and at pH 5 with 30 minutes electrolysis, the final pH of effluent was within allowable limit. In addition, EC process could generate less amount of sludge than conventional chemical treatment option. The sludge generated from this technique is known as electrocoagulated metal hydroxide sludge that can be further used in other purposes, likely building block materials (Adyel *et al.* 2012, 2013).

CONCLUSIONS

From the laboratory bench scale experiments, EC was found effective process for treating Pb from battery industry effluent. The removal efficiency was influenced by various parameters including the voltage, initial effluent pH and electrolysis time. In all working conditions, Pb removal was above 90% but the maximum removal was achieved during passing 30 V of electricity for 15 minutes in the effluent with pH 3. After EC treatment, Pb concentration reached 0.03 - 37.9 μg/l which was within the permissible limit (100 μg/l) for discharging into inland surface water. Therefore, EC would be a promising technology to treat battery industry effluent.

ACKNOWLEDGMENTS

The authors are grateful to the Bangladesh Council of Scientific and Industrial Research (BCSIR) for providing laboratory facilities.

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(*Received revised manuscript on 17 August, 2015*)