

IOP Conference Series: Materials Science and Engineering

PAPER • OPEN ACCESS

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To cite this article: N M Yussuf *et al* 2018 *IOP Conf. Ser.: Mater. Sci. Eng.* **298** 012006

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The enhancement of heavy metal removal from polluted river water treatment by integrated carbon-aluminium electrodes using electrochemical method

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Abstract. The heavy metal removal enhancement from polluted river water was investigated using two types of electrodes consist of integrated carbon-aluminium and a conventional aluminium plate electrode at laboratory-scale experiments. In the integrated electrode systems, the aluminium electrode surface was coated with carbon using mixed slurry containing carbon black, polyvinyl acetate and methanol. The electrochemical treatment was conducted on the parameter condition of 90V applied voltage, 3cm of electrode distance and 60 minutes of electrolysis operational time. Surface of both electrodes was investigated for pre and post electrolysis treatment by using SEM-EDX analytical technique. Comparison between both of the electrode configuration exhibits that more metals were accumulated on carbon integrated electrode surfaces for both anode and cathode, and more heavy metals were detected on the cathode. The atomic percentage of metals distributed on the cathode conventional electrode surface consist of Al (94.62%), Zn (1.19%), Mn (0.73%), Fe (2.81%) and Cu (0.64%), while on the anode contained O (12.08%), Al (87.63%) and Zn (0.29%). Meanwhile, cathode surface of integrated electrode was accumulated with more metals; O (75.40%), Al (21.06%), Zn (0.45%), Mn (0.22), Fe (0.29%), Cu (0.84%), Pb (0.47%), Na (0.94%), Cr (0.08%), Ni (0.02%) and Ag (0.22%), while on anode contain Al (3.48%), Fe (0.49%), C (95.77%), and Pb (0.26%). According to this experiment, it was found that integrated carbon-aluminium electrodes have a great potential to accumulate more heavy metal species from polluted water compare to the conventional aluminium electrode. Here, heavy metal accumulation process obviously very significant on the cathode surface.

1. Introduction

Inorganic pollutants specifically that refer to heavy metals constitute a serious threat for the environment. Most of the metals such as copper, nickel, lead and zinc are harmful when they are discharged without treatment, because they are not biodegradable and tend to accumulate in living organisms, and many heavy metal ions are known to be toxic or carcinogenic [1]. Due to their high toxicity, these contaminated water are strictly concerned to be treated for metals removal. Various



techniques have been employed for the treatment of heavy metals, including precipitation, coagulation/flocculation, adsorption, biosorption, ion-exchange, electrochemical and membrane separation [2].

Among these techniques, precipitation was said to be the most applicable technique and economical. However, this technique produces a large amount of precipitate sludge that requires further treatment [1]. Another techniques such as reverse osmosis, ion-exchange and other membrane separation were limited due to a number of disadvantages such as high material and operational cost and their operational problems [3].

Nowadays, electrochemical technologies have reached such a state that they are not only comparable with other technologies in terms of cost but also are more efficient and more compact [4]. For some situations, electrochemical technologies may be the indispensable step in treating wastewater containing refractory pollutants [5]. Electrochemical remediation involves the application of electricity to the electrodes for contaminants removal and it is used for treating water that, among others, contains COD, oil wastes or metals [6].

Hence, the development of new treatment methods for effluents bearing heavy metals is an urgent issue. Among other methods that have recently been studied, electrochemical proved very efficient in removal pollutants such as organic and inorganic matters from the industrial wastewater [7]. In addition, a number of scientific works have indicated that heavy metals in the free form can be successfully removed by electrochemical using aluminium, iron and stainless steel electrodes [8].

There are few published studies using aluminium electrodes for the treatment of metal removal, no attempt has been made to investigate metal removal by employing an aluminium electrodes integrated with activated carbon. This study primarily aims to investigate the performance of activated carbon as electrodes material in Pb, Zn, Mn and Fe removal and compare its performance with those of conventional aluminium electrodes. The removal efficiency was calculated and the metal accumulation on electrodes were characterized.

2. Materials and method

2.1. Experimental procedure

The experimental equipment schematically is shown in figure 1. It consisted mainly of a Perspex with a specific dimension 30 cm length, 10 cm width and 7 cm height, the cell and electrical circuit. The cell consisted of two aluminium electrodes. The conventional electrodes was a solid aluminium of 10 cm length, 7 cm width and 0.1 cm thick which was supported on the container wall. The integrated aluminium electrodes were fabricated by coating activated carbon slurry onto a sheet of aluminium plate using a doctor knife. Electrode slurry was prepared by mixing activated carbon powder and the polyvinyl acetate (PVAc) and methanol. PVAc was added to bind the coating to the electrode surface and methanol was added to ensure dispersion of the ingredients and allow the application of an even coating onto the electrode surface [9]. The optimum ratio of the mixture was found to be 5g: 2g: 10 mL of AC: PVAc: Methanol. Then the mixture was spread on the aluminium electrodes surface, and the electrode was dried at 100°C for 2h in an oven. Final thickness of integrated electrodes formed was 0.2 cm, with 7cm width and 10 cm length. The gap between the cathode and anode was kept at 3 cm. The electrical circuit consisted of DC power supply and crocodile clippers connected to measure the cell.

2.2. Water sample characteristics

The polluted water sample was collected from *Sungai Senggarang*, Batu Pahat. It was then bring to the laboratory and preserved with nitric acid and stored in a refrigerator at 4°C for metal preservation. The experiment was conducted in a lab-scale size with two different electrode system; one with conventional aluminium electrode plates and the other one with aluminium plate integrated with activated carbon. The reactor cell was filled with 300 mL of river water while DC supply was adjusted to 90 V and the electrolysis time was set for 60 min. Both, pH and temperature were measured with a multiparameter. The multiparameter was calibrated according to the operating manual. The water samples were filtered with a 15 cm filter paper for atomic absorption spectroscopy (AAS) analysis.

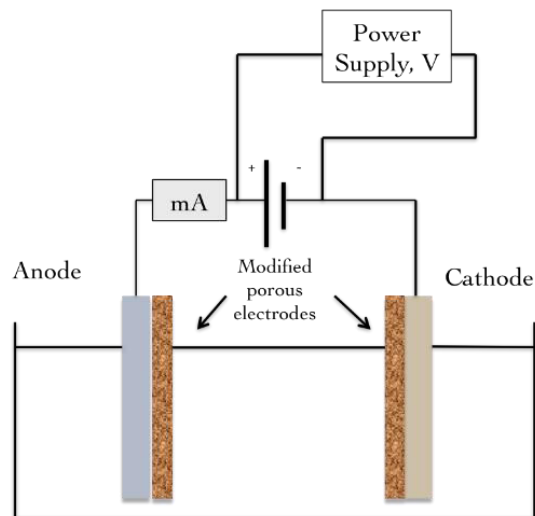


Figure 1. Schematic diagram of the experimental setup.

2.3. Measurements

At the beginning of each experiment ($t=0$), initial pH were measured with a Lutron pH -meter PH-222. The initial concentrations of the selected toxic metals (Mn, Fe, Zn and Pb) were, also, determined by atomic absorption spectroscopy (AAS), as mentioned above. After the 60 min electrolysis process, pH was measured for each liquid sample. Finally, 50 mL of river water samples were measured by AAS for Mn, Fe, Zn and Pb and surface of the electrodes were measured by scanning electron microscopy-energy-dispersive X-ray (SEM-EDX) for pre and post experiments. All experiments were conducted at ambient temperature.

3. Results and discussion

3.1. Removal efficiency of heavy metal

The analysis of Zn, Pb, Mn and Fe were evaluated as shown in table 1. This table gives the results of their concentration, standard deviation of concentration and relative standard deviation of concentration of the four heavy metal in water sample measured by atomic absorption spectroscopy (AAS) and comparison with United States Environmental Protection Agency (EPA) data. For the protection human health, guide lines for the presence of heavy metals in water have been set by International organization such as EPA, thus heavy metal have maximum permissible level in water specified by the organization. The maximum contaminant level (MCL) is an enforceable standard set at a numerical value with adequate margin of safety to ensure no adverse affect on human health [10].

Table 1. Concentration, standard deviation of concentration, relative standard concentration and maximum contaminant level (MCL) of Pb, Zn, Mn and Fe.

Metal	Concentration (mg/L)	SD	RSD (%)	MCL (mg/L) (EPA, 2009)
Pb	0.307	0.038	12.460	0.002
Zn	0.021	0.001	5.920	5.000
Mn	0.378	0.002	0.580	0.050
Fe	5.279	0.045	0.840	0.300

The four elements studied in this research were Pb, Zn, Mn and Fe has the maximum contamination levels of 0.015, 5.0, 0.05 and 0.3 mg/L respectively [10] while the initial pH was 3.51. From the results obtained, the concentration levels for Pb, Mn and Fe were above the specified contaminant level except for Zn. One of the aims of the present work was to check the possibility of reducing or removing the contaminant in water sample by applying electrochemical method. For this reason, an experiment was carried out and the results for metal removal efficiency (%) and final pH are presented in table 2. Figure 2 shows the evolution of the removal efficiency, calculated for operating time 60 mins, as a function of voltage used 90 V. The removal efficiency in percent, is given by:

$$\text{Removal efficiency, \%} = \frac{(C_o - C_t) \times 100}{C_o} \quad (1)$$

where C_o and C_t are respectively the concentration of metal ions before and after the treatment.

Table 2 Metal removal (%) and final pH for conventional and integrated electrodes after 60 minutes electrochemical treatment.

	Conventional electrodes	Integrated electrodes
Removal (%)		
Mn	16.14	47.88
Fe	81.61	91.80
Zn	99.00	99.50
Pb	2.20	8.47
End pH	5.94	6.02

In general, the pH of the medium raises during electrochemical as a result of the electrolysis process and the production of hydroxyl ions in the aqueous solution. According to the results, it was remarkable since all the removal rates are higher for integrated electrodes showing that in these conditions the treatment operation performs better when carbon is used. Zn removal is the highest among all as the removal efficiency is 99.50% while Fe, Mn and Pb are respectively 91.80%, 47.88% and 8.47%. For the conventional electrodes, lower removal percentage were observed for Fe, Zn and Mn (81.61%, 99.0% and 16.14% respectively).

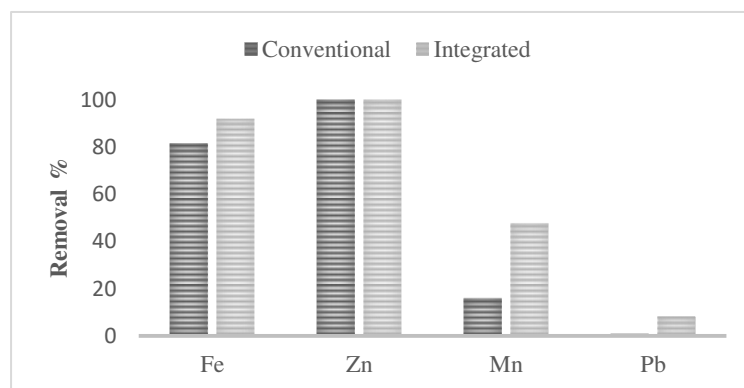


Figure 2. Comparison in metal removal efficiency between a conventional aluminium electrode and integrated electrode.

3.2. Metal accumulation on electrode plates

SEM micrographs and EDX spectra were obtained with HITACHI Horiba SU1510 scanning electron microscope equipped with a LINK analytical system. The electron energy used was 5.9 keV. EDX analysis revealed the higher metal accumulation was on carbon integrated electrode surface for both anode and cathode compared to conventional aluminium electrode, and more heavy metals were observed on the cathode. The atomic percentages of metals distributed on the cathode integrated electrode surface were O (75.40%), Al (21.06%), Zn (0.45%), Mn (0.22), Fe (0.29%), Cu (0.84%), Pb (0.47%), Na (0.94%), Cr (0.08%), Ni (0.02%) and Ag (0.22%), while on anode contain Al (3.48%), Fe (0.49%), C (95.77%), and Pb (0.26%) (Figure 4). In contrast, less heavy metal elements were detected on the conventional electrode surface. Instead, elements that were found on conventional anode surface such as O (12.08%), Al (87.63%) and Zn (0.29%), while on cathode surface are Al (94.62%), Zn (1.19%), Mn (0.73%), Fe (2.81%) and Cu (0.64%) (figure 3).

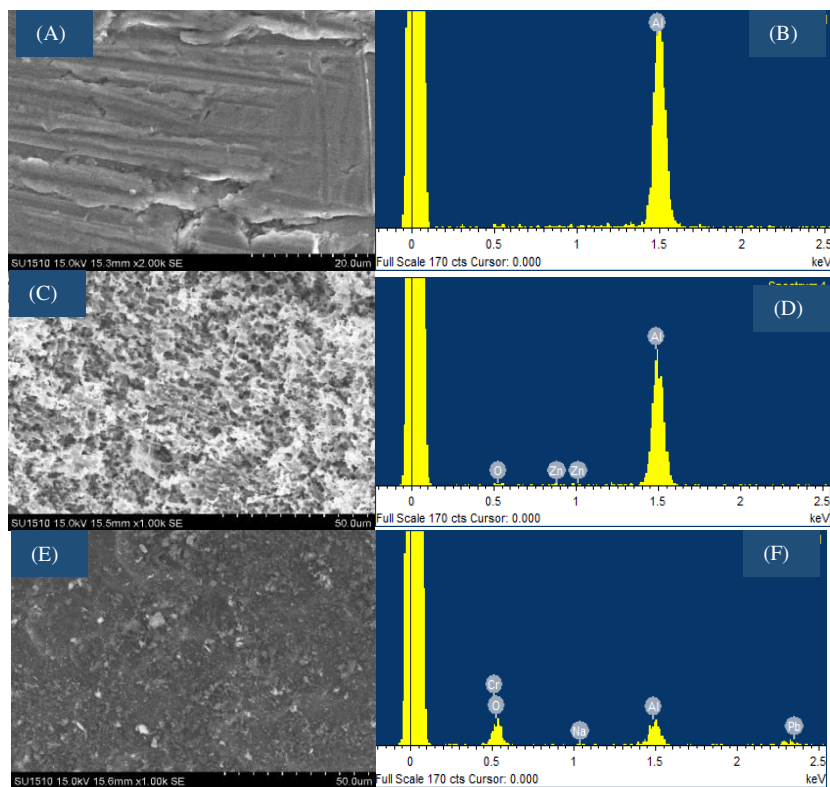


Figure 3. SEM photomicrograph of conventional aluminium electrodes (A) before treatment (C) anode surface after treatment (E) cathode surface after treatment. Corresponding EDX spectra for (A), (C) and (E) are in (B), (D) and (F), respectively.

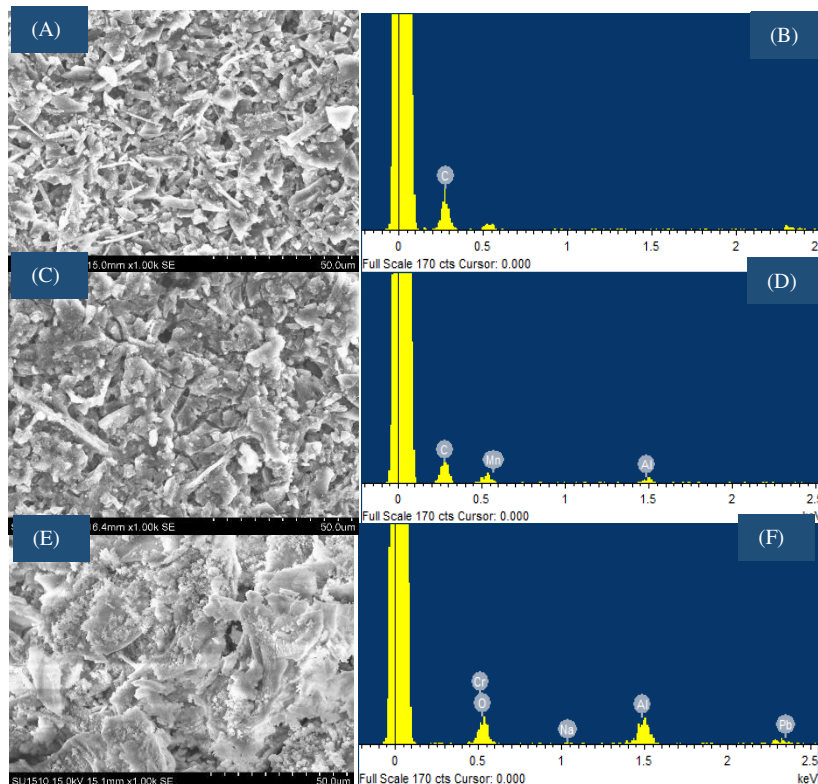


Figure 4. SEM photomicrograph of integrated aluminium electrodes (A) before treatment (C) anode surface after treatment (E) cathode surface after treatment. Corresponding EDX spectra for (A), (C) and (E) are in (B), (D) and (F), respectively.

4. Conclusion

From this work, some conclusion can be summarised as follows :

- Integrated electrode with carbon showed enhanced removal towards metal in water, particularly Fe, Mn, Pb and Zn.
- Fe and Zn showed higher removal efficiency 91.8% and 99.5% for integrated electrode; 81.6% and 99.0% for conventional electrodes, respectively while Pb and Mn removal are less than 50% for both electrodes.
- SEM-EDX analysis revealed the increment of metal accumulation especially on cathode surface.
- To conclude, it is clear that integrated carbon-aluminium electrodes is useful to improve metal removal in water.

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Acknowledgement

This paper was fully sponsored by the Centre for Graduate Studies (CGS), Universiti Tun Hussein Onn Malaysia (UTHM). We also like to show our gratitude to the Research Centre of Soft Soil (RECESS), UTHM to provide us a research laboratory and thanks to Faculty of Mechanical and Manufacturing Engineering, UTHM for assistance with SEM-EDX for morphological study of the electrode surface.