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# Adsorptive Treatment of Landfill Leachate using Activated Carbon Modified with Three Different Methods

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## Abstract

Activated Carbon (AC) is an adsorbent having high surface area which makes the process of removing heavy metals from wastewater (such as landfill leachate) very effective. This study explored the utilization of three methods of modification of AC produced from coconut shell by treating it with nitric acid ($HNO_3$), potassium permanganate ($KMnO_4$) and heating at 600ºC to improve the adsorption capacity. The AC can remove multi-pollutants in the filtration process which was used to treat landfill leachate. The water quality parameters such as pH, TSS, Ammonia-Nitrogen and a few heavy metals were considered in the present study. Results showed that the removal of these parameters was proportional with the increase of contact time and the bed depth of AC. The isotherm analysis of the adsorption of modified AC showed the best Removal Efficiency (RE) can be achieved when AC treated with $KMnO_4$ for $NH_3$-N, zinc, TSS and sulphide. The morphology of the AC was studied through Scanning Electron Microscopy (SEM), Energy Dispersive X-ray spectroscopy (EDX) pattern analysis and Fourier Transform Infrared (FTIR) analysis. It was found that various types of oxygen functional groups were introduced onto the surface of coconut shell derived AC through oxidation using $HNO_3$. FTIR was used to characterize the surface oxygen functional groups. The surface functional groups such as N-H and C-H stretching played a significant role in heavy metals adsorption. Hence, it can be concluded that the hybrid technique by using electrolysis process with AC adsorption be an effective way to remove the suspended solids and heavy metals from landfill leachate and thus able to reduce environmental pollution.

Keywords: *Landfill leachate, electrolysis, activated carbon, adsorption isotherm*

## 1. Introduction

The production of landfill leachate is considered as one of the reasons that lead to the contamination of groundwater which is the main source of potable water in many countries. Many harmful and toxic pollutants, e.g. heavy metals, organic and inorganic compounds are found in surface water streams and groundwater. Various technologies are used to remove these contaminants from water sources, e.g. anaerobic treatment, electrocoagulation, filtration, Activated Carbon (AC) adsorption and wetland (Cossu *et al.*, 1998; Kadlec and Knight, 1996; Alkalay *et al.*, 1998; Welander, 1998; Marselli *et al.*, 2003; Canizares *et al.*, 2005; Wiszniowski *et al.*, 2006; Deng and Englehardt, 2007; Sun and Austin, 2007; Graber *et al.*, 2009; Justin and Zupancic, 2009; Kalmykova *et al.*, 2009; Langergraberet *et al.*, 2009; Faulwetter *et al.*, 2009; Li *et al.*, 2009; Renou *et al.,* 2008; Sonavane and Munavalli, 2009; Vilar *et al.*, 2010; Yuen and Hameed, 2009; Timur *et al.*, 2010).

Adsorption technology is one of the methods used for the treatments of water and wastewater (Hejazifar *et al.*, 2011). The AC is considered as one of the most effective adsorbents which is processed and prepared to have high porosity and very large

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surface area for adsorption. It is generally produced from carbonaceous materials or from biomass substances, e.g. coconut shell, nutshells, peat, wood bagasse, lignite and coal. A great potential form of waste recycling is to covert waste material (e.g. rice husk) to AC. It can improve the human living standards in various ways (Ahsan *et al*., 2013). The characterization of AC and its potential utilization were studied (Rahman *et al*., 2013; Shaheed *et al.*, 2015; Tadda *et al.*, 2016; Ahsan *et al.*, 2015; Khaleel *et al*., 2015). The AC adsorption process can be utilized as a separate unit in wastewater treatment plant. It may be placed after various physicochemical steps such as coagulation, filtration and dissolved air flotation.

The transformation of AC to more polar materials could be done using several methods of treatment that changes the amount of chemical functional groups on the surface of AC. There are various surface modification methods, e.g. chemical (Valdes *et al.*, 2002; Fang *et al*., 2005; Jiang *et al*., 2003; Figueiredo *et al.*, 1999), physical (Pereira *et al.*, 2010), microwave treatments (Liu *et al*., 2010; Menendez *et al*., 1999; Nabais *et al*., 2004) and impregnation (Ahn *et al*., 2009). Depending on the reagents added, the functional sites on the AC are either positively or negatively charged and these functional sites transforms AC to more hydrophilic or hydrophobic and acidic or basic character.

Essentially, hydrophobicity of the AC is associated with the presence of oxygen containing groups on the surface, e.g. carboxylic, phenolic and lactonic groups. Frequently, the most common reagents that used to enhance oxygen content on the surface of AC are nitric acid ($HNO_3$), sulphuric acid, hydrogen peroxide, ozone and potassium permanganate ($KMnO_4$) (El-Hendawy, 2003; Yin *et al*., 2007; Chiang *et al*., 2002).

Liu *et al*. (2007) reported that coconut-based AC showed excellent adsorption performance for Cr (VI), when it was modified by HNO3 and sodium hydroxide. The modification of AC increases the total number of surface oxygen acidic groups and decreases the specific surface area. Oxidisation of $HNO_3$ leads to the production of positively charged acid groups. The acidity can be reduced by treating it with sodium hydroxide which replaces the $H^+$ of the surface acid groups by $Na^+$. The capacity of AC to adsorb Cr (VI) was increased from 7.61 to 13.88 mg/g due to the presence of more oxygen surface acidic groups and suitable surface acidity after modification.

Wibowo *et al.* (2007) modified coal-based AC by chemical treatment with HNO3 and thermal treatment with nitrogen gas flow. The treatment with HNO3 caused the introduction of a significant number of oxygenated acidic surface groups onto the carbon surface, while the heat treatment increases the basicity of carbon. The pore characteristics are not significantly changed after these modifications. The dispersive interactions are the most important factor in this adsorption process.

Heat treatment in gaseous or liquid phase normally increases the concentration of oxygen surface groups (Shen *et al*., 2008). While the heat treatment under inert condition leads to increase in hydrophobicity and pH of the carbon surface (Shafeeyan *et al*., 2010). Usually, hydrogen is the inert gas of choice over nitrogen and argon because it is more effective in removing the oxygen functionalities and also stabilizes the carbon surface. Thermal treatment in $H_2$ at 900°C produced highly stable and basic carbons (Lin and Teng, 2003).

Modification of AC with $KMnO_4$ was investigated in order to improve the adsorption capacity of AC. $KMnO_4$ contributes to the formation of AC with higher specific surface area (Kim *et al.*, 2014). Zhang and Yang (2011) studied adsorption of chromium (VI) by modified AC (MAC) with $KMnO_4$ and they found that the efficiency of AC to adsorb chromium (VI) is about 50%.

The aim of this study was to treat landfill leachate using MAC which was prepared from coconut shell by using three types of modification methods: acid treatment, heat treatment and treatment using $KMnO_4$. The best one was determined by comparing the equilibrium adsorption isotherms for each type of AC. This study also investigated the maximum Removal Efficiency (RE) for organic pollutants and heavy metals from landfill leachate.

## 2. Materials and Methods

The landfill leachate samples used in this study were collected from Jeram Sanitary Landfill (JSL) which is located at Kuala Selangor 20 km North/West of Kuala Lampur, Selangor, Malaysia, the methodology of the study divided into four steps:

### 2.1 Raw Leachate Analysis and Primary Electrolysis Treatment

The samples of leachate were collected in plastic containers and taken immediately to the public health laboratory in Faculty of Engineering, University Putra Malaysia and stored in cold room at 4°C. Laboratory analysis of leachate sample characteristics was carried out based on Standard Methods for the Examination of Water and Wastewater. The analysis included different parameters, e.g. pH, TSS, ammonia–nitrogen and heavy metals (zinc, manganese, sulphide and copper). Before the leachate sample was treated using filter of AC, it was treated in a batch type electrolytic cell made of glass container with dimensions of 300 mm length, 200 mm width and 250 mm height with two pairs of electrodes keeping at a distance of 3 cm between each other. Aluminium is used as cathode and iron as anode. The

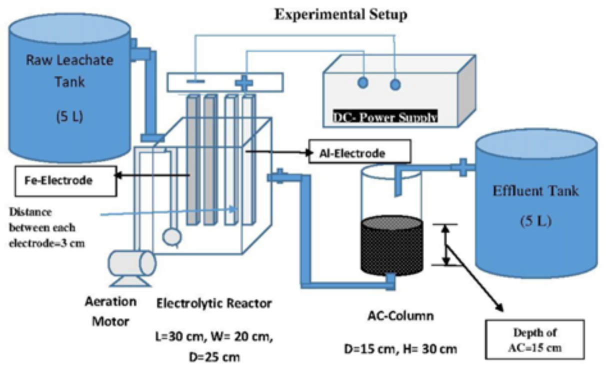


Fig. 1. Schematic of Experimental Setup

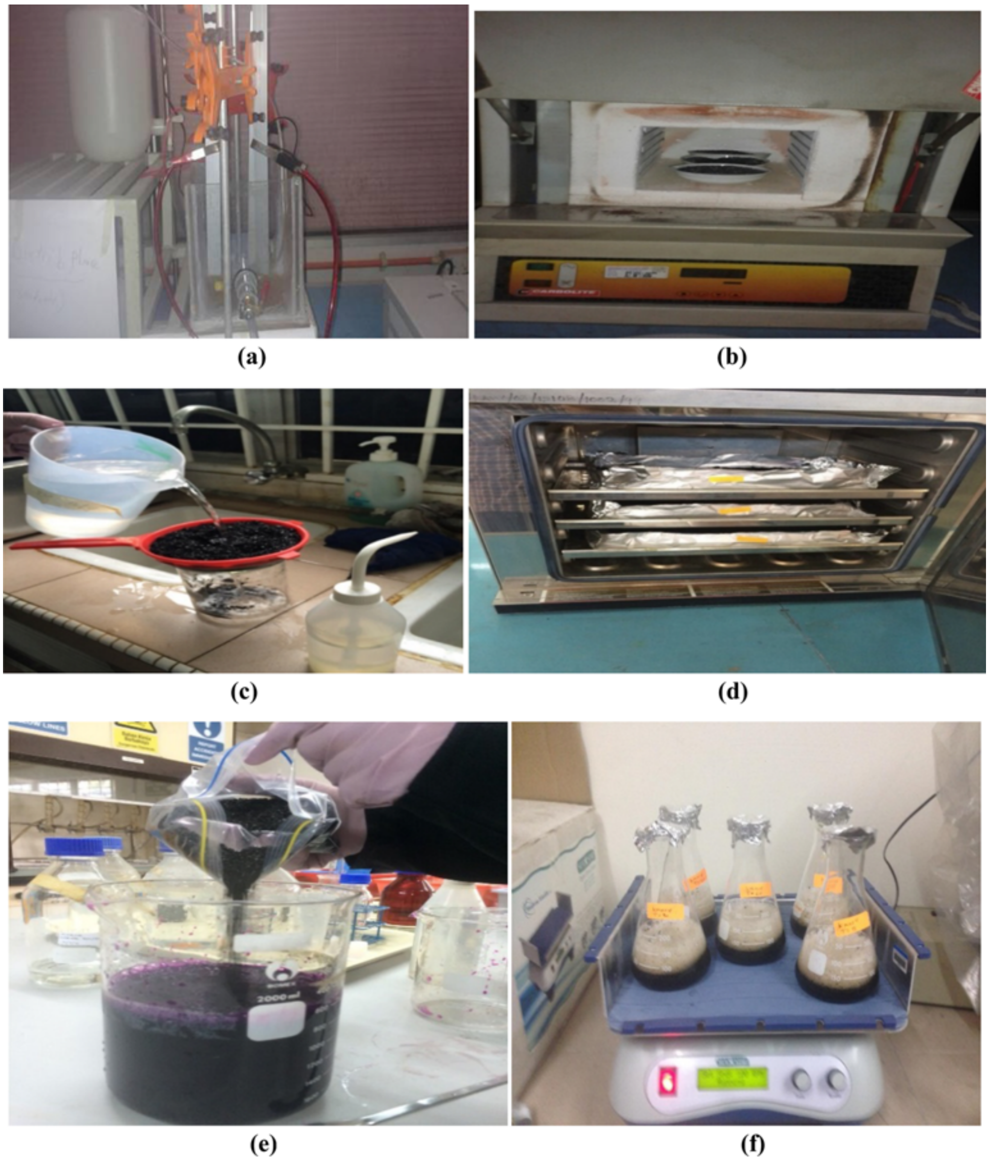

(a) (b) (c) (d) (e) (f)

Fig. 2. Electrolytic Reactor and AC Preparation Arrangement: (a) Electrolytic Reactor, (b) Heating AC in Chamber Furnace with 600ºC, (c) Washing with Distilled Water, (d) Drying AC in Oven with 110ºC, (e) Mixing $KMnO_4$ with AC Sample, (f) Adsorption Test

electrodes were arranged parallel to each other and connected to the negative and positive outlets of the digital DC power supply (Model RXN-3020 D) that regulates the electricity. The pairs of electrodes were submerged in the glass container which contains sample of leachate. The working condition was set with 40 min electrolysis retention time and 24 V power supply. An aerator with an average flow of 1200 $cm^3$/min was placed at the bottom of the batch electrolysis cell in order to achieve proper mixing.

### 2.2 Modification Methods of AC

The Granular Activated Carbon (GAC) with coconut shell as raw material was used in this study. It was directly purchased from local traders, Laqua system Sdn. Bhd., Lot No. 9 & 11, Jalan Industri PBP, Selangor, Malaysia. The GAC has 1150 $m^2$/g surface area, 8 × 30 mesh particle size distribution, 5% moisture content and 0.45-0.50 g/$cm^3$ apparent density. A modification treatment was done on GAC in order to increase the adsorption capacity using three different methods by treating it with nitric acid ($HNO_3$), potassium permanganate ($KMnO_4$) and heating at 600ºC. These three methods of AC modification were selected in this study due to their high performance of adsorption as reported by many researchers (Liu *et al.*, 2007; Shen *et al.*, 2008; Kim *et al.*, 2014).

#### 2.2.1 GAC Modified with $HNO_3$

Acid treatment increases the acidity, the amount of oxygen surface groups and the hydrophilic characteristics of the surface of AC. Common oxidizing acids such as $HNO_3$ and sulphuric acid are used for this purpose. The application of acid treatment is to remove the heavy metals from the aqueous solution. Known quantity of GAC is sieved using 2.35 mm sieve. Oxidation of AC is carried out by boiling it with $HNO_3$ of 65% concentration.

The weight ratio of carbon to acid is 1: 5% and boiled it for 2 hours in drying oven at 60°C in a round bottom flask. After oxidation, the sample is filtered, washed thoroughly with hot distilled water until it is free from nitrate ions. The sample is then dried in oven at 110°C for 2 hours and placed it in a round bottom flask covered with aluminium papers from top and kept in a dry place until it is used.

#### 2.2.2 GAC Modified with $KMnO_4$

Modification process of GAC with $KMnO_4$ is carried out by immersing the GAC (2.0 g) in $KMnO_4$ solution (30 mg/l) for 6 hours. The sample is then rinsed with distilled water until no colour is obtained and then dried under constant temperature of 100°C in drying oven for 16 hours. After cooling, the sample was kept in a glass beaker and covered it with aluminium paper to prevent any pollution and kept in room temperature until it is used.

#### 2.2.3 GAC Modified with Heating

Heat treatment in gases and liquid phase normally increases the concentration of oxygen surface groups, so the character of porous structure changes as the heating temperature increases. Known amount of GAC is sieved using 2.35 mm sieve. The GAC is heated in a thermal furnace at a temperature of 600°C for 2 hours. After heating, GAC is cooled and washed with distilled water several times to remove any impurities that might be present. The GAC is filtered and then it is placed in a drying oven at 110°C for 24 hours and stored in a dry place until it is used.

An AC filter (column) made of fibre glass with 95 mm diameter and 250 mm height is used to carry out the treatment of leachate samples after electrolysis process. The treated leachate was allowed to pass through the filter of different AC bed depths of 5, 10 and 15 cm. The above method allows AC to adsorb more pollutants such as suspended solids and heavy metals from the treated leachate. After adsorption process the treated leachate was collected into a glass beaker and then analysed according to the standard methods for the examination of water and wastewater.

### 2.3 Adsorption Process using MAC

#### 2.3.1 RE and Adsorption Capacity of Parameters

The RE for different water quality parameters was investigated by using a known amount of MAC as adsorbent in leachate sample. The percentage of RE of each parameter at different time intervals was calculated using the following equation.

$$RE = \frac{(C_0 - C_e)}{C_0} \times 100 \tag{1}$$

Where, $C_0$ is the initial concentration (mg/l) and $C_e$ is the concentration at time $t$ (mg/l).

The adsorption capacity (***$q_e$***) of AC can be determined as

$$q_e = \frac{(C_0 - V_e)V}{m} \tag{2}$$

Where, V is the volume of sample (l) and m is the mass of adsorbent (g).

#### 2.3.2 Equilibrium Adsorption Isotherm

An adsorption isotherm which represents the relationship between $q_e$ and $C_e$ was plotted to determine the shape of the adsorption equilibrium curve. Adsorption isotherm is fundamentally essential to explain how solutes interact with adsorbents and is critical in optimizing the use of adsorbents (Hameed *et al*., 2009). The Langmuir (Langmuir, 1916) and the Freundlich (Freundlich, 1906) isotherms were employed in this study for fitting the experimental data at different time intervals of AC. The linearized forms of the two isotherms are as follows.

$$\frac{1}{q_e} = \frac{1}{K_L q_m C_e} + \frac{1}{q_m} \tag{3}$$

$$\ln q_e = \ln K_F + \frac{1}{n} \ln Ce$$

The Langmuir constants, $q_m$ (mg/g) and $K_L$ (l/mg) are related to adsorption capacity and energy of adsorption, respectively. While, $K_F$ and $n$ are the Freundlich constants.

### 2.4 Characterizations of AC

#### 2.4.1 Porosity Characterisation

To characterise the porosity, $N_2$ adsorption-desorption isotherms at -195.6°C need to be determined using an automatic adsorption instrument. Evaluation of surface area for sample was done used BET method. (McEnaney, 1987).

#### 2.4.2 SEM and EDX Analysis

The morphology of the AC was identified by Scanning Electron Microscopy (SEM) (model S-3400N, Hitachi, Japan) which was coupled with Energy Dispersive X-ray spectroscopy (EDX) to carry out the elemental analysis of the AC.

#### 2.4.3 FTIR Analysis of AC

Fourier Transform Infrared (FTIR) spectroscopy is the most commonly used spectroscopic technique for the analysis of chemical structure of AC that can provide information about surface functional groups (Chiang *et al*., 2002). FTIR spectra of three samples of AC treated with three different methods were recorded before and after adsorption for contact time of 2 hours within 400 – 4000 $cm^{-1}$.

Table 1. Characteristics of Electrolyzed Leachate using Al-Fe Electrodes

| Parameters | Unit | Dilution Factor | Initial Concentration (mg/l) |
|---|---|---|---|
| pH | | 10 | 8.44 |
| TSS | mg/l | 0 | 491.67 |
| Ammonia-Nitrogen | mg/l | 10 | 40.00 |
| Zinc | mg/l | 10 | 3.50 |
| Copper | mg/l | 10 | 1.5 |
| Sulphide | mg/l | 10 | 2.30 |
| Manganese | mg/l | 10 | 2.66 |

## 3. Results and Discussion

### 3.1 Primary Treatment of Leachate by Electrolysis

The raw landfill leachate sample was treated first with electrolysis process before the filtration in order to decrease the high concentrations of suspended solids, organic and inorganic matters. Concentrations of various parameters of leachate samples after treated by electrolysis process using a pair of Al-Fe electrodes are shown in Table 1.

### 3.2 Effect of Modification Methods on Porosity Characterizations

Table 2 shows the porosity characterization of MACs after modification with $HNO_3$, $KMnO_4$ and heating treatment. AC-$KMnO_4$ has the highest surface area (1401.07 $m^2/g$) and pore volume (0.509 $cm^3/g$), however, the differences between AC-$KMnO_4$ and AC-Heating are little. While AC-$HNO_3$ has the lowest surface area (1348.21 $m^2/g$) and pore volume (0.486 $cm^3/g$). These results are agreement with other studies conducted by Daifullah *et al.* (2007) and Kim *et al.* (2014). They concluded that the surface area of AC increasing with the increase of the amount of $KMnO_4$.

Table 2. Porosity Characterizations of MAC

| AC type | Surface area ($m^2/g$) | Pore volume ($cm^3/g$) | Micropore surface area ($m^2/g$) | Micropore volume ($cm^3/g$) |
|---|---|---|---|---|
| AC-$HNO_3$ | 1348.21 | 0.486 | 700.18 | 0.371 |
| AC-$KMnO_4$ | 1401.07 | 0.509 | 700.35 | 0.371 |
| AC-Heating | 1392.21 | 0.503 | 682.61 | 0.363 |

### 3.3 Adsorption Treatment of Leachate using MAC

The adsorption performance of three MACs with various contact times was investigated. The intervals of contact time were 0, 15, 30, 60, 90 and 120 min, which were selected according to previous studies (Babel and Kurniawan, 2004; Huang *et al*., 2009; Moreno-Pirajan, 2011; Zhang and Yang, 2011). The final concentrations of various parameters after AC adsorption are shown in Table 3. The parameters considered are pH, TSS, ammonia–nitrogen, zinc, manganese, copper and sulphide. The pH values obtained at different time periods for 3 types of AC vary from 8.20 to 8.55 only and it indicates that the modifications of AC did not affect pH value. The final concentrations of TSS, $NH_3$-N, Zn, Mn, Cu and $S^{2-}$ are decreasing with increasing of contact time for the three types of MAC.

### 3.4 Effect of Contact time on RE

The RE of three types of MAC at different intervals of contact time is shown in Table 4. The RE of TSS, $NH_3$-N, Zn, Mn, Cu and $S^{2-}$ are increased with increasing of contact time for three types of MAC. The TSS indicates the higher removal percentage of 91% using $KMnO_4$ at contact time of 120 min. The maximum removals of $NH_3$-N using AC-$KMnO_4$ are 90, 94, 95, 96, 99 and 99%, respectively at contact time intervals of 0, 30, 60, 90 and 120 min, respectively. Also, Zn, Cu and $S^{2-}$ represented the highest removal efficiencies when using AC-$KMnO_4$ as adsorbent. This can be explained due to the surface modification extent. Modification

Table 3. Final Concentrations of 3 types of MAC at Different Intervals of contact time

| Parameters | Dilution Factor | Type of MAC | Contact Time (min) | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | | 0 | 15 | 30 | 60 | 90 | 120 |
| pH | 0 | AC-$HNO_3$ | 8.51 | 8.41 | 8.36 | 8.35 | 8.34 | 8.33 |
| | | AC-$KMnO_4$ | 8.50 | 8.52 | 8.52 | 8.55 | 8.34 | 8.48 |
| | | AC-Heated (600°C) | 8.22 | 8.44 | 8.44 | 8.50 | 8.28 | 8.53 |
| TSS (mg/l) | 0 | AC-$HNO_3$ | 91 | 81 | 81 | 74 | 69 | 56 |
| | | AC-$KMnO_4$ | 119 | 110 | 103 | 101 | 69 | 44.42 |
| | | AC-Heated (600°C) | 83 | 79 | 77 | 74 | 67 | 67 |
| Ammonia–Nitrogen (mg/l) | 10 | AC-$HNO_3$ | 5.10 | 5.10 | 4.60 | 4.50 | 4.30 | 4.10 |
| | | AC-$KMnO_4$ | 4 | 2.4 | 1.9 | 1.5 | 0.47 | 0.42 |
| | | AC-Heated (600°C) | 5.2 | 4.5 | 4.1 | 3.8 | 3.5 | 3.5 |
| Zinc (mg/l) | 10 | AC-$HNO_3$ | 2.50 | 2.28 | 1.70 | 1.122 | 1.00 | 0.60 |
| | | AC-$KMnO_4$ | 1.7 | 1.5 | 1.4 | 1.2 | 0.88 | 0.49 |
| | | AC-Heated (600°C) | 3 | 2.8 | 2.6 | 2.4 | 2.1 | 2.0 |
| Copper (mg/l) | 10 | AC-$HNO_3$ | 1.10 | 1.00 | 0.90 | 0.80 | 0.70 | 0.55 |
| | | AC-$KMnO_4$ | 1 | 0.9 | 0.7 | 0.5 | 0 | 0 |
| | | AC-Heated (600°C) | 1.2 | 1.1 | 1 | 0.8 | 0.8 | 0.8 |
| Manganese (mg/l) | 10 | AC-$HNO_3$ | 1.44 | 1.82 | 1.75 | 1.72 | 1.61 | 1.47 |
| | | AC-$KMnO_4$ | 1.75 | 1.64 | 1.60 | 1.60 | 1.57 | 1.48 |
| | | AC-Heated (600°C) | 1.61 | 1.60 | 1.48 | 1.47 | 1.10 | 1.14 |
| Sulphide (mg/l) | 10 | AC-$HNO_3$ | 2.00 | 1.90 | 1.50 | 1.20 | 1.10 | 1.04 |
| | | AC-$KMnO_4$ | 1.9 | 1.7 | 1.6 | 1.5 | 1.3 | 1.0 |
| | | AC-Heated (600°C) | 1.7 | 1.7 | 1.5 | 1.5 | 1.43 | 1.36 |

Table 4. RE (%) of 3 Types of MAC at Different Intervals of Contact Time

| Parameters | Type of MAC | Contact Time (min) | | | | | |
|---|---|---|---|---|---|---|---|
| | | 0 | 15 | 30 | 60 | 90 | 120 |
| TSS (mg/l) | AC-HNO$_3$ | 81 | 84 | 84 | 85 | 86 | 89 |
| | AC-KMnO$_4$ | 76 | 78 | 79 | 79 | 86 | 91 |
| | AC-Heated (600°C) | 83 | 84 | 84 | 85 | 86 | 86 |
| Ammonia –Nitrogen (mg/l) | AC-HNO$_3$ | 87 | 87 | 89 | 89 | 89 | 90 |
| | AC-KMnO$_4$ | 90 | 94 | 95 | 96 | 99 | 99 |
| | AC-Heated (600°C) | 87 | 89 | 90 | 91 | 91 | 91 |
| Zinc (mg/l) | AC-HNO$_3$ | 29 | 35 | 51 | 65 | 71 | 83 |
| | AC-KMnO$_4$ | 51 | 57 | 60 | 66 | 75 | 86 |
| | AC-Heated (600°C) | 14 | 20 | 26 | 31 | 40 | 43 |
| Copper (mg/l) | AC-HNO$_3$ | 27 | 33 | 40 | 47 | 49 | 52 |
| | AC-KMnO$_4$ | 33 | 40 | 53 | 67 | 100 | 100 |
| | AC-Heated (600°C) | 20 | 27 | 33 | 47 | 47 | 47 |
| Manganese (mg/l) | AC-HNO$_3$ | 46 | 32 | 34 | 35 | 39 | 45 |
| | AC-KMnO$_4$ | 35 | 38 | 40 | 40 | 41 | 44 |
| | AC-Heated (600°C) | 39 | 40 | 44 | 45 | 55 | 57 |
| Sulphide (mg/l) | AC-HNO$_3$ | 13 | 17 | 35 | 48 | 52 | 55 |
| | AC-KMnO$_4$ | 17 | 26 | 30 | 35 | 43 | 57 |
| | AC-Heated (600°C) | 26 | 26 | 35 | 35 | 38 | 41 |

Table 5. Adsorption Capacity (qe) of 3 types of MAC at Different Intervals of Contact Time

| Parameters | Type of MAC | Contact Time (min) | | | | | |
|---|---|---|---|---|---|---|---|
| | | 0 | 15 | 30 | 60 | 90 | 120 |
| TSS (mg/l) | AC-HNO$_3$ | 2.0034 | 2.0534 | 2.0534 | 2.040 | 2.1134 | 2.1784 |
| | AC-KMnO$_4$ | 1.8634 | 1.9084 | 1.9434 | 1.9534 | 2.1234 | 2.2734 |
| | AC-Heated (600°C) | 2.0434 | 2.0634 | 2.0734 | 2.040 | 2.1234 | 2.1234 |
| Ammonia –Nitrogen (mg/l) | AC-HNO$_3$ | 0.1745 | 0.1745 | 0.177 | 0.1775 | 0.1785 | 0.1795 |
| | AC-KMnO$_4$ | 0.18 | 0.188 | 0.1905 | 0.1925 | 0.1976 | 0.1979 |
| | AC-Heated (600°C) | 0.1740 | 0.1775 | 0.1795 | 0.1810 | 0.1825 | 0.1825 |
| Zinc (mg/l) | AC-HNO$_3$ | 0.005 | 0.0026 | 0.009 | 0.0049 | 0.0125 | 0.0145 |
| | AC-KMnO$_4$ | 0.009 | 0.01 | 0.0105 | 0.0115 | 0.0135 | 0.0165 |
| | AC-Heated (600°C) | 0.0025 | 0.0035 | 0.0045 | 0.0055 | 0.007 | 0.0075 |
| Copper (mg/l) | AC-HNO$_3$ | 0.002 | 0.0 025 | 0.003 | 0.0035 | 0.004 | 0.0045 |
| | AC-KMnO$_4$ | 0.0025 | 0.003 | 0.004 | 0.005 | 0.0075 | 0.0075 |
| | AC-Heated (600°C) | 0.0015 | 0.0020 | 0.0025 | 0.0035 | 0.0035 | 0.0035 |
| Manganese (mg/l) | AC-HNO$_3$ | 0.0061 | 0.0042 | 0.0046 | 0.0047 | 0.0053 | 0.0060 |
| | AC-KMnO$_4$ | 0.0046 | 0.0051 | 0.0053 | 0.0053 | 0.0054 | 0.0059 |
| | AC-Heated (600°C) | 0.0053 | 0.0053 | 0.0060 | 0.0061 | 0.0070 | 0.0076 |
| Sulphide (mg/l) | AC-HNO$_3$ | 0.0015 | 0.002 | 0.0013 | 0.0055 | 0.006 | 0.0021 |
| | AC-KMnO$_4$ | 0.002 | 0.003 | 0.0035 | 0.004 | 0.006 | 0.0065 |
| | AC-Heated (600°C) | 0.003 | 0.003 | 0.004 | 0.004 | 0.0014 | 0.0015 |

of AC with KMnO$_4$ increased the negative surface charge of adsorbent which is a favourable condition for removal of positive charge parameters like NH$_3$-N, Zn and Cu (Babel and Kurniawan, 2004). For Mn, the maximum removal percentage was 57% at 120 min using AC-Heating. This might be due to the high surface area and pore volume of this type of AC.

### 3.5 Effect of Contact Time on Adsorption Capacity ($q_e$)

The adsorption capacity (qe) of three types of MAC at different intervals of contact time is given in Table 5. The maximum adsorption capacities for TSS, NH$_3$-N, Zn, Cu and S$^{2-}$ were obtained as 2.2734, 0.1979, 0.0165, 0.0075 and 0.065 mg/g, respectively at 120 min of contact time by using AC-KMnO$_4$ as adsorbent. It has higher surface area and pore volume than the other two types of MAC.

## 3.6 Equilibrium Adsorption Isotherms

### 3.6.1 Adsorption Isotherm for AC-HNO$_3$

Figure 3 shows the experimental adsorption capacity ($q_e$

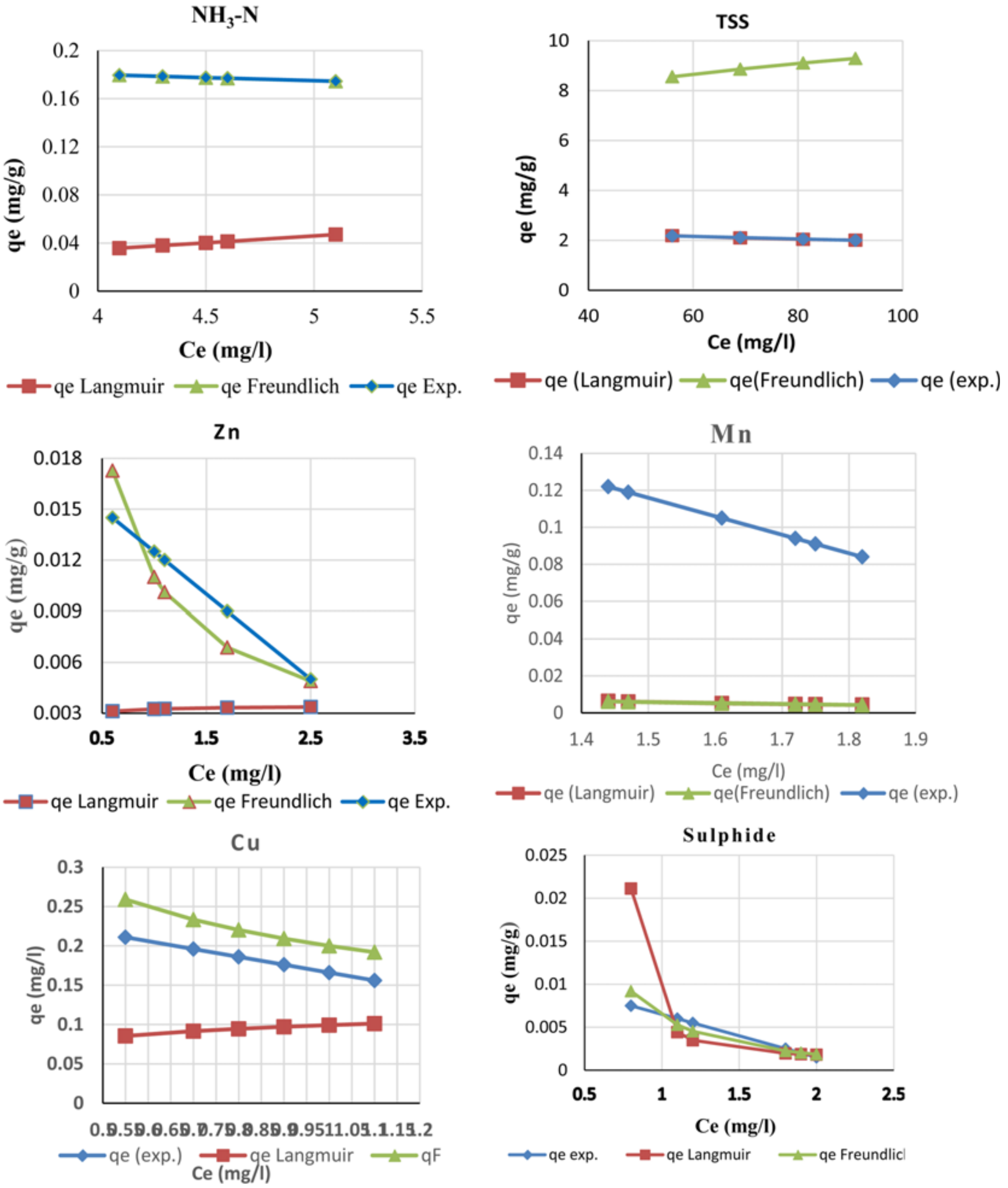


Fig. 3. Adsorption Isotherms of Different Water Quality Parameters using AC-HNO$_3$

exp.), q$_e$ Langmuir and q$_e$ Freundlich for all the parameters. Table 6 summarizes the Langmuir and Freundlich constants of three adsorbents. The adsorption data of AC-HNO$_3$ were well fitted to the Langmuir equation with the high value of correlation coefficients as 0.999, 0.999, 0.962, 0.991 and 1 for the TSS, NH$_3$-N, Zn, Mn and cu, respectively, where S$^{2-}$ has lower correlation coefficient (0.892). The adsorption data of Freundlich isotherm are characterized by the heterogeneity factor 1/n, the magnitude of n gives an indication on the favorability of adsorption. Generally, if n is in the range of 2-10 represents good, 1-2 moderately good, and less than 1 poor adsorption characteristics (Aksu and Kutsal, 1991). The Freundlich isotherm constants of AC-HNO$_3$ are shown in Table 6. The values of n for Mn, Cu and S$^{2-}$ were less than 1, which indicate the poor adsorption characteristics. The Langmuir isotherm indicates the higher correlation coefficients for all parameters than the Freundlich isotherm. Consequently, the Langmuir isotherms showed better fit to the adsorption data than the Freundlich isotherm.

### 3.6.2 Adsorption Isotherm for AC- KMnO$_4$

Figure 4 shows the adsorption isotherms of AC-KMnO$_4$, where the Langmuir isotherm showed a better fit than the Freundlich isotherm for TSS, NH$_3$-N, Zn, Mn, Cu and S$^{2-}$ with the high correlation coefficients of these parameters. These were obtained in the range of 0.915-0.999, which

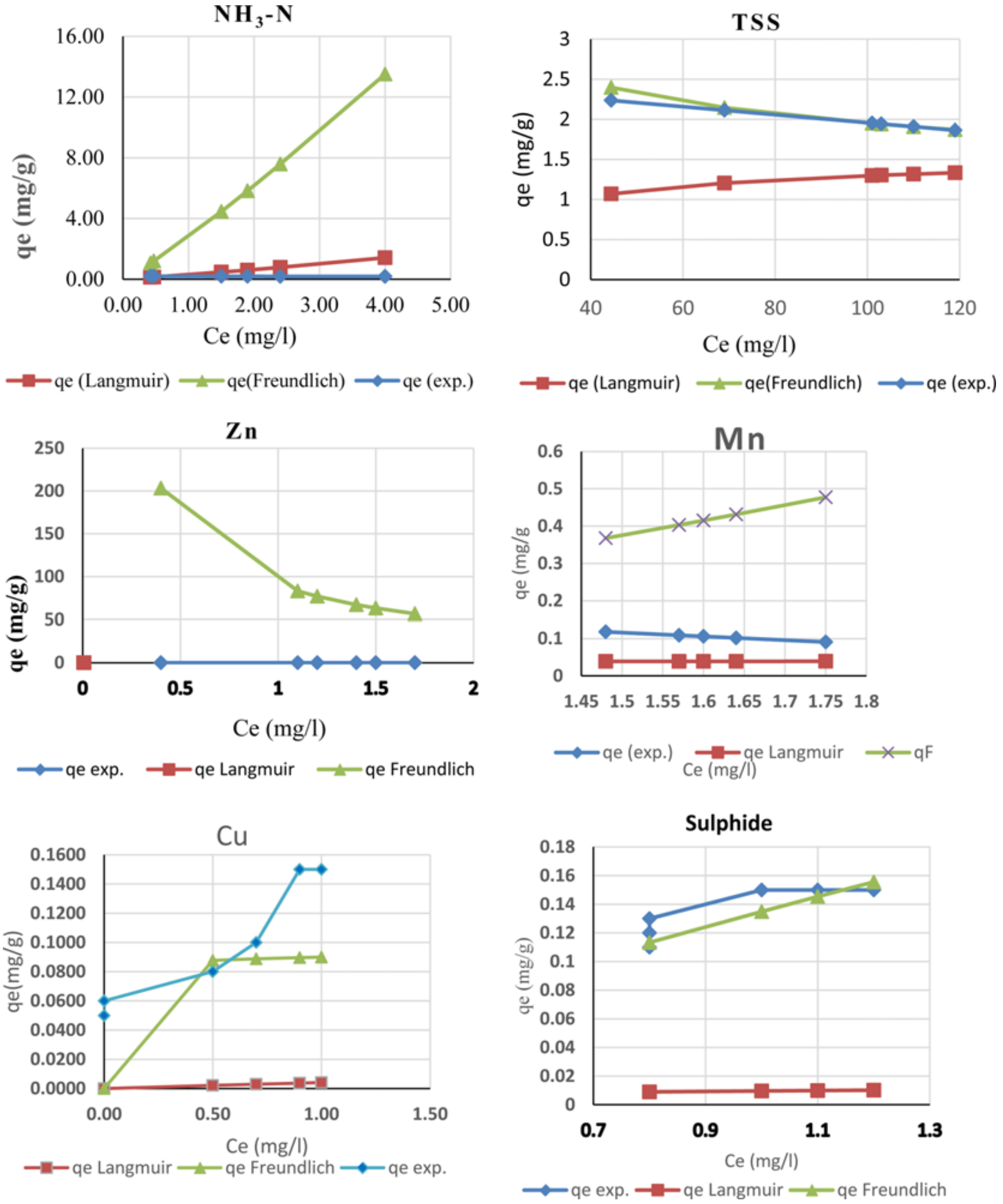


Fig. 4. Adsorption Isotherms of Different Water Quality Parameters using AC- $KMnO_4$

signify the good correlation between them and the adsorption is monolayer.

### 3.6.3 Adsorption Isotherm for AC- Heating

Figure 5 illustrates the adsorption isotherms of AC-Heating and the constants of Langmuir and Freundlich isotherms are indicated in Table 6. The obtained n values (from the Freundlich isotherms) of TSS, $NH_3$-N and Cu are more than 1, which indicate the good adsorption characteristics. For other parameters, e.g. Zn, Mn and $S^{2-}$, the correlation coefficients of Freundlich isotherms are more than the Langmuir isotherms. As a result, Freundlich isotherm shows the better fitting to experimental data than the Langmuir isotherm for AC-Heating, which implies the heterogeneous nature of heating.

## 3.7 Effects of AC Bed Depths on RE

Figure 6 shows the RE of various parameters for the three different bed depths (5, 10 and 15 cm) of AC filter and observed that the 15 cm bed depth gives highest RE for most parameters.

## 3.8 Surface and Chemical Analysis of Adsorbent

The MACs were examined by SEM to analyze the surface of

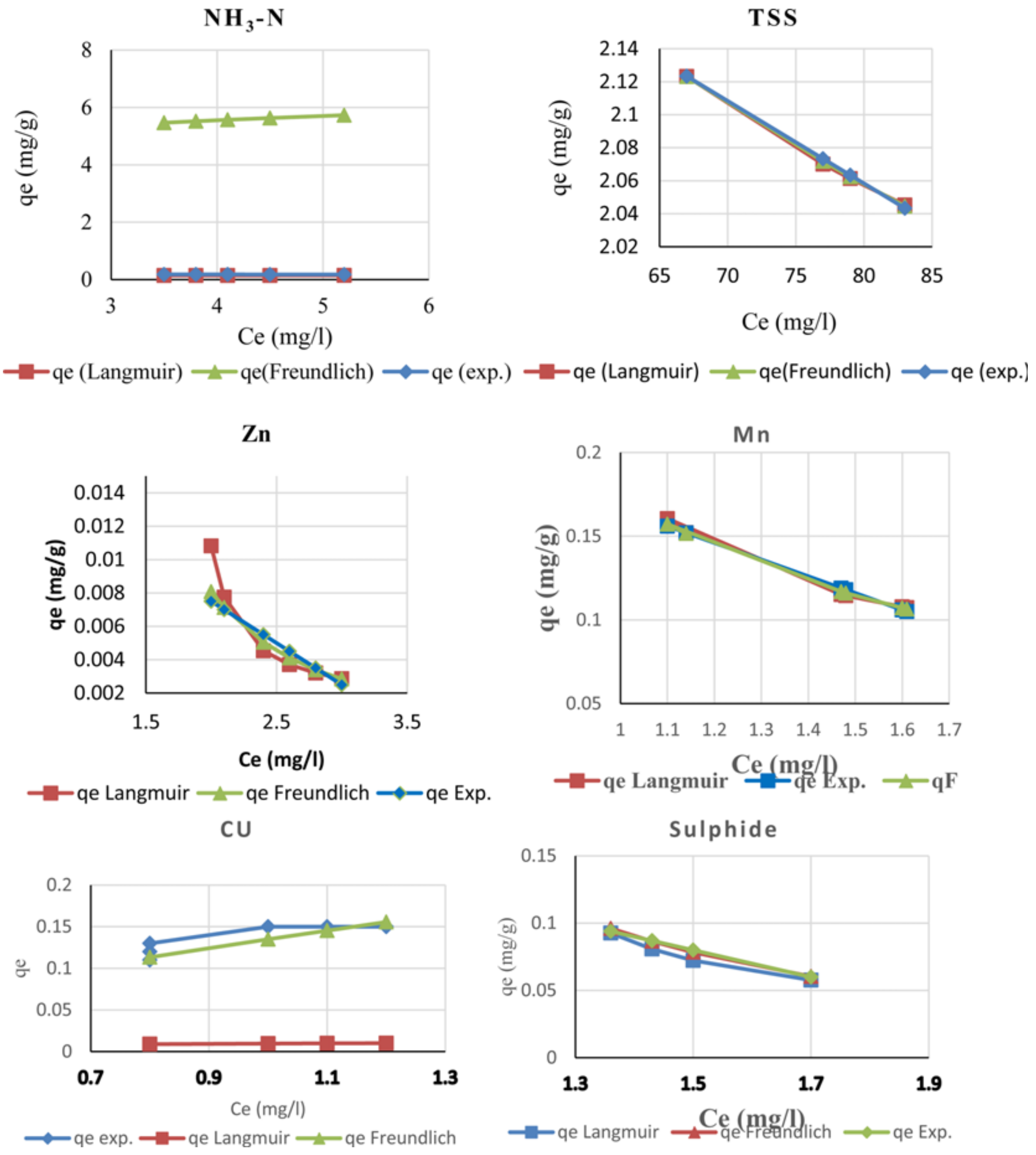


Fig. 5. Adsorption Isotherms of Different Water Quality Parameters using AC- Heating

the adsorbent. The SEM micrographs before and after adsorption for three types of MAC treated with HNO$_3$, KMnO$_4$ and heating are shown in Fig. 7.

In all three cases, well-developed porous surface was observed at higher magnification. The pores observed from SEM images are having diameter in micrometer (μm) range. From the figures, it can be observed that the AC treated with KMnO$_4$ has pores filled with more adsorbent compared to other types of MAC. Fig. 8 shows the EDX analysis of heavy metals such as Mn, Zn, Cu, and S$^{2-}$ which are adsorbed by MAC from the samples of leachate.

### 3.8 FTIR Results

The surface chemistry of the AC modified with three different methods before and after adsorption was explained using FTIR spectroscopy as shown in Figs. 9, 10 and 11, respectively. The spectra of AC were measured within 400- 4000 cm$^{-1}$.

The peak observed at 3713 and 3634 cm$^{-1}$ in Fig. 9 was due to free hydroxyl groups or surface bonded water (Tangjunak *et al.*, 2009). The bands observed at 2917 cm$^{-1}$ suggest the presence of amine groups. The peaks at 2916 and 2847 cm$^{-1}$ indicate the presence of C-H stretching vibrations of methyl group (Farinella *et al.*, 2007).

Figure 10 shows the FTIR analysis before and after adsorption of heavy metals by AC-KMnO$_4$. The peaks at 2916 and -2040 cm$^{-1}$ indicate the presence of C-H stretching vibrations of methyl group. Fig. 11 shows the FTIR analysis before and after adsorption of heavy metals by AC-Heating. The peaks at 1626 and 1383 cm$^{-1}$ can be due to N-H functional group after adsorption. From the FTIR analysis, it can be confirmed that the surface functional groups such as N-H and C-H stretching played a very important

Table 6. Adsorption Parameters of Langmuir and Freundlich Isotherms for Different MAC Samples

| Parameters | TSS | $NH_3$-N | Zn | Mn | Cu | $S^{2-}$ |
|---|---|---|---|---|---|---|
| AC-$HNO_3$ | | | | | | |
| *Langmuir* | | | | | | |
| $K_L$(L/g) | 0.0951 | 0.0454 | 15.634 | 1.0175 | 1.526 | 1.177 |
| $q_o$ (mg/g) | 1.783 | 0.156 | 0.00345 | 0.00197 | 0.00082 | 0.0011 |
| $R^2$ | 0.999 | 0.999 | 0.962 | 0.991 | 1.00 | 0.892 |
| *Freundlich* | | | | | | |
| $K_F$ | 4.322 | 0.216 | 0.0111 | 0.0108 | 0.0024 | 0.0062 |
| 1/n | 0.169 | 0.131 | 0.884 | 1.565 | 1.974 | 1.745 |
| $R^2$ | 0.993 | 0.998 | 0.942 | 0.994 | 0.998 | 0.930 |
| AC-$KMnO_4$ | | | | | | |
| *Langmuir* | | | | | | |
| $K_L$(L/g) | 0.0491 | 1.86 | 3.804 | 20.332 | 0.080 | 31.262 |
| $q_o$ (mg/g) | 1.563 | 0.16 | 0.165 | 0.0402 | 0.060 | 0.137 |
| $R^2$ | 0.998 | 0.999 | 0.968 | 0.991 | 0.915 | 0.977 |
| *Freundlich* | | | | | | |
| $K_F$ | 6.237 | 0.19 | 0.0111 | 0.2 | 0.09 | 0.176 |
| 1/n | 0.252 | 0.04 | 0.884 | 1.554 | 0.04 | 0.073 |
| $R^2$ | 0.979 | 0.886 | 0.942 | 0.994 | 0.915 | 0.807 |
| AC-Heating | | | | | | |
| *Langmuir* | | | | | | |
| $K_L$(L/g) | 0.0905 | 0.109 | 0.561 | 1.4903 | 2.448 | 0.978 |
| $q_o$ (mg/g) | 1.773 | 3.185 | 0.0232 | 0.0626 | 0.0743 | 0.0234 |
| $R^2$ | 0.999 | 0.999 | 0.904 | 0.991 | 0.535 | 0.987 |
| *Freundlich* | | | | | | |
| $K_F$ | 4.456 | 4.709 | 0.0483 | 0.173 | 0.135 | 0.182 |
| 1/n | 0.176 | 0.119 | 2.579 | 1.024 | 0.782 | 2.079 |
| $R^2$ | 0.999 | 0.996 | 0.956 | 0.994 | 0.875 | 0.992 |

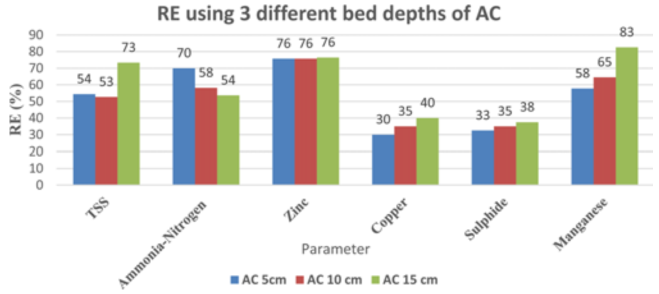


Fig. 6. RE of Various Parameters at Three Different AC Bed Depths

role in heavy metals adsorption.

## 4. Conclusions

Based on the experimental investigation the conclusions derived are as follows:

1. The modified AC (by three different methods) used in the study as adsorbent showed high microporosity, high percentage of pollutants removal and improve the adsorption capacity.
2. Modification of AC with $KMnO_4$ contributed to increase surface area and pore volume of resulted AC more than other modification methods like $HNO_3$ and heating. The high porosity characterizations make the AC effective. It is a cheap precursor to use for various wastewater treatments.
3. The highest removal efficiencies of TSS, $NH_3$-N, Zn, Cu and $S^{2-}$ were obtained as 91, 99 86, 100 and 57%, respectively at 120 min of contact time using AC-$KMnO_4$ due to the highest surface area and pore volume of AC.
4. The Langmuir isotherm showed better fit to the adsorption data than the Freundlich isotherm for AC-$KMnO_4$ with high regression coefficient ranged from 0.915 to 0.999 which means monolayer adsorption.
5. The surface functional groups such as N-H and C-H stretching played a significant role in heavy metals adsorption. The adsorption capacities of heavy metals in landfill leachate increased with increasing contact time of AC adsorption. The maximum adsorption capacities for TSS, $NH_3$–N, Zn, Cu and $S^{2-}$ were obtained as 2.2734, 0.1979, 0.0165, 0.0075 and 0.0065 mg/g, respectively using AC-$KMnO_4$ as adsorbent at 120 min of contact time.
6. It can be concluded that the hybrid technique by using electrolysis process with AC adsorption be an effective way to remove the suspended solids and heavy metals from landfill leachate and thus it can be applied to treat various types of wastewater.

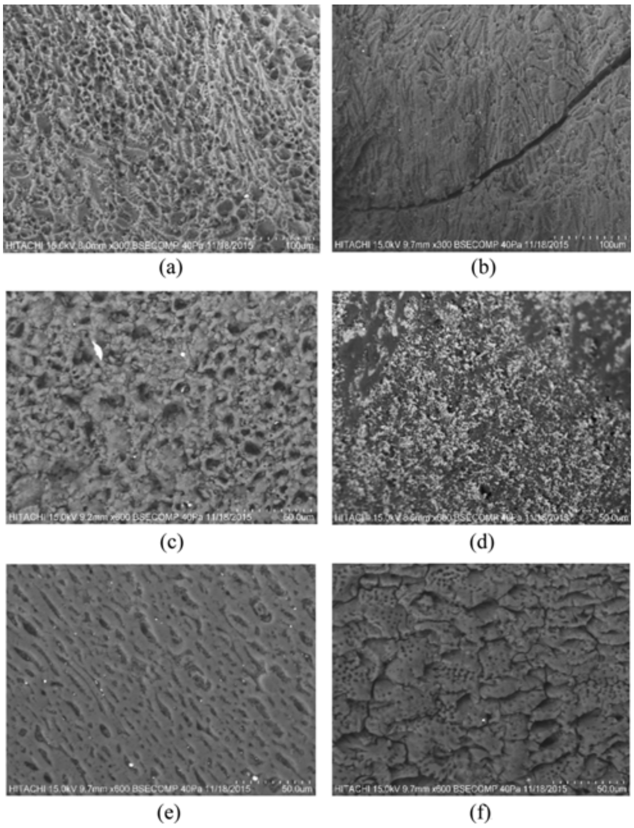
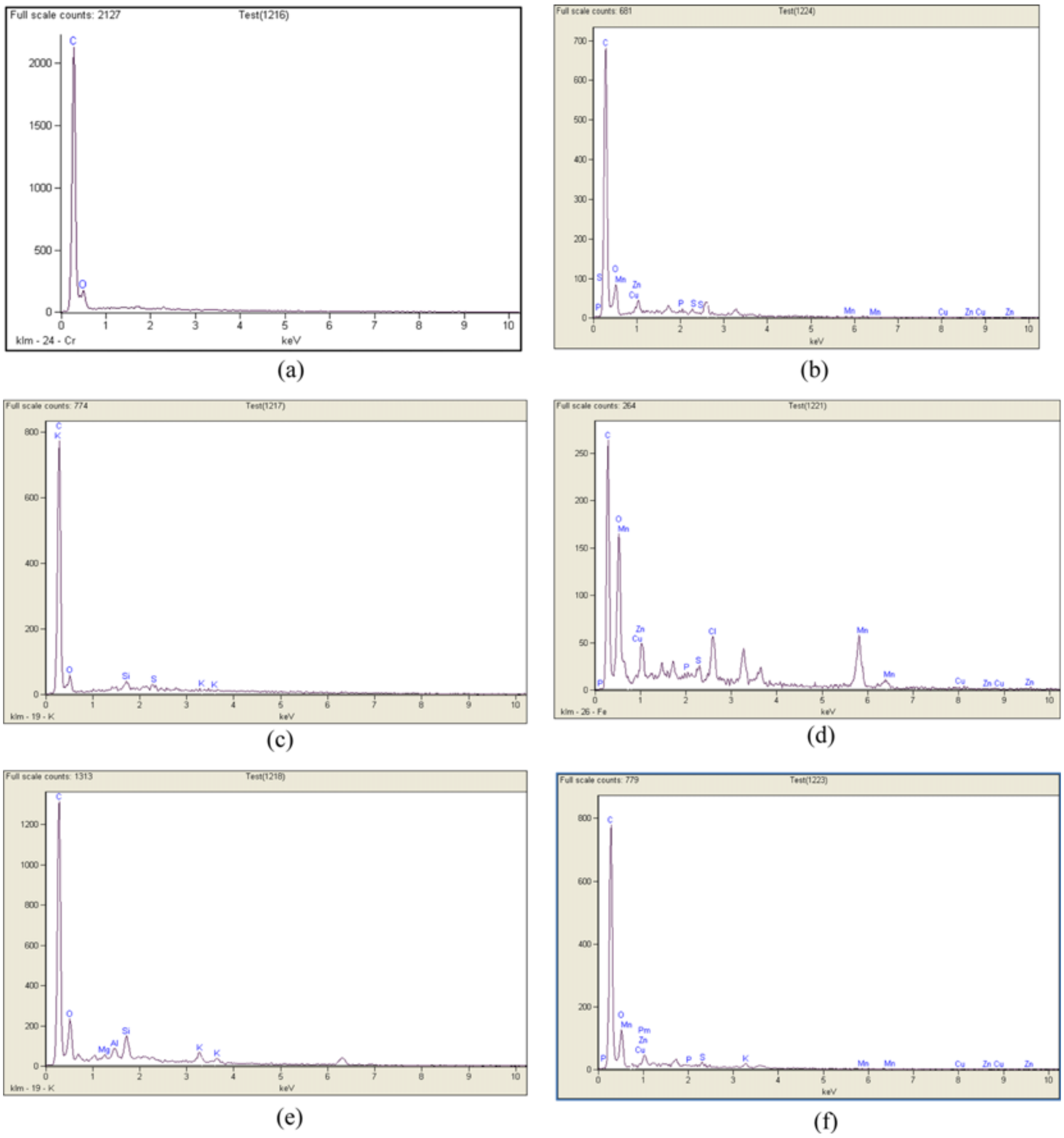


Fig. 7. SEM Images of Three MACs before and after Adsorption: (a) AC-$HNO_3$ (before adsorption), (b) AC-$HNO_3$ (after adsorption), (c) AC-$KMnO_4$ (before adsorption), (d) AC-$KMnO_4$ (after adsorption), (e) AC-Heating (before adsorption), (f) AC-Heating (after adsorption)



(a) (b) (c) (d) (e) (f)

Fig. 8. EDX Analysis of Three MACs before and after Adsorption: (a) AC-$HNO_3$ (before adsorption), (b) AC-$HNO_3$ (after adsorption), (c) AC-$KMnO_4$ (before adsorption), (d) AC-$KMnO_4$ (after adsorption), (e) AC-Heating (before adsorption), (f) AC-Heating (after adsorption)

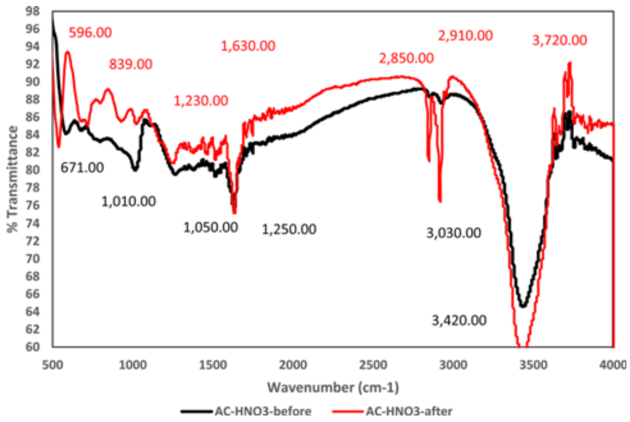


Fig. 9. Surface Functional Groups of AC-$HNO_3$ before and after Adsorption

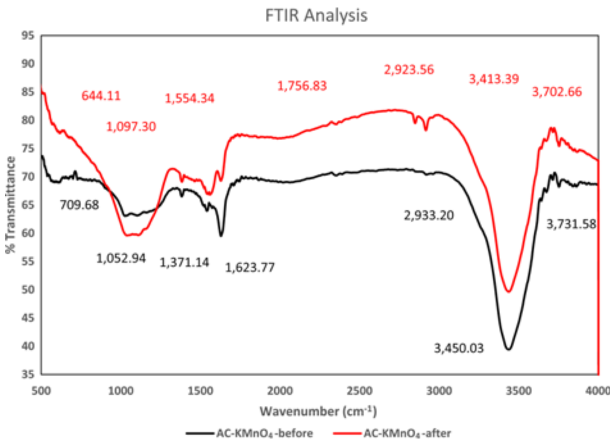


Fig. 10. Surface Functional Groups of AC-$KMnO_4$ before and after Adsorption

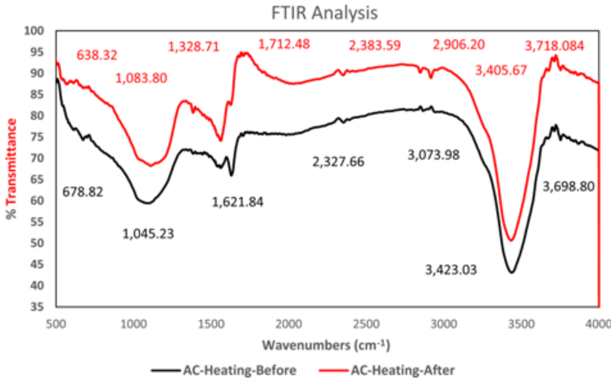


Fig. 11. Surface Functional Groups of AC-Heating before and after Adsorption

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