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TEA BAG DERIVED ACTIVATED CARBON FOR THE REMOVAL OF AS(III) AND PB(II)

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ABSTRACT

Purpose: This paper investigates the application of Activated Carbon derived from Waste Tea Bags (AC-WTB) in the removal of As(III) and Pb(II) ions from aqueous solutions.

Design/methodology/approach: Batch experiments were conducted to determine the effect of varying adsorption parameters on the removal of aqueous As(III) and Pb(II) ions; the data collected were compared to known adsorption isotherms.

Findings: The adsorption of As(III) ions was found to be maximum (80%$\pm$2.33) at pH 5 and that of Pb(II) ions was found to be optimum (58%$\pm$1.89). The adsorption capacity, Q_o, (mg g^{-1}) of As(III) is higher than that of Pb(II) ions and were found to be 7.348 and 3.791 respectively. The affinity of the two metals for the adsorbent surface in terms of b is higher for As(III). The Freundlich level of conformity for As(III) is to a high extent compared to the Langmuir adsorption isotherms as indicated by higher correlation coefficients values of 0.9917.

Original value of paper: A cheap and readily available adsorbent is proposed for tackling harmful metals in aqueous solutions, thus meaning a reduction in the potential of water borne diseases.

Keywords: Activated Carbons; (AC-WTB), Adsorption; Heavy metals; Langmuir and Freundlich isotherms: Lead: Arsenic.

INTRODUCTION

Increased industrialised use of metals and chemicals in process industries has resulted in the generation of large quantities of effluent that contains high levels of toxic heavy metals and their presence poses environmental-disposal problems. Our ecosystem has been contaminated by a high concentration of heavy metals released into the biosphere by human activity. Industrial activities, energy production, construction, urban waste treatment and vehicle exhausts are some of the sources causing large quantities of heavy metal contamination in the atmosphere, water and soil (Naeem et al., 2012).

Arsenic has been listed as one of the human carcinogens by the International Agency for Research on Cancer (IARC) since 1980. There have been many studies in the literature to show the direct relationship between arsenic exposure and cancer in different countries including the USA, Taiwan, Bangladesh and India (Kapaj et al., 2006). Bladder cancer (Steinmaus et al., 2003), skin cancer (Luster and Simeonova, 2004) and lung cancer (Chiu et al., 2004) are some of the cancer types associated with arsenic exposure as reported by many researchers.

In addition to carcinogenic effects, there are some non-carcinogenic effects of chronic arsenic exposure (Kapaj et al., 2006). According to (Tsai et al., 2003), long term accumulated arsenic in adolescence may cause neuro-behavioural effects and exposure to high amounts of arsenic may affect behaviour later in life. In addition, other heavy metals, such as lead, also have effects on health; these effects will be more severe with the synergistic behaviour of arsenic and lead (Tsai et al., 2003). There are many sources of arsenic exposure. One of the big sources for humans to inorganic arsenic exposure is groundwater. High concentrations of arsenic in groundwater have been recognised as a global concern (Kapaj et al., 2006).

Lead pollution in water systems has seriously influenced the quality of life, especially in developing countries. Lead accumulates in tissue and may cause serious health problems for humans and animals. This metal inhibits biosynthesis and affects the kidneys, brain cells and the permeability

of the liver membrane, thereby reducing some of their functions. It can be accumulated in the body and can promote abnormalities such as nausea, vomiting, diarrhoea, sweating and in some cases, convulsions, coma and death (Maranhao et al., 2005; Sharma et al., 2009).

Conventional techniques aimed at removing heavy metals usually include chemical precipitation, ion exchange, membrane process, crystallisation and electro-chemical treatment (Babel and Kurniawan, 2003). Several technologies for the removal of arsenic (Ahmed, 2000) and lead (Babarinde et al., 2006; Andal and Sakthi, 2010; Tuzen et al., 2007) from aqueous solutions have been investigated by many researchers. Adsorption of heavy metals on Activated Carbons (ACs) and modified ACs proves to be one of the most efficient techniques in this application. The use of low cost precursors that are available in large amounts, renewable and contain a high carbon and low inorganic content are recommended. These requirements are found in many agricultural wastes (ElHussien and Isa, 2015c; ElHussien and Isa, 2015b; Elhussien and Hamid, 2015; Elhussien and Isa, 2015a).

The aim of this research was to investigate the use of Activated Carbon derived from Waste Tea Bags (AC-WTB) in removing As(III) and Pb(II) ions from aqueous solutions. The study involved the examination of experimental conditions such as pH of the solution, contact time, adsorbent loading and initial metal ion concentrations on the removal of As(III) and Pb(II) from aqueous solutions. The Freundlich and Langmuir adsorption isotherms were used to investigate the adsorption process.

MATERIALS AND METHODS

Materials

Preparation of the Adsorbate Solutions

All chemicals used were of analytical grade (Merck, Germany). Two stock solutions of the metal ions, each containing 1000mg/L of $Pb(NO_3)_2$ and $NaAsO_2$, were prepared by dissolving the calculated amounts in double distilled water. The experimental solutions were prepared by diluting the metal ion stock solution in accurate proportions to different initial concentrations from 5–17.5mg/L and each of them was stored in 50ml reagent bottles, respectively. The supernatant was carefully decanted into clean polyethylene containers and analysed using a Flame Atomic Absorption Spectrometer (FAAS) Pin AAcle 500 Spectrometer, unit size 1/EA.

Preparation of the Adsorbent (AC-WTB)

The adsorbent used in this study was powdered, Activated Carbon obtained from disposal Waste Tea Bags (AC-WTB), collected from a coffee shop's waste in the Faculty of Education, Nile Valley University, Atbara City, River Nile State (Northern Sudan), labelled as Algazaltain Tea Bags. The tea bag samples were manufactured by the Cofftea Company, Gamhoria Street, Khartoum, Sudan. These waste tea bags were washed thoroughly with hot distilled water until the washing water was clean. After washing was completed the waste tea bags were dried and emptied. The carbonisation was done at high temperature (525°C) for 48hrs and pre-treated according to the method reported in the literature (Babarinde et al., 2006; Elhussien and Isa, 2015a; Porpino et al., 2011).

The sample was screened and washed with de-ionised water to remove dirt impurities, then dried in the oven at 110°C for three hours. The sample was washed again with de-ionised water until the pH was in the range of (6.8−7.3): the pH was determined using a pH meter (model ATPH-6). The sample was then dried in the oven at about 95°C to remove moisture. The dried sample was ground and sieved in the using sieves to collect the fraction of 90−125μm, which was preserved in a desiccator for use.

Methods

Batch Equilibrium Experiments

The adsorption experiments were conducted in an experimental apparatus in an appropriate condition. A rotary shaker at 150rpm using 100ml shaking flasks containing 50ml of the metal ion solutions at different concentrations and initial pH values was used. The corresponding amounts of the adsorbent were added to each flask and then the flasks were sealed to ensure there was no volume change during the experiments. The samples were shaken for predetermined time intervals and then withdrawn; the metal ion solutions were separated from the adsorbent by centrifugation and filtration respectively. The pH values for the separated metal ion solutions were again measured.

The data obtained in the batch modes were used to calculate the equilibrium metal ions adsorptive amounts (mg/g); the calculations were done using the following expressions (ElHussien and Isa, 2015c; Porpino et al., 2011):

$$q_e = \left(\frac{V}{W}\right)(C_o - C_e) \tag{1}$$

$$removal(\%) = \frac{(C_o - C_e)}{C_o} \tag{2}$$

Where q_e is the amount of the heavy metal ion adsorbed by the AC-WTB sample in mg/g, C_o and C_e are the initial and equilibrium concentrations of the metal ion respectively in mg/L, V is the volume of the solution treated (L) and w is the adsorbent mass of AC-WTB, in g. All experiments were duplicated and the mean values are reported. The maximum deviation was observed to be 2.6 and 1.7% for As(III) and Pb(II) ions respectively.

The effect of each parameter (pH, initial metal concentration, adsorbent mass and contact time) was studied in experiments by varying the parameter under consideration, while all the other parameters were maintained as constant. The negative control (with no adsorbent) was used to ensure that the adsorption was only due to the AC-WTB sample and not to the reaction vessels. The data were fitted using Langmuir and Freundlich isotherm models (Rose and Rajam, 2012; Shukla and Pai, 2006; Ahamad and Jawed, 2010; Acemioglu, 2004; Odebunmi and Okeola, 2001; Hobson, 1969). All batch experiments were carried out at a constant temperature (29±0.15°C).

An Fourier Transform Infrared Spectroscopy (FTIR) analysis was done using IPRrestige-21, FTIR-84005, Shimadzu Corporation (Kyoto, Japan). A 0.1g sample of AC-WTB was mixed with 1g of KBr, spectroscopy grade (Merck, Darmstadt, Germany), in a mortar. Part of the mixture was transferred to the FTIR analyser and the corresponding spectrogram was obtained showing wave lengths of the different functional groups in the sample; these were identified by comparing these values with those in the library (Calzaferri and Imhof, 1996).

RESULTS AND DISCUSSION

Sample Characterisations

The physico-chemical parameters of the AC-WTB sample are shown in Table 1. The FT-IR spectrum showed the absorption bands at these regions $2,880-3,460cm^{-1}$, $1,315-1,770cm^{-1}$, $1,100-1,260cm^{-1}$ and $475-760cm^{-1}$.

The sample showed a broad band with two maxima at $2,895cm^{-1}$, which is attributed to a C-H interaction with the surface of the carbon and at $3,440$ cm^{-1}, which is due to an O-H stretching mode. Furthermore, the indicated bands in the range of $3,320$–$3,670cm^{-1}$ have also been attributed to the hydrogen-bonded OH group of alcohols and phenols. However, the band at $1,480cm^{-1}$ may be attributed to the aromatic carbon-carbon stretching vibration. The two peaks at $1,110-1,155cm^{-1}$ showed the fingerprint of this carbon. The sharp absorption band at $1,120$ cm^{-1} is ascribed to either Si-O or C-O stretching in alcohol, ether or hydroxyl groups (Calzaferri and Imhof, 1996; Socrates, 1994). The band at $1,150cm^{-1}$ can also be associated with ether C-O symmetric and asymmetric stretching vibration (-C-O-C- ring) (Calzaferri and Imhof, 1996). This band could also be attributed to the anti-symmetrical Si-O-Si stretching mode as a result of existing silica within the samples (Socrates, 1994).

EQUILIBRIUM ADSORPTION STUDIES

Effect of pH

The effects of pH on metal ion adsorption were monitored over a pH range of $2-9$. Fifty millilitres of separate solutions, 15mg/L As(III) and 12.5mg/L Pb(II) ions, were transferred into 100ml conical flasks agitated at 150rpm for 100min with 1.00 and

TABLE 1   Physico-chemical parameters of the AC-WTB sample

Physical Aspect	Crystallinity (XRD)	Energy Dispersive X-ray Analyses	FTIR Results Surface functional groups
Powder activated carbon	Amorphous	C(82%);O(11%);Si(2%);Cl(3%);Zn(2%)	-C6H5-,-CH2-,-CH3-, -CO-,-CH-,-OH

Source: Authors' present research

1.25g AC-WTB for As(III) and Pb(II) ions respectively. The mixtures were filtered and the filtrate analysed for residual metal ions.

Figure 1 represents the effect of pH on the adsorption of As(III) and Pb(II) ions. The equilibrium adsorption capacities of metals removed from aqueous solutions are influenced by the pH (ElHussien and Isa, 2015c; Zvinowanda et al., 2009). On the other hand, the state of chemically active sites is changed by the solution pH (ElHussien and Isa, 2015c; (ElHussien and Isa, 2015b). The optimum adsorption of As(III) and Pb(II) ions was achieved at pH 5 and 6 respectively. At pH values higher than 7.2, both metals' ions were precipitated. Therefore the removal of metal ions at higher pH values is due to the formation of precipitates rather than adsorption (Bayat, 2002). The adsorption capacity (q_e) increased with increases in pH up to pH 5 for As(III) ions and pH 6 for Pb(II) ions. The increase in metal adsorption with the increase in pH is due to a decrease in competition between hydrogen ions and metal ions for the surface sites, and due to a decrease in positive surface charge.

As the pH increases, a larger area of negatively charged surface becomes available, thus facilitating greater metal adsorption. At low pH, higher concentration and mobility of H^+ ions favour H^+ adsorption compared to metal ions. Metal ions are more soluble in solution at lower pH values and this reduces their adsorption (Onundi et al., 2010). At lower pH, the surface of the adsorbent is surrounded by hydronium ions (H^+) thereby blocking metal ions from binding sites on adsorbents.

Effect of Contact Time

The investigation was done to determine the optimum time for adsorption of both metals As(III) and Pb(II) ions using AC-WTB. Then, 1.0g and 1.25g samples were contacted with 50ml separate solutions of 15mg/L As(III) and 12.5mg/L Pb(II) ions, adjusted to pH 5 and 6 respectively. The solutions were agitated at 150rpm for different contact times (10−100min). The mixtures were filtered and residual metal ions in filtrate were analysed.

The influence of contact time on the adsorption of As(III) and Pb(II) ions is shown in Figure 2. The amount adsorbed increases with increasing time up to 75min for As(III) and 35min for Pb(II). The constant adsorption at equilibration is due to the saturation of adsorption sites of AC-WTB. The rapid adsorption of metal ions during the initial stages was due to the large initial concentration gradient between the adsorbate in solution and the number of available vacant sites on the AC-WTB surface. The adsorption capacity for As(III) ions was higher than for Pb(II) ions.

Effect of Adsorbent Mass

The effect of the AC-WTB mass was investigated by equilibrating separate 50ml solutions of 15mg/L As(III) and 12.5mg/L Pb(II) ions, adjusted to pH 5 and 6 respectively, with different masses of the adsorbent (0.25−1.50g) agitated at 150rpm for 100min. The mixtures were filtered and the residual metal ions in their filtrate were analysed.
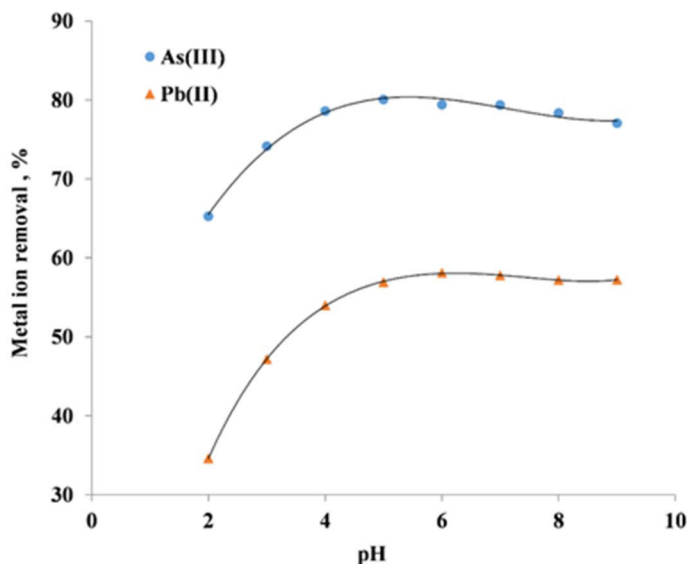
FIGURE 1　Effect of pH on As(III) and Pb(II) removal by AC-WTB: initial concentrations$=$15, 12.5mg/L, time$=$75, 35min., adsorbent masses$=$1.00, 1.25g respectively
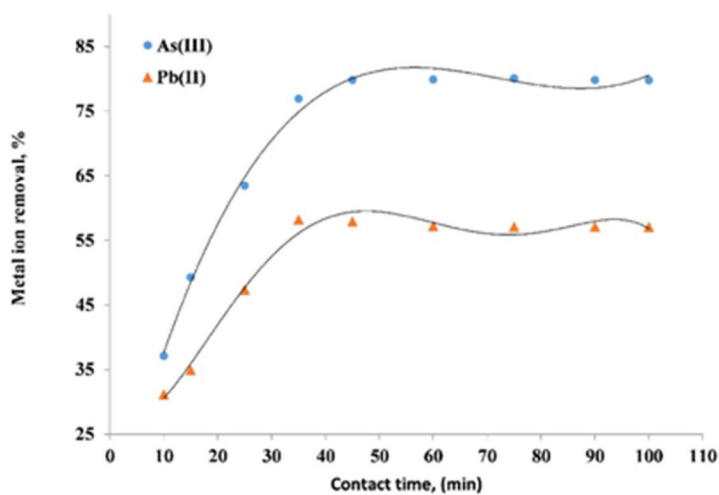
Source: Authors' present research

FIGURE 2　Effect of contact time on As(III) and Pb(II) removal by AC-WTB: pH$=$5, 6; initial concentrations$=$15, 12.5mg/L; adsorbent masses$=$1.00, 1.25g respectively

Source: Authors' present research

Figure 3 represents the effect of adsorbent mass on the adsorption capacity of As(III) and Pb(II). The figure shows an increase in adsorption capacity with increasing adsorbent mass up to a maximum of 1.00g for As(III) and 1.25g for Pb(II) giving the corresponding optimum percentage removal of 80%$\pm$2.33 and 58%$\pm$1.89 respectively. The initial increase in adsorption capacity with increasing adsorbent mass is explained by

the increase in the number of exchangeable sites for metal ion adsorption (Onundi et al., 2010). The decline in adsorption beyond certain adsorbent masses could be due to the aggregation/agglomeration of adsorbent particles at higher dosages which would lead to a decrease in the total surface area of the adsorbent particles available to the metal ions. As a result the adsorption capacity of the adsorbent is not fully utilised (Andal and Sakthi, 2010).

Effect of Initial Metal Concentration

Separate 50ml solutions of As(III) and Pb(II) ions with different initial concentrations (5−17.5mg/L) were contacted with an optimised adsorbent mass (1.00g for As(III) at pH 5 and 1.25g for Pb(II) at pH 6). The mixtures were agitated at 150rpm for 100min. The mixtures were filtered and the filtrate analysed for residual metal ions.
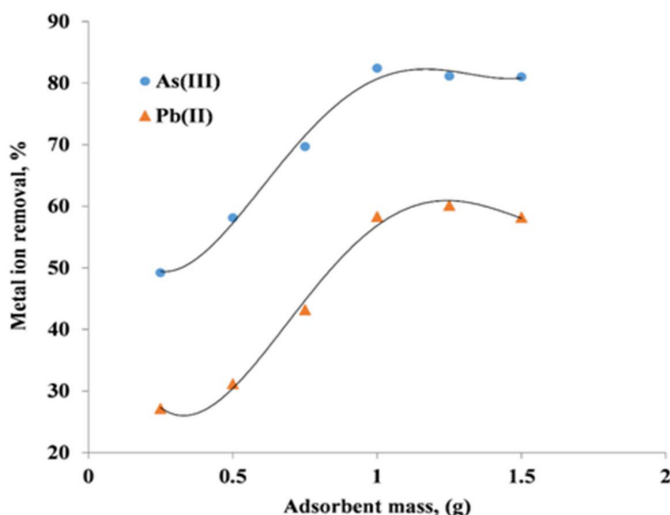

FIGURE 3   Effect of adsorbent mass on As(III) and Pb(II) removal by AC-WTB: pH=5, 6; initial concentrations=15, 12.5mg/L; time=75, 35min respectively

Source: Authors' present research

The effect of initial concentrations of As(III) and Pb(II) ions are shown in Figure 4. For both metal ions, an increase in initial concentration resulted in an increase in the adsorption capacity. This can also be explained in terms of the increase in adsorption sites coverage as the metal ion concentration is increased (Elhussien and Isa, 2015a). High metal concentration saturates the adsorbent sites more quickly, thereby decreasing the overall percentage of metal removal (ElHussien and Isa, 2015c).

Adsorption Isotherms

The Langmuir and Freundlich isotherms models were applied to the equilibrium studies to test data conformity. The linear forms of the isotherms are represented by equations 3 and 4 respectively.
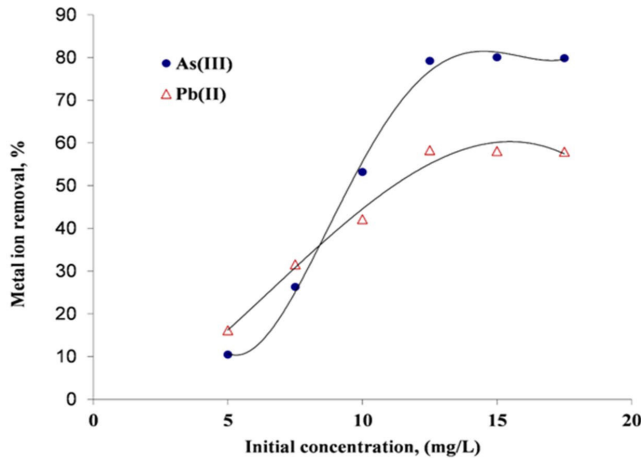
FIGURE 4   Effect of As(III) and Pb(II) initial concentrations on their removal AC-WTB: pH=5, 6; time=75, 35min.; adsorbent dose=1.00, 1.25g respectively

Source: Authors' present research

$$\frac{C_e}{q_e} = \left(\frac{1}{Q_o}\right) C_e + \frac{1}{Q_o} b \qquad (3)$$

$$\ln\left(q_e\right) = \ln\left(k_f\right) + \frac{1}{n}\ln C_e \qquad (4)$$

Where Q_o (mg g^{-1}) is the monolayer adsorption saturation capacity; b (L mg^{-1}) Langmuir constant relating to adsorption energy; q_e (mg g^{-1}) the amount of metal ion bound per gram of the adsorbent at equilibrium and C_e, the residual (equilibrium) metal ion concentration left in the solution after binding. The Q_o and b can be determined from C_e/q_e versus the C_e plot, which gives a straight line of slope (1/Q_o) and intercept of (1/bQ_o). K_f and n are the Freundlich constants characteristic of the activated carbon system.

It was observed that the Langmuir adsorption isotherm satisfactorily describes the adsorption of As(III) and Pb(II) ions from aqueous solutions using AC-WTB. However, according to the correlation coefficients (R^2) of 0.9735 and 0.9213, respectively (Figure 5), the level of conformity for As(III) is high compared to Pb(II) ions. The applicability of the Langmuir isotherm indicates good monolayer coverage of As(III) more than Pb(II) ions on the surface of AC-WTB, which consequently suggests the formation of a mono layer on the adsorbent surface in the given concentration range (Najua et al., 2008). The fact that the Langmuir isotherm fits the experimental data very well for As(III) may be due to the homogenous distribution of active sites on the adsorbent since the Langmuir equation assumes that the surface is homogenous. Thus the applicability of the Langmuir isotherm in the present system indicates the mono-layer coverage of As(III) to a high extent more than Pb(II) ions on the outer surfaces of the adsorbent (Ahmed, 2000; ElHussien and Isa, 2015c; Elhussien and Isa, 2015a). The calculated model parameters with correlation coefficients are shown in Table 2.

TABLE 2   Langmuir and Freundlich Isotherm parameters for the removal of As(III) and Pb(II) by (AC-WTB) sample

Metal ion	Langmuir			Frenudlich		
	Q_0, (mg/g)	b, (L/mg)	R^2	N	K_f,[(mg/g) (L/mg)$^{1/n}$]	R^2
As(III)	7.348	0.275	0.9735	2.396	2.087	0.9917
Pb(II)	3.791	0.237	0.9213	2.440	1.025	0.9195

Source: Authors' present research

The adsorption capacity, Q_o (mg g^{-1}) of AS(III) is higher than that of Pb(II) ions. The affinity of the two metals for the adsorbent surface in terms of b(L/mg) is higher for As(III), 0.275 compared to that of 0.237 for Pb(II). The values imply high surface energy in the process and consequently high bonding between metal ions and the AC-WTB.

The Freundlich isotherms for As(III) and Pb(II) ions adsorption are shown in Figure 6. It was observed that the Freundlich adsorption isotherm also conforms to the adsorption of As(III) and Pb(II) ions from aqueous solutions using AC-WTB. However, the level of conformity for As(III) is to a high extent compared to the Langmuir adsorption isotherms as indicated by higher correlation coefficient values of 0.9917. On the other hand, the level of conformity for Pb(II) is low compared to the Langmuir adsorption isotherms, as indicated by the lower correlation coefficient values of 0.9195. The values of n (2.396 and 2.440) for As(III) and Pb(II)) ions respectively, confirm that the AC-WTB has a heterogeneous surface since the values satisfy the heterogeneity condition where n must be in the range $1<n<10$ [10-18]. The values of 1/n ranging from 0 to 1 are a measure of adsorption intensity or surface homogeneity as the values approach zero. Values for 1/n less than 1 indicate normal Langmuir adsorption isotherm, while those values above 1 are indicative of cooperative adsorption (Calzaferri and Imhof, 1996; Ali and Muhammad, 2008). The values of K_f for As(III) ions (2.087) and Pb(II) ions (1.025) imply that there was low uptake of the metal ions onto the adsorbent surface.
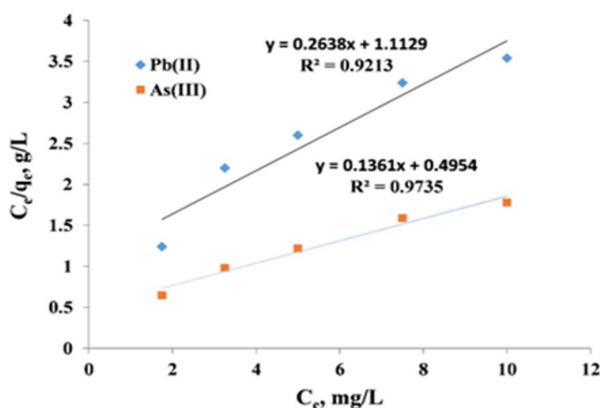

FIGURE 5   Langmuir adsorption isotherm for As(III) and Pb(II) by AC-WTB
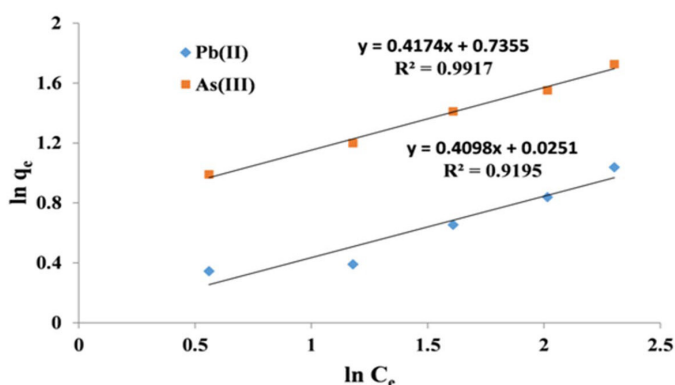
Source: Authors' present research

FIGURE 6   Freundlich adsorption isotherm for As(III) and Pb(II) by AC-WTB

Source: Authors' present research

CONCLUSIONS

This study has demonstrated that AC-WTB has favourable properties for the adsorption of As(III) and Pb(II) ions from aqueous solutions. The adsorption of As(III) and Pb(II) ions were highly dependent on experimental parameters such as contact time, initial metal concentration, adsorbent mass and pH. The adsorption of As(III) was found to be optimum at pH 5, adsorbent dose of 1.0g, metal ion concentration of 15mg/L and contact time of 75min. The adsorption of Pb(II) was found to be optimum at pH 6, adsorbent mass of 1.25g, metal ion concentration of 12.5mg/L and contact time of 35min.

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