

# Removal of Lead and Benzene from Groundwater by Zeolite and Brown Coal: Isotherm and Kinetic Studies

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**Abstract** - The feasibility of using brown coal and zeolite as low-cost adsorbents for the removal of lead and benzene from groundwater in permeable reactive barrier (PRB) was investigated in batch studies. The Freundlich, Langmuir, and Temkin adsorption isotherms were used to analyse the experimental data. The kinetic analyses of the adsorption processes were performed using the pseudo-first order and pseudo-second order kinetic models. The Langmuir isotherm adequately described the adsorption of lead onto the materials, whereas the adsorption of benzene was adequately described by the Freundlich isotherm based on the correlation coefficients of the different isotherms. The Langmuir dimensionless constant, separation factor ( $R_L$ ) determined were between  $0 < R_L < 1$ , indicating favourable adsorption. The heat of adsorption  $b$  ( $\text{kJmol}^{-1}$ ) obtained were generally somewhat low, indicating physisorption of lead to the adsorbates. For the kinetic studies, the pseudo-second order model suitably described the removal of lead by both zeolite and brown coal whereas in the case of benzene the pseudo-first order model gave a better fit.

**Keywords:** Permeable reactive barriers, Reactive materials, Lead, Benzene, Isotherms, Kinetics.

## 1. Introduction

Lead and benzene are among the contaminants commonly found in groundwater. The release of lead into groundwater resources occurs largely via anthropogenic activities such as mining or through industrial production of batteries and antifouling paints. The presence of benzene (often detected together with xylene, ethylbenzene and toluene) in groundwater may be due to accidental releases of petroleum products and their subsequent migration to the subsurface or leakage of these products from underground storage tanks. These contaminants are profoundly detrimental to humans and other life forms. Consequently, their presence in groundwater resources often calls for urgent remediation of the contaminated area or isolation of the contamination source (Bowlen and Kosson, 1995; Adepoju-Bello and Alabi, 2005; USEPA, 2007).

Over the years, concerted efforts have been made to develop innovative methods that are relatively cheap and sustainable for groundwater remediation. Among the innovative technologies currently available, the permeable reactive barrier (PRB) technology has received tremendous attention. With the PRB technology, a carefully selected reactive medium is installed across the trajectory of the contaminant plume to remove the contaminants by a single or a combination of mechanisms such as adsorption, precipitation and or biodegradation (Carey et al., 2002; Henderson and Demond, 2007).

PRBs have been used to treat a broad spectrum of contaminants including chlorinated solvents (TCE, PCE, DCE, and VC), petroleum hydrocarbons (BTEX), heavy metals and radionuclides. The removal of

benzene in PRBs has mostly been accomplished by adsorption and biodegradation processes. Though the latter mechanism is usually the preferred option as it leads to a reduction of contaminant mass load, it can sometimes be a slow process and may not suffice in reducing the risk posed to the environment within the desired time frame. Heavy metals such as lead, on the other hand, are characteristically not biodegradable, thus processes such as adsorption, precipitation and reduction-oxidation have often been utilized for their removal. In the literature, the application of a variety of materials that utilize the adsorption mechanism to remove these contaminants in PRBs has been documented (Scherer et al., 2000). These materials are efficient but costly as well. Therefore, there is currently a growing interest in identifying new, preferably cheaper materials to replace these efficient, but rather expensive adsorbents in PRBs (Ahmad et al., 2007). Among them, the application of materials such as natural zeolites and brown coal (lignite) is becoming increasingly attractive due to their low cost and availability.

The suitability of an adsorbent is not only dependent on its adsorption capacity, but also on the energy and kinetics of adsorption. In the present study, the feasibility of using zeolites and brown coal for the removal of benzene and lead was investigated through isotherm and kinetic studies.

## 2. Materials and Methods

### 2. 1. Reagents

The reagents used were analytical grades of Pb (1000 mg/L  $\pm$  4 mg/L; c-HNO<sub>3</sub> = 2% w/w; Fluka) and benzene (Fluka, 99%). The contaminated solution was prepared by spiking the deionized water with the required amount of lead and benzene to achieve the desired initial concentration.

### 2. 2. Adsorption and Kinetic Study

Amber bottles of 250 mL in volume were filled with the reactive materials and the contaminated solution to achieve a solid to liquid ratio of 1:20. Different initial concentrations: 2.5 to 100 mg/L for lead and 2 to 50 mg/L for benzene were used. The mixtures were then placed on a 150 rpm orbital shaker and agitated for 48 hours at room temperature (22 $\pm$ 1°C), after which 15 mL of the solution was taken for Pb analysis using atomic absorption spectrophotometry (AAS 3, Corp Zeiss). In the case of benzene, 100 mL of the aqueous solution was taken into a 100 mL volumetric flask. One mL of pentane was added to the solution and agitated for 5 min to extract the benzene. The amount of benzene in the pentane was then determined as the final concentration using gas chromatography (Shimadzu GC 17A, PAF/A/5/Sb). The amount of lead and benzene adsorbed per unit weight of the adsorbent  $Q_e$  was calculated using Eq. (1). The Freundlich, Temkin and Langmuir isotherms were then utilized to analyze the experimental data. The coefficient of determination ( $R^2$ ) was used to determine the goodness of fitting the experimental data to the isotherms.

$$Q_e = v \left( \frac{C_0 - C_e}{m} \right) \quad (1)$$

Where:  $Q_e$  is the amount of benzene and lead adsorbed (mg/g) at equilibrium;  $C_0$  and  $C_e$  are the initial and equilibrium concentration in mg/L, respectively;  $m$  is the mass of the adsorbent (g) and  $v$  is the volume of the solution (L).

For the kinetic studies, the amounts of benzene and lead removed at a particular time were calculated using Eq. (2).

$$q_t = v \left( \frac{C_0 - C_t}{m} \right) \quad (2)$$

Where:  $q_t$  is the amount sorbed at time ( $t$ ) (mg/g);  $C_0$  is the initial concentration in mg/L and  $C_t$  is the concentration at time ( $t$ ) in mg/L;  $m$  is the mass of the adsorbent (g) and  $v$  is the volume of the solution (L).

### 3. Results and Discussion

The Freundlich isotherm assumes that the adsorbent has unlimited capacity to sorb the sorbate. It suitably describes adsorption onto heterogeneous surfaces and multi-layer adsorption. However, it does not have a sound theoretical basis, in that it was derived empirically (Juang et al 1996). It is represented mathematically by Eq. (3) (Freundlich, 1906):

$$Q_e = K_{iF} C_e^{1/n_i} \quad (3)$$

Where:  $K_{iF}$  (L/g) is the Freundlich constant or capacity factor, which indicates the steepness of the adsorption;  $1/n_i$  is the Freundlich exponent indicating the curvature of the isotherm or the intensity of adsorption (Akgerman and Zardkoohi, 1996).  $C_e$  (mg/L) represents the equilibrium concentration of the adsorbate in the liquid phase and  $Q_e$  is the equilibrium amount sorbed (mg/g), which is the summation of the adsorption of all sites (Do, 1998). Equation (3) can be transformed into a logarithmic relation to obtain a linear version (Eq. 4) that allows the determination of  $K_{iF}$  (the intercept) and  $1/n_i$  (the slope).

$$\log Q_e = \log K_{iF} + \frac{1}{n_i} \log C_e \quad (4)$$

The higher the  $K_{iF}$  value the greater the adsorption intensity. Igwe and Abia (2007) stated that the magnitude of the exponent gives an indication of the favourability and capacity of the adsorbent/adsorbate system. Values of  $n_i > 1$  represent favourable adsorption conditions whereas those  $< 1$  represent unfavourable adsorption. In most cases, the exponent between  $1 < n_i < 10$  indicate beneficial adsorption (Chantawong et al., 2003). The results of the analysis of the experimental data for both Pb and benzene using the Freundlich isotherm are presented in Table 1 below. The adsorption capacities were generally higher for Pb than benzene. The obtained  $K_{iF}$  values also suggest that brown coal has a higher capacity to sorb both Pb and benzene than zeolite under identical operating conditions. This may be due to the use of multiple removal mechanisms such as adsorption, ion-exchange and/ or complexation by the brown coal. The  $1/n_i$  values obtained for both materials, however, indicate favourable adsorption of benzene ( $1/n_i < 1$ ) and unfavourable adsorption of Pb ( $1/n_i > 1$ ) by the materials.

Table 1. Freundlich isotherm with the correlation coefficient ( $R^2$ ).

Parameter	Pb		Benzene	
	Brown coal	Zeolite	Brown coal	Zeolite
$1/n_i$	1.121	1.096	0.885	0.702
$K_{iF}$ (L/g)	6.910	3.627	0.232	0.029
$R^2$	0.762	0.868	0.992	0.804

The Langmuir isotherm was derived based on two assumptions: (i) the forces of interaction between the adsorbed molecules are negligible, and that, once a molecule occupies a site no further sorption takes place at this site. This assumption suggests the preclusion of sorption of an oncoming molecule through interaction between an adsorbed molecule and the oncoming molecule; (ii) adsorbents have saturation limits beyond which no further sorption takes place. In effect, sorption is limited by the sorption capacity of the adsorbent (Langmuir, 1918). In general, the Langmuir isotherm gives a good fit for monolayer surfaces and is represented by Eq. (5) below (Lui and Lui, 2008).

$$Q_e = \frac{\Gamma_m K_a C_e}{1 + K_a C_e} \quad (5)$$

Where:  $C_e$  is the equilibrium solution concentration (mg/L),  $Q_e$  is the amount adsorbed (mg/g) at equilibrium. The  $\Gamma_m$  is the maximum achievable surface concentration of a given compound in terms of

monolayer adsorption;  $K_a$  is the Langmuir constant related to the energy of adsorption. Several linear versions of the Langmuir isotherm exist as shown in Eqs. (6) - (9) below (Ofomaja, and Ho, 2008; Subramanyam and Das, 2009). By performing linear regressions on the data,  $K_a$  and  $\Gamma_m$  can be obtained.

$$\frac{C_e}{Q_e} = \frac{1}{\Gamma_m} C_e + \frac{1}{K_a \Gamma_m} \quad \text{Plot of } \frac{C_e}{Q_e} \text{ vs } C_e \quad (6)$$

$$\frac{1}{Q_e} = \frac{1}{\Gamma_m K_a} \frac{1}{C_e} + \frac{1}{\Gamma_m} \quad \text{Plot of } \frac{1}{Q_e} \text{ vs } \frac{1}{C_e} \quad (7)$$

$$\frac{C_e}{Q_e} = \frac{1}{\Gamma_m} C_e + \frac{1}{K_a \Gamma_m} \quad \text{Plot of } Q_e \text{ vs } \frac{Q_e}{C_e} \quad (8)$$

$$\frac{Q_e}{C_e} = K_a \Gamma_m - K_a Q_e \quad \text{Plot of } \frac{Q_e}{C_e} \text{ vs } Q_e \quad (9)$$

In this study, Eq. (7) was used to determine the Langmuir isotherm parameters. The results are presented in Table 2 below. The  $\Gamma_m$  and  $K_a$  were both higher for brown coal than for zeolite in the case of both benzene and lead. This may be due to the use of multiple removal mechanisms such as adsorption, ion-exchange and/ or complexation, the smaller particle size resulting in higher surface area and pore volume in the brown coal. The positive values of  $\Gamma_m$  and  $K_a$  imply that the adsorption of lead and benzene onto brown coal and zeolite could be described by the Langmuir isotherm.

The favourability of adsorption of the adsorbate onto the adsorbent was tested using the Langmuir isotherm dimensionless constant called the separation factor  $R_L$ , which is defined by the Eq. (10) below:

$$R_L = \frac{1}{(1+K_a C_0)} \quad (10)$$

Where:  $C_0$  is the initial adsorbate concentration (mg/L) and  $K_a$  is the Langmuir constant (mg/L) (Hall et al., 1966). Values of  $R_L$  between 0 and 1 indicate favourable absorption. Also  $R_L$  values equal to 0 indicate irreversible absorption,  $R_L = 1$  is linear and  $0 > R_L > 1$  indicate that adsorption is not favoured. The  $R_L$  values obtained for both Pb and benzene in this study for both brown coal and zeolite were between 0 and 1, indicating favourable adsorption. Those of benzene were positive and ranged from 0.0173-0.3056 for zeolite and 0.0067-0.1447 for brown coal for initial concentrations of 2-50 mg/L, thus indicating favourable adsorption of benzene onto both brown coal and zeolite. Also in the case of brown coal, the  $R_L$  for Pb ranged between 0.0032-0.1129 and that of zeolite was 0.0045 to 0.1533 for initial concentration of 2.5 mg/L to 100 mg/L, also indicating favourable adsorption. The lower  $R_L$  obtained for Pb for both materials indicate that in the simultaneous removal of both Pb and benzene, the former will be preferentially adsorbed than the latter onto both materials.

Table 2. Langmuir isotherm constants with the correlation coefficient ( $R^2$ )

Parameter	Pb		Benzene	
	Brown coal	Zeolite	Brown coal	Zeolite
$K_a$ (L/mg)	3.144	2.210	2.956	1.136
$\Gamma_m$ (mg/g)	1.664	1.535	0.717	0.118
$R^2$	0.939	0.959	0.967	0.351

The Temkin isotherm was used to determine the binding energies of the sorption process. The derivation of this isotherm is based on the assumption that: (i) the heat of sorption decreases linearly with coverage rather than logarithmically, as implied in the Freundlich equation with coverage due to

adsorbent–adsorbate interactions and, (ii) the adsorption is characterized by a uniform distribution of binding energies. The Temkin isotherm has generally been expressed by Eq. 11 (Qureshi et al., 2009).

$$Q_e = \frac{RT}{b} \ln K_T + \frac{RT}{b} \ln C_e \quad (11)$$

Where:  $K_T$  is the Temkin equilibrium binding constant or the adsorption potential ( $\text{Lmg}^{-1}$ ),  $b$  is related to the heat of adsorption ( $\text{kJmol}^{-1}$ ),  $R$  is the gas constant ( $8.314 \times 10^{-3} \text{ kJK}^{-1}\text{mol}^{-1}$ ) and  $T$  is the absolute temperature (K).

A plot of  $Q_e$  versus  $\ln C_e$  enables the determination of the isotherm constant  $K_T$ , and  $b$ . With physisorption, the adsorbate adheres to the adsorbent only through weak Van der Waals interactions, thus physisorption processes have relatively low adsorption energies. Chemisorption processes on the other hand have relatively high adsorption energies in that the adsorbate adheres to the surface through the formation of a chemical bond. Qureshi et al. (2009) noted typical binding energy for the ion-exchange mechanism to be in the range of 8–16  $\text{kJ mol}^{-1}$ . Generally, the values of  $b$  obtained in the present study for the contaminants indicate somewhat weak ionic interactions (physisorption) (Table 3). The potential of adsorption  $K_T$  also suggests that there is higher potential for the adsorption of Pb than benzene. Moreover, between the adsorbents, brown coal presents a higher adsorption potential for both adsorbates than zeolite.

Table 3. Temkin isotherm constants with the correlation coefficient ( $R^2$ )

Parameter	Pb		Benzene	
	Brown coal	Zeolite	Brown coal	Zeolite
$K_T$ (L/mg)	56.604	36.94	4.180	1.363
$b$ ( $\text{kJmol}^{-1}$ )	7.515	4.483	21.984	0.399
$R^2$	0.342	0.428	0.898	0.794

The interactions between adsorbates and adsorbents that culminate in adsorbate removal are time-dependent. Kinetic studies are often undertaken to enable understanding of the dynamic interactions, predict the fate of contaminants and design efficient management systems (Sparks and Suarez, 1991). To determine the uptake rate of Pb and benzene, two kinetic models such as pseudo-first order and pseudo-second order were applied to the experimental data.

The pseudo-first order equation is one of the most popular kinetic equations (Qin et al., 2006). The differential equation is given by Eq. (12). After integration and applying the initial conditions  $q = 0$  at  $t = 0$  and  $q = qt$  at  $t = t$ , Eq. (12) becomes:

$$\frac{dq}{dt} = k(q_e - q) \quad (12)$$

After integration by applying the initial conditions  $q = 0$  at  $t = 0$  and  $q = qt$  at  $t = t$ , Eq. 12 becomes Eq. (13), which can also be rearranged to obtain a linear form Eq. (14).

$$\log\left(\frac{q_e}{q_e - qt}\right) = \frac{k_1}{2.303} t \quad (13)$$

$$\log(q_e - qt) = \log q_e - \frac{k_1}{2.303} t \quad (14)$$

Where:  $q_t$  and  $q_e$  are the amount of lead adsorbed ( $\text{mg/g}$ ) at time  $t$  and at equilibrium, respectively, and  $k_1$  is the rate constant of the pseudo-first-order adsorption process ( $\text{hr}^{-1}$ )

The adsorption kinetics may also be described by a pseudo-second-order equation. The differential equation is the following (Ho et al., 1999).

$$\frac{dq}{dt} = k_2 (q_e - q)^2 \quad (15)$$

Integrating Eq. (15) and applying the boundary conditions, gives Eq. (16), which can also be rearranged to obtain a linear form to obtain Eq. (17).

$$\frac{1}{q_e - q_t} = \frac{1}{q_e} + k_2 t \quad (16)$$

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{1}{q_e} t \quad (17)$$

The results of the kinetic studies are presented in Table 4. The results indicate that the adsorption of lead onto brown coal and zeolite followed the second order rate equation whereas that of benzene followed the first order kinetic model, when comparing the correlation coefficients ( $R^2$ ). In terms of materials, adsorption of both benzene and lead onto brown coal is faster than adsorption onto zeolite.

Table 4. Parameters of the Pseudo first and second kinetic models with their correlation coefficient ( $R^2$ )

Model type	Parameter	Brown coal		Zeolite	
		Pb	Benzene	Pb	Benzene
Pseudo-1 <sup>st</sup> order	q <sub>e</sub> (exp) (mg/g)	3.066	1.921	2.600	1.366
	q <sub>e</sub> (cal) (mg/g)	1.904	1.798	2.518	1.349
	k <sub>1</sub> (hr <sup>-1</sup> )	0.087	0.124	0.086	0.096
	R <sup>2</sup>	0.949	0.954	0.811	0.992
Pseudo 2 <sup>nd</sup> order	k <sub>2</sub> (g/mg/hr)	0.113	0.089	0.029	0.177
	q <sub>e</sub> (exp) (mg/g)	3.066	1.921	2.600	1.366
	q <sub>e</sub> (cal) (mg/g)	3.026	2.156	3.117	1.043
	R <sup>2</sup>	0.971	0.928	0.822	0.950

#### 4. Conclusion

The feasibility of using brown coal and zeolite as low-cost adsorbents for the removal of lead and benzene from groundwater in a PRB was investigated. Three adsorption isotherms namely: Freundlich, Langmuir, and Temkin adsorption isotherms were used to analyse the experimental data. By considering the correlation coefficients ( $R^2$ ), the adsorption of Pb onto the materials followed the Langmuir isotherm, whereas the adsorption of benzene onto the materials was adequately described by the Freundlich isotherm. The adsorption capacities obtained were consistently higher for brown coal than for zeolite for both contaminants, suggesting that brown coal has a higher capacity in removing the contaminants than zeolite. From the values of the apparent energy of adsorption, the adsorption process was found mostly to be a physiosorption process. For the kinetic studies, the pseudo-second order model suitably described the rate of Pb removal by both zeolite and brown coal whereas in the case of benzene the pseudo first order model gave a better fit. The study, in general, demonstrates the feasibility of applying brown coal and zeolite in PRBs for the removal of both lead and benzene from contaminated groundwater.

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