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Evaluation of Clay Brick and Laterite Brick as Low-Cost Adsorbents for the Removal of Cd^{2+} and Pb^{2+} in Aqueous Solutions

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ABSTRACT

Adsorption is recognized as a low-cost and effective method for treating Cadmium and Lead in wastewater. Therefore, this study focused on investigating the potential of construction demolition wastes (CDWs), laterite bricks (LB), and clay brick (CB) to remove Cd and Pb in synthetic wastewater. A series of batch adsorption experiments were conducted to study the adsorption characteristics and capacities. The results revealed that the maximum adsorption capacity of CB and LB reached 210.85 mg/g and 210.72 mg/g for Pb^{2+}, respectively. The maximum adsorption capacity of Cd^{2+} by CB and LB was 4.52 mg/g and 4.51 mg/g. From adsorption isotherms, the Langmuir and Freundlich models fitted well for $0 \leq C_i \leq 1000$ mg/L of Cd^{2+} adsorption on both CB and LB. Adsorption of Pb^{2+} onto CB and LB is well captured by all isotherm models tested. A similar pattern of adsorption was observed among all particle sizes of CB and LB (<0.5, 0.5-1.0, and 1.0–2.0 mm), indicating that the particle size of tested adsorbents is not affecting the adsorption isotherms of Cd^{2+} and Pb^{2+}. However, the particle size ranges used in this study lay within a narrower range, which might be the reason for not showing any difference in adsorption among the different size fractions. Therefore, it is concluded that both CB and LB are effective and environmentally friendly adsorbents to remove Pb^{2+} and Cd^{2+}in wastewater.

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INTRODUCTION

Accumulation of heavy metals in the environment is a major concern that needs to be addressed. Direct discharging of industrial wastewater containing heavy metals into the environment is the leading cause for this issue, and the removal of such contaminants has become a global challenge over the past few decades. Specific characteristics of heavy metals such as higher atomic weight, higher density, and bioaccumulation led to their wide distribution in the environment, raising concerns over their potential effects on human health and the ecosystem.

The accumulation of Cd^{2+} in the water resources is mainly due to industrial sources such as corrosion of galvanized and cadmium-composing pipelines, untreated effluents of battery processing reactions, mining, manufacturing of phosphate fertilizers, and electronic components (Malakootian *et al.,* 2009; Hai , 2018). Accumulation of Pb^{2+} in the water resources is mainly due to industrial activities such as lead smelting, coal combustion, lead-based paints, lead-containing pipes or lead-based solder in water supply systems, battery recycling, grids and bearings, paper and pulp industries (Flora *et al.,* 2012; Mansoorian *et al.,* 2014). Cadmium (Cd) and Lead (Pb) are highly toxic to humans and the environment: Cd affects kidneys and the skeleton of humans and possesses carcinogenic effects; Pb is a cumulative toxicant particularly hazardous to young children and pregnant women. A higher concentration of lead can cause a severe impact on human life (Fu and Wang 2011; Sarma *et al.,* 2015). Therefore, Cd^{2+} and Pb^{2+} from industrial wastewater must be removed before discharge to the environment (Sewwandi *et al.,* 2014).

Several methods have been used to refine heavy metals in wastewater, such as coagulation, flocculation, reverse osmosis, membrane separation, and ion exchange. All these methods are characterized by high capital and operational cost (Fu and Wang, 2011; Kumara *et al.*, 2018; Kushwaha *et al.,* 2020). Adsorption has emerged to be a better alternative treatment method characterized as a highly efficient, effective, and economically feasible wastewater treatment method especially fit for the developing world (Mondal, 2009; Chiban, 2012; Kumara *et al.*, 2019; Bandara *et al.*, 2020). Adsorption treatment methods have been proved as economical and efficient for removing heavy metals, organic pollutants, and dyes from polluted waters (Karnib *et al.,* 2014).

Construction and demolition waste (CDW) is generally defined as waste that arises from construction, demolition, and renovation activities, including land excavation, site clearance, civil and building construction, demolition activities, roadwork, and building renovation (Ranjan *et al.,* 2014; Tripathi and Rawat Ranjan, 2015). Rapid urbanization, industrialization, and population growth in developing and developed countries produce millions of tons of CDW per year (Kumara *et al.*, 2018). Construction demolition waste materials of Sri Lanka, consisting of 21% of Concrete, 25% of Sand, 14% cement, 14% bricks, 7 % rubble, and 6% cement blocks approximately (Kulatunga *et al.,* 2006). These CDW can be used as adsorbent materials. Therefore, this study aimed to characterize Clay Brick (CB) and Laterite Brick (LB) in terms of Cd^{2+} and Pb^{2+} adsorption properties and to compare the maximum adsorption capacities of Clay Brick (CB) and Laterite Brick (LB) in terms of Cd^{2+}, and Pb^{2+} adsorption in aqueous solutions.

METHODOLOGY

Adsorbent Preparation and Characterization

The laterite bricks were collected from Hanwella, and clay bricks were collected from Anuradhapura. Laboratory experiments were conducted in the Soil and Water Science laboratory in the Faculty of Agriculture, Rajarata University of Sri Lanka, Puliyankulama, Anuradhapura, Sri Lanka,

The raw CB and LB were crushed using clean wooden mortar and pestle to break the aggregated structure. Then samples were air-dried in a cleanroom at room temperature. To facilitate air drying, samples were spread to a thin layer on a clean piece of paper and shallow containers (plastic containers were used to avoid metal contamination). Large impurities such as debris, stones, etc., were removed. Later, the materials were sieved using 0.5, 1.0, 2.0 mm plastic sieves. Then, the samples were analyzed for pH, Electrical Conductivity (EC), Cation Exchange Capacity (CEC), and trace elements (Cd and Pb) following standard procedures. Each analysis was triplicated for particle size <0.5 mm, 1 – 2 mm, and >2 mm.

Preparation of Stock Solution

$Cd(NO_3)_2.4H_2O$ and $PbCl_2$ were used to prepare stock solutions. The solutions, 1000 mg/L, were prepared by dissolving the proper amount of metal salts in deionized water. The various concentrations were prepared by diluting prepared stock solutions in deionized water to obtain the appropriate concentrations (0, 25, 50, 75, 100, 200, 400, 600, 800, 1000 mg/L).

Batch Adsorption Experiment

Batch adsorption experiments were conducted using the standard method recommended by the Organization of Economic Cooperation and Development (OECD, 2000). The method was chosen to determine each adsorbent's adsorption behavior under full contact conditions.

The experiment was conducted in three replicates in each 0.5 mm, 1.0 mm, and 2.0 mm particle size. Sorbates solutions having 25, 50, 75, 100, 200, 400, 600, 800, 1000 mg/L of Cd^{2+} and Pb^{2+} were prepared. The known weight (1.5 g) of sorbents was measured into a 100 ml conical flask separately for each particle size. Thus, 360 samples were prepared for all three particle sizes (9 samples from each particle size). For the batch adsorption experiments, 15 ml of solute solution was pipetted into a centrifuge tube with 1.5 g of adsorbents under the natural pH of the adsorbents. The sample tubes were positioned on a reciprocating shaker under room temperature (25 ^{o}C) and shakes for 24 hours at 100 rpm. After 24 hours, the samples were centrifuged at 6500 rpm for 5 min, and supernatants were removed from the centrifuge tubes by membrane filter (WhatmanTM filter paper 42 µm). Then the residual heavy metals ions concentrations were analyzed using inductively coupled plasma-optical emission spectroscopy (ICP-OES).

Adsorption Isotherms

This study investigated the adsorption capacities for Cd^{2+} and Pb^{2+} concentrations from 0 mg/L to 1000 mg/L using three different isotherm equations. Linear model [Eq. (1)], Langmuir model [Eq. (2)] and Freundlich model [Eq. (3)] were used (Hai, 2018).

$$Q_e = K_d C_e \qquad (1)$$

$$(C_e/Q_e) = (1/bQ_m) + (C_e/Q_m) \qquad (2)$$

$$Q_e = K_f C_e^{1/n} \qquad (3)$$

Where, Q_m (mg/g) = maximum adsorption capacity, C_e (mg/L) = equilibrium concentration of Heavy Metals; Q_e (mg/g) = amount adsorbed per adsorbent at equilibrium; b (g/L) = Langmuir constant related to bindingand K_f (L/g) is the Freundlich adsorption capacity, and 1/n is the adsorption intensity.

Statistical Analysis

The best fitted models for Cd^{2+} and Pb^{2+} adsorption by CB and LB were identified by using OriginPro 2019b graphing and analysis software. Significant differences among the adsorption capacities of tested adsorbents were investigated using paired t-test.

RESULTS AND DISCUSSION

Physicochemical Properties of Adsorbents

Adsorption of heavy metals in the liquid phase into the solid phase mainly depends on the physical and chemical properties of the adsorbent. The basic physicochemical properties of CB and LB are presented in Table 1.

Table1: Physico-chemical properties of Clay Brick (CB) and Laterite Brick (LB).

Adsorbents	Particle size (mm)	Moisture content (%)	Specific gravity	pH	EC (mS/cm)	CEC (Cmol/Kg)
Clay Brick (CB)	0.5	0.036±0.00	2.44	6.04±0.23	0.31±0.01	20.87±0.35
	1.0	0.032±0.00		5.95±0.16	0.28±0.02	19.30±0.75
	2.0	0.016±0.00		6.03±0.11	0.18±0.02	16.80±0.36
Laterite Brick (LB)	0.5	0.004±0.01	3.06	5.08±0.10	3.24±0.10	18.30±0.50
	1.0	0.005±0.00		6.09±0.09	1.85±0.75	14.70±0.44
	2.0	0.005±0.00		6.18±0.05	0.75±0.05	18.23±0.76

There is no significant difference in pH values in different size fractions of CB. Clay is an inorganic colloid resulting in a high buffering capacity. Therefore, pH values observed for different size fractions of clay bricks were more or less similar irrespective of the particle size. However, 0.5 mm fraction in comparison to the other two fractions, 0.5 – 1 mm or > 1 mm, of LB showed significantly lower pH values. It might be due to the greater replacement of H^+ in the smallest fraction due to higher specific surface areas. According to the results, selected adsorbents had an almost neutral pH. As a result, negatively charged adsorbent surfaces can absorb cations from wastewater and, during the adsorption process, may be able to neutralize acidic wastewater (Kumara and Kawamoto, 2019).

The Electrical Conductivity of LB is higher than CB, which may be due to the different mineral compositions in these two materials. The Electrical Conductivity decreases with the particle size increment. The increasing trend of CEC was observed with decreasing soil particle size. Moreover, higher CEC was observed in CB in comparison to LB. It may be due to differences in clay mineral compositions of CB and LB.

Effect of Particle Size on Adsorption

The effect of particle size on selected heavy metal adsorption was investigated for three particle sizes (0.5 mm – 2.00 mm) at room temperature. The measured maximum adsorption capacities for Cd^{2+} and Pb^{2+} of three different fractions are shown in Figure 1. According to the results, Pb^{2+} adsorption by both CB and LB are shown higher adsorption capacity than the Cd^{2+} adsorption. Pb^{2+} adsorption onto CB varied from 19.62 mg/g to 19.55 mg/g, while Pb^{2+} adsorption onto LB shows the same value (19.54 mg/g) among all three fractions.

It is reported that adsorption increases with decreasing particle size. It is probably due to increasing internal surface area with decreasing particle size. According to Huang *et al.,* (2014)fine adsorbent particles can transport heavy metals more than coarser particles due to larger specific surface areas. Furthermore, Al-Anber, (2010) , Al-Senani & Al-Fawzan, (2018) described that most interior particle surfaces are used for adsorption because metal ions transferred along a short path inside adsorbent particles pores, the smaller particle size results in high rates of adsorption. However, the particle size ranges used in this study (<0.5 mm, 0.5–1 mm, and 1-2 mm) lay within a narrower range, which might be the reason for not showing any difference in adsorption among the differentsize fractions.

Effect of Initial Heavy Metal Concentration on Cd^{2+} and Pb^{2+} Adsorption

The effect of initial heavy metal concentration on the adsorption was studied by batch adsorption experiments at room temperature using different initial metal ion concentrations (0, 25, 50, 75, 100, 200, 400, 600, 800, 1000 mg/L).

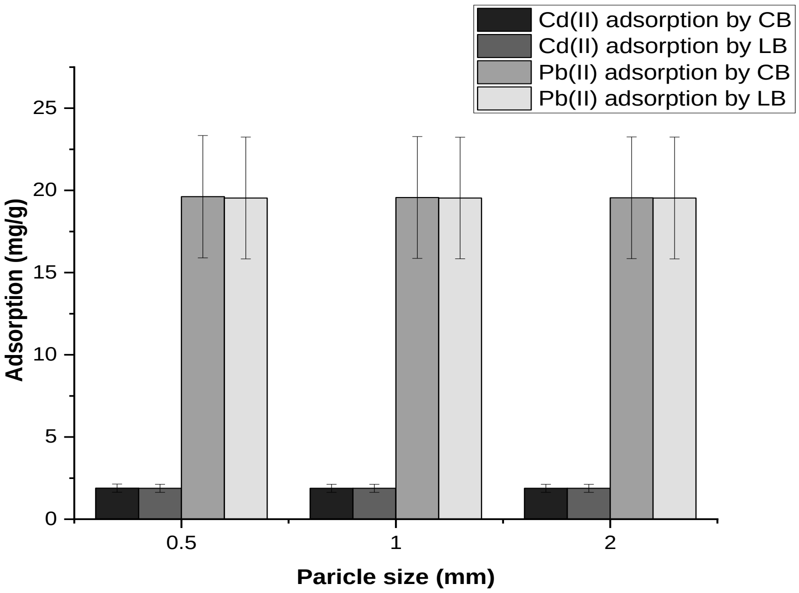

Figure 1: Effect of particle size of the adsorbents on Cd^{2+} and Pb^{2+} adsorption.

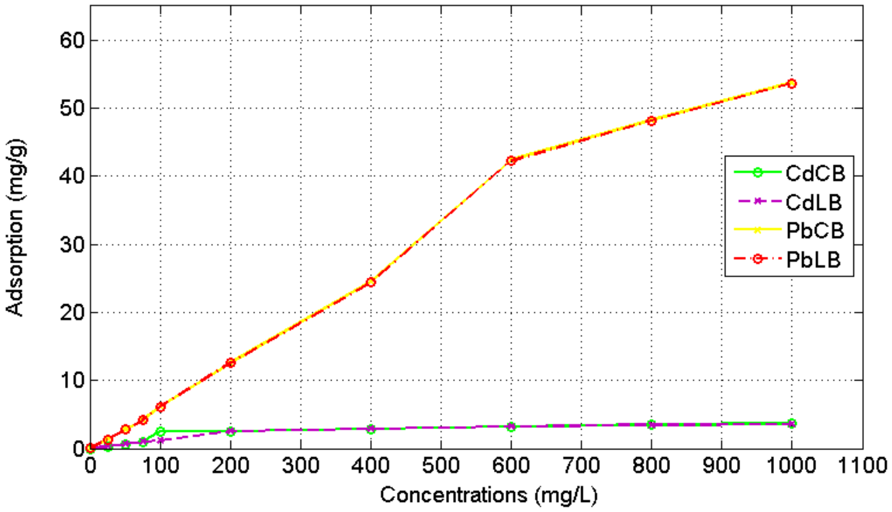

Figure 2: Effect of initial heavy metal concentration on Cd^{2+} and Pb^{2+} adsorption by CB and LB.

As shown in Figure 2, the adsorption capacity sharply increased with the initial concentration of heavy metals ions. The maximum adsorption capacities of $Cd^{2+}$and Pb^{2+} by CB were 3.60 mg/g and 53.72 mg/g, respectively. On the other hand, the maximum adsorption capacities of $Cd^{2+}$and Pb^{2+} by LB were 3.60 mg/g and 53.60 mg/g, respectively. Increasing initial heavy metal ion concentrations increase the driving force at the solid-liquid interface, resulting in increased adsorption capacity until the adsorption sites were saturated (Ouyang *et al.,* 2019).

Adsorption Isotherms

Cd^{2+} Adsorption by CB

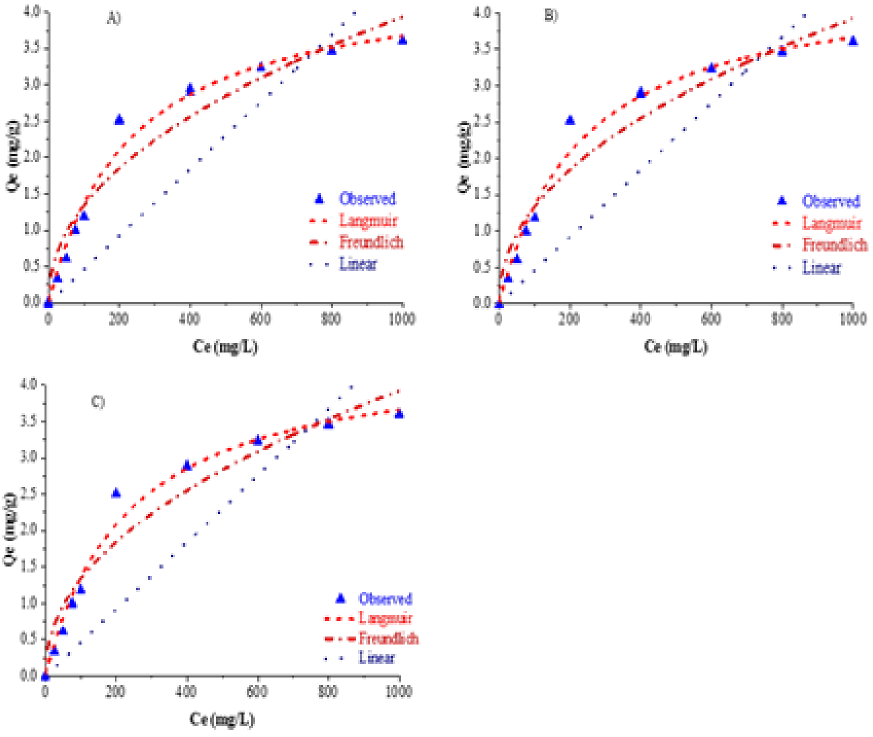

Figure 3. Cd^{2+} Adsorption Isotherms of Clay Brick: A) CB (<0.5 mm), B) CB (1.0 mm), C) CB (2.0 mm).

Figure 3 illustrates the measured data and the fitted adsorption isotherms models. The Langmuir model fitted well (R^2=0.98) for the concentration range of $0 \leq C_i \leq 1000$ mg/L on Cd^{2+} adsorption onto clay brick compared to the other two models. This shows a mono-layer type of adsorption phenomenon in terms of Cd^{2+} onto CB. The maximum adsorption capacity (Q_m) of clay brick was 4.52 mg/g (Table 2).

The Freundlich model's adsorption of Cd^{2+} is well described based on the high correlation coefficients R^2 (0.94), suggesting possible multi-layer adsorption in the concentration range of $0 \leq C_i \leq 1000$ mg/L. Furthermore, K_f of this adsorbent was 0.16 L/g at its particle size of 0.5 mm.

The linear model is empirical; the measured correlation coefficient R^2 value is 0.65 for all three fractions, indicating that the linear model was most suitable for low initial concentrations (Table 2).

Cd^{2+} Adsorption by LB

Figure 4 describes the adsorption of Cd^{2+} onto LB with different particle sizes while the fitted parameters are shown in Table 2. Like CB, the Langmuir model well describes Cd^{2+} adsorption onto LB (R^2 =0.98) followed by the Freundlich model (R^2=0.94). This indicated that mono adsorption in which cadmium ions are adsorbed onto the homogeneous surfaces of LB is dominant. However, in terms of the cadmium adsorption onto LB, the 1/n, calculated from the linear form of the Freundlich model was lower than 1, suggesting that the Freundlich model is favorable.

Table 2: Cd^{2+} Adsorption isotherm parameters of CB & LB

Heavy metals	Particle size (mm)	Linear model		Langmuir model			Freundlich model		
		K_d (L/g)	R^2	b (g/L)	Q_m (mg/g)	R^2	K_f (L/g)	$1/n$	R^2
Cd^{2+}-CB	0.5	0.005	0.65	0.004	4.522	0.98	0.16	0.47	0.94
	1.0	0.005	0.65	0.004	4.521	0.98	0.15	0.47	0.94
	2.0	0.005	0.65	0.004	4.517	0.98	0.15	0.47	0.94
Cd^{2+}-LB	0.5	0.005	0.66	0.004	4.511	0.98	0.15	0.47	0.94
	1.0	0.005	0.66	0.004	4.511	0.98	0.15	0.47	0.94
	2.0	0.005	0.66	0.004	4.513	0.98	0.15	0.47	0.94

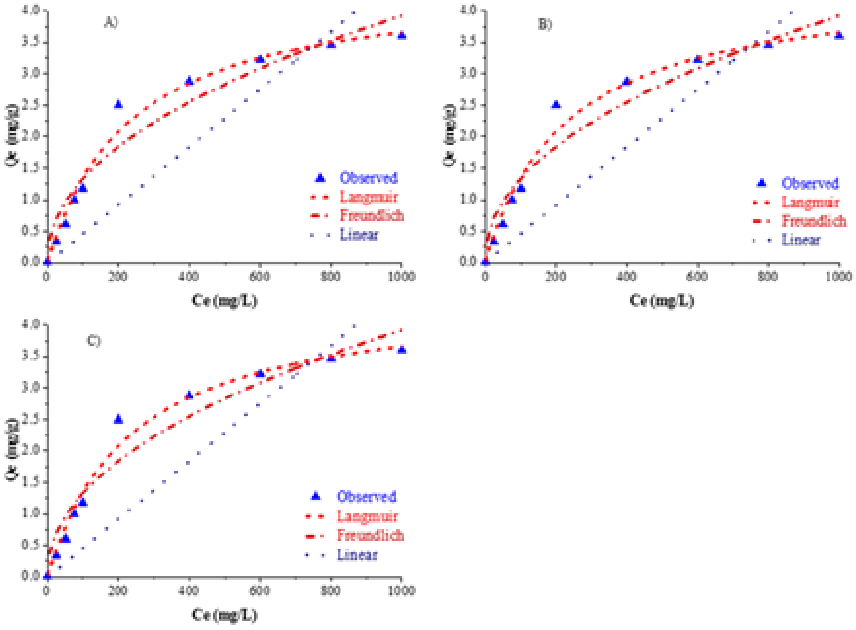

Figure 4. Cd^{2+} Adsorption Isotherm of Laterite Brick: A) LB (<0.5 mm), B) LB (1.0 mm), C) LB (2.0 mm).

The measured *Q_m* value for Cd^{2+} adsorption onto laterite brick was 4.51 mg/g for all three fractions and obtained a similar adsorption pattern, suggesting that particle size did not affect the Cd^{2+} adsorption onto laterite brick this study.

The replicated data which was used to fit the Cd^{2+} Adsorption by CB & LB were shown in Table 3

Table 3: The replicated data used to fit the Cd^{2+} Adsorption isotherm models by CB & LB.

Concentrations (mg/L)	Cd^{2+} adsorption by CB in different particle sizes (mm)			Cd^{2+} adsorption by LB in different particle sizes (mm)		
	0.5	1.0	2.0	0.5	1.0	2.0
0	0.00±0.01	0.00±0.00	0.00±0.00	0.01±0.00	0.01±0.00	0.01±0.00
25	0.34±0.00	0.34±0.00	0.34±0.00	0.34±0.00	0.34±0.00	0.34±0.00
50	0.61±0.00	0.61±0.00	0.61±0.00	0.61±0.00	0.61±0.00	0.61±0.00
75	1.00±0.00	1.00±0.00	1.00±0.01	1.00±0.00	1.00±0.00	1.00±0.00
100	1.19±0.00	1.18±000	1.18±0.00	1.18±0.00	1.18±0.00	1.18±0.00
200	2.52±0.01	2.51±0.01	2.50±0.01	2.50±0.00	2.50±0.00	2.50±0.01
400	2.93±0.03	2.89±0.02	2.88±0.01	2.89±0.01	2.88±0.00	2.88±0.02
600	3.24±0.01	3.23±0.00	3.23±0.00	3.22±0.00	3.22±0.00	3.22±0.00
800	3.47±0.00	3.47±0.00	3.46±0.01	3.46±0.00	3.46±0.00	3.46±0.00
1000	3.60±0.01	3.60±0.01	3.60±0.00	3.60±0.00	3.60±0.00	3.60±0.00

Pb^{2+} Adsorption by CB

The studied adsorption isotherms for Pb^{2+} adsorption onto clay brick with three fractions are shown in Figure 5, while the fitted parameters are shown in Table 4. Adsorption of Pb^{2+} onto clay brick is well captured by all Langmuir, Freundlich, and linear models based on the high correlation coefficients R^2 (>0.98).

The *Q_m* value for Pb^{2+} adsorption onto clay brick is high at 2.0 mm size fractions with a recorded value of 210.85 mg/g. The slopes of the linear form of Freundlich model 1/n were closer to 0, showing that the Freundlich model is favorable.

According to Figure 5, similar adsorption behavior shows for all three fractions, suggesting that the particle size did not affect all adsorption isotherms

Pb^{2+} Adsorption by LB

Figure 6 illustrates the Pb^{2+} adsorption isotherm of laterite brick for all particle size fractions. The three well-known isotherm models Linear, Langmuir, and Freundlich were fitted and experimental data are shown in Table 4. A similar adsorption pattern was observed for all particle sizes, suggesting that particle size does not directly affect the adsorption of Pb^{2+} onto laterite brick.

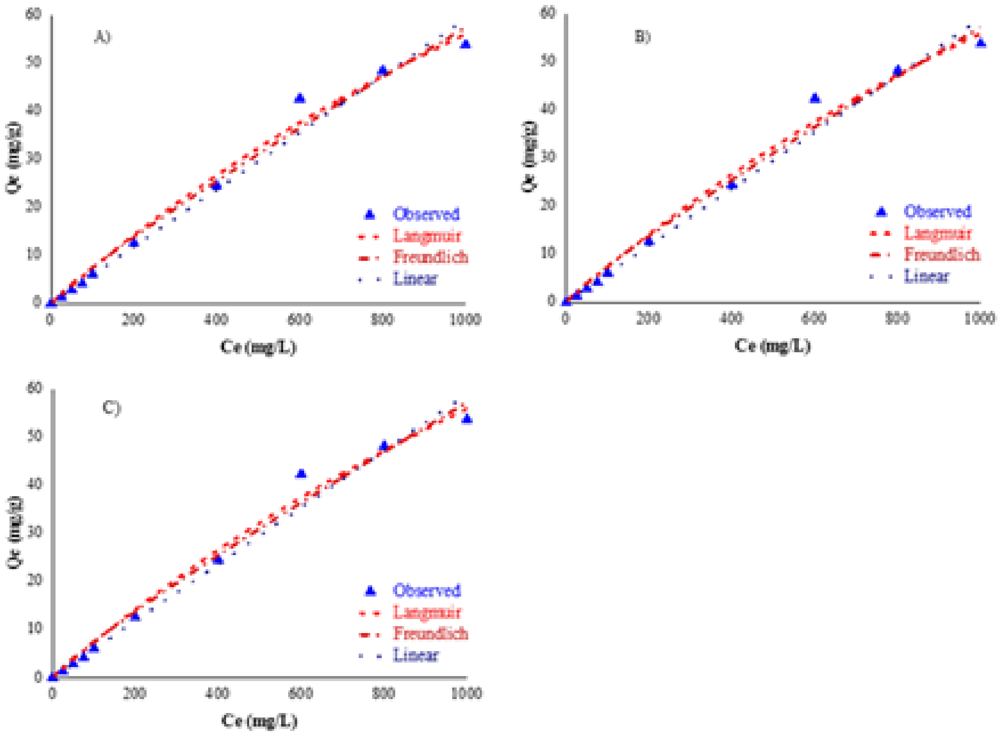

Figure 5. Pb^{2+} Adsorption Isotherm of Clay Brick: A) CB (<0.5 mm), B) CB (1.0 mm), C) CB (2.0 mm).

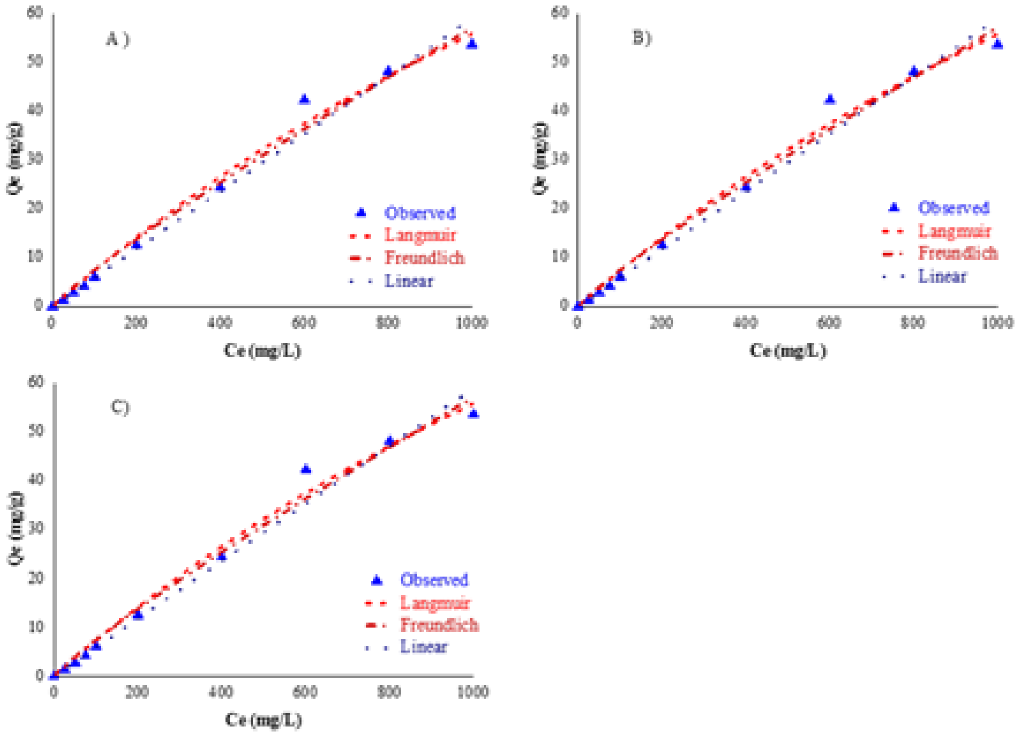

Figure 6. Pb^{2+} Adsorption Isotherm of Laterite Brick: A) LB (<0.5 mm), B) LB (1.0 mm), C) LB (2.0 mm)..

Table 4: Pb^{2+} Adsorption isotherm Parameters of CB & LB.

Heavy metals	Particle size (mm)	Linear model		Langmuir model			Freundlich model		
		K_d (L/g)	R^2	b (g/L)	Q_m (mg/g)	R^2	K_f (L/g)	$1/n$	R^2
Pb^{2+}-CB	0.5	0.059	0.98	3.6×10^{-4}	210.53	0.99	0.13	0.89	0.99
	1.0	0.059	0.98	3.6×10^{-4}	210.16	0.99	0.13	0.89	0.99
	2.0	0.059	0.98	3.6×10^{-4}	210.85	0.99	0.13	0.89	0.99
Pb^{2+}-LB	0.5	0.059	0.98	3.6×10^{-4}	210.45	0.99	0.13	0.89	0.99
	1.0	0.059	0.98	3.6×10^{-4}	210.56	0.99	0.13	0.89	0.99
	2.0	0.059	0.98	3.6×10^{-4}	210.72	0.99	0.13	0.89	0.99

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Table 5: The replicated data used to fit the Pb^{2+} Adsorption isotherm models by CB & LB.

Concentrations (mg/L)	Pb^{2+} adsorption by CB in different particle sizes (mm)			Pb^{2+} adsorption by LB in different particle sizes (mm)		
	0.5	1.0	2.0	0.5	1.0	2.0
0	0.01±0.00	0.01±0.00	0.01±0.00	1.01±0.00	0.01±0.00	0.01±0.00
25	1.37±0.00	1.37±0.00	1.37±0.00	1.37±0.00	1.37±0.00	1.37±0.00
50	2.80±0.00	2.80±0.00	2.80±0.00	2.80±0.00	2.80±0.00	2.80±0.00
75	4.17±0.00	4.17±0.00	4.17±0.00	4.17±0.00	4.17±0.00	4.17±0.00
100	6.10±0.00	6.10±0.00	6.10±0.00	6.10±0.00	6.10±0.00	6.10±0.00
200	12.56±0.00	12.55±0.00	12.50±0.01	12.55±0.00	12.55±0.00	12.55±0.00
400	24.55±0.01	24.50±0.02	24.45±0.02	24.38±0.01	24.37±0.00	24.37±0.00
600	42.52±0.09	42.34±0.02	42.29±0.02	42.27±0.00	42.26±0.00	42.26±0.00
800	48.35±0.13	48.15±0.01	48.12±0.01	48.11±0.00	48.11±0.00	48.12±0.00
1000	53.80±0.01	53.70±0.01	53.66±0.02	53.60±0.00	53.59±0.00	53.60±0.00

The measured Q_m value was slightly lower than clay brick, and a higher value was obtained at fractions of 2.0 mm (210.72 mg/g). The adsorption intensity (1/n) of Pb^{2+} adsorption onto LB calculated from the linear form of the Freundlich model was lower than1.0.

The replicated data which was used to fit the Pb^{2+} Adsorption by CB & LB were shown in Table 5

The Cd^{2+} and Pb^{2+} adsorption by CB and LB compared by paired t-test revealed that Pb^{2+} adsorption into CB is significantly higher than Pb^{2+} adsorption into LB and Cd^{2+} adsorption into CB is higher than LB

CONCLUSIONS

From adsorption isotherms, the Langmuir and Freundlich models fitted well for $0 \leq C_i \leq 1000$ mg/L of Cd^{2+} adsorption on both CB and LB. Adsorption of Pb^{2+} onto CB and LB is captured by all Langmuir, Freundlich and linear models. The maximum adsorption capacity of CB and LB reached 210.85 mg/g and 210.72 mg/g for Pb^{2+} respectively. The maximum adsorption capacity of Cd^{2+} by CB and LB was 4.52 mg/g, and 4.51mg/g respectively. A similar pattern of adsorption was observed among all particle sizes of CB and LB(<0.5, 0.5-1.0, and 1.0–2.0 mm). It suggests that the particle size of tested adsorbents is not affecting the adsorption isotherms of Cd^{2+} and Pb^{2+}. However, the particle size ranges used in this study lay within a narrower range, probably the reason for not showing any difference in adsorption among the different size fractions. Therefore, in conclusion, both CB and LB are effective and environmentally friendly adsorbents to remove $Pb^{2+}$and Cd^{2+}in wastewater.

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