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RESEARCH ARTICLE

Utilizing Thermally Regenerated Diatomaceous Earth from Spent Diatomaceous Earth to Remove Lead Ions from Wastewater

M. W. E. H. Wimalarathna[1], A. N. Navaratne[2] and C. Gunathilake[3,*]

[1]*Department of Environmental and Industrial Sciences, University of Peradeniya, Peradeniya, Sri Lanka*

[2]*Department of Chemistry, University of Peradeniya, Peradeniya, Sri Lanka*

[3]*Department of Chemical and Process Engineering, University of Peradeniya, Peradeniya, Sri Lanka*

*chamilag@pdn.ac.lk

Abstract

This study describes a method of removal of lead ions (Pb^{2+}) in wastewater using thermally regenerated diatomaceous earth formed from spent diatomaceous earth (SDE) under 400 and 800 °C (SDE-400 °C and SDE-800 °C, respectively). SDE is generated as an industrial waste from raw diatomaceous earth (RDE or simply called DE) used in food processing and brewery industries. This study reveals the effectiveness and efficiency of SDE as an adsorbent for removing Pb^{2+} from aqueous solutions with respect to adsorbent dosage and the contact time. A well-organized porous structure with some particles on the surface is shown by the surface morphology of the SDE dry form as determined by field emission scanning electron microscopy (FE-SEM). Surface area (BET) analysis of DE and the dry form of SDE was carried out at liquid nitrogen temperature, and discovered that DE has a higher specific surface area and total pore volume of 3.70 $m^2\ g^{-1}$ and 0.015 $cm^3\ g^{-1}$, respectively, than those of SDE dry form, in which the values were 2.22 $m^2\ g^{-1}$ and 0.010 $cm^3\ g^{-1}$. The optimal conditions for Pb^{2+} adsorption onto SDE were identified. The adsorbent dosage of 50 mg and contact time of 180 min at a pH of 4.8, sustained for 50 mL of lead solution at 25 °C, produced the maximum adsorption. The equilibrium results of the investigated systems were interpreted using the Freundlich and Langmuir adsorption isotherm models. With the maximum adsorption capacities of 92.60, 163.93, 208.33, and 322.58 mg g^{-1} of Pb^{2+}, the Langmuir model best fitted the adsorption properties of Pb^{2+} on DE (or RDE), SDE, SDE-400 °C , and SDE-800 °C. Due to the presence of chemisorption, the present study further suggests that SDE-800 °C is a more advantageous application of SDE as an effective material for the application in removal of Pb^{2+} from aqueous solutions for industrial wastewater treatment than the other three adsorbents.

Keywords: Adsorption, Isotherm, Pb^{2+}, Spent diatomaceous earth, Thermal regeneration

Introduction

Water is an essential requirement for all sorts of growth and development of humans, animals and plants, and it is in great demand due to its scarcity. However, the suitability of water for a selected purpose is often suffering from other substances dissolved or suspended in it. In recent years, with the rapid pace of industrialization, an enormous quantity of wastewater containing heavy metals and dyes is continuously discharged into water bodies, which causes devastating effects on the ecological environment and human health. In addition, almost all heavy metal ions are difficult to be biodegraded showing some resistance to environmental conditions; hence, wastewater treatment is of considerable significance requiring urgent implementation.

Lead is the commonest heavy metal; Lead and its compounds possibly enter the ecosystems through soil, air and water, as a result of industrial processes, such as the production of paint and pigments, insecticides containing lead, batteries, petroleum refining, electrical and electronic products and lead smelters leaching from lead pipes and Pb/Sn soldered joints as well as runoff from land close to heavily travelled highways (Zhou and Haynes, 2011). The practically widespread use of lead compounds in plumbing fixtures and as solder in water distribution systems is significant from the standpoint of drinking water in order to safeguard both people and the environment (Mahdi et al., 2018). Therefore, effective and economical methods for treatment of wastewater are mandatory to reduce pollutant concentrations to an acceptable level. It is a matter of caring for our environment and our own health.

Adsorption technology, one of the most cost-effective and environmentally friendly methods of water purification (Bilgin and Tulun, 2015), was used in this study with diatomaceous earth (DE)/ raw diatomaceous earth (RDE) as the adsorbent due to its unique properties, such as high silica content, high porosity, high surface area and chemical inertness. (Mateo et al., 2017). Additionally, spent diatomaceous earth (SDE), a waste product of DE, was used to prepare thermally regenerated DE utilizing the thermal regeneration method at 400 and 800 °C, respectively. Note that SDE is mostly produced as an industrial waste by the brewing and food processing industries (Tsai et al., 2004).

The main goals of this study were to investigate the efficacy and efficiency of the removal of Pb^{2+} from aqueous solutions by raw diatomaceous earth (RDE), SDE, and two thermally regenerated materials from SDE such as SDE-400 °C and SDE-800 °C, and to further identify the efficiency of SDE by varying the parameters: contact time, adsorbent dosage and initial concentration of adsorbate. For simplicity, we used DE symbol for RDE as well.

Adsorption is usually described by isotherm which is a relationship between the amounts of adsorbate adsorbed on the surface of adsorbent at a constant temperature, and there are several isotherm models reported (Ng et al., 2017). This study is mainly focused on two adsorption isotherm models; Langmuir and Freundlich.

Due to its simplicity and capacity to match a wide range of adsorption data, the Langmuir isotherm is the most widely used isotherm model, which depends on the assumption that the adsorbent surface is made up of active sites with homogeneous energy distribution and that adsorption occurs in monolayers (Ayawei et al., 2017). Due to its simplicity and capacity to match a wide range of adsorption data, the Langmuir isotherm is the most widely used model.

The Langmuir isotherm equation is written as;

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e} \quad (1)$$

where the K_L (L mg^{-1}) is the Langmuir constant, C_e (mg L^{-1}) is the equilibrium concentration, q_e and q_m (mg g^{-1}) are the equilibrium and maximum adsorbed quantities, respectively.

The following equation shows how the linear Langmuir isotherm equation is most frequently used;

$$\frac{1}{q_e} = \frac{1}{K_L q_m} \times \frac{1}{C_e} + \frac{1}{q_m} \quad (2)$$

The parameters q_m and K_L can be calculated by plotting $\frac{1}{q_e}$ vs. $\frac{1}{C_e}$, and the curve fitting can be verified by calculating the correlation coefficient (R^2), which should be a straight line with a slope of $\frac{1}{K_L q_m}$ and an intercept of $\frac{1}{q_m}$ (Akafu et al., 2019). The following expression is used to calculate the quantity of ion adsorbed at equilibrium (adsorption capacity), q_e (mg g^{-1}), by the adsorbent:

$$q_e = \frac{(C_0 - C_t)}{m} \times V \quad (3)$$

where C_0 and C_e are the concentrations of Pb^{2+} solution at initial and at equilibrium, respectively, in mg dm^{-3}, m is the mass of the adsorbent used in g, and V is the volume of the Pb^{2+} solution in dm^3 (Gunathilake et al., 2015).

The separation factor or equilibrium parameter (R_L), determined by the following equation, can be used to explain the key characteristics of the Langmuir isotherm model.

$$R_L = \frac{1}{(1 + K_L C_0)} \quad (4)$$

The values of R_L indicate the type of isotherm, and there are four possibilities for the R_L value. (i) $0 < R_L < 1$ for favourable sorption, (ii) $R_L > 1$ for unfavourable sorption, (iii) $R_L = 1$ for linear sorption, and (iv) $R_L = 0$ for irreversible sorption (Akafu et al., 2019).

The Freundlich adsorption isotherm, which implies that the adsorbate is adsorbed on a heterogeneous surface via multilayer sorption and that the stronger binding sites are occupied first, leads to a reduction in binding strength with increasing site occupancy (Kuang et al., 2020).

This isotherm is given by;

$$q_e = K_F C_e^{1/n} \quad (5)$$

The linear form of the Freundlich isotherm model can be expressed as;

$$\log q_e = \log K_F + \frac{1}{n} \log C_e \quad (6)$$

where K_F [mg g^{-1} (mg $L^{-1})^n$] is the Freundlich coefficient, and the dimensionless constant $1/n$ indicates the intensity of adsorption or surface heterogeneity ($0 < 1/n < 1$). In general, as the K_F value rises, so does the adsorbent's ability to absorb a certain adsorbate. A straight line with a slope of $1/n$ and an intercept that equals the value of $\log K_F$, demonstrating multilayer sorption capacity, is produced by plotting $\log q_e$ against $\log C_e$ (Kuang et al., 2020).

The ion removal efficiency (percentage ion removal) can be calculated by the following expression (Akafu et al., 2019):

$$\text{Ion removal percentage} = \frac{(C_0 - C_e)}{C_0} \times 100 \quad (7)$$

Materials and Methods

Chemicals

The diatomaceous earth (DE or RDE) and its waste, spent DE (SDE), were provided by Lion Brewery (Ceylon) PLC. Lead(II) nitrate [$Pb(NO_3)_2$] with min. purity of 99% was purchased from Sisco Research Laboratories Pvt. Ltd. (India). Sodium hydroxide pellets (above 98.0% purity) were purchased from Daejung Chemicals & Metals Co., Ltd (Korea) and concentrated nitric acid (65%) was obtained from Sigma Aldrich. Both these chemicals were used to adjust the pH of the solutions. All chemicals used in the experiments were of analytical reagent grade, and hence, further purification was not required.

Instrumentation

The field emission scanning electron microscopy (Hitachi SU6600 FE-SEM, Japan) and nitrogen adsorption-desorption analyser (ASAP 2010 volumetric analyser) were used for the characterization of DE and SDE, respectively. Volumetric analyser was used to calculate surface properties, including specific surface area (S_{BET}), total pore volume (V_t), micropore volume (V_{mi}), pore width (W_{max}) and pore size distribution (PSD) of DE and SDE samples.

The thermal regeneration of SDE was performed in a laboratory furnace (Muffle Furnace - Human Lab DMF-12, Medi Lab Research Technologies (Pvt) Ltd., Sri Lanka). at temperatures of 400 and 800 °C. Orion 420A pH meter (USA) was used to adjust the pH of each sample solution. The adsorbent and the adsorbate were allowed to contact with each other for pre-determined time periods using Lab Companion SK-600 Benchtop Shaker (Korea).

The total concentrations of Pb^{2+} were determined by Flame Atomic Absorption Spectrometer (Thermo M series AAS, Germany).

Methodology

Thermal regeneration of SDE: The thermal regeneration process was carried out in a laboratory furnace (maximum temperature is beyond 800 °C). A specific amount of SDE sample (10.0000 g) was heated at a rate of 10 °C min^{-1} to 400 °C in air, and then held at that temperature for 2 h. Similarly, another sample with equal mass was subjected to heating upto 800 °C in flowing air and kept for 2 h. After thermal regeneration, the resulting products were cooled in a desiccator, and labelled as SDE-400 °C and SDE-800 °C, respectively. Then, subsequent experiments and characterization measurements were carried out with four types of adsorbents (RDE, SDE, SDE-400 °C, and SDE-800 °C).

Adsorption isotherms: Stock solution (1000 mg L^{-1}) of Pb^{2+} was prepared by dissolving (1.5985±0.0001) g of $Pb(NO_3)_2$ solid in distilled water in a 1000 mL volumetric flask and diluting up to the mark.

The adsorption isotherm studies of Pb^{2+} were performed in initial concentrations ranging from 50 to 300 mg L^{-1}. The required 50 mg dosage of four different adsorbents (RDE, SDE, SDE-400 and SDE-800 °C) was dispersed in various concentrations of Pb^{2+} solutions (50.0 mL) at pH 4.8 in a reagent bottle with constant shaking of 150 rpm for about 3 h at room temperature.

In order to do this, a stock solution that was appropriately diluted with 50.00 mg L^{-1} of $Pb(NO_3)_2$ solution was first prepared in a 1000 mL volumetric flask. Then, a sample of (0.0500±0.0001) g of each of the four adsorbents was separately added to four bottles, each of which contained (50.00±0.05) mL of 50.00 mg L^{-1} $Pb(NO_3)_2$ solution. Throughout this process, the optimum parameters were maintained (pH of the solution = 4.8, volume of the solution = 50 mL, contact time = 3 h and stirring rate = 150 rpm).

Following settling, the solutions were diluted in a ratio of 1:10 before being used for

the analysis using flame atomic absorption spectroscopy (λ_{max} = 662.5 nm). The initial concentrations of Pb^{2+} 100, 150, 200, 250, and 300 mg L^{-1} were varied while still following the same process.

Results and Discussion

This research work is primarily concerned with the thermal regeneration of SDE due to the ease of performance, short time duration and mainly the utilization of SDE as an adsorbent after thermally regeneration is very scarce in literature. As can be seen from Figure 1, significant colour difference among the four different adsorbents used, such as RDE, SDE, SDE-400 °C and SDE-800 °C, indicates the possible variation of characteristic chemical and adsorption properties among them. The primary components of SDE are SiO_2 and Al_2O_3 which have high melting points of 1710 °C and 2072 °C, respectively. As the sintering process could occur at 950 °C for SDE, heating it up to 400 and 800 °C would show no effect on these main components.

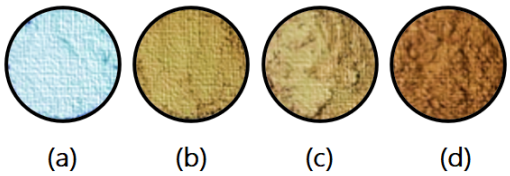

Figure 1. Differences in the colour of four types of adsorbents: (a) RDE (b) SDE (c) SDE-400 °C and (d) SDE-800 °C.

Characterization of adsorbents

SEM Analysis: The surface morphology of SDE was characterized using field emission scanning electron microscopy (FE-SEM) prior to the adsorption of lead ions. After a 15-second gold sputter coating, the sample was mounted onto the sample stub using carbon tapes, and the images were then taken.

Images shown in Figure 2 illustrates a well-organized porous structure with some particles on the surface. Thus, it is apparent that neither the annealing temperature nor the waste particles have sufficient influence to close pores. However, the high temperature has the ability to burn off or remove residues still exist inside the interior pores. It is believed that these apertures will improve the adsorption of Pb^{2+} ions.

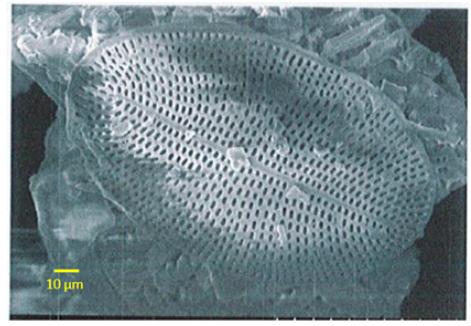

Figure 2. SEM image of SDE before adsorption.

Nitrogen adsorption-desorption analysis: The ASAP 2010 volumetric analyser was used to measure the nitrogen adsorption isotherms at -196 °C (Figure 3). All materials were out gassed at 110 °C for 2 h while under vacuum prior to the adsorption experiments.

The volume of the single-point pores (V_{sp}) was calculated from the amount of material adsorbed at a relative pressure (p/p^0) of ~ 0.98. The modified KJS (Kruk-Jaroniec-Sayari) method calibrated for cylindrical pores was used to compute the pore size distributions (PSD) utilizing the adsorption branches of nitrogen adsorption-desorption isotherms. With a cross-sectional area of 0.162 nm^2 for each N_2 molecule, the Brunauer-Emmett-Teller specific surface areas (S_{BET}) were derived from the N_2 adsorption isotherms in the relative pressure range of 0.05–0.20 (Table 1). BET analysis of DE and the dry form of SDE was carried out at liquid nitrogen temperature, and discovered with respect to silica that DE has a higher specific surface area and total pore volume of

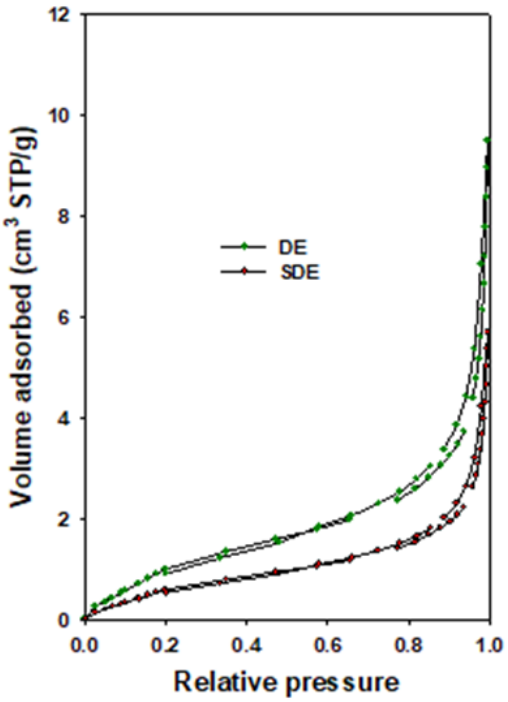

Figure 3. Nitrogen adsorption-desorption isotherms of DE (RDE) and SDE.

3.70 m^2 g^{-1} and 0.015 cm^3 g^{-1}, respectively, than those of SDE dry form, in which the values were 2.22 m^2 g^{-1} and 0.010 cm^3 g^{-1}.

Table 1. Adsorption parameters for DE (RDE) and SDE samples.

Sample	V_{sp} (cc/g)	V_{mi} (cc/g)	S_{BET} (m2/g)	V_t (cc/g)
RDE	0.014	<0.01	3.7	0.015
SDE	0.008	<0.01	2.22	0.01

Single-point pore volume (V_{sp}) calculated at the relative pressure of 0.98; V_{mi} is cumulative pore volume of micropores - pores below 2 nm were calculated on the basis of the KJS method; S_{BET} is specific surface area calculated from adsorption data in relative pressure range 0.05 - 0.20; V_t is the total pore volume calculated by integration of the PSD curve.

Effect of SDE dosage on Pb^{2+} ion removal

A key factor in the adsorption process is the quantity of contact surface between an adsorbent and adsorbate solution. The effect of adsorbent dose on Pb^{2+} removal using SDE was performed by varying the adsorbent dose from 10 mg to 100 mg while maintaining other parameters constant at their respective optimum conditions (pH = 4.8, contact time = 180 min, initial concentration of Pb^{2+} solution = 100 mg L^{-1}, and agitation speed = 150 rpm) and the results obtained was shown in Figure 4. Accordingly, the extent of adsorption was gradually increased with the increase in the SDE dosage up to 50 mg, and thereafter, no more considerable increase was observed.

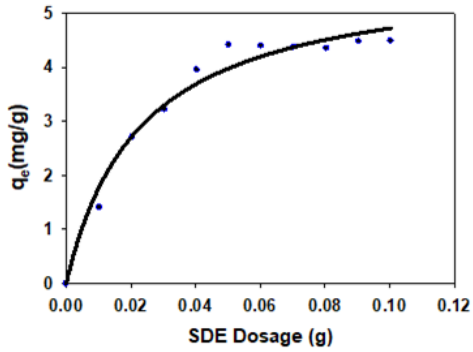

Figure 4. Effect of adsorbent dosage on adsorption capacity of SDE (initial Pb^{2+} concentration = 100 mg L^{-1}, pH = 4.8, contact time = 180 min, and stirring rate = 150 rpm).

These results may be primarily explained by the fact that as the adsorbent dose is increased, more active adsorption sites become available, allowing more adsorbate ions to adhere to it initially. Later, it may reach its saturation level because there are no longer any adsorbates that can be adsorbed at higher dosages.(Elmorsi et al., 2019; Sriram et al., 2020). Therefore, 50 mg was selected as the optimized adsorbent dose of SDE with the maximum adsorption capacity of 4.42 mg g^{-1} at an optimum pH of 4.8 and used for further experiments.

Effect of contact time on Pb^{2+} ion removal

The study of the effect of contact time on the adsorption capacity of SDE on Pb^{2+} removal was carried out by varying it from 30 to 240 min, keeping other parameters constant at optimum values. Figure 5 shows the effect of contact time on the adsorption capacity of the adsorbent and illustrates the gradual increase of the adsorption capacity with the contact time from 30 to 180 min, and afterwards, no more significant increase was observed which further emphasizes that the adsorption capacity was reached equilibrium within 180 min with the maximum value of 5.65 mg g^{-1}.

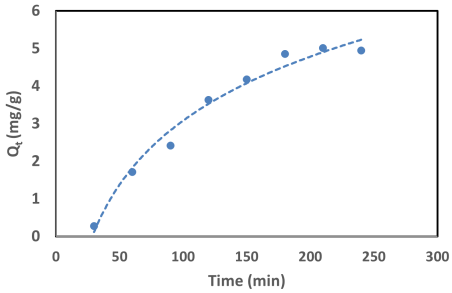

Figure 5. Effect of contact time on adsorption capacity of SDE (adsorbent dosage = 50 mg, initial Pb^{2+} concentration = 100 mg L^{-1}, pH = 4.8 and stirring rate = 150 rpm).

This can be explained by the availability and accessibility of vacant adsorption sites on the SDE surface. However, as time passed and active sites were occupied, the remaining vacant sites became difficult to access due to physical constraints and repulsion forces.

Adsorption isotherms of Pb^{2+} ions

In the present study, two main adsorption isotherm models of Langmuir and Freundlich were applied to the experimental data. Equation 2 was used to obtain the Langmuir isotherm fit to investigate adsorption characteristics of Pb^{2+} on RDE, SDE, SDE-400 °C and SDE-800 °C. Figure 6 shows the fitting of experimental data to the Langmuir isotherm for Pb^{2+} on four different adsorbents. Equation 4 was used to calculate the value of dimensionless equilibrium parameter R_L, which is referred to as separation factor or equilibrium parameter (Bilgin and Tulun, 2015).

In this study, Langmuir isotherm indicates favorable conditions for the adsorption of Pb^{2+} on the adsorbents, because R_L values determined using experimental data are between zero and unity (Bilgin and Tulun, 2015). Equation 6 was used in order to apply the experimental data to Freundlich isotherm and Figure 7 shows the validity of the Freundlich isotherm of the experimental data on Pb^{2+} removal.

The $1/n$ values of four different adsorbents fall between 0 and 1 (Table 2), which indicate the favourable adsorption of Pb^{2+} on such adsorbents (Akafu et al., 2019). The Freundlich isotherm postulates that, in addition to the multilayer sorption of the adsorbate onto the heterogeneous surface, the stronger binding sites are first occupied and that the binding strength diminishes as the degree of site occupation increases.

The K_F (mg g^{-1})(mg $L^{-1})^n$ or (mg g^{-1}) (L $mg^{-1})^{1/n}$ present in the Equation 6 is referred to as the Freundlich coefficient which is a constant indicative of adsorption capacity of the adsorbent, and the dimensionless constant $1/n$ indicates the intensity of the adsorption or surface heterogeneity. Its value ranges between 0 and 1.

The adsorption capacity of the adsorbent for a specific adsorbate increases as the K_F value rises. Indicating multilayer sorption capacity, the plot of $\log q_e$ vs. $\log C_e$ yields a straight line with a slope of $1/n$ and the value of $\log K_F$ can be calculated from the intercept. However, as observed in Table 2, the values of the correlation coefficient of the Freundlich isotherm are lower than those of the Langmuir isotherm.

For the sorption of Pb^{2+} on four different types of adsorbents, the plots of $1/q_e$ vs.

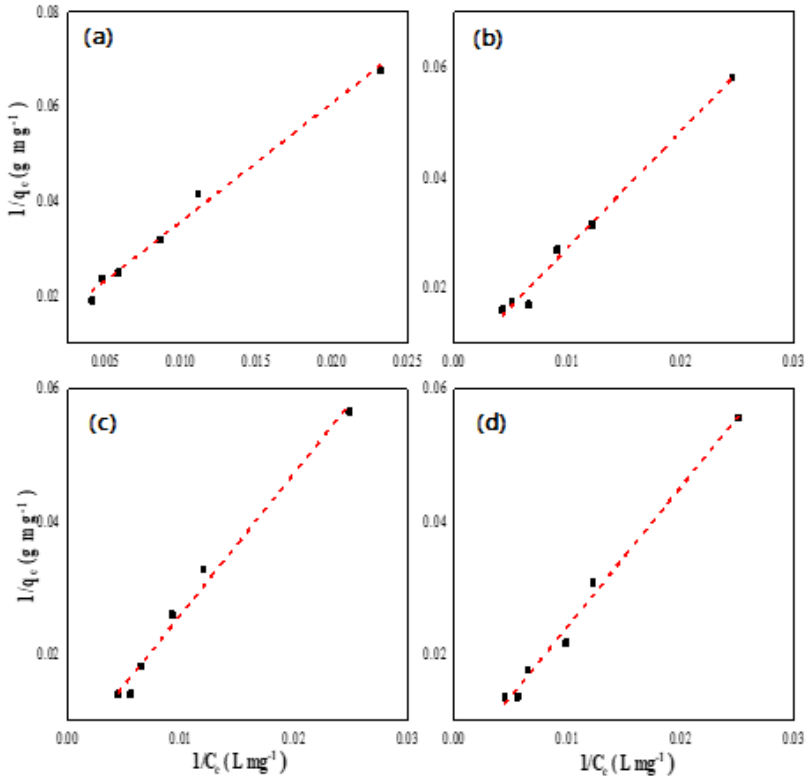

Figure 6. Langmuir isotherm fit of Pb^{2+} adsorption on (a) RDE (b) SDE (c) SDE-400 °and (d) (d) SDE-800 °C adsorbents.

Table 2. Isotherm model parameters for the adsorption of Pb^{2+} on to RDE, SDE, SDE-400 °C and SDE-800 °C adsorbents.

Isotherm		RDE	SDE	SDE-400 °C	SDE-800 °C
Langmuir	q_m(mg g^{-1})	92.59	163.93	208.33	322.58
	K_L (L mg^{-1})	0.0043	0.0029	0.0023	0.0015
	R^2	0.99	0.99	0.99	0.99
Freundlich	$1/n$	0.7101	0.758	0.8648	0.8616
	K_F (mg g^{-1}) (L $mg^{-1})^{1/n}$	1.0181	1.0839	0.6990	0.7684
	R^2	0.99	0.96	0.98	0.97

$1/C_e$ produced straight lines with excellent correlation coefficients, indicating a better fit of the data to the Langmuir isotherm than to the Freundlich isotherm (Figures 6 and 7). The stronger fit of the data to the Langmuir adsorption isotherm shows that the adsorption is monolayer and is based on the presumption that the adsorbent surface is composed of active sites with a homogeneous energy (Akafu et al., 2019). Freundlich isotherm models could not be utilized to explain the adsorption process of Pb^{2+} ions on four different types of adsorbents (RDE, SDE, SDE-400 °C, and SDE-800 °C), according to Table 2, which displays the calculated parameters for both the Langmuir and Freundlich isotherms.

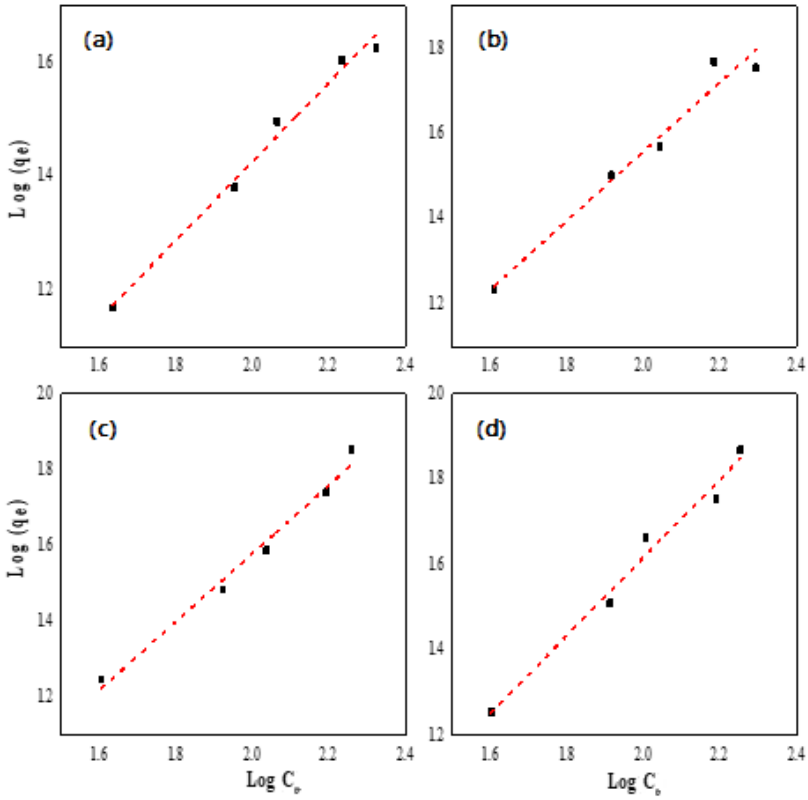

Figure 7. Freundlich isotherm fit of Pb^{2+} adsorption on (a) RDE (b) SDE (c) SDE-400 °C and (d) SDE-800 °C adsorbents.

Conclusion

This research work is focused on the investigation of Pb^{2+} removal using thermally regenerated diatomaceous earth (DE) formed from spent diatomaceous earth (SDE) at 400 and 800 °C due to the good performance, short time duration and mainly the utilisation of SDE as an adsorbent, prepared after thermal regeneration. Use of such biosorbents after thermal treatment has not been much reported.

Field emission scanning electron microscopy (FE-SEM) analysis of the surface morphology of SDE prior to adsorption studies shows a well-organized porous structure with some particles on the surface. Nitrogen adsorption-desorption analysis at -196 °C indicates that RDE has a higher specific surface area and total pore volume of 3.70 $m^2\ g^{-1}$ and 0.015 $cm^3\ g^{-1}$, respectively, as compared to SDE dry form, which have 2.22 $m^2\ g^{-1}$ and 0.010 $cm^3\ g^{-1}$, respectively.

Adsorption experiments conducted by varying each parameter while others being constant indicate that adsorbent dosage of 50 mg and contact time of 180 min at a pH of 4.8 were the optimal conditions for Pb^{2+} adsorption on SDE.

The Langmuir and Freundlich adsorption isotherm models used to analyse Pb^{2+} adsorption on RDE, SDE, SDE-400 °C, and SDE-800 °C indicate that the adsorption data fit well with the Langmuir model with a good correlation coefficient, concluding that

the adsorption is monolayer. The Langmuir isotherm suggests that the conditions were favourable for the adsorption of Pb^{2+} on those adsorbents as $0 < R_L < 1$. The maximum Pb^{2+} adsorption capacities on RDE, SDE, SDE-400 °C, and SDE-800 °C are 92.60, 163.93, 208.33, and 322.58 mg g^{-1}, respectively, according to the Langmuir model. Furthermore, having $1/n$ values ranging between 0 and 1 for four different adsorbents indicates the validity of the Freundlich isotherm as well.

Due to the existence of chemisorption, the study implies that SDE-800 °C is a better material than the other three adsorbents for use in the removal of Pb^{2+} from aqueous solution for the treatment of industrial wastewater. Two surface hydroxyls would release a water molecule and bridge when heated at a high temperature. The degradation of the organic structure, which increases porosity and facilitates pollutant removal would also facilitate adsorption, leading to chemisorption. As a result, the interaction between the adsorbate and the adsorbent surface occurred possibly via electrostatic attractions and H-bonds. In contrast, van der Waals forces between the adsorbate and the accessible pores on the adsorbent surface may cause physisorption to take place.

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