



Adsorption Study for the Removal of Cadmium and Lead from Synthetic Storm Water Using Fired Clay

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Abstract

This work focuses on treating stormwater containing excessive concentrations of Cadmium and Lead to produce acceptable water according to environmental specifications (WHO) and evaluating the performance of fired clay as a low-cost and environmentally friendly water treatment material. The studies were carried out to look into the Cadmium and Leads' room-temperature adsorption isotherm and the effects of various variables. The primary adsorbate concentration, contact time, agitation speed, pH, and mass of the adsorbent have all been studied. According to the results, the optimal experimental conditions pH=6, 0.45mg/L of Cadmium and 0.08 mg/L of Lead as adsorbate, 12 g of adsorbent, 250 rpm of agitation, and 2.0 hours of contact time -reduced Cadmium and Lead by 93 percent for fired clay. The experimental data were examined using the Langmuir and Frenlich isotherm models. The preferred model for Cadmium and Lead adsorption onto fired clay material, according to the results, was the Langmuir adsorption isotherm. This study shows that fired clay particles can be employed as a low-cost adsorbent to take Cadmium and Lead out of water.

Keywords: Stormwater, cadmium, lead, adsorption, fired clay.

الخلاصة:

يركز هذا العمل على معالجة مياه الأمطار التي تحتوي على تركيزات زائدة من الكاديوم والرصاص لإنتاج مياه مقبولة وفقاً للمواصفات البيئية (منظمة الصحة العالمية) وتقييم أداء الطين المحروق باعتباره مادة معالجة مياه منخفضة التكلفة وصديقة للبيئة. تم إجراء الدراسات للنظر في تساوي درجة حرارة الامتزاز للكاديوم والرصاص في درجة حرارة الغرفة وتأثيرات المتغيرات المختلفة. تمت دراسة تركيز المادة الممتزة الأولية، وزمن التلامس، وسرعة التحريض، ودرجة الحموضة، وكتلة المادة المازرة. وفقاً للنتائج، فإن الظروف التجريبية المثلى هي الرقم الهيدروجيني = 6، 0.45 ملغم / لتر من الكاديوم و 0.08 ملغم / لتر من الرصاص كمادة ممتزة، 12 غرام من الممتزات، 250 دورة في الدقيقة من التحريض، و 2.0 ساعة من وقت الاتصال - تقليل الكاديوم والرصاص بنسبة 93 بالمائة للطين المحروق. تم فحص البيانات التجريبية باستخدام نموذج لانجمير وفريدليش للأيسوثرم. النموذج المفضل لامتزاز الكاديوم والرصاص على مادة الطين المحروقة، وفقاً للنتائج، هو ايزوثرم امتزاز لانجمير. توضح هذه الدراسة أنه يمكن استخدام جزيئات الطين المحروقة كمادة ماصة منخفضة التكلفة لإخراج الكاديوم والرصاص من الماء.

1. INTRODUCTION

Because most cities have a lot of paved space and impermeable surfaces like roads and highways, most rainwater quickly enters major rivers during a rainstorm. When it rains, many kinds of contaminants are temporarily soaked up by impermeable surfaces like roads and highways and then carried into the nearby environment by stormwater runoff. Urban stormwater originates from a variety of sources, including direct rainfall, overflowing catchments, and drainages along its path [1].

The many toxins and pollutants that urban stormwater picks up from its sources may endanger the receiving water [2]. Pollutants commonly found in road runoff, parking lots, and activities involving traffic are heavy metals such as zinc (Zn), chromium (Cr), copper (Cu), lead (Pb), nickel (Ni), and Cadmium (Cd), which are mainly emitted

from lead cars [3]. The most dangerous long-term impacts on human health are caused by Cd, which can lead to choking, abdominal pain, anemia, renal failure, and diarrhea. The US EPA has also classified Cadmium as a carcinogen. Cd and Pb are categorized as non-essential heavy metals, which means they are capable of causing death or disease if ingested, even at extremely low doses.

Traffic-related activities, component erosion, fluid intrusion, pavement deterioration, road maintenance, and atmospheric deposition are the major contributing factors to the creation of these components [4]. Materials used in construction (such as copper and zinc from gutters and roof surfaces, or biocides from exterior paint), automobiles (such as benzothiazoles from tire abrasion), and combustion processes (such as polycyclic aromatic hydrocarbons (PAH), Deposition in the atmosphere (such as the herbicide terbuthylazine) or removal of trash (such as nicotine) from butts on cigarettes are a source of pollution of stormwater [5]. Metal is regarded as a potential pollutant, following dye, because it has a lasting effect, is lethal at low concentrations, and is non-biodegradable and permanent. In other words, it is not susceptible to microbial or chemical degradation. In addition to harming many different body organs, including the blood, brain, lungs, kidney, and liver, heavy metals can also affect the neurological system and cause disorders like Alzheimer's. Numerous experts have examined the harmful effects of certain metals. Therefore, it has become necessary to find an effective and inexpensive way to get rid of these toxic materials, purify water, and reduce their damage. Adsorption is frequently used to remove impurities, including metals, from raw water. Even though several methods, including electrochemical deposition, ion exchange, flotation, chemical precipitation, biological, oxidation, membrane, and coagulation, have advantages and disadvantages [6]. The adsorption method has the advantages of being straightforward to use, easily accessible to the adsorbent materials, and without the use of chemicals. Very little sludge is produced. For the heavy metal's adsorption, all-natural, several low-cost and effective adsorbents have been developed, including clay minerals, natural zeolite, tea leaves, tree bark, and fly ash. Clay minerals and natural clay, such as red soils and bentonite, have tiny particle sizes, porous architectures with sizable specific surfaces, and strong adsorption and cation exchange capabilities. Many adsorption investigations use natural clay minerals because they are widely available and reasonably priced [7]. The adsorption of the adsorbent uses batch treatment, continuous fixed bed (up flow or down flow), continuous moving bed, continuous fluidized bed, and pulsed bed. Other than batch and fixed bed, the remaining adsorption treatment methods have several drawbacks, including the need for a large space, high cost, requirement for a bigger volume of adsorbent, feed channeling, pre-required adsorbent storage, and non-uniform residence time.

2. METHODOLOGY

2.1 SYNTHETIC STORM WATER

Surface runoff samples taken from various watersheds in Wasit Governorate were found to contain heavy metals on multiple occasions. Wastewater effluent concentrations vary widely across different watersheds and are frequently detected at levels above local drinking water guidelines. It is obtained primarily from waste generated by related factories (metal, paint, and coating industries), buildings (especially metal roofs), deteriorating pavements, and traffic activities (e.g., tires, gasoline additives, brake pads, exhaust, wiring, fluid leaks). Several factors, such as degradation, road upkeep, surrounding land usage, sewage overflow, etc., may lead to the contamination of receiving water bodies and an increase in the presence of heavy metals there, necessitating the search for effective removal methods. As indicated in Table 1, in the current investigation, neutral distilled water was used to create synthetic stormwater, to which several heavy metals and organic compounds were added in the ratios with a purity of 99%.

Synthetic Stormwater Constituents	Concentration or Value	Chemical
Total Phosphorus (TP)	0.7 mg/L	Dibasic Sodium Phosphate (Na_2HPO_4)
Copper (Cu)	0.08 mg/L	Cupric Sulfate (CuSO_4)
Nitrate	2 mg/L	Sodium Nitrate (NaNO_3)
Organic Nitrogen (Org-N)	3.5 mg/L	Glycine ($\text{NH}_2\text{CH}_2\text{COOH}$)
Cadmium (Cd)	0.08 mg/L	Cadmium Chloride (CdCl_2)
Lead (Pb)	0.45 mg/L	Lead Chloride (PbCl_2)
pH	7	Sodium Hydroxide (NaOH)
Dissolved Organic Carbon (DOC)	14 mg/L	Humic Acid

Zinc (Zn)	0.6 mg/L	Zinc Chloride (ZnCl ₂)
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Table 1 Synthetic Stormwater Composition.

2.2. PROCEDURE OF EXPERIMENTS

2.2.1 Adsorbate and adsorbent

Due to the poor biodegradability, high toxicity, and accumulation of heavy metals in the food chain brought on by industrial and urban activity in the river, this issue is important on a worldwide scale. Among the most significant environmental pollutants associated with Cadmium are batteries, phosphate fertilizers, metal paints, pigments, and pigment stabilizers. Additional sources of lead pollution include smelters, mines, and the textile, paint, and metal-plating liquid waste industries [8]. Thus, in this study, the heavy metals examined were Lead (Pb) and Cadmium (Cd).

We used fired clay as an adsorbent in this study because of its advantages over other commercially available adsorbents, including its widespread availability, low cost, outstanding adsorption capabilities, non-toxic nature, high specific surface area, and strong ion exchange potential. These features have drawn the attention of professionals from all over the world to the use of clay materials, whether natural or modified, as adsorbents for the treatment of water. Particle shape, particle size distribution, moisture content, clay content, density between mineral contaminants and soil matrix, magnetic properties, soil matrix heterogeneity, and the particle surface's hydrophobic properties are some of the variables that affect the effectiveness of adsorption [9]. As seen in Figure 1, Fired Clay (FC) was acquired from the nearby market and subsequently processed and pulverized into fine particles in Wasit University's College of Engineering's Health and Environment Laboratory. The product was next sieved and put through an American mesh (with mesh sizes ranging from No. 30 to No. 200), as indicated in Figure 2, to create particles that were 0.6 microns in size. The particles were properly cleaned many times with filtered water to remove impurities. Next, as indicated in Figure 2, allow it to dry in the sun for a full day.

Following two hours of drying at a temperature below 110 degrees Celsius, the product was stored with the clay particles in a plastic container, as shown in Figure 3.



Fig.1 Images of fired clay used as an adsorbent.



Fig.2 Clay particles after sifting, washing, and drying in the sun.



Fig.3 Clay particles after drying them in the oven and keeping them in a plastic container.

2.2.2. Batch experiments procedure

Adsorption tests were carried out in the current study using a batch system in a 250-ml flask on a magnetic agitator at a speed of 250 rpm. Many variables, including adsorbent quantity, detention time, pH, and primary dosage of Lead and Cadmium, were taken into consideration to control experiments in different conditions. At 2.5 hours of contact time, the initial lead concentrations and Cadmium were 0.45 mg/l and 0.08 mg/l, respectively. The average was dependent, and the whole examination was repeated three times. The mass of Pb and Cd in the filtrate was measured using a spectrophotometer (Spectro Direct model Lovibond, Germany) at a wavelength of 220 nm after each test for both adsorbents by spreading the solution on Whitman 42 filter paper to separate adsorbing particles from the solution. The lab explores the effects of several parameters while measuring the exchange parameter and maintaining the values of other parameters. By selecting the optimum values after each trial, further research into the effects of the other factors will be done.

2.3. ADSORPTION ISOTHERM

To describe the process of adsorption and related mechanisms, isotherm models are typically examined [10]. Freundlich and Langmuir are two extensively employed isotherm models. The Langmuir isotherm is often employed to express laboratory results from earlier investigations. This model's linear form resembles Equation 1 [11-13]:

$$\frac{1}{q_e} = \frac{1}{C_e k_L q_{max}} + \frac{1}{q_{max}} \quad (1)$$

q_e represents the equilibrium adsorbed ion per gram of adsorbent, and C_e represents the concentration of metal ions in equilibrium (mg/L). q_{max} and k_L are the energy of adsorption (l/g) and the capacity of surface adsorption (mg/g), respectively, which are the constants of the Langmuir model. The values are obtained by calculating the slope and intercept of the linear Langmuir equation in C_e/q_e versus C_e diagram.

It is expressed using the following Equation 2:

$$q_e = \frac{C_i - C_e}{m} \times V \quad (2)$$

Where V represents the solution volume in liters, m represents the adsorbent mass in grams, and C_i is the primary (Cd, Pb) mass in mg/l. After adsorption in terms of mg/l, C_e represents the equilibrium mass of the adsorbed substance in the liquid.

Using Equation 3, the Freundlich formula can be represented in linear form [14].

$$\log q_e = \log K_F + \frac{1}{n} \times C_e \quad (3)$$

In Equation 3, q_e represents the amount to be absorbed in the equilibrium concentration of mg/l, C_e represents the absorbed mass equilibrium concentration of mg/l, and K_F and n are Freundlich coefficients discovered by plotting $\log q_e$ versus $\log C_e$. They were found by drawing a straight line with a gradient of and an intercept of $\log K_F$.

3. RESULTS AND DISCUSSION

3.1. POINT OF ZERO CHARGE, PZC

The samples were placed in cone-shaped containers with 0.5 g of baked clay, 60 ml of purified water, and 40 ml of a 0.1 mol/l NaCl solution. The mixture was then staggered for 24 hours. Using NaCl or HCl, the primary pH (2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, and 13) was changed. The difference between the primary pH rates and the terminal pH rates was obtained after 24 hours, as shown in Figures 4 and 5. The terminal pH is then determined. The graph produced by plotting pH against primary pH allows for the detection of the optimal pH from the interruption point of zero [15]. According to Figures 4 and 5, the pHPZC for fired clay is 6.

3.2. EFFECT OF PH

Since pH impacts the solubility of metal ions, it is well-known that this parameter is important in the process of removing heavy metal ions from an aqueous solution. We looked at how pH affected the cadmium removal and lead ions using the pH values (13, 12, 11, 10, 9, 8, 7, 6, 5, 4, 3, and 2), which represent acidic, neutral, and alkaline conditions on the lead and cadmium adsorption by fired clay as an adsorbent. In Figures 6 and 7, the outcomes are displayed. The control conditions were lead concentration 0.45 mg/L and cadmium concentration 0.08 mg/L, water sample volume 100 ml, temperature 25 °C, vibration speed 200 rpm, sorbent dose 12 g, and contact time 2 hours. It was demonstrated that Cadmium's removal efficiency and Lead increased from pH 6 to pH 8, with pH 6 having the highest removal efficiency. This result is consistent with prior studies of lead and cadmium adsorption that Ozer et al. carried out to absorb cadmium sugar beet pulp [16], which found that the optimal pH was 6.3. Furthermore, Venkobachar and Bhattacharya identified the optimal pH. For the cadmium removal from Gredia coal, it was 6.6 [17]. pH 6 is the optimum value, according to Talip et al. [18], for perlite to efficiently absorb cadmium. Mohsen A. Hashem showed that the optimum pH for lead removal on okra waste was 5, close to the present result [19], and the optimum pH for lead adsorption on activated carbon was 6 [20].

Figure 6 shows that at 20 °C, the rise in pH from 6 to 8 increases the efficiency of removal by approximately 77% to 72.5%, after which the efficiency of removal drops from 72.5% to 58.75% as the pH increases from 8 to 13. Figure 7 also shows the highest lead adsorption efficiency that occurs when the pH increases from 6 to 8, as the highest lead removal efficiency reached 80% at pH 6.

This decreasing behavior of cadmium and lead removals at very low pH is owing to the larger concentration of H⁺ ions existing in the solution competing with Cd²⁺ and Pb²⁺ for the adsorption sites of the fired clay, leaving free Cd and Lead ions in the solution. This means that at the top with the H⁺ ions concentration, the fired clay granules become more positively charged. Consequently, the attraction of cadmium and lead cations to the surface of the fired clay is lessened.

As pH increases, fewer H⁺ ions are present in the solution. Therefore, there is a rise in the elimination of cadmium ions in the pH range of 6 to 8. Therefore, this will not allow competition with cadmium and lead ions for adsorption by the fired clay, and thus, the surface of the fired clay granules becomes more negatively charged, attracting positive charges and, thus, higher adsorption.

Then, the ability of the fired clay granules to absorb cadmium and lead ions at high pH values from (9 to 13) begins to decrease. The reason for the decrease is because of the hydrolysis of Cd (OH)₃ and Pb (OH)₃ to Cd (OH)₂ and Pb (OH)₂ that precipitates in the solution.

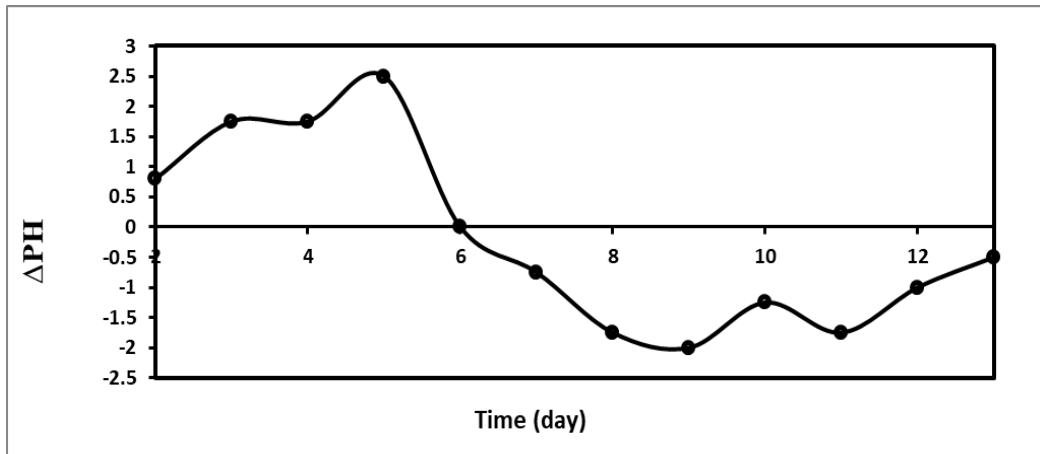


Fig.4 Point of zero charge of Cadmium.

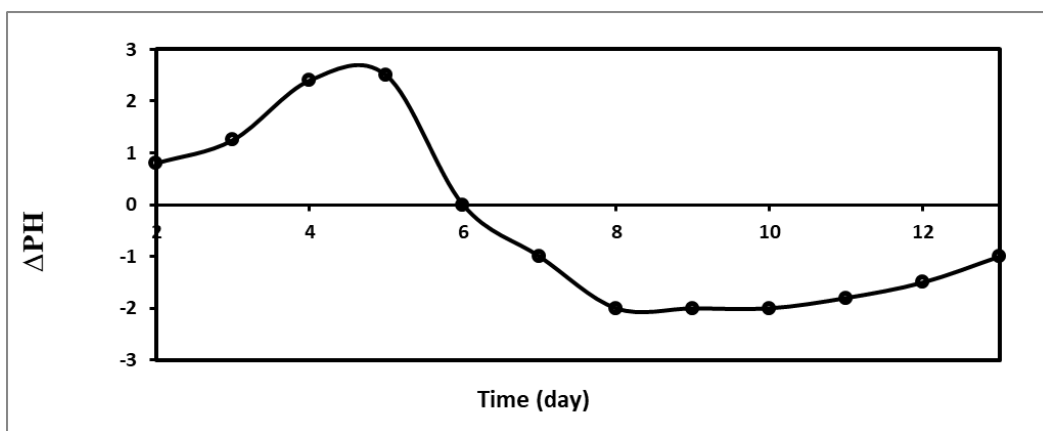


Fig. 5 Point of zero charge of Lead.

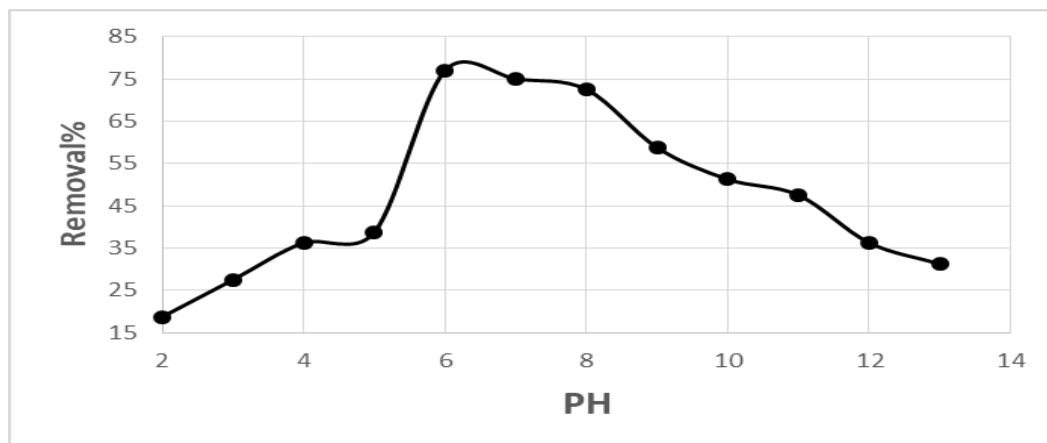


Fig. 6 Cadmium ions adsorbed onto fired clay particles at 20°C at different pH.

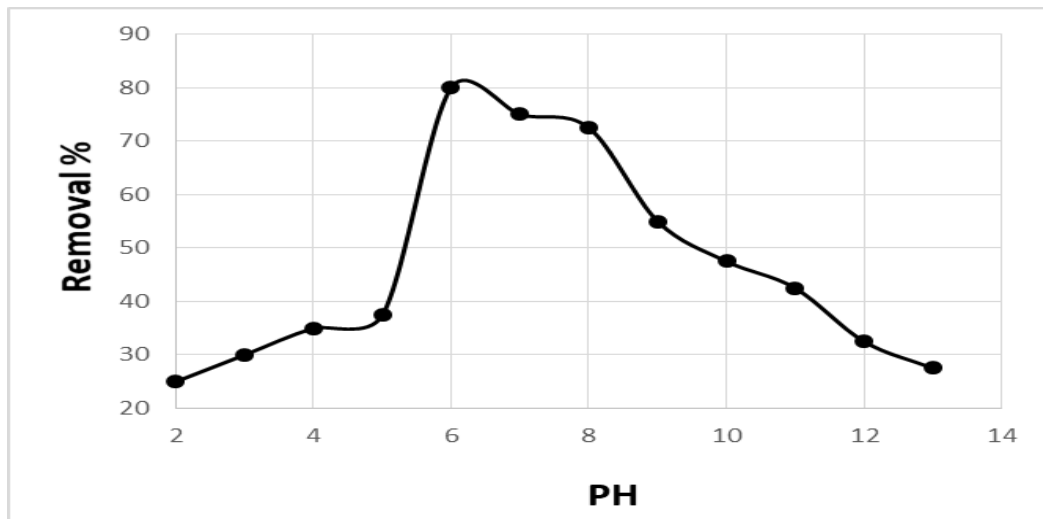


Fig. 7 Lead ions adsorbed onto fired clay particles at 20°C at different pH.

3.3. EFFECT OF AGITATION SPEED

In order to extend the time that Cadmium and Lead ions interacted with the adsorption site on the adsorbent, several agitation speeds were investigated. While keeping all other variables constant, agitation speeds of 100, 125, 150, 175, 200, 225, 250, 275, and 300 rpm were utilized to evaluate the effect of agitation speed on the effectiveness of removing cadmium and lead ions. The removal effectiveness of cadmium ions adsorbed onto fired clay (FC) particles is shown in Figure 8 at various agitation speeds, while the removal effectiveness of lead ions adsorbed onto fired clay (FC) particles is shown at various agitation speeds in Figure 9. By increasing the agitation speed to 200 rpm, Cadmium and lead ions were effectively removed. This might be because it was certain that layer thickness would decrease as speed increases.

The decline in performance removal after 200 rpm can be explained by the ratio of fired clay adhering to the inner surface of the volumetric flask during the mixing phase, which reduces the adsorption of cadmium and lead ions. As a result, 200 rpm will remain the same for every experiment moving forward because it is the best setting for getting the best Cadmium and lead removal.

3.4. EFFECT OF ADSORBENT DOSE

In this investigation, several adsorbent dosages of 2, 4, 6, 8, 10, and 12 have been used to examine their impact on the adsorption process and to achieve the equilibrium isotherm curve while maintaining other parameters constant. The impact of the dose of fired clay particles on the initiative's ability to remove Cadmium is depicted in Figure 10 with a cadmium dose of 0.08 mg/L, pH 6, and 10 hours of contact time. With an adsorbent dose of 12 g/100 mL, the removal efficacy of cadmium ions rose from 21% with an adsorbent dose of 2 g/100 mL to 78%. The capacity of adsorption (q_e), however, was seen to decline from 0.84 to 0.52 mg/g.

At a dose of 0.45 mg/L, a pH of 6, and a contact time of 10 hours, Figure 11 illustrates the outcome of the dose of fired clay particles on lead removal as the removal percentage increased from 23% at an adsorbent dose of 2 g/100 ml to 80% at an adsorbent dose of 12 g/100 ml and the adsorption capacity (q_e) decreased from 5.2 to 3 mg/L.

The reason that cadmium and lead adsorption increase with increased adsorbent mass may be due to the enormous number of effective sites for adsorbing cadmium and Lead. In order to achieve the greatest performance in terms of eliminating cadmium and Lead, 12 g/100 ml of fired clay was sufficient, and this quantity will be fixed for subsequent investigations.

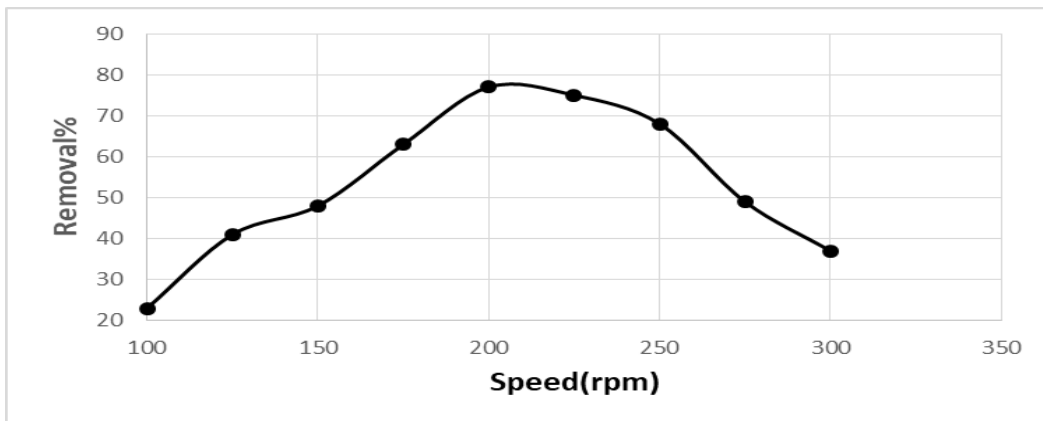


Fig.8 Cadmium ions adsorbed onto fired clay particles at 20°C at different rpm.

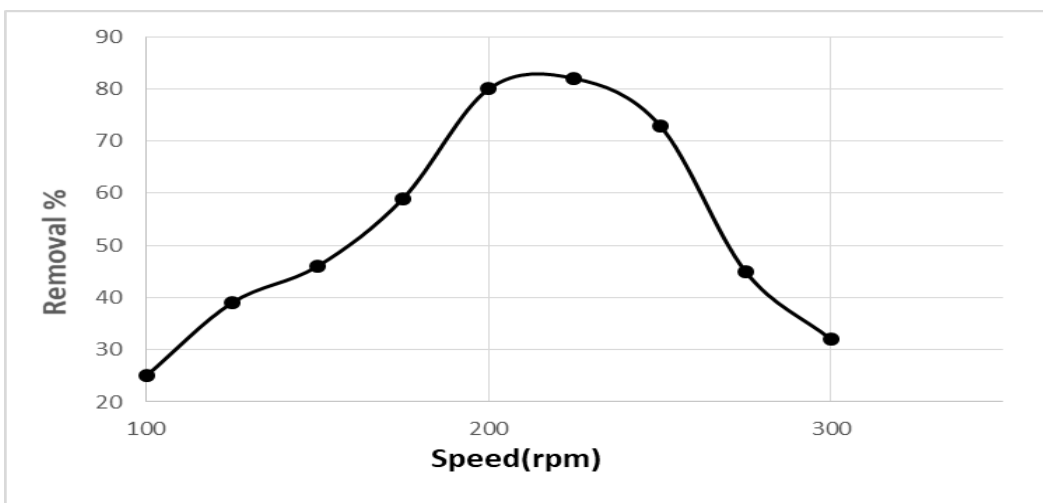


Fig. 9 Lead ions adsorbed onto fired clay particles at 20°C at different rpm.

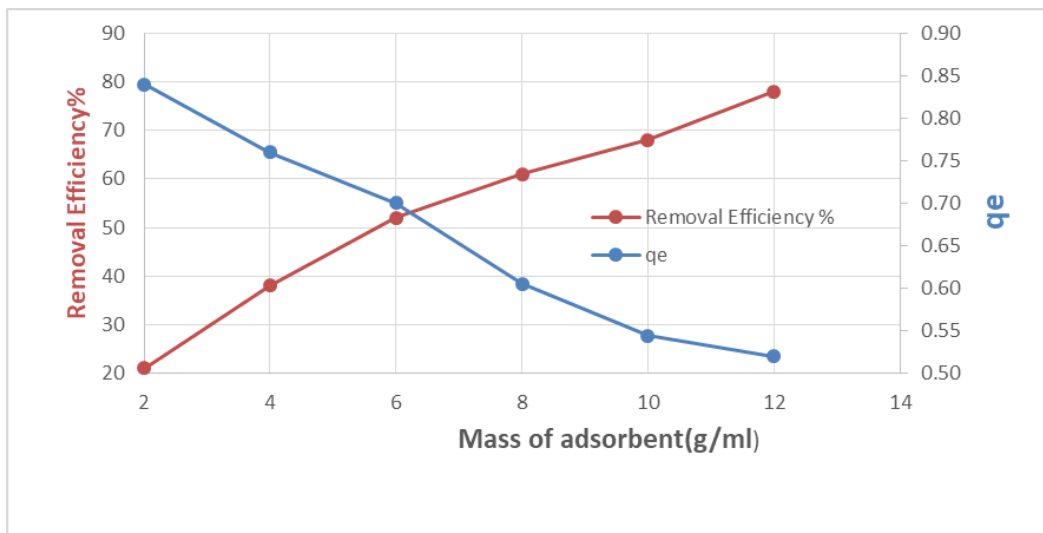


Fig. 10 Effect of fired clay particles dose on Cadmium removal (pH 6, initial Cadmium concentration= 0.08 mg/L. C.T=2.0 hrs, agitation speed).

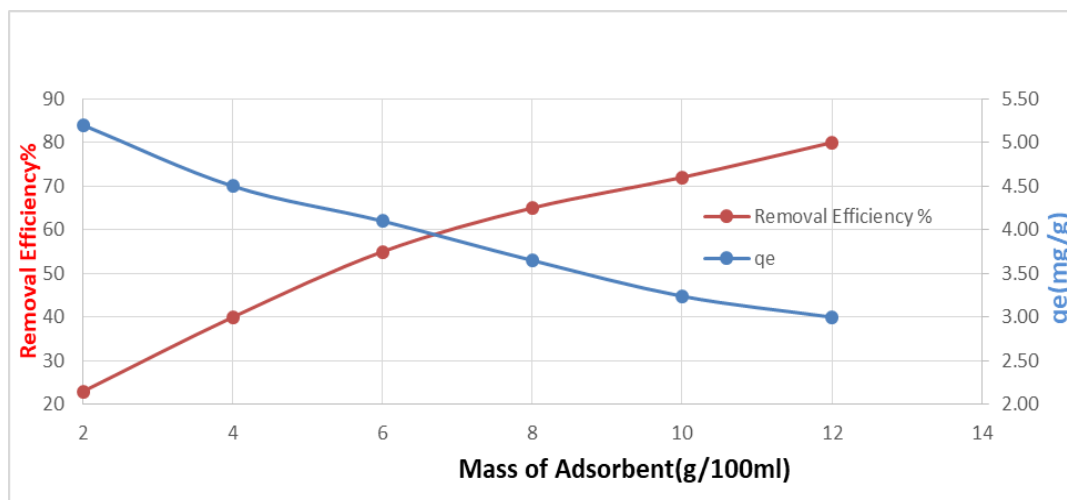


Fig. 11 Effect of fired clay particles dose on Lead removal (pH 6, initial Lead concentration= 0.45 mg/L. C.T=2.0 hrs, agitation speed).

3.5. INFLUENCE OF CONTACT TIME

Using the ideal conditions (velocity = 200 rpm, contact time = 1, 1.5, 2, 2.5, 3, 3.5, 4, 4.5, and 5 h, dosage = 12 g/100 ml, temperature = 25 °C, and pH 6, Lead and Cadmium concentrations 0.45 mg/L and 0.08 mg/L, respectively), the effect of contact time on the adsorption process was examined. The ideal contact time was determined to be the equilibrium time or the point at which no more Cadmium and Lead ions could be eliminated. Figures 12 and 13 depict the impact of the impaction period on the reduction of Cadmium and Lead, respectively. With longer contact times, more cadmium and lead ions are removed. Cadmium and Lead were quickly removed by the fired clays during the first half hour, with removal rates of about 77% for Cadmium and 80% for Lead. In addition to the initial height, this behavior referred to the significant amount of vacant cavity sites on the surface of the fired clay particles. This performance gradually grew to a maximum removal efficiency of 89% for Cadmium and 92% for Lead at the contact time of 2.0 hours, and then the adsorption performance slightly decreased after 2 hours, indicating that the adsorption process started to slow down with time because the active sites on the fired clay are filled with the capacitor.

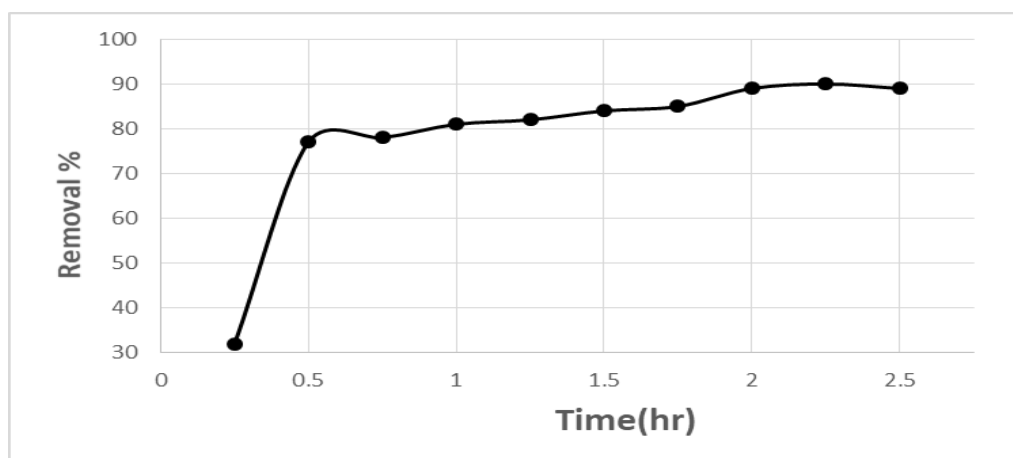


Fig.12 Effect of contact time of fired clay on Cadmium removal.

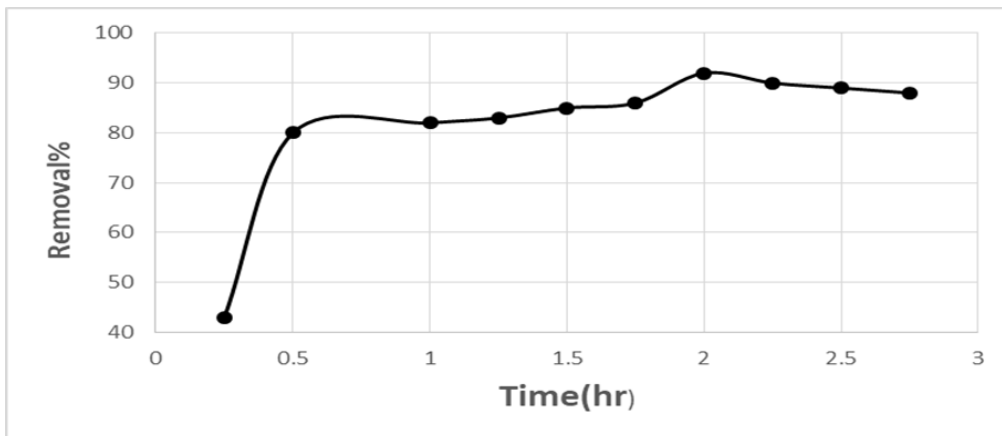


Fig.13 Effect of contact time of fired clay on lead removal.

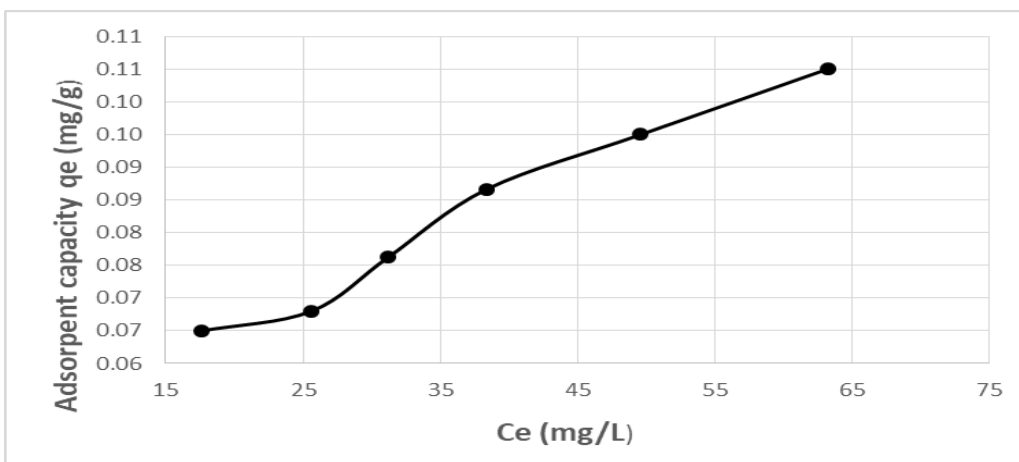


Fig.14 Adsorption isotherm of Cadmium ions onto fired clay particles at pH=6 and 20°C.

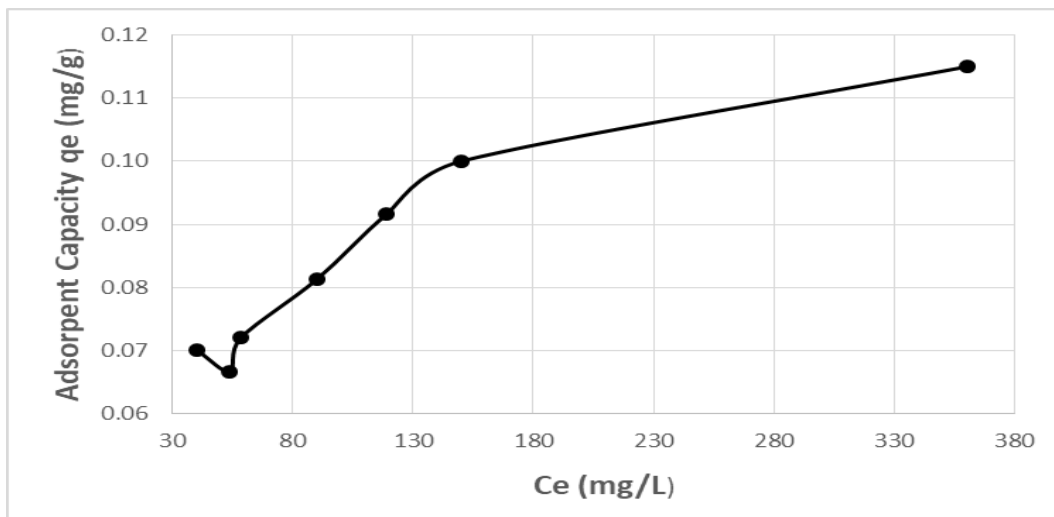


Fig.15 Adsorption isotherm of Lead ions onto fired clay particles at pH=6 and 20°C.

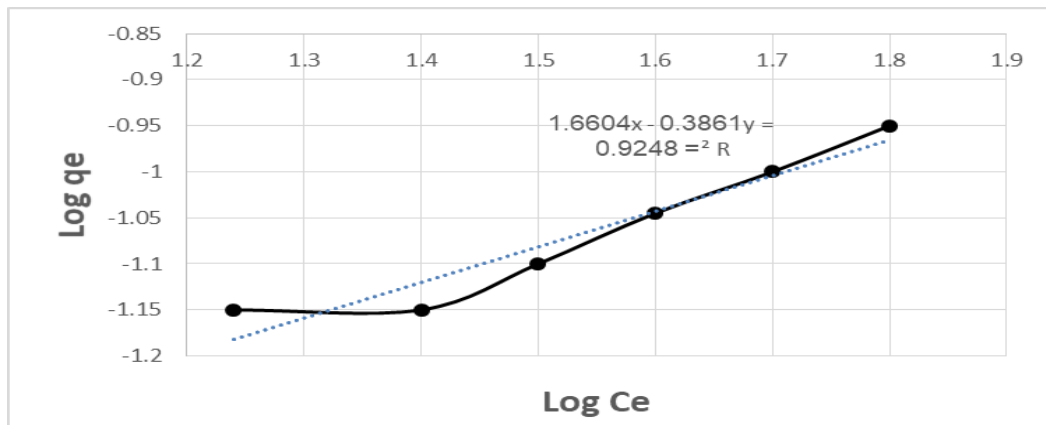


Fig. 16 Freundlich isotherm for Cadmium.

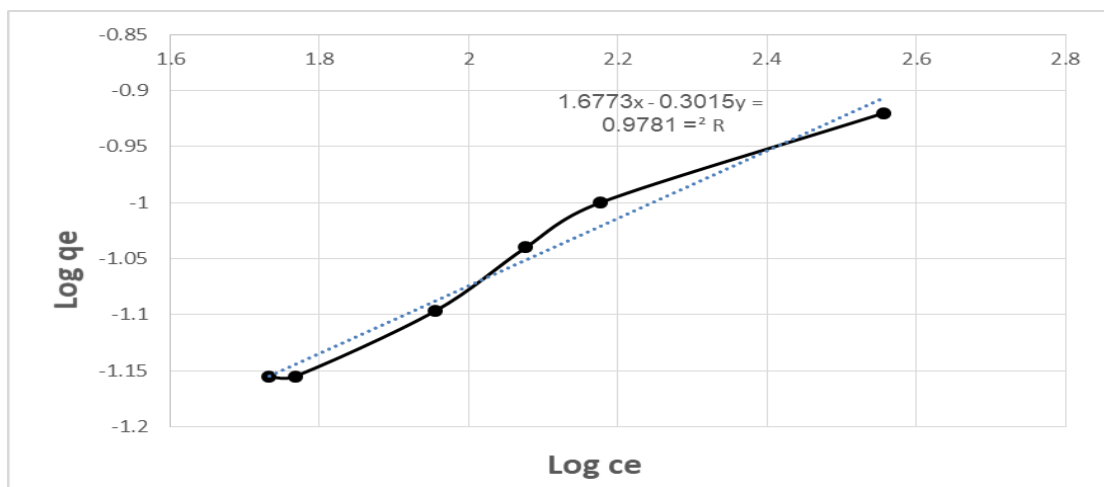


Fig.17 Freundlich isotherm for Lead.

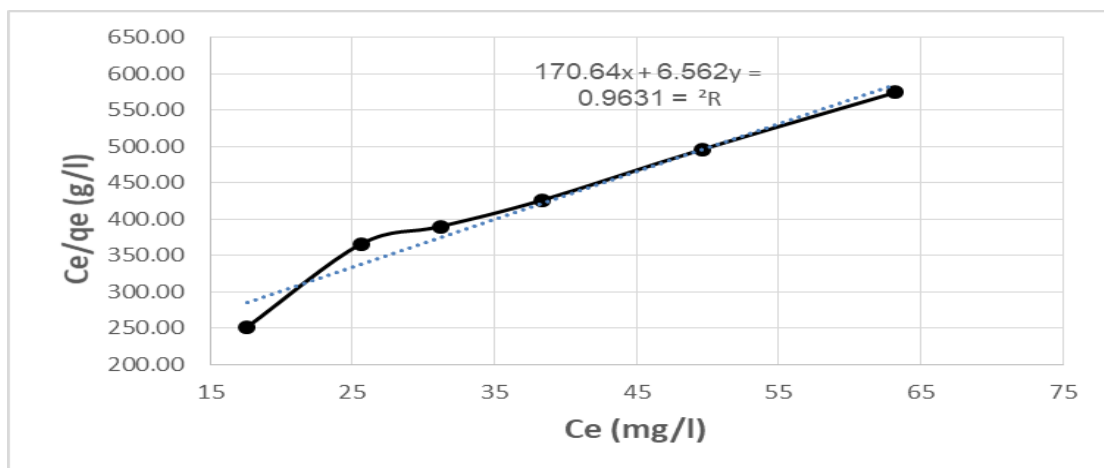


Fig. 18 Langmuir isotherm for Cadmium.

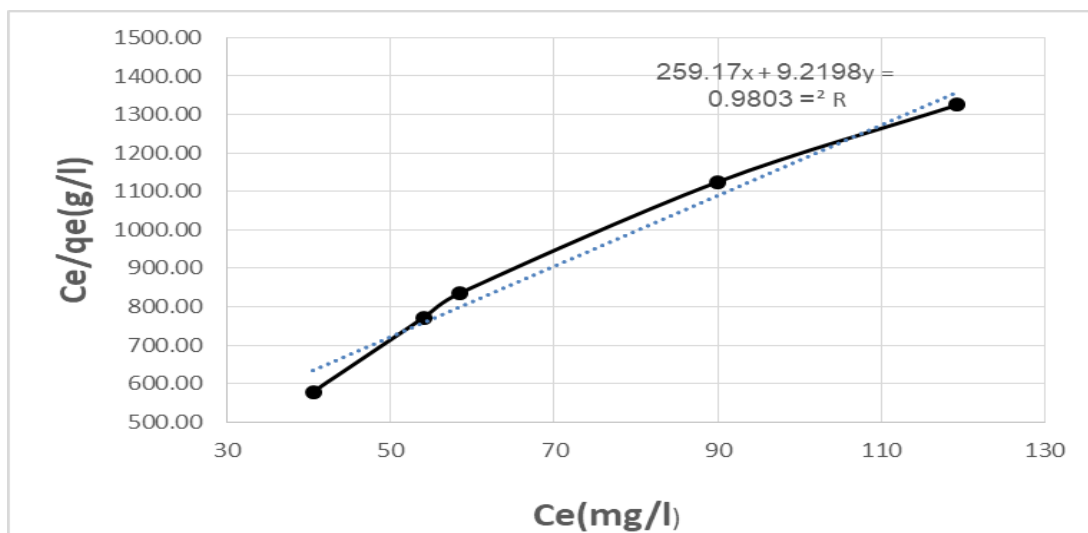


Fig. 19 Langmuir isotherm for Lead.

Langmuir isotherm		Freundlich isotherm	
q_{max}	0.1524	n	2.59
K_L	0.03845	K_F	0.02264
R_2	0.9631	R^2	0.9248

Table 2 Langmuir and Freundlich isotherm constants for Cadmium.

Langmuir isotherm		Freundlich isotherm	
q_{max}	0.10846	n	3.3167
K_L	0.03557	K_F	0.02038
R_2	0.9803	R^2	0.9781

Table 3 Langmuir and Freundlich isotherm constants for Lead.

The fashion adsorption models benefited from the equilibrium information. The Freundlich and Langmuir models, respectively, offer a superior fit to the adsorption data, with R_2 values for Cadmium of 0.9631 and 0.9248 and R^2 values for Lead of 0.9803 and 0.9781. Because the adsorption intensity values ($1/n = 0.386$) and ($1/n = 0.3015$) were found to be smaller than unity, the adsorption processes of Cadmium and Lead are regarded as beneficial.

4. CONCLUSIONS

In this investigation, fired clay was utilized to explore the impacts of adsorbent mass, contact time, pH, initial Cd and Pb content, and adsorption isotherms on the reduction of Cd and Pb via adsorption. The results showed that fired clay might be used as an adsorbent to remove Cd and Pb from synthetic stormwater. The elimination of lead and cadmium increases with longer contact times. With removal rates of roughly 77% for Cadmium and 80% for Lead, the first 0.5 hours saw the most elimination.

At a contact duration of 2.0 hours, this performance increased steadily to a maximum removal efficiency of 93% for Cadmium and 92% for Lead. Thus, it was determined that 2.0 hours was the ideal contact time. The ideal pH for firing clay to remove Cadmium and Lead was 6. As expected, the reduction of Cd and Pb increased as the amount of fired clay rose. The best adsorbent mass was determined to be 12 g of fired clay.

It becomes clear that Cd and Pb reduction by fired clay is a physical fact as the Cd and Pb reduction percentage increases with an increase in the starting Cd and Pb concentration for fired clay adsorbent. The adsorbed amounts of Cadmium ($R_2 = 0.96$) and Lead ($R_2 = 0.98$) satisfy the Freundlich isotherm, demonstrating that the adsorbed

amounts comply with the Freundlich standard. The ideal parameters to yield the maximum adsorption capacity are 12 g of adsorbent material, a collision length of 2.0 hours, primary concentrations of Cd 0.45 mg and Pb 0.08 mg in 100 ml of water, a pH of 6.0, agitation speed 250 rpm, and a Cd and Pb reduction percentage of 93%.

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