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Brick Powder, an Effectual Adsorbent for the Exclusion of Cr(VI) Ion from Aqueous Solution

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Abstract

The adsorption efficiency of finely grinded brick powder on the exclusion of chromium (VI) from the aqueous solution using various experimental situations was successfully assessed. Batch adsorption studies demonstrated the uptake of chromium ions at an assortment of different pH values, adsorbent dosages, contact time and initial concentration of metal ion. Contact time for maximum adsorption required was 120 min at pH 2 with agitation speed 160 rpm and 1 g was optimum dose for adsorption. The maximum adsorption capacity (Q_m) of brick powder was found 82.64 mg/g. The investigational data were snug well with Freundlich isotherm as compared to Langmuir isotherm and Temkin isotherm models. Freundlich model showed heterogeneous multilayer adsorption on brick powder. Freundlich constant n value was found to be 1.35, indicating a heterogeneous surface and favors a physical process of adsorption. Positive Tempkin constant value signifies an exothermic process where adsorption is favored by lower temperatures and heat is released as molecules attach to the surface. The Tempkin constant value < 20 kJ/mol is also evidence of physical adsorption.

Keywords: *Brick powder, chromium, Freundlich isotherm, Tempkin isotherm*

1. Introduction

Water is one of the essential substances for all kinds of life. It should be free from pollution. Water pollution is mainly caused by anthropogenic sources such as industrial activities, domestic effluents. Heavy metals are considered to be one of the major pollutant that contribute for water pollution. Metals which have specific weight usually more than 5.0 g/cm^3 are considered as heavy metals (Garbarino et al., 1995). Some heavy metals such as Cr, Cd, Ni, Cu, Zn, Fe, As arehazardous to environment as well as for living beings. Among these heavy metals, chromium is one of the hazardous metals as reported in literature (Jeyaseelan et al., 2015). Chromium has been extensively applied in industries like electroplating, metal finishing, magnetic tapes, pigments, leather tanning, brass, electrical equipment, catalysis. The effluent from these industries makes the environment contaminated (Mohan et al., 2005).

Chromium occurs in different oxidation states such as 2+, 3+ & 6+. Chromium has both beneficial and detrimental properties. Cr(Ш) is an essential trace element in mammalian metabolism, reducing blood glucose levels, reducing blood cholesterol levels by diminishing the concentration of low density lipoproteins in blood (Dionex, 1996). Cr(VI) compounds are more toxic than Cr(Ш) due to water solubility and mobility. Acute exposure to Cr(VI) causes respiratory, hepatic, kidney and derma problems.. The tolerance of the Cr(VI) to discharge into land surface water 0.1mg/L and in potable water is 0.05 mg/L (Doyurum et al., 2006).

Several treatment technologies have been developed to remove chromium from water and wastewater (Demirbas et al., 2004, Bansal et al., 2005). Among these strategies, adsorption is one of the straightforward fetched successful strategies of the expulsion of chromium from water/waste water containing moderately high concentration (around 100 mg/L) . Using inexpensive materials to adsorb pollutants can be a cheap way to get rid of harmful heavy metals from wastewater. Different types of solid waste, farm waste, and other products can attract and hold heavy metals. Activated carbons are commonly used materials that help to clean polluted water (Bansal et al., 2005) In this study, we tried to remove a heavy metal Cr(VI) from water using brick powder (BP) because it is affordable and works well. Brick powder is a common building material made up of clay, is abundant locally. It contains alumina, silica, lime, oxide of irons, magnesia. Several studies have been conducted to investigate the potential of using crushed clay brick as an adsorbent material (Hemalatha et al., 2014). The literature reported the advantage of using brick powder as adsorbent due to high porosity, high adsorption rate and variation in quality. Excellently powdered clay reserves and oxides exhibited enormous surface area. Clay minerals and oxides adsorb cationic, anionic and neutral species. Though the brick powder showed good adsorption behavior, it has also been reported that the negatively charged surface of brick powder is not very complementary for direct sorption of negatively charged Cr(VI) species.

It is suggested that reduction of Cr(VI) to Cr(III) is important for attracting charges by coulombic attraction. Cr(VI) can be changed into a less harmful form by various substances found in brick clay, like organic matter and minerals that have iron (Fe) in a certain state. Tartaric acid, a natural substance found in soil, can also change chromium from a harmful form (Cr(VI)) to a less harmful form (Cr(III)). . It has also been reported that tartaric acid which is biological reducing agent commonly present in soil is able to reduce Cr(VI) to Cr(III), which is catalyzed by substances present in clay such as TiO2, γ- Al2O3, α- FeOOH (Sontheimer et al., 1988).Therefore, the maximum removal observed is probably due to presence of reducing agents such as organic material containing carboxylic acid functional group and Fe^{2+} ions, which reduce Cr(VI) to Cr(III) followed by adsorption (Priyantha et al., 2011, Negi et al., 1985). Adsorption also influenced by different parameters such as initial concentration of the metal ion, contact time, amount of adsorbent dose, pH of the solution. Present study motivated on brick powder which is considered as low cost ecofriendly material as adsorbent for the exclusion of Cr from water. Adsorption efficiency of brick powder has been studied by applying Langmuir, Freundlich and Tempkin isotherm models.

2. Materials and methods

2.1 Reagents

Analytical reagent grade chemicals have been used in this study. HNO_3, H_2SO_4 used were purchased from Qualigens. $K_2Cr_2O_7$ (99.9%) was purchased from Merck company. DPC (diphenyl carbazide, AR) was obtained from S.D. Fine Company. Brick was collected from local brick industry of Bhaktapur, Nepal.

2.2 Preparation of adsorbent

The brick pieces were collected, washed and dried. Then it was ground to fine powder. It was sopping in distilled water for 24 h and again splashed with distilled water for 4/5 times. Again, it was dried in an oven and sieved by using 0.6 mm sieve.

2.3 Preparation of calibration curve

First of all, λ_{max}, was determined. Then, different calibration standards have been prepared from stock Cr(VI) solution. 1 mL of 5N H_2SO_4 and 1mL of 0.25 % of DPC solution was added in all solutions. The red-violet colored complex of Cr(VI)-DPC was obtained. Then the absorbance of all the solutions were noted using spectrophotometer. Thus obtained absorption data was plotted against concentration of Cr(VI) solution to obtain calibration curve.

2.4 Adsorption study

The adsorption of contaminant onto different adsorbents may be exaggerated by diverse factors such as contact time, pH, initial concentration and quantity of adsorbent dose. The amount of adsorbate loading on adsorbent can be calculated by following **equation (1)**.

$$q_{eq} = \frac{Ci - C_e}{W} \times V \text{ --------------------- (1)}$$

where, C_i & C_e are the initial & equilibrium concentrations of the adsorbate in mg/L respectively, V is the volume of solution in litre and W is weight of adsorbent in g.

The removal percentage or removal efficiency is calculated by using the **equation (2)**.

$$\text{Removal percentage (\%)} = \frac{\boldsymbol{C_i - C_e}}{\boldsymbol{Ci}} \times 100\% \text{ .... (2)}$$

Most of chemicals used in this study were freshly prepared. The pH of the solution was attuned by using 0.1M HCl and 0.1M NaOH solutions. A 25 mL of Cr(VI) with known concentration was taken in 150 mL adsorption bottle and 1 g of adsorbent was added and was stirred for 2 hrs at room temperature, the equilibrium was established. Thus obtained equilibrated solution was filtered and filtrate was used to find the equilibrium concentration of the chromium ions. The equilibrium concentration was determined by spectrophotometer following DPC method. The amount of chromium ions loaded on brick powder was finally intended using **equation (1)**.

3. Results and Discussion

3.1 Preparation of calibration curve

The λ_{max} was obtained at 540 nm which is matched with the reported values (Singh et al., 2013).The calibration curve was plotted and is shown in **Fig.1**. It was found to be linear and obeying Beer-Lambert's law.

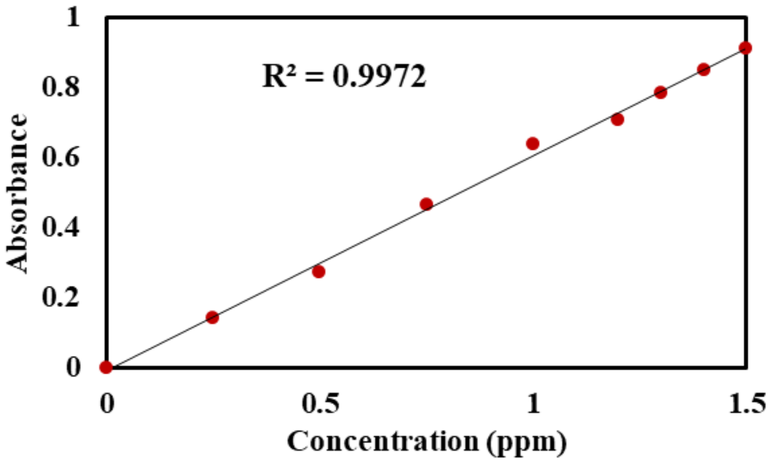

Fig. 1: Calibratiuon curve for the determination of Cr(VI).

3.2 Optimization of operational parameters on adsorption of Cr (VI) ions on brick powder

3.2.1 pH

The pH is one of the imperative causes which distress the adsorption process of heavy metals in water samples. Experimentations were performed utilizing different initial pH values such as 1, 2, 3, 4, 5, 6, 7, 8, 9 &10 to catch the optimum pH on the adsorption of Cr (VI) ions onto brick powder. The experimental parameters such as the amount of dose (1 g), concentration of the ions (5 mg/L), agitation speed (160 rpm) and contact time (2 hr) were retained. The consequence of pH was shown in **Fig. 2**. It clearly showed that the removal efficiency of Cr(VI) ion from the stock solution by the brick powder was definitely pH dependent. The optimum pH was found to be 2 and maximum removal percentage was 67% at pH 2. At low pH condition, the surface of the adsorbent was encircled by hydronium ions that participate with Cr ions. These hydronium ions obviously will prevent metal ions from nearing the binding sites on the adsorbent, resulting in decrease in exclusion of metal ions (Hemalatha et al., 2014). Since Cr(VI) exists in the form of oxyanions such as $HCrO_4^-$, $Cr_2O_7^{2-}$, CrO_4^{2-} etc in acidic medium and the lowering of pH caused the surface of the adsorbent to be protonated to a higher extent and as a result a strong attraction exist between these oxyanions of Cr(VI) and positively charge superficial of the adsorbent. Hence, the uptake upsurges with the diminution of pH value of the solution.

On the other hand, at higher pH 5-10, the negatively charged sites increases and this could boost the repulsion forces prevailing between the sorbent surface and Cr(VI). This is due to the creation of $Cr(OH)_3$ precipitate respectively which decrease adsorption efficiency.

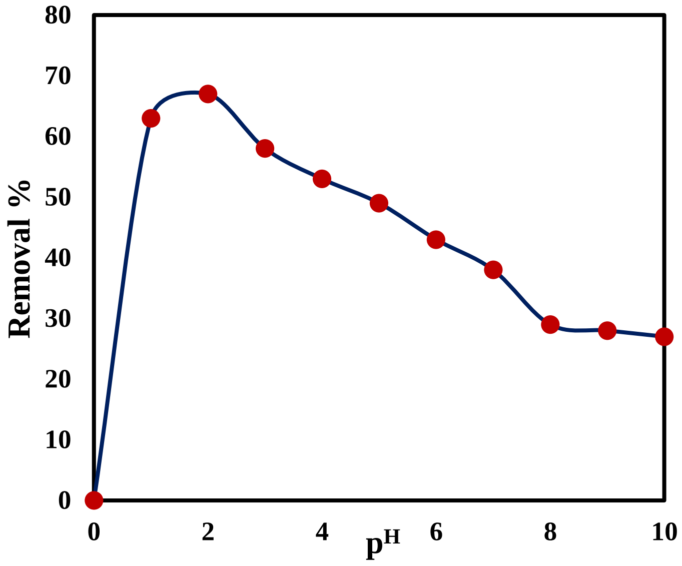

Fig. 2: Percentage removal of Cr(VI) as a function of pH.

3.2.2 Adsorbent dose

The consequence of adsorbent dose on the exclusion of Cr(VI) was studied at pH 2 by varying the adsorbent amounts ranging from 0.25-1.5 g were used. For this experiments, initial concentration of Cr(VI) (5 mg/L), agitation speed (160 rpm) and contact time (2 hr) was maintained. The result revealed is expressed in graph as shown in **Fig. 3.** The curve indicates that when the brick powder dose amplified from 0.25 to 1.5 g, the Chromium adsorption also found to be increased and then little variation was observed with further increment. The initial increment with intensification in adsorbent dose was due to upsurge in binding spots. It is reasonable that with higher the adsorbent dose, greater would be the number of exchangeable sites for chromium ions (Daneshvar et al., 2002). The results revealed that the optimum adsorbent dose was established to be 1 g.

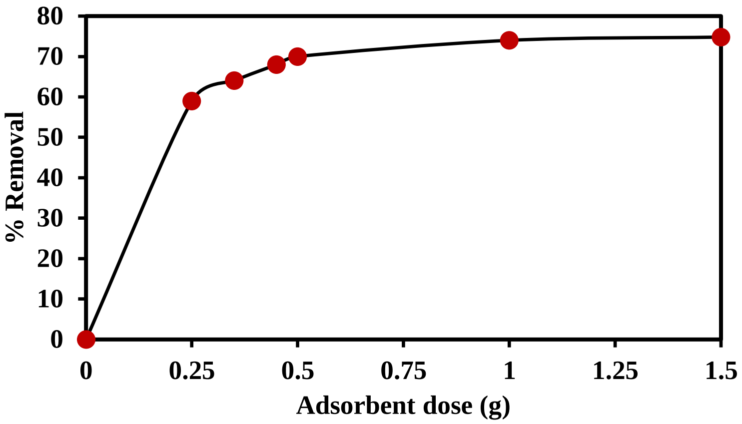

Fig. 3: Percentage removal of Cr(VI) as a function of adsorbent dose (g)

3.2.3 Initial concentration of Cr(VI) ion

It studied by varying their concentrations to 2.5 to 30 µg/mL and protecting other factors constant such as pH 2, dose 1g, agitation speed 160 rpm, time 2 hrs. **Figure 4** showed percentage removal as a function of concentration. **Figure 4** clearly revealed that in 2.5 µg/mL initial Cr(VI) ions concentration, the maximum 90 percentage adsorption was observed.

The results revealed that, the exclusion of Cr(VI) ions was established to be decreased with upsurge in the concentration of Cr(VI) ions. At inferior concentration, the ratio of initial chromium ions concentration to the existing binding site of brick powder was larger. As mentioned in literature, adsorption becomes independent on minimum adsorbate concentration (Yu et al., 2001). However, at greater concentration, maximum adsorption sites present in the brick powder may be occupied by chromium ions and after that no free sites are existing to adsorb. These results evidently presented that a reduction in percentage exclusion of the chromium ions consistent to an upsurge of initial chromium ion concentration (Feng-Chin et al., 2000).

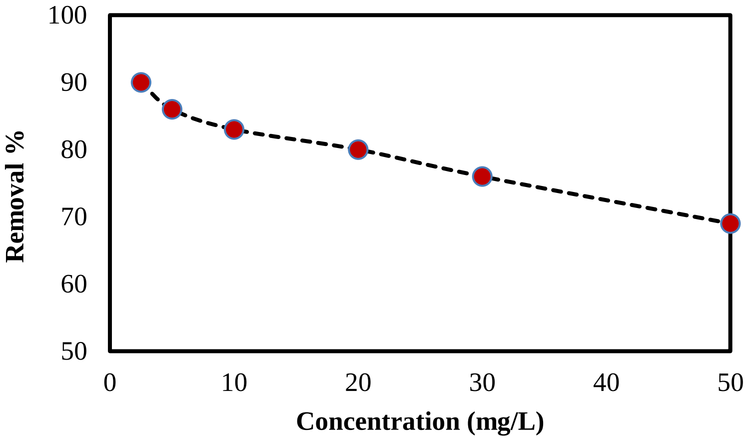

Fig. 4: Percentage removal of Cr(VI) ions as a function of initial concentration of Cr(VI) (mg/L).

3.2.4 Contact time

Exclusion of chromium ions from the water by brick powder adsorbent as a function of time was studied. **Fig. 5** revealed that the optimum removal rate of chromium ions was 120 min. Adsorption kinetics mainly depends on the surface area of the adsorbent and intra- particle diffusion. The early faster rate may be due to the accessibility of the exposed surface area of the adsorbent. As these sites increasingly occupied, the adsorption become more problematic. This is the general characteristic of adsorption of metal ions (Abdus Salam et al., 2005).

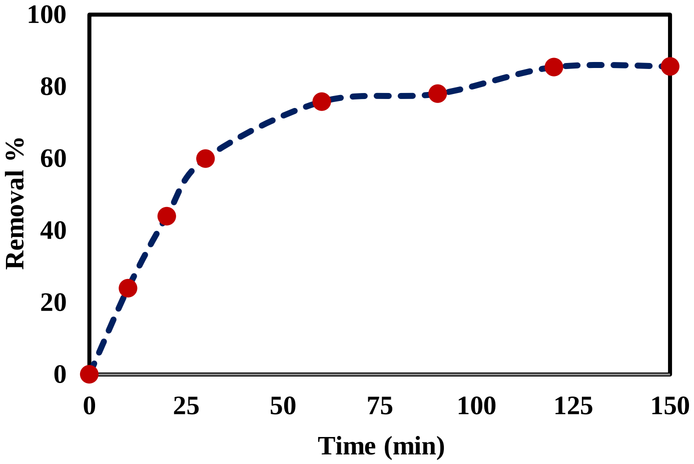

Fig. 5: Percentage removal of Cr ions as a function of contact time (min).

3.3 Adsorption isotherm model

After optimizing the adsorption parameters like adsorbent dose, initial concentration, time of equilibrium, pH and agitation speed, the adsorption models have been evaluated to explore adsorption capacity of brick powder to eliminate chromium ions from water. The Langmuir, Freundlich and Temkin isotherm models (Dada et al., 2012) have been studied and plots are shown in **Figures 6, 7, 8**. The slope and intercept of the linearized Langmuir, Freundlich and Temkin plots were used to estimate the adsorption constants which are exposed in **Table 1**.

a. Langmuir adsorption isotherm model

It is widely applied model to describe experimental adsorption data based on the assumption that maximum adsorption corresponding to saturated monolayer of adsorbate molecule on adsorbate surface with a constant energy and no further adsorption. The Langmuir adsorption equation is represented by **equation (3).**

$$Q_e = \frac{Q_m b C_e}{1+bC_e} \quad \text{.....................}(3)$$

The linear form of Langmuir expression is expressed by **equation (4).**

$$\frac{C_e}{q_e} = \frac{1}{Q_m b} + \frac{1}{(Q_m)} C_e \quad \text{................(4)}$$

Where, Qe (mg/g) is the amount of adsorbate adsorbed per unit mass of adsorbent; Ce (mg/L) is the equilibrium concentration of the adsorbate in solution after adsorption; Qm (mg/g) is the maximum adsorption capacity corresponding to monolayer coverage of adsorbents and b (L/ mg) is the adsorption equilibrium constant. The value of Qm and b are calculated from the slope and intercept of the Ce/Qe versus Ce plot (**Figure 6**).

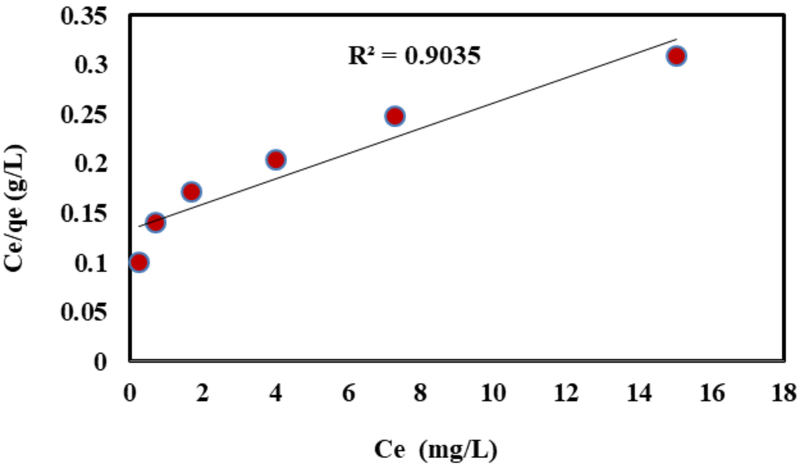

Fig. 6: Langmuir adsorption isotherms for chromium adsorption.

As can be seen in **Fig. 6**, curve was found to be a straight line having a slope ($1/q_m$) = 0.0121, intercept ($1/bq_m$) =0.1332 and the constants (q_m) and (b) were then obtained from the slope and intercept. The maximum adsorption (q_m) was found to be 82.64 mg/g, i.e, 1g of brick powder can adsorbed 82.64 mg of chromium and adsorption constant value (b) is 0.090. The correlation coefficient (R^2) was 0.9035, which indicated that Langmuir isotherm is not good fitted. However, the dimensionless parameter (R_L) which measure of adsorption favorability, was found to be, R_L= 0.181, which lies between zero to one, representing a favorable adsorption.

b. Freundlich adsorption isotherm model

The Freundlich adsorption isotherm model has also been examined using adsorption data. The plot is shown in **Fig. 7**. The model elucidates the non-ideal sorption which comprises heterogeneous surface energy system and is articulated by **equation (5)**.

$$Q_e = K_F C_e^{\frac{1}{n}} \text{............................ (5)}$$

The linear form was expressed by equation (6).

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

Where, K_F and n (dimensionless constants) are the Freundlich adsorption isotherm constants, K_F indicates adsorption capacity and n indicates adsorption intensity. Freundlich adsorption isotherm is shown in **Fig. 7.**

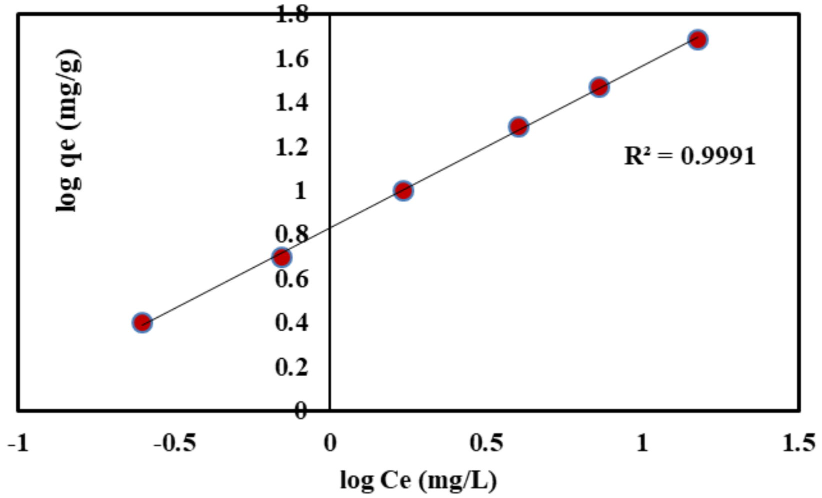

Fig. 7: Freundlich adsorption isotherm for Cr(VI) adsorption.

The isotherm (**Fig. 7)** was found to be a straight line having a slope (1/n) and intercept (log K_f). Here K_F was found to be 2.3 indicating the relative adsorption capacity of the brick powder. The value is quite comparable with literature value (Bai et al., 2003). The value of n was established to be 1.35 which specifies the adsorption intensity. The value of n lies between 1 and 10, indicating the favourable physical adsorption of chromium (VI) on the brick powder. At this point, correlation coefficient value (R^2) was 0.9991 which was established to be higher than correlation coefficient value (R^2) of Langmuir. Hence, Freundlich adsorption isotherm was found to be best fitted. It revealed that the scattering of the sites on the brick powder was non-uniform i.e., heterogeneous multilayer adsorption. It is also an indication of huge number of adsorption sites related with different energy which is in good agreement with literature (Gupta et al., 2001).

c. Temkin adsorption isotherm model

This model take responsibility on the heat of adsorption of all the molecules in the layer which would decline linearly rather than logarithmically with coverage due to adsorbate adsorbent interaction. The adsorption is characterized by uniform distribution of binding energy up to some maximum binding energy. The linearized Temkin equation is specified by **equation** (**7 and 8**).

$$q_e = \frac{RT}{b} logK_T C_e \quad ....(7)$$

$$q_e = BlnK_T + BlnC_e ....(8)$$

Where, (RT/b) = B, K_T = Temkin isotherm equilibrium binding constant (L/g), which corresponds to maximum binding energy, b = Temkin isotherm constant, R = Universal gas constant (8.314 J/mol/K), T = temperature at 298 K, B = constant related to heat of sorption (kJ/mol). The Temkin constants KT and b is calculated from the slope and intercept of Qe as a function of lnCe.

Temkin adsorption isotherm was exposed in **Fig. 8**. The correlation coefficient (R^2) was found to be 0.8623 which was very less than Langmuir and Freundlich value which showed that Tempkin isotherm model was not close-fitting. However, positive Temkin constant **B or** heat of sorption (kJ/mol) value i.e., (+10.95) signifies an exothermic process where adsorption is favored by lower temperatures and heat is released as molecules attach to the surface. The value < 20 kJ/mol is an symptom of physical adsorption.

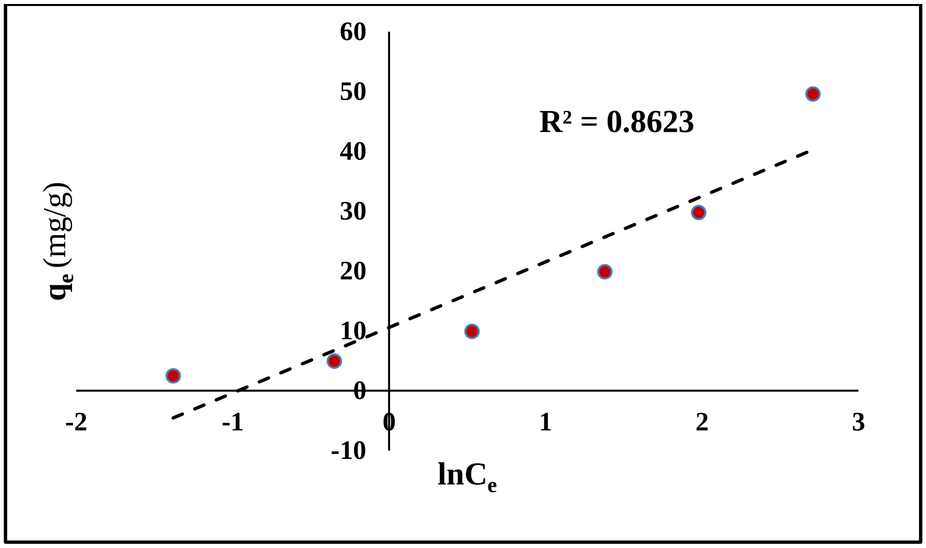

Fig. 8: Temkin adsorption isotherm for Cr(VI) adsorption.

Table 1: Langmuir, Freundlich and Temkin constants obtained from the Cr adsorption on brick powder.

S.No.	Parameter	Cr(VI)
1.	**Langmuir Isotherm**	
	Binding energy b (L/mg)	0.090
	Maximum loading Q_m (mg/g)	82.64
	Correlation coefficient (R^2)	0.9035
	R_L	0.18
2.	**Freundlich Isotherm**	

	Adsorption capacity (K_f)	**2.30**
	Adsorption intensity (1/n)	**1.35**
	Correlation coefficient (R^2)	**0.9991**
3.	**Temkin Isotherm**	
	Equilibrium binding energy A (L/g)	10.64
	Heat of sorption B (kJ/mol)	10.95
	Correlation coefficient R^2	0.8623

3.4 Probable mechanism of adsorption of Cr(VI) on brick powder adsorbent

The surface of brick powder was positively charged when solution pH was 2 and Cr(VI) occurs in the form of oxyanions such as $HCrO_4^-$, $Cr_2O_7^{2-}$, CrO_4^{2-} in acidic medium. In such condition, adsorption is due to electrostatic interaction between positive and negative charge. Besides this, adsorption on the brick powder may also be due to ion exchange process. The clay suspension generally contain metal ions like K^+, Na^+, Ca^{2+}, Al^{3+}, and Fe^{2+}/Fe^{3+} as reported in literature (https:/ en.m.wikipedia.Org>Wiki>Brick). Consequently, the negatively charged Cr(VI) species directly do not adsorbed by brick particles. The dominating mechanism for Cr(VI) removal in most of the studies reported, is reduction of Cr(VI) to Cr(III) followed by adsorption. The reduction of Cr(VI) into positively charged Cr(III) is important for ion exchange. Cr(VI) may also be reduced by organic material and minerals with surface metal species. Deng, Lan, Houston, and Brandy, investigates how clay minerals influence the reduction of Cr(VI) by organic compounds. Study explores mechanisms by which these clay minerals affect the Cr(VI) reduction process, likely involving surface interactions, the presence of catalytic species within the clays like Fe(II), and the role of organic matter (Deng et al., 2003). The tartaric acid may also able to reduce Cr(VI) to Cr(III), which may be catalyzed by TiO_2, γ- Al_2O_3, α- FeOOH present in clay (Deng et al., 2003). Therefore, the maximum removal observed is possibly due to existence of reducing agents like COOH- functional group and metal ions in organic matter. These agents support to reduce Cr(VI) to Cr(III) followed by adsorption. Hence, the probable mechanism is as follows:

$$HCrO_4^- + \text{Organic material} + H^+ \longrightarrow Cr^{+3} + H_2O \ \ldots\ldots\ldots\ldots\ldots (9)$$

$$Cr^{+3} + (M^{n+}\text{-brick matrix}) \longrightarrow \text{Cr- brick matrix} + M^{n+} \ \ldots\ldots\ldots (10)$$

Here, M^{n+} may be K^+, Na^+, Ca^{2+}, Al^{3+}, and Fe^{2+}/Fe^{3+}. These metal ions present in brick clay may get released into solution and then exchanged with Cr^{3+} as described in literature (Mohan et al., 2006).

4. Conclusions

A low cost adsorbent waste brick powder has been used for the removal of Cr(VI). The exclusion of chromium was totally depend on adsorbent dose, pH, initial concentrations of Cr(VI) ions and also contact time. The acidic pH was favorable. The optimum pH was found to be 2. Contact time for maximum adsorption required was 120 min. The uptake of Cr(VI) ion amplified with enhancement of adsorbent dose and it was found that 1 g was optimum dose for adsorption.

The adsorption isotherm results are consistent with the Freundlich isotherm. The correlation coefficients R^2 values calculated for the Langmuir and Temkin fittings are slightly lower than those obtained from Freundlich system. However, the value of n was >1 indicates for favorable adsorption condition.

This process of adsorption—ion exchange would observationally be seen as a multilayer process viewing the validity of the Freundlich isotherm. Tempkin value also suggests that the reaction mechanism may be exothermic process. Heavy metal removal with brick powder adsorbent appears to be technically feasible and eco-friendly too.

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Author's declaration

There is no conflict of interest among the authors. All the figures and data presented in the manuscript are ours.

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