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Synthesis and characterization of novel Co/Bi-layered double hydroxides and their adsorption performance for lead in aqueous solution

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Abstract   The Co/Bi-layered double hydroxides (Co/Bi-LDH) were synthesized by co-precipitation method and used for the removal of lead from aqueous solutions. The Co/Bi-LDH was characterized using X-ray diffraction (XRD), Fourier Transform Infrared spectroscopy (FTIR), Transmission Electron Microscopy (TEM), Selected Area Electron Diffraction (SAED) and BET for textural properties. Adsorption of lead solution by Co/Bi-LDH was carried out using batch experiment by mixing the lead solution and the adsorbent. The effects of various parameters such as contact time, pH, adsorbent dosage and initial concentration were investigated. The optimum pH for lead removal was found to be 4 and the optimum time of lead removal was found to be 120 min. The isotherm data were analyzed using Freundlich and Langmuir. The adsorption isotherms can be well described by the Langmuir model with $R^2 > 0.99$. Its adsorption kinetics followed the pseudo-second-order kinetic model. Thermodynamic parameters were also studied. It was found that the synthesized Co/Bi-LDH can reduce the lead concentration and makes it a potential material for the decontamination of lead polluted water.

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1. Introduction

Layered double hydroxides (LDHs), have been attracting attention because of their wide applications as anion exchangers (Newman and Jones, 1998), catalysts (Hamada et al., 2005), bioactive (Choy et al., 1999) and so on. LDHs also known as anionic clays, constitute a large family of compounds with general formula $M(II)_{1-x}M(III)_x(OH)_2 A^{n-}_{x/m} \cdot nH_2O$, where, M(II) is a divalent cation [Mg(II) and/or Ni(II), Zn(II), Co(II), Mn(II)], M(III) is a trivalent cation [Al(III) and/or Fe(III), Ga(III), Cr(III), V(II) and A^{n-} is an anion with charge n (OH^-, CO_3^{2-} NO_3^-, Cl^-, (SO_4^{2-}) (Auerbache et al., 2004; Jaiswal and Chattopadhyaya, 2011; Cavani et al., 1991; Clearfield et al., 1991; Jaiswal and Chattopadhyaya, 2009).They can be synthesized using different methods (Cavani et al., 1991), the most common being the co-precipitation at constant pH of dilute solutions

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containing M(II) and M(III) cations with solutions containing carbonate and hydroxide ions (Tichit et al., 1995; Reichle et al.,1986; Corma et al.,1994). The most important characteristic of the structural properties of the LDHs are the distance between the neighboring cationic layers, also known as basal spacing. These parameters depend to a large extent on the nature of the anions in the inter layers. Lead is an important compound used as an intermediate in the processing industries, which however has been recognized to be acutely toxic to human beings when present in high amounts in water. Of these, lead ions are attracting wide attention of researchers as one of the heavy metals because all forms of lead are toxic and adversely affect reproductive, nervous, immune, cardiovascular system as well as development process in children (Schwarts, 1994). Studies have shown that young children, infants and pregnant women are particularly susceptible to unsafe Pb(II) levels. For adults, increased levels of Pb(II) have been linked to high blood pressure and damaged hearing. Drinking, eating, inhaling even at low level of Pb(II) can cause other serious health effects (Adelekan and Abegunde, 2011; Kabbashi et al., 2009). The permissible limit of Pb(II) in drinking water and surface water intended for drinking, as set by European Union (EU), United States Environmental Protection Agency (USEPA) and WHO, is 0.010, 0.015 and 0.010 mg/L, respectively (Lalhruaitluanga et al., 2010; Li and Wang, 2009). Adsorption techniques have been successful in removing heavy metals from solutions. As a result LDHs are now being considered as low cost adsorption alternatives because they are easily prepared and regenerated. LDHs are naturally occurring clays which can be easily synthesized by co-precipitation of dilute aqueous solution of cobalt and bismuth. It is also regenerated when treated with 0.1 M NaOH.

The present work deals with the synthesis of Co/Bi-layered double hydroxides by co-precipitation method, characterization by XRD, FT-IR, TEM and their application in the removal of Pb(II) ion from aquatic media. Equilibrium isotherm data for lead ion was analyzed by Freundlich, Langmuir isotherm and effect of various parameters concentration, contact time, pH and temperature on adsorption. Kinetic and thermodynamic parameters were also studied in this work.

2. Materials and methods

2.1. Synthesis of layered double hydroxide

The standard conditions described by Marchi were used (Marchi and Apesteguia, 1998). 100 mL of an aqueous solution containing cobalt nitrate (Thomas Baker) and bismuth nitrate (Guaranteed Reagent) (3:1), was added in 200 mL of water under stirring (Soln. 1). 16 g of NaOH (Laboratory Reagent) and 2.86 g of Na_2Co_3 (Loba Chemie) were dissolved in 1 L of water solution (Soln. 2), Then solution 1 is added drop by drop into solution 2, under vigorous stirring on a magnetic stirrer, at constant pH and at temperature between 60 and 80 °C. The mixed suspension was centrifuged to recover a black precipitate, the solid was washed with deionized water and then the precipitate was dried in an oven for 24 h at 100 °C. The synthesized material was ground to a fine powder using a pestle and mortar.

2.2. Characterization of layered double hydroxides (LDHs)

The synthesized Co/Bi-LDH was characterized by XRD. X-ray diffraction is the most common technique used for the characterization of minerals (Thorez, 1976). XRD was performed on X'PERT-PRO diffractometer operated at 40 kV/ 30 mA, using CuKα$_1$ radiation with a wavelength of 1.54 in the wide angle region from 20 to 80 on 2Θ scale. FT-IR study was carried out on a Perkin Elmer Spectrum Two FT-IR spectrometer to analyze functional groups, using potassium bromide (KBr) disk method. Transmission electron microscopy (TEM) and selected area electron diffraction (SAED) characterizations were performed using FEI TEM electron microscope. The textural analysis was performed using a Micromeritics model ASAP 2020, V3029, single port. The samples were previously treated at 200 °C of temperature in vacuum.

2.3. Preparation of stock solution

An aqueous stock solution (1000 mg/L) of Pb(II) ions was prepared using $Pb(NO_3)_2$ salt (Merck). pH of the solution was adjusted using 0.1 M solution of HNO_3 or NaOH. Adsorption is influenced by many physicochemical factors, including effects of adsorbate concentration, contact time, pH and temperature. The adsorption capacity of Co/Bi-LDH was determined from the contact of various concentrations (10–200 mg/L) of metal solution in conical flasks, with 0.5 g of LDHs. The mixture was shaken in an orbital shaker (Shivam, IS0 900/2000) at 120 rpm followed by filtration. The filtrate containing the residual concentration of metal was determined by Atomic Absorption Spectrophotometer (ECIL 4141). The percentage (%) removal of metal ion and amount of metal ion adsorbed on LDHs (q_e) were calculated by Eqs. (1) and (2), respectively:

$$\%removal = 100 \times C_o - C_e/C_e \qquad (1)$$

$$q_e = (C_o - C_e)V/M \qquad (2)$$

where, q_e is the amount of metal ion adsorbed on adsorbent at equilibrium (mg/g), C_o and C_e are the initial and equilibrium concentration (mg/L) of metal ion in solution, respectively. V is the volume of solution (L) and M is the mass of adsorbent (g).

3. Result and discussion

3.1. Characterization of adsorbent

Fig. 1 shows the XRD spectra of the LDH show peaks at the positions 11.5°, 29.1°, 33.5°, 35.44°, 36.7°, 38.9°, 45.6° and 52.5°, which are in good agreement with JCPDS file (JCPDS 38-0486, $a = 3.076$ Å, $b = c = 22.80$ Å). The XRD spectra showed small kink type peaks and the average particle size was found ∼35 nm.

The FT-IR spectra for Co/Bi-LDH (Fig. 2) showed a broad and strong band in the range 3200–3600 cm^{-1} centered at 3424 cm^{-1} which was due to the O–H stretching vibration of the inorganic layers and interlayer water molecules. Another common wave number for LDH-like material is a band at 1629 cm^{-1} which is assigned to the bending vibration of interlayer water molecules. For Co/Bi-LDH, a strong absorp-
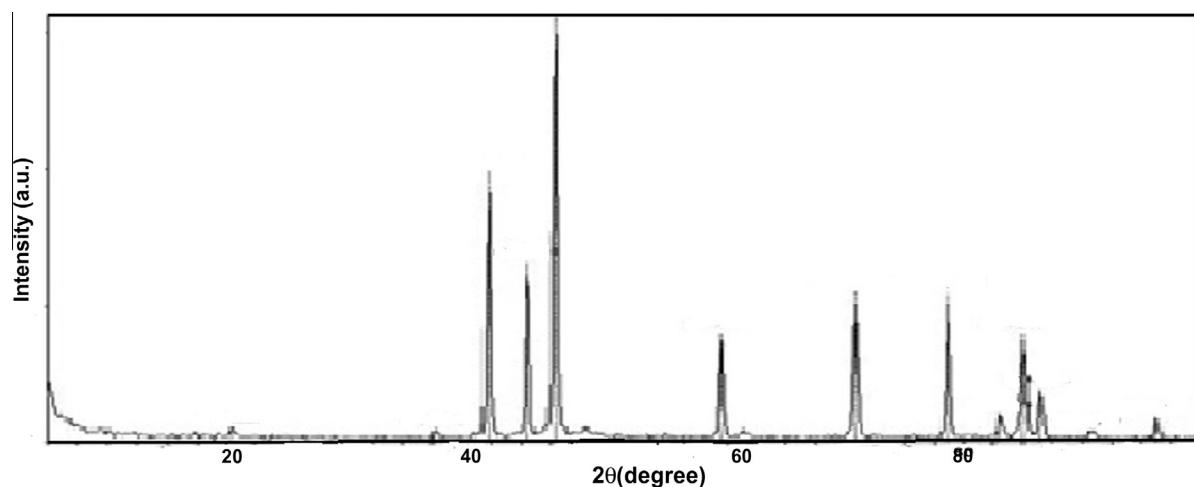
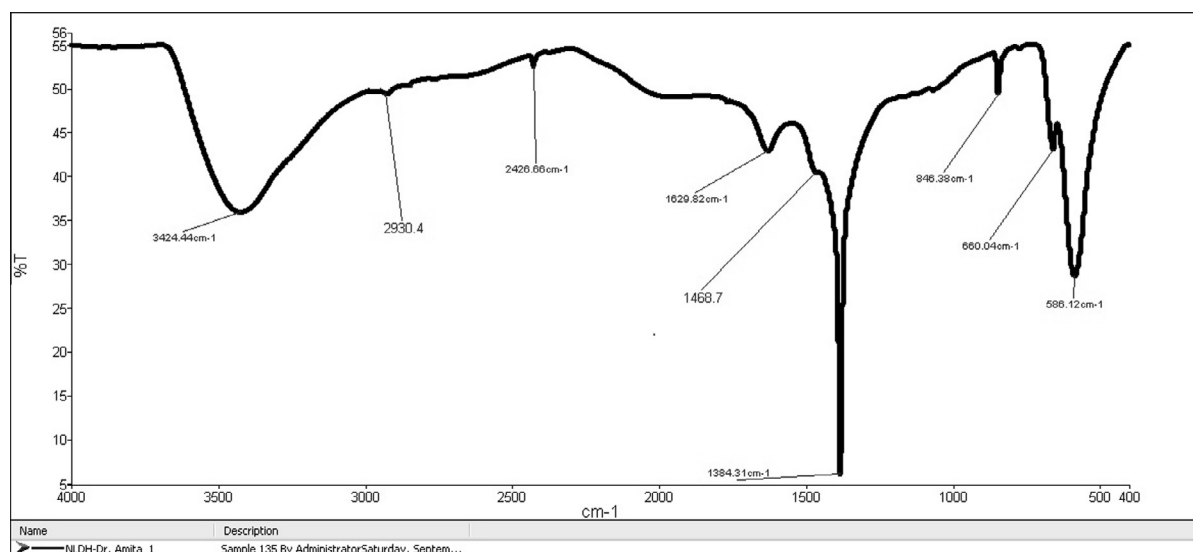**Figure 1** XRD spectra of Co/Bi-LDH.

Figure 2 FT-IR spectra of Co/Bi-LDH.

tion band centered at 1384 cm^{-1} is assigned to the N–O stretching vibration. A band in the lower wave number region corresponds to the lattice vibration mode such as the translation vibrations of Co–OH at 660 cm^{-1} and deformation vibration of OH–Co–Bi–OH at around 586 cm^{-1} (Feng et al., 2006; Zhang et al., 2004). The TEM micrograph of synthesized LDHs is shown in Fig. 3. From Fig. 3, it is clear that the particle size of Co/Bi-LDH is around 50 nm heterogeneous and hexagonal in shape. The SAED pattern (Fig. 4) of individual sheets exhibited hexagonally arranged spots, confirming their single-crystal nature. The hexagonal lattice with $a = 3.06$ Å was compatible with the in-plane structural parameter of Co/Bi-LDH crystals determined from the XRD characterization, Textural properties of the adsorbent were also taken into consideration as the same play significant role in adsorption process. The BET surface area and the pore volume of Co/Bi-LDH were found to be 47.8 m^2/g and 0.46 cm^3/g, respectively.
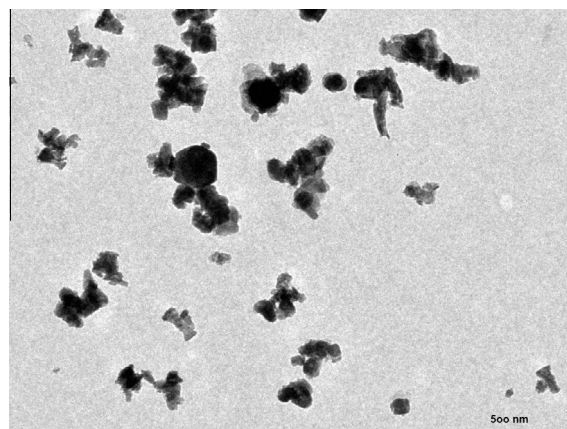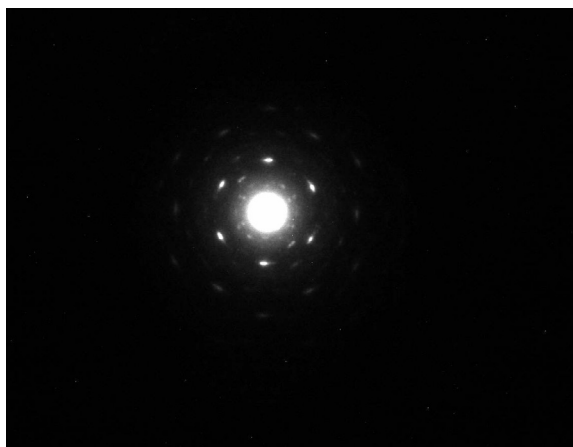

Figure 3 TEM of Co/Bi-LDH.

Figure 4   SAED pattern of Co/Bi-LDH.

3.2. Effect of concentration

The effect of different concentration of Pb(II) (10–200 mg/L) has been investigated at 303 K, Fig. 5 shows that the adsorption of metal ions increased with increase of equilibrium concentration. This trend is due to the progressive increase in the electrostatic interaction between the Pb(II) and Co/Bi-LDH active sites. However, this can be explained by the fact that more adsorption sites were being covered as the metal ion concentration increases (Laraous et al., 2005).

3.3. Effect of contact time

Effect of contact time on adsorption of Pb(II) on the Co/Bi-LDH is shown in Fig. 6. It was found that 89.86% of the metal was removed with in 120 min. After that, contact time has no effect in metal removal percentage. Very high adsorption rates were observed at the beginning because of the larger number of sites available for the sorption and adsorption equilibria were then gradually achieved (Babel and Kurniawan, 2004).

3.4. Effect of pH

The initial pH of the test solution varied from 2 to 5. The amount of Pb(II) adsorbed on the adsorbent increases steadily with increasing pH in range from 2 to 4 (Fig. 7) and after 4 pH Pb(II) was not used in order to avoid the precipitation of metal
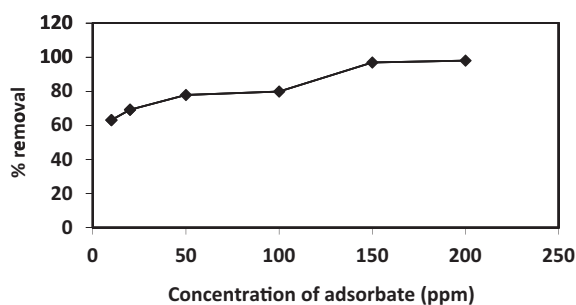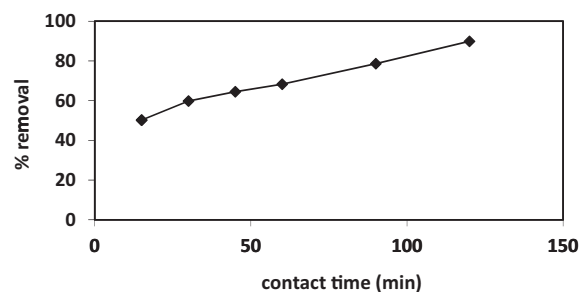

Figure 6   Effect of contact time on adsorption of Pb(II) on Co/Bi-LDH.
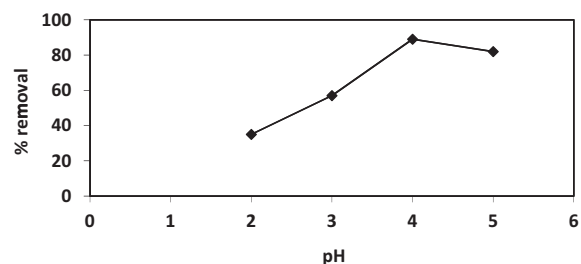

Figure 7   Effect of pH on adsorption of Pb(II) on Co/Bi-LDH.

ions in the form of their hydroxide (Afkhamia et al., 2010). At lower pH, adsorption of metal ions decreased due to increased competition with H^+ ions for active adsorption sites. However, at higher pH values, the presence of H^+ ion in solution decreased and LDH surface also deprotonated, leading to increased adsorption of metal ions.

3.5. Effect of temperature

Temperature is known to have a profound effect on various chemical processes. Temperature affects the adsorption rate by altering the molecular interactions and the solubility of the adsorbate (Ahmaruzzaman and Sharma, 2005). The effect of temperature on the adsorption of Pb(II) on LDH was investigated by experiments for 10 mg/L Pb(II) at 283–313 K. It was observed that on increasing the temperature percentage the removal of metal ions increased (Fig. 8). This showed that the adsorption process was endothermic.
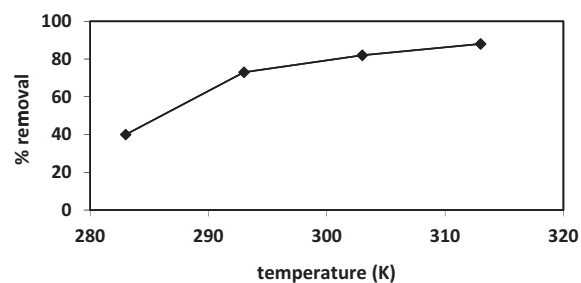

Figure 5   Effect of concentration on adsorption of Pb(II) on Co/Bi-LDH.

Figure 8   Effect of temperature on the adsorption of Pb(II) on Co/Bi-LDH.

3.6. Effect of co-existing ions

The effect of co-existing ions was evaluated by the adsorption efficiency of the adsorbent and various interference ions such as Hg(II), Ni(II), Co(II) and Cu(II) were added with a metal ion concentration of 25 mg/L for each metal into 100 mL volumetric flasks containing Pb(II) metal ion solution at a definite concentration of 25 mg/L. The experimental result shows that presence of these ions adversely affects the uptake of Pb(II) ions on the Co/Bi-LDH. It has been found that the percent removal of Pb(II) decreases from 98.96% to 82.86% and the trend of removal in decreasing order follows this pattern Pb > Hg > Cu = Ni > Co. The electronegativity of each metal is 2.33, 2.0, 1.90, 1.91, and 1.88 for Pb(II) Hg(II), Cu(II), Ni(II) and Co(II), respectively (Huheey, 1978). This order of affinity for metal ion on the Co/Bi-LDH material may be attributed to the differences in electronegativity i.e. more electronegative metal will show higher tendency for adsorption.

3.7. Kinetics of adsorption

Several kinetic models have been proposed to clarify the mechanism of a solute adsorption from aqueous solution onto an adsorbent. The rate constant of adsorption was determined from the pseudo-first order rate equation (Lagergren, 1898);

$$dq_t/dt = k_1(q_e - q_t) \qquad (3)$$

where, q_t is the amount of metal ion adsorbed at time t (mg/g), q_e is the adsorption capacity at equilibrium (mg/g) and k_1 is the pseudo-first order rate constant (min^{-1}). The values of k_1 and q_e (Table 1) were calculated from the slope and intercepts of linear plots of ln $(q_e - q_t)$ vs. t.

The pseudo-second order kinetic model is represented as (Ho and McKay, 1999),

$$dq_t/dt = k_2(q_e - q_t)^2 \qquad (4)$$

where, q_t and q_e are amount of metal ion adsorbed at a time t (mg/g) and at equilibrium (mg/g), respectively and k_2 is the pseudo-second order rate constant (g/mg/min). The values of k_2 and q_e were calculated from the intercepts and slopes of the linear plot of t/q_t vs. t, respectively. The values of regression correlation coefficient (R^2) confirmed that the adsorption of Pb(II) on LDH followed pseudo-second order kinetics (Table 1).

3.8. Adsorption isotherm

Inspections of an adsorption isotherm are relatively simple methods of assessing the feasibility of using LDH for a particular application. The uptake of Pb(II) by LDH has been evaluated in accordance with the linearized form of Ferundlich and Langmuir isotherm equations, respectively (Freundlich, 1906; Langmuir, 1916).

$$\log q_e = \log K_f + 1/n \log C_e \qquad (5)$$

$$C_e/q_e = (1/K_L q_m) + (C_e/q_m) \qquad (6)$$

where, q_e is the concentration in the adsorbed phase, i.e., the amount of adsorbate adsorbed per unit weight of LDH, C_e is the equilibrium concentration of adsorbate in solution after adsorption, K_f, $1/n$ are Freundlich constants and K_L, q_m are

Table 1 Kinetic Parameters for the adsorption of Pb(II) on Co/Bi-LDH.

$q_e^{\exp}$ (mg/g)	1.005
Pseudo first order	
k_1 (min^{-1})	0.008
q_e^{cal} (mg/g)	0.848
R^2	0.904
Pseudo second order	
k_2 (g/mg min^{-1})	0.056
q_e^{cal} (mg/g)	0.816
R^2	0.998

Table 2 Freundlich and Langmuir isotherm parameters for the adsorption of Pb(II) on Co/Bi-LDH.

Freundlich parameters		Langmuir parameters	
K_f(L g^{-1})	0.036	K_L (L mg^{-1})	1.087
$1/n$	1.204	q_m (mg g^{-1})	143.4
R^2	0.981	R^2	0.991

Langmuir constants. The straight line nature of the curve having correlation coefficient 0.9861 and 0.9914 for Freundlich and Langmuir isotherm (Table 2), respectively indicates that the adsorption process followed both isotherm models but fitted well with Freundlich isotherm.

3.9. Thermodynamic parameters

In any adsorption process, both energy and entropy considerations must be taken into account in order to determine what process will occur spontaneously. Values of thermodynamic parameters are the actual indicators for practical application of a process. The equilibrium constant K was calculated (Igwe and Abia, 2007) using following equation,

$$K = q_e/C_e \qquad (7)$$

where, q_e is the amount of metal ion adsorbed at equilibrium (mg/g) and C_e is the amount remaining in solution at equilibrium (mg/L). The values of Gibb's free energy ($\Delta G°$) for the adsorption at different temperatures were calculated using following Eq. (8).

$$\Delta G° = -RT \ln K \qquad (8)$$

where, $\Delta G°$ is the Gibbs free energy change, R is the universal gas constant and T is absolute temperature (K). The standard free energy change is also related to standard enthalpy change ($\Delta H°$) and the standard entropy change ($\Delta S°$) by Eq. (9).

$$\Delta G° = \Delta H° - T\Delta S° \qquad (9)$$

From Eqs. (8) and (9), we obtained:

$$\ln K = -\Delta H°/RT - \Delta S°/R \qquad (10)$$

Hence, from the slope and intercept of plot ln$K$ vs. $1/T$, the value of $\Delta H°$ and $\Delta S°$ was obtained. Values of all the thermodynamic parameters are given in Table 3.

**Table 3** Thermodynamic parameters for the adsorption of Pb(II) on Co/Bi-LDH.

Temp. (K)	Standard Gibbs free energy change (ΔG°)(kJ/mol)	Standard enthalpy change (ΔH°) (kJ/mol)	Standard entropy change (ΔS°) (kJ/molK)
283	−4525.54	−1716.82	9.92
293	−4624.79		
303	−4724.04		
313	−4823.29		

Table 4 Comparison of the adsorption capacities of Pb(II) onto various adsorbents.

S. No.	Adsorbents	Adsorption capacity (q_m mg/g)	References
1	Fly ash	0.368	Yadava et al. (1989)
2	China clay	0.411	Yadava et al. (1991)
3	Ekulu clay	0.557	Dawodu et al. (2012)
4	Wollastonite	1.68	Yadava et al. (1991)
5	Coconut activated carbon	4.38	Gueu et al. (2007)
6	Seed hull of the Palm tree activated carbon	3.77	Gueu et al. (2007)
7	Kaolinitic clay	9.37	Majone et al. (1993)
8	Illite clay	14.1	Farrah et al. (1980)
9	Phosphatic clay	35.82	Singh et al. (2006)
10	Montmorillonite clay	71.8	Farrah et al. (1980)
11	Co/Bi-LDH	143.4	Present work
12	Tartrate-Mg–Al LDH	8.4	Yasin et al. (2013)
13	Mg–Al–NO3 LDH	3.2	Yasin et al. (2013)

4. Regeneration of LDH

Layered double hydroxide which had been exposed to Pb(II) metal ion solution and adsorbed up 89% of Pb(II) were again treated with 0.1 M NaOH for elution of Pb(II). The Pb(II) was almost completely removed from the Co/Bi-LDH which can be explained by following equation,

$$M - Pb + 2NaOH \rightarrow M - Na_2 + Pb(OH)2$$
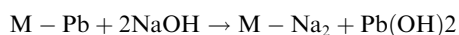
where, M = Co/Bi-LDH

5. Comparative study

A comparative study in terms of adsorption capacity which has been carried out with other reported adsorbents is summarized in Table 4. From Table 4, it is clear that the adsorption capacity of Co/Bi-LDH is greater than that of other adsorbent materials, reflecting a promising future for this LDH utilization in Pd(II) removal from aqueous solutions.

6. Conclusions

This study showed that the novel composition of Co/Bi-LDH adsorbed significant amount of Pb(II) ion. From the experiment we concluded that the percentage removal of Pb(II) adsorption steadily increases with increasing adsorbate concentration, agitation time, pH and temperature. The adsorption kinetics followed pseudo- second order kinetics. The adsorption of Pb(II) onto LDH followed both Freundlich and Langmuir isotherm models but Langmuir Isotherm is best fitted than Freundlich isotherm. The negative value of ΔG° indicates that the adsorption process was feasible and spontaneous in nature and the positive value of ΔH° shows the endothermic nature of the process. The used LDH can be easily regenerated.

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