Adsorptive Removal of Pb (II) From Aqueous Solution Using Pyoaurite Type Sorbent, Cerelite, Activated Carbon, Goethite and Rice Husk.

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Abstract

Removal of divalent lead from aqueous solution, a comparative study of six sorbents was carried out by batch as well as column processes. The order of lead removal capacities for these chemical adsorbents was found to be IRA > PTS> PAC > PLC > FeO(OH) > RH. This is probably due to several factors such as molecular size, molecular polarity, pH lead reactivity and retentivity, sorbent properties and environmental conditions. The leaching order should be reverse of the sorption order. The sorption of lead ions goes on increasing as their concentration decreases from 10 to 0.1 mg/l. Thus, the removal was about 20 % less than what it was in the batch process Leaching increases as the flow rate increases. Solution pH significantly affected the extent of contaminant removal.

Keywords: Removal, Sorption, Activated carbon, Goethite, Rice husk, Cerelite

Introduction

In modern society, capricious discharge of industrial wastewater accommodate heavy metal ions has develop in serious problems sustainable dissymmetry and health risk. Human lead exposure results from lead plumbing materials such as solder, brass fittings, service lines, crumbling and chalking lead-based paint, airborne lead particulates released by industrial or waste elimination emissions and fallout of decades of leaded gasoline emissions. Overall evidence from a metal analysis has established a strong link between low-dose lead exposure and intellectual deficit in children. Above this level, adverse health effects range from upset stomachs, irritability, constipation, and damage to organs, including the nervous system and the brain, high lead levels can be fatal.

The objective of this study is to compare and evaluate different types of adsorbents for lead removal from aqueous media. Adsorbent materials such as activated carbon and naturally occurring goethite and rice husk were selected because of their relative low cost. A cation exchange resin ceralite IR 120 (Na^+ form, 20-50 mesh) was selected because it can provide a basis for comparison with the synthesized sorbent.

Determination of urinary lead, lead and nickel in steel production workers was carried by Horng et al. (2002) Arsenic and lead concentrations in the Pond Creek and Fire Clay coal beds, eastern Kentucky coal field by Hower et. al. (1997) Hu Qiufen C. et al. (2002) determined trace lead, lead and mercury by on-line column enrichment followed by RP-HPCL as metal-tetra-(4-bromophenyl)-porphyrin chelates. Mavropoulos et al immobilization. (2002) studied the mechanisms of lead by hydroxyapatite. Murphy and Hall (2000) examined changes in dust lead loading on walls and ceilings after lead hazard control

interventions. Zhendong and Fiona (2001) presented a thermodynamic approach to ion flotation. II. Metal ion selectivity in the SDS-Cu-Ca and SDS-Cu-Pb systems.

Several reports on lead removal from water have been published (Afshan et al.,2001: Ainsworth et al., 1994; Ajmal et al., 1998; Brown et al.,2000: Christophi and Axe, 2000). several substances have been reported to remove lead from water such as activated carbon (Gajghate et al., 1990; Gajghate and Saxena, 1991), polyethylene glycol-methacrylate Gel beads carring Cibacron blue F3GA (Denizli et al., 1999), peat, coir pith carbon (Kadirvelu and Namasivayam,2000), goethite (Villalobos et al., 2001), petri dishes and filter materials (Voigt et al., 2001), pyroaurite like compound (Seida et al., 2001), palas (Butea monosperma) bark substrate (Rampure and Patil, 1995; Suh and Kim, 2000) etc.

The advanced treatment processes such as ion exchange, reverse osmosis, membrane separation, electrodialysis, chemical precipitation, and activated carbon sorption are prohibitively expensive. The use of microbes such as bacteria, fungi and algae in treating contaminated wastewater is today an attractive technique but yet not suitable for applications on a large scale. The high capital cost and recurring expenses of most of these methods do not suit the small-scale industries. studies on the treatment of wastewaters bearing heavy metals and organics reveal Sorption to be the most promising technique because it is highly effective, cheap, easy and eco-friendly method among all physicochemical processes.The present study has been undertaken to investigate and compare the suitability of synthesized pyroaurite like compound (PLC), modified PLC, i.e., pyroaurite type sorbent (PTS) and adsorbents such as powdered activated carbon (PAC), rice husk (RH) and goethite (FeO(OH)) for the Pb(Il) removal from wastewater using batch and column ex experiments. A cation exchange resin Ceralite IR 120 (Na+ form, 20-50 mesh) was also taken so that the sorption capacity of the synthesized sorbent can be compared (Singh et al., 2001; Suh and Kim, 2000).

Materials and Method
Pb(ll) Solutions

All reagents were of analytical grade. In order to avoid interference by other elements in wastewater, the experiments were conducted with aqueous solutions of pentavalent lead in redistilled water. Synthetic samples of various initial Pb(II) concentrations were prepared from the stock solution that was in turn prepared by dissolving 1.598 g of lead nitrate, $Pb(NO_3)_2$, in a minimum volume of (1:1) HNO, and then diluting it to 1 litre with 1 % (v/v) HNO_3. Four standard solutions of 0.1, 1, 5 and 10 mg/l concentrations of Pb (II) were prepared by diluting the stock solution of 1 g/1. Double distilled water was used to dilute the solution.

Preparation and Characterisation of Pyroaurite like Compound

Mg Cl_2 (o.15 mol) and$FeCl_3$ (0.075 mol) were dissolved into 200 ml of distilled water. 10-wt% NaOH aqueous solution was then added drop wise into the solution while stirring vigorously until the pH of the solution reached above 13. The solution was kept at room temperature for 24h with vigorous stirring. The obtained precipitate was washed with a large amount of distilled water to wash away residual.

Table 1. Characteristics of sorbents

Characteristics	IRA	PTS	PLC	PAC	FeO (OH)	RH
Density (glcc)	2.11	2.20	2.50	1.40	2.49	1.09
Bulk density (g/ml)	1.3	1.6	1.8	1.2	1.7	0.9
Moisture(%)	10.5	11.31	16.31	3.61	15.00	8.20
Ash (%)	10.7	9.65	12.65	3.32	12.32	2.30
Volatile matter (%)	9.3	6.74	5.74	9.39	6.08	10.02
Combustible matter (%)	55.6	64.16	54.16	84.85	57.54	89.02
Loss of ignition	4.52	5.02	4.02	3.83	5.96	9.02
Chemical Analysis						
Sio_2 (%)	9.3	2.00	2.00	7.05	33.92	18.35
Al_2O_3 (%)	12.0	0.02	0.02	2.05	12.44	1.83
CaO (%)	.31	0.12	0.12	0.04	4.15	1.35
Fe_2O_3(%)	0.22	5.32	5.02	1.23	12.26	2.51
MgO (%)	0.11	3.02	3.00	1.10	2.36	1.04
Surface area (m^2/g)	730	590	470	538	378	198

Batch Studies

Batch tests were conducted at constant temperatures under steady state and transient rate batch conditions.

Capacity Studies

The capacity studies were carried out by transient batch tests. In20 numbers (4 concentration of 4 toxics each + 4 controls one for each) of 100- ml capacity PVC bottles with screw caps), 0.5 g of sorbent was added to each bottle of 4 sets, each set having 5 bottles 4 for different concentrations of a toxic and 1 for its control. 100 mi of the toxic solution was added to each bottle. The solution was buffered with 0.07 M sodium acetate – 0.03M acetic acid, pH 8. The bottle was shaken in a reciprocating shaker at 150 rpm for 5 h at room temperature, and the concentration of a toxic element was measured after 1, 3, 5, and 24 h until the solution reaches equilibrium. The controls without sorbent demonstrated toxic uptake due to the sorbent, not form other sources such as the walls of the container, centrifuge tube etc. The contents were centrifuged at 2000 rpm for 10 min and the supernatant liquid was filtered using a 0.45 mm membrane filter. The filtrate was analysed for the studied ions (APHA 1998) The initial and final concentrations of the solutions were determined using a Perkin Elmer Analyst 100 AAS. Experiments were triplicated and results averaged. Unless otherwise stated all experiments were conducted at room temperature.

The uptake of toxic ions on the sorbent at initial concentrations 0.1,1,5 and 10 mg/l were investigated. To study the effects of the sorbent dose 0.1,1,5. 10 and 20 g sorbent/ l of toxic solution was taken. To examine the effect of pH, the pH was varied from 2 to 8 when metal ions largely precipitate. To examine whether the filters used during the tests absorbed or released any Pb, several filtered and unfiltered samples were compared.

Sorption capacity (SC) of the substrate was expressed in term of amount of toxic sorbed on the unit sorbent mass (mg/g) and sorption efficiency of the system (% Removal) was indicated as the percent removal of toxic ions relative to the initial amount.

$$SC = \frac{Co - Ce}{Cs} = \frac{(Co - Ce)V}{m}$$

$$\% \ Removal \ = \frac{Co - Ce}{Co} \times 100$$

Where Co and Ce are respectively, the initial or original and final or equilibrium concentrations of toxic ions in solution (mg/l) and Cs is the concentration of the sorbent in the mixture (g/l), m is the mass of sorbent and V is the volume of the solution. The data were analysed using isotherm, kinetic and thermodynamic equations.

Kinetic Studies

Kinetic studies were conducted in continuously stirred tack reactors having laboratory -scale baffled batch sorbers. Each unit consisted of a 2-litre plastic vessel of internal diameter 0.13 m holding a volume of 1.80-liter toxic solution. Mixing was done by a six- bladed, flat plastic impeller of 0.065 m diameter and blade height of 0.013 mm. A variable speed motor will be used to drive the impeller using a 0.005 m diameter shaft. All the six plastic baffles were evenly spaced around the circumference of the vessel, positioned at 60° intervals and securely in place on top of the vessel that was used to obtain kinetic data and the variables namely effect of initial toxic ion concentration and effect of sorbent mass. The impeller speed was set at 350 rev/min. All components of the batch system design: impellers, baffles and vessels, were constructed from PVC to minimize metal ion sorption by fixtures. In all contact time investigations, 10 g of media was added to a stirred tank that contained 1.8 liters of toxic ion solution and there was started. 4 single component solution of 10 mg/l toxic concentration was drawn with used. At increasing time interval of 1 h, aliquots of 5 ml sample were drawn with a 10 ml plastic syringe up to a maximum of 10 h. The solution was immediately filtered into tubes, preserved with nitric acid and analyzed on the AAS.

Normalized toxic ions removal (Co -Ct) / (CO -Ce) was compared with the overall percentage removal (Co -Ct)/Co for various times throughout the run in the single and multi-toxic systems. The experimental data were analysed using four sorption kinetic models: the pseudo first order, Ritchie second order, the modified second order and Elovich equations.

Column Process

In order to establish the usefulness of PTS for in-line removal of toxins from wastewater, column studies were conducted. Continuous downflow experiments were conducted using PVC column (100) cm height x2.5 cm i. d). for each contaminant. Five columns were placed in series that could provide different empty bed contact times (EBCTs of 10,20,30 and 50 min.) and bed heights (BHs of 15,30,6, and 75 cm.). The EBCT is the average residence time and is obtained by dividing the bed volume including voids by the flow rate. The columns may be inclined differently to vary flow rates naturally without pumping. All the columns were packed with previously analysed sorbent on a small plastic wire mesh support. To study the effect of influent concentration 100 ml of each 0.1.1, 5 and 10 mg/l solutions of a toxic were passed at the rate of 10 ml/min. through columns each having the BH 15 cm at a pH of 7,2 ± 0.2. The effect of flow rate

was investigated using a column of 75 cm BH and varying the flow rate from 10 to 50 ml/min. The effect of pH was studied by changing the pH of the feed from 2 to 8. To determine exhaustive capacity in all the experiments the eluant was collected after breakthrough and analysed for the toxic. The initial amount of the metal in 20 ml feed minus the amount found in the effluent gave the amount of the toxic retained by the sorbent. The process was continued until the amount of toxic is same in 20 ml of the feed and the effluent. The % amount of effluent toxic removal was calculated, and the data were analysed statistically.

Desorption Study

In order to make adsorption process more economical, it is necessary to regenerate the spent sorbent. Attempts were made to desorb toxics from the loaded sorbents using various molarities of HCI, HNO3, NaOH and other chemicals. The regeneration cycle was repeated three times. After each cycle the sorbent was Washed with distilled water and dried.

Results and Discussion

Table 2. Removal % of Pb at different concentrations and at sorbent dose 5 Sg/l, pH 6.0, contact time 5 h, temperature 25"C and rpm 150.

Concentration, mg/l	IRA	PTS	PAC	PLC	FeO(OH)	RH
0.1	98.1	95.6	96.7	94.0	85.3	60.3
1	95.6	96.5	94.3	93.2	81.3	52.5
5	94.3	93.9	92.4	86.6	75.3	51.4
10	93.4	90.3	89.3	85.6	53.4	50.2

Table 3.   Removal % of Pb at different concentrations and at bed height (BH) 60 cm, pH 6.0, flow rate 10 ml/min and temperature 25'C by column process

Concentration, mg/l	IRA	PTS	PAC	PLC	FeO(OH)	RH
0.1	79.2	76.5	75.4	73.5	66.2	36.4
1	76.6	75.1	72.1	70.2	53.4	33.8
5	74.5	71.4	71.9	65.8	43.9	32.1
10	75.4	69.7	71.3	64.1	25.4	23.8

Table 4. Removal % of Pb at different pH values and at initial concentration 10 mg/l, sorbent dose 5g/1, contact time 5 h, temperature 25°C and rpm 150

pH	IRA	PTS	PAC	PLC	FeO(OH)	RH
2	69.3	65.3	66.0	60.1	35.2	29.1
4	84.3	79.4	83.3	78.3	48.3	40.1
6	93.4	90.3	89.3	85.6	53.4	50.2
8	92.4	89.2	88.6	84.8	52.7	48.3

Table 5. Removal % of Pb at different pH values and at initial concentration 10 mg/, BH 60 cm, flow rate 10 ml/l and temperature 25°C by column process

pH	IRA	PTS	PAC	PLC	FeO(OH)	RH
2	50.8	44.3	48.3	41.8	16.5	14.8
4	63.5	58.8	54.4	53.4	27.4	22.5
6	75.4	69.7	71.3	64.1	25.4	23.8
8	73.2	67.8	70.1	61.4	23.4	24.9

Table 6. Removal % of Pb at different sorbent doses and at initial concentration 10 mg/l, pH 6.0, contact time 5 h, temperature 25°C and rpm 150

Sorbent dose(g/l)	IRA	PTS	PAC	PLC	FeO(OH)	RH
0.1	84.1	74.3	80.1	69.8	45.2	45.1
1	84.5	79.4	83.3	78.3	49.2	40.1
5	93.4	90.3	89.3	85.6	53.4	50.2
10	96.6	95.1	97.8	90.3	50.2	44.8

Table 7. Removal % of Pb at different BHs and at initial concentration 10 mg/l, pH 6.0, flow rate 10 ml/min and temperature 25°C by the column process

Bed Height, cm	IRA	PTS	PAC	PLC	FeO(OH)	RH
10	65.4	56.3	57.5	48.4	15.4	13.6
30	67.5	58.6	55.7	53.9	23.3	20.5
60	75.4	69.7	71.3	64.1	25.4	23.8
75	78.8	76.4	78.6	72.6	31.9	30.3

Table 8. Removal % of Pb at different agitation times and at initial concentration 10 mg/l, sorbent dose 5g/1, pH 6.0, temperature 25°c and rpm 150.

Contact time(h)	IRA	PTS	PAC	PLC	FeO(OH)	RH
1	63.5	62.8	64.3	60.1	35.2	32.3
3	75.9	73.3	72.3	70.4	45.2	39.8
5	93.4	90.3	89.3	85.6	53.4	50.2
24	94.6	91.1	90.8	86.3	54.2	51.3

Table 9. Removal % of Pb at different flow rates and at initial concentration 10 mg/l, BH 60 cm, pH 6.0 and temperature 25°C by column process.

Flow rate, ml/min	IRA	PTS	PAC	PLC	FeO(OH)	RH
10	75.4	69.7	71.3	64.1	25.4	23.8
20	71.6	55.4	58.3	57.2	23.5	18.5
30	69.4	51.2	53.6	52.0	22.6	17.1
50	69.2	46.3	45.5	42.3	21.9	15.1

Conclusion

In an attempt to evaluate the suitability of synthesized pyroaurite like compound (PLC), modified PLC, i.e., pyroaurite type sorbent (PTS) and adsorbents such as powdered activated carbon (PAC), rice husk (RH) and goethite(FeO(OH) and a cation exchange resin Ceralite IR 120 (Na form, 20-50 mesh) for the removal of divalent lead from aqueous solution, a comparative study of these six sorbents was carried out by batch as well as column processes. The relative rate for lead adsorption was: IRA > PTS > PAC > PLC >FeO(OH)> RH. Modeling lead adsorption by different adsorbents using the Langmuir and Freundlich isotherm expressions helps compare the affinity of each adsorbent for lead and determine the adsorbents capacity for lead removal from aqueous media. The data best fitted the Langmuir isotherm expression where every adsorption site is theoretically equivalent and independent. The PTS data best fitted the Freundlich isotherm expression where adsorption sites are theoretically nonuniform, either due to pre-existing differences or due to repulsive forces between adsorbed molecules. The effects of various parameters affecting the adsorption such as initial metal concentration, sorbent dose, contact time and pH Were determined. Adsorption decreases with rise in lead concentration and pH but increases with increase in adsorbent dose and contact time. Equilibrium was attained in 5 h and the maximum removal was at pH 6. Thus acidic medium favored the removal. The values of specific reaction rate or velocity coefficient k Calculated for all the sorbents and they were found constant when calculated using the first order equation. The absorption resulted in a significant decrease in Gibbs energy pointing to a spontaneous and energetically favorable process. The average value of standard entropy change were also negative showing that the adsorption of lead on sorbents was accompanied by a decrease in entropy. Thus the adsorption process is exothermic and spontaneous as well as thermodynamically favorable.

In the column process also the order of lead removal capacities for these chemical adsorbents was found to be IRA > PTS> PAC > PLC > FeO(OH) > RH. This is probably due to several factors such as molecular size, molecular polarity, pH. lead reactivity and retentivity, sorbent properties and environmental conditions. The leaching order should be reverse of the sorption order. The sorption of lead ions goes on increasing as their concentration decreases from 10to 0.1 mg/l. Thus, the removal was about 20 % less than what it was in the batch process Leaching increases as the flow rate increases. Solution pH significantly affected the extent of contaminant removal. The results showed variation of percent removal at various pH values. A specific relation of optimum pH was observed. Dilute HNO; can be profitably used to regenerate the spent sorbent.

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