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Adsorptive removal of lead (II) from aqueous solution by low cost adsorbents: A review

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ABSTRACT

The present review paper focused on the removal of toxic lead (II) ions from aqueous solution by various types of adsorbents using cost effective and highly efficient adsorption technology. Industrial waste material, agricultural and forest waste, dead plant residue and biomass, natural minerals, chemically modified substances are employed as low cost adsorbents. Numerous operating parameters like effect of metal ion concentration, pH, adsorbent dose, contact time, stirring rate and temperature on removal efficiency of lead (II) ions from aqueous solution using low cost adsorbents has been presented in this paper. In addition equilibrium, kinetics and thermodynamics of adsorption has also been discussed.

KEYWORDS: Adsorption, Equilibrium, Kinetics, Removal, Contact time

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INTRODUCTION:

Statement of pollution problem:

Heavy metals in water have been a major preoccupation for researchers for many years because they are considered as the most toxic environmental pollutants¹. With the rapid development of industries such as metal plating, mining operations, refining ores fertilizer industries, tanneries, batteries, paper industries and pesticides, etc., heavy metals loaded wastewater are directly or indirectly discharged into the environment increasingly, especially in developing countries and possess a serious threat to environment^{2,3}. The major toxic and hazardous metal for humans as well as other forms of life is zinc, copper, nickel, mercury, cadmium, lead and chromium. These heavy metals are of specific concern due to their toxicity, bio-accumulation tendency and persistency in nature⁴⁻⁶. The effects of heavy metals in water and wastewater range from beneficial through troublesome to dangerously toxic, depending upon the concentration of heavy metal.

Lead is rarely found as the free metal in nature, but it is present in several minerals, principally in galena (PbS), the major source for lead production. It is also found as anglesite (PbSO₄) and cerrusite (PbSO₃).

Lead is one of the most commonly used non-ferrous metals. It is extensively used in process industries, such as acid battery manufacturing, metal plating and finishing, ammunition, tetraethyl lead manufacturing, ceramic and glass production, printing, pigment manufacturing, petrochemicals, fuel combustion and photographic materials⁷. Such industries continue to pose a significant risk to workers, as well as surrounding communities.

Lead can exist in two oxidation states, lead (II) and lead (IV), with the divalent form being the more dominating and stable species in most aquatic environments. The speciation of lead compounds in water is dependent upon a number of factors, principally pH, dissolved oxygen and the concentration of other organic and inorganic constituents in aqueous streams. In surface waters, lead actually exists as hydrated lead (II), or [PbCO₃(aq)]⁰ in the pH range 7–9. However at pH 6, concentrations of lead (II) and Pb(OH)⁺ are equal, whereas at higher pH values there is an increase in the concentration of lead in form of Pb(CO₃)₂²⁻ and [Pb(OH)⁺]⁸.

Health effects of heavy metals:

Heavy metal ions have lethal effects on all forms of life and these enter into the food chain through the disposal of wastes in water channels. Due to non-biodegradability, metal ions accumulate and their amounts are increased along the food chain. Hence, their toxic effects are more pronounced in the animals at higher trophic levels. Toxicity of lead includes Anaemia, brain damage,

anorexia, malaise, loss of appetite, diminishing IQ^{9,10}. Permissible limit of lead ions according to various environmental monitoring authorities are presented in Table 1.

Table 1. Permissible limits of lead (II)

Metal contaminant	Permissible limits for industrial effluent discharge (in mg/l)		Permissible limits by international bodies (µg/l)	
	Into inland surface waters Indian Standards: 2490 (1974)	Into public sewers Indian Standards: 3306 (1974)	WHO	USEPA
Lead	0.10	1.00	10	05

Source: (Reference 11).

Different methods of treatment:

Heavy metal removal from inorganic effluents can be achieved by conventional treatment processes such as chemical precipitation, reverse osmosis, ultrafiltration, coagulation–flocculation, electrodialysis, flotation, ion exchange and adsorption. However, these processes have their inherent advantages and limitations in application in terms of energy and chemical products consumption. The summary of these methods will be discussed in the following sections. Among other processes for removal of heavy metals from industrial effluent, adsorption has been shown as the most appealing in terms of economic and an environmental friendly procedure.

Previous studies:

The adsorption of lead (II) onto the surface of microwave–assisted activated carbon prepared from *Acacia auriculiformis* scrap wood char was studied through a two–layer feedforward neural network¹². The input variables for the proposed network were solution pH, contact time, initial adsorbate concentration, adsorbent dose and temperature, whereas the output variable was the percent lead (II) removal. The model predicted and experimental values of the percent lead (II) removal were found to be in reasonable agreement with each other.

A thermochemical method was performed by Pitsari et al. (2013)¹³ to enhance lead adsorption of unbleached newspaper pulp. Unbleached pulp was then chemically modified with citric acid (CA) at 90 °C for 90 min. The parameters under investigation were the pH of aqueous solution (3–9), the concentration of mechanically and chemically treated pulps (0.1–1 g), the concentration of CA (0.5 M and 1 M), the initial concentration of lead (20 –1000 mg/L) and the pulp–lead solution contact time (0 –24 h). It was observed that maximum adsorption capacity of mechanically treated newspaper pulp (MT–pulp) increased by 35% (25.71 mg/g) after modification with 0.5 M CA and

82% (34.6 mg/g) with 1 M CA. Maximum lead adsorption occurred at pH = 6. The adsorption experimental data fitted well to the Langmuir isotherm model. Kinetic studies for three different concentrations of lead (II) showed that the adsorption process involved two stages and the experimental data followed the pseudo-second order model.

Deng et al. (2013)¹⁴ explored the feasibility of using ethylenediamine-modified attapulgite (EMATP) as the sorbent for lead removal from water. The amine-modified attapulgite clay extracted from Xuyi, Jiangsu in China exhibited a higher sorption affinity for aqueous Lead (II) ions. Moreover, the sorption affinity for lead (II) ions was not affected by the presence of and the alkali metal ions (Na(I). The sorption kinetic of lead (II) was well described by the pseudo-second order kinetic model, while the sorption isotherm followed the Langmuir model. On acid treatment the lead loaded amine-modified attapulgite could be regenerated.

Ramesh et al. (2013)¹⁵ investigated the sorption of lead (II) in aqueous solution onto hydroxyapatite (HA) surfaces. The effect of contact time, HA dosage, and initial pH on removal efficiency were investigated. The optimum dose of HA for lead (II) removal was found to be 0.12 g/L (removal efficiency of 97.3%) at an equilibrium contact time of 60 min. It was reported that the adsorption kinetics of the lead (II) on HA followed the pseudo second-order model. All the isotherms fitted well for experimental data. Capacity of HA was found as 357.14 mg lead (II)/g of HA.

Peganum harmala seeds were assessed as biosorbent for removing lead (II) ions from aqueous solutions¹⁶. The effects of various parameters such as the aqueous solution pH, contact time, initial metal ion concentration and the amount of adsorbent in the process were investigated. The adsorption efficiency increased with pH. It was found that about 95% of lead ions could be removed from 45 mL of aqueous solution containing 20 mg/L of lead cation with 2 g of adsorbent at pH 4.5 after 15 min. Kinetic investigation of the process was performed by considering a pseudo-second-order kinetic model. It was found that experimental data was better described by Freundlich model.

Lang et al. (2013)¹⁷ examined the biosorption of lead using the dead detergent pre-treated chitosan-immobilised and grown fungal beads with initial lead (II) nitrate concentrations ranging from 9.02 to 281.65 mg/L. The adsorption data were best correlated with equilibrium adsorption isotherms in the order Redlich-Peterson, Langmuir, Freundlich and Fritz-Schlünder by non-linear regression method. The biosorption kinetics of lead (II) was better discussed by the pseudo second-order ($R^2 > 0.99$) model. According to Weber and Morris the rate-limiting step was shown to be intraparticle diffusion controlled.

The removal of lead (II) from wastewater was studied by using clay as the adsorbent through a membrane approaching natural adsorption technique¹⁸. The lead adsorption time was about 3 days.

The adsorption of lead (II) onto clay, obeyed the Langmuir, Freundlich isotherms with high regression coefficients.

Gerola et al. (2013)¹⁹ examined the efficiency of residues of passion fruit skin as biosorbent materials and also evaluated their capacity to adsorb lead (II) ions in *natura* skin (SK-N) and two modified skins, with NaOH (SK-S) and with NaOH and citric acid (SK-SCA). Fourier transform infrared spectroscopy confirmed the chemical modification by a peak at $1,730\text{ cm}^{-1}$. After pH 4, the adsorbed percentage was practically constant. Equilibrium was reached after 170 min. Kinetics followed the behavior described by the pseudo-second-order equation. The maximum adsorption capacity was 204 mg/g for the SK-SCA biomass. The residues followed Langmuir adsorption model. Through thermodynamic parameters (negative values of Gibbs' energy) it was verified that adsorption occurs spontaneously. Moreover, desorption studies showed that adsorbed ions may be recovered in two cycles.

Saka et al. (2012)²⁰ made a review work on the application of agricultural and forest waste adsorbents. All the adsorbents were compared with each other by metal binding capacities, metal removal performances, sorbent dose, optimum pH, temperature, initial concentration and contact time. It was shown that these adsorbents had sufficient binding capacity to remove lead (II) ions from wastewater.

Mohamed and Layla, (2012)²¹ reported the use of *Acacia tortilis* leaves as low cost adsorbent to reduce the lead cations from an aquatic environment. The influence of initial pH and contact time on removal efficiency was studied under batch mode. The optimum pH was found to be 4. Isotherm study showed that Freundlich adsorption isotherm was better fitted for adsorption of cations than Langmuir adsorption isotherm. The process was exothermic in nature due to the negative value of ΔH° . At temperatures 303 K and 333 K, the system was spontaneous and the spontaneity decreased as the temperature increased. The maximum adsorption capacities were 704.8, 632.3, 437, 332 mg/g at 293, 303, 313, 323, 333 K, respectively.

Singha and Das, (2012)²² conducted a study to investigate the applicability of six different biosorbents namely rice straw, rice bran, rice husk, coconut shell, neem leaves, and hyacinth roots for the removal of lead (II) ions from aqueous solution and effluent from battery industry. The sorption kinetic data was best described by pseudo-second-order model for all the biosorbents except rice husk which followed intraparticle diffusion model. Maximum monolayer sorption capacities onto the six studied natural sorbents were estimated from the Langmuir sorption model. The Elovich model, the calculated values of effective diffusivity, and the sorption energy calculated by using the Dubinin-Radushkevich isotherm indicated that the sorption process was chemical in

nature. The thermodynamic studies indicated that the adsorption processes were endothermic. Regeneration of biosorbents were carried out by desorption studies using HNO_3 .

The application of poly (acrylamide-co-sodium methacrylate) (AAM/SMA) hydrogel for the removal of lead (II) ions from aqueous solutions has been investigated using batch adsorption technique²³. The extent of adsorption was investigated as a function of solution pH, adsorbent dose, and temperature. The Fourier transform infrared (FTIR) spectra showed that $-\text{NH}_2$ and $-\text{COOH}$ groups are involved in lead (II) ion adsorption. It was found that the lead (II) ion adsorption followed pseudo-first-order kinetic model. Nonlinear regression analysis of six isotherms, Langmuir, Freundlich, Redlich-Peterson, Toth, Dubinin-Radushkevich, and Sips, have been applied to the sorption data, while the best interpretation was given by Redlich-Peterson model. Based on the separation factor, R_L and negative values of ΔG° adsorption was favorable and spontaneous.

The potential of a new green material obtained from *Portulaca oleracea* plant was investigated by Dubey and Shiwani, (2012)²⁴. Various batch experiments were carried out using different experimental conditions such as pH, contact time, adsorbent concentration and metal ion concentration to identify the optimum conditions. Results showed the optimum initial pH for adsorption as 6. Adsorption equilibrium was reached in 120 min. The adsorption data were modeled using both the Langmuir and Freundlich classical adsorption isotherms. Results showed ~78% removal of lead (II) from aqueous solution. The kinetic data fitted well with pseudo second-order model.

Response surface methodology (RSM) and artificial neural network (ANN) models were employed to develop prediction models for lead removal from industrial sludge leachate using red mud²⁵. Adsorbent dosage, contact time and pH were considered as independent experimental factors. Box-Behnken design (BBD) was chosen for the response surface design setup and was also used as Neural Network Training set for comparison purposes. The results of ANN were found to be more reliable than RSM in terms of better statistical parameters.

Adsorption of lead (II) from wastewater effluents using indigenous cellulose based biosorbent, such as nipa palm nut (NPN), palmyra palm nut (PPN), oil palm empty fruit bunch (EFB), oil palm fibre (OPF), and oil palm shell (OPS) have been tested by Nwabanne and Igbokwe, (2012)²⁶. Chemical activation method was used in preparing the activated carbon. The influence of various factors namely pH, adsorbent dosage, contact time, initial metal ion concentration and adsorbent particle size were studied. The amount of lead (II) adsorbed increased with increase in adsorbent dosage, and contact time and decreased with increase in particle size and initial metal ion concentration. The optimum pH was found to be pH 6 for all the studied adsorbents. The result of

adsorption studies showed that activated carbons produced from OPS and NPN were the most efficient adsorbents for the removal of lead (II) from aqueous solutions. Equilibrium data fitted well to the Freundlich, Langmuir and Temkin isotherm models.

The adsorption of lead (II) ions onto chitosan beads and cross-linked chitosan beads has been investigated by Gyananath and Balhal, (2012)²⁷. The epichlorohydrin (ECH) was used as a cross-linking agent. Adsorption experiments were carried out as a function of pH, stirring time, adsorbent dosage and concentration of lead (II) ions. Maximum removal was observed at pH 6. The experimental data of the adsorption equilibrium correlated well with the Langmuir isotherm model. The uptake of lead (II) ions on chitosan beads was of 72.89 mg Lead (II)/g chitosan, while on ECH cross-linked chitosan beads, it was of 39.42 mg lead (II)/g chitosan.

Surchi, (2011)²⁸ conducted a study for the removal of poisonous lead (II) from artificially contaminated water by five low-cost natural adsorbents viz. chaff, rice husk, sesame, sun flower and tea waste. Kinetic study revealed that pseudo-first order model was suitable to explain the kinetics of adsorption and intraparticle transport was not the only rate-limiting step. The adsorption equilibrium data correlated well with Freundlich model with regression, R^2 , range from 0.947–0.993. The results showed that removal efficiencies of chaff, rice husk, sesame husk, sun flower husk and tea waste for lead ion removal were 85%, 90%, 100%, 86%, 98% respectively.

The potential efficiency of three acid treated activated carbons, prepared from plant biomass of *Colas edulis* shell (CAH), *Pentaclethra macrophylla* husk (GAH) and *Aucoumea klaineneia* sawdust (QAH), for the removal of lead (II) ions from aqueous solution, has been investigated by Eba et al. (2011)²⁹. The study was managed using pH, concentration of lead solution, adsorption process equilibrium time and temperature as parameters. The adsorption increased with the increase in pH. The experimental data were found to fit both Freundlich and Langmuir isotherms. CAH (3.52 mg/g) was found to possess a more large heterogeneous surface capacity than GAH (2.19 mg/g) and QAH (1.43 mg/g) for lead (II) ions adsorption, according to the Freundlich model. However, Langmuir model had given QAH (42.19 mg/g) higher monolayer surface capacity than GAH (27.25 mg/g) and CAH (22.62 mg/g). Adsorption kinetics followed pseudo-second-order model. Between 308 and 318 K the adsorption process was found to be spontaneous.

Mouni et al. (2011)³⁰ used low-cost activated carbon prepared from *Apricot stone* material by chemical activation with sulphuric acid for the removal of lead (II) from dilute aqueous solution. The studied parameters include physical and chemical properties of adsorbent, pH, adsorbent dose, contact time and initial metal concentrations. The optimum pH was found to be 6.0. The results indicated that the second-order kinetic model fitted well to the adsorption kinetic data. The

adsorption isotherm data was described by the Langmuir and Freundlich models. The estimated maximum adsorption capacity of lead ions adsorbed by *Apricot stone* activated with sulphuric acid was 21.38 mg/g.

Activated carbon fiber (ACF) was modified with acetates of sodium, potassium and lithium at concentration of 15% and tested as an adsorbent for the removal of lead (II) from aqueous solution³¹. The effect of acetate treatments of ACF was studied in adsorption behavior of lead ion. It was observed that acetate treatment reduced surface area and pore volume of ACF. The adsorption amount of lead ion on the modified ACF (MACF) was greater than that on the pristine ACF. The adsorbed lead could be fully desorbed by using 0.01 mol/L HNO₃ solution. The maximum adsorption capacity of lead ion on acetate lithium modified ACF was 165.1 mg/g and the adsorption isotherm followed Langmuir isotherm model better than the Freundlich isotherm model. The adsorption kinetic data could be described well by the pseudo-second-order kinetic equation.

Activated carbon prepared from the cones of the European Black pine was used as adsorbent for the removal of lead (II) ions from aqueous solutions³². The effect of pH, initial concentration of lead (II) ions, contact time, and adsorbent dosage on the removal efficiency was studied in a batch process. Langmuir isotherm provided the best fit to the equilibrium data with maximum adsorption capacity of 27.53 mg/g. The kinetic data were found to follow closely with the pseudo-second-order model. The microstructures of the activated carbon before and after adsorption of lead (II) ions were observed by scanning electron microscope. Surface structure was characterized by using Fourier transform infrared spectroscopy and Boehm titration.

The adsorption characteristics of lead (II) on pre-boiled treated onion skins (PTOS) and formaldehyde-treated onion skins (FTOS) were evaluated under batch mode³³. The effects of lead (II) initial concentration, agitation rate, solution pH, and temperature were investigated. Lead (II) adsorption was found to increase with increase in initial concentration. The point of zero net charge (PZC) was 6.53. The optimum pH for the maximum removal of lead (II) was 6.0. The adsorption equilibrium data was best represented by the Langmuir isotherm model for FTOS and the Freundlich isotherm model for PTOS. The maximum amounts of lead (II) adsorbed (q_{\max}), was 200 mg/g for FTOS. The efficiencies of PTOS and FTOS for lead (II) removal were 84.80% and 93.5% at 0.15 g/200 mL adsorbent dose, respectively. It was found that the adsorption kinetics of lead (II) obeyed pseudo-first-order kinetic model. Obtained activation energy (E_a) was 25.596 kJ/mol.

Ammonium citrate modified spent *Lentinus edodes* was investigated as a novel adsorbent for lead (II) removal from aqueous solution³⁴. The Fourier transform infrared (FTIR) spectroscopy and scanning electron microscopy (SEM) analysis confirmed that modification by ammonium citrate obviously changed the property of spent *L. edodes*. In batch studies, pH, initial lead (II)

concentration and adsorbent dose were found to significantly affect the removal efficiency of lead (II) by a two-level Plackett–Burman design. The effects of foreign substances (ion strength, inorganic ions (anions, cations) and surfactants) on lead (II) removal were studied. The adsorption kinetics were well described by the pseudo–second–order model. The adsorption equilibrium data followed the Langmuir model and gave the maximum adsorption capacity of 59.17 mg/g at 35 °C. The adsorption mechanism was found to be dominated by ion exchange and electrostatic attraction on exterior, accompanying with the intraparticle diffusion. The adsorbent could be regenerated by 0.1 M HNO₃.

The removal of lead (II) ions from aqueous solutions by chemically modified walnut shells was studied through a 2⁴ full factorial design analysis³⁵. The effects of solution pH, adsorbent dose, initial concentration of lead (II) ions, and temperature on metal removal efficiency were examined in a batch system. Analysis of variance (ANOVA), *F*-test and Student's *t*-test showed that removal of lead (II) ions were only slightly temperature dependent, but markedly increased with adsorbent dose and solution pH. The initial concentration of lead (II) ions had a relatively small negative effect on removal efficiency. Modified simplex method was used for the optimization of the statistically significant factors. The recommended optimum conditions were: adsorbent dosage of 13.5 g/L, solution pH of 6.3, initial lead concentration of 45.3 mg/L, with the lead (II) removal efficiency of 98.2 %.

Al-Zboon et al. (2011)³⁶ utilised synthesized geopolymer from waste coal fly ash as an adsorbent for lead (II) removal from aqueous wastewater. The effect of various parameters including geopolymer dosage, initial concentration, contact time, pH and temperature were investigated. The major components of the used ash were SiO₂, Al₂O₃ and Fe₂O₃ representing 91.53 wt % of its mass. It was found that the synthesized geopolymer has higher removal capacity for lead ions when compared with that of raw coal fly ash. The removal efficiency increased with increasing geopolymer dosage, contact time, temperature, and the decrease of lead (II) initial concentration. The optimum removal efficiency was obtained at pH 5. Adsorption isotherm study indicated that Langmuir isotherm model was the best fit for the experimental data. It was noted also that the adsorption process was endothermic and more favorable at higher temperatures.

The removal of lead (II) from aqueous solutions by modified areca waste with the Fenton reagent was investigated³⁷. Experiments were carried out as function of contact time and initial concentration (4–12 mg/L). The equilibrium adsorption data were well presented by the Freundlich and Langmuir adsorption isotherm models. The monolayer adsorption capacity was found to be 3.37 mg/g at pH 6.6 and 323 K. The negative value of ΔG^0 indicated the feasibility and spontaneity of the adsorption process. The positive ΔH^0 suggested the endothermal nature of the adsorption. The

positive values of ΔS^0 indicated the affinity of adsorbent towards lead (II). Lead loaded adsorbent can be regenerated using HNO_3

A new low-cost adsorbent, fallen *Cinnamomum camphora* leaves (FCCL) can be effectively used for the removal of lead (II) from aqueous solutions³⁸. The adsorption kinetic experiments followed pseudo-second order kinetics and is mainly controlled by the film diffusion mechanism. All equilibrium data obtained at different temperatures fitted perfectly with Langmuir isotherm models compared to Freundlich and D-R isotherm models, and the maximum adsorption capacities of lead (II) onto FCCL were found to be 73.15, 73.58, 74.13 and 75.82 mg/g at 303.2, 313.2, 323.2 and 333.2 K, respectively.

Sakthi et al. (2010)³⁹ carried out a batch adsorption study to investigate the isotherms, kinetics and thermodynamics of lead (II) ions from aqueous solution by activated carbon prepared from *Bombax ceiba* sawdust (SDC). The effects of pH, adsorbent dosage, contact time, initial concentration of lead (II) and temperature on the adsorption were studied. Maximum adsorption occurred at pH 5. The sorptive mechanism followed the pseudo second-order kinetics and intra particle diffusion model. The equilibration data fitted well with both Langmuir and Freundlich isotherm model with maximum sorption capacity of 209 mg/g. The mean free energy of adsorption calculated from Dubinin-Radushkevich (D-R) isotherm model indicated that the adsorption occurred by chemical ion exchange process. Thermodynamic parameters and Tempkin constant showed that the sorption process was feasible, spontaneous and endothermic under studied conditions.

Gorme et al. (2010)⁴⁰ reported that bottom ash could be used as an adsorbent for the removal of lead (II) from aqueous solutions. The adsorption rate increased with decreasing particle size. They showed that pseudo-second order kinetics was the most suitable model for describing the kinetic data, while the Freundlich isotherm best represented the equilibrium sorption onto bottom ash. The maximum adsorption capacity and energy of adsorption of bottom ash were 0.315 mg/g and 7.01 kJ/mol, respectively.

Mousavi et al. (2010)⁴¹ investigated the possibility of the utilization of waste tire rubber ash (WTRA) as a low cost adsorbent for removal of lead (II) ion from aqueous solution. The effect of different parameters (contact time, adsorbate concentration, adsorbent dosage, pH of the medium and temperature) were investigated. Equilibrium was reached after 90 min. Equilibrium data fitted well with the Langmuir model with maximum adsorption capacity of 22.35 mg/g. The adsorption kinetics was best fitted with first-order equation. Thermodynamic results showed that the lead (II) ion adsorption on WTRA was an endothermic and spontaneous process.

Carbonate hydroxyapatite (CHAP) synthesized from eggshell waste has been employed for removing lead ion from aqueous solutions⁴². The maximum uptake of lead ion was obtained at pH

6.0. Adsorption equilibrium was established in 60 min. The pseudo-second order kinetic model provided the best correlation ($R^2 > 0.9991$) of the used experimental data compared to the pseudo-first order and intraparticle diffusion kinetic models. The adsorption of lead ion increased with the initial concentration of lead ion. The maximum lead ion adsorbed was found to be 101mg/g. It was found that the adsorption of lead (II) on CHAP was correlated well ($R^2 = 0.9995$) with the Langmuir isotherm model.

Removal of lead (II) ions by adsorption onto bamboo dust and commercial activated carbons was carried out by Kanan and Veemraj in the year of 2009⁴³. The effect of various process parameters has been investigated by following the batch adsorption technique at $30 \pm 1^\circ\text{C}$. Percentage removal of lead (II) ions increased with the decrease in initial concentration and increased with increase in contact time and adsorbent dose but with the decrease in particle size of the adsorbent amount of lead (II) ions adsorbed increased.

Mehrasbi et al. (2009)⁴⁴ assessed the potential of almond shells for adsorption of lead (II) from aqueous solution. Almond shells were pretreated separately with 0.4 mol/L NaOH, 0.4 mol/L HNO_3 and distilled water and their adsorption abilities were compared. Batch adsorption experiments were carried out as a function of the initial metal ion concentration, pH and adsorbent dosage. The alkali-modified almond shells had adsorption capacities for lead (II) from 2 to 9 mg/g which was much higher than acid- and water-pretreated adsorbents. Experimental results showed that the best pH for adsorption was 5–6 and the extent of adsorption decreased with lowering pH. Isotherm models indicated the best fit for Langmuir model for alkali-modified almond shells.

Huang et al. (2009)⁴⁵ applied Taguchi method to determine the optimum condition for lead (II) removal from aqueous solution by spent *Agaricus bisporus*. An orthogonal array experiment design ($L_9 (3^4)$) which is of four control factors (pH, t (contact time), m (sorber mass), and C_0 (initial Lead (II) concentration) having three levels was employed. The optimum condition for biosorption capacity was found to be pH = 5.00, $t = 5.0$ h, $m = 0.010$ g, and $C_0 = 50$ mg/L. And for percent removal, the optimum condition was found to be pH = 4.00, $t = 4.0$ h, $m = 0.100$ g, and $C_0 = 50$ mg/L. Under these optimum conditions, biosorption capacity and percent removal were reported to be 60.76 mg/g and 80.50%, respectively.

In a study Senthilkumar and Gayathri, (2009)⁴⁶ showed that bael tree (BT) leaf powder could be used as an adsorbent for removal of lead (II) ions from aqueous solutions through batch equilibrium technique. Studies showed that removal efficiency increased with increasing solution pH. The monolayer adsorption capacity was 4.065 mg/g with the correlation coefficient of 0.993. The experiments showed that highest removal rate was 84.93% at solution pH 5, contact time 60 min and initial concentration of 50 mg/L. Thermodynamic parameters such as Gibbs free energy,

enthalpy and entropy suggested that the sorption process was feasible, spontaneous and exothermic in nature. It was also shown that the adsorption of lead (II) ions could be described by the pseudo-second order equation.

Naiya et al. (2009)⁴⁷ conducted a study for the adsorption of lead (II) from aqueous solution on rice husk ash as a function of various experimental parameters like pH, initial concentration, adsorbent dosage, contact time and the effect of temperature. Optimum conditions for lead (II) removal were found to be pH 5, adsorbent dosage 5 g/L of solution and equilibrium time 1 h. Adsorption of lead (II) followed pseudo-second-order kinetic model. The effective diffusion coefficient obtained in the order of 10^{-10} m²/s. The adsorption capacity (q_{\max}) of rice husk ash was 91.74 mg/g. The change of entropy (ΔS^0) and enthalpy (ΔH^0) were estimated at 0.132 kJ/(mol K) and 28.923 kJ/mol respectively. The negative value of Gibbs free energy (ΔG^0) indicated the feasibility and spontaneous nature of adsorption. The value of the adsorption energy (E), calculated using Dubinin-Radushkevich isotherm, was 9.901 kJ/mol.

Hasan et al. (2009)⁴⁸ successfully utilized biomass of *Aeromonas hydrophila* for the removal of lead (II) from aqueous solution. The effect of process variables such as pH, initial lead (II) concentration, biomass dose and temperature on the uptake of lead were investigated using two level four factor (2^4) full factorial central composite design with the help of MINITAB® version 15 software. The predicted results obtained were found to be in good agreement ($R^2 = 98.6\%$) with the results obtained by performing experiments. The multiple regression analysis and analysis of variance (ANOVA) showed that the concentration has positive and temperature and biomass dose have negative effect whereas pH has curved relationship with the uptake of lead (II). The maximum uptake of lead (II) predicted by optimization plots was 122.18 mg/g at 20 °C, initial lead (II) concentration of 259 mg/L, pH 5.0, temperature 20 °C and biomass dose 1.0 g. Langmuir isotherm model was applicable to sorption data and sorption capacity was found to be 163.3 mg/g at 30 °C, pH 5.0 and lead (II) concentration range 51.8–259 mg/L. Dubinin-Radushkevich (D-R) isotherm model showed the chemisorption ($E = 12.98$ kJ/mol) nature.

Biosorption of lead (II) ions from aqueous solutions was studied in a batch system by using *Candida albicans*⁴⁹. The extent of metal ion removal increased with increasing contact time, initial metal ion concentration and temperature. Biosorption equilibrium time was observed in 30 min. The maximum biosorption capacity of lead (II) on *C. albicans* was determined as 828.50 ± 1.05 , 831.26 ± 1.30 and 833.33 ± 1.12 mg/g, respectively, at different temperatures (25, 35 and 45 °C). Biosorption showed pseudo second-order rate kinetics at different initial concentration of lead (II) and different temperatures. From Arrhenius equation the activation energy of the biosorption (E_a) was estimated as 59.04 kJ/mol. Thermodynamic analysis results showed that biosorption of lead (II)

ions on *C. albicans* were endothermic and spontaneous. The optimum pH for lead (II) removal was determined as pH 5.0.

Kinetics adsorption of lead from aqueous solution using silica ceramic has been investigated in batch methods by Salim and Muneke, (2009)⁵⁰. The process of lead adsorption followed pseudo second-order rate expression and obeyed the Langmuir isotherm model with high correlation coefficient ($R^2 > 0.99$). The maximum adsorption capacity was found to be 2.7 mg/g.

Modified kaolinite clay with 25% (w/w) aluminium sulphate and unmodified kaolin were investigated as adsorbents to remove lead (II) from aqueous solution⁵¹. The results showed that amount of lead (II) adsorbed onto modified kaolin (20 mg/g) was more than 4.5-fold than that adsorbed onto unmodified kaolin (4.2 mg/g) under the optimized condition. It was observed that the adsorption data from both adsorbents fitted well to the Langmuir isotherm. The kinetic adsorption of modified and unmodified kaolinite clay fitted well to the pseudo-second-order model.

Singh et al. (2008)⁵² investigated the adsorption of lead (II) from dilute aqueous solution by low-cost activated carbon, prepared from Tamarind wood material by chemical activation with sulphuric acid. The parameters studied include physical and chemical properties of adsorbent, pH, adsorbent dose, contact time and initial concentrations. The kinetic data were best fitted to the Lagergren pseudo-first-order and pseudo-second order models. The isotherm equilibrium data were well fitted by the Langmuir and Freundlich models. The maximum removal of lead (II) was obtained 97.95% (experimental) and 134.22 mg/g (from Langmuir isotherm model) at initial concentration 40 mg/L, adsorbent dose 3 g/L and pH 6.5.

Răţoi et al. (2008)⁵³ carried out the adsorption experiments to evaluate the adsorption capacity of sphagnum moss peat (Poiana Stampei, Romania) for lead (II), as a function of several experimental parameters. The obtained results indicated that for an initial solution pH = 6.0 (acetate buffer), sphagnum moss peat was effective in removing up to 98% of lead. With the increasing of initial lead concentration, the residual lead (II) content from aqueous solution increase the same, but the removal percent was higher than 95% only in case of solutions with an initial concentration lower than 350 mg/L. In addition it was also observed that the retained lead ions could be easily leached from sphagnum moss peat using 0.5 N HCl solutions.. The results of recovery test showed that the constituents of water samples (Na(I), K(I), Ca(II), Mg(II), Copper (II), Zn(II), Co(II), Nickel (II)) did not interfere significantly with the lead removal by sphagnum moss peat.

The adsorption of lead (II) on to activated carbon derived from an indigenous Ethiopian medicinal plant leaves namely Birbira (*Militia ferruginea*) was investigated⁵⁴. The maximum adsorption (97.3 %) took place at 3 h, at a dose of 4.0 g of adsorbent and at pH of 4.0. The amount of lead ion adsorbed per gram of the adsorbent increased with decreasing concentration of lead (II) and

increased with the increasing temperature. The positive value of ΔH^0 and negative values of free energy (ΔG^0) indicated endothermic and spontaneous nature of adsorption. The Freundlich adsorption isotherm best fitted and calculated adsorption capacity was 3.3 mg of lead (II) per g of adsorbent. The adsorption followed the first order kinetics. The pH effect and desorption studies showed that ion exchange mechanism might be involved in the adsorption process. The removal of lead (II) from industrial wastewater sample was also tested and showed that more than 97% removal was attainable.

Adsorption of lead (II) ions from aqueous solution onto tobacco stems has been investigated to evaluate the effects of initial lead ion concentration, adsorbent dosage, contact time, pH and temperature on the removal of lead (II) systematically⁵⁵. The optimal pH value was found to be 5.0. The removal of lead ions for concentrations 10, 30 and 50 mg/L using 0.8 g adsorbent at contact time of 120 min and at temperature of 299K were 94.37%, 92.10% and 90.43%, respectively. The thermodynamics of lead (II) adsorption onto the tobacco stems indicated that the adsorption was spontaneous and endothermic.

Coconut shell carbon (CSC), a low cost sorbent derived from organic waste material, was used by Sekhar, (2008)⁵⁶. The results of the batch sorption studies indicated that the efficiency of lead removal by coconut shell carbon was comparable to that of commercially available activated carbon. From the kinetic and equilibrium studies, the sorptive capacity of coconut shell carbon for lead was found to be 30 mg/g.

The adsorption of lead (II) on mustard husk has been found to be dependent on heavy metal concentration, pH, contact time, adsorbent dose and temperature⁵⁷. The adsorptive behavior of lead (II) on mustard husk satisfied both the Langmuir and Freundlich assumptions. Ion exchange and surface complexation were the major adsorption mechanisms involved.

Badmus et al. (2007)⁵⁸ studied the removal of lead (II) ion from industrial wastewaters by activated carbon prepared from periwinkle shells (*Typanotonus fuscatus*). Activated periwinkle shell carbon (PSC) was prepared. The performance of PSC was compared with that of commercial activated carbon (CAC) and a mixture of PSC and CAC (PSC: CAC) in a ratio 1:1. The effect of various parameters such as adsorbent dose, contact time, pH, agitation speed, and particle size of the adsorbent was studied to optimise the conditions for maximum adsorption. It was found that the adsorption mechanisms in the lead/adsorbent system followed pseudo-second-order kinetics with a significant contribution from film diffusion. The adsorption isotherms model fitted both with Langmuir and Freundlich isotherms. The percentage removal of lead onto PSC, PSC:CAC, and CAC were 82.78 %, 92.68 %, and 88.44 %, respectively.

The removal of poisonous lead (II) from wastewater by different low-cost abundant adsorbents (Rice husks, maize cobs and sawdust) was investigated by Abdel-Ghani et al. (2007)⁵⁹. The adsorption efficiencies were found to be pH dependent, increased by increasing the solution pH in the range from 2.5 to 6.5. The equilibrium time was attained after 120 min and the maximum removal percentage was achieved at an adsorbent loading weight of 1.5 g. The equilibrium adsorption data was well described by Temkin isotherm model.

Hashem, (2007)⁶⁰ performed the adsorption experiments by okra wastes. The optimum pH for lead removal was between 4 and 6, the percentage of lead removal at equilibrium increased with increasing the amount of okra wastes and temperature. The removal of lead (II) ions attained 99%.

The adsorption characteristics of lead (II) onto expanded perlite (EP) from aqueous solution were investigated with respect to the changes in pH of solution, adsorbent dosage, contact time and temperature of solution⁶¹. The Langmuir isotherm model fitted to equilibrium data better than the Freundlich isotherm model. The monolayer adsorption capacity of EP was found to be 13.39 mg/g for lead (II) ions. From Dubinin-Radushkevich (D-R) isotherm model, the mean free energies of adsorption was found as 9.12 kJ/mol for lead (II) indicating chemical ion-exchange mechanism. Thermodynamic parameters showed that the adsorption lead (II) ions onto EP was feasible, spontaneous and exothermic at 20–50 °C. Pseudo-second-order kinetics better described adsorption phenomenon.

CONCLUSION

This review article provides an extensive overview for the removal of lead (II) ions from aqueous solution by different types of adsorbents instead of expensive commercial adsorbents. It was found from the review that lead (II) ions removal is highly sensitive to the variation of solution pH as well as initial lead (II) ion concentration, adsorbent dose and temperature. It was observed that removal percentage gradually increases with adsorbent dose and contact time and decreased with initial lead (II) concentration, pH and temperature. Response surface methodological based review study indicated closeness of experimental removal percentage with that of the software output value.

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