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Chemical Remediation of Soil Contaminated with Heavy Metals by Washing with EDTA

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Abstract. Removal of heavy metals from contaminated soil is very essential because these are toxic to biological systems. Chemical extraction is a rapid, cost-effective and environment friendly ex- situ technique for removal of heavy metals from contaminated soil. Due to industrial processes and improper disposal of garbage soil get contaminated with lead also which has very harmful effect on human being therefore method for removal of lead from contaminated soil by using Ethylene-DiamineTetraacetic Acid (EDTA) solution is optimised. Efficiency of lead extraction by soil washing with EDTA solution is dependent on pH and concentration of EDTA solution, washing time and liquid-soil ratio. Optimum condition for removal of lead from contaminated soil were obtained by adding EDTA solution of concentration of 0.01M and liquid-soil ratio of 20 to 2 g of soil sample and were agitated at room temperature for 2 hours.

Keywords: EDTA, Contaminated Soil, Extraction Efficiency, Heavy Metal

1 Introduction

Lead contamination in soil is due to improper disposal of sewage and wastes from industrial activities such as mining, smelting, reclamation of lead from batteries, automobile applications. Lead may get swept away from contaminated areas to new areas by wind and rain. Due to gravitation and rain, airborne lead of automobiles, shooting range and lead smelting refiners settle on soil. Ground water gets contaminated on leaching of lead from the lead-contaminated soils. In soil, lead and its compounds are very stable due to their low solubility [1-4]. Lead is toxic to human and causes anemia, kidney damage, increase in blood pressure, etc. High levels of exposure to lead may result in coma and may progress to death [5].

Preventing heavy metal contamination of soil is very essential because removing heavy metals from contaminated soils is very difficult and expensive. Methods for remediation of contaminated soil for reducing the harmful effects of heavy metal include excavation, phytoremediation and chemical extraction [6]. Chemical extraction (soil washing) is reported to be a rapid, cost-effective and environment friendly ex- situ technique for removal of heavy metals from contaminated soil [7]. Washing of soil with water can remove only small amount of heavy metals because they occur in sorbed state and are sparingly soluble in water. For effective removal of heavy metals from contaminated soil, some chemicals are added to the washing water which binds with heavy metal to form water soluble compound thus transfer heavy metals from the soil into washing solution [8-10]. The amounts of heavy metal extracted from the contaminated soil depend on the nature of soil and the extracting agent [11,12]. When soil contaminated with heavy metal is washed with aqueous solution of chelating reagent (chitosan, EDTA, nitrilotriacetic acid, ferric chloride), it bind with metal to form complex which is soluble in water therefore metal get removed from the contaminated soil [13,14].

Ke et al. reported that tartaric acid is an environmentally-friendly extractant for remediation of heavy metal contaminated soils. Within 24 h, tartaric acid in the pH range of 3.5-4.0 was effective for removal of 50%-60% of Cd, 40%-50% of Pb, 40 %-50% of Cu and 20%-30% of Zn from the contaminated soil [15]. The best removal efficiencies for Cd, Cu and Zn were obtained when soil was washed with 0.01M FeCl3 as washing solution with liquid-soil ratio of 20 on rotary shaker at 200±5 rpm for 2 h. On increasing washing time from 0.5 to 2 h percent removal of Cd increased from 83.14 to 98.18. Lim et al. reported that 95% of Pb, 89% of Cd and 90% of Ni was removed from contaminated soil [16].

EDTA is widely used for washing of contaminated soil because of its ability to form water soluble complex with almost all heavy metal [17-21]. FeCl3 and EDTA were used by Bilgin et al. to extract Cu, Cd and Zn from the contaminated soil [22]. After 1 h of soil washing, 90.57% of Cd, 73.22 % of Cu and 99.59% of zinc were removed from contaminated soil using 0.01 M EDTA as washing solution. Udovic and Lestan reported that 73 % of Pb, 23 % of Zn and 74 % Cd were removed from contaminated soil by using EDTA as washing solution [23]. Jiang et al. extracted copper and nickel from contaminated soil by washing it with chitosan, EDTA and sodium citrate [24]. Extraction efficiency of any of the chelating agents was higher for nickel than for copper which is consistent with the relative stability of chelate complexes of these two metals.

2 Materials and Method

In the present study, 100 gm of soil (2mm passing) obtained from Jharkhand was spiked with 500 mg lead nitrate. To minimize the discrepancy between the field-contaminated soil and the artificial-contaminated soil, the soil samples were equilibrated for 2 weeks in wet condition followed by dry aging for 4 weeks. Soil sample spiked with lead nitrate was characterized and extraction experiments were carried out with EDTA solution to study the effect of parameters (pH and concentration of EDTA solution, washing time and liquid-soil ratio) on its extraction efficiency for removal of lead from contaminated soil. AR grade EDTA was used for this study. To study the effect of pH on extraction efficiency of EDTA for removal of lead from contaminated soil, three samples were prepared in which 20 ml of 0.01M EDTA solution was added to 2 gm of spiked soil and pH of three samples were adjusted to 4, 7 and 9 respectively by using buffer solution. These solutions were stirred for 2 hours and filtered. Concentration of lead in filtrate was determined by using atomic absorption spectrophotometer. Extraction efficiency of EDTA for removal of lead from contaminated soil was highest at pH 4 so all other experiments were done at pH 4. To see the effect of washing time on extraction efficiency of EDTA for removal of lead from contaminated soil, second set of experiments were done in which 20 ml of 0.01M EDTA solution was added to 2 gm of spiked soil and pH was adjusted to 4 by using buffer solution. These solutions were stirred for 4 and 6 hours respectively and filtered. Concentration of lead in filtrate was determined by using atomic absorption spectrophotometer. Extraction efficiency of EDTA for removal of lead from contaminated soil obtained from the above set of experiment was similar to that obtained for washing time of two hours at pH 4 so other experiments were done at pH 4 and washing time of 2 hours. To see the effect of concentration of EDTA solution on extraction efficiency of EDTA for removal of lead from contaminated soil, third set of experiments were done in which 30 ml, 40 ml and 50 ml of 0.01M EDTA solution was added to 2 gm of spiked soil and pH was adjusted to 4 by using buffer solution. These solutions were stirred for 2 hours and filtered. Concentration of lead in filtrate was determined by using atomic absorption spectrophotometer. To see the effect of liquid-soil ratio on extraction efficiency of EDTA for removal of lead from contaminated soil, fourth set of experiments were done in which 20 ml of 0.01M EDTA solution was added to 2 gm of spiked soil, 10 ml, 20 ml, 30 ml and 40 ml of water was added respectively and pH was adjusted to 4 by using buffer solution. These solutions were stirred for 2 hours and filtered. Concentration of lead in filtrate was determined by using atomic absorption spectrophotometer. Experimental details and result are presented in **Error! Reference source not found.**.

Table 1. Experimental data for removal of Lead from contaminated soil.

Sample no.	Volume of EDTA	Volume of water	Washing time	Ph	% Pb removed
1	20 ml		2hr	4	62.75
2	20 ml		2hr	7	55.63
3	20 ml		2hr	9	49.16
4	20 ml		4hr	4	63.13
5	20 ml		6hr	4	63.47
6	30 ml		2hr	4	67.72
7	40 ml		2hr	4	67.87
8	50 ml		2hr	4	67.93
9	20 ml	10 ml	2hr	4	68.11
10	20 ml	20 ml	2 hr	4	68.87
11	20 ml	30 ml	2 hr	4	68.93
12	20 ml	40 ml	2 hr	4	68.96

3 Results and Discussion

As is evident from the results shown in **Error! Reference source not found.**, removal of lead with EDTA is very efficient because the formation of soluble Pb-EDTA complex is thermodynamically favourable. The experimental results indicate that efficiency of metal extraction by soil washing with EDTA is dependent on pH of washing solution. On comparison of the percentage lead removal of sample no. 1 to 3, it is found that percentage removal of lead from the contaminated soil is increased on decreasing the pH of the wash solution from 9 to 4 and extraction was most efficient when its pH was 4 because availability of heavy metal is increased in acidic condition. Percentage removal of lead at higher pH is decreased because solubility of lead-EDTA complex is decreased as pH increased.

At pH 4, after 2 h of washing of contaminated soil with EDTA solution, 62.75% of lead was removed. Extending the reaction time from 2 h to 6 h did not affect the extraction efficiency significantly. A reaction time of 2 h was chosen as the optimal reaction time for extraction of lead with EDTA solution. On comparing experimental results of sample no. 1 and 6, it is evident that extraction efficiency of lead is increased on increasing concentration of EDTA solution. Percentage removal of lead from contaminated soil increased from 62.75% to 67, 72% when volume of 0.01m EDTA solution increased from 20 ml to 30 ml but as is evident from experimental results of sample no. 7 and 8 percentage of lead removal did not increase on further increasing the volume of 0.01m EDTA solution. On comparing experimental results of sample no. 6, 9 and 10 it is clear that on increasing liquid -soil ratio extraction efficiency of lead is increased from 62.75 % to 68.87 %. Experimental results of sample nos. 11 and 12 show no significant increase in extraction efficiency on further increase in liquid-soil ratio so liquid-soil ratio of 20 can be considered as optimum liquid-soil ratio.

4 Conclusions

Efficiency of lead extraction by soil washing with EDTA solution is dependent on pH and concentration of EDTA solution, washing time and liquid-soil ratio. Optimum condition for removal of lead from contaminated soil were obtained by adding EDTA solution of concentration of 0.01M and liquid-soil ratio of 20 to 2 g of soil sample and was agitated at room temperature for 2 hours. High liquid-soil ratio can prevent clogging of the soil during washing but generate large amount of wastewater, which would increase the cost of soil washing. EDTA is a hexadentate ligand and coordination number of lead ion is six so six-coordinate Pb-EDTA is a very stable complex. Molar concentration of EDTA needed to extract lead from contaminated soil, should be the same as the molar concentration of lead in the soil but EDTA is a non-specific chelating agent and it reacts with other metals present in soil so removal efficiency of lead is reduced. Maximum lead extraction from Pb-contaminated soils can be achieved, if solution with an EDTA molar concentration higher than the molar concentration of lead in soil is used as well as extraction efficiency of lead is also dependent on type of soil. In further studies, effectiveness of other chelating agents such as diethylenetriaminepentaacetic acid, nitrilotriacetic acid, ferric chloride will be examined as extractant for removal of lead from contaminated soil.

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