Extraction of Lead from Artificially Contaminated Soil by EDTA and Optimization of Removal Process Using Taguchi Orthogonal Array Design

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Abstract— **Removal of lead from artificially contaminated soil by ethylene di amine tetra acetic acid was investigated. The effect of operating parameters affecting the extraction efficiency such as liquid to solid ratio, pH, strength of solution and washing time was studied and optimized using Taguchi factorial design. Optimal conditions were achieved as liquid to solid ratio 40 ml/g, pH 4, strength of solution 0.5 g/l and washing time of 3 hours. Also the removal efficiency was obtained 99.08%.**

Keywords- Contamination ,Heavy metal, Soil washing ,EDTA, Taguchi orthogonal design

I. INTRODUCTION

Pollution of the natural environment by heavy metals is a worldwide problem because these metals are permanent and most of them have toxic effects on living organisms when they exceed a certain concentrations. The rapid development of industry and the use of chemical substances in many industrial activities have caused a steady increase of trace- metal pollution in soil. Soils receive potentially toxic elements from both natural and wide range of anthropogenic sources, including the weathering of primary minerals, mining, fossil fuel combustion, the metallurgical, electronic, and chemical industries, and waste disposa [12]l.

The ecological effects of heavy metals in soil are closely related to the distribution of species in the solid and liquid phase of the soil. Metals such as lead, mercury, cadmium and copper are cumulative poisons. These metals cause environmental hazards and are reported to be exceptionally toxic. Plants take up metals by absorbing them from contaminated soils, as well as from deposits on parts of the plants exposed to the air from polluted environments. Prolonged consumption of unsafe concentrations of heavy metals through foodstuffs may lead to the chronic accumulation of heavy metals in the kidney and liver of humans causing disruption of numerous biochemical processes, leading to cardiovascular, nervous, kidney and bone diseases[19]. Total metal content can be a starting point for developing control strategies to obtain better environmental quality and is a key to assess soil quality.

It is technically difficult and expensive to reduce the heavy metal content in soil to an extremely low level. But if a reasonable clean up goal is set, the decontamination would be much easier and more economical. Such a goal could be remove the fraction which may access to food chain. The heavy metal redistributed in soil has increased the chances of people exposed to these metals through inhalation, ingestion and skin contact [3]. Thus, the remediation of heavy metal contaminated soil is urgency especially for cultivated land. To improve soil quality, various in-situ and ex-situ remediation methods have been employed, such as stabilization, solidification, soil washing, electro remediation and phytoremediation [4].

Among these techniques, soil washing is considered as one of the most suitable on-site techniques for removing metals by which the contaminant bonded to soil are eliminated effectively and transferred to the liquid phase by desorption and solubilization. Recently, increased attention has been focused on the use of acidic and chelating agents which are capable of desorbing heavy metal contaminants from soil into solution. Acid washing mainly utilize ion exchange and soil matrix dissolution to solubilize metal and it effectively increase the mobility of heavy metals [10, 15].

The advantages of using chelating agents in soil washing are high efficiency of metal extraction, high thermodynamic stabilities of the metal complexes formed, good solubilities of metal complexes, and normally low adsorption of the chelating agents and their metal complexes on soils. Recent advances in chelating agent applications in soil washing and their recovery and recycling techniques have made soil washing an attractive alternative technology for cleaning up the metal-contaminated sites [9].

With chemical soil washing, soil particles are cleaned by selectively transferring the contaminant from the soil into solution. The effectiveness of washing is closely related to the ability of the extracting solution to dissolve the metal contaminants in soils. However the strong bonds between metal and soil make the cleaning process difficult. Therefore only extractants capable of optimally dissolving the metal must be carefully sought during soil washing. Acids , surfactants, electrolytes and chelating agents can be used in soil washing techniques. Strong acids attack and degrade the soil crystalline structure at extended contact times. For less damaging washes, organic acids and chelating agents are often suggested as alternatives to straight mineral acid uses [15, 21].

Various studies proved that organic acids and chelating agents are excellent to the extraction of heavy metals from soils. But acids washing leads to decreased soil productivity and adverse changes in the chemical and physical structures of soils due to mineral dissolution [16]. Previous studies has proved that EDTA,DTPA,NTA and EDDS were suitable to remediate metal contaminated soils. Chelating agents like diethylenetrimine pentaacetic acid(DTPA), nitriloacetic acid (NTA) are regarded as more attractive alternatives to acids or bases because the can form strong metal ligand complexes and thus highly effective in remediating heavy metal contaminated soils. Among these chelators NTA is class 2 carcinogen and DTPA identified as toxic and potential carcinogen. So some contries prohibit its use in soil washing. EDTA is continuously to be explored extensively for soil remediation because of its ability to mobilize metal cations effectively coupled with only minor impact on the physical and chemical properties of the soil structures [5,22].

The objective of the present study is to investigate the effects of EDTA, a typical chelating agent on the removal of lead from artificially contaminated soil sample and the optimization of the removal process by Taguchi orthogonal array design.

II. MATERIALS AND METHODS

A. Soil Preparation

This study was carried out in Thrissur, Kerala . Kerala, which lies at the extreme southern part of India. The soil investigated in this study was obtained from 0 cm – 20 cm below the ground surface from Government Engineering College Thrissur, Trichur campus. This college is located at 23° 34' 56.4384" N latitude and85° 37' 57.8172" E longitude. Soil samples was collected from different areas within the campus and it mixed well. The soil sample was belong to sandy loam and was free from toxic heavy metals. All the soil samples were air dried, screened through 1.73 mm sieve to remove large particles and stones, then thoroughly mixed to ensure uniformity and stored in a plastic bag at room temperature (20°C - 30°C) for sub-sequent experiments. Required amount of soil was withdrawn after thorough mixing. Properties like pH, conductivity, density, specific gravity, voids ratio, organic carbon, organic matter, cation exchange capacity, lead, content of the collected sample were determined. Since the concentration of lead was very low (below detectable limit), the sample was spiked with known concentrations of heavy metal(1000mg/kg).

B. Soil washing Experiments

Lead removal was performed in a batch method by varying four parameters including liquid to solid ratio (10 ml/g – 40 ml/g), different strength of EDTA (0.3g/l – 0.6 g/l), different pH(2 – 5) and a set of washing time (1 hour – 4 hour).

Batch study was carried out in 250 ml Erlenmeyer flasks containing known weight of soil sample spiked with known concentrations of lead and a measured volume of chelating agent. The solutions were shaken 190 rpm in a temperature-controlled shaker at room temperature for a selected period. The suspensions were centrifuged at the rotating speed of 3500 rpm for 15 min and the supernatants were then filtered through Whatmann filter paper no.1 to remove particulates in the solution. The residues were washed twice by de-ionized water. The supernatants were collected into a 100 ml flask. Finally, the concentration of lead, in the collected samples were analyzed using atomic absorption spectrophotometer [20,23].

The percent of each metal removed were calculated using the following equation

$$\textit{Percent of metal removed (\%)} = (C_1V_1/C_S\, m_S) \text{ X } 100$$

where $C1$ and Cs are the concentrations of metal in supernatant (in mg/L) and soil (mg/kg), respectively; V_l is the volume of supernatant (in L) and m_s is the dry mass of the soil (in kg) [8].

C. Taguchi Method of optimization

Using the Taguchi method, this study presents a systematic optimization approach for removal of lead (Pb) by EDTA chelating agents. Traditional experimental design methods are very complicated and difficult to use. Additionally, these methods require a large number of experiments when the number of process parameters increases. In order to minimize the number of tests required, Taguchi experimental design method, a powerful tool for designing high quality system, was developed by Taguchi. This method uses a special design of orthogonal arrays to study the entire parameter space with small number of experiments only. Taguchi recommends analyzing the mean response for each run in the inner array, and he also suggests analyzing variation using an appropriately chosen signal-to-noise ratio (S/N) [1-2].

III. RESULTS AND DISCUSSION

Selected properties of the studied soil is presented in table 1.This kind of soil shows neutral pH and relative low EC values. Organic matter content is one of soil properties which plays a strong role defining different chemical interactions between the pollutant and soil. The organic content of soil is high, so its cation exchange capacity also high.

Table 1. Characteristics of soil

Parameter	Value
Bulk Density	1338 kg / m^3
Specific Gravity	2.67
Porosity	51%
Organic content	78 mg / gm
pH	6.9
Electrolytic Conductivity	332 µs/ cm
Cation exchange capacity	7.1 cmol / kg
Lead Content	1000 mg / kg (after spiking)

In order to optimize the factors involved in the desorption of heavy metal from soil, 16 sets of experiments with operating conditions according to Taguchi experimental design were performed. The variables and values of their levels selected from batch study and as well as previous studies.

Table 2 Variables and Values of Their Levels

Parameter	Level 1	Level 2	Level 3	Level 4
Liquid to solid ratio	10	20	30	40
Washing time (hour)	1	2	3	4
Strength of chelating agents (mg/l)	0.3	0.4	0.5	0.6
pH	2	3	4	5

A. Results of experimental runs of Taguchi method

Based on 16 different trials of extraction process designed by Taguchi method the maximum lead removal obtained is 98.90%.

B. Statistical analysis

The results of the experiments were evaluated by the analysis of variance (ANOVA). The main purpose of the ANOVA was to determine the effect of each parameter on the variance of the results, regarding the total variance of all the parameters. In ANOVA studies with certain error, error variance determination is very important. Obtained data are used to estimate F Value of Fisher test (F test). Variation observed in an experiment is attributed to each significant factor or interaction is reflected in percentage contribution (P) which shows relative power of factor or interaction to reduce variation [6-7,18].

Table 4 shows the ANOVA results of lead removal efficiency by EDTA. The analysis was evaluated for a confidence level of 95%.

Table 3 Analysis Of Variance for Lead Removal

Source	DF	Seq SS	Adj SS	Adj MS	F	P	P(%)
L/S ratio	3	14.3928	14.3928	4.7976	414.48	0.000	63.32
Washing time	3	0.5975	0.5975	0.1992	17.21	0.021	2.63
Strength of solution	3	5.8646	5.8646	1.9549	168.89	0.001	25.80
pH	3	1.8423	1.8423	0.6141	53.05	0.004	8.10
Error	3	0.0347	0.0347	0.0116			0.15
Total	15	22.7320					100

[DF–degree of freedom; Seq SS- Sequential sum of squares; Adj SS- Adjusted sum of squares; Adj MS- Adjusted mean squares , P% - Percentage contribution]

According to ANOVA table the most important factors contributing to lead removal are liquid to solid ratio followed by strength of solution pH , and finally soil washing time

C. Signal to noise ratio

The S/N was used to measure both the mean value (named signal represent the desirable effect) and the standard deviation (named noise represents the undesirable effect) of a set of data. Higher S/N ratios are desirable in lead removal. The S/N ratios used for this type of response is defined as following [14].

$$S/N = -10 \log [1/n \ \Sigma \ 1/ Yi^2]$$

The results of S/N ratio for various designed experiments after ANOVA calculations are shown in Figure 1

i. Effect of Liquid To Solid Ratio

According to figure 5.1 the best S/N ratios was found at L/S ratio of 40. When L/S ratio decreased from 30 to 10 the S/N ratios decreased significantly.

ii. Effect of pH

The S/N ratio increases with increasing pH of chelating agents. The best S/N was found at pH of 4. When pH was decreased 4 to 2, the S/N ratios decreased significantly. At pH values higher than 4 there is a decline in S/N ratios.

iii. Effect of Strength Of Chelating Agents

Here also S/N ratio increases with increasing strength of solution. The best S/N was found at a strength of 0.5 g/l and S/N ratio almost equal at a strength of 0.6 g/l and the S/N ratio decreased significantly when strength of solution decreases 0.5 to 0.3 g/l.

iv. Effect of washing time

The best S/N ratio was found at a washing time of 3 hour and it decreased significantly with an increasing washing time. When washing time decreases from 3 to 1 hour the S/N ratio is also get decreases.

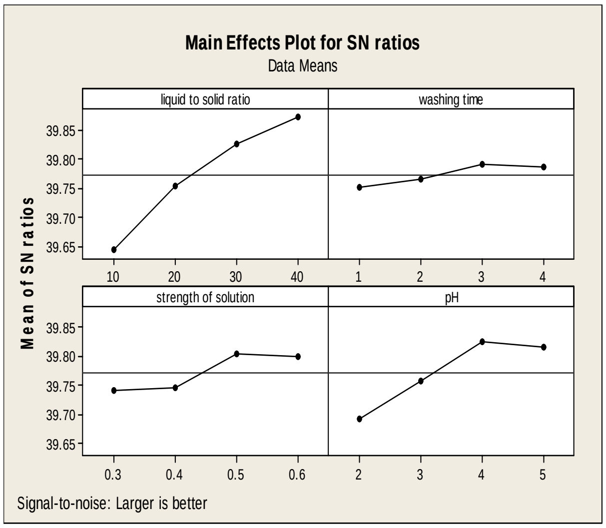

Figure 1. Effect of process parameters on S/N in Extraction process

D. *Interactions in the Lead Removal Process*

The possible interaction between variables were studied using interaction plots. Figure 2 displays a matrix plot for four factors .This graph shows a full interactions plot matrix [11].

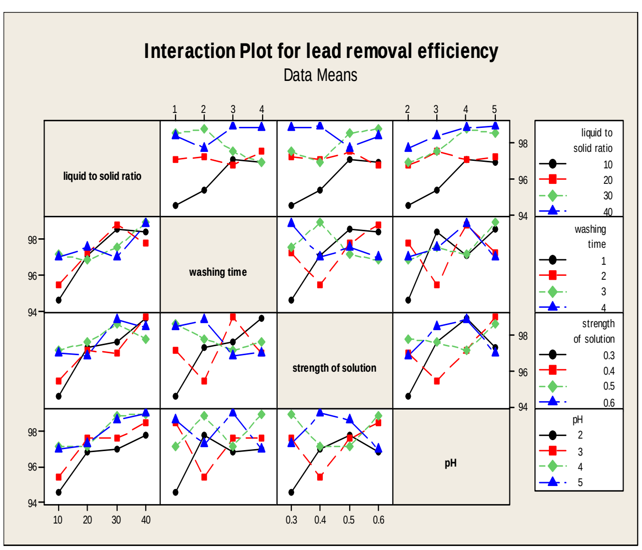

Figure 2. Interaction matrix plot between 4 factors : pH, strength of solution, washing time and liquid to solid ratio

Row 1 and 2 indicate the interaction between L/S ratio and pH . Row 2 and 3 indicate interactions between pH and strength of solution. Row 3 and 4 indicate interactions between strength of solution and washing time. Similarly row 1 and 4 indicate the interactions between liquid to solid ratio and washing time. Row 1 and 3 indicate interaction between liquid to solid ratio and strength of solution. Finally row 2 and 4 indicate the interaction between pH and washing time.

From matrix plot it can be concluded that the interaction between pH factor with L/S ratio, strength of solution and washing time is seem to be little strong at pH 4 and 5, interaction between L/S ratio factor with pH, strength of solution and washing time is seem to be little strong at L/S ratio of 20 and 40, interaction between strength of solution factor with L/S ratio pH, and washing time is seem to be little strong at a strength of 0.5 g/l and 0.6 g/l. Finally interaction between washing time factor with L/S ratio, strength of solution and pH is seem to be little strong at a washing time of 2 and 3 hours.

E. *Confirmation Test*

Confirmation test is a crucial step suggested by Taguchi to verify the experimental results. Figure 1 shows the best conditions for removal of lead from soil. The confirmation experiment was conducted with the optimum parameter obtained and the result were compared with the predicted result obtained using the multiple linear regression model.

Table 4. Optimum conditions for the lead removal

Parameter	**Optimum value**
Liquid to solid ratio	40 ml/g
pH	4
Strength of chelating agent	0.5 g/l
Soil washing time	3 hour

The confirmation test was carried out in optimal conditions and the lead removal efficiency was obtained as 99.08%. The regression equation developed in Taguchi software is

lead removal efficiency = 92.0228 + 0.085 liquid to solid ratio + 0.149 washing time + 2.64 strength of solution + 0.4905 pH

The predicted result for lead removal is 99.15 which is close to the result obtained from confirmation test

IV.CONCLUSION

This study demonstrated the application of fractional factorial design based on Taguchi's method for screening of the significant factors in removing the lead from artificially contaminated soil. An OA_{16} was used to investigate the effect of four factors (liquid to solid ratio, strength of solution, pH and washing time) on lead removal efficiency. The effect of each parameter was evaluated using ANOVA. The result of

ANOVA showed that liquid to solid ratio has significant effect on lead removal efficiency followed by strength of EDTA. The removal efficiency was achieved in optimal condition was 99.08%. But higher liquid to solid ratio would generate waste water and increase the cost of soil remediation.

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