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Recycling of lead-contaminated EDTA wastewater

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Abstract

Ethylene diaminetetraacetic acid (EDTA) is one of the chelating agents used in the soil washing process for the decontamination of lead-contaminated soil. Lead–EDTA complexes in the wastewater from the soil washing process must be removed before the wastewater can be safely discharged. This study outlines a method to recycle Pb–EDTA wastewater by substituting the Pb complexed with EDTA with Fe(III) ions at low pH, followed by precipitation of Pb ions with phosphate or sulfate ions. Fe(III) ions complexed with EDTA were then precipitated at high pH using sodium hydroxide. The resulting solution (Fe-precipitated solution) was tested on three lead-contaminated soils. The Fe-precipitated EDTA solution was found to have similar extraction capabilities as fresh EDTA solution. Experimental results showed that the recycled EDTA solution may be recycled several times without losing its extractive power. Recycled EDTA wastewater with phosphate precipitation was found to be slightly more effective than recycled EDTA solution using sulfate precipitation. The recycling procedure may be applied to wastewater generated during soil washing of lead-contaminated soil, resulting in a reduction in wastewater generated and savings in the amount of EDTA used. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Recycling; EDTA; Lead; Contaminated soil; Soil washing

1. Introduction

The past decade has witnessed the rapid development of various treatment technologies for the remediation of lead-contaminated soils. One of the more effective treatment methods is the extraction of lead from lead-contaminated soils with chelating agents [1–3]. Of the many commercially available chelating agents, ethylene diaminetetraacetic acid (EDTA) has been shown to be the most effective, but the high cost of EDTA has

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hampered its wide use for the remediation of metal-contaminated soils. Another difficulty with using EDTA is that a large volume of wastewater containing metal–EDTA complexes is generated and must be treated before disposal. To reduce the cost of using EDTA, recycling of used EDTA and reduction of the volume of the wastewater for disposal are needed. Unfortunately, there are currently no practical and commercially available methods for recycling EDTA in wastewater, although several recycling methods have been demonstrated on a laboratory-scale.

Some of the earlier metal–EDTA wastewater recycling studies were concerned with the recovery of metals from metal–EDTA complexes. One of the recycling methods investigated was the application of electrolysis in conjunction with a cation-exchange membrane for the recovery of metals in a solution of metal–EDTA complexes [4]. With the current interest in remediation technologies, the electrolysis method has been extended by several researchers for the recycling of wastewater from soil washing [5,6]. Results of the electrolysis method showed that the recovery of metal and EDTA were approximately 99% and 91%, respectively [6]. However, the electrochemical experiments were conducted using bench-scale experiments with clean Pb–EDTA solution. Even though EDTA was recovered in these experiments, the recovered EDTA was not reused to demonstrate the effectiveness of the recovered EDTA for further treatment of contaminated soil. The electromembrane process was not without any problems. According to Allen and Chen [5], the pH in the cathode compartment was found to increase with time as a result of hydroxyl ion production. The high pH (> 10) in the cathode compartment resulted in the degradation of the membrane. In the anode compartment, the pH of the solution decreased as a result of proton production. Precipitation of EDTA on the membrane also resulted in a reduction in EDTA recovery and current efficiency. For these reasons, several operating problems must be addressed before the electromembrane system can be a practical and feasible system for recycling EDTA.

Rudd et al. [7] proposed precipitating lead ions as lead hydroxide or as lead sulfide at high pH to recover the EDTA and to allow the EDTA wastewater to be safely discharged. Although sulfide precipitation was found to reduce lead concentrations to acceptable discharge levels, the effectiveness of the recycled EDTA solution for further treatment of contaminated soil was not demonstrated. Addition of ferrous sulfate at pH 6–9 was found to be effective in treating Cu–EDTA complexes in metal finishing wastewater [8]. Ferrous ions from ferrous sulfate were able to displace the Cu^{2+} in the Cu–EDTA complex by forming a more stable Fe(III)–EDTA complex than the Cu–EDTA complex. Work done by Kennedy [9] showed that addition of approximately 0.25% to 5% by weight of calcium hydroxide to a Fe(III)–EDTA solution resulted in the precipitation of more than 99% of ferric ions from the Fe(III)–EDTA complex. Similar work was done by Tunay and Kabdashi [10] where calcium ions were found to be effective in binding with EDTA at high pH, freeing the heavy metals from the metal–EDTA complex to form hydroxide or carbonate solids.

There are currently no practical means of recycling Pb–EDTA or metal–EDTA wastewater from a soil washing system. The objectives of this study were to assess a method for recycling Pb–EDTA wastewater and its effectiveness for repeated use in the treatment of lead-contaminated soils. The recycling process proposed may also be used to recover EDTA used in the flushing and cleaning of cooling water systems.

2. Recycling approach

When lead-contaminated soil is washed with EDTA solution, the extracted solution will contain Pb–EDTA and various metal–EDTA complexes (generally Al-, Ca-, Fe-, Mg- and Mn–EDTA complexes) and residual EDTA. In general, the relative stability of a metal–EDTA complex in the presence of other metals may be predicted based on the stability constants, the pH of the solution, the concentration of the metals and EDTA, and the presence of other electrolytes [8,11,12]. Fig. 1 provides a comparison of the conditional stability constants for various metal–EDTA complexes as a function of pH. For low pH conditions (less than 3), the tendency for metal–EDTA complexes to form may be assumed to follow the following sequence: $\text{Fe}^{3+} > \text{Cu}^{2+} > \text{Pb}^{2+} > \text{Al}^{3+} > \text{Zn}^{2+}$

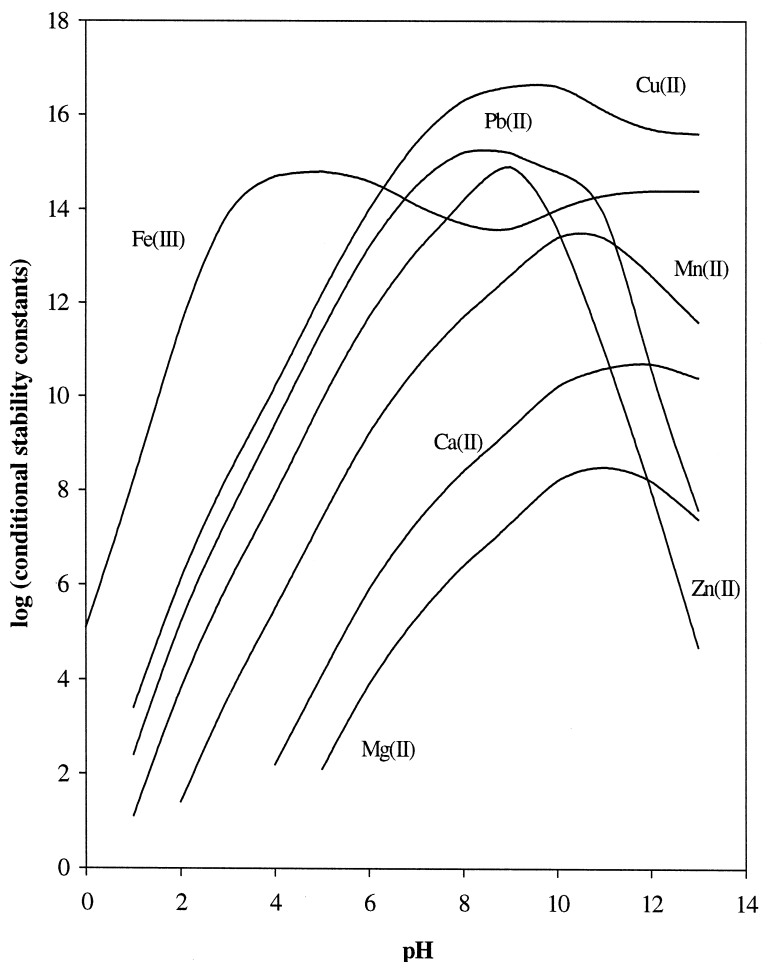


Fig. 1. Comparison of conditional stability constants for various metal–EDTA complexes as a function of pH (adapted from Ref. [12]).

$> \text{Mn}^{2+} > \text{Ca}^{2+} > \text{Mg}^{2+}$ (see Fig. 1). Based on the conditional stability constants of Pb–EDTA complexes, EDTA will complex with lead in preference over the other cations at all pH values except for ferric and cupric complexes. Chemical equilibrium predictions by Norvell and Lindsay [13] showed that at low pH values, ferric ions can substitute for the zinc and manganese ions complexed with EDTA. Similarly, predic-

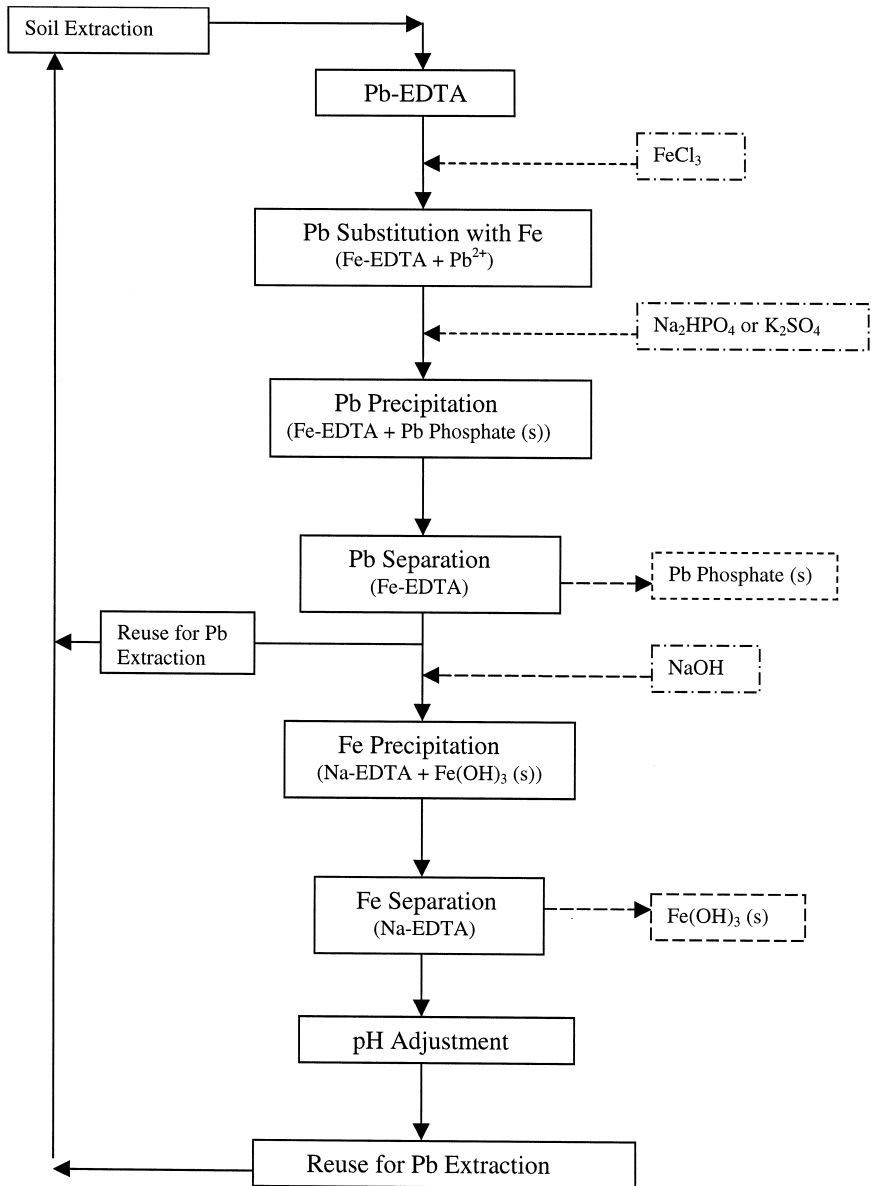


Fig. 2. Flow diagram of the recycling process.

tions by Sommers and Lindsay [14] showed that for a solution with equal molar concentrations of lead, ferric and EDTA ions, Fe(III)–EDTA would be more dominant than the Pb–EDTA complex at pH less than 5.2.

Based on the above observations and work done by Kim and Ong [15] and Bowers and Huang [16], lead in the Pb–EDTA complexes may be substituted at low pH by ferric ions due to the higher stability of Fe(III)–EDTA complexes over Pb–EDTA complexes. Lead ions can then be precipitated at low pH using either sulfate or phosphate ions. For the same molar concentration of sulfate or phosphate ions and for pH less than neutral pH, lead phosphate has a lower solubility than lead sulfate. In the presence of chloride ions, the solubility of lead ions in a phosphate solution may be reduced further through the precipitation of chloropyromorphite $Pb_5(PO_4)_3Cl$ ($\log K = -25.05$). The precipitated lead may then be separated from the solution. Ferric ions in the Fe(III)–EDTA solution may be precipitated at high pH using sodium hydroxide. The resulting solution (called the Fe-precipitated EDTA solution) may then be recycled and used again for the extraction of lead from lead-contaminated soils. Fig. 2 shows the flow diagram of the recycling procedure. Note that the solution after precipitating the Pb may be used as an extraction solution provided the solution pH is adjusted to above pH 7 during extraction. The effectiveness of the Fe(III)–EDTA solution is limited to solution pH greater than 7 while the Fe-precipitated EDTA solution may be applied over a wider pH range. Results are presented which compared the extraction efficiency of recycled Fe(III)–EDTA and Fe-precipitated EDTA solutions. In addition, the effectiveness of recycled EDTA solutions using phosphate and sulfate as precipitants for lead ions were compared.

3. Materials and methods

3.1. Experimental approach

Experiments were conducted in two phases to test the recycling procedure. In the first phase, the substitution and precipitation steps in the recycling procedure were tested using a prepared solution of Pb–EDTA. The recycled EDTA was then applied to a lead-contaminated soil to assess its extraction capability. Ferric chloride was used for the substitution of lead in the Pb–EDTA complexes while lead ions were precipitated using sodium phosphate. The reason for selecting ferric chloride in the substitution process was that chloride ions would be available to enhance the precipitation of lead when phosphate was added. In the second phase, Pb–EDTA wastewaters from the extraction of three different lead-contaminated soils were recycled and reused as an extraction solution. The recycle–reuse procedure was conducted over several cycles to assess the extraction efficiencies of recycled Pb–EDTA solution.

3.2. Precipitation and extraction studies using prepared Pb–EDTA solution

A stock solution containing approximately 0.005 M of lead was prepared by dissolving an adequate mass of lead sulfate in 2000 ml of nanopure water. The dissolved lead concentration in the solution was analyzed using a Smith Hieftje 12 atomic

absorption spectrophotometer. Sodium EDTA crystals, at molar amount slightly less than the dissolved lead concentration present in the solution, were added to the solution. The solution was stirred for 24 h and filtered with a 0.45- μm membrane filter paper to separate any residual lead sulfate particulates from the solution. The lead concentration in the solution was analyzed again and all the lead present in the solution was assumed to be complexed with EDTA.

The optimum amount of ferric ions needed for the substitution of lead in the Pb–EDTA complex was determined by conducting a series of experiments where different amounts of ferric chloride were added to 10 ml of the prepared Pb–EDTA solution. The substitution experiments were conducted in 50 ml polypropylene centrifuge tubes. Adjustment of pH was not needed since the addition of ferric chloride depressed the solution pH to between 2 and 4. The solution was shaken for 6 h. Sodium phosphate was then added to the solution to obtain a phosphate concentration of approximately 0.15 M. The solution was shaken for 10 h and centrifuged at 3000 rpm for 10 min. The solution was filtered with 0.45- μm filter paper. Preliminary experiments showed that a reaction time of 2 h was sufficient for ferric substitution and lead precipitation, respectively. However, longer reaction times as indicated above were used to ensure complete reaction. Lead and iron concentrations in the filtrate were then measured with an atomic absorption spectrophotometer.

A portion of the Fe(III)–EDTA solution was set aside and used as an extraction solution on a lead-contaminated soil. For the other portion of Fe(III)–EDTA solution, the pH was adjusted to 13 by dissolving NaOH pellets in the solution. The solution was filtered using a 0.45- μm filter paper to separate the precipitated ferric hydroxide. The Fe-precipitated EDTA solution was then used as an extraction solution on a lead-contaminated soil.

The extraction experiments were conducted by mixing 1 g (dry weight) of a lead-contaminated soil with 10 ml of Fe(III)–EDTA or Fe-precipitated EDTA solution in a 50-ml polypropylene centrifuge tube for 24 h. The lead-contaminated soil used, Soil A, was obtained from a Superfund site in New Mexico which was a former battery recycling and smelter facility. The physical–chemical properties of Soil A are presented in Table 1. The pH of the slurry for extraction was adjusted using 1 M sodium hydroxide and 1 M HNO_3 solutions. The slurry was then filtered with a 0.45- μm filter paper. The pH and the lead concentration in the filtrate were measured. The difference in lead concentration in the solution before and after the application of the Fe(III)–EDTA or Fe-precipitated EDTA solution was assumed to be the amount of lead extracted from the lead-contaminated soil. A similar set of extraction experiments using fresh 0.005 M EDTA solution was conducted and the lead extraction efficiencies were compared against the results of the recycled EDTA solutions. Based on the lead concentration in Soil A, the EDTA:Pb molar ratio for fresh 0.005 M EDTA solution was 0.78.

3.3. Lead extraction studies using recycled Pb–EDTA wastewater

The extraction capability of recycled Pb–EDTA wastewater was tested on three lead-contaminated soils using batch experiments. The first soil sample was Soil A as described above while the second sample (Soil B) was taken from a military firing range

Table 1
Soil properties and major cation concentrations of lead-contaminated soil samples

Parameters	Soil A	Soil B	Soil C
Organic carbon (%)	2.52	0.18	0.75
Cation exchange capacity (meq/100 g)	17.51	5.5	11.61
Specific surface area (m ² /g)	15.39	0.66	9.68
pH	8.1	8.5	8.2
<i>Cations (mg/kg)^a</i>			
Pb	13,260	6238	2413
Fe	316	328	^b
Mn	2820	21	^b
Al	14,330	2440	^b
Ca	12,410	7450	^b
Mg	5270	436	^b
Cu	30	279	^b
Zn	86	70	^b

^aBy acid digestion method (Method 3005, [17]) except for Fe. Fe is the amorphous iron concentration measured using the method as in Ref. [18].

^bNot tested.

in Florida. The third sample (Soil C) was artificially contaminated with lead. The base material for the artificially contaminated soil was oxidized glacial till obtained from the Iowa State University Agricultural Farm near Ames, IA. The physical–chemical characteristics of the three soils are presented in Table 1. Each soil was air dried and screened through sieve number 25 (0.707 mm) to remove large particles such as bullet fragments in the firing range soil and organic debris. The artificially contaminated soil was prepared by mixing 200 g of soil with adequate amount of lead nitrate solution to obtain a lead concentration of 2413 mg/kg. The slurry was mixed for 48 h and the excess solution was drained. The soil was allowed to dry for over 3 months before being screened.

Table 2
Precipitation of lead from 0.005 M Pb–EDTA stock solution (Pb concentration approximately 1040 mg/l)

Sample no.	Ratio of Fe/Pb	Added phosphate NaH ₂ PO ₄ ·H ₂ O (M)	Final Pb concentration (mg/l)	Lead precipitated (%)	Final pH
1	0.72	0.15	301.4	72	3.5
2	0.72	0.15	291.6	73	3.5
3	1.44	0.15	15.6	99	3.3
4	2.16	0.15	0	100	3.0
5	1.5	0.15 ^a	1.9	99.9	7.6
6	1.41	0.15 ^b	10	99.9	3.1

^aWith Na₂HPO₄·7H₂O.

^bWith K₂SO₄.

Extraction experiments were conducted as described above with 20 g in 200 ml of EDTA solution giving a 10:1 volume:mass ratio as in the earlier extraction experiments. Because of the varying concentrations of lead in the three soils, the EDTA concentrations used were 0.05 M, 0.03 M and 0.03 M for Soil A, Soil B and Soil C, respectively. The EDTA:Pb molar ratios were 7.8, 9.96, and 25.7, respectively. The extraction experiments were conducted over a 12-h period. The filtrate was recycled using the procedure described above with Fe substitution, Pb precipitation, and Fe precipitation steps. After recycling, the Pb and Fe concentrations were measured to ensure that most of the Pb and Fe had precipitated and removed from the solution. The pH of each recycled EDTA solution was adjusted to approximately 8.2 before it was used for lead extraction. The recycle–reuse procedure was conducted over three or four cycles. For lead precipitation, sulfate and phosphate ions were used to determine their effectiveness and impact on the recycling process. Triplicate extraction experiments for each cycle were conducted for Soil C while single extraction experiments for each cycle were conducted for Soil A and Soil B.

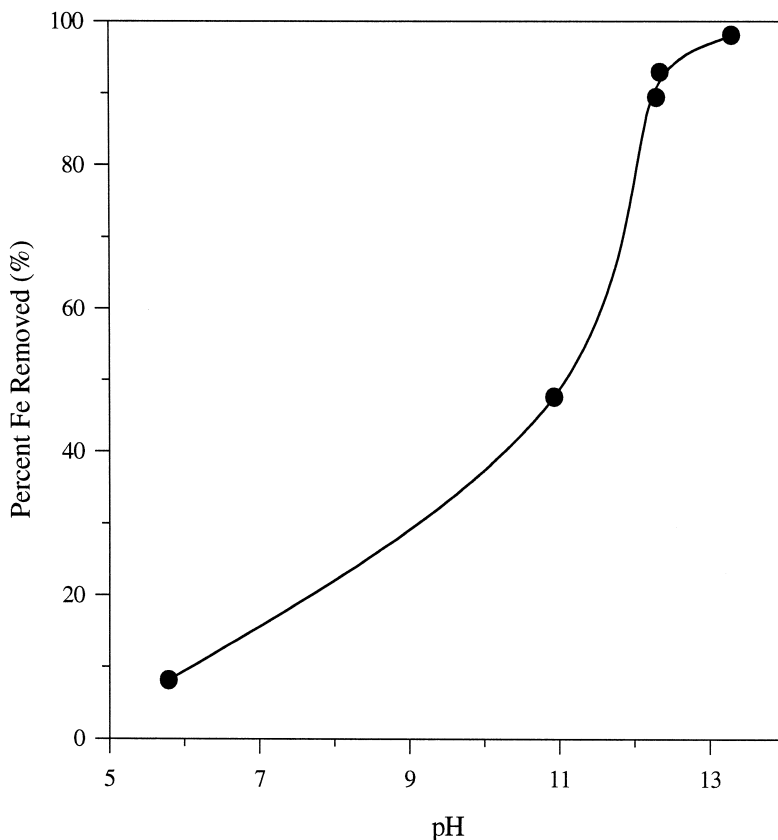


Fig. 3. Percent Fe(III) removal from Fe–EDTA solution at different pH with sodium hydroxide.

4. Results and discussion

4.1. Lead precipitation from Pb–EDTA stock solution

Test results for the substitution of lead with Fe(III) ions in a 0.005 M Pb–EDTA solution and the precipitation of lead using potassium sulfate or monosodium phosphate are presented in Table 2. For a Fe:Pb molar ratio of 1.44 and 1.41, the percent of lead precipitated with phosphate and sulfate were 99% and 100%, respectively. Precipitation results showed that a molar amount of Fe(III) approximately equal to 1.5 times the Pb molar amount (or the applied EDTA molar amount) was needed to remove most of the lead ions from the Pb–EDTA solution. With 0.15 M of disodium phosphate (sample no. 5 in Table 2), the precipitation results were similar to that of monosodium phosphate but with a significant increase in the final pH of the solution. Disodium phosphate may be more appropriate for lead precipitation because the higher pH of the final solution will mean a lower volume of NaOH needed for the precipitation of Fe(III) ions. As such, disodium phosphate was used for the precipitation of lead in the recycling experiments using actual lead-contaminated soils. Also in Table 2, lead precipitation with potassium

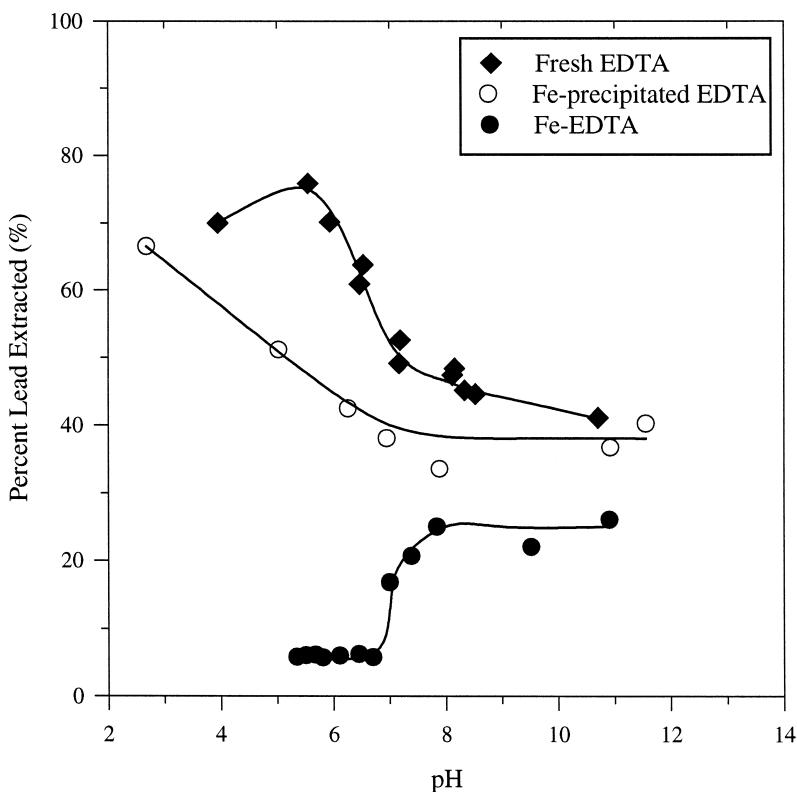


Fig. 4. Percent lead extracted using recycled EDTA solution and fresh 0.005 M EDTA solution at various pH.

sulfate was similar to that of disodium phosphate ions as the precipitant except that the final pH was 3.1. With regard to precipitation of Fe, Fig. 3 shows that most of the Fe(III) ions present in the Fe(III)–EDTA solution were precipitated at high pH, indicating that the hydrolysis of ferric ions at high pH may be thermodynamically more favorable than Fe complexation with EDTA.

4.2. Extraction efficiency of recycled Pb–EDTA stock solution

Fig. 4 shows the extraction efficiencies of Fe(III)–EDTA solution and Fe-precipitated EDTA solution on Soil A for different pH values. The Fe(III)–EDTA and the Fe-precipitated EDTA solutions were prepared from 0.005 M Pb–EDTA stock solution as described earlier. Also plotted on Fig. 4 are the extraction efficiencies of fresh 0.005 M EDTA solution on Soil A. Fig. 4 shows that pH played an important role in determining the lead extraction efficiency of the recycled Fe(III)–EDTA solution. Of the three solutions used, the Fe(III)–EDTA solution had the lowest lead extractive capability. At low pH values, the percent of lead extracted using Fe(III)–EDTA solution was approxi-

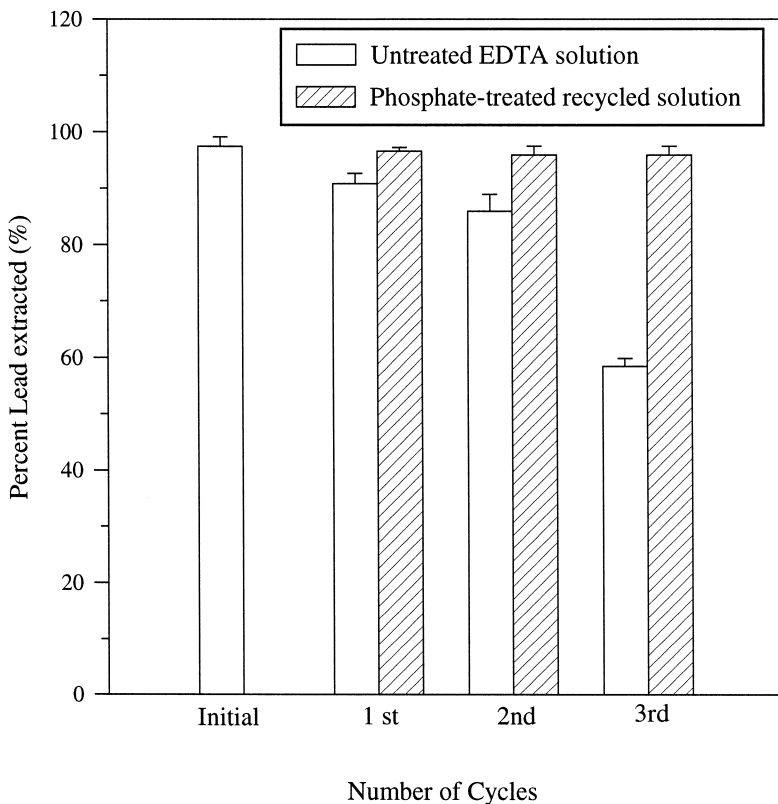


Fig. 5. Percent lead extracted from artificially lead-contaminated soil (Soil C) with 0.03 M EDTA solution (S.D. for triplicates; with and without recycling treatment at pH 8.2).

mately 5–7% (see Fig. 4). The reason was that for pH less than 7, the Fe–EDTA complex would be more favorably formed than the Pb–EDTA complex (see Fig. 1). Above pH 7, the extraction efficiencies of the Fe–EDTA solution improved and were about half of the extraction efficiencies of fresh EDTA solution. This reflects the more favorable complexation conditions for Pb over Fe at pH greater than 7.

As shown in Fig. 4, Fe-precipitated EDTA solution performed better than Fe–EDTA solution and had much higher lead extraction efficiencies over the pH range of 4–11. The extraction efficiency of the Fe-precipitated EDTA solution was about 65% to 95% of the extraction efficiency of the fresh EDTA solution. Precipitating Fe from the Fe–EDTA solution before it is used for extraction increased the extraction efficiency of the recycled EDTA solution.

4.3. Lead extraction using recycled Pb–EDTA wastewater

Fig. 5 shows the extraction efficiencies of lead for artificially lead-contaminated soil (Soil C) with 0.03 M EDTA solution. For each cycle, the EDTA wastewater was treated

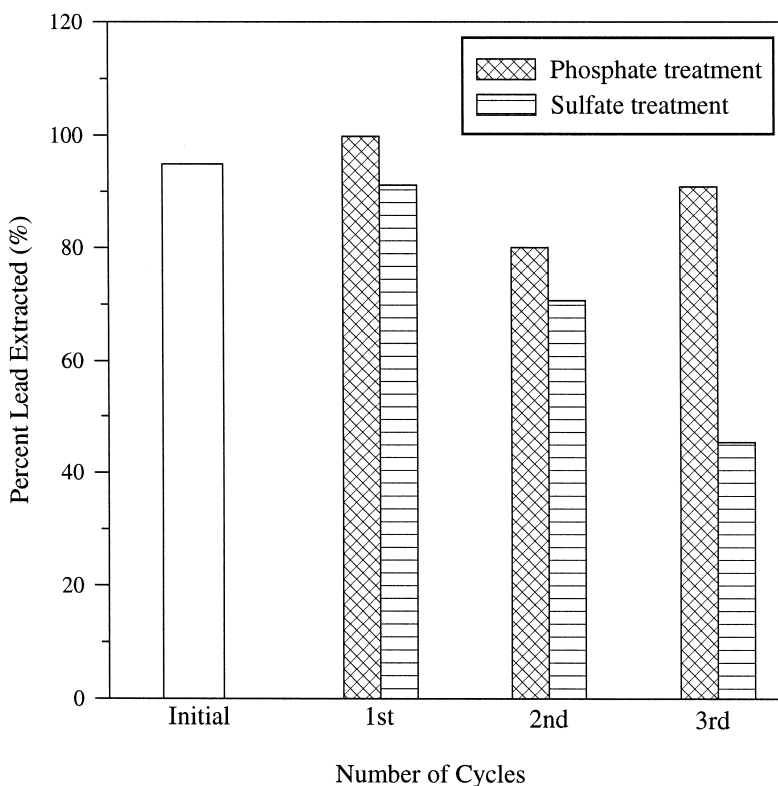


Fig. 6. Percent lead extracted from Soil A with 0.03 M EDTA solution (phosphate vs. sulfate treatment at pH 8.2).

with ferric chloride and disodium phosphate to precipitate lead ions, followed by Fe(III) precipitation. The recycled Fe-precipitated EDTA showed a much higher lead extraction efficiency than consecutive application of the same EDTA solution. The extraction efficiency of consecutive application of 0.03 M EDTA dropped to about 55% after the third application while the extraction efficiency for the recycled EDTA remained the same at approximately 100%. Fig. 6 shows the lead extraction efficiencies for Soil A with 0.03 M EDTA solution over several cycles. For these experiments, either phosphate or sulfate was used to precipitate the lead ions. Even after the third recycle, the extraction efficiency for the recycled Fe-precipitated EDTA wastewater was still high at 90% using phosphate as the precipitant for lead.

Similar results were obtained for Soil B with the recycled Fe-precipitated EDTA solution (see Fig. 7). Extraction efficiencies of the recycled Fe-precipitated EDTA using phosphate ions were about 80% to 100% of the extraction efficiency of fresh EDTA solution. As shown in Fig. 7, the EDTA wastewater continued to maintain extraction efficiencies as high as 65% even after being recycled over three cycles. Similar results were obtained using sulfate as the precipitating ions, although the extraction efficiencies

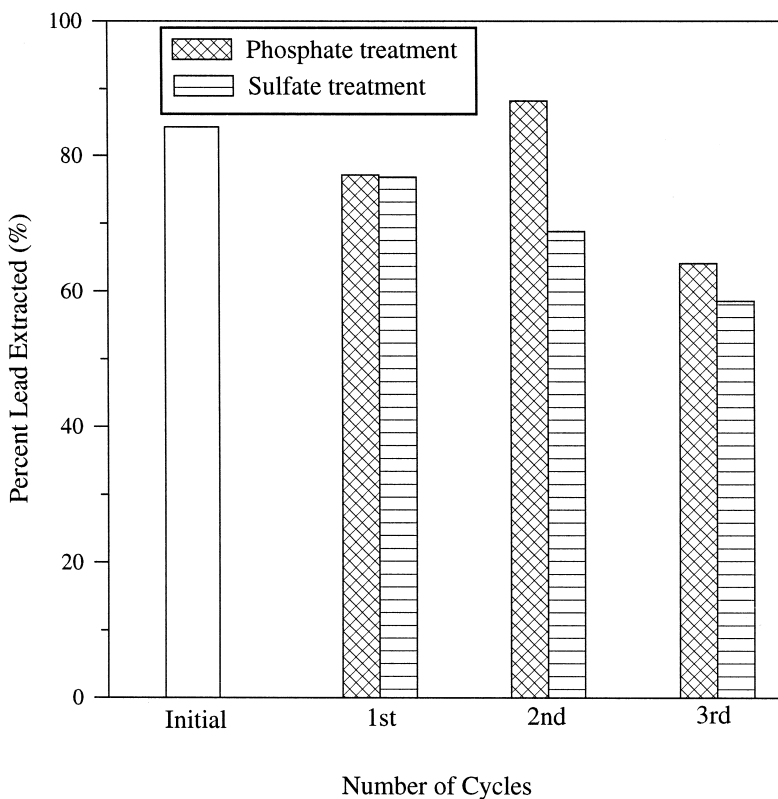


Fig. 7. Percent lead extracted from Soil B with 0.05 M EDTA solution (phosphate vs. sulfate treatment at pH 8.2).

were slightly lower than using phosphate as the precipitating ions. These experiments confirmed that the recycled EDTA solution may be as efficient as fresh EDTA solution for extracting lead from lead-contaminated soil even after the EDTA solution was recycled for the third time.

The merits of recycling used EDTA solution can be illustrated by computing the volume of wastewater generated. For every gram of soil treated, at least 5 g to 10 g of extraction wastewater will be produced (assuming a solution:soil mass ratio of 5 to 10). If the wastewater is recycled over three cycles, the wastewater generated will be one-fourth of the total volume generated when there is no recycling and reuse. Although the experiments were conducted over three cycles, the recycled EDTA can continue to be recycled for several more cycles. Obviously, some of the wastewater will be lost during the recycling process and make-up water and EDTA will be needed. In addition to a reduction in wastewater, there will be savings in the amount of EDTA used. Chemical costs will be incurred regardless of whether there is recycling or not since chemicals will be needed to treat the wastewater before it can be disposed. In summary, the EDTA recycling process can reduce the volume of EDTA wastewater effluent and the amount of EDTA needed.

5. Conclusion

A method to recycle Pb–EDTA wastewater has been presented. Based on the experimental results, the Pb–EDTA wastewater can be recycled several times without losing much of its extractive capabilities. The recycling method involves substituting the Pb complexed with EDTA with Fe(III) ions at low pH values, followed by precipitation of lead ions with either phosphate or sulfate ions. Fe(III) in the Fe–EDTA solution is then precipitated at high pH. The Fe-precipitated EDTA solution was found to be just as effective as fresh EDTA and may be recycled several times without losing its extractive power. If the Fe(III) is not precipitated, the Fe–EDTA solution may be used as an extraction solution but is not as effective as the Fe-precipitated solution and must be applied at pH greater than 7. Precipitation of lead using phosphate ions was found to give a slightly better recycled EDTA solution than precipitation with sulfate ions. The recycle method proposed will provide considerable reduction in the wastewater generated for the soil washing technology and will also reduce the amount of EDTA used.

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