

Extraction of heavy metals from contaminated soils using EDTA and HCl

Hatem Asel Gzar Assistant professor Department of Environmental Engineering College of Engineering- Baghdad University email:hatam_asal@yahoo.com Israa Mohammed Gatea Department of Environmental Engineering College of Engineering- Baghdad University email: sarawemoh@gmail.com

ABSTRACT

The present study examines the extraction of lead (Pb), cadmium (Cd) and nickel (Ni) from a contaminated soil by washing process. Ethylenediaminetetraacetic acid disodium salt (Na₂EDTA) and hydrochloric acid (HCl) solution were used as extractants. Soil washing is one of the most suitable in-situ/ ex-situ remediation method in removing heavy metals. Soil was artificially contaminated with 500 mg/kg (Pb , Cd and Ni). A set of batch experiments were carried out at different conditions of extractant concentration , contact time, pH and agitation speed. The results showed that the maximum removal efficiencies of (Cd, Pb and Ni) were (97, 88 and 24) % respectively using (0.1 M) Na₂EDTA. While the maximum removal efficiencies using (1M) HCl were (98, 94 and 55)% respectively. The experimental data of batch extraction were applied in four kinetic models; first order, parabolic diffusion, two constant and Elovich model. The parabolic diffusion was the most fitted to the experimental data.

Key words: contaminated soil, heavy metals, soil washing, chemical extraction, kinetics.

انتزاع المعادن الثقيلة من التربة الملوثة باستخدام EDTA و HCl

إسراء محمد كاطع	حاتم عسل كزار
قسم الهندسة البيئية كلية الهندسة/ جامعة بغداد	استاذ مساعد قسم الهندسة البيئية / كلية الهندسة/ جامعة بغداد
	الخلاصة

في الدراسة الحالية تم دراسة الانتزاع للرصاص الكادميوم, والنيكل من التربة الملوثة باستخدام (HCl) Hydrochloric acid و Na₂EDTA)Ethylenediaminetetraacetic acid disodium salt استخلاص .

ان طريقة غسل التربة لازالة المعادن الثقيلة هي واحدة من أكثر طرق المعالجة الملائمة والتي تستخدم خارج وداخل المواقع الملوثة. تمت دراسة تربة التي يتم تلويثها مختبرياً بالعناصر التالية: الرصاص ، الكادميوم ، النيكل حيث كان تركيز الملوث 500 ملغم/كغم. في تجارب الدفعة التي اجريت على التربة تم دراسة ظروف مختلفة مثل تركيز المستخلص و زمن التماس والرقم الهيدروجيني وسرعة الاهتزاز. اظهرت نتائج تجارب الدفعة على ان الحد الأقصى لكفاءة الإزالة زمن التماس والرقم الهيدروجيني وسرعة الاهتزاز. اظهرت نتائج تجارب الدفعة على ان الحد الأقصى لكفاءة الإزالة (97،88،24) كل الد الكادميوم و الرصاص والنيكل على التوالي في عند استخدام (0.1) مولاري مولاري اليما الحد الأقصى لكفاءة الإزالة باستخدام (1) مولاري من HCl كان (94، 94، 55)% . تم تطبيق البيانات والنتائج العملية المستحصلة من تجارب الدفعة في أربعة نماذج رياضية(والموذج الرياضي الافضل لتمثيل البيانات والنتائج العملية والمستحصلة من تجارب الدفعة في أربعة نماذج رياضية (والدون مني الافضل المستخلة المستحصلة من المستخلة المعادية ال



الكلمات الرئيسية: تربة ملوثة,معادن الثقيلة، غسل التربة ، الانتزاع الكيميائي ،حركيات.

1. INTRODUCTION

Pollution of the soils with toxic metals is widespread across the globe, and threat in many countries today. It has become a major environmental concern in many parts of the world due to rapid industrialization, increased urbanization, modern agricultural practices and inappropriate waste disposal method. Accordingly, the cleanup of these soils is a difficult task for environmental engineering **Tandy, et al .,2004.**

Heavy metals are chemical elements with a specific gravity that is at least (5) times the specific gravity of water. They are often problematic environmental pollutants, with well-known toxic effects on living systems, **Evanko**, et al., **1997**. They are introduced into the environment during mining, refining of ores, combustion of fossil fuels and industrial processes. They cannot be degraded or destroyed, **Davydova**, **2005**.

The contamination of surface and groundwater with highly mineralized mine waters or with compounds leached from mine dumps or tailings pose a very persistent environmental problem. Harmful contaminants, derived from such sources, enter the surface streams, settle in sediments, dissolve gradually and enter the environment as hazardous substances over long periods of time **,Phelelani,2007.**

As one of the most suitable in-situ / ex-situ physical /chemical treatment technologies, soil washing has been successfully used for the treatment of soils contaminated with heavy metals, hydrocarbons and semi-volatile organic compounds **,Peters, 1999**. Soil washing usually employs different extractants such as acids, bases, chelating agents, electrolytes, oxidizing agents and surfactants **,Reddy,** and **Chinthamreddy, 2000**. Soil washing is fast emerging technologies that is used to treat/clean up contaminated soil with contaminants ranging from heavy metals to hydrocarbons **,Shumba, 2008**.

The techniques are to separate the metal from soil by using chelating agents such as , ethylene diamminotetraacetic acid disodium salt Na₂EDTA, which, can extract heavy metals from contaminated soils with high efficiency. Strong inorganic acid ,also, can be used as washing solutions in terms of reasonable cost and simple handling of the effluent solution. It is an effective solvent due to high its removal efficiency in extraction of heavy metal, especially hydrochloric acid (HCl) **Jsoyama**, and **Wada**, **2007**.

Soil washing is a variable treatment alternative for metal contaminated sites chemical extraction to enhance the efficiency of heavy metals extraction. Process parameters in soil washing include the mode of extraction (batch or continueoud), extraction type and concentration, pH, electrolyte concentration, liquid-to-solid ratio (L/S), retention time. The soil related parameters are pH ,particle size distribution and mineral type of metal to be extracted and their concentration, distribution and physicochemical forms in the soils,**Tandy, et al .,2004**.

2. MATERIALS

2.1 soil

Natural Iraqi soil samples were collected from depth not exceeded 20 cm below the ground surface. These samples were cleaned, dried and well stored with an additional sieving onto 2mm mesh to achieve satisfactory uniformity. **Table 1** summaries the physicochemical properties of the soil. The required tests for specifying the characteristics of soils are carried out at State Company of Geological Survey and Mining/ Ministry of Industry and Minerals.



2.2 Contaminants

To simulate the soil's lead , cadmium and nickel contamination, a solutions of Pb(NO₃)₂, Cd(NO₃)₂.4H₂O , Ni(NO₃)₂.6H₂O, respectively (which are manufactured by Analar Company/made in England) were prepared and added to the specimen to obtain representative concentration.Cd(NO₃) $_{2}$.4H₂O has a molecular weight of 308.47 g/mole and atomic weight of cadmium ions is112.4g/mole. For example, to prepare a soil sample with cadmium concentration of 500 mg/kg , 1.372 g of Cd(NO₃) $_{2}$.4H₂O dissolves in 500 ml of distilled water and added to 1 kg of dry soil.

2.3 Na₂EDTA (C₁₀H₁₄N₂Na₂O₈.2H₂O)

Ethylenediaminetetraacetic acid disodium salt, is a very effective chelating agent. Its ability to bind heavy metal ions can be used to sequester these trace metals, is very stable and can be mixed with just about any phosphate containing fertilizer at basically any pH. It is very resistant to microbial degradation; therefore it remains quite stable in soils. It is of interest that EDTA has been used medically to promote removal of lead from the human body and also as an additive to render floor polishes with zinc binders amenable to detergent washing,Joseph, et al., 1997.Table 2 summaries the Na₂EDTA characteristics used in the present study.

2.4 HCl

Hydrochloric acid is a clear, colorless, highly-pungentsolution of hydrogen chloride (HCl) in water. It is a highly corrosive, strongmineral acid with many industrial uses. HCl is found naturally in gastric acid. HCl is very effective in the removal of heavy metals and achieve high removal efficiency in the case of natural pH with out neutralization to no loss the acidic property that are of high importance for the movement of contaminant sand solubility and thus easily extracted **,SRI, 2001.Table 3**summaries the HCl characteristics used in the present study.

3. Experimental Procedure

Batch equilibrium tests are carried out to specify the best conditions of contact time, pH, concentration of extractant and agitation speed. This means that these tests are suited to identify the activity of the chemical extraction using Na₂EDTA and HCl . Series of 250 ml flasks are employed, each flask contained 2 g of soil and 20 ml of (Na₂EDTA or HCl) extractant with a solid to liquid ratio (S/L) 1/10 , and contamination rate of 500 mg/kg .The Na₂EDTA concentrations were (0.001,0.005,0.01,0.05 and 0.1) M , and HCl concentrations were (0.01,0.05, 0.1,0.5 and 1) M. pH values of the five HCl solutions were (2,1.7,1.44,1.16 and 1.08) respectively .The shaking of solutions were achieved by an orbital shaker(incubated AAH23212K,model S1-600R ,Korea) at a speed of 200 and 250 rpm at temperature ($25\pm1^{\circ}$ C) at different contact time (1,2,3,4,5 and 6 hours).

Then the solution is filtered using a whatman No 42 filter paper. The supernatant was analyzed for heavy metal using atomic absorption spectrometry (AAS) (GBC, SensAA (Australia)). All tests were performed in triplicate and the results were presented as average of the duplicates extracts. Kinetic studies were investigated with different values of pH (4,7,10) for Na₂EDTA .

The removal of each contaminant can be calculated using the following Eq.(1), **Reddy**, and **Chinthamreddy**, 2000. as :



Contaminant removal % = $\frac{\text{contaminant mass in superntant (C_LV_L)}}{\text{initial contaminant mass in soil (C_SM_S)}} \times 100\%$ (1)

where:

 C_L : concentration of contaminant in supernatant (mg/L). C_S : concentration of contaminant in soil (mg/kg). V_L :volume of supernatant (L). M_R : dry mass of the soil (kg)

 M_S : dry mass of the soil (kg).

4. Results and Discussion:

4.1 Effect of pH

Solution pH is an important factor determining the efficiency of extraction because it can influence the soil retention of metals by extraction , **Peters**, **1999**, and effect the capability of extractant to extract the contaminants from soil through different mechanisms.

Fig. 1 shows the effect of pH on removal efficiency. Highest removal efficiencies (88% for Pb, 97% for Cd and 24% for Ni) were obtained at lower pH=4, because of protons (H⁺) can promote oxide dissolution, besides protons (H⁺) can react with soil surface sites (layer silicate minerals and /or surface functional group Al-OH, Fe-OH, and COOH groups) and enhance extraction of metals cations ,which are transferred into washing fluid. Also the mobility of heavy metals increased with the pH value of soil decreased. Hydroxo complexes tend to absorb protons in acidic pH, as a result, the concentration of effective anion in soil surface decreased and the ability of extracting heavy metals was weakened. However, further increase in pH values would cause a decreasing in removal efficiency. This may be attributed to the formation of negative (Pb , Cd and Ni) hydroxides Pb(OH)₂⁻, Cd (OH)₂⁻ and Ni(OH)₂⁻, which are precipitated from the solution making true extract studies impossible.

HCl solution concentrations of (0.01, 0.05, 0.1, 0.5 and 1) M were used in the present study, pH values of these concentrations are (2, 1.7, 1.44, 1.16 and 1.08) respectively. NaOH did not added to neutralize the acid because this will reduce the acidic property or loss will overshadow(OH) instead of (H) and thus will reduce the movement of metals in polluted soil, which are necessary for the transfer of pollutants to the washing solution and therefore will prevent or reduce extraction. This agrees previous studies in the use of HCl. Also at pH < 2 this dissolution process replace ion exchange **,Kuo, et al.,2006.**

4.2 Effect of Contact Time

Extraction time plays a very important role in soil washing. The percentages of removal of (lead, cadmium and nickel) were determined using Na₂EDTA and HCl solutions. Heavy metals were estimated at different contact times (1, 2, 3, 4, 5 and 6 hr) keeping the other parameters (pH, extractant concentration, and agitation speed) fixed. Metal removal efficiency by chemical extraction process depends on soil geochemistry (soil texture, cation exchange capacity, organic matter, particle size, and large surface area of contaminated soils allow extract of pollutants). They were observed that the heavy metals removal increase as the contact time increase and it remains constant after reaching the equilibrium ,**Mohanty**, and **Mahindrakar**, **2011.** Because of a large amount of extractant became exhausted , the extraction rate is controlled by the rate at which the extract is transported from the soil exterior to the interior sites of the extractant, because buffering capacity for soil reaching equilibrium. However, further

increase in contact time had no significant effect on heavy metals removal. Figs.2 and 3 show the effect contact time on extraction (Pb, Cd and Ni) using Na₂EDTA and HCl .

4.3 Effect of Extractant Concentration

Chelating agent modify metal concentration in soil solution by forming various soluble complexes , thus enhance metal removal (**Reddy andChinthamreddy,2000**). Different concentrations (0.001, 0.005, 0.01, 0.05 and 0.1M) of Na₂EDTA were used in the present study. The soil washing with Na₂EDTA extractant was at pH=4. For the extraction of contaminated soil using HCl extractant, different concentrations of HCl (0.01, 0.05, 0.1, 0.5 and 1M) were used . No pH adjustment was made in the experiments of soil extraction by HCl. The extraction of heavy metals from contaminated soil using Na₂EDTA and HCl was at solid/liquid ratio 1/10 and shaking speed 200 rpm.

Removal of the three metals increased with increasing Na_2EDTA and HCl concentrations and this agree with result of **Kirpichtchikova**, et al., 2006. This may due to the soil's properties, such as metals distribution and physicochemical forms in the soil.

High concentrations achievedacidic mediumwhich isessentialforthemovement of pollutants and ease of transmission and exchange of ions with washing solutions. Also this was expected that the higher concentrations of extractant in soil, the greater availability of extraction. The extraction were carried out using different dosage of extractants as shown in **Figs.4** and **5**.

4.4 Effect of Agitation Speed

Effect of agitation on removal efficiency is shown in **Figs .6** and **7**. These figures showed that the removal efficiency decrease after 200 rpm speed. This may due to stick a part of the contaminated soil on inside surface of volumetric flask neck during the operation of thermostatic mixer which lowers the extraction of metals. Therefore, 200 rpm was taken as the best agitation speed where maximum removal efficiency can be obtained .

4.5 Kinetic Models for Soil Washing

In order to examine the heavy metals extraction mechanism, kinetic data were fitted with four mathematical models: first order, parabolic diffusion, two constant and elovich model. **Min, et al., 2008** presented the linear forms of these mathematical models as shown in **Table 4** which were applied for experimental data in the present study.

2.5.1 First order kinetic model

The first-order model is generally expressed as below Eq.(2) (Kue and Lotes, 1973): $lnq_t = lnq_0-k_1t$

Where q_t and q_0 are amount of contaminant desorbed (mg/kg) after time period of extraction , k_1 is first order rate constant (s⁻¹).

2.5.2 Parabolic diffusion kinetic model

Khater and Zaghloul proposed parabolic diffusion kinetic model, which can be written as Eq.(3)

, Khater and Zaghloul, 2002.

 $q_t = q_0 + k_p t^{0.5}$

Where $k_p =$ is diffusion rate constant (mg contaminant/ kg) ^{-0.5}

(2)

(3)



2.5.3 Two-constant kinetic model

The two-constant model is generally expressed as below Eq.(4) , **Dang et al.,1994** : $q_t=at^b$

Where a= is initial contaminant desorption rate constant (mg contaminant/kg/s)^b, b= is contaminant desorption rate coefficient (mg contaminant/kg)⁻¹

2.5.4 Elovich Kinetic Model

A widely used equation to describe the kinetics of chemical desorption is the Elovich Eq.(5)

,Polyzopoulos et al., 1986: $q_t = 1/\beta_s \ln \alpha_s \beta_s + 1/\beta_s \ln t$

(5)

(4)

Where α_s = is initial contaminant desorption rate (mg contaminant/kg/s), β_s =is contaminant desorption constant (mg contaminant/kg)⁻¹

The kinetic constants of each model were obtained using Microsoft excel software. From calculating the coefficient of determination (R^2) of each model ,the best fit model can be found.

This fitting can be intercept by the higher coefficients of determination (R^2) as shown in **Tables** 5 and 6.

The parabolic diffusion model provided the best correlation {coefficient of determination (R^2) } with experimental data.

5. CONCLUSION

1- The batch result indicated that several factors such as extraction or equilibrium time, initial pH of the solution, extractant concentration and agitation speed affect the extraction process. The best values of these factors that will achieve the maximum removal efficiency of heavy metals can be summarized as follow:

- a-Using Na₂EDTA as extractant, the best contact time was three hours for removing lead and four hours for removing cadmium and nickel .
- b- Using HCl as extractant, the best contact time was four hours for removing lead and cadmium, and five hours for nickel.
- c- The best pH value was 4 when using Na₂EDTA .
- d- Maximum removal percentage of lead, cadmium and nickel in batch extraction was obtained at concentrations of 0.1 M Na₂EDTA and 1 M HCl.
- e- The agitation speed 200 rpm gave higher removal efficiencies compared with 250 rpm.

2- The removal efficiency of extraction increase with:

- b- Increase time until reach equilibrium time.
- c- Decrease pH solution.
- d- Increase extractant concentration.
- e- Decrease agitation speed to a certain limit.

3- The sequence of heavy metals removal was Cd >Pb>Ni.

4-The parabolic diffusion model provided the best correlation with experimental data.



REFERENCES

- Dang, Y.P., Dalal, D.G., Edwards ,D.G., Tiller, K.G., 1994, Kinetics of Zinc Desorption from Vertisols, Soil Sci. Soc. Am. J.Vol. 58, PP 1392-1399.
- Davydova, S., 2005, Heavy Metals as Toxicants in Big Cities, Microchemical Journal, Vol.79, PP.133–136.
- Evanko, C.R., and Dzombak, A., D., 1997, Remediation of Metals- Contaminated Soils and Groundwater, Technology Evaluation Report, TE-97-01.
- Joseph, S. H., Wong, R., Hicks, R. E. and Probstein, R. F., 1997, EDTA Enhanced Electroremediation of Metal Contaminated Soils, J. Hazardous Materials. Vol.55, No. 1-3, PP. 61-79.
- Isoyama, M., and Wada, S.I.,2007, Remediation of Pb-Contaminated Soils by Washing with Hydrochloric Acid and Subsequent Immobilization with Calcite and Allophanic Soil, J. Hazard. Mater., Vol.143, PP. 636–642.
- Khater, A.H., and Zaghloul, A.,M., 2002, Copper and Zinc Desorption Kinetics from Soil: Effect of pH, In 'the 17thWorld Conference on Soil Science, Vol. 47, PP.1-9.
- Kirpichtchikova, T.A., Manceau, A., Spadini, L., Panfili, F., Marcus, M.A., and Jacquet, T., 2006, Speciation and Solubility of Heavy Metals in Contaminated Soil Using X-ray Microfluorescence, EXAFS Spectroscopy, Chemical Extraction, and Thermodynamic Modeling, Geochimica et Cosmochimica Acta., Vol. 70, No. 9, PP.2163– 2190.
- Kue, S., and, Lotse, E.G., 1973, Kinetics of phosphate adsorption and desorption by hematite and gibbsite, Soil Science, Vol.116, PP. 400-406.
- Kuo, S., Lai, M.S., Lin, C.W.,2006, Influence of Solution Acidity and CaCl₂ Concentration on the Removal of Heavy Metals from Metal-Contaminated Rice Soils, Environ. Pollut., Vol. 144, PP.918–925.
- Min, Z., Bohan, L., Ming, L., Young, Z., Qingru, Z., Bin, O., 2008 , Arsenic Removal from Contaminated Soil using Phosphoric Acid and Phosphate, Journal of Environmental Sciences, Vol. 20, PP. 75-79.
- Mohanty,B.,andMahindrakar,A.B.,2011,Removal of Heavy Metal by Screening Followed by Soil Washing from Contaminated Soil, International Journal of Technology and Engineering System (IJTES).Vol.2, No. 3,PP.290-293.
- Peters, R.W., 1999, Chelant Extraction of Heavy Metals from Contaminated Soils, Journal of Hazardous materials, Vol. 66, PP.151-210.
- Phelelani,P.,2007,Remediation of Soil and Water Contaminated by Heavy Metals and Hydrocarbons using Silica Encapsulation, MSc. Thesis, Witwatersrand University ,College of Science.



- Polyzopoulos, N.A., Keramidas, V.Z., Pavlatou, A .,1986,On the Limitation of the Simplified Elovich Equation in Describing the Kinetics of Phosphate Sorption and Release from Soils, Journal of Soil Science, Vol.37,PP. 81-87.
- Reddy, K.R., and Chinthamreddy, S., 2000, Comparison of Extractants for Removing Heavy Metals from Contaminated Clayey Soils, Soil and Sediment Contamination, Vol.9, PP. 449–462.
- Shumba,T.,2008,Removal of Heavy Metals from Crud and Slimedam Material using Soil Wshing and Bioremediation, MSc. Thesis ,Stellenbosch University, College of Engineering.
- SRI,2001, Hydrochloric Acid,Chemicals Economics Handbook .733.http://en.wikipedia.org/wiki/Hydrochloric _acid.
- Tandy, S., Bossart, K., Mueller, R., Ritschel, J., Hauser, L., Schulin, R., and ,Nok,B.,2004, *Extraction of Heavy Metals from Soils using Biodegradable Chelating Agent*, Environ.Sci.Technol.,Vol, 38, PP. 937-944.

> NOMENCLATURE

a= is initial contaminant desorption rate constant (mg contaminant/kg/s)^b

b= is contaminant desorption rate coefficient (mg contaminant/kg)⁻¹

A= the intercept ; B: the slope

 C_L = concentration of contaminant in supernatant (mg/L).

 C_S = concentration of contaminant in soil (mg/kg).

 k_1 = is first order rate constant (s⁻¹)

 k_p = is diffusion rate constant (mg contaminant/ kg) ^{-0.5}

 $M_S = dry$ mass of the soil (kg).

 q_t , q_0 = are amount of contaminant desorbed (mg/kg) after time period of extraction

- S_0 = initial pollutant content in soil (concentration of contaminant in soil) (mg/kg).
- S= pollutant removal content at time (concentration of removal from soil with time) (mg/kg).

t: contact time (hour).

 V_L :volume of supernatant (L).

 α_s = is initial contaminant desorption rate (mg contaminant/kg/s)

 β_s =is contaminant desorption constant (mg contaminant/kg)⁻¹

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Property	Soil
Particle size distribution (ASTM D 422)	
Sand (%)	1.5
Silt (%)	63.5
Clay (%)	35
Cation Exchange Capacity (meq/100g)	12.5
Initial pH	8.3
Background concentration of cadmium (mg/kg)	nill
Background concentration of nickel (mg/kg)	3.52
Background concentration of lead (mg/kg)	15
Organic matter (%)	0.49
Organic carbon (%)	0.16
Electrical conductivity EC (µS/cm)	593
Surface area (m ² /g)	22.776
Bulk density (g/cm ³)	1.1317
Porosity (n)	0.493
Specific weight	2.69
Soil classification	Silty
Son classification	clay loam

Table 1.Composition and properties soil used in the present study .

Volume	21	Januarv
volume	41	Januar y

Property	Value
Molecular weight	372.24 g/mole
Density	1.01 g/cm ³
pH (5% solution)	4.3- 4.7
Water – in soluble matter	0.003%
Chloride (Cl)	0.004%
Nitrilotriacetate N(CH ₂ COO) ₃	0.05%
Sulphate (SO ₄)	0.02%
Calcium(Ca)	0.002%
Copper (Cu)	0.0001%
Iron (Fe)	0.0005%
Lead (Pb)	0.0005%
Magnesium (Mg)	0.001%
Potassium (K)	0.002%

Table 2. Physical and chemical properties of Na2EDTA.

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Table 3. Physical and chemical properties of HCl.

Property	Value
Molecular weight	36.46 g/mole
Minimum assay	35%
Specific density	1.19 g/cm^3
Sulphate (SO ₄)	0.0005%
Sulfite (SO ₃)	0.001%
Free chlorine (Cl)	0.0001%
Heavy metals (as Pb)	0.0005%
Iron (Fe)	0.0002%
Arsenic (As)	0.0001%



Model	Linear form
First -order	$\ln(S_0 - S) = A - Bt$
Parabolic diffusion	$S = A + Bt^{1/2}$
Two-constant	$\ln S = A + B \ln t$
Elovich	S = A + Blnt

 Table 4. The mathematical models applying to fit experimental data . Min, et al., 2008

where:

t: contact time (hour).

- S_0 : initial pollutant content in soil (concentration of contaminant in soil) $(mg/kg). \label{eq:solution}$
- S: pollutant removal content at time (concentration of removal from soil with time) (mg/kg).
- A: the intercept ; B: the slope.

Table 5.The coefficient of determination (R^2) of kinetic models for single component systemusing Na2EDTA.

Models	Coefficient of determination (R²)		
	Pb	Cd	Ni
First -order	0.591	0.753	0.896
Parabolic diffusion	0.9286	0.8514	0.8946
Two-constant	0.6048	0.6544	0.7438
Elovich	0.4495	0.7758	0.7758

Table 6. The coefficient of determination (\mathbb{R}^2) of kinetic models for single component systemusing HCl.

Models	Coefficient of determination (R ²)		
	Pb	Cd	Ni
First -order	0.078	0.821	0.333
Parabolic diffusion	0.7818	0.843	0.7646
Two-constant	0.0132	0.7758	0.7146
Elovich	0.0317	0.8523	0.7537

Number 1





Figure 1.Effect of pH on removal efficiency of (*lead, cadmium and nickel*) as a function of contact time using Na₂EDTA (Na₂EDTA concentration = 0.1M ;speed = 200 rpm).



Figure 2.Removal efficiency of *(lead, cadmium and nickel)* as a function of contact time using Na₂EDTA (pH =4; Na₂EDTA concentration =0.1M; speed=200 rpm).



Figure 3.Removal efficiency of *(lead, cadmium and nickel)* as a function of contact time using HCl (pH = 1.08; HCl concentration = 1M; speed=200rpm).



Number 1



(c)Nickel

Time (hr)

Figure 4.Effect of Na₂EDTA concentration on removal efficiency of (*lead, cadmium and nickel*) as a function of contact time (pH =4; speed=200 rpm).







(b)Cadmium



(c)Nickel

Figure 5.Effect of HCl concentration on removal efficiency of *(lead, cadmium and nickel)* as a function of contact time (speed =200 rpm).



Figure 6.Effect of agitation speed on removal efficiency of (*lead,cadmium and nickel*) as a function of contact time using Na₂EDTA (Na₂EDTA concentration =0.1M; pH =4).



Figure 7.Effect of agitation speed on removal efficiency of (*lead,cadmium and nickel*) as a function of contact time using HCl (HCl concentration =1M; pH =1.08).