



OPTIMIZATION OF COPPER REMOVAL FROM CLAY SOIL USING ELECTRO-KINETIC REMEDIATION BY TAGUCHI METHOD

Maryam S. Karbool, Sata K. Ajjam and Hassan A.Z. Al-Fetlawi

College of Engineering, University of Babylon, Iraq.

Abstract

One of the advanced technologies is the treatment technology of electro-kinetic that presents considerable pledge for the cleanup the earth from metals that are heavy. The efficiency of the electro-kinetic method depends on several factors that have been studied many of them in this study by using the “Minitab17” design for the purpose of removal the copper (pollution). The program depended on several factors: (1) pH, (2) concentration of copper and (3) the gradient in voltage. The program present that the soil potential of electrical is stationary with the time and the soil degradation and the temperature effect are negligible. The results of the experiments show that the use of this program has succeeded in using vinegar as a purification resolution and palm fibers to prevent reverse osmosis, in increasing the efficiency of copper removal of clay soil.

Key word: Electro-Kinetic, Copper Removal, Soil Remediation, Vinegar, Palm Fibers, Taguchi method.

Introduction

Heavy metals lead to polluting the surface of the earth and that is truly widespread. To clean up these sites pollution and as this process is big and accelerated, many efforts are done with observance physicochemical and bio sides of the type of polluted (Oyanader, 2004). Even though there are succeed and ecological kindly soil remediation technologies nevertheless they have not been totally calculated and performed. There are numerous mechanisms that have fascinated the attention between scientists and manufacturing administers. These are: biotreatment, extraction the vapor of soil, thermal desorption and washing and flushing of soil (Virikutyte *et al.*, 2002).

The advantage of electro-kinetic treatment as compared with methods of conventional treatment which are (1) easily, (2) safety to avoid workers and people from contaminants, (3) A large area of polluting media which used for soils, mud, sediments and groundwater that proper for low permeability and non-homogeneous soil deposits within the Faduz area where traditional remedial ways have sure unsuccessful or costly, (4) a extensive domain of pollution, applied for metals, radionuclides, or groups of these pollutants, (5) resilience,

applied as an on-site or off-site management way and can be easily inserted with other treatment techniques like as biological treatment (Reddy, 2010).

Materials and Methods

The Sample of Soil

The soil that was imparted of a depth ranged from 30 to 50 cm below the ground surface from an agricultural area in the Al Mahawil district north of Babylon province. It was cleaned by removing any stones and plant roots in it and was further desiccated and sift subsequent with a sieve of 2mm to get satisfying uniformity. Table 1, explained the composition of soil and their properties that applied in this work. The soil has 13% sand, 21% CaCO₃, 24% clay and pH 7.7. The soil hence can be classified a clay soil.

Copper Contaminant

The pollution of soil simulated *via* copper, a Cu(NO₃)₂.3H₂O solution were equipped and additional to the specimen to get a envoy concentration. Because of calculation sample of producing (600 mg/kg) copper to weight of soil and primary humidity satisfied equivalent to 30%, quantity of 2.26g of Cu(NO₃)₂.3H₂O molecular weight of 241.g/mol was dissolved in 500 ml of distilled water and additional dry soil of 400 ml to 1 kg.

Table 1: Soil Composition and Properties.

Property	Value
Particle size distribution(ASTM D 422)	
Sand (%)	13
Silt (%)	61
Clay (%)	24
Cravel (%)	0
Atterberg limit (ASTM D 2487)	
Liquid limit (%)	42.10
Plastic limit (%)	28.48
Plasticity index (%)	14.60
Compaction text	
Max dry density (gm/cm ³)	1.74
pH	7.7
CaCO ₃ (%)	21

The Solutions for Purging

To examine the electro-kinetic treatment method performance (by applying improvement solution), Vinegar inadequate pH 3, 4 and 5 to eliminate copper as of the soil contaminated *via* this metal. Under definite conditions, the used of improvement agents to solubility the contaminant is essential pro cost efficient implementation. Iraqi date vinegar (IDV), liquid that including a water, 5-20% acetic acid and some trace chemicals was called Vinegar.

Date Palm Fibers (DPF)

DPF were composed as of the tronks and cut into little pieces, distilled water washed to eliminate grime and dehydrated at 80°C to take away the humidity satisfied. It was argument and sifter to dissimilar particle size by vibratory sieve after drying process. By the SEM testing, the good bio sorbent DPF show that contain a many physical and chemical properties (Mohamed *et al.*, 2013).

Reactor Setup

The treatment of EK includes the low level D.C.

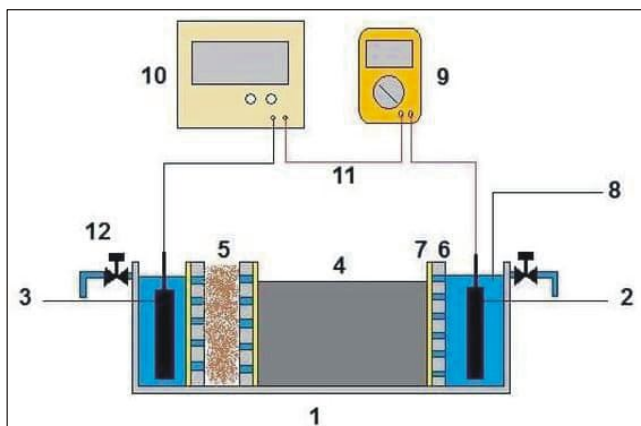


Fig. 1: Electro-Kinetic Cell Schematic diagram; 1. Glass basin, 2&3. Graphite electrodes, 4. Soil, 5. pdf, 6. Perforated plastic plates, 7. Filter paper, 8. Purging solution, 9. Multi-meter, 10. DC, 11. Electrical wires, 12. Valve.

current applying or low voltage slope a traverse electrodes that are at polluted soil. A nearly everyone significant electron move reactions which happen at the electrodes through the way are the water electrolysis (Alkilabi, 2005) and the EK trial system that utilized at the work was show in fig. 1. The setup contains a cell of electro-kinetic, two electrode sections, multi-meter and a power supply. The inter dimensions of glass cell of electro-kinetic equal to (40 cm × 8 cm × 10 cm high) but the soil specimen real length equal to 20cm. The electro-kinetic cell barrier is a DPF that is between the cathode section and soil having 4cm as a thickness. The valve of each electrode is to control the cell flow.

Perforated Plastic Plates were utilized to divide the DPF blockade as of one end soil and to the other end cathode electrode. The plates contain dimensions of 8 cm × 12 cm and contain 6 mm holes diameter, at 1 cm space starting hole center to another hole center.

Experimental Design

In this study, we used the “Taguchi Method” to calculate the previous variables and select the best conditions and variables that effect the removal of copper.

These variables: pH (3, 4, 5), concentration of copper (600mg/kg, 900 mg/kg, 1200 mg/kg), voltage (20 V, 25 V, 30 V) and used vinegar as abolition solution and DPF as a reverse osmosis avoid.

• Taguchi Design of Experiments DOE: Taguchi methods are statistical methods developed by Genichi Taguchi in Japan to optimize the procedure factors and get better the feature of products, afterward its purpose was extended to a lot of extra grounds in Engineering, such as Biotechnology. This technique is able to decrease the experiments number capable for covering the scheme assortment similar to all other investigational design methods (*i.e.* Factorial, Full Factorial etc.) (Alrubaye, 2013). The traditional trial design ways require a large number of experiments therefore becomes complex with increasing number of factors, while Taguchi approach reduces number of experiments compared to the other systems which have same number of control variables (Kapil, 2012).

The following steps for implementing Taguchi Experimental design (Madaeni, 2006):

1. Choosing the changeable reaction to be optimized.
2. Recognizing features (the input variables) and selecting the levels.
3. Choosing the proper orthogonal collection design (numbers of experiment).
4. Performing the experiments.

5. Analyzing results by means of signal-to-noise ratio (S/N).

6. Determining the optimum levels.

In the orthogonal matrixes in the Taguchi process, is proposed to employ a low number of trails to get practical numerical in sequence. An illustration process for orthogonal matrixes is $(La = nw)$, where (L) is the Latin squares of orthogonal matrix, (a) is the experimental number, (n) is the factor levels number, (w) is the factors number (parameters) (Tseng, 2013).

• Identify Factors (variables) and Their Levels: Three factors which are pH, voltage and copper concentration with three levels for each were selected. This study orthogonal collection (Three factor and three levels) is OA =L9 (Eriksson *et al.*, 2000) table 2 shows the factors with the levels used.

Chemical Analysis

The total copper content into dissimilar soil parts was acid absorption extorted (Haswell, 1991). The oven-dried samples were ground into fine particles by a soil grinder mill. 1 g of specimen was scaled and set in a 50 ml Teflon tube. 15 ml of HNO₃ and 5 ml of HCl with concentration of 69.9% were added to the sample. The mixture was heated, for about (45-60) minutes, in the arenaceous bath awaiting the brown steams vanished and a specimen reaches to dry case. Then the beaker was left for (5-10) min in the lab temperature, added a 5 ml of HCl centered, heated for a second time in the sandy bath, then cooled and added 5 ml of HCl centered and 50 ml of heated distilled water to clean the beaker from residue dissolved specimen and the mix was heating to the boiling points for (2-3) min. The specimen was filtered on the filter paper No. 42 and then saves it in volumetric vial ability of 100 ml. Then the precipitation washing with distilled water and the before washed water addition to filtration and full the size to 100 ml. lastly, the copper concentration was calculated

Table 2: Results based on Taguchi traditional design for copper removal.

SNRA1	Removal (R%)	Voltage €	Concentration ©	pH	Series
34.6639	54.1	20	600	3	1
33.7327	48.6	25	900	3	2
31.8435	32.1	30	1200	3	3
34.1514	51.0	25	600	4	4
32.5473	42.4	30	900	4	5
26.1070	20.2	20	1200	4	6
30.9555	40.3	30	600	5	7
26.8878	22.1	20	900	5	8
25.8893	19.7	25	1200	5	9

by atomic absorption spectrophotometer (AAS).

Results and Discussion

Application of Taguchi method

The results investigation based on Taguchi process needs to approximation signal-to-noise ratio (R%) and in this study signal-to-noise was selected for larger-the-enhanced (pH, Voltage, Concentration). In this work was the best (pH = 3, Voltage = 30, Concentration = 600).

Analysis of the results is done using MINITAB version 17.0 statistical software, these result are shown in table 2 and fig. 2.

Effect of pH, Copper concentration and Voltage gradient

Fig. 3, 4, 5 and 6 illustrate the whole concentration of copper, electro-osmosis and EC difference every part of the treated soil for EX1, EX2 and EX3 at pH of 3 by using Vinegar. Whenever a few value of the pH (the acidity increased) whenever the removal is better, because the more acidity has increased the editing of hydrogen ions that help analyze complex compounds in soil and thus increasing electro migration. In these three tests the pH value was constant (pH =3), but the voltage and concentration of Copper were variable in each test, EX1 (pH=3, C=600 mg\kg, E=20 V), EX2 (pH=3, C=900mg\kg, E=25 V) and EX3 (pH=3, C=1200

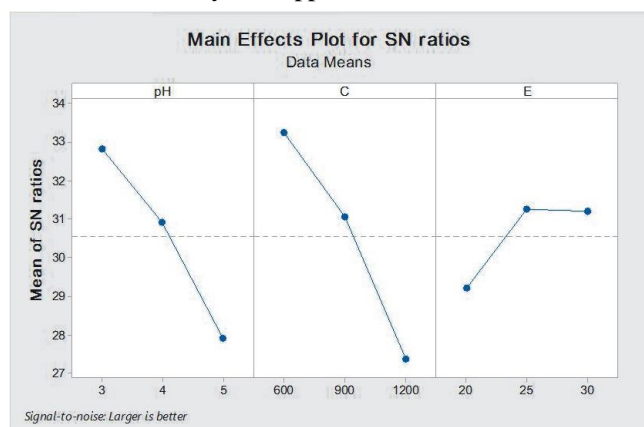


Fig. 2: The trend for pH, C and E in final removal present for copper.

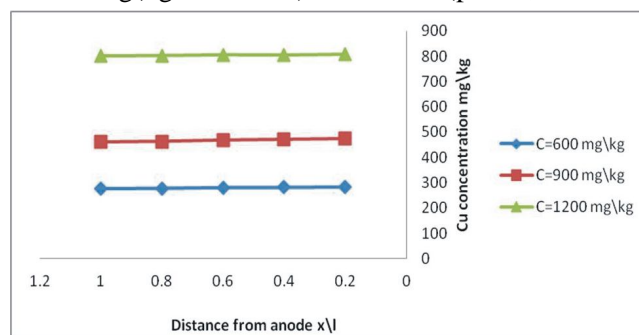


Fig. 3: The Cu concentration (mg/kg) against to the distance from anode at pH 3.

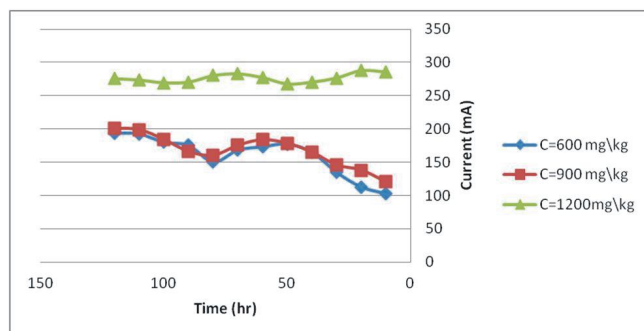


Fig. 4: The current (mA) vs. to the time (hr) at pH 3.

mg/kg, $E=30$ V). The produced hydrogen and hydroxyl ions are elated during the sample to the reverse directions via electro-movement, dispersion and advection. The stand front progress is slower than the acid front progress due to the offset electro-osmotic stream and too as the H^+ ions contain higher ionic mobility values than OH^- (Acar and Alshawabkeh, 1993) as soon as the acid and stand fronts get together, water was shaped consequential in a sharp vary in pH, that influenced the solubility of polluted and their soil particles adsorption. One more problem is symbolized *via* the pH effect on the minerals of soil, a raise in definite ions concentrations (Mg, Al and Fe) are frequently shown consequently of the raised acidity and others (Si and Al) of the greater than before alkalinity. Al ions are usually noticed in soils aperture solutions that have submitted electro-kinetic treatment.

Adjust in the pH profiles all along the soil had a huge influence on pollutant movement. Trails behavior in this work appeared that the pH has an important shock on Cu movement in the soil. In low PH at the anode, the Cu concentration reduced. The high pH indicates that a few OH^- ions were getting the soil and probably interfere with the copper ions movement.

The high pH surroundings was positive to create $Cu(OH)_2$ that precipitate close to the cathode parts and reduce the conductivity of the soil.

The current relative to the concentration of the metal pollutant, as the pollutant ions donate to the ionic strength

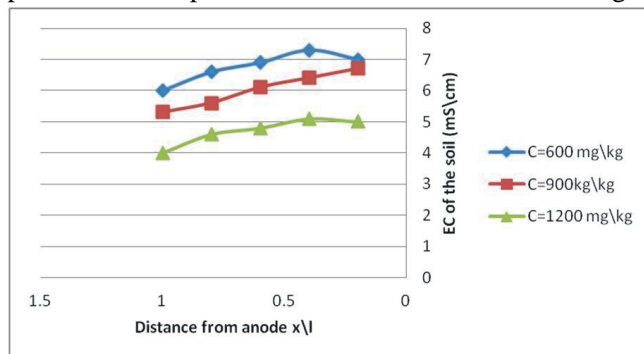


Fig. 5: The EC of the soil (mS/cm) versus the distance from anode (x\l) at pH3.

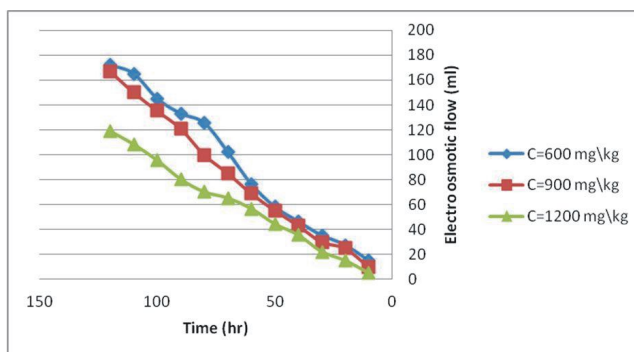


Fig. 6: The volume of electro osmotic flow (ml) versus the time (hr) at pH 3.

and the pore solution conductivity, the fig. 4, explains that, like predictable. And the raise in elimination efficiency by the raise in voltage gradient was according to the raise in together electro-osmotic and electro-movement rates that are linaerly relative to the practical electrical incline. On the other hand, at higher voltage gradients, higher current densities are produced that raise the electrolysis reaction rate at the electrodes. These electrolysis reactions create H^+ and OH^- at a quicker speed than the speed at that soil be able to permit them to move (Reddy and Shirani, 1997).

Cumulative effluent collected from all the tests decreased when increase the initial concentration of Copper.

The electro osmotic flux is straight relative to current and suspends pollutants in attendance in the hole liquid.

Effect of Palm Fiber

Based upon this experimental work, (DPF) have swimmingly vetoed the undo electro-osmotec flux, that has a negative consequence on the path and scale of copper movement through EK procedure. (DPF) were efficient in adsorbing copper metal as of the aqueous solution. Because of the high adsorption ability of (DPF), little quantity of copper ions were adsorbed through (DPF); since the salts ions opposition previously obtainable in the soil specimens as in table 1 with copper ions (Shukla *et al.*, 2003). But, the copper ions quantity that adsorbed through (DPF) was in the variety of 105 mg/kg, 96 mg/kg and 93 mg/kg in the Vinegar (EX-1, EX-4 and EX-7) correspondingly.

Conclusions

The most important conclusion of the present study is so as to the elimination efficiency of copper raises *via* declining pH of removal solution since in high pH values, the metal ions inclination to grow to be adsorbed on to the earth particles too raises. And the elimination rose through raises voltage. The Copper elimination efficiency *via* utilizing Vinegar is the most excellent; since vinegar involves CH_3COOH , C_2H_5OH , sugar and further

compounds and thus can perform as a tough natural chelating instrument. Additionally, vinegar is readily obtainable, ecology kindly and doesn't relate or has every negative collision on soils. Vinegar and as in mainly natural compounds, be able to decrease pH in calcareous soils (the kind that is utilized in this work) and get better soil setting for better microbial actions, nutrients accessibility and too soil gathering constancy and construction. DPF were establish to be an efficient ecological bio sorbent matter in avoiding the electro-osmosis method in sandy soils; particularly as the cumulative electro osmotic effluent developed into high in such procedure. Though, EK method *via* with both obtainable date palm goods shows as appropriate solution pro the treatment of heavy metals dirty soils in Iraq.

References

- Alkilabi, J. (2005). "Role of Mycorrhiza, different of Phosphate Sources and Compost on the Availability of Phosphors and Copper in soil, growth and yield of Maize (*Zea mays* L)."
- Acar, A.N. and Y.B. Alshwabkeh (1993). "Principles of electro-kinetic remediation," *Environ. Sci. Technol.*, **27**: 2638-2647.
- Alrubaye, R. (2013). "Generation and Characterization of Catalytic Films of Zeolite Y and ZSM-5 on FeCrAlloy Metal," MSc Thesis, University of Manchester., 2013.
- Haswell, S.J. (1991). *Atomic absorption spectrometer; theory, design and applications*. Amsterdam: Elsevier.
- Kapil, K. (2012). "Effect of Ti Loading on the HDS and HDN Activity of KLGO on NiMo/TiSBA-15 Catalysts, *Microporous and Mesoporous Materials.*, **152**: 224-234.
- Madaeni, S.S. (2006). "Application of Taguchi Method in the Optimization of Waste Water Treatment Using Spiral-wound Reverse Osmosis Element," *Chem. Eng. J.*, **119**: 37-44.
- M.J. Shukla, S.S., L.D. Kenneth and A. Shukla (2003). "Adsorption of Chromium from Aqueous Solutions by Maple Sawdust," *J. Hazard. Mater.*, **100**: 53-63.
- Oyanader, M.A. (2004). "Physicochemical and hydrodynamics Aspects of Electro-Kinetics in Soil Remediation," Ph.D. Thesis, Florida State University, College of Engineering.
- Reddy, K.R. (2010). "Technical Challenges to In-Situ Remediation of Polluted Sites," *Geotech. Geol. Eng.*, **28**: 211-221.
- Reddy, A.B. and K.R. Shirani (1997). "Electro-kinetic remediation of metal contaminated glacial tills," *Geotech. Geol. Eng.*, **15**: 3-29.
- S. Eriksson, L., E. Johansson, N. Kettaneh-Wold, C. Wikström and Wold (2000). *Design of experiments. Principles and Applications, Learn ways AB*. Stockholm.
- Tseng, K.H. (2013). "Optimization of Microwave Based Heating of Cellulosic Biomass Using Taguchi Method," *Materials (Basel)*, **6**: 3404-3419.
- Virkutyte, P., J. Sillanpaa and M. Latostenmaa (2002). "Electro-Kinetic Soil Remediation - Critical Overview," *Sci. Total Environ.*, **289**: 97-121.