Effect of Low Frequency AC Supply & Purging Solution on Remediation of Nickel Contaminated Soil using Electrokinetics

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Abstract. The complete earth is prone to natural hazards, cannot be prevented, but through an understanding of the earth condition and the processes which could culminate in damage to life and property, it is possible to minimize the damage through proper disaster management measures. Man-made hazards have today taken over natural hazards in terms of the loss of life & the long term effect on mankind and the ecosystem on the whole. Reddy et al. (1996) reported on laboratory electrokinetic experiments that were conducted on different soils, namely, commercial kaolin and Na-montmorillonite as well as a fieldderived glacial till. The presented work is an attempt to study effect of various variables i.e. type of anodic & cathodic purging solution, periodic supply as well as improvement of soil with application of low frequency AC supply instead of DC supply. The complete study has been carried out on models having different length with the Nickel (Ni(II)) & commercially available Kaolinite being artificially spiked using Nickel Chloride salt. This study on feasibility of using different extracting solution at various use of 0.1M Ethylenediamine tetra acetic acid (EDTA) better as compared to 1M Sodium Nitrate. The Nickel removal efficiency of organic acid such as citric acid was found to be low as compared to other solution, the best removal being achieved with 1M NaCl.

Keywords: Reactor Size, Low Frequency AC Supply, Electrokinetics, Remediation of Nickel Contaminated Soil

1 Introduction

There are many remediation technologies available for treating contaminated soils and groundwater that are classified as either ex-situ or in-situ techniques (Reddy et al., 1999). Ex-situ techniques treat the contaminated soil and/or groundwater after it has been removed from the subsurface, whereas in-situ techniques treat the soil and/or groundwater within the subsurface itself.

Electrokinetics is a process that has shown a great potential for remediating heavy metal contaminated soils, including low permeable clays and/or heterogeneous soils. (Fig. 1) Electrokinetic remediation technique uses the combined effect of electric,

chemical and hydraulic potential for remediation of problematic/ contaminated soil. In this method, a low DC voltage is applied to soil through carbon/sacrificial electrodes and the metals are extracted from contaminated soil in aqueous form. This method is one of the most promising remediation processes; it also has low operational cost and potential applicability to a wide range of contaminant types.



Fig. 1 Process of electrokinetic remediation

2 Literature Survey

The interaction between the soil system and contaminants is of course dependent on soil conditions other than organic matter content. For example, the solubility of metals can vary with pH - lead solubility increases as pH drops (Alshawabkeh and Acar, 1996).

Generally, clay has a net negative charge on its surface. This negative surface electric charge of a clay particle is due to the presence of isomorphous substitution and broken structure. The negative charge on the clay surface is balanced by excess positive charge distributed in the fluid zone adjacent to the clay surface. This distribution adjacent to the clay surface is called the diffuse layer. A diagram of the effect of these charges around an idealized spherical particle is shown in Fig. 2.2 and a qualitative graph of the variation of electrical potential with distance from particle surface is presented in Fig.2.3 along with the idealized distribution of charge. (Michael Harbottle,2003)



Fig. 2.2 Idealized Representation of Water Around A Clay Particle (Michael Harbottle 2003)

Electroosmosis is the transport of pore fluid in soils under the influence of an electric field. Several theories have been formulated to describe the flow of water by electroosmosis. (Fig. 2.11) The most widely accepted model is the Helmholtz Smoluchowski (H-S) theory. According to this theory, the net water flow results when the excess positive charge is transported to the cathode (Probstein et al., 1993). Characteristics of the soil, which influence kinetics of contaminant removal, include adsorption, ion exchange, and buffering capacity (Grim, 1968; Sposito, 1984). Soil contaminant interaction is especially high in fine-grained soils.

3 Material Characteristics

The present work is carried out on Kaolinite sieved through 425μ sieve and then tested for basic geotechnical properties, and found to be CH type, as per IS classification system. Geotechnical properties of Kaolinite were found out using standard methods prescribed in relevant IS codes (IS 2720), and are summarized in **Table 1**. This soil is dried and then mixed with 1.012gm of Nickel Chloride/kg soil with water content kept equivalent to liquid limit. The mixed soil was kept to mature for 1 month before filling them in the models. Model for the study are prepared using PVC pipes of pressure capacity 20kg/cm^2 , with flange end connections. For the entire set of tests, 0.1M Ethylene Diamine Tetra Acetic Acid solution is used at cathode and 1M Sodium Chloride solution is used at anode. Voltage applied for all the experiments is kept at 25 volts in DC and AC mode.

4 Effect of Periodic Supply

Experiments in this phase were to determine contaminant mass removal by using a periodic voltage application in different combinations i.e. one day on & one day off, one day on and two day off and two day on and one day off. The applied voltage and purging remains constant during all tests as mentioned above. Other details are as per **Table 2**

Table 1. Geotechnical Properties of Virgin Soil

Table 2 Details of Different Phases of Work

Property Liquid Limit(%)	Value 73.8	Type of Supply	Frequency (Hz)	Duration of Supply	
Plastic Limit(%)	33.2		Effect of Periodic Supply		
Plasticity index(%)	40.6	DC	0	1 day on – 1 day off	
Shrinkage limit(%)	24.0	DC	0	1 day on – 2 day off	
Specific Gravity	2.3	DC	0	2 day on – 1 day off	
Maximum Dry Density(gm/cc)	1.38	Eff	Effect of Low Frequency AC Supply		
Optimum moisture content(%)	32.3	AC	0.25	Continuous	
Free Swell (%)	96	AC	0.50	Continuous	
Cohesion, c (kg/cm ²)	1.12	AC	0.75	Continuous	
Angle of internal friction, ϕ	3.65	AC	1.00	Continuous	

4.1 One Day On-One Day Off

This study is carried out to study the effect of supply period on remediation of contaminated soil keeping the purging solutions as 0.1M Ethylenediamine tetra acetic acid (EDTA) at cathode and 1M Sodium Chloride (NaCl) at anode. **Fig. 2** shows variation of voltage at anode, middle and cathode compartment along with applied voltage with respect to time. Sudden increase in the voltages is observed subsequent to addition of solutions in the anode and cathode compartments and/or when there was high sacrificial behaviour of electrode at anode. **Fig. 3** shows variation of current and current density. During electrokinetics, the fluctuation in currents can occur due to different reason. The current drop with the shutting down of the supply & shows a low value the next day when reading are noted down after the supply is restarted. The temperature profile also almost follows the current & voltage patterns drop in temperature on the off days. **Fig. 4** shows variation of pH of solution at anode and at cathode at the end of every week.

3.2 One Day On-Two Day Off

This is the second variation used for study where in the shutoff time was double of the supply on time. **Fig. 5** shows variation of voltage near anode, cathode and at middle section along with applied voltage with respect to time. Sudden increase in the voltages is observed subsequent to addition of solutions in the anode and cathode compartments, the current also drops to zero in the off time, the second day shows a current reading since it was taken once the supply was switched on once again. **Fig. 6** shows variation of current and current density. Initially, when the voltage gradient was first applied, the current is low and starts build up slowly because it takes time for the solution migrates into the electrode reservoirs and for the salts and/or contaminants to dissolve. The variation continues showing no stabilizing trend due to the supply off period. **Fig. 7** shows variation of pH of solution at anode and at cathode at the end of every week.



Fig. 2 Variation Of Voltage At Different Sections Along Length Of Sample & Applied Voltage With Time for one day on and one day off model



Fig. 3 Variation Of Current And Current Density With Respect To Time for one day on and one day off model



Fig. 4 Variation Of Temperature & Ambient Temperature With Time for one day on and one day off model



Fig. 5 Variation Of Voltage At Different Sections Along Length Of Sample & Applied Voltage With Time for One day on and two day off model



Fig. 6 Variation Of Temperature & Ambient Temperature With Time for One day on and two day off model



Fig. 7 Variation Of pH At Every Week for One day on and two day off model

4.3 Two Day On-One Day Off

This is the third variation wherein the supply on time was double as compared to the supply off time. **Fig. 8** shows variation of voltage at anode, middle and cathode compartment along with applied voltage with respect to time. **Fig. 9** shows variation of current and current density. During electrokinetics, the fluctuation in currents can occur due to different reason. The graphs follows the trend with the supply on time, there being a decreases in the current, current density , & voltage at various sections as well as temperature during this period. **Fig. 10** shows variation of pH of solution at anode and at cathode at the end of every week, which shows no appreciable change.

After completion of the experiment three soil samples were collected near anode, mid section and near cathode from all the models and then tested on EDS. **Fig. 11** shows concentration of Nickel(II) after electrokinetic remediation and **Fig. 12** shows the percentage removal with respect to the original concentration of 250 mg/kg.

Research has shown that the applied electric potential produces complex physical, chemical, and electrochemical changes within clay soils that affect mass transfer and overall efficiency. From the graphs it can be concluded that greater removal of Nickel is achieved during two day on-one day off supply as compared to one day on-one day off and one day on-two day off periodic supply.



Fig. 9 Variation Of Current And Current Density With

Respect To Time for Two days on and One day Off

model

Fig. 8 Variation Of Voltage At Different Sections Along Length Of Sample & Applied Voltage With Time for Two days on and One day Off model



Fig. 10 Variation Of pH At Every Week for Two days on and One day Off model



Fig. 11 Concentration Of Metal After Electrokinetic Remediation



5 Effect of Low Frequency AC Supply

For this phase the required information is as per **Table 2**, four different frequency of sinusoidal waveform i.e. 0.25, 0.5, 0.75 and 1 Hz has been used in the four models to study the effect of low frequency AC supply. Purging solution used is same as 1st Phase.

4.1 Frequency = 0.25 Hz

Sinusoidal waveform of 0.25Hz frequency was set in AC supply instead of DC supply. **Fig. 13** shows effect of temperature near anode and cathode and at middle section with ambient temperature with time. During experiment, at an interval of one week, samples from both anode and cathode compartments were collected, filtered and tested on pH value. **Fig. 14** shows variation of pH of solution at anode and at cathode at the end of every week. Reading for current could not be taken since the variation was too fast to be recorded. Since it is sinusoidal waveform the voltage cannot be also recorded.

4.2 Frequency = 0.5 Hz

Sinusoidal waveform of 0.5Hz frequency was set in AC supply next for the study. **Fig. 15** shows effect of temperature near anode and cathode and at middle section with ambient temperature with time. **Fig. 16** shows variation of pH of solution at anode and at cathode at the end of every week. The low variation in pH shows that the movement of particle is effected due to change in anode & cathode respectively due to AC supply.

4.3 Frequency = 0.75 Hz

Sinusoidal waveform of 0.75Hz frequency was set in AC supply after 0.5 Hz further carrying on the study further. **Fig. 17** shows effect of temperature near anode and cathode and at middle section with ambient temperature with time. **Fig. 18** shows variation of pH of solution at anode and at cathode at the end of every week. The pH follows the same trend showing no appreciable change in the values, but temperature variation shows appreciable change of 4 to 5°C giving an idea that some flow of ions does definitely occur.

4.4 Frequency = 1.0 Hz

Sinusoidal waveform of 1.0 Hz frequency was set in AC supply instead of DC supply. **Fig. 19** shows effect of temperature near anode and cathode and at middle section with ambient temperature with time. **Fig. 20** shows variation of pH of solution at anode and at cathode at the end of every week.

After completion of the experiment three soil samples were collected near anode, mid section and near cathode from each model and then tested on EDS. **Fig. 21** shows concentration of Nickel(II) after electrokinetic remediation. Nickel concentrations were low in the sections closer to the anode and higher near the cathode. **Fig. 22** shows highest percentage removal of Nickel when the used 0.25Hz frequency AC supply.





Fig. 15 Variation Of Temperature & Ambient Temperature With Time for 0.5 Hz Frequency



Fig. 14 Variation Of pH At Every Week for 0.25 Hz Frequency



Fig. 16 Variation Of pH At Every Week for 0.5 Hz Frequency



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pH at Anode

pH at Cath

Enkl at Cat



Frequency

Fig. 19 Variation Of Temperature & Ambient Temperature With Time for 1 Hz Frequency

Fig. 20 Variation Of pH At Every Week for 1 Hz Frequency



Fig 21Concentration Of Metal After Electrokinetic Remediation



Fig 22 Total Percentage removal of Nickel After Electrokinetics Remediation Using Low Frequency AC Supply

The remediation using low frequency AC supply is relatively small as compared to the DC supply. When using an AC supply even at a low frequency as low as 0.25Hz

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there is change in anode and cathode supply once every four seconds which affects and hinders the movement of ions, thereby the electrical gradient not helping in the movement of the metal ions. This same effect does not allow electromigration and/or migration potential to develop fully neither do other process work as efficiently as with DC supply. AC supply also does not allow the water to break up as speedily as in DC supply also hindering the electroosmosis process which helps in keeping a balance of the pH at the anode and cathode.

6 Conclusions

The following conclusions are deduced from the experimental results:

• It can be concluded that greater removal of Nickel is achieved during two day on-one day off supply as compared to one day on-one day off and one day on-two day off periodic supply. The applied electric potential produces complex physical, chemical, and electrochemical changes within clay soils that affect mass transfer and overall efficiency

• The low frequency AC supply at 0.25 Hz gives a percentage removal of 75% only, since an AC supply even at a low frequency of 0.25Hz has change in positive and negative amplitude once every four seconds which affects and hinders movement of ions. This effect also does not allow electromigration and/or migration potential to develop fully; neither do other processes work as efficiently. It also does not allow the water to break up as speedily as with DC supply, hindering the electroosmosis process which helps in keeping a balance of the pH at the anode and cathode.

• The above results can be used for treating grounds contaminated by various heavy metals caused due to spilling of waste or untreated disposal of plating waste. As opposed to the traditional electrokinetics which uses DC current use of AC supply is easier to work with. The use of AC current reduces the operational hazards on the field. The use of AC current gives an added advantage of breaking of the pH and ion barriers formed in the soil when using DC supply.

• Application of intermittent supply i.e. periodic supply has an added advantage of allowing the reversing of the created pH barrier which in DC supply becomes a major drawback in increasing the removal percentage. It gives an advantage of reducing the energy consumption without major decrement in removal percentage.

6 **References**

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