

Removal of Heavy Metal and Organic Substance in Contaminated Soils by Electrokinetic and Ultrasonic Remediation

동전기 및 초음파 복원기술에 의한 오염지반내의 중금속 및 유기오염물질 제거

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요 지

본 논문에서는 동전기초음파정화기술을 이용하여 오염지반내의 중금속 및 유기물질을 제거하는 연구를 수행하였는데, 지반내 오염물질의 이동 및 제거에 대한 동전기기술과 초음파기술의 복합효과 분석에 초점을 두었다. 일반적으로 오염지반내에서 동전기기술은 중금속을 제거하는데 탁월하며 초음파기술은 유기물질을 제거하는데 탁월한 것을 보고되어 있는바 이들 두 기술의 장점을 이용한 복합기술을 고안하게 되었다. 특수하게 고안된 실험장비를 이용하여 동전기기술과 초음파기술을 결합한 실내토양세척실험을 실시하였다. 본 실험에서는 단순, 동전기, 초음파, 동전기&초음파의 4조건에 대하여 토양세척실험을 실시하였다. 오염물질로는 중금속으로는 납, 유기물질로는 에틸렌글리콜을 사용하였다. 실험결과 동전기기술과 초음파기술을 도입한 경우 유출량, 투수계수, 오염물질 제거율이 단순기술에 비하여 상대적으로 크게 증가하는 것으로 나타났다. 본 연구를 통하여 동전기초음파정화기술을 현장에 적용할 수 있는 가시적인 결과를 얻게 되었다.

Abstract

The electrokinetic technique has been applied to remove mainly the heavy metal and the ultrasonic technique to remove mainly organic substance in contaminated soil. In this study, the combined electrokinetic and ultrasonic remediation technique was studied for the removal of heavy metal and organic substance in contaminated soils. This study emphasized the coupled effects of electrokinetic and ultrasonic techniques on migration as well as remediation of contaminants in soils. The laboratory soil flushing tests combining electrokinetic and ultrasonic technique were conducted using specially designed and fabricated devices to determine the effect of both of these techniques. A series of laboratory experiments involving the simple, electrokinetic, ultrasonic, and electrokinetic & ultrasonic flushing test were carried out. A soil admixed with sand and kaolin was used as a test specimen, and Pb and ethylene glycol were used as contaminants of heavy metal and organic substance. An increase in out flow, permeability and contaminant removal rate was observed in electrokinetic and ultrasonic flushing tests. Some practical implications of these results are discussed in terms of technical feasibility of in situ implementation of electrokinetic ultrasonic remediation technique.

Keywords : Contamination, Electrokinetic, Ground, Pollutant, Remediation, Ultrasonic

1. Introduction

Natural concentrations of heavy metals and organic

substances in soil deposits are not high; however, studies have indicated that many areas near urban complexes, metalliferous mines or major roads display

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abnormally high concentrations of these elements. In response to the demand for developing effective and economical cleanup techniques, numerous studies have been conducted over the years. Several clean-up techniques have been developed; examples include pump-and-treat, soil vapor extraction, soil washing, water flushing, steam extraction, and bioremediation, etc. The pump-and-treat method is ineffective due to the requirement of large equipment with high energy and small removal effect of strong adsorptive contaminants with soil particles. The soil vapor extraction method is not applicable to remove contaminants from saturated soil deposits and ground water. The effectiveness of bioremediation depends greatly on suitable microorganism and nutrients in the subsurface. Therefore, much still remain to be done in order that a generally accepted methodology can be developed for a broad range of applications.

A variety of options may exist in selecting a cleanup method at a site, however the efficiency and costs of these options may vary widely. Most of the existing remediation technologies are limited to soils with high hydraulic conductivities and are not effective in removing heavy metals adsorbed in soil particles, particularly fine-grained deposits. There exists a need to introduce cost-effective, innovative, and preferable in-situ remediation technologies.

Electrokinetic soil processing is a new, innovative, and cost-effective remediation technology that employs conduction phenomena under electric currents for transport, extraction, and separation in soils. The driving mechanisms for species transport are ion migration by electrical gradients, pore fluid advection by prevailing electroosmotic flow, pore fluid flow due to any externally applied or internally generated hydraulic potential difference, and diffusion due to generated chemical gradients. As a result, cations are accumulated at the cathode and anions at the anode, while there is a continuous transfer of hydrogen and hydroxyl ions across the medium. Various laboratory and field studies on the feasibility of the electrokinetic process have shown that heavy metals and other cationic species can be removed from the contaminated soil as illustrated in Figure 1. The feasibility

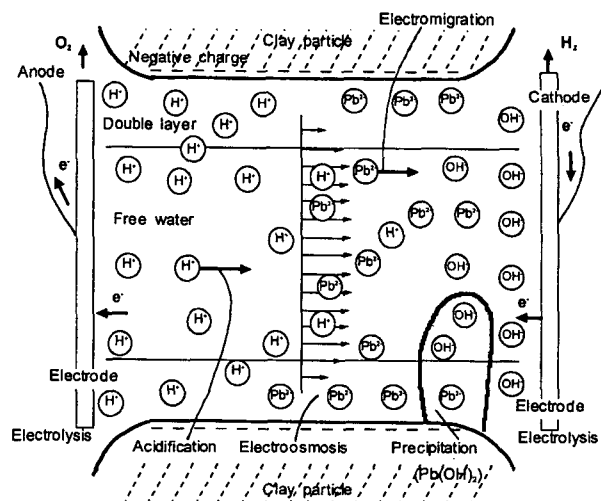


Fig. 1. The effects of electrokinetic phenomena on porous soil media

and cost effectiveness of electrokinetics for the extraction of heavy metals such as lead, copper, zinc, and cadmium from soils have been demonstrated by many researchers (Mitchell 1986; Runnels and Larson 1986; Hamed 1990; Pamukcu et al. 1990; Acar et al. 1989, 1992, 1993 and 1994, Acar and Alshawabkeh 1993, Acar and Gale 1986; Acar and Hamed 1991; Yeung et al. 1994; H. Lee 2000; S. Han 2000).

Numerous researchers have proposed possible mechanisms for the effect of stress waves on fluid flow through porous media. The cavitation and capillary forces are principally responsible for the movement of fluid in porous media. Capillary forces play an important role in liquid percolation through fine pore channels. The liquid films adsorbed onto pore walls during the percolation process can be destroyed by mechanical vibration. The seismoacoustic fields lower the capillary pressure and seismoacoustic wave affects the increase of water saturation and flow in soil stratum. An increase in hydraulic conductivity and contaminant removal was observed from the effect of acoustic excitation on soils. Various researchers have shown the enhancement of the contaminant migration and recovery by acoustic waves through laboratory and field tests (Simikin and Verbitskaya 1989; Suslick 1988; Frederic 1965; Murdoch et al. 1998; Iovenitti et al. 1995, Kim 2000).

2. Laboratory Experimental Methodology

In this study, 4 types of experiments including simple soil flushing experiment, electrokinetic soil flushing experiment, ultrasonic soil flushing experiment and electrokinetic & ultrasonic soil flushing experiment were carried out. Traditional simple soil flushing experiment was combined with electrokinetic and ultrasonic experiment. The schematic view of the experimental setup is shown in Figure 2.

The simple soil flushing processor consisted of three parts: test chamber, inlet and outlet, supply water reservoir. The test chamber is made of a plexiglas cylinder having an inner diameter of 10cm with a height of 30cm. The cylinder was filled with contaminated soil. The inlet and outlet tubes were installed in the lower and upper side part of the cylinder. The inlet tube is connected to a reservoir and the outlet tube is used to maintain constant heads by allowing overflow of excess water through soil specimen. Outlet tube is connected to a burette for measuring the outflow quantity. A reservoir was filled with de-aired and de-ionized water, and mariotte system was installed inside of this reservoir. The mariotte bottle system was used to maintain the constant hydraulic head during experiment from the water level in reservoir to the top of soil specimen.

The electrokinetic soil flushing processor consisted of three parts: anode electrode, cathode electrode, and electric power supplier. The graphite electrode is used and situated on the top (cathode part) and the bottom (anode part) of the soil specimen. A constant voltage

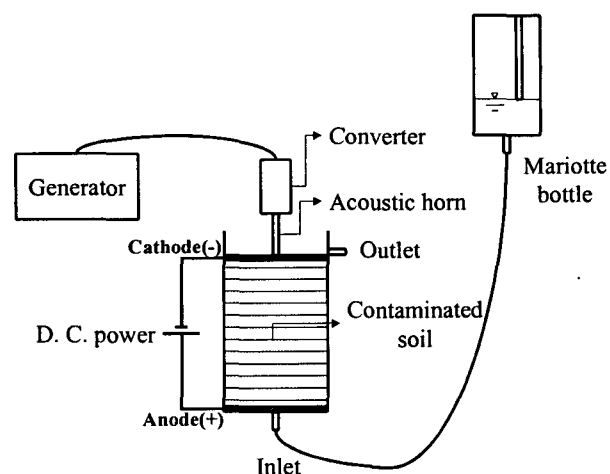


Fig. 2. Test setup for soil flushing experiment

gradient of 2.0 V/cm was applied to the anode and cathode electrodes. Two sheets of filter paper were placed on the upper and lower graphite electrode.

The ultrasonic soil flushing processor consisted of three parts: a generator, a converter, and an acoustic horn. The transmitting acoustic horn, which is mounted on top of the soil sample, is used for generating ultrasound. The ultrasonic processor has a maximum power output of 200W with a frequency of excitation equal to 30kHz.

The electrokinetic & ultrasonic soil flushing processor consisted of three parts: simple soil flushing processor, electrokinetic soil flushing processor, ultrasonic soil flushing processor, that is, the combination of three experiments.

The summary of test conditions is shown in Table 1. An admixed soil in the ratio of sand (90%) and kaolin (10%) was used as a soil specimen, Pb and ethylene

Table 1. The summary of test program for soil remediation by electrokinetic and ultrasonic process

Test No.	Test method	Contaminant	Concentration	Electric gradient (V/cm)	Ultrasonic frequency(kHz)	Hydraulic gradient	Anode reservoir fluid
Test 1	Simple soil flushing	Pb & ethylene glycol	500 mg/kg respectively	-	-	0.5	De-ionized water
Test 2	Electrokinetic soil flushing	Pb & ethylene glycol	500 mg/kg respectively	2.0	-	0.5	De-ionized water
Test 3	Ultrasonic soil flushing	Pb & ethylene glycol	500 mg/kg respectively	-	30	0.5	De-ionized water
Test 4	Electrokinetic & ultrasonic soil flushing	Pb & ethylene glycol	500 mg/kg respectively	2.0	30	0.5	De-ionized water

glycol were used as a surrogate contaminant to demonstrate the soil contaminated by heavy metal and organic substance. The soil flushing tests were conducted for four conditions: simple soil flushing, electrokinetic soil flushing, ultrasonic soil flushing, electrokinetic & ultrasonic soil flushing. Hydraulic gradient for the flushing process was constant to 0.5 during experiment. For the preparation of contaminated soil, the admixed soil specimens were thoroughly mixed with lead of 500mg/kg concentration and ethylene glycol of 500mg/kg concentration. The test specimen was then subjected to ultrasonic waves at 30kHz frequency from ultrasonic test setup and to electric power at 2.0V/cm from electrokinetic test setup.

3. Experimental Results and Discussion

3.1 Outflow Rate Through Soil Specimen

Under the actions of advective flow by hydraulic gradient, electroosmotic flow by electric power, fractured flow by ultrasonic waves, the pore water and contaminant in soil are allowed to flow and migrate from the inlet (bottom of specimen, anode side) to outlet (top of specimen, cathode side). Thus, the advective flow was directed towards inlet connected water reservoir from outlet, the electroosmotic flow was directed towards the cathode from the anode, and fractured flow was directed towards the advance of ultrasonic wave.

The effluent was collected in a 500ml polypropylene cylinder. The quantity of outflow with elapsed time through soil specimen is presented in Figure 3. In this figure, net flow in y-axis means the quantity of outflow

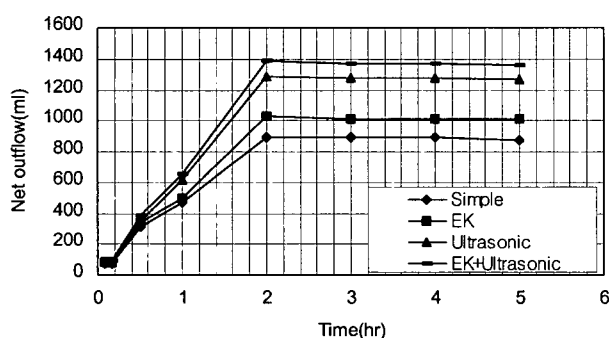


Fig. 3. The quantity of outflow with time

with elapsed time through the outlet of test chamber. These results demonstrate that the outflow rate steadily increased in the beginning stage with an increase in the operating duration up to 1 hour during experiment, and continuously constant to the end of experiment. The pore water began to flow out through the outlet of test chamber after a little while in the beginning stage of experiment on the application of hydraulic head, electrical and ultrasonic energy due to time elapse, on the development of their phenomena in soil porous media. The outflow rate after 2 hours was 890ml/hr for simple soil flushing, 1030 ml/hr for electrokinetic soil flushing, 1290 ml/hr for ultrasonic soil flushing, and 1390 ml/hr for electrokinetic & ultrasonic soil flushing. After approximately 2 hours, the flow rate stabilized like the current reached a nearly constant value.

In the experiments with 4 different operating conditions, the outflow rate was lowest in the case of simple soil flushing, second in the case of electrokinetic soil flushing, third in the case of ultrasonic soil flushing, and highest in the case of electrokinetic & ultrasonic soil flushing. The outflow rate was higher in the cases of 3 enhanced experiments (electrokinetic, ultrasonic, electrokinetic+ultrasonic flushing) compared with unenhanced experiment (simple flushing), and the outflow rate was highest in the case of electrokinetic+ultrasonic flushing among 3 enhanced experiments by the coupled effect of electrokinetic and ultrasonic process. These phenomena were caused by generation of electro osmotic flow due to electric power and turbulence flow due to ultrasonic wave.

3.2 Cumulative Outflow Through Soil Specimen

The accumulated flow volume with time is presented in Figure 4. The basic pattern of flow was relatively consistent for each experiment. The figure shows the accumulative water flow is varied and increased with time. The accumulated flow volume with time is higher for ultrasonic soil flushing, electrokinetic & ultrasonic soil flushing than for simple soil flushing and electrokinetic soil flushing. It means that the ultrasonic process

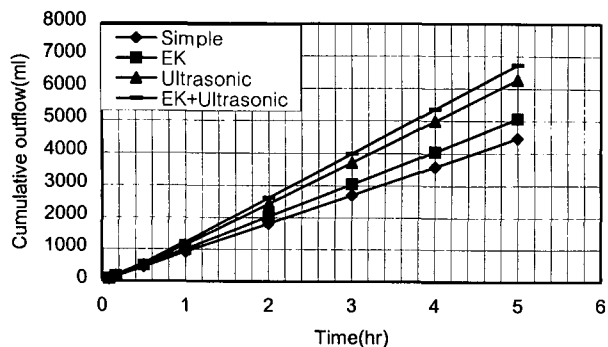


Fig. 4. Accumulated flow volume with time

plays a big role in increasing the liquid outflow due to sonication effects, but electrokinetic process plays a small role in the increase due to short test duration and sandy soil. Normally, electroosmosis by electric power is not or little developed in a short time and in sandy soil. The basic patterns of flow and current behavior were similar, but the magnitude of the flow and current differed. This is because there are many additional factors that affect electroosmosis besides the electric current.

The accumulated quantity of flow increased linearly with an increase in the operating duration. The accumulated quantity of outflow after 5 hours was 4460 ml/hr for simple soil flushing, 5060 ml/hr for electrokinetic soil flushing, 6260 ml/hr for ultrasonic soil flushing, and 6715 ml/hr for electrokinetic & ultrasonic soil flushing. The final accumulated quantity of outflow increased to 113% for electrokinetic soil flushing, 140% for ultrasonic soil flushing, 151% electrokinetic & ultrasonic soil flushing compared with that for simple soil flushing.

3.3 Permeability of Soil Specimen

The permeability of specimen relatively increased by applying the ultrasonic process. This can be attributed to the effect of ultrasonication. The calculated mean permeability is shown in Figure 5. In this Figure, we can see that the mean permeability of experimental soil is 0.00163cm/sec for simple flushing, 0.00185cm/sec for electrokinetic flushing, 0.00229cm/sec for ultrasonic flushing, 0.00245cm/sec for electrokinetic & ultrasonic flushing, respectively. The permeability is very high in the tests using the ultrasonic process compared with that

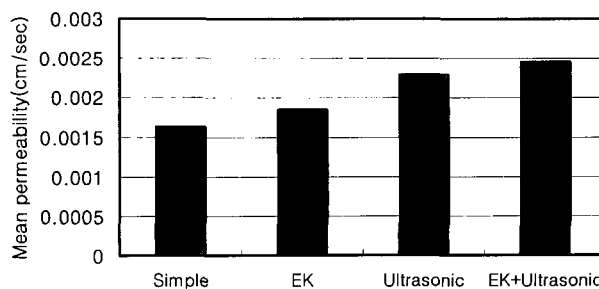


Fig. 5. Mean permeability for 4 experimental methods

in the tests not using the ultrasonic process, and the permeability is slightly high in the tests using the electrokinetic process compared with that in the tests not using the electrokinetic process. The mean permeability is highest in the case of electrokinetic & ultrasonic soil flushing due to the coupled effect of electrokinetic and ultrasonic technique.

3.4 pH of Outflow

The pH of outflow was measured by pH meter from the effluent passing the soil sample. Figure 6 shows a plot of the effluent pH with respect to time. The pH of outflow ranged from 6.6 to 7.0 for all experiments. The initial pH of soil specimen is about 6 to 7. There is no change in pH of effluent. The hydraulic gradient and ultrasonic wave did not contribute to the change of pH. And then the pH remained relatively constant and similar to initial pH condition.

The oxygen gas and hydrogen ion are generated and decreased the pH at the anode, on the other hand hydrogen gas and hydroxide ion are generated and increased pH at the cathode by electrolysis. The acid fluid adjacent anode electrode at the bottom of soil

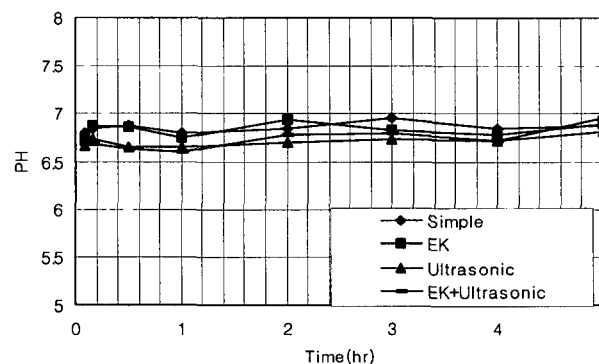


Fig. 6. pH with time

specimen flowed toward the base zone at the top of soil specimen by electroosmosis. Thus, the acid fluid meets the base fluid at the base zone, and the effluent neutralized due to mixing of the acid fluid and the base fluid.

3.5 Electric Conductivity of Outflow

Conductivity is inversely related to the resistance offered to current flow. This resistance would change due to pore size (porosity), tortuosity in the porous medium, and variations in pore fluid and double layer electrolyte concentration. Conductivity was measured by measuring device from the effluent in all experiments.

Figure 7 represents the change in conductivity of outflow with time. Since the experiments were performed under constant voltage conditions, the decrease in conductivity with time reflects an increase in the resistance offered and decrease in the current required to maintain the given voltage. In all experiments, the conductivity steadily decreased from 1076-1856(S at the start of the experiments) to 31-51(S at the end of the

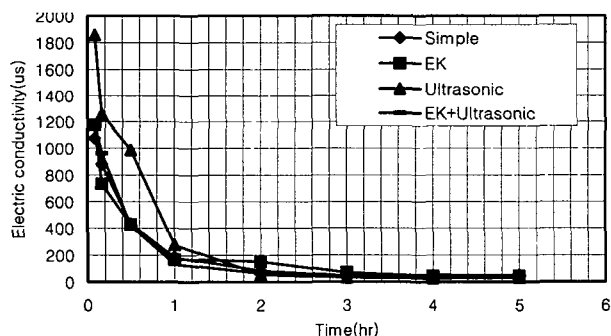


Fig. 7. Electric conductivity with time

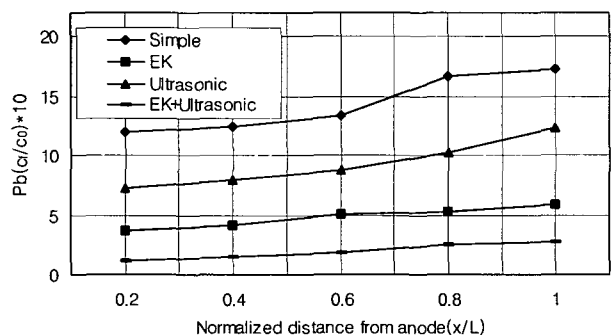


Fig. 8. Normalized concentration of Pb in soil specimen

experiments). Similar phenomena were obtained by Acar et al (1992).

3.6 Removal Effects of Pb and Ethylene Glycol in Soil Specimen

1) Removal Effects of Pb

The soil specimen was extruded and cut into 5 small sliced specimens with same length after conducting the experiment to measure water content, pH, and concentration throughout total soil specimen length. Pb concentrations of each sliced soil specimen were measured by laboratory chemical analysis facilities. The contaminant was allowed to migrate from the inlet (bottom of specimen, anode side) to outlet (top of specimen, cathode side) under the actions of advection by hydraulic gradient, electroosmosis and electromigration by electric fields and fractured migration by ultrasonic waves. From the results of these phenomena, finally the contaminant is accumulated to the cathode zone or wholly passed the cathode zone and gone outside from the soil specimen.

Figure 8 demonstrates the normalized Pb concentration (final concentration/initial concentration) profile across the soil specimen with different experimental conditions. The results show that the Pb ion migrates and is transported toward the cathode zone, and removed from the soil specimen. Thus the soil specimen is cleaned due to extraction of contaminant compared with initial condition. The Pb concentration is relatively higher at the cathode zone than at the anode zone. Overall, considering the initial concentration of Pb in the soil, 4 types of flushing techniques removed significant amounts of Pb.

The normalized Pb concentration is varied with experimental conditions. The normalized Pb concentration in soil specimen is low in the case of electrokinetic and electrokinetic & ultrasonic flushing process compared with in the case of simple and ultrasonic flushing process. The heavy metal contaminant such as Pb is easily migrated and removed by electromigration phenomena induced from electrokinetic process. The removal efficiency of heavy metal by electrokinetic technique is higher than that by ultrasonic technique.

Electrokinetic process is the most effective technique to remove heavy metal in contaminated soil. The residual concentration of Pb in soil specimen is 15% for simple soil flushing, 5% for electrokinetic soil flushing, 10% for ultrasonic soil flushing, and 2% for electrokinetic & ultrasonic soil flushing.

2) Removal Effects of Ethylene Glycol

Ethylene glycol concentration in soil specimen was measured from 5 small sliced specimens in each experiment. The ethylene glycol was allowed to migrate from the inlet to outlet under the actions of advection, electroosmosis, electromigration, and ultrasonic wave. Since ethylene glycol has a high aqueous solubility, it is expected to fully mix with water and migrate simultaneously with water flow.

Figure 9 demonstrates the normalized ethylene glycol concentration profile across the soil specimens determined at the conclusion of experiments. The results show that the ethylene glycol migrates and is transported toward the cathode zone, and removed from the soil specimen. The ethylene glycol concentration is relatively higher at the anode zone than at the cathode zone. Considering the initial concentration of ethylene glycol in the soil, significant amounts of ethylene glycol were removed for all flushing techniques.

The normalized ethylene glycol concentration in soil specimen is low in the case of ultrasonic and electrokinetic & ultrasonic flushing process compared with that in the case of simple and electrokinetic flushing process. The organic substance such as ethylene glycol easily

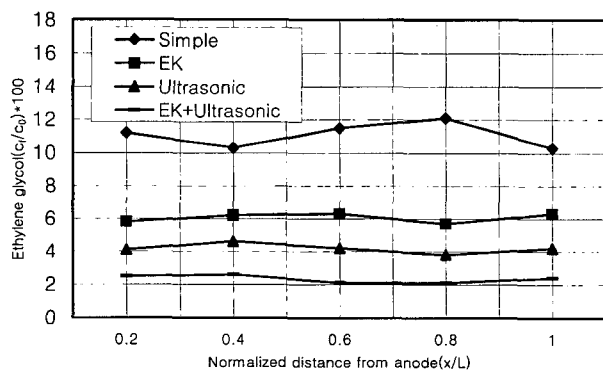


Fig. 9. Normalized concentration of ethylene glycol in soil specimen

migrates and is removed by fractured phenomena induced by ultrasonic process. The removal efficiency of organic substance by ultrasonic technique is higher than that by electrokinetic technique. Ultrasonic process is the most effective technique to remove organic substance in contaminated soil.

Reddi et al (1993) also investigated the effect of ultrasonic energy on enhancement of the permeability of soils. They observed an increase in the permeability of all experiments. They attributed the increased permeability to the removal of particles smaller than clay and colloidal size particle in the experimental specimens due to sonication. Therefore, for the test soils, the increased permeability due to sonication can be attributed primarily to particle agitation and dislodging. The residual concentration of ethylene glycol in soil specimen is 11% for simple soil flushing, 6% for electrokinetic soil flushing, 4% for ultrasonic soil flushing, and 2% for electrokinetic & ultrasonic soil flushing.

3) Comparison of Contaminant Removal Rate

The contaminant removal efficiency is calculated from the inverse of residual concentration of contaminant in soil specimen. The removal rate of Pb in soil specimen is 85% for simple soil flushing, 95% for electrokinetic soil flushing, 90% for ultrasonic soil flushing, and 98% for electrokinetic & ultrasonic soil flushing. And the removal rate of ethylene glycol in soil specimen is 89% for simple soil flushing, 94% for electrokinetic soil flushing, 96% for ultrasonic soil flushing, and 98% for electrokinetic & ultrasonic soil flushing. From these results, it can be suggested that the electrokinetic process is efficient for the removal of cation ionic matter such as heavy metal, and the ultrasonic process is efficient for the removal of nonionic organic substance such as ethylene glycol. Thus, the contaminant removal rate is highest in electrokinetic & ultrasonic soil flushing system due to the coupled effect of electrokinetic and ultrasonic technique

The removal rate is over 90% for enhancement technique such as electrokinetic and ultrasonic soil flushing process, on the other hand the removal rate is

under 90% for unenhancement technique such as simple soil flushing process. This could suggest that introduction of such enhancement techniques, addition of ultrasonic process and electrokinetic process onto simple soil flushing, could be effective for increasing pollutant removal rate from the contaminated soil.

4. Conclusions

The objective of this laboratory investigation was to evaluate the coupled effect of electrokinetic and ultrasonic technique for extraction of heavy metal and organic substance from contaminated soils. A series of tests were conducted for simple soil flushing, electrokinetic soil flushing, ultrasonic soil flushing, electrokinetic & ultrasonic soil flushing. The main conclusions drawn from the results of this investigation were as follows:

- 1) The water and contaminant in porous soil media are allowed to flow and migrate under the actions of advective flow by hydraulic gradient, electroosmosis and electromigration by electric power, and fractured flow by ultrasonic waves.
- 2) Outflow rate, accumulated outflow, permeability, contaminant removal rate is lowest in the case of simple soil flushing process and highest in the case of electrokinetic & ultrasonic soil flushing process.
- 3) The accumulated quantity of outflow after 5 hours is 4460 ml/hr for simple soil flushing, 5060 ml/hr for electrokinetic soil flushing, 6260 ml/hr for ultrasonic soil flushing, and 6715 ml/hr for electrokinetic & ultrasonic soil flushing.
- 4) The contaminant removal rate is over 90% for enhancement technique such as electrokinetic and ultrasonic soil flushing process, on the other hand the removal rate is under 90% for unenhancement technique such as simple soil flushing process.
- 5) The ultrasonic technique can enhance the removal of organic substance from contaminated soils, the electrokinetic technique can enhance the removal of heavy metal from contaminated soils.
- 6) The new remedial technique combined with electrokinetic and ultrasonic processes can be effectively

applied for the removal of contaminants in the ground contaminated with various organic and inorganic pollutants.

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