

Effect of a surfactant on enhancing efficiency of the electrokinetic method in removing anthracene from a clay soil

A. R. Estabragh

Assistant Professor. Faculty of Soil and Water Engineering, University of Tehran, PO BOX 4411 Karaj 31587-77871, Iran

Tel: +98 261 2241119

Fax: +98 261 2226181

Email: raeesi@ut.ac.ir

M. Lahoori

Postgraduate Student, Faculty of Soil and Water Engineering, University of Tehran, PO BOX 4411 Karaj 31587-77871, Iran

Tel: +98 261 2241119

Fax: +98 261 2226181

Email: lahoori.m@ut.ac.ir

A. A. Javadi

Professor, Computational Geomechanics Group, Department of Engineering, University of Exeter, Devon, EX4 4QF, UK

Tel: +44 1392 723640

Fax: +44 1392 217965

Email: a.a.javadi@exeter.ac.uk

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Abstract

This paper presents a study of the removal of anthracene from a clay soil using a surfactant and investigates the effects of electric field on the anthracene removal efficiency. A non-ionic surfactant, Tween 80, was fed to the anode reservoir and the tests were conducted under voltage gradient of 1.5 v/cm and different times (3, 7 and 10 days). A reference test was also conducted with distilled water in anode and cathode reservoirs under the same voltage with duration of 7 days. During each test the pH, EC (electrical conductivity) and volume of outflow discharge were measured. At the end of each test, a number of soil samples were extracted from different distances of anode and removal of anthracene from them was measured by using HPLC (High Performance Liquid Chromatography) apparatus. The results show that by using Tween 80 the removal of anthracene is increased. Also, by increasing the duration of test until 7 days the removal is increased. However, after 7 days the rate of increase in the removal of anthracene is not considerable.

Key words: anthracene, clay soil, electrokinetic remediation, surfactant

Introduction

Contaminants are substances that are on, in or under the ground, and which have potential to cause harm, and/or pollution to specific receptors ([1]). Contaminants can be broadly categorized into organic and inorganic contaminants. Organic contaminants are carbon based and could be natural or anthropogenic in existence. Most organic contaminations are associated with accidental spills and leaks, originating from equipment cleaning, maintenance, storage tanks, residues from used containers and outdated material ([2]- [3]). Transport and fate of organic contaminants are important, and models are used to assess migration plumes based on the target goals of the developer. Organic contaminant migrations are due to advection (by fluid flow through soil) and diffusion, but other forms of transport e.g. infiltration may also contribute to migration ([4]).

Polycyclic aromatic hydrocarbons (PAHs) are organic contaminants that are formed of carbon and hydrogen that are composed of multiple aromatic rings. They are nonpolar and insoluble in water persistent in soil and many of them are recognized as carcinogenic ([5]- [6]). The source of them usually is oil production and petrochemical complex industry. Anthracene is a chemical matter from PAHs group that is commonly found in soil through industrial activities and causes contamination of soil.

The electrokinetics method is of the techniques for remediation of contaminated soil. It comprises of three major processes namely electrophoresis, electroosmosis and electrolysis. Electrophoresis describes the transport of large colloidal or other charged material through soil mass. Electroosmosis is the main mechanism of water flow through fine-grained porous media under the influence of an electric field. Electro-migration

explains the movement of ionic species in pore fluid toward oppositely charged electrodes ([7]). Electrokinetics is a low cost process for remediation of contaminated soil but suffers from several disadvantages such as generation of gases and acid at the anode that reduce the efficiency of the process ([8]). Researchers such as ([9]-[11]) reported that the electrokinetics technique can successfully remove heavy metals and organic matters from contaminated soil.

Today the adsorption method has attracted the attention of researchers for removing metal ions from aqueous environment that may be polluted by the release of metal ions from chemical industry. This method is easy to apply and low in cost ([12]). Nano composites are extensively used as adsorbent materials due to their characteristics such as small size and high specific surface. Zheng et al ([13]) reported that the adsorption potential of carbon nanotubes (CNTs) is limited because of the lack of functional groups. They enhanced the behaviour of amine functionalized CNTs and their potential absorption was increased. Zeng et al. ([14]) indicated that both nanomaterials and polymers have advantages in adsorption of heavy metal ions. Qiao et al. ([15]) reported the effectiveness of molybdenum disulfide (MoS_2) in removing the methylene blue from aqueous solution. A composite of MoS_2 and polymerization of levodopa (DOPA) was made by Huang et al. ([16]) which had a higher adsorption potential than MoS_2 . Huang et al. ([17]) found that functionalization of SiO_2 particles could increase the adsorption potential of the composite. Zhang et al. ([18]) showed that the Mussel-inspired surface chemistry can be used for functional materials for removing organic dyes, heavy metal ions and other pollutants. Zeng et al. ([19]) prepared a two-dimensional nanomaterial by coating MoS_2 with polydopamine (PDA) and nanoparticles of Ag. They found that its

adsorption potential is more than MoS₂. Huang et al. ([20]) modified the MgAl-layered double hydroxide with polyethylene polyamine (PP) and tannic acid (TA) which increased the potential of adsorption in heavy metals. Liu et al. ([21]) reported that graphene oxide (GO) and its composites with organic and inorganic materials is one of the most popular carbon nanomaterials for environmental application.

The removal of PAHs from contaminated soil is rather complex because of their low solubility and their tendency to remain attached to soil particles and organic matters in soil. Several methods have been proposed for remediation of soil contaminated with this kind of organic contaminant. Electrokinetic technique is one of the methods that has been suggested and examined for the removal of PAHs from soil ([22]). Due to the low water solubility of most organic contaminants and the neutrality of their molecules, it is not possible to remove hydrophobic organics from the soil by electrokinetic technique ([23]). In order to overcome these limitations, the degree of solubility of organics can be increased by the use of surfactants as flushing solution in anode and/or cathode ([24]). Surfactants are surface active agents that are used to reduce interfacial tension and increase solubility of non-aqueous phase liquids ([25]). The process relies on the use of surfactants to increase the solubility of hydrophobic organic pollutants and hence increase the removal efficiency on application of the washing fluid. Surfactants are typically classified according to the nature of their head group, as cationic, anionic and nonionic. Surfactants increase the efficiency of electrokinetics in remediation of contaminated soils ([26]- [28]). Park et al. ([29]) reported that removal of phenanthrene is increased up to 70% by using APG and Brij 30 surfactants. Yang et al. ([30]) used APG and Calfax 16L-30 for removal of phenanthrene from contaminated soil. They reported

that the removal of phenanthrene is increased as high as 98% by using APG and 25% by Calfax 16L-30

A review of the literature shows that investigation on the removal of anthracene from a contaminated soil is relatively rare. Therefore, the aim of this work is to investigate the applicability of electrokinetics in soil flushing with ionic surfactant Tween 80 to remove anthracene from a clay contaminated soil. The tests were conducted with voltage gradient of 1.5 v/cm at times of 3, 7 and 10 days. The results were analyzed and compared with a reference test and with each other.

Materials and Methods

Materials

Soil

The soil used in this experimental work was a clay. It was composed of 23% sand, 52% silt and 25% clay. It had a liquid limit of 48.0% and plasticity index of 22.0 %. The optimum water content and maximum dry unit weight in standard compaction test were 17.90% and 17.75 kN/m³ respectively. The specific gravity of solids (G_s) was 2.71. According to the Unified Soil Classification System (USCS), the soil can be classified as clay with low plasticity (CL). The chemical properties of the soil are summarized in Table 1. The results of XRD (X-ray diffraction) tests conducted on samples of this soil show that the minerals of the soil include quartz, calcite, clay mineral, feldspar (Na, Ca) and feldspar (K). The clay minerals of the clay are illite, chlorite and montmorillonite.

Anthracene

Anthracene is a chemical substance from PAHs (Polycyclic aromatic hydrocarbons) group with chemical formula $C_{14}H_{10}$ and molecular weight and density equal to 178.23

g/mol and 0.7407 g/mL respectively. Its chemical structure is composed of three fused benzene rings. The color of it can vary in appearance from a colorless to pale yellow crystal like solid. Its water solubility at 25°C is 0.044g/L and its boiling and melting points are 340°C and 218°C respectively. The major application of anthracene is in production of dyes, plastics and pesticides. PAHs components are relatively resistant to biodegradation and can remain in the environment for a long time. Researchers such as ([31]- [36]) indicated that existence of anthracene in soils may exhibit a toxic activity towards different biological elements of the environment such as plants, microorganisms and invertebrates.

Surfactant

Surfactants can increase the solubility of organic contaminants in the aqueous phase by solubilizing hydrophobic organic contaminants into micelles. However, the adsorption of surfactant onto soil or soil organic substances can decrease the efficiency of remediation. Non-ionic surfactants are appropriate for the electrokinetic process because their neutral charge does not affect electroosmotic flow, they are biodegradable and in general their toxicity is low ([37]). Tween 80 was selected as non-ionic surfactant in this work with chemical formula of $C_{18}H_{37}-C_6H_9O_5-(OC_2H_4)_{20}OH$. The average molecular weight of Tween 80 is 1309 and HLB (Hydrophilic- Lipaphitic- Balance) is equal to 13.4 ([38]). The main factors that should be considered in selecting surfactants include biodegradability, low toxicity, solubility at ground water temperature, low adsorption to soil and effectiveness at concentrations lower than 3% ([39]). Anionic and nonionic surfactants are less likely to be absorbed to the soil ([40]). Tween 80 has the above mentioned characteristics and hence was selected in this work.

Electrokinetic apparatus

The electrokinetic test setup used in this work was similar to those used by researchers such as Mohamedelhassan and Shang ([41]) and Ritirong et al. ([42]) for simulating contaminant transport. The schematic plan of the apparatus is shown in Fig.1. This apparatus consists of a Plexiglas with 1 cm thickness as the main cell, a loading frame and a D.C. (Direct Current) power supply. The main cell measured 30 cm in length, 10 cm in width and 25 cm in height. Two reservoirs as anode and cathode were added on the two sides of main cell and they were connected to the main cell through perforated Plexiglas sheets. These reservoirs can be filled by desired fluid and the total hydraulic head in them can be controlled by adjusting two identical standing tubes through valves. Two electrodes (EVDS) made of copper foil covered with conductive polymer were vertically placed at a distance of 5 cm from the soil in the main cell. The front and back of the soil samples were covered by two pieces of saturated geotextile in order to inhibit the migration of colloidal particles of soil into the electrode components as used by Jeon et al. ([43]) and Kim et al. ([44]). A number of voltage probes were installed at the bottom of the main cell at the distances of 2, 5, 8 and 11 cm from anode to measure the electrical potential variation intervals during the tests. A loading system was made for applying load to the soil in the main cell. It was made of a plate that was placed at the top of the sample and connected to another plate at the bottom of it by a bar. The bar was covered with a foam material to isolate it against electrical current. The load was applied through the bottom plate on the sample. A dial gauge was mounted on the top plate for measuring the vertical deformation due to the settlement of soil. The power supply for D.C. current

consisted of a generator to produce various ranges of voltage and connected to the EVD through a special connection.

Sample preparation and testing procedure

For preparing samples of soil contaminated with anthracene, the amount of 500 mg anthracene per kg of soil was considered because according to USEPA (2000), the typical PAH concentration is 500mg/kg near source zones of contaminated sites. Saichek and Reddy ([26]) and Delgado-Balbuers et al. ([45]) used phenanthrene with concentration of 500 mg/kg in their research works. The solubility of anthracene in water is very low but it is completely dissolved in acetone ([46]). Therefore, the mixture of acetone anthracene (10g anthracene per one liter acetone) was subsequently mixed as spray with the measured amount of soil. The soil-acetone-anthracene mixture was placed in a tray for one week until the acetone completely evaporated and the contaminated soil was dry. After that the contaminated dry soil was mixed with a measured amount of distilled water and mixed manually until the water content of it reached more than liquid limit (i.e. in saturation condition). The moist soil was kept in a seal cover for 48 hours for uniform distribution of water. The prepared soil was then poured into the main cell of apparatus in several layers and each layer was tamped in the cell so that the entrapped air could go out and the space between particles reached to minimum. After the soil was placed in the main cell, the apparatus was assembled and other accessories were connected to it. The anode and cathode reservoirs were then filled with desired solution so that the level of fluid in the reservoirs was the same as the level of soil in the main cell. A total of 4 tests were conducted on the contaminated soil samples with different times. Test 1 was considered as the reference test in which the anode and cathode reservoirs were filled

with distilled water. For the rest of the tests (2, 3 and 4) the anode was filled with solution of 3% Tween 80. A constant voltage gradient of 1.5 V/cm was applied for durations of 3, 7 and 10 days for all tests. During the tests pH, EC, and discharge volume of fluid outflow from the cathode were measured periodically. At the end of test soil samples were extracted at the middle section of the main cell at certain distances from anode (7, 14, 21 and 28 cm). For chemical analysis of the extracted samples, after each test the procedure of sample preparation was followed according to the EPA 3540 standard. In this method the extracted soil samples were dried. Then 2 g of each sample was selected and mixed by 5 cm³ of hexan and acetone mixture with ratio of 2:1. It was shaken for 2 minutes for uniform mixing; it was then put in a centrifuge with 3500 rpm for about 5 minutes for settlement of particles and separation of the liquid-solid phases. The liquid above the sample was passed through a filter with mesh size of 0.45 um. The extracted liquid was then injected to the HPLC apparatus. The HPLC was calibrated before performing the chemical analyses. A standard pure compound was injected to the apparatus and the peak in the chromatogram was assigned based on the retention time of the standard. The peak areas or heights were used to determine the concentration of released contamination from the sample. Comparing the response of the unknown concentration to that of the known (standard) concentration was used to determine the amount of concentration in the released leaching liquid.

Results and discussion

Enhanced electrokinetic remediation tests were conducted on a contaminated clay soil to investigate the efficiency of removal of anthracene from the soil. Fig.2 shows the variations of pH with time for the anode and cathode reservoirs. As shown in this figure

there is a rapid drop in pH from initial values at the anode reservoir for all tests, indicating that the anode reservoir gets acidic. The values of pH in the cathode are increased so that at the end of the tests the final value of pH reaches to 12 or more. The pH increases rapidly until about 50 hours after which, the rate becomes slower until it reaches to about 12. This value of pH shows that the liquid of the cathode reservoir is alkalian. Therefore, acidic and alkaline fronts are formed and penetrate to soil sample which change the properties of the soil such as pH. These changes in the value of pH are resulted from electrolysis of water that leads to the formation of H^+ ions and oxygen at the anode and OH^- ions and hydrogen at the cathode. The immediate results are change in the initial value of pH of the pore solution so that acidity increases with time at anode and alkalinity increases at the cathode ([47]). An acid front moves across the soil sample from the anode toward the cathode and a base front moves from cathode to the anode. Thses movements cause changes in the behaviour of the soil ([48]).

The variations of EC at anode and cathode for different tests are shown in Fig.3. As shown in this figure, the values of EC increase with time in both anode and cathode reservoirs but at a specific time, the value of EC at anode is less than cathode for all tests. The results of these tests provide information about the concentrations of ions in the reservoirs. The fabric of soil is composed of clay minerals that may dissolve due to the changes in composition of pore solution. This leads to a significant increase in the electric conductivity in the cathode reservoir, indicating the accumulation of dissolved ions as shown in Fig.3.

The cumulative outflow of fluid from the anode reservoir for different tests is shown in Fig.4. For the test with anode reservoir filled with distilled water, the final volume of

discharge fluid is 575 cm³ but it changes to 1380 cm³ for the test in which the anode reservoir was filled with solution of 3% Tween 80 for duration of 3 days. Adding Tween 80 caused the outflow of fluid from the cathode reservoir to increase about 2.4 times compared with the case of distilled water as the anode reservoir fluid. Comparing the results of the tests with the same anode reservoir fluid (solution of 3% Tween 80) but with different test durations shows that by increasing the duration of test, the amount of fluid outflow from the sample increases. For example, for duration of 3 days the volume of outflow is 1380 cm³ but it changes to 1640 cm³ and 1720 cm³ for durations of 7 and 10 days respectively.

Fig.5 shows the variations of k_e with time for different tests. k_e is the electro-osmotic permeability. It governs the water flow in a soil mass under an electrical gradient in a similar way as the hydraulic conductivity governs the flow in soil under hydraulic gradient. As shown in this figure there is a sharp drop in the values of k_e at early stages of all tests, but then the rate of reduction decreases significantly. For the test with distilled water as the anode reservoir fluid, the final value of k_e is 2.11E-06 cm²/v.sec while it is 8.68E-06 cm²/v.sec for the test with Tween 80 solution at three days. The results show that distilled water is more effective than the Tween 80 solution in decreasing k_e . In addition, increasing the duration of test is effective in reducing k_e . At a constant voltage gradient and time, the amount of discharge fluid from cathode is dependent on the value of k_e . For the test where the anode reservoir is filled with distilled water the values of discharge fluid and k_e are less than the tests with anode reservoir filled with solution of 3% Tween 80.

Fig.6 shows the results of remediation of soil at different distances (7, 14, 21 and 28 cm) from the anode reservoir. As seen in this figure, the percentage of remediation for all tests is reduced by increasing the distance from anode. The maximum remediation is near the anode reservoir (7 cm from anode) and the minimum is near the cathode reservoir (28 cm from anode). Comparison of the results shows that near the anode reservoir the percent of remediation for the distilled water as the anode reservoir fluid is 2.5% but for the test with 3% solution of Tween 80 and duration of 3 days it is changed to 30.72%. It is seen that the Tween 80 solution (as anode fluid) increases the percent of remediation about 12 times more compared with distilled water. It is also seen from this figure that by increasing the duration of test from 3 days to 7 or 10 days the percent of remediation is increased and this increase is more considerable at points 14 and 21 cm from anode. It is resulted that the electrokinetic remediation of hydrophobic organics is not possible by using water as the processing fluid as also indicated by [23]. The low solubility of anthracene and its tendency to remain attached to soil particles make the unenhanced electroremediation ineffective ([49]). The use of Tween 80 modifies the surface tension of the aqueous solution and favors the extractability and solubility of the hydrophobic organics which have been entered the aqueous phase and stabilized in the micelles that were formed by surfactant ([26]). Tween 80 is a non-ionic surfactant that penetrates the soil by electroosmosis from the anode and removes the contaminant. Therefore, the removal of hydrophobic organic contaminants from soil with an enhanced electrokinetic treatment depends on the ability of surfactant to extract or desorb the contaminant from the soil particle surfaces and maintain the contaminant in solution forming micelles. Those micelles are then usually removed from the soil by electro-osmosis. The results

indicated the removal of contaminants from the soil is dependent on the volume of fluid discharge from the sample and solubility of contaminant with surfactant. As shown in Fig.3, with Tween 80 solution the volume of fluid discharge is increased in comparison with water. Therefore, Tween 80 can be a favourable candidate for removal of anthracene from soils because of the large cumulative volume of fluid during electrokinetic remediation ([23]). The difference in the volume of fluid discharge for the cases of distilled water or Tween 80 as anode fluid can be explained through the interaction of the Tween 80 with soil particles. This interaction leads to the modification of the zeta potential that is an important factor for inflow of fluid from cathode. Kaya and Yukselen ([50]) studied the effect of ionic and non-ionic surfactants on the zeta potential of clay particles. Their results showed that the surfactants produce negative zeta potential and the amount of fluid discharge is increased because the zeta potential and volume of fluid discharge are directly related based on the following equation:

$$q_{eo} = nA \frac{D\zeta}{\eta} E_x \quad (1)$$

where n , A , D , ζ , η and E_x are porosity, area perpendicular to the direction of flow, dielectric constant of the fluid, viscosity of the fluid and the gradient of voltage in the direction of the flow (x in this case).

Therefore, as reported by Kaya and Yukselen ([50]) an increment in the value of zeta potential of soil leads to increase in their fluid discharge out of the soil and the percentage of removal of contaminants is higher in comparison with distilled water as anode reservoir fluid.

Conclusion

The removal of anthracene from a clay soil was studied through experimental tests by using distilled water or solution of Tween 80 as anode reservoir fluid. The tests were performed under constant gradient of voltage and different times. The results showed that distilled water is not effective in removal of anthracene from contaminated soil. The addition of the Tween 80 solution to the anode reservoir increases the effluent of fluid from cathode. By using Tween 80 the rate of removal of anthracene is increased and this increase is dependent on the duration of test. The percent of removal is decreased with increasing the distance from anode. However, using surfactant improves the contaminant (anthracene) removal efficiency.

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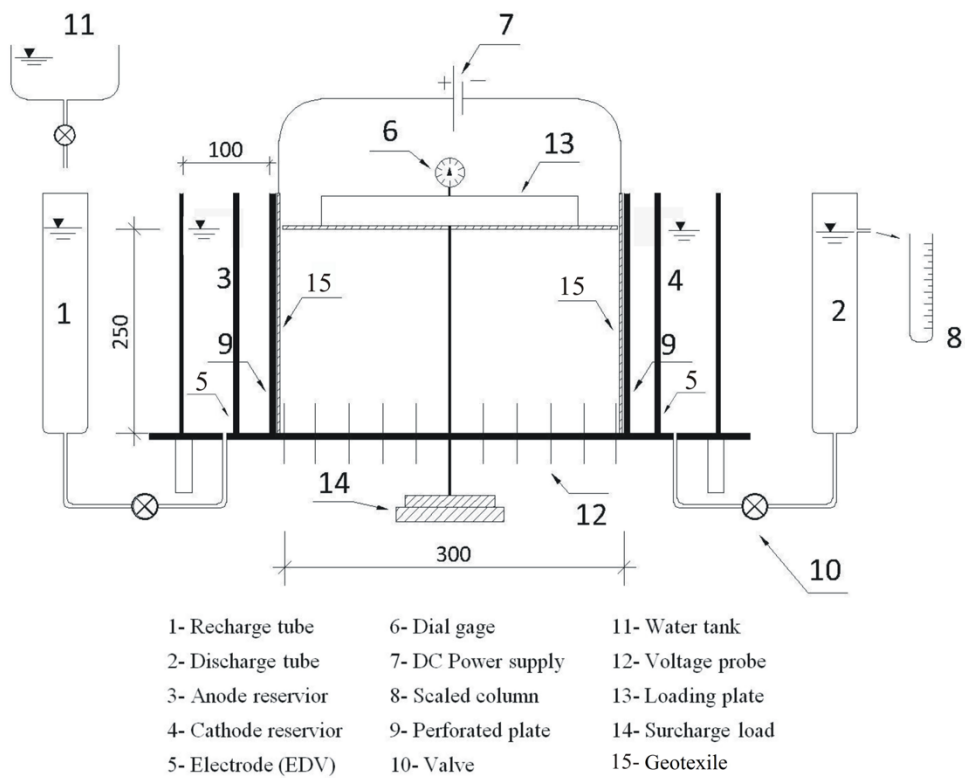


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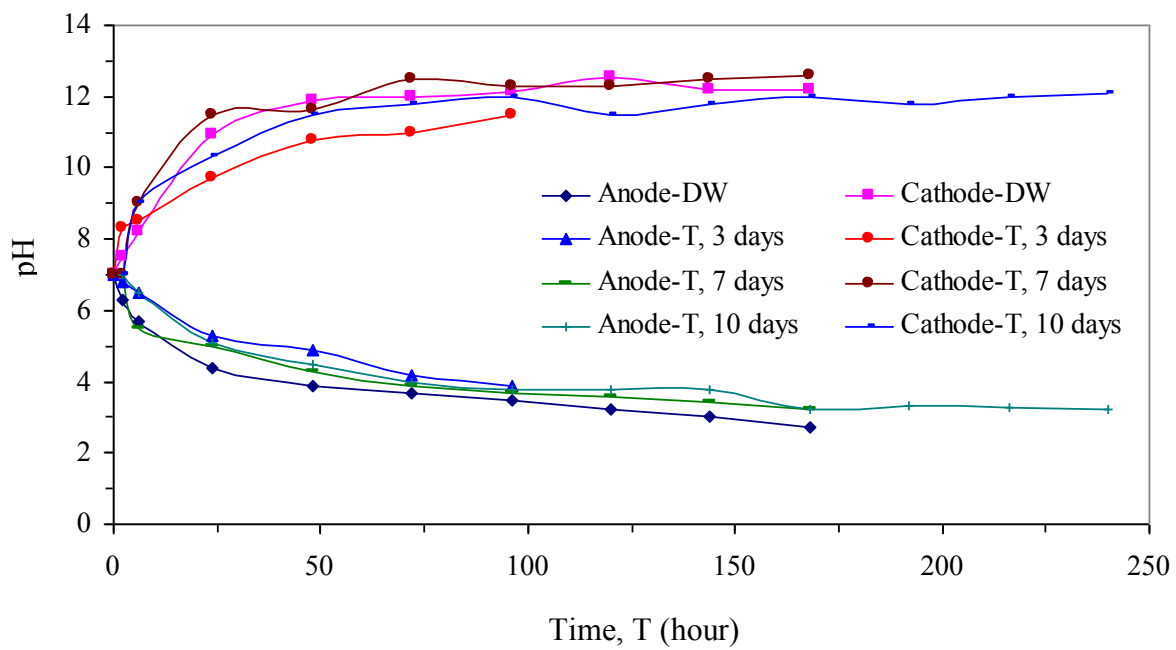


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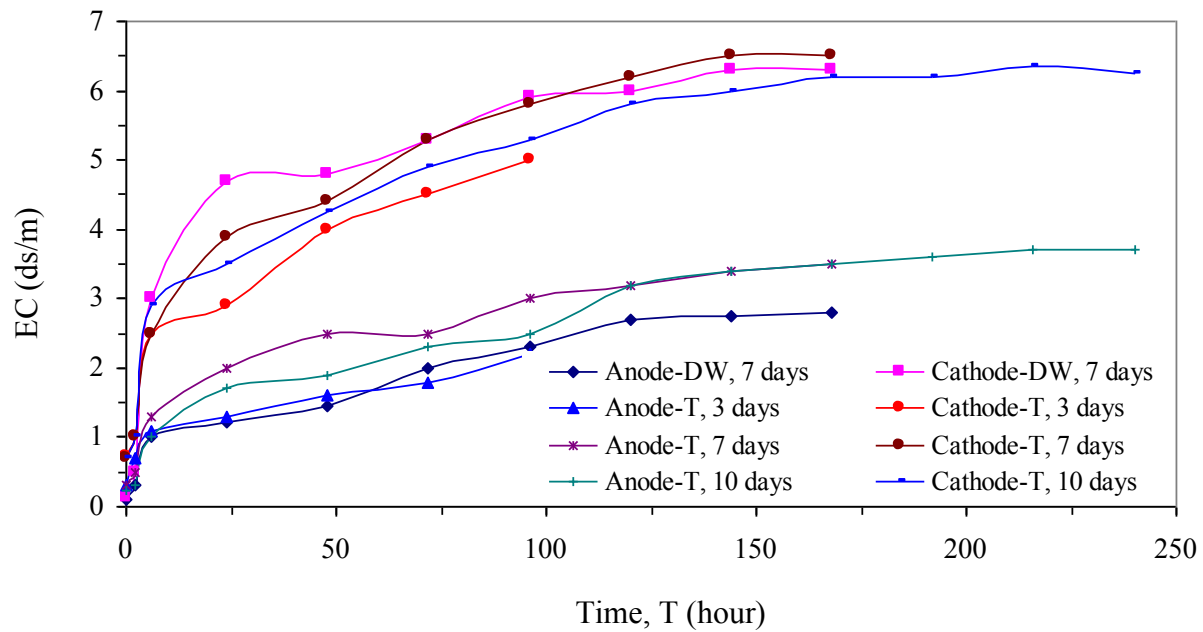


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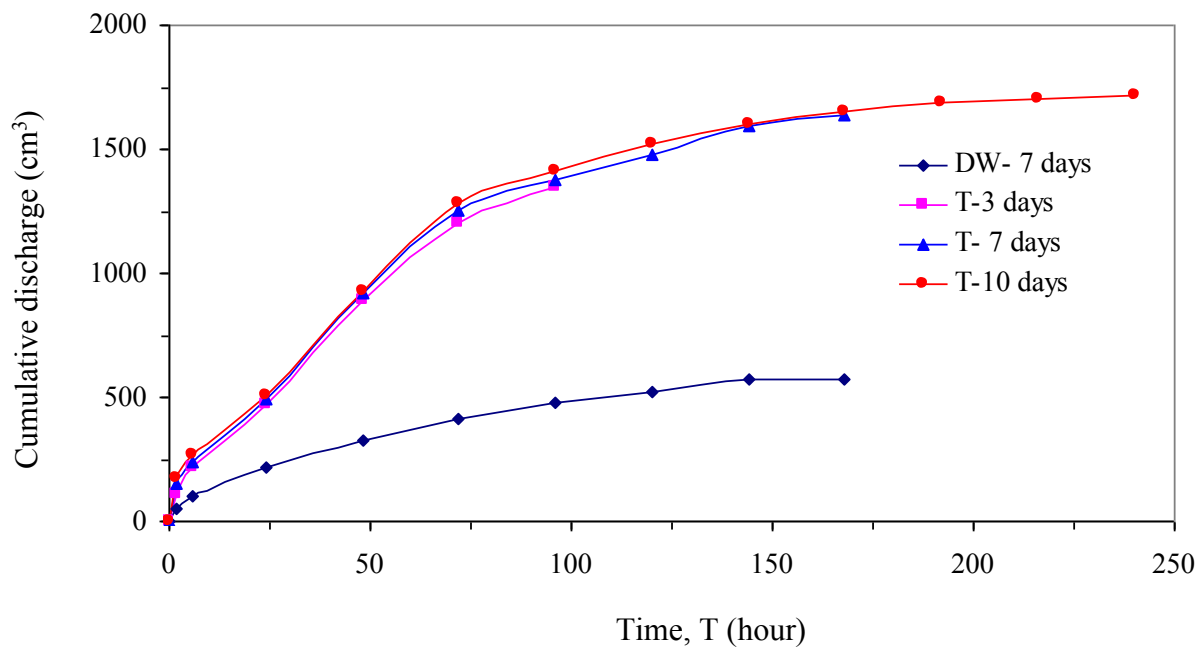


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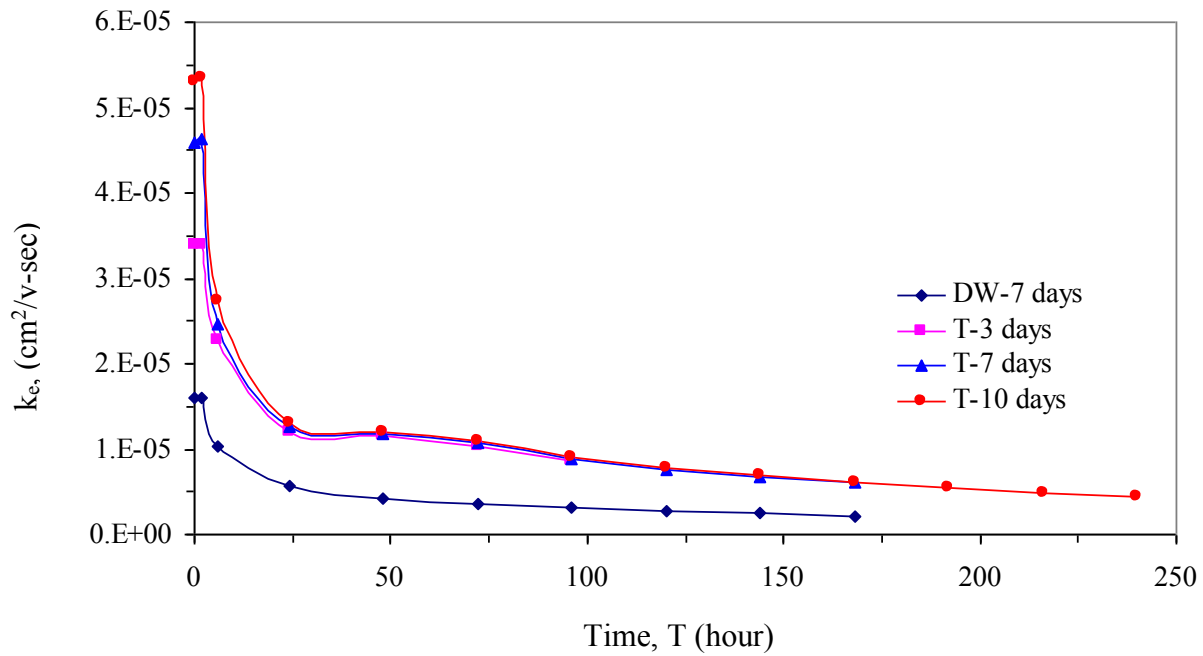


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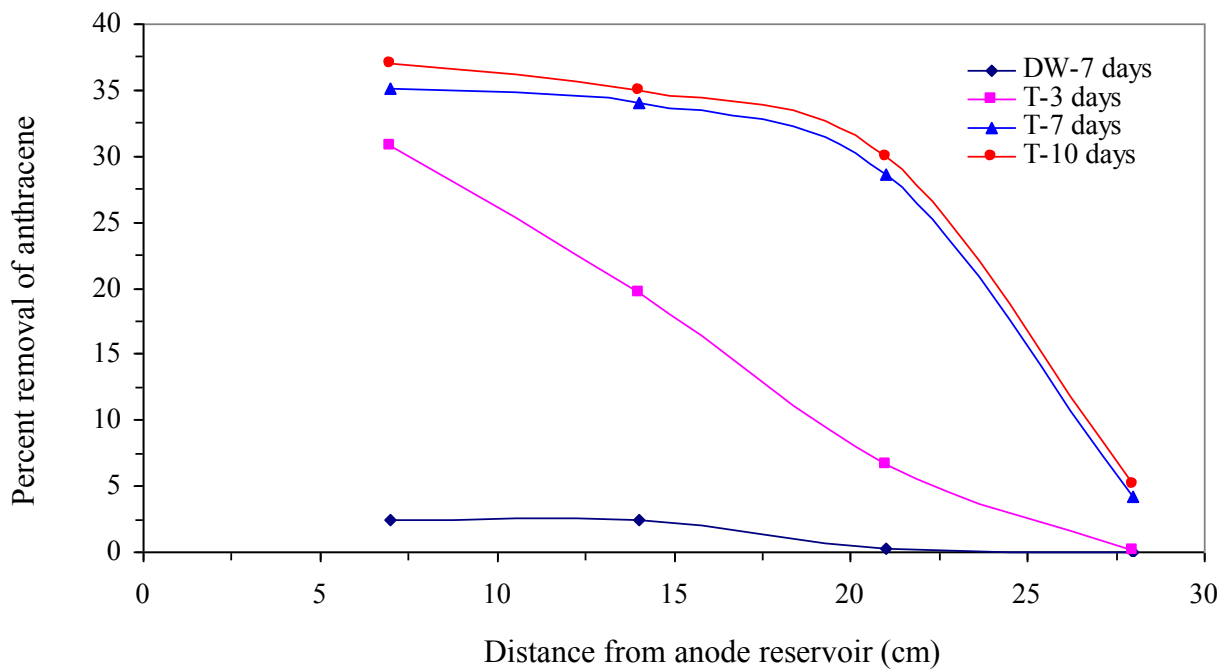


Fig.6. Percent removal of anthracene from various points of soil sample for different tests

Table 1. Chemical composition of soil

Chemical component	Amount	Chemical component	Amount
pH	7.8	Mg ²⁺ (meq/L)	11.25
EC ^a (dS/m)	13.2	Cl ⁻ (meq/L)	62.5
Na ⁺ (meq/L)	108.69	CO ₃ ²⁻ (meq/L)	0.6
K ⁺ (meq/L)	0.20	HCO ₃ ⁻ (meq/L)	5.0
Ca ²⁺ (meq/L)	35.0	SO ₄ ²⁻ (meq/L)	72.91
Ca CO ₃ (%)	10.2	O.C. ^b (%)	0.11
C.E.C. ^c (meq/100g)	8.42		

a- Electric Conductivity

b- Organic Content

c- Cation Exchange Capacity