Iranian (Iranica) Journal of Energy & Environment
Journal Homepage: www.ijee.net
IJEE an official peer review journal of Babol Noshirvani University of Technology, ISSN:2079-2115

Analysis of Dense Non-aqueous Phase Liquid Contaminants Effects on Soil Permeability by Response Surface Methodology

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P A P E R I N F O

Paper history:
Received 11 January 2022
Accepted in revised form 10 March 2022

Keywords:
Naphthalene
Permeability
Phenol
Response surface methodology
Soil contamination

A B S T R A C T

Soil contamination is considered a controversial issue in most countries. Nowadays, it is important to clearly understand how pollutants influence the soil from different sources. Today, hydrocarbons are one of the most important sources of soil contaminants, which is considered as a fundamental issue at the global level. The current study aims to analyze and model the effect of simultaneous parameters (time and concentration) of phenols and naphthalene with different percentages (10, 15, 20 and 25%) together with the amount of bentonite in fine-grained sandy soil. The designed experiments made use of response surface methodology (RSM) and Design-Expert software to carry out a computer-based simulation. According to the proposed model, the amount of bentonite is most affected by the permeability of the soil. The obtained results also showed that the permeability significantly decreases in the light of increasing the percentage of phenol and naphthalene coupled with the amount of bentonite and the age of contamination. On average, an 80% reduction of permeability was observed in contaminated soil, which was found in the soil contaminated with naphthalene. According to the results of the synergistic effects of time, the considerable impacts of both the percentage of hydrocarbon pollutants and the amount of bentonite on the reduction of permeability are quite evident.

doi: *10.5829/ijee.2022.13.02.05*

INTRODUCTION

Soil contamination is among the most crucial environmental issues in the world. A major environmental problem is the hydrocarbon contaminations caused by various human and industrial activities [1, 2]. Oil pollutants and their derivatives are among dangerous sources of environmental contamination, also the resulting products are highly toxic [3]. Soil contamination can be occurred by the presence of chemicals or other changes in the natural environment [4].

Generally speaking, contamination is the form of chemical compounds whose short or long-term consequences take their toll on the human body and environment. The soil pollutants are divided into two types of organics and minerals, holding different and complex functions in the porous media [5]. Polycyclic Aromatic Hydrocarbons (PAHs), which show a high degree of resistance to degradability and are also placed in the category of persistent organic pollutants, are considered to be priority pollutants by the Agency for Toxic Substances and Disease Registry (ASTDR) [6].

Non-aqueous Phase Liquids (NAPLs), which are classified as an important hydrocarbon contaminant and also widely observed in the soil environment, penetrate the soil and create a permanent contamination resource for both soil and water [7, 8]. According to their density, these contaminants are categorized as either dense non-aqueous phase liquids (DNAPLs) or light non-aqueous phase liquids (LNAPLs). NAPLs originate from different factors in the environment, especially leakage from reservoirs and transmission lines, waste caused by production and refining of crude oil, excavation and

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Please cite this article as: H. Tamadon Ghareghie, M. Yazdi, D. Yousefi Kebria, H. Aminirad, 2022. Analysis of Dense Non-aqueous Phase Liquid Contaminants Effects on Soil Permeability by Response Surface Methodology, Iranian (Iranica) Journal of Energy and Environment, 13(2), pp. 141-150. Doi: 10.5829/ijee.2022.13.02.05

extraction sites [9–11]. Hydrocarbon contamination depending on the permeability of soil and pollutant viscosity can result in changes in soil behavior [7, 12, 13].

Phenol and naphthalene are the most popular hydrocarbon, and are also known as DNAPLs [14]. Naphthalene ($C_{10}H_8$) is an aromatic hydrocarbon as well as the main ingredient of tar, which is a solution of white escape, is soluble in toluene, alcohol, benzene, and ether. Naphthalene is one of the prioritized aromatic pollutants classified by the United States Environmental Protection Agency as mutagenic and carcinogenic [15, 16]. Moreover, Phenol is among the most famous pollutants of aromatic hydrocarbons with molecular structure of aromatic as C_6H_5OH. This organic substance possesses high solubility in petroleum, glycerin and alcohol, and very small amount is dissolved in water [17].

The effect of hydrocarbon contamination on soil properties has always been discussed by researchers. Fine-grained soils are more susceptible to contamination than coarse-grained ones [5]. In the geotechnical investigation of sandy and clay oil-contaminated soils the reduction of permeability, strength, maximum dry density, optimum moisture content and Atterberg limits are observed [18]. Hanaei et al. [19] studied different percentages of hydrocarbon pollutants (0, 5, 10 and 15) on two types of sandy soils. According to the results, the permeability of contaminated soil has decreased. Also, the maximum dry density increased, and the optimum moisture content is reduced in contaminated specimens.

Direct cutting experiments also show an increase in cohesion and a reduction in internal friction angle after contamination exposure [19]. The results of this study are in accordance with Nasehi et al. [20] and Al-Aghbari et al. [21], representing that its internal friction angle experiences a decrease, and both cohesion and Atterberg limits increase due to increasing the percentage of hydrocarbons, maximum dry density and optimum moisture percentage. Aghajani et al. [22] investigated the effect of crude oil spills on the geotechnical properties of silty sandy soil using Taguchi method. The results showed that by raising the leak time, shear strength parameters and permeability significantly decreased. Also, with increasing the porosity of soil samples, the shear parameters and permeability increased. The content of the water brings about a reduction in permeability and improvement in shear parameters [22].

One of the methods of remediation and clearing the soil is the use of bentonite. Bentonite is a fine-grained clay that is often produced from the formation of volcanic ash form and mainly contains the mineral smectite and often montmorillonite [23, 24]. The main feature of bentonite is the absorption of water; the high inflation feature of bentonite is significantly higher because it has a double penetration layer. Bentonite is employed for high adsorption capacity, low hydraulic conductivity and low cost as a suitable adsorbent in the refining of contaminated soils [25, 26]. It has been proven that the addition of bentonite to contaminated soils leads to contamination removal [27]. Most of the previous researches focus only on studying the effect of the content and quality ratios in the behavior of the sand-bentonite mix behavior. The results of the previous researches in the study of physical and mechanical properties of sand with different percentages of bentonite show that compounds containing 20 to 30 % bentonite and 70-80 % of sand are classified to be an optimum composition as a fine-grained soil [28]. Due to the researches done in the previous studies, which have been studied extensively on the geotechnical behavior of hydrocarbons pollutants, it can be observed that the modeling of effective parameters and the simultaneous effect of parameters in soil contaminated with dense non-aqueous phase liquids hydrocarbon has not been investigated. In this study, by making use of experimental results, the statistical analysis of the effects of phenol and naphthalene contaminants as DNAPLs on the soil permeability is investigated in the light of Response Surface Method (RSM). Also, the interaction and simultaneous effect of pollutants, bentonite, age of contamination and soil permeability are analyzed.

MATERIAL AND METHODS

The experiments were performed on a fine-grained sand obtained from the southern coasts of the Caspian Sea (Babolsar, Mazandaran, Iran). The gradation tests were conducted on the sand according to the ASTM D422 [29]. The standard ASTM D854 [30] was used to determine the density of solid soil particles. The density of solid soil particles was obtained by 2.75. Based on the results of granulation experiments and the unified soil classification system, the used soil and bentonite were classified as SP (poorly-graded sand) and CH (clay of high plasticity). Figure 1 shows the sand granulation curve and Table 1 shows the physical characteristics of the soils used.

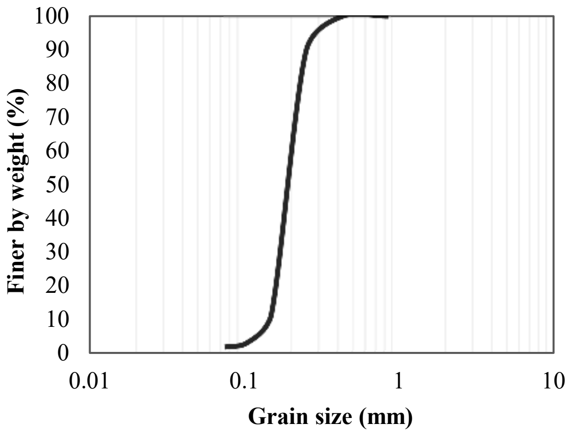


Figure 1. Grain size distribution of the soil sample

Table 1. Parameters of soil characteristics

Parameter	Sand	Bentonite
Liquid Limit (%)	-	135
Plastic Limit (%)	-	40
Plasticity Index (%)	-	95
G_S (g/cm^3)	2.75	2.7
Soil type (USCS)	SP	CH
ρ_d (min) (g/cm^3)	1.498	-
ρ_d (max) (g/cm^3)	1.767	-
D_{50} (mm)	0.22	-
e_{min}	0.526	-
e_{max}	0.8	-
C_u	1.6	
C_s	1.1	

The naphthalene used from the Tehran Oil Refinery with density of 1.14 g/cm^3 and a boiling point of 218°C and as well as phenol supplied by Merck (Darmstadt, Germany) with a density of 1.07 g/cm^3 and a boiling point of 181.7 °C as the hydrocarbon pollutants used in this study.

Preparation of contaminated soil

In order to prepare the sample, first to increase the uniformity of the sample , the values of soil that are required at each stage to build the sample are rejected from sieve 4 to make the soil uniform. The soil that was passed from sieve was then cooled to dry for 24 h at a temperature of 105°C. The soil prepared with weight percentages (20 %, 40 % and 60 %) of sodium bentonite and weight percentages (10 % , 15 % , 20 % , and 25 %) of phenol and naphthalene weere mixed [31]. Contaminated soil mixtures prepared at 30°C were maintained for at least one week to obtain homogeneous and uniform samples [32]. The experimental variables are summarized in Table 2.

Table 2. Parameters tested in this study

Domain of change	Number of changes	Variable type	Unit	Parameter
DNAPLs: Phenol, Naphthalene	2	Independent	-	NAPLs
10, 15, 20, 25	4	Independent	% wt	Concentration of NAPLs
20, 40, 60	3	Independent	% wt	Bentonite clay
3 months	3	Independent	day	Age

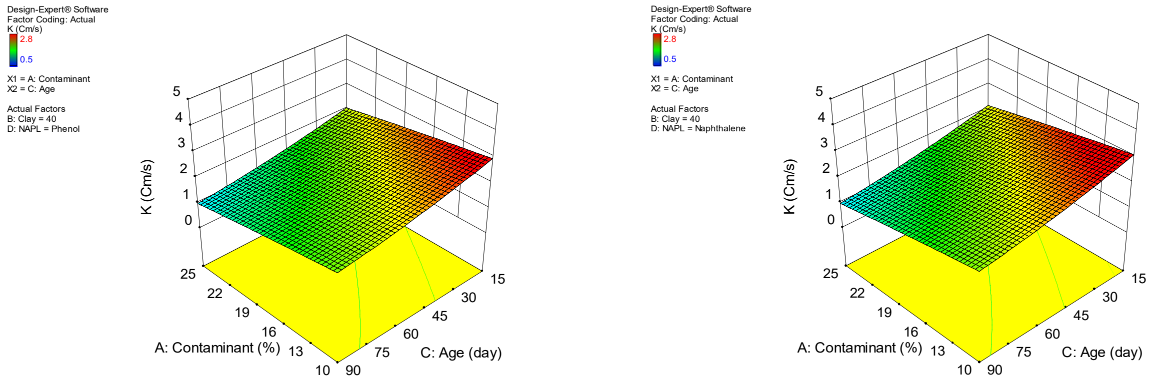
Response surface methodology

Response surface methodology (RSM) is a set of mathematical and statistical methods used to optimize the various processes, so that by applying a multivariate equation to the data set with the objective of statistical prediction, it describes the behavior of the set [33]. Response surface methodology is useful for analysis of experiments where one or more independent variables are influenced by many variables and the purpose is to optimize the response. One of the advantages of this approach and Design Expert software is in reducing the number of experiments provides the possibility of providing a mathematical relationship between the independent variable and dependent variables. In this method, in addition to the numerical variables, it is possible to investigate the effects and interations of variables [34]. In order to design the experiments and to analyze the results, the Design-Expert software (version: 10.0.6) was used.

RESULTS AND DISCUSSION

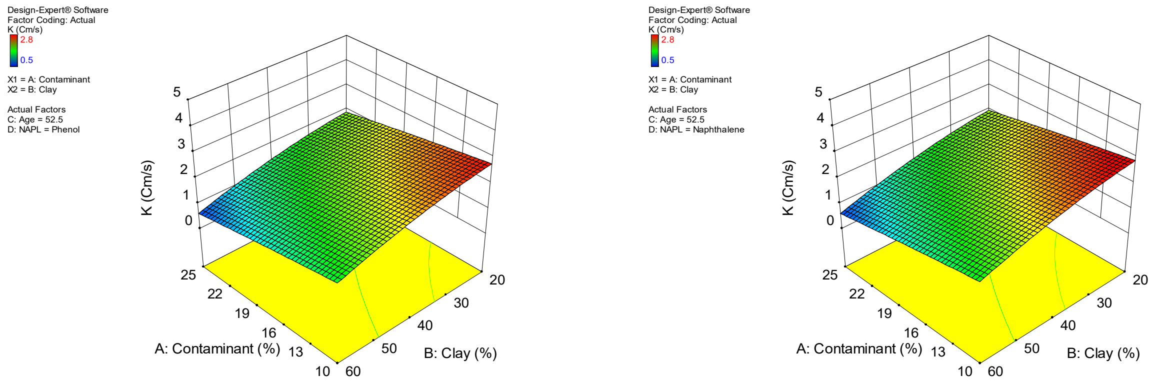
Simultaneous effect of parameters

According to Figures 2, 3 and 4 the effect of independent parameters, the age of infection and the percentage of bentonite were investigated. Based on obtained results shown in Figure 2, by increasing the amount of hydrocarbon contamination and the percentage of the content of sodium bentonite clay increased. It is observed that the surface of the contaminated soil with the phenol as well as the exposure of the naphthalene contaminated soil was observed by the reduction of permeability. According to Figure 2, per 20% bentonite and 10% contaminants, the permeabilities for phenol-contaminated soil and naphthalene-contaminated soil were 2.38 cm/s and 3.55 cm/s, respectively. This trend was gradually reduced, so that for 60% of bentonite clay and 25% of the contaminant, the permeabilities of the contaminated soil with naphthalene and phenol contaminated soil were 0.07 cm/s and 0.45 cm/s, respectively. The simultaneous effect of contamination age and the percentage of pollutants on the permeability of soil is shown Figure 3. According to the obtained results by increasing the rate of contamination and the percentage of pollutants, the soil permeability decreases. At the age of 15 days, per 10% of the contaminant, the permeability rate for phenol-contaminated soil was 2.5 cm/s, which decreased by 40% with respect to time and at the age of 45 days. Over the course of 90 days, for 25% of the contaminant, permeability was reduced to 1 cm/s. This rate for naphthalene contaminated soil at 15 days and 10% naphthalene was 2.8 cm/s, which at 90 days and 25% contaminant reduced to 0.8 cm/s. Based on Figure 4, the simultaneous effect of time and percentage of bentonite clay content based on permeability is observed.



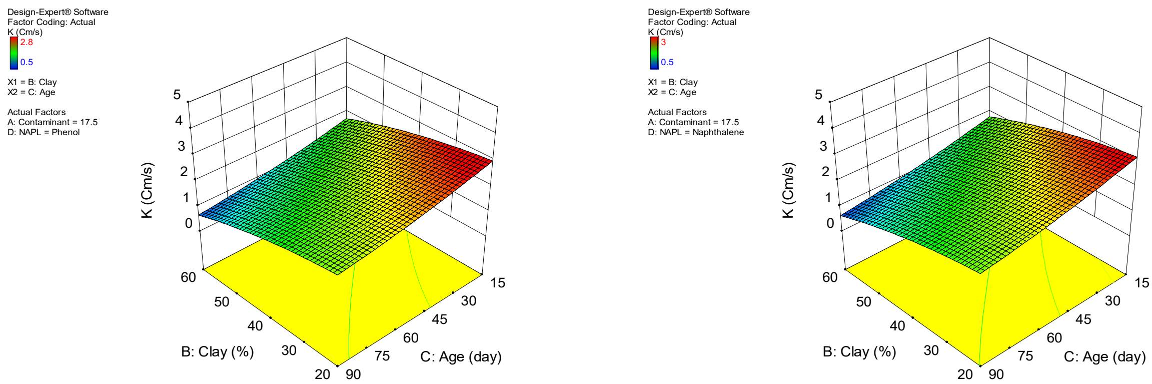
a) Phenol b) Naphthalene

Figure 2. Effect of contaminant percentage a) phenol, b) naphthalene and bentonite clay on soil permeability



a) Phenol b) Naphthalene

Figure 3. Effect of contaminant percentage a) phenol, b) naphthalene and age on soil permeability



a) Phenol b) Naphthalene

Figure 4. Effect of bentonite clay percentage and age on soil permeability a) phenol, b) naphthalene

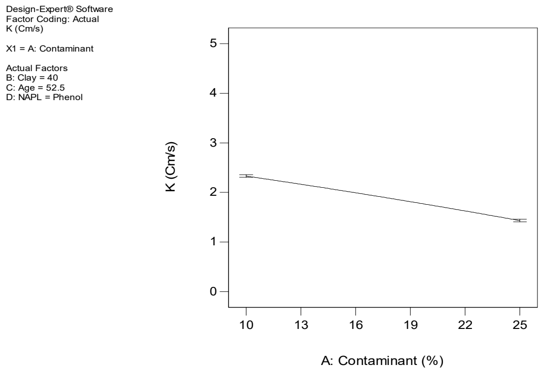
According to previous cases, by increasing the age of infection and the percentage of bentonite, permeability was significantly decreased. Thus, in the early age per 20% bentonite permeability in naphthalene-contaminated soil was decreased by 3 cm/s, which after 90 days and the 60% percentage of bentonite reached to to 0.5 cm/s. This trend for phenol-contaminated soil was from permeability of 2.8 cm/s to 0.5 cm/s, which was reduced by 82%. The obtained results were due to the low cohesion, sealing, and permeability of bentonite. Al-Sanad et al. [35] observed the continuous reduction in the permeability of a poorly-graded sand. Khamehchiyan et al. [12] also reported the continuous reduction in the permeability of a poorly-graded sand and a silty sand caused by hydrocarbon contamination and attributed it to the reduced porosity of the contaminated soil. Sarmadi et al. [32] and Oyediran et al. [36] also observed lower permeability and angle of internal friction angle and higher cohesion in a sandy soil contaminated with hydrocarbons. The decrease rate of permeability in naphthalene-contaminated soil was higher than phenol. Shin and Das [37] and Al-Aghbari et al. [21] reported that hydrocarbons with higher kinematic viscosity result in higher reductions in the permeability, which completely agrees with the results of the present study. The reduction in the hydraulic permeability may be due to the fact that diesel and benzene fill the void spaces of the soil, facilitating the fluid flow. The reduction in the permeability of soils contaminated with hydrocarbons has been reported by other researchers [12, 18, 37]. According to the results, the percentage of contaminants, bentonite clay content and age of contamination are the most important parameters affecting the permeability of polluted soil to hydrocarbons. According to the obtained results and previous studies, the model has sufficient validity [38].

Individual effect of parameters

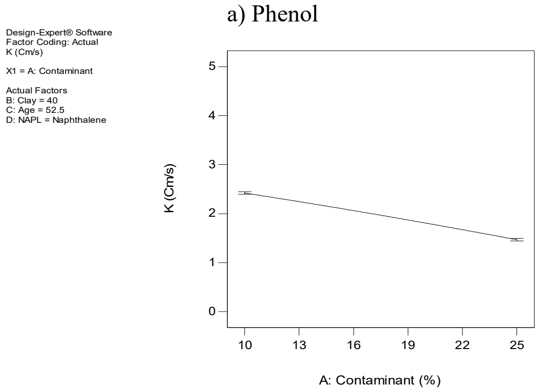
At this stage, by drawing some one-dimensional diagrams, the relationship between each effective parameter and the permeability is separately investigated.

According to Figure 5, the permeability of phenol and naphthalene pollutants is shown in different percentages (10, 15, 20, and 25%). Due to an increase in pollutants, permeability rates have decreased with relatively high gradient. It can be estimated that the relation between permeability coefficient and the percentage of pollutants is roughly linear. The permeability of naphthalene-contaminated soil per 2.5% of pollutant is 2.5 cm/s and faces an increase in 25%, in pollutant, to 1.5 cm/s, experiencing decreased of 40%. In the case of the phenol contaminated soil, per 10% phenol, permeability coefficient equal to 2.3 cm/s, increasing by the percentage of phenol to 25% to 1.7 cm/s, representing corresponding permeability decreased by 26%. Figure 6 shows the relationship between permeability and the percentage of bentonite clay. Due to the shape, the permeability has decreased nonlinearly with increasing percentage of bentonite. Permeability coefficient for naphthalene contaminated soil is 2.5 cm/s for 20% bentonite, 1.8 cm/s for 40% bentonite and also 1.2 cm/s for 60% bentonite. In total, it has encountered a decrease of by 40%. This trend has increased from 2.2 cm/s to 1.2 cm / s for phenol-contaminated soil, which has decreased by 45%.

Figure 7 also shows the relationship between permeability and age of contamination. In the light of increasing age of contamination, permeability decreases with a gentle slope. In the phenol-contaminated soil sample at an early age, the permeability was 2.5 cm/s, which reaches to 1.8 cm/s after 90 days, illustrating a decrease of 18% with respect to permeability.

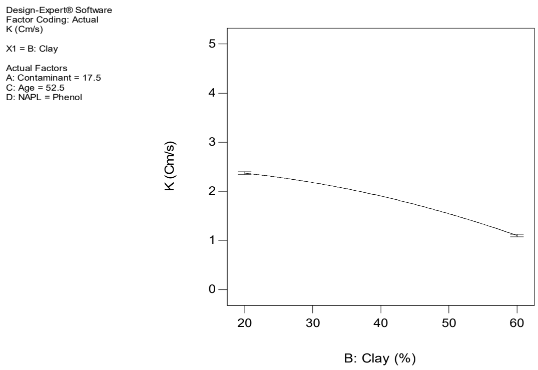


a) Phenol

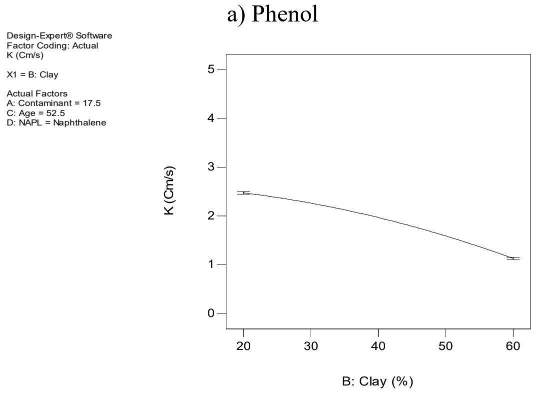


b) Naphthalene

Figure 5. Effect of contaminant percentage on soil permeability a) phenol, b) naphthalene

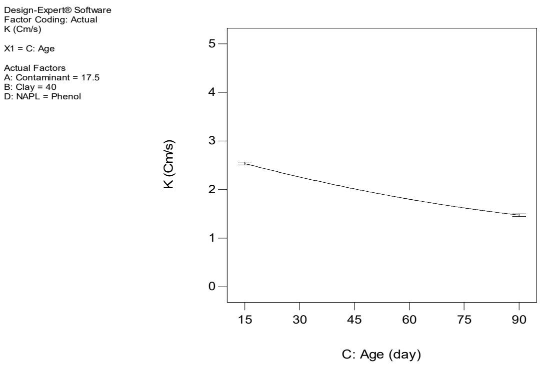


a) Phenol

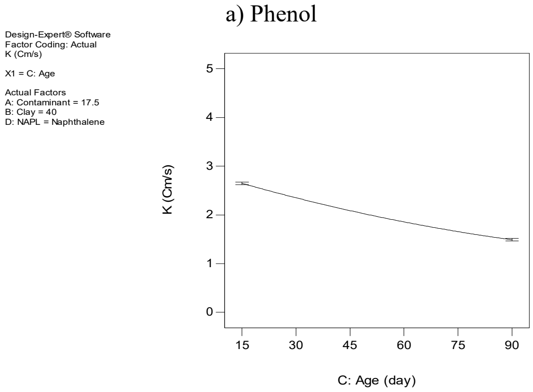


b) Naphthalene

Figure 6. Effect of bentonite clay percentage on soil permeability a) phenol, b) naphthalene



a) Phenol



b) Naphthalene

Figure 7. Effect of age on soil permeability a) phenol, b) naphthalene

Also, for naphthalene contaminated soil, the permeability decreased from 2.8 cm/s to 1.7 cm/s, accompanied by a decrease of approximately 40%. Based on obtained results, the effect of pollutant content, the amount of bentonite and time on the soil permeability is quite obvious.

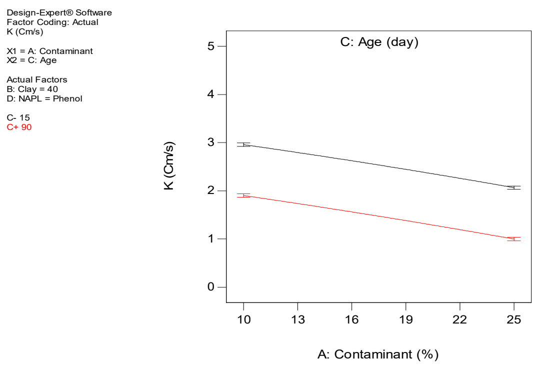
Interaction and perturbation graphs

The interaction and simultaneous amount of pollutant, percentage of bentonite clay and time on the permeability are shown in Figures 8, 9 and 10. The purpose of these graph is to investigate the synergistic effect of two variables on the other one.

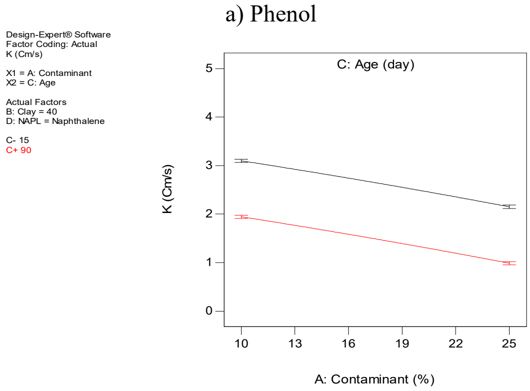
In Figure 8, the synergistic effects of two parameters, contamination percent and time on permeability is illustrated. As mentioned in the previous sections, owing to increasing in the percentage of hydrocarbons and with time, the soil permeability declines. According to Figure 8, in the early age of 15 days, with an enhancement in the percentage of contaminants from 10 to 25%, the permeability for phenol-contaminated soil has increased from 3 cm/s to about 2.3 cm/s. In 90 days, the permeability has decreased from 1.8 cm / s to 1.4 cm/s as well. This trend has increased for naphthalene contaminated soils in the early stages of contamination from 3.2 cm/s with an enhancement of 25% to 2.2 cm/s. Also, in 90 days, the permeability increased from 2 cm/s to 1 cm/s, showing a significant decrease in permeability by 50%. The interaction of two variables, the amount of pollutant and the percentage of sodium bentonite clay, is also shown in Figure 9. For 20 % of the value of phenol with increasing phenol from 10 to 25 %, the permeability from 2.8 cm/s to 2.2 cm/s was reduced by 22%.

For 60% bentonite, the permeability is changed from 1.5 cm/s to 0.5 cm/s. Also, for 20% bentonite, the permeability of naphthalene-contaminated soil decreased from 2.9 cm/s to 2.1 cm/s. According to the results, the significant effect of bentonite on reducing the permeability is vividly proved.

The synergistic effect of time and the amount of bentonite was also investigated in Figure 10. Due to the shape of both hydrocarbon-contaminated soil samples, the increase in the percentage of bentonite, the decrease in permeability is occurred with an almost steep slope. At the age of 15 days, with enhancing the percentage of bentonite from 20 to 60%, the permeability of phenol-contaminated soil decreased from 3 cm/s to 2 cm/s. While, by the time of 90 days, the rate of permeability experiences an enhancement from 2.5 cm/s to 0.5 cm/s. The trend of changes for naphthalene-contaminated soils is cut down from 3.1cm/s to 2.1 cm/s by increasing the concentration of bentonite from 20 to 60% at the early age. On the other hand, over 90 days, which the trend decreases more than 2 cm/s has reached to 0.45 cm/s.

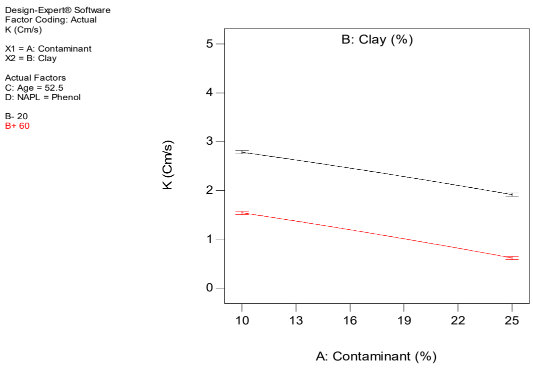


a) Phenol

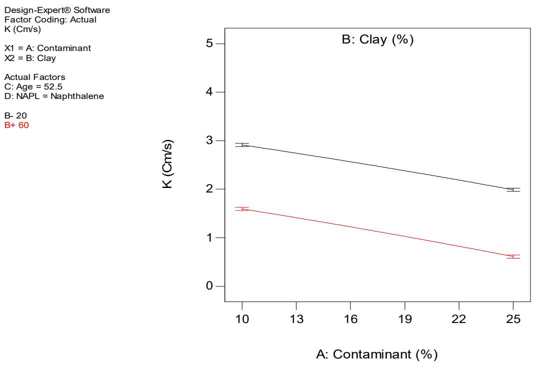


b) Naphthalene

Figure 8. Effect of contaminant and age on soil permeability a) phenol, b) naphthalene

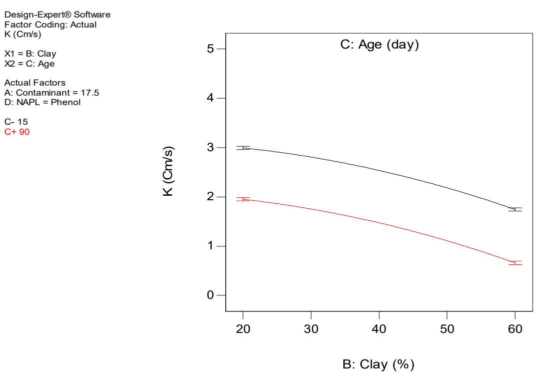


a) Phenol

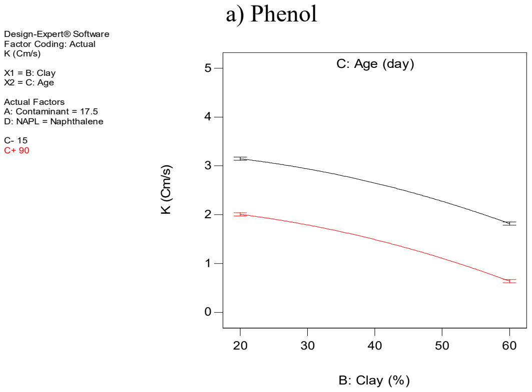


b) Naphthalene

Figure 9. Effect of contaminant and bentonite clay on soil permeability a) phenol, b) naphthalene



a) Phenol



b) Naphthalene

Figure 10. Effect of bentonite clay and age clay on soil permeability a) phenol, b) naphthalene

Variance analysis and mathematical model

After full analysis of the results, the corresponding model for phenol and naphthalene is presented in Equations (1) and (2), respectively. According to the model, bentonite had the greatest effect on permeability. As shown in Table 3, the results of the variance analysis demonstrate the validity of the proposed model with p-value of 0.0001.

Also, the variables of contamination percentage and time, which are statistically p-value, are less than 0.0001. Therefore, the hypothesis that these two factors have no impact on permeability is rejected, implying that these two parameters have a statistically significant effect on permeability. Therefore, according to the obtained model, the permeability is inversely related to the percentage of contamination, the amount of bentonite and the age of contamination, which has a slightly greater effect on naphthalene-contaminated soil.

$$K\left(\frac{cm}{s}\right) = +4.18676 - 0.035995 \times \text{Contaminant} + 5.69712\text{E}-003 \times Clay - 0.020415 \times Age - 9.58333\text{E}-005 \times \text{Contaminant} \times Clay - 8.88889\text{E}-006 \times \text{Contaminant} \times Age - 1.33333\text{E}-005 \times Clay \times Age - 5.58423\text{E}-004 \times Contaminant^2 - 4.38250\text{E}-004 Clay^2 + 6.60779\text{E}-005 Age^2 \quad (1)$$

$$K\left(\frac{cm}{s}\right) = +4.45963 - 0.039675 \times \text{Contaminant} + 3.75712\text{E}-003 \times Clay - 0.021690 \times Age - 9.58333\text{E}-005 \times \text{Contaminant} \times Clay - 8.88889\text{E}-006 \times \text{Contaminant} \times Age - 1.33333\text{E}-005 \times Clay \times Age - 5.58423\text{E}-004 \times Contaminant^2 - 4.38250\text{E}-004 Clay^2 + 6.60779\text{E}-005 Age^2 \quad (2)$$

CONCLUSION

The present work investigated the effect of dense non-aqueous phase liquid contaminants, phenol and naphthalene on soil permeability. For this purpose, different percentages of 10, 15, 20 and 25% of contamination for mixing percentages of 20, 40 and 60% of bentonite on fine-grained sandy soils was investigated over a period of 90 days. Experiment design is also performed in Response Surface Methodology (RSM) method and by making use of the Design-Expert software expert. According to the obtained model, the amount of bentonite had the greatest effect on soil permeability. The results also illustrate that, in general, permeability significantly decrease owing to an increasing the percentage of hydrocarbon pollutants, bentonite content and age of contamination. Moreover, in the light of the results of the synergistic effect of time, the percentage of pollutants and the amount of bentonite on the reduction of permeability is quite evident.

Table 3. ANOVA analysis for response surface quadratic model

Source	Sum op squares	df	Mean squares	F-value	p-value prob > F	
Model	62.90	df	3.00	727.45	< 0.0001	
A-Contaminant	7.65	21	7.65	1857.32	< 0.0001	Significant
B-Clay	22.79	1	22.79	5535.75	< 0.0001	
C-Age	17.54	1	17.54	4260.72	< 0.0001	
D-NAPL	12.70	1	4.23	1028.32	< 0.0001	
AB	6.612E-003	3	6.612E-003	1.61	0.2101	
AC	2.000E-004	1	2.000E-004	0.049	0.8263	
AD	0.26	1	0.088	21.26	< 0.0001	
BC	3.200E-003	3	3.200E-003	0.78	0.3817	
BD	0.21	1	0.071	17.31	< 0.0001	
CD	0.084	3	0.028	6.79	0.0005	
A2	0.041	3	0.041	9.88	0.0026	
B2	1.27	1	1.27	307.64	< 0.0001	
C2	0.36	1	0.36	86.44	< 0.0001	
Residual	0.24	1	4.118E-003			
Lack of Fit	0.17	58	4.535E-003	1.36	0.2315	
Pure Error	0.066	38	3.324E-003			Not Significant
Cor Total	63.14	20	3.00	727.45	< 0.0001	

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Persian Abstract

چکیده

آلودگی خاک به‌عنوان مسئله‌ی بحث‌برانگیز در اغلب کشورها محسوب می‌شود. در سال‌های اخیر درک چگونگی نفوذ آلاینده‌ها از منابع مختلف به خاک از اهمیتی ویژه‌ای برخوردار است. امروزه آلاینده‌های هیدروکربنی از مهم‌ترین منابع آلوده‌کننده خاک به شمار می‌آیند که به‌عنوان موضوع اساسی در سطح جهانی شناخته می‌شود. هدف از این پژوهش، بررسی آماری و مدل‌سازی اثر همزمان پارامترهای زمان و غلظت آلاینده‌های فنول و نفتالین به‌عنوان آلاینده‌های (DNAPL) با درصدهای مختلف (۱۰، ۱۵، ۲۰ و ۲۵ درصد) و میزان بنتونیت موجود در خاک ماسه‌ای ریزدانه با درصدهای مختلف (۲۰٪، ۴۰٪ و ۶۰٪) می‌باشد. طراحی آزمایش به روش سطح پاسخ (RSM) و با استفاده از نرم‌افزار Design-Expert انجام شد. بر اساس مدل ارائه‌شده، مقدار بنتونیت بیشترین تأثیر بر میزان نفوذپذیری خاک را داشته است. نتایج نشان داد، به‌طورکلی با افزایش درصد فنول و نفتالین، مقدار بنتونیت و سن آلودگی نفوذپذیری به‌طور قابل‌توجهی کاهش می‌یابد. به طور متوسط کاهش ۸۰ درصدی نفوذپذیری در خاک آلوده مشاهده شد که این کاهش در خاک آلوده به نفتالین بیشتر بوده است. همچنین با توجه به نتایج اثر هم‌افزایی زمان، درصد آلاینده هیدروکربن و مقدار بنتونیت بر کاهش نفوذپذیری کاملاً مشهود است.