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Permeability of Two Clayey Soils Exposed to Petroleum Products and Organic Solvents

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ABSTRACT: Clayey soils are the most common material used for water sealing and undertake an important role in controlling landfill-related pollution. Organic liquids can adversely affect the effectiveness of clay liners by drastically increasing their hydraulic conductivity. The aim of this study is to investigate and compare the permeability in two types of clay with different plasticity, exposed to the flow of kerosene and diesel as non-polar immiscible liquids and ethanol as a miscible liquid with an intermediate dielectric constant. The effects of plasticity and water content for a given compactive effort are also investigated. Two different clayey soils with different plasticity were provided and their physical properties determined. Next, modified constant-head permeability tests were conducted on the samples. Results show that the lower dielectric constant of the organic fluids, leads to an increase in hydraulic conductivity. Research has shown that organic fluids shrink the diffuse double layer due to their lower dielectric constant and reduce its thickness. Shrinkage of the double layer leads to higher permeability and lower plasticity in the soil. As a result, the void space for the passage of the fluid increases. With the decrease the dielectric constant from 80.1 to 1.8, permeability is increased up to 1800 times. On the other hand, results show that for a clay with a higher liquid limit and plastic limit, permeability for all the liquids investigated in the research is lower.

Keywords: Atterberg Limits, Clay Soil, Dielectric Constant, Permeability, Plasticity.

INTRODUCTION

Soils have long been used as the first choice to block any undesirable flow in civil engineering projects. In the design of impermeable layers for landfills and bottom of lakes used as disposal places for chemical reservoirs during emergency evacuation, it is necessary that the soil layer be able to prevent the permeation of any kind of contaminant. Many researchers have focused on the use of various additives to soil in order to reduce its permeability and/or enhance its performance as barrier (Abdi and Parsapazhouh, 2009; Qiang et al., 2014; Mousavi and Wong, 2015). Based on the regulations imposed by the US Environmental Protection Agency, permeability of these clay layers in the floor of landfills must be lower than 10^{-9} m/s. Similar values are used as reference in Germany, Brazil and China. When the blocked liquid is a type of aqueous solution,

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compacted clayey soils can be used as barriers. However, the performance of these soils against non-polar liquids and solvents like hydrocarbons or other organic solvents is still a controversial subject.

Non-aqueous phase liquids or NAPLs are liquid solution contaminants responsible for a significant portion of soil and water contamination. These include petroleum products. NAPLs do not dissolve in or easily mix with water due to their different density and chemistry. They are either spilled on the soil surface or leaked underground. Such fluids with densities lower than that of water (light NAPLs) tend to float on top of the groundwater. Their movement is controlled by the gradient of the top of the groundwater and the permeability for the fluid and soil combination. Fluids with densities greater than that of water (dense NAPLs) tend to flow vertically or sub-vertically through the groundwater until their movement is arrested by an impervious soil stratum. Movement is then largely controlled by the gradient of the top of the impervious stratum and the permeability for the fluid and the soil.

As previously mentioned, soil permeability is a key parameter in waste and contamination control. Factors and parameters influencing the permeability of soil can be classified into three main groups:

1. Factors associated with the permeating fluid (physical and chemical properties such as viscosity, pressure, density and concentration)

2. Factors associated with physical and chemical properties of the soil (flow path tortuosity, void ratio, degree of saturation, water-soil potential, shape and size of soil particles, pore size distribution, soil structure and texture, temperature)

3. Factors affecting forces in the double layer and the interaction of clay with water (water-soil interaction, ionic concentration, double layer thickness, etc.)

In cases of organic fluid flowing through clay, properties of the permeating fluid, chemical and mineral composition of the soil and the natural absorption of the permeating fluid by the surface of soil particles are some of the important and effective factors.

The clay exchange of ions and unsatisfied charges caused by the broken bonds specially in the edges, result in the presence of negative charges on the surface of clay particles. Due to the existence of the mentioned charge, clay soil particles are chemically active and have a high ion exchange capacity and their behavior is essentially affected by the environment, depending on the type of clay minerals. Using the double layer theory, it is possible to justify the dependence of behavior on environment properties and the changes in clay properties caused by changes in physical and chemical characteristics of the fluid around the particles, to some extent. Based on this theory, the charged clay particle surface and the distribution of electric charges around it is known as the diffuse double layer.

Based on the Gouy-Chapman theory, thickness of the double layer is equal to:

$$T_h = (\frac{\varepsilon.K.T}{8.\pi.n.e^2.v^2})^{1/2} \tag{1}$$

T_h: is thickness of the double layer; ε: is dielectric constant; K: is Boltzmann constant; T: is temperature; n: is electrolyte concentration; e: is elementary charge and v: is ionic valence.

According to the above formula, the thickness of the double layer around the clay particles has a direct relationship with the dielectric constant of the permeating fluid. Therefore, fluids with lower dielectric constants bring about the shrinkage of the double layer.

Based on Bolt (1956); Fernandez and Quigley (1988) and many other findings, permeability of clay for organic fluids can increase up to tens of times that of aqueous solutions.

Van Olphen (1963) suggested that a reduction in the dielectric constant will induce flocculation (a change in fabric), thereby providing more pore space for fluid flow.

Mitchel et al. (1965) showed that the hydraulic conductivity of compacted clays, depends highly on the compactive effort and the soil water content. As seen in Figure 1, an increase in water content and compactive effort causes a decrease in the hydraulic conductivity. Similar results have been reported by Lambe (1955) and Benson and Daniel (1990).

Mesri and Olson (1971) measured a large increase in permeability when fluids other than water were used in consolidation tests. They suggested that the differences in polarity between water and organic permeants are responsible. The reasoning was that the water molecule, with a large dipole moment, forms strong hydrogen bonds with the surface of silicate minerals in the soil. In clayey soils, which have small pores and often contain high surface area minerals, most of the pore fluid exists in an adsorbed film held in place by intermolecular attractions of various kinds. The existence of an adsorbed layer of water on the surfaces of the soil particles effectively reduces the size of the pore channels, decreasing the ability of the fluid to move through the soil. Fluids lower in polarity than water, as is the case for many organic liquids, are less strongly bonded to the soil minerals, and the fluid flows more easily through the pore network.

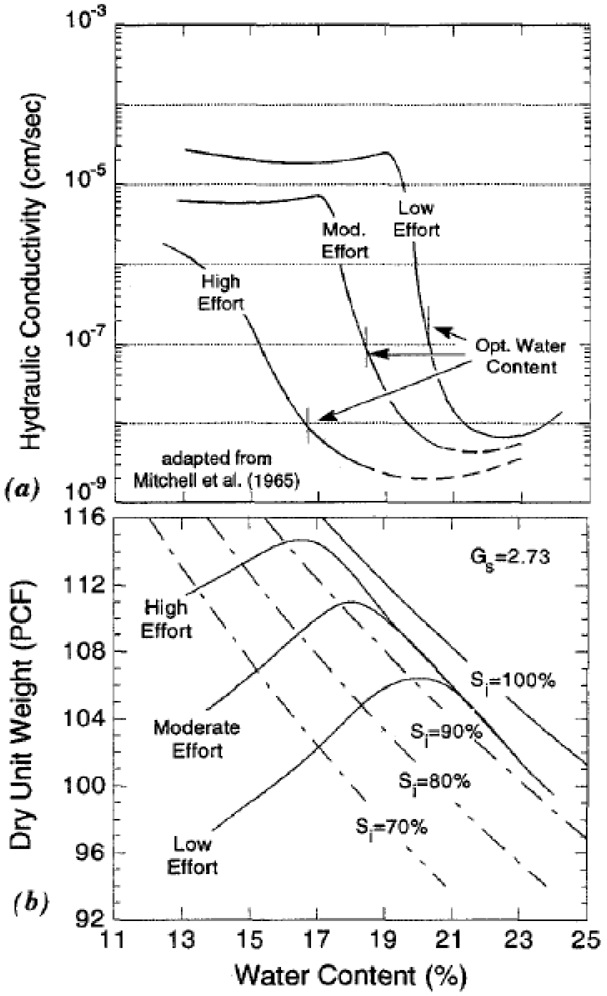

Fig. 1. Relationship between hydraulic conductivity, water content and compactive effort (Mitchell et al., 1965)

The second explanation, and perhaps the more widely accepted at present, is that the interaction of organic fluids with the soil minerals causes the soil to change its fabric in a manner that provides an easier passage for the pore fluid.

Mersi and Olson (1971) and Gilligan and Clemence (1984) reported that when clay is saturated with an organic fluid, its permeability is much higher than when it is saturated by and exposed to the flow of water.

Based on the findings of Mitchell (1976), permeability of clay can be highly affected by factors such as particle size or specific surface, particles arrangement, degree of saturation, electrolyte type and concentration, electro-chemical properties of the clay part of the soil and external pressure. For organic fluids, the connection between physical and chemical properties of the fluid and permeability are not as obvious and harder to predict.

Gilligan (1983) and Gilligan and Clemence (1984) suggested that flow of an organic fluid through soil rich with clay, results in the formation of tactoids in the soil which leads to the development of a grain structure that results in an increase in void space and therefore, easy passage for the flow of the organic fluid through the soil.

Experimental results by Quigley and Fernandez (1988) show that clay is highly permeable by non-polar organic liquids, the reason being the cracks formed as a result of the decrease in the thickness of the absorbed water layer.

Benson and Trast (1995) investigated the effect of factors such as liquid limit and plasticity index on clay permeability by studying the permeability of 13 types of clay.

Figure 2 presents the variation of hydraulic conductivity with liquid limit and plasticity index. As shown in Figure 2, with the increase in plastic and liquid limit, hydraulic conductivity decreases.

Research conducted by Ahangar et al. (2011) confirmed the notion that as soil plasticity increases, its permeability decreases.

Amarasinghe et al. (2012) used a newly developed porous rigid wall, flexible wall permeability device to evaluate permeability and consolidation characteristics of Na-montmorillonite clay with five fluids with different dielectric constants ranging from 2.4 to 110. The coefficient of permeability was reported to be of the order of 4-5 × 10^5 times higher, 3 × 10^3 higher and 2 × 10^2 lower than that of water for samples permeated with low-/non-polar fluids, medium-polar fluid, and highly polar fluids respectively. The vast range of variations in the permeability values for the same clay was attributed to the clay-fluid molecular interactions that affect the mobility of fluid molecules and drastically alter the clay microstructure.

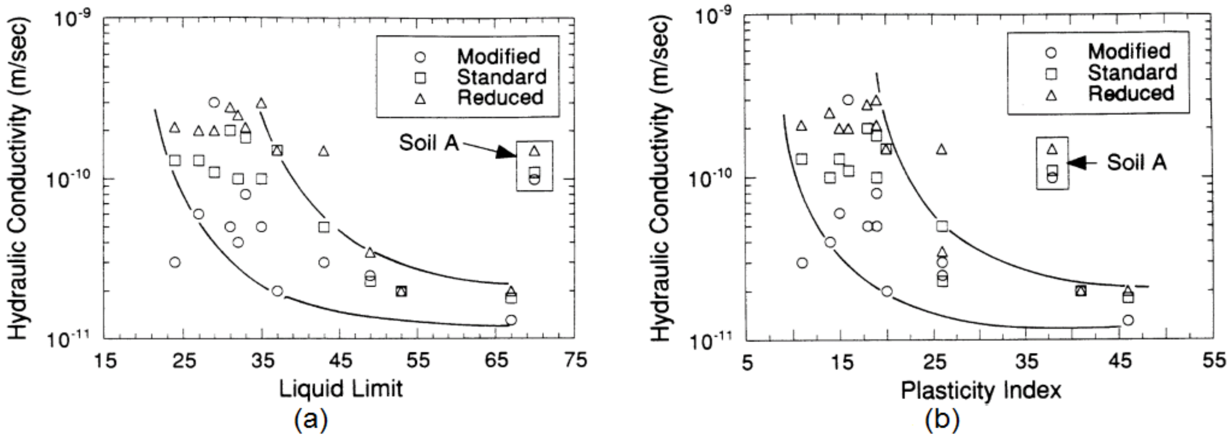

Fig. 2. Variation of hydraulic conductivity versus: a) liquid limit, b) plasticity index (Benson and Trast, 1995)

Many studies on clay behavior have pointed out changes in the soil behavior brought about by changes in the attractive and repulsive forces when an organic pollutant is the permeant as opposed to water. (Olgun and Yildiz, 2012; Spagnoli et al., 2012; Mosavat and Nalbantoglu, 2013; Liang et al., 2015; Balaban et al., 2015).

Mosavat and Nalbantoglu (2012) reported changes in the geotechnical properties of clay permeated with hazardous liquids including flocculation and aggregation of the soil particles due to the lower dielectric constant of the pore fluid which led to the faster sedimentation of the soil particles and a non-plastic granular texture in the soil when permeated with ethylene glycol, toluene and the sea water.

Goodarzi et al. (2016) investigated the influence of different organic pollutants of various concentrations including methanol, acetone, acetic acid, and citric acid on the macro and microstructure responses of Na+-Bentonite. They reported a reduction in the plastic properties of the soil as well as an increase in the permeability of Na+-Bentonite. These changes were ascribed to the collapse of the diffuse double layer, reduction in the surface charge density of the particles and clustering and aggregation of particles in the presence of the organic chemicals. However, it was reported that changes in soil behavior may not be completely explained by the diffuse double layer theory since these changes are not precisely comparable only with reference to the changes in the dielectric constant of pore fluid.

Machado et al. (2016) recognized that the influence of the permeating liquid polarity or its dielectric constant on the soil is significant. Induced changes including shrinkage of clusters, localized cracks and changes in the soil structure can modify the intrinsic permeability of the soil in a way that conventional equations may no longer apply. In addition, they reported that the high solids/water interactions in high plasticity soils that reduce the interstitial water availability for flow, are much less pronounced in the case of low-polarity fluids. As a result, Machado et al. (2016) developed a model based on experimental soil permeability data obtained for different fluids in a variety of soils to predict soil permeability of organic fluids based on soil and fluid properties. The proposed empirical equation was defined based on the following parameters: soil permeability for water, plasticity index, degree of water saturation, relative dielectric constant of the fluid, density and viscosity of the fluids.

The interactions between fine-grained soils and various types of inorganic fluids have been investigated in the recent years by multiple researchers including Siddiqua et al. (2011), Cui et al. (2012), Chen and Huang (2013), Zhu et al. (2013) and Goodarzi and Akbari (2014). In this paper, the aim is to investigate and compare the variation of clay permeability against the flow of kerosene and diesel as non-polar and immiscible liquids and ethanol as a polar and miscible liquid with an intermediate dielectric constant. In addition, Atterberg limits are important parameters that can provide knowledge into the chemical reactivity of clay. Mishra et al. (2011) indicated that the Atterberg limits can be used as an indirect indicator providing insight into the performance of clayey soils as barriers. Therefore in this paper, the effect of soil plasticity and soil water content in a given compactive effort, is also investigated.

MATERIALS AND METHODS

Soils used in this research are collected from the Saravan area in Guilan Province. This area is highly important as the largest dumpsite in the northern Iran is located in this region and the clayey soils in the region play an important role in blocking and reducing leachate ad contaminant migration.

The collected soils were passed through sieve number 200 and the finer portions were used in the research. To determine the soil types and their properties, several preliminary experiments were carried out. The geotechnical properties of soil were determined based on the ASTM Standard. The properties of the two clays are summarized in Table 1.

The water used in the experiments was tap water. Table 2 presents the physical and chemical properties of the fluids used in this research. The most remarkable difference between the fluids is their dielectric constant.

The laboratory device used for determining the permeability of the soil samples in this research, was designed and manufactured in the Guilan University. The main body of the device includes reservoir, pressure pump, fluid flux adjustment valves, barometers, molds/permeameter cells, graduated tubes and a fiberglass compartment to put the molds on. The molds are cylindrical-shaped with diameters of 10 and heights of 17 centimetres. The inner diameter of the molds was equal to the standard Proctor mold in order for the applied energy to conform with the standard Proctor energy as suggested by the ASTM standard. Molds are rigid-bodied and made from aluminium and they provide the possibility of compacting the soil sample inside. Therefore, samples are compacted and subjected to permeability tests within the same molds.

Two metal lids are used on top and bottom of the molds and liquid transfer pipes pass through the middle of them. Soil samples used in this research have a low permeability and need a long time to reach saturation. To reduce the required time, a high gradient is needed to be applied to the sample. A high gradient causes a large flowing force which can cause leakage from the space between the sample and the inner wall of the mold. To prevent the fluid from moving between the soil and the mold inner wall and also to increase the friction between the sample and the mold wall, the inner wall of the mold is equipped with 12 grooves with thickness equal to 5 millimetres and 2 millimetres depth. In addition, the fluid moves from the bottom of the sample upwards. The grooves of the mold are shown in Figure 3.

Table 1. Properties of the used soils

Title	Soil A	Soil B
% Finer than #200 sieve	100	100
Liquid limit (%)	62	40
Plastic limit (%)	30.1	21.3
Plastic index (%)	31.9	18.7
USCS classification	CH	CL
Specific gravity	2.74	2.69
Maximum dry unit density (gr/cm^3)	1.708	1.648
Optimum water content (%)	25.60	19.61

Table 2. Physical and chemical properties of permeant liquids at 20 ˚C

Type of Liquid	Dielectric Constant	Unit Weight (gr/cm^3)	Viscosity (mPa.s)
Water [a]	80.1	1	1
Ethanol [a]	24.3	0.81	1.24
Diesel [a]	2.13	0.82-0.86	3-4
Kerosene [b]	1.8	0.82	2.42

a: After Machado et al. (2016)

b: After Park and Fan (2007)

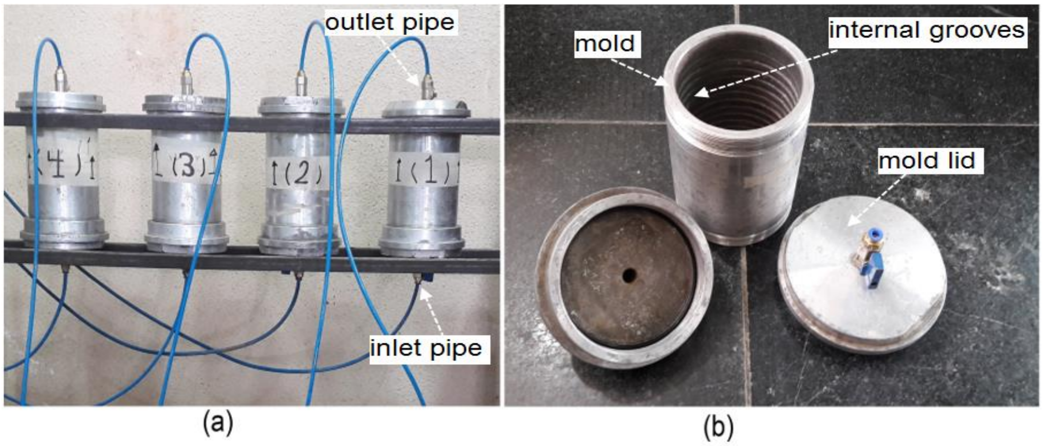

Fig. 3. a) Permeability device, b) Components of the testing mold

In the current research, two different clays (CH and CL) were used at four different water contents (two points drier than optimum, one point at optimum water content and one point wetter than optimum) to study the effect of the water content at compaction on soil permeability.

Soil was mixed with the desired amount of water and then stored in plastic bags for 24 hours for water content equalization. Samples were compacted in three layers with each layer receiving 25 blows, according to the standard Proctor compaction method (as recommended by ASTM D698). At the end of the compaction, the free height over the sample was measured to accurately obtain the sample length. In order for the fluid to enter into the sample at a balanced rate and to prevent the obstruction of the inlet plastic tap, a 2-cm filter made of two layers of geosynthetic with gravel in between was placed at the bottom of the mold. On top of the sample, a similar filter was placed to prevent the fine soil particles from exiting the sample with the fluid. After installing and adjusting the device, fluid enters the tubes from the reservoir and enters the sample from the bottom of the mold. The fluid is extracted from the sample at different rates, in a way that lower rates of fluid outflow correspond to less permeable samples.

The inflow of liquid into the sample continued until the sample was completely saturated, which is when the amount of the extracted liquid from the sample reaches a fixed amount at a fixed time interval. After complete saturation of the sample, measurements of the liquid outflow with the passage of time is initiated, and the fluid is entered into a graduated cylinder after exiting through the top tap of the mold. The amount of fluid accumulated in the cylinder from every sample at different time intervals was recorded. Tests were stopped when the variation of 3 consecutive measurements of permeability was lower than 10%.

By measuring the time and volume of the liquid outflow and by knowing the height difference between the bottom of the mold and the fluid level in the reservoir which is the applied hydraulic head, and the length and the cross section of the sample, the permeability of the samples can be calculated according to the following equation.

$$k = \frac{QL}{Aht} \tag{2}$$

where Q: is the amount of the liquid outflow; A: is the cross section of the soil sample; L: is the sample length and t: is the fluid collecting time.

Full saturation is a time-consuming process due to the size of the samples. To reduce the duration of the test, it is necessary to apply a high gradient to the sample.

Therefore, conducting the tests according to simplified falling head (SFH) method was not appropriate. One of the advantages of the constant head method is the application of large hydraulic gradients to the samples. Therefore, the constant head method was adopted for the permeability tests.

RESULTS AND DISCUSSION

Effect of Compaction on Permeability

Soil compaction causes changes in the soil structure that affect the mechanical properties of the soil, particularly permeability. Studying the Kozney-Carman equation leads to a better understanding of the impact of porosity and various parameters on the permeability of soil. The permeability of saturated soil can be defined as follows:

$$k = \frac{1}{k_0 T_f^{\,2} S_0^2} \left(\frac{e^3}{1+e}\right)\left(\frac{\gamma}{\mu}\right) \tag{3}$$

where k: is the hydraulic conductivity; k_0: is pore shape factor; T_f : is tortuosity; S_0: is particle specific surface area; e is void ratio; γ: is unit weight of the permeating fluid and μ: is viscosity of permeating fluid.

The above equation states:

1) Permeability decreases with decreasing void ratio which is itself a function of soil water content at compaction.

2) Permeability depends on the unit weight and viscosity of the permeating fluid.

3) By increasing tortuosity, permeability will decrease; i.e. the fluid flows through a more zig-zag path.

4) By increasing the specific surface area, permeability will decrease.

Figure 4 shows the permeability of compacted samples with different water content and void ratio but the same compactive effort (standard compactive effort according to ASTM D698).

Based on the figure, with increasing water content, permeability decreases for all the fluids in both soils. This reduction reflects the effect of changes in void ratio and porosity with the increase in water content. As water content is increased, essentially moving from the dry side of the compaction curve towards the wet side, maximum dry density of the soil is increased while void ratio and permeability are decreased. Soil compaction reduces the free space between the soil particles and consequently reduces porosity. Needless to say, soil with lower porosity will have a lower hydraulic conductivity. In addition, when the soil water content moves from the dry side of the compaction curve to the wet side, the soil structure changes from a flocculated state to a dispersed state. Lambe (1995) reported that at similar void ratios in a clay, the permeability is many times greater in a flocculated state than in a dispersed state. This is due to the disordered placement of clay particles in the flocculated structure and the creation of larger pores in the soil. Large pores have a great impact on the permeability of the soil. Clay particles form small clusters together, which will then form larger clusters with each other.

Another type of porosity is due to cracks and fissures that naturally occur in clay. These cracks form the largest pores in soil through which most of the flow is transferred. Therefore, during clay compaction, cracking should be prevented as much as possible. Soil water content during compaction, the compaction method and energy, size of the clay clusters, and the adherence of the layers are factors that influence the structure of the compacted clay.

For both soils with different plasticity, the highest permeability is obtained on the dry side of the compaction curve. The lowest value of permeability happens at the water content higher than the optimum water content. Measuring the hydraulic conductivity at a specific water content state, is only valid for that specific state.

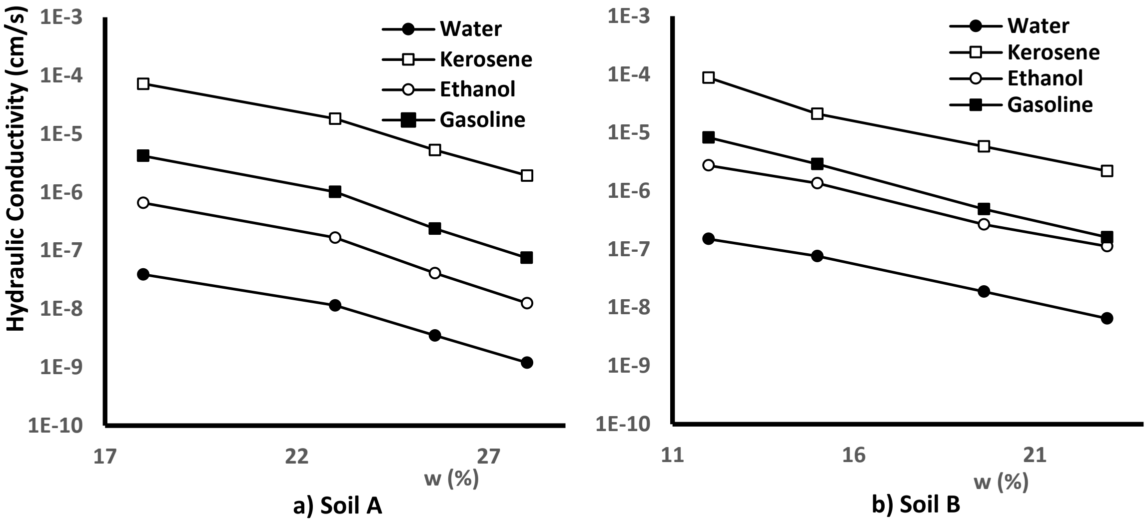

Fig. 4. Variation of soils A and B permeability against water content

For all of the water contents, kerosene permeability is higher than the other three fluids. Therefore, kerosene is the most critical organic fluid in this experiment. If permeability for a soil against kerosene satisfies the required criteria, it is definitely suitable for the three other fluids as well.

Effect of Clay Plasticity and Properties of the Permeating Fluid on Its Permeability

Permeability is influenced by the physical and chemical properties of the soil and the permeating fluid.

The interaction between the organic fluid and clay minerals is affected by three factors:

1. Lower dielectric constant of the fluid leading to a decrease in the thickness of the double layer.
2. Fluid viscosity.
3. Interactions between the organic compositions and clay minerals.

Organic compositions react with clay minerals in three ways:

- They are absorbed by the surface of clay (via hydrogen bonding and exchanging ions)
- They are absorbed by large organic molecules through van der Waals forces and are entered into the space between silicate layers
- The effect of organic molecule weight on its interaction with clay.

Figure 5 show the permeability of samples compacted at optimum water content against the flow of the fluids in this study.

As seen in Figure 5, the permeability of soil A is lower than soil B against the flow of all the fluids. By increasing the Liquid Limit (LL) and Plastic Limit (PL) of the clay, its permeability against all the fluids is decreased, but the rate of decrease is different for different fluids.

Comparing the results of this study shows that the effect of increasing soil plasticity is higher for polar fluids, which means, with the liquid limit increasing, permeability for polar fluids is decreased more in a way that changes in permeability in both soils is higher against the flow of water and ethanol. The double layer is created due to the polarity of the permeating fluid. Ethanol is miscible and has relatively small and polar molecules, which is why it binds to the water molecules and is absorbed by the surface of clay minerals and has a lower shrinking effect on the double layer. As soil plasticity increases, the permeability decreases against the flow of ethanol.

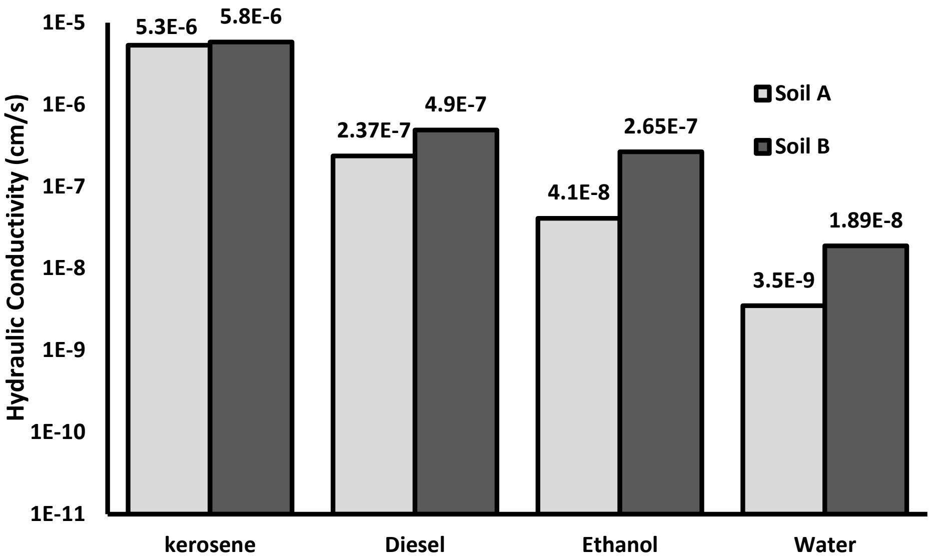

Fig. 5. Comparison of hydraulic conductivity of soils A and B at optimum water content

Kerosene and diesel are nonpolar fluids. The permeability of the two soil types against the flow of these two fluids is not different from each other. Kerosene and diesel have large and nonpolar molecules which greatly affect the double layer which is created due to the polarity of the permeating fluid. This will result in the shrinkage of the double layer. Since the plasticity of clay minerals which is due to the water molecules is eliminated, clay behaves similar to silty soils and the increase of clay plasticity does not significantly affect the permeability values for these two fluids.

Comparison of the permeability results of the two soils shows that the higher plasticity of soil A in comparison to soil B results in its better performance against the flow of water and ethanol as polar fluids. Increasing the soil plasticity for the flow of nonpolar fluids reduced permeability as well, but these changes are not significant.

Table 3 shows the permeability of soil A at optimum water content (w = 25.6%). Figure 6 presents the variation of permeability against the fluid dielectric constant for soil A. In addition, soil intrinsic permeability defined by the following equation has also been presented.

$$K = \frac{k\mu}{\rho g} \tag{4}$$

where k [LT^{-1}]: is the soil hydraulic conductivity, K [L^2]: is intrinsic permeability, g [LT^{-2}]: is gravity acceleration, ρ [ML^{-3}]: is the fluid density and μ [$ML^{-1}T^{-1}$]: is fluid dynamic viscosity. The intrinsic permeability depends solely on properties of the solid matrix. Changes induced in the soil structure as a result of the permeating liquid dielectric constant can modify the intrinsic permeability of the soil.

Table 3. Permeability of soil A at optimum moisture content

Type of Liquid	Dielectric Constant	Hydraulic Conductivity (cm/s)	k/k_w	Intrinsic Permeability K (cm^2)	K/K_w
Water	80.1	3.5×10^{-9}	1	3.57×10^{-14}	1
Ethanol	24.3	4.1×10^{-8}	11.71	6.40×10^{-13}	18
Diesel	2.13	2.37×10^{-7}	67.7	1.01×10^{-11}	282
Kerosene	1.8	5.3×10^{-6}	1514	1.59×10^{-10}	4469

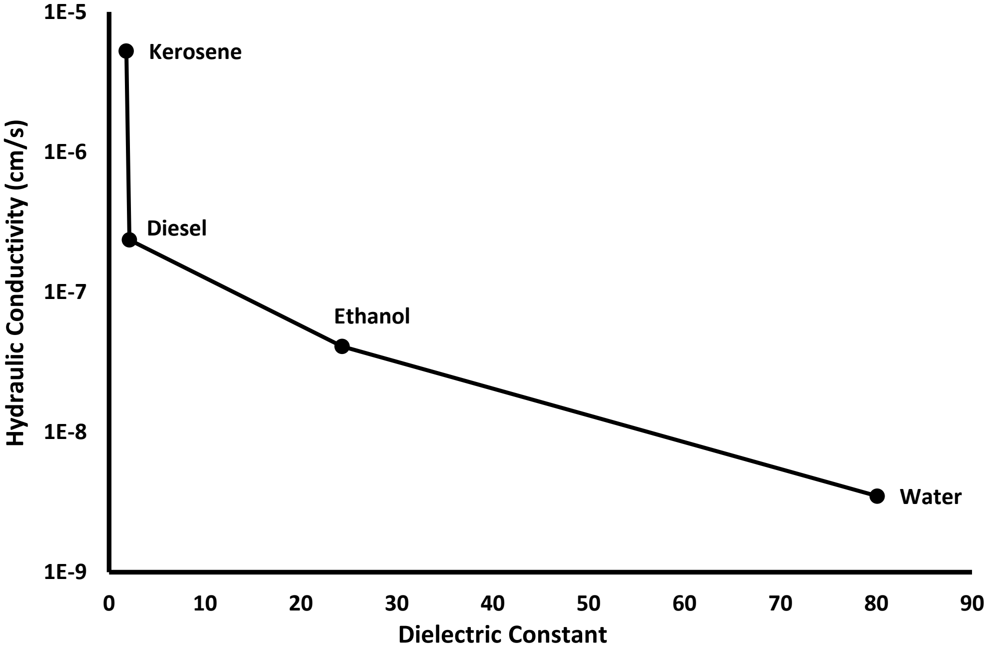

Fig. 6. Variation of permeability against the fluid dielectric constant for soil A

Based on the results shown in Table 3 and Figure 6, for a constant void ratio at the optimum water content of soil A, with decreasing dielectric constant from 80.1 to 1.8, the hydraulic conductivity has increased from 10^{-9} to 10^{-6} which means permeability has increased to 1500 times the permeability of water. Moreover, the intrinsic permeability of soil A has increased to over 4400 times the value of water.

Ethanol as a relatively polar and miscible fluid with a dielectric constant of $\varepsilon = 24.3$, has increased permeability to 12 times that of water. The double layer is created due to the polarity of the permeating fluid. Ethanol has relatively small and polar molecules, which is why it binds to the water molecule and is absorbed by the surface of the clay minerals and has a lower shrinking effect on the thickness of the double layer.

Diesel with a dielectric constant of $\varepsilon = 2.13$ increases the hydraulic conductivity up to 70 times that of water. Kerosene with a dielectric constant of $\varepsilon = 1.8$ increases the permeability to 1500 times that of water. This is because kerosene and diesel have large molecules. Decreasing the dielectric constant, results in the reduction of the repulsion between the particles; however, the magnitude of the attractive van der Waals forces in less aqueous environments is mainly dependent on the size and composition of the particles and are in effect, virtually independent of the characteristics of double layer. Since for the same repulsion, heavier particles have stronger van der Waals forces of attraction; the presence of heavier molecules changes the dispersed structure of clay into a flocculated structure and increases permeability. On the other hand, the repulsive forces have traditionally been described in the context of the diffuse double layer (Spagnoli et al., 2012). The nonpolar kerosene and diesel molecules, strongly affect the double layer by shrinking it. Since the plasticity of clay minerals which is due to the presence of the water molecules in the double layer is eliminated, clay shows a behavior similar to that of silty soils which leads to an increase in permeability.

Since decreasing the dielectric constant, results in the reduction of the double layer thickness, the repellent forces between the particles are also decreased and therefore due to the accumulation and flocculation of clay particles, the structure of the soil changes from dispersed to flocculated. The resulting increase in large pores, decrease in tortuosity, shrinkage and cracking of the skeleton of the soil leads to an increase in permeability.

But it is not just the decrease of the dielectric constant that causes the behavior observed in kerosene and diesel. Viscosity of the liquid directly affects the permeability of clay as well. Low fluid viscosity, facilitates the passage of the liquid among the soil particles and increases the permeability of clay. The vast difference between the permeability of diesel and kerosene, which have similar values of dielectric constant, can be explained by the difference in their viscosity. The low viscosity of kerosene facilitates its movement among the soil particles and further increases the soil permeability.

The higher viscosity of diesel compared to kerosene has played an important role in reducing its permeability. Therefore, the hydraulic conductivity for diesel is lower than kerosene due to its higher dielectric constant and viscosity.

The same trend was observed for tests conducted at other water contents. To avoid repetition, permeability at optimum water is presented.

Table 4 presents the hydraulic conductivity as well as the intrinsic permeability of soil B at optimum water content (w = 19.61%). Figure 7 presents the variation of permeability against the fluid dielectric constant for soil B.

Table 4. Permeability of B soil at optimum water content

Liquid	**Dielectric Constant**	**Hydraulic Conductivity (cm/s)**	k/k_w	**Intrinsic Permeability K (cm^2)**	K/K_w
Water	80.1	1.89×10^{-8}	1	1.93×10^{-13}	1
Ethanol	24.3	2.65×10^{-7}	14	4.14×10^{-12}	21
Diesel	2.13	4.9×10^{-7}	26	2.08×10^{-11}	108
Kerosene	1.8	5.8×10^{-6}	307	1.74×10^{-10}	905

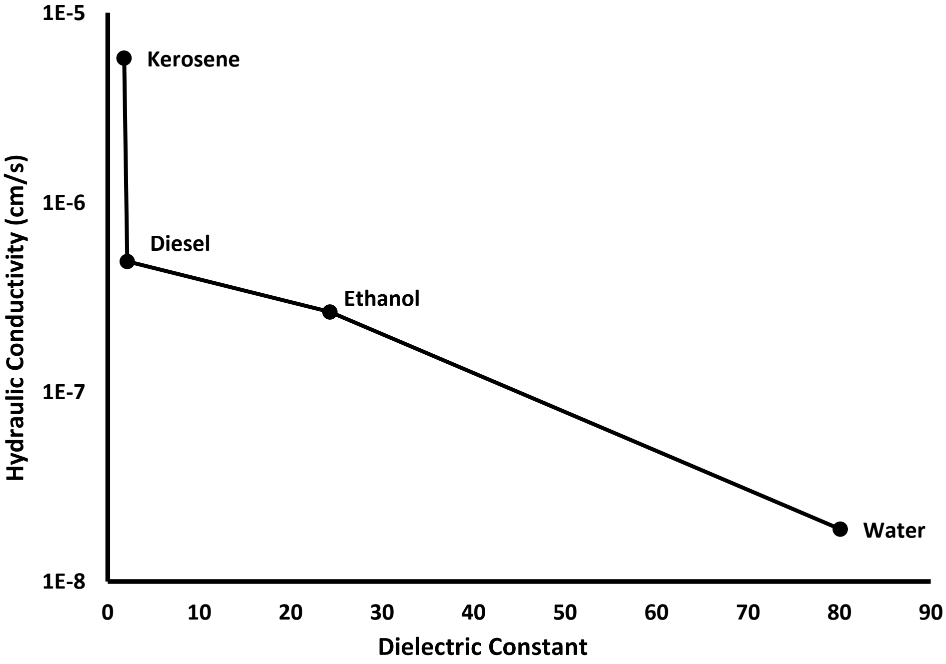

Fig. 7. Variation of permeability against fluid dielectric constant for soil B

Based on the results shown in Table 4 and Figure 7, for a constant void ratio at the optimum water content of soil B, with decreasing dielectric constant from 80.1 to 1.8, the hydraulic conductivity has increased from 10^{-8} to 10^{-6} which means permeability has increased to 300 times the permeability of water. Moreover, the soil intrinsic permeability has also increased up to 900 times that of water.

As previously mentioned, the double layer is created due to the polarity of the pore fluid. Ethanol as a relatively polar and miscible fluid with a dielectric constant of $\varepsilon = 24.3$, has increased permeability to 12 times that of water.

Diesel with a dielectric constant of $\varepsilon = 2.13$ increases the hydraulic conductivity up to 26 times that of water. Kerosene with a dielectric constant of $\varepsilon = 1.8$ increases the permeability to 300 times that of water.

In both types of clay with different plasticity values, the lowest permeability pertains to water, a polar fluid with a dielectric constant of 80.1; ethanol a relatively polar fluid with a dielectric constant of 24.3 is in the middle, and high values of hydraulic conductivity pertain to diesel and kerosene which are nonpolar fluids with dielectric constants of $\varepsilon = 2.13$ and $\varepsilon = 1.8$ respectively.

The passage of these fluids through soil A has increased the permeability for kerosene up to 1500 times and for diesel up to 70 times that of water. However, the passage of these fluids through soil B has led to increasing the permeability for kerosene up to 300 times and for diesel 26 times the permeability obtained for water. As soil plasticity is increased, permeability has decreased against the flow of polar fluids, but no significant changes in the permeability of nonpolar fluids have been observed. Thus, the difference between the permeability in polar versus nonpolar fluids in soil A is greater than soil B.

The dielectric constant is an important parameter with regards to the interactions between clay minerals and the organic fluid in a way that when the dielectric constant decreases, thickness of the double layer is decreased as well. Intrinsic permeability can reflect the changes in the soil matrix that are due to the differences in the dielectric constant of the permeating fluids. As seen from Tables 3 and 4, the increase in the dielectric constant of the permeating fluid has caused increase in the intrinsic permeability.

However, changes in the hydraulic conductivity against the flow of an organic fluid cannot be solely attributed to the reduction of the dielectric constant. Another important parameter in the interaction of clay and organic fluid is viscosity and molecular weight of the permeating fluid. Hydraulic conductivity of soil depends on several properties of clay, and these properties are not independent of each other, therefore, it is not readily possible to obtain a relationship between the hydraulic conductivity of soil and any of the aforementioned variables. Rather, a set of variables should be used to estimate the soil hydraulic conductivity.

Many authors recognize the influence of the liquid polarity or dielectric constant on the soil permeability. Fernandez and Quigley (1985) investigated the hydraulic conductivity of a clayey soil permeated by 9 different fluids. Results by Fernandez and Quigley (1985) show that with decreasing dielectric constant from 80 to 2, the hydraulic conductivity has increased from 10^{-8} to 10^{-3}. Highest permeability belongs to the fluid with the lowest dielectric constant. Fernandez and Quigley (1985) distinguished three regions on the hydraulic conductivity versus dielectric constant chart: a low permeability region for polar water of dielectric constant 80; an intermediate region for relatively polar alcohols ($\varepsilon = 20\text{-}35$); and a high permeability region for the nonpolar aromatics ($\varepsilon = 2$). Machado et al. (2016) presented the results of permeability tests conducted for different fluids (water, gasoline, commercial gasoline with 24% ethanol by volume, ethanol, diesel and carbon tetrachloride) in a variety of soils in order to derive a model to predict soil permeability of organic fluids based on soil and fluid properties. Figure 8 presents a comparison of the results presented by Fernandez and Quigley (1985) and Machado et al. (2016) and the current study.

It can be seen from Figure 8 that liquids with higher values of dielectric constant generally have a lower hydraulic conductivity and that the results of the current study are in agreement with the trend of results presented by Fernandez and Quigley (1985) and Machado et al. (2016). In comparison to Fernandez and Quigley (1985) and Machado et al. (2016) lower values of hydraulic conductivity have been obtained in the current study. This difference could be due to the presence of coarser grains in the soil samples of Machado et al. (2016) and Fernandez and Quigley (1985). These soil samples contained sand content in the range of 4 to 40 %.

CONCLUSIONS

The permeability in two types of clay with different plasticity, exposed to the flow of kerosene, diesel and ethanol was investigated by conducting modified constant-head permeability tests. The effects of clay plasticity and water content for a given compactive effort on clay permeability was investigated as well. The results derived from the current research suggest that:

1- As the water content of the samples increases, the permeability for all the fluids in both soils decreases, which points out the effect of soil structure on its permeability.

2- Organic fluids affect the behavior of clay soils through various mechanisms. The Gouy-Chapman double layer theory can’t fully describe the behavior change caused by the presence of organic fluids in clay soils, but provides a good interpretation of these changes in clay behavior.

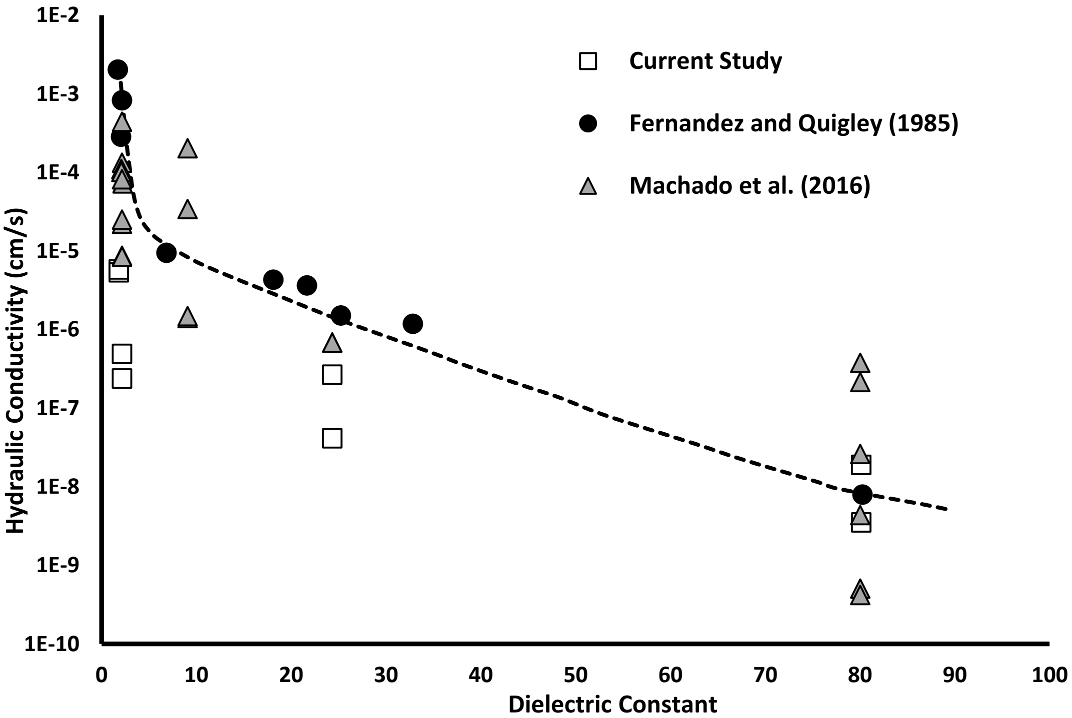

Fig. 8. Comparison of results of the current study with Fernandez and Quigley (1985) and Machado et al. (2016)

3- In the interaction between organic fluid and clay minerals, dielectric constant, kinematic viscosity and molecule size, control the behavior changes of clay in the presence of organic fluids. In this regard, reduction in the dielectric constant causes a reduction in the double layer thickness and therefore, results in changes in soil structure that increase the pore space for passage of the organic fluid, effectively increasing permeability. With a decrease in dielectric constant from 80.1 to 1.8, permeability is increased up to 1800 times. Maximum permeability belongs to the fluid with the lowest dielectric constant which is kerosene. In this condition, accordance of the double layer theory with the presented results is more obvious.

4- Viscosity of the fluid directly affects the permeability of clays as well. Low viscosity of the fluid facilitates its flow among the soil particles and increases its permeability.

5- Clay of higher Liquid Limit (LL) and Plastic Limit (PL) presented lower values of permeability against the passage of all the fluids in this experiment, but the decrease rate is different for different fluids. Increasing soil plasticity, causes an increase in the double layer thickness and results in the reduction of permeability.

6- In this experiment, permeability pertaining to kerosene is higher than that of the other three fluids. Therefore, kerosene is the most critical organic fluid among all the fluids in this research. If the soil permeability against kerosene satisfies the required criteria, the soil would definitely be suitable for the other three liquids.

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