Materials Research Proceedings 31 (2023) 504-513 https://doi.org/10.21741/9781644902592-52

One dimensional oedometer laboratory testing for expansive clay submerged with hydrocarbon fluids

Mousa Bani Baker[1,a,*], Adel Hanna[2,b], Maria Elektorowicz[2,c], Tahar Ayadat[3,d], Batool Al-Shorman[4,e] Raed Abendeh[1,f]

[1] Department of Civil and Infrastructure Engineering, Al-Zaytoonah University of Jordan, Amman, Jordan

[2] Department of Building, Civil and Environmental Engineering, Concordia University, Montreal, Canada

[3] Department of Civil Engineering, Prince Mohammad Bin Fahd University, Al-Khobar, Saudi Arabia.

[4] Department of Civil Engineering, Jordan University of Science and Technology, Irbid, Jordan

[a]m.banibaker@zuj.edu.jo, [b]hanna@civil.concordia.ca, [c]mariae@civil.concordia.ca, [d]tayadat@pmu.edu.sa, [e]byalsharman17@eng.just.edu.jo, [f]r.abendeh@zuj.edu.jo

Keywords: Hydrocarbon Liquids, Expansion, Fuels, Bentonite Clay, Swelling

Abstract. Landfills are currently one of the most effective ways to dispose of waste. Underground storage tanks (UGST) are also used to store hydrocarbon fluids that include different types of fuels. The bottom part of the landfills and UGST is critical. This liner material and its composition prevent heavy metals and leachate from infiltrating the groundwater table. Failure of this layer presumably causes most landfill failures. Bentonite clay is used to build such liner because of its properties including high cation exchange capacity and swelling index. The swelling of bentonite is sensitive to the type of liquid and load. It swells under low loads when submerged with water and to a lesser extent for ethanol. However, it undergoes consolidation when penetrated by biofuel. Test results indicate that bentonite undergoes swelling in water under high load (40 kPa) and consolidates for both alternative fuels (biofuel and ethanol). Under very high loads (100 kPa) bentonite consolidates for all kinds of liquids including water.

1. Introduction

Environmental engineering investigates hazardous waste contaminants, their pathways, transport, fates and disposition. It also explores ways of protecting groundwater, thereby protecting humans and the environment from hazardous wastes. Petroleum products, for instance, account for sixty-nine percent of soil contamination in Quebec [1].

Many liners are made watertight with clayey materials, such as sand-bentonite mixtures, that retain liquid and solid toxic wastes. Although many cases were reported for high leakage of bentonite liners [2], they remain in use. Over time, the principal function of liners is reduced and contaminants leak through them. As these toxic contaminants infiltrate the subsoil and the groundwater, they have serious ramifications on the stability of constructions and the safety of humans and animals.

Failure of landfill liners is a problematic issue for engineers and a costly one for governments, societies and the environment. To protect groundwater, the structure of a natural landfill liner needs to preserve its properties in harsh conditions for a sustained period. In addition, the type of disposal materials in landfills and storage tanks is changing and it is starting to include alternative fuels such as biofuel or ethanol fuel.

Containment barriers commonly use clays for their low permeability as liners in landfills and settling ponds. The properties of the fluid passing through liner material affect the liner's structure.

Thus, in the presence of alternative fuels, the dispersed orientation of clay particles alters the clay's permeability. The percolation of fluids might result in changes in the microstructure of clay fabrics. Leaking fluids destroy clay barriers, even though the liners are designed to prevent these infiltrations. In practice, when subjected to organic liquids, the clay liner leaks and destroys the clay microstructure, thereby increasing the permeability of the liner. [3] conducted a test on clayey soil permeated by liquid hydrocarbon. They reported a significant increase in hydraulic conductivity from 10-8 cm/s to 10-4 cm/s. [4] conducted research using a batch test to determine the adsorption coefficient of benzene, toluene, and 2-fluorotoluene on three soils. They found that hydrophobicity is an important factor in the relative adsorption of benzene and toluene in the materials investigated. They also supported the idea of incorporating organophilic clay to sand-bentonite mixtures to prevent compacted liner composed of these materials from desiccating when exposed to diesel fuel.

[5] investigated the correlation between permeability; microstructure and surface chemistry of geosynthetic clay liner (GCL) interacted with leachate. They concluded that when (GCL) containing sodium bentonite is in contact with fluids containing other cations, the latter exchanges with the sodium present between clay layers. This modification in clay surface chemistry changes the clay microstructure, therefore, changing the hydraulic conductivity. They also suggested that the cation selectivity of clay surface exchange sites depends also on the swelling and site occupation history of the clay. [6] conducted tests to evaluate how ammonium attenuates municipal solid waste (MSW) landfill leachate by adsorption into bentonite in a landfill liner. The bentonite adsorption capacity was measured in two types of laboratory tests. The first was a standardized test ASTM D4646 (1987) using bentonite in the dust state. The second test used compacted bentonite to reproduce actual landfill bottom conditions. Parameters were monitored such as; ammonium, heavy metals (Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn) and COD. Based on risk simulations, they concluded that the maximum capacity of adsorption in traditional landfill liners is exhausted in a few years (2-10 years). Thereafter, chemicals pass through the liner at the concentration found in the leachate.

As the chemical concentration increase, the Diffuse Double layer (DDL) shrinks in bentonite clay, then the bentonite clay swelling decreases as a result of flocculation of the clay particles. Some of the clay tests show that there are volume changes of bentonite clay when exposed to the NaCl, CaCl or KCl solutions and then reexposed it to the water, large volume decrease was investigated as exposure the specimens to the saturated solutions [7]. It is worth mentioning that there are some numerical modeling recently used to study the swelling of clay layers such as the model of Alonso which was proposed by [8] which showed a great economic value to study the parameters of soil's swelling.

Brazilian bentonite clay was prepared to utilize quaternary ammonium salt by [9] to study its effect on the orientation of petroleum-derived fuels, the results of swelling tests indicate increasing in basal spacing and formation of absorption bands associated with the CH2 and CH3 groups. Some techniques can be used to improve the role of the bentonite clay layer, one of them is increasing the adsorption capacity of the bentonite clay, [10] studied the effect of organic modification on the adsorption behavior of bentonite and found that the adsorption capacity increases with increase in the concentration of the organic contaminant up to 500 mg/l for the modified clays. The Functionalized Bentonite clay-sPEEK was used as composite membranes for direct methanol fuel cells and found that these composite membranes exhibit lower methanol permeability in comparison to pristine membrane. [11] found that there is a long term effect of water chemistry (Distilled water and synthetic water) on the swelling pressure of a bentonite-based material experimentally, their research study results show that the swelling pressure decrease for the samples specially for the sample saturated with synthetic water. The hydraulic performance of organo clay enhanced sand bentonite as the secondary liner was investigated by [12] and the results indicate that the permeability coefficient was assessed both for loading and unloading cases. A decrease in the coefficient of permeability with diesel by an order of two was observed on increasing the organo clay content from 0 to 10% in SB mix, whereas a reverse trend was noted with water as permeant. The adsorption of ionic liquids can also change the aggregation behaviors of the bentonite particles by which the "house of cards" structures in the bentonite scuttled are improved or destroyed [13]. The compacted bentonite swelling behavior can be enhanced due to the aging effect, the axial strain and the swelling pressure decreased as the aging time increased for samples prepared at both dry and wet sides [14]. The technological voids also affect the compacted bentonite by decreasing the swelling pressure anisotropy with its increase [15].

[16] studied the volume changes of bentonite which was exposed to NaCl, KCl or CaCl solutions and re-exposed to water. Presentation of specimens to saturated solutions resulted in great volume decreases. [17] investigated the effect of water chemistry on the swelling pressure and found that there is no clear effect of it in the short term (100h) due to the high dry density of the bentonite. On the other side, considering the long term period (700 days) the swelling pressure decreases with a non-negligible ratio (9%). Other studies such as [18], which are concerned with exploring the permeability variation of the Sabkha soil during the distilled water leaching and found that there is a direct relation between the rate of ion dissolution and soil permeability. The raise of soil permeability is because of the dissolution of Cl^{-1}, $SO4^{-2}$ and Na^{+1} ions into the Sabkha soil layer.

2. Materials

2.1 Bentonite clay

Montmorillonite clay (bentonite) was used which is some commercial powdered rich in Na-montmorillonite. It was used in this study for its high swelling capacity, high cation exchange capacity (CEC) and high surface area [19]. The isomorphous substitution occurs mainly in the alumina sheet of the montmorillonite minerals, with magnesium or iron substituting for aluminium in the dioctahedral minerals. Since there are no potassium ions to bond the layers together, therefore, water can enter easily between layers causing swelling.

The used bentonite possesses the following properties: Liquid limit of 440%, Plastic limit of 65%, Shrinkage limit of 8.2%, Free swelling index is 487.5%, the optimum moisture content is 25.6%, CEC (meq/100g) equals to 95, pH in water is 9.96, size passing sieve number 325 (D=44 micrometre), the specific surface area is 600 m^2/g. The mineralogical composition of natural bentonite (X-Ray Analysis) is given in Table 1.

Table 1. *Mineralogical composition of natural bentonite (X-Ray Analysis)*

Element	Percentage (by weight)
Montmorillonite	85
Quartz	5
Feldspars	2
Cristobalite	0.35
Illite	2
Calcium and Gypsum	1
LOI	4.65

2.2 Liquids

Water was used as the permeate reference, ethanol (100%), and biofuel was used as permeates with dynamic viscosities of 0.89, 1.071 and 61 cP respectively, as measured in the laboratory at room temperature.

3. Methodology

Bentonite is the material that undergoes swelling. It is, therefore, responsible for the swelling of the liner composition when permeated with different liquids. The one-dimensional free swelling test was conducted as per the ASTM D 2435-90 using the fixed-ring oedometer test as shown in figure 1. After preparing the compacted bentonite at the optimum moisture content of 25.5% by weight using the proctor, undisturbed samples were taken from the compacted bentonite using consolidation ring cells with sharp edges from one side, a 60 mm diameter and a 20 mm height. Vertical confining pressures of 7 kPa, 40 kPa, and 100 kPa were applied on the specimen with different liquids (water, ethanol, and biofuel). Three samples were tested using the same kind of liquid and load (a total of 27 samples were prepared).

At first, the load was applied on the samples until consolidation ceased and the consecutive readings were close. Then, the liquid was added to the cell. Deformations started in the sample and displacements were obtained using a linear variable differential transformer (LVDT). It was connected to the computer and three cells to perform three simultaneous tests using the same pressure and the same liquid in each set of samples. Each time, new samples were prepared and tested using the desired liquid. Results were compiled in an Excel datasheet and an average of the three readings was calculated accordingly. The percent swell was defined as the ratio of a percent increase in the thickness to the actual thickness of the specimen.

Fig. 1. *Test setup.*

4. Test Results

Figures 2, 3, and 4 show the swelling/compression of bentonite under low pressure of 7 kPa, medium pressure of 40 kPa, and high pressure of 100 kPa using water, biofuel, and ethanol fuel. The highest swelling of bentonite during 4 hours was achieved when water was used for both loads 7 kPa and 40 kPa. Swelling of bentonite submerged with ethanol was also observed at the lowest load (7 kPa). The highest load (100 kPa) prevented the swelling of the bentonite samples, a compression of bentonite for all liquids was observed. The bentonite submerged with ethanol under 100 kPa expressed the highest compression. Results indicate that for all types of pressures biofuel causes the bentonite clay to compress during loading.

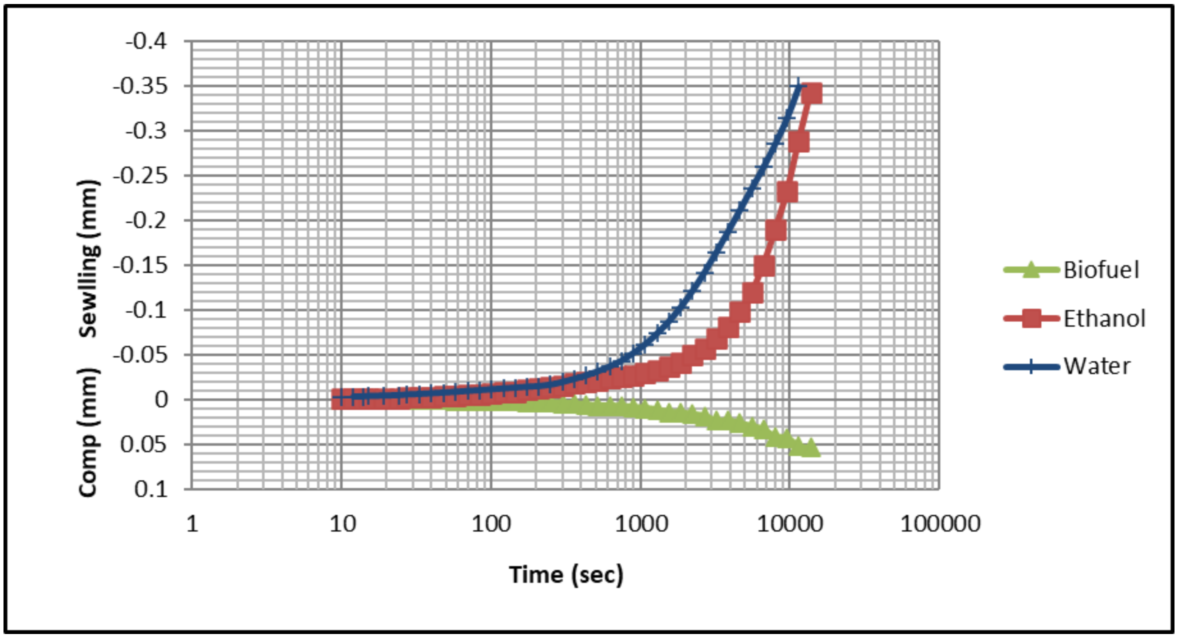

Fig.2. Swelling of bentonite under a pressure of 7 kPa using different liquids.

5. Analysis and Discussions

The results showed that bentonite swelling behaviour depends on the type of permeate as well as pressure. Many physic-chemical phenomena can be responsible for such behavior. Under low pressure (7 kPa), bentonite swelled the most in the contact with water. The swelling was also observed in the case of ethanol. However, when submerged by biofuel, the bentonite behaved differently. It underwent compression (consolidation). Under an increased load of 40 kPa, only the samples tested by water swelled. The ones tested by biofuel and ethanol underwent compression (consolidation) and biofuel had a higher rate of consolidation than ethanol. When applying a high pressure of 100 kPa, all samples are compressed. The most consolidation occurred in samples interacting with ethanol, then biofuel, leaving the samples that came in contact with water with the least compression. Very high pressure prevents the expansion of montmorillonite parts for all types of liquids.

The bentonite material submerged with ethanol (polar liquid) expressed the highest sensitivity to compression. Montmorillonite (the major component of bentonite) consists of tetrahedral and octahedral sheets. The space between these sheets can be expanded up to 10 water molecules thickness (10-9 m). In no load or low load conditions, the ethanol can provoke a similar expansion like water. However, the process of swelling, in this case, is lower.

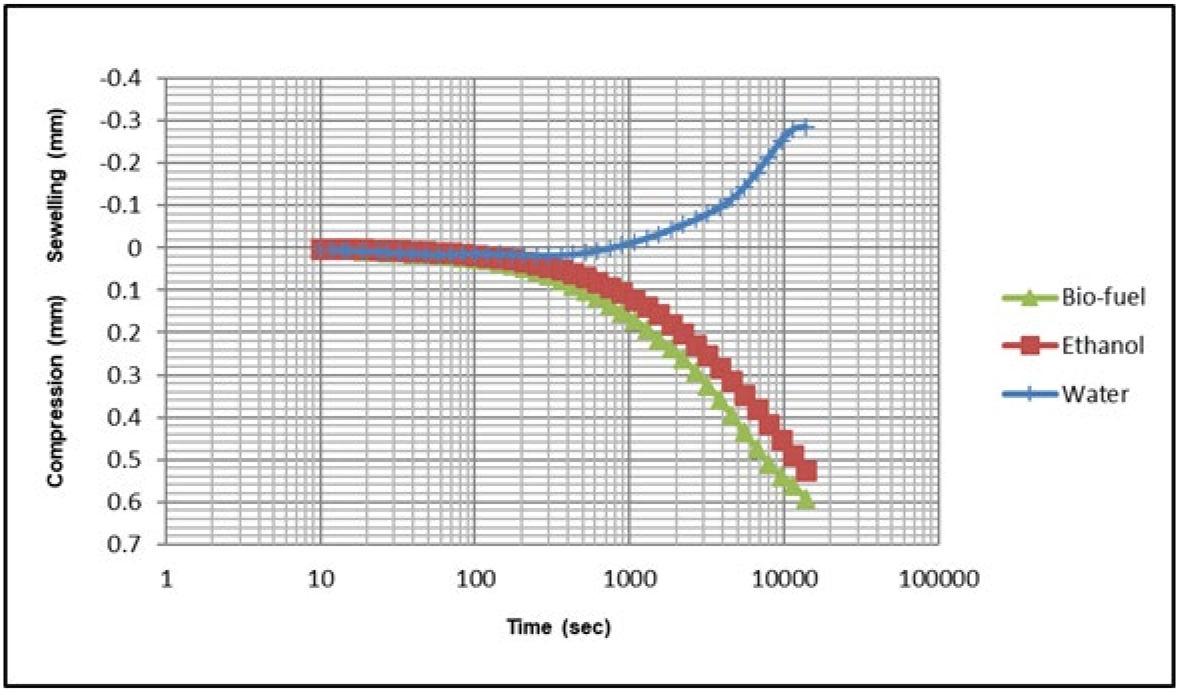

Fig.3. *Swelling of bentonite under a pressure of 40 kPa using different liquids.*

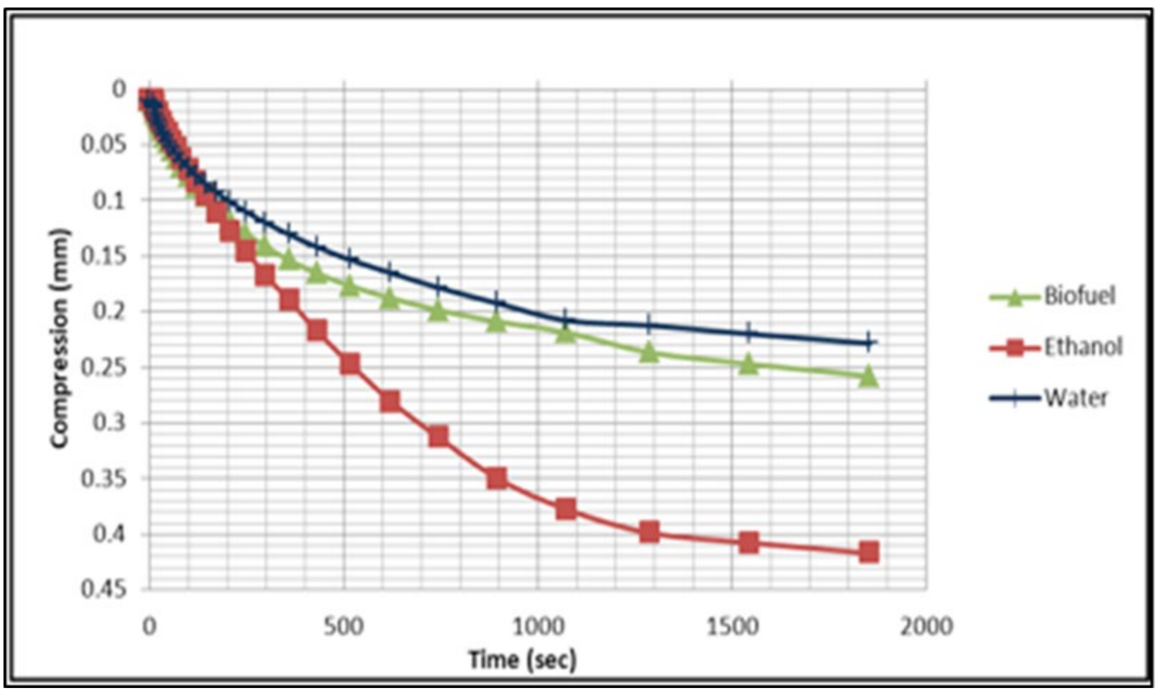

Fig.4. *Swelling of bentonite under a pressure of 100 kPa using different liquids.*

The bentonite swelling occurs when polar water molecules enter between the adjacent platelets and separate them so that van der Waals forces are no longer sufficient to keep the montmorillonite particles intact. This, in turn, causes the swelling of bentonite, decreasing pore voids, thus, reducing the hydraulic conductivity.

As water enters between the particles, it may displace the high-valent cations, which help the clay particles to the cluster. In turn, this may create repulsive forces between clay particles, causing the dispersion and additional swelling of bentonite and resulting in the swelling of the whole liner mixture.

The surface capillary forces, which are involved in attracting the soil particles decrease when an additional volume of water fills the pores (e.g. during saturation) and the water film surrounding the grain increases. Subsequently, clay can expand.

Biofuel operates differently, it acts oppositely, it prevents the expansion of clay, it does not permit for the tetrahedral and octahedral sheet to be expanded; montmorillonite submerged with biofuel undergoes compression with time in all tested conditions. Under the load condition, liquid films around the grains undergo deformation. This deformation is enhanced when biofuel is present. Biofuel causes the montmorillonite particles adhesion and the formation of flocs, consequently, it makes the bentonite liner coagulate and consolidate increasing pore voids, leading to fracture or tunnelling phenomena.

Biofuel has a convex meniscus (contrary to water concave meniscus) which creates higher surface tension forces (strong attractive forces between the grains) with the increase of additional biofuel liquid in pore voids. Subsequently, oil-particle flocs can be created. An extension of this phenomenon depends on temperature due to biofuel viscosity dependence.

Some studies indicated that polar hydrocarbons are required for flocculation to occur. However, experimental tests in this study indicate that both polar (ethanol fuel) and non-polar (biofuel) liquids cause the flocculation of clay fine particles. Furthermore, clay particles normally have negative charges in a stable clay solution then the repulsive forces are predominant. However, when biofuel is in contact with negatively charged bentonite in the liner's mixture, a thin layer is formed around the clay particles making them isolated, reducing the repulsive forces and allowing particles to form flocs. Therefore, two phenomena are present in this research: one for polar organic liquids (ethanol) and the other for non-polar (biofuel) forming of oil film filling the pores.

The results permitted us to conclude that the hydraulic conductivity of liners can increase under a load of waste after the percolation of alternative fuel leachates. This increase is due to many factors about the changes in the structure and composition of sand-bentonite mixtures. For instance, erosion of fine particles, as liquids infiltrate through the liner, affects the hydraulic conductivity. Also, the amount of fine particles flushed out of the mixtures increased with higher pressures, during the leaching of liquids, especially for water.

The fact that particles wash out of a liner exposed to high hydraulic pressure had already been observed. While water causes the highest rate of erosion due to its high solubility, its low density and viscosity enhance the erodibility of fine clay particles during the leaching process. The washing out of clay particles from the soil matrix increases with hydraulic pressure. This phenomenon, known as suffusion or piping, increases the final hydraulic conductivity of the liner due to the creation of larger pores (Kaoser et al., 2006).

In contact with the liner, water causes swelling of the liner's mixture through the swelling of bentonite clay in the liner composition and due to the dispersion of negatively charged clay particles. As water enters between the particles, it may displace the high-valent cations, which help the clay particles to the cluster. In turn, this may create repulsive forces between clay particles, causing the dispersion and additional swelling of bentonite and resulting in the swelling of the whole liner mixture.

Biofuel has a convex meniscus (contrary to water concave meniscus) which creates higher surface tension forces (strong attractive forces between the grains) with the increase of additional biofuel liquid in pore voids. Subsequently, oil-particle flocs can be created. An extension of this phenomenon depends on temperature due to biofuel viscosity dependence.

It was also observed that biofuel caused more cracks and higher surface fractures when percolating liners compared to ethanol fuel and water. In this study, biofuel grain size distribution and the mean size of particles increased drastically when liners were percolated by alternative fuels (ethanol or biofuel). This is due to the coagulation of fine particles in their interaction with fuel as they adhere together forming larger oil-clay clusters that do not separate in emulsion used for particle size analysis. However, many flocculated and single mineral fine particles remained, as shown in the particle size analysis.

Clay-ethanol flocs were also observed in the case of using ethanol as a permeate to percolate through the liner sand-bentonite mixtures. In the lab experiments, the clay-fuel floc aggregates were formed, which was also found by other researchers who observed clay-oil floc aggregates under an optical microscope and concluded that many flocculated single mineral fine particles were still present. A variety of crude oils ranging from light crude oils to heavy crude oils were, therefore, able to interact with micron-sized mineral fines to form ―clay-oil flocs‖ consisting of solids-stabilized oil-in-water emulsions.

In addition, it is known that the adsorption behaviour appears to be inversely proportional to the solubility of the compound and directly proportional to the percentage of organic matter in the mixture. Biofuel, the least soluble of the three liquids used (water, ethanol, biofuel), appears to have a propensity to be adsorbed in the bentonite clay particles. Thus, bigger particles form through the coagulation of fine particles, causing fractures in the sand-bentonite mixture. This might explain the higher surface fracture in the liner mixtures percolated by biofuel. It ia appears that polar hydrocarbons are required for flocculation to occur. However, experimental tests in this study indicate that both polar (ethanol fuel) and non-polar (biofuel) liquids cause the flocculation of clay fine particles.

Clay particles normally have negative charges, and in a stable clay solution, repulsive forces predominate. But when biofuel is in contact with negatively charged bentonite in the liner's mixture, a thin layer forms around the clay particles making them isolated, reducing the repulsive forces and allowing particles to form flocs. Distinctive phenomena can be related to polar organic liquids (ethanol) and non-polar liquids (biofuel) forming oil films [20, 21].
During lab experiments, biofuel flow through the 100% sand liner took more time to reach the column outlet. High biofuel viscosity creates higher friction forces between the liner and the fluid.

Conclusions

The swelling of bentonite is sensitive to the type of liquid and load. It swells under low loads (e.g. 7 kPa) when submerged with water and to a lesser extent for ethanol. However, in the case of submerging the bentonite by biofuel, it undergoes consolidation. For higher load (e.g. 40 kPa) bentonite undergoes swelling in water, however, it consolidates for both alternative fuels (biofuel and ethanol). Under very high loads (e.g. 100 kPa) bentonite consolidates for all kinds of liquids including water.

References

[1] Enviroment Québec, 1994, Dix ans des restauration des terrains contamines‖, Enviro.doc EN950098.

[2] ASTM D2435-90 Standard test method for one-dimensional consolidation properties of soils. Chapuis R. P., (2002), The 2000 R.M. Hardy Lecture: Full-scale hydraulic performance of soil-bentonite and compacted clay liners. Canadian Geotech. J. 39: 417-439. https://doi.org/10.1139/t01-092

[3] Fernandez F., and Quigley R. M., (1985), Hydraulic conductivity of natural clays permeated with simple liquid hydrocarbons‖. Canadian Geotechnical Journal. 22: 205-214. https://doi.org/10.1139/t85-028

[4] Headley V., Boldt-Leppin E.J., Haug D., and Peng J., (2001), Determination of diffusion and adsorption coefficients for volatile organics in an organophilic clay-sand-bentonite liner‖ Canadian Geotechnical Journal. 38: 809-817. https://doi.org/10.1139/t01-017

[5] Guyonnet D., Gaucher E., Gaboriau H., Pons C.-H., Clinard C., Norotte V., and Didier G., (2005), Geosynthetic clay liner interaction with leachate: correlation between permeability, microstructure, and surface chemistry‖. Journal of Geotechnical and Geoenvironmental Engineering ASCE June 2005: 740-749. https://doi.org/10.1061/(ASCE)1090-0241(2005)131:6(740)

[6] Pivato A. and Raga R., (2005), Tests for the evaluation of ammonium 163 attenuation in MSW landfill leachate by adsorption into bentonite in a landfill liner‖. Waste Management 26: 123-132. https://doi.org/10.1016/j.wasman.2005.03.009

[7] Arasan S., (2010), Effect of Chemicals on Geotechnical Properties of Clay Liners: A Review‖. Research Journal of Applied Sciences, Engineering and Technology 2(8): 765-775.

[8] Aissa Mamouns, S.M., and Bekkouche, A., (2011). Modeling of the Behavior of Expansive Soils. Jordan Journal of Civil Engineering, Volume 5, No. 1, 2011.

[9] Bertagnolli C., and Carlos da Silva M. G., (2012), Characterization of Brazilian Bentonite Organoclays as Sorbents of Petroleum-derived Fuels‖. Materials Research; 15(2): 253-259. https://doi.org/10.1590/S1516-14392012005000017

[10] Sreedharan V., and. Sivapullaiah P. V., (2012), Effect of Organic Modification on Adsorption Behaviour of Bentonite‖. Indian Geotech J 42(3):161-168. https://doi.org/10.1007/s40098-012-0014-6

[11] Wang Q. et al., (2014), Long-term effect of water chemistry on the swelling pressure of a bentonite-based material. Applied Clay Science 87: 157-162. https://doi.org/10.1016/j.clay.2013.10.025

[12] Sreedharan V., and Sivapullaiah P. V., (2017), Hydraulic Performance of Organo Clay Enhanced Sand Bentonite as Secondary Liner. Indian Geotech J. https://doi.org/10.1007/s40098-017-0232-z

[13] Ren Y. et al., (2019), Adsorption of imidazolium-based ionic liquid on sodium bentonite and its effects on rheological and swelling behaviors. Applied Clay Science 182: 105248. https://doi.org/10.1016/j.clay.2019.105248

[14] Estabragh A.R., Amini M., and Soltanian M.R., (2019), Aging effects on the swelling behavior of compacted bentonite. Bulletin of Engineering Geology and the Environment. https://doi.org/10.1007/s10064-019-01708-7

[15] Zeng, Z., Cui, Y.-J., Zhang, F., Conil, N., & Talandier, J. (2020). Effect of technological voids on the swelling behaviour of compacted bentonite/claystone mixture. Canadian Geotechnical Journal. 9. https://doi.org/10.1139/cgj-2019-0339

[16] Di Maio, C., (1996). Exposure of bentonite to salt solution: Osmotic and mechanical effects. Geotechnique, 46(4): 495-707. https://doi.org/10.1680/geot.1996.46.4.695

[17] Sasikala, S., Meenakshi, S., Bhat, S.D., Sahu, A.K. (2014). Functionalized Bentonite clay-sPEEK based composite membranes for direct methanol fuel cells, Electrochimica Acta. https://doi.org/10.1016/j.electacta.2014.04.180

[18] Fahad A.A., (2020), Variation of Sabkha Soil Permeability Associated with Ions' Dissolution during Distilled Water Leaching. Jordan Journal of Civil Engineering, Volume 14, No. 4, 2020.

[19] Bani Baker, M., Abendeh, R., Sharo, A., Hanna, A. Stabilization of Sandy Soils by Bentonite Clay Slurry at Laboratory Bench and Pilot Scales. Coatings/Functional Building Materials and Pavements Coatings, 2022, 12 (12). https://doi.org/10.3390/coatings12121922

[20] Bani Baker, M., Elektorowicz ,M., Hanna, A., (2018), Elctrokienetic Nondestructive In-situ Technique for Rehabilitation of Liners Damaged by Fuels, Journal of Hazardous Materials. Vol. 359. Pp. 510-515. https://doi.org/10.1016/j.jhazmat.2018.07.113

[21] Sharo, A.A., Khasawneh, M.A., Bani Baker, M., Al Tarawneh, D.M. Sonicated waves procedure effect on stabilizing expansive soil by nano-clay: Treat with cause. Frontiers in Built Environment, 2022, 8, 975993 https://doi.org/10.3389/fbuil.2022.975993