Investigating the influence of alternative fuels on the properties of sand-bentonite liners

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Abstract. Landfills are one of the most effective ways utilized to dispose the wastes and usable up today, most of their failures were suspected due to the failure of their protecting natural liners. Landfill liners failure is one of the most problematic issues that face engineers and expose governments, societies, and the environment to high costs. Protection of groundwater requires that natural landfill liner structure be able to reserve its properties in harsh conditions and over a long period. However, the composition of disposed of residuals might change in the following contemporary trends and it could contain alternative fuels and their impact on natural liners requires further investigations. This paper focuses on the changes in sand-bentonite liner due to the interaction and infiltration of hydrocarbon liquids (alternative fuels); biofuel and ethanol-fuel. In this investigation, an experimental program was carried out to examine the influence of chemical properties of alternative fuels on the hydraulic conductivity, erodibility, swelling potential and shrinkage behaviour of natural liners. Series of laboratory tests were conducted in 20 cm-high PVC columns to investigate the hydraulic conductivity of the liners, swelling behaviour of bentonite when subjected to water, biofuel or ethanol-fuel was assessed by oedometer tests, and shrinking behaviour and cracking patterns of samples taken from the PVC columns were examined employing digital photo analysis. The results can be useful in designing liners, barriers as well in assessing the behaviour of clayey soil in case of accidental spills or intentional discharges.

1. Introduction

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 [1], they remain in use. In Quebec, for example, 450 projects built since 1980 involved more than 1000 liners. 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. This undoubtedly leads to the elimination of the principal function of the liners over a while, leading to the migration of contaminants through these liners. Tunnelling and the migration of fine particles throughout the liner mixture might be one of the reasons for increasing the hydraulic conductivity. Despite the existing municipal solid waste (MSW) landfill disposal practices, leaching still occurs, leading to heavy metal migration towards the groundwater, whether the liners are built of synthetic materials or sand and bentonite mixtures [2].

Failure of landfill liners is a challenging issue for engineers and a costly one for governments, societies and the environment. The increase in hydraulic conductivity may lead to the failure of the liner. The infiltration of liquid hydrocarbon has a significant increase in hydraulic conductivity from 10-8 cm/s to 10-4 cm/s [3]. To protect groundwater, the structure of a natural landfill liner needs to preserve its properties in harsh conditions for a sustained time. Also, the type of disposing of materials in landfills and storage tanks is changing and it is starting to include alternative fuels such as biofuel or ethanol fuel. [4], studied the function of liquid chemistry on the compressibility and swell characteristics. Two types of bentonites are mixed with pure silica sand and the results indicate that swell potential, swell pressure, swell time, and volume compressibility decreases with the increase of chemical concentrations of the sand-bentonite mixture. As research shows that the coefficient of hydraulic conductivity changes when soil barriers are infiltrated with organic fluids [5]. Various problems appeared in several case histories such as extreme leakage of a stable soil–bentonite mix, internal erosion of a natural clay liner through an insufficient sand filter, washing of bentonite in a soil–bentonite mix, and effects of freezing on the total leakage of a clay liner, [1] and these leading to misuse of the landfill due to decreasing of its linear function. [6] results showed that hydraulic conductivity of bentonite saturated with gasoline was two orders of magnitude 10-5 cm/s higher than the requirement for earthen liner10^{-7} cm/s. Soil/clay liner was suspected to deteriorate when exposed to organic fluids, resulting in a major increase in hydraulic conductivity. [7] observed that compaction conditions of the sand-bentonite liners affect values of the coefficient of permeability of the liner (k). The compaction conditions are resultant in moisture content slightly wet of optimum lead to the smallest values of (k). The particle size of the sand may affect the behaviour of the sand-bentonite liners, [8] found that unheeding of the sand particle size, the sand which mixed with bentonite content less than 20 % showed a general lack of considerable swelling, for the same bentonite content, mixtures with fine sand displayed relatively higher swelling pressure and lower hydraulic conductivity. [9] studied the strength and hydraulic conductivity of the sand-bentonite linear and found that the safest liner carries an 8% of bentonite with sand mixture had a hydraulic conductivity below 1×10^{-7} cm/s. At times researchers typically left out volumetric shrinkage in their researches, but field studies have shown that desiccation can induce harsh cracking of unprotected soil barriers, [10].

Therefore, in this research, the coefficient of hydraulic conductivity is used as an indicator of liner performance (including failure, disintegration, shrinking, fracture, etc). Hence, this paper focuses on the changes within the sand-bentonite liner structure due to the infiltration of biofuel and ethanol fuel.

2. Methodology

The research examines the infiltration of leachate through sand-bentonite barriers. Three liquids were used as leachate: first, water, as a control; then, ethanol fuel, and biofuel. The sand-bentonite barriers (i.e. liners) were composed of sand and bentonite in proportions varying by weight and mixed using the optimum moisture content. Then, each liquid was leached through each liner under specific pressure. The research consisted of laboratory work to observe the infiltration of the three different liquids through the sand-bentonite liners using column leaching test, under different pressures, to test hydraulic conductivity, erodibility, particle size distribution, surface fracture, and changes of some geotechnical properties of the clay materials when exposed to water and alternative fuels. Three different mixtures (100% sand, 95% sand: 5% bentonite, and 90% sand: 10% bentonite) were prepared and tested. First of all, preparing a set of samples (18 liners) tested under 40 kPa and 100 kPa to study the hydraulic conductivity and particle size distribution changes before and after the infiltration of the liquid through the liners. 6 liners were studied using biofuel as permeate and the other 6 ones were studied using ethanol fluid and the last 6 infiltrations by water.

Then set of tests were done as the water was introduced from the bottom part until saturation is achieved, then water was introduced from the top part under the pressure of 20 kPa. The same test will be done for unsaturated samples. Four samples will be taken from different depths 2, 8, 12, and 18 cm for analysis. Digital images will be taken from the top part, middle parts (after cutting the liner into two parts using a guitar wire) and the bottom part; hydraulic conductivities will be tested along the liner depths after the percolation using tubes.

2.1. Preparation of Materials

Two types of minerals were used: silica sand and bentonite clay (Dried sand from Quebec was sieved and sand passed mesh # 40 was used, as per the ASTM standards, ASTM E 11-70 -1995). Bentonite commercial powdered rich in Na-montmorillonite from Houston TX was used in this study for its high swelling capacity, high cation exchange capacity (CEC) and high surface area. The properties of sand were: CEC (meq/100g) =3 as measured in the lab, pH in water (measured in lab) =8.52, size (mesh# =40, D= 0.42 mm), specific surface area (m^2/g, [11]) = 0.1, and percentage of organic matter =0.07 (as measured in the laboratory using ASTM D 2974). The properties of bentonite were: 95, 9.96, 325 (D= 0.044mm), 600, and 4.1, respectively. The mineralogical composition of natural bentonite (X-Ray Analysis: [11]) were: 85% Montmorillonite, 5% Quartz, 2% Feldspars, 0.35% Cristobalite, 2% Illite, and 1% Calcium and Gypsum. Liquids: Water (tap water-Montreal) was used as the permeate reference for all tests, 100% ethanol (source: Quebec) and biofuel (originated from corn oil- Quebec) were used as permeates as well with dynamic viscosities of 0.89, 1.071 and 61 cP respectively, as measured in the laboratory at room temperature. The properties of ethanol were:density (g/cm^3) =0.789,Solubility in water= miscible, and surface tension (mN/m) at 20º C = 22.39. The properties of biofuel were: 0.992, immiscible, and 20-25, respectively.

Materials were blended in different proportions by weight. Then water was added to soil mixtures to reach the optimum moisture content. The optimum moisture contents are 20%, 19%, and 10% for (85%:5%), (90%:10%), and (95%:5%); (sand:bentonite), respectively. Stopping at a maximum of 10% bentonite was deemed sufficient as it satisfies the hydraulic conductivity required to construct liners. A rammer similar to the standard proctor test was used to compact the liner samples. The soil was compacted in the PVC column leaching mold in three equal layers. It delivered 19 blows to each layer as per the ASTM D 698-78 to obtain the equivalent compaction energy due to the change of column diameter (the diameter of the column is less than that in the standard proctor used to find the optimum moisture content). Constructing liners of 100% sand is impractical: it was only used here for comparison purposes. Since sand alone is hard to compact, it was fully saturated to be placed later in the column with compaction by a metallic rod. Liners were tested for hydraulic conductivity for leachate infiltrating downward, representing most cases in the field. Samples were collected from different depths along the liner samples to test for grain size distribution.

2.2. Experimental Method

Column leaching test simulates a clay liner in a landfill or an underground storage tank. Liner materials were prepared and placed in the column in two parts. Each layer was compacted for optimum moisture content using the standard proctor test procedure. Then, the column was leached using different mixtures, liquids and pressure values.

To perform the test, a PVC wall permeameter was used. A PVC column consists of two equal parts; each has a height of 98.00 mm and an inside diameter of 96.00 mm used to contain the liner samples during the experiments. In addition, two PVC heavy-duty flanges were used and held to continuously threaded stainless steel rods and the PVC cell-wall cylinder, as explained in Fig 1. Both ends of the column were equipped with perforated stainless steel plates to support the liner sample and facilitate the flow. Another stainless steel mesh was placed between the stainless steel plate and the liner to confine the liner and prohibit it from swelling. A filter paper (opening diameter of 110 µm) was placed between the mesh and the liner, facing the liner sample to prevent migration of fine particles. The stainless steel plates rest on another permeameter composed of two parts: 1. A bottom part with the same characteristics and a height of 40 mm includes cylinder support open from both sides with openings on the wall bottom sides to facilitate the flow of fluids, support the bottom plate and consequently support the liner. 2. A top part consists of a PVC permeameter with the same diameter and a height of 40 mm. These two parts can be fixed to the rest of the column using O-N Buna rubber rings. The top plate consists of thick PVC heavy-duty flanges with a valve on the top to control the flowing fluid. Another opening, which can be blocked, allows air to be released when starting the flow (bleeding valve). The bottom plate is equipped with an elbow to be used as an outlet opening.

The upper and bottom PVC plates can be fixed to the rest of the column using the rubber rings and threaded stainless steel rods by assembling all parts of the column and pressuring with a hydraulic pressure machine. This prevents leakage from the column.

Three typical cells were manufactured and connected with a pressure vessel, designed using two PVC heavy-duty flanges and a PVC pipe (20.3 cm inside diameter and 50 cm height). The flanges and the pipe were connected by threaded stainless steel rods and rubber rings to be placed between the plates, the top and the bottom parts of the pipe to prevent the fluid from leaking. The top part of the vessel has a pressure regulator and is connected to a pressure cylinder and a safety valve. An inlet opening-ball valve, which can be blocked, is used to fill the liquid in the vessel. The bottom part is connected to the cell via the outlet (valve). The pressure vessel is connected to a transparent tube, from bottom to top, that measures the elevation of liquid in the vessel and displays it during experiments. Fig. 2 shows the experimental setup of the column leaching test.

3. Test Results and Analysis

3.1. Coefficient of Hydraulic Conductivity

The liners of pure sand and sand-bentonite composition with different proportions which infiltration by water, biofuel, and ethanol-fuel; 18 tests were done and the following results were found (Table 1).

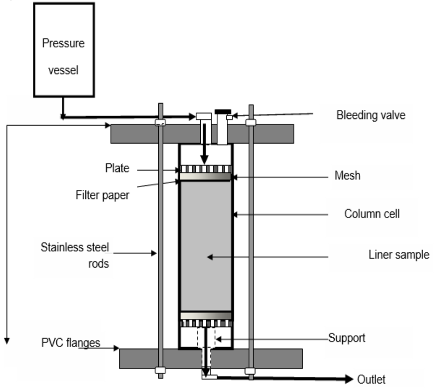


Fig.1. *Setup for column leaching test*

Fig.2. *Column leaching test cells with pressure vessel setup*

The coefficient of permeability or hydraulic conductivity is defined as a constant proportional to the ease with which a fluid passes through a porous medium. The constant head method was used to test for hydraulic conductivity according to ASTM D 2434-68.

Table 1 shows the coefficient of hydraulic conductivity for different liners saturated downward (with the same kind of permeate used as leachate) then leached by water, ethanol and biofuel under pressures of 40 kPa and 100 kPa.

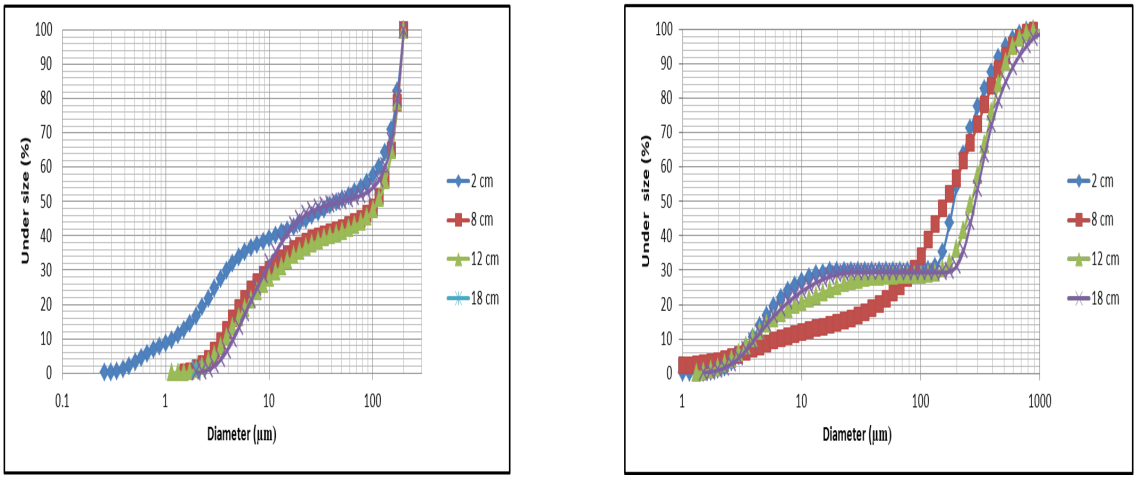
Table 1. *Coefficient of hydraulic conductivity (k, cm/s) for the liners infiltrated by water, ethanol and biofuel under pressures of 40 kPa and 100 kPa*

Liquid %Bentonie	Water	Ethanol	Biofuel	Pressure (kPa)
0	6.35×10^{-05}	5.72×10^{-05}	1.69×10^{-05}	40
5	1.57×10^{-07}	2.42×10^{-06}	5.10×10^{-06}	
10	1.37×10^{-11}	1.53×10^{-11}	3.12×10^{-08}	
0	7.20×10^{-05}	1.17×10^{-04}	2.11×10^{-05}	100
5	2.30×10^{-07}	8.13×10^{-06}	4.52×10^{-06}	
10	1.90×10^{-11}	1.39×10^{-11}	2.73×10^{-08}	

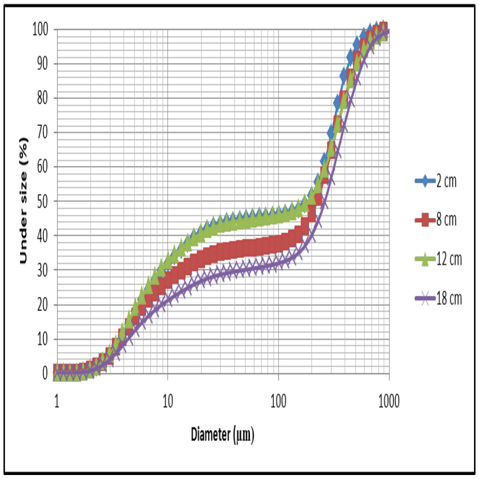
3.2. Grain Size Distribution

After dismantling the column, disturbed samples were taken from different depths to be tested for particle size distribution using the HORIBA LA-950V2 laser scattering particle size distribution analyzer. Four samples from each liner were tested to evaluate the changes in particle size distribution in the liner under different pressures infiltrated by water, ethanol and biofuel as illustrated in the following figure (Fig. 3).

Fig. 3 showed the variation of the particle size distribution of a liner composed of 90% sand and 10% bentonite.



a. b.



c.

Fig. 3. *Particle size distribution for the liner of 90% sand and 10% bentonite permeated by water, biofuel and ethanol under 40 kPa (a.water, b. biofuel, and c. ethanol)*

Under 40 kPa, the particle size distribution for a liner infiltrated by water ranged from 0.1 to 200 μm at different depths (2, 8, 12 and 18 cm). For a liner infiltrated by biofuel or ethanol, the particle size distribution ranged from 1 to 1000 μm. Particle size distribution curves along the liner's depths behaved in the same manner under 100 kPa. Black spots were detected at 10 to 14 cm down from the liner surface. This could be attributed to the presence of anoxic microorganisms which used ethanol as a carbon source causing ethanol biodegradation, and therefore, no free droplets of an organic liquid (ethanol) were detected by the laser particle size analyzer. The maximum particle size detected was 250 μm. It could be concluded that fine particles in the sand-bentonite mixture were highly susceptible to agglomeration when interacting with fuels. As a result, the sand grains and bentonite clay particles formed bigger flocs which lead to an increase in the liner hydraulic conductivity, an increase in the rate of infiltration, and thus a remarkable fracture along with the liner's depth as shown in Fig 4. The lab test demonstrated changes in liner behaviour depending on bentonite content and the type of permeate. Several Physico-chemical phenomena can be responsible for such behaviour. The results permitted us to conclude that the hydraulic conductivity of liners can increase under a load of waste after the infiltration of alternative fuel leachates.

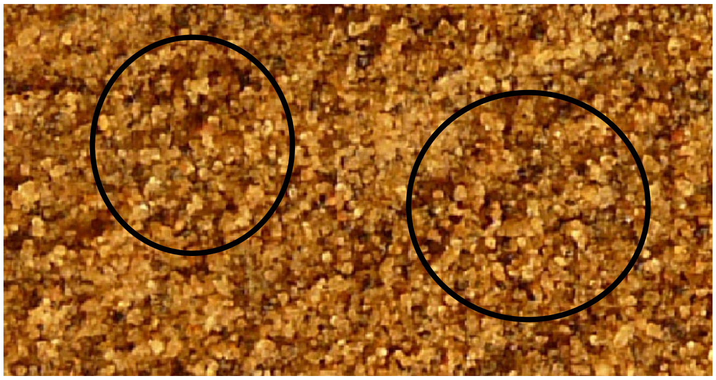


Fig 4. *Digital image for the upper surface of a 95% sand and 5% bentonite liner leached by biofuel under a pressure of 100 kPa (cracks are showed in circles)*

This increase is due to many factors pertaining to 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 [12]. While water causes the highest rate of erosion of soil due to its high solubility, besides, water has low density and viscosity which 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 hydraulic conductivity of the liner due to the creation of larger pores [13]. In contact with the liner, water causes swelling of the liner's mixture through the swelling of bentonite clay [14] 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 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 (Fig. 5), which are involved in attracting the soil particles decrease when the additional volume of water fills the pores (e.g. during saturation) and the water film surrounds the grain increases leading to the point of rupture of the liquid bridge due to the increase of its volume. Subsequently, clay can expand. The concave meniscus of water creates repulsive forces between clay particles and leads to the expansion of the clay soils. Biofuel has a convex meniscus (contrary to water concave meniscus) which creates higher surface tension forces (strong attractive forces between the grains and clay particles) 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 on it.

It was also observed that biofuel caused higher cracks and surface fractures when infiltrating liners compared to ethanol fuel and water. In this study, the grain size distribution of particles increased drastically when liners were infiltrated 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.

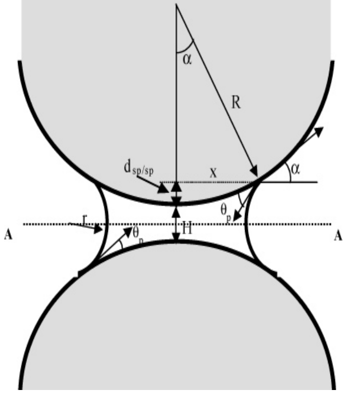


Fig 5. *Capillary forces of water surrounding soil particles, Geometry of the sphere/sphere interaction with a liquid bridge. AA is the plane of symmetry. (after [15])*

Where: γ is the liquid surface tension, R is the particle radius, X is the radius of curvature, θ_p is the contact angle between grain and liquid surface, α is the "embracing angle", and H is the shortest distance between the two spheres.

Clay-ethanol flocs were also observed in the case of using ethanol as a permeate to infiltrate through the liner composed of sand-bentonite mixtures. In the lab experiments, the clay-fuel floc aggregates were formed, which was also found by [16] 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. Also, it is known that the adsorption behaviour appears to be inversely proportional to the solubility of the compound [17] and directly proportional to the percentage of organic matter in the mixture [18]. 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 explains the higher surface fracture in the liner mixtures leached by biofuel. [19] 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. 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 resulting in their isolation, 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. During lab experiments, biofuel flow through the 100% sand liner took more time to reach the column outlet [20, 21]. High biofuel’s viscosity creates higher friction forces between the liner and the fluid. Formation of microorganism colonies in the case of using ethanol fuel as permeate was observed in an anoxic part of the leaching column. These microorganisms used organic carbon as a source of energy to multiply. It might influence the performance of the liner.

Conclusion

Alternative fuels have adverse impacts on sand-bentonite liners through which pores are filled with miscible and immiscible or polar and non-polar liquids/residuals. Thus, the properties of the soil matrix were subject to several physical changes such as surface tension and capillary forces. These changes would lead to the formation of oil-clay flocs, provoking fractures, erodibility, and consequently increased the hydraulic conductivity and finally liners failure. Based on the experimental results the following conclusions can be drawn:

1- The voids in sand-bentonite mixtures increase due to the infiltration of alternative fuels, creating channels that enhance the flow of liquids, resulting in a higher coefficient of permeability of the liner.
2- The biological growth of anoxic microorganisms was observed according to the reported substrate (ethanol) consumption and hence influenced the liner’s performance.
3- The swelling of bentonite is sensitive to the type of leachate.
4- The migration of fine particles depends on the flow direction and has a direct influence on the hydraulic conductivity of the liner.Fine particles migrate the most in the case of water used as the permeate, less for ethanol and then biofuel.
5- Fine particles flushed out the liner is reversely proportional to the amount of bentonite. In the case of biofuel, there are almost no fines flushed out of the liners, when bentonite is higher than 5% of the liner composition.
6- Liner’s surface has more fracture when infiltrated by biofuel compared with ethanol and water. The hydraulic conductivity increases drastically when the liner leached by biofuel.

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