

## Dielectric Dispersion Characteristics of Unsaturated Sand Contaminated by Diesel

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### Abstract

Generally the methods were used to characterize soil contamination include collecting samples of soil then analyzing them to recognize contaminants in the laboratory. Commonly, this method of characterizing the contamination of a soil system is the only one agreeable to regulatory societies. However, sample analysis in the lab faces important problems such as soils sampling is really time consuming and costly, sampling is not constant with time and the samples be able to contaminate through sampling and carrying to the lab. Thus several geophysical techniques have been developed which used the dissimilarity in the soil physical properties after soil contamination. Dielectric technique shows high conceivable for characterization diesel contaminated soil. Further use of this technique; rely upon the availability of information about the dielectric properties of the contaminated soil. In this study, the effects of induced by a diesel presence in an unsaturated soil, on the complex dielectric properties were sought. It has been shown experimentally that the diesel presence in an unsaturated soil is traduced by an increase of both dielectric constant and loss factor. A comparison with the existing results in this study and in the literature for saturated soils shows an opposite effect on the complex dielectric properties. The importance of the influence induced by the diesel on the dielectric properties of an unsaturated soil was noticed and compared to a saturated soil. On the basis of the theoretical dielectric mixture models, a justification to these opposite behaviors and their importance has been presented and various models for the two cases have been developed.

**Keywords:** Diesel; Soil contamination; Dielectric properties; Permittivity

### Introduction

The undesired release of petroleum products via leaking underground storage tanks is a widespread and costly problem. The world demand for fuel has led to the exploration and production of an increasing number of petroleum hydrocarbons (crude oil) reserves [1]. Among hydrocarbon pollutants, diesel oil is a complex mixture of alkanes and aromatic compounds that frequently are reported as soil contaminants leaking from storage tanks and pipelines or released in accidental spills [2], and the effects of spilled oil many times pose serious threats to the ecosystem and effects on marine life [3-9]. Leaking underground and aboveground storage tanks, improper disposal of petroleum wastes, and accidental spills during the transportation and storage of petroleum and petroleum products are major routes of soil and groundwater contamination with petroleum products and have a role in climatic change [10-13].

The toxicity of petroleum products in general, to human and laboratory animals is high due to presence of hemotoxic, carcinogenic and teratogenic components [8,10,14-16]. Diesel fuel as petroleum product is toxic when ingested it cause acute or chronic health problems due to their carcinogenic and mutagenic effects [8,16,17].

Most commonly used methods to characterized subsurface contamination involve collecting representative samples of soil and pore fluid and analyzing them for targeted species in the laboratory. In general, this way of characterizing the contamination of a soil-fluid system is the only one acceptable to regulatory agencies [18]. However, sample collection and analysis in the laboratory pose significant problems: (1) Sampling of the soils is extremely time consuming and expensive;

(2) Sampling is destructive in the case of removing the soil samples;

(3) The sampling is not continuous with time;

(4) Samples can be contaminated during sampling, transportation, and analysis in the laboratory.

Thus several geophysical methods have been developed which utilizes the contrast caused by the contaminant on physical properties of the soil [19,20]. Electromagnetic and dielectric methods shows high potential for characterization hydrocarbon contaminated soil and determination of the level of contaminant. Further use of this method; rely upon the availability of information about the dielectric properties of the contaminated soil. Development of such information has formed a focus of research by many authors around the world over several years [20-22].

Chenaf and Amara [20] evaluate the dielectric properties of unsaturated soil contaminated with diesel using TDR. The measured dielectric properties have been found to increase when diesel concentration increases. These finding are in disagreement with what has been found in previous studies when dealing with diesel-contaminated soils that are fluid (water and diesel) saturated. Chenaf and Amara [20] study the hydrocarbon contaminated at 15% moisture content only, therefore there is a need to evaluate the dielectric properties of hydrocarbon contaminated at various level of moisture

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ranging from dry to saturated conditions in order to understand the dielectric behavior of hydrocarbon contaminated soil.

Since the application of electromagnetic method to hydrocarbon contaminated soil is restricted to saturated soils [23,24]. Therefore, these finding, highlight the need for more research needed for electromagnetic and dielectric characterization of unsaturated soils contaminated with hydrocarbon.

## Dielectric Properties

The dielectric constant of a material is a complex parameter and is composed of real and imaginary parts. The real part results from the polarization of material, while the imaginary part is due to ohmic and polarization losses. Following are brief definitions of the important variables of the dielectric properties of materials. If a capacitor of a plate area of  $A$  and separation of  $d$  contains no matter between its plates, its capacitance  $C_0$  will be:

$$C_0 = \frac{\epsilon_0 A}{d} \quad (1)$$

Where  $\epsilon_0$  is the permittivity of a vacuum.

If a material of complex permittivity ( $\epsilon$ ) is inserted between the plates, the capacitance will increase to:

$$C = \frac{C_0 \epsilon}{\epsilon_0} \quad (2)$$

If a capacitor is leaky, that is, if there is some energy dissipation mechanism inherent in it, there will be a loss current,  $I_p$  that lags the charging capacitor,  $I_c$ , and is separated from the charging current by a loss angle,  $\theta$ . The dissipation factor,  $D$ , is defined as:

$$D = \tan \theta = I_p / I_c \quad (3)$$

If the loss current arises from a process other than the migration of charge carries, a complex dielectric constant is defined:

$$\epsilon = \epsilon' - j\epsilon'' \quad (4)$$

Where  $\epsilon$  is the complex permittivity,  $\epsilon'$  is the real part (dielectric constant),  $\epsilon''$  is the imaginary part (loss factor) and  $D = \tan \theta = \frac{\epsilon''}{\epsilon'}$ . In terms of the dissipation factor,  $D$ :

$$D = \tan \theta = \frac{\epsilon''}{\epsilon'} \quad (5)$$

The dielectric material increases the storage capacity of a capacitor by neutralizing charges at the electrode surface. The neutralization process may be visualized as the result of the orientation or creation of dipoles to oppose the applied field. Such a polarization is proportional to the ease with which the dielectric can be polarized. The polarizability is the average induced polarization per unit field strength. As the soil-fluid system becomes conductive, the system will act as a leaky capacitor, in other words, there will be of current, which will result in a higher dissipation. In the presence of ions, the ions will move under

an externally applied electrical field if there is enough time for them to orient themselves in the direction of applied field. However, if there is not enough time for orientation of the ions, polarization will not take place, which will lower the dielectric constant.

## Materials and Methods

Quartz sand was used as a benchmark. A sand sample of 500 g was used to determine the porosity of the soil. The total volume ( $V$ ) of the dry sample is 315.65 cm<sup>3</sup>, the volume of solid is 189.39 cm<sup>3</sup> and the volume of voids ( $V_v$ ) is 126.26 cm<sup>3</sup> therefore the porosity  $n$  of the soil sample is 0.4, the specific gravity is 2.64 the dry bulk density is 1.584.

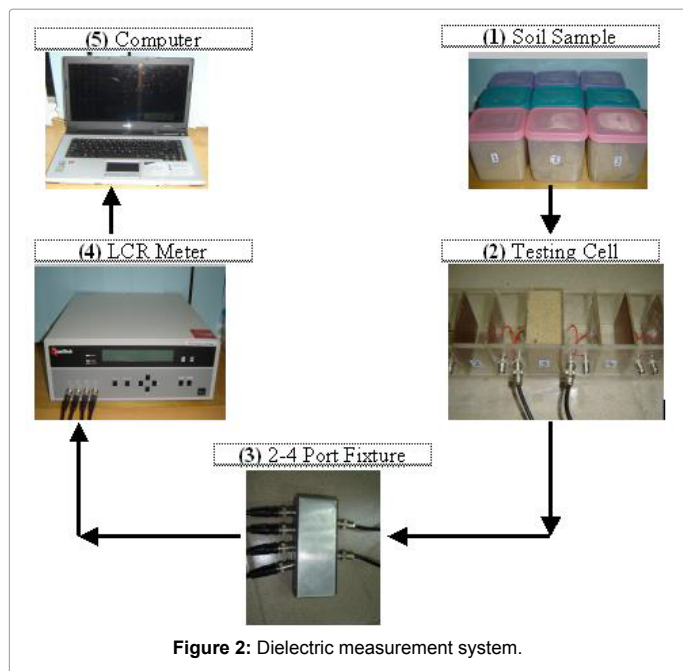
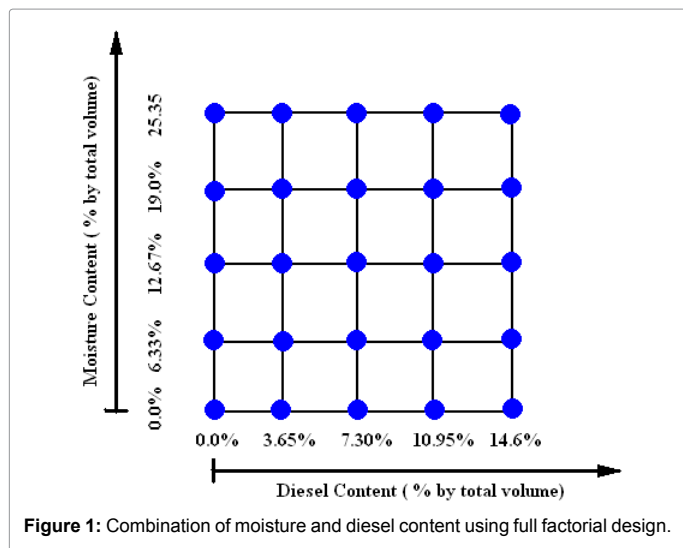
To evaluate the effect of hydrocarbon contamination of saturated sand soil, five samples were prepared. The total liquid (water and diesel) was 40% by volume in order to saturate the sand soil with porosity 0.4. Five different diesel content were used in order to contaminate the sand soil. The details of these five samples are shown in Table 1.

To determine the diesel contamination levels of unsaturated soil designed in this study, 25 soil samples were prepared from dry sand. The weight of each sample was 500 g. The sand samples were divided into 5 groups each group consist of five samples. First group were kept dry (Moisture Content 0%). Water was added to the other four sets of sand samples 2, 3, 4, and 5. The amount of water for set 2, 3, 4, and 5 were 20 g, 40 g, 60 g and 80 g respectively in order to have moisture contents of 4%, 8%, 12%, and 16% by weight of dry sand. The five samples in each set were contaminated by diesel contents 0%, 2%, 4% 6% and 8% by weight of dry sand. The amounts of diesel were 0 g, 10 g, 20 g, 30 g and 40 g. The soil samples are shacked for 15 min then kept for three hours in sealed plastic container to avoid any evaporation. The aim of this operation is to insure a uniform diesel and moisture distribution within the soil sample and a well diesel and water adsorption by the soil grains. The soil is then placed in the cell test 80×80 mm and 40 mm thick. Three impedance measurements were obtained at each frequency and the average value was determined. Total of diesel contaminated soil samples were 25 covers a 5 full factorial analysis presented in Figure 1.

After soil samples were prepared, the soil is then placed in the cell test 8080 mm and 40 mm thick. Three impedance measurements were obtained at each frequency and the average value was determined. The test cells were of internal dimensions 80 × 80 × 40 mm. Copper electrodes with dimensions of 80×80×2 mm were attached to two opposite faces of the cell. Copper connections were passed through the cell walls and connected to the electrodes. All impedance measurements were acquired using the QuadTech 1900LCR meter, operating in voltage-drive mode, with the signal voltage being 1000 mV. A linear sweep over the frequency range of 1 kHz to 1000 kHz was used with the data, recorded at 21 frequency points within this range. The connection to the LCR meter was by means of short, individually screened coaxial cables to the voltage (V), high/low and current (I) output/input terminals. Cable impedance, plate impedance and fringing impedance were determined using an appropriate model circuits. From the measurements, the impedances of soil sample were

Sample Code	Weight (gram)				Volume Fraction			
	Soil	Water	Diesel	Air	Soil	Water	Diesel	Air
D0WS	500	126.26	0	0	0.6	0.4	0	0
D1WS	500	114.72	10	0	0.6	0.3635	0.0365	0
D2WS	500	103.18	20	0	0.6	0.327	0.073	0
D3WS	500	91.64	30	0	0.6	0.2905	0.1095	0
D4WS	500	80.1	40	0	0.6	0.254	0.146	0

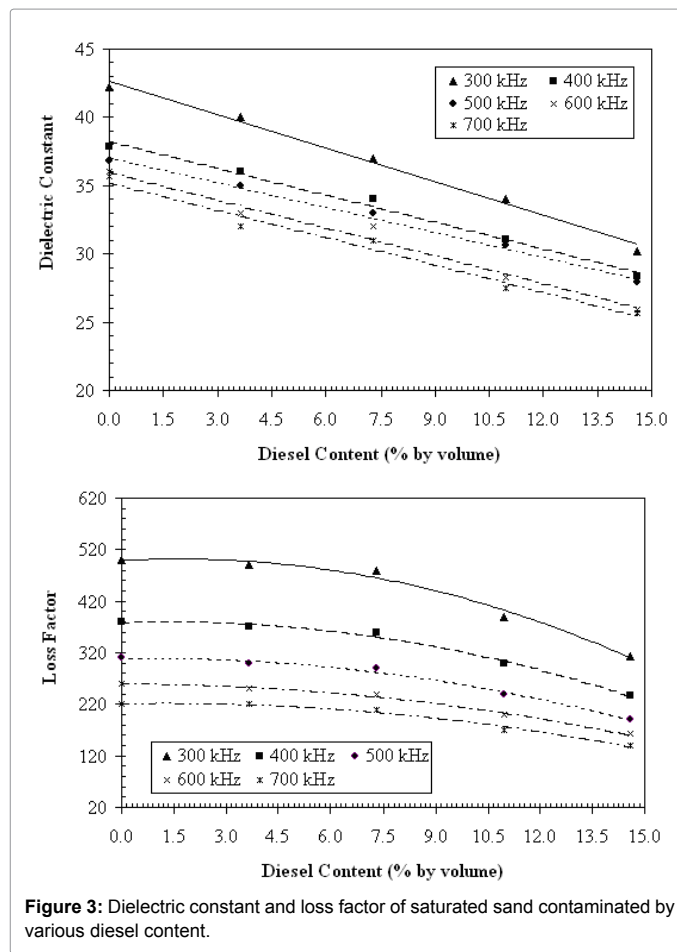
Table 1: Samples of diesel contaminated soil-saturated samples.



calculated and a PC logs all data. The complex permittivities (real part dielectric constant and imaginary part loss factor) of soil samples were deduced from the impedance of the soil. The setup of the system is shown in Figure 2.

## Results and Discussions

The results of dielectric constant and loss factor of saturated soil contaminated with five level of diesel hydrocarbon versus frequency are shown in Figure 3. It is clear from the results of diesel contaminated soil at saturation condition that both dielectric constant and loss factors of soil are decreased with increasing diesel content. The dielectric constant shows linear decrease with increasing diesel content while loss factor shows high rate of decrease with increasing diesel content. This can be attributing to the decrease of conductivity of the soil when the high conducting water is replaced by a very low conductivity of hydrocarbon (diesel). These results can be attributing to the high resistivity of diesel



and reducing of current conductance at high frequency. These results are in agreement of the results reported in literature [25].

The variations of dielectric constant and loss factor of unsaturated contaminated soil with diesel content are shown in Figures 4 and 5. The results show an opposite trend of saturated soil discussed in previous section. In general, the dielectric constant (real part of permittivity) increased with increasing diesel content while the loss factor (imaginary part of permittivity) showed three different trends similar to the trends observed for resistivity. The loss factor of unsaturated contaminated soil indicates an opposite result in compare with the result of saturated soil. The result of loss factor of unsaturated soil contaminated with hydrocarbon (diesel) depends on the relative resistivity of water and hydrocarbon, the relative volume fraction of the three phases in the pore structure of the soil (water, diesel and air). To draw a clear picture about the effect of diesel content on the loss factor of unsaturated soil, unsaturated soil were divided into low, medium and high moisture content.

The loss factor of low moisture contaminated soil increased with increasing diesel content up to certain level then the trend changed and a decreasing in the loss factor were recorded at high diesel content. The loss factor of medium moisture contaminated shows an increase with increasing diesel content. The resistance of high moisture contaminated soil increase with low diesel content then decrease with further increasing diesel content. Therefore, the result indicates that the electromagnetic properties of soil are dependent on the interaction of the various phases (water, diesel and air) in the voids of unsaturated

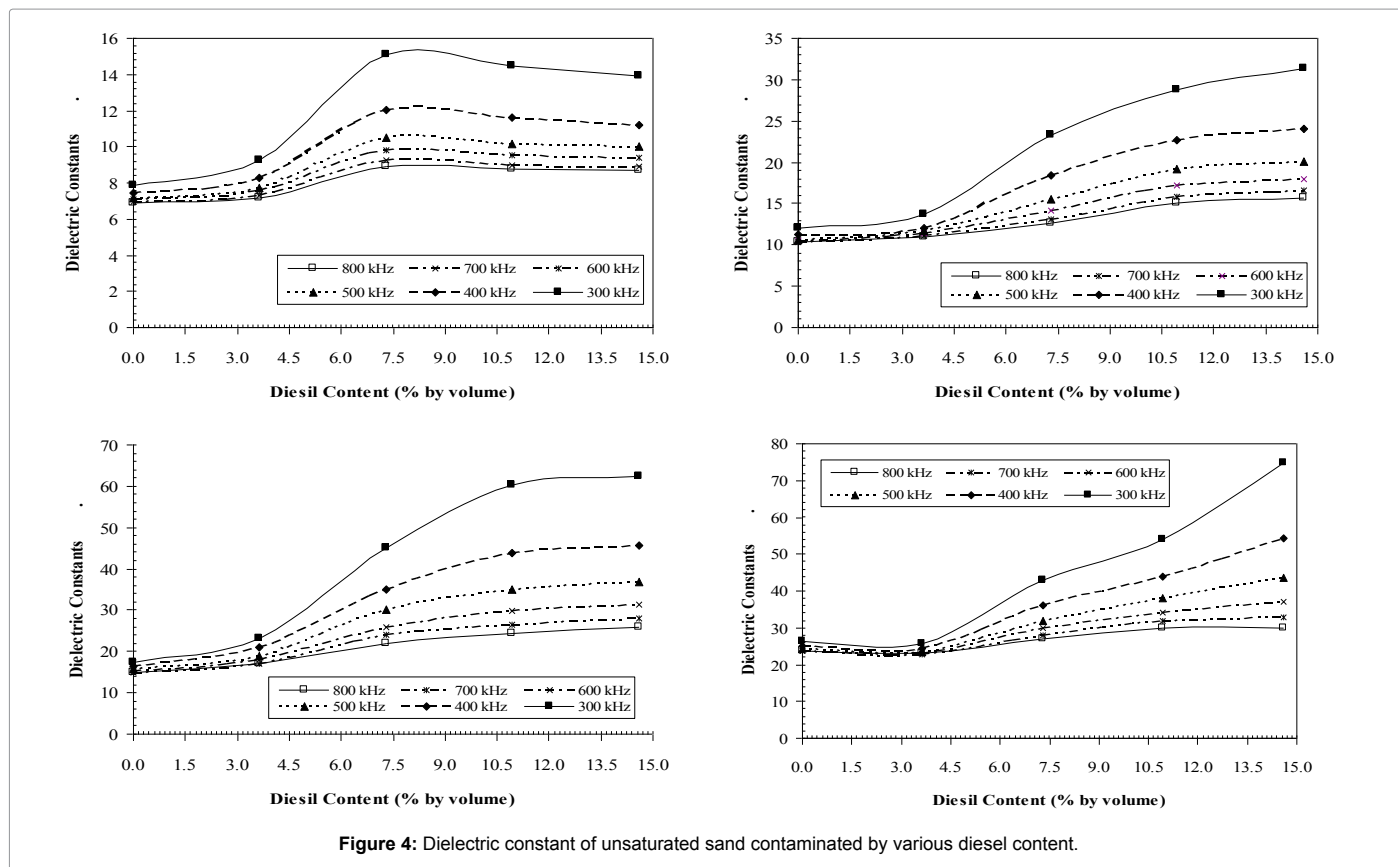


Figure 4: Dielectric constant of unsaturated sand contaminated by various diesel content.

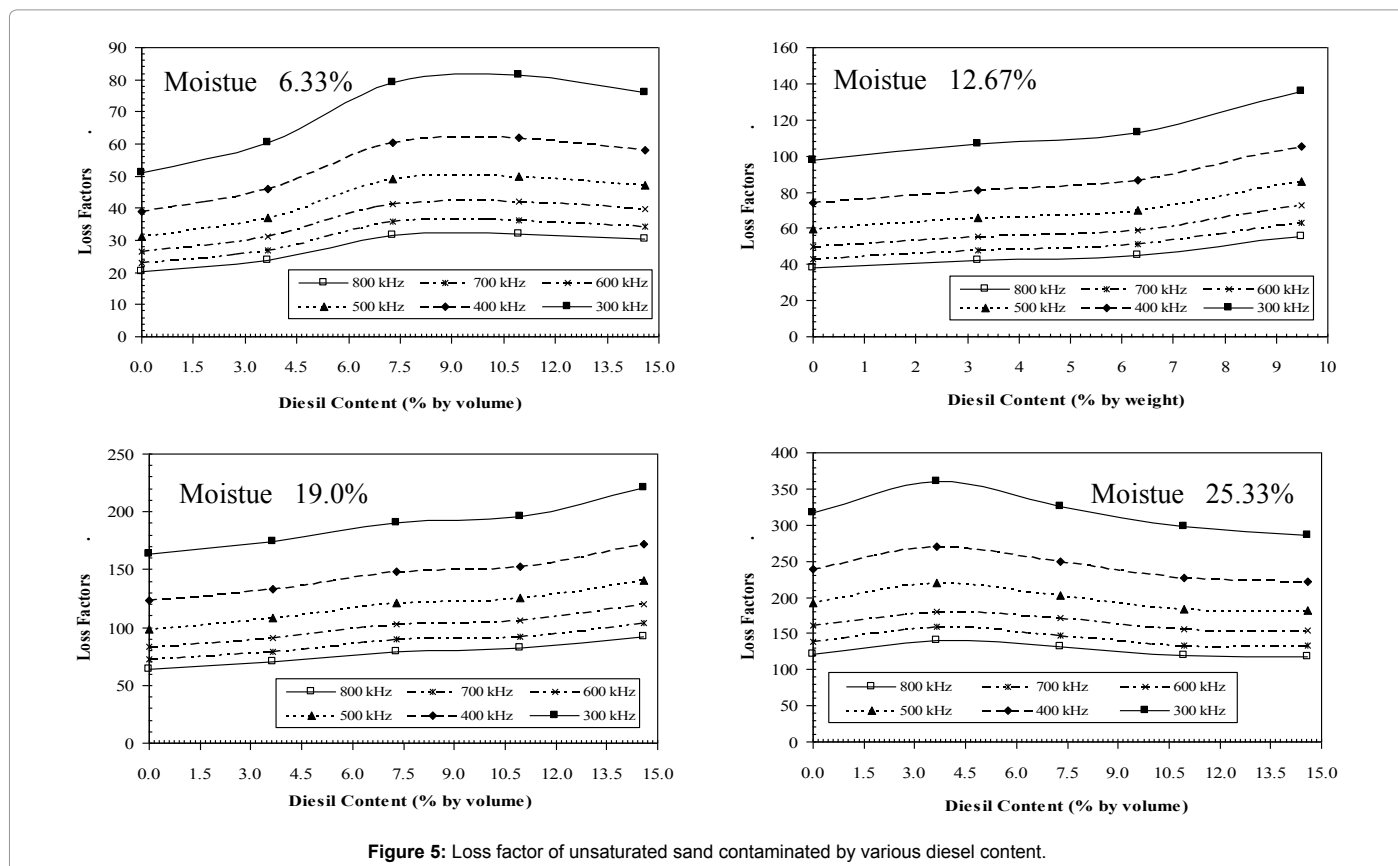


Figure 5: Loss factor of unsaturated sand contaminated by various diesel content.

soil. In addition, these results may be attributing to the conducting channels forms in the pore space of soil.

Low moisture content of unsaturated soil tends to form few conducting channels through the pore structure of soil and several other channels were terminated by air voids. Adding diesel liquid to this soil will fill some of the air voids and some of diesel will displaced water and give more chance for the water to form more conducting channels. This can attribute to the decreasing of resistivity of the soil with increasing the conductance channel formed by water. Additional increasing of diesel will face a limited space of air voids and start to interrupt the existing conducting channels of water phase, this result on an increase in the resistivity of the soil material.

Adding diesel to the unsaturated contaminated soil having medium moisture content help to form more conducting channels by water phase from the large amount of water available in the unconnected channel in the pore structures of soil. This phenomenon can be used to explain the continuous decrease of resistivity of soil with increasing diesel content as a result of increasing the conductivity of soil by water.

Unsaturated contaminated soil having high moisture content tends to have very high conducting water channels and less air voids available.

Adding small amount of diesel will increase the conducting channels of water from the few available non-conducting water channels. Increasing the diesel content furthermore, the diesel start to cut the water conducting channels in the soil which will leads to increase the resistivity of the soil similar to the saturated soil.

The results obtained from the analytical models for soil-contaminated samples showed low predictability of the analytical models for hydrocarbon contamination soil. This can be attributed to the not expected result of unsaturated contaminated soil. In addition the local peaks and curvature of the results presented in Figures 4 and 5 are difficult to consider by these forms of analytical models. These results raised the need for empirical models which can offer butter predictability of the dielectric properties of contaminated soil, therefore, the inverse of these models can be used more accurately to determine the contamination level.

Since the available mixture model show low predictability of the dielectric properties of contaminated soil with hydrocarbon, several empirical model using linear, interaction and nonlinear quadratic model were developed using modules in MATLAB software. The detail analysis of these models for dielectric constant and loss factor of contaminated soils are shown in Figures 6 and 7.

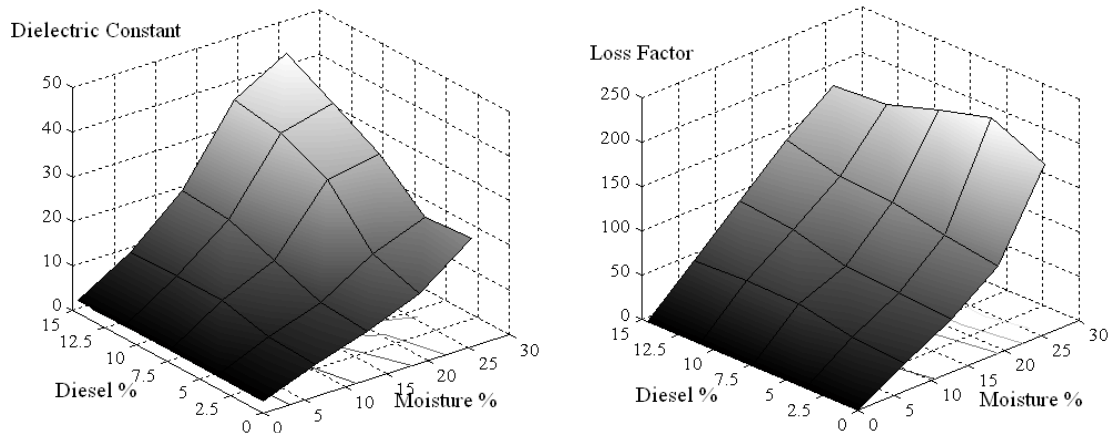


Figure 6: Response surface of dielectric constant and loss factor of contaminated soil at various moisture content and diesel content.

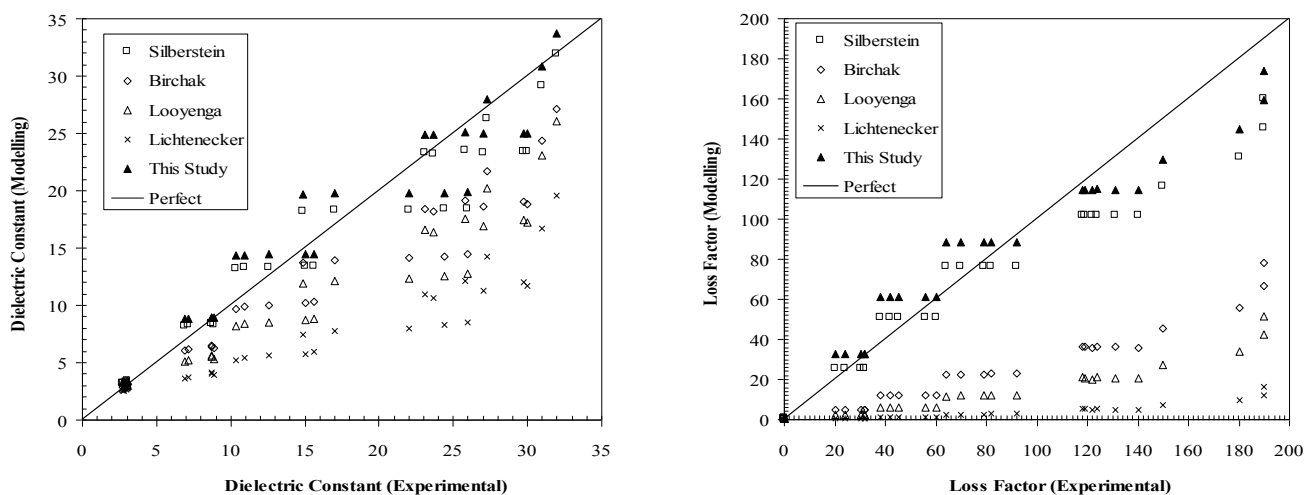


Figure 7: comparison of experimental and theoretical dielectric constant and loss factor of diesel contaminated soil.

From the result of regression analysis, it is clear that the best fit model was the interaction model. This model has the higher values of adjusted square of the correlation coefficient and also has the minimum mean square error.

$$Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_{12} X_1 X_2 \text{ with } R^2 = 98.2\% \quad (6)$$

## Conclusions

In this study, the effects of induced by a diesel presence in an unsaturated soil, on the impedance (resistance and reactance) and permeability (dielectric constant and loss factor) were sought. It has been shown experimentally that the diesel presence in an unsaturated soil is traduced by an increase of both dielectric constant and loss factor. A comparison with the existing results in this study and in the literature for saturated soils shows an opposite effect on the complex impedance and dielectric properties. The importance of the influence induced by the diesel on the dielectric properties of an unsaturated soil was noticed and compared to a saturated soil. On the basis of the analytical and the empirical models, a justification to these opposite behaviors and their importance are presented and various model for the two cases are developed.

The contaminated soils exhibit different complex dielectric dispersion from the uncontaminated soils. The differences of the dielectric behavior with contaminant content suggest that the monitoring of complex dielectric constant has the potential to quantify contaminants. The additional analysis for the imaginary part of the dielectric constant can be recommended to obtain the clear information about the state of contaminants in soil material.

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