Dielectric Dispersion Characteristics of Unsaturated Sand Contaminated by Diesel

Rabah M Ismail1, Hashem M Al-Mattarneh2, Muhd Fadhil Nuruddin3, Nasir Shafiq4 and Mohamad A Dahim2

1Faculty of Engineering, Universiti Teknologi Petronas, Malaysia
2Faculty of Engineering, Najran University, Saudi Arabia
3Faculty of Engineering, King Khalid University, Saudi Arabia

Abstract

Generally the methods were used to characterize soil contamination include collecting samples of soil then analyzing them to recognize contaminates 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.
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 findings, 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:

\[
C_0 = \frac{\varepsilon_0 A}{d}
\]

Where \( \varepsilon_0 \) is the permittivity of a vacuum.

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

\[
C = \frac{C_0 \varepsilon}{\varepsilon_0}
\]

If a capacitor is leaky, that is, if there is some energy dissipation
mechanism inherent in it, there will be a loss current, \( I_L \), 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:

\[
D = \tan \theta = I_L/I_c
\]

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

\[
\varepsilon = \varepsilon' - j\varepsilon''
\]

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

\[
D = \frac{\tan \theta}{\varepsilon} \frac{\varepsilon''}{\varepsilon'}
\]

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\(^3\), the volume of solid is 189.39 cm\(^2\) and
the volume of voids \( V_v \) is 126.26 cm\(^3\) 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
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 shook 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 80×80 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  | 91.84 | 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 formed 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 better 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.](image)

![Figure 7: Comparison of experimental and theoretical dielectric constant and loss factor of diesel contaminated soil.](image)
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.

\[
Y = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_1 \cdot \beta_2 X_1 \cdot X_2 \text{ with } R^2 = 98.2\%
\] 

(6)

Conclusions

In this study, the effects of induced by a diesel presence in an unsaturated soil, on the impedance (resistance and reactance) and permeativity (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.

References

1. Bierkens J, Geerts L (2014) Environmental hazard and risk characterisation of petroleum substances: A guided “walking tour” of petroleum hydrocarbons. Environment international 66: 182-193.

2. Gallego JL, Loredo J, Llamas JF, Vázquez F, Sánchez J (2001) Bioremediation of diesel-contaminated soils: evaluation of potential in situ techniques by study of bacterial degradation. Biodegradation 12: 325-335.

3. DeLeo DM, Ruiz-Ramos DV, Baums IB, Cordes EE (2016) Response of deep-water corals to oil and chemical dispersant exposure. Deep Sea Research Part II: Topical Studies in Oceanography 129: 137-147.

4. Sagerup J, Nahrgang J, Frantzen M, Larsen LH, Geraudie P (2016) Biological effects of diesel marine oil exposure in red king crab (Paralithodes camtschaticus) assessed through a water and foodborne exposure experiment. Marine environmental research 119: 126-135.

5. Varjani SJ, Srivastava VK (2015) Green technology and sustainable development of environment. Renewable Research Journal 3: 244-249.

6. Ron EZ, Rosenberg E (2014) Enhanced bioremediation of oil spills in the sea. Current Opinion in biotechnology 27: 191-194.

7. Hentati O, Lachhab R, Ayadi M, Kabi M (2013) Toxicity assessment for petroleum-contaminated soil using terrestrial invertebrates and plant bioassays. Environmental monitoring and assessment 185: 2899-2998.

8. Chandra S, Sharma R, Singh K, Sharma A (2013) Application of bioremediation technology in the environment contaminated with petroleum hydrocarbon. Annals of microbiology 63: 417-431.

9. Chase DA, Edwards DS, Qin G, Wages MR, Willimg MM, et al. (2013) Bioaccumulation of petroleum hydrocarbons in fiddler crabs (Uca minax) exposed to weathered MC-252 crude oil alone and in mixture with an oil dispersant. Science of the Total Environment 444: 121-127.

10. Souza EC, Vessoni-Penna TC, de Souza Oliveira RP (2014) Biosurfactant-enhanced hydrocarbon bioremediation: An overview. International Biodeterioration & Biodegradation 89: 88-94.

11. Prince RC, McFarlin KM, Butler JD, Febbo EJ, Wang FC, et al. (2013) The primary biodegradation of dispersed crude oil in the sea. Chemosphere 90: 521-526.

12. Abdullah MZB, Saat AB, Hamzah ZB (2012) Assessment of the impact of petroleum and petrochemical industries to the surrounding areas in Malaysia using moses as bioindicator supported by multivariate analysis. Environmental monitoring and assessment 184: 3959-3969.

13. Wassmann P, Duarte CM, Agusti S, Serj MK (2011) Footprints of climate change in the Arctic marine ecosystem. Global change biology 17: 1235-1249.

14. Zhang XW, Yang LS, Li YH, Li HR, Wang WY, et al. (2011) Estimation of lead and zinc emissions from mineral exploitation based on characteristics of lead/ zinc deposits in China. Transactions of Nonferrous Metals Society of China 21: 2513-2519.

15. Costa AS, Romão LPC, Araújo BR, Lucas SCO, Maciel STA, et al. (2012) Environmental strategies to remove volatile aromatic fractions (BTEX) from petroleum industry wastewater using biomass. Bioresouce Technology 105: 31-39.

16. Meckenstock RU, Boll M, Mouttaki H, Koelschbach JS, Cunha Taruco P, et al. (2016) Anaerobic degradation of benzene and polycyclic aromatic hydrocarbons. Journal of molecular microbiology and biotechnology 26: 92-118.

17. Mrozik A, Pietrowska-SEGEL Z (2010) Bioaugmentation as a strategy for cleaning up of soils contaminated with aromatic compounds. Microbiological research 165: 363-375.

18. Okoye CN, Cotton TR, O’Meara D (1995) Application of resistivity cone penetration testing for qualitative delineation of creosote contamination in saturated soils. In Geoenvironment 2000: Characterization, Containment, Remediation, and Performance in Environmental Geotechnics pp: 151-166. ASCE.

19. Olhoeft GR (1986) Direct detection of hydrocarbon and organic chemicals with ground penetrating radar and complex resistivity. In Proc. NWWA/API Conf. Petroleum Hydrocarbons and Organic Chemicals in Ground Water-Prevention, Detection and Restoration.

20. Chenell D, Amara N (2001) Time domain reflectometry for the characterization of diesel contaminated soils. In Proceedings of the second international symposium and workshop on time domain reflectometry for innovative geotechnical applications, Northwestern University.

21. Arcena JM, Rutherford PM (2005) Properties of hydrocarbon-and salt-contaminated flire pit soils in northeastern British Columbia (Canada). Chemosphere 60: 567-567.

22. Hilhorst MA (1998) Dielectric characterisation of sol. Doctoral Thesis, Wageningen Agricultural University, Wageningen, The Netherlands.

23. Robinson DA, Friedman SP (2001) Effect of particle size distribution on the effective dielectric permittivity of saturated granular media. Water Resources Research 37: 33-40.

24. Redman JD, Kueper BH, Annan AP (1991) Dielectric stratigraphy of a DNAPL plume: Application to subsurface detection. Ground Water Management 5: 1017-1030.

25. Son Y, Oh M, Lee S (2009) Influence of diesel fuel contamination on the electrical properties of unsaturated soil at a low frequency range of 100 Hz–10 MHz. Environmental geology 58: 1341-1348.