



Dielectric dispersion characteristics of sand contaminated by heavy metal, landfill leachate and BTEX (02-104B)

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Abstract

Experiments have been performed to investigate the dielectric properties of contaminated sand. The separate real and imaginary parts of a dielectric constant were investigated in the frequency range of 75 kHz to 12 MHz. The contaminated soils exhibit different complex dielectric dispersion from the uncontaminated soils. The variation in the real dielectric constant can be explained by a polarization mechanism while that in the imaginary dielectric constant by ionic conductivity loss mechanism. The differences of the dielectric behavior with contaminant types suggest that the monitoring of complex dielectric constant has the potential to classify contaminants. The additional analysis for the imaginary part of the dielectric constant can be recommended to obtain the clear information about the state of ionic contaminants in subsurface.

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1. Introduction

Leachate leaked from municipal or industrial landfill, petroleum products leaked from underground storage tanks, and heavy metals in the acid mine drainage are the major components of subsurface contamination. Physical and chemical analysis for the detection of contaminated soil and groundwater are essential processes, but conventional sampling and analysis methods are extremely time consuming and expensive [1]. Therefore, new in situ

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methods must be developing a simple, quick and cost-effective detecting technique. One of the techniques utilized to assess the subsurface contamination is electrical resistivity measurement, which can be performed rapidly and nondestructively in situ. But, electrical resistivity measurements alone will lead to some degree of ambiguity. To overcome the ambiguity inherent in electrical resistivity measurements, measurement of the dielectric constant of the system has also been suggested to characterize subsurface contamination [1,2]. The dielectric property becomes an important parameter in remote sensing and geophysical exploration [3].

In early stage research for dielectric properties, efforts have been made to estimate the quantity of water contained in soil and/or the soil structure, particle shape, soil mineralogy and anisotropy by measuring dielectric constant of soil [4–7]. It has been recognized that the dielectric properties of soils can serve as a sensitive and accurate indicator of soil moisture. Recently, research utilizing dielectric properties for the investigation of subsurface contamination has been main theme [1,8,9]. However, the dielectric behavior of contaminated soil has not yet been fully established.

In this study, the separate real and imaginary parts of dielectric constant were investigated in the range of 75 kHz to 12 MHz since most of the previous studies were of unspecified measurement frequency or measured only the real part of dielectric constant. Thus, the objectives of this research was to investigate the behavior of dielectric dispersion of contaminated sand and then to demonstrate the feasibility of the dielectric measurement technique for characterizing subsurface.

2. Background

2.1. Basic concept of dielectric property

The relative permittivity or dielectric constant is a measure of the extent to which the electrical charge distribution in a material can be polarized by the application of an electrical field. In an alternating electric field, a soil sample is characterized by a complex dielectric constant (ϵ^*):

$$\epsilon^* = \epsilon' - i\epsilon'' \quad (1)$$

where ϵ' is the real part of dielectric constant or the common dielectric constant of the material and ϵ'' is imaginary part of dielectric constant.

The real part of dielectric constant represents the capacitive behavior or polarizability of the material, while the imaginary part represents the energy losses due to polarization and conduction [1,10,11].

When an alternating electric field is applied, a polarization is produced. That polarization can be measured in terms of capacitance. In measuring the dielectric constant, a basic understanding of capacitance theory is beneficial. Capacitance (C) is defined as the ability of two electrodes to store a charge (Q) when a potential (V) is applied across them. If the region between the two parallel electrodes is a vacuum at parallel plate capacitor, then the capacitance (C_0) is found by Eq. (2):

$$C_0 = \frac{Q}{V} = \epsilon_0 \frac{A}{d} \quad (2)$$

where ε_0 is the permittivity of free space (8.854×10^{-12} F/m), A the area of the electrodes and d is the distance between the two electrodes.

If a material with a permittivity of ε'_m is inserted between the plates, the capacitance is calculated by Eq. (3):

$$C = \varepsilon'_m \frac{A}{d} = C_0 \frac{\varepsilon'_m}{\varepsilon_0} = C_0 \varepsilon' \quad (3)$$

where ε' is the real part of dielectric constant or relative permittivity. Thus, real part of dielectric constant is defined as the ratio of the permittivity of the material to the permittivity of free space and dimensionless.

If there is some energy dissipation mechanism inherent in a capacitor, there will be a loss current, I_1 , that lags the charging current, I_c , and is separated from the charging current by a loss angle, δ . Dissipation factor D or loss tangent $\tan \delta$ can be expressed by the ratio of loss current to charging current as shown in Eq. (4) [1,12].

$$D = \tan \delta = \frac{I_1}{I_c} = \frac{1}{\omega RC} = \frac{\varepsilon''}{\varepsilon'} \quad (4)$$

where ω is the angular frequency (Hz) ($2\pi f$), R resistance (Ω) and C is capacitance (F).

Thus, the imaginary part of dielectric constant can be written as Eq. (5):

$$\varepsilon'' = \varepsilon' \tan \delta = \frac{\varepsilon'}{\omega RC} \quad (5)$$

By measuring the real and imaginary parts of the dielectric constant for frequency, the dielectric behavior of a material could give information on the characteristics of the material [1,8].

2.2. Polarization mechanisms

Due to the various kinds of charge carriers that exist within dielectric materials that are able to be displaced and polarized by an electric field, there are several types of polarization mechanisms that tend to dominate certain frequency ranges. When an external electric field is applied, the charge distribution realign in materials. This phenomenon is called dielectric polarization or polarization. Polarization arises due to the existence of atomic and molecular forces, and appears whenever charges in a material are somewhat displaced with respect to one another under the influence of an electric field. The number of charges per unit volume multiplied by the average displacement is the polarizability of the medium. The magnitude of polarizability of a material is reflected by the dielectric constant [4].

The total polarization of a dielectric arises from four sources of charge displacement: (1) electronic polarization, (2) atomic polarization, (3) orientation/dipole polarization, (4) space charge polarization [1,12]. These four mechanisms are illustrated in Fig. 1.

Electronic polarization is common to all materials, as it involves distortion of the center of charge symmetry of the basic atom. Under the influence of an applied field, the nucleus of an atom and the negative charge center of the electrons shift, creating a small dipole. This

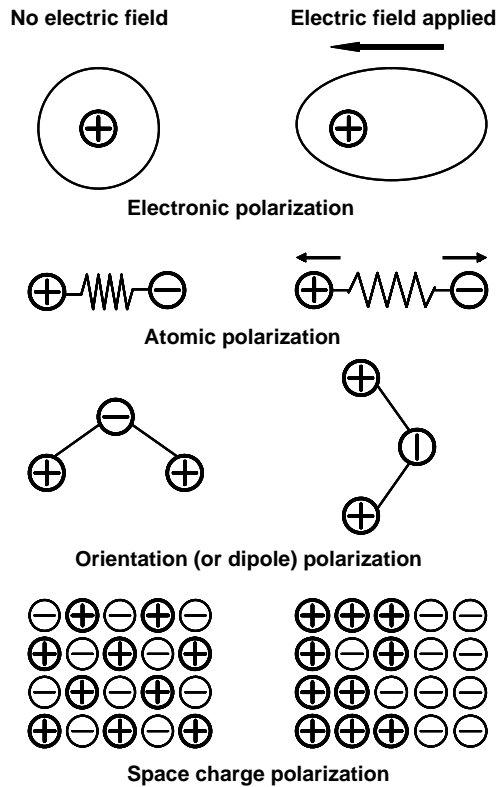


Fig. 1. Mechanisms of polarization (modified after Von Hippel, 1954).

polarization effect is small, despite the vast number of atoms within the material, because the moment arm of the dipoles is very short.

Atomic polarization is common in an ionic bond or covalent bond materials, which consist of crystal lattices occupied by cations and anions. Under the influence of an electric field, dipole moments are created by the shifting of these charged atoms (or ions) towards the opposing polarity. The displacement or moment arm of the dipoles can be relatively large in comparison to the electronic displacement, and therefore can give rise to higher dielectric constants than that of electronic polarization.

Orientation (or dipole) polarization is a phenomenon involving rotation of permanent dipoles under an applied field. Orientation polarization is more common in polymers which by virtue of their atomic structure permit reorientation. This mechanism of permanent dipoles is different from that of induced dipoles of ionic polarization.

Space charge polarization is extrinsic to any crystal lattice and common in multiphase system. The phenomenon arises due to charge carriers that can migrate for some distance through the dielectric. Compared with other mechanisms, this polarization effect is very large. But the effect of orientation polarization or space charge polarization is a negligible quantity at high frequency (MHz range) because the mobility of charge carrier is relatively small.

3. Experimental section

Jumunjin sand, used in this study, was classified as poorly graded sand (SP) under the Unified Soil Classification System. Table 1 shows the index properties of this sand. Heavy metals including lead (Pb) and cadmium (Cd), and aromatic hydrocarbons including benzene, toluene, ethylbenzene and xylene (BTEX) were used as contaminants. Lead and Cadmium standard solutions (Cica-reagent, Kanto Chemical Co., Inc., Japan), and BTEX (ACS reagent, Sigma–Aldrich, USA) were diluted in deionized water (DI water), respectively.

Landfill leachate was collected from a municipal landfill site in Korea was used as a contaminant solution. The Gimpo landfill is located at Incheon Metropolitan City and Gimpo-city, Gyunggido on coastal reclaimed land around $126^{\circ}36'$ of east longitude and $37^{\circ}47'$ of north latitude. The Gimpo landfill is the largest single landfill in the world. It is on the reclaimed land of about 20,700,000 m² in Gimpo district in the west sea coast. The constituents and their concentrations of the Gimpo landfill leachate are shown in Table 2. Gimpo landfill leachate used here could be considered as representative for the typical landfill leachate in its constituents and concentration because their constituents and concentration fall into the general range of the reported leachate data [13].

Acrylic mold was designed for this study. Fig. 2 shows the schematic diagram of acrylic mold used in this study. The acrylic mold employed the parallel plate capacitor method to

Table 1
Index properties of Jumunjin sand

Coefficient of gradation	1.01
Uniformity gradation	1.51
Specific gravity	2.64
Plasticity index	Non-plastic
USCS	SP ^a
Maximum dry density (g/cm ³)	1.66
Minimum dry density (g/cm ³)	1.33

^a SP: poorly graded sand.

Table 2
Constituents and concentrations of Gimpo landfill leachate

Constituents	Concentration	
	Ranges (mg/l)	Average (mg/l)
T–N	1602–2148	1964
T–P	10.90–16.59	13.13
NH ₃ –N	1529–2018	1814
TS	11790–16360	13620
Cl [–]	3617–5177	4313
Cu	0.06–1.07	0.20
Cd	<0.17	0.02
Pb	<0.17	0.02
As	<0.21	0.05
Cr ⁶⁺	0.04–0.16	0.10
Zn	0.60–2.15	1.30

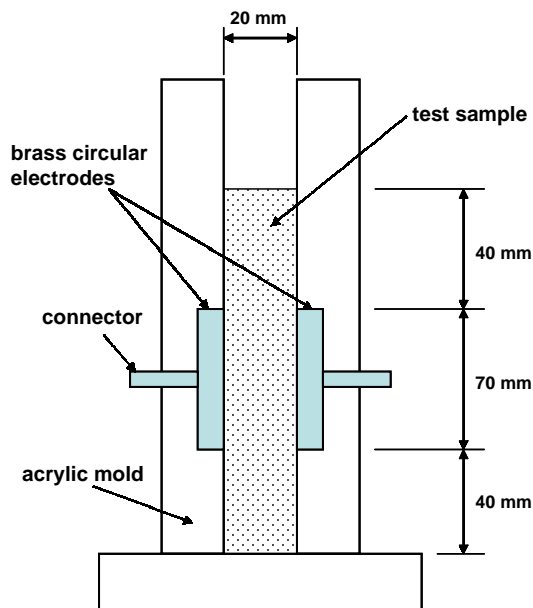


Fig. 2. Schematic diagram of acrylic mold.

measure the dielectric property of samples. The brass electrodes are 7 cm in diameter and they are 2 cm apart. The soil sample was placed between the two electrodes in acrylic mold. The dielectric properties were measured in acrylic mold were calibrated in air and DI water before measurement of the dielectric constant of the samples to eliminate the fringing effect of measurement system.

Jumunjin sand was uniformly mixed with DI water or contaminant solutions. Then the soil-solution mixture was directly compacted in an acrylic mold. The dielectric properties were calculated from the measured capacitance and resistance of the soil sample with the known area and thickness. The real and imaginary parts of the dielectric constant were calculated using Eqs. (3) and (5), respectively. The capacitance and resistance of the soil sample were measured using HP 4285A Precision LCR Meter which is capable of determining the capacitance of soil samples in the frequency range from 75 kHz to 12 MHz. Because dielectric properties of materials are affected by temperature [3,14], the measurements were performed rapidly at room temperature (19–21 °C) to minimize the effect of temperature variations.

4. Results and analysis

4.1. Dielectric dispersion characteristics

It has been recognized that the dielectric constant of soil is greatly influenced by the volumetric water content, which is defined as the ratio of the water volume in a soil mass

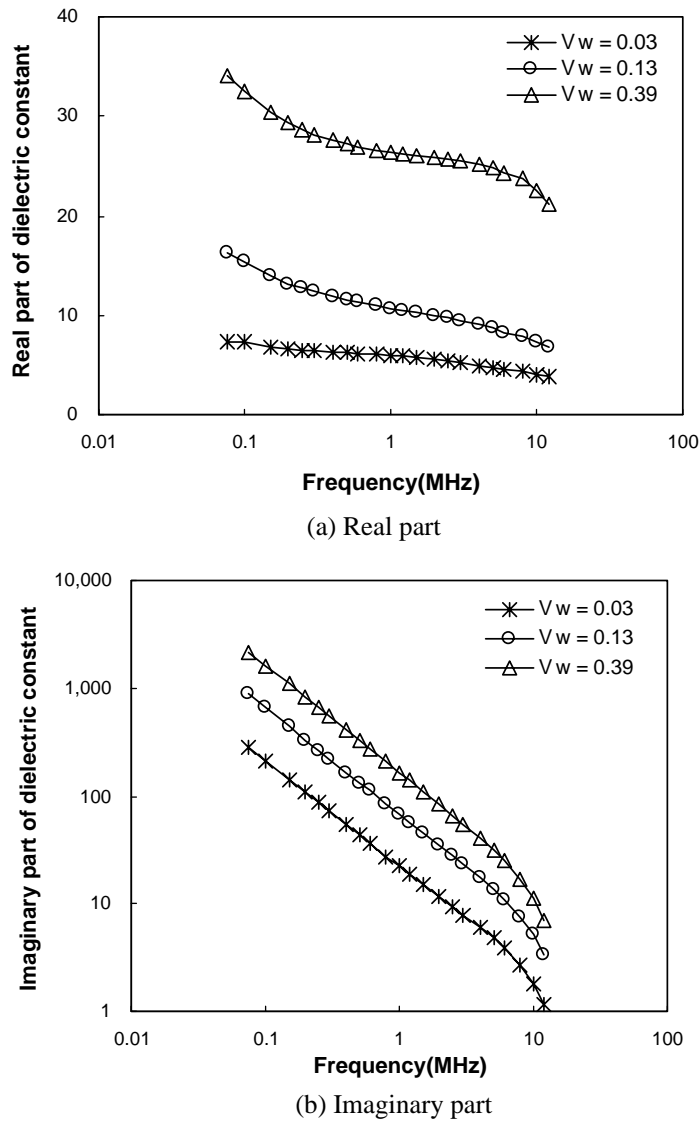


Fig. 3. Dielectric dispersion characteristics of Jumunjin sand with DI water.

to the soil mass volume [15–18]. The test results reported in this paper are based on the volumetric water content (V_w) of the soil sample. Fig. 3 shows the dielectric properties of Jumunjin sand at 0.03, 0.13 and 0.39 of volumetric water contents. The real part of dielectric constant of soil increased with its volumetric water content by the fact that the real part of dielectric constant of water is 78 that is larger than that of soil particle or air [3,19–22]. The increases of the imaginary part of the dielectric constant with volumetric water content can be explained by the increase of the conduction loss caused by the increase of continuous

current path in soil sample. In addition, both real and imaginary part of dielectric constant of soil sharply decrease with frequencies at higher volumetric water content. This result is in agreement with other research. Campbell [3] showed that the decreases in dielectric constant with frequency depend on volumetric water content.

The real and imaginary part of soil dielectric constants had a tendency to decrease with frequency, known as dispersion, that is similar to the reported dielectric behaviors in the literature [3,4]. When an external electric field is applied in soil sample, water molecules (permanent electric dipole) in the soil sample realign in the direction of electric field and cause the orientation polarization. The water molecules, which possess an electric dipole moment, tend to orient themselves in a strong electric field so that the positive poles face the negative electrode and the negative poles face the positive electrode. Also, the negative electron cloud around the oxygen nucleus is deformed or polarized in an electric field. Thus, an induced dipole is formed and adds its moment to that of the permanent dipole. But, as the frequency increases, the dielectric constant decreases because the polarized dipoles do not have enough time to realign and only a small number of water molecules can cause orientation polarization. If the frequency reaches a certain value and the period of electric field gets shorter than the smallest time that the dipoles can rearrange, the dielectric constant of soil is not affected by this polarization [1].

4.2. Dielectric properties with contaminant types

The dielectric behavior as a function of concentration for heavy metals (lead and cadmium) and leachate at 12 MHz are presented in Fig. 4. The real part of the dielectric

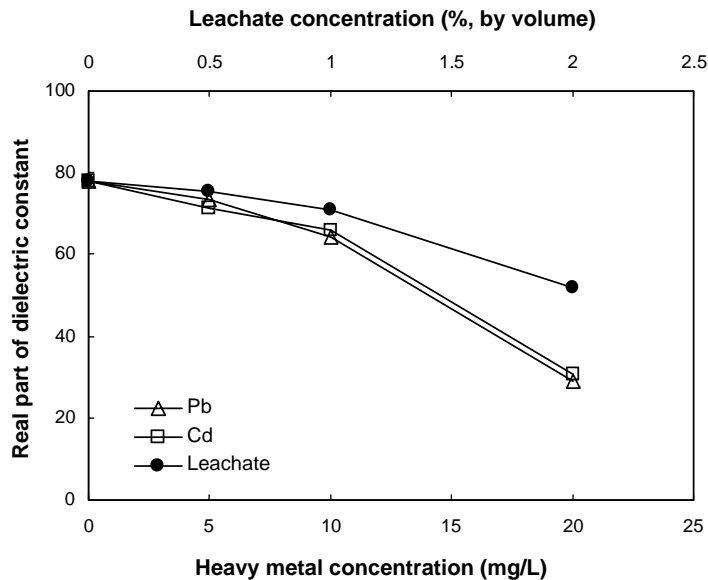


Fig. 4. Real part of dielectric constant of DI water in presence of heavy metals or landfill leachate at 12 MHz.

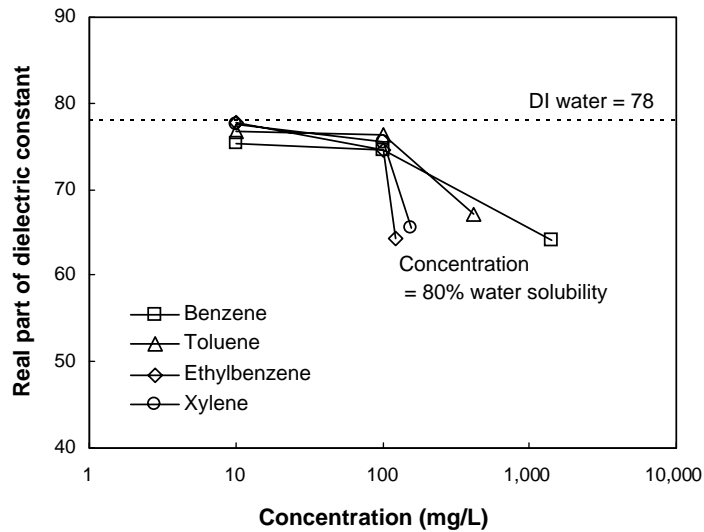


Fig. 5. Real part of dielectric constant of DI water in presence of BTEX at 12 MHz.

constants of three solutions decreased distinctly as their metal concentration increased. These results will provide a useful basis in the detection of subsurface contamination using dielectric properties. Fig. 5 shows the dielectric behaviors of BTEX solutions at three different concentrations, 10 ppm, 100 ppm and 80% (benzene = 1424 ppm, toluene = 412 ppm, ethylbenzene = 121.6 ppm and xylene = 158.4 ppm) of their respective water solubility at 12 MHz. Although the dielectric constants values of pure BTEX are low (benzene: 2.3, toluene: 2.4, ethylbenzene: 3.0 and xylene: 2.4 at 20 °C), the distinct drop of the real part of dielectric constant appeared at the concentration of 80% of their respective water solubility.

Fig. 6 illustrates a comparison of the dielectric dispersion behaviors of various contaminants. The real part of dielectric constant of DI water is almost constant ($\epsilon'_{\text{water}} \approx 78$ at 20 °C) for all measured frequencies. But the real part of dielectric constant of 1424 ppm benzene began to decrease at 3 MHz and was 16% less than that of DI water at 12 MHz. Random motion of benzene molecules in water seems to interrupt water molecules resulting in orientation polarization. The interruption of benzene molecules hardly affects the orientation polarization of water molecules at low frequency because water molecules have enough time to rearrange in direction of electric field.

In Fig. 6, the real part of dielectric constant of lead, cadmium and leachate decreased with increasing metal ion concentration. In the case of ions interacting with water molecules, cations should be attracted to the negative end of the water dipole and anion should attract one of the two protons of the water molecule rendering rotation of the water molecule possible only about the molecular dipole axis; this is orientation polarization. Since orientation around the molecular dipole axis would not give any dipole contribution to the real part of dielectric constant, the contributions of the molecules are fixed, and consequently, the real part of dielectric constant was decreased [1]. Fig. 7 illustrates the interaction between water molecules and heavy metals or anion in leachate. The rotation of water dipole is restricted

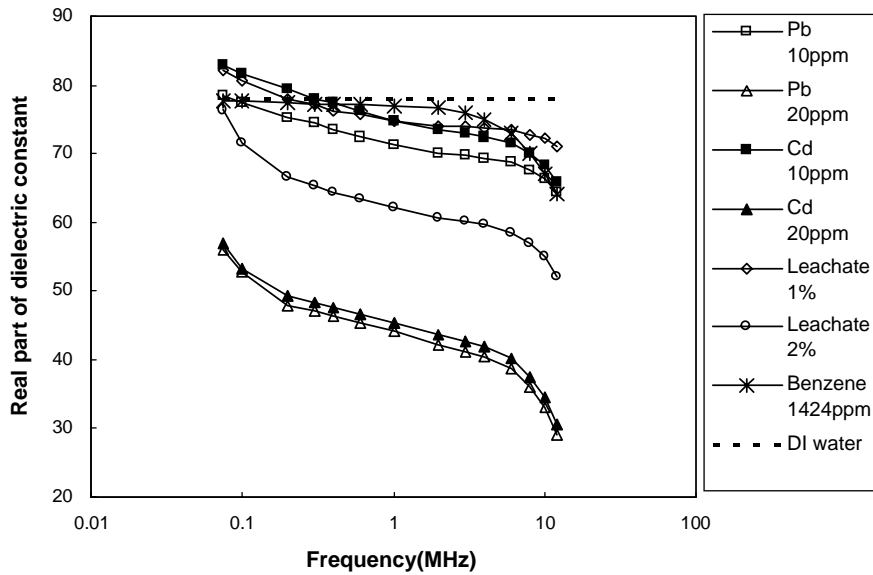


Fig. 6. Real part of dielectric constant of heavy metal, landfill leachate and benzene.

by cationic heavy metals which are attracted to the oxygen of water molecules and chloride ions which are attracted to the hydrogens of water molecules. This condition results in a lower orientation polarization. Consequently, the presence of heavy metals or landfill leachate in water causes lower dielectric constants. Therefore, the significant differences between contaminants and clean water in dielectric constant indicate that the monitoring of dielectric constant has a great potential for investigating the subsurface contamination.

4.3. Dielectric properties of contaminated soil

In this section, the results of dielectric dispersion behavior of contaminated sand are presented. Because the real and imaginary part of dielectric constant of soil is dependent on the volumetric water content of soil, the results are shown by dividing the volumetric water content into unsaturated and saturated part. The real and imaginary parts of the dielectric

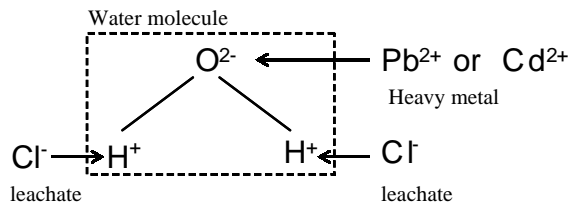
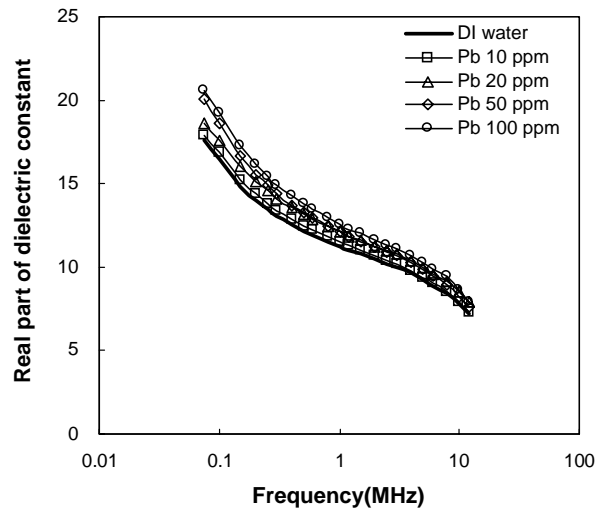


Fig. 7. Illustration of interaction of heavy metals and ionic constituents in leachate with water molecules (modified after Kaya and Fang [1]).

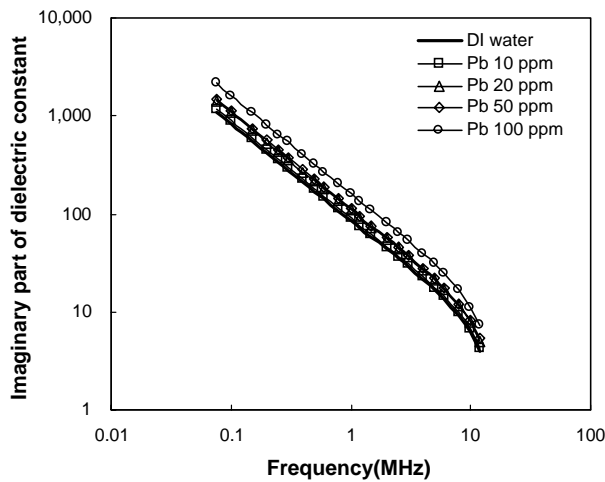
constant were shown to depend on the contamination for soils having same volumetric water content. The analysis of real part of dielectric constant is focused here, the imaginary part of dielectric constant of contaminated sand will be analyzed and discussed in the next section.

4.3.1. Heavy metals

The dielectric dispersion behavior of soil contaminated with lead is presented in Figs. 8 and 9. The results for cadmium showed a similar pattern of dielectric dispersion as well as

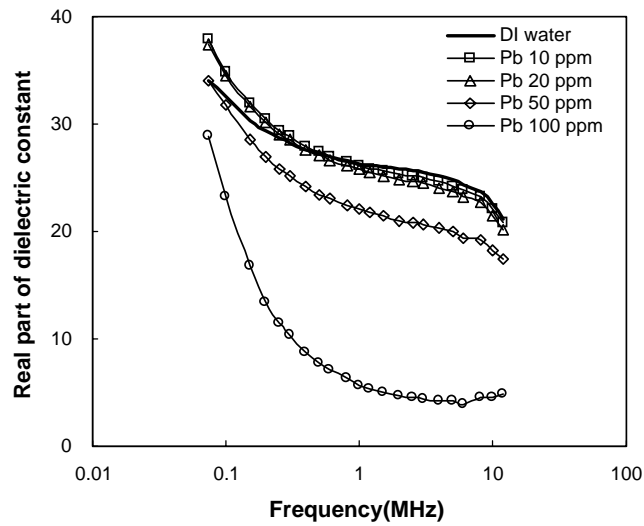


(a) Real part

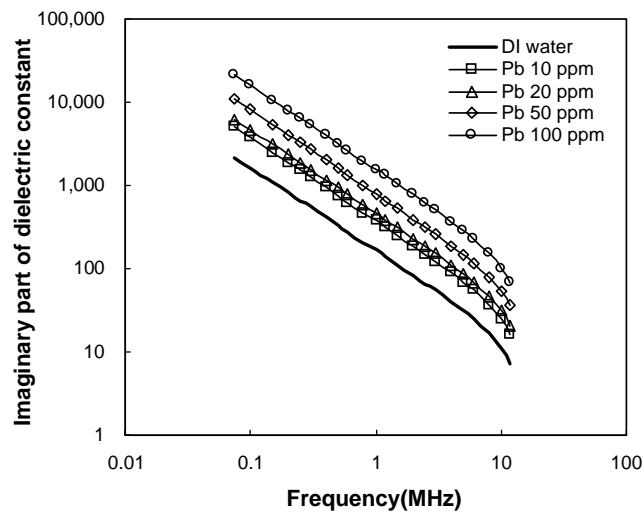


(b) Imaginary part

Fig. 8. Dielectric dispersion of unsaturated sand with lead ($V_w = 0.13$, $S = 0.28$).



(a) Real part



(b) Imaginary part

Fig. 9. Dielectric dispersion of saturated sand with lead ($V_w = 0.39$).

dielectric constants in real and imaginary parts to results for lead. This result could be due to cadmium and lead having the same ionic valence. The graphs of dielectric dispersion of contaminated soil with cadmium are not shown here in this paper.

As the concentrations increases, the imaginary part of dielectric constant increases for the entire measuring frequency range in both saturated and unsaturated soil. The imaginary part of dielectric constant had a larger increase in saturated soil than in unsaturated soil.

However, the real part of dielectric constant in saturated and unsaturated soil showed the opposite results. In the case of unsaturated sand (volumetric water content (V_w) = 0.13, degree of saturation (S) = 0.28, Fig. 8), the addition of heavy metals caused a slight increase of the real part of dielectric constant at low frequency. In the case of saturated sand (V_w = 0.39, Fig. 9), the addition of solutions of heavy metals containing over 50 ppm caused the significant decrease of the real part of dielectric constant although the real part of dielectric constant showed a little increase in the case of 10 and 20 ppm at low frequency (75–500 kHz).

In unsaturated sand, water coexists with air in the pore of soil. Although the ionic interaction of heavy metals interrupts the orientation polarization of water molecules in pore water, space charge polarization is developed in the interface between pore fluid and air or soil particles due to ionic migration. Because the increase in the dielectric constant due to space charge polarization is higher than the decrease in the dielectric constant due to orientation polarization, it is thought that the addition of space charge polarization causes only a slight increase of the soil dielectric constant. However, in saturated sand, water only exists in the pores and the interface between pore fluid and air disappears. Then, the amount of space charge polarization decreases and the decrease of orientation polarization increases due to the increase of heavy metal cations. Therefore, the dielectric constant of contaminated soil decreases as the concentration of heavy metals increase.

4.3.2. Landfill leachate

To analyze the dielectric dispersion behavior of soil containing leachate, 1, 2, 3 and 10% (by volume) of Gimpo municipal landfill leachate diluted in DI water, were added to soil. The dielectric dispersions of sand contaminated with leachate are shown in Figs. 10 and 11.

In unsaturated sand (volumetric water content (V_w) = 0.13, degree of saturation (S) = 0.28, Fig. 10), the addition of leachate caused only a small increase of the real part of the dielectric constant for the same reason as the dielectric behavior of contaminated soil with heavy metals. In saturated sand (V_w = 0.39, Fig. 11), on the basis of 250 kHz, the real part of dielectric constant of the contaminated soil increased at the lower frequency whereas the real part of dielectric constant decreased at the higher frequency as the concentration of leachate increased. The increase of space charge polarization at the lower frequency might be more than with heavy metals, because Cl^- which has higher mobility than that of Pb^{2+} or Cd^{2+} is abundant in leachate. Also, the decrease of orientation polarization is less than the case of heavy metals because the decrease of dipole moment by Cl^- is less than that by Pb^{2+} or Cd^{2+} . Therefore, the dielectric constant of the contaminated soil may increase at a lower frequency as the concentration of leachate increases. However, there is a little increase of space charge polarization at the higher frequency because ions have not sufficient time to migrate. Therefore, the dielectric constant of the contaminated soil may decrease at the higher frequency as the concentration of leachate increases because the decrease of the orientation polarization increases. Additionally, the imaginary part of dielectric constant increased with leachate concentration for complete frequency range and this will be discussed later.

4.3.3. BTEX

Jumunjin sand was mixed with BTEX solution, at 80% of the contaminants respective water solubility (benzene: 1424 ppm, toluene: 412 ppm, ethylbenzene: 121.6 ppm and xylene:

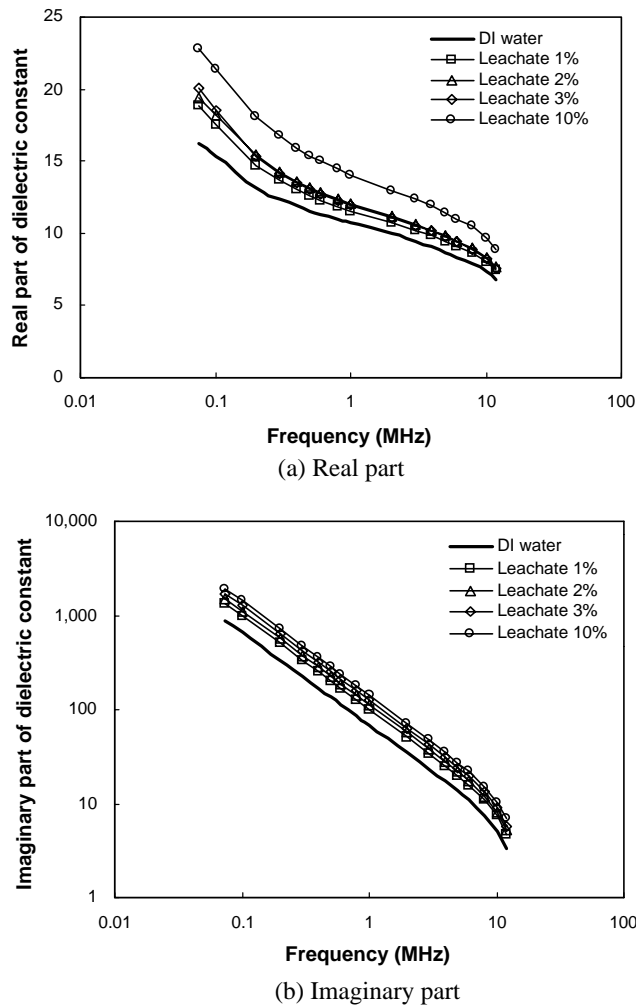


Fig. 10. Dielectric dispersion of unsaturated sand with landfill leachate ($V_w = 0.13$, $S = 0.28$).

158.4 ppm) because the dielectric dispersion behavior of 10 and 100 ppm BTEX solution has not shown a clear difference compared to the dielectric dispersion behavior of DI water. The dielectric dispersion characteristics of contaminated sand with BTEX are shown in Fig. 12. The imaginary part of dielectric constant of contaminated sand with BTEX was not different from that of uncontaminated sand.

In the case of real part of dielectric constant in unsaturated sand (volumetric water content (V_w) = 0.13, degree of saturation (S) = 0.28), the addition of BTEX had little effect because the number of BTEX molecules was small, although the real part of dielectric constant slightly increased at a low frequency (75–300 kHz). In saturated sand ($V_w = 0.39$), the real part of dielectric constant of the contaminated soil was from 6% (at 75 kHz) to 12% (at

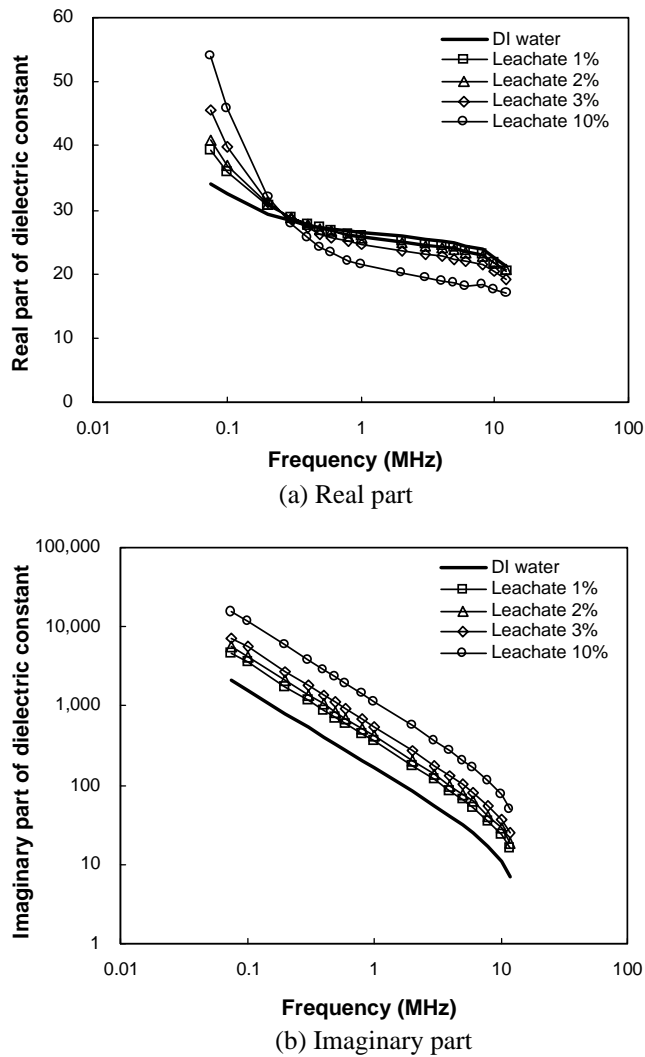
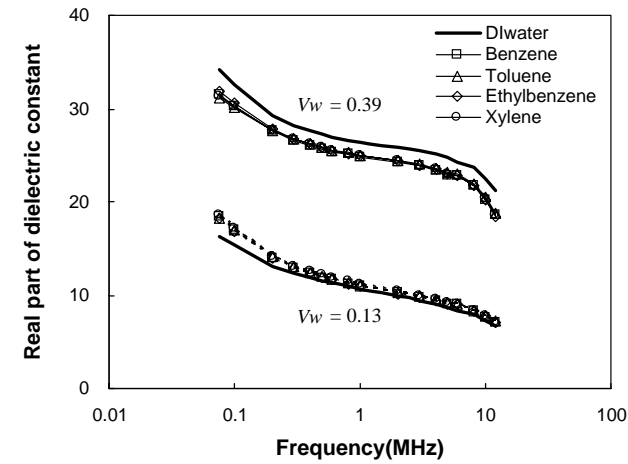


Fig. 11. Dielectric dispersion of saturated sand with landfill leachate ($V_w = 0.39$).

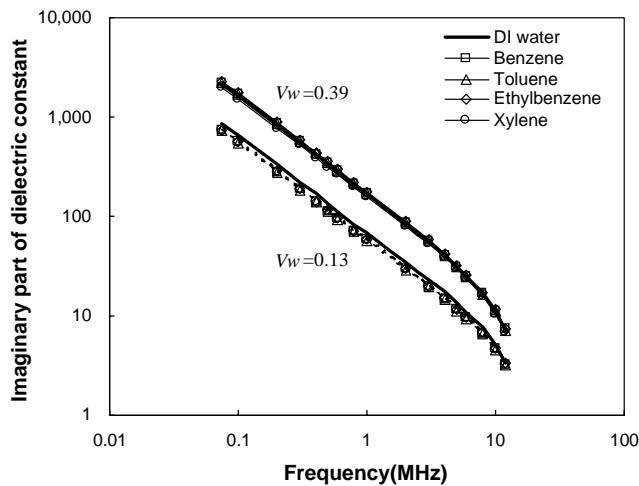
12 MHz) less than that of soil with DI water over the whole measuring frequency range due to the interruption of BTEX molecules against the orientation polarization of water molecules. However, there was not much difference between the dielectric behavior of soil with DI water and the dielectric behavior of the contaminated soil in spite of high concentration of BTEX.

4.4. Imaginary part of dielectric constant

The imaginary part of dielectric constant represents energy losses. There are a number of possible mechanisms for energy losses in soil including charged double layers,



(a) Real part

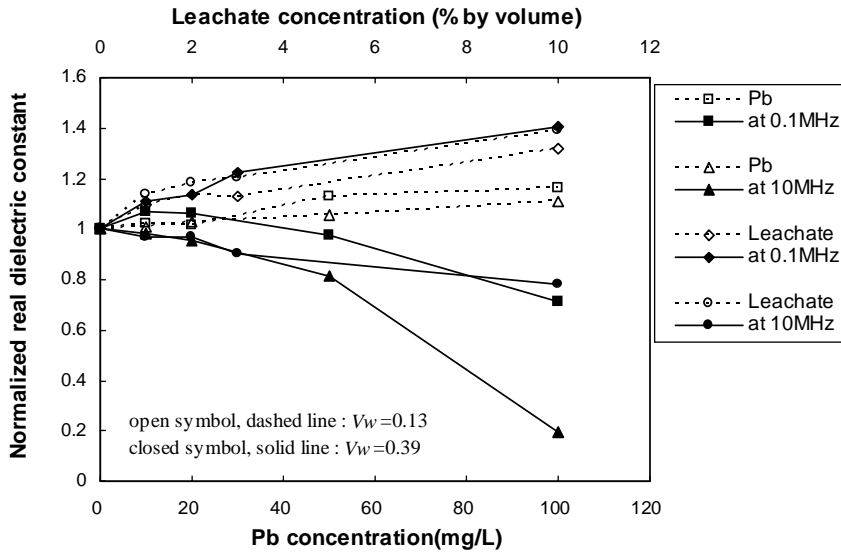


(b) Imaginary part

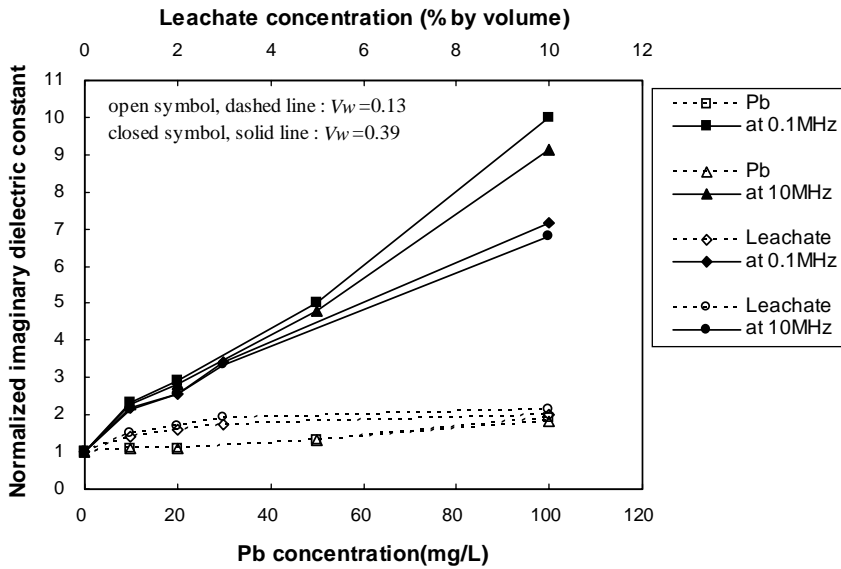
Fig. 12. Dielectric dispersion of sand with BTEX at concentration of their 80% water solubility.

Maxwell–Wagner effect, surface conductivity, bound-water relaxation and ionic conductivity [3]. Ionic conductivity might be the most proper mechanism to explain the results of this study using Jumunjin sand. As the ionic conductivity increases, electrical conduction in sample is enhanced. Ions will move under an externally applied electrical field if there is enough time for them to orient themselves in the direction of applied electric field [1]. Consequently, amount of energy lost in the measurement of dielectric constant is increased, and the imaginary part of the dielectric constant is increased.

The imaginary part of the dielectric constant of contaminated sand with both lead and leachate increased with their concentration for whole frequency range at both unsaturated



(a) Normalized real part of dielectric constant ($\epsilon'_{cont.} / \epsilon'_{uncont.}$)



(b) Normalized imaginary part of dielectric constant ($\epsilon''_{cont.} / \epsilon''_{uncont.}$)

Fig. 13. Normalized real and imaginary part of dielectric constant of sand with lead and leachate with their concentration at 0.1 and 10 MHz.

and saturated conditions. These results are caused by the increase of energy loss due to the enhancement of conduction in soil with ionic concentration. In addition, the values of imaginary part of dielectric constant in unsaturated sand (Figs. 8 and 10) was lower than that in saturated sand (Figs. 9 and 11) because the ion constituents mobility is restricted due to a reduction in the long-range connectivity of pore water at low volumetric water content [3].

BTEX cannot enhance conduction in soil because BTEX compounds are nonconductive materials whereas heavy metals and leachate are conductive materials. Therefore, conduction loss of contaminated soil with BTEX was not changed compared to that of soil with DI water. As shown in Fig. 12, the imaginary part of dielectric constant of soil with BTEX was not greatly different from that of soil with DI water for both unsaturated and saturated condition over whole frequency range utilized.

To evaluate the effectiveness of detecting contaminants such as heavy metals and leachate as a function of their concentration using dielectric constants, the normalized values, defined as the ratio of dielectric constant of the contaminated soil to that of uncontaminated soil, with the concentration of lead or leachate are shown in Fig. 13.

In Fig. 13(a), the variations of real part of dielectric constant with concentration did not clearly characterize the contamination at low concentrations of lead and leachate in both unsaturated and saturated sand and the behavior of the real part of the dielectric constant with concentration was frequency dependent. For example, in the case of saturated sand with leachate, the normalized value of the dielectric constant increased at 0.1 MHz whereas it decreased with concentration at 10 MHz. This result indicates that the measurement of real part of dielectric constant at a single frequency leads to false information about the subsurface contamination. However, in Fig. 13(b), the imaginary part of dielectric constant clearly increased, especially in saturated sand. Although, in unsaturated sand, the increment of normalized value of the imaginary part of the dielectric constant sand was small, the increase of imaginary part of dielectric constant with concentration was clearer than that of real part of dielectric constant.

In conclusion, the important findings shown in Fig. 13 follows. Measurement of real part of dielectric constant alone will lead to some degree of ambiguity in the results. To overcome the ambiguity in measuring only the real part of the dielectric constant, an additional measurement and analysis for the imaginary part of dielectric constant is required to characterize subsurface contamination besides real part of dielectric constant, especially in the case of ionic contaminants. The analysis of dielectric dispersion behavior for a wide frequency range is recommended because of the frequency dependent characteristics of the dielectric constant. Additionally, because the complex (both real part and imaginary part) dielectric constant is dependent on the volumetric water content, to utilize the dielectric measurement method for investigating subsurface contamination, the evaluation of volumetric water content of subsurface is also required.

5. Conclusions

The following conclusions are drawn from the analysis of complex (both real part and imaginary part) dielectric behavior of contaminated sand with heavy metal, landfill leachate and BTEX based on our experimental laboratory work.

1. In the case of contaminated sand with heavy metal or leachate, the real part of dielectric constant in unsaturated soil slightly increased due to the development of space charge polarization. However, the real part of dielectric constant in saturated soil significantly decreased (except for leachate) due to the decrease of orientation polarization. In the case of BTEX at high concentrations, the real part of dielectric constant of saturated sand slightly decreased due to interruption of orientation polarization of water molecules by BTEX molecules, whereas the real part of dielectric constant in unsaturated soil remains constant.

The distinct difference of the dielectric behavior with contaminant types, such as heavy metal, landfill leachate and BTEX, implies that the monitoring of complex dielectric constant has a potential to evaluate contaminants in the subsurface.

2. In saturated contaminated sand with leachate, on the basis of 250 kHz, the real part of dielectric constant of the contaminated soil increased at the lower frequency whereas the real part of dielectric constant decreased at the higher frequency as the leachate concentration increased. When the dielectric constant method is applied to the investigation of subsurface contamination, the measurement of dielectric constant at one frequency can yield unreliable information about the contaminant. Therefore, one must evaluate the dispersive behavior of real part of dielectric constant for wide frequency range.
3. The imaginary part of dielectric constant of soil contaminated with lead or leachate significantly increased with concentration of the contaminants in both unsaturated and saturated media for the entire frequency range, whereas the real part of the dielectric constant gives somewhat ambiguous results at low concentrations. The increases of the imaginary part of the dielectric constant can be explained by the ionic conductivity loss mechanism. Therefore, the additional measurement and analysis for the imaginary part of the dielectric constant in addition to the real part of the dielectric constant is needed to obtain accurate information about the state of ionic contaminants in subsurface.

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References

- [1] A. Kaya, H.Y. Fang, Identification of contaminated soils by dielectric constant and electrical conductivity, *J. Environ. Eng.* 123 (1997) 169–177.
- [2] S. Thevenayagam, Environmental soil characterization using electric dispersion, in: *Proceedings of the ASCE Special Conference of the Geoenvironment 2000*, ASCE, New York, NY, 1995, pp. 137–150.
- [3] J.E. Campbell, Dielectric properties and influence of conductivity in soils at one to fifty megahertz, *Soil Sci. Soc. Am. J.* 54 (1990) 332–341.
- [4] J.K. Mitchell, K. Arulanandan, Electrical dispersion in relation to soil structure, *J. Soil Mech. Found. Div.* 94 (1968) 447–471.
- [5] J.R. Lundien, Laboratory measurement of electromagnetic propagation constants in 1.0–1.5-GHz microwave special region, Report 5, *Terrain Analysis by Electromagnetic Means*, Technical Report No. 3-693, US Army Engineer Waterways Experiment Station, 1971.

- [6] K. Arulanandan, S.S. Smith, Electrical dispersion in relation to soil structure, *J. Soil Mech. Found. Div.* 99 (1973) 1113–1133.
- [7] S. Thevenayagam, Frequency-domain analysis of electrical dispersion of soils, *J. Geotech. Eng.* 121 (1995) 618–628.
- [8] M. Carrier, K. Soga, Dielectric measurements of clay as a potential method of contaminant detection, in: *Proceeding of the Second Material Congress on Environmental Geotechnics*, Osaka, Japan, 1998, pp. 491–496.
- [9] S. Darayan, C. Liu, L.C. Shen, D. Shattuck, Measurement of electrical properties of contaminated soil, *Geophys. Prospect.* 46 (1998) 477–488.
- [10] M. Carrier, K. Soga, A Four Terminal Measurement System for the Investigation of the Dielectric Properties of Clay at Low Frequencies, *Geoenvironmental Engineering*, Thomas Telford, London, 1997, pp. 3–10.
- [11] J.Q. Shang, J.W. Scholte, R.K. Rowe, Multiple linear regression of complex permittivity of a till at frequency range from 200 to 400 MHz, *Subsurf. Sens. Technol. Appl.* 1 (2000) 337–356.
- [12] A.R. Von Hippel, *Dielectric Materials and Applications*, Wiley, New York, 1954.
- [13] P.B. Bedient, H.S. Rifai, C.J. Newell, *Groundwater Contamination*, PTR Prentice-Hall, New Jersey, 1994, p. 89.
- [14] H.Y. Fang, *Introduction to Environmental Geotechnology*, CRC Press, Boca Raton, Florida, 1997.
- [15] G.C. Topp, J.L. Davis, A.P. Annan, Electromagnetic determination of soil water content: measurements in coaxial transmission lines, *Water Resources Res.* 16 (1980) 574–582.
- [16] M. Ansoult, L.W. De Backer, M. Declercq, Statistical relationship between apparent dielectric constant and water content in porous media, *Soil Sci. Soc. Am. J.* 49 (1985) 47–50.
- [17] D.A. Robinson, C.M.K. Gardner, J.D. Cooper, Measurement of relative permittivity in sandy soils using TDR, capacitance and theta probes: comparison, including the effects of bulk soil electrical conductivity, *J. Hydrol.* 223 (1999) 198–211.
- [18] J.O. Curtis, Moisture effects on the dielectric properties of soils, *IEEE Trans. Geosci. Remote Sens.* 39 (2001) 125–128.
- [19] H. Ella, A. Denote, A capacitive soil moisture sensor, *J. Hydrol.* 185 (1996) 137–146.
- [20] J.O. Curtis, R. Narayanan, Effects of laboratory procedures on soil electrical property measurements, *IEEE Trans. Instrum. Meas.* 47 (1998) 1474–1480.
- [21] C.M.K. Gardner, T.J. Dean, J.D. Cooper, Soil water content measurement with a high-frequency capacitance sensor, *J. Agric. Eng. Res.* 71 (1998) 395–403.
- [22] T. Saarenketo, Electrical properties of water in clay and silty soils, *J. Appl. Geophys.* 40 (1998) 73–88.