Electrical properties speculation of contamination by water and gasoline on sand and clay composite

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ABSTRACT

Effects of temperature, frequency, and molarity on electrical conductivity have been studied for sand-clay samples contaminated by water and gasoline. Electrical properties changes according to frequency, contaminant type and structure of components at the sample. A comparison was performed for these contaminated sand-clay samples. Changes of dielectric constant ($\in'$), conductivity (σ) and complex impedance with frequency (0.1 Hz 2×10^{7} Hz), with different contaminant concentration (at constant room temperature ~21°C) have been studied. The sand-clay samples were mutually wetted gradually using gasoline and distilled water. Then, electrical properties were measured sequentially. Water is a conductive liquid and the contaminant gasoline is an insulator. The experimental results indicate that, conductivity of samples increases with increase of water concentration while with the additions of the

contaminant gasoline, the sample conductivity decreases. The permittivity, decreases with reduction of conductive links between grains and with the progressive increase of frequency. The conduction of electrical conductivity, commonly, increases with increase of connectivity between different links between grains. Also, conduction increases with progressive increase of frequency. Comparison of both outcomes from lab electrical measurements gives a speculation about the picture in the field.

Keywords: Electrical properties; conductivity; water saturation; gasoline saturation; permittivity; frequency.

INTRODUCTION

There are many physical, chemical and mechanical properties that can affect the electrical properties. These parameters may be grain shape, size, orientations, interface boundaries, porosity, inhomogeneity, combination made by mixing substances together, and structural interconnected pores of the conducting and insulating fractions of the grains ... etc. Also, wetting the samples with liquid is an efficient factor for the changing of electrical characteristics (Gomaa and Abou El-Anwar 2015). The systematic investigation of ac conductivity at various temperatures and frequencies gives valuable information on conduction mechanisms at samples according to its electric charge carriers (Gomaa 2013). In general, the gradual additions of conductor concentration raise the conductivity value (Abou El-Anwar and Gomaa 2016). Also, the gradual additions of conductor concentration decrease the distance spaces separating two grains and, accordingly, raises the

permittivity values (Gomaa and Elsayed 2009). The latter increase of conductivity and permittivity is present until the samples reach the percolation concentration threshold (Gomaa 2008). Percolation threshold of concentration is the conductor concentration fraction that permits complete contact between electrodes.

Increase of conductivity is the outcome of growing of uninterrupted conduction links of conducting elements (water). The conduction values of conductivity are faint at relatively low overall conductor amount. With progressive increase of overall conductor amount the conductivity becomes greater (Gomaa et al. 2018, Gomaa and Kassab 2017). Generally, with increase of overall conductor amount and with progressive increase of frequency more continuous conductor links start to be established between the different grains (Gomaa et al., 2009). Increase of continuous conductor links leads to the general increase of conductivity values (Knight and Endres 1990). The raise of frequency initiates particles to overcome energy barriers between atoms and therefore the conductivity value is increased. The growing of frequency makes the permittivity decreases. Also, decrease of permittivity is initiated from decrease of conductor concentration. When the conductor concentration decreases, this leads, generally, to increase of insulator material along the space separating the conducting grains. Permittivity values increase with increase of total conductor concentration until it achieves the dispersion limit. At that limit, the permittivity may decrease again above that percolation limit (Gomaa and Alikaj 2009, Gomaa and Abou El-Anwar 2017). The small pores

among grains result in relative decrease of permittivity in samples (Barabanova et al. 2013). Permittivity changes from one specimen to another due to alteration of combinations, volumes and amounts of the conducting and insulating materials in specimen. Also, there may be the texture between grains (Gomaa et al., 2015 a and b). Conductivity, in general, has a monotonic attitude of increase with the successive increase of frequency. The permittivity, in general, has a tendency to increase with decrease of the frequency (Kassab et al. 2017, Gomaa 2006).

Frequency behavior of conductivity is may resulted from orientation and translational hopping mechanisms. Orientation mechanism means hopping and jumping of electron in the space separating two charged defects. The translation hopping means that there is D C conductivity at zero frequency. Imaginary impedance, which depends on frequency, point to the existence of different reactions in the system.

Comparing effect of water as a conductor and that of gasoline as an insulator, with both frequency and wetting at lab was done in this work. To achieve this study, a sand-clay sample was measured (electrically) at laboratory. The samples was measured dry and at different contamination saturations, of water and gasoline, at constant room temperature (~21 °C), at frequency range of (0.1 Hz up 2×10^{7} Hz). Electrical measurements; permittivity, conductivity and complex impedance compared to frequency, declare effect of contaminants and texture on the studied samples. The water is conductive liquid, because miscibility of salts found in its structure and the

gasoline is insulator (Abou El-Anwar and Gomaa 2013). In the laboratory, conductivity of samples increases with sample's water content while with additions of gasoline the sample conductivity decreases. Wetting the samples with liquid is effective factor in the change of electrical responses (Gomaa and Kassab M., 2016**)**.

In the current paper we will compare both outcomes from electrical measurements from different contaminants (water and gasoline) and we will discuss wetting conditions of water, gasoline and frequency on electrical laboratory measurements.

Experimental Details

Electrical measurements

Dielectric was measured using broadband dielectric spectrometer concept, Novo Control, Germany, at wide frequency range (0.1 Hz 2×10^{7} Hz). We apply testing voltage of 1 Vrms at constant room temperature (~21 ^{0}C). Sample constituents are some small sand grains that are naturally mixed with some clay impurities. The disc shaped sample holder has dimensions of 8 mm diameter and 3.623 mm height with total volume of 0.182 cm^{3}. Data were automatically measured and studied by the instrument software WinDeta. The sample was enclosed between two copper electrodes and a Teflon cylinder. The sample then placed in its proper position inside the instrument. Measurements were done in a row, pure sand-clay mixture and then 10 μL of filtered water or gasoline was added to sample and measurement repeated. The last step was repeated until the water volume in the specimen reaches 60 μL of filtered

water. The sample sand cell is present at Fig. (1), 1: is the Teflon cylinder and 2: indicates the two copper electrodes. Dielectric constant was calculated by the formula $\in' = \frac{C_p d}{\epsilon_0 A}$ (Jonscher 1973, 1975, 1977 and 1999, Hill and Jonscher 1983). The ac conductivity is given by $\sigma = w \in' \epsilon_0$ (Hill and Jonscher 1983), where w is the angular frequency.

Results and Discussion

Electrical properties of any mixture of samples are affected by:

1- The change of sample arrangement during pumping of the water or gasoline. This may render inaccurate measurements as the field sample has changed in its structure. This gives misleading data. Consequently, it would be better to remove washout of the measured samples.

2- Using samples of large volume is suitable for electrical field measurements. However, this may contradict the condition for laboratory electrical measurements which is instituted on using of small volumes with large areas. Physically, the laboratory sample must have small thickness to form electric field with proper strength to minimize errors.

1) Chemical assembly of gasoline

Gasoline is a reiterated component of a liquid mixture of hydrocarbons. It is consisted of mixtures of hydrocarbons, blending agents, and additives. Commonly, the chemical constituents of gasoline hydrocarbons changes extensively (IARC 1989). This dependence is affected by crude oils concentrations, the filter procedure used, and the product precise requirement.

The gasoline composition range from 6 to 13 % of alkanes; 25 to 40 % iso-alkanes; 3 to 7 % cyclo-alkanes; l to 4 % cyclo-alkenes; and from 20 to 50 % of overall aromatics (0.5-2.5% benzene). Detailed information regarding the Chemical analysis, particle size analysis, hydrogeological properties are located at Table (l). The sand used is identified by the properties at Table (l).

Table 1 - Chemical analysis, Particle size analysis, Hydrogeological properties

Chemical analysis results										
	SiO_2	TiO_2	Al_2O_3	MnO	Fe_2O_3	MgO	CaO	Na_2O	K_2O	CO_2
%	81.17	0.06	4.75	0.05	0.56	0.13	6.67	0.83	1.15	4.02

Particle size analysis of sand							
d (mm)	0 – 0.074	0.75 - 0.104	0.105– 0.149	0.150– 0.420	0.421– 0.840	0.841– 2.000	>2.001
%	0.8	0.03	0.20	14.27	70.67	10.6	3.43

Hydrogeological properties		
d (mm)	K_{max} (m/s)	ρ (%)
0.09	$5*10^{-3}$	35

2) Laboratory electrical measurements

Figure (2) shows the series resistance (R_s) with frequency spectrum and distinct concentrations of filtered water (10 µL, 20 µL, 30 µL, 40 µL, 50 µL and 60 µL). The dry sand-clay samples have the highest resistance (Song et al. 1986, Leroy et al., 2008, Chew and Sen 1982, Chelidze and Guéguen 1999). With the consecutive changes of disttled water, the resistance decreases. Series resistance steeps down when adding the 1st and 2nd successive additions of disttled water. The decrease is slowly with the 3rd, 4th and 5th successive additions. The last successive addition, 6th dose, had a mild decrement reaching

minimum values of sreries resistance for sand-clay composite (e. g. Chelidze et al. 1999, Sen 1984, Sen 1981, Olhoeft 1977). This is nearly the full saturation of sand-clay sample. The sample series resistance descended three decades with successive additions from dry to full saturation (Shaltout et al, 2012, Gomaa 2009). When specimen is dry (0 μL), decrease of the resistace value with frequency is fast and requires nearly more than one decade for the whole frequency range (Olhoeft 1976, Olhoeft 1980, Olhoeft 1985). When specimen is wetted with the other successive additions of water (10 μL, 20 μL, 30 μL, 40 μL, 50 μL and 60 μL), decrease of the resistace with frequency is slow and takes nearly half decade in the whole frequency range (Minaw et al. 1972, Levitskaya and Sternberg 1996 a, b, Levitskaya 1984).

The dry sample does not reach saturated (flat) resistance value, even at very high frequency (2×10^{7} Hz), while the other wet samples (10 μL, 20 μL, 30 μL, 40 μL, 50 μL and 60 μL) reaches the saturated (flat) resistance value at relatively lower frequencies (10^{3} Hz), and differs gradually from one saturation to the other (Gomaa and Elsayed 2006, Gomaa et al. 2000). As the satuartion concentration increases, the saturated (flat) resistance value at definite frequency is decreased (Mendelson and Cohen 1982, Last and Thouless 1971, Knight and Nur 1987). The sample series resistance nearly takes three decades from the dry condition to the full saturation in the whole frequency range.

Figure (3) displays the series resistance (R_s) with frequency dry and at various saturations of gasoline. The dry sand-clay samples have the lowest resistance. Series resistance (R_s) increases with adding gasoline concentrations

(10 μL, 20 μL, 30 μL, 40 μL, 50 μL and 60 μL).. Subsequent gasoline additions are quite different from water additives. The series resistance is directly related to gasoline addition. However, the whole of growth of the series resistance was within one decade at the whole frequency range. Series resistance raised with raise of gasoline concentration (Jonscher 1973, 1975, 1977 and 1999, Hill and Jonscher 1983). The dry sample has very low resistance while successive gasoline concentration increases the resistance sequentially. The series resistace of the dry sample ($0\mu L$) decreases with frequency increase (Garrouch and Sharma 1994, Dias 2000, Chinh 2000). The variation of the series resistance, nearly, takes more than one decade in the whole frequency range. When specimen is contaminated with gasoline (10 μL, 20 μL, 30 μL, 40 μL, 50 μL and 60 μL), the resistace is higher. Resistace is raised with raise of the gasoline concentration with more faster degree (with frequency), consuming nearly two decades in the whole frequency range for high concentration (60 μL). All samples does not accomplish saturation resistance of frequency even at very high frequency (2×10^{7} Hz). The curves will not reach that saturation resistance frequency (with frequency increase, the resistance is still decreasing, Chelidze 1979, Knight and Abad 1995). Clear difference is found between the series resistace value at Fig. (2) ($2X10^{5}$) for high water saturation and that value of series resistace at Fig. (3) ($1X10^{6}$) for high gasoline saturation. This difference is completely attached to change of the contaminent (Bussian 1983, Khalafalla and Maegley 1973). The gasoline saturation is quitely increases the resistance and water saturation is quitely decreases the resistance.

Fig. (4) Shows the permittivity (ε') with frequency dry and at various concentrations of filtered water (10 μL, 20 μL, 30 μL, 40 μL, 50 μL and 60 μL). The sucsessive additions of water concentrations increases the value of sample permittivity by nearly one decade, at low frequency, and this value decreases at relatively high frequency. At low frequency, permittivity (ε') has relative high values that decrease with frequency increase and finally at relatively high frequencies becomes independent of frequency. The the permittivity decreases rapidly at relatively lower frequencies and decreases slowly at relatively higher frequencies. Normal permittivity (ε') behaviourshow that there values decreases with increasing frequency untell it reacches a constant value. Above definite frequency range, the exchange interaction of electron between the ions cannot keep track of applied electric field. At relatively high frequency regions, the charge carriers would barely have started to move before the field reversal occurs and the permittivity ε' falls to its smallest values. Space charge polarization arises between grain interfaces and grain-electrode interface due to inhomogenety of permittivity structure at these interfaces. This is what is called double layer; semi-conducting grains separated by insulating or poor conducting grains. The permittivity of a material comes from electronic, ionic and space charge accumulation. The electronic contribution is the main factor that are affected by the present frequency range, while at other frequency ranges other mechanisms may be the dominant factors.

Figure (5) Shows the permittivity (ε') with frequency, dry and at various saturations of gasoline. The sucsessive additions of gasoline decreases the value of sample permittivity by less than one decade, for lower frequency ranges.

Figure (6) shows the complex series impedance (Z_s) for the sand-clay sample dry and at various concentrations of filtered water (10 μL, 20 μL, 30 μL, 40 μL, 50 μL and 60 μL). For the dry condition case, measured impedance shows nearly an arc (or a part of another semicircle, for lower frequencies) in contact with another semicircle (at relatively higher frequency). This indicates two different polarization mechanisms within the specimens. First semicircle (at high frequency) shows the properties of specimen materials (bulk property). The other arc or the other part of semicircle (at low frequency) shows the properties of interface (grain boundary) between the grains. This may be resulted from transport of charges across the grain boundaries. Complex impedance was gradually decreasing with frequency increase which resulted from space charge increase at relatively higher frequencies. With increase of conductor concentration (distilled water), the relaxation peak proceeds to higher frequency and its strength decreases. This effect that is similar in behaviour, more or less, to rise of temperature.

Figure (7) shows the complex parallel impedance (Zp) for the sand-clay samples, dry and at various saturations of gasoline (10 μL, 20 μL, 30 μL, 40 μL, 50 μL and 60 μL). For dry sample, measured impedance plane shows nearly an arc (at low frequency) that has low impedance than the other

saturated curves. This arc describes the total conductor connections at the specimen. Also, we can find two different polarization mechanisms within the samples (bulk at relatively higher frequency and grain boundary at relatively low frequencies). As gasoline concentration increases, the impedance peaks move to high values of complex impedances. Also, sequential additions of gasoline concentrations cause the peaks widths to be broader and more expanded. The expansion of peaks means that the barriers that the ions have to overcome, due to increment of resistivity, become bigger with the additions of gasoline concentrations. As insulator contaminant increases (gasoline), the relaxation peak proceeds to lower frequencies and increases in its strength and become broader.

Figure (8) displays the Nyquist representation of conductivity of sand-clay with different amounts of water concentrations. Figure (8) can be compared with figure (6), for impedance representation.

Figure (9) displays the Nyquist representation of conductivity of sand-clay with different amounts of gasoline concentrations (10 μL, 20 μL, 30 μL, 40 μL, 50 μL and 60 μL). The behavior of the samples in figure (9) can be compared with the behavior of figure (7), for impedance representation.

The water contamination decreases the whole sample impedances. The gasoline contamination increases the the whole sample impedances.

3) CONCLUSION

In the laboratory, conductivity of samples increases with adding water concentration while with the additions of gasoline causes a conductivity to decrease. Electrical conduction, in general, is raised with rise of overall conductor continuous links and with frequency increase. There is a link between hydrocarbons contamination and geophysical signal and the electrical resistivity. The bulk conductivity observed in contaminated samples is linked to contaminant fluid conductivity and its amount and consequently changes according to the type of contaminant. So we could infer that type of contaminant is important at altering geophysical properties of contaminated sediments. Hence, influence of type of contaminant on physical properties cannot be ignored in geophysical contaminated sites. Frequency effect of conductivity is reflected from orientation and translation hopping. The dependence of impedance on frequency indicates distribution of relaxation in the samples. Imaginary impedance peaks are shifted to higher frequency side with increase of filtered water. Finally, the electrical measurements are good support for analysis to define qualitatively the type of contaminant.

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Table Caption

Table (1) Chemical analysis, Particle size analysis, Hydrogeological properties (d_m: average diameter; K_{max}: hydraulic permeability; r: porosity).

Figure Caption

Fig (1) Displaying sample cell, where 1 is a Teflon cylinder a 2 are the two electrodes enclosing the sample.

Fig. (2) Shows the series resistance (R_s) as a function of frequency dry and at different saturations of distilled water.

Fig. (3) Shows the series resistance (R_s) as a function of frequency dry and at different saturations of gasoline.

Fig. (4) Shows the permittivity (ε') as a function of frequency dry and at different saturations of distilled water.

Fig. (5) Shows the permittivity (ε') as a function of frequency dry and at different saturations of gasoline.

Fig. (6) Shows the complex series impedance (Zs) for the sand-clay sample dry and at different saturations of distilled water.

Fig. (7) Encloses the complex series impedance (Zp) for the sand-clay sample dry and at different saturations of gasoline.

Fig. (8) displays the Nyquist plot of the conductivity of sand-clay as a function of different amounts of water.

Fig. (9) displays the Nyquist plot of the conductivity of sand-clay as a function of different amounts of Gasoline.

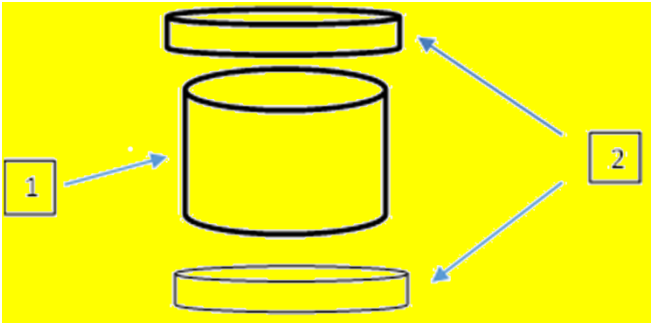
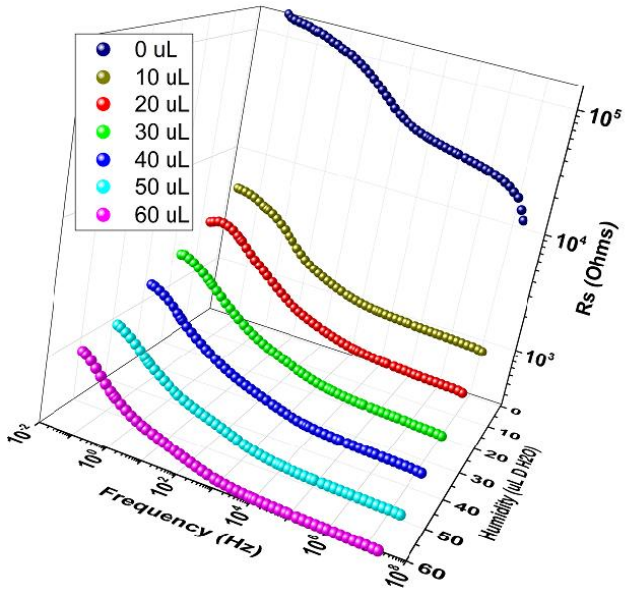


Fig. (1) Show sample cell holder, 1) is the Teflon cylinder and 2) two electrodes enclosing the sample.



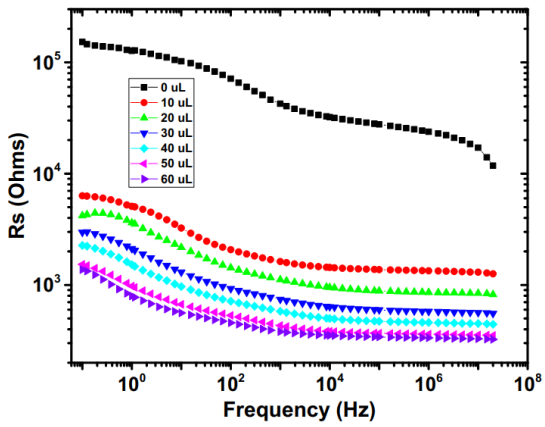
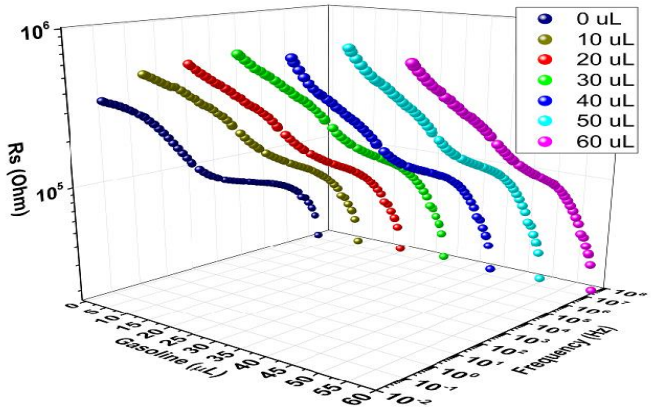


Fig. (2) Shows the series resistance (R_s) with frequency, dry and at various saturations of distilled water.



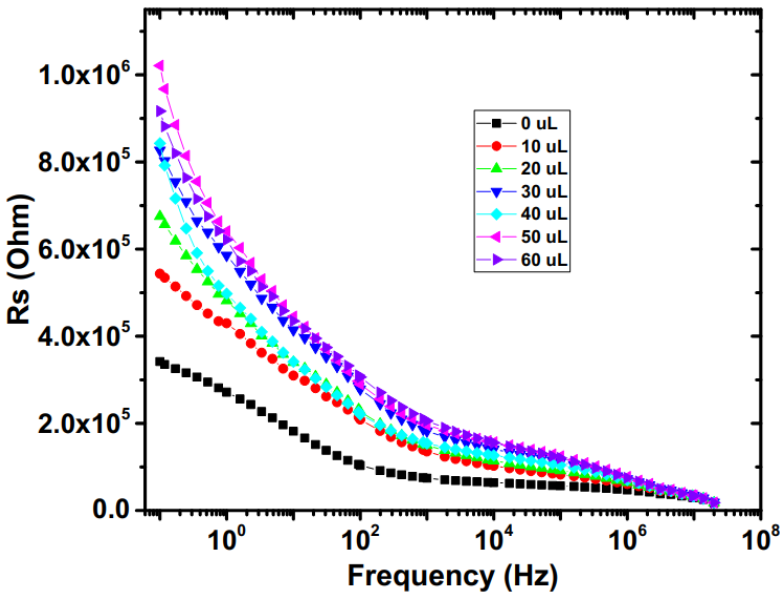
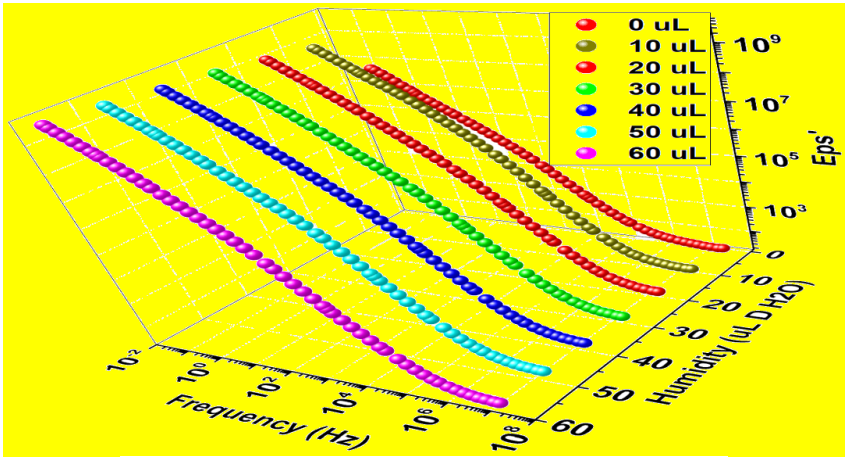


Fig. (3) Shows the series resistance (R_s) with frequency, dry and at various saturations of gasoline.



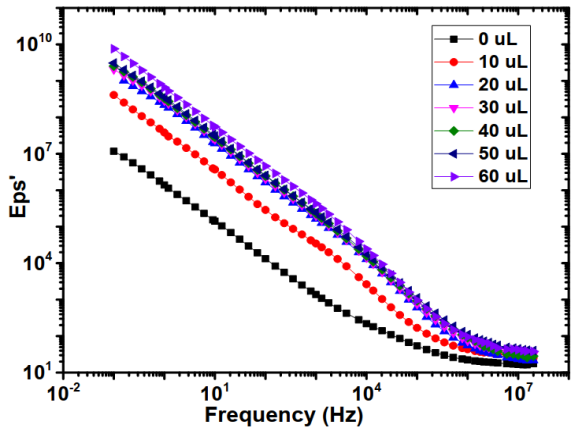


Fig. (4) Shows the permittivity (ε') with frequency, dry and at various saturations of distilled water.

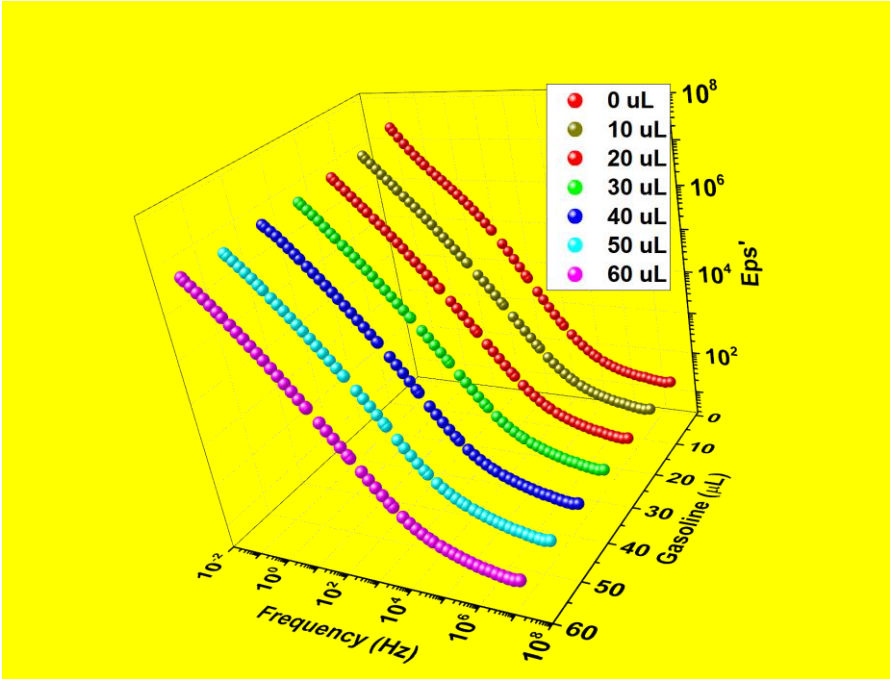


Fig. (5) Shows the permittivity (ε') with frequency, dry and at various saturations of gasoline.

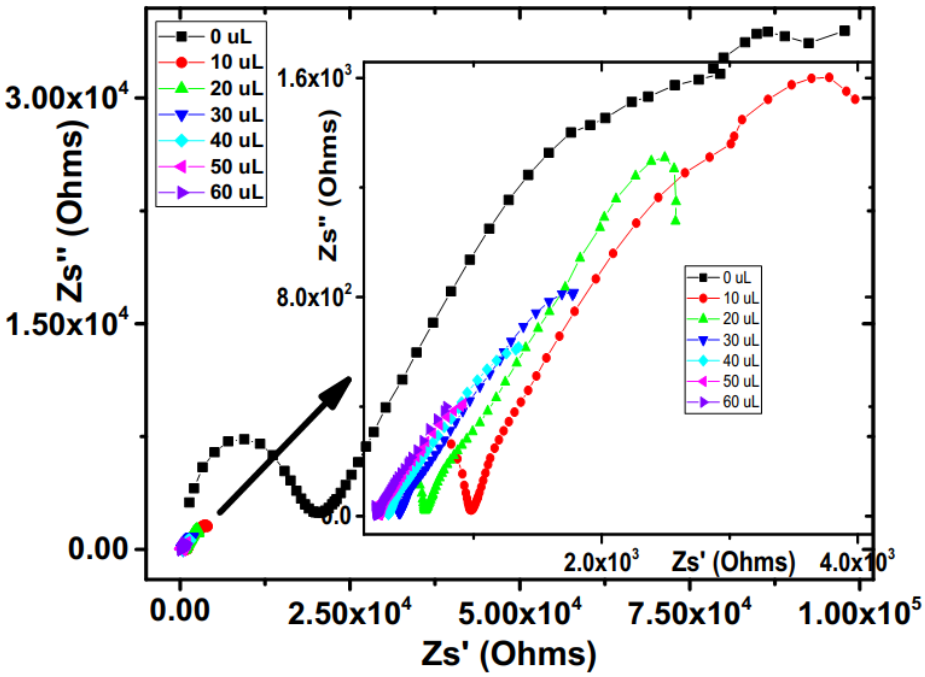


Fig. (6) Shows the complex series impedance (Zs) for the sand-clay sample dry and at various saturations of filtered water.

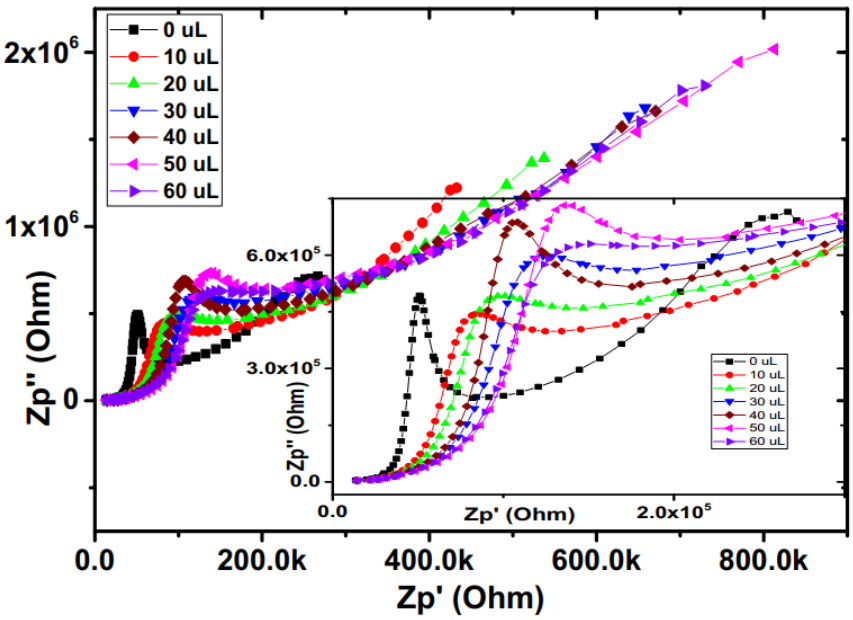


Fig. (7) Encloses the complex series impedance (Zp) for the sand-clay sample dry and at various saturations of gasoline.

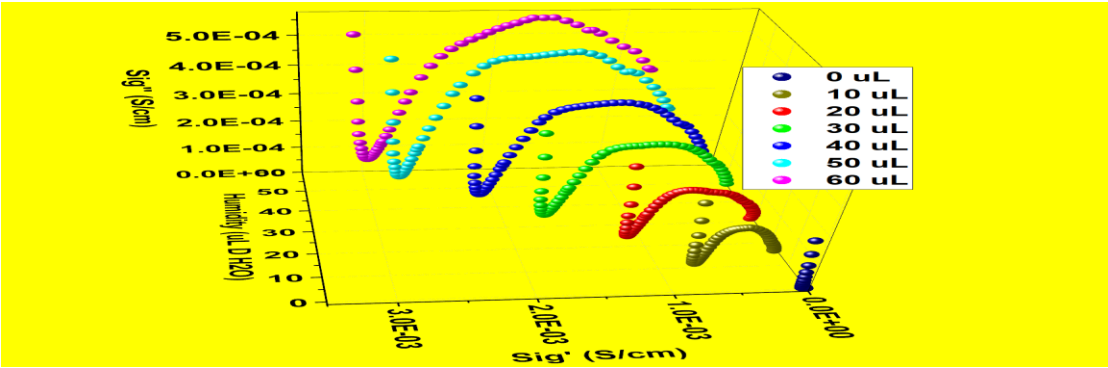


Fig. (8) displays the Nyquist representation of conductivity of sand-clay with different amounts of water.

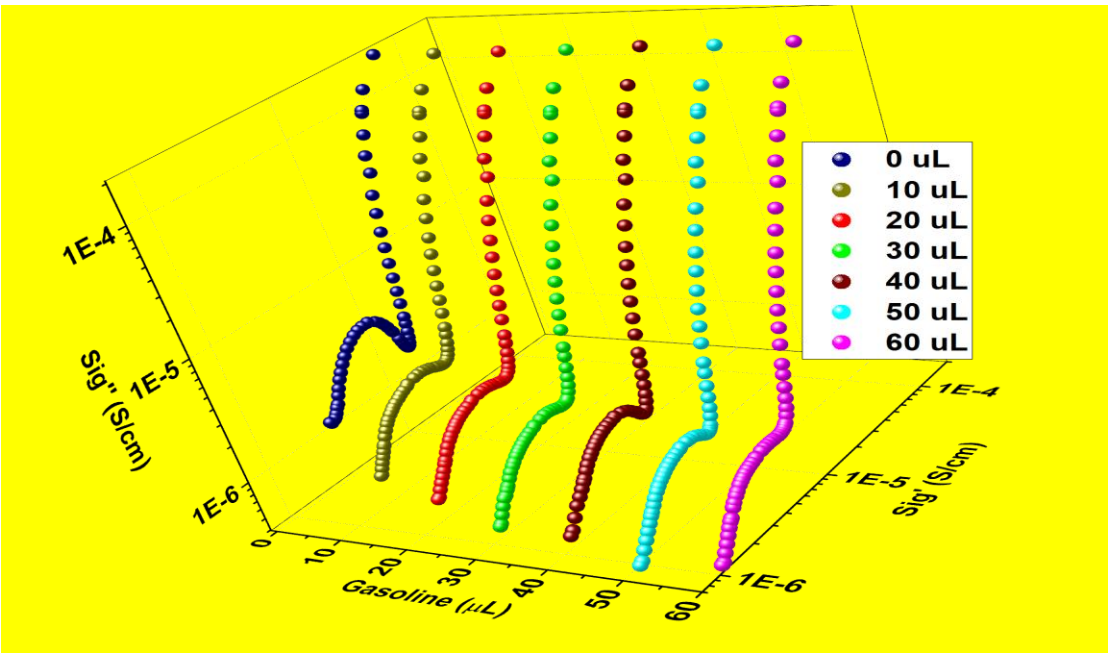


Fig. (9) displays the Nyquist representation of conductivity of sand-clay with different amounts of gasoline concentrations.