THE INFLUENCE OF CLAY FRACTION ON THE COMPLEX IMPEDANCE OF SHALY SANDS

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ABSTRACT

The influence of clay fraction on the electrical properties of shaly sands has been studied using synthetic samples in which control is exercised over the porosity, clay content, and clay distribution. Samples were fabricated having different clay types (montmorillonite, kaolinite and illite), in various concentrations from 3-15%. Clean control samples were also tested for comparison. The samples were subsequently placed in a hydrostatic testing cell, to simulate reservoir overburden stress conditions, and were then consolidated by a sequence of loading and unloading cycles. During and after compaction, the electrical properties were measured over a range of frequencies from 10 Hz to 2 MHz, using an impedance analyzer. Electrode polarization effects occurred in the low frequency range from 10 Hz to 20 kHz. The critical frequencies (f_c) of these samples do not show any correlation with clay content, but are influenced by clay type. A direct linear relationship was observed between the clay fractions and the complex impedance, with clay type also having an effect. The bulk sample impedance is constant over the frequency spectrum from 20 kHz to 500 kHz. The dielectric constant is more sensitive to frequency, and a relationship exists between the dielectric constant and clay content and type.

INTRODUCTION

The influence of clay on the electrical response of reservoir rocks, and problems associated with its interpretation, have been major issues of investigation in the petroleum industry for many years. Traditionally, only low-frequency d.c. properties have been used in the analysis. However, a new generation of logging tools that are capable of measuring electrical properties over a wide range of frequencies have recently become available. Consequently, attention is shifting towards the possibility of using the frequency-varying electrical response as a method of extracting information about clays present in reservoirs. In this paper, the effect of clay content and type on the electrical properties is investigated. Specifically, attention is focussed on the relationship between clay type and concentration and the complex impedance and dielectric constant. Electrical properties of any material are characterized completely by three physical properties: electrical conductivity, dielectric permittivity and magnetic permeability [1].

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In the case of the petroleum industry, complex impedance has the attraction of being a non-invasive technique, which measures the electrical response of rocks over a range of frequencies.   The impedance value, the dielectric constant and their frequency dependencies have been used as tools to estimate various rock properties, such as grain shape, permeability, porosity, saturation and wettability [2-8].

Much work has been conducted on the effect of clay on the d.c. electrical response [9-14].   On the other hand, the past few decades have seen a gradual shift towards the possibility of using frequency varying electrical response as a method of determining reservoir properties. Complex electrical data was initially used in the gigaHertz range to distinguish between hydrocarbons (*i.e.*, oil and gas), water and different rock lithologies. However, very little attention has been given to the lower frequency range from 10 Hz to 10 MHz [15]. In this frequency range the various petrophysical properties such as, clay content/type, salinity, saturation and wettability become more dominant in influencing this frequency dependency.

BACKGROUND THEORY

The resistivity of fully- and partially-saturated brine filled reservoir rocks is typically related to porosity and saturation using the Archie [16] resistivity equations. According to Archie's first law, the ratio (*FF*) of the resistivity of a fully brine-saturated sample, ρ_o, to the resistivity of the brine itself, ρ_w, is related to the porosity ϕ via the equation

$$FF = \frac{\rho_o}{\rho_w} = \frac{a}{\phi^m},\tag{1}$$

where *a* is the tortuosity parameter and *m* the cementation exponent, both of which are related to the degree of compaction and pore-space tortuosity between the sample grains. The higher the value of *FF*, the less well-connected the pore space. Archie's equation only applies to clay-free samples, or samples where the electrolyte solution is the dominant conductive pathway. For shaly sands, Waxman and Smits [10] proposed the following equation:

$$\frac{1}{\rho_o} = \frac{1}{FRF}\left(\frac{1}{\rho_w} + BQ_v\right),\tag{2}$$

where *B* is the equivalent ionic conductance of clay exchange cations, and Q_v is the cation exchange capacity of the rock per unit volume.

In the laboratory, the complex impedance of a sample, *Z*, is measured in the presence of a time-varying electric field. It consists of a resistive component, *R*, which gives rise to a current that is in-phase with the voltage, and the reactive component, *X*, which gives rise to a current that is out-of-phase with the voltage. Thus,

$$Z = R + iX,\tag{3}$$

where $i = \sqrt{-1}$ is the imaginary unit number. The phase angle, θ, by which the voltage and current are out-of-phase is given by

$$\theta = \tan^{-1}\left(\frac{X}{R}\right). \tag{4}$$

Effective electrical permittivities and conductivities can be extracted from the Z and R values measured in the laboratory. The electrical permittivity (ε^*) is a complex frequency-dependent parameter that describes the microscopic electromagnetic properties of the material. Thus, from the effective permittivity, the dielectric constant (K) can be deduced:

$$K = \frac{\varepsilon^*}{\varepsilon_o}. \tag{5}$$

LITERATURE REVIEW

Previously, investigators have studied the effect of clay on the electrical response as part of a package of properties being investigated. Sen [2] studied how the dielectric constant of sedimentary rocks depends on clay content, frequency, salinity and textural variables. Knight and Nur [4] investigated the effect of saturation on the dielectric constant of various sandstones. The surface area-to-volume ratio of the pore space appeared to be the controlling factor on the dielectric constant values. Knight et al. [17] deduced that the power law exponent derived from the data was inversely related to the surface area-to-volume ratio. Vinegar and Waxman [18] developed a model and equations of the complex conductivity of shaly sands in the frequency range from 3 Hz to 1 kHz. Petrophysical variables and equations consistent with the Waxman-Smits equations and model were used. The model and equations developed accounted for effects of shaliness on the complex conductivity of the rocks. A direct relationship between the shaliness parameter and the complex conductivity was observed. Taherian et al. [19] created a database of the dielectric response of 312 sedimentary sandstones in the frequency range from 10–1300 MHz. It was concluded that both the Cole-Cole and power law functions successfully modeled the frequency dependence of the samples in this frequency range, within experimental error.

Börner [20] investigated the frequency dependence of complex electrical conductivity in the frequency range from 1 mHz to 10 kHz, on a variety of shaly sandstones saturated and partially saturated with brines of various salinities. It was seen that the complex nature of the conductivity was caused by the capacitive behavior between the solid matrix and the electrolyte solution. A power law dependence of the real and imaginary parts of the conductivity was observed, and the exponent found to be related to surface area to porosity ratio, water salinity and water saturation. The dielectric measurements using two- and four-electrode techniques were performed on fourteen shaly sand samples by Garrouch and Sharma [21]. It was shown that clay influenced the dielectric spectra of rocks in the frequency range from 10 Hz to 10 MHz for the two-electrode configuration.

Garrouch and Sharma [22] concluded that water saturation, clay content and grain size could be obtained from electrical measurements made above 2 MHz.

Denicol and Jing [6] investigated the complex resistivity and phase angle as a function of salinity, water saturation and clay content, and concluded that frequency dispersion consistently increased with clay concentration, particularly in the frequency range from 10 kHz to 100 kHz. Li *et al.* [8] investigated the dielectric behavior of silica sand and kaolinite samples with the wettability ranging from water–wet state, through states with various amounts of sorbed oil. It was found that the presence of contaminants on the solid surface can influence the dielectric response of partially saturated materials, especially for materials having high surface areas.

MATERIALS AND METHODS

Core Sample Preparation
Our investigations were carried out on synthetically prepared shaly sandstones. The reason for using synethetic samples was to achieve control over the variations in clay content, clay type and clay distribution, thereby allowing systematic investigation of the effect of clay, frequency, salinity and saturation on the electrical properties. Twenty-eight synthetic samples were used, with various clay concentrations and clay types, although only eight are discussed in this paper; see Al-Mjeni [23] for full details.

Sample preparation began by vigorously mixing sand of a particular grain size with the required amounts of clay, thus ensuring a good homogeneous mixture of quartz and clay. This mixture was poured into a shrink-sleeve having equally-spaced wires along its length. Filter paper and stainless steel meshes were placed at the top of the sample to prevent any fine migration, after which the samples were capped. Subsequently, the samples were weighed, and then saturated with the required brine, 5% sodium chloride and 1% potassium chloride. After saturation they were weighed again, thus giving the effective pore volume of the samples. At this stage the samples were ready to be placed into the viton sleeves and loaded into the pressure vessel (Figure 1). Table 1 and 2 present the various properties of the materials used to prepare the samples, and the basic sample properties, respectively.

Experimental Apparatus
The experimental apparatus consists of a stainless steel multi-sample pressure vessel with a maximum working pressure of 69 MPa and maximum working temperature of 200°C. Confining pressure is controlled by a hydraulic assembly module pressure intensifier. The pore pressure system consists of two interfaces, a gas/oil and an oil/brine interface. The bottom of the unit is connected to the oil/brine interface unit, where as the top is connected to a controllable gas pressure supply. The oil/brine interface contains five calibrated glass burettes, partially filled with oil and brine, and connected to the core samples. They are used for volume displacement measurement during application of stress, in order to calculate the sample pore volume at each stress point, as well as for

volume measurements during drainage/imbibition cycles. Finally, pressure transducers and regulators comprise the rest of the system (Figure 2) used to control and monitor the flow of fluids into and out of the samples.

The complex impedance is measured using a Quadtech model 7600 RLC impedance analyzer, which spans a frequency range from 10 Hz to 2 MHz. The meter is equipped with four coaxial BNC terminals on its front panel, which locate the calibration plane. The calibration plane is the position in which the instrument measures within its specified accuracy of 0.05%. Test fixtures and cables are used to interconnect the sample to the instrument. Parasitic phenomena related to fixturing, cabling and connections are frequency dependent and add to the sample response; they are eliminated by the open/short compensation technique.

Experimental Procedure
The samples were repeatedly loaded and unloaded over successive loops, following the procedure outlined in [13,24,25]. A total of twelve loops were performed, covering a hydrostatic pressure range from 50 psi to 4000 psi (0.35-27.6 MPa). The samples were subjected to two compaction cycles. During the first cycle, the volume of fluid leaving each sample was monitored. In subsequent cycles, the permeabilities were measured at a target pressure of 1000 psi (6.9 MPa). The samples were then unloaded, a porous stone was placed at the outlet of the samples, and the second compaction cycle was performed. During this cycle the volume of brine exiting the sample was measured, as well as the electrical resistance. Thus, using the electrical resistance data, the cementation exponents (*m*) over the successive stress cycles were calculated.

RESULTS AND DISCUSSION
Figure 3 presents the value of *m* as a function of stress. A large amount of hysteresis between the loading and unloading paths was observed during the first two cycles. The hysteresis was greatly reduced in the subsequent loops, to the extent that subsequent paths almost tracked each other. Consequently, the stress cycles were stopped at 1000 psi. Figure 3 indicates a stress sensitivity effect on the value of *m*. This is attributed to changes in the pore throat geometry and in the electrochemical double layer of the clays as the hydrostatic pressure changes [13].

The cementation exponent is also influenced by clay content and type as shown in Figure 4. Thus, the value of *m*, in addition to being stress-sensitive, is also clay-sensitive. It increases with clay concentration, but at a rate that varies with clay type. This is consistent with the variation in the cation exchange capacity (*CEC*) of the various clays. It is observed that as the *CEC* of the clays decreases, the cementation exponent increases (Table 1). The values of the cementation exponent found in our work are within the range of previously published values [13,24].

The samples used in this investigation were all saturated with 5% sodium chloride and 1% potassium chloride brine. Figures 5 and 6 illustrate the effect of clay content and

concentration, respectively, on the complex impedance spectra. The impedance spectrum is a combination of electrode polarization and bulk sample response, and can be divided into three regimes. The first is the low frequency regime between 10 Hz and 10 kHz, the second is the medium frequency regime from 10–500 kHz, and the third is the high frequency regime above 500 kHz. In Figure 5 it is observed that the complex impedance increases with increasing clay concentration, over the entire frequency range, which is in agreement with previous findings. There is also a strong frequency-dependence of the complex impedance at the low and high ends of the spectra. In the low frequency range of the spectra, sample/electrode polarization takes place. This is a major experimental concern, and failing to account for it would introduce errors when analyzing the data. This sample/electrode polarization results from materials of differing conductivities or dielectric permittivities coming into contact. This produces an interfacial polarization on the grain surface, which results in the strong frequency dependence. Figure 6 illustrates the effect that clay type has on the complex impedance spectra, and shows that, for the same concentration, different clay types give different responses. This is related to the *CEC* values of the various shaly samples, and the precise relationship is being investigated further.

The electrode/sample polarization and bulk sample response are isolated from each other by plotting the real component versus the imaginary component of the complex impedance response (i.e. an Argand diagram). An example is presented in Figure 7, where the reactance is plotted versus the resistance. The frequencies on the Argand plot increase down the tail segment of the plot, then counterclockwise around the remaining part. The critical frequency (f_c) shown in Figure 7 is the point where the electrode sample polarization is separated from the bulk sample response [7,8,17]. Note that clay fraction seems to have no influence on (f_c), but the type of clay does have an influence. However, more samples need to be tested in order to establish a trend. Frequencies from around 500 kHz to 1 MHz are characterized by the relaxation frequency of the polarization process, which occurs at 800 kHz [6]. Hence, 500 kHz was selected as a cut-off point to ensure that no high-frequency effects were incorporated into the analysis. The intermediate region from 20-500 kHz was selected for investigating the effect of clay on the complex impedance spectrum. The bulk sample response in the intermediate frequency range was characterized by an almost-negligible frequency effect. However, a direct linear relationship between the clay content and the average impedance value is clearly seen in Figure 8. It is therefore concluded that the complex impedance value is very sensitive to clay content. It is also sensitive to clay type. This may be related to the different *CEC* value of the samples.

Figures 9 and 10 show the dielectric constant of various shaly samples. Frequency dependence is observed, showing that the dielectric constant is much more strongly influenced by the frequency than is the impedance. The dielectric constant (Figures 9 and 10) are consistent with those presented by Garrouch and Sharma [21]. They observed the same trend for outcrop samples and modeled the data using an electro-diffusional model [26]. Thus, it is concluded that this frequency dependence is somehow

affected by the double layer polarization phenomena. The figures also suggest that the dielectric constant is influenced by both clay concentration and clay type.

CONCLUSIONS
The effects of clay concentration and clay type on shaly sandstones have been investigated in a systematic manner, using synthetically-prepared samples with known clay contents. The cementation exponents (m) of the artificial shaly samples are consistent with those found in the literature for natural un-cemented sandstones. This exponent m is sensitive to stress, clay fraction and clay type. The complex impedance values were found to be sensitive to clay fraction and clay type, and insensitive to frequency, over a wide range of intermediate frequencies. The critical frequency (f_c) was found to be sensitive to clay type, but not to clay concentration *per se*, although more samples need to be tested. There is a strong linear relationship between the average impedance value in the intermediate frequency range, and the clay content. The dielectric constant (K) was strongly influenced by clay fraction and type, and varied with frequency. The double-layer polarization phenomenon of the samples may explain this frequency dependency.

NOMENCLATURE

a	Tortuosity factor	Z	Complex impedance, Ω
B	Equivalent ionic conductance, (S cm^2 meq^{-1})	ε_o	Dielectric permittivity of free space (8.854 x 10^{-12} F/m)
f_c	Critical frequency, Hz	ε^*	Effective permittivity, F/m
FF	Formation resistivity factor	K	Dielectric constant
i	$\sqrt{-1}$, imaginary unit number	ρ_w	Brine resistivity, Ω m
m	Cementation exponent	ρ_o	Resistivity of fully saturated sample, Ω m
Q_v	CEC of rock, meq m^{-1}		
R	Resistance, Ω	θ	Phase angle, radians
S_w	Brine saturation		
X	Capacitive reactance, Ω		

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Table 1. Properties of the clay material used to prepare the synthetic samples ([a]Grim [27]; [b]van Olphen and Fripiat [28]).

Material Type	Density [g/cm^3]	Grain Size [μm]	Surface Area[b] [m^2/g]	CEC[a] [mmols/100g]
USA Sand	2.64	45-425	-	-
Japan Sand	2.64	0-125	-	-
Montmorillonite	-	0.1-0.3	46	80-150
Kaolinite	-	0.3-4	6.6	3-15
Illite	-	-	101	10-40

Table 2. Description of the synthetic samples used in this investigation.

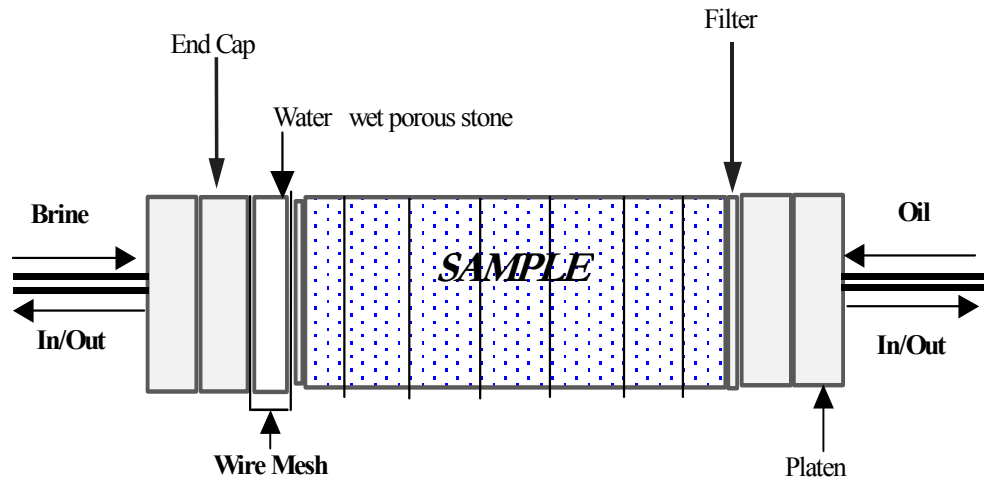| Sample Name	Clay Type	Clay Concentration [%]
SQ	0	0
SM3	Montmorillonite	3
SM5	Montmorillonite	5
SM8	Montmorillonite	8
SM10	Montmorillonite	10
SM15	Montmorillonite	15
SK3	Kaolinite	3
SK5	Kaolinite	5
SK8	Kaolinite	8
SK10	Kaolinite	10
SI5	Illite	5
SI10	Illite	10

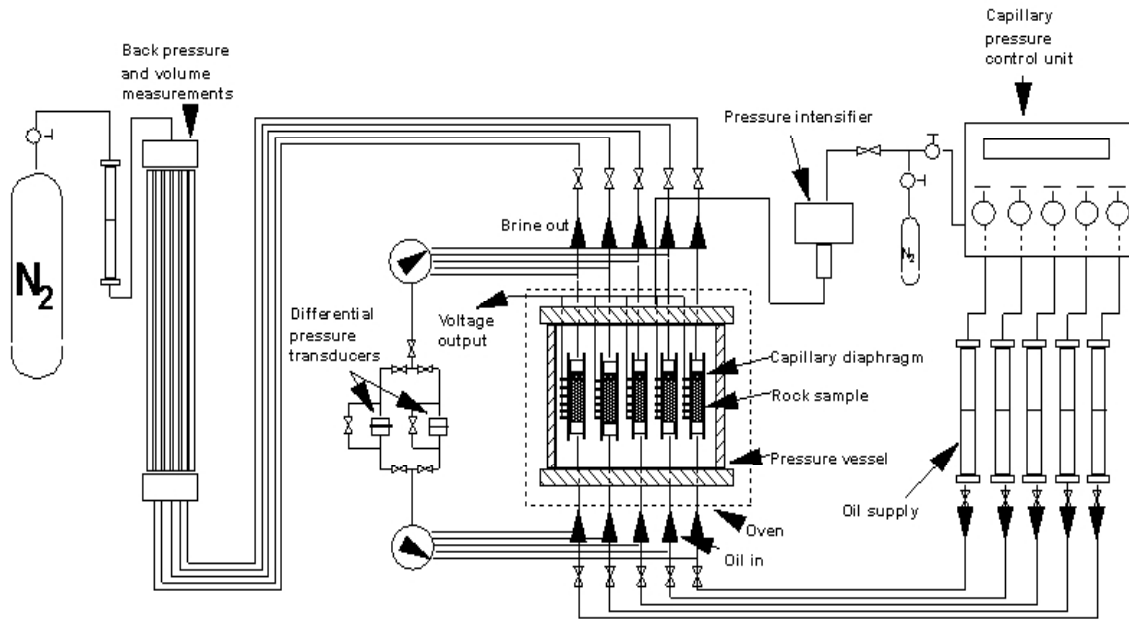**Figure 1. Schematic diagram of sample in the core sleeve with multi-electrodes.**

Figure 2.   Schematic diagram of the multi-sample cell for measuring the electrical properties, capillary pressure and permeability of the rock samples at different confining pressures (after [7]).
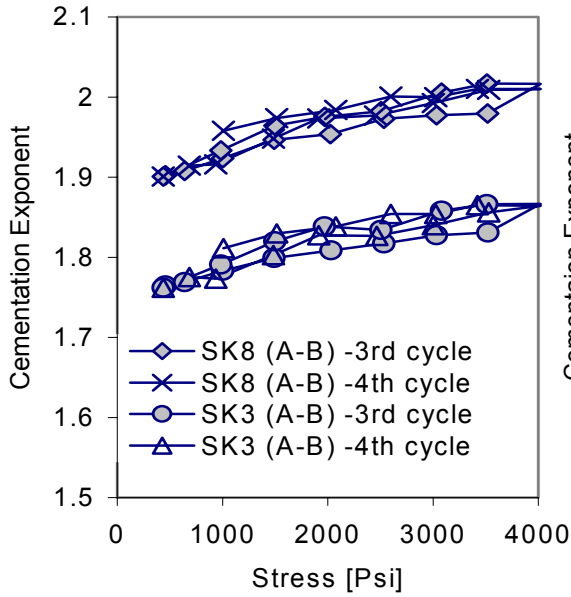
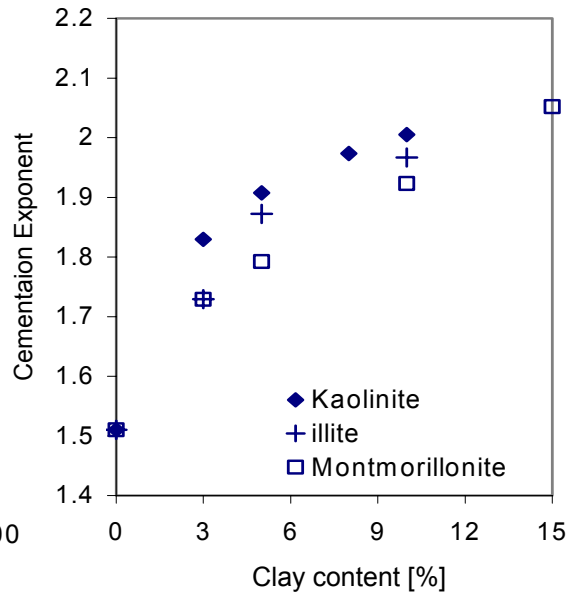**Figure 3. Cementation exponent vs. stress for Kaolinite clay samples.**

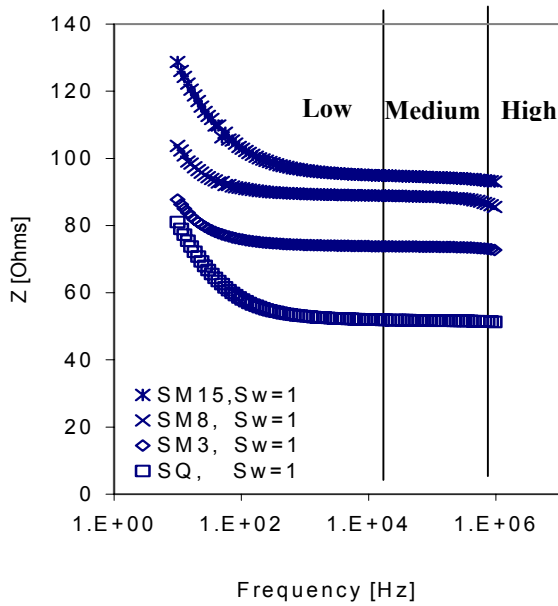**Figure 4. Cementation exponent as a function of clay concentration and type.**

Figure 5. Effect of clay content on the complex impedance spectra of montmorillonite shaly sands.
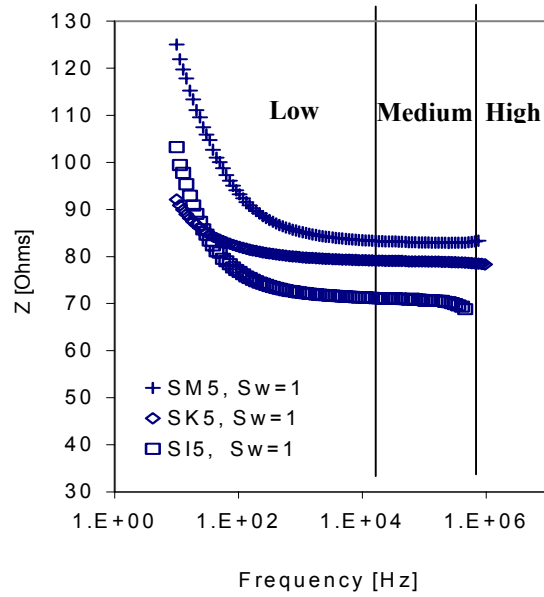
Figure 6. Effect of clay type on the complex impedance spectra of shaly sands.
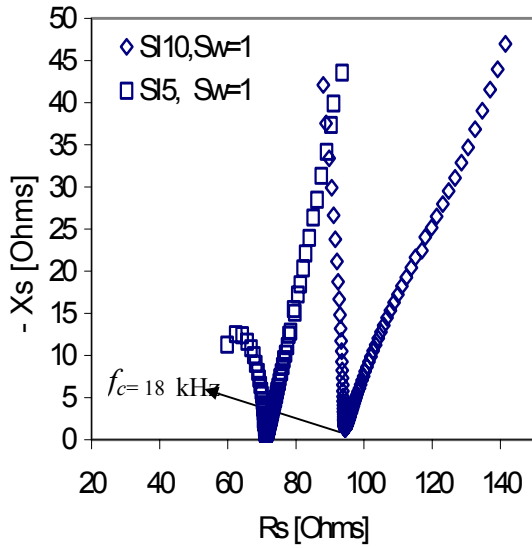
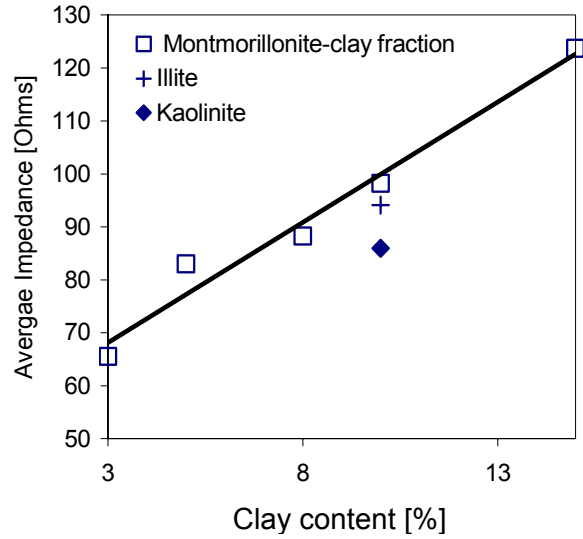**Figure 7. Argand plots of the illitic shaly samples fully saturated with brine.**

Figure 8. Plot of average complex impedance as a function of clay content and type.
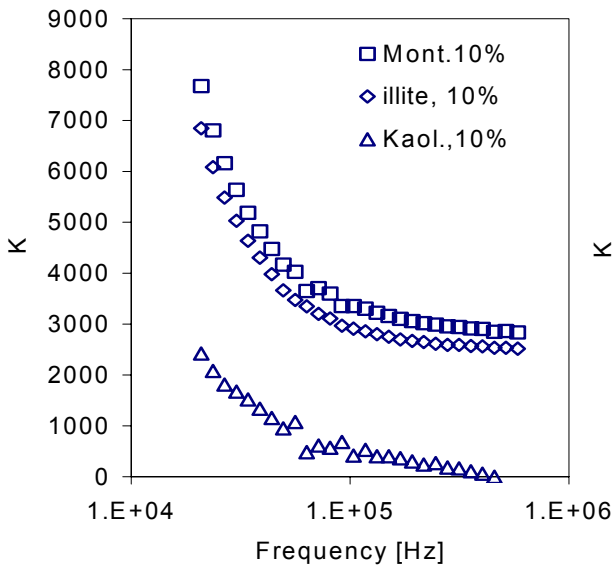

Figure 9. Dielectric constant as a function of frequency for various clay types.
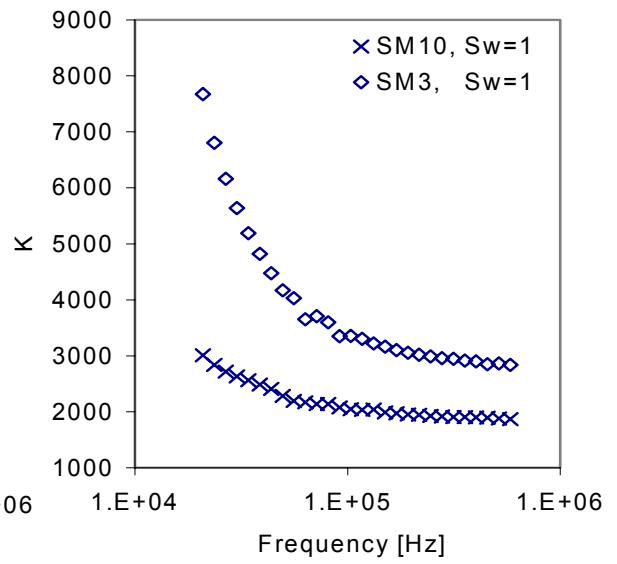

Figure 10. Dielectric constant as a function of frequency for two clay concentrations.