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Electrical properties of oilshale rocks

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The characterization of electric properties of oilshale deposit is reported. The scanning electron microscopy and the results of the dielectric constants show that the deposit is a heterogeneous composite material containing some conducting and insulating microstructural units. The components of the equivalent input impedance measured at microwave frequencies (8 to 12 GHz) and the impedance at low frequencies (0.5 Hz to 30 kHz) showed frequency dependence. The real and imaginary components of the permittivity determined from the impedance data obtained at low frequencies showed an exponential decrease with frequency. The observed frequency dependence of the rock permittivity is ascribed generally to a Maxwell–Wagner type of mechanism. Also it was observed that the conductivity is almost independent of the frequency below 1 kHz and increases with frequency above this range. The values obtained for the conductivity at low frequencies indicate that the rock has an intermediate electrical conduction due to some mineralized metallic complexes and carbon content.

1. Introduction

Oilshale deposit is a composite material whose electric characterization depends on the physical properties of its porous structure consisting mainly of oil fluid and sedimentary rock components such as organic carbon matter, calcite, quartz and dolomite crystallites [1, 2]. The elemental constituents, carbon content and metallic inclusions made the deposit a conducting material with a high-frequency electrical conductivity of average value 3×10^{-7} mho m^{-1}. Some measuring techniques such as microwave impedance and dielectric measurements on oil-reservoir rocks are presently receiving a great deal of attention from the scientific community in the study of the conducting behaviour and the dispersion phenomena shown by these rocks [3–7]. These nondestructive measurements could be successfully applied to oil exploration, determination of hydrocarbon content and evaluation of the deposit as a source of energy. Oilshale rock deposits are distributed in locations all over the world, such as China and USA. About two decades ago, huge amounts of oilshale deposit with an average yield of 5% to 15% crude oil by weight, were discovered in Jordan.

In a recent study [2], some structural and microwave properties of the Jordan deposit were reported in the X-band (8 to 12 GHz) by measuring the insertion loss, return loss, and equivalent input inpedance. Here we reconsider further analysis of some of the microwave results and extend the study to the electrical properties of the deposit at low frequencies.

2. Experimental procedure

Samples of the oilshale rocks were obtained from the El-Lajjun oilshale area located in the southern part of Jordan, near Karak city. The deposit exists at a depth of about 32 m under sedimentary structure of limestones, marls, cherts, shales and phosphate. The constituents of the oilshale deposit are moisture, oil, sulphur complexes and organic matter. From the bottom to top structures, the oil content varies from 12.6% to 4.3%, the moisture from 1.8% to 3.5%, sulphur from 3.5% to 1.9%, and the organic matter from 14.5% to 6.3 wt% as reported by the National Resources Authority of Jordan [1]. Disc and plate-shaped specimens, 2 to 7.15 mm thick, were cut randomly from the oilshale deposit with dimensions suitable for the desired measurements. A Leitz SEM was used to examine the texture of the deposit samples taken from different depths. The test specimen was mounted on stubs and coated with gold for SEM observations.

The equivalent impedance of the oilshale deposit specimens was measured, at different microwave frequencies in the X-band, using the slotted line method [8, 9]. Disc-shaped specimens were cut from the deposit to perform low-frequency impedance measurements. Two copper electrodes, about 500 nm thick, were evaporated under vacuum on both faces of the disc surfaces and two fine copper wires were soldered to the electrodes by silver dag. The test specimen was shielded and connected to an electronic bridge described elsewhere [10, 11]. A gain-phase meter was used to measure the phase and the amplitude ratio of the input to the output signal for the test specimen. Impedance measurements were performed as a function of frequency in the range 0.5 Hz to 30 kHz. The uncertainty in the measured values was approximately less than ±5%.

3. Results and discussion

The scanning electron micrographs shown in Fig. 1 reveal the mineral components and the texture of two

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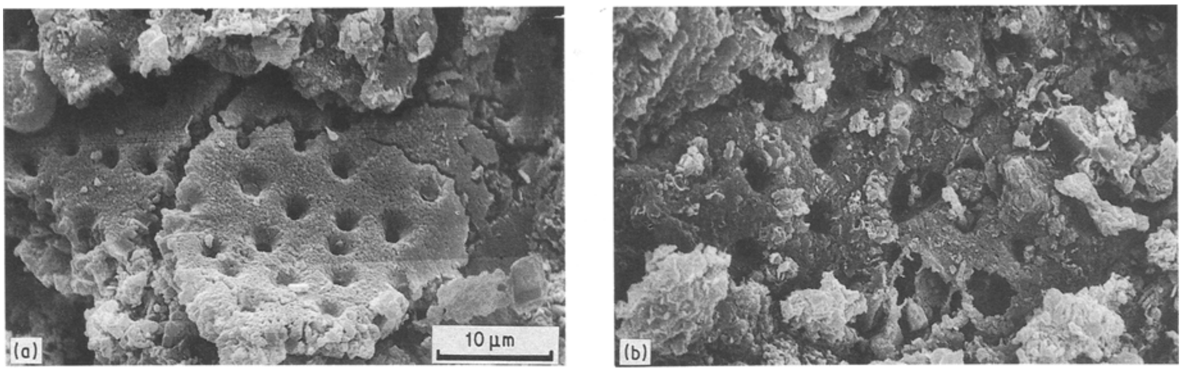

Figure 1 SEM micrographs: (a) upper layer rock, (b) lower layer rock.

test specimens taken from the top and bottom bed of the oilshale rock. The overall rock microstructure consists of a mixture of minerals such as calcite, quartz, kaolinite, apatite and dolomite crystallites and a small percentage of complex organic carbon matter. A small difference in the mineralogical structure between the top and the bottom deposit structures is observed [2]. But a difference in the porous texture of the two deposit layers is clearly seen in the scanning electron micrographs. Fig. 1a, for the top deposit layer, shows small black pores of average diameter 2 to 3 µm, while Fig. 1b, for the bottom layer (at about 20 m depth from the top layer), shows black pores of average diameter 4 to 5 µm. Hence the lower layer has a higher pore space than the top one. However, these black pores containing some traces of crude oil reveal that the oilshale rock has an inhomogeneous porosity.

The equivalent input impedance $Z = Z_R - Z_X$, at microwave frequencies (8 to 12 GHz), is measured using the slotted waveguide method. The data obtained for Z show that there is a slight difference between the behaviour of impedance and that of frequency for specimens of the same thickness (5.1 mm) taken from the upper and lower deposit layers. The impedance shows a nonlinear increase with the frequency. The real part, Z_R, and the imaginary part, Z_X, of the equivalent impedance are plotted as a function of frequency in Fig. 2. A first look at this plot shows that the dependence of Z_X and Z_R on frequency is not similar to semisolids or biomaterials [13]. The real part of Z increases sharply with frequency and the imaginary part increases slowly to a maximum around 11 GHz and then decreases rapidly. It is worthwhile to note that the intrinsic impedance of the waveguide for the dominant mode propagating in the guide (i.e. TE_{10}) is more than 600 Ω around 8 GHz and decreases to more than 400 Ω around 12 GHz. This variation in the guide impedance can be used to normalize the equivalent input impedance of the oilshale rock which will indicate steeper variation in the real part and smoother variation in the imaginary part. The observed data of Fig. 2 show that there is no difference between the impedance behaviour of the two specimens. However, the dependence of impedance on frequency represents an inductive behaviour.

The complex impedance plot of the data for the same specimens is shown in Fig. 3 where Z_X (imaginary part of Z) is plotted against Z_R (real part of Z). The plots obtained at microwave frequencies are almost identical semicircular arcs and are similar to those found in many studies of ionic conductors and selective electrodes [11, 13, 14]. However, the slight observed difference in the semicircular shapes is attributed to small variations in the microstructure of the two specimens such as the microgeometry and pore space [6].

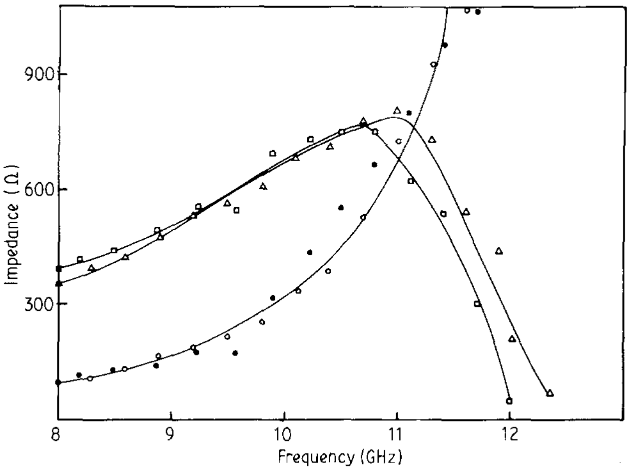

Figure 2 The complex impedance as a function of microwave frequencies for (○, △) upper and (●, ◇) lower deposits. (○, ●) Z_R, (△, □) Z_X, $t = 5.1$ mm.

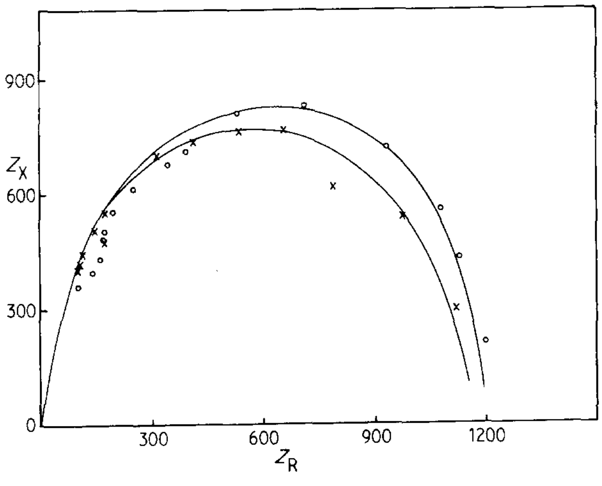

Figure 3 Complex impedance plot data from (○) upper and (×) lower deposits at microwave frequencies. $t = 2$ mm.

The dependence of the measured phase angle, ϕ, on the low-frequency range (0.5 Hz to 30 kHz) for two specimens of the same thickness (2 mm) taken from the upper and lower rock layers is shown in Fig. 4. There is a pronounced difference in the phase angle behaviour of the two samples. The phase angle for both specimens increases rapidly and saturates in the frequency range 1 to 10 kHz. The variations of the measured impedance with frequency are shown in Fig. 5 where the impedance, Z, decreases rapidly in the very low frequency range and takes small values at high frequencies. This suggests that the oilshale specimens and the electrodes operate as a capacitive network. This capacitive behaviour can be explained by a simple physical model based on networks of grain–grain, moisture or oil–grain, pore space–grain capacitors which contribute to the observed behaviour of the impedance, Z, and the phase angle, ϕ, with frequency. The dielectric constant, ε, related to Z and ϕ is given elsewhere [15]. The data presented in Figs 6 to 8 are calculated from the following equations [13, 14]

$$Z = Z_R - jZ_X \qquad (1)$$

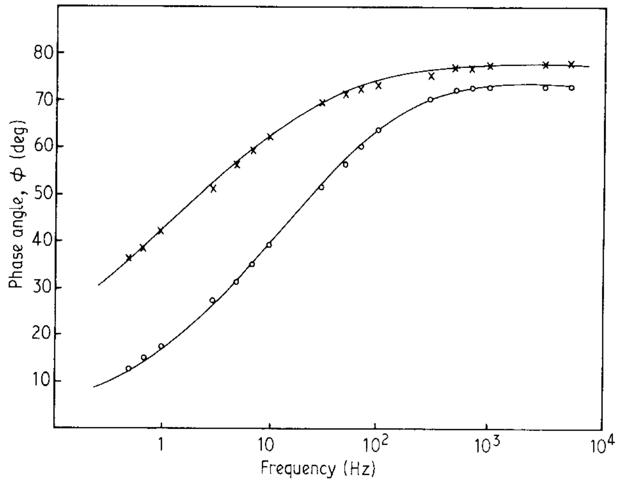

Figure 4 The phase angle as a function of frequency for (○) upper and (×) lower deposits. $t = 2$ mm.

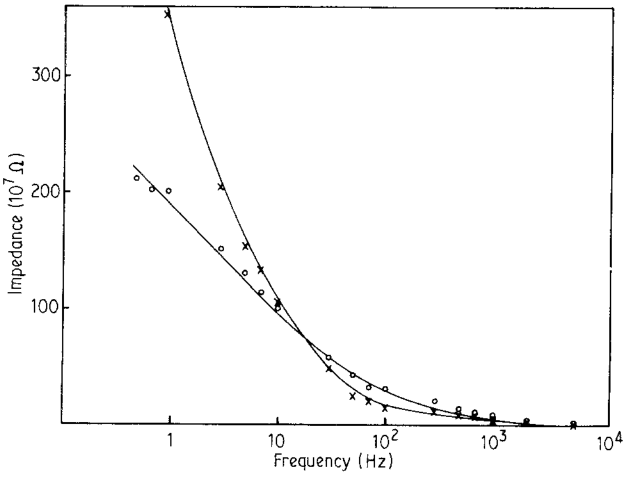

Figure 5 The impedance as a function of frequency for (○) upper and (×) lower deposits. $t = 2$ mm.

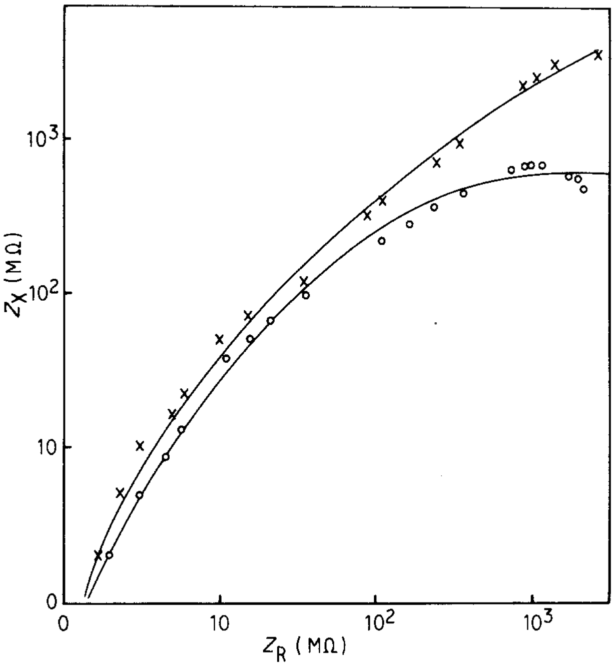

Figure 6 The complex impedance plot data from (○) upper and (×) lower deposits at low frequency range. $t = 2$ mm.

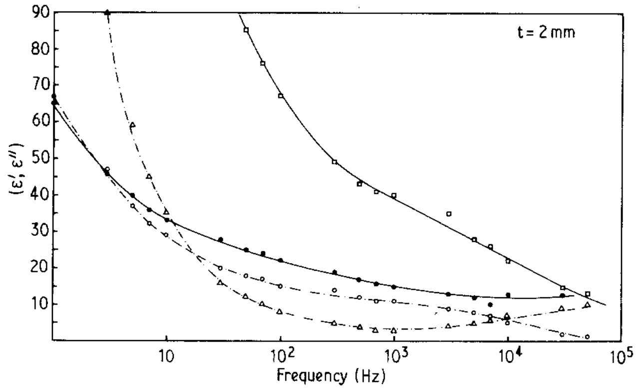

Figure 7 The complex dielectric constant as a function of frequency. (○) ε_1' (upper), (△) ε_1'' (upper), (●) ε_2' (lower), (□) ε_2'' (lower).

and

$$\varepsilon = \varepsilon' - j\varepsilon'' \qquad (2)$$

where Z_R and Z_X are the real and the imaginary parts of the complex impedance Z; ε', and ε'' are the real and imaginary parts of the relative dielectric constant. Their values are given by

$$\varepsilon' = Z_X/(2\pi f C_0 Z^2) \qquad (3)$$

and

$$\varepsilon'' = Z_R/[2\pi f C_0 Z^2] = \sigma/2\pi f \varepsilon_0 \qquad (4)$$

where σ is the conductivity, ε_0 is the permittivity of free space, C_0 (= 0.25 pF) is the capacitance of the electrodes, and f is the frequency.

The complex impedance plot of the real component, Z_R, and the imaginary component, Z_X, in the range 0.5 Hz to 30 kHz is shown in Fig. 6. This Cole–Cole plot shows greatly distorted semicircles for the upper and the lower (surface and deep) deposit specimens. These observed semicircles are due to the specimen bulk and the distortion may be due to the complex

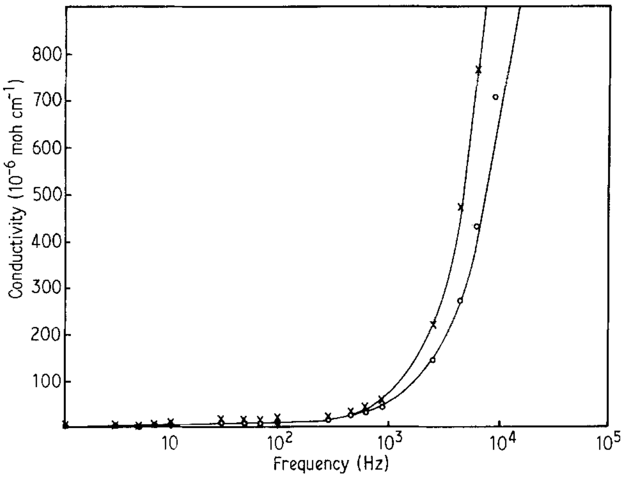

Figure 8 The dependence of conductivity on frequency for (○) upper and (×) lower deposits.

behaviour of the dielectric permittivity at low frequencies where a distribution of relaxation times is expected from some local microinhomogeneities within the specimen bulk [14, 15]. The difference between the plots of the two specimens, especially at very low frequencies, is also due to some anomalous behaviour of ε, as will be seen later.

Oilshale rock is a complex heterogeneous composite containing different mineral components, organic matter and moisture (water content). Long ago, the oil diffusion into the rock structure led to the deposition of minute amounts of metallic elements, complex oxides and sulphides which settled as conducting blocks or inclusions. Hence, this type of rock composite is expected to have permittivity characteristics related to these structural components. The expected sources of deposit permittivity at low frequencies are a collection of Maxwell–Wagner behaviour, dipolar and atomic charge polarization, in addition to the interfacial polarization. Other physical properties which can affect the rock polarizability are the density and the porosity plus the grain-boundary microcracks and pore space existing in the bulk specimen [6, 7, 16].

The real part, ε′, of the complex permittivity is a measure of the amount of polarization in a material and it is much influenced by the water or moisture content. On the other hand, ε″ is a measure of the amount of the dielectric hysteresis for this material at low frequencies, or it is equivalent to the electrical conductivity, σ. The dependence of ε′ and ε″, calculated from the impedance data using the above equations, on frequency is shown in Fig. 7. Clearly, there are large differences in the behaviour of the real and imaginary parts of the upper and lower deposit specimens in addition to both of them having high values at low frequencies. From Fig. 7, some information can be drawn about the physical properties of the rock deposit by looking at the dependence of ε′ and ε″ on frequency. The values of ε′ for both specimens indicate that the oilshale deposit contains a quantity of moisture or water which enhances ε′, while the high value of ε″ indicates that the deposit has an equivalent electrical conductivity at low frequencies. The values of ε″ for the lower (or deep) specimen is very much higher than those values for the upper (surface) specimen, i.e. the lower deposit specimen is more conducting because it contains more mineral components, metallic complexes and organic carbon content. The final information which may be drawn from ε′ and ε″ is that the overall behaviour of ε exhibits a complex or anomalous permittivity behaviour with a broad distribution of relaxation times due to some local microinhomogeneities and microcracks with the grains of the deposit microstructure.

The variation of the conductivity, σ, calculated from the values of the imaginary part ε″ using Equation 3 for both upper and lower specimens, is shown in Fig. 8. The value of σ remains almost constant ($< 10^{-7}$ mho m^{-1}) at frequencies below 1 kHz; above this frequency, σ becomes a strongly increasing function and attains a value of about 7.5×10^{-6} mho m^{-1}.

4. Conclusion

The SEM and impedance behaviour of the oilshale deposit existing in Jordan have been considered. The dielectric permittivity, ε, and the conductivity, σ, are determined through impedance measurements in the low-frequency range 0.5 Hz to 30 kHz. From the data obtained, the following conclusions were drawn.

1. The oilshale deposit is a heterogeneous composite which contains some minerals, complexes and carbon content with a porous texture.
2. The equivalent input impedance showed a complex dependence on microwave frequency. Both of its components vary with frequency and the imaginary part reaches its maximum around 11 GHz.
3. The determined complex dielectric constant showed a frequency dependence. Its real and imaginary parts showed very large values at very low frequencies with large frequency dependence. This also indicates that the rock has a very heterogeneous structure with moisture content. It was found that its polarizability behaviour is controlled by the Maxwell–Wagner polarization mechanism.
4. The conductivity is almost independent of frequency below 1 kHz and increases exponentially up to 30 kHz. The determined values for conductivity indicate that the deposit is a relatively low-conducting material due to multiminerals, complexes and carbon content.

Acknowledgements

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