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Laboratory studies of the DC – induced polarization in saturated sand models of rock

Introduction

Usage of The Induced Electrical Polarization technique over the past years has proven to be one of the most successful geophysical methods in providing in situ information about rock mineral composition especially in search for disseminated minerals with electronic conductivity. In this case, the technique is being applied in quite a new field, which concerns materials that do not contain conductive minerals but rather clay minerals. However the method has been used, rarely in the field of hydrogeology [8, 23], oil and gas field exploration [19] and in environmental studies such as mapping of polluted land areas [22]. The induced polarization method has been recognised as a useful method in the detection of organic liquids electrical response both in field [24] and laboratory conditions [6, 25, 26]. Several environmental studies have used geophysical methods to delineate oil-contaminated sites [1, 2]. More recently [21] obtained power law dependence for IP measurements carried out on clay free unconsolidated sediments from a sandy aquifer and unsaturated sands, respectively.

Polarization is a geophysical phenomenon which measures the slow decay of voltage in the ground after the cessation of an excitation current pulse (time domain method) or low frequency variation of the resistivity of the earth (frequency domain method) [9, 20]. The IP effect reflects the degree to which the subsurface is able to store electric charge, analogous to a leaky capacitor. If the current is interrupted, a difference in potential, which decays with time, is observed. The IP phenomenon can be modelled with equivalent parallel circuits composed by two resistances (R_1 and R_2) and one capacitor (C) (Fig. 1). In this case, R_2 represents the behaviour of resistive ionic conduction in unmineralized current paths near a metallic mineral.

The resistor R_1 represents the resistance due to blocked conduction paths within mineralized rock, and the capacitor C can be associated with the double-layer capacitance and Warburg impedance [9, 10, 20]. The rate of decay of this potential (IP potential) depends on the lithology of the rock, its pore geometry, degree of liquid saturation and type of liquid. The IP effect in earth materials having non metallic minerals is usually referred to as membrane polarization. This requires the presence of particles such as clays, with a negative surface charge, which in the presence of an electrolyte like water, attracts cations and forms an electrical double layer on the particle surface. The volume property of soil related to this phenomenon is the cation exchange capacity [12] which is proportional to the surface charge density and the specific surface area of the solid phase [3]. Polarization and the complex nature of electrical rock conductivity are attributed to zones of unequal ionic transport properties along the pore channels caused by charged interfaces and constrictions. Interactions at and near the contact area of the solid and liquid phases are the main causes of the formation of an electrical double layer. They depend on pore space structure and microstructure of the internal rock boundary layer. Therefore, they contain information about internal surface area, rock porosity, rock permeability and fluid properties.

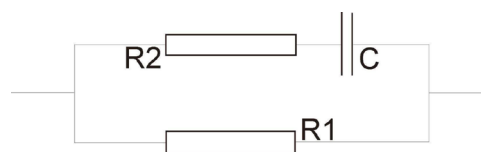


Fig. 1. Equivalent circuit to simulate the IP phenomenon. The circuit is composed by two resistances (R_2 and R_1) and one capacitor (C) [After 20]

The application of an external physical field on a saturated heterophase medium intensifies filtration and diffusion processes in it and results in changes in its electrical, magnetic and acoustic properties [15–18]. When an electric field is applied to rocks, all kinds of effects may appear, e.g. dislocation friction, impact on electrons, and oscillation of crystalline cells and so on, contributing to secondary effects [11, 13]. Polarization properties of rocks depend on both the nature of the rocks and of the filler (pore fluid). Depending on the chemical composition and petrophysical parameter of a rock, and the type of saturated medium (oil, electrolyte), the mechanism of (DC – induced) polarization is different. This means that depolarization after the field action ends must differ in a few parameters, e.g. in the maximum polarization amplitude, the relaxation time and the character of signal re-

laxation decay. The Author emphasizes that such complex structures are characterized by very different polarization mechanisms, from electronic to electroosmotic with very different characteristic relaxation times (from 10^{-9} s to minutes). By using macroscopic experimental technique the Author actually measures the average value of the electric potential. Therefore, in order to properly describe the polarization relaxation one needs to know the relationship between the macroscopic electrical characteristics of the medium and its components (phases). In view of this, Author considers the relaxation time characteristic for the materials in question.

A better insight into this effect may lead to a new investigative parameter for geophysical prospecting. The investigations yielded the results partly discussed now and partially presented earlier [14].

Mathematical deliberation

Consider the limitless saturated porous layer (sand model), which covers Euclidean space area $0 < x < l$ [14]. Material of the layer – insulator with open porosity (sand). In the following will be considered fluid saturated layers of different nature. Attached to the surface layer $x = 0, x = l$ of constant potential difference U_0 and it runs along the OX axis with the constant current density j_0 [5].

$$j_0 = \sigma_e E_0 \quad (1)$$

Where: E_0 – macroscopic (averaged) electrostatic intensity of the electric field in the layer,

$$\sigma_e = (\Phi_1 \sigma_e^{(1)} + \sigma_{es} s_{12}) f_E^{(1)} \quad (2)$$

– coefficient of electrical conductivity of the model, Φ_1 – porosity, $\sigma_e^{(1)}$ – coefficient of electrical conductivity of pore fluid, σ_{es} – coefficient of surface electrical conductivity of the hard phases-fluid contact (double electrical layer), s_{12} – surface area of phases (double electrical layer):

$$s_{12} = \sqrt{\frac{\Phi_1^3}{\theta \Psi d_p}} \quad [7] \quad (3)$$

Where: θ, Ψ, d_p – (accordingly): pore geometry, pore tortuosity and permeability coefficient of the model.

$$f_E^{(1)} = \frac{\varepsilon - \varepsilon^{(2)}}{\Phi_1 (\varepsilon^{(1)} - \varepsilon^{(2)})} \quad (4)$$

Here: ε – permittivity of the layer, $\varepsilon^{(1)}, \varepsilon^{(2)}$ – permittivity of fluid and the hard phase, $E_0 = U_0/l$ – the macroscopic (averaged) electrostatic intensity of the electric field in the layer.

At the moment $t = t_0 + 0$ attached to the surface layer potential difference becomes equal to zero $U_0 = 0$. The electric field in the layer must satisfy the equation:

$$j + \frac{dD}{dt} = 0 \quad (5)$$

Where: j – current density, D – component of the vector of electrostatic induction.

The solution of equation (5) for the initial condition: $E = E_0$ and $t = t_0$ has the form:

$$E = E_0 e^{-\frac{\sigma_e}{\varepsilon}(t-t_0)} \quad (6)$$

Formula (4) can be rewritten as:

$$E = E_0 e^{-\frac{t-t_0}{\tau_e}} \quad (7)$$

Where, $\tau_e = \varepsilon/\sigma_e$.

Equation (6) or (7) will describe the process of relaxation of electrostatic intensity of the model.

From formulas (2), (3) and (4) we can see that the coefficient of electrical conductivity of the layer: $\sigma_e = \sigma_e(\Phi_1, \sigma_e^{(1)}, \sigma_{es}, s_{12}, \varepsilon^{(1)}, \varepsilon^{(2)}, \varepsilon)$ is depend from: Φ_1 – porosity, $\sigma_e^{(1)}$ – coefficient of electrical conductivity of pore fluid, σ_{es} – coefficient of surface electrical conductivity of the contact hard phases-fluid (double electrical layer), s_{12} – surface area of double electrical layer, ε – permittivity of the layer, $\varepsilon^{(1)}, \varepsilon^{(2)}$ – permittivity of fluid and the hard phase.

Specific conductivity of liquid depends on the nature and concentration of solution. If the liquid is a mixture of water and oil – its conductivity depends on water solution concentration. Permittivity of saturated porous medium is a mixture permittivity. If the liquid is a water – oil mixture it has a mixture permittivity too. Permittivity of a layer will also depend on the size and shape of grains of the hard phase. For harmonic in time processes (like a polarization) a dielectric permittivity is a complex value which accounts for the different mechanisms of relaxation.

Let us consider that a pore fluid – symmetric binary electrolyte solution. Then the coefficient of electrical conductivity can be written as:

$$\sigma_e^{(1)} = qn(v_+ + v_-) \quad (8)$$

Where: q , n – (accordingly) module of charge and ions concentration of electrolyte, $n = N/V$, N – the quantity of molecules in the bulk of electrolyte – V , v_+ , v_- – the mobility of positive and negative ions.

Formula (8) can also be written as:

$$\sigma_e^{(1)} = q_m^+ C_+ v_+ + q_m^- C_- v_- \quad (9)$$

Where (accordingly): q_m^+ , q_m^- i C_+ , C_- – the absolute values of specific charge and mass density of positive and negative ions, or:

$$\sigma_e^{(1)} = FC_M(v_+ + v_-) \quad (10)$$

Where: (accordingly): F , C_M – Faraday constant and molar concentration of electrolyte.

Methodology of investigations

Author investigated the depolarization process of saturated sand model of rock after they had been long time acted on with a constant electric field with different parameters (modified the time domain method).

The investigative procedure was as follows. As a sample porous medium the Author considers purified sand placed in a 0.52 m · 0.2 m · 0.22 m thick-walled viniduril (polyvinyl chloride) box (Fig. 2). Sieved commercial quartz sand (0.1 – to 0.3 mm grain size) was cleaned from clay and dust particles by repeated washing. Depending on the needs, the model or the rock was saturated with a brine of

different salinity or with water-petroleum emulsion. Let us consider that in our investigations the model did not contained any additional clay. Power feeding carbon electrodes and high-quality stainless steel electrodes were used. The electrodes were submerged down to the whole depth of the model. The distance between the electrodes in the model was changeable from 0.15 to 0.35 m. Each time the porous medium model was saturated with a different liquid (in the fixed amount) from the liquids mentioned above. First the medium was saturated with distilled water, then with a 0.1÷6% table salt solution, then, in turn, with diesel oil or crude oil (taken from the Barnówko-8 bore-hole and having the following parameters: density – 816.3 kg/m³, kinematic viscosity – 5.18 mm²/s, dynamic viscosity – 4.22 mPa · s, saturated hydrocarbons content – 79.8%, aromatic hydrocarbons content – 15.3%, asphaltene content – 0.5%). Each time the hard phase (sand) of the investigated medium was replaced. The amount of liquid in the model was in the range of 250÷500 ml. The model's moisture stability was monitored by performing regular measurements of its electrical resistance.

A constant potential difference was applied to a model which caused a constant electric current in it. One of the main problems was the choice of proper measuring electrodes (made from carbon, very good quality stainless steel and hand made non polarizable potential electrodes assembled from pen plastic containers, copper wires and saturated copper sulphate solution. Different electrodes were tested for self-polarization and it was

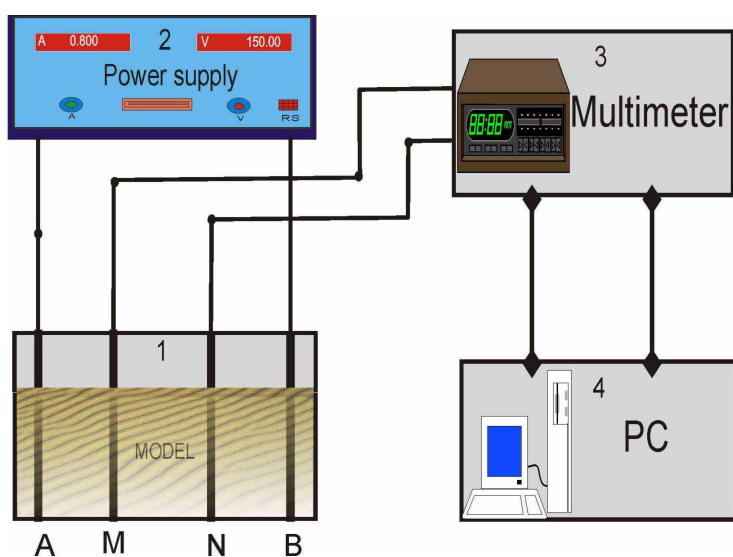


Fig. 2. A block diagram of the stand used for laboratory investigations of the relaxation of electrically induced (with DC) polarization signals: 1 – a porous medium model with electrodes (AB, MN), 2 – DC power supply, 3 – a digital automatic multimeter with an RS-232 interface, 4 – PC-computer

found that carbon electrodes are characterized by the most stable parameters, but their self polarization potential is the highest whereby relaxation effects in their final phase (above 30÷50 seconds) cannot be examined in detail. With standard non polarizable potential electrodes another problem occurs – initial current flow limitation was observed. Therefore stainless steel electrodes were employed even though this entails additional check measurements. At this stage of the experiments a two-electrode measurement methodology was employed, i.e. the same electrodes were used to introduce a polarizing voltage and to register the relaxation signal. The use of the two-electrode system was justified by the less complicated (and so easier to interpret) current distribution in the medium. When four electrodes were used, a momentary change in the sign of the signal was observed. This can be explained by, for example, electrode polarization in such a measuring system (the electric field's direction being opposite to the direction of the relaxation field) and by the fact that under the action of an external electric field liquid molecules as they move in capillaries orient their axes in the direction in which the field propagates. Naturally, such orientation of the molecules creates its own electric field which when the external field is removed, will decrease over time, resulting in a complicated character of the investigated signal [18].

Experimental results

At the initial stage, the most difficult problem was to synchronize the instant of polarization current switch off with the start of relaxation signal recording. This is due to the very short duration (from a few microseconds to a millisecond) of the initial phase of relaxation when a sharp drop in voltage to a level of a few volts is observed. At this stage the largest differences in the character of relaxation signals for similar media saturated with different liquids exist. It was noticed, however, that in the case of the petroleum saturated models, as opposed to the medium saturated with water, the relaxation time in this stage would lengthen and tend towards to the right limit of the millisecond range.

Experimental studies have shown that the filler significantly influences the nature and the speed of the relaxation of the electric field. The smallest relaxation times, initial polarizations and the largest disappearing speed were observed for the distilled water, while the largest initial values of the polarization were observed for the model filled with the electrolyte solution. Our

A special custom-built computer-controlled power supply unit was used as the source of electric current. The voltage or the current could be stabilized. The values of the potential difference and current were carefully controlled. The Author measured the dynamics of the potential difference between some internal sample points using the special measuring system with the RS-232 interface, hooked up to a PC, and automatically recorded the transmitted measurement data. These measurements were automatically transferred to the computer for processing. The experiments were conducted for different initial values of the external DC voltage from 20 V to 400 V, with the voltage increased by 10 V steps and a current up to 6 A, without pulsation. Normally the voltage was stabilized. The time of application of the low constant voltage causing no noticeable heating is 15 min (in a comparison to traditional time domain IP method this time is very long). For larger voltages the heating can become significant, and the time of application of the voltage was reduced to 2÷3 min. The initial low DC voltage (20 V) was subsequently increased stepwise up to 400 V. At the same time the current flowing in the medium was registered. Normally the electric field acted on the model for 15 minutes and polarization relaxation was registered for about 5 minutes (its value was automatically read every half of second with a simultaneous printing obtained curve on a paper).

results also show that the largest times for the voltage drop are obtained for a mixture of water and oil used as filler with the relaxation curves highly depending on the concentration of oil.

Fig. 3 shows the results of the automatic registration of the second phase (above 0.5 sec) of polarization signal relaxation, obtained for the model saturated with different liquids for the stimulating voltage of 120 V. According to the results, the system saturated with distilled water is characterized by the lowest potential after polarization and a very short relaxation time (curve 5). For models saturated with petroleum with the addition of a salt water solution (up to value 5% of the filler total volume) the lowered polarization potential which markedly decreased with the petroleum content in the model and the extended relaxation time {curves 3 (150 ml of petroleum) and 4 (300 ml of petroleum), respectively}, are proper. The models saturated with a table salt solution have the highest initial potential. It was noticed, however, that the absolute signal relaxation time in the latter models is shorter in comparison with the

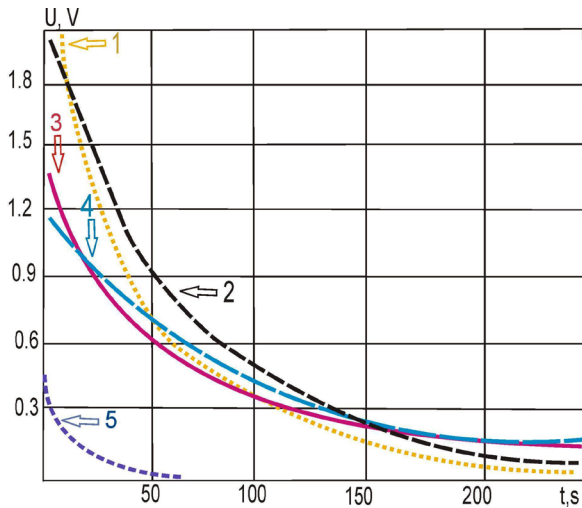


Fig. 3. The automatic registration of the second phase (above 0.5 sec) of polarization signal relaxation, obtained for the model saturated with the different liquids for the stimulating voltage of 120 V: 1, 2 – NaCl saturation, 3, 4 – oil emulsion saturation, 5 – distilled water saturation

models saturated with petroleum {curves 1 (6% solution) and 2 (3% solution) respectively}.

Quite interesting results were obtained by changing stepwise (every 10 V) polarization voltage amplitude while the other system parameters remained constant. In this case, for each model the initial system potential increases linearly and its relaxation time is clearly dependent on the polarization voltage amplitude and it lengthens as the latter is increased, particularly in a range of 60÷200 V. Fig. 4 shows an example of the variation in the polarization relaxation of the models saturated with water salt solution (3%) for different polarization voltage amplitudes: (1) – 180, (2) – 140, (3) – 100, (4) – 60 V (curves 1–4, respectively).

Fig. 5 shows the automatic registration of the second phase (above 0.5 sec) of the relaxation of polarization signal, obtained for the model saturated with the petroleum emulsion with different proportional with water salt solution. Let us note, however, that relaxation time lengthens the most for the models saturated with petroleum in a comparison to models saturated with a brine of different salinity.

Also the system memory effect, consisting in the lengthening of polarization relaxation time after several cycling training of the model, was observed for constant polarization voltages, but this phenomenon requires more detailed investigations.

In the author’s opinion the different relaxation effects in the polarized model of saturated rocks can be explained by the different physical properties of the investigated media, which are associated with their type of conductance. In the

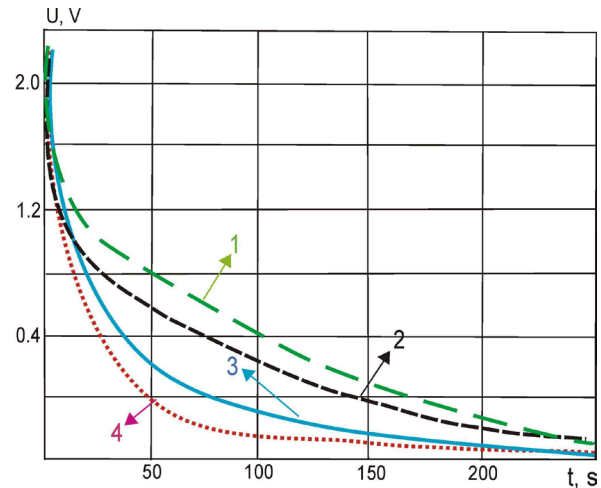


Fig. 4. An example of the variation in the polarization relaxation of the models saturated with water salt solution for different polarization voltage amplitudes: (1) – 180, (2) – 140, (3) – 100, (4) – 60 V

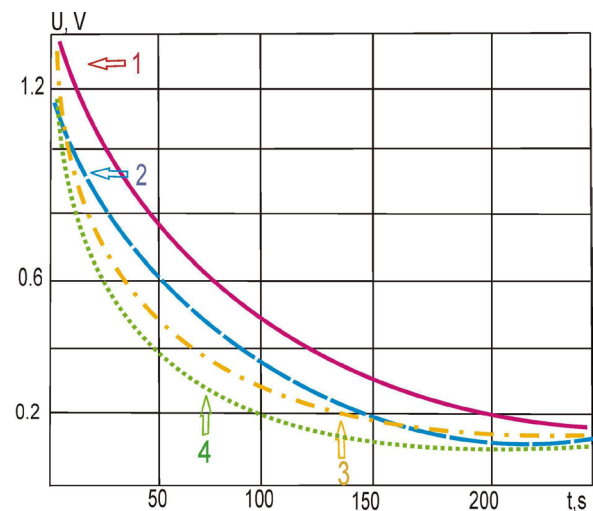


Fig. 5. The automatic registration of the second phase (above 0.5 sec) of polarization signal relaxation, obtained for the model saturated with the petroleum emulsion with different proportional with water salt solution (5% of the filler total volume) for the stimulating voltage of 120 V: 1 – 150 ml of petroleum, 2 – 200 ml of petroleum, 3 – 250 ml of petroleum, 4 – 300 ml of petroleum

models saturated with a salt solution, the faster electron-ion conductivity occurs, whereas in the petroleum saturated models the slower molecular conductance takes place. The mechanism of the latter is connected with the destruction of molecular compounds under the action of an electric field and probably with the formation and decay of free radicals, similarly as in the case of the action of a very strong ultrasonic field [17].

The obtained results are characterized by good repeatability and can be easily reproduced.

Final remarks

Schlumberger discovered the IP effect in 1920. The nature of the phenomenon is rather complex and still not understood completely after nearly 90 years [27]. Special electrochemical investigations are needed to explain the complex nature of the IP. Organic electrochemistry is not well studied, compared to other types of reactions that cause the IP effect and still, there is insufficient study of how the IP parameters correlate with hydrocarbon saturation. Many unsolved problems remain related to the IP effect that require more research. Organic chemical liquids form a problematic group for geophysical studies because their poor conductivity restricts the use of conventional electric and electromagnetic methods [4]. But the IP method can be used to detect organic contaminants because soil – organic mixtures are polarizable even with very low contents of clay minerals [24]. The mechanisms that cause the polarization effects in these mixtures can be related with interaction be-

tween minerals and organic molecule and produce changes in the chargeability values. Thus, from the chargeability value is possible to understand these mechanisms, which is very important. As it was mentioned before, IP effects have been approached by different types of equivalent circuits. That is, real systems are very complex and it is clear that further research, beyond the scope of this work, has to be done.

The above experiments were the first step in investigations of the phenomenon of the IP signal relaxation using non-traditional laboratory research methods. Similar measurements are to be conducted in the future, but using other medium hard phase models, in order to study in more detail the development of the effects and to propose a diagnostic parameter which will make it possible to distinguish sedimentary rocks with regard to the type of saturating medium.

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