Impedance Spectroscopy on Carbonates

Matthias Halisch⁽¹⁾, Andreas Weller⁽²⁾, Mohamed A. Kassab⁽³⁾

⁽¹⁾Leibniz Institute for Applied Geophysics (LIAG), Dept. 5 Petrophysics & Borehole Geophysics, Stilleweg 2, D-30655, Hannover, Germany
⁽²⁾Clausthal University of Technology, Institute of Geophysics (IfG), Arnold-Sommerfeld-Straße 1, D-38678, Clausthal-Zellerfeld, Germany
⁽³⁾Egyptian Petroleum Research Institute (EPRI), Nasr City, Cairo, Egypt

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ABSTRACT

With impedance spectroscopy (IS), or spectral induced polarization (SIP) as it is also termed, the complex electrical conductivity of rocks is measured over a wide frequency range, typically ranging from 1 mHz to 10 kHz. Originally developed for the exploration of ore deposits, today's SIP measurements are used for a wide range of applications within geophysics. On the field scale, this technique is (e.g.) used to characterize the hydraulics of shallow aquifers, to assess pollution for environmental protection purposes, or even to support archaeological investigations. On the laboratory scale, SIP is commonly used for the investigation of electric rock properties for clastic reservoir rocks in general, and sandstones in particular.

The frequency dependent complex electrical conductivity gives access to an advanced rock characterization, even beyond a more qualitative and in most cases general pore space characterization. Parameters particularly relevant for hydraulics – as for example specific internal surface, (dominant) pore size and permeability – can be estimated. Some robust empirical relations between induced polarization (IP) parameters and petrophysical parameters (BET surface, surface conductivity, cation exchange capacity) have been identified for sandstones and sandy material. However, for carbonates these relations do not seem to work and some of the resulting parameters show a quite different behavior. More systematic studies are needed to evaluate the potential of SIP for characterization of carbonates. The authors would like to show the results of a first case study on a limestone sample set originating from shallow wells of the Tushka area, south Western Desert, Egypt. Besides basic principles of SIP in general, adaptation of theory and evaluation technique will be highlighted in particular.

INTRODUCTION

Electric resistivity, as well as electric conductivity is described as complex quantity values [1]. As explained by [2], complex conductivity (σ^*) of a rock sample is determined by measuring the magnitude of conductivity ($|\sigma|$) as well as the phase shift (φ), relative to a reference resistor. The real (σ') and imaginary (σ'') part of σ^* , which represent the ohmic conduction and polarization charge transport mechanisms, respectively, are directly determined from impedance measurements. The phase shift thereby is defined as:

$$\varphi = \arctan\left(\frac{\sigma''}{\sigma'}\right) \sim \frac{\sigma''}{\sigma'} \text{ (for } \varphi < 100 \text{ mrad}),$$
 (E1)

which is - by convention - defined as a positive value in conductivity space. Generally, complex conductivity models are based on two contributing conductivity terms, as denoted amongst others by [3]:

$$\sigma^* = \sigma_{el} + \sigma_{surf}^* \,. \tag{E2}$$

The first term (σ_{el}) represents the conductivity of the electrolyte filled interconnected pore space, whereas the second term (σ_{surf}^*) reflects conduction and polarization within the Electrical Double Layer (EDL) of the interconnected pore (matrix) surface, respectively. With this, polarization solely is connected with surface conductivity at low frequencies [4]. Accordingly, conductivity can be expressed in terms of the real and imaginary part as follows [2]:

$$\sigma' = \frac{1}{F}\sigma_{el} + \sigma'_{surf} , \qquad (E3)$$

$$\sigma'' = \sigma''_{surf} . \tag{E4}$$

As a result it may be concluded that the boundary layer between the grains and the pore fillings is of utmost importance for the description of the IP-effect. The development of this boundary layer is described in detail amongst others by [5]. Summarized, this boundary layer is formed by two different layers in particular: first, by the fixed ions, which are adsorbed at the surface of mineral grains. This layer is also known as the Sternlayer. The transition zone between this fixed layer and the electrolyte within the pore is described as a diffusely distributed area of ions, whose concentration decreases exponentially with distance from the grain surface. This area is known as the Gouy-layer. Together, Stern- and Gouy-layer form the so called *Electrical Double Layer* (EDL). In addition, also electromagnetic coupling effects at frequencies > 1 kHz, which are caused by inductive and capacitive reciprocities, need to be taken into account for ISmeasurements [6]. Because of this, SIP-measurements are mostly performed in between frequencies of 1 mHz to 1 kHz or even lower (100 Hz). The SIP method itself has developed from classic frequency domain IP, and takes both, electric resistivity as well as phase shift into account. This method has become a potentially good tool for the enhanced characterization of rocks as well as of soils, due to the dependence of the measurements on the internal surface area (Spor) of the investigated materials [5; 7; 8; 9].

Because of the evidence of such a relationship, strong efforts have been taken into account to predict permeability by using SIP method [10; 11; 12]. Nevertheless, concerning this method for solid rocks, all of these investigations have been performed for (porous) sandstones. Consequently, transferring this knowledge for the application of SIP on carbonates is the next substantial progress in order to assess the feasibility of this method for other types of porous rocks.

EXPERIMENTAL SETUP & RESULTS

For a first study, 41 carbonate samples originating from seven shallow wells from the Tushka area of the western desert, Egypt, have been investigated. The main geomorphologic unit recognized in the study area is the Sinn El-Kaddab limestone plateau. Extensive geological and stratigraphical information for this area can be found in [13; 14]. The studied cores are composed of molluscan chalky limestone. According to the stratigraphic succession and the petrographical studies, the samples are taken from subsurface occurrences of late Cretaceous. Three microfacies associations (MFA) are described and have been used in previous studies [15; 16]. The first group of 26 samples (MFA1) includes bio-cortoids intraclastic packstones or grainstones with moldic pores. Bio-dolomitic molluscan intraclastic glauconitic floatstones and packstones belong to MFA2, which is represented by seven samples. The pore space of this association is related to dolomitic replacement. Bio-glauconitic floatstones and packstones are assigned to MFA3, which is characterized by intergranular pores (containing eight samples in total). The glauconite content for the investigated samples varies between 3.4 % and 26 %. To describe the complex experimental workflow for this study in detail would be beyond the scope of this paper. Extensive details concerning SIP multi-salinity experiments and data processing techniques, which have been performed for this study (e.g. related correlations, Debye-Decomposition, etc.), can be found amongst others in [2; 9; 17; 18].

Figure 1 exemplifies the results for the SIP measurements of the Tushka carbonates. In total, four different types of curves can be identified: minimum type (fig.1, A), maximum type (fig.1, B), max-min type (fig.1, C) and constant slope (fig.1, D). The green line indicates the first measurement, the blue line the second measurement (one day later). These different types of curves clearly indicate polarization effects within the samples. However, against first expectations, the specific curve types occur randomly within the sample set and do not correlate with the MFA, which have been described at the beginning of this chapter.

Figure 2 (left hand side) shows the relationship between σ'' and σ'_{surf} (evaluated at a frequency of 1 Hz which is standard for single frequency IP) including data for sandstones as the observed and described by [18]. For both rock types, data can be well explained by a single linear gradient: 0.042 for sandstones, and 0.019 for carbonates with a coefficient of determination, R², of 0.911 and 0.853, respectively.



<u>Figure 2</u>: σ'' and σ'_{surf} (@ 1 Hz) relationship for sandstones (after [18]) and for the investigated carbonate samples from Tushka (left hand side) and σ'' (@ 1 Hz) and S_{por} relationship for the investigated carbonate samples and in comparison with several sandstone data sets (modified after [9], right hand side).

This result indicates that surface conductivity dominates the conductivity and polarization mechanism of this sample set, as it has been observed for sandstones before (compare: [18]). That suggests to correlate the specific pore space related internal surface (S_{por} , determined with N_2 adsorption) with the imaginary part of conductivity. The right hand side of figure 2 shows this relationship for the carbonate sample set in combination with sandstone data as collected and published by [9]. Actually, MFA1 and MFA2 together form one cluster whereas MFA3 forms another, clearly shifted towards higher surface areas but without being shifted towards higher imaginary conductivity values. Despite clustering, fitting and trend predicting does not seem feasible. Furthermore, if this data

set is included into the well described correlation for sandstones, the result is again unexpected. Due to the higher specific surface area (roughly ranging from 11 to 400 $1/\mu$ m) of the carbonates it may be expected, that these samples would sustain the correlation of the sandstones. Obviously, they do not follow this trend, as they tend to lower imaginary conductivity values (about two orders of magnitude lower than expected by surface data). Figure 3 highlights the correlation between measured sample permeability and imaginary part of conductivity and in addition measured vs. predicted permeability of this sample set in comparison with data from Egyptian sandstones. Surprisingly, a positive correlation between sample permeability and σ'' has been found (fig.3, left hand side). The calculated permeability has been estimated from parameters ρ_{DC} and m_t resulting from Debye decomposition of the SIP data as described by [17] and has been accordingly added into the sandstone correlation plot (fig.3, right hand side) as described by [12]. As a result, calculated carbonate permeability is over-estimated by one order of magnitude for measured permeabilities equal or smaller to 0.1 mD. For measured permeabilities in range of 0.1 to 10 mD, calculated data fit well into the correlation. For measured permeabilities greater than 10 mD, calculated K tends to be under-estimated by one order of magnitude.



<u>Figure 3:</u> K and $\sigma''(@ 1 \text{ Hz})$ relationship for the investigated carbonate samples (left hand side) and comparison of predicted vs. measured permeability between the studied carbonates and several sandstone data sets also originating from Egypt (right hand side, modified after [12]).

CONCLUSIONS & OUTLOOK

The authors have shown that carbonates clearly indicate polarization effects that have been observed and described before for different types of sandstones (fig.1). Accordingly, there is a strong relationship between the real and imaginary part of surface conductivity for both, sandstones as well as for carbonates (fig.2). However, the expected relationship between imaginary part of conductivity and specific internal surface area has not been observed (fig.2, right hand side), which may be an effect of different MFA, different pore network development and / or mineralogy. Consequently, common permeability prediction models over- or under-estimate measured permeability (fig.3). It is planned to

greatly extend this first applicability study for SIP measurements on carbonates. More carbonate sample sets from different type locations and also with larger and broader range of petrophysical parameters (e.g. permeability range from 0.01 mD up to 2 D) will be investigated. In addition, more and also more different microfacies associations will be taken into account as well as different types of pore networks (vuggy as well as well interconnected pores), in order to find conclusive explanations for the different surface conductivity – surface area relationship. The final goal is to understand the (dominating) polarization effects for carbonates in close comparison to the well known mechanisms for sandstones, as well as to adapt and improve SIP data processing for better correlation and prediction of petrophysical parameters from SIP data.

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