

A new and modular laboratory core holder for high-precision measurements with low frequency impedance spectroscopy on natural rocks

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ABSTRACT

Investigating complex electrical properties of natural rocks and soils by using spectral induced polarization (SIP, i.e. “low frequency impedance spectroscopy, typically from 1 mHz up to 100 kHz) is of high research interest for all pore space and boundary surface specific processes and properties between the matrix and the pore fluid. The deduction of hydraulic and pore space related structural properties, as well as the correlation of SIP-data with CAL and SCAL data is recently in the focus of research. In the past ten years, especially the development of new measurement equipment has greatly improved the quality of the results. Nevertheless, no uniform standards and technical requirements exist for adequate core holders for the determination of SIP related parameters. Furthermore, most of the core holders do not take different materials (unconsolidated, consolidated) or different core geometries (diameter, length, cross-section shape) into account. Within the department Petrophysics and Borehole Geophysics of the Leibniz Institute for Applied Geophysics (LIAG), a new and modular core holder for measuring complex electrical properties has been developed, in order to address the mentioned needs and functionalities. In addition, the core holder is characterized by a significantly reduced residual noise, so that also rock materials, which feature low frequency phase effects (such as Fontainebleau or Nivelsteiner Sandstone), can be investigated with high measurement accuracy and excellent reproducibility. The core holder is currently pending for patent and already commercially available for interested working groups.

INTRODUCTION

Spectral Induced Polarization (SIP) measurements (i.e. “low frequency” impedance spectroscopy, typically conducted in a frequency range from 1 mHz up to 100 kHz) are used in many different ways to characterize natural rocks and soils. Main foci of interest are the enhanced characterization of the causes of polarization effects in sedimentary rocks. The interactions between the matrix-fluid-system and within the electrical double layer as well as the correlation with petrophysical parameters, such as specific surface area, permeability, and pore radii distribution as derived from mercury intrusion capillary pressure data are considered. A variety of polarization models, either grain based or pore based, have been developed over the past years to describe the polarization effects of

sedimentary rocks [e.g.: 1; 2; 3; 4; 5; 6; 7]. The electric resistivity, as well as electric conductivity are both described as complex quantity values. As explained by [8], 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:

$$\varphi = \arctan(\sigma''/\sigma') \approx \sigma''/\sigma' \text{ (for } \varphi < 100 \text{ mrad)}, \quad (\text{E1})$$

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 [9]:

$$\sigma^* = \sigma_{\text{el}} + \sigma^*_{\text{surf}}. \quad (\text{E2})$$

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. 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 impedance spectroscopy (IS) measurements [10]. 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 (S_{por}) of the investigated materials [e.g.: 11]. Additionally, strong efforts have been taken into account to predict permeability by using SIP method [12; 13;].

Hence, it is favorable to study a wide frequency range with SIP-measurements, in order to cover the majority of IP related parameters, effects and processes. Nevertheless, most of the SIP devices produce accurate and reliable data only within a limited frequency range. These issues have been addressed by the manufactures, which have been improving their equipment consequently over the last years [14]. Unfortunately, the type of electrode system used leads to restrictive limitations, even if the electronics of the instrumentation fulfills state of the art demands [15]. For 2-electrode systems, the electric coupling between sample and electrodes may cause transfer impedance effects, which furthermore cause frequency dependent disturbances on a wide frequency range (from 1 mHz up to the kHz domain). For 4-electrode setups, electromagnetic coupling, especially between connection wires, also lead to frequency dependent errors, typically above frequencies of 100 Hz to 1 kHz. Besides these instrumental influences, specific core holder effects, such as coupling issues of the sample-electrode system, self-polarization effects of the electrode material, leakage currents alongside the core surface and evaporation of the saturating fluid have severe impact upon the quality of SIP data. Taking this as motivation, the concept for the new core holder was created accordingly.

CONCEPT

According to these challenges, the sample holder needs to be constructed completely out of non-polarizable and brine-resistant material (i.e. free of any metal), including non-polarizable potential and non-oxidizing (i.e. electro-chemically inert) current electrodes. Besides these technical requirements, more universal demands exist and need to be addressed as best as possible. Hence, such a core holder should be easy to handle, as far as possible maintenance free, compactly designed and able to fit different core sizes without changing the entire measurement setup. Figure 1 showcases a schematic exploded assembly drawing of the general concept and design of the SIP core holder.

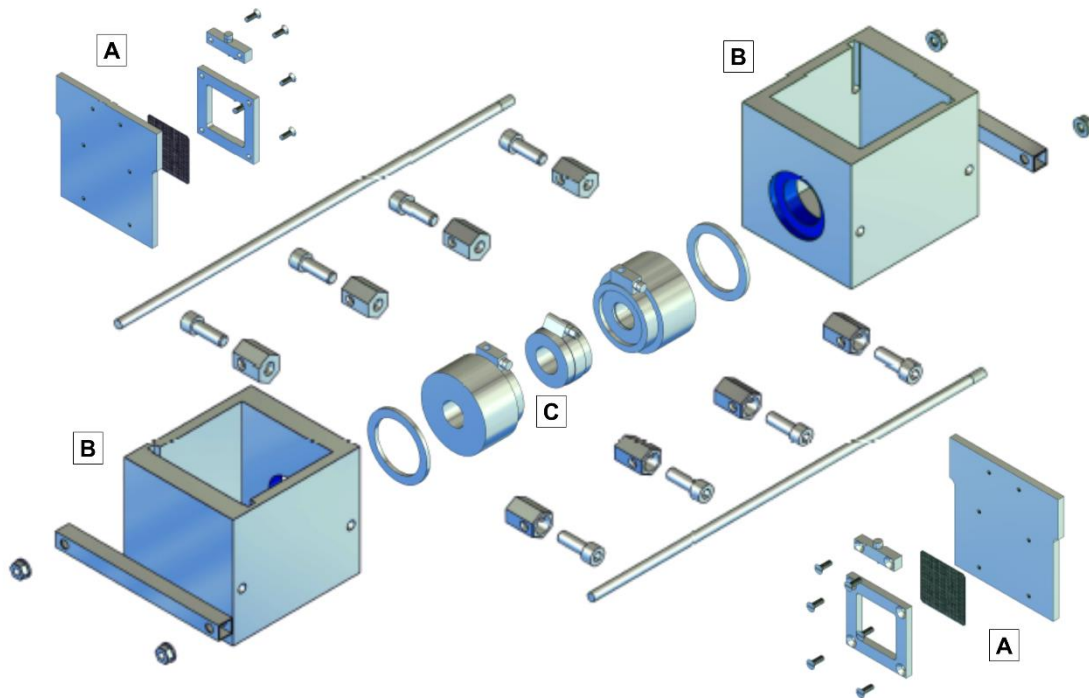
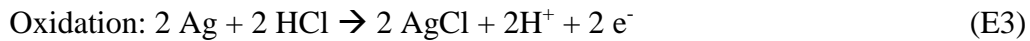


Figure 1: Schematic exploded assembly drawing of the core holder / cell for SIP measurements. (A): current electrode holder with platinum net electrodes; (B): acrylic glass electrolyte reservoirs; (C): potential electrode adapter for silver chloride ring-electrodes and core holder sleeve.

The SIP core holder is basically made out of three main modules: the current electrodes (A), the electrolyte (brine) reservoirs (B), and the core sleeve with the potential electrode adapters (C). The current electrodes are made out 8 x 8 cm of platinum net, with a mesh aperture of 125 μm to guarantee a highly uniform distributed current density along the medial core holder axis. The electrode holders are made out of acrylic glass and can be connected to the power generator with a 2 mm socket plug. The assembled current electrodes are raked into a guide rail, located at the edge of the electrolyte reservoirs (B). The reservoir tanks are connected with two guide rails on each side, which allows to adjust the electrode distance flexibly, depending upon the length of the core plug. Main part of the SIP cell is the sample sleeve (C), including two adapter for the potential electrodes, which are made as a ring-electrodes. Thus we can achieve the best possible signal-to-noise

ratio as well as the best possible coupling towards the sample material and the saturation fluid, respectively. This module can be easily replaced to address a variety of plug lengths and diameters (length: 30 mm to 80 mm; diameter: 20 mm to 45 mm). The electrodes are made out of silver wire (99.9 % purity; diameter 0.4 mm), which can be chlorinated in order to prevent polarization effects on the electrode surface. For this, the electrode can be connected to a power supply by using the 2 mm socket plug, which usually is used to measure the potential signal. Then, the adapter module is placed in a vessel filled with hydrochloric acid and a small alternating potential is applied (1 – 2 V for about 5 – 10 minutes). By doing this, the silver is electrochemically oxidized:



This chloride layer depletes over time and several measurements, and hence needs to be reproduced occasionally. By experience, every 5 samples this procedure needs to be repeated. Additionally, the sample is placed in a chemically inert silica rubber sleeve, which is tightened with a small hose clamp. By doing this, the rubber adapts towards the sample surface and prevents creeping currents alongside the skin surface. Furthermore, this sleeve seals the entire potential electrode module towards saturation fluid leakages, as soon as the reservoir tanks are clutched along the guide rails. Figure 2 shows the core holder as side face view. The position of the electrodes and of the sample are highlighted.

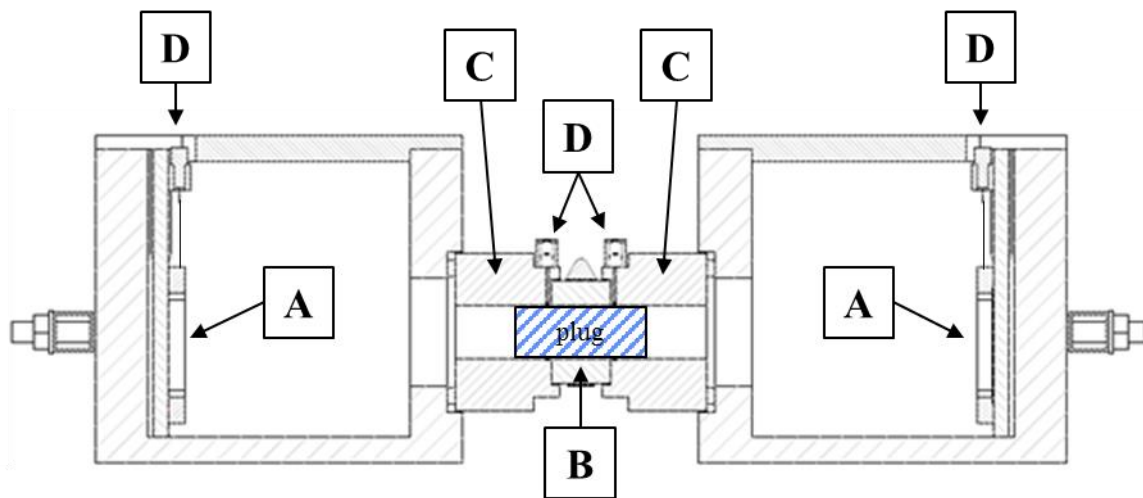


Figure 2: Schematic side view of the SIP sample cell. The current electrodes (A) are located in the electrolyte reservoirs. The sample is fixed within the silica rubber sleeve (B), which is fixed by the potential electrode adapters (C). Current as well as electric potential can be measured by 2 mm socket plugs (D).

RESULTS & DISCUSSION

For testing of the new sample cell, a simple experiment has been conducted. The main goal of this improved cell was to reduce the “noise”, i.e. measuring errors caused by coupling, creeping currents and self-polarization of the electrodes, to the largest possible extent. Hence, a non-polarizable reference material should be used, in this specific case, a sodium-chloride solution with approximately 100 mS/m specific conductivity. In an ideal case, the measured phase shift should be equal to zero. A small round robin test between three different institutions (LIAG, Bundesanstalt für Materialforschung und –prüfung = BAM, Bundesanstalt für Geowissenschaften und Rohstoffe = BGR, and Clausthal University of Technology = TUC) has been performed to achieve independent results for different 4-electrode SIP core holders. The results for the phase shift measurement for a frequency range from 1 mHz to 100 kHz (according to equation E1) are presented in figure 3.

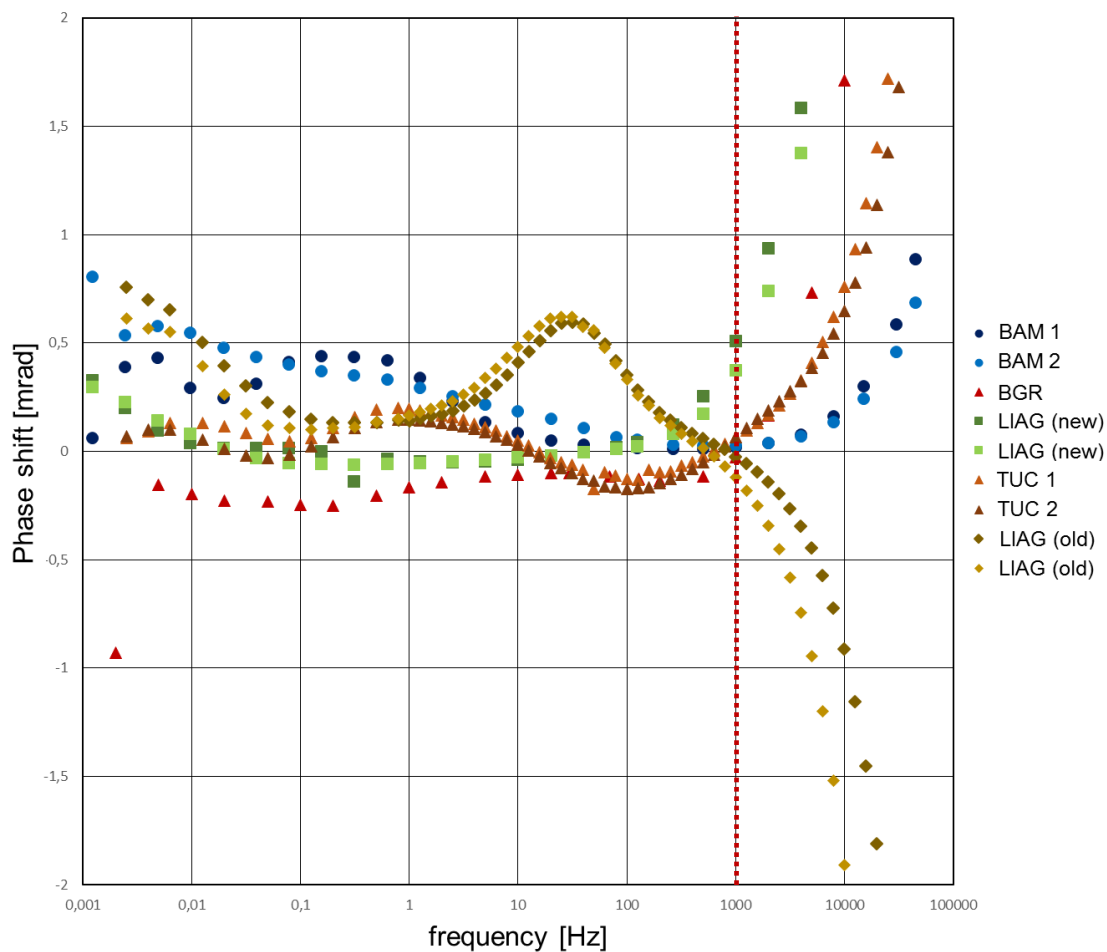


Figure 3: Representation of the results of the phase shift measurements on sodium-chloride solution with a specific conductivity of approximately 100 mS/m for the small round robin test amongst three different institutions (LIAG, BAM and TUC). The dashed red line indicates the threshold frequency, from which upwards electro-magnetic coupling effects dominate the measurements.

As showcased, all three core holders feature low to very low “noise-effects” (± 0.5 to 1 mrad). For frequency ranges > 1 kHz, electro-magnetic coupling effects dominate the results, and hence should be neglected. The new LIAG SIP core cell features the overall lowest “noise-level” (< 0.25 mrad) for all of the used cells, closely followed by the TUC (< 0.5 mrad) and the BAM (< 1 mrad) core holder. For comparison, the old LIAG core holder (also a 4-electrode core holder, but with non-optimized electrode configuration and platinum electrode material) has been measured as well (brownish diamonds), showcasing the most pronounced noise (phase maximum around 20 Hz) for all of the investigated cells.

SUMMARY & CONCLUSIONS

For this paper, the authors have showcased the motivation and the concept for the development of a new core holder for high-precision spectral induced polarization measurements on natural rocks (figure 4). The modular construction enables flexible handling of different sample sizes and geometries, combined with high user-friendliness and low maintenance efforts. The results of the round robin test have shown, that the LIAG cell has pushed the edge of “low-noise” SIP core holders a little bit further, featuring the lowest phase shift of all investigated cells. Hence, this new setup is particularly suitable for investigating rocks that feature low phase effects such as the Fontainebleau Sandstone as presented in [16, this SCA proceedings] with high accuracy and with high reliability.

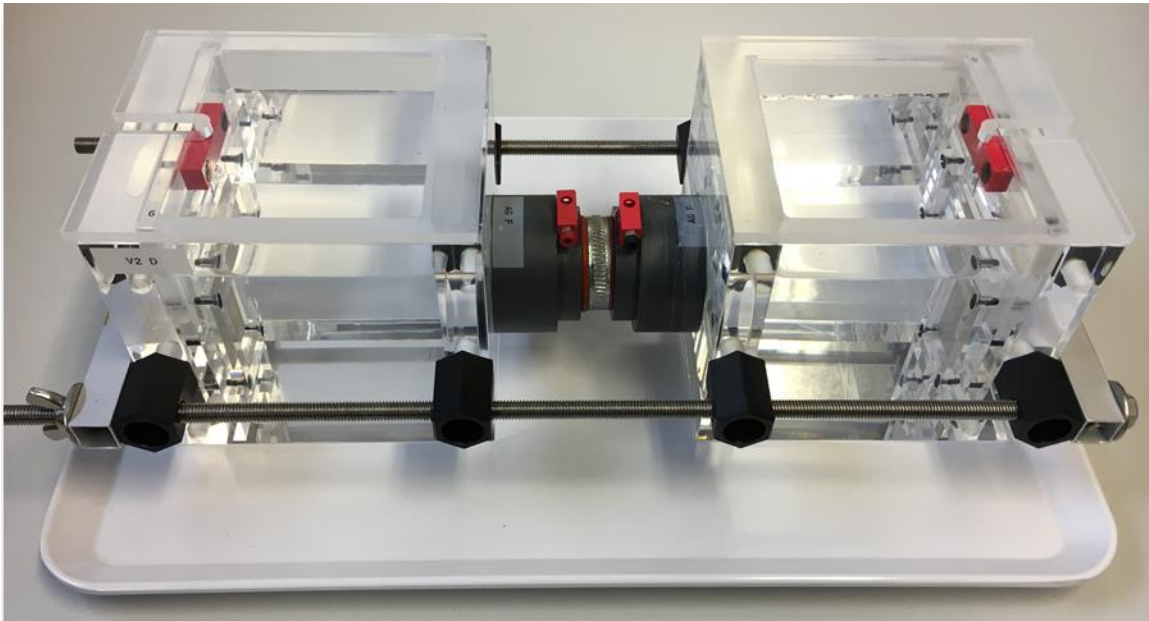


Figure 4: The actual version (Mk. II) of the LIAG 4-electrode core holder and measuring cell for high-precision SIP-measurements.

OUTLOOK

The ongoing development and improvement of this type core holder for spectral induced polarization measurements is one main task for the Petrophysics working group at the LIAG. Consequently, the current version of this core holder has been registered for patent, and the successful announcement is expected to arrive soon. Already now, the SIP cell is commercially available at the LIAG. In a next development step, this core holder is being adopted and re-configured (e.g. the potential electrode layout) for measuring the complex electrical properties of unconsolidated material (soils, unconsolidated sands and clayey materials). Additionally, this core holder will be prepared to fit the requirements to measure NMR spectra without removing the sample material from the SIP sample holder (i.e. no metal parts are allowed to be placed inside of the NMR magnet). Furthermore, the parallel measurement of the streaming potential as well as the application of overburden stress (triaxial, up to 300 bar, i.e. approx. 4400 psi) are under development.

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