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Hydrology and Earth System Sciences Discussions

Interactive comment on "Impact of controlled changes in grain size and pore space characteristics on the hydraulic conductivity and spectral induced polarization response of "proxies" of saturated alluvial sediments" *by* K. Koch et al.

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We found the comments of the referees very insightful, constructive, and useful and hence we shall make a concerted effort to accommodate them in their entirety in the course of the revised version of this manuscript. In the following, we outline in detail our proposed reactions to these reviews.

Replies to the Comments by Referee André Revil:

The manuscript by Koch et al. discusses the effect of the texture of very simple porous media (proxies of saturated alluvial sediments) upon the spectral induced polarization response in the laboratory. I am not aware of the publication of such a work in the literature. I especially like the compaction experiments as they show that something is going on in the relaxation time that is not described in available models. This manuscript is well written.

The description of the EDL is a bit fuzzy: we can read "towards the outer limit of the EDL, where ions are in equilibrium with the solution" is not correct. Indeed each element of the diffuse layer is in equilibrium with the neutral part of the pore water. Actually the Bolzmann distributions for the concentration profiles of the cations and anions are derived from the equality of the electrochemical potentials between any distance in the diffuse layer and infinity (for which the electrostatic potential is zero), see for instance Revil, A., & Glover, P.W.J., Theory of ionic surface electrical conduction in porous media, Physical Review B., 55(3), 1757-1773, 1997.

To address this issue, we propose to replace the existing text: “Beyond the Stern layer, positively charged ions continue to be attracted by the negatively charged mineral surface, but at the same time are repelled by each other and the Stern layer. The resulting dynamic equilibrium is referred to as the diffuse layer and represents the transition zone towards the outer limit of the EDL, where ions are in equilibrium with the solution and distributed in a random manner.”,

By the following: “Beyond the Stern layer, positively charged ions continue to be attracted by the negatively charged mineral surface, but at the same time are repelled by each other and the Stern layer. The resulting dynamic equilibrium is referred to as the diffuse layer and represents the transition zone between Stern layer and the neutral part of the pore water. The individual elements of the diffuse layer are hence in equilibrium with the neutral part of the pore water as is the basis for the concentration

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profiles of anions and cations from the Boltzmann distributions derived via the equality of the electrochemical potentials between any distance in the diffuse layer and infinity (e.g., Revil & Glover, 1997)".

Another sentence that is really wrong is the following: Much of today's conceptual understanding is based on the work of Schwarz" Actually (i) Schwarz never mentioned the Stern layer in his seminal paper. His model was supposed to represent the entire double layer. (ii) the standard model used in colloidal chemistry is the Dukhin and Shilov model based on the polarization of the diffuse layer, and (iii) most of the geophysicists still believe that the membrane polarization is the dominant mechanism of polarization. This is only since the work of Revil and co-workers (Leroy et al, 2008, Leroy and Revil, 2009, Jougnot et al., 2010, and Revil and Florsch, 2010) that the Stern layer has been considered to be the potential main contributor to low frequency complex resistivity. There was no work published previously in geophysics that were pointing out a dominant role to the Stern layer. De Lima and Lesmes in several papers pointed out the potential role of the Stern layer but the model they used was based on the Dukhin and Shilov theory of diffuse layer polarization. This historical note needs probably to be put in relief in this manuscript because one may believe that the Stern layer polarization model has always been something obvious to geophysicists, which is grossly untrue.

As the referee pointed out, "[It] is only recognized since the work of Revil and co-workers (Leroy et al, 2008, Leroy and Revil, 2009, Jougnot et al., 2010, and Revil and Florsch, 2010) that the Stern layer has been considered to be the potential main contributor to low frequency complex resistivity." In the above mentioned part of the introduction we did not actually consider the Stern layer, but the whole EDL which than further explains our discussion of the work by Titov (2004) and his attempt to provide a visualization of either pore size or grain size based characteristics and his quick review of granular and capillary models.

To clarify this issue, we propose to replace the existing text: "The EDL provides the conceptual background for the electrochemical processes considered to be responsible for much of the observed SIP response, as documented, for example by the recent study of Leroy et al. (2008). Much of today's conceptual understanding of the origin of the SIP response is based on the work of Schwarz (1962) and his interpretation of the SIP effect as a result of the redistribution of counter-ions surrounding spherical particles in suspension. The only geometric factor involved in his model is the size of the sphere. Translating this geometrically simple analytical model to texturally complex porous media is not evident. Titov et al. (2004) attempted to provide a visualization of the two basic conceptual views on the origin of the SIP effect in porous media linking it either to the grain size distribution or to the pore size distribution. A number of studies have attempted to gain further insight into these matters by attributing the polarization to the EDL surrounding individual grains (e.g., Lesmes & Morgan, 2001) and to excesses and deficiencies in ion concentrations along pore throats (e.g., Titov et al., 2002)."

With the following: "The EDL provides the conceptual background for the electrochemical processes considered to be responsible for the observed SIP response. Today's conceptual understanding of the origin of the SIP response is strongly based on the work of Schwarz (1962) and his interpretation of the polarization effect as a result of the redistribution of counter-ions surrounding spherical particles in suspension. This model comprises the entire double layer and does not differentiate between the Stern layer and the diffuse layer. The only geometric factor involved in this model is the size of the sphere. Translating this geometrically simple analytical model to texturally complex porous media is not evident, as it simply is too strong a simplification. The commonly applied models are based on polarization of the diffuse layer in the pore system (e.g., Dukhin and Shilov, 1974), where Titov et al. (2004), amongst others, attempted to provide a visualization of the two basic concepts with regard to the origin of the SIP effect in porous media by linking the relaxation length scale to either granular or capillary models. A number of studies have tried to gain further insight into these matters by attributing the polarization to the EDL surrounding the individual grains (e.g., Lesmes & Morgan, 2001) and to excesses and deficiencies in ion concentrations along pore

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throats (e.g., Titov et al., 2002). It is only based on the recent work of Leroy et al. (2008), Leroy & Revil (2009), Jougnot et al. (2010), and in particular Revil & Florsch (2010) that a more and more refined picture of polarization processes taking place in the Stern layer has started to emerge. Indeed, this work provides strong evidence to suggest that the Stern layer is potentially the region where the most important processes associated with the observed SIP response take place."

The model of Titov is a membrane polarization mechanism, it should not be lisleded with a Stern layer polarization mechanism as presently written in the text. Both contributions are however compatible (they can exist simultaneously). I therefore do not understand the discussion of the Tiitov model after the point raised that the polarization of the Stern layer is the dominant mechanism. The experiments and the interpretation of the results in term of the ColeCole parameters is good.

Please see above.

This sentence is going backward: These findings corroborate (...) are consistent with the original findings of Kozeny and Carman in that the specific surface area of a porous material is in general the determining parameter for permeability" I would say that this is simply untrue and 70 years of works in petrophysics have demonstrated that the specific surface area is certainly not the relevant parameter characterizing the permeability (think about dead ends for instance, you should read the following paper and references therein: Revil, A., and Cathles, L.M., Permeability of shaly sands, Water Resources Research, 35(3), 651-662, 1999. The authors should make for their benefit a short review of the available literature on this subject and this idea has been strongly discussed and fighted by many researchers.

We agree that the Kozeny-Carman comparison is misleading the way it was written. The authors think that the reference does not help the overall comprehension of the discussion of the results, and hence propose to remove it from the revised version of the manuscript.

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I fullly agree with the conclusions of the compaction experiments that are great and puzzling results. In conclusion, I think this ms can be published with minor revision. It is a very timely work.

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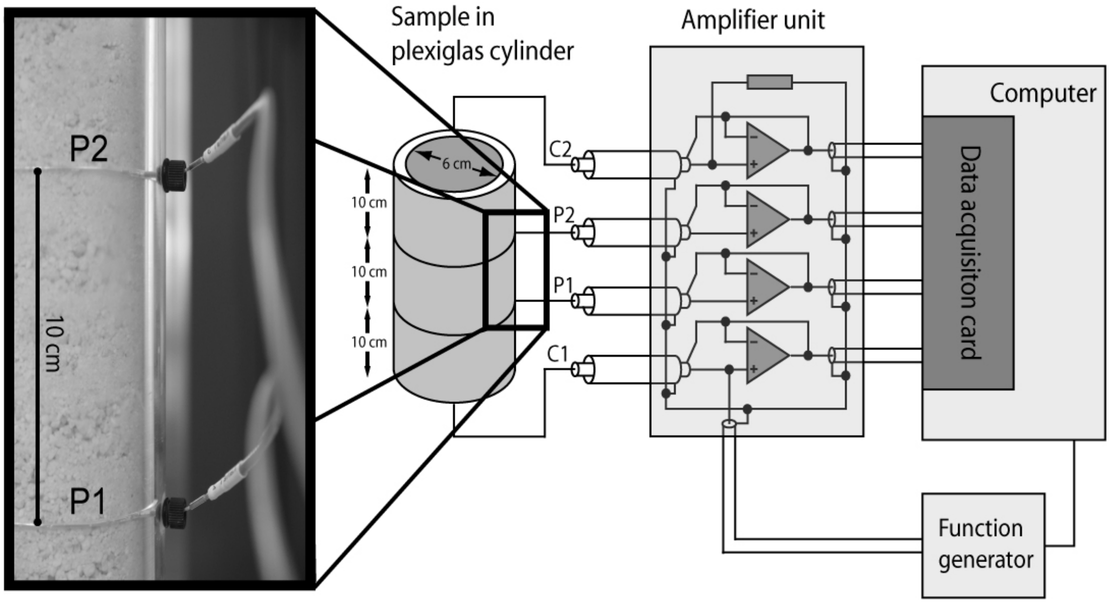

Fig. 1. Experimental setup and schematic illustration of the high-sensitivity impedance spectrometer used for the SIP measurements presented in this study.