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Reply to the comment by A. Revil on “Review of Self-potential methods in Hydrogeophysics”, by L Jouniaux et al. [C.R. Geoscience 341 (2009) 928-936]

Laurence Jouniaux[1], Alexis Maineult[2], Véronique Naudet[3], Marc Pessel[4], and Pascal Sailhac[1]

1. Institut de Physique du Globe de Strasbourg, UMR7516, CNRS et Université de Strasbourg (EOST), France
2. CNRS and PRES Sorbonne Paris Cité – Institut de Physique du Globe de Paris UMR7154, Paris, France
3. Université Bordeaux 1, Laboratoire GHYMAC-EA 4134 (Géosciences Hydrosciences Materiaux Constructions), Talence, and BRGM (French Geological Survey), Orléans, France
4. Laboratoire I.D.E.S., UMR 8148 CNRS/Université de Paris-Sud XI, 91405 Orsay, France.

Corresponding author : L. Jouniaux, CNRS/IPGS, 5 rue René Descartes, 67000 Strasbourg, France. Ph : 33 3 68 85 03 34/ fax : 33 3 68 85 01 25. l.jouniaux@unistra.fr

We did not discuss subjects that are still not well understood and still being debated since the title of our paper includes “review”. We did not review and discuss all aspects of the theoretical background based on microscaled thermodynamics because we focused on field applications in hydrogeophysics. Moreover we submitted our manuscript in 2008 and thus did not acknowledge any paper published in 2009. However this comment is of interest because it gives us the opportunity to provide more precision by answering some points.

We agree that there is an increasing number of publications in this field. Recently, electrokinetic models have been proposed for reservoir geophysics and petroleum investigation [28, 49]. New applications have also been developed, such as the use of SP measurements in boreholes as an electrical flowmeter [44]. Initially, we did not include it, but the electrokinetic coupling is also directly involved in seismoelectromagnetics effects [9, 10,16, 20, 24, 27, 52]. For hydrological applications, it is difficult to infer a steady link between SP intensity and water flux [11, 15, 30] or deformation [25, 29], although some authors have proposed to predict the permeability by using electrokinetic theory [22, 23].

We obviously agree that our eq. (1), which expresses the phenomenological law between fluxes and forces, is valid only close to thermodynamic equilibrium. This has been known for a long time and was already mentioned by Onsager and Nourbehecht [40, 41]. Indeed, these constitutive laws are used by many authors to interpret and to model self-potential observations [18, 19, 26, 35, 36, 42, 51, 55]. Far from equilibrium, high-order terms must be considered, but the linear case is sufficient for the interpretation of most of the field situations encountered by hydrogeophysicists. Finally, to our knowledge, A. Revil did not develop any theory describing the constitutive laws valid far from the thermodynamic equilibrium. Indeed, even when A. Revil considers chemical potentials and combines several thermodynamic forces, he uses fluxes related through linear relationships in a symmetric Onsager coupling matrix; this basically follows classical theory of irreversible thermodynamics near equilibrium [13, 14, 53].

The fact that the flow state can affect the streaming potential was shown experimentally before the work cited by A. Revil (his reference 4). For instance, an experimental study performed by Watanabe and Katagishi [54] on granular materials showed that the fluid flow began to deviate from Darcy's law at a Reynolds number larger than 3, and that streaming potentials began to deviate from the linear relation at larger Reynolds numbers, leading to a lower than expected streaming potential. Another study from Kuwano et al. [31] showed that the streaming current coefficient (electrokinetic coefficient multiplied by the rock conductivity) was constant at low Reynolds numbers, but decreased at Reynolds numbers larger than 1-10. They showed that this decrease was much smaller than that of permeability, leading to the conclusion that non-linear laminar flow induces more streaming current per unit flow rate. Nevertheless, as stated by Fetter [17], "under most natural ground-water conditions, the velocity is sufficiently low for Darcy's law to be valid" (with Reynolds numbers of less than 1 to 10). One major exception can be the vicinity of pumping wells (e.g., reference 21 of the comment).

We proposed eq. (5) for the electrokinetic coefficient as a function of the saturation. We wrote precisely: *the few observations published up to now [Guichet et al, 2003] show…eq. (5)*. Therefore this equation is based on the observations. The previous sentence was exactly: *Based on theoretical considerations it has been proposed [8, 54] that the electrokinetic coefficient is inversely proportional to the effective saturation*, which is not the eq. (5) we proposed, since this equation shows that the electrokinetic coefficient is proportional to the effective saturation. It is true that we refered to an experimental study, which is not verified by the model proposed by A. Revil. The model proposed by A. Revil uses a strong assumption, which is that the volumetric charge density in the water-saturated pore is inversely

proportional to the water saturation. However, we do not know if this assumption is valid. The model proposed by A. Revil (in his reference 8) is able to explain some of his measurements when extrapolating the value of the electrokinetic coefficient at full saturation from values obtained at various saturations using the model that the author wants to prove. However, this model cannot explain the values at full saturation of his measurements, previously published (in his reference 9), but unfortunately not reported in his last work (his reference 8). Moreover, other models were proposed for the electrokinetic coefficient as a function of water saturation [28, 43, 49]. Recently, for the first time, an experimental study presented continuous records of the electrokinetic coefficient as a function of water saturation. It showed that the electrokinetic coefficient does not depend monotonously on the saturation [1]. Therefore, we still think that further studies are needed in this domain.

We do not challenge the general formulation proposed by A. Revil (his eq. 1) for the current densities associated with the chemical potential. But we point out that this formulation is not easy to use for modelling and interpretation, in particular because the Hittorf numbers depend on the concentration, which can evolve with space and time. The form derived from the well-known generalized ionic diffusion equations underlines more clearly the link between the electrical potential gradient (measured in the field) and its source, I.e., the relative gradient of concentration (to be deduced from the potential gradient). It has been successfully used to explain the response of a NaCl, $FeCl_2$ and Kcl front flowing through sand [32, 33, 34].

We would also emphasize that the theoretical foundation of A. Revil's eq. (5) was first used by Arora et al. [2] from an experimental deduction by Naudet et al. [38].

Concerning the inverse problem of SP data, we do not expect great advances from the choice of the optimization method itself. Actually this choice helps, but is not the fundamental issue for breaking the non uniqueness of solutions: one should improve the accuracy of the data and the accuracy of the theory.

The recent success in connecting SP ground surface measurements to piezometric height is due essentially to the theory (use of Fournier's equation), and only partially results from the choice of the optimization algorithm ("PSO" versus "Simplex") [39]. Indeed, other authors have previously obtained realistic results by solving equations similar to that of Fournier by deconvolution [7, 8] or simply by fitting sliding proportionality factors [3, 4, 5]. Besides, using the wavelet method is discussed because this is one step in the improvement of the theory: the relationship between the stream function of groundwater flows and SP analytic signals can be better understood when

SP wavelet transforms are used to characterize ground water flow singularities [46]. This also allows one to reduce the non-uniqueness of the inverse problem without the need for classical regularization techniques: Tikhonov's additional terms to the data misfit function actually results in smoother models. Concerning the merit of the cross-correlation approach over wavelet-based techniques, we still invite our colleague to read Gibert and Sailhac [21]: one should not consider these methods as opposed because coefficients of "wavelet" and "cross-correlation" are essentially the same; moreover one should not use the inappropriate expression "tomography of probability" that was introduced by mistake for naming the plots of these coefficients.

Besides, we agree that improvements of SP data inversion will also come from the integration of SP data with different kinds of parameters sensitive to groundwater and more specifically to the electric conductivity tomography; this is the guideline of the work performed at the experimental site of La Soutte [6, 48]. Nevertheless, we consider that further improvements of the theory are necessary. Although the accuracy of future SP inversion techniques might be improved by a general Bayesian framework, one needs deeper insight into the joint hydro-electric potentials; there is still a number of undefined parameters involved in the joint phenomena of actual ground water flows and electric potentials. It is true that there exist experiments of inversion methods based upon joint hydro-electric modelling tackling hydraulic parameters. Some of these joint inversion methods use analytic formulas [12, 47] or numerical modelling [37, 45, 50]. There are still limitations related to necessary improvements in modelling the behaviour of the coupling coefficients in real soils, for instance in heterogeneous or unsaturated media, multiphase or multi-fluid flows, and with various dynamics. One still needs further developments and experiments for the theory to improve hydraulic flow accuracy when inverting SP data; this is why we consider that "moving tomography of the electrokinetic source term into the tomography of some hydraulic parameters (...) is the real challenge of the next decade".

This is IPGP contribution n°3007.

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