



Induced polarization dependence on pore space geometry: Empirical observations and mechanistic predictions



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ABSTRACT

We use an extensive database to compare empirical observations and previously proposed empirical models against recently developed mechanistic formulations for the induced polarization (IP) response in porous media as a function of pore space geometry and interfacial chemistry. These comparisons support the argument that the pore-volume normalized internal surface (S_{por}) is the most important geometric parameter influencing the polarization. The specific polarizability derived from the empirical relationship between imaginary conductivity σ'' and S_{por} is independent of the porosity. By contrast, equivalent specific polarizability terms in recently proposed mechanistic models are found to be significantly correlated with porosity, and thus do not appear to represent an interfacial chemistry factor independent of the pore space geometry. Furthermore, the database shows no evidence for a significant decrease in the counterion mobility of clayey materials relative to clay-free materials, as postulated in recent studies. On the contrary, a single value of c_p is consistent with no significant differences in ionic mobility given that all samples were saturated with a NaCl solution close to a common salinity of about 100 mS/m.

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1. Introduction

The understanding of the physicochemical controls on induced polarization (IP) measurements in non-metallic soils has increased considerably over the last decade as a result of the development of mechanistic models (Leroy et al., 2008; Revil and Florsch, 2010; Revil, 2012) and empirical studies based on extensive datasets (Weller et al., 2010b, 2011, 2013, 2015b). This parallel development of mechanistic models and empirical observation/model testing has confirmed the dependence of IP measurements on key physical properties, including the pore-volume normalized internal surface S_{por} and the closely related cation exchange capacity (CEC). However, these parallel research efforts have also led to some inconsistencies between the predictions of mechanistic models and empirical observations obtained for a wide range of unconsolidated sediments and consolidated sandstones.

In particular, the form of the mechanistic or empirical parameters describing the effect of the interfacial electrochemistry on the measured IP response has generated recent discussion (Revil, 2014; Weller et al., 2014). These parameters have been represented as a specific surface conductivity in mechanistic models (Leroy et al., 2008; Revil

and Florsch, 2010; Revil, 2012, 2013) or more generally as a specific polarizability in empirical relationships developed from observed relationships between pore space geometric parameters and IP measurements (Weller et al., 2011). Both mechanistic and empirical models recognize the need for a parameter defining the role of pore fluid chemistry and surface mineralogy on the interfacial electrochemistry (e.g., ion mobility and charge density in the electrical double layer (EDL)), and thus the IP response. Accurate understanding of this parameter is critical to reliable prediction of pore geometrical properties, and hence permeability, under conditions of variable groundwater chemistry (and possibly changing lithology or mineralogy).

In this study we use an extended database to compare empirical observations and previously proposed empirical models against recently proposed mechanistic formulations, focusing on the implications for the form of the term describing the control of the interfacial chemistry on polarization. When correctly defined, we argue that this term should be independent of any geometric properties of the pore space. We find that this term is independent of pore space geometry in empirical models fit to observations, but is not independent of pore space geometry in recent mechanistic models. We also demonstrate that, contradictory to recently proposed arguments made in mechanistic models, the magnitude of this pore geometry-independent term identified in empirical models is not strongly controlled by the mineralogy.

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2. Recent electrical models for induced polarization

2.1. Empirical models

The empirical approach to improving the understanding of the controls of pore geometry and pore fluid chemistry on induced polarization measurements has focused on compiling extensive databases where original data for both the IP measurements and the geometrical and chemical properties of interest are available (Weller and Slater, 2012; Weller et al., 2010b, 2011, 2013, 2015b). Numerous early studies based on specific sediment types and sandstone formations emphasized the critical role of S_{por} in controlling the magnitude of the IP response (Börner et al., 1996). Weller et al. (2010b), compiled a database of 114 samples of sandstone and unconsolidated sediments which were saturated with a NaCl solution with a fluid conductivity approximately equal to 100 mS/m, to derive a single empirical relationship between the imaginary conductivity σ'' (taken at 1 Hz) and S_{por} derived from nitrogen Brunauer, Emmett and Teller (BET) (Brunauer et al., 1938) measurements. This relationship takes the form,

$$\sigma'' = c_p S_{por}. \quad (1)$$

with a single value of the specific polarizability $c_p \approx 10^{-11}$ S fitting the entire 114 samples (coefficient of determination $R^2 = 0.84$). Weller et al. (2011) introduced the specific polarizability $c_p = \frac{\sigma''}{S_{por}}$ to represent the lumped effects of the interfacial chemistry of the surface on the IP response independent of the pore geometry. Weller et al. (2015b) showed additional strong evidence for the relationship given in Eq. (1) by investigating sands and sand–clay mixtures (again at a fluid conductivity close to 100 mS/m). They showed that the relationship given in Eq. (1) is even stronger when the methylene blue approach for determining S_{por} (Kahr and Madsen, 1995; Yukselen and Kaya, 2008) is used as the internal surface area of clays inaccessible to nitrogen BET measurements is included. This empirical formulation for the specific polarizability assumes that S_{por} alone is the appropriate geometric factor controlling σ'' i.e.,

$$c_p = \frac{\sigma''}{S_{por}}. \quad (2)$$

The electrochemical factors controlling c_p have also been studied, again using extensive databases of sandstone and unconsolidated sediment samples (Weller and Slater, 2012; Weller et al., 2011). This work has shown that the approximation, $c_p \propto (\sigma_w)^{0.5}$ describes the salinity dependence well up to some high salinity asymptote. This increase has been attributed to increases in surface charge density due to ion exchange with the pore fluid (Lesmes and Frye, 2001; Weller and Slater, 2012). At salinities higher than this asymptote, strong evidence exists for a reduction in c_p that has been attributed to the dominance of the competing effect of ionic mobility reduction with increasing salinity (Weller et al., 2015c). Empirical studies indicate that c_p is only weakly dependent on mineralogy (e.g. quartz versus clay) and that c_p is slightly lower for clean sands (Weller et al., 2010b, 2015b).

2.2. Mechanistic models

A mechanistic model to describe the IP dependence on pore geometry and interfacial chemistry has evolved in recent years (Leroy et al., 2008; Revil and Florsch, 2010; Revil and Skold, 2011; Revil, 2013, 2012; Skold et al., 2011). This model combines a macroscopic conductivity model for the polarization of a porous medium with a triple layer model that includes the specific surface conductivity of the Stern layer. The surface polarization is attributed to the ions in the Stern layer surrounding the mineral grains, although surface conduction is assumed to occur in both the diffuse and Stern layers. The model is

ambitious and potentially powerful as it attempts to provide a fully mechanistic explanation for the IP response of unconsolidated sediments and rocks.

Here, we omit a detailed explanation of the model as it is the subject of numerous papers. Instead, we focus in on the mechanistic equivalent of Eq. (1) that is predicted by this model. This mechanistic model is continually evolving and we therefore consider two recent formulations of the predicted relationship between the imaginary conductivity and the pore geometry found in recent papers. The first is the formulation found in the POLARIS model (Revil, 2012) and the second is the more recent model proposed by Revil (2013).

2.2.1. The POLARIS model

A mechanistic relationship between σ'' and S_{por} is developed in the POLARIS model starting from an expression for a direct relationship between σ'' and cation exchange capacity (Equation 43 in Revil, 2012),

$$\sigma'' \approx \frac{2}{3} \rho_s f \beta_{(+)}^S CEC, \quad (3)$$

where ρ_s is the mineral density, f is the partitioning coefficient describing the relative fraction of counterions in the Stern layer versus the total number of counterions in the EDL and $\beta_{(+)}^S$ is the mobility of ions in the Stern layer (which is assumed salinity independent for simplicity). We note that Equation (43) of Revil (2012) defines σ'' (and hence the phase of the complex conductivity) as negative; we define it as positive here for consistency with the more common convention of the IP exploration community where the phase of the complex resistivity is usually defined as negative. The approximation in Eq. (3) results from the assumption that the ratio $(F-1)/F$, where F is the electrical formation factor, is close to unity in most materials. Using the formulation for the charge density per unit surface area Q_s being equal to the ratio of the CEC to the mass normalized specific surface area S_m (in m^2/kg) of the material, $Q_s = CEC/S_m$, Revil (2012) identified a linear relationship between σ'' and S_m ,

$$\sigma'' \approx \frac{2}{3} \rho_s f \beta_{(+)}^S Q_s S_m, \quad (4)$$

suggesting that S_m is the only geometric parameter controlling the quadrature conductivity.

From the expression for the specific surface area per unit pore volume,

$$S_{por} = \rho_s \frac{1-\phi}{\phi} S_m, \quad (5)$$

the relationship,

$$\sigma'' \approx \frac{2}{3} f \beta_{(+)}^S Q_s \frac{\phi}{1-\phi} S_{por}, \quad (6)$$

can be formulated (Equation 48, in Revil (2012)). Adhering to the concept that the specific polarizability represents the lumped effects of the interfacial chemistry of the surface independent of the pore geometry (i.e. S_{por} and ϕ in Eq. (6)),

$$\sigma'' \approx c_{m1} \frac{\phi}{1-\phi} S_{por}, \quad (7)$$

where subscript m_1 differentiates this mechanistic definition from c_p defined above. In this study the equivalent specific polarizability from this

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