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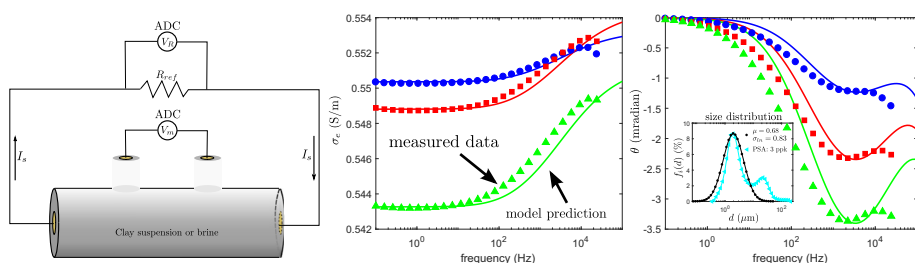
Low frequency complex dielectric (conductivity) response of dilute clay suspensions: Modeling and experiments



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GRAPHICAL ABSTRACT



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ABSTRACT

In this work, we establish an effective medium model to describe the low-frequency complex dielectric (conductivity) dispersion of dilute clay suspensions. We use previously obtained low-frequency polarization coefficients for a charged oblate spheroidal particle immersed in an electrolyte as the building block for the Maxwell Garnett mixing formula to model the dilute clay suspension. The complex conductivity phase dispersion exhibits a near-resonance peak when the clay grains have a narrow size distribution. The peak frequency is associated with the size distribution as well as the shape of clay grains and is often referred to as the characteristic frequency. In contrast, if the size of the clay grains has a broad distribution, the phase peak is broadened and can disappear into the background of the canonical phase response of the brine. To benchmark our model, the low-frequency dispersion of the complex conductivity of dilute clay suspensions is measured using a four-point impedance measurement, which can be reliably calibrated in the frequency range between 0.1 Hz and 10 kHz. By using a minimal number of fitting parameters when reliable information is available as input for the model and carefully examining the issue of potential over-fitting, we found that our model can be used to fit the measured dispersion of the complex conductivity with reasonable parameters. The good match between the modeled and experimental complex conductivity dispersion allows us to argue that our simplified model captures the essential physics for describing the low-frequency dispersion of the complex conductivity of dilute clay suspensions.

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1. Introduction

The unusually strong dispersion of the low-frequency complex permittivity, or, equivalently, complex conductivity, of clay sus-

pensions has been used to infer the electronic and microscopic structure of the surface of clay grains immersed in an electrolyte [1–3]. Because bare clay layers carry negative surface charge when exchangeable cations disassociate from the clay into the surrounding electrolyte, it is natural to invoke the electric double layer structure, in which there is a layer of diffuse, freely-moving positive ions surrounding the clay. Due to the surface chemistry of clay

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minerals, the Stern-Gouy double layer theory is often used to more realistically describe the ionic structure at and around the clay surface [4]. The Stern-Gouy theory includes the Stern layer where cations are bound to the clay surface, in addition to the cations in the diffuse layer. Various models, based on the Stern-Gouy double layer theory have been developed for explaining the dispersion of the complex conductivity. Lockhart had argued that only the cations in the diffuse layer contribute to the enhancement of the conductivity in the presence of clay, while the dielectric polarization or, effectively, the phase response of the complex conductivity occurs solely at the Stern layer [1,2]. The ionic polarization in the Stern layer is often referred to as Stern layer polarization. To match the dispersion of the conductivity, Révil had further assumed that cations in the Stern layer would not contribute to the real part of the conductivity at lower frequencies but can contribute to it at higher frequencies [5]. In contrast, Leroy and Révil have proposed a triple-layer model (a modification of the Stern-Gouy double layer theory) to describe the surface conductivity of the clay [6,7]. In their model, both the Stern layer and the diffuse layer will contribute to the surface conductivity while the polarization response at low frequencies is mainly dominated by the Stern layer polarization. Due to the existence of multiple and sometimes contradictory models, the interpretation of the unusually strong complex conductivity dispersion is often model-dependent and the real cause of it is still illusive.

To further complicate matters, strong low-frequency dispersion of the complex conductivity of clay suspensions can occur without the Stern layer polarization. By carefully continuing the potential from the electric double layer into the ionic neutral regime (or effectively using a proper boundary condition between the two regimes) in the presence of an applied oscillatory electric field, the frequency-dependent polarization coefficient for a charged sphere immersed in an electrolyte can be obtained theoretically in the thin double layer limit [8–12]. This is largely verified numerically [13–16]. Explicitly, Chew and Sen have shown that a strong dielectric enhancement, and, hence, the dispersion in the complex conductivity at low frequencies, is caused by an out-of-phase current in the ionic neutral region [9]. However, these previously obtained polarization coefficients for the charged sphere do not directly describe the dielectric response of charged platy grains. To address this issue, some of the authors of this paper have previously established analytic solutions for the low-frequency polarization coefficients which capture both the size and the shape effects of a charged oblate spheroidal grain immersed in an electrolyte [17]. Valuable solutions have also been established in Refs. [18,19], with their applicable regime constrained to near spherical particles. Those solutions show that a strong dielectric dispersion can be observed for dilute suspensions of charged oblate spheroids due to the out-of-phase current in the ionic neutral regime.

The main goal of the current study is to understand the mechanism that leads to the strong low-frequency complex dielectric (conductivity) response for dilute clay suspensions. Challenges to our efforts can come from many facets. Experimentally, measuring the complex conductivity dispersion over a wide frequency range is a challenging task because most instruments also have intrinsic frequency responses. To better extract the sample response, one must develop a scheme for calibrating the instrument, as illustrated in Appendix A. For sample preparation, because non-purified source clay often comes with a fraction of non-clay minerals, precise control of the clay fraction in the suspension becomes difficult. In addition, clay suspensions can form gels for swelling clays or settle too fast before reliable measurements can be performed. These can affect the measured signal and whether our simplified model can really describe the measured sample.

In the present work, we will use the polarization coefficients obtained in Ref. [17] to establish an effective medium model based

on the Maxwell Garnett mixing formula to describe the low-frequency complex dielectric (conductivity) dispersion of the dilute clay suspension. As mentioned above, there exist many possible competing mechanisms to explain the observed, strong dispersion of the low-frequency complex conductivity. Because all the models have different levels of phenomenological, adjustable parameters, to distinguish which model better captures the underlying physics of the measured signal is not a straightforward task. Hence, a model that can describe the observed phenomenon with the least non-measurable parameters can have great practical use.

Our model consists of six key parameters: the brine salinity, the clay cation exchange capacity (CEC), the volume fraction of clay, the effective aspect ratio of clay particles, and the mean, as well as the variance, of the log-normal function describing the particle size distribution of the clay. Besides the effective aspect ratio, the rest of the parameters are either well-constrained or measurable quantities. To further benchmark our model, we measure the low-frequency complex conductivity of clay suspensions with various brine salinities and clay volume fractions for two different clay minerals with contrasting CEC values: smectite and illite. We also measure particle-size distributions of the clay suspensions. The selection of our measurements provide a wide range in parameter space for us to test whether our model can describe the dielectric response of clay suspensions.

Our current study could eventually further our understanding of the low frequency electromagnetic measurements and response of rock formations for geological characterization and petroleum exploration. Clay minerals, which are ubiquitous in nature, often pose challenges for interpreting electromagnetic signals. For instance, the presence of clay minerals reduces the formation resistivity. As a result, the CEC or, equivalently, the excess cations per unit pore volume, Q_v , is required as an input for determining the water saturation from resistivity measurements [20,21]. In addition, a strong conductivity dispersion together with an unusually large quadrature conductivity is also observed for clay-bearing formations in the low frequency regime, $f < 10$ kHz. This phenomenon is often referred to as the spectral induced polarization (SIP) response for the measurement of geological materials [22–24]. In terms of the complex dielectric constant, the SIP effect implies an enhancement of the permittivity (the real part of the complex dielectric constant) at lower frequencies. Often, the permittivity of the clay-bearing formation can be on the order of 10^5 or higher at frequencies lower than 1 kHz, which is several orders of magnitude higher than that of the constituents of the formation [25]. More surprisingly, the presence of clay minerals affects the complex dielectric dispersion not only in the low frequency regime, but also in the microwave frequency range at $f > 10$ MHz, where the Stern layer polarization is expected to be negligible [23,26–28].

This manuscript is organized as follows: In Section 2, we first establish a model for describing the low-frequency dielectric dispersion of dilute clay suspensions based on the solutions obtained previously for the low-frequency polarization coefficients of the charged oblate spheroid and the Maxwell Garnett effective medium formula. To account for the particle size distribution of the clay grains, we will assume that the size of the charged oblate spheroids follows a log-normal distribution. In Section 3, we discuss some limitations and assumptions of our model. We set numerical values for some physical and material parameters required in our model. In Section 4, we illustrate the response of our simplified complex dielectric (conductivity) model for dilute clay suspensions to key parameters. In Section 5, we present the measurements of the complex conductivity of 18 dilute clay suspension samples along with the fits to the model established in Section 2. Some discussion on how well our simplified model

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