EXPRESS LETTER

On the use of the Cole–Cole equations in spectral induced polarization

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SUMMARY

Two different equations, both of which are often called 'the Cole–Cole equation', are widely used to fit experimental Spectral Induced Polarization data. The data are compared on the basis of fitting model parameters: the chargeability, the time constant and the exponent. The difference between the above two equations (the Cole–Cole equation proposed by the Cole brothers and Pelton's equation) is manifested in one of the fitting parameters, the time constant. The Cole–Cole time constant is an inverse of the peak angular frequency of the imaginary conductivity, while Pelton's time constant depends on the chargeability and exponent values. The difference between the time constant values corresponding to the above two equations grows with the increase of the chargeability value, and with the decrease of the Cole–Cole exponent value. This issue must be taken into consideration when comparing the experimental data sets for high polarizability media presented in terms of the Cole–Cole parameters.

Key words: Electrical properties; Electromagnetic theory; Hydrogeophysics.

1 INTRODUCTION

Over the last two decades, an interest to the use of the spectral induced polarization (SIP) method has been growing across the different geological applications: hydrogeology (e.g. Slater & Lesmes 2002), mineral exploration (e.g. Vanhala & Peltoniemi 1992) and biogeophysics (e.g. Abdel Aal *et al.* 2010). The SIP parameters are used to predict physical properties of soils and sediments (the hydraulic conductivity, see e.g. Binley *et al.* 2005; Zisser *et al.* 2010; the pore or grain size distribution, see e.g. Scott & Barker 2003; Revil & Florsch 2010) and the content and grain size of disseminated sulfide minerals (e.g. Zhdanov *et al.* 2012; Gurin *et al.* 2013).

Induced polarization (IP) is manifested by frequency-dependent behaviour of electrical conductivity. This phenomenon is also called the 'low-frequency dispersion' of the electrical conductivity (or its inverse, the electrical resistivity). So far, no universal physical–chemical model has been developed to describe the frequency dispersion; therefore, the most commonly used approach is to fit the experimental data on the basis of phenomenological models (e.g. Kemna 2000; Binley *et al.* 2005).

One of the commonly used phenomenological models is the Cole–Cole model (CCM), which was originally formulated for the complex dielectric constant (Cole & Cole 1941):

$$\varepsilon^* = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{1 + (\mathrm{i}\omega\tau)^{1-\alpha}}, \tag{1}$$

where ε_0 and ε_∞ are the low-frequency and the high-frequency dielectric constants values, respectively, τ is the central relaxation

time and $c = 1 - \alpha$ is a so-called CCM exponent, which describes the broadness of the relaxation time distribution.

Based on the electrostatic analogy, for dielectric materials with losses, it is easy to re-formulate eq. (1) for the complex electrical conductivity (by substituting the complex electrical conductivity for the complex dielectric constant):

$$\sigma^* = \sigma_\infty + \frac{\sigma_0 - \sigma_\infty}{1 + (\mathrm{i}\omega\tau)^c}, \tag{2}$$

where σ_0 and σ_∞ are the low-frequency and the high-frequency electrical conductivity values, respectively.

The complex electrical resistivity is often used in practical applications because it is directly calculated from the measured electrical impedance. The complex resistivity model, proposed by Pelton *et al.* (1978) (PM), can be obtained directly from the original CCM equation by substituting the complex electrical resistivity for the complex dielectric constant, which, strictly speaking, is not supported by physical considerations

$$\rho^* = \rho_\infty + \frac{\rho_0 - \rho_\infty}{1 + (\mathrm{i}\omega\tau)^c}, \tag{3}$$

where ρ_0 and ρ_∞ are the low-frequency and the high-frequency electrical resistivity values, respectively.

Both eqs (2) and (3) are typically used to approximate the experimental SIP data. Pelton *et al.* (1978) named their resistivity model the 'CCM'. However, considering the complex resistivity, ρ^*, as the inverse of the complex conductivity, σ^*, $\rho^* = 1/\sigma^*$ and rewriting the resistivity equation, eq. (3), for conductivity, one does not actually recover the initial CCM equation (see e.g. Binley *et al.* 2005;

Kruschwitz *et al.* 2010, where Pelton's model is presented in the conductivity form).

The CCM had been discussed exhaustively, and had been used in many papers to fit experimental data. Lesmes *et al.* (2000) presented physical–chemical interpretation of the IP parameters based on the CCM equation in its original form. Revil & Florsch (2010) also partly based their analysis of the IP phenomenon on the CCM equation. Conversely, Kemna (2000), Binley *et al.* (2005) and Slater *et al.* (2006) used Pelton's formulation. Kruschwitz *et al.* (2010) compared the different data sets on the basis of the Cole–Cole parameters retrieved by fitting the experimental data with the generalized Pelton's equation. One can expect that, in the near future, the data obtained by the various research groups will be compared on the basis of the Cole–Cole parameters. However, the use of the different equations can lead to inconsistence of the compared parameters. In this context, it is very important to understand the difference between the two formulations (the original equation by the Cole brothers and Pelton's equation). This difference has been recently highlighted by Florsch *et al.* (2012), although it has not been the focal point of their paper.

In this paper, we establish a link between the parameters obtained on the basis of the two formulations: the original formulation, CCM (Cole & Cole 1941) and the PM equation (Pelton *et al.* 1978). In the second section, we briefly present the theoretical background. In the third section, we compare CCM and PM analytically. In the fourth section, we present an experimental example of the so-called CCM fitting procedure that is often used to describe the experimental spectra by the model parameters (chargeability, time constant and exponent) and we show a significant difference in the relaxation time values obtained with the use of CCM and PM.

2 THEORETICAL BACKGROUND

Application of electrical filed to a medium causes displacements of the bound charges and directional movements of the free charges. Displacements of the bound charges are characterized by the dielectric constant, ε, while the directional movements are characterized by the electrical conductivity, σ. The electrical displacement, **D**, is related to the electrical field, **E** and to the polarization, **P**

$$\mathbf{D} = \varepsilon_a \mathbf{E} + \mathbf{P}, \tag{4}$$

where $\varepsilon_a = 8.854 \times 10^{-12}$ F m^{-1} is the vacuum permittivity.

The polarization of material, **P**, is defined as the induced dipole moment per unit volume. The polarization is assumed to be proportionate to the electrical field,

$$\mathbf{P} = \varepsilon_a \chi \mathbf{E}, \tag{5}$$

where χ is the dimensionless dielectric susceptibility. Considering eq. (5), eq. (4) can be rewritten as

$$\mathbf{D} = \varepsilon_a (1 + \chi) \mathbf{E} = \varepsilon_a \varepsilon \mathbf{E}, \tag{6}$$

where $\varepsilon = (1 + \chi)$ is the relative dimensionless dielectric constant of the material.

The total current density, **J**, resulting from an alternating electrical field (whose time dependency is $\mathbf{E} = \mathbf{E_0} \exp(i\omega t)$, with the angular frequency $\omega = 2\pi f$) is the sum of the conduction current density, $\mathbf{J}_c$, and the displacement current density, $\mathbf{J}_d$,

$$\mathbf{J} = \mathbf{J_c} + \mathbf{J_d}, \tag{7}$$

where $\mathbf{J}_c = \sigma \mathbf{E}$ according to Ohm's Law, and $\mathbf{J}_d = \dfrac{\partial \mathbf{D}}{\partial t} = i\omega \varepsilon_a \varepsilon \mathbf{E}$ according to the first Maxwell equation.

Hence, the total current density can be expressed in the form of Ohm's Law, considering the electrical conductivity as a complex value

$$\mathbf{J} = \sigma \cdot \mathbf{E} + i\omega \varepsilon \varepsilon_a \mathbf{E} = \sigma^* \mathbf{E}, \tag{8}$$

where $\sigma^* = \sigma + i\omega \varepsilon_a \varepsilon$ is the effective complex electrical conductivity, which fully describes the electrical behaviour of a medium with electrical conductivity and polarization (an imperfect conductor). Alternatively and equivalently, the electrical behaviour of such a medium can be expressed through the complex dielectric constant:

$$\mathbf{D} = \varepsilon \varepsilon_a \mathbf{E} + (\sigma/i\omega)\mathbf{E} = (\varepsilon_a \varepsilon + \sigma/i\omega)\mathbf{E} = \varepsilon^* \varepsilon_a \mathbf{E}, \tag{9}$$

where $\varepsilon^* = \varepsilon + \sigma/i\omega \varepsilon_a$ is the complex dielectric constant, which also fully describes the behaviour of a dielectric with losses. Therefore, the electrical behaviour of a medium can be expressed through one of three complex parameters: the dielectric constant, the electrical conductivity or the resistivity

$$\sigma^*(\omega) = \frac{1}{\rho^*(\omega)} = i\omega \varepsilon_a \varepsilon^*(\omega). \tag{10}$$

Which parameter to be used to describe the electrical behaviour of the studied media is a matter of tradition and of experiment design. In geophysics, data are often presented in the form of conductivity or resistivity, and in the colloidal sciences the dielectric constant is traditionally used.

3 THE COLE–COLE EQUATION AND PELTON'S EQUATION

As an extension of the Debye equation (Debye 1929), the CCM equation converges to Debye's, when $c = 1$. The Debye model describes the orientation polarization of polar molecules in continuous media with macroscopic viscosity. In the Debye model, a single value of the relaxation time is assumed. In contrast, CCM uses a distribution of the relaxation time values; the broadness of that distribution is defined by the exponent, c. The exponent value varies between 0 and 1. For rocks, soils and disseminated ores, its typical values range from 0.1 to 0.6 (e.g. Kemna 2000). When c decreases, the relaxation time distribution becomes broader, the transition between the low- and the high-frequency values becomes wider, and the peak on the imaginary part of the dielectric constant or electrical conductivity also becomes wider. Therefore, the CCM can be viewed as a superposition of multiple Debye models (e.g. Lesmes *et al.* 2000). The CCM central relaxation time, τ, is an inverse of the peak position on the imaginary part of dielectric constant versus frequency (the critical frequency).
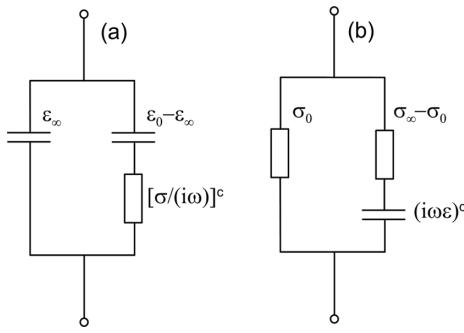
Considering the CCM equation as a result of superposition of multiple Debye relaxations with a probability distribution, $F(\tau)$, one obtains (Cole & Cole 1941; Lesmes *et al.* 2000):

$$\varepsilon^* = \varepsilon_\infty + \int_0^\infty \frac{F(\tau)}{1 + i\omega\tau} d\tau, \tag{11}$$

where

$$F(x) = \frac{1}{2\pi} \frac{\sin(\alpha\pi)}{\cosh[(1 - \alpha) \cdot x] - \cos(\alpha\pi)}, \tag{12}$$

$x = \ln(\tau/\tau_0)$ and τ_0 is the geometric mean of the distribution, eq. (12).

Figure 1. Equivalent electrical circuits for the Cole–Cole equation in the dielectric constant (a) and conductivity (b) formulations.

The phenomenological models are often interpreted in terms of equivalent electrical circuits. As was shown in the original paper by the Cole brothers (1941), the dielectric CCM (eq. 1) corresponds to the equivalent circuit with two capacitances and one conductance (Fig. 1a). The capacitances model the polarization, and the conductance models the dielectric losses. Two serial elements are connected with the third element in parallel. With this circuit, the relaxation time is given by

$$\tau = (\varepsilon_0 - \varepsilon_\infty)/\sigma^{1/c}. \tag{13}$$

In the circuit (Fig. 1a), substituting two conductances for two capacitances, and one capacitance for one conductance, one obtains the equivalent circuit (Fig. 1b) for the conductivity CCM, (eq. 2). Similarly, the relaxation time for the conductivity CCM is

$$\tau = \varepsilon/(\sigma_\infty - \sigma_0)^{1/c}. \tag{14}$$

The equivalent circuit (Fig. 1b) corresponds, for example, to two pores (polarized and non-polarized) connected in parallel. When $c = 1$, eq. (2) also describes polarization of a single spherical grain surrounded by a surface layer with frequency-dependent surface conduction (Schwarz 1962). The grain gives the Debye response with the relaxation time

$$\tau = \frac{r^2}{2D}, \tag{15}$$

where r is the grain radius and D is the surface diffusion coefficient. As was shown by Revil & Florsch (2010), assuming a log normal distribution of the grain radii and using eq. (15), one obtains the relaxation time distribution that is close to the one in CCM, eq. (12). Consequently, CCM can be used to approximate the electrical conductivity dispersion associated with a log normal distribution of grain sizes.

Eq. (2) can be expressed using the high-frequency or the low-frequency electrical conductivity and the polarization magnitude (chargeability, defined by Seigel 1959)

$$\sigma_{CC}^* = \sigma_\infty \left(1 - \frac{m}{1 + (i\omega\tau)^c} \right)$$

$$= \sigma_0 \left(1 + \frac{m}{1-m} \cdot \left(1 - \frac{1}{1 + (i\omega\tau)^c} \right) \right), \tag{16}$$

where $m = \frac{\sigma_\infty - \sigma_0}{\sigma_\infty}$ is the chargeability, the '*CC*' index denotes the CCM.

We defined the critical frequency as the peak frequency of the imaginary conductivity. However, it is also often defined as the peak frequency of the conductivity (or resistivity) phase distribution (e.g. Kruschwitz *et al.* 2010),

$$\varphi = \arctan \left(\frac{\sigma''}{\sigma'} \right). \tag{17}$$
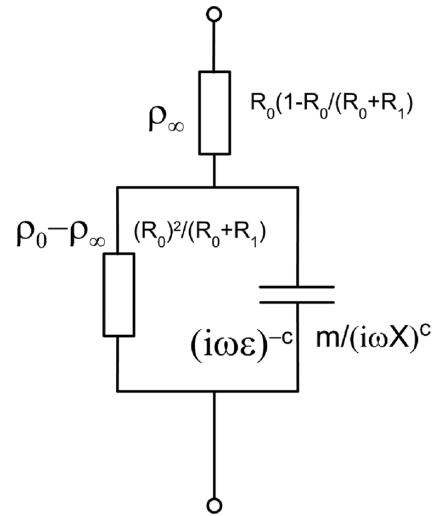
Figure 2. Equivalent electrical circuits for Pelton's equation in terms of resistance and resistivity.

By differentiating eq. (17), and considering eq. (16), one can obtain, after simple algebraic operations, a relationship between the phase peak position and the relaxation time

$$\omega_{\varphi CC}^{\text{peak}} = \frac{1}{\tau}(1-m)^{\frac{1}{2c}}. \tag{18}$$

Eq. (18) shows that the phase peak shifts towards lower frequencies relative to the imaginary conductivity peak with the increase of chargeability and with the decrease of the exponent.

In geophysical applications, the frequency-depended electrical impedance is usually obtained. Pelton *et al.* (1978) proposed the following original equation for impedance:

$$Z(\omega) = R_0 \left[1 - m \left(1 - \frac{1}{1 + (i\omega\tau)^c} \right) \right], \tag{19}$$

where R_0 is the DC (low frequency) resistance,

$m = \dfrac{R_0}{R_0 + R_1}$ is the chargeability,

$\tau = X \left(\dfrac{R_0}{m} \right)^{1/c} = X(R_0 + R_1)^{1/c}$,

$\frac{R_0 R_1}{R_0 + R_1}$ is the high-frequency limit of the resistance and X is the effective capacitance.

Pelton *et al.* (1978) believed that eq. (19) described the equivalent circuit for the CCM (Fig. 1b), where R_0 corresponded to $1/\sigma_0$, R_1 corresponded to $1/(\sigma_\infty - \sigma_0)$, m corresponded to Seigel's (1959) chargeability and $(i\omega\varepsilon)^{-c}$ corresponded to $(i\omega X)^{-c}$ (their Fig. 1). However, simple analysis shows that this is not true. Eq. (19) corresponds to another circuit, shown in Fig. 2. This circuit illustrates that PM can be used to describe the impedance of two serially connected pores (a 'passive' non-polarized pore and an 'active' polarized pore) (see, e.g. Marshall & Madden 1959).

Pelton's equation can be expressed in terms of the complex resistivity. Using Seigel's (1959) definition of the chargeability, based on the resistivity, $m = (\rho_0 - \rho_\infty)/\rho_0$, and substituting these resistivities for the resistances in eq. (19), one obtains (e.g. Kemna 2000)

$$\rho_P^* = \rho_\infty + \frac{\rho_0 - \rho_\infty}{1 + (i\omega\tau)^c} = \rho_\infty \cdot \left[1 + \frac{m}{1-m} \left(\frac{1}{1 + (i\omega\tau)^c} \right) \right]$$

$$= \rho_0 \cdot \left[1 - m \left(1 - \frac{1}{1 + (i\omega\tau)^c} \right) \right], \tag{20}$$

where ρ_0, ρ_∞ are the low-frequency resistivity limit and the high-frequency resistivity limit, respectively, index P denotes Pelton's model. The relaxation time of the PM equation is

$$\tau = \varepsilon \cdot (\rho_0 - \rho_\infty)^{1/c}. \tag{21}$$

The phase peak of the complex resistivity is also shifted relative to the peak of the imaginary part of the resistivity. However, contrary to the case of conductivity, it shifts towards the higher frequencies with increase of m and with decrease of c (Major & Silic 1981)

$$\omega_{\varphi P}^{\text{peak}} = \frac{1}{\tau} \cdot \frac{1}{(1-m)^{1/2c}}. \tag{22}$$

Obviously, eq. (20) is not identical to eq. (16). Pelton's equation is often presented in terms of electrical conductivity (e.g. Binley *et al.* 2005; Slater *et al.* 2006; Kruschwitz *et al.* 2010). Algebraic operations show that the following forms of Pelton's equation, written with the use of the low-frequency or high-frequency limits of the conductivity, are equivalent

$$\sigma_p^* = \sigma_\infty \cdot \left[1 - \frac{m}{1 + (i\omega\tau)^c \cdot (1-m)} \right]$$

$$= \sigma_0 \cdot \left[1 + m \cdot \left(\frac{(i\omega\tau)^c}{1 + (i\omega\tau)^c \cdot (1-m)} \right) \right]$$

$$= \sigma_0 \left[1 + \frac{m}{1-m} \left(1 - \frac{1}{1 + (i\omega\tau)^c \cdot (1-m)} \right) \right]. \tag{23}$$

Direct comparison of eq. (16) and the right-hand side of eq. (23) shows that they are formally identical if

$$\tau_{CC} = \tau_P (1-m)^{1/c} \tag{24}$$

(see Florsch *et al.* 2012).

Therefore, the PM equation converges to the CCM equation when the chargeability approaches zero.

4 DIFFERENCE IN RELAXATION TIME BETWEEN THE COLE–COLE AND PELTON'S EQUATIONS

In the CCM equation, the relaxation time is directly related to the critical frequency, which we defined as the peak position on the imaginary part of the complex conductivity

$$\omega^{\text{peak}} = \frac{1}{\tau_{CC}}. \tag{25}$$

However, eq. (24) shows that this is not true for the PM equation. Differentiating the imaginary part of eq. (23) by frequency, and equating the result to zero, one obtains the critical frequency for the PM equation

$$\omega^{\text{peak}} = \frac{1}{\tau_P} \cdot \frac{1}{(1-m)^{1/c}}. \tag{26}$$

As follows from eq. (26), contrary to CCM, the relaxation time in PM depends not only on the critical frequency, but also on the chargeability and the exponent value. The higher the chargeability, and the lower the exponent, the more the relaxation time τ_p increases as compared to the value of $\tau_{CC} = 1/\omega^{\text{peak}}$.

Eq. (24) enables us to easily obtain the numerical difference between the relaxation time values according to CCM and PM (Fig. 3). At low chargeability values ($m < 0.1$), typical of common soils (e.g. Zisser *et al.* 2010), the τ_{CC}/τ_P ratio varies between 0.6 and 1 depending on the exponent value. This means that the PM application leads to relatively small systematic overestimation of the relaxation time value as compared to that obtained with CCM.
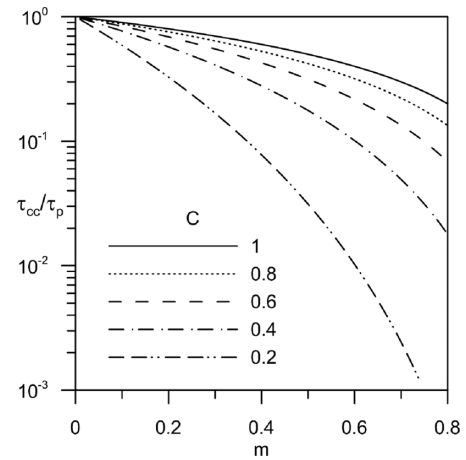

Figure 3. The ratio of the Cole–Cole relaxation time and Pelton's relaxation time versus chargeability for the different exponent values.

At higher chargeability values, the differences between τ_{CC} and τ_p become more noticeable. High chargeability values are typical for the media containing metallic particles (e.g. Slater *et al.* 2006). In these cases, it becomes critical to indicate which model (CCM or PM) was used to fit the experimental data. We recommend using eq. (24) to compare the results of the Cole–Cole fitting when different equations (CCM and PM) are used.

Theoretical models link the relaxation time to permeability. For example, Revil *et al.* (2012) derived an equation

$$k = \frac{\tau_{CC} D_i}{4F}, \tag{27}$$

where k is the permeability, D_i is the ion diffusion coefficient and F is the formation factor. They based eq. (27) on CCM. Let us assume the relaxation time is obtained on the basis of PM. For typical chargeability values ($m < 0.2$) and the Cole–Cole exponent in the range of 0.2–1, the minimum difference between the CCM and PM relaxation time values is 3 times (see Fig. 3). This results in overestimation of the permeability value by the factor of up to 3, which is not very significant, considering its typically log normal distribution.

Nordsiek & Weller (2008) based their Debye decomposition approach on the superposition of Pelton's equations with the exponent c equal to 1, the relaxation time values equally distributed in logarithmic scale, and with the magnitude values named 'specific chargeability' (their eq. 2). In principle, this also can be done on the basis of CCM. The specific chargeability values are typically low (< 0.1) for both the ion-conducting media (Tarasov & Titov 2007) and the media containing metallic particles (Nordsiek & Weller 2008; Gurin *et al.* 2013). According to eq. (24) and Fig. 3, for these values of the specific polarizability and with $c = 1$, the relaxation time values obtained with PM and CCM are close to each other. At higher specific chargeability values, one can see how the peak of relaxation time distribution shifts depending on the used model in Fig. 3 (the graph corresponding to $c = 1$). Our numerical experiments (not shown here) reveal that for the total chargeability, $m < 0.5$, the peak position shifts by a factor of less than 2.

In the next section, we use experimental data to illustrate the difference between the relaxation time values derived from CCM and PM.

5 COLE–COLE RELAXATION TIME FROM EXPERIMENTAL DATA

We use the data from Slater *et al.* (2006), where IP spectra were measured on a synthetic sample obtained by mixing Ottawa sand with
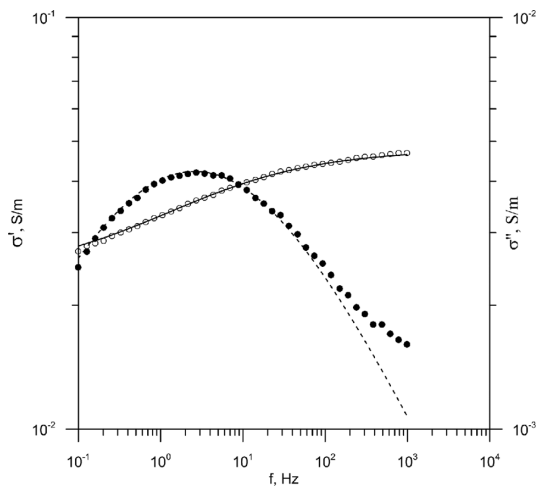
Figure 4. Real and imaginary parts of the electrical conductivity versus frequency obtained on a mixture of Ottawa sand and 10 per cent iron fillings (data from Slater *et al.* (2006)). Open symbols are the real conductivity, and solid symbols are the imaginary conductivity. The data fits for PM and CCM are shown by the solid (magnitude) and dashed (phase) lines. Three model parameters are the same for Pelton's and the Cole–Cole models ($\sigma_0 = 0.0271$ S m^{-1}, $m = 0.51$; $c = 0.424$) but the relaxation time values are very different ($\tau_P = 0.33$ s, and $\tau_{CC} = 0.061$ s).

10 per cent iron filings (Fig. 4). To fit these data with CCM and PM, we applied an algorithm based on the least-squares approach with Marquardt regularization, which was proposed by Kemna (2000). Three model parameters (the low-frequency conductivity, the chargeability and the exponent) were the same for CCM and PM, and corresponded to the parameters determined by Slater *et al.* (2006). However, the relaxation time values for the two formulations showed significant difference ($\tau_p = 0.33$ s, and $\tau_{CC} = 0.061$ s) according to eq. (23).

6 CONCLUSIONS

In this paper, we show that two formulations, both called the 'CCM' in geophysics (CCM and PM) are formally and computationally different. In terms of equivalent circuits, these formulations are based on serial and parallel connections of polarized and non-polarized elements. These models converge at low chargeability values.

The Cole–Cole fitting procedure produces three equal model parameters (DC conductivity, chargeability and exponent) regardless of the used formulation. However, the relaxation time value depends on the applied model. Pelton's relaxation time differs from the Cole–Cole relaxation time by the factor of $(1 - m)^{1/c}$. CCM is easier to use in practice because the inverse of the critical angular frequency of the imaginary conductivity is equal to the relaxation time, regardless of the chargeability and exponent values.

When experimental data are compared in terms of the Cole–Cole parameters, it is important to use the same model, especially for the cases where the chargeability values are high. Eq. (24) presents the link between the two models, and can be used to convert the relaxation time values from one model to the other.

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