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Anomalous-diffusion approach applied to the electrical response of water

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We investigate the electrical response of Milli-Q deionized water by using a fractional diffusion equation of distributed order with the interfaces (i.e., the boundary conditions at the electrodes limiting the sample) governed by integrodifferential equations. We also consider that the positive and negative ions have the same mobility and that the electric potential profile across the sample satisfies Poisson's equation. In addition, the good agreement between the experimental data and this approach evidences the presence of anomalous diffusion due to the surface effects in this system.

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I. INTRODUCTION

One of the most important phenomena in physics is the diffusion which permeates several fields of science. In fact, it can be found, for instance, in atom deposition into a porous substrate [1], diffusion of high molecular weight polyisopropylacrylamide in nanopores [2], highly confined hard disk fluid mixtures [3], fluctuating particle fluxes [4], diffusion on fractals [5,6], ferrofluid [7], p-doped poly(3,4-ethylenedioxythiophene) modified electrodes [8], colloids [9], and many others. Depending on the stochastic process connected to the diffusion we are led to either a convectional [10] or an anomalous diffusion [11–13]. The first scenario is characterized by a time-linear dependence for the mean square displacement [i.e., $\langle (z-\langle z\rangle)^2\rangle \sim t$], which is typical of a Markovian process and the dynamic of the system may be connected to a diffusion equation or Langevin equation with white noise [10]. On the other hand, systems with, for example, memory [12,13], fractality [5,6], and long-range interactions [14–16] are characterized by a different time dependence for the mean square displacement [e.g., $\langle (z-\langle z\rangle)^2\rangle \sim t^{\vartheta}$], which is evidence of the non-Markovian nature of the processes present in these systems. Surface effects on the system may also lead us to scenarios characterized by anomalous diffusion as, for example, the situations reported in Refs. [17,18] where the effect of the adsorption-desorption process of the surface produces an anomalous spreading of the system depending on the time scale considered. These effects play an important role in the electrical response of a system and lead us to an anomalous behavior which is not suitably described by the usual models. In fact, the anomalous electric response can be found in several systems such as fractal electrodes [19], nanostructured iridium oxide [20], and water [21]. To face these anomalous responses, several approaches have been proposed such as the extension of the Warburg model [22,23] and fractional diffusion equations [24–27] by accomplishing the Poisson equation [28–30]. Other developments can be found in Refs. [31,32].

The plan of this work is to investigate the electrical response of Milli-Q deionized water (i.e., ultrapure water of Type 1 according to the standards) by using a fractional diffusion equation of distributed order with the interfaces (i.e., the boundary conditions at the electrodes limiting the sample, governed by integrodifferential equations). This phenomenological model is compared with experimental data and the very good agreement suggests the presence of anomalous diffusion in the electrolytic cell. Indeed, this feature provides us with a relation between the electrical response of the system and the influence of the dynamics aspects of the ions motion (i.e., the diffusive processes). These developments and the comparison between the theoretical predictions and experimental data are performed in Secs. II and III. The last section (Sec. IV) is devoted to the discussions and the conclusion.

II. ANOMALOUS DIFFUSION AND ELECTRICAL RESPONSE

Let us describe the phenomenological model used to investigate the experimental data present in the next section. Following the formalism developed in Ref. [30], we start by considering the fractional diffusion equation of distributed order [33] for the bulk density of ions n_α ($\alpha = +$ for positive and $\alpha = -$ for negative ones)

$$\int_0^1 d\overline{\gamma}\;\overline{\tau}(\overline{\gamma})\frac{\partial^{\overline{\gamma}}}{\partial t^{\overline{\gamma}}}n_\alpha(z,t) = -\frac{\partial}{\partial z}j_\alpha(z,t), \tag{1}$$

where $\overline{\tau}(\overline{\gamma})$ is a distribution function of $\overline{\gamma}$ with the current density given by

$$j_\alpha(z,t) = -\mathcal{D}\frac{\partial}{\partial z}n_\alpha(z,t) \mp \frac{q\mathcal{D}}{k_B T}n_\alpha(z,t)\frac{\partial V}{\partial z}. \tag{2}$$

In Eq. (2), $\mathcal{D}$ is the diffusion coefficient for the mobile ions (the same for the positive and negative ones) of charge q, V is the actual electric potential across a sample of thickness d with the electrodes placed at the positions $z = \pm d/2$ of a Cartesian reference frame in which z is the axis normal to them, k_B is the Boltzmann constant, and T the absolute temperature. The fractional operator considered here is Caputo's one, that is,

$$\frac{\partial^\gamma}{\partial t^\gamma}n_\alpha(z,t) = \frac{1}{\Gamma(k-\gamma)}\int_{t_0}^{t} dt'\frac{n_\alpha^{(k)}(z,t)}{(t-t')^{\gamma-n+1}}, \tag{3}$$

with $k-1<\gamma<k$ and $n_\alpha^{(k)}(z,t) \equiv \partial_t^k n_\alpha(z,t)$. In particular, we consider $t_0 \to -\infty$ to analyze the response of the system to the periodic applied potential defined later on, as indicated by Ref. [34]. Equation (1) can be related to a rich class of diffusive processes depending on the choice performed to the distribution $\overline{\tau}(\overline{\gamma})$, in particular the non-Markovian processes manifested by the system which lead us to an anomalous spreading of the system. Simple cases can be

recovered by means of suitable choices for $\overline{\tau}(\overline{\gamma})$, such as $\overline{\tau}(\overline{\gamma}) = \tau\delta(\overline{\gamma} - 1)$, which corresponds to the normal diffusive process, and $\overline{\tau}(\overline{\gamma}) = \tau_{\gamma}^{\gamma}\delta(\overline{\gamma} - \gamma)$, $0 < \gamma \leqslant 1$, related to a pure fractional diffusion equation of order γ. Of special interest may be the choices $\overline{\tau}(\overline{\gamma}) = \tau\delta(\overline{\gamma} - 1) + \tau_{\gamma}^{\gamma}\delta(\overline{\gamma} - \gamma)$ and $\overline{\tau}(\overline{\gamma}) = 1$ and the mixing between these cases, which correspond to a situation with two or more different diffusive regimes [33,35]; these may be related to the situations discussed in Ref. [32] for the motion of the ions. In this manner, we expect to produce two different behaviors for the impedance, one of them governing the low frequency regime and the other manifested in the high frequency limit, as, for example, the regimes presented in Figs. 1 and 3 for the real part of the impedance which have a different behavior from the usual situation [36]. This feature could be verified by analyzing the mean square displacement which in this case is expected to have two different behaviors, one for small times and the other for large time related to the dynamic aspect of the ions. In this sense, as pointed out in Ref. [32] this quantity may be useful to establish a bridge between the experimental data and the theoretical model. To cover the influence of the surface on the ions in a more general situation, we consider Eq. (1) subject to the boundary conditions

$$j_{\alpha}(z,t)\big|_{z=\pm\frac{d}{2}} = \pm\int_0^1 d\vartheta\, \widetilde{\tau}(\vartheta) \times \int_{-\infty}^{t} dv\overline{\kappa}(t-v)\frac{\partial^{\vartheta}}{\partial v^{\vartheta}} n_{\alpha}(z,v)\bigg|_{z=\pm\frac{d}{2}}. \tag{4}$$

This boundary condition may be related to an adsorption-desorption process at the surfaces with an unusual dynamics which, for the particular choice $\overline{\kappa}(t) = \kappa e^{-t/\tau}$ with $\widetilde{\tau}(\vartheta) = \delta(\vartheta - 1)$, recovers the adsorption-desorption processes at the surfaces governed by a kinetic equation corresponding to the Langmuir approximation [36]. Others choices of $\overline{\kappa}(t)$ and $\widetilde{\tau}(\vartheta)$ can be performed to incorporate memory effects such as the situations worked out in Ref. [30] for $\widetilde{\tau}(\vartheta) = \delta(\vartheta - 1)$ and, consequently, anomalous relaxation processes [18] and the mixing between the usual and anomalous relaxation processes. In this sense, it is also interesting to note that, for $\overline{\kappa}(t) \propto \delta(t)$ with $\widetilde{\tau}(\vartheta)$ arbitrary, we can relate the processes at the surface with fractional kinetic equations [37,38]. From this point of view, Eq. (4) may interpolate several contexts which play an important role in the description of the electrical response of the system and, consequently, may be useful to investigate the behavior of the impedance in the low frequency limit where the nonusual behavior is manifested. In fact, the changes produced by incorporating the fractional time derivative of distributed order are useful to investigate the real part of the impedance and the boundary conditions play an important role for the imaginary part of the impedance. The potential is determined by Poisson's equation

$$\frac{\partial^2}{\partial z^2}V(z,t) = -\frac{q}{\varepsilon}\left[n_{+}(z,t) - n_{-}(z,t)\right], \tag{5}$$

which depends on the difference between the densities of charged particles. A solution for the previous equations and, consequently, an expression for the electrical impedance may be found in the linear approximation by considering that $n_{\alpha}(z,t) = \mathcal{N} + \delta n_{\alpha}(z,t)$, with $\mathcal{N} \gg \delta n_{\alpha}(z,t)$ where $\mathcal{N}$ represents the number of ions. In addition, we also consider $\delta n_{\alpha}(z,t) = \eta_{\alpha}(z)e^{i\omega t}$ to analyze the impedance when the electrolytic cell is subjected to the time-dependent potential $V(z,t) = \phi(z)e^{i\omega t}$, with $V(\pm d/2,t) = \pm V_0 e^{i\omega t}/2$. After substituting these quantities in Eqs. (1), (4), and (5), we obtain a set of four coupled equations which may be decoupled by introducing the functions $\psi_{+}(z) = \eta_{+}(z) + \eta_{-}(z)$ and $\psi_{-}(z) = \eta_{+}(z) - \eta_{-}(z)$. The first two equations are

$$\frac{d^2}{dz^2}\psi_{\pm}(z) = \alpha_{\pm}^2\psi_{\pm}(z), \tag{6}$$

where $\alpha_{-}^2 = \Lambda(i\omega)/\mathcal{D} + 1/\lambda^2$ and $\alpha_{+}^2 = \Lambda(i\omega)/\mathcal{D}$, in which $\Lambda(i\omega) = \int_0^{\infty} d\overline{\gamma}\overline{\tau}(\overline{\gamma})(i\omega)^{\overline{\gamma}}$ and $\lambda = \sqrt{\varepsilon k_B T/(2\mathcal{N}q^2)}$ is the Debye's screening length. The other two equations are

$$\left(\mathcal{D}\frac{d}{dz}\psi_{-}(z) + \frac{2q\mathcal{N}}{k_B T}\mathcal{D}\frac{d}{dz}\phi(z)\right)\bigg|_{z=\pm\frac{d}{2}} = \mp\Phi(i\omega)\psi_{-}(z)\big|_{z=\pm\frac{d}{2}}, \tag{7}$$

$$\mathcal{D}\frac{d}{dz}\psi_{+}(z)\bigg|_{z=\pm\frac{d}{2}} = \mp\Phi(i\omega)\psi_{+}(z)\big|_{z=\pm\frac{d}{2}}, \tag{8}$$

at $z = \pm d/2$, with $\Phi(i\omega) = e^{-i\omega t}\int_0^1 d\vartheta\widetilde{\tau}(\vartheta)(i\omega)^{\vartheta} \int_{-\infty}^{t} d\overline{t}\,\overline{\kappa}(t-\overline{t})e^{i\omega\overline{t}}$. The solution of Eq. (6) is $\psi_{\pm}(z) = \mathcal{C}_{\pm,1}e^{\alpha_{\pm}z} + \mathcal{C}_{\pm,2}e^{-\alpha_{\pm}z}$, where $\mathcal{C}_{\pm,1}$ and $\mathcal{C}_{\pm,2}$ are determined by the boundary conditions and the symmetry of the potential: $V(z,t) = -V(-z,t)$, which implies $\mathcal{C}_{-,1} = -\mathcal{C}_{-,2}$ and, consequently,

$$\psi_{-}(z) = 2\mathcal{C}_{-,1}\sinh(\alpha_{-}z), \tag{9}$$

$$\phi(z) = -\frac{2q}{\varepsilon\alpha^2}\mathcal{C}_{-,1}\sinh(\alpha_{-}z) + \overline{\mathcal{C}}z. \tag{10}$$

The constants $\mathcal{C}_{-,1}$ and $\overline{\mathcal{C}}$ are determined by using the boundary condition for $\psi_{-}(z)$ [i.e., Eq. (7)] and the condition imposed on the potential, for example, in $z = d/2$. Using these results and the procedure presented in Ref. [36], we can obtain the current and the impedance, $\mathcal{Z} = V/I$, of the electrolytic cell. In particular, for the case discussed above, it is given by

$$\mathcal{Z} = \frac{2}{i\omega\varepsilon\mathcal{S}\alpha_{-}^2} \times \frac{\tanh(\alpha_{-}d/2)/(\lambda^2\alpha_{-}) + \mathcal{E}d/(2\mathcal{D})}{1 + \Phi(i\omega)(1 + i\omega\lambda^2/\mathcal{D})\tanh(\alpha_{-}d/2)/(\alpha_{-}i\omega\lambda^2)} \tag{11}$$

with $\mathcal{E} = \Lambda(i\omega) + \alpha_{-}\Phi(i\omega)\tanh(\alpha_{-}d/2)$ and $\mathcal{S}$ is the electrode area.

III. THEORETICAL AND EXPERIMENTAL RESULTS

Now, we compare the experimental data obtained for the electrical response of Milli-Q deionized water with the impedance spectroscopy technique and the model discussed in the previous section. The measurements of the real and imaginary parts of the electrical impedance were performed using a Solartron SI 1296 A impedance-gain phase analyzer. The frequency range used was from 10^{-2} Hz to 5 MHz. The amplitude of the ac applied voltage was 20 mV. The Milli-Q water was placed between two circular surfaces spaced 1.0 mm from each other. The area of electrical electrodes was

3.14 cm^2. We used two kinds of electrical contacts. One of them was of brass and other one made of stainless steel. The Milli-Q water we used has resistivity $\rho = 18.2\ M\Omega$ cm and $pH = 6.1$. We did not add a concentrated potassium chloride solution as a buffer in the pure Milli-Q water. Before starting the measures we adopted the following procedure for cleaning. First, the electrodes were washed with detergent and deionized water. After that, the electrodes were polished with fine sandpaper. Finally, the electrodes were placed in an ultrasonic bath for 10 min. After this procedure, we put the Milli-Q water between the electrodes (1.0 mm thickness). All the measurements of the real $R = \text{Re}\,(\mathcal{Z})$ and imaginary $X = \text{Im}\,(\mathcal{Z})$ parts of the electrical impedance were made at room temperature. It is important to mention that the electrical response obtained in this experiment has also the contribution of the device, which is very small when compared with the electrical response of the water. Figure 1 shows the experimental data for the real part of the impedance and the model when the sample is between brass electrodes. We also show in this figure (see the inset) the usual model by taking perfect blocking electrodes into account to show the effect of the boundary conditions given by Eq. (4). Note that the usual model which corresponds to Eq. (4) with $\overline{\kappa}(t) = 0$, $\widetilde{\tau}(\vartheta) = 0$ and $\overline{\tau}(\overline{\gamma}) = \delta(\overline{\gamma} - 1)$ cannot reproduce the behavior of the impedance for $f < 1$ KHz. However, it has a good agreement with the experimental data for $f > 1$ KHz where the system is properly described in terms of the usual diffusion equation. This feature can be verified by the agreement between the experimental data and the usual model based on the standard diffusion equation with blocking electrodes [36]. For $f < 1$ KHz, the experimental data obtained for the real or imaginary part of the impedance evidence an anomalous diffusion process related to the ions. This unusual diffusion process may be produced by bulk and/or surface effects which influence the dynamic of the ions. To quantify these effects in the experimental data for the real and imaginary parts of the impedance, we fit the experimental data by using the previous model and testing the sensibility of the parameters present in the model when compared with the experimental data. Our best fit is obtained for $\varepsilon = 83\varepsilon_0$ [where $\varepsilon_0 = 8.815 \times 10^{-12}\ \text{C}^2/(\text{Nm}^2)$], $\mathcal{N} = 4.1 \times 10^{21}\ \text{m}^{-3}$, $\mathcal{D} = 5.88 \times 10^{-8}\ \text{m}^2/\text{s}$, $\gamma = 1$, $\tau(\vartheta) = \delta(\vartheta - 1)$, $\overline{\kappa}(i\omega) = \kappa\tau[(i\omega\tau)^{\vartheta_1} + (i\omega\tau)^{\vartheta_2}]/2$, $\kappa = 1.8 \times 10^{-6}$ m/s, $\vartheta_1 = 4\vartheta_2 = -0.88$, and $\tau = 1$ s which evidences that the dynamic of the ions in the electrolytic is governed by the model presented in the previous section based on anomalous diffusion approach. Note that the parameter τ has the dimension of time, whereas κ has the dimension of a length/time. Consequently, $\kappa\tau$ represents an intrinsic thickness related to the surface effect

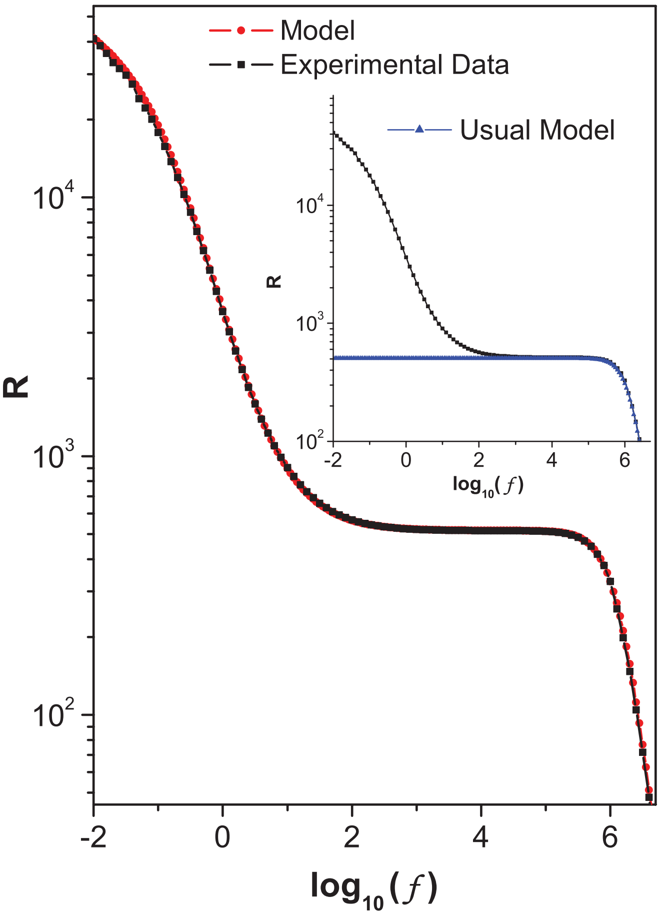

FIG. 1. (Color online) Real part of the electrical impedance of the cell versus the frequency of the applied voltage $f = \omega/2\pi$. The inset compares the behavior of the usual model with the experimental data of the real part of the impedance.

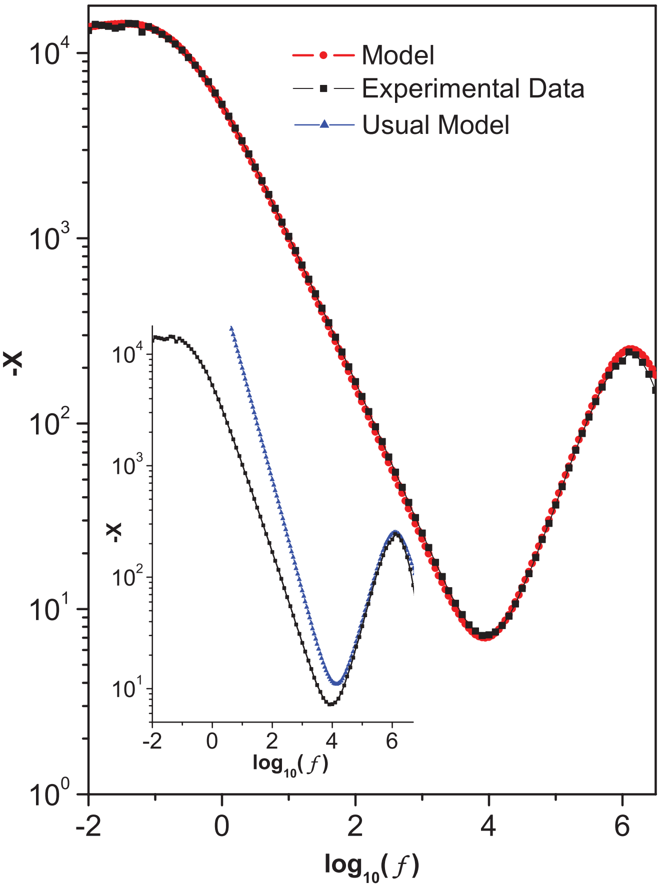

FIG. 2. (Color online) Imaginary part of the electrical impedance of the cell versus the frequency of the applied voltage $f = \omega/2\pi$. The inset compares the behavior of the usual model with the experimental data of the imaginary part of the impedance.

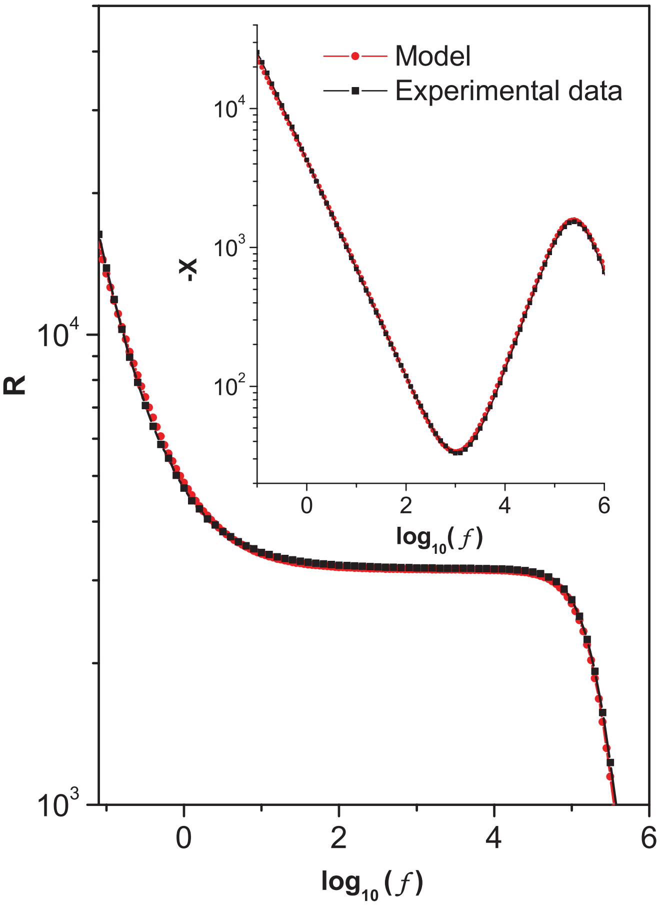

FIG. 3. (Color online) The real part of the electrical impedance of the cell versus the frequency of the applied voltage $f = \omega/2\pi$. The inset compares the experimental data of the imaginary part of the impedance with the theoretical prediction of Eq. (11).

on the ions. The parameters γ, ϑ_1, and ϑ_2 are connected with the effects produced by the bulk and surfaces on the dynamic of the ions. Figure 2 shows the agreement between the model and experimental data for the imaginary part of impedance at 10 mHz $< f <$ 5 MHz. The inset (Fig. 2) shows the deviation of the usual diffusion process at frequency $f <$ 10 KHz. In the region of high frequency, the imaginary part of impedance behaves according to the prediction of the usual model [36].

Figure 3 shows the experimental data for the real and imaginary parts of the electrical impedance of Milli-Q water for stainless steel electrodes. The behavior of R and $-X$ is the same observed in Fig. 1. The model has a good agreement with the experimental data. In this figure we have considered $\varepsilon = 76\varepsilon_0$, $\mathcal{N} = 2.258 \times 10^{21}$ m^{-3}, $\mathcal{D} = 1.52 \times 10^{-8}$ m^2/s, $\tau(\vartheta) = \delta(\vartheta - 1)$, $\overline{\kappa}(i\omega) = \kappa\tau[(i\omega\tau)^{\vartheta_1} + 0.75(i\omega\tau)^{\vartheta_2}]$, $\overline{\tau}(\overline{\gamma}) = 0.9\delta(\overline{\gamma} - 1) + 0.1\delta(\overline{\gamma} - 0.98)$, $\kappa = 7.3 \times 10^{-6}$ m/s, $\vartheta_1 = -0.34$, $\vartheta_2 = -0.14$, and $\tau = 0.3$ s to obtain the theoretical prediction from the model.

IV. DISCUSSIONS AND CONCLUSION

We have investigated the electrical response of Milli-Q water by using the impedance spectroscopy technique and the fractional diffusion equation of distributed order with the interfaces (i.e., the boundary conditions at the electrodes limiting the sample, governed by integrodifferential equations). The results have shown a good agreement between the experimental data and the theoretical results obtained from Eq. (11) in the frequency range 10^{-2} Hz to 5 MHz. Thus, the dynamic of the ions in the sample is anomalous for $f <$ 1 KHz and could be related to an anomalous diffusion by considering the agreement of the experimental data and the model. For $f >$ 1 KHz the usual approach based on the diffusion with blocking electrodes can describe the behavior of the experimental data. These features may be related to the diffusive regimes manifested by the ions in the sample due to the surface and bulk effects which have a direct influence on the electrical response of the system. Similar situations are discussed in Ref. [32] for disordered solids and point out that the dynamics of the ions may be characterized by different diffusive regimes and exhibit and anomalous behavior. Finally, we also hope that the results presented here can be useful to investigate the electrical response of other systems and their connection with the anomalous diffusion.

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