Electrode Polarization Effects in Broadband Dielectric Spectroscopy

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In the present work, we provide broadband dielectric spectra showing strong electrode polarization effects for various materials, belonging to very different material classes. This includes both ionic and electronic conductors as, e.g., salt solutions, ionic liquids, human blood, and colossal-dielectric-constant materials. These data are intended to provide a broad data base enabling a critical test of the validity of phenomenological and microscopic models for electrode polarization. In the present work, the results are analyzed using a simple phenomenological equivalent-circuit description, involving a distributed parallel RC circuit element for the modeling of the weakly conducting regions close to the electrodes. Excellent fits of the experimental data are achieved in this way, demonstrating the universal applicability of this approach. In the investigated ionically conducting materials, we find the universal appearance of a second dispersion region due to electrode polarization, which is only revealed if measuring down to sufficiently low frequencies. This indicates the presence of a second charge-transport process in ionic conductors with blocking electrodes.

1. INTRODUCTION

Electrode polarization (EP) leading to blocking electrodes (BEs) is a frequently encountered phenomenon when investigating ionic conductors with impedance or dielectric spectroscopy. This effect, giving rise to giant values of the dielectric constant and a strong drop of conductivity towards low frequencies, arises when the ions arrive at the metallic electrodes and accumulate in thin layers immediately beneath the sample surface forming a so-called space-charge region. It occurs in solid-state electrolytes as well as in aqueous solutions, ionic liquids, and in many biological systems (e.g., [1–18]). EP in ionic conductors often hampers the determination of the intrinsic dielectric properties but it also is of high technical relevance, e.g., in the design of fuel cells or double layer capacitors.

EP also plays an important role in the dielectric spectroscopy of electronic conductors (e.g., [19–31]). In semiconducting samples it is caused, e.g., by the formation of Schottky diodes at the electrode-semiconductor interface. In this case the blocking of the electrode arises from the depletion layer at the semiconductor/metal contact in the blocked state of the diode. Recently these effects came into the focus of interest due to their presumed importance in the so-called "colossal dielectric constant" (CDC) materials [24–27, 28, 30–32].

The many suggestions made to account for electrode effects in dielectric measurements can be classified into two categories, namely, (i.) avoiding the electrode polarization by specialized measurement techniques or (ii.) modeling these contributions, e.g., by equivalent circuits. The first approach, using, for example, four-electrode methods or varying the electrode distances (e.g., [1–33, 34]), quite generally implies considerable additional experimental efforts compared to standard dielectric measurements. Thus, often the second approach is adopted and numerous, partly quite sophisticated phenomenological and microscopic models have been proposed to model EP effects in dielectric spectra (e.g., [3–7, 12–16, 24, 35–43]). In dielectric studies, one often deals with situations where the detected EP effects are not in the focus of interest but still cannot be ignored since they have to be taken into account for an unequivocal determination of the intrinsic properties. In such cases, for a straightforward modeling of EP effects, simple equivalent circuits are usually employed, which are assumed to be connected in series to the intrinsic bulk contribution of the sample material. The most common ones are a parallel RC circuit [16, 19, 22, 23, 44, 45] or a so-called constant phase element (CPE) [4, 6, 7, 10, 33, 39, 46], the latter being mainly applied for modeling ionic conductors. Both models work relatively well if only the onset of EP effects is seen, arising close to the low-frequency boundary of the covered frequency range. However, in our experience both approaches have certain drawbacks when analyzing broadband dielectric spectra, where contributions from EP are observed over several frequency decades. Instead we found that a distributed RC equivalent circuit (referred to as DRC circuit henceforth), based on a Cole-Cole (CC) distribution of relaxation times [21, 47–50], is best suited to describe EP. This is true for such different materials as aqueous solutions, biological systems, solid-state electrolytes, ionic liquids and melts, and all kinds of electronic conductors.

The purpose of the present work is twofold: i.) To provide broadband experimental spectra with strong EP effects in a variety of very different materials that may be helpful for future tests of models of EP (for this purpose, the experimental data are provided in electronic form in the supplementary information. ii.) To demonstrate the universal applicability of the DRC circuit for the modeling of EP effects.

2. MATERIALS AND METHODS

Low-frequency measurements up to \( \nu \approx 3 \) MHz were performed using frequency-response analyzers (Novocontrol Alpha-A and Schlumberger 1260 combined with the
The CPE is often employed to account for EP in ionic conductors [6, 7, 10, 11]. Its admittance is defined by 
\[ Y^* = A(i\nu)^n \] with an exponent \( n < 1 \) and the prefactor \( A \). A theoretical rationale for the CPE can be obtained when considering a fractional geometry of the electrode/sample interface [4, 5]. A more intuitive modeling of EP effects is provided by a parallel connection of a resistance \( R_E \) and a capacitance \( C_E \), connected in series to the bulk impedance \( Z_B \) [16, 19, 22, 23, 44, 45]. Here \( C_E \) models the high capacitance of the thin blocking layers and \( R_E \) accounts for their high resistance (\( R_E = \infty \) for complete blocking). This equivalent circuit leads to a total impedance of

\[ Z_{\text{total}} = Z_B + \frac{R_E}{1 + i\omega \tau_E} \] (1)

with \( \tau_E = R_E C_E \) and \( \omega = 2\pi\nu \) the circular frequency. Assuming, e.g., a frequency-independent bulk resistance and capacitance (i.e., another RC circuit) for the bulk response, this results in so-called Maxwell-Wagner relaxation. It leads to a frequency dependence of the total complex capacitance (and thus of the permittivity \( \varepsilon^* = \varepsilon' - i\varepsilon'' \)) that is fully equivalent to the Debye relaxation law [54] describing the relaxational response of an ideal dipolar system [32]:

\[ \varepsilon^* = \varepsilon_\infty + \frac{\Delta\varepsilon}{1 + i\omega\tau} - i\frac{\sigma_{dc}}{\omega\varepsilon_0} \] (2)

\( \tau \) denotes the relaxation time (not identical with \( \tau_E \) [32]) and \( \Delta\varepsilon = \varepsilon_s - \varepsilon_\infty \) the dielectric strength. \( \varepsilon_s \) and \( \varepsilon_\infty \) are the limiting values of the real part of the permittivity for frequencies well below and above the relaxation frequency \( \nu_{\text{relax}} = 1/(2\pi\tau) \), respectively. The last term in eq. (2) was included to account for the contribution of dc charge transport to the dielectric loss \( \varepsilon'' \propto \sigma/dc \), with \( \sigma_{dc} \) the dc conductivity. Typical spectra of \( \varepsilon' \) and the conductivity \( \sigma' \) arising from the Maxwell-Wagner relaxation mechanism, described above, are schematically shown by the solid lines in Figs. 1(a) and (b), respectively. It should be noted that in case of the validity of eq. (3), an intrinsic dipolar relaxation and a Maxwell-Wagner relaxation caused by EP cannot be distinguished based on the measured spectra alone.

However, we found that in many cases the above approach does not lead to a satisfactory description of the experimental data, especially if the spectra extend to frequencies sufficiently low to reveal more than just the onset of the EP effects. Then the spectral features are often found to be more smeared out than expected for this simple equivalent circuit. Interestingly, intrinsic relaxations, e.g., in dipolar glass forming liquids [51], usually exhibit a similar behavior, i.e. a broadening of the observed spectral features. In these cases, the use of an empirical extension of the Debye function, the Havrilak-Negami function [56], is well established to account for the observed deviations from Debye behavior:

\[ \varepsilon^*(\nu) = \varepsilon_\infty + \frac{\Delta\varepsilon}{[1 + (i\omega\tau)^{1-\alpha}]^\beta} - i\frac{\sigma_{dc}}{\omega\varepsilon_0} \] (3)

The above function with \( 0 \leq \alpha < 1 \) and \( 0 < \beta \leq 1 \) includes the special cases with \( \beta = 1 \) and \( \alpha = 0 \), termed Cole-Cole [57] and Cole-Davidson [58, 59] function, respectively. These two additional parameters lead to a broadening of the spectral features, which is commonly ascribed to a distribution of relaxation times. While
the Havriliak-Negami and the Cole-Davidson functions are purely empirical, the Cole-Cole distribution of \( \tau \) can be approximated by the microscopic model of Gaussian-distributed energy barriers \([60]\). This formula leads to a symmetric broadening (compared to the Debye case) of the relaxation peak in \( \varepsilon' \) and, e.g., is often used for the description of secondary relaxations of glassy matter \([61, 62]\). However, we found that a direct application of eq. (3) to Maxwell-Wagner relaxations arising from EP \([61, 62]\). However, we found that a direct application of the description of secondary relaxations of glassy matter of the relaxation peak in a symmetric broadening (compared to the Debye case) is assumed to be distributed. In the equivalent-circuit case, the corresponding quantity is \( \tau = R_B C_E \) (\( R_B \) denotes the bulk resistance), which determines, e.g., the loss peak position via \( \nu_\tau = 1/(2\pi\tau) \), just like \( \tau \) in eq. \([3, 24, 32]\). However, the quantity that is distributed in the equivalent-circuit case is \( \tau_E = R_E C_E \), thus leading to different curve shapes.

Finally it should be mentioned that the DRC circuit for the Cole-Cole case and a parallel connection of a CPE and a resistor can produce identical frequency dependences of the dielectric quantities \([50]\). The DRC circuit seems to be the more physical one because fitting the experimental data with this model provides direct access to the resistances and capacitances of the blocking surface layers. However, as will become clear in the further course of this work, the extended CPE element may find an explanation in a second, slower hopping transport process within the BE layers \([14]\).

Figure 1 shows typical spectra of \( \varepsilon' \) (a) and \( \sigma' \) (b) calculated by assuming a DRC equivalent circuit to account for the EP. For comparison, spectra using a plain RC circuit (solid lines in (a) and (b)) and a CPE (frames (c) and (d)), instead of the DRC, are provided. In all cases for the bulk response a simple parallel RC circuit was assumed, corresponding to a frequency independent bulk \( \varepsilon' \) and \( \sigma' \). For the distribution, the CC case (i.e., \( \beta = 1 \) and \( \alpha \neq 0 \) in eq. \( [4] \) was chosen. The curves for the undistributed RC circuit (solid lines), which are identical to Debye behavior, eq. \( [2] \), show the typical dispersion steps in \( \varepsilon' \) and \( \sigma' \). With increasing \( \alpha \), these steps become smeread out for both quantities. In addition, the low-frequency plateau of \( \varepsilon'(\nu) \) develops into a weak power law. The low and high-frequency plateaus of \( \sigma'(\nu) \) remain unaffected (however, we found that for very high values of \( \alpha \) the \( \sigma'(\nu) \) curve can cross these limits). Comparing this behavior with that of the CPE case (Fig. 1(c) and (d)), the most obvious difference is the missing low-frequency plateau in \( \sigma'(\nu) \) for the latter (d). This would correspond to completely blocking electrodes. However, adding a parallel resistance to the CPE leads to such a plateau and, as mentioned above, in this case both circuits produce identical curves.

In the following, several experimental spectra being governed by strong EP effects, obtained on partly very different materials, will be fitted by the models introduced above. All fits were simultaneously performed for \( \varepsilon'(\nu) \) and \( \sigma'(\nu) \). For the experimental data, statistical errors were assumed.
4. RESULTS AND DISCUSSION

![Graph](image)

**FIG. 2:** (a) Dielectric constant and (b) real part of the conductivity of CKN at 379 K as function of frequency. The lines represent fits using different ways to account for the EP effects: equivalent circuits with a CPE (dash-dotted lines), a parallel RC circuit, eq. (1) (dotted lines), or a DRC circuit, eq. (4) with $\beta = 1$ (solid lines), connected in series to the bulk. For the latter, a power-law increase in conductivity at low frequencies is assumed. This approach is indeed observed in several of the investigated materials.

As demonstrated by the solid lines in Fig. 2 modeling the BEs by a DRC circuit yields satisfying fitting curves of $\varepsilon'(\nu)$ and $\sigma'(\nu)$ in the whole frequency range, including the onset of a low-frequency plateau in the conductivity. The resistance arising from the second charge-transport mechanism is taken into account by the resistor within the parallel RC equivalent circuit, modeling the EP effects. We find $\alpha = 0.26$ and $\beta = 1$, indicating a moderate, symmetrical distribution of relaxation times $\tau_E$ (Cole-Cole case). Broadband spectra obtained at other temperatures in CKN also can be well modeled in this way. In fact, in an earlier work a sophisticated model that included a DRC circuit (termed "ZC" there) to account for EP, was used to obtain excellent fits of spectra in CKN at four temperatures between 342 and 361K.

Figure 3 provides results on BMIM-BMSF, an ionic liq-
A good description of the present data. Instead, the

Admittedly, the mentioned features are not well pro-

longed and rather smeared out. However, we found

final blocking of a second charge-transport mechanism.

As an example for a classical electrolyte solution, Fig.

FIG. 4: (a) Dielectric constant and (b) conductivity of a

0.018 % LiBr/water solution at 295 K as function of frequency.

The meanings of the lines are the same as in Fig. 3.

As an example for a classical electrolyte solution, Fig.

FIG. 3: (a) Dielectric constant and (b) real part of the con-

ductivity of BMIM-BMSF at 252 K as function of frequency. The

The lines represent fits assuming different models for the EP
effects: the sum of two Cole-Cole functions, eq. (8) with \( \beta = 1 \)
(dashed lines), two parallel RC circuits, eq. (1), connected in
series to the bulk (dotted lines), and two DRC circuits, eq.
(4) with \( \beta = 1 \), also connected in series to the bulk impedance
(solid lines). For the latter two cases, the bulk is modeled by
a parallel RC circuit.

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FIG. 4: (a) Dielectric constant and (b) conductivity of a

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As an example for a classical electrolyte solution, Fig.
the intrinsic ion conductivity of this solution, dominated by the main charge-transport process (see discussion of the CKN data). This system exhibits a high conductivity and an onset of the EP effects at rather high frequencies (about \(10^5\) Hz in \(\sigma' (\nu)\), compared to \(3 \times 10^3\) Hz in BMIM-BMSF at 252 K and \(10^2\) Hz in CKN at 379 K, cf. Figs. 3 and 2). Probably for this reason, the appearance of a second plateau (at about \(10^2\) Hz and \(\sim 10^{-5} \text{ } \Omega^{-1}\text{-cm}^{-1}\)) is well pronounced in this electrolyte and, in addition, the final blocking is well seen (further decrease of \(\sigma' (\nu)\) below \(\sim 10\) Hz). Both effects clearly indicate a second conductivity mechanism that becomes blocked at lower frequencies, as already suspected in the discussion of the CKN and BMIM-BMSF data.

The lines in Fig. 4 again are fits with different empirical functions to account for the BEs. As demonstrated by the dotted lines, two parallel RC equivalent circuits (eq. (1)), both connected in series to the bulk impedance, obviously cannot account for the experimental data. Here again the bulk was assumed to have frequency-independent \(\varepsilon'\) and \(\sigma'\). The sum of two Cole-Cole functions (eq. (3) with \(\beta = 1\)) leads to much better, but still not perfect fits (dashed lines). In contrast, assuming two DRC elements, eq. (4), with \(\beta = 1\), connected in series to the bulk (solid line), provides nearly perfect fits. The values of the distribution parameter \(\alpha\) are 0.32 (for the low frequency EP effect) and 0.28 (for the high frequency EP effect).

As an example for the temperature-dependent development of EP effects, Fig. 5 shows \(\varepsilon' (\nu)\) (a) and \(\sigma' (\nu)\) (b) of a 0.73% LiBr/glycerol solution for various temperatures between 220 K and 410 K. For the lowest temperature shown (220 K) only the intrinsic \(\alpha\)-relaxation of glycerol (cooperative reorientation of the dipolar glycerol molecules \([51]\)) and the bulk conductivity arising from the ionic charge transport can be seen in this frequency range. This relaxation is characterized by steps in \(\varepsilon' (\nu)\) and in \(\sigma' (\nu)\) (the latter corresponding to the well-known loss peaks in \(\varepsilon'' (\nu)\) \([51]\)). The bulk conductivity leads to the low-frequency plateau in \(\sigma' (\nu)\) of about \(10^{-10} \text{ } \Omega^{-1}\text{-cm}^{-1}\). Due to the low ionic mobility at this temperature, the EP effects lie outside the investigated frequency range. With increasing temperature, the mobility rises and the EP shows up as a strong increase in \(\varepsilon' (\nu)\) at low frequencies that finally develops into a steplike curve. In addition, a decrease in the conductivity for decreasing frequency arises above 240 K. At even higher temperatures, a second, smeared-out step is revealed in \(\varepsilon' (\nu)\), accompanied by a weak corresponding feature in \(\sigma' (\nu)\). Similar to the behavior in the aqueous LiBr solution (Fig. 4), this further dispersion indicates a second charge-transport mechanism as discussed above. As demonstrated by the lines in Fig. 5, perfect fits of all these spectra are achieved by assuming a DRC circuit (eq. (4)) with \(\beta = 1\) for the description of the observed EP effects, connected in series to the bulk response. For the latter, the \(\alpha\)-relaxation is accounted for by the Cole-Davidson function, eq. (3) with \(\alpha = 0\) \([51, 70]\). The distribution parameters \(\alpha\) of the slower EP effect vary between 0.1 and 0.33; for the second one we obtained values between 0.1 and 0.29.

Further examples of dielectric spectra, dominated by EP are provided in Fig. 6. It shows the dielectric properties of a variety of partly very different materials, including a highly concentrated salt solution, two biological samples, and three electronic conductors. The squares represent the results obtained on a 17% LiCl/water solution at 290 K. Its frequency spectra reveal two strong dispersion steps in \(\varepsilon' (\nu)\) (a) and \(\sigma' (\nu)\) (b). As demonstrated by the lines, the spectra are well fitted with two Cole-Cole DRC elements in series to the sample impedance, which is modeled by a simple RC circuit. The broadening of the low-frequency dispersion step is characterized by \(\alpha = 0.22\); the faster one could be modeled with a pure RC equivalent circuit (i.e. \(\alpha = 0, \beta = 1\)).

The triangles in Fig. 6 represent the broadband data of human blood at 290 K \([18]\). Blood can be considered as an aqueous solution of a variety of different salts with the addition of a considerable fraction of suspended “particles”, mainly red blood cells. As treated in detail in ref. \([18]\), at \(\nu > 10^5\) Hz, those data are dominated
by the well-known $\beta$- and $\gamma$-dispersions of blood\[18,71\]. The $\beta$-dispersion (at about $10^6$ - $10^7$ Hz) arises from a Maxwell-Wagner relaxation caused by the membrane of the red blood cells\[71\]. The $\gamma$-dispersion (at about $10^{10}$ Hz) is caused by the cooperative reorientational motion of water molecules\[18,71\]. At $\nu < 10^5$ Hz, strong EP effects show up in the blood spectra of Fig.\[6\]. The complete broadband spectra are well fitted with a single Cole-Cole DRC circuit (eq.\[3\] with $\beta = 1$), connected in series to the bulk impedance. The latter is modeled by the sum of two "normal" Cole-Cole functions (i.e. they are defined in $\varepsilon^*$; eq.\[3\] with $\beta = 1$) to account for the $\beta$- and $\gamma$-dispersions. The finding that a single DRC circuit is sufficient to account for the data may be ascribed to the fact that the onset of the EP effects in blood occurs at rather low frequencies, comparable to the results in CKN (Fig.\[2\]).

The diamonds in Fig.\[6\] show the results for a 0.009\% (5 mmol/l) Lysozyme solution. Protein solutions usually contain free ions and indeed EP effects show up in the present spectra, leading to strong dispersion in both quantities. The very gradual transition to a low-frequency plateau in $\varepsilon^*(\nu)$ at $\nu < 10^2$ Hz indicates the presence of a second, smaller step on top of the main dispersion. Indeed, excellent fits of the experimental spectra were achieved by using two DRC elements in series to the bulk. The DRC circuit leading to the smaller, low-frequency step is Debye-like ($\alpha = 0$, $\beta = 1$); the broad main dispersion step is characterized by $\alpha = 0.46$ and $\beta = 1$.

Finally, Fig.\[6\] also provides some typical data on electronically conducting materials, where the EP arises from the formation of Schottky diodes at the electrode/sample interface. Even when the polarity of the oscillating field changes, one of the two oppositely poled diodes at both sides of the sample always is blocked and dominates the response at low frequencies. In recent years, especially EP effects in non-metallic transition-metal oxides have gained increasing significance: Many of these materials show effects like magnetocapacitance, multiferroicity or the occurrence of very large dielectric constants that have triggered numerous dielectric investigations\[32,72,73\]. However, EP effects can considerably hamper the detection of their intrinsic dielectric properties and even be misinterpreted as being of intrinsic origin\[24,25,74,75\]. The examples provided in Fig.\[6\] include single-crystalline La$_2$CuO$_4$, a parent compound of high-$T_c$ superconductors\[19,21\], single crystalline La$_{15/8}$Sr$_{1/8}$NiO$_4$, a charge-ordered CDC material\[53\], and polycrystalline Pr$_{2/3}$Cu$_3$Ti$_4$O$_{12}$\[51\], a material structurally closely related to the well-known CDC material CaCu$_3$Ti$_4$O$_{12}$\[76\]. For all these materials, strong EP effects are revealed in Fig.\[6\] which look qualitatively similar to those in the ionic conductors. In all cases, the spectra can well be fitted by using a DRC circuit to account for the EP (lines). For La$_{15/8}$Sr$_{1/8}$NiO$_4$ a second DRC circuit was used to cover the second dispersion step seen below about $10^8$ Hz. In ref.\[53\], by measurements with different contact types the high-frequency step was clearly identified to arise from EP. The origin of the low-frequency step is not clarified yet but it was speculated that it may be connected to the inhomogeneous charge distribution in this prototypical charge-ordered material\[53\]. We also found the DRC to be applicable for the description of EP effects in Cu$_2$Ta$_4$O$_{12}$\[25\] and CaCu$_3$Ti$_4$O$_{12}$, another CDC material\[77\].

5. SUMMARY AND CONCLUSIONS

In the present work, we have provided dielectric spectra showing strong EP effects measured in a variety of materials. They partly belong to very different material classes like salt solutions, ionic liquids and melts, biological materials, and electronic semiconductors. Our results are intended to provide a comprehensive data base for comparing current and future approaches for the mod-

![Figure 6](image_url)
eling of EP effects in dielectric spectra. For this purpose, the data are made available in electronic form in the supplementary information. In addition, in the present work we have demonstrated that the DRC equivalent circuit is an easy-to-apply phenomenological model for fitting EP effects in dielectric spectra, which works well for all the different material classes investigated. However, this model certainly cannot be regarded as an alternative to microscopic models as discussed, e.g., in refs. [12, 14, 15, 41–43]. On the other hand, it has proven useful for a quick, phenomenological description of EP effects, enabling fits of experimental data in the complete investigated frequency range, which can help in the determination of the intrinsic bulk properties of the investigated material.

In all the investigated ionically conducting materials, we find evidence for a second charge-transport process that leads to a second plateau in the frequency-dependent conductivity at frequencies below the commonly observed decrease of $\sigma'(\nu)$ due to blocking. In addition, in most of these materials we find a further decrease of $\sigma'(\nu)$ at the lowest frequencies investigated, indicating final blocking of the charge carriers involved in this second process. These seemingly quite universal features have been only rarely detected so far: In most works on ionic conductors only the onset of the EP effects is observed in the spectra, and the data can be fitted by simple models like an undistributed RC circuit or a CPE. It is unlikely that the second dispersion, observed in measurements extending to sufficiently low frequencies, is due to a second, much slower ion species or due to electronic conduction. Instead, to us an explanation in line with a second, much slower ion species or due to electronic transport process in the region close to the electrode. Thus, using the DRC circuit is in line with the model considered in [15].

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