Extracting the contribution of conduction in permittivity measurements using the Kramers-Kronig relations

C. Margo¹, E. Géron¹, T. Ditchi, S. Holé² and J. Lucas¹*
Laboratoire de Physique et d’Etude des Matériaux, CNRS UMR8213, France
* Corresponding author: jerome.lucas@espci.fr, +33 1 40 79 45 33
1ESPCI-ParisTech,
2UPMC Sorbonne Universités

Abstract
The measurement of the material permittivity is often performed via an impedance measurement. In this case the measured permittivity includes the conduction contribution. Most of the time, the impedance-meters performing such measurement do not perform static measurement even if they can go to low frequencies. When the dipolar relaxation of the material leads to low frequency relaxation, separating the relaxation phenomenon from the conduction phenomenon can be difficult, especially when multiple relaxation phenomena superimpose. In this work we use the Kramers-Kronig relations to perform that separation by considering the frequency response aspect of the permittivity. The practical aspect of the method is presented and demonstrated on real measurements.

1. Introduction
Measuring the conductivity of poorly conducting material such as cement is a difficult task. In this paper, we consider measurements performed using an impedance meter. The measurement of these devices are performed by exiting a dielectric sample with potential at given frequencies and measuring the resulting current. A dielectric constant measurement can then be elaborated from the probe geometry and sample thickness [1]. Whatever the nature of the sample, the measured current is the sum of the conduction current and of the displacement current [2]. In a resistive and linear material, the conduction current is preponderant at low frequencies when the influence of the displacement current is effective around the dipolar relaxation frequencies of the material [3, 4]. Furthermore, when the relaxation frequencies are low as in the case of cement, both effect are superimposed making their separation difficult. In this paper, after a short review of the Debye model that we use as a reference, we show that the Kramer-Kronig relations [5] allow to split those effects with no assumptions on the relaxation model even when the relaxation frequencies are very low. Finally, as an illustration, the method is applied to some typical measurements obtained with a cement sample using the technique developed in [6].

2. Theoretical Background
2.1. Complex permittivity of dielectrics
Permittivity \( \varepsilon(\omega) \) corresponds to the response of a material to the electrical field. Dipoles get oriented more or less rapidly depending on their interactions with the lattice. As the displacement field \( \vec{D} = \varepsilon\vec{E} \) is the response of the material, it is necessarily out of phase with the electrical field. Permittivity \( \varepsilon(\omega) \) must be then a complex function to describe the phenomenon correctly.

For an isotropic homogeneous material whose response to the electrical field is linear and containing only one kind of dipoles, the Debye model applies and the permittivity can be written for one relaxation frequency as

\[
\varepsilon_D(\omega) = \varepsilon_\infty + \frac{\varepsilon_s - \varepsilon_\infty}{1 + j\omega/\omega_{rel}}
\]  

where \( \omega \) is the circular frequency, \( \omega_{rel} \) the dipolar relaxation circular frequency, \( \varepsilon_s \) is the static dielectric constant, \( \varepsilon_\infty \) is the optic region permittivity and \( j = \sqrt{-1} \). Notice that there may be more than one relaxation frequency if there are other kinds of dipoles involved in the process. The dipolar relaxation phenomenon appears at low frequency for large molecules. It appears at higher frequencies for smaller molecules or fractions of large molecules and even at higher frequencies at the level of atoms [3].

2.2. Impedance measurement of permittivity
The displacement field \( \vec{D} \) is difficult to measure directly. The permittivity is then often measured through a current measurement. The studied material is placed in a sample holder of known geometry, and the build capacitor is polarized under \( V(\omega) \). By measuring the current \( I(\omega) \), one can obtain the capacitance from \( j\omega C(\omega) = I(\omega)/V(\omega) \) and then deduces
\(\varepsilon(\omega)\) knowing the capacitor geometry. Unfortunately, the measured current consists in a contribution of the displacement current density \(\partial \vec{D}/\partial t\) and a contribution of the conduction current density \(\sigma \vec{E}\) due to the free charges present in the material event if they are few. Consequently the measured permittivity can be written as:

\[
\varepsilon_M(\omega) = \varepsilon_\infty + \left[\varepsilon'_P(\omega) + j \varepsilon''_P(\omega)\right] - j \frac{\sigma}{\omega} \tag{2}
\]

where \(\sigma\) is the static conductivity and \(\varepsilon'_P + j \varepsilon''_P(\omega)\) is the complex permittivity resulting from the polarization. As an illustration, when the material follows the Debye law as for the standard FR4 epoxy substrate [7], one obtains the plots presented in figure [1] for the real and imaginary parts of the permittivity.

In this figure one can see that the imaginary part effectively measured at 1 GHz for instance is nearly thrice the contribution of polarization because of the conduction. Nevertheless concerning FR4, as the resonance occurs at high frequency, whereas the conduction contribution decreases with the frequency, isolating both effect is not really difficult. It would not be the case if the resonance occurred at 100 MHz for instance.

\[\text{Real part of } \varepsilon(\omega)/\varepsilon_0\]

\[\text{Imagery part of } \varepsilon(\omega)/\varepsilon_0\]

![Figure 1: Permittivity of FR4 substrate accordingly to the Debye Model.](image)

The Debye model presented above applies when the considered material presents only one relaxation. In sophisticated material such as polymers or cements, empirically modified versions of the Debye law are often used such as the Cole-Cole model [8] to describe properly the superposition of many relaxations at low frequencies. In all cases the measured permittivity can be written as in equation (2).

Considering only one relaxation frequency, the parameters of the Debye model can be estimated through optimization [9]. Considering equation (2), one can search for the parameter set \((p_1, p_2, p_3, p_4)\) that minimizes the criterion

\[
\mathcal{C} = \sum_{\omega_k} \left| \varepsilon_M(\omega_k) - \left( p_1 + \frac{p_2 - p_1}{1 + j \omega_k / p_3} - j \frac{p_4}{\omega_k} \right) \right|^2 \tag{3}
\]

In this equation, \(\varepsilon_M(\omega_k)\) are the measured values of the permittivity at the circular frequencies \(\omega_k\). The minimization of \(\mathcal{C}\) gives good results when testing materials such as FR4 since the relaxation frequency is sufficiently high in these materials so that its effect can be easily separated from conduction effect. However it is much less efficient with multiple superimposed relaxations when the Cole-Cole or more complicated laws [10] apply.

3. Estimation of conduction from Kramers-Kronig relations

The displacement field \(\vec{D}\) is the material polarization response to the electrical field \(\vec{E}\). One has

\[
\vec{D}_P(\omega) = \varepsilon_P(\omega)\vec{E}(\omega) \tag{4}
\]

The inverse Fourier transform of this relation leads to

\[
\vec{D}_P(t) = \varepsilon_P(t) * \vec{E}(t) \tag{5}
\]

where operator * stands for the convolution product.
3.1. General principle

The material studied is considered linear and invariant in time. Because equation (5) is valid for any temporal evolution of \( E(t) \), \( \varepsilon_P(t) \) can be interpreted as an impulse response. Therefore the causality principle implies:

\[
\varepsilon_P(t) = u(t) \times \varepsilon_P(t)
\]

where \( u(t) \) refers to as the unity step function. Using the Fourier transform and considering the Principal Value, one obtains from (5):

\[
\varepsilon_P(\omega) = -\frac{2j}{\omega} \ast \varepsilon_P(\omega) = j \times H_T(\varepsilon_P(\omega))
\]

where \( H_T \) is the Hilbert transform. The minus sign in (7) may change accordingly to the definition used for the Fourier transform. In this paper we use the sign that is relevant with the FFT algorithm. Relation (7) is known as the Kramers-Kronig (K-K) relation. It is often referred to as the K-K relation pair by using separately the real part \( \varepsilon'_P \) and the imaginary part \( \varepsilon''_P \) of \( \varepsilon_P(\omega) \) as

\[
\begin{align*}
\varepsilon'_P(\omega) &= H_T(\varepsilon'_P(\omega)) \\
\varepsilon''_P(\omega) &= -H_T(\varepsilon''_P(\omega))
\end{align*}
\]

Since Equation (2) can be rewritten as

\[
\varepsilon_M(\omega) = \varepsilon_\infty + \varepsilon'_P(\omega) + j (\varepsilon''_P(\omega) - \frac{\sigma}{\omega})
\]

and because \( H_T(\varepsilon_\infty) = 0 \) since \( \varepsilon_\infty \) is constant, one has

\[
\varepsilon''_P(\omega) = H_T(\Re(\varepsilon_M(\omega))).
\]

It is thus now possible to retrieve the conductivity \( \sigma \) from the measurements by

\[
\sigma = j\omega \left[\Im((\varepsilon_M(\omega)) - H_T(\Re(\varepsilon_M(\omega))))\right]
\]

It is possible to verify the consistency of calculation (11) by verifying whether if \( \sigma \) is constant over a large range of frequencies.

3.2. Calculation using the Fast Fourier Transform

Hilbert transform is a convolution product. It can therefore be calculated using a Fourier transform. Numerically speaking, the discrete Hilbert transform \( H_T(S) \) can be computed for a given sampled signal \( S(x) \) using the FFT algorithm as

\[
H_T(S) = \text{IFFT}(j \times \text{SGN}(k_i) \times \text{FFT}(S))
\]

In this equation \( \text{SGN}(k_i) \) emulates the signum function. It is 1 when \( k_i > 0 \), -1 when \( k_i < 0 \) and 0 when \( k_i = 0 \). It is also possible to use a direct convolution algorithm to compute the Hilbert transform. It does not yield better results than using the FFT algorithm and it is much slower.

4. Application to real measurement

We have applied the method presented above to measurements performed on one-month dry doped cement samples made of MIPLACOL EN 13813-CT16F4 from Bostik SA. The measurements were carried out using a SOLARTRON 1260 impedance meter in the frequency range from 1 Hz up to 1 MHz. To reduce the measurement time, they were performed at logarithmically spaced frequencies. In order to use the FFT algorithm, they were evenly re-sampled at 0.2 Hz using a linear interpolation between measurement points. The imaginary and real parts of the measured permittivity are shown in figure 3.

In this figure, \( \varepsilon_\infty \) is the vacuum permittivity. Considering the shape of the real part of the measurement, one can see that the relaxation circular frequency is very low and therefore superimposes with the conductivity contribution. This measurement is thus a typical case where the proposed method presented in this paper is interesting.

The result of the Hilbert transform applied to the real part of the re-sampled measurements is presented in red dashed line in figure 2. It presents a bell like shape as expected. By localizing its maximal value one can see that relaxations occur around \( f_{rel} = 2 \) Hz which is indeed low. The conduction contribution to the imaginary part of the permittivity is presented in solid blue in the same figure. A straight line is obtained as expected. One can notice that the contribution of conduction is not negligible in regard to the overall imaginary part of the permittivity below 2 Hz.

The conductivity calculated using equation (11) is presented in figure 3. The values obtained are roughly the same up to 1 kHz (red part). As the contribution of conductivity is very small for higher frequencies, the values obtained above 1 kHz are no longer relevant. At even higher frequency (above 100 kHz) one only obtains the contribution of the multiplication by \( \omega \) of equation (11). The conductivity estimated in figure 3 is the mean value of the curve tagged as \( \sigma \) estimation value. The standard deviation to this mean value is also indicated.

It is worth noting that the slope of the asymptotic behavior at infinity of the polarization of figure 2 (dashed red line) is less than 20 dB/decade. This means that the Debye model does not strictly apply.
5. Conclusion

In this paper, the nature of the permittivity measurement is discussed to show that it can always be split between conduction and polarization effects. Because of the causal aspect of polarization, which is the material response to the electrical field, we have proposed a method to extract the material conductivity independently from any behavioral models of that material. This makes the method interesting compared to optimization or fitting methods which require a model. The method is applied to complex cement samples and the results show its efficiency to retrieve the value of the conductivity of such samples. In MIPLACOL EN 13813-CT16F4, the conductivity estimated to be $5.2 \times 10^{-9}$ S/m.

References

References

[1] Agilent Impedance Measurement Handbook. Agilent Technologies, fourth edition, October 2013. www.agilent.com/find/impedance.

[2] C. Kittel. Introduction to solid state physics. John Willey & sons, fifth edition, 1976.

[3] Jackson J.D. Classical Electrodynamics. John Wiley & sons, sixth edition, 1967. Library of Congress Catalog Card Number: 62-8774.

[4] Ramo S, Whinnery J.R, and Van Duzer T. Fields and waves in communication electronics. John Wiley and Sons, 1984.

[5] C.F. Bohren. What did Kramers and Kronig do and how did they do it? Eur. J. Phys, 31:573–577, 2010. doi : 1088/0143-0807/31/3/014.
[6] J. Lucas, E. Géron, T. Ditchi, and S. Holé. A fast Fourier transform implementation of the Kramers-Kronig relations: Application to anomalous and left handed propagation. *AIP Advances*, 2012. AIP Advances 2, 032144 (2012); doi: 10.1063/1.4747813.

[7] M.Y Kolendisteva, K.N Rosanov, A. Orlandi, and J.L Droniak. Extraction of Lorentzian and Debye of dielectric and magnetic dispersive materials for FDTD modeling. *Journal of Electrical Engineering*, 53(9/S):97–100, 2002.

[8] S. K. Cole and H. R. Cole. Dispersion and Absorption in dielectrics. *Journal of Chemical physics*, 9:341–351, April 1941.

[9] Lagarias J.C, Reeds J.A, Wright M.H, and Wright P.E. Convergence Properties of the Nelder-Mead Simplex Method in Low Dimensions. *SIAM Journal of Optimization*, 9(1):112–147, 1998.

[10] S. Hawriliak and S.Negami. A complex plane representation of dielectric and mechanical relaxation processes in some polymers. *Polymer*, 8:161–210, 1967.