

Short Note

A new computational technique for processing transmission-line measurements to determine dispersive dielectric properties

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INTRODUCTION

Precision measurements of dielectric material properties over a wide frequency range have advanced considerably with the use of network analyzers and time-domain spectrometers (Cole and Cole, 1941, 1942; Fellner-Feldegg, 1969; Cole, 1975a, b; Cole et al., 1980; Feldman et al., 1992; Berberian, 1993; Feldman et al., 1996; Berberian and King, 2002; Huisman et al., 2004). The frequency range of these methods (1–10 000 MHz) encompasses those of ground-penetrating radar (GPR) and time-domain reflectometry (TDR) systems and the relaxation frequencies of polar liquids and dispersive clays. Some of these materials, especially the swelling clays (e.g., bentonite, montmorillonite), appear to have discrete relaxation times rather than the continuum thought to be characteristic of complex materials (Ishida et al., 2000). In this note we test an interpretational procedure that incorporates a genetic optimization algorithm to model dispersive dielectric properties using a Debye model with discrete relaxation times. Using network analyzer measurements of bentonite (scattering coefficients), we produce a Debye model that is physically reasonable and self-consistent, and it satisfies the Kramers-Kronig relationships.

Theoretical models have been proposed for dielectric permittivity as a function of frequency (Debye, 1929; Cole and Cole, 1941; Havriliak and Negami, 1966, 1967). Traditional commercially available methods from Hewlett-Packard (1985, 1992) for measuring dielectric properties of such media might experience severe problems. These techniques directly calculate complex permittivity for every measured frequency without being stabilized through the Kramers-Kronig relations (Kittel, 1976) between the real and imaginary parts of the complex, frequency-dependent permittivity. Recently, the accuracy of frequency-domain methods, as opposed to time domain, for measuring dispersive dielectric permittivity has been identified to provide the highest accuracy (Huisman et al., 2004). While these authors use TDR probes with three and

seven wires, we employ a coaxial cell because it is a very well-defined microwave system. In this note we describe a robust approach for measuring complex dielectric permittivity over frequency and ohmic conductivity of natural materials by assuming a Debye-like behavior of the medium.

METHODS

Our approach has been inspired by Schäfer et al. (1996), who assume that the complex dielectric permittivity $\epsilon_r(\omega)$ as a function of angular frequency ω is known, and we reconstruct the distribution of relaxation frequencies. Our measurement method starts from scattering parameters, which can be measured directly with a microwave vector network analyzer; this is a definite advantage for dielectrics exhibiting both high-conductive loss and dielectric dispersion.

The approach is based on a generalization of the Debye model (Debye, 1929; Schäfer et al., 1996) and global optimization techniques that overcome the limitations of traditional methods.

Dielectric material model

We start out with Debye's material model equation for the relative complex permittivity $\epsilon_r(\omega)$ of a homogeneous medium (Debye, 1929):

$$\epsilon_r(\omega) = \epsilon_\infty + \frac{(\epsilon_s - \epsilon_\infty)}{1 + j\omega\tau}. \quad (1)$$

In equation 1, ϵ_s denotes the static limit, ϵ_∞ the infinite frequency limit, and τ the inverse of the angular relaxation frequency ω_r :

$$\tau = \frac{1}{\omega_r} = \frac{1}{2\pi f_r}, \quad (2)$$

where $\omega = 2\pi f$ and f are angular and standard frequency, respectively. In laboratory measurements a superposition of multiple relaxation processes with different values of τ or even

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a continuous τ distribution is observed (Ishida et al., 2000; Zorn, 2002). Then, the Debye model can be extended either by additional, phenomenological parameters (Cole and Cole, 1941; Havriliak and Negami, 1966, 1967; Scaife, 1989) or by an ensemble-like superposition of weighted, multiple, discrete elementary Debye-type models (Böttcher, 1973a, b; Jonscher, 1983; Scaife, 1989). In terms of physical interpretation, the latter approach is more intuitive, particularly when interpreting relaxation frequencies according to the model given by Hilhorst et al. (2001). In the latter case,

$$\epsilon_r(\omega) = \epsilon_\infty + (\epsilon_s - \epsilon_\infty) \sum_{n=1}^N \frac{g_n}{1 + j\omega\tau_n}, \quad (3)$$

where N denotes the order of the generalized Debye model and g_n the weights for each of the corresponding relaxation frequencies f_r^n . The g_n are constrained

$$\sum_{n=1}^N g_n = 1 \quad \text{and} \quad g_n > 0. \quad (4)$$

Additionally, in a preliminary study (Oswald, 2000) we investigate modeling the distribution of g_n with Gaussians, corresponding to a continuous distribution of relaxation times. This yielded almost identical results but with many more parameters. The ohmic conductivity σ of the medium is kept separate, rather than being included in the dielectric losses because it is a physically different loss mechanisms; σ is included in the model of the complex characteristic impedance of the transmission line sections (see equations 5 and 10).

Measurement procedure

To determine dielectric properties at microwave frequencies, we use a vector network analyzer (VNA) to measure the complex scattering matrix. The measurement set up consists of a section of a coaxial transmission line with ample space to contain the sample material between the inner and outer conductors, both having a circular cross section (Nyfors and Vainikainen, 1989). In this study we use a coaxial line with inner radius $r_i = 13$ mm, outer radius $r_o = 30$ mm, and a length $\ell = 200$ mm. For a coaxial transmission line, the scattering matrix is formulated in closed form as a function of the dielectric and ohmic properties of the material between the conductors (Ramo et al., 1984). The characteristic impedance Z_w of any transmission line is given by

$$Z_w = \sqrt{\frac{R' + j\omega L'}{G' + j\omega C'}}, \quad (5)$$

where R' is resistance, L' inductance, G' conductance, and C' capacitance, all per-unit length of the transmission line, denoted by the prime. For a coaxial transmission line,

$$R' = \frac{\sqrt{\pi f \mu \rho}}{2\pi} \left(\frac{1}{r_o} + \frac{1}{r_i} \right) \quad (6)$$

and

$$L' = \frac{\mu}{2\pi} \ln \left(\frac{r_o}{r_i} \right), \quad (7)$$

with $\mu = \mu_r \mu_0$, where μ_r and μ_0 are the relative and the absolute magnetic permeabilities of vacuum, respectively. For a complex, relative permittivity so that

$$\epsilon_r = \epsilon'_r - j\epsilon''_r, \quad (8)$$

we obtain

$$C' = \frac{2\pi \epsilon'_r \epsilon_0}{\ln \left(\frac{r_o}{r_i} \right)} \quad (9)$$

and

$$G' = \frac{2\pi}{\ln \left(\frac{r_o}{r_i} \right)} (\epsilon''_r \epsilon_0 \omega + \sigma), \quad (10)$$

where ϵ_0 is the absolute dielectric permittivity of vacuum. The propagation constant γ of any transmission line, guiding the transverse-electric-magnetic (TEM) mode, is (Ramo et al., 1984)

$$\gamma = \alpha(\omega) + j\beta(\omega) = \sqrt{(R' + j\omega L')(G' + j\omega C')}. \quad (11)$$

The coaxial section with length ℓ and characteristic impedance Z_w is inserted between two transmission-line sections with characteristic impedance Z_{ref} , whose terminations are matched. The standard-reference impedance Z_{ref} in microwave measurements is 50 ohms. The scattering matrix of a coaxial transmission-line section is symmetric with coefficients

$$s_{11} = s_{22} = \frac{(Z_w^2 - Z_{\text{ref}}^2) \tanh(\gamma \ell)}{2Z_w Z_{\text{ref}} + \tanh(\gamma \ell)(Z_w^2 + Z_{\text{ref}}^2)} \quad (12)$$

and

$$s_{12} = s_{21} = \frac{2}{2 \cosh(\gamma \ell) + \sinh(\gamma \ell) \left(\frac{Z_w}{Z_{\text{ref}}} + \frac{Z_{\text{ref}}}{Z_w} \right)}. \quad (13)$$

Dielectric model parameter extraction

We extract the $2N + 3$ model parameters for ϵ_s , ϵ_∞ , σ , f_r^n , and g_n by minimizing the difference between calculated scattering coefficients based on dielectric model parameters and scattering coefficients measured with the network analyzer. The order N of the model is selected to be as small as possible so that it still gives the best fit; it is kept fixed during the optimization. The relationship between the scattering coefficients and the dielectric and ohmic properties is strongly nonlinear (see equations 12 and 13). Therefore, the objective function formulated for the optimization has several local minima, which forbids using gradient-type optimization procedures such as Levenberg-Marquardt (Huisman et al., 2002). Instead, we adopt a global-optimization approach based on genetic algorithms (Rahmat-Samii and Michielssen, 1999); this is greatly facilitated by the availability of the PGA-Pack (Portable Genetic Algorithm Package) library (Levine, 1996), which provides the basic operators.

Genetic algorithms are computer programs imitating the evolutionary process in nature, guided by a fitness criterion f that is roughly proportional to the inverse of a measure for the residual error r . They rely on the collective learning process within a population of individuals, each of which represents a search point in the space of potential solutions. The set of the model's parameters — real numbers, in our case — is encoded into an individual as a bit string. A population encompasses a

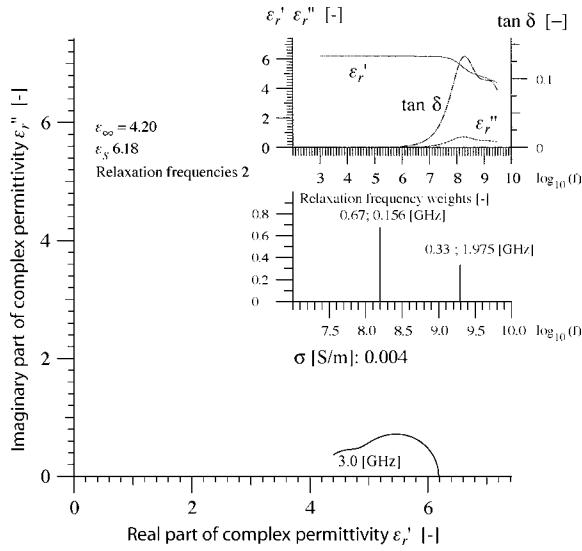


Figure 1. Results for bentonite at 10° C and $s = 0.0$. We plot the dielectric properties as a function of frequency, the real part, the imaginary part, the dielectric loss tangent $\tan \delta$, and the weighted relaxation frequencies f_r^n .

number of different bit strings. Novel solutions are obtained from the existing parameter sets by switching the bits (mutation) in an individual's bit string and/or by exchanging parts of the bit strings between two individuals (crossover). Genetic algorithms generally avoid the common pitfalls of local-minimization algorithms and hold the promise of finding novel solutions.

Our model does not require a starting guess for the optimization routine, except for physically reasonable limits such as $\epsilon_s \geq \epsilon_\infty \geq 1$. Based on physical arguments, we require that the relaxation frequencies f_r^n of a solid-aqueous mixture be below the value of bulk water (Hoekstra and Delaney, 1974). The residual error r is formulated

$$r \stackrel{\text{def}}{=} \sum_{i,j} \mu_{ij} \left| |s_{ij}^c| - |s_{ij}^m| \right| + \sum_{i,j} \lambda_{ij} \left| \arg(s_{ij}^c) - \arg(s_{ij}^m) \right|, \quad (14)$$

where μ_{ij} and λ_{ij} are the weights for the deviation between calculated c and measured m values of magnitude and argument of the scattering coefficients s_{ij} , respectively. Fitness f is roughly proportional to the inverse of the residual r :

$$f = \frac{f_0}{r + r_0}, \quad (15)$$

where f_0 is a scale factor and r_0 is a residual error to prevent division by zero. The algorithm is described in more detail by Oswald (2000).

RESULTS AND DISCUSSION

To test the algorithm, we use scattering coefficients measured for bentonite (ENRESA, 1998; Garcia-Gutierrez et al., 2001) with two different water saturations $s = \{0.0, 0.15\}$ at a temperature of 10°C with a dry density of 1.5 g/cm³. We plot the

dielectric parameters for the two data sets in Figures 1 and 2, a comparison between measured and calculated scattering coefficients and the learning curve in Figures 3 and 4, respectively. For bentonite containing water, the extracted relaxation spec-

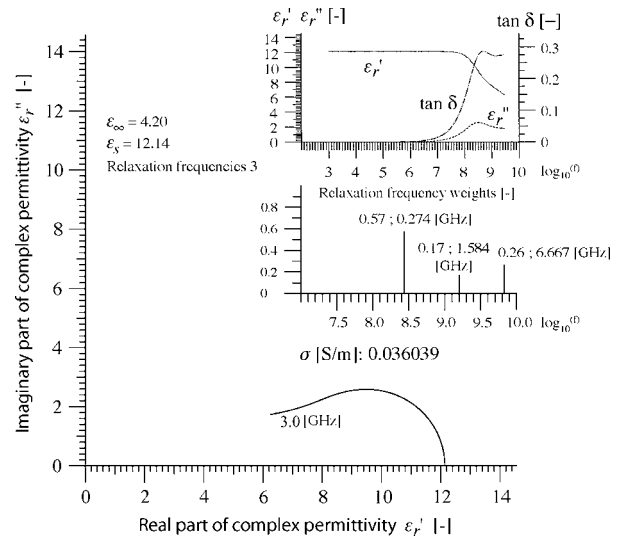


Figure 2. Same as in Figure 1 but for bentonite at 10° C and $s = 0.15$.

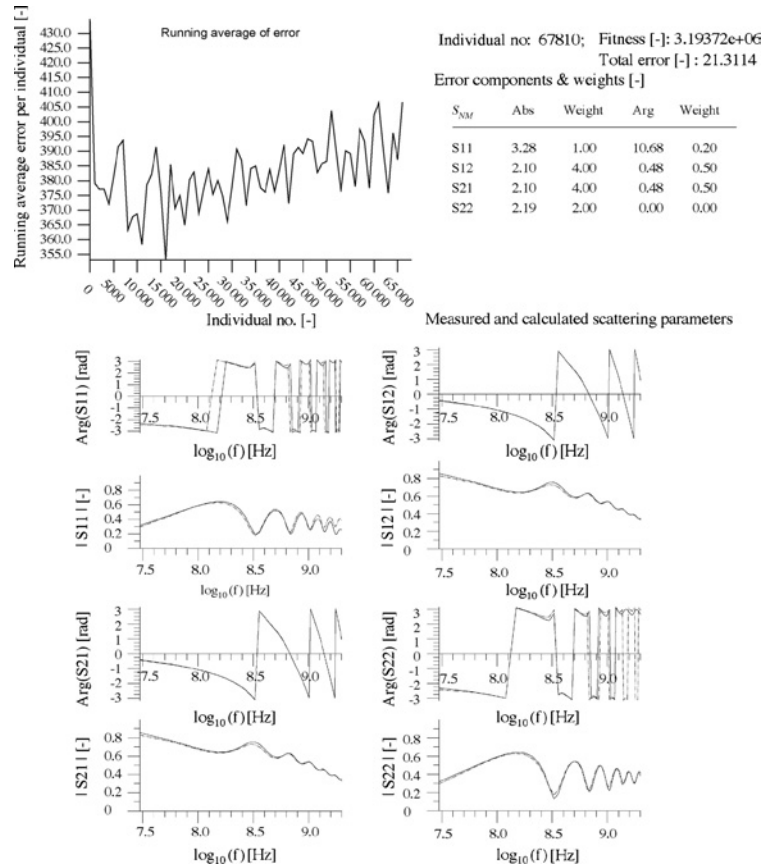


Figure 3. Plot of the absolute values and the argument of measured and calculated scattering coefficients. Note the learning curve, i.e., a down-sampled representation of the individuals' running averages of the residual for bentonite at 10° C and $s = 0.0$.

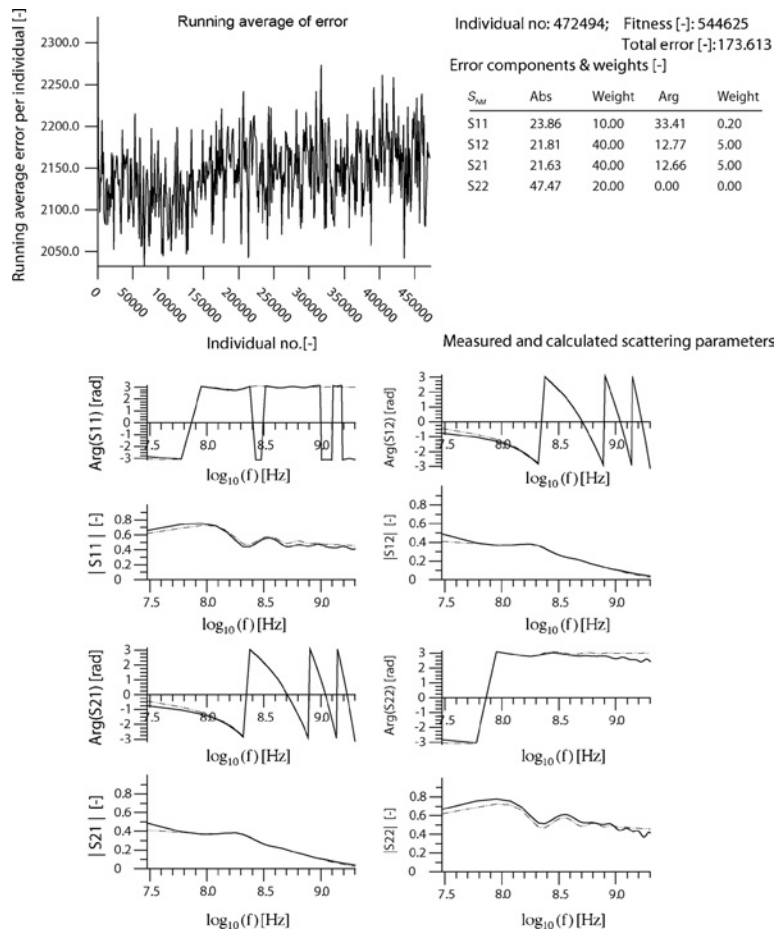


Figure 4. Same as in Figure 3 but for bentonite at 10° C and $s = 0.15$.

tra are consistent with Ishida et al. (2000) in that we also obtain three clearly different relaxation frequencies: namely, at 0.274, 1.584, and 6.67 GHz. Quantitatively, these values differ from Ishida et al. (2000) but are consistent with Hillhorst et al. (2001). We use a relatively large volume of bentonite to achieve a mixture as homogeneous as possible, while the volume used by Ishida et al. (2000) appears to be smaller. While Ishida et al. use a triple Cole-Cole model, we merely require a triple Debye model, additionally providing ohmic conductivity σ .

We also tested our algorithm by extracting dielectric properties of known materials, such as water and several alcohols. Our approach requires an assumption on the number of different Debye relaxation frequencies. While this appears to introduce a bias, the decision on this number is made relatively easily by observing the fit between measured and calculated scattering coefficients, depending on the number of relaxation frequencies. This is also done in Ishida et al. (2000). Heterogeneous mixtures that might exhibit a larger number of relaxation frequencies thus can be modeled also.

Our approach extracts parameters of the dielectric material model by adjusting model parameters through parameter optimization and thus ensures physically consistent results under the assumption of a Debye-like material model with

a prespecified number of relaxation frequencies. The Debye model constrains the real and imaginary parts through the Kramers-Kronig relations (Kittel, 1976). Inclusion of ohmic conductivity to model conduction currents provides one more degree of freedom and is required because ohmic and dielectric loss mechanisms are of different physical origins (Nyfors and Vainikainen, 1989). Taking into account the considerable dielectric and ohmic losses of bentonite — a challenging material for electrical characterization — the agreement between scattering coefficients calculated based on the model parameters and scattering coefficients measured with the network analyzer is good with respect to argument and magnitude. From this argument we conclude that modeling highly dispersive and simultaneously lossy materials with a number of discrete relaxation frequencies is justified if the agreement between measured and calculated scattering coefficients is used as a quality criterion.

We note that the measured s_{11} and s_{22} scattering coefficients are not completely identical as postulated by theory. We suspect that the distribution of the bentonite-water mixture filled into the coaxial transmission line is not completely homogeneous. Dry bentonite is best modeled with $N = 2$, while wetted bentonite with $s = 0.15$ is best modeled with $N = 3$.

CONCLUSIONS

We present a technique to determine the frequency-dependent dispersive dielectric properties using a generalized, physically justified Debye model together with ohmic conductivity σ from scattering coefficients only, measured by a network analyzer. The strongly nonlinear problem demands a globally optimizing algorithm. We use a genetic algorithm that does not require any starting guess; only physically reasonable limits are imposed. The extracted dielectric parameters are consistent with published data.

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REFERENCES

- Berberian, J. G., 1993, Time domain reflectometry: Bilinear corrections and extending the range of analysis beyond the quarter and half wavelength conditions: *Journal of Molecular Liquids*, **56**, 1–18.
- Berberian, J. G., and E. King, 2002, An overview of time domain spectroscopy: *Journal of Non-Crystalline Solids*, **305**, 10–18.
- Böttcher, C. J. F., 1973a, *Theory of electric polarization*, vol. I: Dielectrics in static fields: Elsevier Scientific Publishing Company.
- , 1973b, *Theory of electric polarization*, vol. 2: Dielectrics in time-dependent fields: Elsevier Scientific Publishing Company.
- Cole, K. S., and R. H. Cole, 1941, Dispersion and absorption in dielectrics: I — Alternating current characteristics: *Journal of Chemical Physics*, **9**, 341–351.
- , 1942, Dispersion and absorption in dielectrics: II — Direct current characteristics: *Journal of Chemical Physics*, **10**, 98–105.

- Cole, R. H., 1975a, Evaluation of dielectric behavior by time domain spectroscopy: 1 — Dielectric response by real time analysis: *Journal of Physical Chemistry*, **79**, 1459–1468.
- , 1975b, Evaluation of dielectric behavior by time domain spectroscopy: 2 — Complex permittivity: *Journal of Physical Chemistry*, **79**, 1469–1474.
- Cole, R. H., S. Mashimo, and P. I. Winsor, 1980, Evaluation of dielectric behavior by time domain spectroscopy: 3 — Precision difference methods: *Journal of Physical Chemistry*, **84**, 786–793.
- Debye, P., 1929, *Polare molekeln*: Verlag von S. Hirzel.
- ENRESA, 1998, FEBEX bentonite: Origin, properties and fabrication of blocks: Technical Publication 05/98, Empresa Nacional de Residuos Radiactivos S.A. (ENRESA).
- Feldman, Y., A. Andrianov, E. Polygalov, I. Ermolina, G. Romanychev, Y. Zuev, and B. Milgotin, 1996, Time domain dielectric spectroscopy: An advanced measuring system: *Review of Scientific Instruments*, **67**, 3208–3216.
- Feldman, Y. D., Y. F. Zuev, E. A. Polygalov, and V. D. Fedotov, 1992, Time domain dielectric spectroscopy: A new effective tool for physical chemistry investigation: *Colloid & Polymer Science*, **270**, 768–780.
- Fellner-Feldegg, H., 1969, The measurement of dielectrics in the time domain: *Journal of Physical Chemistry*, **73**, 616–623.
- García-Gutiérrez, M., T. Missana, M. Mingarro, J. Samper, Z. Dai, and J. Molinero, 2001, Solute transport properties of compacted c-bentonite used in FEBEX project: *Journal of Contaminant Hydrology*, **47**, 127–137.
- Havriliak, S., and S. Negami, 1966, A complex plane analysis of α -dispersion in some polymer systems: *Journal of Polymer Science*, **14**, 99–117.
- , 1967, A complex plane representation of dielectric and mechanical relaxation processes in some polymers: *Polymer*, **8**, doi:10.1016/0032-3861(67)90021-3.
- Hewlett-Packard, 1985, Measuring dielectric constant with the HP 8510 network analyzer: The measurement of both permittivity and permeability of solid materials: HP product note 8510-3.
- , 1992, Basics of measuring the dielectric properties of materials: HP application note 1217-1.
- Hilhorst, M. A., C. Dirksen, F. W. H. Kampers, and R. A. Feddes, 2001, Dielectric relaxation of bound water versus soil matric pressure: *Soil Science Society of America Journal*, **65**, 311–314.
- Hoekstra, P., and A. Delaney, 1974, Dielectric properties of soils at UHF and microwave frequencies: *Journal of Geophysical Research*, **79**, 1699–1708.
- Huisman, J. A., W. Bouten, and J. A. Vrugt, 2004, Accuracy of frequency domain analysis scenarios for the determination of complex dielectric permittivity: *Water Resources Research*, **40**, no. 2, 1–12.
- Huisman, J. A., A. H. Weerts, T. J. Heimovaara, and W. Bouten, 2002, Comparison of travel time analysis and inverse modeling for soil water content determination with time domain reflectometry: *Water Resources Research*, **38**, no. 6, doi:10.1029/2001/WR000259.
- Ishida, T., T. Makino, and C. Wang, 2000, Dielectric-relaxation spectroscopy of kaolinite, montmorillonite, allophane, and imogolite under moist conditions: *Clays and Clay Minerals*, **48**, no. 1, 75–84.
- Jonscher, A. K., 1983, *Dielectric relaxation in solids*: Chelsea Dielectrics Press.
- Kittel, C., 1976, *Introduction to solid state physics*, 5th ed.: John Wiley & Sons, Inc.
- Levine, D., 1966, Users guide to the PGAPack parallel genetic algorithm library: Argonne National Laboratory Technical Report 95/18.
- Nyfors, E., and P. Vainikainen, 1989, *Industrial microwave sensors*: Artech House, Inc.
- Oswald, B., 2000, Full wave solution of inverse electromagnetic problems — Applications in environmental measurement techniques: PhD dissertation, Swiss Federal Institute of Technology.
- Rahmat-Samii, Y., and E. Michielssen, 1999, *Electromagnetic optimization by genetic algorithms*: John Wiley & Sons, Inc.
- Ramo, S., J. R. Whinnery, and T. V. Duzer, 1984, *Fields and waves in communication electronics*, 2nd ed.: John Wiley & Sons, Inc.
- Scaife, B., 1989, *Principles of dielectrics: Monographs on the physics and chemistry of materials*: Oxford University Press.
- Schäfer, H., E. Sternin, R. Stannarius, M. Arndt, and F. Kremer, 1996, Novel approach to the analysis of broadband dielectric spectra: *Physical Review Letters*, **76**, 2177–2180.
- Zorn, R., 2002, Logarithmic moments of relaxation time distribution: *Journal of Chemical Physics*, **116**, 3204–3209.