

Cole–Cole Dispersion Models for Aqueous Gelatin-Syrup Dielectric Composites

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Abstract—A dielectric composite is developed with permittivity ranging from 8 to 75 with selectable conductive and dielectric losses. The composite comprises gelatin, high fructose corn syrup (HFCS), NaCl, and water, and can be used to model soils, loams, and sands in the 200-MHz to 20-GHz range. A single-term Cole–Cole dispersion equation is developed with frequency-independent parameters being functions of component concentrations. Fits are provided for various soil samples and surrogate concentrations.

Index Terms—Cole–Cole, dielectric composite, gelatin, soil.

I. INTRODUCTION

A dielectric composite of gelatin, high fructose corn syrup (HFCS), salt (NaCl), and water has been developed that has dielectric properties similar to that of soils. While the design of dielectric surrogates for biological tissues is well established, the dielectric constants of these are too high to model soils which typically have permittivities of 5 to 35 from 200 MHz to 20 GHz [1].

Various polarization mechanisms [2] in composite materials have response times related to the size of the dipole moments and affects opposing rotation. Soils in particular also have interfacial polarization when mesoscale structures of isolated inclusions are conducting [2]. These conducting regions act as macromolecules (known as the Maxwell–Wagner effect) causing an increase in the polarization at low frequencies. The combined effects yield complex variation of dielectric constant and loss with respect to frequency. The contribution of this work is the development of a Cole–Cole model of the permittivity of the dielectric surrogates. The Cole–Cole model has frequency-independent parameters expressed as functions of component concentrations.

II. DEBYE AND COLE–COLE RELAXATION MODELS

An alternating electric field acting on a polar liquid causes a reorientation of dipoles and consequently determines the permittivity profile. The effect on the permittivity $\epsilon^* = \epsilon' - j\epsilon''$ is captured by the Debye equation [3]

$$\epsilon^* - \epsilon_\infty = \frac{\epsilon_s - \epsilon_\infty}{1 + j\omega\tau} \quad (1)$$

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which can be decomposed into real and imaginary parts as

$$\epsilon' - \epsilon_\infty = \frac{\epsilon_s - \epsilon_\infty}{1 + (\omega\tau)^2} \quad \epsilon'' = \frac{(\epsilon_s - \epsilon_\infty)\omega\tau_0}{1 + (\omega\tau)^2}. \quad (2)$$

Here ω is the angular frequency, τ_0 is a characteristic constant describing the relaxation time, and ϵ_0 and ϵ_∞ are the static and infinite frequency dielectric constants, respectively. The magnitude of the dispersion can be expressed as $\Delta\epsilon = \epsilon_s - \epsilon_\infty$. Debye's equation models molecular dipoles when the response to applied electric fields is a relaxation process with a relatively long time constant. However, dispersion in many liquids and solids results in frequency broadening captured by a modified form of the Debye equation known as the Cole–Cole equation [4]

$$\epsilon^* - \epsilon_\infty = \frac{\epsilon_s - \epsilon_\infty}{1 + (j\omega\tau)^{1-\alpha}}. \quad (3)$$

The real and imaginary parts are now

$$\epsilon' - \epsilon_\infty = \frac{(\epsilon_s - \epsilon_\infty) [1 + (\omega\tau)^{1-\alpha} \sin(\frac{1}{2}\alpha\pi)]}{1 + 2(\omega\tau)^{1-\alpha} \sin(\frac{1}{2}\alpha\pi) + (\omega\tau)^{2(1-\alpha)}} \quad (4)$$

$$\epsilon'' = \frac{(\epsilon_s - \epsilon_\infty)(\omega\tau)^{1-\alpha} \cos(\frac{1}{2}\alpha\pi)}{1 + 2(\omega\tau)^{1-\alpha} \sin(\frac{1}{2}\alpha\pi) + (\omega\tau)^{2(1-\alpha)}}. \quad (5)$$

When $\alpha = 0$, the Cole–Cole equation reduces to the Debye equation while dispersion broadening is captured by $\alpha > 0$. For conductive materials, conductivity σ must be included in the Cole–Cole expression, leading to

$$\epsilon^* - \epsilon_\infty = \frac{\epsilon_s - \epsilon_\infty}{1 + (j\omega\tau)^{1-\alpha}} + \frac{\sigma}{j\omega\epsilon_0}. \quad (6)$$

Materials with several separated relaxation time constants in the frequency range of interest can be modeled by the multiterm Cole–Cole model

$$\epsilon^* - \epsilon_\infty = \sum_n \frac{\Delta\epsilon_n}{1 + (j\omega\tau_n)^{1-\alpha_n}} + \frac{\sigma}{j\omega\epsilon_0} \quad (7)$$

where n discrete polarization mechanisms are captured.

III. GELATIN SOLUTIONS

Composites of gelatin (type A, 250 bloom), water and NaCl were prepared and characterized from 200 MHz to 20 GHz as previously described [5]. Gelatin is a heterogeneous mixture of water-soluble proteins of high molecular weight and, on a dry basis, consists of 98% to 99% protein. It contains coils of amino acids joined together by peptide bonds and has a unique protein structure that forms a compound helix in aqueous solutions [6].

Gelatin is described in terms of Type and bloom. Bloom is an ascending index used to measure gel strength in grams. Type can be either *A* or *B*, with Type *A* being porcine gelatin (made from pigskins) and Type *B* being bovine gelatin (made from cattle). Gelatin is tasteless and odorless and swells and absorbs 5–10 times its weight of water to form a thermoreversible hydrogen-bond friendly gel in aqueous solutions between 30 °C to 35 °C [6], [7]. For our purposes, we use Type *A*, 250 bloom gelatin in concentrations of 5%, 10%, 15%, 20%, 25%, and 30% by weight. Laboratory grade NaCl is used in the mixture to vary ionic conduction effects in concentrations of 0%, 1%, 2%, and 3% by weight.

The surrogate measurements were made using an Agilent 85 070E open-ended coaxial dielectric probe kit and the Agilent 8510C Vector Network Analyzer (VNA). Complex permittivity measurements were acquired from 200 MHz to 20 GHz. The dielectric probe kit uses well established inverse models that correlate the reflection coefficient from the probe aperture/material interface to the complex permittivity of the material under test. Models can range from more narrow-band lumped circuit models to more accurate and computationally expensive variational techniques [8]. The advantage to using the open-ended coaxial probe is that a wide frequency spectrum can be measured, contrary to more traditional techniques, such as the resonant cavity and waveguide methods.

The accuracy (typical) of the dielectric constant, ϵ'_r measurements are $\pm 5\%$ and the loss tangent, ϵ''_r/ϵ'_r measurements are ± 0.05 . Repeatability and resolution (typical) per manufacturer's specifications, are 1% to 2% and for this reason, all measurements were made using a single VNA calibration in order to minimize calibration variation error during data acquisition.

The water or water/HFCS solution was heated to 65 ± 5 °C in a water bath and mixed using an electric mixing tool to eliminate air bubbles. The appropriate amount of NaCl was added to the water or water/HFCS solution prior to adding the gelatin powder.

The gelatin was added slowly to reduce clotting and required around ten minutes to dissolve completely for higher gelatin concentrations. The mixtures were then slowly cooled to room temperature and then refrigerated for 24 h to complete the gelation process. Once gelled, the samples were removed from refrigeration and allowed to reach room temperature (approximately 22 °C) prior to making measurements. A total of four measurements were made on various locations of each resulting gelatin-syrup sample and the results averaged. No temperature variance measurements were made during this effort.

A genetic algorithm (GA) was used to simultaneously fit the real and imaginary parts of the measured complex permittivity data to one-, two-, and three-term Cole–Cole equations. The GA was chosen because of its heuristic ability to quickly scan the vast solution set for each Cole–Cole parameter (ϵ_∞ , $\Delta\epsilon$, τ , α , σ). A single-term Cole–Cole model was found to fit the measured data (see Fig. 1) as accurately as the two- and three-term models. ϵ_∞ has been left to assume negative values for best-fitting purposes. Note that as the gelatin concentration increases, the dielectric constant decreases and the dielectric absorption behavior broadens. This indicates a distribution of relaxation

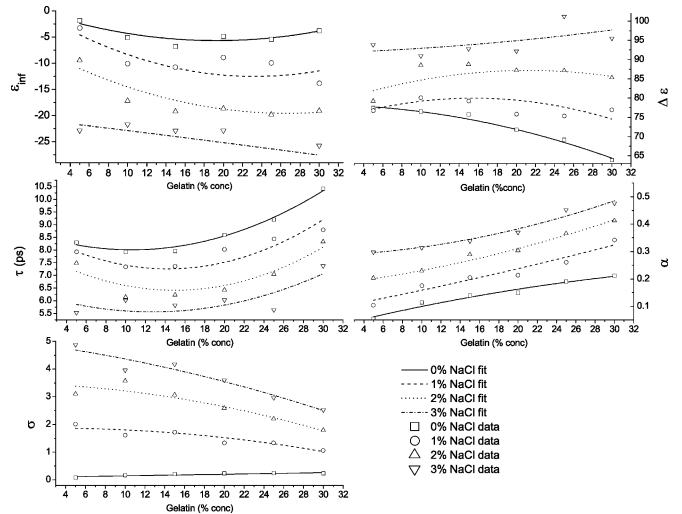


Fig. 1. Parameter fits of the one-term Cole–Cole equation with respect to NaCl (N) and gelatin (G) concentrations by weight.

times that increases with increasing gelatin concentration. Also, the ratio of bound-water to free-water increases as the gelatin concentration increases, which results in lower polarization at 20 GHz [9].

Levenberg–Marquardt optimization was used to extract polynomials of the form

$$x = (a_0 + a_1N + a_2N^2) + (b_0 + b_1N + b_2N^2)G + (c_0 + c_1N + c_2N^2)G^2$$

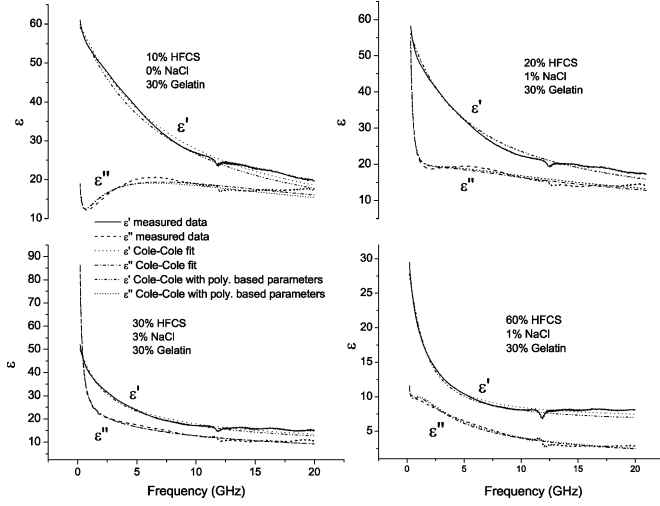
for each of the Cole–Cole parameters in terms of gelatin (G) and NaCl (N) concentrations by weight; see Table I. For 24 datasets the fitting error averaged 0.07% from 200 MHz to 20 GHz, with maximum error of 2.8%, for concentrations ranging from 5% to 30% gelatin and 0% to 3% NaCl.

Gelatin and NaCl alone do not provide the needed ranges of dielectric constant and loss factor to model all soil types. The required suppression of dielectric constant and loss was achieved with the addition of HFCS (42% fructose, 52% glucose, 6% higher sugars). Additionally, HFCS softens the mixture, eases the blending process and acts as a bactericide and fungicide to increase sample shelf life. Now suitable coverage is obtained with a fixed gelatin concentration of 30% which also yields a manageable mixture and provides adequate firmness following gelation. The measured dielectric properties for various HFCS and NaCl concentrations are given in Fig. 2 along with the one-term Cole–Cole fit and the permittivity calculated using polynomial parameter fits. The parameter fits are shown in Table II. The fitting error, compared to measurements, averaged 0.5% over the frequency range 200 MHz to 20 GHz with a maximum error of 11.3% for concentrations ranging from 10% to 60% HFCS and 0% to 3% NaCl.

The dielectric loss decreases significantly with increasing HFCS concentration as does the ionic conduction. The relaxation frequency decreases with increasing HFCS concentration as well and ranges from approximately 7 GHz at 10% HFCS concentration down to 1.5 GHz at 60%.

TABLE I
 EMPIRICAL FITS TO COLE-COLE PARAMETERS FOR GELATIN-SALT SOLUTIONS

y	$y = (a_0 + a_1N + a_2N^2) + (b_0 + b_1N + b_2N^2)G + (c_0 + c_1N + c_2N^2)G^2$								
	a_0	a_1	a_2	b_0	b_1	b_2	c_0	c_1	c_2
$\epsilon_\infty (\times 10^{-3})$	249.9	3605.75	-3526.85	-615.55	-800.19	311.21	15.96	14.37	-6.62
$\Delta\epsilon (\times 10^{-3})$	78263.85	-9130.65	4540.25	-38.58	1255.63	-405.36	-14.12	-20.99	9
$\tau (\times 10^{-15})$	8706.72	633.82	-470.4	-131.425	-141.827	47.6693	6.19357	2.74	-1.04214
$\alpha (\times 10^{-6})$	20290	57610	10220	8730	-1880	-201.241	-80.6589	138.591	-17.2768
$\sigma (\times 10^{-3})$	88.3	1840.72	-73.45	5.5	9.63	-9.07	0.00695357	-1.52	0.377018


 Fig. 2. Cole-Cole parameter fits with respect to NaCl and HFCS concentration. Shown are the measured data for ϵ' and ϵ'' , the single-term Cole-Cole fit, and the Cole-Cole fit with polynomial-based parameters.

IV. DISCUSSION

A primary motivator for this work was the development of a time-invariant, conveniently prepared, easily reproducible electrical surrogate for soils. A secondary objective was the development of a scaled equivalent with higher permittivity so that smaller sample sizes could be used while the frequency profiles of permittivity and loss tangent are maintained. The effective wavelength λ_{eff} is related to the dielectric constant by $\lambda_{\text{eff}} = c/f\sqrt{\epsilon}$, with f being frequency, such that scaling the dielectric constant by S reduces λ_{eff} and the effective sample dimensions by the factor

$$S' = \sqrt{\frac{1}{S}}. \quad (8)$$

A. Electrical Properties of Soil

Soil is an extraordinarily complex medium made up of a heterogeneous mixture of solid, liquid, and gaseous materials. Soil has time-dependent electrical properties resulting from variation in moisture content, temperature change and other compositional transformations making it a difficult material to use in measurements for validating theoretical predictions. The solid phase of soil contains particles of vastly different sizes, spanning the lower limits of the colloidal state to the coarsest fractions of sand and gravel [10].

The components of soil are classified into groups based on the effective diameter of the constituent particles. Sand in-

cludes particles with diameters between 0.05 and 2.0 mm, silt includes particles with diameters in the 0.002–0.05 mm range, and clay includes particles with diameters smaller than 0.002 mm. Several soil types and their composition are described in Table III.

Soil has the ability to retain water and the amount of water retention is dependent on the forces that act against the water in the soil matrix such as gravitational force, ionic forces from solid surfaces, and geometric arrangement of the particles [10]. Water in soils is considered to be bound or free and these states have different orientational and rotational polarization responses to an applied electric field [2]. Bound water refers to the water molecules contained in the first few molecular layers surrounding soil particles where water molecules tend to stick to hydrophilic surfaces [1]. Free water molecules, located several molecular layers away from the soil particles, are able to travel within the soil matrix with relative ease. The proportion of bound water is directly proportional to the surface area of the soil particles and thus the soil particle size distribution. Consequently the proportions of bound and free water, and thus electrical properties, are related to the textural structure of the soil [1], [11].

B. Correlation of Soil and Surrogate

The soil surrogates developed here can be made to look electrically similar to a variety of soils over wide frequency ranges although better matches are obtained over constrained ranges of a half octave. In Figs. 3 and 4 the performance of surrogates is compared to available measured data [12], [13]. Fig. 3 presents comparisons for two surrogates of sandy loams with moisture levels of 17% and 24%, respectively. Similarly good (but generally better) fits are obtained for similar soils with higher clay concentrations and with the same or higher moisture content. The loss is relatively high under these conditions and high loss is captured by decreasing the HFCS concentration and increasing the NaCl concentration to capture conduction effects.

With lower clay or lower moisture concentrations, scaling is required as the permittivity of the soils drops significantly below the level that can be obtained with the water-based gelatin composite. This is because of the high intrinsic permittivity of water alone. In any case scaling is an advantage because of the size reduction achieved. The performance of the surrogate in matching low moisture content is shown in Fig. 4. Here the dielectric constant scale factor is 15.3 reducing the dimensions of the surrogate model by 75% compared to soil. Note that the Maxwell-Wagner effect is evident as the slow rise of the imaginary component of the soil permittivity as frequency reduces is modeled by the surrogate.

TABLE II
EMPIRICAL FITS TO COLE-COLE PARAMETERS FOR 30% GELATIN SOLUTIONS AS A FUNCTION OF SALT (N) AND HFCS (S) CONCENTRATIONS

y	$y = (a_0 + a_1N + a_2N^2) + (b_0 + b_1N + b_2N^2)S + (c_0 + c_1N + c_2N^2)S^2$								
	a_0	a_1	a_2	b_0	b_1	b_2	c_0	c_1	c_2
$\epsilon_\infty (\times 10^{-3})$	-394.37	-6743.43	-998.07	385.69	299.13	48.77	-4.59	-3.22	-0.521682
$\Delta\epsilon (\times 10^{-3})$	64351.9	12631.9	2159.5	-589.24	-595.67	-79.78	-1.16	6.32	0.666964
$\tau (\times 10^{-15})$	25336.6	-7633.45	524.25	-545.79	611.317	54.3384	33.4119	-6.31313	-1.55045
$\alpha (\times 10^{-6})$	316910	165460	-8830	-4640	-5270	396.161	42.175	45.3804	-5.34821
$\sigma (\times 10^{-6})$	198000	875300	-23970	-4250	-24880	406.671	30.3322	175.764	2.75848

TABLE III
PHYSICAL PARAMETERS FOR THE FIVE SOIL TYPES USED IN THE DOBSON AND HALLIKAINEN STUDY

Soil Designation:	1	2	3	4	5
Textural Class:	Sandy Loam	Loam	Silt Loam	Silt Loam	Silty Clay
% Sand	51.51	41.96	30.63	17.16	5.02
% Silt	35.06	49.51	55.89	63.84	47.60
% Clay	13.43	8.53	13.48	19.00	47.38
Specific Surface, $A_s(m^2/g)$	52	49	66	119	252
Specific Density, $\rho_s(g/cm^3)$	2.66	2.70	2.59	2.66	2.56

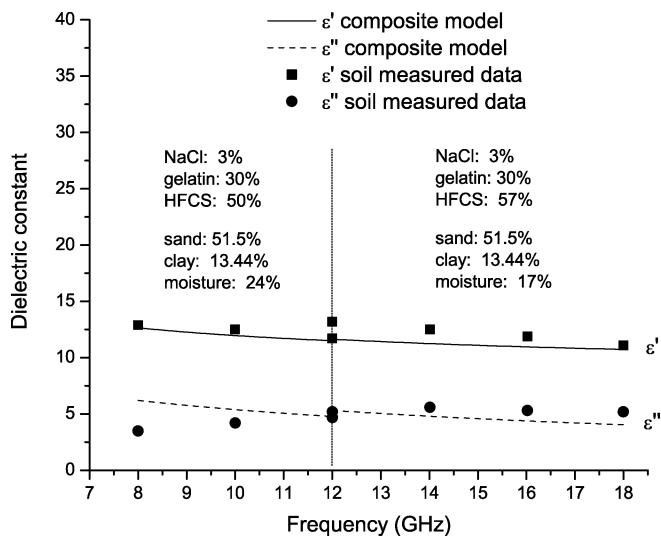


Fig. 3. Comparison of the dielectric surrogate with a sandy loam in the frequency range of 8–18 GHz. The measured data are from Hallikainen *et al.* [12].

V. CONCLUSION

A cost-effective dielectric surrogate can be constructed from water-based gelatin-syrup gels that provides an extensive range of real and imaginary permittivities from 200 MHz to 20 GHz. These surrogates provide a homogenous, stable, and easily reproducible substitute for time-dependent materials such as soils. A particular advantage of using HFCS is that water is bound to the fructose resulting in low variations of dielectric properties due to low evaporative effects. The fructose also acts as a bactericide and fungicide to increase sample shelf life. Frequency-independent models were developed using a one-term Cole–Cole model with parameters expressed as polynomial functions of constituent concentrations. Important effects such as the Maxwell–Wagner effect are modeled correctly.

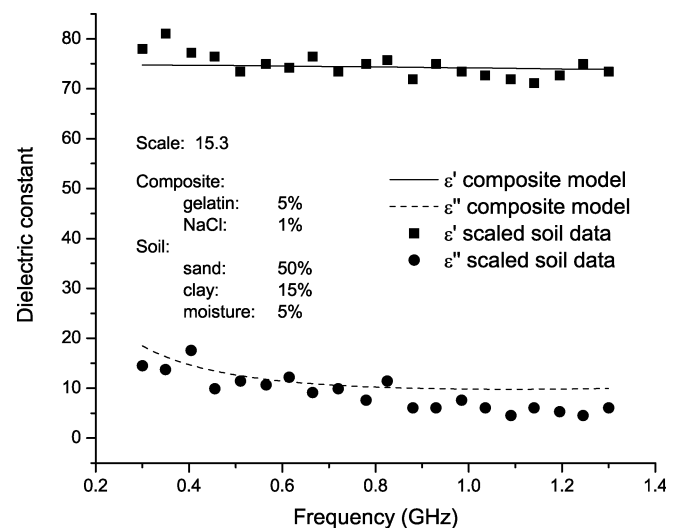


Fig. 4. Comparison of the dielectric surrogate with a sandy loam in the frequency range of 0.3–1.3 GHz with a scale of 15.3. The surrogate properties are: gelatin—5% and NaCl—1%. The loam properties are: sand—50%, clay—15%, and volumetric moisture—5%. The measured data are from Peplinski *et al.* [13].

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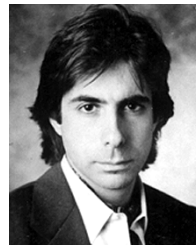


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