Dielectric mixtures
Importance and theoretical approaches

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Abstract
Physics of dielectric mixtures are presented to stimulate discussion and to provide information on the recent advances in this topic.

Keywords: Dielectric mixtures, bounds on dielectric properties, spectral density representation

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1 Introduction
Many engineering materials used in electrical insulation are composites [1]. As Dakin stated ‘Composite materials are at least an order of magnitude more diverse and perhaps as much more complex in their behavior than simple one-component materials.’ Books and review papers on dielectric mixtures or electrical properties of composites [2–14] discuss the properties of these systems with little or no focus on in their electrical insulation properties. The importance of the insulation properties would arise from the frequency dependent dielectric losses and changes in the dielectric breakdown strength of the composite system. The latter case is mainly dependent on the defect density and could be separated in to different branches as stated by Nelson [15]; by the motion of free charges, charge injection from the electrodes, charge multiplication and space charge formation, and the dissipation energy in the material (electro-thermal effect). Some of these effects would be temperature activated that can make the analysis cumbersome [16–18]. However, they are
effected by the local electric field and the energy landscape, which again brings the dielectric properties of
the composite. We will here focus on the dielectric relaxation in dielectric mixtures by taking in to account
the intrinsic properties of components and the geometrical arrangement of phases [11, 12, 19–33].

2 Background

The question to answer in designing an insulation system or material is how we can predict the effective
electrical properties of the system; notice that similar approaches can be applied to mechanical and thermal
properties. We would also like to decompose contributions of the constituent properties for determination of
aging and degradation from non-destructive measurement methods and diagnostic tests. A comprehensive
understanding of the electrical properties of insulation systems would be tremendously beneficial for new
equipment design and assessing already installed equipment maintenance. A chart regarding the relation
ship between material properties is shown in Fig. 1. The presented method later in text would make it possible to
obtain the required information in a dielectric mixture, such that one can estimate (i) the effective dielectric
permittivity of a mixture, or (ii) estimate the permittivity of one of the constituents, or (iii) estimate
topology of the mixture.

There have been analytical approaches for dielectric mixtures starting from the early days of electro-
technical sciences, see Sihvola [3] for review and numerous dielectric mixture formulas. For example Maxwell
[34] derived the mixture expression for layered binary structure; this was something interesting since the
mixture illustrated a new loss peak (or a dielectric dispersion) due to differences in the electrical properties of
the constituents, their permittivity and conductivity [35]. Wagner [36] expended the mixture electrical prop-
erties for a system with spherical inclusions. Both Fricke [37] and Sillars [38] improved Wagner’s approach
to ellipsoidal shape inclusions. This relaxation due to the mixture was later called as the Maxwell-Wagner-
Sillars (MWS) relaxation corresponding to the interface between the constituents. For dilute mixtures the
MWS relaxation is narrow, while for complex mixture topologies and high concentration of inclusions the
inter-facial relaxation can be broad [24]. While analytical models are of great use, they have limitations due
to the assumption in their derivation; they can not be applied to many real systems.

A novel approach to dielectric mixtures was first proposed by Fuchs [39] and then by Bergman [40],
which considers a summation rule related to the depolarization factors; it is called the spectral density
representation. Several improvements in the method were proposed throughout the years [7, 12, 23, 26, 41–
49]. This approach does not rely on one particular shape of inclusions; it considers distribution of shapes
or total topology of the mixture. Numerical approaches to resolve the topology of mixtures using spectral
density approach was employed previously [24, 25, 50–58]. Actually the spectral representation is analogous
to the distribution of relaxation times approach in the theory of dielectrics [12, 26]. Examples of mixture
formulas were shown by Boyd et al. [10], which is a nice illustration of the topologies as shown in Fig. 2. The
upper left and bottom right topologies are for the Wiener bounds [59]. The topology on the upper right is for
Bruggeman’s derivation [60] which represents a bi-percolating structure. The bottom left topology indicates
a mixture with spherical inclusions. The importance of the length scales should not be disregarded in the
effective material property calculations, such that the diameter of the inclusions should be much smaller than
the wavelength of the applied electric field. For example one can apply the effective medium approximations
to nanodielectrics in optical frequencies, however, one cannot apply the same to composites with sub-micron
size fillers due to similarities in the length scale of the applied electromagnetic wavelength and particle size
dimensions. A list of mixture expressions for spherical inclusions were given by Widjajakusuma et al. [61],
which illustrated the variety of formulas for a similar problem.

Numerical simulations of dielectric mixtures have been an approach to study the effective electrical
properties of these systems [3, 8, 11]. The computational methods of the finite elements and finite differences
have been employed numerous times in the literature [20, 22, 24, 29, 31, 33, 35, 62, 67]. The advantage of
numerical methods is that there are no limitation in model parameters except the limitations in computation
time; depending on the size of the problem computation can be costly.
3 Dielectric mixtures

Factors effecting the effective properties of a mixture can be listed as follows in a first approximation.

- Intrinsic electrical properties of the constituents, \( \varepsilon_r \), ohmic conductivity \( \sigma_0 \) and dielectric or conductive relaxation processes represented with complex susceptibility \( \chi^* \).
- Concentration of phase.
- Distribution of phases; topology of the mixture.
- Chemical inter-phase between constituents.

Fornes and Paul [88] summarized the issues related to some of the discrepancies between theory and experiments from the mechanical aspect of mixtures, however, they are valid for electrical properties as well. As mentioned the theories consider uniform shape and constant dimensions. While in reality we have polydispersed particles with distribution of sizes/lengths/thicknesses. Inclusions are often not unidirectional. Interface between the phases are considered perfect in mixture models, however in reality the interface is not perfect and chemical techniques are employed to improve the interface, such as with employment of surfactants; none is included in the model calculations. The intrinsic properties of the materials are assumed to be isotropic and linear, however in reality these assumptions can not be true. In polymer matrix composites the properties of the polymer are assumed to be isotropic, however, depending on the length scale there will be local changes in the density, crystallinity and chain orientation. Most of the theories do not consider interaction between inclusion particles and particle agglomeration, it is well-known that special care must be given to produce well-dispersed particles in fabricated composites.

4 Generalization of effective medium approaches

Some generalization efforts for the effective medium approaches previously proposed [20, 89]. In these approaches the effective dielectric permittivity of the mixture \( \varepsilon_e \) is expressed as

\[
\frac{\varepsilon_e - \varepsilon_1}{\varepsilon_e + (n-1)\varepsilon_1} = q \frac{\varepsilon_2 - \varepsilon_1}{\varepsilon_2 + (n-1)\varepsilon_1}
\]

(1)

where, subscripts ‘\( e \)’, ‘\( 1 \)’ and ‘\( 2 \)’ denote the effective, phase 1 and phase 2; the concentration of phase 2 is represented with \( q \); \( n \) is the shape factor. This expression reduces to the Wiener bounds with \( n = 1 \) and \( n = \infty \) for parallel laminates and perpendicular laminates to electric field. The relationship between various shapes and \( n \) are shown in Fig. 3. Spheres and cylinders yield \( n = 3 \) and \( n = 2 \), respectively. The one thing that is important with this expression is that one can obtain the similar relative effective permittivity \( \varepsilon_e \) with different combination of \( n \) and \( q \), which is represented in Fig. 4. It is clear that for design purposes one have the option to select specific filler (permittivity of phase), concentration and shape of inclusion phase to optimize for the required permittivity value. For example for large discrepancy in permittivities the region of interest has a larger area, see bottom right graph in Fig. 4. If the relative effective permittivity is represented as the scaled permittivity \( \xi \) [12, 23, 26],

\[
\xi = \frac{\varepsilon_e - \varepsilon_1}{\varepsilon_2 - \varepsilon_1},
\]

(2)

then the relationship between the ratio of permittivities and the concentration for a constant shape factor can be visualized as in Fig. 5. The boundaries in Fig. 5 indicate the highest and the lowest scaled permittivities that can be achieved with a given set of materials. For large permittivity discrepancies the shape of the curve is like a rotated ‘L’. Observe that we have not considered particle interaction up to this point, therefore the representation at high filler concentrations would not be accurate.

When frequency dependent properties of constituents are considered the problem is a little bit complicated due to the inter-facial polarization and dispersion coupling between phases, see Tuncer [90] for details. It has been shown that the representation of the dielectric data in effective complex resistivity level \( (\rho_e^* = (\omega\varepsilon_0\varepsilon_e)^{-1}) \), the bounds can be observed clearly, unlike in the effective complex permittivity.
5 Spectral density representation

To go a little bit further with the modeling of mixtures, the spectral density representation will be discussed here. Let us assume we have several different shapes of inclusions in the system with known shape parameters \( n \), then by using a summation one can add the contributions of these entities to the overall effective permittivity. In a distribution of shapes similar approach would be derived with a distribution of shape factors. Using the scaled permittivity representation, the scaled permittivity \( \xi \) is then expressed as

\[
\xi = \xi_p + \int_0^1 g(x)(1 + \varpi x)^{-1}dx.
\]

Here \( x \) and \( g(x) \) are the spectral parameter and the distribution of spectral parameters; \( \varpi \) is the spectral frequency; \( \xi_p \) is the concentration of phase 2 that is not contributing to polarization, in other words the percolating strength or the infinite clusters. The function \( g(x) \) contains the information related to shape and their concentration so,

\[
\xi_n = \int g(x)dx.
\]

The total concentration of phase 2 for example will be the sum of \( \xi_p \) and \( \xi_n \), which is the non-percolating portion of the materials; the finite clusters. The most expected spectral parameter \( \bar{x} \) (shape) can be estimated from \( g(x) \) as well,

\[
\bar{x} = \int xg(x)dx.
\]

Examples of application of the method and resolving different distributions were shown in the literature. In some cases the distribution of spectral parameters were resolved with individual peaks indicating a discrete structure. Application of the method as a micro-structural analysis was proposed which might be a good possibility to electrically characterize composites. However, one needs dielectric property information of the system in advance to perform the analysis, as shown in Fig. 1.

6 Resistor-capacitor network and its spectral density

To better illustrate the strength of the spectral density analysis results from an resistor-capacitor circuit modeling were studied. For system and modeling information, please refer to Hamou et al. In the analysis different sizes of circuits composed of 50-50 resistor and capacitor occupancy were simulated. The sizes of the circuits were \( 2^j \times 2^j \) with \( j = \{ 4, 5, 6, 7, 8, 9 \} \). Example of a \( 3 \times 3 \) circuit is shown in Fig. 6. The dielectric relaxation of the system with randomly arranged resistors and capacitors show very different responses depending on the backbone and percolation paths. The responses from several circuit cases studied with \( 512 \times 512 \) lattice size are shown in Fig. 7. Depending on the structure of the systems, some indicate percolation due to the lining up resistors and the others had broad relaxations; none of the cases were similar.

When we apply the spectral density representation we can see that the randomness and the structure of the spectral densities with respect to considered lattice sizes, Fig. 8 First the largest lattice considered had the broadest distribution of spectral parameters. This is expected due to the size of the system and the possibilities of generating intricate shapes. While the smallest lattice structure had a narrow distribution. The other striking observation is for large spectral parameters \( x \) the structure spectral densities of each set of lattice coincide for the studied cases. While for small spectral parameters we see differences. This is mainly due to the long range order and increasing size of the lattice generates slight differences in the long range correlations. The short range order is not influenced meaning that the arrangement of the resistors and capacitors would not change the structural information when we consider small lengths scales. One way to apply this observation to dielectric breakdown is in order to fail a material charge species need to
correlated with long range order to initiate the breakdown. One interesting example is the electrical treeing where no breakdown is observed until the tree bridges between two electrodes. Taking this example further for those cases where the long range correlation between resistors exists there is a risk to fail the material. Therefore one needs to study the spectral density of the system to determine the existence of any long range correlations. Perhaps it sounds impossible for today's material characterization techniques, but once a library of material properties are built there would be possibilities to study the structure-property relationships in engineering composites.

7 Bounds on dielectric permittivity: circles of interest

Before we finish our topic on the dielectric mixtures, I would like to point our attention towards an important analysis technique for predicting or guessing dielectric properties of composites. Milton [2, 97] has illustrated a graphical technique to study dielectric mixtures. The technique is based on the complex dielectric properties of mixture components. Once an observer gathers information on the composite system it becomes easy to predict the effective properties of the composite. The method was built as follows. It starts with the largest bounds presented by Wiener [59], Eq. (1), and includes another one presented by Hashin and Shtrikman [98],

\[
A \equiv (1 - q)\varepsilon_1 + q\varepsilon_2
\]

\[
B \equiv ((1 - q)/\varepsilon_1 + q/\varepsilon_2)^{-1}
\]

\[
X_{d_2} \equiv \varepsilon_1 + \frac{dq(\varepsilon_1 - \varepsilon_2)}{d\varepsilon_1 + (1 - q)(\varepsilon_2 - \varepsilon_1)}
\]

\[
Y_{d_2} \equiv \varepsilon_2 + \frac{d(1 - q)\varepsilon_2(\varepsilon_1 - \varepsilon_2)}{d\varepsilon_2 + q(\varepsilon_1 - \varepsilon_2)}
\]

Here \(d\) is the dimensionality and \(q\) is the concentration of phase 2. Observe that the mixture expression of Hashin and Shtrikman [98] for \(C\) and \(D\) are the same expressions with interchanged phase 1 and phase 2, and \(q\) and \((1 - q)\). Once can similarly use Eq. (1) for this purpose. Once we have the points \(A\) to \(Y\) we can now start the graphical analysis as shown in Fig. 9. So the electrical properties of the constituents \(\varepsilon_1\) and \(\varepsilon_2\) determine the absolute limits; \(\varepsilon_1 A \varepsilon_2\) is the lower bound and \(\varepsilon_1 B \varepsilon_2\) is the upper bound. Concentration information would yield the positions of \(A\) and \(B\), then we can narrow the region of interest for the effective permittivity. If we have information on the shape of the inclusions, let us say spherical, then \(d = 3\), we would get an even narrower region than before marked between \(X_{d_2} Y_{d_2}\). Now we can test our regions for the resistor-capacitor network as shown in Fig. 10 with dots. For each frequency of interest the limits for the effective permittivity is different due to the complex permittivities or we should the impedance. Since we did not have any information on the size and shape of resistors, some of the points are not inside the smallest region \(X_{d_2} Y_{d_2}\). However all the data are located within the region \(AX_{d_2} BY_{d_2}\), which indicates that the resistor-capacitor network simulation results were acceptable.

8 Summary

Different approaches to dielectric mixtures are presented with numerous research work related to the topic. The importance of the dielectric properties to the overall insulation design was previously discussed by Dakin [1]. Here recent approaches and the methods to study dielectric mixtures were shown. No insulation systems were considered because of the number of such systems available. As known by the dielectric mixture community and mentioned by Dakin [1], dielectric behavior in composite materials is a complex phenomenon due to involvement of different physical parameters other then the intrinsic properties of the constituents. However, with novel numerical methods and improved fabrication techniques many of the parameters influence the over all mixture properties could be studied to better control the electrical properties of these systems.
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