

Dielectric Properties of Three Component Mixtures of Nonlinear, Resistive, and Non-conducting Grains

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Abstract: The nonlinear dielectric properties of a three component system consisting of nonlinear resistive, linear resistive, and nonconducting cubes of equal size is studied to determine the range of nonlinear grading properties which can be achieved. The system is modeled on a statistical basis, with confirmation through Monte Carlo simulations. The objective is to gain insight into the range of field grading properties which should be possible as a function of the characteristics of the nonlinear component and the ratio of the three components.

INTRODUCTION

Nonlinear grading materials are used in high voltage applications in order to control stress concentration. Such materials often take the form of filled polymers, i.e., mixed nonlinear filler and resistive filler in a polymer matrix. Thus such materials can be treated as a disordered electrical network constructed from nonlinear, resistive and non-conducting elements. To date, no method has been developed to calculate the resistance of such a random electrical network. However, percolation theory is one of the principal tools for the study of such systems. For any such three-dimension network, (i) when the non-conducting fraction, P_1 , is less than a critical fraction P_c , percolation paths will exist, and (ii) the overall resistance converges as the network size increases to infinity [1]. Based on these two theorems, we can study this problem with Monte Carlo simulations and statistical approaches.

In our study, all components are treated as consisting of equal sized cubes, and the leakage current through non-conducting grains is assumed to be zero. We also assume percolation paths are isolated, i. e., current does not transfer from one percolation path to another. Obviously, this approximation is good over only a limited range of nonconducting fraction. ZnO-based sintered nonlinear metal oxide ceramic is often used as a nonlinear component [2]. As is well known, intergrain boundaries in such material provide a voltage drop of roughly 3V per intergrain boundary. However, we can still treat the nonlinear elements as cubic grains, since the high conductivity of ZnO (100 to 1000 S/m) makes

the voltage drops in the bulk of the ZnO small enough to be ignored.

The objective of this contribution is to demonstrate a method to derive random electrical network properties and investigate the effect of components properties and volume ratios on the overall material performance.

STATISTICAL APPROACH

Percolation Model

Percolation is a mathematical concept of modeling the spread of a fluid through a medium [3]. Suppose we setup our 3-D model with electrodes in the x-y plane and the electric field applied in the $-z$ direction. The medium between the electrodes consists of three components, nonconducting, non-linear and resistive grains with volume fractions P_1 , P_2 , and P_3 , respectively. We treat all grains as being cubes of the same dimension with sides of length a , which is typically in the range of 0.1 to 10 μm [2,4]. The cubes are distributed randomly to form a dense medium. From any starting point, the percolation path (if it exists) is defined as the path of minimum length through which current can pass from one electrode to the other, i.e., which consists of a continuous string of resistive or nonlinear grains between the electrodes. The percolation number is the number of grains in the percolation path. The minimum percolation number, L , is given by:

$$L = T / a \quad (1)$$

where T is the distance between electrodes.

Monte Carlo Simulations

We assume statistical independence of the components and construct the percolation path based on volume fractions. A program was written in QBASIC to simulate the situation. Grain types are determined by a random number according to their volume fractions. Percolation paths start from every conducting grain on the positive electrode surface. Paths are extended through non-insulating grains or blocked if no non-insulating path is available. We attempt to extend path in the field ($-z$) direction. If that is not possible, then the path is extended in the x or y direction (sideways). If

all paths are blocked, we move backward one grain and attempt to extend the path from that location. When a connection between two electrodes is created, the path is completed. If the path goes back to the starting surface, the path is considered incomplete. Even though we cannot guarantee paths constructed in this way are the shortest possible, they should be close, since we make extension in the direction of the electric field the first priority. Simulation results are shown in Figures 1-3.

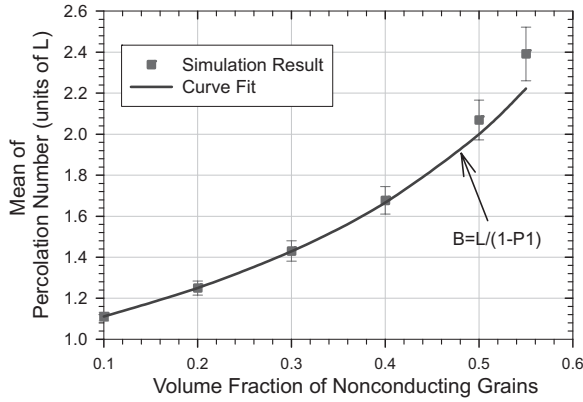


Figure 1. Mean of the percolation number vs. volume fraction of nonconducting grains. One thousand tests were performed for each $P1$ value, from which the mean number and the $\pm 2\sigma$ confidence limits are calculated.

To a good approximation, the percolation number is normally distributed with mean B and standard deviation σl given by the following formulas. K is the fraction of completed paths.

$$B = \frac{L}{1 - P1} \quad (2)$$

$$\sigma l = \sqrt{\frac{B \cdot P1}{1 - P1}} = \sqrt{\frac{L \cdot P1}{(1 - P1)^2}} \quad (3)$$

$$K = 1 - \frac{P1^5}{0.23} \quad (4)$$

Since percolation paths are constructed with resistive and nonlinear grains only, the number of nonlinear grains within one percolation path obeys the binomial distribution with mean, μ , and standard deviation, σ , given by [5]

$$\mu = n p \quad (5)$$

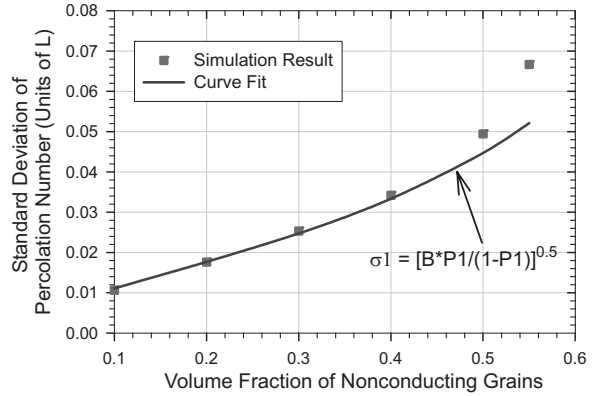


Figure 2. Standard deviation of the percolation number vs. volume fraction of nonconducting grains.

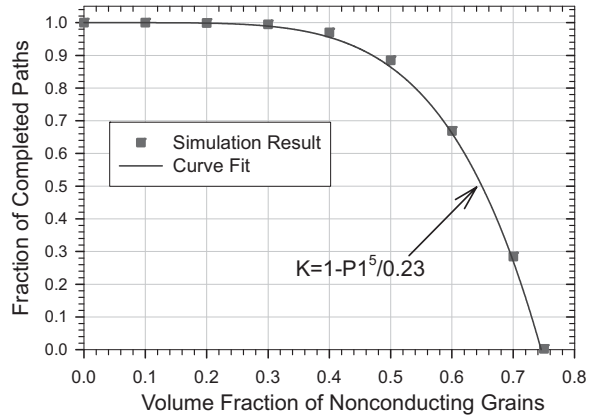


Figure 3. Fraction of complete paths vs. volume fraction of nonconducting grains. From this plot, we can see critical fraction P_c is about 0.75.

$$\sigma = \sqrt{n p (1 - p)} \quad (6)$$

where n is the percolation number, which is typically in the thousands, and p is the probability of a nonlinear grain, which is given by $P2/(1-P1)$. To a good approximation, we could use the mean value of the percolation number for every percolation path, in which case, $N1$ and $N2$, the number of nonlinear grains and resistive grains are given by

$$N1 = N \cdot P2 / (1 - P1) \quad (7)$$

$$N2 = N - N1 \quad (8)$$

where N is the mean percolation number. Monte Carlo simulations indicate that the effect of the binomial distribution on current-voltage (I-V) characteristics is negligible.

Computation of I-V Characteristics

For many applications, a nonlinear grading material should have high dielectric constant and low loss during normal operation at power frequency, and a field-dependent conductivity under impulse conditions. We adopt the following equation for the nonlinear grains [4]:

$$J(V) = 10^{\frac{18}{13} + 4.5 \tanh[50 \log_{10}(V+0.7) - 28] + \frac{10}{9} \log_{10}(V+0.7)} \quad (9)$$

where J is the current density and V is the voltage applied to the grain. This function is plotted in Figure 4.

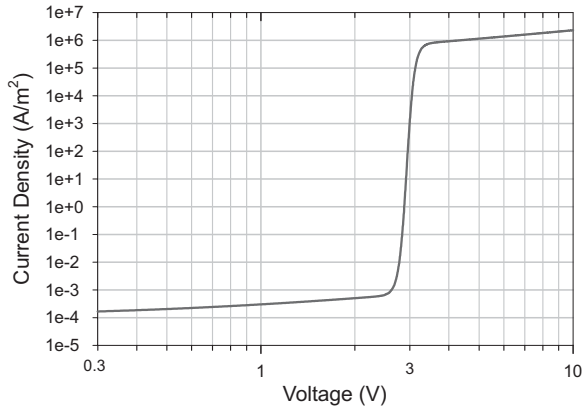


Figure 4. J-V characteristics assumed for nonlinear grains.

The total number of percolation paths, TP , is given by the number of conducting grains under the electrode surface times the pass rate K :

$$TP = A \cdot K / a^2 \quad (10)$$

where A is the electrode area. Nearly all possible path lengths are included in the range between $B-5\sigma l$ and $B+5\sigma l$. For the number of percolation paths of each percolation number, we have:

$$i = [1, 2, \dots, 10\sigma l + 1] \quad (11)$$

$$N_i = B - 5\sigma l + i - 1 \quad (12)$$

$$TN_i = TP \cdot PN_i \quad (13)$$

where N_i , PN_i and TN_i are the number of elements in path i , the probability density for each path number, and the number of path number N_i . Obviously,

$$\sum_{i=1}^{10\sigma l} PN_i = 1 \quad (14)$$

For a given percolation path, the current through the resistive and nonlinear grains must be the same. Thus

$$V_{tot} - N1_i \cdot VB - N2_i \cdot J(VB) \cdot a / \sigma 2 = 0 \quad (15)$$

where V_{tot} , VB and $\sigma 2$ are the applied voltage on the sample, the voltage drop on each nonlinear grain, and the conductivity of the resistive grains, respectively. By substituting Eq. (9) into Eq. (15), VB can be solved numerically for a fixed grain size a [4]. Then the overall resistive property of the sample, i.e., the I-V characteristics, or the conductivity as a function of the electric field, can be computed.

Discussion

If the applied voltage is less than the voltage drop across the nonlinear grains, the current in the percolation path is negligible. Above the voltage which drives the nonlinear grains into conduction, the voltage drop across the nonlinear grains is always about 3V per grain. According to Eqs. (2)-(8), the transition field can be calculated as

$$E_{trans} = \frac{B}{T} \cdot \frac{P2}{1-P1} \cdot 3V = \frac{P2}{a(1-P1)^2} \cdot 3V \quad (16)$$

The voltage drop across the nonlinear grains remains approximately constant at 3 V/grain above this transition field. The remaining voltage is applied to the resistive grains in the percolation paths. Thus the voltage across the resistive grains is given by

$$V_2 = V - V_{trans} = V - E_{trans} \cdot a \cdot B \cdot \frac{P2}{1-P1} \quad (17)$$

From Eq. (16) and (17), we can see that the number of nonlinear grains and the conductivity of the resistive grains together, determine the nonlinear characteristics of the material.

A typical grading material might be capacitively graded at power frequency but resistively graded under lightning impulse conditions. This means that the transition from capacitive to resistive grading should be in the range of 4.5 kV/mm. To design such a material, we start from a particle size of 1 μm and compute the characteristics for a 1 mm thick sample. Since we want a rapid change in conductivity at about 4.5 kV/mm, we need a mean of about 1400 nonlinear grains in the percolation path. If we assume that we want about 50% nonconducting (polymer) grains to provide adequate binding material and flexibility, then From Eq. (16), we know the transition field will be in the range of 4.5

kV/mm if we have 35% nonlinear grains, which leaves 15% resistive grains. The properties of such a material are plotted in Figure 5 for two values of resistive grain conductivity, σ_2 , viz., 0.01 and 1 S/m. Material conductivity vs. field is plotted in Figure 6, with σ_2 of 1 S/m. The conductivity of the resistive grains determines the high field conductivity of the material. This conductivity must be sufficiently high that $\sigma \gg \epsilon\omega$ for a lightning impulse.

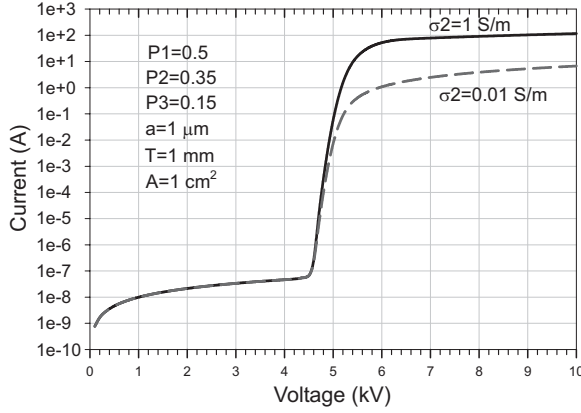


Figure 5. Computed I-V characteristics, with a transition field of 5 kV/mm. Other sample parameters are listed in the plot.

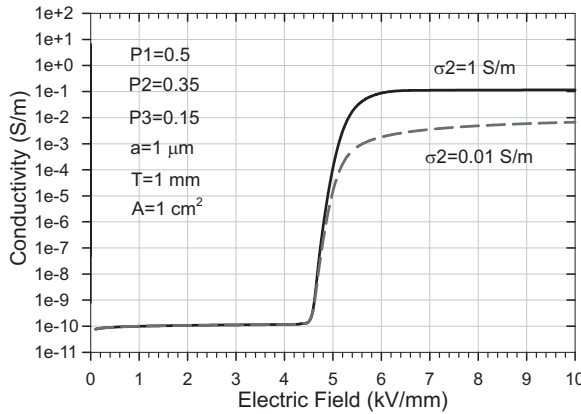


Figure 6. Computed conductivity vs. field. Other sample parameters are listed in the plot.

We can derive a formula for the high field limit of the conductivity based on the assumption that the voltage drop across the nonlinear grains is negligible. In this limit, the conductivity is given by

$$\sigma = \sigma_2 \left[\frac{K(1-P_1)^3}{P_3} \right] \quad (18)$$

This equation does not predict the characteristics of Figure 6 very well, as the voltage drop across the

nonlinear grains is not negligible, and the voltage drop continues to increase with field above the transition region (Figure 4). The latter effect is quite significant.

Suppose the relative dielectric constant for nonconducting grains is 2.3, and 300 for the nonlinear grains. To first order, the effect of cubic resistive grains, which will polarize fully at any reasonable frequency, can be treated as reducing the effective thickness of the sample. Hence the effective dielectric constant of the material should be

$$\epsilon_{rel} \approx (2.3 \cdot 0.5 + 300 \cdot 0.35) / 0.85 = 124.88 \quad (19)$$

so that $\epsilon\omega$ is of order of 10^{-7} S/m at power frequency and about 10^{-3} S/m for a lightning impulse (300 kHz). Thus this would be a good capacitive grading material at low field, and, according to Figure 6, would become resistive grading at about 5 kV/mm ($\sigma \gg \epsilon\omega$) for the case of 1 S/m resistive grain conductivity, well above the power frequency operating field for a termination but within the range of a lightning impulse. A resistive grain conductivity of 0.01 S/m would not be sufficiently high to provide the required high field conductivity for effective nonlinear resistive grading under impulse conditions.

CONCLUSIONS

The electrical characteristics of a random network can be predicted, computed, and engineered to provide effective grading materials based on knowledge of the component dielectric properties. Such knowledge is important in designing grading materials, as it provides the ability to estimate the grading material properties required for transient nonlinear finite element analysis of devices, such as terminations, which employ such materials.

REFERENCES

1. G. Grimmett, *Percolation*, 2nd Ed., Vol. 321, Springer, pp. 380-382, 1999.
2. M. Ellsworth, "Synthesis of Multi-Component Nanophase Particles and Their Use as Functional Fillers", Workshop on Organic/Inorganic Hybrid Materials, The Division of Polymer Chemistry, American Chemical Society, Sonoma County, CA, 14-17 November 2001.
3. R. Smythe and J. Wierman, *First-Passage Percolation on the Square Lattice*, Springer-Verlag, pp. 1, 1978
4. S. Boggs and H. Andoh, "A statistical Approach to Prediction of ZnO Arrester Element Characteristics", IEEE Transactions On Power Delivery, Vol. 16, No. 4, October, 2001.
5. R. Lyman Ott, *An Introduction to Statistical Methods and Data Analysis*, 4th Ed., pp. 153, Wadsworth, Inc. 1993.