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Conductive Nanoparticles in Dielectrics: A Comparative Study

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Abstract— The Maxwell-Garnett method is used to predict the effective dielectric constant and the tangent loss of various composites consisting of a PVDF-TrFE-CFE-matrix and conductive microsphere fillers made of Cu, Ni, W, Zn, or Fe. Simulation results demonstrate that for small filler fraction values and at low frequencies, the electrical properties of the resulting composite do not depend on the conductivity of the filler. These findings show that composites fabricated using cheaper metal nanoparticle fillers are as effective as those fabricated using expensive ones.

Keywords— Composite materials, conductive nanoparticles, Maxwell-Garnett mixing rule.

I. INTRODUCTION

A capacitor's energy storage capacity depends on its size as well as the dielectric permittivity of the spacer/filling material used [1-5]. In recent years, due to the advancements in fabrication technologies, the size of electronic systems and their individual components have significantly shrunk. Consequently, to obtain adequate levels of energy storage, materials with high permittivity values, commonly referred to as high K, have to be used [2-6]. One way of fabricating such materials is through reinforcing dielectrics, such as polymers, with highly conductive metallic fillers (Fig.1). In addition to high permittivity, these composites have several desired mechanical properties, which has led to their popular use in embedded circuits, e.g. in flexible capacitors [6-11].

In this work, electrical properties of metal-dielectric composites are predicted using the Maxwell-Garnett method [12]. More specifically, the effective dielectric constant and the tangent loss of various composites consisting of a PVDF-TrFE-CFE-matrix and conductive microsphere fillers made of Cu, Ni, W, Zn, or Fe are computed in the dilute regime (before percolation regime start). Comparison of the predicted values show that at low frequencies, electrical properties of the composites are same for different type fillers as long as their shape and size and the filler fraction of the mixing are the same. In other words, under these conditions, the conductivity of the fillers does not have any effect on the permittivity and tangent loss of the resulting composite.

These findings might help with fabrication procedures in determining the “best” type of conductive filler when other factors such as the cost of metals need to be considered.

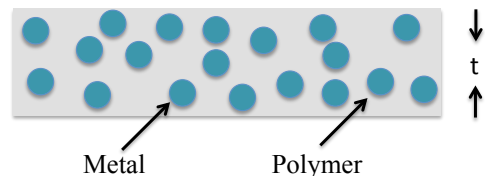


Fig. 1. Representation of a polymer composite with spherical fillers.

II. METHOD AND MATERIALS

The permittivity of a dielectric can be mathematically modeled by [13]

$$\epsilon_e = \epsilon_{\infty,e} + \frac{\epsilon_{s,e} - \epsilon_{\infty,e}}{(1 + (j\omega\tau)^{1-\alpha})^\beta} \quad (1)$$

where $\epsilon_{s,e}$ and $\epsilon_{\infty,e}$ are static and high frequency (optical) relative permittivities, α and β are Cole-Cole and Cole-Davidson constants, and τ is the relaxation time. Similarly, the permittivity of a conductor is given by [13]

$$\epsilon_i = \epsilon_{\infty,i} + \frac{\sigma}{j\omega\epsilon_0} \quad (2)$$

where $\epsilon_{\infty,i}$ is the optical permittivity (for “perfect” metals, such as Copper, $\epsilon_{\infty,i} = 1$), σ is the conductivity, and $\epsilon_0 = 8.85 \times 10^{-12}$ F/m is the permittivity of the vacuum. At low frequencies, the imaginary part of the permittivity is dominant.

The Maxwell-Garnett formula is used to obtain an analytical expression for the effective permittivity of a composite with a dielectric matrix and spherical conductive fillers:

$$\epsilon_{eff} = \epsilon_e + 3\epsilon_e \left(\frac{FF(\epsilon_i - \epsilon_e)}{\epsilon_i + 2\epsilon_e - FF(\epsilon_i - \epsilon_e)} \right) \quad (3)$$

Here, ϵ_e [see (1)] and ϵ_i [see (2)] are permittivities of matrix/host and filler materials, and FF is the volume fraction of the fillers to the overall volume of the composite.

III. RESULTS AND DISCUSSION

The parameters used in (1) for the PVDF-TrFE-CFE matrix are $\alpha = 0.45$, $\beta = 0$, $\epsilon_{s,e} = 50$, $\epsilon_{\infty,e} = 4$, and $\tau = 1.75 \times 10^{-6}$ s and the conductivities of the filler microspheres made of Cu, W, Zn, Ni, or Fe are given in Table I. Diameters of all fillers are 800 nm.

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TABLE I. CONDUCTIVITIES OF FILLER MATERIALS

Filler	Cu	W	Zn	Ni	Fe
$\sigma[MS/m]$	59.6	17.9	16.9	14.3	10.1

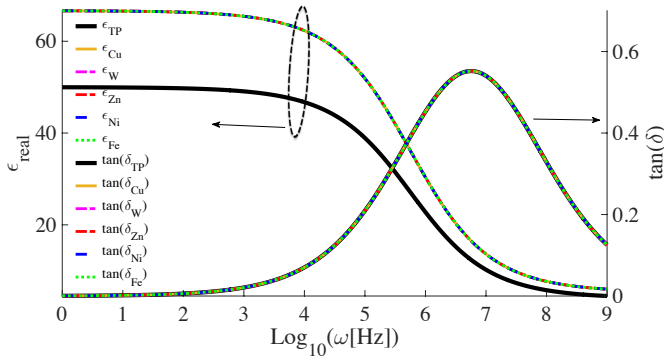


Fig. 2. Complex permittivity of the composites with different type fillers within the frequency band 1Hz and 1GHz. Filler fraction FF=0.10

For the first simulation, filler fraction FF=0.10 while the frequency is varied from 1 Hz to 1 GHz. The effective permittivity of the composites is computed using (3) at every frequency sample. Fig. 2 plots the real part of the permittivity and the tangent loss of PVDF-TrFE-CFE and each composite versus frequency. The results clearly show that the conductivity of the metal fillers does not have any effect on the electrical properties of the resulting composite for a rather broadband of frequencies. This result can be explained as follows. The permittivity of the metallic fillers is significantly larger that of the polymer ($|\epsilon_i / \epsilon_e| \gg 1$). Consequently, the ratio $|\epsilon_i / \epsilon_e|$ used in the Maxwell-Garnett rule (3) dominates all other terms when the frequency is low enough. This makes the effective permittivity a function of only the filling fraction FF.

For the second simulation, frequency is kept constant at 10 KHz while filler fraction FF is varied from 0 to 0.20. Fig. 3 plots the real part of the permittivity and the tangent loss of each composite versus filler factor FF. These results also agree with the explanation above. Effective electrical properties of the composites vary only with the filler fraction and do not depend on the conductivity of the fillers. The results in Figs. 2 and 3 suggest that Fe can effectively replace (the more expensive) Cu as a filler without affecting the electrical properties of the resulting composite even though the conductivity of Cu is six times greater.

IV. CONCLUSION

The effect of metal filler's conductivity on the electrical properties of polymer-metal composites is numerically characterized using the Maxwell-Garnett method. Simulation

results show that, for small values of filler fraction and at low frequencies, electrical properties of the polymer-metal composites do not depend on the conductivity of metal filler.

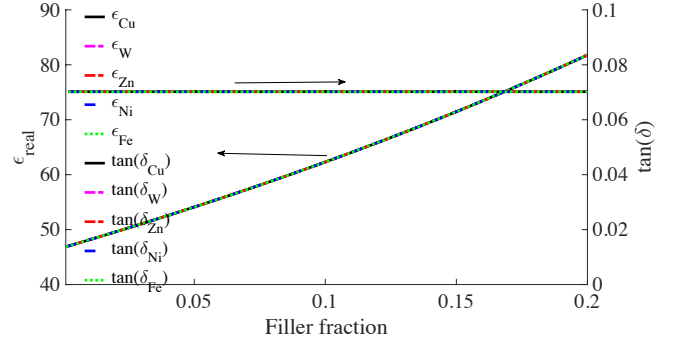


Fig.3. Complex permittivity of the composites for filler fraction changing between 0 and 0.20 at 10kHz.

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