Tuning of dielectric constant of DGEBA polymer using graphite powder

Tony Joseph[1]*, Ambily Krishnan*[1]*, Anlin Lazar K*[2]
[1] *Dept. of Physics, Govt. Victoria College, Palakkad*
[2] *Dept. of Physics, Mercy College, Palakkad*

Abstract

Epoxy resin based on diglycidyl ether of bisphenol A (DGEBA), a widely used thermosetting polymer, was employed as the matrix material, while graphite powder served as the filler in the fabrication of epoxy–graphite composites. The composites were prepared at room temperature using a moulding technique. Key physical properties, including density, moisture absorption (wt%), and thermal conductivity, were evaluated. Dielectric properties were measured using an LCR meter for graphite powder loadings ranging from 0 to 0.50 vol%. Additionally, attempts were made to predict the dielectric behavior of the composites using established theoretical models. The microstructural characteristics of the composites were examined by scanning electron microscopy (SEM).

1 Introduction

During the past decade, there has been an avalanche of new materials suitable for microelectronic and wireless communication industry. The most important properties of materials useful for electronic and wireless communication industry are dielectric constant (εr), dielectric loss (tan δ), thermal conductivity etc. Low dielectric constant materials are used in substrate applications, whereas high dielectric constant materials can be used for capacitor applications. Several researchers have contributed to the theoretical and experimental studies on the critical behavior of metal-insulator composites near the percolation threshold (Doyle ,1990; Ambegaokar, 1971; Sheng 1978). Percolation is result of the formation of continuous network of conducting phase that runs throughout the polymer matrix. In terms of concentration of the conductive filler (f), percolation threshold concentration (fc) and relative

◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇◇

Received : 09 October 2025
Revised : 01 November 2025
Accepted : 30 November 2025

permittivity of the polymer matrix (εD), the effective permittivity (ε) of polymer-graphite composites can be expressed as

$$\underline{\varepsilon} = \varepsilon_D (f_c - f)^{-s} \qquad (1)$$

Here s is a scaling factor related to the microstructure and connectivity of the insulator-conductive filler interface (Grannan, 1981). In a polymer-metal composites, the ease of processibility of polymers can be combined with the high electrical conductivity of conducting phase. In this study we use a low-cost thermosetting polymer named Epoxy (DGEBA) as polymer matrix and graphite power as the conducting filler phase. Graphite has good electrical conductivity, thermal conductivity and low density. Graphite has unique atomic structure, which has delocalized electrons that are free to move. Each carbon atom in graphite is bonded to three other carbon atoms, leaving one electron per atom to be shared and move freely within the hexagonal layers of the material. (Bigg, 1986)

2 Experimental

Different volume fractions ($v_f = 0$–0.5) of graphite powder were added and mixed mechanically with epoxy resin using an agate mortar and pestle. The curing agent added was in the ratio 5:1. The thoroughly mixed epoxy and graphite powder were poured into moulds of different shapes and then kept at room temperature for 12 h in a desiccator. We use the following equation for the calculation of the theoretical density (ρ_c) of the polymer-graphite composite

$$\rho_c = v_f \rho_f + (1 - v_f)\rho_m \qquad (2)$$

where, ρ_c is the density of composite, ρ_f is the density of Graphite (2.1 g/cm^3) and ρ_m is the density of epoxy (1.15 g/cm^3). v_f represents the volume fraction of graphite dispersed in the epoxy matrix. The actual density values of the samples were measured by Archimedes method. The disc shaped samples were made into parallel plate capacitors with the help of silver paste and copper leads. An LCR meter (Hioki 3532-50, Japan) was used to compute dielectric properties and electrical conductivity. Moisture absorption of the samples was obtained by measuring the increase in weight of the samples after keeping them in distilled water for 24 hours. The microstructure of the samples was analyzed using scanning electron microscopy and thermal conductivity of the composites was measured using photopyroelectric technique (Menon, 2000; Marinelli, 1990).

3 Results and Discussion

3.1. Density

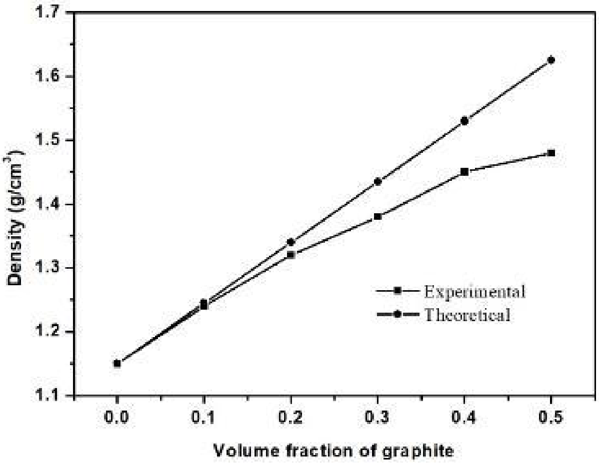

Fig. 1. Variation of density of epoxy-graphite composites as a function of volume fraction of filler.

Fig. 1 shows the experimental and theoretical values of density of the polymer-graphite composites from vf = 0 to vf = 0.5. It is observed that the density increases with the increase in filler content. This is because of the fact that graphite has higher density compared to epoxy. It is found that the experimental density matches with the theoretical density values only in the range 0-0.2vf of filler concentrations. The clustering of graphite particles at higher filler loadings significantly affects the dielectric properties of the composite (Doyle 1990). The mechanical mixing method used in this study is not very effective to form a homogenous mixture, especially when vf is more than 0.2 because of agglomeration of graphite powder and formation of porosity. Higher the filler loading, the more dominant is the effect of agglomeration and porosity. In the calculation of theoretical density, the effect of porosity is not taken into account. For low volume fraction of graphite loading, the epoxy matrix completely covers the graphite particles and hence porosity will be less. However, as the amount of graphite increases beyond 0.2 vf, the packing will be less effective and hence porosity will be more.

3.2. Moisture absorption

Fig. 2 shows the variation of moisture absorption (in weight percentage) and porosity content (%) of the composites as a function of graphite powder concentration. It is found that moisture absorption and porosity increase with the concentration of the conducting phase. The composite properties are highly sensitive to humidity because of the polar nature of water molecules. Hence the values of dielectric constant (εr) and dielectric loss (tan δ) of the composite increases with moisture absorption (Sebastian, 2008; Gonon, 2005). Mechanical strength of the composite materials also adversely affected by moisture absorption. Hence, moisture absorption is an important parameter to be determined prior to practical applications and the amount of filler loading has to be carefully optimized to get the best results.

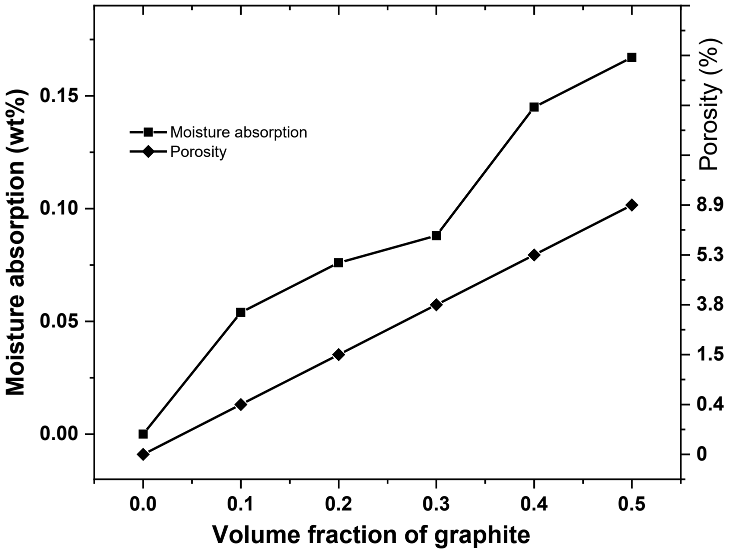

Fig. 2. Variation of moisture absorption and porosity of epoxy-graphite composites as a function.

3.2. Microstructure

Fig. 3 (a) and (b) shows the SEM images epoxy-graphite composites for volume fraction of 0.2 and 0.4 respectively. Irregularly shaped particles with size in the range 1-5 μm are visible in the figures. For a filler loading of 0.2, the particles are well separated, whereas they are much more crowded in the samples loaded with 0.4 volume fraction of the filler. Occasional agglomeration can be observed even for low filler loading (Fig. 3(a)). This is attributed to the lack of effectiveness of mechanical mixing. As the filler content increases, the inter-particle distance decreases, which in turn increases the connectivity among the graphite particles. The aggregation of the graphite particles with the increase in volume fraction in the polymer matrix also results in the formation of porosity.

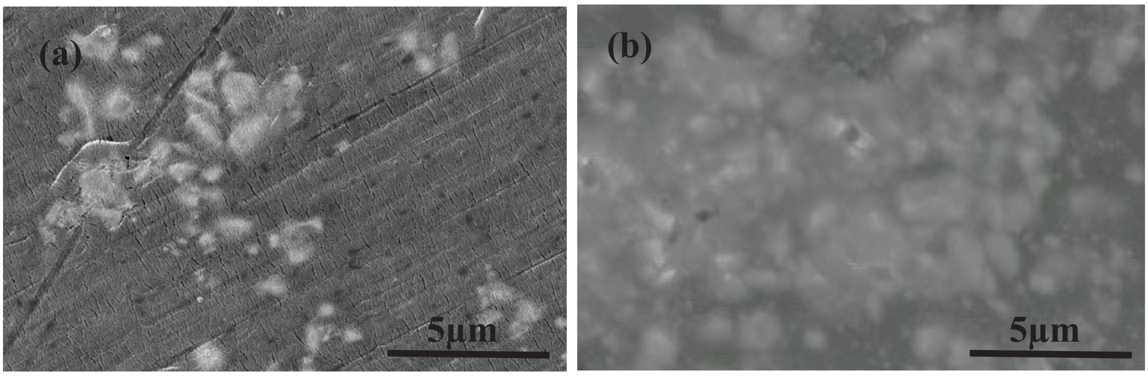

Fig. 3 SEM images of Epoxy-graphite composites for filler loadings (a) 0.2 volume fraction (b) 0.4 volume fraction

3.4. Dielectric properties

The variation of dielectric constant at 10 kHz and 1 MHz of epoxy-graphite composites as a function of different volume percentage of graphite is shown in Fig. 4. Dielectric constant increases from 4.7 for pure epoxy with the addition of graphite and reaches a maximum value of 47.2 at 0.50 volume fraction of graphite at 10 kHz. Similarly at 1 MHz, the variation is from 4.3 to 38.2. Percolation was not observed in the composition range under study. The percolation threshold may possibly be observed at some concentration greater than 0.50 volume fraction of graphite. This value is much higher than the theoretical prediction of percolation for polymer-metal composites. The dielectric constant enhancement at lower concentration is attributed to interfacial polarization. It is also referred to as the Maxwell-Wagner-Sillars (MWS) effect or polarization, (Dat, 1996;Song, 1986) a phenomenon that appears in heterogeneous media consisting of phases with different dielectric constant and conductivity, due to the accumulation of charges at the interfaces. When the graphite content is low, the graphite particles are isolated and well separated such that there is no interaction between them. As the graphite content is enhanced, the number of particles per unit volume and its interfacial area with the polymer increases. This in turn enhances the average polarization associated with the particles and provides greater contribution to the dielectric constant of composite.

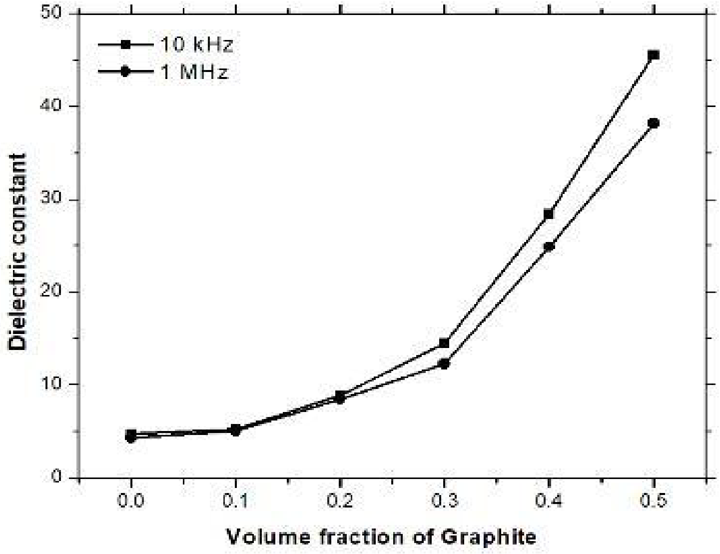

Fig. 4. Variation of dielectric constant as a function of volume fraction of graphite

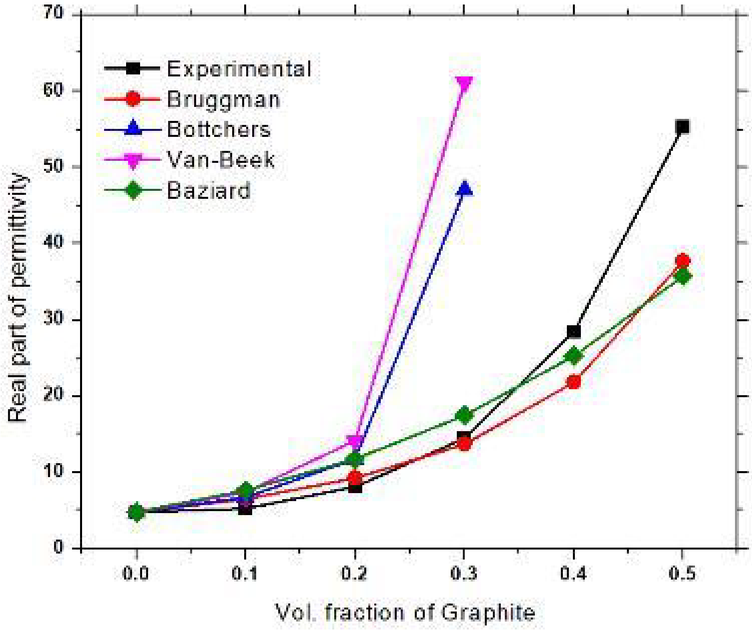

Fig. 5. Comparison of dielectric constant (at 10 kHz) for epoxy-graphite composites with theoretical models

Fig.5 compares the experimental values of dielectric constant of epoxy-graphite composites with some theoretical predictions of the composite materials. The models used are (Baziard, 1988, Ambegaokar, 1971, Sebastian, 2008),

Bruggman's formula:$\varepsilon^* = [\varepsilon 2/(1-v1)3]$ (3)

Bottcher's formula: $\varepsilon^* = \varepsilon 2/(1 - 3v1)$ (4)

VanBeek's formula: $\varepsilon^* = \{[\varepsilon 2(1 + v1)]/(1 - 4v1)\}$ (5)

Bizard's formula, $\varepsilon^* = \varepsilon 2(1+v1)5$ (6)

where ε*,ε1, and ε2 represent the dielectric constant of the composite, the dispersed phase, and the matrix, respectively; v1 denotes the volume fraction of graphite, and v2 denotes the volume fraction of the polymer. The equations (3-6) were derived by taking the ideal case, that is the relative permittivity of the conducting phase as infinite. This assumption is not completely valid as the electrical conductivity of graphite is not as good as metals. Also, theoretical equations are usually framed by assuming that the filler particles are spherical in shape and are well separated. These assumptions are also weak, especially at higher filler loading. This could be the reason for discrepancy between the experimental and theoretical values.

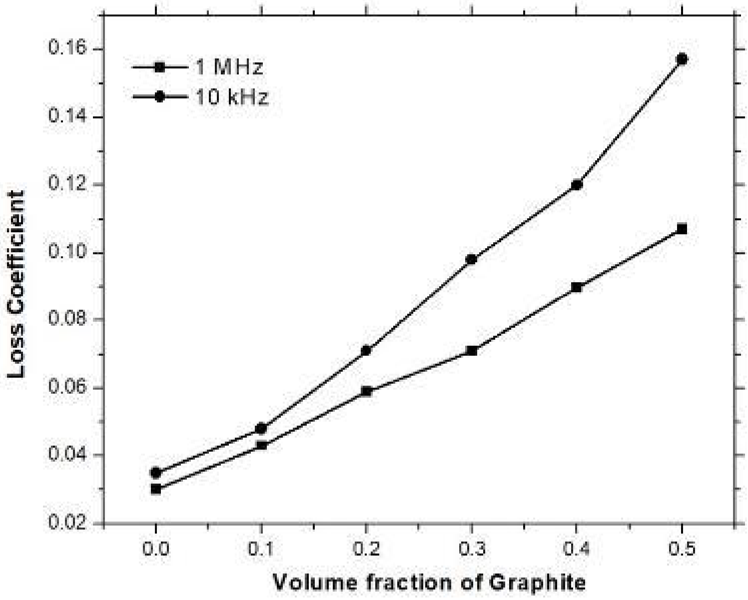

Fig.6. Variation of Loss Coefficient as a function of composition

Fig.6 illustrates the variation of dissipation factor as a function of volume content of graphite for 10 kHz and 1 MHz. It can be seen from this figure that as more filler is added to the matrix, the dissipation factor, in general, shows an increase from 10-2 to 10-1. The increase in dissipation factor with increase in filler concentration, which is also shown by several other conductor–insulator systems, is considered as a consequence of interfacial polarization (Sebastian, 2008).

3.4. Electrical conductivity and thermal conductivity

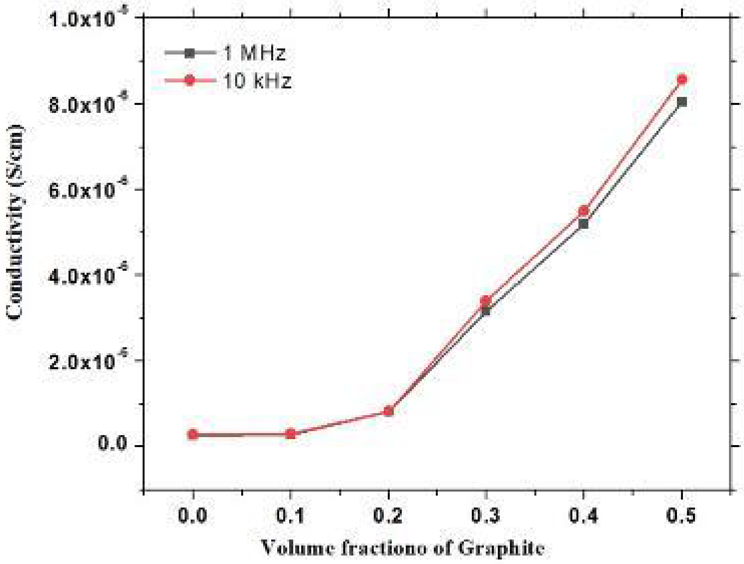

Fig 7. Variation of conductivity as function of filler concentration

Fig. 7 shows the variation of conductivity of the composite as a function of filler loading. Graphite particles are well separated at lower concentrations and hence the electronic wave functions are localized within the grains of graphite. At higher concentration of graphite, the distance between some of the randomly distributed particles gets reduced in such a way that the energy difference will be small enough to delocalize the electron waves and thus promotes tunneling of electrons between the conducting grains of filler particles. (Sheng, 1973; Toker, 2003) The probability of electron tunneling increases with increase in the volume fraction of filler. Fig. 8 shows the variation of thermal conductivity of the composite with volume fraction of the filler. Epoxy, being a polymer has low thermal conductivity values (Zhou, 2024), which causes accumulation of heat in the electronic components, leading to reduced life span. The thermal conductivity can be considerably increased by the inclusion of graphite powder in the epoxy matrix. The observed values of thermal conductivity increased from 0.2 W/mK for pure epoxy to 9.8 W/mK for a filler volume fraction of 0.5. This is in accordance with previously reported results (Park, 2004)

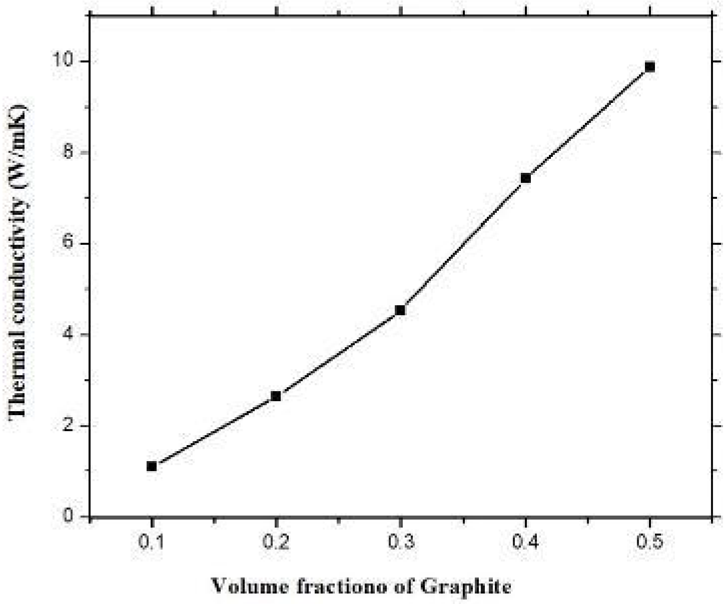

Fig 8. Variation of thermal conductivity as function of filler concentration

4 Conclusion

Epoxy-graphite composites were prepared using moulding technique for a filler loading of 0-0.50 volume fraction. The theoretical density varies from the experimentally observed values as the filler loading increases. This is also the case for moisture absorption. The dielectric properties of the composites were analyzed in the whole composition range at frequencies 10 kHz and 1 MHz. It was observed that the dielectric constant increases with filler loading. At 0.5 vf of filler loading, the values obtained are 47.2 and 38.2 respectively. Theoretical modeling was performed for dielectric constant and none of the models gives adequate fit with the experimental values. The dielectric loss tangent increases as expected for higher filler loading from 10-2 to 10-1. The thermal conductivity of the composites increased from 0.2 W/mK to 9.8 W/mK at 0.5 vf of filler loading.

References

1. Ambegaokar, V., Halperin, B. I., and Langer, J. S.(1971). *Theory of hopping conductivity in disordered systems, Phys. Rev. B, 4, 2612.*

2. Baziard, Y., Breton, S., Toutain, S. and Gourdenne, A. (1988). *Dielectric properties of aluminium powder-epoxy resin composites, Eur Polym J, 24, 521.*
3. Bigg, D. M. (1986). *Graphite-filled Polymers: Properties and Applications; Bhattacharya, S. K., Ed.; Marcel Dekker: New York,;165.*
4. Dat, R, Auciello, O., Lichtenwalner, D. J. and Kingon, A. I. (1996). *Pulsed laser ablation-deposition of* $La_{0.5}Sr_{0.5}CoO_3$ *for use in electrodes in non-volatile ferroelectric memories, J. Mater. Res. 11, 1514.*
5. Doyle, W. T. and Jacobs, I. S. (1990). *Effective cluster model of dielectric enhancement in metal-insulator composites, Phys. Rev. B, 42, 9319.*
6. Gonon, P., Hong, T. P., Lesaint, O., Bourdelais, S. and Debruyne, H. (2005*). Influence of high levels of water absorption on the resistivity and dielectric permittivity of epoxy composites, Polym. Test., 24, 799.*
7. Grannan, D. M., Garland, J. C. and Tanner, D. B. (1981). *Critical behavior of dielectric constant of a random composite near percolation threshold, Phys. Rev. Lett., 46, 375.*
8. Marinelli, M., Murtas, F., Mecozzi, M. G., Zammit, U., Pizzoferrato, R., Scudieri, F., Martellucci, S. and Marinelli, M. (1990). *Simultaneous determination of specific heat, thermal conductivity and thermal diffusivity at low temperature via the photopyroelectric technique. Appl. Phys. A. Mater. Sci. and Process.* ***51****(5): 387.*
9. Menon, C.P. and J. Philip (2000). *Simultaneous determination of thermal conductivity and heat capacity near solid state phase transitions by a photopyroelectric technique. Measurement Science & Technology. 11(12): p. 1744-1749.*
10. Park, S. J., Kim, K. S. and Lee, J. R. (2004). *Thermal and mechanical interfacial properties of expanded graphite/epoxy composites, J. Korean. Ind. Eng. Chem 15[5] 493.*
11. Sebastian, M. T. (2008) *Dielectric materials for wireless communication, Elseiver Publishers, Oxford, U.K.*
12. Sheng, P., Abeles, B. and Arie, Y. (1973). Langer, J. S. (1971). *Hoping conductivity in granular metals, Phys. Rev. Lett. 31, 44.*
13. Sheng, P., Sichel, E. K. and Gittleman, J. I. (1978). *Fluctuation induced tunnelling conduction in carbon-polyvinyl chloride composites, Phys. Rev. Lett., 40, 1197*
14. Song, Y., Noh, T. W., Lee, S. I. and Gaines, J. R. (1986). *Experimental study of the three-dimensional ac conductivity and dielectric constant of a conductor-insulator composite near the percolation threshold, Phys. Rev. B 33, 904.*
15. Toker, D., Azulay, D., Shimoni, N., Balberg, I. and Millo, O. (2003) *Tunneling and percolation in metal-insulator composite materials, Phys. Rev. B 68, 041403.*
16. Zhou, M.H., Yin, G.Z. and Prolongo, S. G. (2024) *Review of thermal conductivity in epoxy thermosets and composites: Mechanisms, parameters, and filler influences, Advanced industrial and engineering polymer research, 7[3], 295.*