

Evaluation of Dielectric Mixing Models for the Relative Permittivity of Epoxy/Barium Hexaferrite ($\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$) Particulate Composites

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How to cite this paper: Tashmukhanbet, B.B. (2026) Evaluation of Dielectric Mixing Models for the Relative Permittivity of Epoxy/Barium Hexaferrite ($\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$) Particulate Composites. *Advances in Materials Physics and Chemistry*, **16**, 263-271.
<https://doi.org/10.4236/ampc.2026.167014>

Received: June 15, 2026

Accepted: July 12, 2026

Published: July 15, 2026

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Abstract

Dispersion-filled polymer composites allow the dielectric response of an insulating matrix to be tailored through the type, size and loading of an embedded second phase. In the present work, the relative permittivity of a two-phase system, consisting of an ED-20 epoxy matrix ($\epsilon_1 = 3$) filled with micron-sized barium hexaferrite ($\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$, $\epsilon_2 = 15$, effective density $5200 \text{ kg} \cdot \text{m}^{-3}$) particles, was studied as a function of filler content and particle size. Composites with spherical filler particles of diameter $2 \mu\text{m}$ and $20 \mu\text{m}$ were measured at filler fractions of 0%, 20%, 40% and 60%. Seven classical effective-medium and mixing relations, the Maxwell-Garnett, Lichtenecker, Wagner, Birchak (square-root), Hashin-Shtrikman, Spichnyak-Novikov and Lorentz-Lorenz models—were compiled and evaluated against the data. The effective permittivity increased monotonically with filler content, from 3 for the neat resin up to 9.2 ($2 \mu\text{m}$) and 5.2 ($20 \mu\text{m}$) at 60% loading. The fine-particle system is described well by the square-root mixing rule, whereas the coarse-particle system falls below all classical isotropic predictions, indicating a pronounced particle-size (interfacial-area) contribution that the classical size-independent rules do not capture.

Keywords

Polymer Composite, Barium Hexaferrite, Relative Permittivity, Dielectric Mixing Rules, Effective Medium Theory, Epoxy Resin

1. Introduction

Composite materials based on polymer matrices that are structured with a dis-

persed filler are of considerable practical importance, because such systems combine a unique set of chemical, physico-mechanical and service properties. The characteristics of these materials depend in a complex way on the grade of the polymer and on structural parameters such as the composition, the shape and the size of the filler particles, and the manner in which the particles are distributed throughout the volume of the matrix [1] [2].

Numerous experimental and theoretical studies have shown that the electrophysical properties of composites are particularly sensitive to changes in the packing of the particles, especially when external magnetic or thermal fields are applied during fabrication and structure formation. The targeted design of polymer composite materials (PCMs) with prescribed properties therefore relies on controlling the structural organization of the filler within the matrix. Nevertheless, the regulation of electrophysical behaviour through field-assisted structuring, and the influence of the orientation of the filler ensemble, remain insufficiently studied.

Most polymers are dielectrics and, under an applied field, behave as electrically insulating materials. In practice, however, the application of a constant voltage produces a small leakage current, which grows as the volume resistivity of the material decreases; for the majority of organic polymeric electrical-insulating materials, the volume resistivity lies in the range $\rho = 10^{10} - 10^{16} \Omega \cdot \text{m}$. In unfilled polymer dielectrics, ionic, polarization and electronic mechanisms of conduction may coexist [2]. The dielectric permittivity itself depends on a number of factors, including the aggregate state of the sample, the structure and chemical composition of the substance, the structure and polarity of the molecules, the hygroscopicity and porosity of the material, and the presence of impurity or adsorbed atoms; in each particular case, the dominant mechanism is established experimentally.

Among inorganic fillers, M-type barium hexaferrite ($\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$, equivalently $\text{BaFe}_{12}\text{O}_{19}$) is attractive because it combines high electrical resistivity and low eddy-current loss with a relatively high permittivity, together with a strong magnetocrystalline anisotropy and good chemical stability [3] [4]. These properties make hexaferrite-loaded composites of interest for microwave devices, electromagnetic-interference shielding and antenna miniaturization. Epoxy resin is a convenient matrix for such composites: it is easy to process, thermochemically stable, has a high dielectric breakdown strength and is inexpensive [5].

The aim of the present work is to obtain and to evaluate a calculation procedure for the relative permittivity of a two-phase system, an epoxy (ED-20) matrix filled with micron-sized barium hexaferrite particles, as a function of the shape, dispersity and content of the filler. To this end, seven classical mixing and effective-medium relations are compiled and their predictions are compared with permittivity data for composites containing 2 μm and 20 μm filler particles at loadings from 0% to 60%.

2. Materials and Modeling

2.1. Structure of the Composite and Distribution of the Filler

In its most general form, the structure of a polymer composite material can be

represented as one continuous polymer phase (the matrix) together with one or more dispersed phases (the filler) distributed within it in a definite manner. The principle of obtaining a composite material thus consists in forming a predetermined combination of two distinct phases by means of an appropriate technological process. According to the way in which the components are distributed, composites can be divided into matrix (regular) systems, in which the filler particles occupy the sites of a regular lattice; statistical systems, in which the components are distributed at random and form no regular structure; and structured systems, in which the components form chain-like, planar or three-dimensional networks. Typical filler distributions are shown in **Figure 1**.

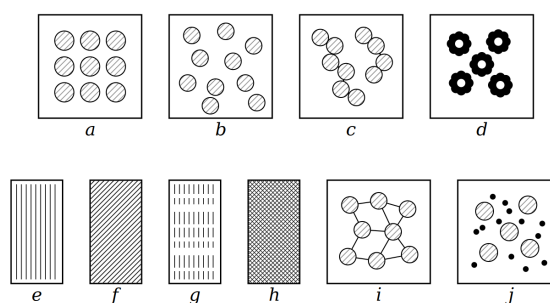


Figure 1. Typical structures of composite materials and distribution of the filler within the matrix: (a) regular (matrix) packing; (b) statistical (random) distribution; (c) (d) structured (chain-like and network) arrangements; (e)–(j) fibre-reinforced and mixed morphologies.

When a composite with special properties is designed, the filler is introduced in order to impart the desired electrophysical, thermal or sensing characteristics, and the particles are distributed within the matrix by one route or another. A whole range of fillers of different nature, both dispersed and fibrous, is used for this purpose.

2.2. Model System and Parameters

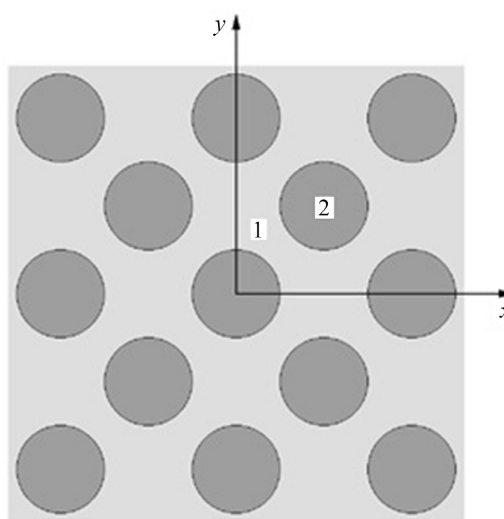


Figure 2. Schematic of a two-component dielectric material with a uniform distribution of the filler: 1: polymer matrix; 2: barium hexaferrite particles.

For the study of the relative permittivity, samples were prepared consisting of a polymer matrix, ED-20 epoxy resin, filled with spherical barium hexaferrite ($\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$) particles of diameter $2\ \mu\text{m}$ and $20\ \mu\text{m}$. The filler content was set to 0%, 20%, 40% and 60% by volume of the total composite, *i.e.* the filler volume fraction $\varphi_2 = V_{\text{filler}} / V_{\text{composite}}$, with $\varphi_1 + \varphi_2 = 1$. The permittivity of the neat epoxy matrix was taken as $\varepsilon_1 = 3$ [2] and that of the barium hexaferrite filler $\varepsilon_2 = 15$ [3]; the effective density of the barium hexaferrite was $5200\ \text{kg} \cdot \text{m}^{-3}$ [4]. The density is not an input to the mixing models themselves; it is reported because it was used to convert the gravimetrically batched filler content into the volume fraction φ_2 employed below. A two-component dielectric with a uniform distribution of the filler is shown schematically in **Figure 2**.

2.3. Determination of the Relative Permittivity

The relative permittivity was determined from a capacitance measurement on a flat (plane-parallel) specimen. The relative permittivity is obtained as the ratio of the capacitance of the specimen to that of the empty (air) cell of the same geometry:

$$\varepsilon_r = \frac{C}{C_0} \quad (1)$$

where C_0 is the capacitance of the empty (air) cell and C is the capacitance with the specimen in place. The capacitances were measured with a UNI-T UT603 handheld RLC meter using a two-electrode parallel-plate (contact) method. Specimens were discs of 20 mm diameter and 2 mm thickness, giving an electrode area of approximately $3.14\ \text{cm}^2$. Reliable electrode-specimen contact was ensured by applying “Kontaktol” conductive adhesive to the disc faces, which suppresses the series air-gap that would otherwise lower the apparent capacitance. Measurements were carried out at ambient laboratory temperature ($\approx 22^\circ\text{C} - 24^\circ\text{C}$). The UT603 applies a fixed internal test signal whose frequency is not specified by the manufacturer; because the mixing relations evaluated here are quasi-static (frequency-independent), the comparison is made in the low-frequency limit. Two to three discs were measured per loading; the reported permittivity is the mean over the specimens, and the measurement uncertainty is taken as the instrument’s rated capacitance accuracy, $\pm(1\% + 5\ \text{digits})$, combined with the observed inter-specimen scatter (**Figure 3**).

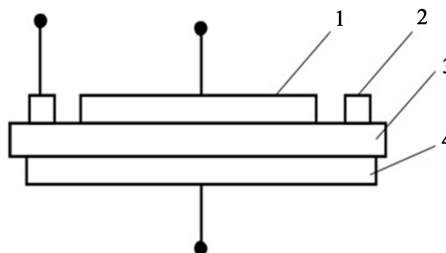


Figure 3. Arrangement of the electrodes on the flat specimen: 1 and 4: electrodes for measuring C_0 and C ; 2: lead of the upper measuring electrode; 3: the polymer specimen filled with barium hexaferrite powder.

2.4. Dielectric Mixing Models

To predict the effective relative permittivity ε_r of the two-phase composite, seven classical mixing and effective-medium relations were compiled from the literature on heterogeneous dielectrics [1] [6] (see also the Bruggeman symmetric effective-medium theory [7]). In all expressions ε_1 and ε_2 denote the permittivities of the matrix (epoxy) and of the filler (barium hexaferrite), respectively, while φ_1 and φ_2 are the corresponding volume fractions, with $\varphi_1 + \varphi_2 = 1$. The models are summarized in **Table 1**.

Table 1. Classical mixing and effective-medium relations for the effective relative permittivity ε_r of a two-phase dielectric composite.

No.	Model	Effective relative permittivity ε_r
1	Maxwell-Garnett [8]	$\varepsilon_r = \varepsilon_1 \frac{(2\varepsilon_1 + \varepsilon_2) + 2\varphi_2(\varepsilon_2 - \varepsilon_1)}{(2\varepsilon_1 + \varepsilon_2) - \varphi_2(\varepsilon_2 - \varepsilon_1)}$
2	Lichtenecker [9] [10]	$\varepsilon_r = \sqrt{\frac{\varepsilon_2 \varepsilon_1 (\varphi_1 \varepsilon_1 + \varphi_2 \varepsilon_2)}{\varphi_2 \varepsilon_1 + \varphi_1 \varepsilon_2}}$
3	Wagner	$\varepsilon_r = \varepsilon_2 \left[1 + \frac{3\varphi_1(\varepsilon_1 - \varepsilon_2)}{\varepsilon_1 + 2\varepsilon_2} \right]$
4	Birchak (square-root) [5]	$\varepsilon_r = \left(\varphi_1 \sqrt{\varepsilon_1} + \varphi_2 \sqrt{\varepsilon_2} \right)^2$
5	Hashin-Shtrikman [11]	$\varepsilon_r = \varepsilon_1 \left[1 + \frac{\varphi_2}{\frac{\varepsilon_1}{(\varepsilon_2 - \varepsilon_1)} + \frac{\varphi_1}{2}} \right]$
6	Spichnyak-Novikov	$\varepsilon_r = \varepsilon_1 \varphi_1^2 + \varepsilon_2 \varphi_2^2 + 4\varphi_1 \varphi_2 \frac{\varepsilon_1 \varepsilon_2}{\varepsilon_2 + \varepsilon_1}$
7	Lorentz-Lorenz	$\varepsilon_r = \varepsilon_1 \left[1 + \frac{\varphi_2(\varepsilon_2 - \varepsilon_1)}{\varepsilon_1 + \frac{1 - \varphi_2}{3}(\varepsilon_2 - \varepsilon_1)} \right]$

The Maxwell-Garnett relation (model 1) treats the filler as well-separated spherical inclusions in a continuous matrix and is most accurate at low loadings [8]. The Lichtenecker relation (model 2) is a power-type mixing law for randomly distributed phases [9] [10], while the Wagner relation (model 3) is the classical dilute-suspension approximation. The Birchak square-root law (model 4) corresponds to a refractive ($\alpha = 1/2$) mixing rule [5]; the Hashin-Shtrikman expression (model 5) provides a variational bound for an isotropic two-phase medium [11]; and the Spichnyak-Novikov (model 6) and Lorentz-Lorenz (model 7) relations describe statistical and polarizability-based mixing, respectively.

3. Results and Discussion

The permittivities used in the calculations are listed in **Table 2**. The resulting relative permittivity of the composite, measured for the two filler particle sizes at

filler contents from 0% to 60%, is given in **Table 3** and plotted in **Figure 4**.

Table 2. Input parameters used for the calculation of the effective permittivity.

Parameter	Value
Matrix permittivity, ε_1 (ED-20 epoxy)	3
Filler permittivity, ε_2 (BaO-6Fe ₂ O ₃)	15
Effective filler density	5200 kg·m ⁻³
Filler particle diameter, d	2 and 20 μm
Filler content, φ_2	0, 20, 40, 60 vol %

Table 3. Relative permittivity ε_r of the epoxy/BaO-6Fe₂O₃ composite as a function of filler content φ_2 and particle diameter d .

Filler content φ_2 (vol %)	0	20	40	60
$\varepsilon_r (d = 2 \mu\text{m})$	3.0	4.4	6.3	9.2
$\varepsilon_r (d = 20 \mu\text{m})$	3.0	3.7	4.6	5.2

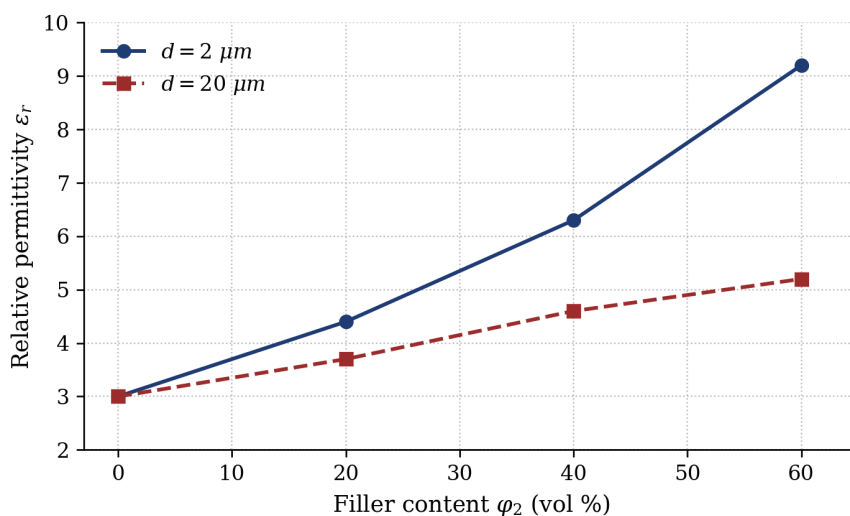


Figure 4. Relative permittivity ε_r of the epoxy/BaO-6Fe₂O₃ composite versus filler content φ_2 for filler particle diameters of 2 μm and 20 μm .

For both particle sizes the relative permittivity rises monotonically with filler content, as expected when a high-permittivity phase progressively replaces the low-permittivity matrix. The increase is markedly stronger for the fine (2 μm) filler: at 60% loading ε_r reaches 9.2 for the 2 μm particles but only 5.2 for the 20 μm particles, although both systems start from the same value ($\varepsilon_r = 3$) for the neat resin. The classical mixing rules of **Table 1** depend only on the permittivities and the volume fractions of the phases and therefore predict a single $\varepsilon_r(\varphi_2)$ curve, independent of particle size; the observed size dependence consequently lies outside the scope of those relations and points to an additional interfacial contribution.

Table 4. Comparison of the measured permittivity with the predictions of three representative mixing rules ($\varepsilon_1 = 3$, $\varepsilon_2 = 15$).

ϕ_2 (vol %)	20	40	60
Lichtenecker	4.39	5.87	7.67
Maxwell-Garnett	4.16	5.67	7.70
Birchak (square-root)	4.67	6.70	9.10
Measured, $d = 2 \mu\text{m}$	4.4	6.3	9.2
Measured, $d = 20 \mu\text{m}$	3.7	4.6	5.2

Table 4 compares the data with three representative mixing rules. The Lichtenecker and Maxwell-Garnett relations give similar predictions, which agree closely with the $2 \mu\text{m}$ data at low and intermediate loadings but underestimate it at 60%. The Birchak square-root rule reproduces the fine-particle data remarkably well over the whole range, predicting 9.10 against the measured 9.2 at 60% loading. In contrast, the coarse ($20 \mu\text{m}$) composite lies below all three isotropic predictions at every loading.

Table 5. Predictions of all seven mixing rules and their mean absolute error (MAE) across the three loadings (20, 40, 60 vol %) relative to the measured data.

Model	20%	40%	60%	MAE ($2 \mu\text{m}$)	MAE ($20 \mu\text{m}$)
Maxwell-Garnett	4.16	5.67	7.70	0.79	1.34
Lichtenecker	4.39	5.87	7.67	0.66	1.48
Wagner	1.91	5.18	8.45	1.45	1.88
Birchak (square-root)	4.67	6.70	9.10	0.26	2.32
Hashin-Shtrikman	3.92	5.18	7.00	1.27	0.87
Spichnyak-Novikov	4.12	5.88	8.28	0.54	1.59
Lorentz-Lorenz	4.16	5.67	7.70	0.79	1.34
Measured, $d = 2 \mu\text{m}$	4.4	6.3	9.2	—	—
Measured, $d = 20 \mu\text{m}$	3.7	4.6	5.2	—	—

Quantitatively, the mean absolute error in **Table 5** is smallest for the Birchak square-root rule for the fine $2 \mu\text{m}$ system (MAE = 0.26), confirming the qualitative agreement noted above. For the coarse $20 \mu\text{m}$ system the smallest error is obtained with the Hashin-Shtrikman relation (MAE = 0.87); nevertheless every model still overestimates the $20 \mu\text{m}$ data at the higher loadings, so the coarse-particle response remains below the isotropic mixing predictions. This behaviour is consistent with an interfacial-polarization (Maxwell-Wagner) interpretation. At a fixed volume fraction, reducing the particle diameter by an order of magnitude increases the total filler–matrix interfacial area by roughly the same factor, which enhances the accumulation of charge at the phase boundaries and therefore raises the effective permittivity. The fine-particle system, with its larger interfacial area and more efficient polarization, is well captured by the refractive (square-root)

mixing rule, whereas the coarse-particle system, where interfacial effects are weaker and the filler approaches the behaviour of isolated inclusions, falls below the isotropic estimates. These observations indicate that an adequate description of dispersion-filled hexaferrite/epoxy composites requires a mixing relation in which the particle shape and dispersity enter explicitly, in addition to the permittivities and volume fractions of the constituent phases [4] [12].

4. Conclusion

A two-phase composite consisting of an ED-20 epoxy matrix filled with micron-sized barium hexaferrite ($\text{BaO} \cdot 6\text{Fe}_2\text{O}_3$) particles was studied, and seven classical mixing and effective-medium relations were compiled and evaluated as a means of predicting its relative permittivity. The relative permittivity increases monotonically with filler content for both particle sizes investigated, rising from 3 for the neat resin to 9.2 (2 μm) and 5.2 (20 μm) at 60% loading. The fine-particle composite is described well by the Birchak square-root mixing rule, while the logarithmic Lichtenecker and Maxwell-Garnett relations slightly underestimate it at high loadings. The coarse-particle composite lies below all isotropic mixing predictions. The pronounced dependence of the permittivity on particle size, which the classical size-independent rules cannot reproduce, is attributed to interfacial (Maxwell-Wagner) polarization, whose magnitude scales with the filler-matrix interfacial area. The results confirm that the dielectric response of dispersion-filled polymer composites can be tuned through both the loading and the dispersity of the filler, and that a predictive description should incorporate the shape and size of the filler particles explicitly. Such tailored high-permittivity, low-loss composites are promising for electrical-engineering, electronic and communication-equipment applications.

Acknowledgements

The author thanks the staff of Al-Farabi Kazakh National University, Almaty, for support of this work, and acknowledges the guidance of the project supervisor.

Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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