



Effective permittivity of composite elastomer with account of electric conductivity of phases and imperfect interface



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ABSTRACT

The homogenization problem of micromechanics in application to the effective permittivity of particulate composite elastomer with conducting constituents and imperfect interface is studied. The available in literature theoretical models are critically reviewed and the typical problems with interpretation of experimental data are addressed. The imperfect dielectric phases is pointed out as a probable reason of the discrepancy between the theory and experiment. It has been shown that taking the electric conductivity of the composite constituents into account greatly improves accuracy of the predictive models. The Maxwell–Wagner model has been extended to composite dielectrics with imperfect interface. Some possible directions of further research are outlined.

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1. Introduction

Dielectric composite elastomers are now widely recognized as a highly promising material for a variety of applications including energy harvesting and robototechnics (e.g., [Pelrine et al., \(1998, 2000\)](#)). Performance of these materials is affected greatly by their electromechanical coupling which is, in turn, a quadratic function of permittivity ([Landau & Lifshitz, 1984](#)). According to [Bortot, Springhetti, and Gei \(2014\)](#), combination of dissimilar materials is an effective way to produce high permittivity soft composites, as demonstrated both experimentally ([Gallone, Carpi, De Rossi, Levita, & Marchetti, 2007](#); [Molberg et al., 2010](#); [Risse, Kussmaul, Kruger, & Kofod, 2012](#); [Stoyanov, Kollasche, Risse, McCarthy, & Kofod, 2011](#)) and theoretically ([Gei, Springhetti, & Bortot, 2013](#); [Ponte Castañeda & Siboni, 2012](#); [Rudykh, Lewinstein, Uner, & deBotton, 2013](#); [Tian, Tevet-Deree, deBotton, & Bhattacharya, 2012](#)). Macroscopic, or effective permittivity of composite may be greatly improved by the purposeful tailoring the microstructure of composite. In so doing, it is important to identify the leading factors which affect composite performance and ensure their proper values in the manufacturing process. This requires a clear understanding the “structure – properties” relationships which can be established by the means of micromechanics.

Effective permittivity of heterogeneous solids has been paid a considerable attention starting from the works of [Clausius \(1858\)](#), [Maxwell \(1873\)](#) and [Mossotti \(1850\)](#). Since that time, the advanced analytical and numerical homogenization schemes developed for the effective conductivity of composite (see, e.g. [Böhm \(2004\)](#) for a review). Noteworthy, the equations describing several transport phenomena (thermal and electrical conductivity, mass diffusivity, dielectric permittivity, magnetic permeability, optical refractivity) are known to be mathematically equivalent. This means that the results

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obtained for one transport property are transferrable to other transport properties of composite including the dielectric constant. However, these advanced and scientifically substantiated results do not find, as a rule, application to composite dielectrics.

Moreover, it turns out that the theoretical models expressing the dielectric constant of heterogeneous solids in terms of the dielectric constants of components fail when it comes to prediction of the properties of ferroelectric ceramic/polymer composites. For example, the experimental data on permittivity of composite reported in [Furukawa, Fujino, and Fukada \(1976\)](#), [Gallone et al. \(2007\)](#), [Ng, Chan, and Choy \(2000\)](#), [Yamada, Ueda, and Kitayama \(1982\)](#) and several other publications show quite a large systematic deviation from the [Maxwell \(1873\)](#) theory.

The possible reason for the observed discrepancy is that the above mentioned theoretical models consider the composite constituents as perfect dielectrics whereas many materials (in particular, polymers) along with dielectric properties possess also electrical conductivity. In this case, both the permittivity and conductivity of constituents affect the overall electric behavior of composite. [Wagner \(1914\)](#) was first to extend the [Maxwell \(1873\)](#) formula to composites made of imperfect dielectric constituents under action of harmonic far field. This and subsequent ([Fricke, 1953](#); [Sillars, 1937](#); [Van Beek, 1960](#); among others) publications explain a nature of such features as dielectric loss and frequency dependence of the effective dielectric constant of composite.

Interface is well known as an important factor affecting the composite's behavior in real world. This is fully applicable to composite dielectrics where the interfaces play a particularly important role. Noteworthy, a vast majority of the theoretical models of composites involve the "perfect interface" assumption which is rather exception than a rule. The real interface may be imperfect due to several reasons including the atomic lattices mismatch, poor mechanical or chemical adherence, surface contamination, oxide and interphase diffusion/reaction layers, debonding, etc. These phenomena affect the bulk properties of composite and make them size-dependent. One can expect that the total interface contribution can become very significant as the particle diameter is reduced. According to [Lewis \(2004\)](#), interfaces are the dominant feature of dielectrics at the nanometer level so the nanocomposites are expected to possess exceptional dielectric properties ([Lopez-Pamies, Goudarzi, Meddeb, & Ounaies, 2014](#)).

In this paper, the macroscopic, or effective permittivity of composite elastomer with imperfect dielectric phases and imperfect interface is addressed. The paper is organized as follows. First, we consider/compare the available in literature experimental data on effective permittivity of composite elastomers and observe their systematic deviation from the well-established theoretical models. Second, we discuss briefly the homogenization problem of micromechanics in application to composites with dielectric constituents and show that the abovementioned discrepancy between theory and experiment is readily eliminated by assuming electro conducting properties of constituents which is supported by the experimental data (e.g., [Furukawa, Ishida, & Fukada, 1979](#)). Next, the Maxwell–Wagner model is extended to composites with imperfect (both low- and highly conducting) interface. Parametric study of this basic model has been performed and the possible ways of its refinement/generalization are outlined.

2. Effective permittivity: experimental data and interpretation

In this Section, we analyze the experimental data on effective permittivity ε^* of composite elastomers reported by several authors. To ensure comparability, the data obtained for similar materials and test conditions were selected for this study. Specifically, a the polymer matrix (0–3 connectivity type) composites are considered. Ferroelectric ceramic powder is used as a reinforcing phase so we have a high phase contrast $\varepsilon_2/\varepsilon_1$. Here and below, ε_1 and ε_2 denote permittivity of matrix and particle materials, respectively. The powder particles of approximately the same size and a near-to-spherical shape are randomly and evenly distributed in the matrix, which provides macroscopic isotropy of composite. All the considered works include production of composite samples, measurement their permittivity in low frequency ($10 \div 10^3$ Hz) range by means of broadband dielectric spectroscopy and fitting the obtained data by that or another theoretical model.

We start with the paper by [Furukawa et al. \(1976\)](#) who studied the composite of epoxy resin and lead zirconate titanate (PZT) ceramic powder of 0.2–2 μm in diameter. Phase contrast $\varepsilon_2/\varepsilon_1 = 1180/4 \approx 300$, measurement frequency 10 Hz. They found that "...magnitude of ε^* was 2 times... greater than the theoretical prediction by [Maxwell \(1873\)](#) theory. The fact that the observed constants were greater than the predictions suggests that PZT ceramics have distorted shape or forming one-dimensional aggregations". However, no evidences in support of this hypothesis were provided.

A binary system consisting of polyvinylidene fluoride (PVDF) and PZT fine powder was studied by [Yamada et al. \(1982\)](#). Phase contrast of composite $\varepsilon_2/\varepsilon_1 = 1850/8.9 \approx 200$, measurement frequency 35 Hz. To fit the experimental data, [Yamada et al. \(1982\)](#) have suggested the Maxwell type formula for effective dielectric constant of composite with ellipsoidal inclusions and found that the best fit is provided by the model with the ellipsoid axes ratio 2.8 (the shape parameter of model $n = 8.5$) and the long ellipsoids of PZT powder oriented in the applied electric field direction.

The experimental data by [Hopkins, Cannell, and Thomas \(1992\)](#) on permittivity of composite with phase contrast of $\varepsilon_2/\varepsilon_1 = 225/3 = 75$ are taken from the paper by [Jayasundere and Smith \(1993\)](#) who modified the Kerner (in fact, Maxwell) formula for effective permittivity by including (as they believed) pair wise interactions between the neighboring spheres into account. The formula they proposed appears to fit the [Yamada et al. \(1982\)](#) experimental data as well.

[Ng et al. \(2000\)](#) studied the composites of PZT powder with particle size of 1–3 μm dispersed in a vinylidene fluoride-trifluoroethylene copolymer [P(VDF-TrFE)] matrix prepared by compression molding. Phase contrast $\varepsilon_2/\varepsilon_1 = 120$. Permittivity

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