1

Introduction

1.1 What is an electronic composite?

"Composite" is a well-accepted word, generally referring to structural components with enhanced mechanical performance. There are a number of textbooks and review articles on these types of composites (e.g., Kelly, 1973; Tsai and Hahn, 1980; Hull, 1981; Chawla, 1987; Clyne and Withers, 1993; Daniel and Ishai, 1994; Gibson, 1994; Hull and Clyne, 1996).

Historically, composites with enhanced mechanical performance have been in existence from ancient-Egyptian time, *c*. 2000 BC, when bricks were made of mud, soil and straw (*Exodus*, Chapter 5, verse 7). Structural composites are designed primarily to enhance the mechanical properties of a matrix material by introducing reinforcement; the primary mechanical properties to be enhanced are strength, stiffness, and fracture resistance.

Normally, a composite consists of a matrix material and one kind of filler, but sometimes more than one kind of filler can be used, forming a "hybrid composite". Depending on the matrix material, one can group composites into three basic types: polymer matrix composites (PMCs), metal matrix composites (MMCs), and ceramic matrix composites (CMCs). Among these, PMCs are the most popular type for electronic composites due to their low processing temperatures and associated cost-effectiveness.

An "electronic composite" is defined as a composite that is composed of at least two different materials and whose function is primarily to exhibit electromagnetic, thermal, and/or mechanical behavior while maintaining structural integrity. Thus, "electronic" should not be interpreted narrowly as referring only to electronic behavior, but instead be understood in much broader terms, including physical and coupling behavior. In this sense, electronic composites are distinguished from structural composites whose primary function is to enhance mechanical properties.

Among various applications, electronic composites have been extensively used as the major component materials in electronic packaging: printed circuit boards (PCBs), thermal interface materials (TIMs), encapsulants, etc., most of which are polymer-based composites providing ease of fabrication and cost-effectiveness. As an extension of electronic packaging, electronic composites are used now in micro-electromechanical systems (MEMS) and BioMEMS, where their functions are multi-fold: active, sensing and housing materials.

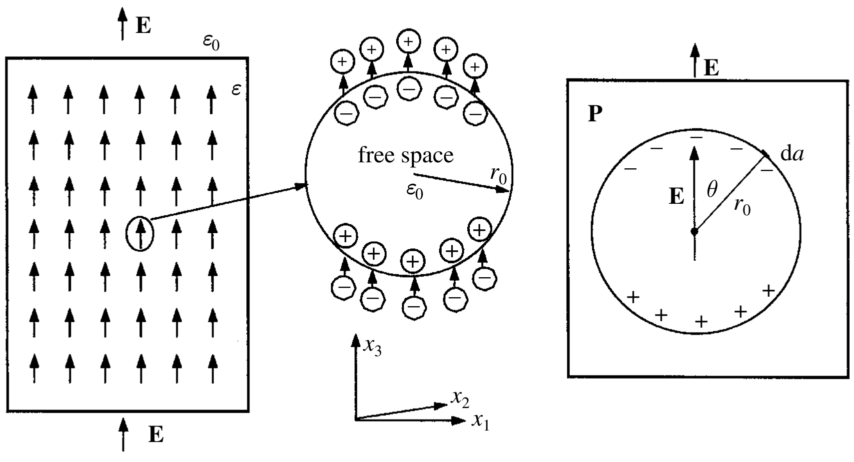
The properties of electronic composites can be tailored to meet specific applications. Thus, prediction of the composite properties at an early stage of designing electronic composites is a key task. Normally the composite property is expected to fall between those of the matrix and filler, following the law-of-mixtures type formula, and depends greatly highly on the microstructural parameters: volume fraction, filler shape and size distributions; the properties of the matrix and filler; and also the properties of the matrix–filler interface. Sometimes, the property of an electronic composite becomes quite different from those of the matrix material and filler, and is far from that based on the law-of-mixture type formula. Such a unique property of the composite can be designed purposely or found accidentally in the course of development of a functional composite; it is termed a "cross product" exhibiting "coupling behavior" (Newnham *et al.*, 1978). Since coupling behavior between various physical properties is included in the definition of electronic composites, composites with such coupling behavior are often referred to as "smart composites" or "multi-functional materials," which are the key materials systems for use in smart structures and devices ranging from bio-micro-electromechanical systems (BioMEMS) through sensors to actuators. Therefore, construction of accurate models for the macro-property–microstructure (or –nanostructure) relation is strongly desired. These models are multi-scale, i.e., covering nano-, micro-, meso-, and macro-levels. If these models at different scale levels are interconnected smoothly, one can establish a hierarchical model which will be useful for many scientists and engineers who want to design new smart (or intelligent) materials, MEMS and BioMEMS devices, and multi-functional structures. The main body of this book is devoted to presenting a number of such models. In the remainder of this chapter, we shall review earlier models of electronic composites.

1.2 Early modeling of electronic composites

Modeling of electronic composites in the nineteenth century and early part of the twentieth century focused on the prediction of the dielectric constant ε and electrical conductivity σ or resistivity ρ of a composite composed of spherical fillers with ε_f, σ_f (ρ_f) and matrix with ε_m and σ_m (ρ_m) where the subscripts f and m denote filler and matrix, respectively. Landauer (1978) made an extensive literature survey of early models for the electrical conductivity of composites that were proposed in the nineteenth century through to the mid twentieth century. We shall review some of the early models used to predict the electromagnetic properties of composites, i.e., (1) Lorentz sphere problem, (2) demagnetization in a ferromagnetic body, and (3) concepts of thermal, electric, and magnetic circuits. The first two provide the background for modeling based on the effective medium theory, the last for the resistor network model. Both models will be discussed in detail in later chapters.

1.2.1 Lorentz sphere problem

Consider a dielectric material with dielectric constant ε which is subjected to a uniform electric field **E**, Fig.1.1(a). At a macroscopic level, the dielectric material is considered to consist of a uniform electric dipole moment with polarization **P** (per unit volume). At the atomic level, one can find a free space with ε_0 dielectric constant between lattice points (atoms) or molecules that constitute the dielectric medium. If we consider a spherical domain of radius r_0 between the atoms or molecules, Fig. 1.1(b), a layer of electric charges (positive and negative) is distributed on the inner surface of the



(a) (b) (c)

Fig. 1.1 Lorentz sphere problem: (a) macro-level model in a dielectric material subjected to uniform electric field resulting in uniform polarization **P**, (b) nano-level model where free space is polarized by pairs of positive and negative charges. (c) Lorentz sphere problem to idealize (b). (After Ishimaru, 1991, with permission from Pearson Education, Inc.)

sphere, which is called the "Lorentz sphere", Fig. 1.1(c). The net (or total) electric field $\mathbf{E}^{\mathrm{t}}$ in the sphere is expected to be larger than the applied field by an amount $\mathbf{E}_{\mathrm{p}}$, i.e.,

$$\mathbf{E}^{\mathrm{t}} = \mathbf{E} + \mathbf{E}_{\mathrm{p}}. \tag{1.1}$$

We shall compute the magnitude of $\mathbf{E}^{\mathrm{t}}$ using a model developed by four pioneers in the modeling of electric composites, Mossotti (1850), Clausius (1879), Lorenz (1880) and Lorentz (1909), which is summarized by Ishimaru (1991).

The magnitude of the charge P_r in the radial direction on the surface element da is given by

$$P_r = \mathbf{P} \cdot \mathrm{d}\mathbf{a} = P\cos\theta \;\mathrm{d}a, \tag{1.2a}$$

where

$$\mathrm{d}a = 2\pi r \sin\theta \; r \;\mathrm{d}\theta, \tag{1.2b}$$

and P is the magnitude of $\mathbf{P}$.

This charge P_r induces an electric field dE_{p} in the radial direction given by the following formula

$$\mathrm{d}E_{\mathrm{p}} = \frac{P_r}{4\pi\, r_0^2 \varepsilon_0}. \tag{1.3}$$

Thus, the magnitude of the electric field, E_{p}, in the x_3-direction, contributed by the layer of distributed charge on the entire inner surface of the sphere, is obtained as

$$\begin{aligned} E_{\mathrm{p}} &= \int_s \frac{P_r \cos\theta}{4\,\pi\, r_0^2 \varepsilon_0} \mathrm{d}a \\ &= \int_0^{\pi} \frac{P\cos^2\theta}{4\,\pi\, r_0^2\varepsilon_0} 2\pi r_0 \sin\theta \; r_0 \mathrm{d}\theta = \frac{P}{2\varepsilon_0} \int_0^{\pi} \cos^2\theta \;\sin\theta \;\mathrm{d}\theta \\ &= \frac{P}{3\varepsilon_0}. \end{aligned} \tag{1.4a}$$

Since the components of $\mathbf{E}_{\mathrm{p}}$ along the x_1- and x_2-directions are zero, the result of Eq. (1.4a) can be written in vector form as

$$\mathbf{E}_{\mathrm{p}} = \frac{\mathbf{P}}{3\varepsilon_0}. \tag{1.4b}$$

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Macroscopically, the dielectric medium is governed by

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P}, \tag{1.5}$$

where **D** is the electric flux density vector (coulombs [C]/m^2). The first term in Eq. (1.5) is the flux density in free space (i.e., if there were no atoms, or molecules) and the second term is the electric polarization vector resulting from electric dipole moments that exist in the dielectric medium. The electric flux density vector in a medium with dielectric constant ε is also proportional to the applied electric field, i.e.,

$$\mathbf{D} = \varepsilon \mathbf{E}. \tag{1.6}$$

For isotropic materials, **P** is proportional to the field **E** as

$$\mathbf{P} = \varepsilon_0 \chi_e \mathbf{E}, \tag{1.7}$$

where χ_e is the electric susceptibility. From Eqs. (1.5)–(1.7)

$$\begin{aligned} \varepsilon &= \varepsilon_0 (1 + \chi_e) \\ \text{or} \qquad \varepsilon_r &= \frac{\varepsilon}{\varepsilon_0} = 1 + \chi_e, \end{aligned} \tag{1.8}$$

where ε_r is the relative dielectric constant (non-dimensional) and ε_0 is the dielectric constant of free space, see Appendix B1.

At the microscopic level, the polarization vector **P** is composed of a number N of elemental dipole moments **p** which are in turn considered to be proportional to the net local field $\mathbf{E}^t$ in the sphere, i.e.,

$$\mathbf{P} = N\mathbf{p} = N\alpha \mathbf{E}^t, \tag{1.9}$$

where α is the polarizability. From Eqs. (1.1), (1.4), and (1.9), we have

$$\begin{aligned} \mathbf{E}^t &= \mathbf{E} + \frac{\mathbf{P}}{3\varepsilon_0} \\ &= \mathbf{E} + \frac{N\alpha \mathbf{E}^t}{3\varepsilon_0}. \end{aligned} \tag{1.10}$$

Equation (1.10) provides the relation between $\mathbf{E}^t$ and **E**,

$$\mathbf{E}^t = \frac{\mathbf{E}}{\left(1 - \frac{N\alpha}{3\varepsilon_0}\right)}, \tag{1.11}$$

which can be rewritten in terms of ε_r by using Eqs. (1.7), (1.8) and (1.10):

$$\mathbf{E}^{\mathrm{t}} = \frac{(\varepsilon_{\mathrm{r}} + 2)}{3}\mathbf{E}. \tag{1.12}$$

Equating the right-hand side of Eq. (1.11) to that of Eq. (1.12) and using Eq. (1.8), we obtain

$$\chi_{\mathrm{e}} = \frac{\dfrac{N\alpha}{\varepsilon_0}}{1 - \left(\dfrac{N\alpha}{3\varepsilon_0}\right)} \tag{1.13a}$$

and

$$\varepsilon_{\mathrm{r}} = \frac{1 + 2\left(\dfrac{N\alpha}{3\varepsilon_0}\right)}{1 - \left(\dfrac{N\alpha}{3\varepsilon_0}\right)}. \tag{1.13b}$$

Equations (1.13) provide the relation between polarizability α and relative dielectric constant ε_{r}:

$$\alpha = \frac{3\varepsilon_0}{N}\frac{(\varepsilon_{\mathrm{r}} - 1)}{(\varepsilon_{\mathrm{r}} + 2)}. \tag{1.14}$$

The above formulation, established by the four pioneering physicists named above, is known as the “Clausius–Mossotti” formula or “Lorentz–Lorenz” formula. Among these physicists, Lorentz summarized the formulae of his predecessors, and the model of Fig. 1.1 is called the “Lorentz sphere.”

1.2.2 Other models for dielectric constants

Let us extend the case of Fig.1.1 to that of a filler material with dielectric constant ε_2 embedded as spheres in a matrix material with constant ε_1, Fig. 1.2. The effective dielectric constant ε_{c} of the composite is given by modifying Eq. (1.13b):

$$\frac{\varepsilon_{\mathrm{c}}}{\varepsilon_1} = \frac{1 + \dfrac{2N\alpha}{3\varepsilon_1}}{1 - \dfrac{N\alpha}{3\varepsilon_1}}, \tag{1.15}$$

where the polarizability α is now replaced by

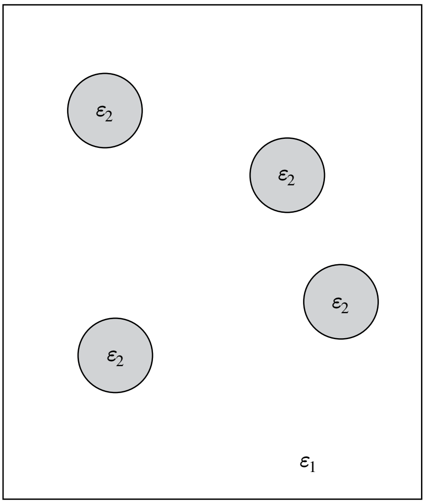

Fig. 1.2 Filler with dielectric constant ε_2 embedded in a matrix with constant ε_1.

$$\alpha = \frac{3\varepsilon_1(\varepsilon_2 - \varepsilon_1)}{(\varepsilon_2 + 2\varepsilon_1)} V \tag{1.16}$$

and V is the total volume of filler material in a sphere. If the spheres occupy a volume fraction f and N is interpreted as the number of the spheres per unit volume,

$$f = NV . \tag{1.17}$$

Note that $NV = 1$ in Eq. (1.14) where, effectively, the spheres occupied the entire space.

After substituting Eqs. (1.16) and (1.17) into Eq. (1.15), the composite dielectric constant ε_c is obtained as

$$\varepsilon_c = \varepsilon_1 \frac{1 + 2f \dfrac{(\varepsilon_2 - \varepsilon_1)}{(\varepsilon_2 + 2\varepsilon_1)}}{1 - f \dfrac{(\varepsilon_2 - \varepsilon_1)}{(\varepsilon_2 + 2\varepsilon_1)}} \tag{1.18}$$

which can be rewritten as

$$\frac{(\varepsilon_c - \varepsilon_1)}{(\varepsilon_c + 2\varepsilon_1)} = f \frac{(\varepsilon_2 - \varepsilon_1)}{(\varepsilon_2 + 2\varepsilon_1)} . \tag{1.19}$$

The formula Eq. (1.18) was first derived by Maxwell (1904), who considered the case of concentric spheres where an inner sphere with electric conductivity

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σ_1 is embedded in an outer sphere with conductivity σ_2. By replacing ε_i by σ_i in Eq. (1.18), one can obtain a formula to predict the electric conductivity σ_c of a composite, which is called the "Maxwell–Garnett mixing formula." In Eqs. (1.18) and (1.19), one can recover special cases, i.e., if $f = 0$, $\varepsilon_c = \varepsilon_1$ (matrix dielectric constant) and, if $f = 1$, $\varepsilon_c = \varepsilon_2$ (filler dielectric constant). Even though the Maxwell–Garnett mixing formula appears to cover the entire range $0 \leq f \leq 1$, the model is based on the assumption of a small volume fraction.

We can further extend our reasoning to the case of two materials (ε_1, ε_2) embedded in a composite matrix material (ε_c), Fig 1.3. Then, in the background composite material (ε_c), each phase (ε_1, ε_2) can be viewed as an inclusion having an electric dipole moment with its polarizability (α_1, α_2). The polarization **P** created by these two different dielectric phases is given by

$$\mathbf{P} = (N_1\alpha_1 + N_2\alpha_2)\mathbf{E}^{t}. \tag{1.20}$$

If **P** is integrated over the entire composite domain, it vanishes because the electric field in the background composite material is the same as the local total field $\mathbf{E}^{t}$. This requires

$$N_1\alpha_1 + N_2\alpha_2 = 0, \tag{1.21}$$

where α_i is the polarizability of the *i*th dielectric material (ε_i) with respect to the composite medium (ε_c). Thus, from Eq. (1.16), α_i ($i = 1, 2$) are given by

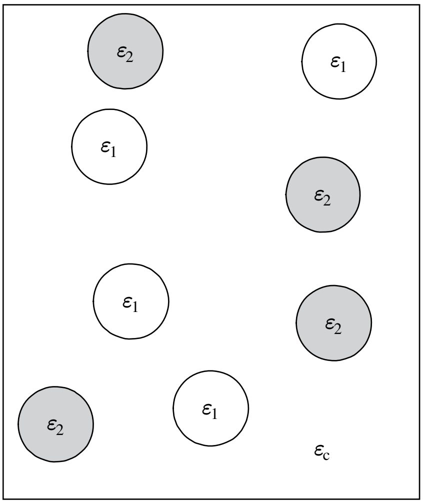

Fig. 1.3 Materials with dielectric constants ε_1 and ε_2 embedded in a background composite with constant ε_c.

$$\alpha_1 = \varepsilon_c \frac{3(\varepsilon_1 - \varepsilon_c)}{(\varepsilon_1 + 2\varepsilon_c)} V_1, \tag{1.22a}$$

$$\alpha_2 = \varepsilon_c \frac{3(\varepsilon_2 - \varepsilon_c)}{(\varepsilon_2 + 2\varepsilon_c)} V_2 \tag{1.22b}$$

and N_i, the number of spheres of the ith dielectric material, is related to the volume fraction f_i of the ith material, by

$$N_1 V_1 = f_1, \tag{1.23a}$$

$$N_2 V_2 = f_2. \tag{1.23b}$$

V_i in the above equations is the volume of each sphere of the ith dielectric material. Upon substituting Eqs. (1.22) and (1.23) into (1.21), and canceling a common factor ε_c, we obtain the formula

$$f_1 \frac{(\varepsilon_1 - \varepsilon_c)}{(\varepsilon_1 + 2\varepsilon_c)} + f_2 \frac{(\varepsilon_2 - \varepsilon_c)}{(\varepsilon_2 + 2\varepsilon_c)} = 0. \tag{1.24}$$

This formula is called “Bruggeman’s symmetric formula” (Bruggeman, 1935) since the interchange of dielectric materials 1 and 2 gives the same formula.

We can consider a simpler model than those of Figs. 1.2 and 1.3, i.e., one which we call the “law of mixtures,” having two cases: (1) the parallel model, Fig. 1.4(a), and (2) the series model, Fig. 1.4(b).

In the parallel model, Fig. 1.4(a), a composite is composed of material 1 with ε_1 and material 2 with ε_2 which are aligned in a parallel manner, and the

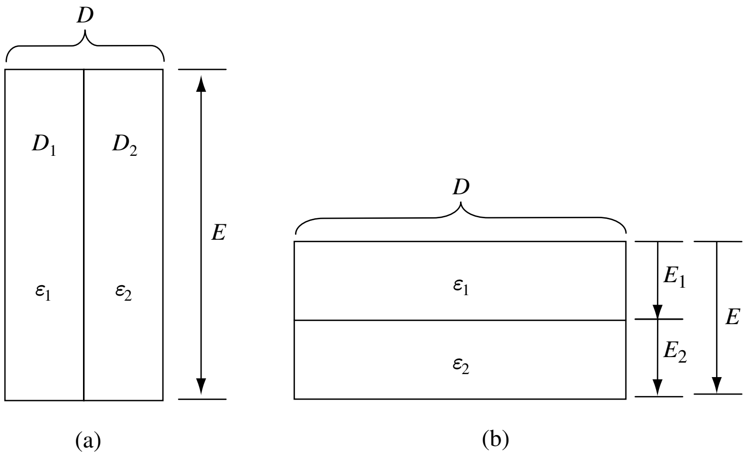

Fig. 1.4 Law-of-mixtures model: (a) parallel, (b) series.

electric field E is applied to the top and bottom surfaces of the composite in such a way that it is common to the two materials, i.e.,

$$D_1 = \varepsilon_1 E, \tag{1.25a}$$

$$D_2 = \varepsilon_2 E. \tag{1.25b}$$

The total electric flux density D_c of the composite is the volume-averaged sum of the electric flux density in each material.

$$D_c = (1 - f)D_1 + fD_2, \tag{1.26}$$

where f is the volume fraction of material 2 which is treated as "filler" here. The composite must obey the following constitutive equation:

$$D_c = \varepsilon_c E. \tag{1.27a}$$

A substitution of Eqs. (1.25) into Eq. (1.26), which is then substituted into Eq. (1.27a), gives the law-of-mixtures formula based on the parallel model,

$$\varepsilon_c = (1 - f)\varepsilon_1 + f\varepsilon_2. \tag{1.27b}$$

If the composite of Fig. 1.4(a) is subjected to an applied electric field E in the transverse direction, then one can create the series model, Fig. 1.4(b). In this case the electric flux density is continuous through materials 1 and 2; thus in the two materials

$$D_c = \varepsilon_1 E_1, \tag{1.28a}$$

$$D_c = \varepsilon_2 E_2. \tag{1.28b}$$

The applied electric field E is the volume-averaged sum of materials 1 and 2,

$$E = (1-f)E_1 + fE_2. \tag{1.29}$$

From Eqs. (1.27) – (1.29), one can derive the law-of-mixtures formula based on the series model,

$$\frac{1}{\varepsilon_c} = \frac{(1 - f)}{\varepsilon_1} + \frac{f}{\varepsilon_2}. \tag{1.30}$$

The law-of-mixtures formulae of Eqs. (1.27) and (1.30) are interpreted as the longitudinal and transverse dielectric constants, respectively, of a continuous fiber composite.