Modeling of Electrical Percolation Threshold of Carbon Graphite Composite

F. D. Senghor[1], G. Wasselunck[1], D. Trichet[1], and G. Berthiau[1]

[1]Institut de Recherche en Energie Electrique de Nantes Atlantique (IREENA), 37 Boulevard de lUniversit, BP 406, 44600 Saint-Nazaire Cedex, France

In this paper, a method to determine the effective electrical conductivity of composite constituted of two distinct phases with different physical property is presented. The conductive particles are randomly distributed in the thermoplastic polymer matrix. Homogenization method based on the equivalent representative volume is used to calculate the effective electrical conductivity. Results are then compared with analytical models based on the inclusion problem of Eshelby. The percolation threshold of the electrical conductivity is then discussed.

***Index Terms*—Composite materials, effective electrical conductivity, inclusion, homogenization, finite element method (FEM).**

I. Introduction

DUE to their excellent mechanical properties, carbon graphite composite materials are widely used in aeronautic applications. These materials are asked to meet new functionalities such as good electrical conductive properties to convey current and replace metallic material. The conception of these materials needs numerical modeling to evaluate the percolation threshold regarding the different distributions of phases. In this paper, the studied composite material consists of a polymer matrix and graphite inclusion of few tenth of micrometer. An example of such a composite is shown in Fig. 1. The electrical properties of these composite strongly depend on the volume fraction of inclusions and the random inclusions distribution in the micro structure. Moreover, as the matrix is poorly conductive, the electrical properties in these composite is closely related to the connectivity of the particles. Due to the high scale factor between the composite size and inclusion size, homogenization methods have to be used. These methods have to take into account the specific distribution of inclusions and the high electrical conductivity contrast between the inclusions and the matrix. The purpose of this paper is to deduce the effective electrical conductivity of a homogeneous equivalent material using the physical properties of these composites components. Moreover, the influence of the inclusions distribution and the electrical conductivity contrast will be studied.In this paper, homogenization approach is proposed using the method of equivalent representative volume. The results obtain by this method will be compared with analytical results given by Maxwell Garnett and Bruggeman model based on inclusion problem of Eshelby.

II. Proposed Modeling Methodology

Modeling of heterogeneous composite materials is very complex; therefore we propose analytical and numerical methods to model the effective electrical conductivity. The limitations and the potential problems of these methods will be distinguished.

A. Analytical Modeling

The most widely used analytical models to estimate the effective electrical conductivity of disordered mixture derived

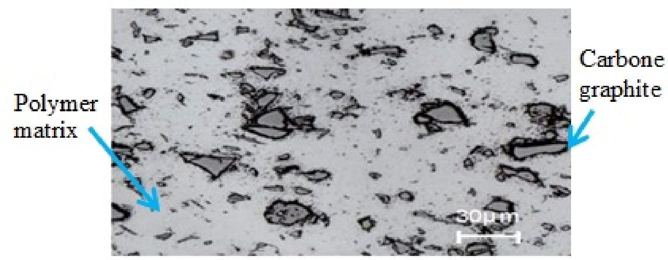

Fig. 1. Scanning electron microscope picture of carbon graphite mixed with thermoplastic matrix. Carbon graphite inclusions are the scale of micrometer.

from the inclusion problem Eshelby [1]. The method for homogenizing the inclusion problem are based on solving a submerged inclusion in an infinite medium, both isotropic electrical conductivity σ_i and σ_{ref} respectively. The electric field in the inclusion E^i (1) [2] is assumed linear, homogeneous and deducted from the field applied E^o

$$E^i = [(I + N_i\, \sigma_{ref}^{-1}(\sigma_i \,-\, \sigma_{ref})))^{-1}]\, E^o \tag{1}$$

The effective behavior of a heterogeneous medium can be defined through the definition of its current density $\overline{J}$ by equation (3).By overlaying the n problems of inclusion (Fig.2.), we deduce the effective electrical behavior of the composite by equation (2) [2].

$$\sigma_{eff} = \sum_{i=1}^{n} \left\langle (f_i \sigma_i [I + N_i\, \sigma_{ref}^{-1}(\sigma_i \,-\, \sigma_{ref})]^{-1}) \right\rangle \cdot \left\langle [(I + N_i \sigma_{ref}^{-1}(\sigma_i - \sigma_{ref}))^{-1}]^{-1} \right\rangle \tag{2}$$

$$\overline{J} = \sigma_{eff}\, \overline{E} \tag{3}$$

where N_i is the depolarization tensor, *I* the identity tensor, f_i the *i* inclusion volume fraction, and *n* number phases *i* (see Fig. 2.).

Therefore, with a judicious choice of the reference medium, the model can take into account the inter-inclusions interactions and theirs geometry. However, Maxwell Garnett [3], [4] and Bruggeman [5] models deduce in the inclusion problem, will be studied more closely.

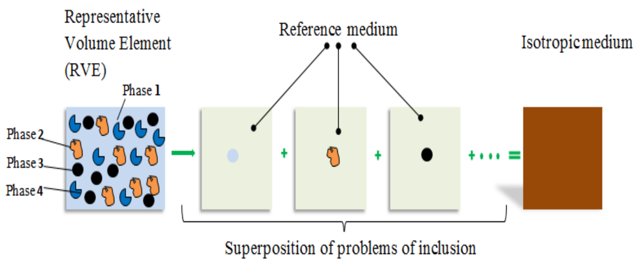

Fig. 2. Homogenization model based on problems of inclusion Eshelby.

B. Numerical Modeling

Due to the complex nature of composites (inclusion with different shapes and sizes, random distribution of these inclusions and large scale factor) direct modeling is impossible. It is then necessary to use a homogenization technique which takes into account these difficulties. The real material is divided into N representative volumes as shown in Fig.2a. The size of these volumes is selected to contain enough particles to be statistically representative of the overall volume while keeping an acceptable computational complexity. In order to generate a representative volume, it is meshed with regular elements of small sizes whose electrical properties are affected by a random algorithm (Monte Carlo) to satisfy the fill rate and law distribution. Fig. 2b shows an example of construction of a representative volume with a uniform distribution of the inclusion. The size of inclusion is a few tens of micrometers and the filling rate is 80%. This geometry generation algorithm is launched P times in order to have a satisfactory confidence interval. For each cells, electrokinetics finite element simulation is done to calculate the electrical conductivity by imposing a current source. The formulation is given by:

$$\begin{cases} \mathrm{div}([\sigma]\,\vec{\mathrm{grad}}(\mathrm{V}) = 0 & (\Omega) \\ [\sigma]\left(\dfrac{\mathrm{dV}}{\mathrm{dn}}\right) = \pm J_s & (\Gamma_1) \\ [\sigma]\left(\dfrac{\mathrm{dV}}{\mathrm{dn}}\right) = 0 & (\Gamma_1), \end{cases} \tag{4}$$

where σ is the electrical conductivity tensor, V the electrical potential and (Γ_1) the electrode boundary. The electrical conductivity is calculated knowing the source current, the cells dimension and the mean value of electrical potential in each electrode. The P simulations can to obtain the confidence interval of the electrical conductivity. In the final section the methodology evaluation of the size unit cell and the number of draws necessary P will be explained.

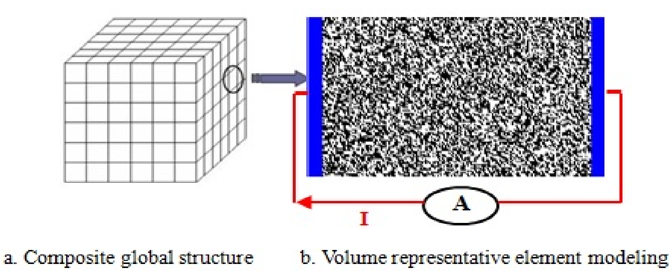

Fig. 3. 2D representation of the numerical model geometry (Black: inclusions, White: polymer matrix and Blue: the electrodes).

III. Results

The analytical and numerical methodology was applied to a two-phase heterogeneous micro structure. The phases are uniformly distributed with in the material and the mean size of particles is 10 micrometer. Fig.3 and Fig.4 show the effective electrical conductivities obtained for 10 simulations with random inclusions position for each volume fraction. Both variation of this volume fraction leads a low variation of electrical conductivity. After, the percolation threshold is reached; we can observe a high correlation between the volume fraction and the conductivities.

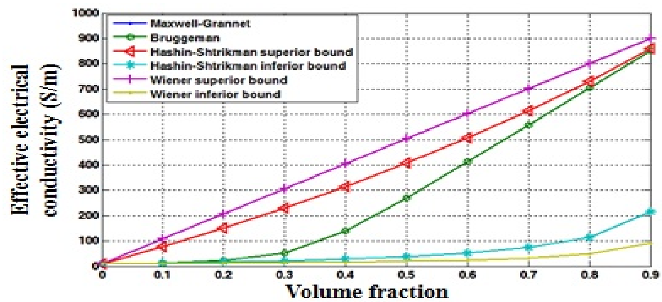

Fig. 4. Effective electrical conductivity and bounds as a function of volume fraction of theoretical mode.

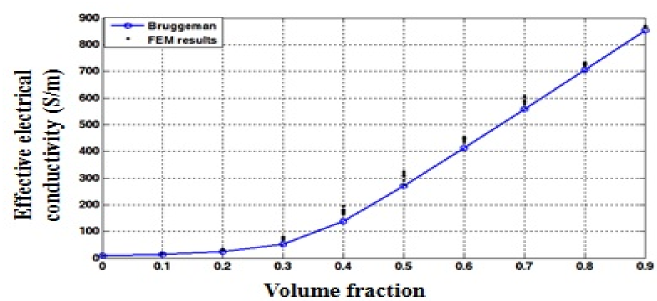

Fig. 5. Effective electrical conductivity as a function of volume fraction of Bruggeman theoretical model compared with FEM results.

In this paper, a homogenization method based on equivalent representative volume is proposed and give good results in comparison with analytical method. The effect of percolation threshold is perfectly simulated. The proposed method takes into account the specificities of composite materials: random distribution, large scale factor, different sizes and shapes of inclusions. In the final paper, the integration of different shapes and distribution of inclusions will be more detailed. In the case of elongated particles, the influence of their orientations will be discussed.

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