Electrical properties of metallic iron particle reinforced polymeric composite materials

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The paper aims to present the experimental research done on metallic reinforced polymeric composite materials with the aim of retrieving some electrical properties of the samples subjected to stress states: electric field, and temperature variation and mechanical load. The experimental curves will underline the temperature dependence of electrical resistance, as well as the variation of the latter with pressure. These curves will help material characterization and structural optimization from electrical point of view as well as identification of potential applications within the field of electric and electronics.

1. Introduction

As are well known a composite material represents a multiphase structure formed from a combination of materials that differ in composition or shape, remain bonded together and retain their identities and properties. Inside the mixture will always exist an interface between the constitutive components but they act in concert to provide improved specific and synergistic characteristics not obtainable by any of the original components acting alone.

The experimentally studies carried out by the authors represent a natural consequence of the extensive studies on different composite material structures, ranging from long or short, uniform or random distributed fiber reinforced composites or metal/nonmetal particle reinforced polymeric composites with the aim of mechanical, electrical, thermal properties retrieving.

The variations of electrical conduction with particle volume fraction, the intensity vs. voltage characteristic for different values of external presure and electrical resistance vs. temperature are being the major aspects approached experimentally.

2. Theoretical aspects on electrical conduction within the particle reinforced composite materials

2.1. Electrical conduction at room temperature

Technical literature provides several approaches on this phenomenon considering that the conduction process may take place due to different mechanisms [3-5]:
- percolation into a continuum network made up from conductive particles into electrical contact;
- electrical transition trough the dielectric spaces within the particles – a tunneling effect within the conductive particles electrical insulated one from the other.

In figure 1.a) is being represented a chain of spherical metallic particles for which the electrical connections are being represented by lines. The bold lines corresponds to the *d* spaces lower than the *R* medium values of the particles through which takes place a transfer based on the percolation phenomenon. The thin lines represent the separation of particles.

Technical literature provides several outstanding reviews of mixing laws and effective media theories for electro-composites, a "brand name" for the electrical conductive composite materials. In the dilute limit, all such equations are reduced to *Maxwell's* equation:

$$\frac{\sigma}{\sigma_m} = 1 + \frac{3 \cdot (r-1)}{r+2} \cdot V_p + higher\ order\ terms, \quad (2)$$

where σ is the conductivity of the overall composite material, σ_m is the conductivity of the matrix phase, r is the ratio of particle conductivity (σ_p) to that of the matrix, V_p is the volume fraction of particles, and the higher-order terms are neglected. The previous notations and symbols have the same meaning for all the following derived theoretical models.

The other models extend calculations beyond the dilute range. For example, the *Maxwell-Wagner* equation (also known as the *Maxwell-Garnett* equation or *Wiener*'s rule based on the well-known *Clausius-Mossoti* equation), is given by:

$$\frac{\sigma}{\sigma_m} = \frac{2 \cdot (r-1) \cdot V_p + (r+2)}{(r+2) - (r-1) \cdot r}. \quad (3)$$

This model is formally equivalent to the *Hashin-Strikman* lower bound (*conductive particles*) and upper bound (*insulating particles*) and sometimes is

referred to as the "*Maxwell&Wagner - Hashin&Strikman*" equation(s).

Bruggeman's asymmetric medium theory for *conducting spheres* is given by:

$$\frac{\sigma}{\sigma_m} = \left(1 - V_p\right)^{-3}, \qquad (4)$$

where as for *insulating spheres* the *Bruggeman*'s asymmetric equation is:

$$\frac{\sigma}{\sigma_m} = \left(1 - V_p\right)^{3/2}. \qquad (5)$$

Finally, *Meredith* and *Tobias* extended *Fricke*'s treatment of ellipsoidal particles, within a *Clausius-Mosotti* framework, by mixing half of the spheres at a given volume fraction, calculating the composite conductivity, and using this as the matrix for a new composite made with the addition of the other half of the spheres. The resulting equations are:

$$\frac{\sigma}{\sigma_m} = \frac{\left(1 + V_p\right) \cdot \left(2 + V_p\right)}{\left(1 - V_p\right) \cdot \left(2 - V_p\right)} \qquad (6)$$

for *conducting spheres* and

$$\frac{\sigma}{\sigma_m} = \frac{8 \cdot \left(2 - V_p\right) \cdot \left(1 - V_p\right)}{\left(4 + V_p\right) \cdot \left(4 - V_p\right)} \qquad (7)$$

for *insulating spheres*, respectively.

2.2. Electrical conduction vs. temperature variation

Electrical conductivity of composite materials function of temperature variation represents an indicator regarding the conduction mechanisms from activation energy point of view.
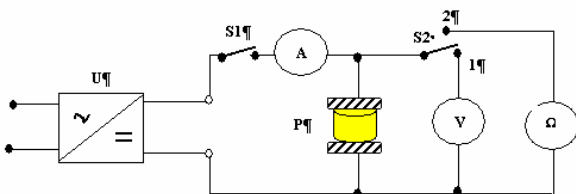

Fig. 2 The experimental measuring circuit.

The resistance variation can be considered the same as semiconductor materials:

$$R(T) = A \cdot \exp\left(\frac{\Delta W_i}{k \cdot T}\right) \qquad (8)$$

where $R(T)$ represents the resistance dependence function of temperature T [^{0}C], A is a constant, k - *Boltzman* coefficient and ΔW_i - the activation energy. The previous expression was adapted considering the experimental values. It can be observed that the variation is closer to the one provided in the literature for semiconductors.

3. Experimental research

The experimental research was carried out on several samples (cylindrical - D×H) of particle reinforced polymeric composite materials for which (see table 1):

▪ particles – conductive phase – metals (e.g. technical pure iron) embedded in different volume fraction (60..80 %); average size – 100 μm;

▪ polymeric matrix – two different polyester resins (Heliopol 9431 and Heliopol 2195), an epoxy vinyl ester resin (Derakane 411-350) and a silicone resin for different combinations and comparisons.

Table 1 The sample characteristics.

Sample	D [mm]	H [mm]	ρ [Ω m]	V_p[%] & resin typ
1	20	24	0.589	70% Fe (2195)
2	20	17.2	0.548	60% Fe (epoxy)
3	20	22	0.389	70% Fe (epoxy)
4	20	21	0.621	60% Fe (2195)
5	20	22	0.345	60% Fe (9431)
6	20	19.8	0.259	70% Fe (9431)

The experimental set-up (fig. 2) used for retrieving the following characteristics: voltage vs. intensity, resistance vs. intensity and resistance vs. temperature, respectively, consist of:

▪ adjusting continuous voltage source U (0..60 V/5 A);

▪ A, V, Ω – stands for ampere meter, voltmeter, and ohm meter measuring devices;

▪ P – composite sample;

▪ S$_1$, S$_2$ – interrupter, commutation;

▪ a temperature controlled owe.

4. Discussion

4.1. R(I) dependences

The electrical conductivity of the metallic particles reinforced polymeric composites depends on the current. This dependence is due in a large measure from the material self-heating. The equivalent resistance of composite

samples is decreasing along with an increase of applied current as can be shown in figure 3.
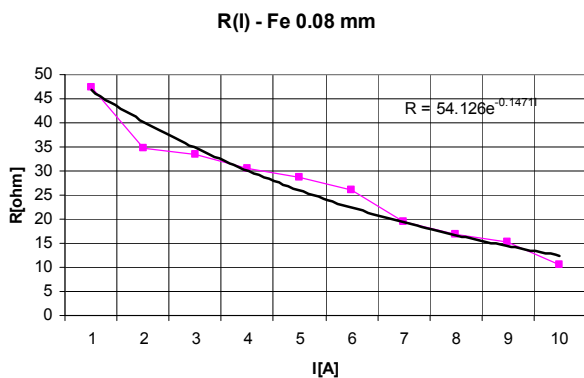
R(I) - Fe 0.08 mm

$R = 54.126e^{-0.1471I}$

Fig. 3

Another influencing factor on the electrical conductivity variation is due to the applied voltage. For small voltage values the voltage vs. current variation is approximately linear. For a certain value, called threshold voltage or critical voltage, the conductivity rises and the dependence become abruptly. The threshold value depends on the chemical composition and physical parameters of constitutive in the composite mixture and the phenomenon responsible for this deviation from linearity is due to *Joule* heating.

4.2. U(I) dependence

In figure 4 is being plotted the current vs. voltage dependence for two different composite samples having different volume fraction particles. The threshold value of voltage decreases along with the increase of particle volume fraction.
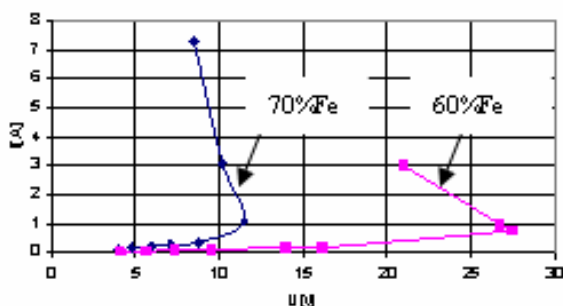

Fig. 4 The U(I) dependence for two different composite samples having 60% and 70% Fe particles.

Supplementary, during the experiments was underlined another influencing factor on the electrical conductivity of

particle reinforced polymeric composites, due to a reheating treatment.

This process was applied to the composite samples up to 200 ^{0}C with the purpose of stress release due to the external loads. The measurements lead to an increase with 20 % of the electrical conductivity values with respect to the previous. A possible explanation for variation can be regarded to the diminishing of the average distance between the particles of the composite structure.

Another important observation made during the experimental research is the fact that the intensity commutation phenomenon can be reversible if the composite's polymeric matrix will not breakdown and carbonized.

4.3. R(T) dependence

In case of the experiments concerning the electrical characteristics temperature dependence, the values reveal a decrease of resistance along with an increase of external applied temperatures (Fig.5). As was mentioned in 2.2 this behavior resembles the semiconductor materials behavior with two distinct ranges. The first temperature variation range goes up around 60 ^{0}C and the electrical resistance shows a slowly decrease with temperature, whereas for higher values (up to 100 ^{0}C) the decrease became abruptly.
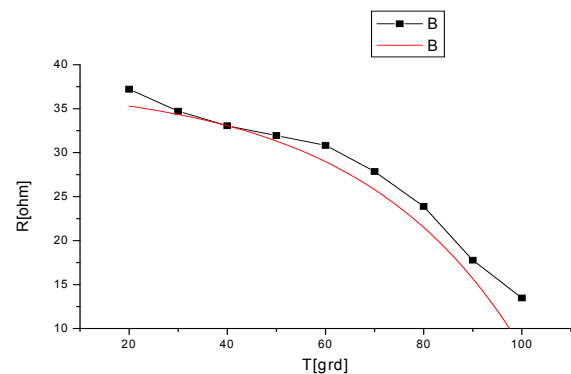

Fig. 5 R(T) dependence for a composite sample having 70% Fe particles.

This experimental dependence *R(T)* is in good concordance with the exponential relationship (8).

4.4. R(F) dependence

The variation of the electrical resistivity, R(F), with this external applied force were measured and plotted as can be seen in figure 6.

The electrical resistance was measured using the ampere meter-voltmeter method. For an epoxy matrix composite sample having 100 μm dimensions of the iron particles the relative variation of the electrical resistance function of applied force is presented in figure 7.
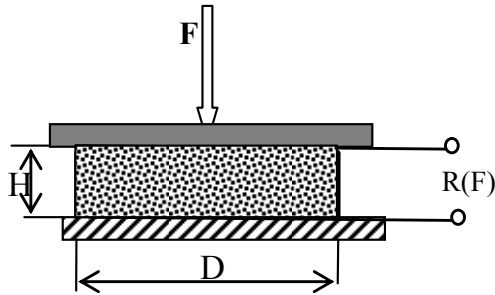
Fig. 6 Schematic layout of the experimental set up used for electrical resistivity measurements.
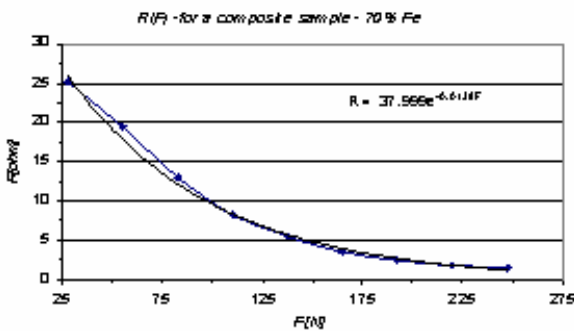

Fig. 7 R(F) dependence for 70% Fe particle – epoxy matrix

Also in this case is a exponential dependence as seen an approximation of the analytical function.

5. Conclusions

The electrical conduction within a composite structure is a complex phenomenon due to the fact it that takes place within a network made up from two phases having different electrical properties: conductive phase– metallic particles and the embedding phase – polymeric matrix. A particle reinforced composite material is electro-resistive, but its conductivity depends of multiple factors, like:

▪ material combinations (chemical compatibility, volume fractions, dimensions, etc.);

▪ manufacturing technology used and reheating treatments applied for stress releasing;

▪ voltage applied during the measurements;

▪ stress states applied on samples;

The experimental material characteristics will help material characterization and structural optimization from electrical point of view as well as identification of potential applications within the field of electric and electronics.

Acknowledgments

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