PREDICTING, MEASURING AND TAILORING ELECTRICAL PROPERTIES FOR NEW COMPOSITE MATERIALS USED IN AUTOMOTIVE SENSORS

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ABSTRACT - The paper focuses on a micromechanical approach for predicting the electrical conductivity of a new multiphase polymeric composite materials developed with the purpose of achieving improved electrical properties vs. reduced manufacturing costs for automotive sensors. The composites were developed by embeding different particle size, types and volume fraction of conductive particles into a polymeric matrix material.

The electrical property retrieved from measurements on several composite samples is the electrical resitivity, at low current densities, and its variation in time by the aid of a Wayne-Kerr analyzer. Simple mathematical manipulation will enable electrical conductivity estimation and further comparisons with the theoretical predicted values. Structural composition, particle distribution and boundary related aspects will aid electrical path characterization, percolation threshold identification and particle-particle combination tailoring measures.

1. INTRODUCTION

Polymer based composites have shown a large widespread due to the variety of polymers and the combinations with different types of compatible materials. Particle reinforced composites, in terms of material properties, were given attention for their outstanding and ability to tailor their thermo-mechanical properties. Electrical properties of composites, generally, and in particular of particle reinforced ones have began to be investigated quite recently [1-2]. Metal matrix composites (MMC) were emerged based on the needs to develop materials withstanding relatively high temperatures or to replace those composite structures reinforced by phases having an organic nature.

Metals are characterized by having material properties that recommends them as the best chose as matrix material: high mechanical properties, high thermal and electrical conductivity, dimensional stability, and good processing capacity.

A particle reinforced polymeric composite material is named electro-resistive if its conductive particles are being uniformly dispersed in the matrix material and the particle volume fraction exceeds a percolation threshold. In the literature is stated the fact that the transition from an insulating to a conducting composite as a function of filler concentration is named percolation and the critical concentration at which this drop occurs is called percolation threshold [3].

Particle reinforced polymeric composite materials are promising candidates for replacing the linear or dependent (e.g. temperature, electrical current or energy, mechanical pressure, etc.) resistive or electro-magnetically radiation attenuation elements. Because these materials are exhibiting electromagnetic properties may have different applications in electronics. In many paper this composite material class is being called “electronic composites”. Electronic particle reinforced composites, whose properties can be controlled by thermal or electromagnetic means, play an important role in micro- and nano-electromechanical systems (MEMS/NEMS) such as sensors, actuators, filters and switches. Some of them can be designed to exhibit shielding effects due to biological safety reasons as a natural consequence of the ever-growing applications of advances electrical and electronic devices.

The paper herein approaches a multiphase particles polymeric based composite combination in order to underline the previous mentioned statements and as a natural consequences of the authors research concerns and focused objectives toward developing composite structures attaining certain mechanical/electrical and thermal properties [3,4] - [8].

2. THEORETICAL APPROACHES

The conductivity of a composite material having an insulating matrix depends not only on the type of the filler, but also on the filler-matrix interface. A stronger interface is associated with a lower contact resistivity, resulting in a lower volume resistivity for the composite. Below the percolation threshold, the filler-matrix interface plays a particularly important role. The contact resistivity of an interface is a quantity that does not depend on the area of the interface or contact, but only on the nature of the interface. These statistical based theories generally assume a random filler particle distribution and furthermore do not include the possibility of particle movement through the matrix [5].

The electrical conduction processes in particle reinforced composite materials can be described using the micro contact theories developed around the electrical conduction phenomenon within the crystalline structure and the contacts that are being established within, case in which the current depends on the amount of energy flow and thermal effects at the grains level. In case of appearance of new conducting paths (contacts established between particles as the results of local thermal strains) within the composite structure causes a sudden increase of the measured electrical current. Since these contacts have a limited stability, transitions are fluctuating.

The electrical current through this structure presents fluctuations depending on the variation of electrical resistivity of the composite sample. The combination of these effects allows the retrieval of resistivity variation with the density of the conduction current or with the electrical energy that is being dissipated within the composite sample.

The contacts belonging to the conductive path are being formed between the composite's phases during the manufacturing steps (e.g. phases mixing, sample forming etc.). The microscopic dust is also being present among these contacts leading to shifts of the heat generated as consequence of electrical signal propagation. In general, metallic particle reinforced composites with concentrations exceeding the threshold associated to the phenomenon of electrical conduction, causes a resistivity decrease along with the increasing of the current density through the sample. This behaviour resembles the conduction mechanisms within the semiconductor materials [7].

3. EXPERIMENTAL RESEARCH

3.1. MATERIALS

The composite samples were manufactured using a self-developed manufacturing technology, by mixing together conductive particles (e.g. iron and black carbon) in different volume fraction, into a polymeric material namely polyvinyl acetate. The inclusions were characterized as having a medium size of 80 µm in case of the Fe particle and 10 µm in case of the carbon particles and were mixed to form a two or multi-phase structure. The samples were formed as having a cylindrical shape (diameter – 20 mm, height – 5 mm), 5 pieces for each class of composite materials as a minimum requirement in statistical data analysis. The composite classes are as follows:

- 80 % Fe, 20% polymer resin;
- 5% C, 75% Fe, 20% polymer resin;
- 10% C, 70% Fe, 20% polymer resin;
- 15% C, 65% Fe, 20% polymer resin.

In the previous, the polymeric resin is considered to include the chemical compounds contained into the additives added to help the polymerization process. The carbon particles were impossible to be mixed solely into the polymeric resin to form a sample by using the manufacturing technology developed and used herein.

3.2. EXPERIMENTAL RESEARCH

The experimental research aimed to retrieve the composite sample's resistivity vs. density current from the measurement set-up as well as resistivity vs. electrical energy transferred to the samples. Simple statistics was used to size the above variations and data analysis.

4. RESULTS AND DISCUSSIONS

In Fig. 1 is being presented a digital microscopic view (x500) for a composite sample manufactured and measured, namely a multiphase configuration made as a mix of Fe and C conductive particles embedded into the polyvinyl acetate matrix. As it can be seen, at this magnification level the Fe particles are clearly differentiable from the C particles. The latter are forming clusters and are contributing to the forming of conductive electrical paths.

In Fig. 2 is being plotted the electrical resistance variation with the current retrieved experimentally, for different polymeric composite structures. As it can be seen the variation can be approximated as an exponential decrease that may further explained by the phenomenon that takes place within the structure and which are characteristics to the class of extrinsic semi-conductive materials, actually explained as a Joule-Lentz effect.

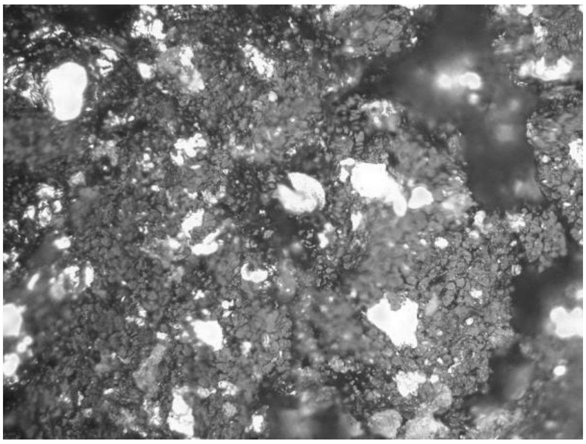

Fig. 1 Optical microscopic view (x500) for a 70% Fe, 10% C and 20% polyvinyl acetate polymeric composite material

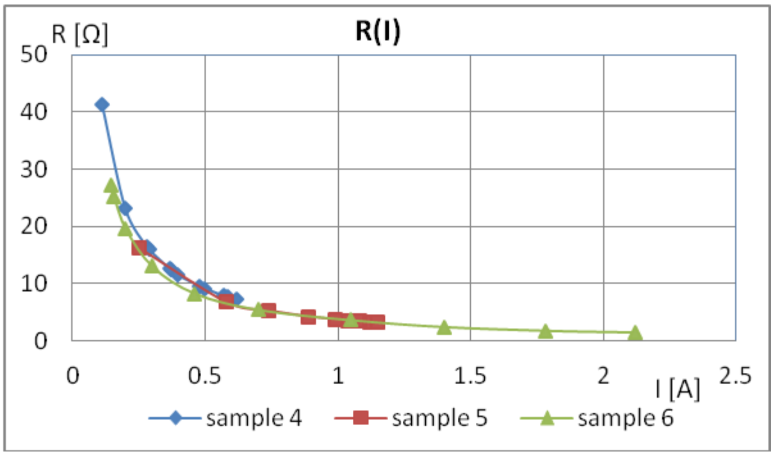

Fig. 2 R(I) dependence for conductive composite samples having different volume fraction of carbon and iron particles

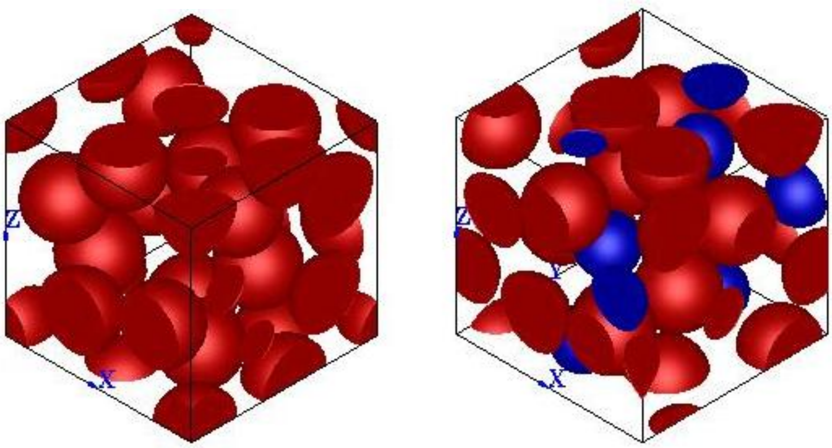

Fig. 3 Representative volume elements for random particle reinforced polymeric composite (left) and multiphase particle-particle reinforced polymeric composites (right)

In case of *adding carbon* (C) particles into the previous mixture – 80% iron (Fe) and 20% polymer – by keeping constant the matrix volume fraction (20%) and varying the particle content accordingly, the samples' resistivity are experiencing a modification along with this increase in carbon (C) particles volume fraction. This can be sized in the Fig. 4 where were plotted the resistivity variation vs. current density within a representative samples from each class of composite samples like the previous case, a two phase polymeric composite, the same exponential decrease occurs.

Moreover, the electrical resistivity decreases along with the decrease of the carbon (C) particles volume fraction, from 15% to 0%. This variation reveals the fact that the energy of the band gap is experiencing the same decrease with the decrease of the volume fraction of carbon particles and increase in the iron particles concentration.

Another influencing factor on the composites resistivity is the *electrical energy transferred* to the measured samples. The phenomenon depends again on the temperature shifts within the structure and time required to the electrical current to pass through the sample. At small values of energies ($W<10\ J/cm^3$) the mean values of the resistivity in case of multiphase composite having a higher concentration of carbon particles are small.

The fluctuating trend on the time dependence and electrical energies of the equivalent composite's resistivity (see Fig. 5) can be regarded to the variations of the resistance corresponding to the contact surfaces among the constitutive particles as well as to the equivalent resistance of the conductive particles.

During the reproducibility step of the measurement process, the electrical conduction process is more stabilized at the same values of the current densities (J), the fluctuation are attenuated and regarded to a temperature field more uniform at the particles' contact surfaces. Consequently, the resistivities values are experiencing an decrease with aproximately 10% and fluctuation's amplitute by half. These it can be seen in the Fig. 5, where the values were retrived for the same values of the current density (J=322 A/m^2) for a multiphase composite sample having the highest values of carbon particles considered in this experimental research.

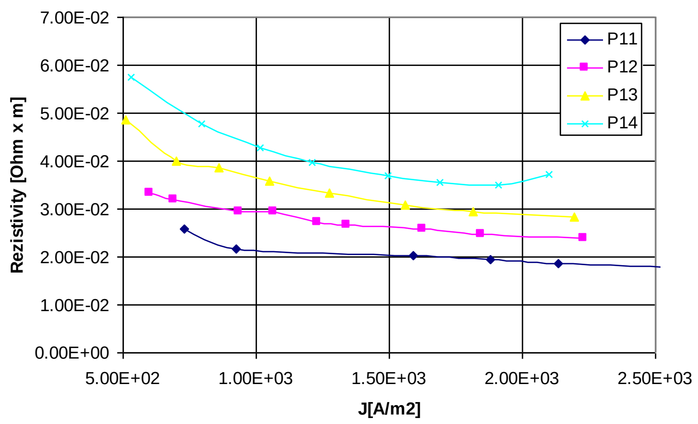

Fig. 4 Electrical resistivity variation with the current density (J) for a representative samples for each class of particle reinforced composites developed and tested

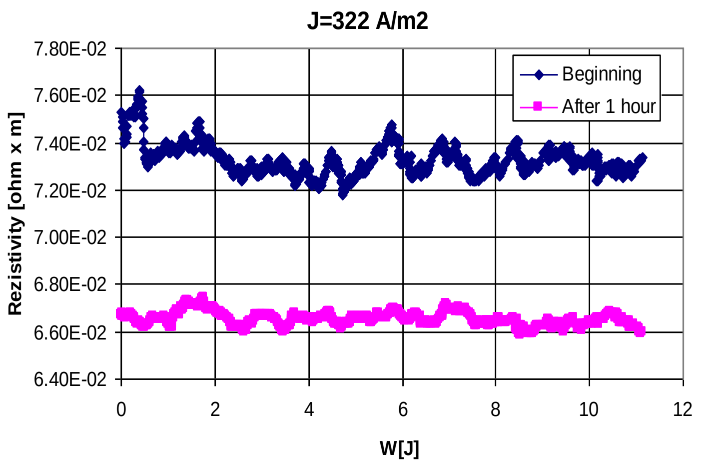

Fig. 5 Resistivity variation with the energy applied in case of the 65% Fe, 15% C and 20% polymer composite – to time instants (1 hour shift)

With respect to the micromechanical based prediction theoretical models, the ones encompassing the percolation factor are far the most comprehensive models to be used with the purpose of retrieving the electrical conduction or electrical resistivity properties of the composite materials under discussion. Authors of the herein paper will not present such theoretical variations or models based on the fact that they already done it in their previous work, but will draw the attention on a delicate issue they faced up during the studies on electrical behaviour of multiphase polymeric composite materials.

Among the micromechanical theoretical models the mean field homogenization concept is usually employed due to its characteristics and simplicity. Based on the representative volumes depicted in Fig. 3, a double inclusion method developed under the mean field homogenization concept was used to retrieve the electrical conductivity/resistivity of the composite samples under investigation, with and without a percolation threshold. In Fig. 6 are being given few of the theoretically retrieved values particular to the samples' electrical conductivity based on the double inclusion concept.

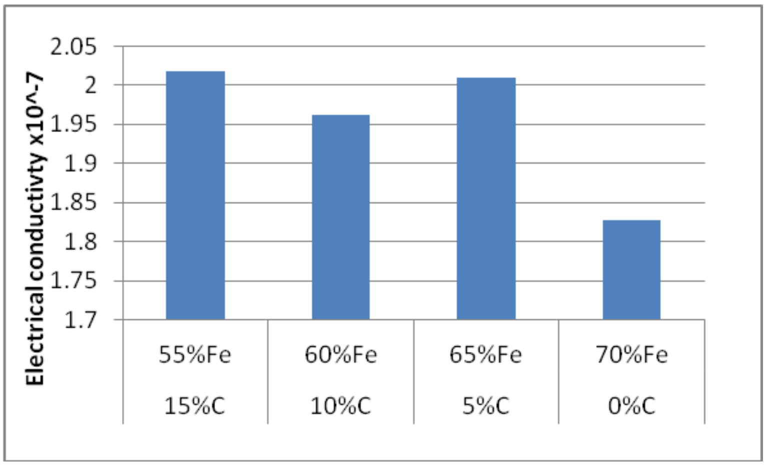

Fig. 6 Electrical conductivity vs. particles volume fraction based on double inclusion micromechanical approach

5. CONCLUSIONS

Particle reinforced polymeric composites characterized as having small particle dimensions (e.g. 100 ÷ 200 μm) and a metallic nature are showing similarities with semiconductor materials from electrical conduction point of view. Thus, their resistivity falls between $1 \div 10^2$ [Ωm], values that corresponds to the extrinsic semiconductor materials, and their resistivity temperature dependence are indicating an intrinsic semiconductor-like behaviour. The differences that arise are those revealed at high densities of the current through the composite structure, case in which the intermediate contacts are characterized by a certain tension that cause a abrupt decrease in the measured resistivity.

Based on data retrieved during the experimental measures of the electrical conduction it can be concluded that the structures of metallic particles reinforced composite materials can be used to manufacture resistive elements, as well as in other application areas as temperature sensors. The lately, can be assigned to the linear dependence of resistivity with temperature. Furthermore, controlling the particles volume fraction can be designed cheaper conductive elements, without loosing their intrinsic quality as conductive materials.

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