Nondestructive Evaluation of Metallic Fillers Embedded in Polyester Matrix

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Composites made from metallic fillers embedded in polymer matrix have good shielding properties as electrical point of view and tribology properties from mechanical point of view. These properties depend on amounts, shapes, sizes, and combinations of fillers and matrix. This paper proposes an electromagnetic method for determination of metallic fillers concentration in composite materials from metallic fillers embedded in polyester matrix. The bulk resistivity and dielectric permittivity were measured but these measurements are less influenced by the fillers concentration. The amplitude measurement of e.m.f induced in the reception coil of the focalized electromagnetic transducer and their averaging for the scanning of a relative large zone of the composite has proven to be an efficient method. The average amplitude linearly decreases with the increasing of fillers concentration, and has different values for Cu and AISI 316 fillers.

Keywords: Polyester, metallic fillers, composite, electromagnetic evaluation, S parameters

Polymer composites materials constitute a very active area for research in material science and nondestructive evaluation. The composites hold improved properties based on the advanced features of each individual component, sustaining new industrial applications which cannot be obtained based on individual components performances [1-3]. It is well known that the thermal and mechanical behavior of polymers is strongly influenced by factors such type, size, content and shape of the fillers [1-3]. Furthermore, these properties have a very complex dependence on the chemical structure and the crosslink density when thermoset resins and metallic fillers are combined [4, 5].

Polymer matrix composites containing conductive fillers are interesting for applications as shielding due their processability which helps to reduce or eliminate the seams in the housing that is the shield [6, 7]. The polymer matrix is commonly electrically insulating and does not contribute to shielding, though the polymer matrix can affect the connectivity of the conductive fillers; this is enhancing the shielding effectiveness. Metallic fillers such as copper, brass, iron, steel, aluminum etc. are added essentially in the various amounts, shapes, sizes, and combinations also in friction materials, [8-10]. These fillers not only improve the thermo-physical properties but also play an important role to enhance the tribo-performance. The use of fillers made from recycled materials increase the energy saving and solve few environmental issues [11].

This paper proposes an electromagnetic method for nondestructive evaluation of fillers concentration in polymer matrix, important feature in applications as construction materials - laminate plates from plastic-metallic fillers (PMF) composites colored by different procedures or friction materials in automotive industry.

Experimental part

Studied samples

Disks from PMF with 125mm diameter and 4.5mm thickness having the characteristics given in table 1 were taken into study.

The matrix is COLPOLY 7201 polyester, unsaturated resin with addition of styrene in amount up to 50%. Two types of samples are presented in figure 1.

Table 1

SAMPLE CHARACTERISTICS

Metallic fillers	Length [mm]	Diameter [mm]	Fillers content [mass %]
Copper	5	1	20
	5	1	30
	5	1	50
AISI 316	5	0.4	15
	5	0.4	35

Fig. 1. Studied samples: a) Cu filler, volume ratio 50%; b) AISI 316 filler, volume ratio 35%

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The fillers have been added by the producer in order to increase the flexural and tensile strength of the laminate [12] to improve thermal transfer during lamination when a considerable exothermic heating can occur, as well as for the increasing the fire resistance of the laminate.

For better characterization and determination of optimum methods for evaluation of fillers content, different types of measurements have been performed.

Bulk resistivity in DC was determined using a 25kV DC source - HV 350 - Amherst USA and a picoammeter Keithley 487 USA [13].

The properties in the range of radio frequency were determined by the measurement of S parameters into a coaxial guide that contains the samples of the composite material, using S Parameter Test Kit 87511A Agilent coupled at Network/Spectrum/Impedance Analyzer 4395A Agilent USA.

In order to minimize the effect of the loss and phase shift in the network between the sample and the measurement reference planes, the length of the 50Ω sample holder was the same as length of the sample, (fig. 2a). The length of the coaxial sample was 11.6mm and the diameter of the inner tube was 3mm.

Once the S – parameters are known (fig. 22b), the transmission and reflection coefficients T and respective R are computed, leading towards the determination of the electrical permittivity of the composite.

The following set of equations are used [14]

$$R = k \pm \left(k^2 - 1\right)^{1/2} \quad (1)$$

$$k = \frac{\left(S_{11}^2 - S_{21}^2\right) - 1}{2S_{11}} \quad (2)$$

$$T = \frac{S_{11} + S_{21} - R}{1 - \left(S_{11} + S_{21}\right)R} \quad (3)$$

$$\varepsilon = \frac{\frac{c}{\omega d}\ln\left(\frac{1}{T}\right)}{\sqrt{\frac{1+R}{1-R}}} \quad (4)$$

where c is the velocity of light in free space, ω is the angular frequency and d is the length of the material sample.

The electromagnetic measurements were effectuated using a special electromagnetic sensor designed in this purpose [15]. The emission part is made from a coil with 50 turns, wound on a ferrite cup core -18mm outer diameter and 5.5mm height. The reception coil has 50 turns and is wound perpendicularly on the emission coil.

The transducer is connected to the Network/Spectrum Impedance Analyzer, the relative displacing transducer-composite to be controlled being assured by a XY motorized stage Newmark USA (fig. 3). The command of equipment and acquisition of data are made by a routine developed in Matlab 2014a.

The determination of metallic fillers concentration by using the information regarding the number of fillers detected by electromagnetic measurement is not realistic due to the relative high number of fillers and their random distribution. Therefore, a more eloquent indicator might be the average value of the amplitude of e.m.f. induced in the reception coils of the electromagnetic transducer.

Results and discussions

In figure 4 is presented the dependency of the electrical resistivity logarithm of composite in function of the fillers concentration, for the two types of fillers considered.

A continuous decrease of the resistivity in function of fillers content can be observed; also a high resistivity is noticed, showing that the percolation does not appear nor at 50% Cu fillers concentration. Because the resistivity modifications are small and taking in consideration the simplicity of the measurement method, the processing/ interpretation implies precautions, as this measure cannot constitute an indicator of the filler content.

In figure 5a and b are presented the dependency of the dielectric permittivity (modulus) and the tangent of the loss angle (tanδ) by frequency using the methods of the coaxial cable and eqs. (1)-(4).

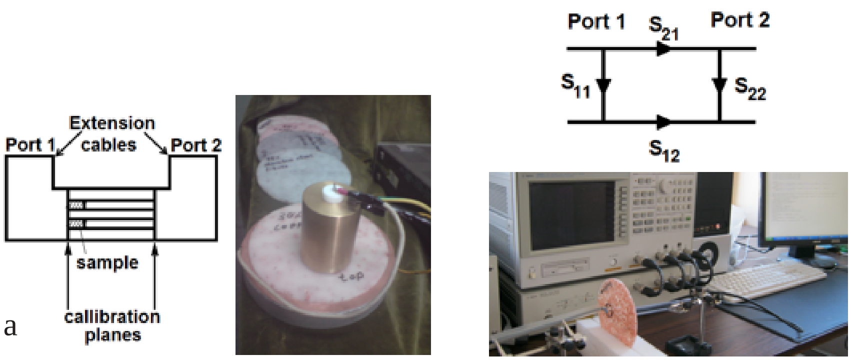

Fig. 2. Test fixture:
a) scheme and coaxial sample holder;
b) S-parameter flow graph and experimental set-up

Fig. 3. The experimental set-up

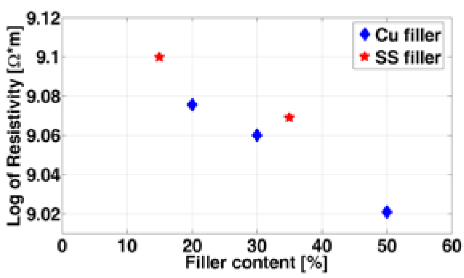

Fig. 4. Electrical resistivity of composite *vs* filler content

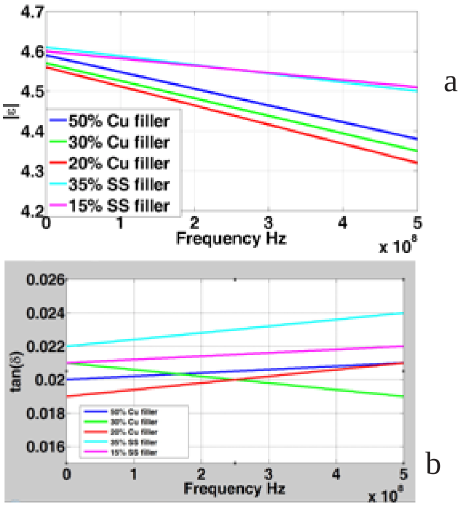

Fig. 5. Dielectric permittivity *vs* frequency: a) modulus; b) tanδ

The analysis of data from figure 5 shows a decrease of the permittivity modulus with the frequency, in a linear trend. The permittivity has high values in the case of AISI 316 fillers composite and slightly lower for the Cu fillers. The value of tand is practically constant with the frequency in the range 100kHz-500MHz.

The electromagnetic measurements allows the visualization of metallic fillers from the studied composite materials, the resolution being good enough at 2MHz frequency, as presented in figures 6a and b for the case of Cu 50% fillers content.

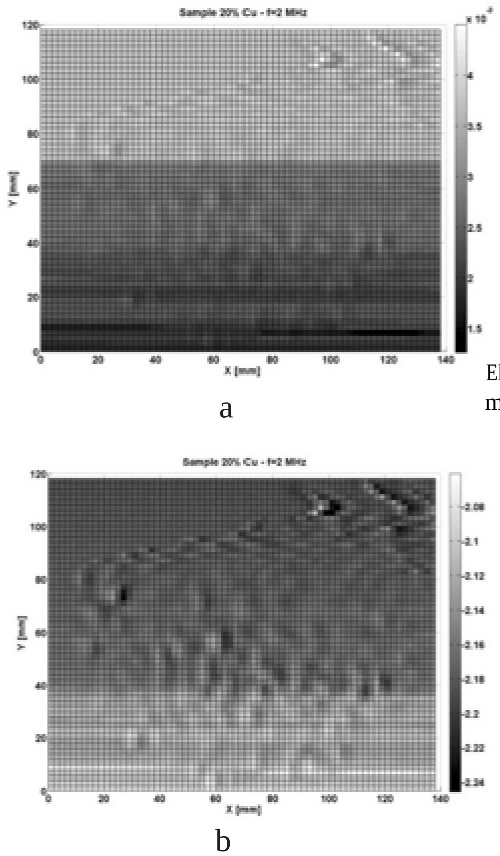

Fig. 6. Electromagnetic measurement at frequency of 2MHz: a) amplitude; b) phase

Even at a thin scan steps (e.g. of 0.2mm), due to the fact that some fillers can be screened by others, the count of fillers is an operation accompanied by large errors; therefore this type of measurements cannot be used as indicator of fillers content in the examined composite.

A much simpler and more efficient method is represented by the mean value of the e.m.f. amplitude induced in the reception coil at a frequency conveniently chosen, when the transducer scans a relatively large area of the composite.

In figure 7 is shown the dependency between the average values of the e.m.f amplitude induced in the reception coil of the electromagnetic transducer by the concentration of fillers when it scans a region of 70x70mm from composite with the step of 0.2mm in both directions at 0.2mm lift-off.

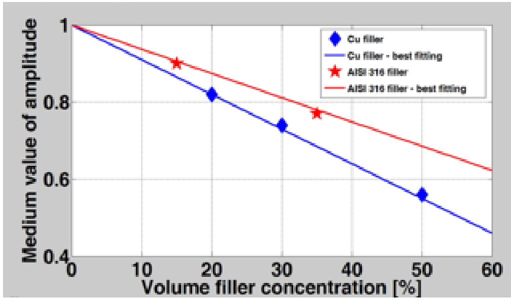

Fig. 7. Dependence of average value of e.m.f. amplitude induced in the reception coil of the transducer with orthogonal coils by the fillers concentration

The work frequency was 500kHz. It can be observed that the dependency is practically linear in both the case of Cu fillers and AISI 316 fillers. The value has been normalized to the amplitude of e.m.f. induced in the reception coil when the transducer is placed on a sample of COLPOLY 7201 polyester resin.

It is observed that the e.m.f. induced in the reception coil decreases more rapidly with the increase of the concentration in the case of Cu filler than in the case of AISI 316 fillers. For fillers concentrations that exceed 70%, the percolation phenomenon is possible to appear and the proposed methods is not efficient anymore.

Conclusions

The bulk resistivity, as well as dielectric permittivity measurements shows that these measures are relatively less influenced by the concentration of Cu and AISI 316 fillers having 1mm and respective 0.4mm diameter and 5mm length. The value of tand practically does not vary with the filler concentration.

The electromagnetic measurements using focalized transducer allow the visualization of the metallic fillers in composite. Due to the effect of shielding of the neighboring fillers, the filler count from the electromagnetic images can lead to large errors in the determination of fillers concentration.

The average value of the amplitude of e.m.f induced in the reception coil can serve as indicator of the fillers concentration, this decreasing with the increasing of the concentration.

Further studies will imply determination of mechanical properties in order to use the composite plastic-metallic fillers as friction materials.

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