

# C/P carbon composites - reinforcement volume effect on the electrical properties

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**Abstract.** Transmission of electrical charges in polymer composites depends not only on quality of their structure but also on electrical properties of individual components. That are a lot of possibility of fillers composites, for example chopped or milled carbon fibres, over multiwalled carbon nanotubes, singlewalled carbon nanotubes, graphene nanopellets and carbon black or fullerenes. The advantages of milled carbon fibres are affordability of starting materials. The main goal of the paper is analysis of densification influence on electrical properties of fabricated samples. All data were statistically tested for statistical significance and their interdependencies.

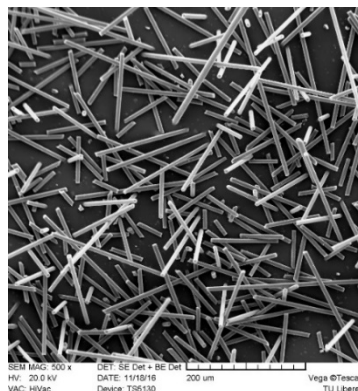
## 1. Introduction

Carbon fibres are very appropriate as electrically conductive fillers composite. This study examines the influence of the concentration milled carbon fibres of electric properties of epoxy composites. Current electrical properties indicate changes in structure fibres, which could be an interesting indicator in case of a degradation of C/P composites.

## 2. Experimental

### 2.1 Material

The epoxy resin system, MGS LR 285 (epoxy)/ H 508 (hardener), mixing ratio 100:40 by weight [2]. One specimen was neat epoxy the others were filled with carbon particles (CF) in following concentrations: 0,5wt%, 1wt%, 1,5wt%, 2wt%, 2,5wt%, 3wt%, 3,5wt% and 4wt%. The used fillers were commercially available Carbiso Milled Carbon Fibre with average diameter about 7 µm, and average length 100 µm [3], see Figure1.



**Figure 1.** Milled Carbon Fibres used as an epoxy fillers.



The mixture was stirred at room temperature for 10 mins by magnetic stirrer Hanna HI190, then cured first for 24 hours at room temperature and followed by curing for 15 hours in 60°C.

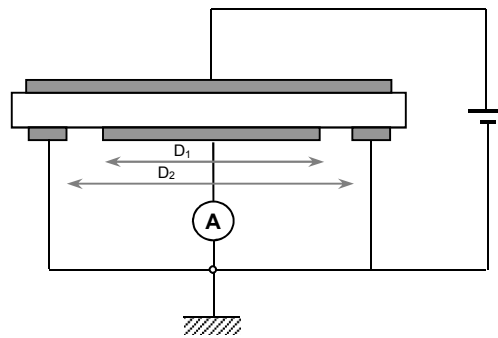
The samples were painted with ELEKTRODAG<sup>®</sup> 1415 silver film (Agar scientific) for maximum adhesion and conductivity.

## 2.2 Method of measurement

Surface conductivity of specimens was analysed using HP 4339B High Resistance Meter, which is designed for measuring very high resistances and related parameters of insulation materials. Materials were measured according to standard IEC 61340-2-3:2000 "Electrostatics - Part 2-3: Methods of test for determining the resistance and resistivity of solid planar materials used to avoid electrostatic charge accumulation", the test voltage was 100 V. Surface Resistivity  $\rho_s$  is defined as resistance per unit surface area. When the 16008B Resistivity Cell is used surface resistivity is given as:

$$\rho_s = \frac{\pi(D_1 + D_2)}{(D_1 - D_2)} R_s \quad (1)$$

where  $D_1$  is main electrode diameter,  $D_2$  is guard electrode diameter and  $R_s$  measured surface resistance.



**Figure 2.** Principle of measurement Surface Resistivity.

In this case, the conductivity is calculated using the following expression:

$$\sigma = \frac{1}{\rho_s} \quad (2)$$

Ac conductivity and dielectric properties samples were measured using AGILENT 4294. The measurements were carried out in the frequency range 100 Hz – 3MHz. A precision analyser was used to measure the sample capacitance  $C$  and the loss tangent  $\tan \delta$  directly. The total conductivity  $\sigma_{AC}$  was calculated from the equation

$$\sigma_A = 2\pi \epsilon_0 \epsilon'' \quad (3)$$

where,  $f$  is the frequency and  $\epsilon_0$  free space permittivity. The dielectric loss  $\epsilon''$  was calculated from the relation

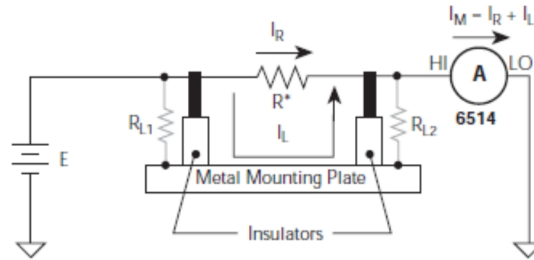
$$\epsilon'' = \epsilon' \tan \delta \quad (4)$$

where  $\delta = 90^\circ - \Phi$ ,  $\Phi$  is the phase angle which has been measured using the same bridge. The permittivity was calculated using the relation

$$\varepsilon' = \frac{c}{\varepsilon_0 S} \quad (5)$$

where  $d$  is the sample thickness and  $S$  the sample cross-sectional area.

DC Conductivity was measured by electrometer Keithley 6514. The conductivity was taken with the frequency at 50 Hz. The two opposite surfaces were silver-painted and tightly pressed with electrodes. The DC conductivity was characterized by two-point method described in [4], respectively.



**Figure 3.** Measurement of DC conductivity [4].

$$\sigma_D = \frac{I_M 4d}{U D^2} \quad (6)$$

The  $R_{L1}$  and  $R_{L2}$  represent the leakage resistance of sample. Measuring were the leakage current  $I_L$  and the current  $I_R$  through sample. Where  $R^*$  is resistance of sample.

Permittivity of specimens was analysed by RCL meter INSTRON-821. The frequency of measurement was 1 kHz according to IEC 62631-2-1 ED. 1.0. "Dielectric and resistive properties of solid insulating materials - Part 2-1: Relative permittivity and dissipation factor - Technical Frequencies, AC Methods". The permittivity was calculated from the equation (5).

### 3. Data analysis

In this section, there have been provided statistical algorithms and tools for organizing, analysing and modelling data. Then it has been used regression for predictive modelling, statistical plots for exploratory data analysis and regression analysis. For analysing multidimensional data there were used partial least-squares regression including analysis of variance (ANOVA), correlation maps and descriptive statistics. We took statistical visualization to convey essence and to allow for further processing. Therefore, it has been applied assumption of the normal distribution and estimation of statistical characteristics of random statistical selection [11].

#### 3.1 Theoretical background

**3.1.1 Coefficient Covariance and Confidence Intervals.** Estimated coefficient variances and covariances capture the precision of regression coefficient estimates. The coefficient variances and their square root, the standard errors are useful in testing hypotheses for coefficients. Covariance quantifies the strength of a linear relationship between two variables in units relative to their variances. Correlations are standardized covariances, indicating a dimensionless quantity that measures the degree of a linear relationship, separated from the scale of either variable.

The coefficient confidence intervals provide a measure of precision for linear regression coefficient estimates. A  $100(1-\alpha)\%$  confidence interval indicates the range that the corresponding regression coefficient will be at  $100(1-\alpha)\%$  of confidence [11].

**3.1.2 Correlation Maps.** Correlation quantifies the strength of a linear relationship between two variables. When there is no correlation between two variables, then there is no tendency for the values of the variables to increase or decrease in tandem. Two variables that are uncorrelated are not necessarily

independent, however, because they might have a nonlinear relationship. You can use linear correlation to investigate whether a linear relationship exists between variables without having to assume or fit a specific model to your data. Two variables that have a small or no linear correlation might have a strong nonlinear relationship. However, calculating linear correlation before fitting a model is a useful way to identify variables that have a simple relationship [11].

**3.1.3 Analysis of Variance.** Analysis of variance (ANOVA) is a procedure for assigning sample variance to different sources and deciding whether the variation arises within or among different population groups. If variations within groups are small relative to variations between groups, a difference in group means may be inferred. Hypothesis tests are used to quantify decisions.

The purpose of two-way ANOVA is to find out whether data from several groups have a common mean. One-way ANOVA and two-way ANOVA differ from that the groups in two-way ANOVA have two categories of defining characteristics instead of one. Two-way ANOVA is a special case of the linear model. The two-way ANOVA form of the model is

$$y_{ijk} = \mu + \alpha_j + \beta_i + \gamma_{ij} + \varepsilon_{ijk} \quad (7)$$

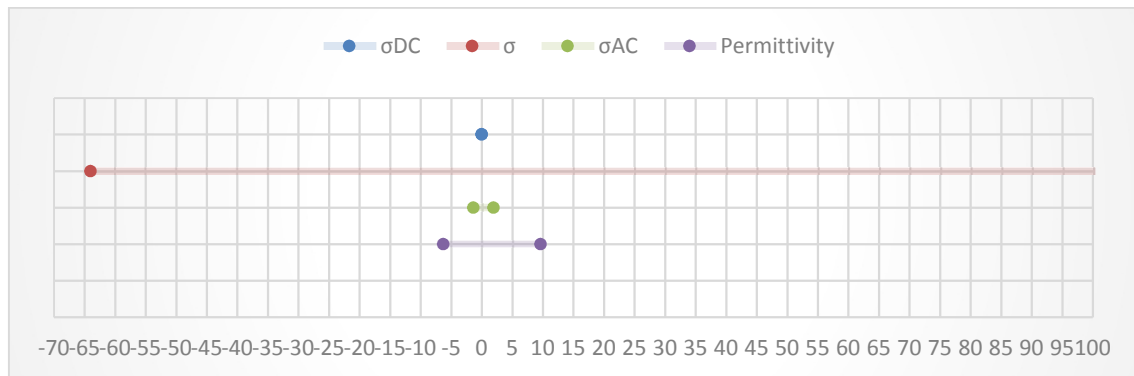
where  $y_{ijk}$  is a matrix of our observations (with row index  $i$ , column index  $j$ , and repetition index  $k$ ),  $\mu$  is a constant matrix of the overall mean,  $\alpha_j$  is a matrix whose columns are variables,  $\beta_i$  is a matrix whose rows are the deviations of each variables,  $\gamma_{ij}$  is a matrix of interactions. The values in each row of  $\gamma_{ij}$  sum to 0, and the  $\varepsilon_{ijk}$  is a matrix of random disturbances [11].

#### 4. Results, Related Discussion

**Table 1.** Measured data of electrics properties

Volume fraction (%)	$\sigma_{DC}(10^{-10}\text{Sm}^{-1})$	$\sigma_{AC}(10^{-5}\text{Sm}^{-1})$	$\sigma(10^{-15}\text{S})$	Permittivity (-)
0.0	0.004	1.71	45	4.23
0.5	0.007	1.10	34	5.48
1.0	0.006	1.23	73	5.87
1.5	0.010	1.33	128	6.33
2.0	0.010	1.72	117	8.02
2.5	0.011	2.35	100	10.57
3.0	0.018	1.65	6335	10.67
3.5	0.019	2.18	9892	10.07
4.0	0.021	1.67	9734	10.50

Table 1 contains a summary of the electrical properties for all the samples studied. These data allow a comparison between neat and filled epoxy.



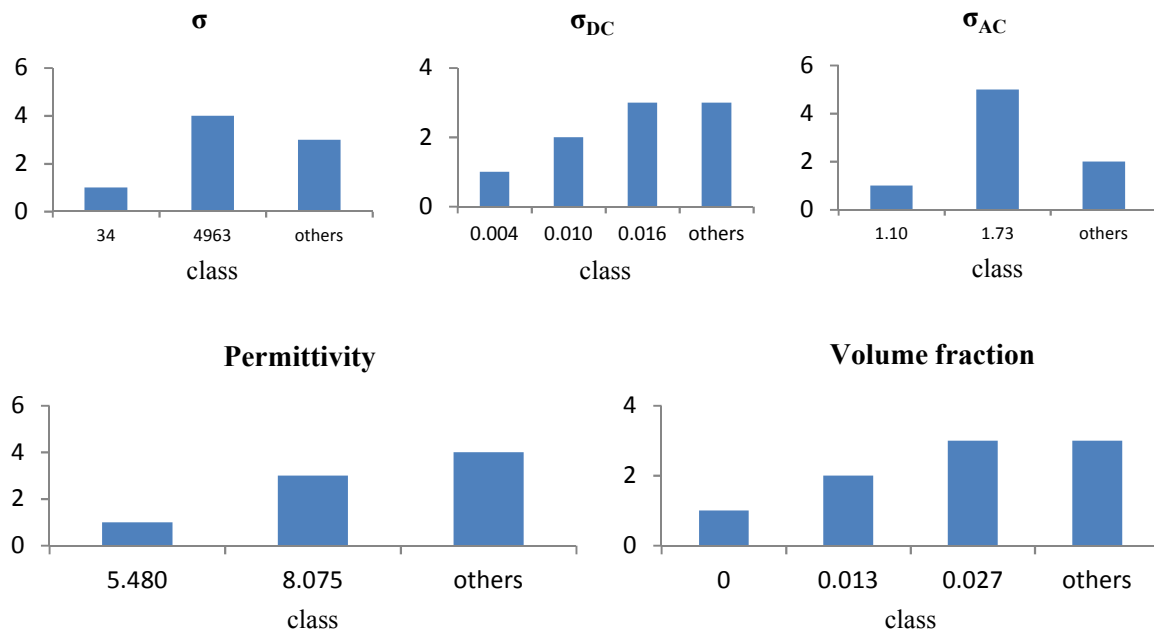
**Figure 4.** Calculated confidence intervals of mean estimate.

From the results shown on Fig. 4 it is clear all confidence interval are in interaction and all measured data are, from first point of view, statistical insignificant with recommendation to eliminate  $\sigma$  data.

**Table 2.** Covariances

	Volume fraction (%)	$\sigma_{DC}(10^{-10} \text{ Sm}^{-1})$	$\sigma(10^{-15} \text{ S})$	$\sigma_{AC}(10^{-5} \text{ Sm}^{-1})$	Permittivity
Volume fraction (%)	0.00017				
$\sigma_{DC}(10^{-15} \text{ Sm}^{-1})$	7.29111	3.39025			
$\sigma(10^{-15} \text{ S})$	44.90333	22.06650	17222509.78		
$\sigma_{AC}(10^{-5} \text{ Sm}^{-1})$	0.00274	0.00097	573.71	0.15113	
Permittivity	0.02927	0.01236	6902.78	0.61543	5.79452

According to the results on Fig.4 are shown results on Tab 2, only  $\sigma$  data are different, which implies elimination  $\sigma$  data.



**Figure 5.** Histograms with data tendencies.

On Fig. 5 we can see data histograms with tendencies to suggested distributions. It is appropriate to verify assumption of normal distribution for volume fraction,  $\sigma_{DC}$ , and permittivity.

**Table 3.** Correlation maps

	Volume fraction (%)	$\sigma_{DC}(10^{-10} \text{ Sm}^{-1})$	$\sigma(10^{-15} \text{ S})$	$\sigma_{AC}(10^{-5} \text{ Sm}^{-1})$	Permittivity
Volume fraction (%)	1				
$\sigma_{DC}(10^{-15} \text{ Sm}^{-1})$	0.97	1			
$\sigma(10^{-15} \text{ S})$	0.84	0.91	1		
$\sigma_{AC}(10^{-5} \text{ Sm}^{-1})$	0.55	0.43	0.36	1	
Permittivity	0.94	0.88	0.69	0.66	1

Table 3 contains correlations map including correlation coefficients, only  $\sigma_{AC}$  has weak dependence on volume fraction. For additional analysis is important to analyse relationship between dependent variables ( $\sigma_{DC}$ ,  $\sigma_{AC}$ ,  $\sigma$  and permittivity) with focus on  $\sigma_{AC}$ .

**Table 4.** ANOVA 2N for AC conductivity

Source	SS	dif.	MS	F	p-value	F krit
rows - frequency	225.086	6	37.514	145.506	4.7258E <sup>-29</sup>	2.295
columns-vol. fraction	9.102	8	1.138	4.413	0.0005	2.138
error	12.375	48	0.258			
total	246.563	62				

Table 4 contains Analysis of Variance for measured data AC conductivity. For frequencies -  $F > F_{krit}$  and  $p\text{-value} < 0,05$ , it follows null hypothesis about mean values concordance isn't true and some of frequencies level is statistical important for AC conductivity. For volume fraction of reinforcement is the same interpretation, null hypothesis about mean values concordance isn't true and some of volume fraction levels is statistical important for AC conductivity. According to the fact volume fraction are equidistant and frequencies not, add focus to volume fraction. It is important do more levels of volume fraction and adjust.

## 5. Conclusion

According to literature [6-10] is obvious that there are two interdependent structures that co-exist in different dimensions and characteristics, respectively. We made a new composite where one-dimensional CF acted as bridges was attributed to the formation of a strong three-dimensional network structure in epoxy matrix. Furthermore, the conductivity of C/P composites was enhanced due to the formation of 3D conductive pathways from CF within the matrix.

The strategy of adding recycled CF into insulated polymers refine electrical conductivity may provide a potential route towards novel functional materials.

Descriptive statistics and confidence intervals did not show statistical significance of the measured data. Statistical analysis ANOVA demonstrated that they are statistically significant, therefore it is advisable to continue in a more detailed statistical testing, e.g. multiple comparisons.

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