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Multifunctional material design for strain sensing: carbon black effect on mechanical and electrical properties of polyamides

Alberto Ciampaglia^{a,*}, Raffaele Ciardiello^a, Federico Cesano^b, Giovanni Belingardi^a,
Valentina Brunella^b

^a*Department of Mechanical and Aerospace Engineering, Politecnico di Torino, Turin, Italy*

^b*Department of Chemistry, Università di Torino, Turin, Italy*

**Corresponding author*

Email address: alberto.ciampaglia@polito.it (A. Ciampaglia)

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Abstract

This work investigates the coupled effect on the mechanical and electrical properties of carbon black (CB) dispersed in polyamide 6,6 matrix. The amount of CB added to the polymer matrix was varied between c.a. 5 and c.a. 25 wt %. The presented results reveal the capability of conductive carbon nanostructured particles, known as carbon black, to functionalize the thermoplastic polymers by activating conductive percolation networks, as obtained over the percolation threshold. The electrical resistivity of the composite compounds under static and dynamic conditions was determined. In this domain, the

22 variation of the electrical properties with the applied stress, is assessed through
23 mechanical tests with in-situ electrical measurements. The conductivity sensitivity of
24 reinforced composites to the applied strain is analyzed within direct (DC) and alternate
25 current (AC) regime, revealing the effect of frequency on the composite strain sensitivity.
26 An investigation on the morphology of particle dispersion at the microscopic level
27 highlights the role played by microstructural arrangements on the homogenized bulk
28 properties of composites. Mean field homogenization models for the estimation of
29 composite stiffness are compared to the experimental results.

30

31 **1 Introduction**

32 Polymer matrix composite (PMC) materials are gaining high interest in the industry
33 and scientific community: their unique properties have paved the way to their usage for
34 numerous applications. However, polymers are insulating, meaning that cannot conduct
35 current. During the last decade, researchers have proved the capability of carbon
36 nanoparticles of modifying the properties of PMC even at the macro-scale. In this regard,
37 great accomplishment can be found in several characteristics, including stiffness
38 enhancement(1–4), strength increase(5), induced plasticity(6), toughening
39 mechanism(7,8), electrical conductivity(9–11), strain sensing (12–23). The evolution of
40 the micro-mechanic alongside with material science knowledge has opened the doors to
41 a boundless portfolio of materials, which properties can be tailored on the design
42 specification by properly defining material constituents and architecture from the nano
43 to the micro scale. The spread of these “smart materials” has not dispensed industrial
44 sectors such as aerospace(24) or automotive(25,26), where functional material can help

45 designers cutting costs, producing lighter structures, and adding new functionalities as
46 structural health monitoring(27–30) (SHM). SHM is an inverse technique aiming at
47 assessing the health state of a structure by processing signals from sensors mounted on
48 the monitored part. The adoption of innovative materials (fiber-reinforced composites,
49 sandwich structures, multi-material laminates), for which the study on fatigue response
50 and damage tolerant design have not been consolidated yet, put the requirement for a
51 reliable, but affordable and lightweight SHM. In accomplishing this, smart self-sensing
52 materials are ideal candidates. Indeed, self-sensing materials shows a change in the
53 electrical conductivity when a stress (or strain) is applied; this phenomenon is usually
54 activated by highly conductive particles (e.g., carbon nanotube, carbon fibers) dispersed
55 into the polymer(31).

56 The adoption of such materials in SHM allows monitoring large structures without
57 any sensor: every point of the smart material structure is monitorable and the cost
58 increase only relies on the functionalizing particle addition to the neat polymer. The goal
59 of this work is to comprehensively study the effect of Carbon Black (CB) particles loading
60 on the mechanical, electrical and strain sensing properties of polyamide 6 and polyamide
61 66 (PA6 and PA66, respectively). First, we manufactured CB-PA composites with
62 injection molding as described in Section 2.1. In Section 2.2 particles concentration and
63 morphology are analyzed with Scanned Electron Microscopy (SEM) and
64 Thermogravimetric Analysis (TGA), while the percolation threshold of CB reinforced PA
65 is established; in Section 3.1, the effect of particles concentration on the stiffness and
66 strength of composites is experimentally investigated through tensile tests and correlated
67 with different homogenization models. In Section 3.3, we present the results of tensile

68 tests with in-situ resistance and impedance measurements on conductive and propose a
69 novel model for the estimation of the material Gauge Factor (GF) variation with the
70 applied electric frequency. At the best of authors knowledge, this work is the first
71 revealing the effect of the excitation current frequency on the self-sensing material gauge
72 factor.

73

74 **2 Materials and methods**

75 **2.1 Materials**

76 Composite specimens were manufactured starting from commercial PA6/CB
77 masterbatch (Cabelec CA3178, polyamide 6 with 27,5 wt.% of CB, MFI= 10 g/10 min at
78 275°C) with addition of PA6 (polyamide 6 TECHNYL C 206, Solvay) or PA66 (polyamide
79 66 TECHNYL A 205F, Solvay). Polymers and the compound were melt-compounded by
80 using a Babyplast 6/10P micro injection molding machine (Rambaldi srl, Italy) having a
81 type-V tensile test specimen mold (ASTM D638). Prior to use, the polymers and
82 masterbatch were dried at 80°C for 4 h, while barrel, injection, nozzle, and mold
83 temperatures were set at 255, 250, 245, and 60°C for PA6, or 280, 275, 270 and 70°C for
84 PA66 compositions, respectively.

85

86 **2.2 Experimental tests**

87

88 *2.2.1 TGA*

89 Thermogravimetric measurements were performed with a TA Q500 instrument, TA
90 Instruments, by using c.a. 8-10 mg of sample. The temperature was increased from 50 °C
91 to 700 °C under nitrogen (N₂) gas flow, then to 800 °C in air (heating rate: 10 °C/min in
92 both steps). This method was adopted to determine the polymer and CB contents after
93 compounding.

94

95 *2.2.2 SEM*

96 Field-emission Scanning Electron Microscopy (FeSEM), Tescan Mira 3 (Tescan, Brno,
97 Czech Republic) was used to assess the particle distribution of the specimens and the
98 corresponding fracture surfaces. An accelerating voltage of 5 kV was used together with
99 a secondary emission signal. The specimen surfaces were not coated with metals since
100 the specimens were electrically conductive and to avoid possible contaminations.

101

102 *2.2.3 DC electrical properties*

103 DC electrical properties were obtained under static conditions by using a conventional
104 two-point probe technique with the specimen connected with a Keithley 2420 source
105 meter. Compounds were cut to obtain smaller specimens c.a. 20 × 14 × 2 mm in size. Both
106 faces of specimens were polished to eliminate the outermost surface and Ag conductive
107 paste was used to make round shaped electrodes with an average diameter of 2mm.
108 Electrical resistivity (ρ) was obtained by using the equation: $\rho = R \times (A/d)$, where R is the
109 measured resistance, A is the sample cross-section, and d is the distance between

110 electrodes. Hence, obtained values were referred to as volume resistivity. Measurements
111 were obtained by placing the specimens in a homemade sample holder consisting of a
112 fixed side and a spring-loaded electrode.

113

114 *2.2.4 Tensile test with in-situ conductivity measure*

115 The tensile tests have been conducted with the universal test machine Zwick/Roell Z5.0
116 with a maximum load of 5kN in the displacement control mode with a crosshead rate of
117 5 mm/min. The strain is then computed as the ratio of the crosshead displacement over
118 the initial grip distance.

119 To measure the in-situ electrical response, the Hioki 3538 impedance meter is connected
120 to the silver epoxy electrodes on the specimen's surface through a 4-point probe
121 connector. Four-terminal sensing is an electrical impedance measuring technique using
122 separate pairs of current and voltage sensing electrodes, eliminating the contact
123 resistance from the measurements. The electrical frequency response in the range of 0-
124 10 kHz is measured at each displacement increment of 50 μm , while keeping the
125 crosshead position fixed. The dynamic range is sampled within evenly logarithmic spaced
126 frequency intervals. Due to the visco-elasto-plastic nature of the polymer, a stress
127 relaxation can be observed while the strain is fixed, resulting in a decrease of the
128 measured force during the measurement window. The effect has been attenuated by
129 applying a low displacement rate of 5mm/min.

130 2.3 Homogenization models

131 2.3.1 Voigt model

132 Due to the heterogeneous nature of the composites under study, the effect of CB
133 loading in the polyamide matrix can be analyzed by mean-field homogenization theories.
134 Under the assumptions of constant strain in the Representative Volume Element (RVE),
135 negligible interaction between inclusions and homogeneously dispersed particles - the
136 Voigt model(32) can estimate the composite elastic modulus E_c from the volume fraction
137 of particles:

$$138 \quad E_c = E_m \cdot v_m + E_p \cdot v_p$$

139 Where E_m and E_p are the Young's Modulus of the matrix and the particles respectively,
140 and v_m and v_p the volume fractions. Since the particle content has only been measured as
141 weight percentage, the volume fraction is computed by means of the density ratio between
142 matrix and inclusions. The density of PA6, PA66 and Carbon Black(4) are assumed to be
143 1.1, 1.1 and 1.8 g/cm^3 , respectively.

144

145 2.3.2 Strain gradient elasticity theories

146 The classical estimates (bounds) of the Hashin-Shtrikman type for materials with an
147 isotropic distribution of the phases(33) read:

148

$$149 \quad \underline{\mu} = \mu^{(1)} + \frac{c_2(\mu^{(2)} - \mu^{(1)})}{1 + c_1 \frac{\mu^{(2)} - \mu^{(1)}}{\mu^* + \mu^{(1)}}} \quad \text{and} \quad \underline{k} = k^{(1)} + \frac{c_2(k^{(2)} - k^{(1)})}{1 + c_1 \frac{k^{(2)} - k^{(1)}}{k^* + k^{(1)}}} \quad (3)$$

150

151 with μ^* and k^* the shear and bulk moduli of the Hill constraint tensor:

152

153
$$\mu^* = \frac{\mu^0(9k^0 + \mu^0)}{6(k^0 + 2\mu^0)} \quad \text{and} \quad k^* = \frac{4}{3}\mu^0 \quad (4)$$

154

155 The choice $\mu^0 = \mu^{(1)}$ and $k^0 = k^{(1)}$ corresponds to the lower Hashin-Shtrikman bound
156 which coincides with the Mori Tanaka (MT) model (34) while the choice $\mu^0 = \mu^*$ and
157 $k^0 = k^*$ defines the self-consistent (SC) estimate (35).

158 Homogenization methods utilizing classical elasticity-based Eshelby tensors cannot
159 capture the particle size effect experimentally observed in particle–matrix composites at
160 the micron and nanometer scales. A simplified strain gradient elasticity theory (SSGET)
161 for spherical inclusion has been proposed by Gao and Ma(36), that introduced one
162 material length scale parameter instead of two classical elastic constants.

163 Equation 4 is rearranged as follows:

164

165
$$\underline{k} = k^{(1)} + k^{(1)} \frac{c_2(k^{(2)} - k^{(1)})}{k^{(1)} + c_1(k^{(2)} - k^{(1)})S_p} \quad \text{and} \quad \underline{\mu} = \mu^{(1)} + \mu^{(1)} \frac{c_2(\mu^{(2)} - \mu^{(1)})}{k^{(1)} + c_1(k^{(2)} - \mu^{(1)})S_s} \quad (5)$$

166

167 where S_p and S_s are the Eshelby tensor components:

168

169
$$S_p = \frac{\nu+1}{3(1-\nu)} \left\{ 1 + \frac{3}{2} \left(\frac{L^{(0)}}{R} \right)^3 \left[1 - \left(\frac{R}{L^{(0)}} \right)^2 - \left(1 + \frac{R}{L^{(0)}} \right)^2 e^{-\frac{2R}{L^{(0)}}} \right] \right\} \quad (6)$$

170
$$S_s = \frac{8-10\nu}{15(1-\nu)} \left\{ 1 + \frac{3}{2} \left(\frac{L^{(0)}}{R} \right)^3 \left[1 - \left(\frac{R}{L^{(0)}} \right)^2 - \left(1 + \frac{R}{L^{(0)}} \right)^2 e^{-\frac{2R}{L^{(0)}}} \right] \right\} \quad (7)$$

171

172 $L^{(0)}$ and R are the material length scale parameter and the inclusion radius,
173 respectively.

174

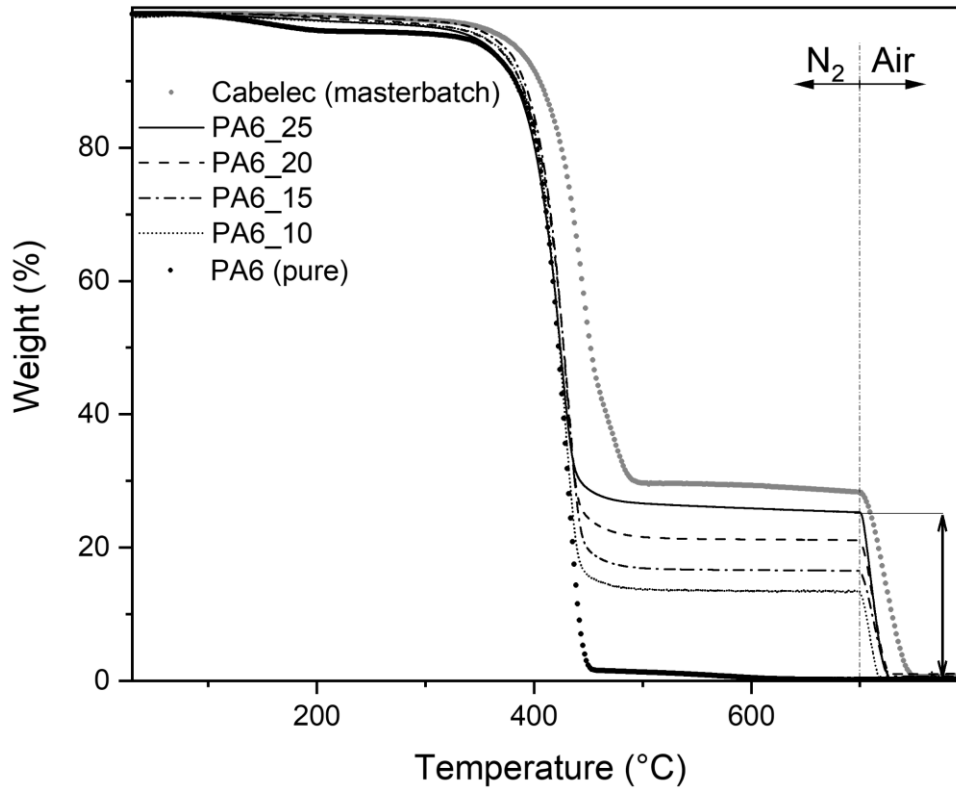
175 **3 Results and discussion**

176 **3.1 Particle dispersion and morphology analysis**

177 **3.1.1 TGA**

178 TGA plots of PA6-based compounds are shown in Figure 1. Due to the thermogram
179 similarities, they are to be considered as representative of both, PA6 and PA66
180 compositions.

181 From the weight loss of the TGA curves, the percentage of polymer inside each composite
182 in the same temperature range (c.a. 320-700°C) can be evaluated and by difference, the
183 percentage of filler (37). The real compositions of the compounds are summarized in
184 Table 1. Besides PA66_25, which contains 22.6 wt.% of CB, from the other compositions
185 it can be concluded that they are close to the predicted compositions.



186

187

188

189

Figure 1 - TGA plots of the PA6-based compounds (gray circles: Cabelec; black solid line: PA6_25, black dashed line: PA6_20; black dotted-dashed line: PA6_15; black dotted line: PA6_10, black circles: PA6, respectively) obtained under N₂ (up to 700 °C) and air flow (from 700 °C up to 800 °C).

190

191

In the following sections, authors will refer to different composites by the following nomenclature: PL_wt. Where *PL* indicates the neat polymer (PA6 or PA66) and *wt* the weight percentage of carbon black (e.g., PA6_10 and PA66_10 refer to polyamide 6 and polyamide 66 loaded with CB 10 wt.%, respectively).

195

196

Table 1 - Composition of samples as obtained from TGA measurements

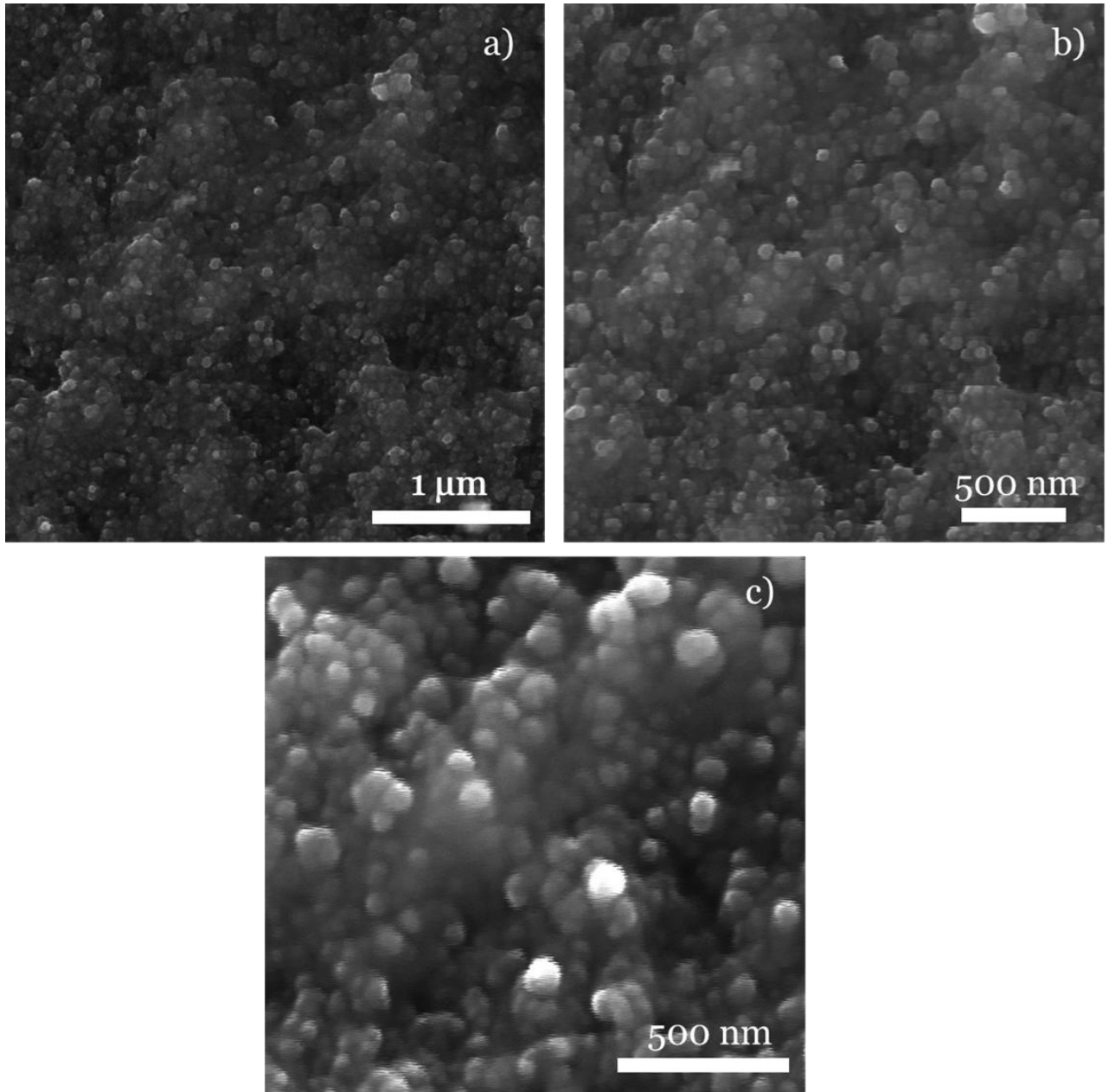
Label	CB (wt.%) ¹	Polymer (wt%)
Cabelec	27,5	72,5
PA6_10	12,1	87,9
PA66_10	9,7	90,5
PA6_15	15,5	84,6
PA66_15	13,7	86,3

PA6_20	19,5	80,5
PA66_20	20,0	80,0
PA6_25	23,6	76,4
PA66_25	22,6	78,4

197 ¹ As obtained from weight loss between 800°C (air flow) and 700°C (N₂ gas flow).

198 3.1.2 SEM

199 The particle distribution was assessed for both PA6 and PA66 with the three different
200 particle concentrations (c.a. 15%, 20% and 25% wt.). All the samples reported a uniform
201 particle distribution. SEM images of PA6 with 25% wt. of CB are reported in Figure 2 at
202 different magnifications: 40kX (Figure 2a), 80kX (Figure 2b) and 120kX (Figure 2c).
203 Figures 2a, 2b and 2c are representative of the particle distribution of the whole
204 specimen. At least 5 different zones of the specimen were analysed that reported the same
205 distribution of the particles. Figure 2a and 2b are at lower magnification and show that
206 the particle distribution is homogeneous and there are no areas with a lower presence of
207 particles or agglomerates. Figure 2c shows the 120 kX magnification that highlights the
208 particle shape and size of the carbon black particles. Figure 2c displays the presence of
209 both spherical and elliptical shapes that are typical of CB particles. A digital image
210 correlation software, Tescan Mira Software (Tescan, Brno, Czech Republic), has been
211 used to study the sizes of the particles. 30 different particles were measured to assess the
212 sizes of the elliptical and spherical shapes. The elliptical particles present an average
213 major axis of 113 nm (± 18 nm) while the minor axis is 74 nm (± 15 nm). The average area
214 of the elliptical particles is 6600 nm². On the other hand, the spherical particles present
215 an average diameter of 83 nm (± 16 nm) and an average area of 5400 nm².



216

217

218

219

Figure 2 - SEM images of the sample PA6_25% wt. at 40 kX magnification (a), 80 kX magnification (b) and 120 kX magnification (c) Figure 3 presents SEM images of PA6_15% (3a and 3b), PA6_20% (3c and 3d) and PA6_25% (3e and 3f) at two different magnifications, 50 kX (3a, 3c and 3e) and 100 kX (3b, 3d and 3f).

220

The SEM images are representative of the two different materials adopted PA6 and PA66

221

and of the three different adopted concentrations. The lower magnifications, 3a, 3c and

222

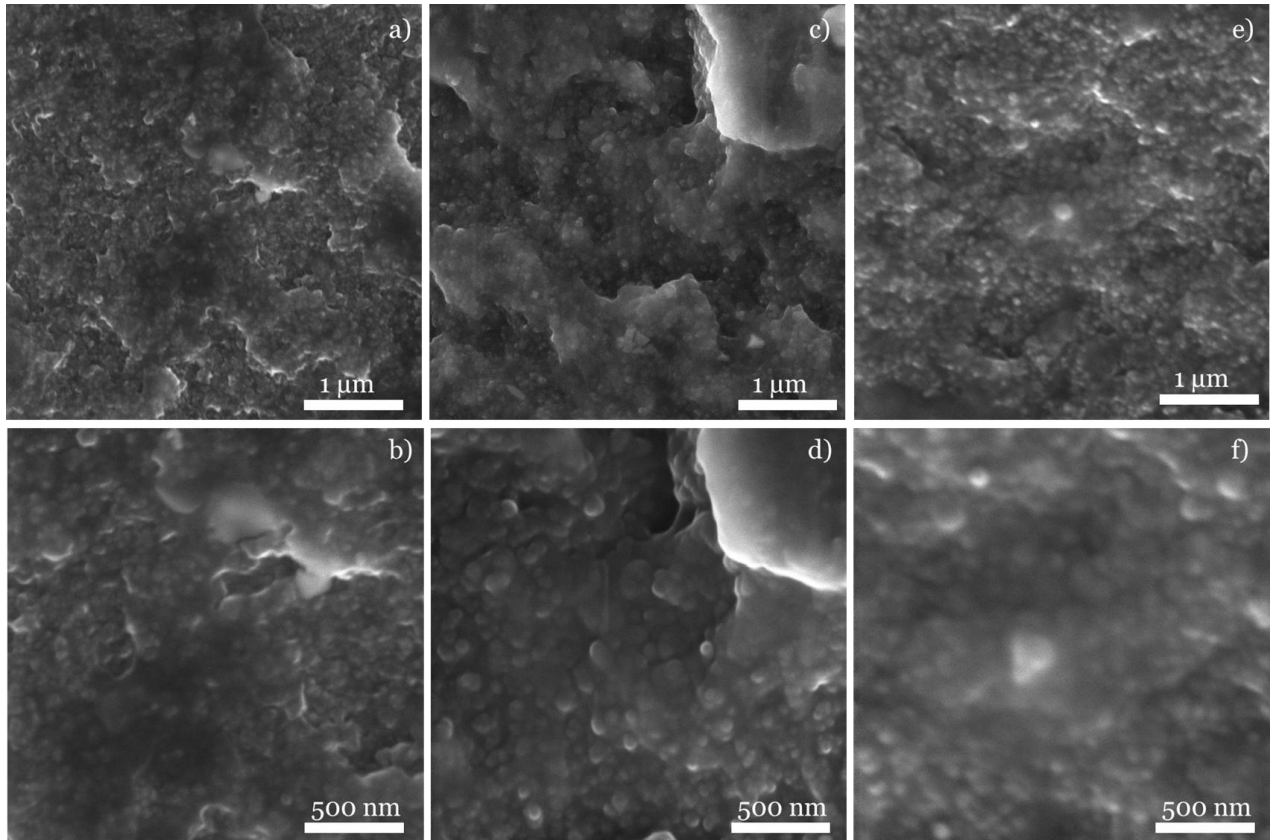
3e of Figure 3 show that the particle distribution is homogeneous. The higher

223

magnifications show that the particles are very close and there are no areas with a lower

224 presence of particles because all the analysed compounds are higher than the percolation
225 point. This verification was the aim of this analysis.

226



227

228 *Figure 3 - SEM images at two different magnifications 50 kX (2a, 2c and 2e) and 100 kX 2b, 2d and 2f) of the samples*
229 *PA6_15% (1a and 1b), PA6_20% and PA6_25%.*

230

231 3.2 Percolation threshold

232 DC electrical resistivity of the CB-filled PA6 and PA66 composites (10-28 wt.%),
233 measured at room temperature are shown in **Errore. L'origine riferimento non è**
234 **stata trovata..** In this figure, electrical resistivity increases with the decreasing CB
235 loading. Notwithstanding the different injection process temperatures, the two types of

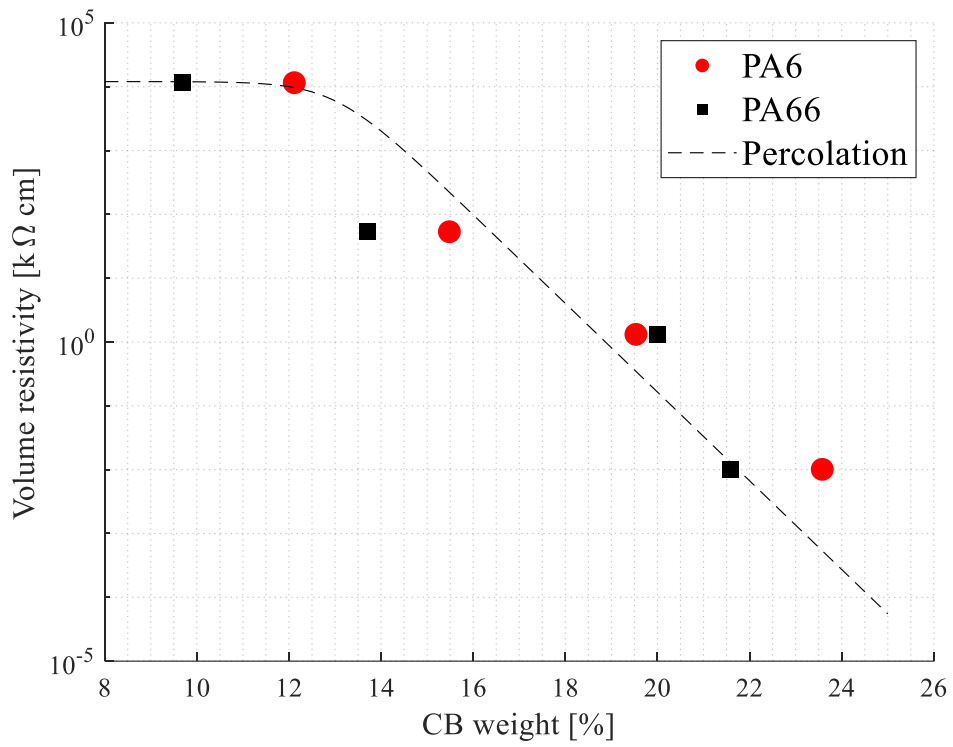
236 polymer composites exhibit a similar electrical behavior. Both Pa6 and PA66 based
237 composites exhibits a resistivity increase by 7 orders of magnitude from 3,9 to $1,2 \times 10^5$
238 and $5,2 \times 10^5 \text{ k}\Omega \text{ cm}$, respectively.

239 The percolation threshold, defined as the particle concentration above which the
240 composite goes from being a perfect insulator to a conductive, can be described by a
241 sigmoid function in the form:

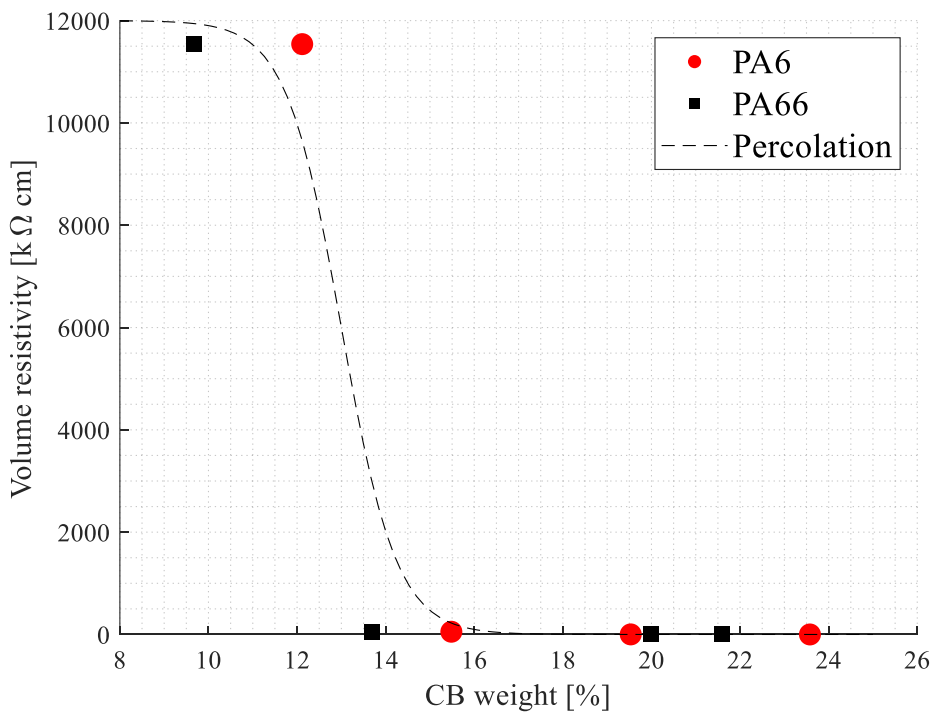
$$242 \quad R = R^0 \cdot \frac{1}{1 + e^{a(x - \bar{x})}} \quad (\text{xx})$$

243 Being R^0 the resistance of the insulating composite, a the coefficient describing the
244 resistance variation with the filler loading and \bar{x} the percolation threshold. Figure 4 shows
245 the results of volumetric resistance measurements.

246



247



248

249 *Figure 4. Change in volume resistivity of CB reinforced PA6 and PA66*

250 The sigmoid function that best approximates the resistance variation with the particle
251 concentration has a percolation threshold of 13% with an R^0 of $12^5 \text{ k}\Omega\text{cm}$ and a
252 coefficient α equal to 1.8.

253 3.3 The effect of carbon black on mechanical properties

254 Although the main goal of the present study is to analyze the effect of CB particles on
255 the strain sensitivity of conductive PA/CB composites, it is fundamental to assess the
256 composite mechanical response at first. In fact, any modification to the material
257 composition with the goal of functionalization, should not degrade the stiffness and
258 strength of the material itself. On the assumption of material response symmetry in
259 tension and compression, only tensile tests are performed to evaluate the elastic modulus
260 of composites. Experiments have been performed following the ISO D638 standards,
261 adopting the specimen shape of type V with a minimum section of 3x3 mm and grip
262 distance of 50 mm.

263 The Young Modulus and Ultimate Tensile Strength (UTS) of neat PA6 are experimentally
264 measured as $2.4 \pm 0.1 \text{ GPa}$ and $42 \pm 4 \text{ MPa}$, respectively; the bulk Young's modulus for the
265 PA66 is found in literature as 2.5 GPa (38).

266 The bulk modulus of CB reinforced PA66 is monotonically increasing in the range of
267 10–25% weight percentage, showing a small deviation from the average value among
268 different repetitions Figure 5 shows the measured values, the error bar and the linear
269 trend approximating the experimental results.

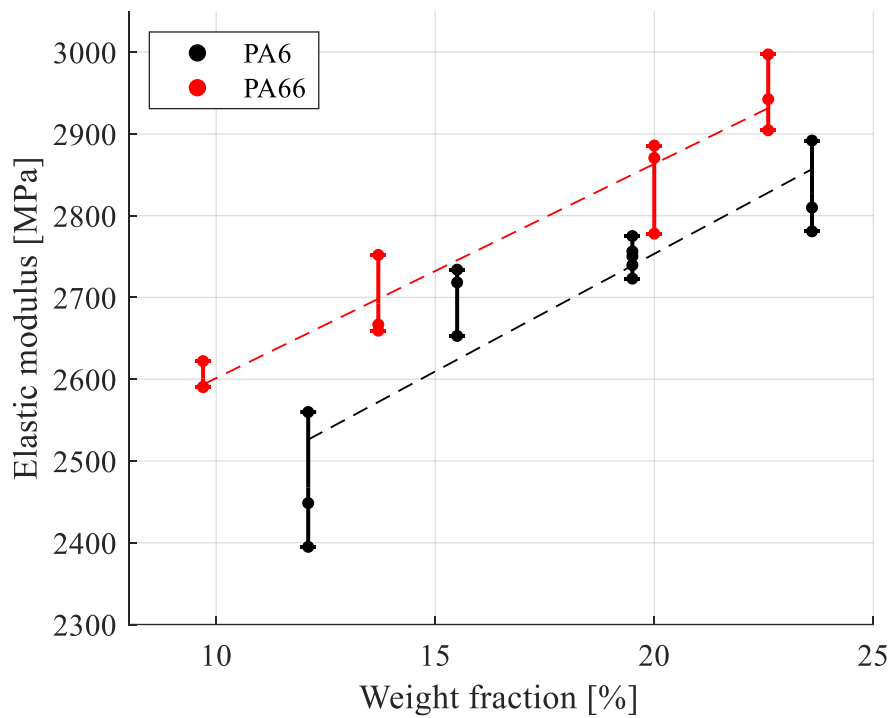


Figure 5 - Young's modulus of composites with different neat polymer and CB concentration.

270

271

272

273 Compared to the neat polymer, the bulk modulus of CB composite increases up to
 274 20%. In PA6 composites, the particle weight percent increase from 10% to 25% lead to a
 275 13% increase in the elastic modulus. In the case of PA66, the stiffness increases of 13%
 276 within the investigated weight percentage range. This demonstrates that the effect of the
 277 CB on the two polymers, PA6 and PA66, is consistent.

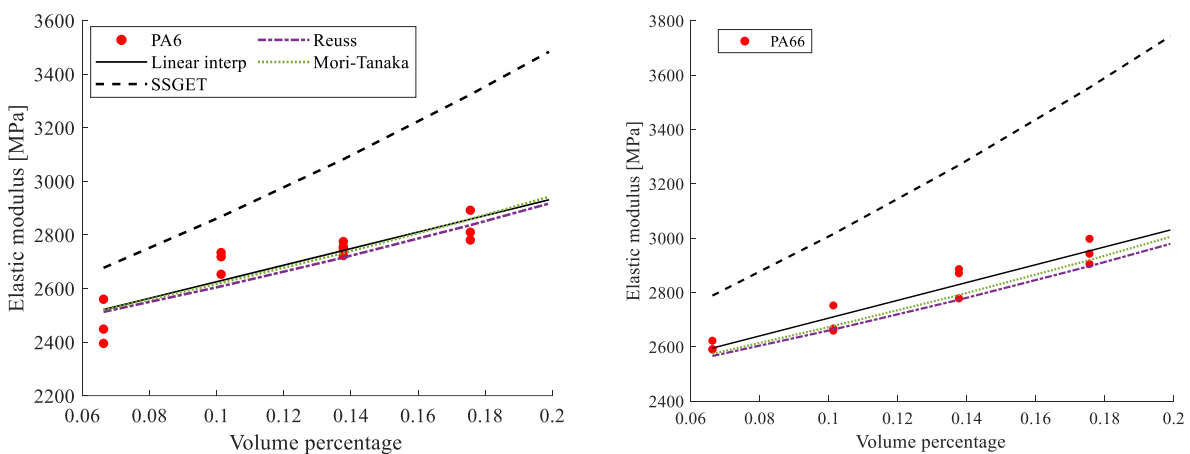
278

279 3.3.1 Mean-field homogenization models

280 Results from tensile tests are compared with homogenization theories described in Section
 281 2.3. The Voigt model predicts an elastic modulus increase on average twice larger than that
 282 observed experimentally. The limit of this theory relies in the assumption of perfectly

283 dispersed inclusions, neglecting any interaction between particles, and any clustering
284 carbon black spheres. Microscopies of CB morphology of the tested specimens shows a
285 clear agglomeration formation in composite with particle weight concentration higher
286 than 10%, yielding to the non-conformity of the Voigt hypothesis with the case under
287 study.

288 As showed in Figure 6, both the Mori-Tanaka and Reuss models correctly predicts the
289 effect of carbon black spherical inclusions on the elastic modulus increase, with a
290 maximum error of 4% respect to the mean value. The SSGET based homogenization model
291 is fitted with a size radius of $83 \mu m$, as observed in SEM image in Section 2, and a material
292 length parameter of $8.3 \mu m$ for the PA66 and $5 \mu m$ for the Nylon. Figure 6 shows that the
293 SSGET do not correctly accounts for the particle size on the stiffening effect of CB particles
294 for PA66 based composites. In can be concluded that the Mori-Tanaka and Reuss models
295 can be adopted to effectively predict the elastic modulus of polyamides composites loaded
296 with carbon black particles.



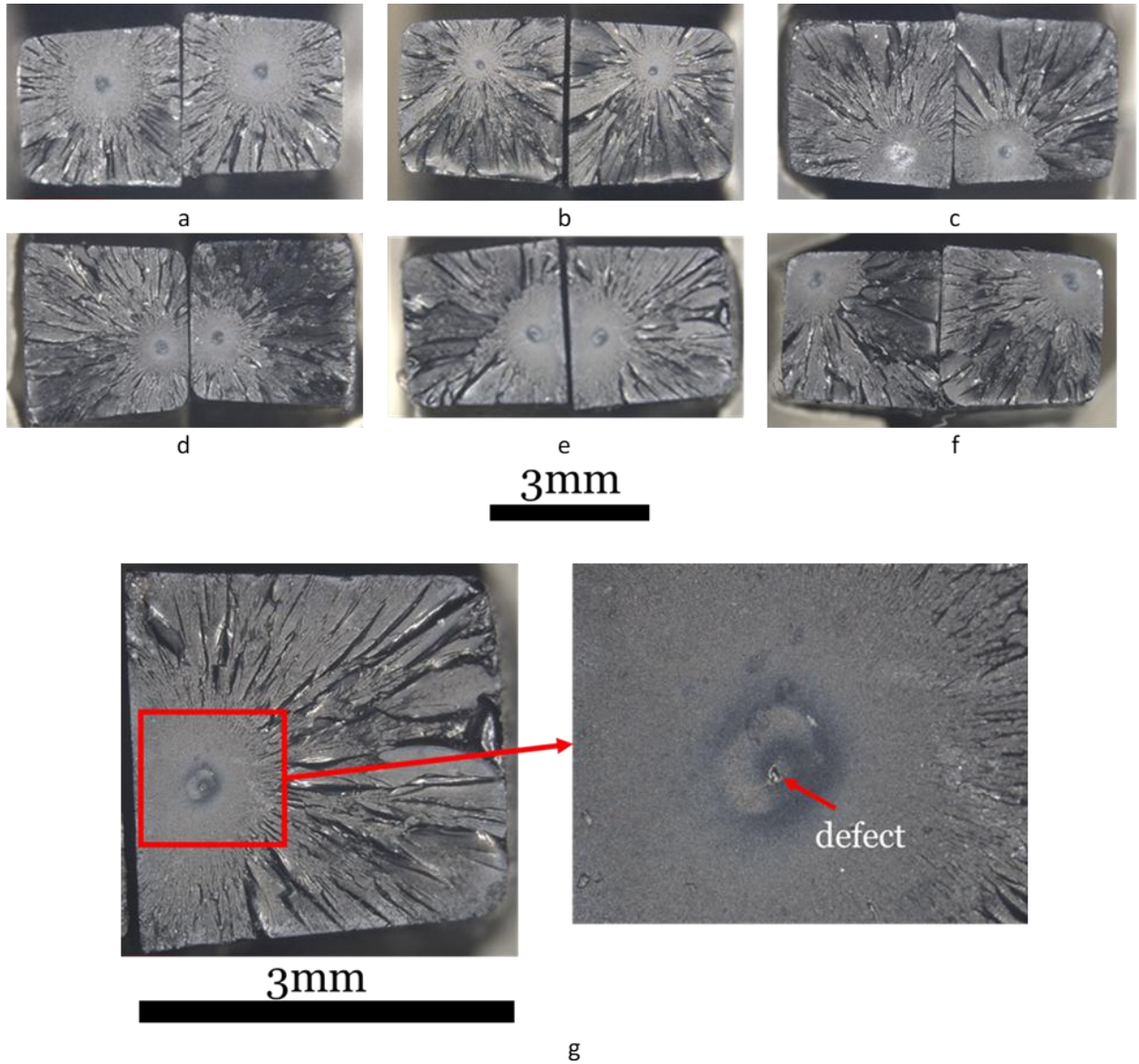
297

298 *Figure 6 - Comparison of homogenization theories prediction with experimental measurements for PA6 and PA66*
299 *with different CB mass fraction*

300

301 *3.3.2 Fracture surfaces (SEM)*

302 Figure 6 shows representative cryofractured surfaces of the PA6 (6a, 6b and 6c) and PA66
303 (6d, 6e, and 6f) for the three different concentrations, 15%, 20% and 25%, respectively. All
304 the fracture surfaces are very similar and display a reproducible failure surface. There is a
305 central area where the crack starts to propagate and appears quite smooth. Besides this
306 area, the fracture is irregular with the presence of clear and large cracks. These two
307 peculiar areas are more evident in Figure 6g that reports a higher magnification of the
308 fracture surface of the specimen PA66 with 20% wt. of CB. As it is well visible in all the
309 subfigures of Figure 6, the crack starts to propagate in the darker point of the smoother
310 circular area. The higher magnification of Figure 6g reports the presence of one defect that
311 was found in that area. These defects trigger the crack propagation initiation. The
312 microscope analysis conducted on all the specimens showed that one or two defects were
313 detected in the darker area for the two polymers prepared with the three particle
314 concentrations. Twelve voids were measured using a digital image correlation software.
315 These voids present a circular shape and an average diameter of 26 μm with a standard
316 deviation of 6 μm .



317
 318 *Figure 7 - Representative fracture surfaces of the PA6 and PA66 compounds*
 319

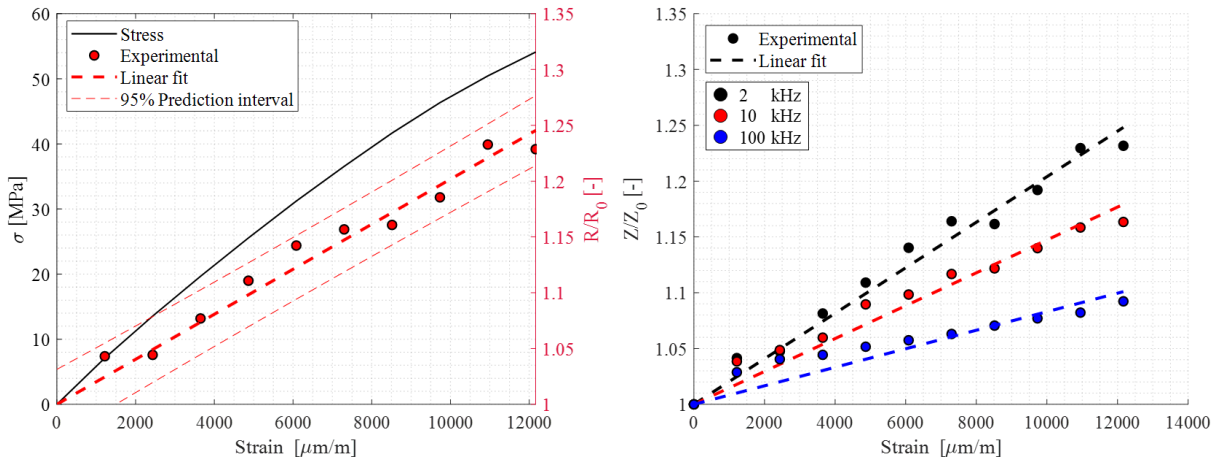
320 **3.4 Strain sensitivity**

321 The main goal of the present work is to study the variation of the electric passive
 322 properties induced by the mechanical strain in Carbon Black reinforced polyamides. The
 323 resistivity of composites is studied in direct current (DC) and alternate current (AC)

324 regime within a frequency spectrum ranging from 0 to 10 kHz. The electrical frequency
 325 response of each coupon is measured at fixed displacement increments of 0.05 mm.

326 From the frequency response measured at each displacement increment, it is possible to
 327 reconstruct the strain variation with respect to the applied strain for each sampled
 328 excitation frequency. First, the direct current electrical response is compared with the
 329 stress- strain curve of the material (Figure 8a) and a linear equation with null offset is
 330 fitted with the experimental measurements with a least-squares method. Figure 8b shows
 331 a comparison of the linearized strain sensitivity of dynamic impedance Z versus strain
 332 curves at three different frequencies. It is clearly highlighted the fact that strain sensitivity
 333 of the conductivity changes with the excitation frequency.

334



335

336 *Figure 8 - a) Variation of electrical resistance compared with stress in the material, b) Variation of impedance for*
 337 *different frequencies. Results shown in figure are referred to PA6_20*

338

339 The gauge factor GF is then computed at each excitation frequency \bar{f} as:

340

$$GF^{\bar{f}} = \min_G \sum_{\varepsilon} (\Delta Z_{\varepsilon}^{\bar{f}} / Z_{\varepsilon=0}^{\bar{f}} - G * \varepsilon) \quad (8)$$

341

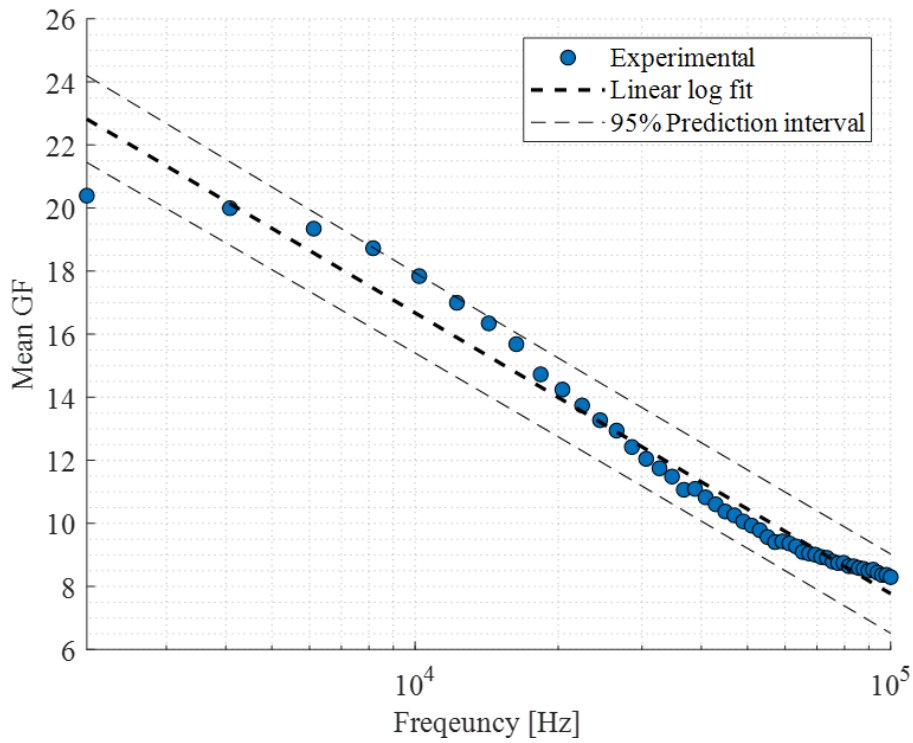
342 The conductivity of composites linearly varies with the logarithm of the current
343 frequency, yielding to the equation:

344

$$GF = GF^{DC} + a \cdot \log(f) \quad (9)$$

346

347 where a is fitted with a least-square method. Figure 9 shows the accuracy of equation 9
348 in predicting the dynamic gauge factor for PA6_20.



349

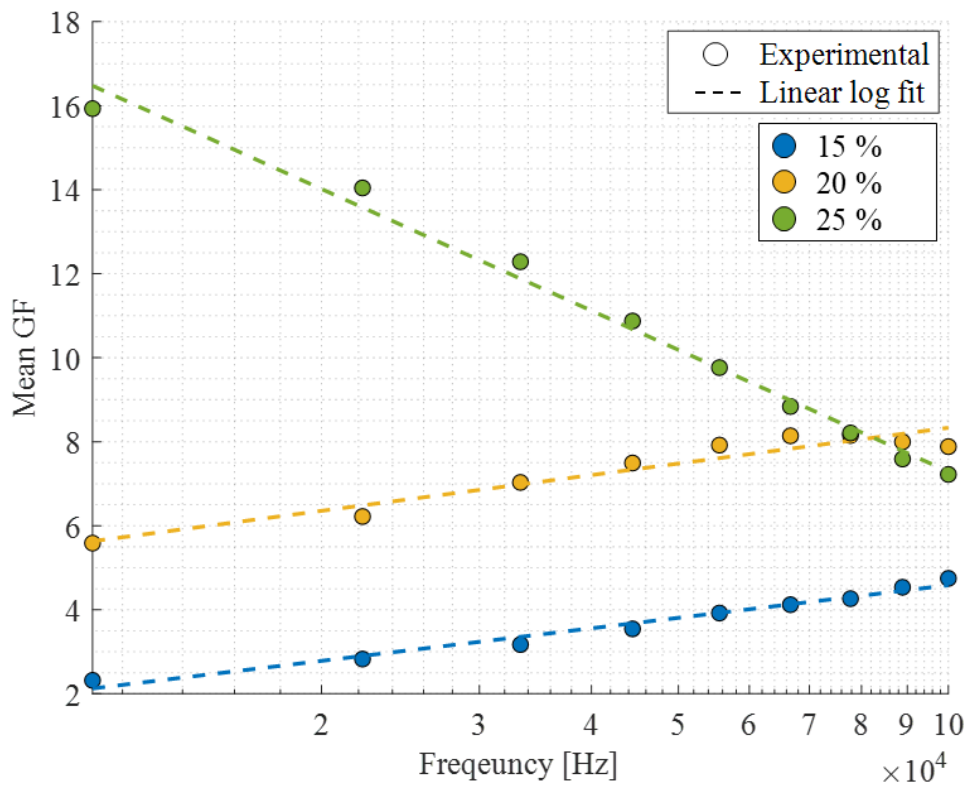
350 *Figure 9 - Linearized Gauge Factor vs. excitation frequency for PA6_20 composite, with $a=-3.9$ and $GF^{DC} = 23$.*

351 To assess the effect of particle concentration on the strain sensitivity, the mean GF
352 and its standard deviation can be computed for each composite composition at each

353 excitation frequency. Eventually, the mean GF variation with excitation current
354 frequency of different composites is compared in Figure 9. Every GF measured and
355 computed across the analyzed frequency spectra, are predicted with an error lower than
356 5% by the linear logarithmic model. Figure 10 shows that there is a linear variation of the
357 dynamic gauge factor with the logarithm of excitation current frequency for each of the
358 conductive composites under study (PA66 with CB weight content of 15, 20 and 25 %).
359 Since the GF dynamic law is linear, few dynamic measures are then necessary to fit the
360 model and assess the frequency effect on the composite strain sensitivity. The slope of the
361 model goes across a sign inversion when shifting from 20% to 25% of conductive particles
362 weight content. The phenomena may be associated to the formation of inductive
363 structures of CB particles that can activate a magnetic field enhancing the current flow in
364 the composite material. This will be further investigated with experimental test for
365 concentrations between 20% and 25%, and visual analysis for the reconstruction of the
366 particles 3D arrangement at high concentrations.

367 At the best of authors knowledge, this is the first research revealing the relation
368 between the gauge factor and the alternate current excitation frequency, evidencing that
369 there is a clear influence of the AC frequency on the composite resistivity and that it is
370 different at different filler content.

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Figure 10 - Effect of carbon black particle dispersion on the strain sensitivity of PA66 in the frequency domain

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Conclusions

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The authors experimentally analyze the effect of carbon black on the mechanical and

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electrical properties of PA6 and PA66 polymers. Composite specimens are produced by

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injection molding, mixing the neat polymer with different weight fractions of PA6/CB

379

compound. The evaluation of the quantity and the dispersion degree of carbon black is

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obtained from thermogravimetric analysis (TGA) and Field-emission Scanning Electron

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Microscopy (FeSEM). The effect of CB filler quantity on the elasto-plastic response is

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evaluated with quasi-static tensile tests and fracture surfaces are investigated with

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FeSEM to assess the influence of CB morphology on the composite strength. The

384 percolation threshold of different compositions has been determined by conductivity
385 measurements; conductive specimens have been tested under axial loading while in-situ
386 measuring the strain-induced change in the composite resistivity and impedance. The
387 gauge factor (GF) has been defined for different composites with different matrices at
388 different volume fractions within an electrical frequency range up to 10kHz.

389 The presented results of the experimental study demonstrated that the dispersion of
390 Carbon black particles within a PA6 and PA66 matrix enhances the stiffness and strength
391 of composite when the volume fraction is up to 20%. Nevertheless, higher concentration
392 could lead to CB agglomeration, yielding to the embrittlement of the composite. The
393 percolation threshold of polyamide/CB composite is found at 13% volume fraction of
394 carbon particles. Overall, the composite electric resistance exhibits high strain sensitivity
395 (gauge factor from 15 to 25). By analyzing the strain sensitivity of PA6 and PA66
396 composite with different Carbon Black concentration excited with different current
397 frequency, this study established that the current frequency does indeed have a
398 significant effect on the composite gauge factor, with a strong correlation to the particle
399 loading. This suggests that, while filler concentration does influence the overall
400 conductivity of the composite, the interplay of CB dispersion and excitation current
401 frequency defines the strain sensitivity of these materials. Carbon black is an ideal
402 candidate for the functionalization of fiber reinforced polyamides since it activates self-
403 sensing feature that can be enhanced by varying the current frequency, while enhancing
404 its mechanical properties. Application can be found in Structural Health Monitoring of
405 FRP parts, conductive plastic for HMI and others requiring an integrated strain sensor.

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412 **Data availability**

413 The raw/processed data required to reproduce these findings cannot be shared at this
414 time as the data also forms part of an ongoing study.

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