# Carbon–polymer composites with extreme electrical conductivity

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**ABSTRACT:** The aim of this study was to develop carbon–polymer composites with extreme electrical conductivity (100 S/cm) combined with good flexural strength. Despite the many optimization methods described in the literature, no comprehensive optimization procedure was to be found because the formulation did not control by itself the final properties. This study showed the major influence of the processing conditions with these peculiar materials. A detailed study of the influence of the processing conditions on the microstructural and macroscopic properties was performed. We, thereby, proposed a comprehensive way to optimize the properties of the final product. © 2015 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2015**, *132*, 42274.

## INTRODUCTION

A very large number of publications have been devoted to elaborate polymer conductive composites because they are used in a wide variety of industrial applications, such as batteries, fuel cells, aeronautique, antistatic media, and corrosion-resistant materials. To enhance the electrical conductivity, carbonaceous fillers are regularly used in the production of the polymer composites. Current carbonaceous fillers include graphite, carbon black (CB), carbon fiber (CF), and carbon nanotubes (CNTs), they exhibit tremendous contrasts in size and shape and, therefore, allow endless combinations of possible formulations. The aim of this study was to develop highly conductive carbon–polymer composites. The set of requirements was issued from fuel-cell application for materials used for bipolar plate application.[1–3] These were characterized by in-plane electrical conductivity above 100 S/cm with excellent mechanical properties (flexural strength > 30 MPa) and a good durability.

To reach these targets, an unusually large amount of additives with different types of fillers was used in the composite. Many attempts have been described in the literature,[4–12] and focused on the optimization of the formulations. Most of the studies have shown little improvement in the key properties cited previously. In fact, the formulation did not seem to be the only parameter that needed to be adjusted to reach the properties required by applications. Several studies[13–16] have also shown that the process can influence the final properties of heavily filled polymer composites. In this article, we describe an experimental study of the influence of the processing conditions on the final properties. We propose a somewhat systematic method for reaching the values of the set of requirements in terms of the electrical and mechanical properties.

A scouting study of the local electrical conductivities of two composite materials processed by compression is first presented. Coupled with microstructural analyses, these preliminary results revealed the influence of the process. A series of experiments was then performed by the variation of the temperature and pressure to obtain the optimal material and shed some light on the parameters that controlled the final properties.

## EXPERIMENTAL

### Materials

Three polymer-based composite formulations were examined in this study. The first one, called material A, was a commercial formulation with a very high electrical conductivity. Its filler content was close to 80 wt %, and it contained several types of carbon fillers (graphite, CB, CFs). The exact formulation was, however, unknown. The two last formulations were both experimental. As shown in a previous study,[16] graphite, because of its micrometer size, its aspect ratio close to one, its low specific surface area, and its excellent electrical conductivity, were incorporated in a large amount in the formulation. However, a large addition of graphite in a polymer greatly improves its electrical properties, even if it rarely reaches the level previously required. Nevertheless, the mixture of different types of conductive filler

is a very promising way to improve the electrical behavior: a well-chosen combination of graphite and others, even less conductive, carbonaceous fillers (CB, CFs, CNTs), may improve the electrical conduction as compared to graphite alone. With a generic approach for the optimization of the composite formulations, it seems that a mix of graphite and CF or a mix of graphite, CFs, and CB (with a low specific surface) would be a good reinforcement system to obtain the best electrical properties.[16] That is why the first formulation, material B, was based on vinyl ester resin filled with 69 wt % of a binary system of graphite (intrinsic electrical conductivity $(\sigma) = 625$ S/cm[16]) and CFs (length $(L) = 6$ mm, diameter $(D) = 7$ μm, $\sigma = 0.03$ S/cm [17]). Material C was similar to material B and was based on the same thermoset resin and binary system of fillers. The only difference concerned the nature of the CFs, which were milled in formulation C.

To reveal the effect of the process on the electrical properties, sheets with relatively large dimensions were processed: formulations A, B, and C were compression-molded at 180°C for 3 min under 40 MPa. The dimensions of the sheets were 200 × 190 × 2–3 mm³.

Then, for process optimization, small sheets were molded: formulations A and B were processed by compression with a hydraulic press (Carver 3912, maximum force = 15 tons) with two heating plates and a temperature regulator (Specac Atlas series, maximum temperature = 300°C, maximum force = 4 tons). A brace of 80 × 80 × 1 mm³ inserted between two steel plates (100 × 100 × 1.5 mm³) was used to mold a sheet of 60 × 60 × 1 mm³. An amount of 10 g of the uncured formulation was first homogeneously distributed in the mold. Then, the mold was pressed for 15 s under 6 MPa. This step was performed at room temperature. The excess material was removed. Then, the mold was pressed again under 4 MPa. During this step, the temperature and time of curing were fixed. For the cooling step, the mold was removed and placed under 2 kg. After 10 min, the material was kept at room temperature.

### Methods

**Microstructural Characterization: Dynamical Mechanical Analysis.** Dynamic mechanical measurements were performed with a forced oscillation pendulum (SMD2000, ACOEM Metravib 01DB, Limonest, France) working in torsion and with small amplitude; this ensured a linear response of the strain with respect to stress. The dimensions of the studied specimens were 30–35 × 7 × 1 mm³. For each formulation, two specimens were tested. To detect relaxation processes in the polymer, the measurements were performed at a frequency of 1 Hz in a wide range of temperatures from −150 to 200°C, including the glass transition. The data [elastic shear modulus $(G')$, dissipative modulus $(G'')$, and loss factor (tan $\delta = G''/G')$] were plotted versus the temperature for the first heating scan. The α transition could be observed with its principal characteristics: α-relaxation temperature $(T_\alpha;$ °C) and intensity, which was defined as the maximum of tan $\delta$.

**Morphological Characterization: Microscopic Observations.** To study the filler dispersion, one specimen of each of the materials A and B was extracted and was embedded in the

epoxy resin and polished. The surfaces were analyzed with a light optical microscope (DMLM, Leica) in reflection mode.

### Electrical Properties

**Local Measurement of the Electrical Conductivity.** As presented in ref. 17, local measurements of the electrical conductivity were performed on each sheet with dimensions of 200 × 200 × 2–3 mm³. $X$ and $Y$ have been arbitrarly defined for each plate. For these measurements, a four-point probe (FPP) device, composed of four aligned probes with a spacing of 1 cm, was used. At each point $[M(X,Y)]$, the device could be placed in two different ways, parallel to $X$ or $Y$, respectively, to give two resistances: $R_X(X,Y)$ and $R_Y(X,Y)$. Thanks to a source measurement unit (Keithley 2602), a constant current $[I$ (A)], which varied between 100 mA and 1 A, was applied through two outside probes, and the steady voltage across the other inside two probes was determined. For each measurement at $M(X,Y)$, the actual local thickness was measured accurately, and the factor $K_m(X,Y)$, which allows a correction of thickness and edge effects, was estimated with the analytical model proposed by Planes *et al.*[17] Finally, the conductivities $[\sigma_X(X,Y)$ and $\sigma_Y(X,Y)]$ and the anisotropy $[A(X,Y)]$ were determined:

$$\sigma_X(X,Y) = \frac{K_m(X,Y)}{R_X(X,Y)}, \; \sigma_Y(X,Y) = \frac{K_m(X,Y)}{R_Y(X,Y)}; \; A(X,Y) = \frac{\sigma_X(X,Y)}{\sigma_Y(X,Y)}$$
(1)

**Electrical and Microstructural Characterization of Specified Areas.** With the previous electrical measurements, the highest and lowest conductivity areas were determined (Figure 1). The samples were eventually cut off to confirm the conductivity with the four-electrode (FE) method.[17,18] The standard electrode assembly for the measurements of resistivity by the FE method had a distance $[d$ (cm)] between potential needles, and the current electrodes were copper strips attached to the sheet thickness. $I$ was applied on a surface $[S$ (cm²)] of the sheet specimen through the two outside electrodes with the help of a source measurement unit (Keithley 2602), and the steady voltage across the others inside two electrodes was determined. A variation of $I$ between 100 mA and 1 A allowed the deduction of the resistance $[R_{FE}$ (Ω), where $R_{FE}$ is the resistance determined with the four-electrode method] and conductivity $[\sigma_{FE}$ (S/cm), where $\sigma_{FE}$ is the conductivity determined by the four-electrode method] of the studied sample:

$$\sigma_{FE} = \frac{d}{R_{FE}S}$$
(2)

Three specimens of each sample were extracted. The density and filler content of each specimen were determined. The density was then determined with the Archimedes method. With a density kit (Mettler Toledo AB), the weights of the specimen in air $(M_A)$ and water $(M_B)$ were measured. The density of the sample $(\rho_{ap})$ was found with the following formula:

$$\rho_{ap} = \frac{M_A}{M_A - M_B}\rho_0$$
(3)

where $\rho_0$ is the density of the water at a fixed temperature.

On the same sample, the amount of fillers was then determined by thermogravimetric analysis (TA Instrument TGA 2050). The
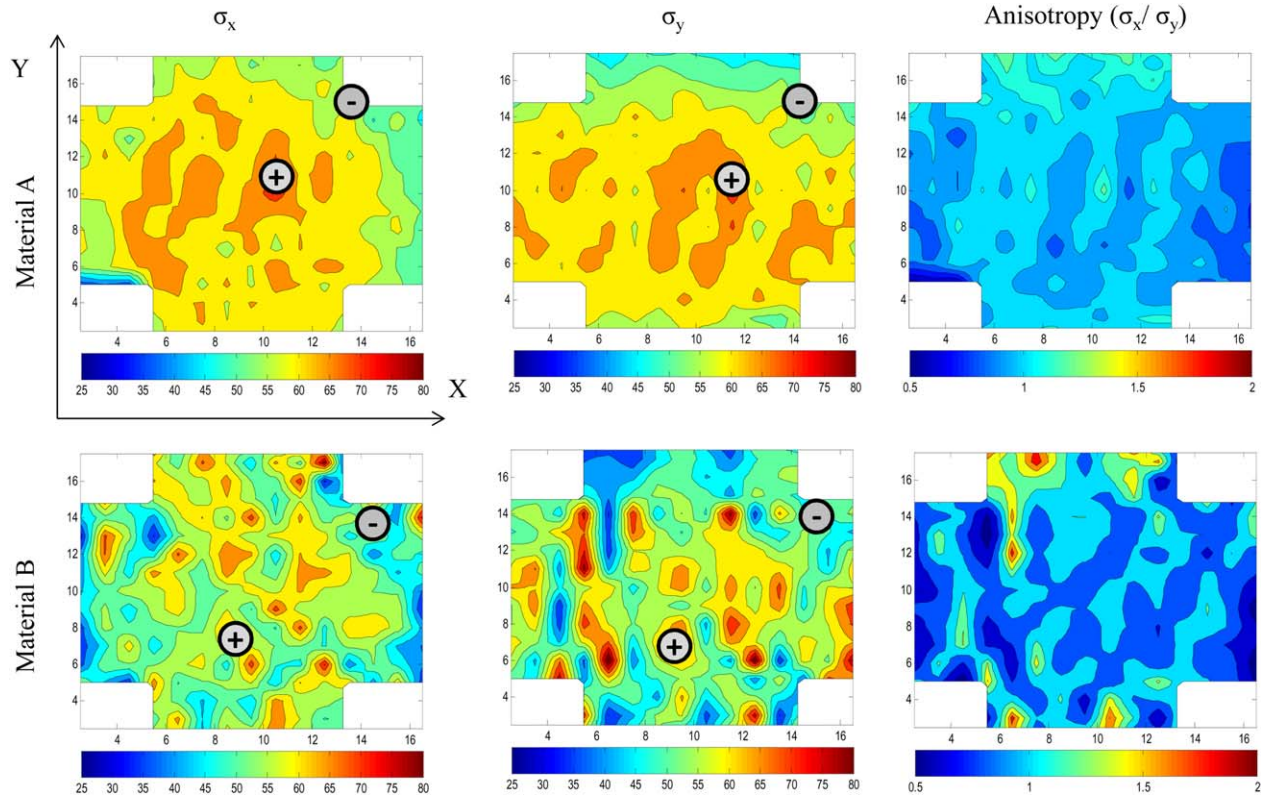
**Figure 1.** Conductivity and anisotropy maps for materials A and B with the position of the characterized areas (+ and − area). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

sample was submitted to an increase in the temperature at 20°C/min from room temperature to 800°C under an inert atmosphere (nitrogen). From the graph of the weight loss versus the temperature, two degradation steps were observed: the first one was attributed to polymer phase degradation, whereas the second one concerned carbon filler degradation. It was, thereby, possible to evaluate the weight contents of the matrix and the carbon filler.

**Electrical Conductivity Measurements.** The FPP method previously presented was also used to determine the in-plane conductivity of small sheets (60 × 60 × 1 mm³). For a specimen with a finite thickness [$w$ (cm)] and a placement of this device at the center of the sheet (where the edge effect can be neglected), the conductivity [$\sigma_e$ (S/cm)] simply becomes

$$\sigma_e = \frac{K_T}{R} \text{ with } K_T = \frac{\ln 2}{\pi w} \tag{4}$$

where $K_T$ is the correction factor (S/cm) and $R$ is the measured resistance (Ω). For each specimen, 10 measurements were performed.

**Flexural Properties**
The flexural properties of the composites were also evaluated. For a sheet with a thickness $w$ (mm), the width [$b$ (mm)], length, and bearing length [$L$ (mm)] were, 2.5 mm, 20 x$w$, and 16 x$w$, respectively. The tests were performed with an MTS20-MH mechanical testing machine equipped with a 5-kN load cell. A constant rate of loading, which was equal to $w/2$ (mm/min), was chosen. The force [$F$ (N)] and the displacement (mm) measured at the sample

center were recorded during the test. For each specimen, the ultimate force [$F_R$ (N)] was determined to give the ultimate bending stress [or flexural strength; $\sigma_m$ (MPa)] with

$$\sigma_m = \frac{3LF_R}{2bw^2} \tag{5}$$

For each formulation, five specimens were tested.

## RESULTS AND DISCUSSION
**Effect of the Process on the Electrical Properties**
**Homogeneity of the Electrical Properties Within the Material.**
The conductivity maps obtained for materials A and B (on large sheets) are presented in Figure 1. The sheet corners were disregarded because these areas were submitted to both parallel and perpendicular edge effects; this resulted in an unknown $K_T$.[17] A rather large distribution in conductivity was evidenced within material A. This material, processed this way, could, therefore, not be used as bipolar plates in reels systems.

To better quantify the homogeneity in the electrical properties and to perform a comparison between materials, the distribution of the conductivities $\sigma_X$ and $\sigma_Y$ values measured with the FPP method were adjusted with a 0 function (Figure 2):

$$y = y_0 + \frac{A}{w\sqrt{\pi/2}} \exp\left(\frac{-2(x-x_c)}{\omega^2}\right) \tag{6}$$

where $x_c$ is the vertical center, $y_0$ is the shift factor, $A$ is the Gaussian area, and $\omega$ is a parameter linked to the full width of half-maximum (fwhm) of the distribution peak.
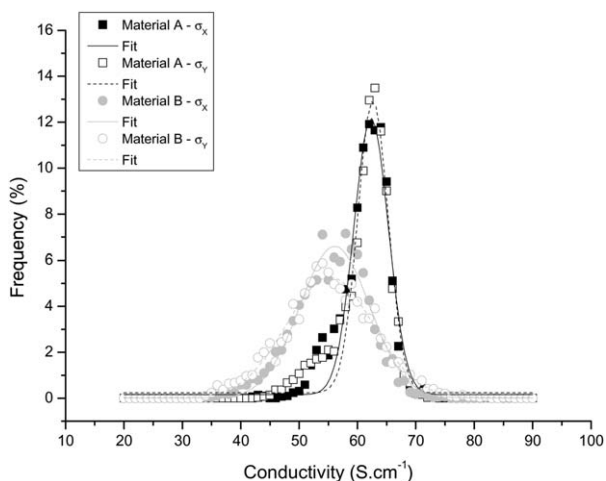
**Figure 2.** $\sigma_X$ and $\sigma_Y$ distributions measured on sheets of materials A and B.

$$\omega = \frac{\text{fwhm}}{\sqrt{2\ln 2}} \qquad (7)$$

The Gaussian parameters $x_c$ and $\omega$ are given in Table I for the two studied materials. For material A, the ratio between the largest conductivity measured at the sheet center and the lowest conductivity on the sheet edges was close to 2. Only the center of the sample presented the electrical properties required by the study (100 S/cm). This very intriguing result has not been mentioned in the literature to our knowledge. This result was further confirmed with the two tested directions, with very little anisotropy throughout the samples.

The electrical properties of material B were found to be even more heterogeneous (Figures 1 and 2). Instead of presenting a gradient from the center to the edges, the sheet made with material B presented several areas with a high electrical conductivity. They were, however, preferentially placed at the sheet center. This was confirmed by the conductivity distributions (Table I), which showed that the sheet of material B presented a lower electrical conductivity on average than that of material A (Table I). Material B also showed a broader distribution and a significant anisotropy. The latter effect was attributed to the filler orientation, which resulted from the presence of fibers.

The overall better properties in the center area likely resulted from the way the material was initially put into the mold, with a larger amount in the center. This could have induced both a poor filler dispersion and porosity formation at the edges, where the pressure might have been lower. To confirm this assumption, the areas with the highest and lowest performance were extracted and further characterized.

**Study of the Extreme Cases.** The conductivity of each sample was determined after extraction with the rigorous FE method. The averaged conductivity obtained by the FPP method was plotted versus the conductivity measured by the FE method for the same area (Figure 3). For all studied areas, the FE method largely confirmed the nondestructive tests performed on the sheets and, especially, the important dispersion in the electrical properties and the large distribution in conductivity for a given sample.

The density and filler content of each specimen were tested to examine the most simple hypothesis of a bad filler dispersion and porosity formation. This plausible idea was, however, not confirmed by the experiments, which could not differentiate the lowest and the highest conductor areas (Table II). The tiny differences could not explain the large difference in conductivity because the latter basically showed a linear variation with the amount of filler far from the percolation transition. In others words, the formulation seemed to have less influence on the electrical properties than the processing conditions,[16] and the filler dispersion and porosity seemed to be secondary parameters to the electrical properties. The optimization of the time, temperature, and pressure of curing in compression indeed largely altered the electrical properties of the polymer composites.[15,19] The application of sufficient mechanical stress with appropriate conditions of time and temperature improved the contacts between the fillers and favored the dewetting of the resin at the filler vicinity; this induced improvements in the percolating network and, thereby, the conductivity of the composite.

**Influence of the Processing Conditions on the Electrical Properties: Relationship Between the Microstructure and the Mechanical Properties**

From the A and B uncured formulations, a series of sheets 1 mm in thickness were processed by compression. The temperature and time were varied with a given pressure of 4 MPa. The size of the samples in this section induced boundary effects that prevented meaningful measurements of the distribution in the electrical conductivity. The influence of the temperature on the electrical conductivity was first studied for material A [Figure 4(a)]. A bell shell curve was observed with a maximum conductivity of 175°C. Although the perfect set of time, temperature, and pressure is likely to not only depend on the formulation but also on the processing tools and manhandling, this first experiment clearly revealed the unexpectedly large sensitivity of these heavily filled composites. In addition, the conductivity reached 110–120 S/cm, which was above the requirements of this study. The influence of curing time was also investigated [Figure 4(b)]. For all of the tested temperatures, an important increase in the electrical conductivity was observed between
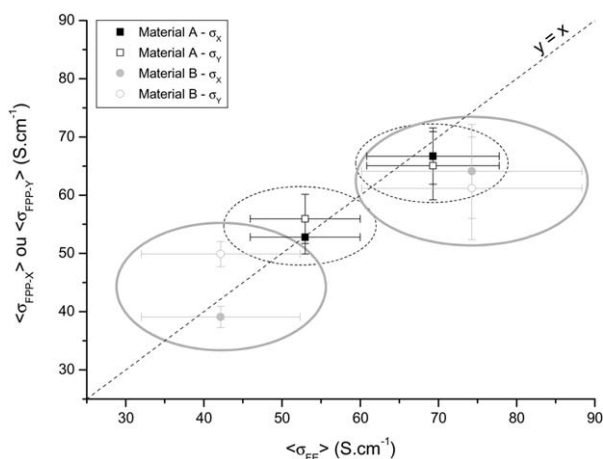
**Table I.** Results of Distribution Fitting with the Gaussian Function

| | $\sigma_X$ | | $\sigma_Y$ | |
| --- | --- | --- | --- | --- |
| | $x_c$ | $\omega$ | $x_c$ | $\omega$ |
| Material A | $62.3 \pm 0.1$ | $5.9 \pm 0.2$ | $62.6 \pm 0.1$ | $5.2 \pm 0.2$ |
| Material B | $56.1 \pm 0.1$ | $12.0 \pm 0.3$ | $55.3 \pm 0.2$ | $14.8 \pm 0.4$ |

**Figure 3.** Comparison between of the conductivities by two methods: FPP and FE methods. $\sigma_{FPP-X}$ and $\sigma_{FPP-Y}$ were the conductivities measured with the FPP method respectively in the X and Y direction.

curing times of 2 and 10 min. After 10 min of curing, the conductivity remained somewhat stable. The curing time should thus be fixed between 5 and 10 min. In summary, for a roughly optimized curing time (10 min) and temperature (175°C), the electrical conductivity of the obtained final product was above the requirements of this study.

Material B was processed the same way for compression. The curing time was fixed at 10 min. Differential scanning analyses showed previously that this time was sufficient to ensure complete degradation of the crosslinking agent (peroxide in this case) and, thus, to complete the reaction of curing within the composite. The conductivity versus curing temperature is presented in Figure 5 for comparison. In contrast to that of material A, the electrical conductivity of material B seemed to be stable and was close to 50 S/cm for all of the curing temperatures. The two formulations were based on vinyl ester resin heavily filled with similar carbon fillers. The qualitatively and quantitatively different behaviors between the two similar materials was a confirmation that the electrical properties were controlled by other parameters. The initial texture of material A (bulk molding compound under powder form) was very different from that of material B (bulk molding compound with fibers), and we, thus, decided to further scout the difference by a more detailed microstructural and morphological analysis.

**Table II.** Results of the Microstructural Characterization of the Lowest and Best Conductor Areas

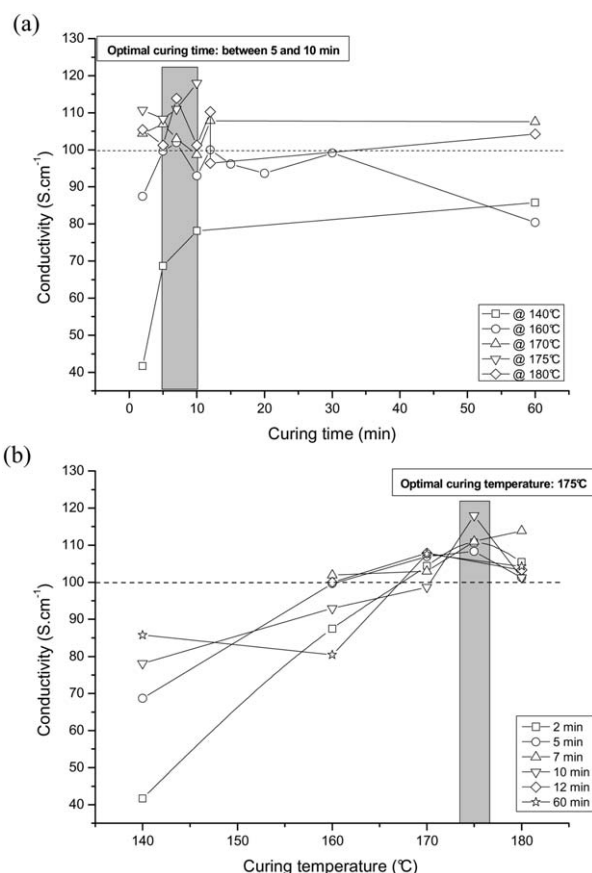|  | Area | Density | Filler content (wt %) | Porosity (%) |
|---|---|---|---|---|
| Material A | + | 1.81 ± 0.03 | 80.8 ± 0.2 | |
|  | − | 1.84 ± 0.03 | 80.9 ± 0.1 | |
| Material B | + | 1.69 ± 0.01 | 71.6 ± 0.5 | 0.7 ± 0.7 |
|  | − | 1.69 ± 0.01 | 71.5 ± 0.7 | 0.3 ± 0.8 |



**Figure 4.** Conductivity of material A processed by compression versus the (a) curing temperature and (b) curing time.

Dynamic mechanical analysis is an efficient way to characterize the molecular mobility of polymers, even with a low volume fraction. The loss factor (tan $\delta$) is particularly appropriate for probing the principal and subglass molecular relaxations. We decided to follow the $\alpha$ relaxation of the vinyl ester resin. The temperature and intensity of this relaxation could be directly linked to the matrix mobility: a higher mobility in the thermoset
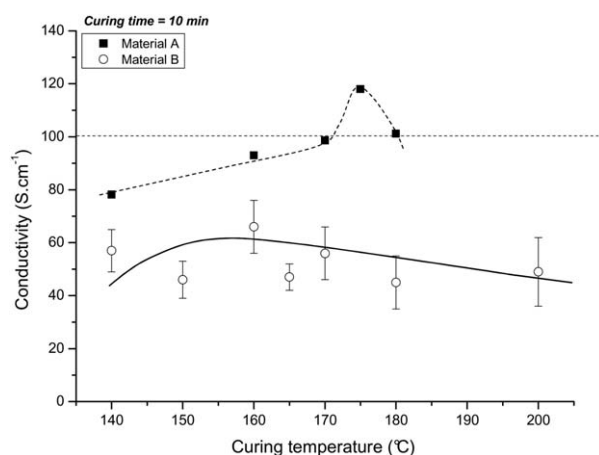


**Figure 5.** Conductivity of materials A and B versus the curing temperature for a fixed curing time (10 min).
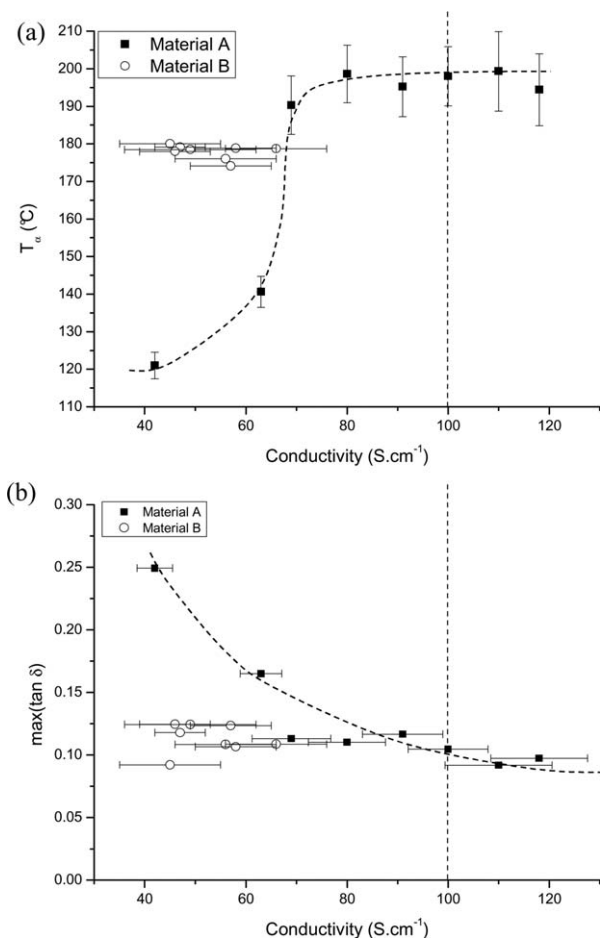
**Figure 6.** (a) Glass-transition temperature ($T_\alpha$) and (b) intensity versus the conductivity for materials A and B.

The results obtained for material B were very different from those obtained for material A [Figure 6(a,b)]. Regardless of the processing conditions, the conductivity was not altered by $T_\alpha$ or its intensity. They both were almost constant in all cases tested. This different feature was a hint for poor process optimization.

The flexural properties of these materials were also characterized at room temperature. The flexural modulus versus conductivity is presented in Figure 7(a) for materials A and B. For material A, an increase in the flexural modulus with increasing conductivity was noted. Whatever their flexural properties, all of the studied composites did not retain flexibility, but they remained flexible enough for the application. Thus, the improvement in the electrical properties also induced an increase in the mechanical properties. In contrast to the formulation where a tradeoff is usually observed,[26] the optimization of the processing conditions thus improved all of the properties at the same time. This result further underlines the correlation between the electrical conductivity and the strengthening of the polymer phase in the composite. As previously shown, the results obtained for material B were different because no variation in the flexural modulus was observed for any of the processing conditions tested. This confirmed the failure of the process optimization for this material.
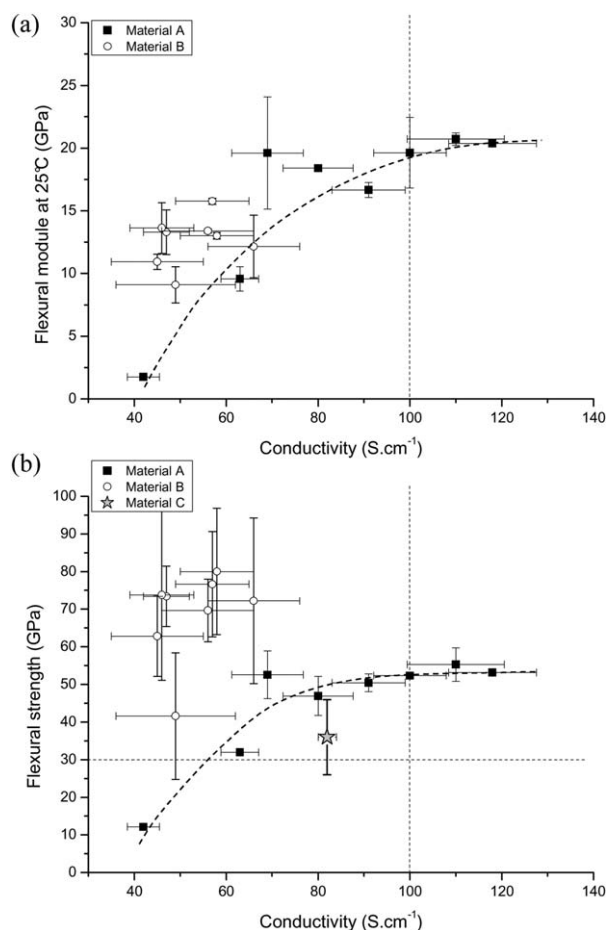


**Figure 7.** (a) Flexural modulus and (b) flexural strength versus the conductivity for materials A and B.

was revealed by a low temperature and large amplitude relaxation. The vinyl ester resin of material A presented an $\alpha$ relaxation between 140 and 200°C for all of the processing conditions tested. The electrical conductivity was then reported as a function of $T_\alpha$ and its amplitude max (tan $\delta$), as presented in Figure 6(a,b). This graph shows that the material with a high conductivity also presented a high $T_\alpha$. In other words, the quality of the contact between the carbon fillers, for a similar arrangement, was improved by the reduction in the polymer mobility. This was further evidenced by a decrease in the intensity of $\alpha$ relaxation. The reduction in the polymer mobility of the polymer phase could be explained either by a densification of thermoset network or a strong immobilization of the polymer phase at filler vicinity. This has already been observed for composites filled with nanoscopic fillers close to percolation threshold.[6–9] However, in this case, the main reinforcement agent was a micrometer-sized graphite filler, and the filler content was relatively high, close to 80 wt %. Thus, this decrease in the polymer mobility could be explained by the very large amount of filler, which induced a fictitious increase in the specific surface area of the filler in the composite. This mechanical response was also observed in the literature for a polypropylene composite heavily filled with Mg(OH)$_2$ or CaCO$_3$.[24,25]
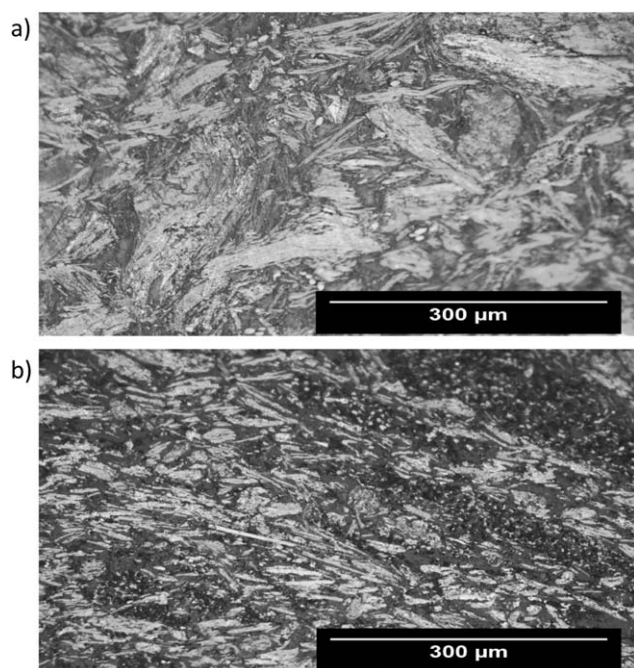
**Figure 8.** Microscopic observations of materials (a) A and (b) B.

The flexural strength versus the electrical conductivity is presented in Figure 7(b) for materials A and B. The mechanical and electrical requirements of the BPPs are also presented. This figure highlights the great interest of process optimization. For a given formulation, as for example for material A, the obtained sheets presented a very large range of electrical and mechanical properties. This optimization step allowed us to obtain materials with electrical and mechanical properties required by the study. In contrast, the process optimization of material B was not sufficient.

The dissimilar behavior between the two formulations (A and B) was likely to result from the larger amount of CFs in material B. Optical microscopy indeed showed that the CF dispersion was poor in the latter case. We could easily find large clusters of fibers that prevented the optimization of the formulation (Figure 8). Similar observations of material A revealed a very low content of CFs. This favored the compactness of the filler network. A last formulation based on the same resin used for formulation B and filled with graphite and milled CFs was processed by compression. As shown in Figure 8(b), an increase in the electrical conductivity up to 82 S/cm was observed. The incorporation of milled CFs in the formulation enabled us to improve the percolation network and, thus, the electrical properties. A slight decrease in the flexural strength was observed, but the samples remained above the DOE requirement. The process conditions (180°C, 3 min, and 40 MPa) were not optimized, and as shown previously, process optimization certainly induced improvements in the electrical and mechanical properties.

## CONCLUSIONS

Local measurement of the electrical conductivity allowed us to highlight the heterogeneity in the electrical properties of the sheets. Two different formulations were studied: one commercial and another based on vinyl ester resin and filled with graphite and CFs. We showed that the processing conditions primarily influenced the final properties of the composite. The proper optimization of the formulation and processing conditions induced spectacular changes in these materials. Not only did one reach a significantly higher electrical conductivity, but the glass-transition temperature also increased, and the mechanical properties were improved.

A coupled optimization of the formulation and process is, therefore, necessary to develop highly conductive polymer composites. With these heavily filled materials, the exact formulation impacts the electrical conductivity less than the processing conditions.

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