

Electrical ac and dc behavior of epoxy nanocomposites containing graphene oxide

Paolo Mancinelli,¹ Davide Fabiani,¹ Andrea Saccani,² Maurizio Toselli,³ Michel F. Fréchet⁴

¹Dipartimento di Ingegneria dell'Energia Elettrica e dell'Informazione "Guglielmo Marconi", viale Risorgimento 2, 40136 Bologna, Italy

²Dipartimento di Ingegneria Civile, Ambientale e dei Materiali, via Terracini 28, 40131 Bologna, Italy

³Dipartimento di Chimica Industriale "Toso Montanari", via Terracini 28, 40131 Bologna, Italy

⁴Institut de Recherche d'Hydro-Québec, 1800 boul Lionel-Boulet, Varennes, Quebec, Canada

Correspondence to: A. Saccani (E-mail: andrea.saccani@unibo.it)

ABSTRACT: This article deals with the investigation of electrical properties of epoxy-based nanocomposites containing graphene oxide nanofillers dispersed in the polymer matrix through two-phase extraction. Broadband dielectric spectroscopy and dc electrical conductivity as a function of electric field have been evaluated in specimens containing up to 0.5 wt % of nanofiller. Nanocomposites containing pristine graphene oxide do not show significant changes of electrical properties. On the contrary, the same materials after a proper thermal treatment at 135°C, able to provoke the *in situ* reduction of graphene oxide, exhibit higher permittivity and electrical conductivity, without showing large decrease of breakdown voltage. Moreover, a nonlinear behavior of the electrical conductivity is observed in the range of electric fields investigated, i.e. 2–30 kV mm⁻¹. A new relaxation phenomenon with a very low temperature dependence is also evidenced at high frequency in reduced graphene oxide composites, likely associated to induced polarization of electrically conductive nanoparticles. © 2015 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* **2015**, *132*, 41923.

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INTRODUCTION

Graphene has definitely arisen as a huge interest in the field of materials science: according to its remarkable mechanical, thermal, and electrical performance, it is intensively used also in the formulation of polymer nanocomposites.^{1–5} More recently graphite oxide, which consists of a layered structure of graphene oxide (GO), is becoming an important intermediate in the preparation of graphene-based nanocomposites. Because of the presence of functional polar groups such as hydroxyl and epoxide on basal planes in addition to carboxyl and carbonyl groups, mostly at the sheet edges, GO becomes hydrophilic and readily dispersible in water and in other polar organic solvents to form stable colloidal suspensions.^{6–9} However, functional groups and defects on GO make it thermally unstable and electrically less conductive than graphene. Reduction process can partially restore the outstanding properties of graphene. The exfoliated sheets of GO can be reduced into graphene in different ways: chemically, electrochemically, or thermally. Chemical reduction performed with several reducing agents, including hydrazine or sodium borohydrate is the most frequently used.^{10–12} One of the main drawbacks is that these processes, altering the hydro-

philic character of GO, increases its tendency to agglomerate irreversibly. A possible solution to avoid this problem can be found in a thermal treatment able to reduce the already dispersed GO sheets directly into a rigid matrix.^{13–17} In some published papers, researchers have described the *in situ* thermal reduction of GO dispersed in polymeric matrix at temperatures near or below the melting point of some thermoplastics^{18–22} or even during the polymerization process.²³ Some authors have underlined that the reduction temperature and reduction degree of GO inside the polymeric matrix depend on the chemical structure of the surrounding polymer.²⁴

Nanocomposites consisting of dispersed graphene into an epoxy matrix have potential application in the electronic industry. In high voltage systems, the composite material has to behave as electrical insulation, thus showing very low electrical conductivity, e.g. < 10⁻¹² S m⁻¹. However, it could be required to modulate conductivity of electrical insulation over a few decades of values. In particular, cable accessories, e.g. joints or terminations, make use of insulation systems having variable conductivity / permittivity with the aim of grading the electric field and avoiding field concentration, which can eventually lead to the

failure of the insulator.^{25,26} To date, conductivity modulation is achieved by doping insulating polymers with a relatively large amount of micrometer-size carbon black (up to 30 wt %). This high amount of filler can create disadvantages such as scanty mechanical properties or a strong reduction of the dielectric strength.²⁷ The use of graphene could, at the same time, enhance the mechanical properties and locally increase the electrical conductivity/permittivity by using much lower filler content because of the extremely high surface area of the filler.

Literature is still lacking regarding researches on graphene nanocomposites for high-voltage application. In particular, silicon rubber-based composites have been investigated recently,²⁸ but a very few studies on epoxy-based composites are available in literature to date.^{29–31}

According to its chemical structure, hydrophilic GO containing many epoxy and carboxyl groups, represents in principle an ideal filler in the preparation of epoxy resins. On account of the presence of these functional groups, the *in situ* polymerization should enable the covalent bonding of the GO sheets between either the epoxy prepolymer or the amine through various condensation reactions.^{29,30,32} Once the GO sheets are dispersed in the thermosetting matrix, it should be possible to reduce them *in situ* through a proper thermal treatment.³¹ In the present work, composites containing up to 0.5 wt % of GO have been prepared by a two-phase extraction process involving the ultrasonic dispersion of GO in water and the subsequent phase exchange with the bisphenol-A based thermosetting epoxy.^{33,34} The modified based thermosetting epoxy has been subsequently cured by adding an aliphatic amine as hardener and submitted to a mild thermal treatment *in vacuo*. The dielectric properties of the systems so far obtained (ac permittivity, loss factor and dielectric strength) as well as their dc electrical conductivity have been investigated and discussed.

EXPERIMENTAL

Materials

Graphite Oxide powder has been prepared from graphite powder by Hummer's method and purchased from Punto Quantico (Italy). The lateral dimensions of the platelets is about 2–6 microns and the C/O ratio is 1.63. Epoxy resin based on diglycidylether of bisphenol A (DGEBA) with an epoxy equivalent weight of 172–176 g eq⁻¹ (DER 332) and poly(propylene glycol) bis(2-amino-propyl ether) with average $M_n = 230$ g mol⁻¹ as hardener were purchased from Sigma-Aldrich and used as received.

GO Homogeneous Dispersion in Based Epoxy

This procedure closely follows the preparation reported by other researchers.^{33,34} Graphite oxide powder was sonicated in water for 1 hour obtaining a brown homogeneous water dispersion with the concentration of 5 mg mL⁻¹. Subsequently, 20 g of DGEBA was mixed with 16.1 or 26.9 mL of GO water dispersion in 100 mL glass flasks. The two-phase mixtures were tip sonicated for about 1 hour by using a 7.0 mm diameter ultrasonic probe UP200S (Hielscher, Germany). Afterward, water was removed under vacuum at 80°C in a rotary evaporator and further stirred overnight at 60°C to complete water removal. At the end of the process, the modified epoxy base appeared

Table I. Epoxy-Resin Composite Sample Description

Sample code	wt % of GO on based epoxy	wt % of GO on final resin	T_g (°C)
Neat resin	–	–	93
GO_03	0.403	0.300	94
GO_05	0.672	0.500	94

homogeneous at visual inspection at all the investigated GO amounts with a brown coloration whose intensity was a function of the GO content. The GO concentration in the based epoxy is reported in Table I.

Samples Preparation

GO-epoxy composites were formed adopting the following procedure: (1) mixing the two components (GO-based epoxy:amine) in a 1000 : 344 weight ratio; (2) degassing in the vacuum oven at 40°C for 20 min and casting in steel moulds; and (3) curing for 6 hours at 105°C. Flat sheets of about 170 × 130 mm² with a thickness of 0.55 ± 0.03 mm were obtained. In order to perform the *in situ* thermal reduction of the GO, some specimens, out of all the investigated compositions, were subjected to a postcuring thermal treatment in a vacuum oven at 135°C for about 48 h.

Measurements and Characterization

Cutting the sample using ultramicrotomy with a diamond knife produced samples having a thickness of 90 ± 10 nm. The thin sample was deposited on a metal grid and submitted to Transmission Electron Microscopy (TEM), a Hitachi HD2700 with a Bruker EDX SSD.

A Differential Scanning Calorimeter (DSC TA Q20) was used to determine the glass transition temperature. The measurements were carried out between 0°C and 200°C with 10°C min⁻¹ ramps. Samples were tested following a process of heating, cooling and heating again to eliminate the effects of previous thermal history.

Dielectric properties were analyzed between 10⁶ and 10⁻² Hz using a Novocontrol broadband dielectric spectrometer with a nitrogen-flow temperature control. *In vacuo* measurements were instead performed between 10⁴ and 10⁻² Hz by means of Solartron Analytical Model 1253A, which is frequency limited up to 20 kHz. For dielectric measurements, samples were set between two electrodes of 40 mm diameter and 2 mm thickness. All the dielectric measurements have been performed supplying a voltage of 3 V.

Electrical dc conductivity was measured applying a 20 kV mm⁻¹ electrical field to the sample sandwiched between metallic electrodes. In this case, the HV electrode has a 27 mm diameter and the lower electrode, connected to the electrometer, has a 15 mm diameter. Current flowing in the circuit was measured by a Keithley mod. 617 electrometer while the field was applied by means of High-Voltage Dielectric Current generator model FUG HCN35–35000. In order to avoid the influence of surface currents on the measurement, the lower electrode is surrounded

by a guard ring connected to ground. Samples have been gold coated replicating the shapes of electrodes and guard ring.

AC dielectric strength was measured on a sample of five specimens by means of a sphere-sphere test cell, using a voltage ramp of 2 kV s^{-1} . The breakdown fields were then processed through the Weibull probability function obtaining the α -parameter, representing the 63.2th percentile of dielectric strength, and the β -parameter, which is inversely proportional to standard deviation.

RESULTS AND DISCUSSION

The resin codes, the corresponding compositions and the glass transition temperatures of investigated materials, determined by DSC, are reported in Table I.

Figure 1 shows the TEM morphology of GO_05 composite. The image shows a good dispersion without large aggregates similarly to that reported in other GO epoxy nanocomposites.^{23,35} The homogeneous structure formed by well exfoliated GO layers confirms the efficiency of the preparation method.

Figure 2 shows real ϵ' (a) and imaginary ϵ'' (b) part of relative permittivity of neat epoxy and of GO composites at 20°C before the thermal reduction, as a function of frequency. A broad peak in the imaginary part of permittivity, corresponding to the β transition of the epoxy network, is clearly visible for the neat epoxy as reported elsewhere.³⁶ The presence of the filler slightly changes the position of the peak and the intensity of the relaxation, and its main effect is detectable as a very small decrease of the ϵ' values (about 1.2%). This slight decrease can possibly be linked to a reduced mobility of the epoxy segments deriving from the interaction with the rigid sheets of GO.

To study the influence of the thermal-reduction treatment on the dispersed GO, a GO_05 sample was tested during the postcuring process (135°C in a vacuum oven) at different time steps. The sample was set *in vacuo*, heated at 135°C for 2 hours and slowly

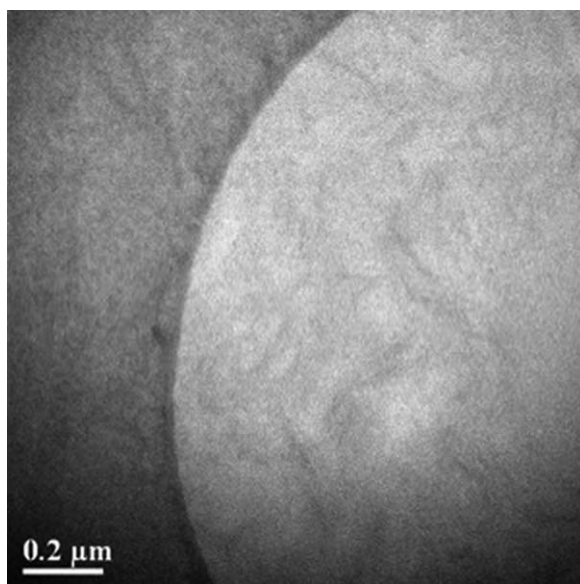


Figure 1. TEM image of GO_05 sample.

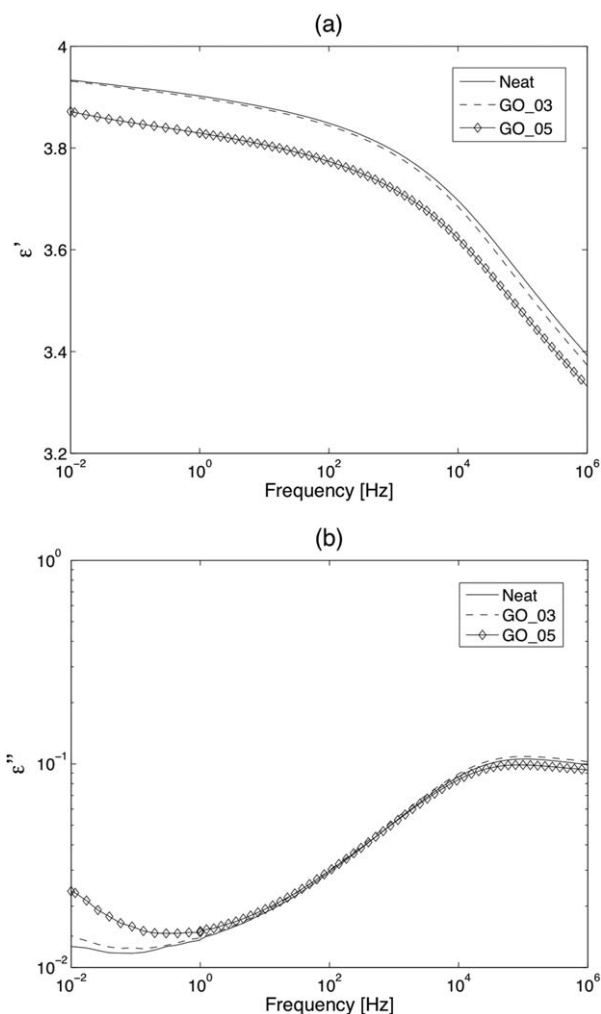


Figure 2. (a) Real part and (b) imaginary part of relative permittivity at 20°C of neat epoxy and composites containing 0.3 and 0.5 wt % of GO before thermal treatment.

cooled at 60°C (measurement temperature) in order to measure dielectric properties below glass transition temperature.³⁷ This procedure was repeated keeping the sample under vacuum, thus making the process much closer to the postcuring treatment.

Figure 3 illustrates the changes in the values of ϵ' at the reported times. A clear and progressive increase in the values is detectable as the thermal treatment proceeds. It should also be underlined that a strong change in color takes place during the first 6 hours of the treatment, leading to a dark black hue of the specimen. These results are a clear evidence of the chemical reduction of the GO platelets inside the matrix leading to the loss of oxygenated groups, according to the available literature^{10–13,32} and the earlier studies described by the authors elsewhere.³¹ Based on the results shown in Figure 3 and previous work, a longer duration of the thermal treatment has been selected in order to complete the thermal reducing process. Hereafter, GO_x resins submitted to thermal treatment at 135°C for 48 h will be referred to as TRGO_x.

Figure 4 shows the real ϵ' (a) and imaginary ϵ'' (b) part of relative permittivity of neat epoxy and TRGO_x composites at

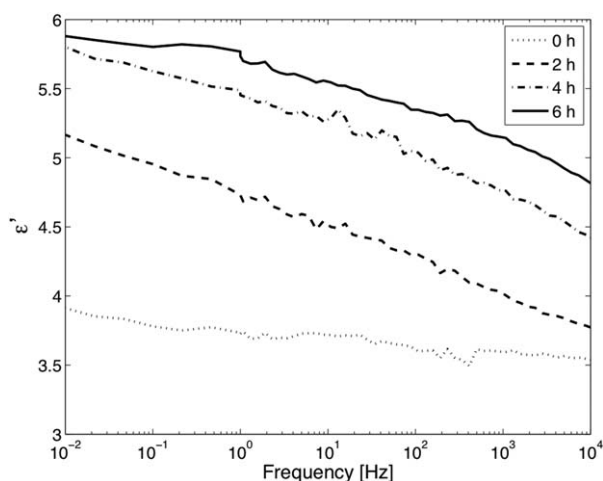


Figure 3. Changes in the real part of permittivity measured at 60°C of a GO_0.5 composite during the first hours of thermal reduction treatment at 135°C.

20°C, as a function of frequency. A significant increase in the ϵ' values takes place. The increase is active already at the highest investigated frequencies. On analyzing the ϵ'' plot, a further change is detected: a new relaxation process overlapping the β segmental movements appears.

Further dielectric spectroscopy measurements have been performed at different temperature steps, from -100°C to 180°C to analyze this phenomenon. First of all, it should be noted that the dielectric response is stable over time and cycling. In Figure 5, three 3-D maps of ϵ'' as a function of temperature and frequency are displayed in a reduced temperature range, i.e. -100°C to 80°C , taking into consideration only phenomena occurring under glass transition temperature to better evidence relaxation peaks. The β -relaxation process of neat epoxy is clearly visible in Figure 5(a). This plot confirms the temperature and frequency correlation of this polarization process which is well known for this type of dielectrics.^{36,38}

Figure 5 shows the polarization processes in neat epoxy and in TRGO_05 composite. The β -peak is not significantly influenced by the presence of the filler. However, a second polarization peak can be highlighted in the plot. This peak appears at high frequency, around 10^6 Hz, and is not present in the GO_05 composite, as can be observed comparing Figures 2(b) and 4(b). The new relaxation, showing a very weak temperature dependence [see Figure 5(b)], could be associated with the thermally reduced GO. Indeed, this new polarization process cannot be attributed to dipolar relaxation, even if the occurrence frequency is typical of such polarization, because the very weak temperature dependence suggests a different explanation. After the thermal treatment, in fact, reduced GO nanoparticles dispersed in the matrix become more conductive. Hence, the applied electric field can induce charge separation in the conductive particles, thus giving rise to microscopic induced dipoles, as shown in Figure 6, and already described in literature when micro composites are involved.³⁹ The relaxation time of each induced dipole is related to conductivity, size, and orientation of each nanoparticle. The corresponding contribution to

polarization is located in the high-frequency region of the permittivity spectrum, thus resulting in a significant increase of imaginary part of permittivity at high frequencies, as can be seen in Figure 4(b) in TRGO_05 composite. It is worth noting that the overall macroscopic effect on polarization, because of the contribution of all the induced dipoles, consists in the formation of a polarization peak located at frequencies close to 1 MHz.

Table II presents the DC electrical conductivity of neat epoxy and composites measured at room temperature and electric field of 20 kV mm^{-1} . First of all, electrical conductivity increases slightly with the amount of GO filler, becoming two times the value of neat epoxy at 0.5 wt % GO loading. This behavior could be expected considering the almost insulating nature of GO, even if more conductive than base epoxy, and the small fraction of the filler amount.

It is worth noting that thermal *in situ* reduction of GO affects significantly electrical conductivity of nanocomposite. A large increment of conductivity is measured in TRGO_x samples compared to those containing GO, likely because of the

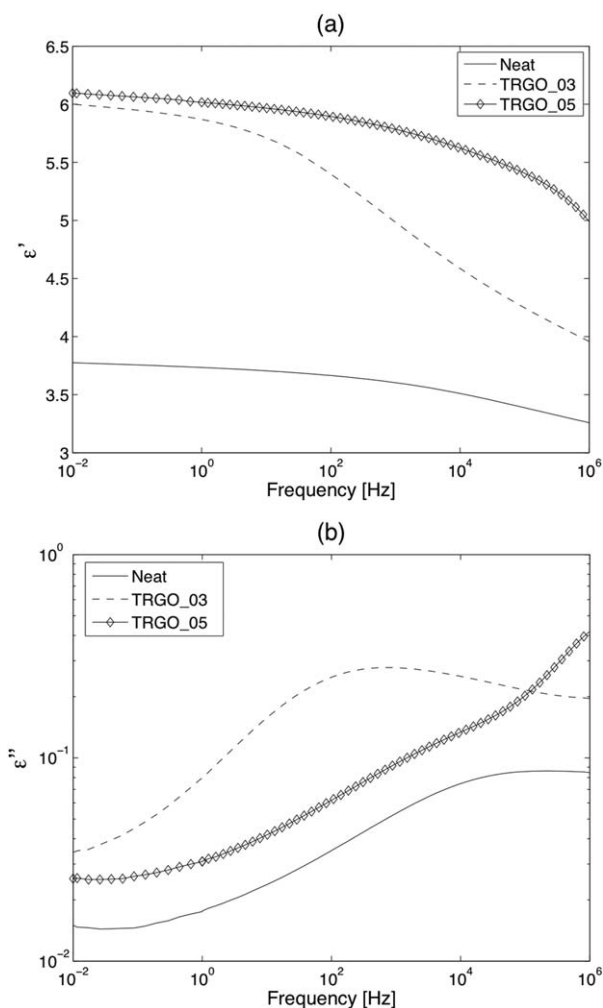


Figure 4. (a) Real part and (b) imaginary part of relative permittivity at 20°C of neat epoxy and TRGO_X composites after the thermal reduction treatment at 135°C for 48 h.

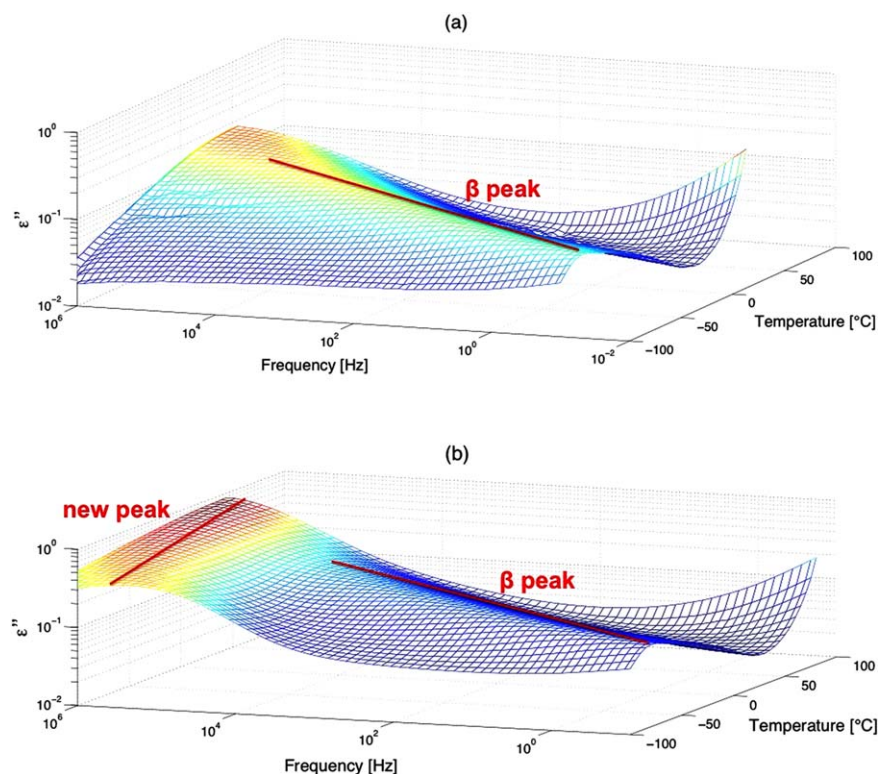


Figure 5. Imaginary part of electrical permittivity as a function of temperature and frequency: (a) base epoxy and (b) TRGO_05 nanocomposite. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

conductive nature of reduced GO. Indeed, TRGO_03 conductivity is about 55 times that of neat epoxy, while the increase shown by TRGO_05 is about 80 times.

The behavior of electrical conductivity of neat epoxy and TRGO_05 as a function of electric field is reported in Figure 7. For the sake of brevity, among the composites we focused atten-

tion on TRGO_05, which provided the largest variation of conductivity.

As expected, the conductivity of neat epoxy is almost constant up to 15 kV mm^{-1} , indicating an ohmic behavior, while significant increase can be observed for larger fields, likely because of charge injection from the electrode that turns the conduction



Figure 6. Scheme of the induced dipoles under electric field originated by the nanoparticles dispersed into the matrix. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Table II. Electrical Conductivity, σ (Mean Value and Confidence Interval at 95% Probability), at Room Temperature of the Epoxy Composites Before and After the Thermal Reduction Treatment at 20 kV mm⁻¹

Material	σ (S m ⁻¹)
Neat resin	$(3.2 \pm 1.2) \times 10^{-15}$
GO_03	$(6.6 \pm 2.5) \times 10^{-15}$
TRGO_03	$(1.7 \pm 0.9) \times 10^{-13}$
GO_05	$(7.6 \pm 1.7) \times 10^{-15}$
TRGO_05	$(2.5 \pm 1.0) \times 10^{-13}$

mechanism from ohmic to Space Charge Limited Current (SCLC).⁴⁰

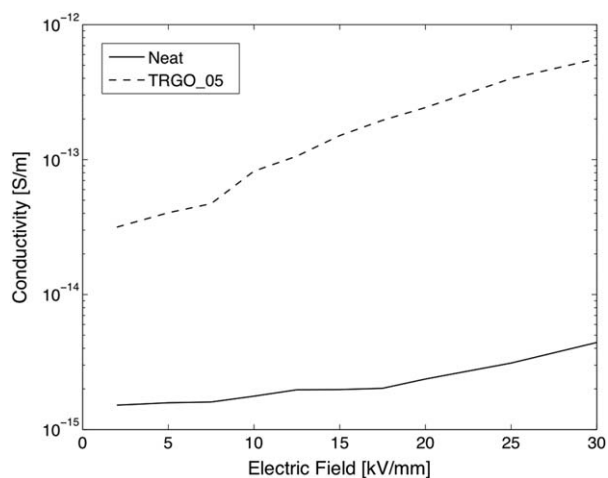
The specimens containing TRGO_05, on the contrary, show a different behavior. Indeed, electrical conductivity of TRGO_05 exhibits a large variation with electric field even at very low field values, suggesting nonohmic conduction mechanism.

This behavior could be associated with the effect of the matrix-particle interface, which would introduce distributed Schottky barriers in the polymer matrix, thus altering both charge injection and transport. This provides a strong nonlinear conduction characteristic, broadening the nonohmic region even to lower fields.²⁸

Dielectric strength results reported in Table III indicate a 25% decrease of the 63.2 th percentile (α -parameter) of breakdown field for the composites with respect to the neat epoxy. This reduction is, however, acceptable for application in high-voltage grading systems, being much smaller than that observed in thermoplastic copolymers doped with carbon-black fillers, i.e. breakdown voltage drop of about 90% with a filler content of 5 vol %.⁴¹

CONCLUSIONS

Through two-phase extraction, a proper dispersion of GO into epoxy-based component has been obtained. The efficient dispersion has been maintained also in the final cross-linked materi-

**Figure 7.** Electric field influence on electrical conductivity at 20°C of the epoxy and TRGO_05.**Table III.** Dielectric Strength Results: Weibull α and β Parameters of Breakdown Fields

Material	Weibull α -parameter (kV mm ⁻¹)	Weibull β -parameter
Neat resin	82.1 \pm 4.5	18.2
TRGO_03	65.9 \pm 3.2	21.6
TRGO_05	61.3 \pm 3.9	19.8

The confidence intervals at 95% probability are also reported for α .

als. Nanocomposites did not show relevant change in electrical properties, unless a thermal *in situ* reduction of GO is performed. The chemical change on GO leads to an enhancement of permittivity (more than 50%) and conductivity, (almost two orders of magnitude) even with very low nanofiller content (<0.5 wt %), with a very limited decrease of breakdown field, lower than 25%.

The low content of nanofiller used, as well as the simple and eco-friendly way to treat the nanocomposites (no solvent or chemical reagent are involved in the process), allow us to envisage the possibility of producing insulating materials with locally tailored properties using very low GO content. In particular, the strong nonlinear behavior of the conductivity with electric field could be exploited successfully in field-grading applications, e.g. in cable accessories and electric motor end-winding.

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