

Dielectric breakdown model for conductor-loaded and insulator-loaded composite materialsP. Bergero,¹ F. Peruani,² G. Solovey,² I. M. Irurzun,^{1,2} J. L. Vicente,¹ and E. E. Mola^{1,*}¹INIFTA (CONICET, CICPBA, Universidad Nacional de La Plata), Sucursal 4, Casilla de Correo 16, (1900) La Plata, Argentina²Departamento de Física, "Juan José Giambiagi," FCEyN, UBA, Buenos Aires, Argentina

(Received 7 July 2003; revised manuscript received 9 October 2003; published 30 January 2004)

In the present work we generalize the dielectric breakdown model to describe dielectric breakdown patterns in both conductor-loaded and insulator-loaded composites. The present model is an extension of a previous one [F. Peruani *et al.*, Phys. Rev. E **67**, 066121 (2003)] presented by the authors to describe dielectric breakdown patterns in conductor-loaded composites. Particles are distributed at random in a matrix with a variable concentration p . The generalized model assigns different probabilities $P(i,k \rightarrow i',k')$ to breakdown channel formation according to particle characteristics. Dielectric breakdown patterns are characterized by their fractal dimension D and the parameters of the Weibull distribution. Studies are carried out as a function of the fraction of inhomogeneities, p .

DOI: 10.1103/PhysRevE.69.016123

PACS number(s): 05.70.Ln, 61.41.+e, 81.05.Qk, 02.50.-r

I. INTRODUCTION

The dielectric breakdown phenomenon in solid materials has been widely studied both theoretically and experimentally due to its importance in the electrical industry. The design of insulators bearing high electric strength is highly desirable, and in the past years composite materials such as resin matrix filled by fibers or strong particles have been widely used with such purpose in many industrial applications [1–4]. For example, high density polyethylene is one of the most widely used materials for the production of insulators, and composites containing carbon black and titanium dioxide have recently been tested experimentally [5] to determine the influence of such particles on the dielectric properties of the material. It has been shown that dielectric breakdown still produces branching structures, such as those in homogeneous materials but with an extension of damage and a distribution of breakdown times dependent on the concentration and electrical characteristics of the filler. We note that while both a low failure probability and a small damage are desirable, our results suggest that this goal is not always attainable.

From the theoretical point of view, dielectric breakdown in homogeneous materials has been described as a stochastic process producing fractal structures that are called electrical trees. The most widely used model is the dielectric breakdown model (DBM), first introduced by Niemeyer, Pietronero, and Wiesmann [6], which assumes that the dielectric is homogeneous, i.e., the electrical tree propagates in a dielectric medium without inhomogeneities. The main feature in the DBM is the dependence of the breakdown probability on the local electric field in the material, a fact that attempts to consider the basic mechanism underlying breakdown in real materials. Stochastic fluctuations produce breakdown channels that damage the material increasing the local electric field and eventually producing new channels. Since its intro-

duction the DBM has been broadly studied to describe experimental results [7–13], though the physical origin of such stochastic fluctuations is beyond the DBM and cannot be explained by it [14].

The present paper addresses the problem of dielectric breakdown in composite materials. While breakdown phenomena in conductor-loaded dielectric have been studied in recent years from the standpoint of percolation theory [15,16], insulator-loaded dielectric materials have received much less attention. Theoretical efforts have concentrated on lattice models in an attempt to see whether the basic physical mechanism of breakdown can be identified in these materials. Some efforts have focused on the breakdown of fuse networks, while others have concentrated on dielectric breakdown in networks.

Our interest, however, is mainly focused on the study of fractal dielectric trees. Considering that percolation conditions impose a limit to the breakdown processes we are interested in the study of the breakdown phenomenon below that limit, i.e., how the concentration of particles in the composite material modifies the characteristics of the dielectric trees. In a previous paper [17,18] we applied the DBM to describe dielectric breakdown patterns in conductor-loaded composites and in this work we generalize the DBM to describe dielectric breakdown patterns in insulator-loaded composites. Insulating particles are distributed at random in the resin matrix, and the dielectric breakdown propagates according to new rules to take into account electrical properties and particle sizes. In this way we extend the DBM to take into account material inhomogeneities from the point of view of electrical properties.

The extension of the DBM model presented in this paper also allows us to describe dielectric breakdown patterns by means of their fractal dimension and their Weibull distribution parameters [19].

In Sec. II we present a description of the DBM, and in Sec. III the model generalization is introduced and a brief description of the model for conductor-loaded composites is also included for comparison. Results are presented in Sec. IV, and our conclusions are summarized in Sec. V.

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II. THE DIELECTRIC BREAKDOWN MODEL

In the DBM [6] the dielectric is represented by a rectangular lattice where each site corresponds to a point in the dielectric. Microscopic examination of electrical tree growth shows that branch extension occurs in increments typically of 5–10 μm , while the interelectrode gap is 1–2 mm [20,21]. This implies that a gap of 100 lattice units will represent the experimental situation adequately, and accordingly 100×100 lattices were employed in this work (the separation between nodes then represents a distance of about 10 μm). The DBM assumes that the tree grows stepwise, starting in an electrode with electric potential $\phi=0$ and ending in the counterelectrode where $\phi=1$. The discharge structure has zero internal resistance, i.e., at each point of the structure the electric potential is $\phi=0$. The tree channel growth is governed stochastically by the electric field. The probability P of a tree channel growth at each site of the electrical tree neighborhood is chosen to be proportional to a power η of the electric field E at such site ($P \propto E^\eta$). The electric field E can be written from ϕ , and therefore

$$P(i,k \rightarrow i',k') = \frac{(\phi_{i',k'})^\eta}{\sum (\phi_{i',k'})^\eta}. \quad (1)$$

The sum in the denominator refers to all of the possible growth sites (i',k') adjacent to the electrical tree.

The electric field distribution is obtained by solving the Laplace equation considering that the tree structure has the electric potential of the electrode ($\phi=0$).

Breakdown patterns generated by this model have a fractal structure that has broadly been dealt with in the literature [7–13]. The fractal structure of the trees is highly dependent on the value of the exponent η and can be characterized by their fractal dimension D and the probability of dielectric failure, $R(t)$.

The fractal dimension is defined from the correlation function $C(r)$, which is the quotient of the *average* number of lattice sites that belong to the tree, divided by the total number of lattice points that can be found within a circle of radius r . The average is performed over the set of circles of radius r centered on every point of the electrical tree. The scaling behavior of $C(r)$ with r is given by the following equation:

$$C(r) = C_0 r^{D-2}, \quad (2)$$

where D is the fractal dimension. For practical purposes, D can be considered measuring the extension of damage produced in the material, at high values of D the damage is larger.

The propagation time t is measured as the number of channels incorporated into the tree (the incorporation of a new channel represents a unit of time).

The cumulative probability of failure, $R(t)$, of a family of trees generated by computer simulations satisfies a two-parameter Weibull distribution [11,12,19], such as those observed in experimental studies, given by

$$R(t) = 1 - \exp[-(t/\alpha)^\beta], \quad (3)$$

where α is the characteristic propagation time, i.e., if we denote the gamma function Γ , the mean time to failure $\mu = \alpha\Gamma(1 + 1/\beta)$. α can be considered to show the degree of branching of the tree. β is a shape parameter, for instance, when $\beta=1$ we have the exponential distribution, when $\beta = 3.602$ we have a distribution close to a normal one, and when β is less (greater) than 3.602 the distribution has a long right (left) tail. Also, by calling the variance of the propagation time to failure σ^2 , then σ/μ is a function of β only.

III. THE COMBINED MODEL

In a previous paper [17] we extended the DBM to consider dielectric breakdown in conductor-loaded composite materials. We represented these materials by a matrix with a variable concentration p of randomly distributed conducting inhomogeneities. Inhomogeneity characteristics were introduced assigning different probabilities $P(i,k \rightarrow i',k')$ to breakdown channel formation. The model was then simplified and a numerical study comparing both was performed. We refer the reader to Ref. [17] for details about the model. Here, we consider a variation of the simplified model to also take into account dielectric breakdown in insulator-loaded composite materials. To compare it with our present results, in Sec. III A we briefly describe our previous simplified combined model with conducting particles (SCMC). Then, in Sec. III B, the simplified combined model with insulating particles (SCMI) is presented.

A. Conducting particles (SCMC)

In the DBM the probability $P(i,k \rightarrow i',k')$ of the breakdown channel growth between two nodes is chosen to be proportional to a power η of the electric field, according to Eq. (1).

In our simplified model, SCMC, $P(i,k \rightarrow i',k')$ is modified according to the following rules [17,18]

If (i',k') is a site of the polymeric matrix,

$$P(i,k \rightarrow i',k') \propto (\phi_{i',k'})^\eta, \quad (4)$$

as in the DBM [see Eq. (1)] but we assume the simultaneous and instantaneous incorporation of conducting particles when they are reached by the electrical tree. Then these incorporations are not counted in the propagation time t , measured as the number of channels incorporated into the tree. Thus, if in a step of the tree growth, sites (i',k') exist adjacent to the structure and occupied by conducting particles, they are incorporated simultaneously and instantaneously into the electrical tree.

The SCMC describes the dielectric breakdown phenomenon in conducting-loaded materials (such as carbon black-filled polymers), by considering particles with a diameter just slightly less than the length of the dielectric breakdown channel (10 μm) [18] in reasonable agreement with the experimental results. In such case, the assumption of simultaneous and instantaneous incorporation of conducting particles to the tree becomes reasonable, since the potential

gradient between either two neighboring conducting particles or a tree node and a neighboring conducting particle is very high.

B. Insulating particles (SCMI)

In this case, we assume that the presence of an insulating particle in a given site prevents the breakdown of the material at that site, i.e., we assume that the filler permittivity ϵ_f is much greater than polymer permittivity ϵ_p . Then, we again modified the probability $P(i,k \rightarrow i',k')$, according to the following rules

If (i',k') is a site of the polymeric matrix,

$$P(i,k \rightarrow i',k') \propto (\phi_{i',k'})^\eta, \quad (5a)$$

as in the DBM [see Eq. (1)], but if (i',k') is occupied by an insulating particle, then

$$P(i,k \rightarrow i',k') = 0. \quad (5b)$$

According to Eq. (5), sites (i',k') , which are occupied by an insulating particle, are not incorporated into the electrical tree.

The SCMI attempts to describe the experimentally observed effect of the inclusion of high-permittivity particles in polymer matrices. Such particles increase the resistance of the materials excluding the electric breakdown path from them, a property that is well represented by the SCMI. The SCMI also assumes a diameter particle slightly less than the dielectric breakdown channel.

IV. RESULTS

The SCMI was numerically explored simulating electrical trees and characterizing them by the fractal dimension D and the α and β parameters of the Weibull distribution [see Eq. (3)]. Their dependencies on both the concentration of particles p and the exponent η were studied. Trees were simulated on 100×100 lattices and sets of 100 trees were generated for each value of p and η .

Figure 1 shows three electrical trees generated with different values of p . When the filler fraction p is sufficiently high, electrical trees cannot propagate across the matrix, i.e., they never reach the counterelectrode. Therefore, a critical concentration exists, p^* , beyond which dielectric breakdown does not occur. From our simulations $p^* = 0.42 \pm 0.03$ for 100×100 lattices. This value of p^* can also be derived from percolation concepts. Considering a percolation problem in which the clusters are defined as the set of particles being both first and second nearest neighbors, a critical value ~ 0.42 is obtained, instead of the value 0.59 that is obtained considering only first nearest neighbors. Physically, the phenomenon occurs when all possible growth sites for the tree are occupied with insulating particles.

Below p^* , the cumulative probability of failure follows a Weibull distribution with α and β parameters depending on p and η . Figure 2 shows both behaviors. In Fig. 3 we quoted the behavior for the SCMC for comparison.

Finally, in Fig. 4 we show the dependence of fractal di-

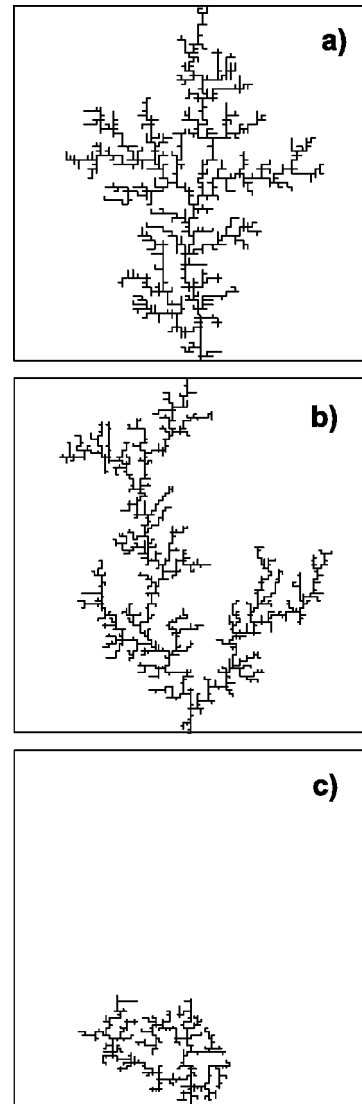


FIG. 1. Electrical trees grown in composite materials with an increasing fraction p of insulating particles. (a) $p=0$ is an electrical tree such as those simulated in Refs. [10], (b) $p=0.20$ ($p < p^*$), (c) $p=0.42$ ($p = p^*$). A tree that does not reach the counterelectrode is shown.

mension D on p and η for both models, the SCMI and the SCMC.

V. CONCLUSIONS

The DBM produces fractal electrical trees that can be well compared with those obtained in experiments. Our purpose here was to study the effect of the concentration of filler in composite materials on the fractal characteristics of the trees. The DBM was then modified assigning different probabilities to the breakdown channel formation. Two cases were considered: conducting and insulating particles in comparison with the matrix, leading to the SCMC and the SCMI, respectively. Insulating particles are distributed at random and have the effect of excluding the electric breakdown path from them, a behavior that can be well described by the SCMI.

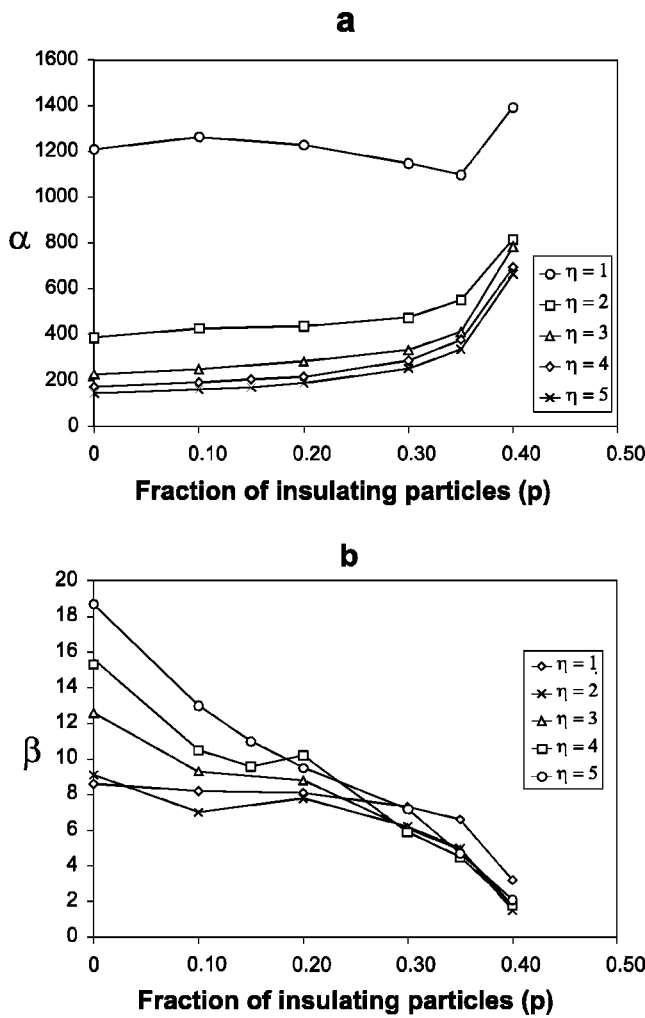


FIG. 2. Dependence of Weibull distribution parameters (a) the characteristic time α and (b) the shape factor β , on the fraction of insulating particles, p and η , calculated from a set of 100 electrical trees by employing the SCMI.

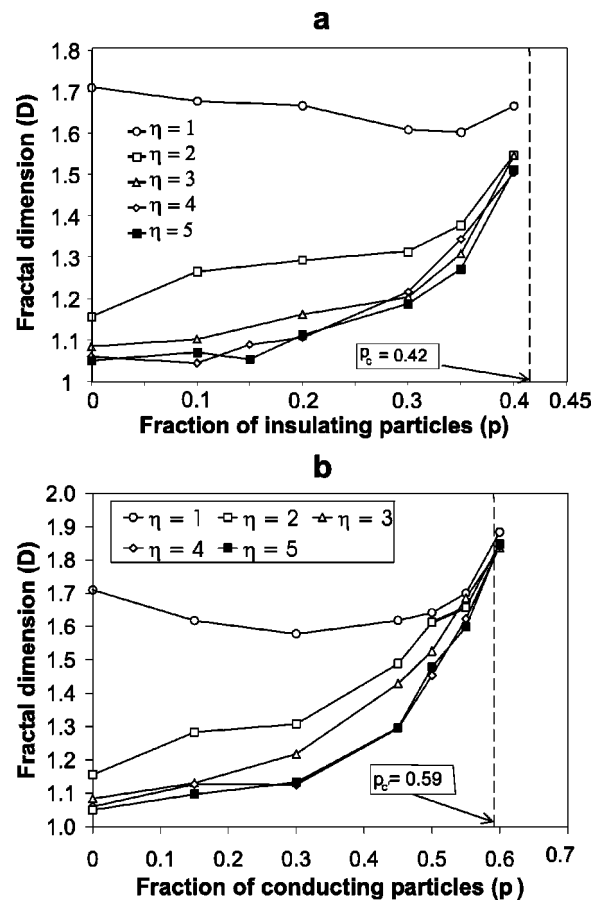


FIG. 4. Dependence of the fractal dimension D on the fraction of particles p and η calculated from a set of 100 electrical trees by employing the SCMI (a) and the SCMC (b).

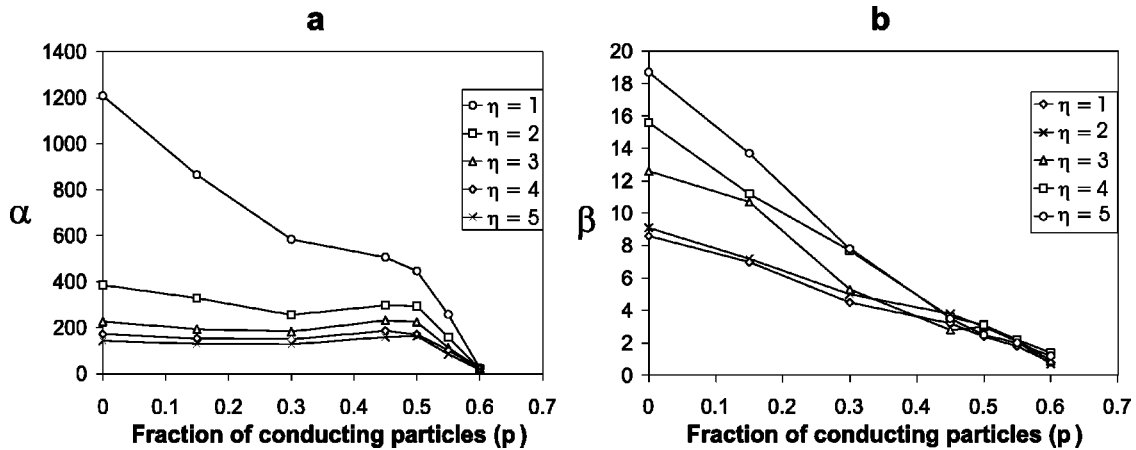


FIG. 3. Dependence of Weibull distribution parameters (a) the characteristic time α and (b) the shape factor β , on the fraction of conducting particles, p and η , calculated from a set of 100 electrical trees by employing the SCMC.

A numerical study comparing both the SCMC and the SCMI was performed. Electrical trees are characterized by their fractal dimension D and the α and β parameters of the Weibull distribution. Their dependencies on both the concentration of particles p and the exponent η were studied.

In the SCMI a critical concentration exists ($p^*=0.42 \pm 0.03$) beyond which dielectric breakdown does not occur. This phenomenon happens when all possible growth sites for the tree are occupied with insulator particles. Below p^* the propagation time distribution follows a Weibull distribution with α and β parameters depending on p and η . Also, the fractal dimension of electrical trees, D , depends on p and η , below p^* . Qualitatively, similar behaviors of α , β , and D as a function of p are obtained for all values of η , except $\eta = 1$.

The fractal dimension D behaves monotonically with p (see Fig. 4) when $\eta > 1$. Whereas for $\eta = 1$, D presents a minimum. Electrical trees simulated with $\eta = 1$ are characterized by a higher degree of branching compared with those simulated with greater values of η . Insulating particles basically act as obstacles inhibiting possible paths for branching, and therefore D decreases in the interval $0 \leq p < 0.35$. While the branching capacity of trees with $\eta = 1$ is enough to find a path to reach the counterelectrode, D will decrease only accounting for the reduction of growing paths. In the interval $0.35 \leq p < p^*$ the reduction in the number of breakdown channels is so strong that the electrical trees have to increase their degree of branching to reach the counterelectrode and D increases. Accordingly, α also exhibits a nonmonotonic behavior for $\eta = 1$ diminishing even below its value for $p = 0$.

On the other hand, insulating particles in electrical trees simulated with $\eta > 1$ have the effect of increasing the degree of branching in the whole interval $0 \leq p \leq p^*$, because dielectric trees themselves have a low capacity for branching. Breakdown structures now must explore more alternative paths as p is increased and, consequently, α increases with p for $\eta > 1$.

As p approaches p^* ($p \rightarrow p^*$) the values of α seem to merge together for $\eta > 1$. α roughly represents the branching degree of the trees, which diminishes as η increases. One could speculate then about the existence of a common value for α in $p = p^*$ (and for $\eta \gg 1$) because the number of possible growing paths for the tree decrease strongly.

The main difference in the breakdown process between a polymer matrix with conducting or insulating filler is in the behavior of the characteristic propagation time α . While in the SCMC a reduction in α is observed when the fraction of conducting particles, p , is increased [Fig. 3(a)], in the SCMI α increases (for $\eta > 1$) with the fraction of insulating particles, p [Fig. 2(a)].

The dependence of β which is the shape parameter of the Weibull distribution, on p and η is qualitatively similar for both the SCMI and the SCMC. As p increases, the distribution becomes broad approaching an exponential distribution

($\beta \sim 1$). In the SCMI this behavior indicates a large proportion of breakdowns at short times. It is important to note that although the inclusion of insulator filler in the composite increases the average failure time and α (which is a desirable property), it also produces a strong reduction of β and a broad distribution of failure time that in practical applications means a loss in the reliability of the material.

In the SCMC the reduction of β is accompanied by a reduction of α and is a consequence of the instantaneous incorporation of the conducting particles to the tree.

For practical purposes the extension of damage also has an economic impact, and in this sense, the variation of D must also be considered.

Finally, it is interesting to note that in the SCMC and due to the presence of conducting particles, electrical damage must be distinguished from mechanical damage. For example, the reduction of α with the increase of p indicates that the material rapidly becomes a conductor but in fact, it also shows that the number of breakdown channels is very small. Since the conducting particles are incorporated instantaneously to the tree, they are not included in the calculation of the propagation time; note, however, that D measures the extension of the structure including conducting particles. Since the direction of the electrical tree propagation is known, such property can be useful to detect small mechanical failures in the material.

The model presented in this paper mimics quite well fillers with very high permittivity and mechanical strength, a combination that makes it extremely difficult for them to be penetrated by an electrical tree. Treeing breakdowns therefore avoid the fillers whenever possible, even to the extent of adopting extremely low field tortuous paths. Calculations show that for materials whose dielectric breakdown is described by η values greater than one, the onset of tortuosity defines the smallest p value (≈ 0.30) for effective breakdown inhibition. This can be followed by looking either at the dependence of η (Fig. 4) on p or at the dependence of α (Fig. 2) on p .

In practical insulation systems the quality of the polymer-insulation interface will clearly assume a major importance. Poor binding or contributory mechanical stresses will cause the interface to fail rapidly and hence facilitate the path of the breakdown around the filler particle thereby reducing its effectiveness.

ACKNOWLEDGMENTS

This research project was financially supported by the Consejo Nacional de Investigaciones Científicas y Técnicas, the Comisión de Investigaciones Científicas de la Provincia de Buenos Aires, and by the Universidades Nacionales de La Plata (UNLP) and Buenos Aires (UBA). E.E.M. acknowledges the useful discussions held with Professor L. A. Dissado, University of Leicester, United Kingdom, during his visits at that University.

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