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Dielectric breakdown model for composite materials

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This paper addresses the problem of dielectric breakdown in composite materials. The dielectric breakdown model was generalized to describe dielectric breakdown patterns in conductor-loaded composites. Conducting particles are distributed at random in the insulating matrix, and the dielectric breakdown propagates according to new rules to take into account electrical properties and particle size. 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 conducting inhomogeneities, p. The fractal dimension D of electrical trees approaches the fractal dimension of a percolation cluster when the fraction of conducting particles approximates the percolation limit.

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I. INTRODUCTION

Polymers are in general insulated materials, with low values of electrical conductivity and a low value of dielectric constant. One of the ways to increase electrical conductivity is to introduce high electrical conductivity fillers such as metal powders or carbon black in the polymer matrix. Then, the value of the electrical conductivity of the compound can be increased several orders of magnitude depending on the volume fraction and the dispersion of this second phase in the matrix.

When introduced in the polymer, filler particles can adopt different types of structures that are sometimes characterized by a fractal geometry [1], and a transition from insulating to conductive behavior is observed when the filler volume fractions are about 25%, depending on the type of filler [2]. In compounds with metal powder as a second phase, a percolation threshold is experimentally confirmed by a sharp change in the electrical conductivity. In the case of carbon black compounds, this change is not so sharp and a transition for filler volume fractions in the range between 15% and 35% is expected. This behavior would be the consequence of the filler network formed during the different steps in the mixing process [3–7].

In the past decades, models of the electrical conductivity of filled composites were proposed in the frame of three classes: the composite medium approach based on Maxwell equation [8], the discrete medium approach based on Kirkpatrick's ideas [9], and the percolation approach. This last approach was analyzed by Pike and Seager [10], who investigated the problem of percolation and conductivity with computer simulation.

This paper addresses the problem of dielectric breakdown in composite materials. Breakdown phenomena in conductor-loaded dielectrics have received some attention in recent years from the standpoint of percolation theory [11,12]. Theoretical efforts have concentrated on lattice models in an attempt to see whether the basic physical mechanisms of breakdown in these materials can be identified. Some efforts have focused on the breakdown of fuse networks, while others have concentrated on dielectric breakdown in networks.

Experimentally, high-density polyethylene (HDPE) composites containing carbon black and titanium dioxide have recently been tested [13]. The results of the dielectric breakdown test were analyzed by their Weibull distribution, and it has been concluded that the shape parameter β of the distribution may be used to evaluate the dispersion of carbon black agglomerations in HDPE compounding formulations.

HDPE is one of the most widely used materials for the production of insulators, spacers, and also for coating conducting cables used in electric power distribution networks, and, in this type of application, the dielectric strength is one of the properties that must be taken into account in order to check the ability to withstand high electric fields [14]. On the other hand, the development of formulations containing additives to protect polymers against property decay (e.g., mechanical and thermomechanical) during the processing stages and/or in service [15] is highly desirable technologically, and in the case of applications in electrical insulation these additives may impair electrical properties.

Dissado and co-workers [16] studied a narrow size distribution of irregular aluminum particles blended into power cable insulation-grade polyethylene. The failure statistics of the loaded polymers were then determined under ac ramped stress. They demonstrated the validity of the percolation model expression for the characteristic breakdown strength, i.e., a reduction in the characteristic value of the applied field E with an increasing particle volume fraction p.

In this paper we generalize the dielectric breakdown model (DBM) to describe dielectric breakdown patterns in conductor-loaded composites. The DBM was introduced by Niemeyer, Pietronero, and Wiesmann [17] and assumes that the dielectric is homogeneous, i.e., the electrical tree propagates in a dielectric medium without inhomogeneities. In the DBM, material electrical properties are represented by the exponent η.

In the present work, conducting particles are distributed at random in the insulating matrix, and the dielectric breakdown propagates according to new rules to take into account

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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 by their Weibull distribution parameters.

In Sec. II we present a description of the DBM, and in Sec. III the new model is introduced. Results are presented in Sec. IV, and our discussion and conclusions are summarized in Secs. V and VI.

II. DIELECTRIC BREAKDOWN MODEL (DBM)

In the DBM [17] 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, [18]. This implies that a gap of 100 lattice units will represent the experimental situation adequately and, accordingly, 100 $\times 100$ lattices were employed in this work (therefore, the separation between nodes represents a distance $L = 10\ \mu\text{m}$). 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

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

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 [17–21]. The fractal structure of the trees is highly dependent on the value of the exponent η.

Experimental and simulated electrical trees can be characterized by their fractal dimension D and failure probability.

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:

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

where D is the fractal dimension.

The probability of dielectric failure is usually determined as a function of the propagation time t, 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, $P(t)$, of a family of trees generated by computer simulations satisfies a two-parameter Weibull distribution [22], such as those observed in experimental studies, given by

$$P(t) = 1 - \exp[-(t/\alpha)^{\beta}], \tag{3}$$

where α is the characteristic propagation time and β is a shape parameter.

III. COMBINED MODEL (CM)

Previous models of dielectric breakdown [17–19] were developed for homogeneous materials. We now want to discuss the basic aspects that should be taken into account to study composite materials. These kinds of material could be represented by a matrix with randomly distributed inhomogeneities. In a real material the matrix could be represented by a polymer, and the inhomogeneities by carbon black, aluminum, or titanium dioxide, i.e., a highly insulating matrix surrounding conductor inhomogeneities.

In order to build up a model for composite materials, we should first define some characteristics of inhomogeneities, such as electrical properties, shape, size, etc.

As a first approximation we assume conducting inhomogeneities of circular shape (with a diameter not much less than the length L_0 of a breakdown channel) randomly distributed in a two-dimensional geometry. Note that inhomogeneities are centered at the matrix nodes, and therefore, inhomogeneities do not form equipotential clusters. This assumption simplifies the calculation of the electric potential during tree growth.

In the DBM and according to Eq. (1), the probability P of breakdown channel growth between two nodes is chosen to be proportional to a power η of the electric field, and therefore

$$P(i,k \rightarrow i',k') = \frac{\left(\dfrac{\phi_{i',k'}}{L_i}\right)^{\eta}}{\sum \left(\dfrac{\phi_{i',k'}}{L_i}\right)^{\eta}}, \tag{4}$$

where we have explicitly introduced L_i as a breakdown channel length (Note that in the DBM all channels have the same length.)

The extension of the DBM presented in this work introduces inhomogeneity characteristics, assigning different probabilities P to the breakdown channel formation, according to the conducting characteristics at each site. The situation can be rationalized introducing different values of L_i in Eq. (4). We note that this modification affects only the probability P assigned to each site adjacent to the electrical tree. As indicated in Fig. 1, L_i is written as

$$L_i = L_0 - \frac{L_0}{a_i}, \tag{5}$$

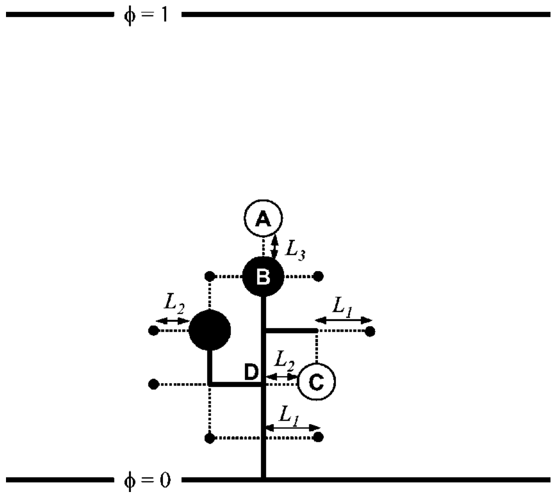

FIG. 1. This figure represents the dielectric breakdown model in a composite material. Large circles represent conducting particles. The black circles are already incorporated in the electrical tree, whereas the white circles are not.

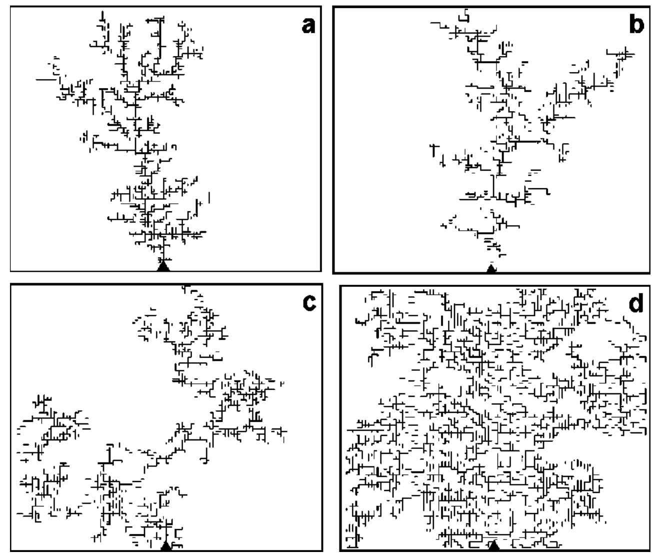

FIG. 2. Electrical trees grown in composite materials with an increasing fraction p of conducting particles. (a) $p=0$ is an electrical tree such as those simulated in Refs. [20,22]. Electrical trees in (b)–(d) were simulated with SCM [Eq. (6)], with a fraction of conducting particles, $p=0.15, 0.45$, and 0.60, respectively.

where a_i $(i=1,2,3)$ is a parameter taking three different values:

(1) $a_i=a_1\rightarrow\infty$, for a channel connecting the electrical tree with a lattice point nonoccupied with a conducting particle. This process is equivalent to the DBM (see Sec. II).

(2) $a_i=a_2=2$ if the channel is connecting a conducting particle that belongs to the electrical tree with a lattice point, or an electrical tree node (that is not a conducting particle) with a conducting particle that does not belong to the electrical tree (see sites C and D in Fig. 1). We emphasize that we are considering conducting particles with a diameter not much less than L_0.

(3) $a_i=a_3\gtrsim 1$ if the channel is connecting two conducting particles, where one of them belongs to the electrical tree. Note that this channel is very short.

Therefore, in our model, tree growth is still governed stochastically by the electric field, as in the DBM, with a probability P given by Eq. (4), but with L_i given by the following:

(i) If (i,k) and (i',k') are sites of the polymeric matrix, $L_i=L_0$.

(ii) If (i,k) is an electrical tree node and if in (i',k'), there is a conducting particle, $L_i=L_0/a_2=L_0/2$.

(iii) If in (i,k) there is a conducting particle and (i',k') is a site of the polymeric matrix, $L_i=L_0/a_2=L_0/2$. (iv) If in (i,k) and in (i',k') there are conducting particles, $L_i=(a_{3-1}/a_3)L_0$. Thus in this combined model, P depends not only on the electric field, but also on the conducting characteristic of the site.

We now perform an extension to the previous model, introducing some simplifying assumptions. L_i is equal to $L_0/2$ either in cases (ii) or (iii). However, from the physical viewpoint there is no justification to assume different probabilities whether the channel begins in a conducting particle or not. We are in favor to assume, as an extension of the previous model, that probabilities in cases (i) and (iii) (named P_1 and P_3, respectively) are equal, i.e., $P_3=P_1$. Also, in fact, these probabilities will now be much smaller than those in cases (ii) and (iv) $(P_2$ and $P_4)$. To simplify the numerical simulations we assume that both of them are roughly equal, and as a first approximation $P_2=P_4=1$.

Therefore we come to the following simplified combined model (SCM):

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

$$P(i,k\rightarrow i',k')\propto(\phi_{i',k'})^{\eta}, \tag{6.1}$$

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

$$P(i,k\rightarrow i',k')=1. \tag{6.2}$$

According to Eq. (6), sites (i',k'), which are occupied by a conducting particle, are incorporated with probability 1 into the electrical tree. We also assume that the incorporation of such particles is instantaneous, i.e., they are not counted in the propagation time t, measured as the number of channels incorporated into the tree. Thus, if in a step of tree growth sites (i',k') are adjacent to the structure and occupied by conducting particles, they are incorporated simultaneous and instantaneously into the electrical tree.

In the following section we will compare results obtained from both CM and SCM according to Eq. (6), for different fractions p of conducting particles.

IV. RESULTS

We will now present a study of electrical trees simulated with the model developed in the preceding section. The dependence of their fractal dimension and propagation times on the conducting particle fraction will be studied. We will begin with results obtained with the SCM, Eq. (6), and then

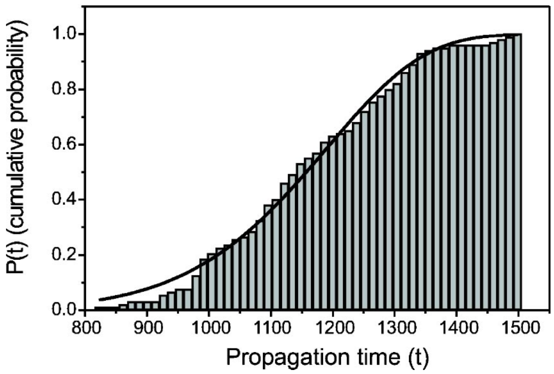

FIG. 3. Normalized cumulative probability of propagation times calculated from a set of 100 electrical trees by employing SCM, Eq. (6). Time is measured as the number of bonds incorporated in the tree. Parameter values employed were $\eta = 1$ and $p = 0.45$.

continue with the results derived from the CM. Finally, a comparison of results obtained from the CM and the SCM will be performed.

Figure 2 shows four electrical trees simulated on lattices with growing concentration p of conducting particles and $\eta = 1$. The propagation time of these electrical trees also follows a Weibull distribution like those grown on lattices with $p = 0$, see Ref. [21] and references therein. Figure 3 shows the dependence of the cumulative probability of failure, $P(t)$, on the propagation time t of a set of 100 electrical trees grown with the set of parameters $p = 0.45$ and $\eta = 1$. The Weibull distribution parameters α and β, see Eq. (3), depend on the concentration of conducting particles p and on the parameter η (see Fig. 4). It is interesting to point out that shape parameter β decreases monotonically by increasing p, therefore as $p \rightarrow p_c$, the Weibull distribution is narrowed around α.

Electrical trees are characterized by their fractal dimension D obtained by a log-log plot of their average correlation function $C(r)$ versus r [see Eq. (2)]. Figure 5 shows the dependence of fractal dimension D on the set of parameters p and η.

The corresponding results obtained from CM are shown in Figs. 6 and 7. In Fig. 6 we show the Weibull parameters α and β as a function of p and η. In Fig. 7 we show the dependence of the fractal dimension D (of a set of 100 electrical trees) on the fraction of conducting particles p for different values of the parameter η.

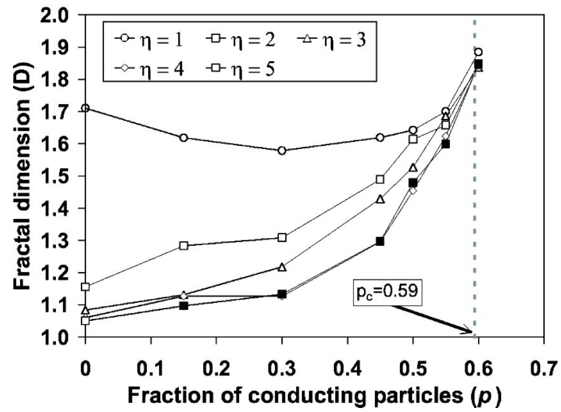

FIG. 5. Dependence of the fractal dimension D on the fraction of conducting particles, p, calculated from a set of 100 electrical trees by employing SCM, Eq. (6).

A comparison of the fractal dimension of electrical trees simulated with either SCM or CM (Figs. 5 and 7, respectively) shows that there is a remarkable agreement in the fractal dimension of electrical trees in the interval $0 < p < 0.3$.

V. DISCUSSION

To understand the dependence of the fractal dimension D of electrical trees on the fraction p of conducting particles (see Figs. 5 and 7), we will resort to some elements from the percolation theory.

As particles are randomly added to the lattice, nearest neighbor particles will form clusters as in the percolation model. The size of clusters grows by increasing the fraction p of conducting particles. Therefore, for a sufficiently large value of p, the cluster size will be of the order of the interelectrode gap. This cluster size limit is known as the *percolation cluster*. From percolation theory it is well known that the greatest cluster size N scales with the fraction p of conducting particles as follows:

$$N \propto \begin{cases} \ln(p) & \text{for } p < p_c \\ p^{Dp} & \text{for } p = p_c \\ p_\infty & \text{for } p > p_c, \end{cases} \tag{7}$$

where p_c is a critical concentration. If $p < p_c$, there exist only clusters of finite size, whereas if $p \geqslant p_c$ there exists a

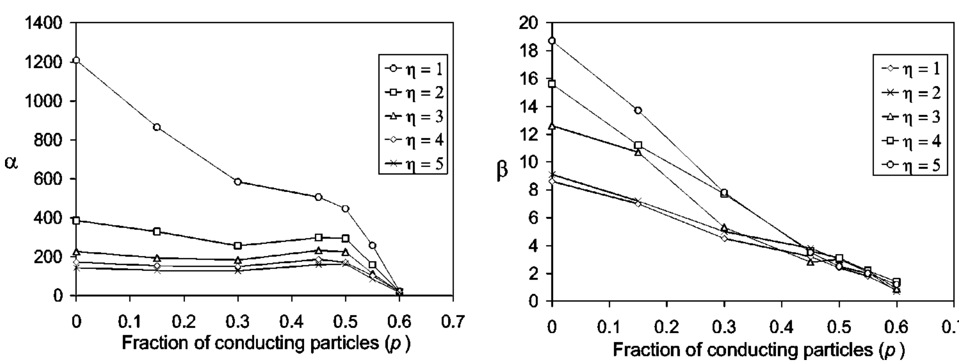

FIG. 4. Dependence of the Weibull distribution parameters α (characteristic time) and β (shape factor) on the fraction of conducting particles, p, calculated from a set of 100 electrical trees by employing SCM, Eq. (6).

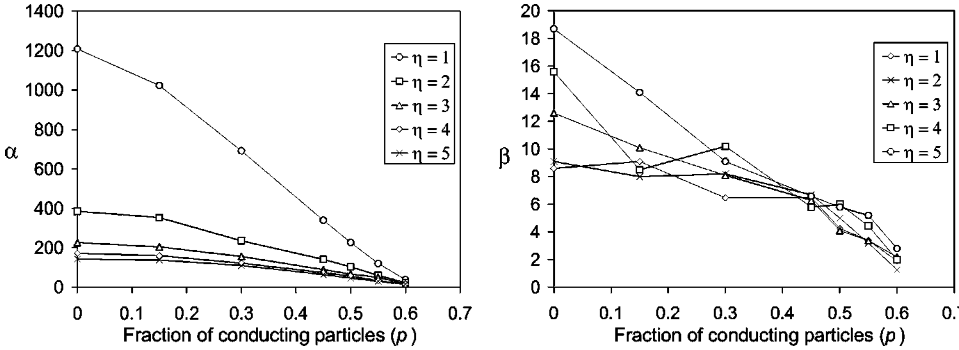

FIG. 6. Dependence of the Weibull distribution parameters α (characteristic time) and β (shape factor) on the fraction of conducting particles, p, calculated from a set of 100 electrical trees by employing CM.

cluster that bridges the interelectrode gap. The percolation limit observed in our simulations with a matrix size 100×100 was $p_c=(0.59\pm0.01)$, in remarkable agreement with the values found in the literature [23], whereas the percolation dimension of the clusters was $D_p=(1.89\pm0.03)$. Figure 8 shows the dependence of percolation probability $P(p)$ (i.e., the fraction of percolating clusters) on the fraction p of conducting particles. The critical fraction p_c was estimated by adjusting $P(p)$ to the function $G(p)$ defined as follows:

$$G(p)=\frac{p^{\alpha}}{(p_c)^{\alpha}+p^{\alpha}}. \tag{8}$$

In the SCM, described by Eq. (6), when a growing electrical tree incorporates a conducting particle, it will also incorporate all their conducting particle nearest neighbors, see particles A and B in Fig. 1. From Figs. 5 and 7 we learn that the fractal dimension of our simulated electrical trees obeys the expected percolation behavior when the fraction of conducting particles approaches the critical fraction p_c.

To investigate the dependence of the fractal dimension D of electrical trees on the fraction of conducting particles in the limit $p\to0$ (see Figs. 5 and 7), we study how strong the perturbation produced by the conducting particles on the electrical tree structure is. The following procedure was applied.

(1) An electrical tree is simulated using the dielectric breakdown model (Sec. II) with a given η value.

(2) A fraction p of conducting particles is then "randomly added" to the lattice employed to perform the simulation.

(3) Those clusters of conducting particles that are nearest neighbors to the electrical tree are added to it (see particles A and C in Fig. 1).

(4) Once step (3) is fulfilled, the correlation function of this "new" electrical tree is determined. For every p value investigated, 100 simulations were performed. From the correlation function $C(r)$ a fractal dimension D is evaluated.

The procedure described in steps (1)–(4) is repeated for every η value investigated, the results obtained are shown in Fig. 9.

We learn from Fig. 9 that for rather low p values ($p<0.3$) there is a remarkable agreement with the results shown in Figs. 5 (CM) and 7 (SCM). Agreement between these figures is also observed when the fraction of conducting particles approaches the critical value p_c.

VI. CONCLUSIONS

In this paper, we generalized the DBM to describe dielectric breakdown patterns in conductor-loaded composites. Conducting particles are distributed at random in the insulating matrix, and the dielectric breakdown propagates according to new rules to take into account electrical properties and particle size.

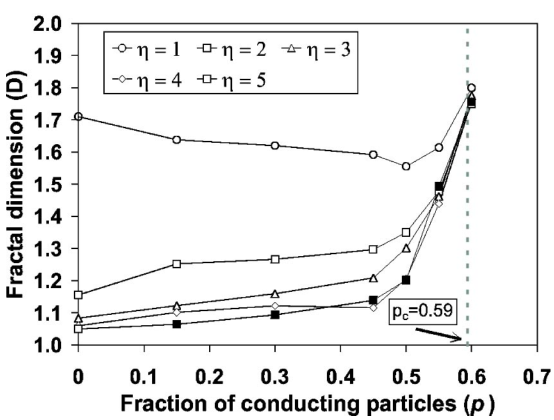

FIG. 7. Dependence of the fractal dimension D on the fraction of conducting particles, p, calculated from a set of 100 electrical trees by employing CM with parameter values $a_2=1/2$ and $a_3=100/99$.

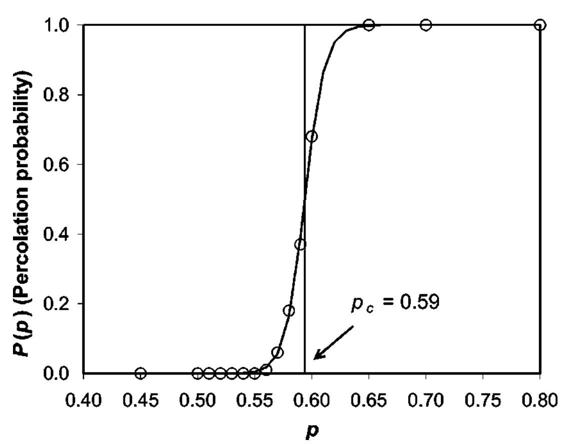

FIG. 8. Probability of percolation on a 100×100 lattice. Circles represent the simulation results. The continuous curve is an adjustment of the simulation performed with the function $G(p)$, Eq. (8).

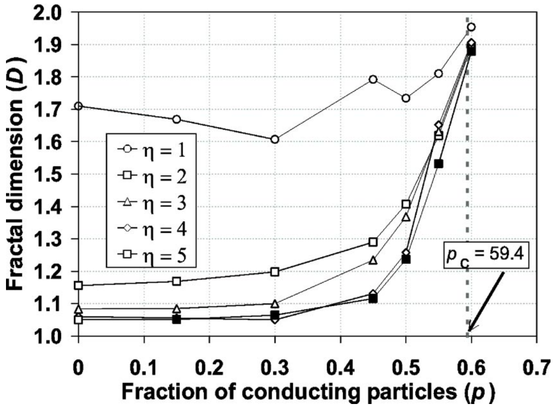

FIG. 9. Dependence of the fractal dimension D on the fraction of conducting particles, p, when electrical trees are simulated by the procedure indicated in the Discussion section [see steps (1)–(4)]. This figure should be compared with Figs. 5 and 7.

Dielectric breakdown patterns are characterized by their fractal dimension and the parameters of Weibull distribution. Studies are carried out as a function of the fraction of conducting inhomogeneities.

A reduction in the characteristic propagation time α is observed when the fraction p of conducting particles is increased (Figs. 4 and 6). This reduction is particularly noticeable when $\eta=1$. Consequently, as the fraction of conducting particles and η are increased, β (Weibull shape parameter) and its dispersion are smaller (Figs. 4 and 6). Therefore, as η is increased, the breakdown time distribution becomes sharper around its mean value.

The fractal dimension D of electrical trees approaches the fractal dimension of a percolation cluster when the fraction of conducting particles approaches the percolation limit $p \rightarrow p_c$ (Figs. 5 and 7), independent of the η value employed to perform the simulation.

In addition, if $p<0.3$, conducting particles do not significantly affect electrical tree growth, from the standpoint of their fractal dimension, see Figs. 5, 7, and 9.

Finally, the two approaches named SCM and CM show a remarkable agreement, see Figs. 5 and 7. Additional studies continuously changing a_3 in the CM, as well as incorporating a distribution of values of a_3 to simulate particles of different sizes, are in progress. Also, although stochastic models are useful for a qualitative description of breakdown processes, they leave unanswered questions concerning the origin and growth of the dielectric breakdown. In this sense, work is being done in order to develop more deterministic models at the Universities of Leicester, Buenos Aires, and La Plata.

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