Fractal analysis of electrical trees in a cross-linked synthetic resin

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A statistical picture of dielectric breakdown in cross-linked polyester resins for a two-dimensional geometry is presented and discussed in this paper. A connection is established between the dielectric breakdown model (DBM) and the physical properties of the resin. Distribution propagation times of simulated trees obey a Weibull statistics, as was experimentally found. This adjustment is achieved by a redefinition of the unit of time, which is different from the one employed up to date. The experimental dependence of characteristic propagation times on the fractal dimension D can be reproduced in the range $1.2<D<1.5$. A relationship is established between the glass transition temperature T_g and the DBM parameter η, which is in agreement with thermodynamical considerations. It is suggested that fractal characteristics of electrical trees should be related to a basic material property, such as the cross-linking density, which implies a notion of universality that deserves to be explored.

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I. INTRODUCTION

Dielectric breakdown of insulator materials leads to stochastic branching patterns or fractal structures. Fractal structures are characterized [1] by the relationship between $N(r)$, the total number of branches inside a circle of radius r, and the radius r itself. This relationship should be a power law with a noninteger exponent D

$$N(r)\sim r^D, \tag{1}$$

where D is called the fractal dimension.

In 1982 Sawada *et al.* [2] introduced the stochastic model to describe the fractal nature of branching structures that are similar to those obtained by dielectric breakdown experiments. Later on Niemeyer, Pietronero, and Wiesmann [3,4] developed the dielectric breakdown model (DBM), in which the growing probability on each site was considered to be proportional to an electric field power at that site. In 1986 Wiesmann and Zeller [5] introduced modifications to the DBM model in order to apply it to very different environments of the stepwise propagation of those damaged structures known as electrical trees.

The introduction of stochastic models has allowed the comparison between experimental structures and those generated by numerical simulation. Generally, it is considered that the fractal dimension depends on the physical and chemical characteristics of the material, the applied voltage, the presence of mechanical forces, etc.

Cooper and Stevens [6] have studied the relationship between the fractal dimension and the cross-linked level in polyester resins. They observed that an increase in the postcure temperature (and then the cross-linked level) increases the fractal dimension and the characteristic propagation time of electrical trees. Similar results have been obtained by Maruyama *et al.* [7] in cross-linked polystyrene. Other factors, such as the strength and polarity of the applied voltage, also modify the fractal dimension, showing that it strongly depends on the internal characteristics of the material. An extensive compilation of the experimental evidence has been performed recently by Kudo [8]. However, a deep analysis of the relationship between the fractal nature of the electrical trees and properties of the material is required.

In this paper, we make a complete comparison between the trees generated by the DBM model and those experimentally obtained by Cooper and Stevens [6] in cross-linked polystyrene materials. A description of the experimental data is given in Sec. II . The DBM model and the modifications performed in this work are given in Sec. III. We also discuss the different methods used in the determination of the fractal dimension, especially that utilized by Cooper and Stevens [6] in the analysis of experimental structures.

Finally, in Sec. IV we compare experimental data with numerical simulations and found a description and interpretation for them, with an adequate redefinition of the basic parameters.

II. DIELECTRIC BREAKDOWN PATTERNS: EXPERIMENTAL EVIDENCE

The electrical discharge process in insulator materials occurs in three steps: initiation, propagation, and termination. The tree patterns are formed during the propagation stage and propagation time is defined as the time from initiation (i.e., first appearance of a tree channel) to the complete bridging of the electrode gap. Cooper and Stevens [6] have studied the influence of the physical properties of a cross-linked polyester resin on its electrical tree behavior in the point-plane geometry. Blends of an unsaturated linear polyester with styrene monomer ($C_6H_5CH=CH_2$) were used. The addition of a methylethylketona hydroperoxide catalyst causes simultaneous styrene polymerization and cross-linking. As the reaction proceeds the cross-link density increases beyond the gel point. To obtain a range of network characteristics and physical properties, the resin systems were initially cured to cessation of the cross-linking reaction at 25 °C followed by a postcure at a range of temperatures up

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to 80 °C. These polymer networks, which are initially formed at 25 °C, have the ability to continue to cross-link as the postcure temperature (T_{post}) is raised. At 70 °C, the network has fully reacted and the cross-link density is at its maximum value. Thus, T_{post} is correlated with the cross-link density (see Cooper and Stevens [6] for details).

Cooper and Stevens [6] have also reported measurements of the glass transition temperature (T_g) as a function of postcure temperature made by differential scanning calorimetry (DSC). Electrical trees were studied in the point-plane geometry, using 5-μm pin tip radius steel needles and a 2-mm separation between electrodes.

For each postcure temperature, the propagation time distribution was measured. Cooper and Stevens [6] have analyzed the experimental data using two-parameter Weibull statistics [9],

$$P(t)=1-\exp[-(t/\alpha)^{\beta}], \tag{2}$$

where $P(t)$ is the cumulative probability, α is the characteristic propagation time, and β is a shape parameter.

Hence, α and β values were reported as a function of postcure temperature. An increase in α with T_{post} was observed, while β was nearly constant.

Tree patterns were structurally characterized measuring their fractal dimension. The authors employed a simple radial method, in which an origin is chosen at the needle point and arcs of known radii are drawn from the origin. The effective length of all tree segments within each arc was counted and the cumulative number of segments L as a function of radius R was obtained. The scaling law

$$L(R)\sim R^{D} \tag{3}$$

was used to obtain the fractal dimension D.

In Sec. III we compare this method with the more exhaustive correlation function method. However, in order to compare our simulated results with experimental data, we used the radial method to calculate the fractal dimension D. Cooper and Stevens [6] have determined D as a function of T_{post} and found an increasing value of D as the postcure temperature T_{post} is raised.

For theoretical comparisons that will be developed in Sec. IV, in Fig. 1 we plotted the experimental characteristic propagation time α and the glass transition temperature T_g obtained by Cooper and Stevens [6] as a function of the fractal dimension D.

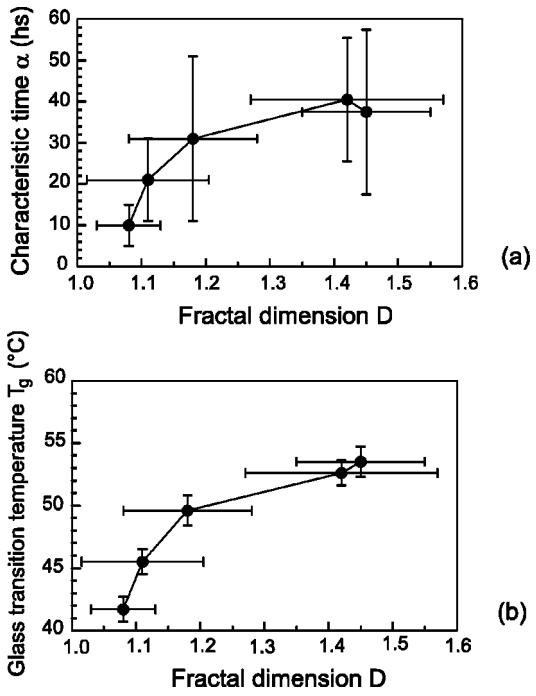

FIG. 1. (a) Characteristic propagation times α as a function of fractal dimension D. (b) Glass transition temperature T_g as a function of fractal dimension D. Data from Ref. [6].

III. FRACTAL ANALYSIS OF DIELECTRIC BREAKDOWN PATTERNS

A. The model

A two-dimensional square lattice, in which two opposite sides represent the two electrodes, is considered [10]. We adopted this model because boundary conditions imposed by a planar geometry sample allow the study of dielectric breakdown without any loss of generality. The interelectrode gap is typically 1–2 mm. Microscopic examination of electrical tree growth shows that branch extension occurs in increments typically of 5–10 μm. This implies that a gap of 100 lattice units will represent the experimental situation adequately. In this work, 100×100 lattices and, in some cases, 200×200 lattices were employed.

Breakdown starts at a point of a high local field and this enhancement is usually attributed to electrically conducting inclusions. The conducting inclusions are represented by electrode pins. Discharge simulations begin at one of the electrode pins where a short filament represents the tip of one pin. The rules assumed for the growth of the discharge pattern (the electrical tree) are the following.

(1) The electrical tree grows stepwise. The discharge structure has zero internal resistance, i.e., at each point of the structure the electric potential ϕ is $\phi=0$, whereas at the counterelectrode it is $\phi=1$.

The discrete form of the Laplace equation

$$\phi_{i,k}=\frac{1}{4}(\phi_{i-1,k}+\phi_{i+1,k}+\phi_{i,k-1}+\phi_{i,k+1}) \tag{4}$$

is solved with the above boundary conditions.

(2) The probability that a bond will form between a point, which is already part of the electrical tree, and the new adjacent point is a function of the local field between the two points (i.e., the potential difference between the two points). A power-law dependence with exponent η is assumed and the probability associated with points i,k at the structure and points i',k' adjacent to but outside it, is given by

$$P(i,k\rightarrow i',k')=\frac{(\phi'_{i',k'})^{\eta}}{\sum_{(j',l')\in\Gamma}(\phi_{j',l'})^{\eta}}. \tag{5}$$

The sum in the denominator refers to all of the possible growth sites (j',l') adjacent to the electrical tree, where Γ is

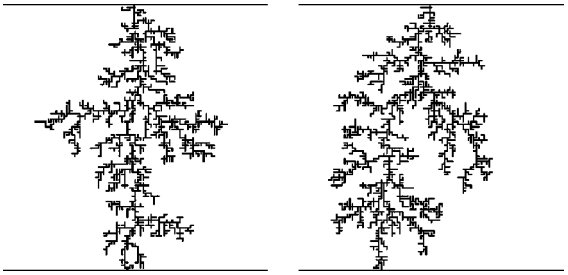

FIG. 2. Two fractal structures simulated with the dielectric breakdown model (DBM), presented in Sec. III A with $\eta=1$.

the set of possible candidates to be incorporated into the electrical tree.

(3) A new bond is chosen randomly and added to the electrical tree.

(4) With the new electrical tree and boundary conditions the process starts again.

The trees generated by this model have a fractal structure that has been broadly dealt with in the literature [10–13]. According to the DBM growing rules, for each value of η we obtain a family of trees that have statistically the same fractal dimension D. Two trees obtained with $\eta=1$ are shown in Fig. 2.

The probability of dielectric failure is usually determined as a function of the propagation time, measured as a function of the number of bonds incorporated into the tree t_s, where the incorporation of a new bond represents a unit of time.

The cumulative probability of failure in the trees generated by computer simulations of the model satisfies a two-parameter Weibull distribution such as those observed in experimental studies [see Eq. (2)]. The histogram showed in Fig. 3(a) represents the cumulative probability of failure obtained from a 500-tree family with $\eta=1$, simulated on a 100×100 lattice. The histogram is adjusted by a Weibull distribution [continuous curve, see Eq. (2)].

Figures 4(a) and 4(b) shows the Weibull distribution parameters α and β, respectively, as a function of the model parameter η. The mean values μ and μ/σ, calculated from the normal distribution with mean μ and standard deviation σ are also shown in these figures. The behavior of these values is similar to α and β, respectively [see Eq. (2)].

In order to find an appropriate comparison with the experimental data, we redefined the time unit mentioned above. The new unit of time needed to reproduce experimental data indicates the existence of a relaxation time related to this Γ set. In other words, the old unit of time based on the time elapsed from the incorporation of a new bond to the next one, does not reproduce experimental data. However, a unit of time based on the time elapsed to incorporate a number of bonds equal to the number of elements of the Γ set, indeed reproduces Cooper's results. This unit of time implies $t=k$ ($k=1,2,3,\ldots$), see Fig. 5, an average value in material relaxation time, which is the time needed to incorporate a number of bonds equal to the whole members of the Γ set. The electrical potential is calculated when each bond is incorporated into the tree. In Sec. IV we compare the propagation time calculated by the two methods (called step time t_s, and

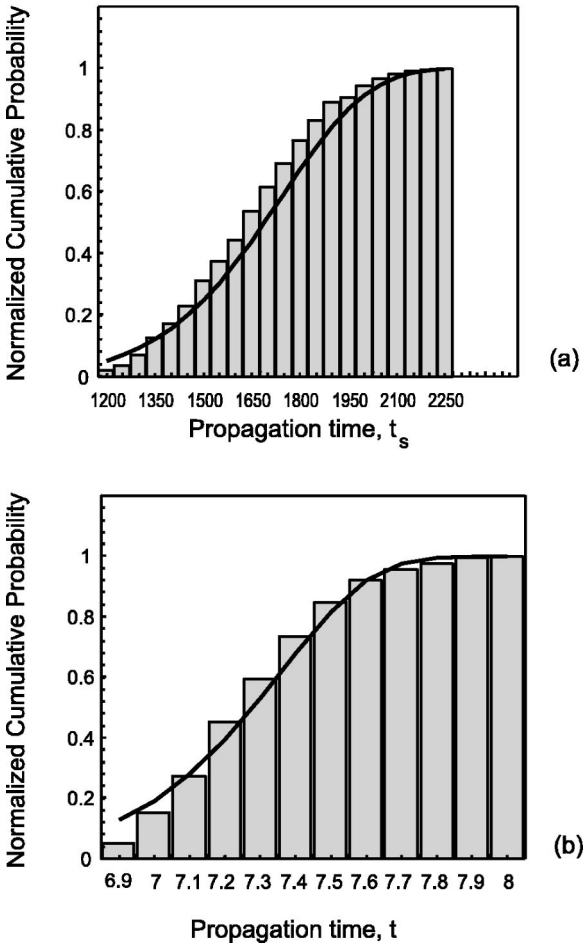

FIG. 3. Normalized cumulative probability of propagation times. (a) Time is measured as the number of bonds incorporated into the tree (t_s). (b) Time is measured as the number of periods required for the tree to incorporate a number of bonds equal to the number of candidates incorporated in the previous period t.

period time t, respectively) and those experimentally obtained.

The new time unit also gives a Weibull distribution with parameters (α,β) different from the previous ones (α_s,β_s). Figure 3(b) shows the cumulative probability of failure for a 500-tree family with $\eta=1$ simulated on a 200×200 lattice.

B. The fractal dimension

There are different methods to estimate the fractal dimension of the electrical tree [Eq. (1)] recently classified by Kudo [8] in the following methods.

(a) The box counting method.

(b) Fractal measure relations.

(c) The correlation function.

(d) The distribution function.

The radial method employed by Cooper and Stevens [6], Eq. (3), is shown in Fig. 6 in a $\log L(R)$ vs $\log R$ plot, applied to two electrical trees simulated by the dielectric breakdown model, with $\eta=1$ and $\eta=5$. When the increments in R are very small, the $\log L(R)$ vs $\log R$ curves have S shapes because of incomplete sampling. Following Cooper and

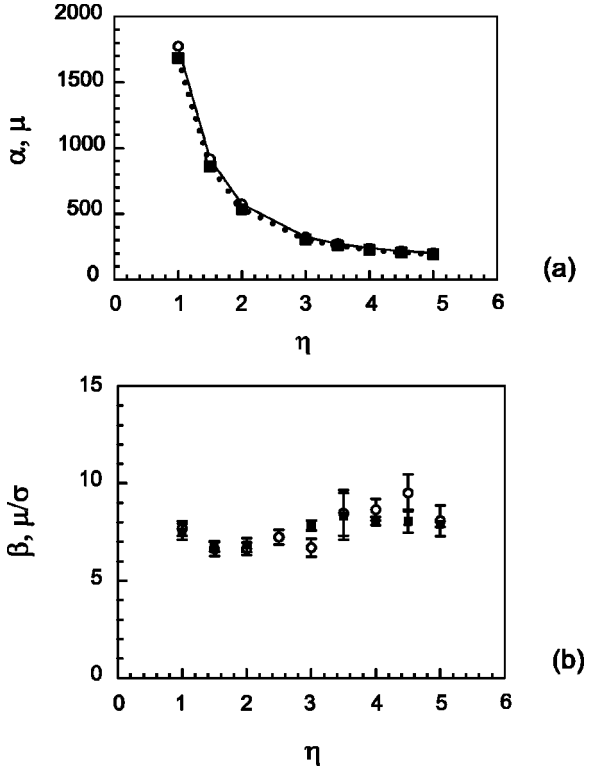

FIG. 4. (a) Dependence of characteristic propagation time α, ○ [Weibull distribution, see Eq. (2) and the mean time μ, ■ (normal distribution) on the DBM parameter η. (b) Dependence of shape parameter β, ○ [Weibull distribution, see Eq. (2)] and the relative mean value μ/σ, ■ (normal distribution) on the DBM parameter η.

Stevens [6], the ends of these curves were not taken into account. Within the 95% of the confidence interval, the linear relationship is satisfied.

The correlation function method gives distributions with smaller standard deviations of the fractal dimension D and, for this reason, it is one of the most frequently employed in the study of simulated trees [13]. However, its use in real situations is limited by the complexity of the method. On the other hand, the radial method, see Eq. (3), has the advantage of simplicity. We compared both methods in 500-tree families generated on a 100×100 lattice.

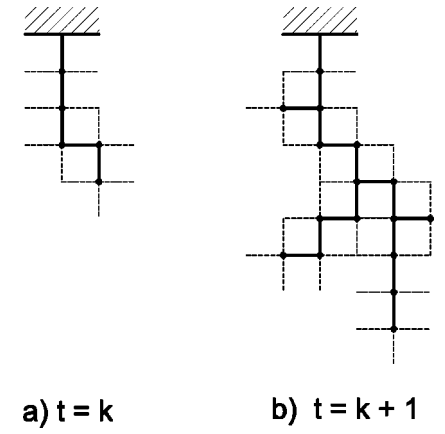

FIG. 5. The new unit of time is the period required for the tree to incorporate a number of bonds equal to the number of candidates to be incorporated (dashed lines) at (a) $t=k$; (b) $t=k+1$, $k=1,2,3,\ldots$. From (a) to (b) t increases by one unit, whereas t_s increases by 11 units.

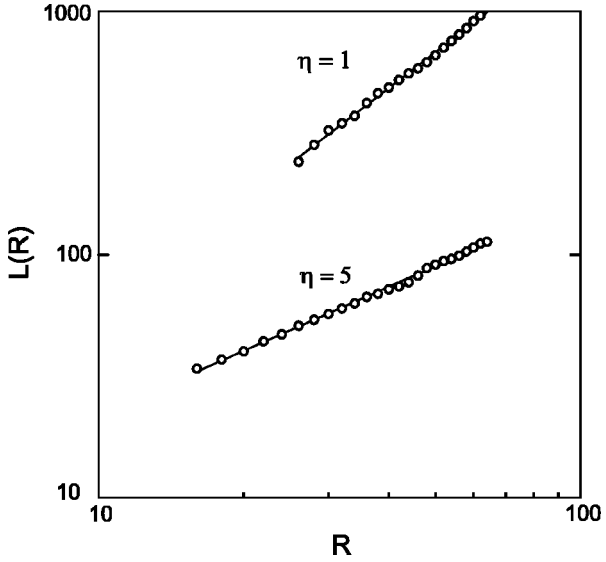

FIG. 6. Range of validity of the radial method, see Eq. (3), to evaluate fractal dimensions D on 100×100 lattices for a couple of simulated electrical trees.

Figure 7 shows a comparison between the radial and the correlation method as a function of the η value. Both methods give similar fractal-dimension mean values, but the radial method has very large dispersion. It is necessary then to take a large number of samples in order to have a better estimation of the fractal dimension D.

IV. RESULTS

Figures 8(a) and 8(b) show principal branches of electrical trees experimentally obtained by Cooper and Stevens [6],

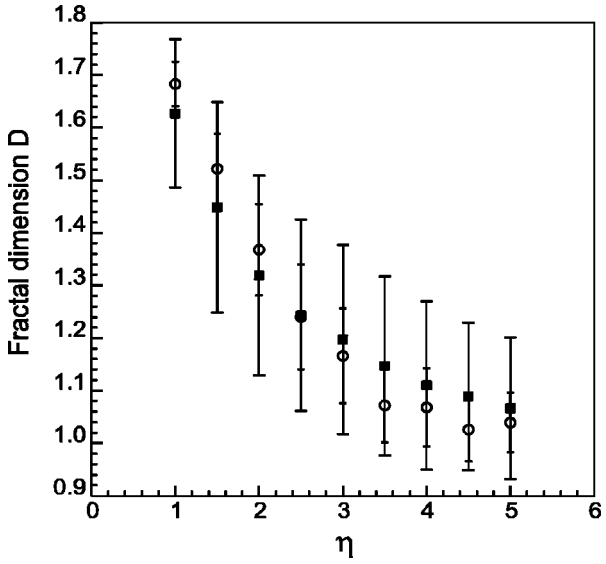

FIG. 7. Comparison between the fractal dimension D evaluated with the function correlation method, ○, and the radial method, ■.

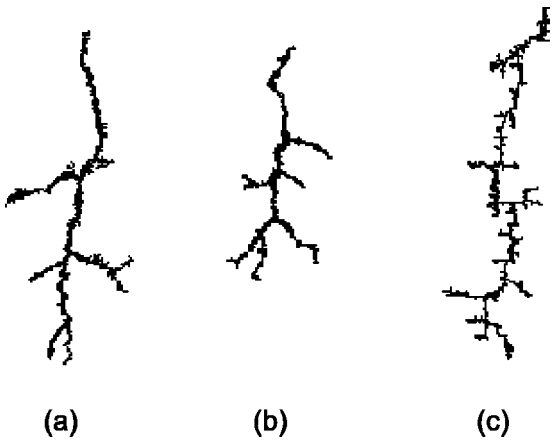

FIG. 8. Comparison between an experimental electrical tree (on the left, from Ref. [6]) and a simulated electrical tree (on the right) from the DBM model (η=3) developed in Sec. III A . Both structures have the same fractal dimension $D=1.18$ evaluated with the radial method.

whereas Fig. 8(c) shows a fractal structure generated by the model developed in Sec. III. A visual comparison between experimental and simulated electrical trees enabled us to appreciate their remarkable similarity.

Now let us compare the dependence of the characteristic propagation time α on the fractal dimension of the electrical trees generated by the model developed in Sec. III, and the experimental dependence obtained by Cooper and Stevens shown in Fig. 1.

Electrical trees were simulated on a 200×200 lattice (size). Due to the fact that characteristic lengths of electrical channels are roughly 5 to 10 μm, the above-mentioned lattice size is adequate to simulate the 2-mm-thick samples used by Cooper and Stevens [6].

Propagation time was evaluated by using two different approaches (see Sec. III A), called t_s step and t period, respectively. If the model was adequate to describe the experimental information both propagation characteristic times (theoretical and experimental) would differ by a constant, $\alpha_{theor}/\alpha_{expt}=cte$.

Figure 9(a) shows the dependence of α, α_s, and α_{expt} on the fractal dimension D. Both, α and α_s were multiplied by a constant factor for comparison purposes. Ratios α/α_{expt}, α_s/α_{expt} as a function of the fractal dimension D were evaluated and plotted in Fig. 9(b), where α_{expt} stands for the experimental characteristic propagation time obtained by Cooper and Stevens [6]. We learn from that figure that the ratio α/α_{expt} approaches a constant value when the fractal dimension (or the cross-linking density, see Sec. II) is increased, whereas the ratio α_s/α_{expt} grows without any limits as the fractal dimension increases.

Characteristic propagation time α seems to describe satisfactorily the experimental behavior within the fractal dimension interval $1.1<D<1.5$. Therefore, we can conclude that the unit of time proposed in this paper to evaluate propagation time t clearly reproduces the dependence of characteristic propagation time α on the fractal dimension observed experimentally, see Fig. 9(a). Hence, a propagation time

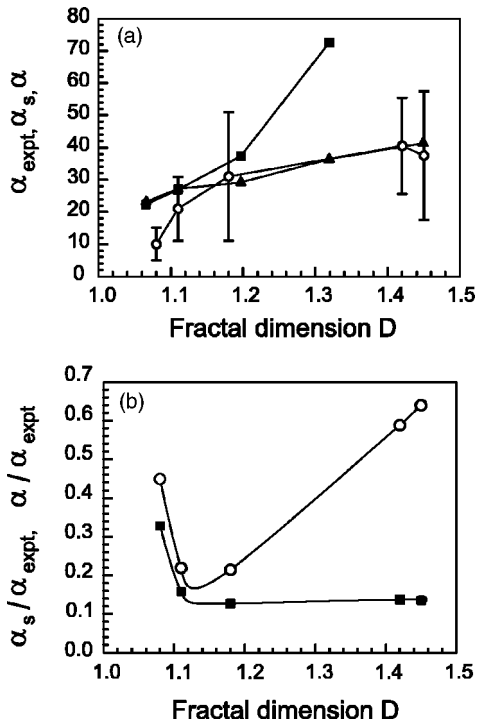

FIG. 9. (a): Dependence of characteristic propagation times α_{expt}, ○; α_s, ■; and α, ▲ on fractal dimension D. (b) Dependence of characteristic propagation times α_s and α relative to the experimental value α_{expt} on fractal dimension D. α_s/α_{expt} (○); α/α_{expt} (■).

based on the number of bonds incorporated into the electrical tree t_s, as was proposed up to now by different authors [14], fails to reproduce the experimental facts, see Fig. 9.

The glass transition temperature T_g of cross-linked polymers is proportional to the cross-linked density (n),

$$T_g(n)=T_g(0)+Zn, \tag{6}$$

where Z is a material-dependent constant. Equation (5) was proposed in the 1950s by Ueberreiter and Kaming [15]. Glans and Turner [16] have tested the validity of such assumption on samples of cross-linked polystyrene.

We have recently proposed a thermodynamical interpretation for the fractal structures obtained by the DBM model on a linear two-dimensional geometry [10,17,18], following the pioneer paper of Elezgaray *et al.* on an open-planar geometry [19]. According to the growth rule of the model, a collection of electrical trees C_M, where M is the number of branches is obtained and the branching structures C_M give the state of damage in terms of the number of branches. In this context, a probability $p(C_M,\eta)$ for each C_M can be assigned to each value of η. Based on a number of numerical simulations, the following expression can be obtained [18]

$$\ln[p(C_M,\eta)]=S(C_M)+A(C_M)\omega(\eta)+\gamma(\eta,M), \tag{7}$$

where $\omega(\eta)$ and $\gamma(\eta,M)$ are two universal functions, $A(C_M)$ plays the role of the energy of the electrical tree, and $S(C_M)$ is a history degeneracy factor. The above expression for $p(C_M,\eta)$ has the form of a Boltzmann weight if $\omega(\eta)$ is

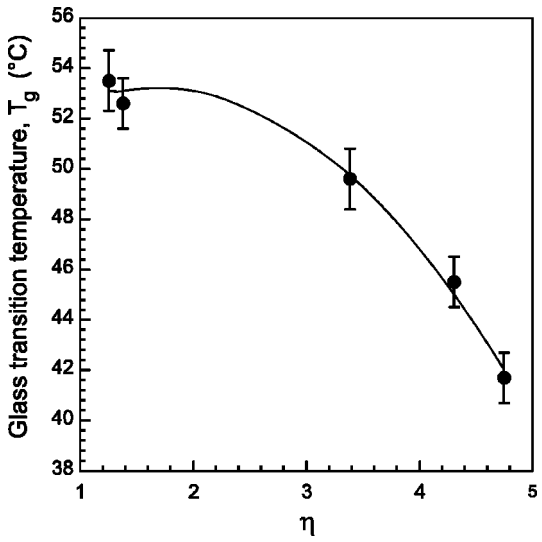

FIG. 10. Dependence of glass transition temperature T_g on the DBM parameter η, see Sec. IV.

identified as a ratio with a ''reference energy '' and $\gamma(\eta,M)$ as the free energy.

Cooper and Stevens [6] have determined that the cross-linking reaction is exothermic with a total reaction enthalpy related to the concentration of styrene monomer and unsaturated polyester sites. As the reaction proceeds the cross-link density increases beyond the gel point. This was monitored by measuring the residual enthalpy of the reaction and corresponding increases in the glass transition temperature.

In the present situation, the universal function $\omega(\eta)$ could be identified with the inverse of the cross-linked density energy (the above-mentioned ''reference energy''), or, see Eq. (6), with the glass transition temperature. Figure 10 shows the relationship between T_g and η that was deduced by observing their dependence on the fractal dimension D, see Figs. 1(b) and 7. An inverse relationship between glass transition temperature and η is observed.

V. CONCLUSIONS

In this paper we studied the fractal nature of electrical trees obtained by a dielectric breakdown of cross-linked materials by numerical simulations based on the DBM model in a two-dimensional geometry. We extended the one parameter model (DBM) by redefining the unit of time. The new unit of time is based on the time elapsed to incorporate a number of bonds equal to the number of elements of the Γ set, see Fig. 5 and Eq. (5).

With the model developed in Sec. III we found the following:

(i) The model produces fractal structures that are similar to those experimentally observed (see Fig. 8 and [6]), characterized by a fractal dimension D.

(ii) An adequate definition of the unit of time allowed us to reproduce the dependence of the characteristic time α, experimentally found, on the fractal dimension D, which is valid in the interval $1.2<D<1.5$, see Fig. 9.

(iii) Propagation distribution times of the simulated electrical trees obey a Weibull statistics [see Fig. 3(a)] characterized by two parameters; a shape factor β and a characteristic propagation time α, as experimentally found [6].

(iv) From Figs. 1(a) and 1(b) we learn that the dependence of the propagation characteristic time α on the fractal dimension D should be related to a basic material property such as the cross-linking density. Based on variations in residual enthalpy with the postcure temperature, Cooper and Stevens have come to the same conclusions, see Fig. 2 in Ref. [6] . Anyhow, more experimental information would be necessary to test this hypothesis. In particular, chemically different cross-linked polymers should be investigated.

(v) Finally, a relationship between the parameter η of the DBM model and the glass transition temperature T_g was found, which is in agreement with derivations based on a thermodynamical model of electrical trees [10,18].

The present model involves a notion of universality that deserves to be explored; work is currently in progress at La Plata University to unravel it.

ACKNOWLEDGMENTS

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