

A New Integrated Approach for Fast Intrinsic Dielectric Breakdown

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Abstract: A non-equilibrium phenomenological description for a complete dielectric breakdown sequence in perfect solid electric insulators, from the dynamics of instability to the growth of branching patterns, is implemented. It represents an original treatment which allows overcoming the current lack of such a complete physical description, involving thermodynamics, and in particular equations on maximum dissipation energy for dielectric breakdown are developed. From the assumption that there is a time delay in the energy rates and propagation speeds between the system input and output, a principle of maximum energy dissipation is postulated and corresponding dynamic evolution equations were obtained. It is shown that the delay in those speeds gives rise to the appearance of iterated equations which in turn leads to the logistic maps describing the dynamic evolution of the system. The results can describe the instability process along with dissipation patterns formation. The vast theoretical and experimental analogies between mechanical fracture and dielectric rupture, lead us to foresee the potential applicability of this model in the prediction of dielectric rupture patterns. This work opens then the possibility to predict the consequent branching patterns by using a maximum energy dissipation principle, contributing in a significant way for energy efficiency engineering.

Keywords: Dielectric breakdown, Breakdown branching, Maximum energy dissipation, Energy efficiency

I. INTRODUCTION

The search for an integrated physical interpretation of failure mechanisms in solid dielectrics under electric stress (dielectric breakdown) can represent a decisive step towards the ability to predict rupture patterns, which is of a crucial importance in science and engineering [1]. It involves phenomena that represent a response to an extreme energy input event originating from instability behaviour and giving rise to breakdown structures and patterns formation. Such structures and patterns typically exhibit a branched geometry that can be characterized as a fractal, between a finite range of scales $\epsilon_{\min} \leq \epsilon \leq \epsilon_{\max}$. Despite a great deal of research into solid dielectrics conducted for nearly a century now has led to a basic understanding of the breakdown processes in solids, there are a number of theories, and none explain all the complete phenomena. Furthermore, there is not a complete physics framework able to describe all this phenomenology, from the unstable dynamics until the growth of a branched structure or the fractal pattern formation [2]. The motivation of this work is based on such absence of a non-equilibrium description for the complete phenomenological sequence, from the dynamics of instability to the growth of the branched structure or formation of fractal patterns.

The phenomenon of energy transfer between micro and macro level is the subject of many discussion in physics and engineering [3] and dielectric breakdown is a case that deserves a special approach although it has been the subject of several discussions. In the aim of irreversible thermodynamics, situations of instability with nonlinear processes happening far from equilibrium, cannot be described with the linear theory of flows and quadratic potentials (Onsager-Gibbs), because this is limited to the near-equilibrium conditions in the so-called stable or quasi-static growth processes. In this work, a system of equations was developed using velocities associated with the flows of energy injection and branched patterns desynchronized by a time delay between them. Therefore, our proposal is an extension of the theory presented by Onsager [4-6] providing a continuous phenomenological view of the problem that goes from the non-equilibrium thermodynamic relations to the construction of logistic maps. Furthermore, there is a need to formulate a

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principle of instability enabling to describe the consumption of energy and the formation of fractal patterns in these situations. It should also evidence the functional dependence of the growth rate of these structures, so that the "most probable path" and the time can be inferred from the theory, and then overcoming the gap that exists in the previous thermodynamic near-equilibrium description.

Dielectric breakdown resembles the mechanical fracture of solids from various aspects and the analogy between electric field and mechanical stress has been noticed [7]. Dielectric rupture often generates a tubular conductive channel just like a crack; electric field is concentrated near the tip of the conductive channel just like the mechanical stress concentration at a crack tip; and only materials at the vicinity of a conductive channel or a crack is damaged while the major part of the bulk material remains intact. By assuming a maximum energy dissipation due to the maximization of entropy at time intervals increasingly smaller (in the case of a mechanical fracture system far from equilibrium), it is possible to obtain the dynamic evolution equations [8]. In addition, any solicitation imposed to the system above the rate at which this transfer is possible, will produce instability which tends to bring the system to a maximum dissipation and utter chaos. The class of systems for which this principle is obeyed concerns those where maximum dissipation corresponds to the fastest energy decrease. In a previous work we have proposed a modified maximum energy dissipation principle for explaining crack instabilities studies in fracture dynamics [9]. It assumes that there is a time delay in the energy rates and propagation speeds between the system input and output, and corresponding dynamic evolution equations were obtained. Therefore, there is a full agreement with the intuitive expectation that the system chooses the path on the energy landscape along which energy decays in the fastest way.

We intend here to reconsider the problem in dielectric rupture, this time paying attention to various microlevel phenomena and their role in the energy dissipation and develop a dynamic model starting from instability and leading to formation of the tubular conductive structures/patterns. Using a dynamic fractal description of the phenomena, a coupling macromicro model of a dielectric rupture process was presented. All equations written for the micro level can also be written to the macro level, and so the coupling between the dissipated energy in the macro and micro levels is established by the scale dynamic relationship of the formed pattern (assuming in principle as a fractal), since the instability factor defined by Sander [10], relates the speeds at the micro and macro scales. Furthermore, it is shown that the above proposed delay of velocities in the description of the instability is adequate to describe the process of energy dissipation, leading naturally to the mathematical description of fractal patterns formation, and explaining the dielectric rupture instability in high speed regime.

II. THEORETICAL FRAMEWORK

Intrinsic Dielectric Breakdown Mechanism

The maximum electric field that a pure insulating solid material can withstand (electric breakdown strength) under ideal conditions without experiencing failure of its insulating properties is at a constant temperature an intrinsic property of the material and independent of its geometry or that of the electrodes with which the field is applied. The unavoidable presence of a few free electrons will be accelerated by the external electric field and then collide with the atoms/molecules in the lattice. If the field is high enough, a large number of electrons can detach themselves from their chemical bonds that form the material and move therefore, across the forbidden gap from the valence into the conduction band (CB). The localized charge density reaches a critical value, originating the disruption of chemical bonds. The free electrons under the influence of the electric field can collide with atoms/molecules in the lattice giving rise to a very fast avalanche process causing further breakdown of chemical bonds [11]. Ultimately, through continuation of these processes a gaseous channel can grow in the dielectric and eventually bridges the electrodes. The initial conduction electrons for the avalanche are either present in the CB or are injected from the metal into the CB as a result of field-assisted thermal emission from the Fermi energy in the metal to the CB in the dielectric. Typically, it has been experimentally verified that electrical conductivity increases by a factor of 10 in a time of about 10 nanoseconds [12]. Such time is required because some current flows in the dielectric when the electric field is

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applied. The progress of the conductive gaseous channel needs to overcome the mechanical resistance imposed by the cavity walls, and its growth is thus dependent on the electron density in the gas and on the wall area of the cavity. Also there is some total energy released, partially as kinetic energy of the individual particles and partially as electron excitation, when a molecule of the solid is converted into the gas phase. Since a heat flow from the gas takes place while the cavity is growing, the inner temperature and pressure will vary in time; this in turn will cause a change in the density of the neutral and ionic species as well as of the kinetic coefficients of the physical processes involved (recombination, electron attachment/detachment and ionization). Therefore, the mechanical resistance against the channel growth are dependent on the equations that determine the physical state of the cavity system (kinetic rate equations, electron density and heat balance).

A typical experimental dielectric breakdown branch-tree structure in polymer (Fig.1) is thus the result of hollow gas-filled tubules and chemical degradation. These local electron avalanches damage the material over a limited distance from their initiation point. Once a critical level is reached along a given avalanche path, the local polymer chains fragment under the combined action of electromechanical stresses and the thermal energy deposited by the culminating avalanche. This results in an extension to the existing structure in the form of a chain in that region. The field strength at which breakdown occurs typically increases with the time the voltage is applied. The disruptive electric current causes heat; some of which is dissipated and the other raises locally the temperature of the material.

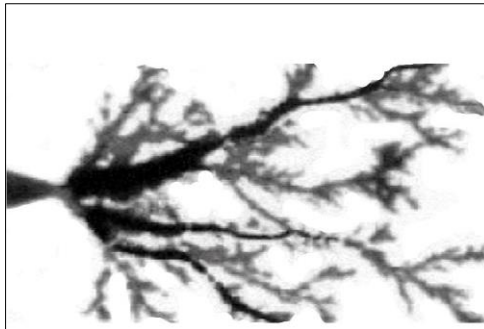


Fig.1. Typical experimental dielectric breakdown branch tree.

Fracture and Dielectric Rupture Analogies

A Griffith-type energy criterion has been proposed for the propagation of a damaged conductive channel [13-16]. A J integral has been defined [13] (when the dielectric breakdown process is idealized as the growth of a conductive tubular channel embedded in a dielectric material- Fig. 2) given by:

$$J = \int_s (Wn_1 - n_i \sigma_{ij} u_{j,i} + n_j D_j E_1) dS \quad (1)$$

W , σ_{ij} , u_i , D_i and E_i are the electric enthalpy density, the stress, the displacement, the electric displacement and the electric field, respectively; n_i is the unit vector of the outward normal to the surface enclosing the channel head and S is the surface embracing the conductive channel. The subscript comma (,) denotes a partial derivative with respect to the Cartesian coordinates.

Since equilibrium equations for mechanical stresses and electric displacements are given respectively by $\sigma_{ij,i}=0$, $D_{i,j}=0$ and making use of the divergence theorem, it is possible to show that the path-independent surface integral around the tip of a thin conductive channel (in analogy with the J -integral of fracture) is

$$J = \int_{S_h} (Wn_1 + n_j D_j E_1) dS \quad (2)$$

Where, S_h is the surface of the channel head (Fig. 2b). The authors [13] also showed that the energy release per unit length of the conductive channel due to its growth, G is exactly equal to J , and so shown to be equal to the breakdown

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energy at the critical point [14]. According to thermodynamics, the energy balance for an incremental increase in the channel length can be expressed as Γ_{db} , where b is the channel length and Γ the work to create a unit length of the channel (known by breakdown energy, which is a material property analogous to the mechanical fracture energy, related to the first ionization energy of the atoms or molecules making up the material, and also to its density). The theory assumes [14,15] that when the electric energy release rate (i.e. the decrease in electrostatic energy associated with the unit length extension of a conductive channel) exceeds the energy consumption in creating unit length of a conductive channel, this channel will propagate. Thus, the energetic criterion is that a conductive channel will grow only if the electrostatic energy released per unit length of the channel is greater than that dissipated through damage. In short words, the channel cannot grow when $G < \Gamma$.

As the damage evolution and charge redistribution are much slower than the speed of light, the energy emission in the form of electromagnetic waves is neglected. We assume the system to be in electrostatic equilibrium at all times of consideration. The energy dissipation through electric conduction, on the other hand, is considered a part of the breakdown energy.

At this stage, let us resume several analogies between intrinsic dielectric breakdown and fracture mechanics [7,14]:

i) Mechanical stress σ is the analog of the electric displacement D , and strain ϵ is the analog of the electric field E ; $D(t) = \epsilon_0 E(t) + P(t)$, where P is the polarization of the dielectric,

ii) The critical electric field E_0^c is a function of the channel length L_0 , dielectric constant ϵ and breakdown surface energy Γ ; it is a point of unstable equilibrium, given by $\left[\frac{4\Gamma}{\pi\epsilon L_0} \right]^{1/2}$, and so for electric fields larger than it, dielectric

breakdown will spontaneously occur,

iii) Also exists an electric field intensity factor, $K = E_0 (\pi L_0)^{1/2}$ and an electrostatic energy release rate, $G = \epsilon(K)^2$, where $G dL_0$ is the amount of electrostatic energy release when the channel extends by dL_0 (remind that in fracture mechanics $G = K^2/Y$, where Y is the Young modulus). The critical value of G is 4Γ and so $K_c = E_0^c (\pi L_0)^{1/2}$; the contour line integral J can be demonstrated to be equal to $(-K^2 \epsilon / 2)$ [7,14],

iv) Lattice models of dielectric breakdown allow comparison with other predictions [17] when transposing the Griffith-Irwin fracture mechanics framework, Inspired by the phase-field model of brittle fracture [18,19] it has also been developed a phase-field model for the damage evolution in solid dielectrics during breakdown [20]. Such model represents the breakdown-associated damage by a continuous field and the breakdown energy by the surface energy between the damaged and intact phases. It can be readily implemented into numerical codes to simulate the breakdown processes and calculate the effective breakdown strength, as well as energy dissipation. The phase-field model [20] uses empirical parameters to reflect the degree of damage (insulation quality) on the electric permittivity [21] without a real physical meaning, although prone to be used in numerical calculations and simulations. To have coexisting damaged and undamaged phases, the model introduces an energy function which varies with the critical electrostatic energy needed to initiate the damage (Γ/L_0^2). This energy function can be imagined as a buffer that artificially stores the energy consumption during the formation of conductive channel, i.e., the material damage process. Using variational calculus to minimize the total potential energy of the system as a function of the electric potential and damage parameter, one can obtain the Euler equations for the equilibrium state. The J integral can also be written as a function of the phase field, giving rise just to a function of the radial coordinate and proportional to Γ/L_0 [20]. The degradation decays exponentially around the conductivity channel and is highly localized in a region of radius $\approx L_0$, so then J is obtain as a function of Γ , L_0 and the phase field: $J \approx 0.982\Gamma$. The J integral around a conductive channel in a linear dielectric material has then a logarithmic dependence on the tip radius of curvature. Due to the reversible assumption of the damage field, the equilibrium corresponds to the critical state for the growth of the conductive channel, when the driving force J equals the resistance (Γ is thus approximately the breakdown energy of the material). In the case of propagation of a conductive channel when the energetic driving force is

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supercritical, a new empirical parameter is introduced to address the speed of damage in the material and compute the temporal evolution of the electric field and phase field (assuming a linear kinetics, using the governing partial differential equations) [20].

Once initiated the dielectric breakdown in a parallel circular plates capacitor with a thin conductor parallel to the electric field (placed in the axis and connected to one of the electrodes), the conductive channel extends the pre-existing field concentration, leading to even stronger field concentration. The electric field needed to further extend that channel thus decreases with its extension. The propagation of the conductive channel is unstable and can be illustrated in COMSOL simulations [20]. In addition, these simulations show that the spontaneously formed damage zone takes a 1D tubular form along the direction of the electric field, and when the size of the conductive channel is still sub-critical it will not grow; the catastrophe breakdown event takes place when the channel beyond the critical length is formed via growth and then branching occurs [20,21]. Furthermore, this numerical method when applied to simulations in materials with defects allows recreating the inverse power law in a plot of dielectric strength versus thickness of the sample, typical of several experimental results [22]. In short words, the phase-field model employs an energy criterion in analogy with the one of the crack propagation in linear fracture mechanics: the electrostatic energy released equals or exceeds the energy consumption in growing the conductive channel by unit length; this criterion is equivalent to that given by the path-independent J integral.

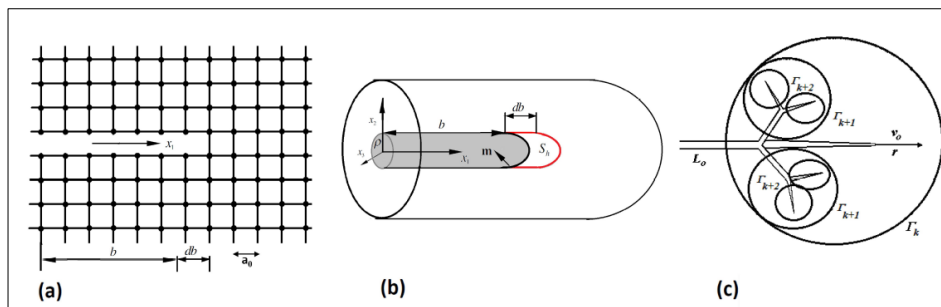


Fig. 2. Timeline for the growth of dielectric breakdown-tree structures. (a) Breakdown bonds forming a conductive channel in a crystalline 2D atomic lattice of periodicity a_0 ; (b) Growth of a tubular conductive channel; (c) Envelope surfaces over a branched structure, for calculating the integral of excess M in a model of non-interacting mutual branches.

Dielectric Breakdown Propagation Process and Instability

The internal energy of a system can be increased indefinitely but as far as new energy levels are reached, dissipation can happen in different ways, because different energy barriers encountered at each hierarchical level of energy accumulation (energy barriers at multiple scales) can be overcome as more energy is provided to the system. In this case the system must be considered at least closed so that the externally injected energy generates alternative forms of dissipation as the barrier of each phenomenon can be overcome. The thermodynamics of the system says that it should go to the minimum lower energy state and the kinetics of the phenomenon says that it must dissipate the more "efficient" and as quickly as possible. The result is therefore a state in which such alternate forms of dissipation as heat, sound, etc. are created including generating branched patterns in the form of dielectric breakdown channels. In dielectric rupture dynamics, the system is able to accumulate a certain amount of energy till an upper limit of saturation, or rupture critical value. Above it, the triggering of the catastrophic electrical disruption occurs. In practical terms this means that, the dielectric rupture strength, as well as other properties, will specify the volumetric capacity for the accumulation of electrical energy in the material. Such systems are prone to generate instabilities when the external request reaches its maximum load limit.

The Energy Fluxes in Dielectric Breakdown

In fracture mechanics simulations of fast crack propagation correlated with crack tip stress in 2D hexagonal atomic lattices [23] show that branching and oscillation phenomena inevitably occur when the crack speed reaches a critical

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value, which is closely related to the stress field near the crack tip. Results reveal that the crack propagation strongly depends on the stress and displacement fields near the crack tip. It is likely that a nonlinear zone exists at the tip of dynamic propagating crack playing a controlling role on the onset of oscillation and instability during rapid fracture. It is known from experiments on fracture mechanics that the classical solution of elastodynamic non-stationary energy released rate is unable to explain the instability and the branching phenomenon; in addition, those experiments allowed the determination of the velocity at which that instability begins as well as a delay of the order of the mechanical stress relaxation time [9]. Considering the analogies between the fast crack growth and the dielectric breakdown, thus in the case of dielectric rupture, where the nonstationary energetic balance is given by

$$\dot{\phi} \equiv \frac{d\phi}{dt} = \frac{d\Phi}{dt} - \left(\frac{dU}{dt} + \frac{dK}{dt} \right) \quad (3)$$

with the stationary classical solution

$$\dot{\phi}(L_0, v_0, t) = G_{OD}(L_0, v_0, t) v_0(L_0(t)) \quad (4)$$

is not also expected to explain the instability and the branching phenomenon either. Notice that now, $d\phi/dt$ stands for the balance in the injected free energy flux giving rise to the conductive channel with a growth velocity v_0 , and G_{OD} is the elastodynamic energy release rate; $d\Phi/dt$ is the injected energy flux itself, dU/dt the energy flux of electric strength and dK/dt the kinetic energy flux (sum of energy released in the formation of the conductive channel and in its growing); the subscript "0" is used to represent all the quantities that are considered in the projected plane on the direction of the dielectric breakdown growth.

When the flux of electric energy injected in the system is larger than the one it can support, there is a delay between the input and output energy fluxes. The instability gives rise to a shift of the system from its linear pathway and so the conductive channel can grow by branching as a more efficient dissipation way, increasing the entropy and relieving the concentration of electric strengths. Actually, like it happens in fracture mechanics, the electric "treeing" phenomenon is very common in the dielectric rupture experimental observations (Fig. 1). The process of electron avalanche breakdown in electron-atom/molecule collisions gives rise to a conductive channel during the first few microseconds after electrical breakdown, as it was demonstrated in experimental measurements [22], where an acoustic shock wave expands at a constant velocity while the expanding radius of the channel is proportional to the 4th root of the energy and the square root of time. These dependencies are predicted by modelling the breakdown channel as an expanding adiabatic ideal gas with an instantaneous input of energy [22]. Such expansion takes place in the form of adiabatic waves with a limited velocity given by the Rayleigh wave's velocity, R_c (according to classical theory of fracture mechanics). Actually, the so-called "rate-determining process" in kinetics of sequential processes is always the slowest one; therefore, comparing the speeds of the physical and chemical processes involved, the slowest corresponds to the fracture propagation. Since this process is limited by the speed of Rayleigh waves, then dielectric rupture (as a sequential process) will also be limited by that speed. The formulae of conducting microcrack growth and lifetime prediction were applied by Ding et al. [24] to the experimental data of SiO_2 films. It is the nature of cohesive bonding between the constituent atoms in the material which ultimately determines the resistance to the passage of a microcrack (Fig. 2) [23].

Using the Griffith-like energy balance, the breakdown energy required to create a unit area of conducting microcrack is given by:

$$\Gamma(L_0, v_0, t) = \frac{1}{2} \frac{dU_s(L_0, v_0, t)}{dL_0} \quad (5)$$

Where U_s is the surface energy of conducting microcrack patterns, and the electrostatic energy release rate is:

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$$G = \frac{\pi \epsilon E_0^2}{L_0} \quad (6)$$

GdL_0 is the amount of electrostatic energy released when the microcrack extends by dL_0 (it can be viewed as the conducting microcrack extension force). In ideal conditions, the total energy difference between a cracked (conducting) and uncracked (insulating) material becomes

$$dU = -GdL_0 + 2\Gamma dL_0 \quad (7)$$

The critical value of dG is thus:

$$G_c = 2\Gamma \quad (8)$$

For $G > G_c$, the conducting microcrack will grow spontaneously, as well as successive bond breakdown. It is then interatomic potentials that ultimately determine the breakdown energy barrier against conducting microcrack extension, and this barrier reflects the nonlinearity of the bond rupture. Let us then consider a conducting microcrack growing by the sequential breakdown of lattice bonds (Fig. 2) [24]. Using a kink model of kinetic conducting microcrack growth [24] it is possible to calculate its growth rate, making use of the temperature T and Boltzmann constant k_B :

$$v_c = \frac{dL_0}{dt} = \nu_0 a_0 \exp\left[\frac{\alpha \pi \epsilon E_0^2 L_0 - U_0^*}{k_B T}\right] \quad (9)$$

Where, the basic lattice vibration frequency ν_0 is related with Planck constant $h(\nu_0 = k_B T/h)$, U_0^* , stands for the activation energy of breakdown and α is an adjustable constant for the particular system under study. The time necessary to complete the build-up stage is then obtained by integration. Generally, the lifetime τ (time-to-failure) can be shown to be estimated by:

$$\tau = \frac{\exp\left(-\frac{\alpha \pi \epsilon E_0^2 L_0}{k_B T}\right)}{\nu_0 a_0 \exp\left(-\frac{U_0^*}{k_B T}\right) \frac{\alpha \pi \epsilon E_0^2}{k_B T}} \quad (10)$$

This expression illustrates how lifetime increases exponentially as E increases or U_0^* decreases. This conclusion is in perfect agreement with experimental results in SiO_2 films [24].

Energy Transfer between the Micro and Macro Scale

Assuming that as in fracture mechanics dielectric rupture experiments also show that there are intermitences in the plot of crack growth velocity versus time and versus crack length, then one may conclude that those oscillations can make the velocity reaching values below $v_L(v_0 \rightarrow v_L)$ and when the energy injected in the crack tip corresponds to a speed larger than it, there is the possibility to create new steps (branching). As long as an energy surplus exists in the crack tip, that process can be repeated indefinitely giving rise to multifractality. The time delay τ between the injected flux of energy in the tubular channel tip, and the spent energy flux (or dissipated power, Ψ_s) to form the surfaces of the conductive channel, produces with origin in the critical velocity, a decoupling between the velocity of the growth channel, $v_\phi(t) = v_c(t)$, and the rate of formation of the channel surfaces:

$$v_s(t + \tau) = \frac{dL_0(t + \tau)}{dt} \quad (11)$$

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This decoupling is responsible for the indetermination of the channel growth velocity, and on its turn for the instability in the shape of the oscillations of that velocity. The instability gives rise to a rough channel that increases the consumption of energy, and so limits its growth velocity to a value $v_0 \leq c_R$; this way, a new delay is produced in the formation of the breakdown surfaces, and increasing even more the decoupling between the injected flux and the rate of energy needed to form the conductive surfaces, and so forth. The resulting pattern can sometimes be associated with a fractal or multifractal geometry, where each successive branching is self-similar or self-affine to the previous one, showing that its propagation reveals a memory effect at scale or branching levels. Therefore, geometrical aspects of the dissipation pattern surface, can be analysed and studied in order to obtain information about the process that generated it [10]. The growth dynamics of those surfaces can be abstracted from a purely geometric study of the surface structure, with the purpose of including such dynamics within a class of phenomena already known from nonlinear dynamics and chaos theory [9].

The analogies between mechanical fracture and dielectric rupture, have been also revealed at the experimental level in which concerns patterns growth and dynamics of fast dielectric rupture propagation [25-27].

In order to explain those experimental analogies it is developed in this article a physical model where the time delay is explicitly introduced, generalizing the classical formulation of the fast intrinsic dielectric rupture. It is possible to formulate the variation of the applied electric field and the electric displacement in a time interval, while a disruptive channel and the contour around it are progressing. The branching happens as a consequence of the energy release being so high that not all of it can be absorbed in a unique disruption zone [3].

Therefore, a non-equilibrium thermodynamic principle must exist when the instability conditions and the formation of the branching structure can be related.

In processes occurring far from equilibrium, the system will seek to dissipate excess energy by moving away from the thermodynamically favourable for the equilibrium by a path where the efficiency in energy dissipation is not just about the change in the geometry of a path, but also in search of a more efficient way of energy dissipation. Therefore, several unstable physical systems that have extreme energy dissipation, shows branch patterns. Consequently, the geometrical aspect of a dissipation pattern can tell us how the dissipative process occurred, and phenomenology indicates that a general non-equilibrium principle is responsible by the most of it. Therefore, to study the growth of dissipative patterns it is necessary to know its causes by mean of a phenomenological model where evolution equations can be obtained and a general principle for non-equilibrium thermodynamics is applicable to preview them.

It is possible to show that a simple non-trivial stochastic model of dielectric breakdown naturally leads to fractal structures for the discharge pattern [28]. Wiesmann and co-workers introduced a stochastic model to simulate the growth of a fractal structure which resembles a Lichtenberg figure. They determined the Hausdorff dimension and other fractal properties of the structure and showed that there are close relations to other fractal structures (e.g., diffusion limited aggregation). Multifractal analysis of Lichtenberg figures in connection with physical conditions of electrostatic discharges experiments have found that fractal dimension of the total structure clearly depends on those physical conditions [29]. However, since their model does not include a breakdown field, it was necessary to introduce a critical field for growth, E_c , in a way that the growth probability is assumed proportional to the local field E_{loc} if $E_{loc} \geq E_c$, and zero if $E_{loc} < E_c$ [28]. In addition, an internal field in the structure, $s E$, was introduced. The electrostatic potential in the structure is no longer equal to the Φ_0 of the connecting electrode but $\Phi_0 + sE$, where s is the length of the path along the structure which connects the point to the electrode. Breakdown occurs only if the voltage exceeds the product $E_s d$, where d is the electrode spacing. The essence of the stochastic procedure assumes that growth probability depends on the local electric field determined by the equipotential discharge pattern. The discrete form of the Laplace equation, $\nabla^2 \phi = 0$, for the 2D lattice, where ϕ is the electrostatic potential, can be written as

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$$\phi_{i,k} = \frac{1}{4}(\phi_{i+1,k} + \phi_{i-1,k} + \phi_{i,k+1} + \phi_{i,k-1}) \quad (12)$$

and given the appropriate boundary conditions, the potential is obtained by iterating this equation.

Authors who first recognized fractal features of a surface discharge channel structure and tried to estimate its fractal dimension [30], used a computer model for solving the problem of ramification by introducing the growth probability p dependent on a local electric field E :

$p \sim E^\eta$, where η is a model parameter. They presented a dielectric breakdown model where they assumed that the breakdown structure had the same potential as the high voltage source and considered the breakdown process to proceed in a stepwise manner with a stochastic choice of extension from a field-weighted ($\sim E^\eta$) probability distribution. Fractal branched structures were produced whose fractal dimension depended upon the value chosen for η . The branching tree propagation has been shown [31] to exhibit deterministic chaos with the Lyapunov exponent inversely proportional to the fractal dimension of the tree. While such model produces structures with the shapes that are observed experimentally in solids, it lacks physical justification.

Later on, a deterministic model for the electrical-tree breakdown mechanism [32], enables to simulate branched structures in homogeneous polymers. It is non-linear in the local electric fields and contains feedback features, which generate deterministic chaos in the discharges driving the breakdown, such as have been observed in laboratory experiments. It was surmised that it is this chaotic behaviour which is responsible for branch structures that it generates.

Analysis of the computed gas discharge data [1], where the amount of damage produced is assumed to be proportional to the number of ionisations caused by the avalanche, and hence the fraction f_{av} of critical damage generated is given by

$$f_{av} = \frac{Nb}{N_c} \left\{ \exp \left[\frac{Lb}{\lambda} \exp \left(-\frac{IL_b}{\lambda \Delta V} \right) \right] - 1 \right\} \quad (13)$$

N_c is the critical number of ionisations for formation of a new tubule, N_b is the number of initiating electrons, λ is the minimum distance for impact-ionisation, I is the ionisation potential, and ΔV is the potential difference over the path-length L_b of the avalanche. Values for L_b and I are taken from experiments, ΔV and N_b are computed, and λ is assumed to be progressively increased from a material dependent value λ_0 as the damage is accumulated according to

$$\lambda_{new} = \lambda_{old} + f_{av} L_b \quad (14)$$

The initial value of λ i.e. λ_0 may be as low as the scattering path-length for electrons (5 nm in polyethylene [1], but could be much higher depending upon the material morphology. For instance, it has been shown that the model reproduces the experimental field dependence of electrical-tree growth with mean values of λ in the range 50-150 nm. Computations show that the smallest values of ΔV are $\Delta V \sim 50-100$ V and hence when $\lambda \approx \lambda_0$ the avalanches are very rare. Thus the average damage per avalanche given by equation (13) is small. However, the gradual increase of λ with damage, as expressed through equation (14) leads eventually to large avalanches and the occurrence of tree extension. The mechanism therefore contains the possibility of a regime of deterministic chaos, through the non-linearity of the field dependence of equation (13), the positive feedback of equation (14) and the complex feedback processes of the discharge-avalanche charge redistribution.

The model is implemented for an ac voltage of defined peak value applied to the point electrode. The point electrode is taken to a hyperboloid of revolution with a chosen radius of curvature. It is useful to define the damage within a tree of length L as proportional to the sum of the lengths of all individual branches (channels) comprising the

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tree structure which will be denoted by $\sigma(L)$. It will be shown that $\sigma(L)$ is dependent on a non-integer power of the longitudinal tree length L .

A common method of estimating the fractal dimension of a tree that has been projected onto two dimensions is to construct arcs from the inception point; then the sum $\sigma(L)$ of the lengths of all the branches within an arc may be plotted as a function of the arc radius L to yield a log-log straight-line plot with a slope equal to the fractal dimension. This procedure is valid only when it is possible to represent the trees using a 2D plane. Defining a nondimensional damage S by $\sigma(L)/L_b$, where L_b is the average length of a newly-created channel, then since trees are fractal structures, $S=(L/L_b)^{-d_f}$, and they are considered branched trees if the fractal dimension d_f is in between 1.2 and 1.8, or bush trees if $2.4 < d_f < 3$. In AC fields it is known that $S(t)=(L(t)/L_b)^{d_f}=t/t_{ch}$ where t_{ch} is a characteristic time for channel formation. In an avalanche (as in gas discharge), an electron gains kinetic energy as it is accelerated by the field. If the mean free path between collisions in the direction of the electric field is sufficiently long and the field sufficiently high, then the kinetic energy gained may be enough to cause ionization upon impact with a molecule so that another electron is liberated. Tow send defined a primary ionization coefficient α as the number of electrons liberated by an initial electron in passing through unit distance of gas in the direction of the field. In solids this coefficient can have different forms of functional dependence on electric field. In passing through a distance dx , the increase in the number of free electrons dn is therefore $n(x)\alpha dx$ where $n(x)$ is the number of free electrons at x .

Thus, $dn/dx=\alpha n$, and $n(x)=n_0 \exp(\alpha x)$ where n_0 is the number of initiating electrons. In order to formulate an expression for the conditions required to form a new discharging electrical tree-branch from an avalanching charge tree-trunk, it is assumed that the amount of damage caused by an avalanche is proportional to the number of impact ionizations and that both the avalanche length and newly-formed tree branch length are the same (L_b):

$$S_{aval} \sim n_0 [\exp(\alpha L_b) - 1] \quad (15)$$

This length L_b is determined by various factors of which one of the most prominent is the short duration of the discharge ≈ 20 ns. By inserting this equation into the above equation of $S(t)$, and replacing α with the appropriate function of electric field, it is possible to relate tree length to field and time. Furthermore, by imposing the condition that breakdown will occur soon after a tree has reached a critical length, it is possible to relate time-to-breakdown to applied voltage [33].

Modified Maximum Energy Dissipation Principle and Instability Dynamics

Due to the nature of matter at the nanoscale, the interaction between particles (atoms or molecules) admits characteristic times, lengths or speeds at which energy transfer should occur. For this reason, the physical process of dielectric rupture is microscopically limited by these fundamental characteristic quantities of energy transfer within the system.

The analogies between the intrinsic dielectric breakdown and fast fracture mechanics have been still explored here, in order to include the recent modified maximum energy dissipation principle [9]. Considering the irreversible process of a structure formation as shown in the Fig. 2, where the excess of energy that flows to the external medium of the system, is calculated, making use of an excess energy integral, M , along the entire contour, Γ , of the dissipation structure formed [3]; namely, assuming that the outward flow exists in the form of sound, heat, radiation, etc., and all the internal process given by the difference between the injected energy flux, $d\phi/dt$, and the energy rate used to form the pattern, $\Psi_s \equiv dU_s/dt$, i.e. $d\phi/dt - dU_s/dt$, which contributes to the formation of a branched fractal structure. Integrating over a contour Γ of a curve that involves the entire structure we have, that the integral of the energy excess rate is given by:

$$\oint_{\Gamma} \phi(L_{\phi}, \vec{v}) ds = \int_{\Gamma} \left[\frac{d\phi(L_{\phi}, \vec{v}_{\phi})}{dt} - \frac{dU_s(L_s, \vec{v}_s)}{dt} \right] ds \geq 0 \quad (16)$$

Where $\phi(L_{\phi}, \vec{v})$ is the free energy injected into the system and $U_s(L_s, \vec{v}_s)$ is the energy used to form the dissipation pattern.

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In order to create one conductive channel branch a thermodynamic special condition must exist as it will be shown in the next section. One can admit that the above mentioned decoupling between the two velocities (as responsible by instability process), is given by a time delay τ between them. This time delay can be calculated by the input and the output balance of the energy of the system using thermodynamics arguments, in terms of ratio between the minimal characteristic channel length L_{0min} and the maximal possible channel growth velocity, $v_{\phi max}$ as:

$$\tau_{\phi} = \frac{L_{0min}}{v_{\phi max}} = \frac{GL_{0min}}{M(L, \vec{v})_{max}} = \frac{L_{0min}}{[C_R - v_s / \alpha]} = \text{characteristics} \quad (17)$$

where $v_{\phi max} \approx 2/3c_R$ and c_R the limiting velocity given by Rayleigh waves velocities and $\alpha=G/R$, where G is the electrostatic energy released rate and R is the conductive channel growth resistance. Another way to calculate the time delay can be made considering the maximum velocity of the pattern formation, $v_{s max}$ giving a similar result as follow:

$$\tau_s = \frac{L}{v_{s max}} = \frac{RL}{M(L, \vec{v})_{max}} = \frac{L}{[c_R - \alpha v_s]} = \text{characteristic} \quad (18)$$

The decoupling of velocities by a characteristic time delay can be verified by this equation. This characteristic time is likely to be typical of processes where there is a maximization of power [34]. Considering the existence of a minimum conductive channel size, L_{0min} and that also exists a maximum growth velocity, v_s , it can be concluded that there is a characteristic time as shown in the expression above.

Therefore, it can be conjectured that, for each energy flow, $d\phi(L, \vec{v})/dt$, imposed to the system, the size of the elementary structure formed, L_0 , (where $L_{0min} \leq L_0 \leq L_{0max}$) changes with the flow, generating a multifractal structure in the case where the fractal dimension, D_q , or the magnitude $\tau(q)$, analogous to the free energy density [31,35,36], also varies instantaneously. Then the fractality shall be local and not global, but with a fractal dimension, D_q , different for each region. Since the energy dissipation is described by a growth process of multifractal nature, entropy production is no longer a (global) extensive magnitude. In this case the entropy becomes the one given by a not extended type (local) entropy [28,37], for example.

Considering the excess $M(L(t), \vec{v}(t))$ between the supplied energy rate, $d\phi(L_{\phi}(t), \vec{v}_{\phi}(t))/dt$ and the dissipated energy rate, $\psi_s(L_s(t), \vec{v}_s(t))/dt$, with each one respectively given by:

$$\bar{\phi}(L_{\phi}(t), \vec{v}_{\phi}(t)) = GD(L_{\phi}(t), \vec{v}_{\phi}(t)) \cdot \vec{v}_{\phi}(t) \quad (19)$$

and

$$\psi_s(L_s(t'), \vec{v}_s(t')) = \vec{R}_s(t') \cdot \vec{v}_s(t') \quad (20)$$

The integrating of Eq. (16) is made in the following way: each stage k the system increases the injected energy, consequently the pattern formed changes its geometrical aspect by a quantity L_k in its length. Therefore the excess between the injected and the dissipated energy spent in the formation of the pattern is calculated in each stage, where the new excess is used in the next stage $k+1$, as shown in the Fig. 2c.

$$\overbrace{(\bar{\phi}_k - \psi_{sk})}^{M_k \geq 0} \rightarrow \overbrace{(\bar{\phi}_{k+1} - \psi_{sk+1})}^{M_{k+1} \geq 0} \rightarrow \overbrace{(\bar{\phi}_{k+2} - \psi_{sk+2})}^{M_{k+2} \geq 0} \quad (21)$$

The conductive branching are then caused by

$$\varphi(L, \vec{v}) \equiv \int_{\Gamma} M(L, \vec{v}) ds = \int_{\Gamma} [\bar{\phi}(L_{\phi}, \vec{v}_{\phi}) - \psi_s(L_s, \vec{v}_s)] ds \geq 0 \quad (22)$$

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The conductive branchings are then caused by

$$\underline{R}_s(t') = u_s \rho_s(t') \tag{23}$$

which is the dynamic resistance to the conductive dielectric patterns growth, where u_s is the unitary energy spent to form the pattern, and $\rho_s(t')$ is a vectorial density describing the number of elements in the pattern of a given direction and $L_s(t')$, $v_s(t')$ are the length used to form the pattern and its respective speed developed by the dissipation pattern as the branched conductive dielectrics in its growth.

From the instantaneous energy balance between the input and output of a dissipative system, and considering the instantaneous energy excess, $\phi(L,v)$, delimited by a Γ curve that involves the structure, the modified maximum dissipation energy principle (MDEP) is [3,9]:

$$\begin{aligned} \delta \left[\varphi(L, \vec{v}) \right] &= \int_{\Gamma} \delta M(L, \vec{v}) ds = \\ \int_{\Gamma} \delta \left[\vec{G}_{\phi}(L_{\phi}(t), \vec{v}_{\phi}) \cdot \vec{v}_{\phi} - \vec{R}_s(L_s(t'), \vec{v}_s(t')) \cdot \vec{v}_s(t') \right] ds &= 0 \end{aligned} \tag{24}$$

The structure considered is generated in different steps of iteration, resulting possibly in invariant structure by scale transformation. In order that the above integral is maximal we must have:

$$\frac{\partial M}{\partial \vec{v}_{\phi}} = \frac{\partial \bar{\phi}}{\partial \vec{v}_{\phi}} = \frac{\partial \psi_s}{\partial \vec{v}_{\phi}} = \frac{\partial (\vec{G}_{\phi} \vec{v}_{\phi})}{\partial \vec{v}_{\phi}} - \frac{\partial (u_s \rho_s \vec{v}_s)}{\partial \vec{v}_{\phi}} = 0 \tag{25}$$

Once there is a time delay the conservation of energy theorem must be observed, and it can be deduced from the equation where one has:

$$\int \frac{\partial (\vec{G}_{\phi} \vec{v}_{\phi})}{\partial \vec{v}_{\phi}} d\vec{v}_{\phi} = \int \frac{\partial (u_s \rho_s \vec{v}_s)}{\partial \vec{v}_{\phi}} d\vec{v}_{\phi} \tag{26}$$

Or

$$\vec{G}_{\phi} \vec{v}_{\phi} = u_s \rho_s \vec{v}_s \tag{27}$$

Considering the existence of this time delay corroborated by the experimental correlations measurements in dielectric rupture between the oscillations in the channel growth velocity and the damage profile [9,38] the pattern formation velocity \vec{v}_s in the time t is equal to the velocity permitted by the instantaneous deformation \vec{v}_{ϕ} in a time after $t'=t+\tau$, i.

e. considering the hypothesis of a time delay, we have:

$$\vec{v}_s = \vec{v}_{\phi}(t + \tau) = \vec{v}_{\phi}(t) f(t/\tau) \tag{28}$$

and

$$\vec{G}_{\phi} \vec{v}_{\phi} = u_s \rho_s \vec{v}_{\phi}(t/\tau) \tag{29}$$

since the flow of energy $\bar{\phi}(L, \vec{v})$ is the amount defined in the Eq. (19). One can obtain from the variational calculation of the Eq. (20) the following energy conservation principle given from Eq. (27) as

$$\bar{\phi}(L_{\phi}(t), \vec{v}_{\phi}(t)) = \psi_s(L_s(t'), \vec{v}_s(t')) \tag{30}$$

or

$$\bar{\phi}(L_{\phi}(t), \vec{v}_{\phi}(t)) = \psi_s(L_s(t + \tau), \vec{v}_s(t + \tau)) \tag{31}$$

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This is the Conservation Energy Theorem for systems with a time delay which relates the injected power with the dissipated power.

Since there is a delay between the input and output energy fluxes or velocities, the clue is to consider a self-similar or self-affine time solution for the problem, as follow:

$$\bar{\phi}(L_\phi(t), \vec{v}_\phi(t)) = \vec{G}_\phi(L_\phi(t), \vec{v}_\phi(t)) \cdot \vec{v}_\phi(t) \rightarrow \psi_s(L_s(t'), \vec{v}_s(t')) = \vec{R}_s(L_s(t'), \vec{v}_s(t')) \cdot \vec{v}_s(t') \quad (32)$$

or

$$\bar{\phi}(L_\phi(t), \vec{v}_\phi(t)) = \vec{G}_\phi(L_\phi(t), \vec{v}_\phi(t)) \cdot \vec{v}_\phi(t) \rightarrow \psi_s(L_s(t'), \vec{v}_s(t+\tau)) = \vec{R}_s(L_s(t'), \vec{v}_s(t+\tau)) \cdot \vec{v}_s(t+\tau) \quad (33)$$

Therefore, one assumes that the instability in the dielectric breakdown dynamics occurs as a consequence of the time delay between the injected energy flux $d\phi/dt$ and the dissipated energy flux $\psi_s(t')$, in the form of conductive channel surfaces, as the injected energy flux interacts with the dissipated energy flux $\psi_s(t)$ after a time delay, given by the relaxation process into the material, where, $t'=t+\tau$.

By the Maximum Dissipation Principle [9] for $\vec{v}_{\phi max} = c_R$ (the Rayleigh waves velocity) one has that:

$$\psi_s(t+\tau) = \frac{2\Gamma}{\left[1 - \frac{v_\phi(L_0(t))}{c_R}\right]} \frac{dL_s}{dL_0(t)} v_\phi(L_0(t+\tau)) \quad (34)$$

applying the Conservation Energy Theorem for systems with a time delay and using (19) in (31)

$$v_\phi(L_0(t+\tau)) = \frac{\vec{G}_D(L_\phi(t), \vec{v}_\phi(t)) \cdot \vec{v}_\phi(t)}{2\Gamma \frac{dL_s}{dL_0(t)}} \left[1 - \frac{v_\phi(L_0(t))}{c_R}\right] \quad (35)$$

Therefore, the time delay between the velocities where $v_{s v_\phi(t+\tau)}$, from Eq. (34) one obtains the logistic equation:

$$v_\phi(t+\tau) = \mu \vec{v}_\phi(t) \left[1 - \frac{\vec{v}_\phi(t)}{\vec{v}_{\phi max}}\right] \quad (36)$$

Where $\mu = 2\Gamma dL_s/dL_0(t)$.

In terms of a discretised process that happens in steps of time τ , for $x_{k+1} = \vec{v}_\phi(t+\tau)$ and $x_k = \vec{v}_\phi(t)/\vec{v}_{\phi max}$, one has

$$x_{k+1} = (1 - x_k) x_k \quad (37)$$

This equation shows that the energy dissipation can also be written in the form of a logistic map with the consequence that critical velocity, instability, bifurcation and channel branching, are not explained by the classical theory of intrinsic dielectric breakdown. The interpretation of this equation for the dynamic channel growth is shown in the Fig. 3. In this figure the injected flux $N_k = G_\phi v_k$ interacts with the dissipated flux $P_k = 2\gamma v_{k+1}$ in a way that the partial fraction of the velocity μx_k is balanced by the remaining partial fraction of the velocity $1 - x_k$.

The energy flux to the channel tip can be represented by Fig. 4. Observe from this that the MEDP (Slepyan Principle) along with decoupling in time between the velocities provide the mathematical solution for the dielectric rupture problem as logistic iterated system of equations with a time delay.

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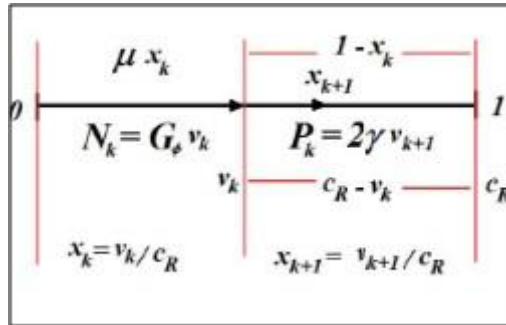


Fig.3. Interpretation of the logistic equation in terms of the degradation channel growth.

In the Fig. 4 one can also observe the point where the power and the efficiency is optimized when the Eq. (37) is satisfied. For more feed-backed irreversible iterations a doubling period can be obtained from the recursive Eq. (37). Successive iterations of this function can generate a bifurcation map.

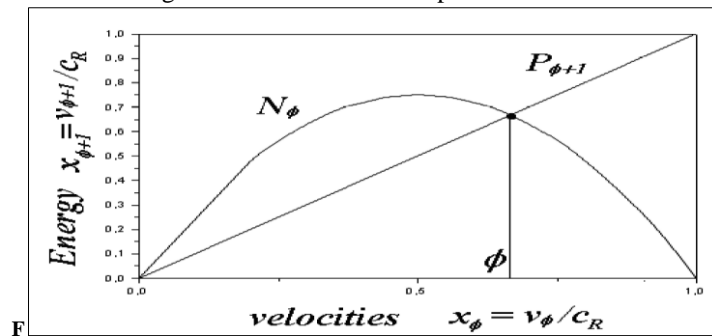


Fig.4. Maximum Dissipation Principle in terms of iterated function of $\phi_{\phi+1}=Gx_{\phi+1}$ versus $P_{\phi}=Rx_{\phi}$, for logistic equation.

III. DISCUSSION AND CONCLUSIONS

Therefore, it was shown from the results obtained with this proposed model that bifurcation process is in agreement with the assertion made by Gross: “The bifurcation is a response to the system being driven further from equilibrium and trying to find new ways to dissipate the forcing” [25]. This means that studying the details of the pattern can lead both to an increased understanding of the bifurcation and to the system’s dissipation process”. This seems to be a general feature of fast rupture in matter, as a dielectric breakdown, where dissipation geometric patterns are generated during the electric discharge like the spokes on stormy days, for example.

Still notice that time delay between the input and output fluxes in the system, originated the iterated equations and that on its turn gave rise the instability by means of bifurcation maps. Since Feigenbaum constant is universal and common to all of bifurcation maps, one can conclude that a universality of these maps is also associated with the universality of the principle of maximum energy dissipation. As a general principle in Nature dissipation of energy should be maximum while the excess is maximal. The logistic equation (37) results from the modified version of the Slepian’s maximum dissipation principle expressed by Eq. (24) assuming the ideal case of electric equivalent to the Griffith semi-infinite plate.

In short words, the branched electrical conductive channels present in the intrinsic dielectric rupture of materials are a consequence of instability and extreme dissipation of energy. Through a modification of the non-equilibrium

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thermodynamics Principle of Maximum Dissipation in which a local roughness and a delay time are added in its formulation, it is possible to include fractal structures in order to obtain the dielectric rupture dynamic equations derived from this principle. The iterated functions provide a bifurcation map of all possibilities of the dynamical system, explaining the pattern formation associated to dissipation in the considered system. The theoretical and experimental analogies between mechanical fracture and dielectric rupture, take for granted the applicability of this model in the prediction of dielectric rupture patterns. The results show a clear picture and a phenomenological sequence of complete non-equilibrium thermodynamics, capable of explaining the instability of the dynamics equations until the dissipation structure growth or the formation of fractal patterns in the branching conductive channels.

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