proceedings

MDPI

Proceedings

Nonequilibrium Thermodynamics and Entropy Production in Simulation of Electrical Tree Growth †

Adrián César Razzitte [1,*], Luciano Enciso [2], Marcelo Gun [2] and María Sol Ruiz [1]

1 Department of Chemistry, Universidad de Buenos Aires, 1063, Buenos Aires, Argentina; mariasol1984@gmail.com
2 Department of Electrical Engineering, Universidad de Buenos Aires, 1063, Buenos Aires, Argentina; luloenciso@gmail.com (L.E.); marcelo.gun@gmail.com (M.G.)
* Correspondence: adriancr2007@gmail.com
† Presented at the 5th International Electronic Conference on Entropy and Its Applications, 18–30 November 2019; Available online: https://ecea-5.sciforum.net/.

Published: 17 November 2019

Abstract: In the present work we applied the nonequilibrium thermodynamic theory in the analysis of the dielectric breakdown (DB) process. As the tree channel front moves, the intense field near the front moves electrons and ions irreversibly in the region beyond the tree channel tips where electromechanical, thermal and chemical effects cause irreversible damage and, from the nonequilibrium thermodynamic viewpoint, entropy production. From the nonequilibrium thermodynamics analysis, the entropy production is due to the product of fluxes $\boldsymbol{J}_i$ and conjugated forces $\boldsymbol{X}_i$: $\boldsymbol{\sigma = \sum_i J_i X_i \geq 0}$. We consider that the coupling between fluxes can describe the dielectric breakdown in solids as a phenomenon of transport of heat, mass and electric charge.

Keywords: entropy production; dielectric breakdown; nonequilibrium thermodynamics

---

1. Introduction

All materials conduct electricity to a greater or lesser extent and all suffer some form of breakdown in a sufficiently strong electric field.

For low field strengths the conduction process is ohmic, but as the field strength is increased, the conductivity usually becomes field-dependent; if the field strength is increased further, some form of destructive irreversible conduction takes place [1]: the dielectric breakdown.

Gases and liquids due to the mobility of their molecules do not present permanent damage (channels) when the application of the field ceases, something that does happen in solids. The permanence of these channels in the dielectric breakdown of solids is attributed to thermal effects: local fusion of the material that originates the formation of the channel and gives rise to the transport of ion or electron carriers within it.

Dielectric solids usually exhibit a permanently damaged discharge track.

In a recently published work [2], we have made an analysis of the entropy associated with the structure of electric trees through the behavior of four entropies in the field of statistical mechanics in the framework of the DBM Dielectric Breakdown Modell [3].

Historically, the treatment of the simulation of the dielectric breakdown has placed emphasis only on the electric field, particularly in the local field, as the sole cause of the dielectric breakdown, omitting perhaps other factors such as the thermal effect or the presence of moisture. The latter usually affects the structure of the insulating material in high-voltage transformers.

In this work, we will try to correlate the entropy production associated with the simulation process of the structure formed by means of the DBM model with observables of real systems. In other words, we will try to give physical meaning to the simulated discharge within the framework of an "abstract" structure, where the discharge structure, the electric tree, is characterized by the parameters of fractal geometry with fractal dimension $\mathbf{d_f}$: $1 < \mathbf{d_f} < 2$ for branched trees.

Given that the dielectric breakdown is a clear example of a nonequilibrium process, we believe that the originality of this work lies in the combination of a stochastic model based on the DBM for the description of the local field and the purely electrical contribution and others effects associated with the dielectric breakdown process, such as the ionic or electronic conduction, or the presence of moisture on the insulator material, or thermal instability. For this purpose we will use the methods of linear nonequilibrium thermodynamics.

2. Electrical Conductivity of Dielectrics

The electrical conductivity of the dielectrics can be ionic, electronic or both. It is very difficult, from an experimental point of view, to be able to separate these contributions, particularly to high fields.

The ionic conductivity (σ) is simply due to the migration of positive and negative ions. The basic expression of all electrical conduction is given by:

$$\sigma = \sum_i n_i e_i \mu_i \quad (1)$$

Where n_i is the density of carriers i and e_i and μ_i are the corresponding charge and mobility, respectively.

In the usual analysis of low-field ionic conductivity one can write:

$$\sigma = \sigma_0 e^{-\frac{\phi}{kT}} \quad (2)$$

where σ_0 and Φ are determined experimentally (constants in a certain range of T).

Leaving aside the theoretical models that can be adopted to explain conduction in insulating solids, experimental data can be adequately described by Equation (2).

The basic equation of steady-state driving is:

$$J = \sigma E \quad (3)$$

Where J is the current density, E is the field strength and σ is the conductivity defined by Equation (1).The current and the potential difference V are experimentally measured. A simple procedure establishes the link between theory and experiment:

$$j = \frac{I}{A} \quad (4a)$$

$$E = \frac{V}{d} \quad (4b)$$

Where A is the cross section and d the thickness of the sample. Since the rupture path usually extends over a small area of the cross section, the current density in the rupture channel is much greater than that found with the set of Equations (4a) and (4b).

It is well known that the relaxation time associated with the current distribution in the steady state is given by:

$$\tau = \frac{\varepsilon}{4\pi\sigma} \quad (5)$$

where ε is the dielectric constant and σ is the conductivity. If the current is due to charge carriers of different species, the conductivity is given by Equation (1). In this case, a relaxation time is obtained:

$$\tau = \frac{\varepsilon}{4\pi n_i e_i \mu_i} \tag{6}$$

The relaxation time is associated with each species of cargo carriers where the eventual approach to the steady state will be determined by the value of the corresponding time constants. It can be easily verified that small concentrations of relatively immobile species can lead to relaxation times of hours. This is not important if the spatial charge density due to the carriers is small enough to produce a weak distortion of the electric field.

To estimate this effect, let us see, for example: $\mu = 10^{-10}\, cm^2 V^{-1} seg^{-1}, \varepsilon = 6$ with a relaxation time of 3600 s. This calculation gives a concentration of carriers $n_i = 10^{13}\, cm^{-3}$.

The Poisson equation for the effect of the special charge of the *i*-th species is:

$$\frac{\partial \phi}{\partial x} = 4\pi n_i e_i \tag{7}$$

which gives a value of 10^7V cm^{-2} for the speed of field variation.

3. The Model

We think of a dielectric breakdown model due not only to the effect of pure dielectric breakdown but also to the contribution of other effects, with the effect of moisture on the disturbance of the electric field during the pre-breaking process coming to mind.

In a rigorous analysis of the rupture process, we should take into account all possible causes: electric field, thermal effect and transport of matter.

In the formalism of linear nonequilibrium thermodynamics [4,5], the phenomenological equations that would describe the flows and coupled forces that characterize this complex phenomenon would have this form:

$$\begin{aligned} J_q &= L_{qq}(\nabla \frac{1}{T}) + L_{q\mu}(-\frac{1}{T}\nabla\mu) + L_{q\phi}(-\frac{1}{T}\nabla\phi) \\ J_\mu &= L_{\mu q}(\nabla \frac{1}{T}) + L_{\mu\mu}(-\frac{1}{T}\nabla\mu) + L_{\mu\phi}(-\frac{1}{T}\nabla\phi) \\ J_\phi &= L_{\phi q}(\nabla \frac{1}{T}) + L_{\phi\mu}(-\frac{1}{T}\nabla\mu) + L_{\phi\phi}(-\frac{1}{T}\nabla\phi) \end{aligned} \tag{8}$$

Where $\boldsymbol{J_q}$, $\boldsymbol{J_\mu}$ and $\boldsymbol{J_\Phi}$ in the set of Equation (8) are the fluxes of heat, matter and electric charge, respectively. Those forms obey $\boldsymbol{J_i = \Sigma_k L_{ki} X_i}$. The forces $\boldsymbol{X_i}$ of transport conjugate to the fluxes that are the thermal force [***grad* 1/*T***], the chemical force **[−1/*T grad*μ_T]** and the electrical force: **[−1/*T gradΦ*]. *L-coefficients***are the phenomenological coefficients, $\boldsymbol{L_{ij}}$ are the coupling coefficients; $\boldsymbol{L_{ij} = L_{ji}}$ by the Onsager Theorem. The diagonal coefficients are described by $\boldsymbol{\lambda}$ (thermal conductivity of the material), ***D*** (diffusion coefficient of the material) and $\boldsymbol{\kappa}$ (the electrical conductivity) following the classical laws of Fourier, Fick and Ohm respectively. In the framework of local equilibrium hypothesis we consider that the temperature of the system is constant during the breakdown process and equal to the melting point temperature (T = Tm) of the material in the site where the branch of the tree grows.

Then we consider only the dielectric breakdown in terms of the electrical force **[−1/*T gradΦ*]** and chemical force **[−1/*T grad*μ_T]** in a system with no transport of heat ***Jq* = *0***. Where T=Tm is the melting point of the material in the zone of branch growth, finally the equations of DB simulated are reduced to:

$$\begin{aligned} J_{\mu} &= L_{\mu\mu}\left(-\frac{1}{T}\nabla\mu\right) + L_{\mu\phi}\left(-\frac{1}{T}\nabla\phi\right) \\ J_{\phi} &= L_{\phi\mu}\left(-\frac{1}{T}\nabla\mu\right) + L_{\phi\phi}\left(-\frac{1}{T}\nabla\phi\right) \end{aligned} \tag{9}$$

In the set of Equation (9) we will consider T = Tm.

The simulation of the electric field effect was done using the classic DBM model in a 2D grid [2]. The used model is a two-dimensional square lattice (Figure 1) in which the opposite sides represent the two electrodes. Breakdown starts at a point of a high local field, and this is usually attributed to electrical conducting inclusions [6]. The conducting inclusions are represented by electrode pins.

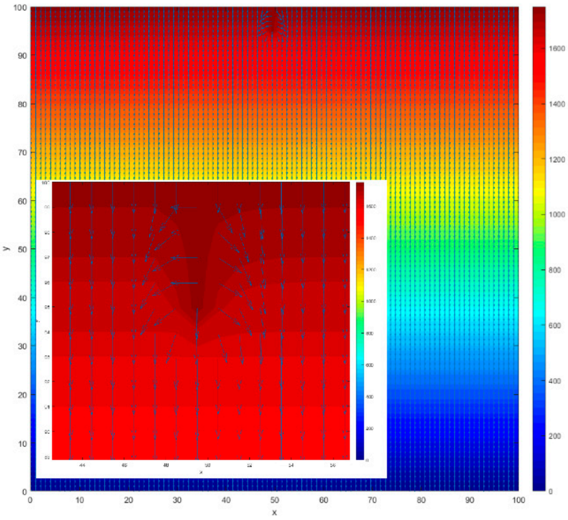

Figure 1. Potential and electrical field of grid with tip in potential electrode. Inset corresponds to magnified area in tip, on the side of potential electrode.

The equations of this model are [2,3]:

$$\begin{aligned} &\nabla^2\phi = 0 \\ &\phi_{i,k} = \frac{1}{4}(\phi_{i+1,k} + \phi_{i-1,k} + \phi_{i,k-1} + \phi_{i,k+1}) \\ &\mathrm{P}(\mathrm{i,k} \rightarrow \mathrm{i}', k') = \frac{(\phi_{i',k'})^{\eta}}{\sum_{j,i\in\Gamma}(\phi_{j,i})^{\eta}} \end{aligned} \tag{10}$$

For the model, see [2].

When we apply the model according to Equation (10) we obtain several results related to the fractal structure of the electric tree. In this opportunity we will try to relate some physical results of the model, such as the applied field strength and the number of branches of the structure formed with the characteristics of a real insulating system. Then we will relate these observables of applied field and conductivity with the characteristics of a given material (it can be cellulose with oil).

In the same way, we will relate the effect of the transport of water through the insulating material, which reduces the breaking stress with the number of branches (channels) formed.

In the framework of nonequilibrium thermodynamics, the entropy production is a bilinear form of conjugated fluxes and conjugated forces.

$$\sigma = \sum_i J_i X_i \geq 0 \tag{11a}$$

For isothermal systems and low values of the electric field E, the current density ***J*** is given by Ohm's law (Equation (3)), then ***J* = *σE* = *σT*(*E*/*T*)**, which means

$$L_{\Phi\Phi} = T\sigma \tag{11b}$$

By approximation to the Fick's law the diffusion coefficient $\boldsymbol{D = L_{\mu\mu}/T}$, which means

$$L\mu\mu = TD \tag{11c}$$

For the system to obey the second law:

$$L_{\mu\mu} L_{\phi\phi} \geq L_{\mu\phi}^2 \tag{12}$$

$$T(\sigma D)^{1/2} \geq L_{\mu\phi} \tag{13}$$

If we know the conductivity of the insulating material and the diffusion coefficient of the species that diffuses within it for certain field values, in the model given by the value of $\boldsymbol{\eta}$ we can estimate the entropy production by Equation (11a).

In the next figures (Figures 2 and 3) we see the behavior of the field strength and the number of channels formed according to the $\boldsymbol{\eta}$ parameter.

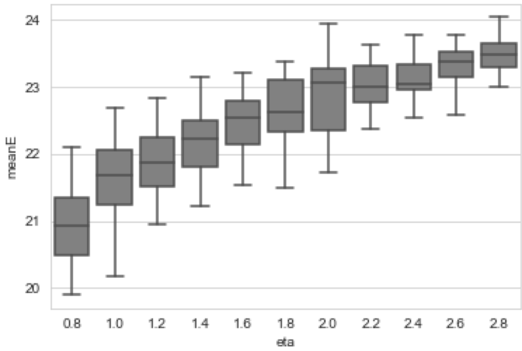

Figure 2. Increasing of electrical field in arbitrary units as a function of $\boldsymbol{\eta}$.

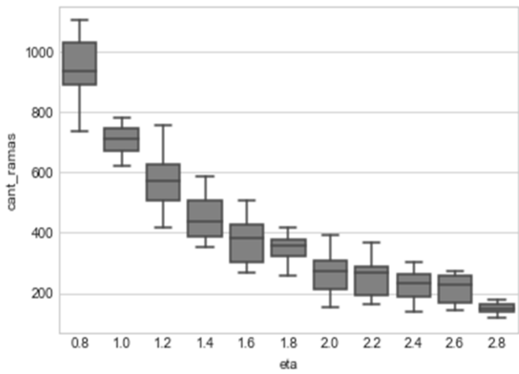

Figure 3. Number of branches of the tree as a function of $\boldsymbol{\eta}$.

In the simulation of the model we consider the number of branches (***Nb***) related to the diffusion coefficient of the contaminant inside the dielectric or insulator: $\frac{1}{T}\nabla\mu \approx \frac{1}{T}Nb$.

Combining the values of the field for each value of the η parameter with the number of branches obtained we can construct the equations of the model to simulate, modifying Equation (9), combined with Equations (11b) and (11c) as:

$$\begin{aligned} J_\mu &= TD(-\frac{1}{T}Nb) + L_{\mu\phi}(\frac{E}{T}) \\ J_\phi &= L_{\phi\mu}(-\frac{1}{T}Nb) + T\sigma(\frac{E}{T}) \end{aligned} \tag{(14)}$$

4. Conclusions

The presented model constitutes an attempt to associate the pure dielectric rupture with other effects that can also affect the phenomenon of rupture in insulators, such as the presence of small amounts of water that accumulate or perhaps are formed by degradation of products in the insulators of high voltage. We see through a model based on nonequilibrium thermodynamics presented in Equation (14) as it influences the presence of the contaminant in the production of entropy of the system by the use of the Equation (11a).

References

1. O'Dwyer, J. *The Theory of Electrical Conduction and Breakdown in Solids Dielectrics*; Clarendon Press: Oxford, UK, 1973.
2. Enciso, L.; Gun, M.; Ruiz, M.S.; Razzitte, A.C. Entropy in multifractal non equilibrium structures of dielectric breakdown. *J. Stat. Mech. Theory Exp.* **2019**, *2019*, 094011.
3. Niemeyer, L.; Pietronero, L.; Wiessmann, H.J. Fractal dimension of dielectric breakdown. *Phys. Rev. Lett*. **1984**, *52*.
4. De Groot, S.R.; Mazur, P. *Non-Equilibrium Thermodynamics*; Dover: Mineola, NY, USA, 1984.
5. Prigogine, I.; Kondepudi, D. *Modern Thermodynamics*; Wiley: Hoboken, NJ, USA, 1998.
6. Dissado, L.A. Understanding Electrical Trees in Solids: From Experiments to Theory. *IEEE Trans. Dielectr. Electr. Insul.* **2002**, *9*, 483–497.