# Percolative Model Of Electric Breakdown In Liquid Dielectrics

J. Qian, R. P. Joshi, K. H. Schoenbach, and M. Laroussi

Dept. of Electrical & Computer Engineering

Old Dominion University, Norfolk, VA 23529-0246

E. Schamiloglu and C. Christodoulou

Dept. of Electrical and Computer Engineering

University of New Mexico, Albuquerque, NM 87131.

A percolative approach, useful in modeling liquid dielectric breakdown, is presented. The dielectric is treated as a network of resistors having random values and breakdown characteristics based on a specified statistical distribution. The method is quite general, and lends itself to the inclusion of internal fluctuations and localized heating. It should also be applicable to mixtures and facilitate the inclusion of air-bubbles. Despite its simplicity, the model successfully characterizes fractal structure in dielectric breakdown. In particular, the fractal dimension for a 2D lattice as given by the exponent of a power law, agrees with the theoretical value. The dependence of critical external voltage on the internal disorder is also investigated. It is shown that the overall breakdown process consists of the successive breakdown of individual elements to finally form a percolation cluster. The clusters have a typical dendrite structure. Also, in keeping with qualitative expectations, it is shown that $V_{br}$ decreases with the disorder and increases with resistor variance.

## INTRODUCTION

There is considerable interest in the study of electrical breakdown in water (and other liquids) for a variety of applications. For example, water filled gaps are used to design acoustic equipment [1,2], for the insulation of high-voltage devices [3], as the medium in spark erosion machines [4], and as energy storage and switching elements for pulsed power systems. Dielectric breakdown in liquids, gases, and solid insulators is frequently characterized by two features: (i) The occurrence of narrow discharge channels, and (ii) a strong tendency of these channels to branch into complicated stochastic patterns. Typical examples include lightning [5], surface discharges leading to Lichtenberg figures [6], and treeing in polymers [7]. Since electrical breakdown (at least for voltages near the threshold range) appears to be a variable process, a statistical description of the entire assembly seems necessary for an accurate and physical representation. From a dielectrics breakdown standpoint, liquids seem to have some advantages over solids and gas systems. For example, the electric breakdown strengths are higher as compared to gases. Their ability for circulation leads to greater heat dissipation and homogeneity as compared to solids, and liquid dielectrics are well suited for applications involving complex geometries.

The theoretical study of breakdown in liquids under high electric fields has largely relied on the use of perturbation theory for calculation of transport parameters. Lekner advanced a theory of electron motion in liquid argon [8]. He postulated that electrons scatter off an effective potential comprised of a superposition of screened-electron-large-argon atom potentials. Later Jahnke et al. [9], showed that the single-center effective potential theory disagreed with the experimental scattering length, its dependence on density and the slope. Davis et al. [10] similarly demonstrated the difficulty in developing a satisfactory theory for electron motion starting from scattering cross sections with liquid molecules. The emphasis of most calculations has been on transport parameters such as the drift velocity and diffusion coefficient. For example, Basak and Cohen [11] calculated the electron mobility in a rare liquid gas assuming the existence of a conduction band and taking density fluctuations as the only scattering mechanism. Ascarelli included the role of phonon scattering [12]. The scattering cross section based approach for electron transport in liquids was applied more recently by Kunhardt [13] based on a Monte Carlo (MC) scheme. The advantage of the kinetic MC scheme is that it overcomes the assumption made by several researchers that a state of quasi-equilibrium exists between electronic states [11,12,14,15]. Despite the improvement, a number of issues and problems remain. For example, scattering calculations are based on perturbative theory that assumes the existence of Bloch-like states for electronic transitions. However, in the absence of long-range order for liquids, this assumption becomes questionable. Also, in reality, both continuum and discrete states need to be considered. Moreover, these techniques do not include random inhomogeneities, nor are they capable of predicting the stochastic patterns and streamers observed in liquid breakdown. Finally, it has recently been shown that gas phase streamer mechanisms and breakdown physics cannot be applied to liquids since electron impact ionization is insignificant at fields for which breakdown is observed [16].

Here, we attempt to develop a simple percolative model to describe transport and breakdown in liquids. Percolation theory since its inception [17] has become a major statistical tool for interpreting and predicting morphological [18-20] and transport properties [21-24] of disordered media. Description of transport based on drift-diffusion approaches (both equilibrium and non-equilibrium treatments) are valid at distances much larger than the correlation length of percolation theory. For smaller distances, the diffusion equation is no longer applicable and the process becomes non-Markovian [25]. Thus, diffusion or Boltzmann-type approaches cannot be used for systems that are disordered, have small length scales, or the phenomena of interest occurs over small distances. Diffusion theory is also inadequate in describing systems whose elements change position or characteristics with time. Breakdown in liquid water is a case in point since: (i) The system is heterogeneous, consisting of dissolved gases, solutes, and contains a mixture of hydrogen (H) and deuterium (D) atoms to form both $H_2O$ and $D_2O$. (ii) Also, asperities and density fluctuations exist at the contacts [26] . This is known to affect injection at metal-liquid interfaces, and lead to the nucleation of streamers and "tree-like structures" at high applied voltages [27]. (iii) Thermal expansion that can result from inelastic energy transfers and/or external energy injection (e.g. in laser driven situations). In gases, the heat absorption aspect can be ignored due to the low density, but not in liquids and solids. The thermal expansion can potentially lead to density gradients, form localized "bubbles", and create pressure waves [28]. (iv) Growth and transport of internal bubbles which make the system characteristics dynamic, and result in instabilities at the vapor-liquid interfaces [29].

In this contribution, a percolative approach to modeling dielectric breakdown is presented. Conduction is treated in terms of current flows through a network of resistors having random values and breakdown characteristics based on a specified statistical distribution. The method relies on first carrying out self-consistent evaluation of the local potentials. The conductivity of individual resistive elements is computed from the potentials in keeping with assigned current-voltage characteristics. Failure of a constituent resistor element occurs if the local fields exceed a critical threshold. However, breakdown of the overall structure will take place if a "failure channel" percolates all the way from one electrode to the other. The electrical model uses the stochastic concepts proposed by Niemeyer et al. [30]. Random models have also been applied to treat transport in sintered composites and materials containing grain-boundaries [31], and for quenched random media [32]. Water could be a typical liquid dielectric of interest. It is shown in this study that despite its simplicity, the model can successfully characterize fractal structure in dielectric breakdown. The dependence of critical external voltage on the internal disorder, and the fractal behavior has also been investigated.

## SIMULATION DETAILS

The simulation model is developed and implemented by considering a two-dimensional (2D) rectangular lattice in which the two sides (left and right) represent the electrodes. A 2D representation has been used for simplicity, but the approach can easily be extended for three-dimensional analysis as well. The approach relies on initially generating an array of random seed points within a rectangular simulation lattice. Each of these seeds can be associated with a cell (a 2D polygon in this case) containing all regions that are nearer to it than any other seed. Such elemental Voronoi polygons [33] are formed by intersecting perpendicular bisectors of lines connecting neighboring seeds. The 2D polygon surrounding each seed point then represents a local region within the liquid dielectric. Resistors, having random values are placed between every pair of seed points. Physically, the resistor values depend on the resistivity and geometric characteristics of each 2D polygon. For time-dependent modeling, this approach can easily be extended to include capacitors. The attributes of each resistive element can, in principle, be assigned based on material physics to include all details to the level desired. Hence, it becomes possible to include effects such as local density fluctuations arising from variable solute concentrations, the presence of bubbles, and fractional compositions of each component in case of mixtures such as oils, thermal effects, etc. Here, the resistor magnitudes at the start of the simulation were assigned randomly based on a specified distribution function (DF). For concreteness, a Gaussian DF has been used here with the mean $<R>$ and variance $\sigma_R$ being user specified input parameters for the simulation. Thus, each resistor between pairs of seed points can be made identical to or different from the other, depending of the variance value specified (i.e. $\sigma_R \geq 0$.)

Figure 1 depicts a simple rectangular lattice with uniformly distributed seed points. As shown, the resistor network is periodic, but each element has a different random value. For an N x M rectangular geometry (M longitudinal and N transverse segments), the total number of resistors would be: $2*N*M - M - N + 1$. Voltage (Dirichlet) boundary conditions are applied to all nodes on each side. The Kirchhoff current equation (KCE) at each of the internal nodes (i.e. seed points) yields a set of coupled simultaneous equations that can be solved if the current-voltage (I-V) characteristic of each resistive element is specified. Here the following simple, non-linear I-V relationship was chosen to test the percolative model:

$$R(E) = R_{oi} \ , \quad \text{for } E < E_{crit} \ , \qquad (1a)$$

$$\text{and,} \quad R(E) = R_{of} \ , \quad \text{for } E \geq E_{crit} \ , \qquad (1b)$$

where, E is the local electric field within each resistor segment, $E_{crit}$ the critical threshold electric field for breakdown, $R_{oi}$ the initial random value assigned to the $i^{th}$ resistive segment based on the DF, and $R_{of}$ the final value upon breakdown. Physically in this simple nonlinear model, the resistance drops dramatically upon breakdown from $R_{oi}$ to $R_{of}$. Here, the magnitude of $R_{of}$ was chosen to be 1 percent of the initial value. Thus, a relatively simple I-V nonlinearity was implemented to model a transition between a high resistance "current blocking" state to a low resistance "conducting" for $E \geq E_{crit}$. In principle, though, other more complicated non-linear I-V characteristics [34-39] could easily be implemented. The overall calculation would proceed as outlined here, but the computational time for solving the set of coupled KCEs would increase for complex I-V characteristics.

It should be pointed out that the issue of thermal perturbations and localized heating, if important, can also be addressed based on the current scheme. The heating aspect is important since it can lead to localized heating, expansion, bubble formation that results in density fluctuations. As recently proposed, a lowering of density in localized regions can be a mechanism for enhanced electron impact ionization [16]. The rarefied zones can then, in principle, act as seed sites for triggering eventual breakdown. In the present approach, as discussed above, solution of the KCEs would yield the current distribution in each branch of the network, and the node voltages. Power dissipation density within each segment can thus be computed from their product. From this the temperature profiles can be obtained by solving the heat diffusion equation using the power density, subject to appropriate boundary conditions. Then by explicitly including a temperature dependence of the resistance [i.e. $R_{oi}(T)$ and $R_{of}(T)$ ], effects of thermal heating would be automatically included. Determining the details of the resistance variations with temperature, however, is not a trivial task. For an accurate physical representation, one would need to take account of density fluctuations, phonon scattering, possible electronic trapping-detrapping effects in the liquid, and variations in impact ionization due to changes in the mean free path. A simple analytical approximation for R(T) would provide a quick solution. Here, the thermal heating aspect was ignored, and will be addressed elsewhere.

In the present model, the following procedure has been implemented for determining the breakdown voltage ($V_{br}$) for a given network size. A relatively low trial voltage is used as the external biasing value. The internal node voltages and branch currents are then computed by solving the KCE. If the local electric field exceeds the critical value $E_{crit}$ (assigned as an input parameter), the resistor values change in accordance to equation (1). Using the updated resistance network, a

KCE solution is recomputed. The procedure repeats until a percolative path is established between the two electrodes, or no further transitions in the resistance occur at any of the elements for the applied bias. The value of the applied voltage sets $V_{br}$ in the former case. In the latter situation, with an absence of a complete percolation channel, the biasing voltage is increased, and the above procedure repeated until an eventual a complete percolative path is achieved. It must be mentioned that the $V_{br}$ value thus obtained for a given network array is not unique since it depends on the initial random selection of the $R_{oi}$ value set. The uncertainty and statistical variation can be eliminated by repeating the full procedure numerous times, with different random-number seeds. This effectively yields multiple networks, each of which yields a breakdown value. Upon averaging, the mean $<V_{br}>$ value can be determined.

**RESULTS AND DISCUSSION**

Simulations for breakdown in a dielectric (such as purified/de-ionized water which is of interest to the pulsed-power community) were carried out based on the procedure outlined in the previous section. The value of $E_{crit}$ was set to $2 \times 10^5$ $Vcm^{-1}$ in keeping with experimental reports on breakdown in water [16,40], while $\sigma_R$ was retained as an adjustable parameter. For simplicity, all of results obtained here were for N = M. Simulation results showing the evolution of breakdown and the systematic development of a percolation cluster are shown in Fig. 2 for a 40 x 40 network with $<R>$ = 5000 Ohms. The total physical dimension for the network on each side was taken to be 3 centimeters. Thus, under ideal conditions (i.e. identical segments), a breakdown voltage of $3 \times 2 \times 10^5 = 0.6$ MV is expected. However, in the presence of internal inhomogeneities, one expects the overall breakdown voltage to be lower since percolation via a connected path of least resistance can conceivably form. A similar logic lies behind the well-known reductions in overall "strength of chains" containing non-uniform elements, based on the weakest internal links. Here, two $\sigma_R$ values of 50 Ohms and 250 Ohms were used to probe the impact of non-uniformities within the constituent resistors on the overall breakdown strength. The results yielded a breakdown voltage of 0.545 MV for $\sigma_R$ = 250 Ohms, and 0.588 MV for the $\sigma_R$ = 50 case. For the $\sigma_R$ = 250 Ohms simulation, shown in Figs. 2(a)-2(d), the initial segment with co-ordinates around (5,30) is the first to breakdown. This is followed by a solitary increase to the left, a branching around this section, the development of other scattered breakdown sections, and finally a percolation with many failures on the right side. Results for the $\sigma_R$ = 50 Ohms case, shown in Figs. 2(e)-2(g) exhibit a similar breakdown pattern, and the occurrence of random trees. Thus, these simulations demonstrate two important aspects. First, breakdown involves the development of filamentary trees with associated random structure, followed by an eventual percolative path. Second, there is an inherent reduction in breakdown strength with increases in the internal non-uniformity, as manifested through a $\sigma_R$ higher value.

The dependence of breakdown voltage on the variance of resistor elements was simulated next. The results obtained for a 40 x 40 network are given in Fig. 3. Three points are evident from the figure. (i) First, the breakdown voltage decreases monotonically as the internal non-uniformity (i.e. $\sigma_R$) is increased. (ii) The variation and error range increases with $\sigma_R$ . This is expected since the variability in possible resistances associated with the electrical network increases, and so do the inherent fluctuations. (iii) An extrapolation of the mean $V_{br}$ values tends to the maximum theoretical value of 0.6 MV in the $\sigma_R$ –> 0 limit. The plot clearly demonstrates that in practical situations, the presence of strong local imhomogeneity (such as a bubble, the presence of localized laser excitation, or solute/solvant clusters) would work to dramatically lower the overall breakdown capability of the liquid dielectric. This is in keeping with experimental observations.

Another practical feature of interest in systems containing internal non-uniform segments is its scaling behavior. For a given mean resistance, $\sigma_R$ value, and $E_{crit}$ , the network is most likely to fail or breakdown when the distance between electrodes is the shortest. This is expected under such conditions, because of a higher probability of forming a percolation path connecting elements of least resistance. With increasing physical separation, the likelihood of a continuous connected channel at a given external bias decreases, and so the hold-off voltage can be expected to increase. The results of Fig. 4 bring out this feature more quantitatively. Shown in Fig. 4 are the mean $V_{br}$ values obtained from the simulation for different network lengths and $\sigma_R = 100$. Network sizes ranging from 10x10 to 40x40 were used, with a fixed primitive unit of 0.1 cm. Numerous random seeds were used for each N x N network to obtain the average $V_{br}$ value. A nearly linear scaling (for constant $\sigma_R$) of the hold-off voltage with the distance between electrodes is apparent. This trend is in keeping with the expected behavior for $\sigma_R = 0$ (i.e. with no variations), and validates the current approach. It must be mentioned, however, that the linearity obtained here is partly a result of the simplified I-V characteristics chosen. Use of more realistic expressions, such as polynomial I-V relations with variable "soft" thresholds $E_{crit}$ , would have led to deviations from linear scaling behavior. Physically though, variability in $E_{crit}$ is not generally expected, except in cases of heterogeneous mixtures.

Another scaling characteristic, in this context, is the behavior of $V_{br}$ on the feature size of the constituent elemental resistances. In the simulations of Fig. 4, an elemental unit length of 0.1 cm was used, while the earlier simulations for the 40 x 40 networks of total length 3 cm had equal segments of 0.075 cm (=3 cm/40) each. Physically, this elemental unit, Δ, represents the characteristic length scale over which the resistor values (or conduction characteristics of the liquid) fluctuate. From a practical standpoint, actual values of $\Delta$ would depend on such factors as the size of air bubbles, the extent of internal inhomogeneities etc.. In the limiting case of very large $\Delta$ (i.e. only one segment between the two opposite electrodes), the mean $V_{br}$ value would be at its highest level of 0.6 MV. With increasing number of fragments (i.e. smaller $\Delta$ parameter), the breakdown voltage is expected to decrease as the probability for a completed path via an inter-connection of low resistance segments increases. Fig. 5 shows the variation in mean hold-off voltage with elemental length for a fixed 3 cm electrode separation and a $\sigma_R$ of 100. Networks ranging from 10x10 to 40x40 were used to vary the number of elements placed along the 3 cm distance. The highest value is for the 10 x 10 network, with a monotonic decrease as expected.

Finally, the relationship between the number of "broken" resistors and the total resistive elements in the network, at the breakdown threshold, was examined for possible fractal structure of these networks. As is well known for fractal structures, the total number of branches N(R) within a circle of radius "R"follows a power law [41] : $N(R) \sim R^D$ , with D being a non-integer exponent. Alternatively, the power law can also be applied by varying the network size N x N, and plotting the relationship between the number of broken resistors ($N_B$ ) to $N_T$ the total network elements [42]. Following this procedure based a number of different initial random seeds, the curve shown in Fig. 6 was obtained. The values of <R> and $\sigma_R$ were chosen to be 5000 and 100 Ohms, respectively. For convenience the numbers have been plotted on a log-log scale to the base 2. A curve fitting procedure to the points yielded the following least-square error relationship: $N_B = 0.25\ N_T^{0.96} \equiv k\ N_T^{\eta}$. Following Mandelbrot's area (A)-length (L) relation [41] of: $A^{0.5} \propto L^{1/D}$ , leads to $D = 2\eta$. Here, $\eta = 0.96$ which yields: $D = 1.92$. This value is very close to the fractal dimension of 1.89 for randomly diluted percolation clusters on a two-dimensional (2D) lattice [43]. The close agreement between our calculation and the theoretical 1.89 value for a 2D geometry is very encouraging, and underscores the inherent fractal structure.

## SUMMARY AND CONCLUSIONS

There is considerable interest in the study of electrical breakdown in water (and other liquids) for a variety of applications. These include water filled gaps for acoustic equipment, the insulation of high-voltage devices, as the medium in spark erosion machines, and as energy storage elements for pulsed power systems. However, the dielectric breakdown phenomenon in liquids is not very well understood, and the development of physical models to simulate the processes remains a germane issue. The breakdown is usually characterized by the occurrence of narrow discharge channels, and a tendency for these channels to branch into complicated stochastic patterns. Hence, any attempts to develop a physically based model needs to include, at the very least, this stochastic element. Most previous model studies of breakdown have relied on the use of perturbation theory. Perturbation approaches become questionable since they do not incorporate randomness, nor does the system have the inherent periodicity necessary for determining the eigenstates.

In this contribution, a percolative approach to dielectric breakdown has been presented. Conduction is treated in terms of current flows through a network of resistors having random values and breakdown characteristics based on a specified statistical distribution. The conductivity of individual resistive elements is computed from the potentials in keeping with assigned current-voltage characteristics. Failure of a constituent resistor element occurs if the local fields exceed a critical threshold. However, breakdown of the overall structure occurs if a "failure channel" percolates all the way from one electrode to the other. It has been shown that the method is quite general, and lends itself to the inclusion of all the internal physics and microscopic details. The model is capable of treating internal fluctuations, inhomogeneities and localized heating. It should be applicable to mixtures and facilitate the inclusion of air-bubbles. Improvements to the basic simulation scheme presented here can be achieved by coupling calculation segments for the current-voltage characteristics, thermal dependence of resistance, and details of “soft-breakdown”.

It has been shown here that despite its simplicity, the model can successfully characterize fractal structure in dielectric breakdown. In particular, the fractal dimension for a 2D lattice as given by the exponent of a power law, agreed with theoretical value. The dependence of critical external voltage on the internal disorder was also investigated. It has been shown that the overall breakdown process consists of the successive breakdown of individual elements to finally form a percolation cluster. The cluster was shown to have a typical dendrite structure. Also, in keeping with qualitative expectations, it has been shown that $V_{br}$ decreases with the disorder and variance in resistor values. In the limit of zero variance, the maximum breakdown value was recovered. This has obvious implications of a lowered hold-off voltage for mixtures or liquids containing local fluctuations in solutes or air-bubbles.

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## FIGURE CAPTIONS

Fig. 1. A simple rectangular lattice of resistors for uniformly distributed seed points.

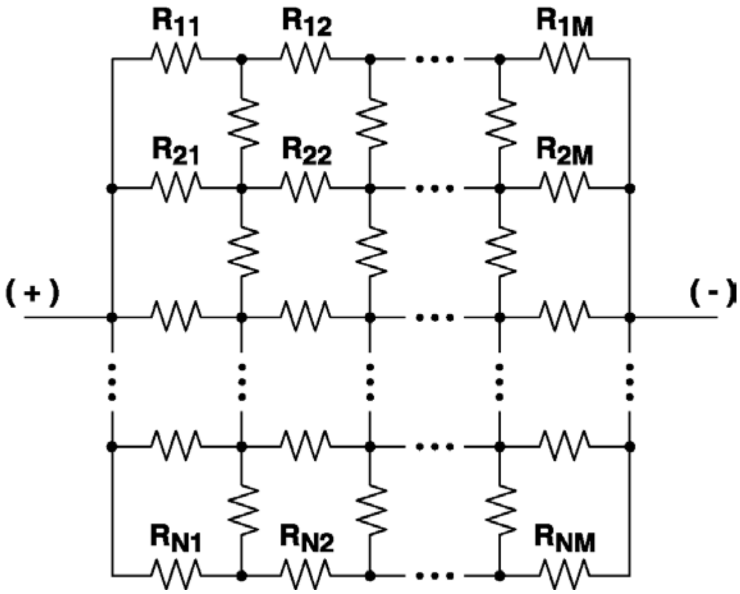
Fig. 2 Breakdown evolution examples for a 40 x 40 network with <R> = 5 kOhms. (a)-(d) Different network states from intact to percolative breakdown for $\sigma_R$ = 250 Ohms, and (e)-(g) the various network states for $\sigma_R$ = 50 Ohms.

Fig. 3 Dependence of the breakdown voltage on the resistor variance.

Fig. 4 Scaling behavior of hold-off voltage with separation for a fixed $\sigma_R$ = 100 with a primitive unit of 0.1 cm.

Fig. 5 Variation in the mean hold-off voltage for a fixed 3 cm electrode separation and $\sigma_R$ = 100 with the number of elements along each side. Networks ranging from 10x10 to 40x40 were used.

Fig. 6 Relationship between number of "broken" resistors ($N_B$ ) versus the total number of resistor elements in network ($N_T$) to probe fractal properties.


R11
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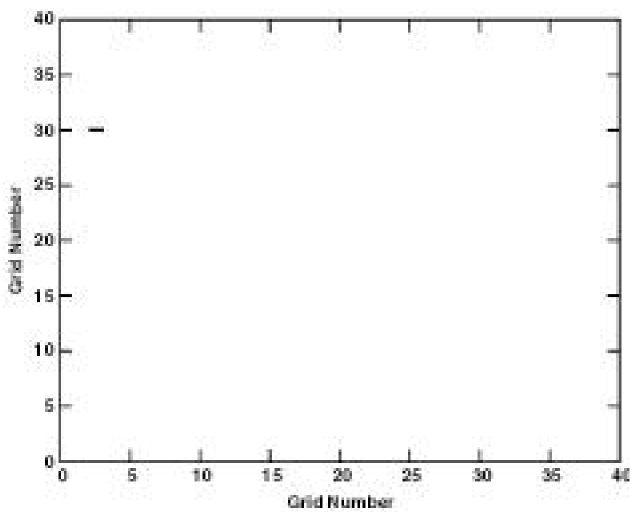
RN1
RN2
RNM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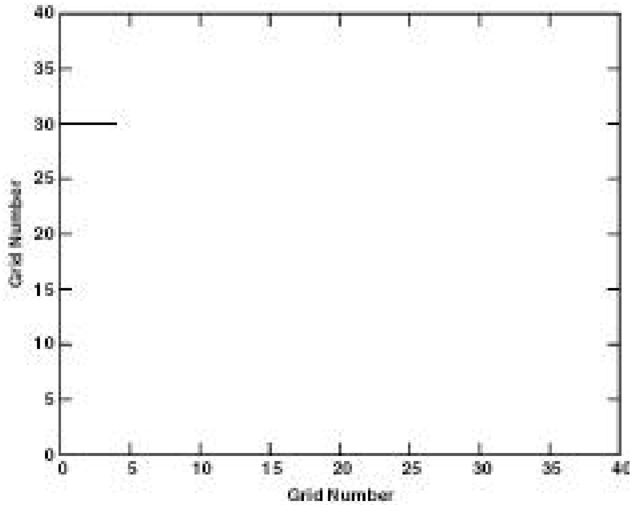
Grid Number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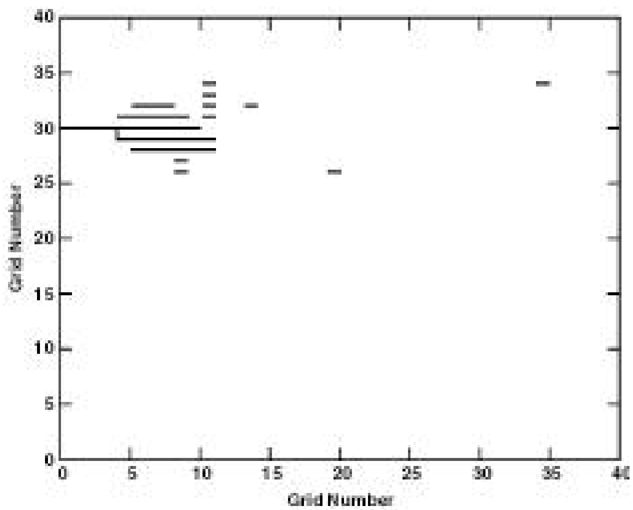
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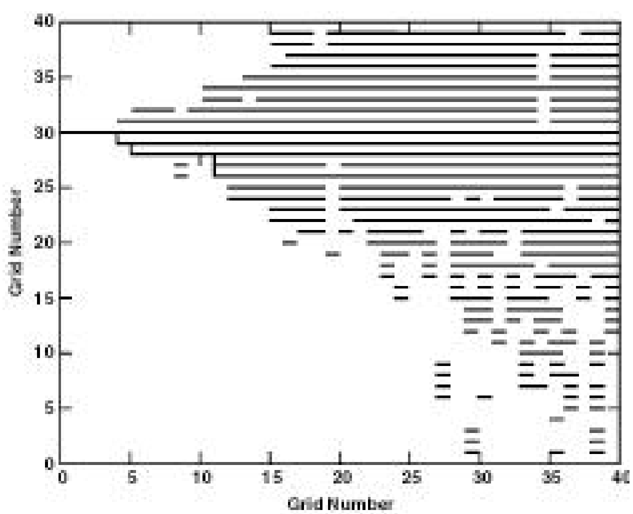
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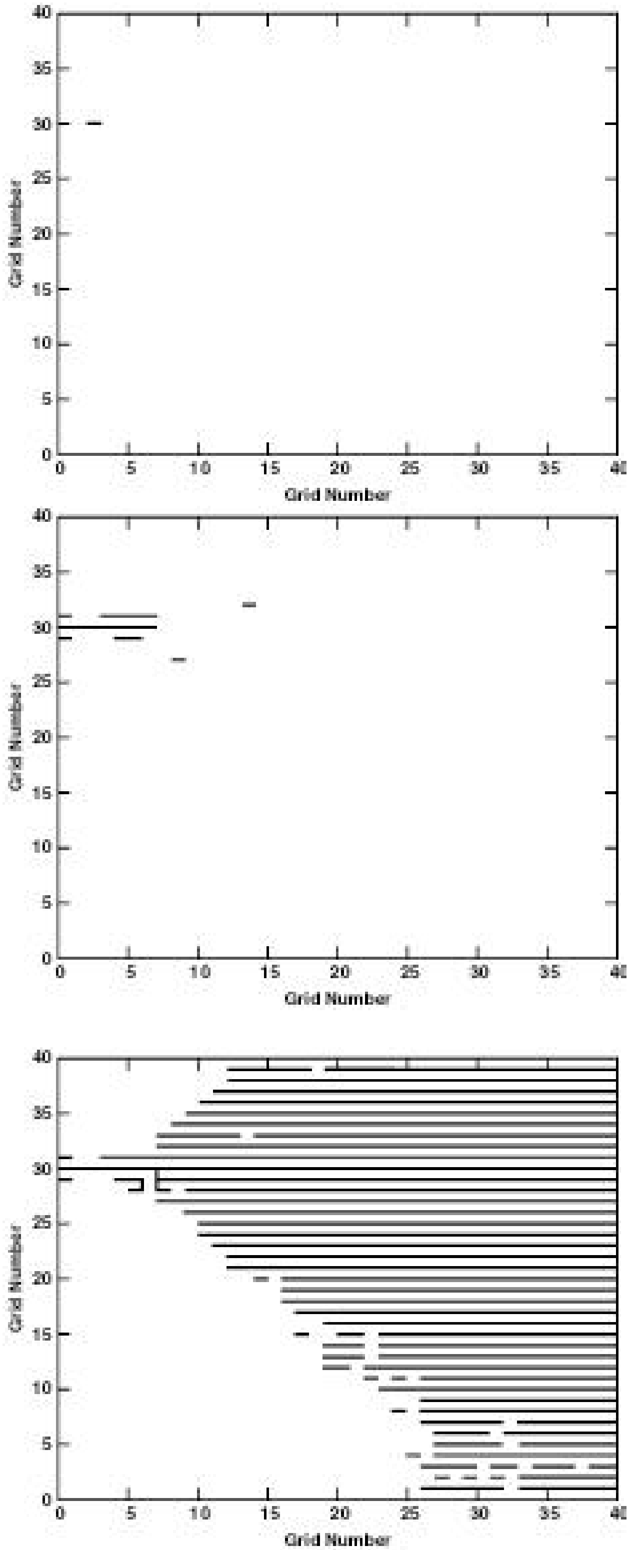
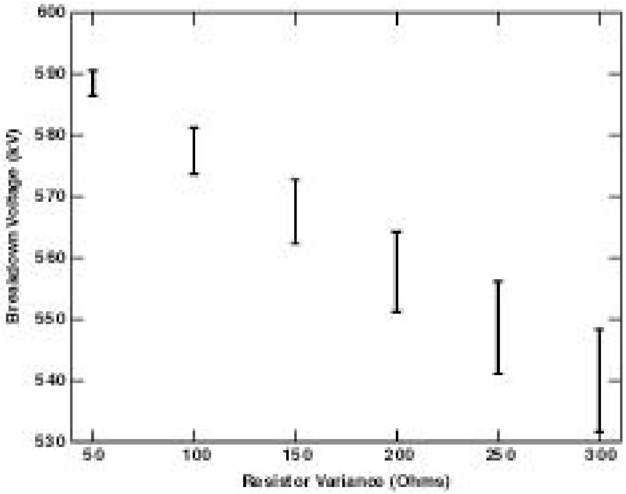
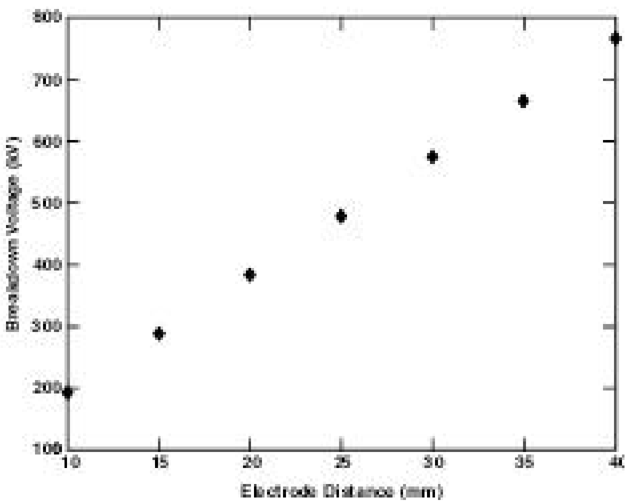
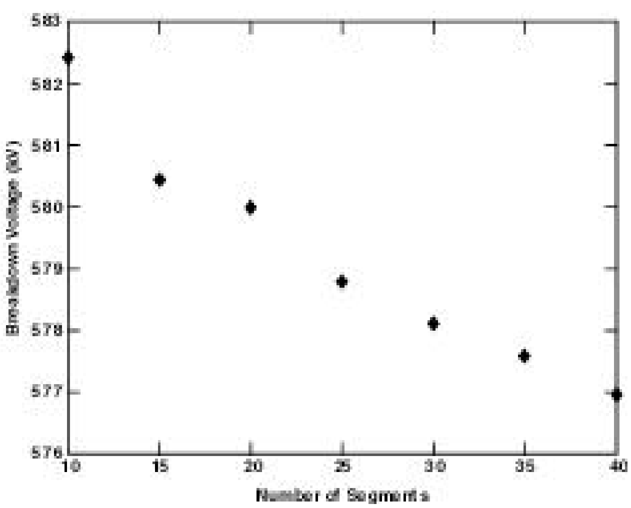
Grid Number

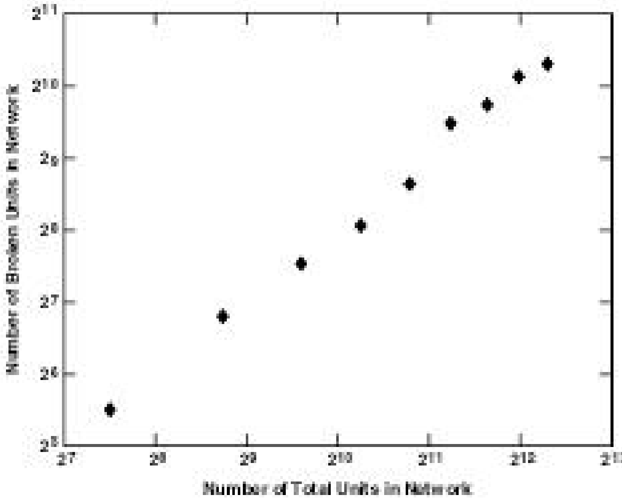


Grid Number
Grid Number
Grid Number
Grid Number
Grid Number
Grid Number


Breakdown Voltage (kV)
Resistor Variance (Ohms)
600
590
580
570
560
550
540
530
50
100
150
200
250
300


Breakdown Voltage (kV)
Electrode Distance (mm)
800
700
600
500
400
300
200
100
10
15
20
25
30
35
40


Breakdown Voltage (kV)
Number of Segments
583
582
581
580
579
578
577
576
10
15
20
25
30
35
40


Number of Broken Units in Network
Number of Total Units in Network