Conductivity and Dielectric Breakdown of Heterogeneous Materials. Application of the Renormalization Group Approach

Vitaly V. Novikov and Oleg P. Poznansky

Odessa Polytechnic University, 270044 Odessa, Ukraine

and

Valery P. Privalko

Institute of Macromolecular Chemistry, National Academy of Sciences of Ukraine, 253160 Kiev, Ukraine

INTRODUCTION

In recent years, there has been a growing interest in the treatment of strongly disordered systems as fractal objects /1,2/; typical in this respect is the fractal geometry approach to the so-called percolation conductivity /3,4/ and dielectric breakdown /5,6/ problems. This approach treats stochastically heterogeneous systems consisting of conducting and non-conducting phases; the geometrical phase transition is assumed to occur at the volume concentration of the former $p = p_c$, and the conductivity σ in the vicinity of the percolation threshold p_c to scale with the difference $(p\text{-}p_c)$ as

$$\sigma \sim (p\text{-}p_c)^t, \tag{1}$$

where t is the critical conductivity index.

The model of dielectric breakdown (DB) is representative of a family of generalized models of random growth. It is assumed that a model system consists of dielectric and conducting phases separated by a stochastic dynamic boundary, the dynamics of the latter being controlled by the growth rate of "damaged particles" at the cluster periphery. Properties of dielectric and conducting phases should obey the following physical conditions, eq. (2) and (3), respectively.

$$\Delta\varphi = 0;\ E = -\nabla\varphi, \tag{2}$$

$$\Delta\varphi = -Q;\ E = -\nabla\varphi, \tag{3}$$

where φ is the potential, E is the electric field voltage and Q is the charge density. The boundary problem is complemented by boundary conditions, $\varphi|_B = const$, where B means boundary.

Disregarding for the sake of simplicity the complex pattern of the processes of charge nucleation, transfer and annihilation is equivalent to assuming $\varphi = const$ throughout the conducting phase (i.e., the conductivity of this phase is ideal); in this case, the charge density will be non-zero and proportional to the field voltage in a dielectric phase at the interfacial boundary. The initially small fluctuation of the field variable may grow deterministically up to the onset of the cluster of "damaged particles" bridging the opposite electrodes. The breakdown voltage V in the vicinity of the percolation threshold is assumed to behave asymptotically as

$$V \sim (p\text{-}p_c)^t p, \tag{4}$$

where t_p is the critical index for breakdown voltage.

In analogy with thermodynamic phase transitions, one may assume scaling invariance for effective conductivity in the critical region and estimate the relevant critical indices with the aid of the real space renormalization group (RSRG) technique /7/, treating a disordered structure at the percolation event as a statistically self-similar (fractal) object, the mass of which, M_f, scales with its characteristic linear dimension L, as $M_f \sim L^{d_f}$ (where d_f is the non-integer fractal dimensionality).

RSRG TRANSFORMATION

Let the model system comprise a high-conductivity phase (of conductivity σ_c^0 and concentration p_c) and a low-conductivity phase (of conductivity σ_d^0 and concentration $(1-p_c)$), and let the ratio of conductivities be $\sigma_d^0/\sigma_c^0 = c << 1$. Let both phases be randomly distributed on the square unit cell lxl according to the following partition density function:

$$P_0(\sigma) = (1-p_0)\,\delta(\sigma-\sigma_d^0) + p_0\,\delta(\sigma-\sigma_c^0) \qquad (5)$$

This function (keeping $p_0 = const$) may be realized by different ways with many possible bond configurations on a given unit cell. Thereafter, such a representative set of cells will be treated as a renormalization cluster (RC) which may change its size, lxl, but ought to maintain a symmetrical shape in each RSRG step.

The main idea of the RSRG approach, that is, replacement of an initial non-homogeneous cell by its homogeneous counterpart with effective properties, may be realized by a series of successive averaging over the sizes of RC, each step involving formulation of a new partition function as a combination of δ-functions weighted in accordance with the step number. The disordered nature of RC, however, is preserved in each RSRG step in the scale interval $l << \xi << L$, where l and L are the smallest and largest size of RC, respectively, and ξ is the RC size in the course of the RSRG procedure. The result of evolution of the partition function after n steps of renormalization may be expressed as

$$P_n(\sigma) = (1-p_n)\,\delta(\sigma-\sigma_d^n) + p_n\delta(\sigma-\sigma_c^n), \qquad (6)$$

where

$$p_n = R(p_{n-1}) \qquad (7)$$

is the p_n change according to RSRG transformation. The function $R(p)$ is defined as the ratio of the number of bonded configurations to the total number of all possible tries in a given RSRG step. As a result, the cluster size ξ_n of T_n cell is related to that (ξ_{n+1}) in the next step (T_{n+1}) as

$$\xi_{n+1} = \xi_n/l. \qquad (8)$$

It should be clear that the iteration process is completed if ξ_n tends to zero after n steps in so far as the cell T_n becomes homogeneous and the partition function transforms to $P_n(\sigma) = \delta(\sigma-\sigma_{ef})$, where σ_{ef} is the effective conductivity of a percolation system with p_0 as the concentration of a conducting phase.

The RSRG transformation technique outlined above was used to analyze the conductivity and dielectric breakdown in model percolation systems. The starting cells were 3x3, 4x4 and 9x9 (two-dimensional case) or 3x3x3 (three-dimensional case). The computed R-function is shown in Fig. 1. The percolation threshold p_c for each model was associated with an inflexion point of the corresponding R-function (i.e., the abscissa of the point of intersection of the R-function with a bisectrix). The fractal dimensionality d_f for each model was determined as $d_f = d(\ln M_f)/d(\ln L)$, where M_f is the fractal mass at the percolation threshold (cf. Table 1).

EFFECTIVE CONDUCTIVITY

The conductivities σ_c^n and σ_d^n of bonded and non-bonded RC in the n-th step (two-dimensional case) were estimated using the relevant formula for lower and upper conductivity bounds /8/, i.e.,

$$\sigma_c^{n+1} = \sigma_d^n[1-(1-p_n)^{1/2} + c^n(1-p_n)^{1/2}(c^n + (1-c^n)(1-p_n^{1/2})^{-1}], \qquad (9a)$$

$$\sigma_d^{n+1} = \sigma_d^n(1-p_n^{1/2} + p_n^{1/2} + p_n^{1/2}[1+(c^n-1)p_n^{1/2}]^{-1}, \qquad (9b)$$

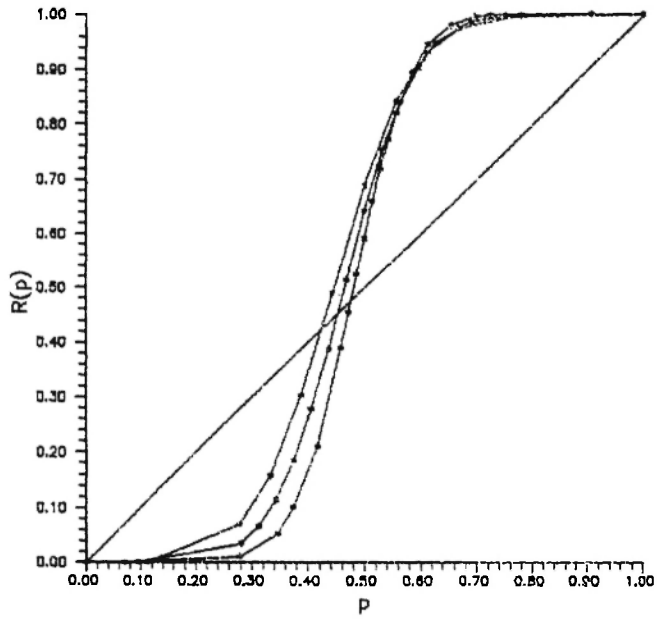

Fig. 1: *R*-functions for the cells of dimensions: 3x3 (1), 6x6 (2) and 9x9 (3).

Table 1
Geometrical parameters of fractal lattices

Model, *lxl*	p_c	d_f
3x3	0.4267	1.623
6x6	0.4766	1.790
9x9	0.4883	1.843
3x3x3	0.2109	1.793

where $c^n = \sigma_d^n/\sigma_c^n$.

In the three-dimensional case, Hashin-Shtrikman's formulae for upper and lower bounds /9/ were used assuming conductivities of bonded and non-bonded RC to be given by the following expressions, respectively:

$$\sigma_c^{n+1} = \sigma_c^n p_n+\sigma_d^n(1-p_n)+p_n(1-p_n)(\sigma_c^n-\sigma_d^n)^2/[p_n\sigma_d^n+(1-p_n)\sigma_c^n+\sigma_c^n], \quad (10a)$$

$$\sigma_d^{n+1} = \sigma_c^n p_n+\sigma_d^n(1-p_n)+p_n(1-p_n)(\sigma_c^n-\sigma_d^n)^2/[p_n\sigma_d^n+(1-p_n)\sigma_c^n+\sigma_d^n]. \quad (10b)$$

The step-by-step averaging for a conductivity problem was carried out making use of eq. (7) and Fig. 1. As can be seen from Fig. 2, the conductivity scales with L as $\sigma \sim L^{d}c$, where d_c is the corresponding exponent. The critical conductivity index t in the vicinity of the percolation threshold determined as $t = d\ ln(\sigma)/d\ ln(p-p_c)$ (Table 2) was in good agreement with $t \sim 1.27$, as reported in /10/.

DIELECTRIC BREAKDOWN

The relevant model is the Z^d lattice /5,6/ with the *S*-structure as a conducting phase, and its supplement $Z^d \backslash S$ as a dielectric phase. It is assumed that the *S*-structure grows by one unit in each DB step until the

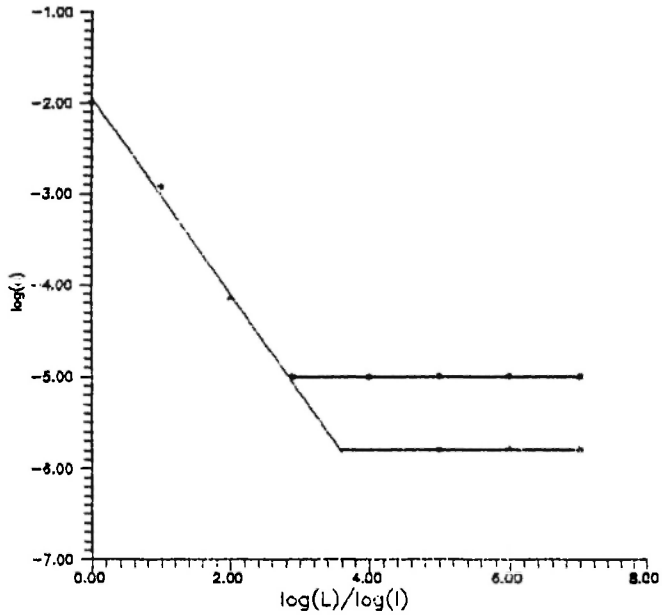

Fig. 2: Dependence of effective conductivity on the RSRG transformation step at the concentrations $(p-p_c)$. 0.001 (1), 0.01 (2).

Table 2
Critical conductivity indices for a fractal cluster

Model, *lxl*	t	t_p
3x3	1.19	1.19
6x6	1.21	1.08
9x9	1.23	---
3x3x3	2.12	---

opposite electrode is touched at the onset of bonded configuration. Discretizing eq. (3), one obtains

$$\Delta\varphi = \sum_{(i,j)} \delta_{i,j}(\varphi_i - 2d\varphi_j), \qquad (11)$$

where (i,j) are the nearest neighbors in $Z^d \backslash S$, $d = 2$, and

$$\delta_{i,j} = \begin{cases} \sigma, Z^d \backslash S \\ \lambda.\sigma, S \end{cases} (\lambda >> 1)$$

Before DB, the conductivity σ is assumed to be distributed over the square lattice according to the following partition function,

$$\rho(\sigma) = (1-p)\delta(\sigma) + p\delta(\sigma - \sigma_c), \qquad (12)$$

where p is the concentration of resistors of conductivity σ [thus, $(1-p)$ will be the concentration of resistors of zero conductivity].

The elementary acts of transition from one phase into another will be simulated by irreversible changes of conductivity. Assume that a constant voltage $V < V_c$ is applies to a resistor before DB (where V_c is a certain "critical" voltage); the DB at $V > V_c$ is accompanied by a conductivity increase from σ to $\lambda\sigma$ and remains thereafter unchanged at all values of V.

The system of linear equations (11) may be solved numerically using the following algorithms /6/

i. Calculate the conductivity σ with partition function (12) for each bond of the square lattice so as to ensure the onset of a bonding cluster between opposite electrodes;
ii. Apply the boundary condition U to opposite electrodes;
iii. Solve eq. (11) for φ_i;
iv. Calculate the conductivity of a system before DB;
v. Check every resistor (excluding those in which DB has already occurred) and replace $\delta_{ij} = \sigma$ by $\delta_{ij} = \lambda\sigma$ in the conditions ensuring DB, $(\varphi_i - \varphi_j) > V_c$;
vi. Stop the process if the electrodes are connected by a cluster of "damaged particles"; otherwise, relax the boundary condition so as to ensure the DB condition at one of the resistors available and return to item iii.

Let the limiting voltage corresponding to the onset of the first cluster of "damaged particles" be named "DB voltage" (DBV) of a given unit cell; the mean DB voltage will be obtained by averaging the function (12) over different ways of its realization. The effective DBV is estimated in each step of the RSRG transformation; in the next step, both effective conductivity σ_n and DBV, V_c^n, are assigned to each bond and the process of DBV renormalization with the number of iteration steps n is continued until V_c^n eventually becomes independent of n.

As can be seen from the numerical results of this procedure (Fig. 3), the DBV in the vicinity of the percolation threshold obeys the following asymptotic law, $V_c \sim L^{d_p}$ (where d_p is the corresponding critical index). The critical index for concentration dependence of DBV, $V_c \sim (p-p_c)^t p$, was arrived at by a standard procedure as $t_p = d\ ln(V_v)/d\ ln(p-p_c)$; a similar procedure was used to estimate the critical index for conductivity, t. The values of the cited critical indices for different lattices, lxl, are listed in Table 2.

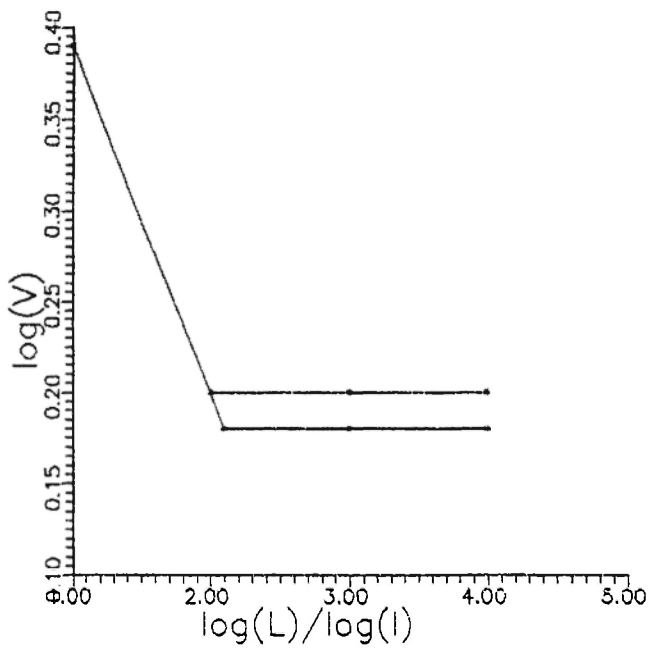

Fig. 3: Dependence of effective breakdown voltage on the RSRG transformation step at the concentrations $(p-p_c)$: 0.001 (1), 0.01 (2).

CONCLUSIONS

Application of the real space renormalization group transformation technique to the problem of conductivity of percolating systems permitted estimation of the corresponding critical indices and prediction of the effective conductivity over the entire interval of concentrations of components at any ratio of their conductivities.

The same approach was used to prove the difference in the scaling behavior of dielectric breakdown voltage and conductivity in the vicinity of the percolation threshold for a model disordered percolation cluster.

REFERENCES

1. B.B. Mandelbrot, *Fractals: Form, Chance, and Dimension*, San Francisco, Freeman, 1977.
2. B.B. Mandelbrot, *The Fractal Geometry of Nature*, San Francisco, Freeman, 1982.
3. S. Kirkpatrick, *Phys. Rev. Lett.*, **27**, 1722 (1971).
4. R. Stinchcombe and B. Watson, *Phys. Rev.*, **C9**, 3221 (1976).
5. L. Niemeyer, L. Pietronero and H. Wiessman, *Phys. Rev. Lett.*, **52**, 1033 (1984).
6. H. Takayasu, *Phys. Rev. Lett.*, **54**, 1099 (1985).
7. K.G. Wilson and J. Kogut, *Phys. Rep., Ser. C*, **12**, 75 (1974).
8. G.N. Dulnev and V.V. Novikov, *Transport Processes in Non-Homogeneous Media*, Leningrad, Energoatomizdat, 1991 (in Russian).
9. T. Shermergor, *Theory of Elasticity of Non-Homogeneous Media*, Moscow, Nauka, 1977 (in Russian).
10. J.M. Ottino and N. Shah, *Chem. Eng. Sci.*, **41**, 283 (1986).