Percolative Shunting on Electrified Surface

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The surface discharge of electrified dielectrics at high humidity is considered. The percolative nature of charge transport in electrets is established. Particular attention is given to the phenomena of adsorption and nucleation of electrically conducting phase in the cause of percolation cluster growth on electrified surface. The critical index of the correlation length for percolation cluster is found, and its value is in good agreement with the known theoretical estimations.

I. INTRODUCTION

The percolation theory is most useful in dealing with charge transport when the local electric conductivity has a large dispersion. Such a situation is inherent in surface discharge of electret under the action of humidity of the ambient atmosphere. It is just the surface discharge which is responsible for electret stability under the real operating conditions for electronic devices, whereas the charge transport through a bulk of dielectric occurs mainly when physical properties of electret materials are studied, especially, using various techniques of thermally stimulated discharge.

The basis for consideration is formed by the percolation model of electret charge relaxation originally proposed in [1]. This model is based on the combined application of percolation theory [2]-[4] and the Kolmogorov model [5] for 2D nucleation kinetics. Such an approach has allowed to get a quantitative description for charge decay on the electrified surface under the action of electrically conducting external agent, primarily, the ambient air humidity.

II. BASIC CONCEPTS OF PERCOLATION APPROACH TO CHARGE RELAXATION IN ELECTRETS

The water adsorption gives rise to the conducting channels on the electrified surface that modifies significantly its electric properties. Since water is much more better electrical conductor than the electret material, these channels shunt the surface of the specimen. Such a physical situation cannot be described correctly in terms of the usual specific surface resistance of the electret material, because the uniformity of surface conductivity is perturbed strongly. When the surface fraction covered by the adsorbate reaches the percolation threshold, the conducting channels will form an infinite percolation cluster along which the excess charge flow off to be subsequently neutralized. At the instant an infinite cluster is arisen, the percolation transition occurs. This cluster shunts the entire surface of the specimen, so all the parts of electrified surface that are connected to it does not contribute to the surface potential of the electret $U$. Thus we can write: $U \propto 1 - P$, where $P$ is the percolation cluster density. The latter quantity fulfills the role of an order parameter in percolation problems, and gives the probability that a randomly located point of the surface belongs to an infinite cluster. The percolation cluster density depends on the fraction of the surface occupied by adsorbate $\theta$ at time $t$: $P = P(\theta)$, where $\theta = \theta(t)$. To find the adsorption kinetics we shall use the Kolmogorov theory [6], which has been developed for a treatment of the nucleus growth in steady-state reaction space of an arbitrary dimensionality. All initial assumptions of this theory are assumed to be satisfied for electret discharge process. For the case of 2D nucleus growth this means that: (i) the reaction space is unbounded, so that the mean area of incipient nuclei is infinitely small compared to the total area of electret surface; (ii) Poisson law holds for the nucleation process, so all the nucleation centers are generated uniformly on the electret surface in a random way at a finite rate of $\alpha = \alpha(t)$ for the expected number of arising centers per unit time and per unit area; (iii) all of the nuclei have the same convex shape so they are geometrically similar; (iv) the growth is uniform so at any stage of the growth process all the nuclei have the same rate of the growth, which is a function of time: $\upsilon = \upsilon(t)$.

Under these assumptions we have the following kinetics for the growth of adsorbed phase for all time after electrization occurred at time $t = t_0$:

$$
\theta(t) = 1 - Q(t_0,t) \exp \left( - \int_{t_0}^{t} d\zeta \alpha(\zeta) S(R_2(\zeta,t)) \right)
$$

(1)
where

\[ Q(\tau_0, t) \equiv \exp\left(- \int_0^{\tau_0} d\zeta \alpha(\zeta) S(R_1(\zeta, \tau_0) + R_2(\tau_0, t)) \right) \]  

(2)

The functional \( R(\zeta, t) \equiv \int_0^t \nu(\eta) d\eta \) gives the radius at the time \( t \) of the nucleus arisen at time \( \zeta \); \( S = S(R) \) is the area of an isolated nucleus of radius \( R \); \( \alpha = \alpha(t) \) is the nucleation rate. The relationship \( R = R(\zeta, t) \) specifies the geometrical features of adsorbate nucleus growth. The nucleus arisen after electrization has the radius \( R = R_2(\zeta, t) \); otherwise, the radius is \( R = R_1(\zeta, t) \) while the nucleus grows before electrization, and \( R = R_1(\zeta, \tau_0) + R(\tau_0, t) \) when its growth is going on thereafter. The function \( Q(\tau_0, t) \), as given by equation (2), describes the growth of all the nuclei arisen before electrization which continue to grow at a later time. The value \( Q(\tau_0, t) \) is equal to the fraction of electret surface remained clear of such nuclei at time \( t \). Over a period of time from the preparation of the future electret material \( (t = 0) \) until the electrization \( (t = \tau_0) \) the interaction with the ambient medium is taking place. After thermodynamic equilibrium has been established, the fraction of the surface occupied by the adsorbate will remain constant provided that the specimen is kept in controlled steady-state conditions. In this case \( Q(\tau_0, t) \) may be replaced by the time-independent function \( Q(\tau_0) \), which gives the fraction of the surface left free of adsorbate before the electrization. A large amount of new nucleation centers are generated in the course of electrization, so it is precisely these ones that greatly reinforce adsorption process, which is activated by the proper electric field of electret itself. A good agreement with the experimental data on the surface potential decay of teflon electrets has been achieved using such a simple representation for the nucleation rate: \( \alpha(t) = \beta \delta(t - \tau_0) + \alpha_0 \), where \( \beta \) is the concentration of nucleation centers appeared in the course of electrization, \( \delta(t) \) is Dirac pulse function, \( \alpha_0 \) is the constant rate of spontaneous nucleation, which is going on over the entire process of electret discharge. Thus, both instantaneous nucleation during electrization and the continuous one during subsequent storage (or, really, operation) of the electret are taken into account.

Thus, upon shifting in time by the interval \( \tau_0 \), the Equation (1) may be simplified as

\[ \theta(t) = 1 - Q(0) \exp\left(- \int_0^t d\zeta \alpha(\zeta) S(R_2(\zeta, t)) \right) \]  

(3)

Now electrization is assumed to be over at time \( t = 0 \) so the discharge is started just thereafter. When the electret surface fills with adsorbate, a percolation cluster forms there. As soon as an infinite cluster appears, a percolation transition takes place so the surface potential of electret starts to decay. There are three different types of percolation transitions. The first one corresponds to the case when infinite cluster forms after a lapse of some time since electrization. Up to this moment the fraction of the surface covered by adsorbate is below the percolation threshold, so the surface potential of the electret remains constant and the greatest stability of the electret charge is thereby achieved. For percolation transition of the second type the infinite cluster arises just by the end of electrization, therefore the surface potential begin to decay at once, with no plateau on its time-dependence curve. In the case of percolation transition of the third type the infinite cluster has already existed during electrization, resulting in a catastrophic discharge of electret (percolation breakdown). The percolation transitions of all the above-mentioned kinds were observed experimentally.

III. SCALING OF SURFACE POTENTIAL NEAR PERCOLATION THRESHOLD

The distinctive feature of percolation cluster growth is that adsorption of conducting phase is activated by electric field: the polar molecules of water are attracted by the additional adsorption centers arising from the charges on electrified surface. Immediately after electrization dielectric is in highly non-equilibrium state, so the relaxation processes are going especially fast. As the electrified surface is getting covered by the network of conducting channels, the further discharge slows down since the appearance of new paths for current flow has been hampered. The curve of electret discharge has a saturation then: the surface potential of electret first falls off steeply, whereupon it stays on some stable level for a long time. Such a saturation of the surface potential occurs when the correlation length of percolation cluster becomes much shorter than the characteristic geometrical size of the electret, so that the backbone network of infinite cluster has sufficiently small cells.

The percolation nature of the surface discharge of electrets is confirmed by the good agreement between the experimental and theoretical values of the critical index of the correlation length. According to the scaling conception \( |4|, |7| - |9| \) the correlation length is the only geometrical size that is intrinsic to percolation cluster near the percolation transition.
threshold. Let us suppose that the correlation length $\xi$ at the saturation of surface potential is proportional to the characteristic size of electret $D$. This allows us to define the surface potential $U_s$ at the saturation point as $U_s = U(t = t_s, \xi \propto D)$, where $t_s$ is the time when saturation is reached. Thus, the correlation length at the saturation point is equal, up to a constant factor $\lambda$, to electret size: $D = \lambda \xi$. Hence it follows that the larger is electret, the greater is its surface potential at saturation. Furthermore, the surface potential is uniquely determined by the percolation cluster density, which is given by the fraction of electret surface covered by adsorbate. So it is possible to find the relationship between $\lambda \xi$ and $\theta$ from experimental data for the discharge of electrets of various sizes. Such data have been obtained for teflon electrets exposed to 98%-humidity at room temperature over a period of three months. Teflon has been chosen as one of the best electret material. The results are shown in Fig. 1.

All the points have been plotted after abscissa conversion from the surface potential at saturation $U_s$ to the difference between the fraction of the surface covered by adsorbate $\theta$ and the percolation threshold $\theta_c$ for this quantity. ($\theta_c = 0.5$ for the case of 2D continual percolation). The initial surface potential after electrization was equal to 300V for all the specimens, whereas its value at saturation has fallen within the range between 30V and 210V according to the electret size.

The correlation length near the percolation threshold obeys the following scaling law [7], [8]:

$$\xi \propto |\theta - \theta_c|^{-\nu}, \quad 0 < \theta - \theta_c << 1$$

(4)

where $\nu$ is the critical index of the correlation length for 2D percolation. Thus, the value of this index can be found from the slope of the scaling curve $\lambda \xi$ vs $(\theta - \theta_c)$ plotted in logarithmic scale. An appropriate procedure is illustrated in Fig. 1. The optimal approximation allows us to get the slope of the scaling asymptote in the region near a percolation threshold, where scaling behavior of Eq. (4) is valid. The critical index of the correlation length estimated in this way was $\nu = 1.4 \pm 0.1$, which is in good agreement with the known theoretical estimates for 2D percolation [4], [7], [10].

Scaling behavior of the surface potential near a percolation threshold is closely related to the peculiarities of adsorbate nucleus growth. In the present work the charge transport on electrified surface has been studied for the different laws of nucleation. The relationship between the local growth rate of conducting phase nuclei and the integral propagation velocity of the potential jump attendant on the formation of conducting channels on the electrified surface has been cleared up. This potential jump travels as fast as the different points of electrified surface are getting electrically connected when adsorption is going on. It has been found that the integral velocity of potential jump propagation $V$ far exceeds the local growth rate of an isolated nucleus $v$: $V = (2n - 1)v$, where $n$ is the number of nucleation centers over a distance $L$ traveled by the potential jump, which can be written as $L(t) = \int_0^t V(\eta) d\eta$. Further, the utmost distance that the potential jump can reach on the electrified teflon surface at high humidity has been measured. The data for the water adsorption on teflon have been analyzed by means of three principal adsorption isotherms: of Brunauer-Emmet-Teller, Langmuir, and Henry. It has been found that the growth rate of
adsorbate nucleus is well approximated by the exponential law: $v(t) = v_0 \exp(-t/\tau)$, where $\tau$ is a relaxation time, and $v_0 \equiv 1/(2\tau \beta^{1/2})$. For exponential relaxation of the growth rate the utmost distance $L(\infty)$ traveled by the potential jump over an infinitely long period of time is finite; whereas if the growth rate was constant or obeyed the hyperbolic decay, that propagation length would be infinitely large. Exponential relaxation of the growth rate has been also verified directly by the fact that experimental data for the utmost distances of potential jump propagation can be linearized in the co-ordinates: $\{\ln(L(\infty) - L(t)) \text{ vs } t\}$. Thus, the found exponential law for the growth rate adequately describes the main features of the charge transport on electrified teflon surface.

IV. CONCLUSION

The charge transport on an electrified surface is of a percolative nature as a result of the adsorption of a conducting phase stimulated by the electric field. The percolation approach has allowed to get a quantitative description of the surface discharge of electrets, so it is possible now to predict the stability of electret devices under the real operating conditions.

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