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The structure of field-induced near-wall charged layers arising in weakly conducting liquids near the surface of solid dielectrics

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ABSTRACT

The present paper investigates space charge formation that occurs near the surface of a solid dielectric in a weakly conducting liquid and takes place under the action of the strong external electric field and due to constant dissociation. The cases of 1D and a more complex 2D macro scale geometries are studied numerically using Comsol Multiphysics software. In the general case, the charge accumulates in a diffuse layer and can get in the bulk in consequence of its transport along the dielectric surface; and there is also a dissociation-recombination layer of ion deficit (same as in the electrohydrodynamic conduction pumping).

1. Introduction

Liquid dielectrics have very small ionic conductivity and are considered to be very weak electrolytes, in which the formation of ions either occurs on the surface of the electrodes due to the injection [1,2] or results from the dissociation in the bulk, including the case of dissociation enhanced by the electric field [3]—the Wien effect. When considering an electrode, a charged double layer [4] can form on its surface and, if there is a strong electric field, the non-equilibrium dissociation-recombination near-wall layer [5–7] of co-ion deficit also forms. The latter changes as the injection current raises [8] until the layer becomes that of injected charge (when the charge polarity reverses). When there are elements made of solid dielectrics in the interelectrode gap, like barriers partially blocking the gap, the charged layers do also form near their surfaces under the action of the electric field. The structure of such layers is not trivial and its specifics is to be studied.

If the conductivity of the solid dielectric is much smaller than that of the liquid, the surface of the former accumulates electric charge that can modify the electric field distribution in the system significantly. Moreover, additional regions of the space charge can form in the bulk near the surface; they undergo the action of the Coulomb force and lead to formation of electrohydrodynamic (EHD) flows or a change in the structure of already existing ones. Thus, since there are EHD systems that include solid dielectrics as essential parts [9–11], the study of the structure of near-wall charged layers at a solid-dielectric surface becomes actual.

There are several processes that happen at and near the interface

between a solid dielectric body and a weakly conducting liquid, which can lead to the formation of the net charge, and need to be described in detail here. First of all, the polarization charge should be commented on. This charge emerges due to difference in polarizabilities of the two media and causes a jump in the normal component of the electric field. Therefore, the distribution of the field strength in the system depends on the polarizability (hence, on the permittivity) of the introduced solid insulator. However, any theoretical or numerical calculations easily account for this charge using the condition of the continuity of the normal component of the electric displacement vector. Most of the present paper discusses the results obtained for the case of equal permittivities but the effect of various permittivity ratios is commented in certain cases.

The near-wall charged layers in the liquid emerge due to the following phenomena.

First, the exchange of charged particles between the media and ion adsorption result in an electrical double layer [4] where its diffuse part is located in the bulk of the liquid near the surface. The double layer modifies the electric field only inside the layer whereas the field remains the same elsewhere in the system. Nevertheless, there are two ways for the double layer to affect an EHD-system: a part of the diffuse layer can be carried away by an external flow (flow electrification [12]) or an electroosmotic flow [13] can emerge as the Coulomb force pushes the charged diffuse layer along the surface. There are many works aimed at studying the double layers; however, these layers are disregarded in the present paper.

Another mechanism of the diffuse layer formation is the ion accumulation near the surface under the action of the external electric field

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that passes through the surface. For simplicity, hereinafter, it is assumed that the solid dielectric is perfect (has zero electrical conductivity) and the conductivity of the liquid is finite. In this case, the electric field lines pass through the surfaces of the solid dielectric while those of the electric current density do not, which make the ions accumulate near the surface and form a diffuse layer that screens the normal component of the electric field in the bulk of the liquid. The ion migration towards the surface is balanced by their diffusion in the opposite direction within the layer; and its size can be smaller than Debye length in liquid dielectrics [6]. Additionally, charge transport along the surface occurs in the presence of the tangential component of the electric field, which can affect both the value of the total current passing through the system and the distribution of the electric field strength. Finally, diffuse layers of this kind can also serve as a source of electroosmotic flows [14] (with such flows considered in micro sizes). When EHD flows are studied on a scale of millimeters or more, the relatively thin diffuse layer is frequently disregarded [9,10] in computer simulation and the charge is instead assumed to be superficial and motionless and to screen the normal component of the electric field completely, which can be done by applying certain boundary conditions on the dielectric surfaces. An approach like that allows reducing computational recourses needed but makes it impossible to account for and to analyze the impact of the near-surface phenomena onto the whole system. In particular, the diffuse layer structure and the charge transport along the surface are disregarded. At the same time, some works like [15] account for the charge accumulation but do not analyze its details and effects directly.

Lastly, high-voltage phenomena can cause the loss of electro-neutrality of weakly conducting liquid near the solid-body surface. They are actively being investigated in the context of electrohydrodynamics and are either the injection or the formation of dissociation-recombination layers of co-ion deficit [8] (these layers are also frequently called heterocharge layers). The former can only take place on electrode surfaces while the latter is shown in the present paper to be also possible near solid dielectrics.

In conformity with the foregoing, the purpose of the present work is to study the structure of the charged layer in a weakly conducting liquid near a solid dielectric surface and to analyze the need in accounting for related phenomena when the electric field strength and the system sizes are typical for electrohydrodynamics. In this paper, theoretical results are presented and analyzed but the main research tool is computer simulation. Section 1 describes the mathematical and computer models and is followed by Section 2 that analyzes the theoretical solution of the governing equations in the 1D stationary approximation. Further, in Section 3, the transient process of charge accumulation is investigated in 1D model with the use of computer simulation. Finally, Section 4 considers a two-dimensional computer model corresponding to the system from Refs. [9,10] where the process of charge transport along the surface is proved to be of importance in the case of low (compared to [9,10]) liquid conductivity. Unlike Sections 3, Section 4 considers steady-state regime; however, the structure of the layers turns out to be the same in these two quite different cases.

2. Mathematical and computer models

The low-voltage conductivity of a liquid dielectric is caused by the presence of ions—charge carriers, mobile in electric field. This work assumes the simplest case when there are only two, positive and negative, ion species that are univalent and have the same mobilities and diffusion coefficients. The free ions appear and disappear due to dissociation of ion pairs and ion recombination, respectively. Though the strong electric field can enhance the dissociation [3], this phenomenon (the Wien effect) was disregarded in the present study for the sake of simplicity. What can happen as a result of the field-enhanced dissociation in the liquid dielectric is discussed in Refs. [9,10], for example. Thus, to describe the electric current passage through the liquid

dielectric, the following set of equations should be used (see the list of notations at the end of the paper):

$$\nabla^2 \varphi = -\rho/(\varepsilon\varepsilon_0) \quad (1)$$

$$\partial n_{\pm}/\partial t + \operatorname{div}(\pm b \vec{E} n_{\pm} - D \nabla n_{\pm}) = W_0 - \alpha_r n_+ n_- \quad (2)$$

These are Poisson's equation (1) and Nernst-Planck equation (2) that have the dissociation and recombination intensities on the right-hand side; and there are the ion flux densities under the divergence:

$$\vec{j}_{\pm} = \pm b \vec{E} n_{\pm} - D \nabla n_{\pm} \quad (3)$$

The indices “+” and “-” refer to positive and negative ions in Eq. (2), and there is also a difference between Eq. (2) for the two ion species in the sign of the first term in ion flux density \vec{j}_{\pm} owing to the ion valencies that are ± 1 . The case of motionless liquid is considered in the present work therefore there is no convective term in Eq. (3).

Equations (1) and (2) should be supplemented with the following relationships:

$$\vec{E} = -\nabla \varphi \quad (4)$$

$$\rho = e_0(n_+ - n_-) \quad (5)$$

$$D = b\varphi_0 \quad (6)$$

$$\varphi_0 = k_B T/e_0 \quad (7)$$

$$W_0 = \alpha_r n_{\text{eq}}^2 \quad (8)$$

$$\alpha_r = 2be_0/(\varepsilon\varepsilon_0) \quad (9)$$

$$n_{\text{eq}} = \sigma_0/(2e_0b) \quad (10)$$

Equations (6) and (7) represent the Einstein relation, and Eq. (9) is the Langevin formula.

The numerical solution of Eqs. (1) and (2) in a conservative form is carried out with the help of the Comsol Multiphysics software that implements the finite element method. In this method, the domain of the computer model is divided into a set of subdomains—elements (that form the mesh); and the solution is sought in the form of a combination of basis functions (polynomials) within each element. In this case, the solution should most accurately correspond to the exact solution of Eqs. (1) and (2), which requires the sizes of the elements to be sufficiently small in the regions where the sought quantities and their derivatives undergo significant spatial variations. This requirement is especially relevant for the element sizes across the near-wall layer; and their sizes were adjusted so that they were as large as possible but their reduction caused no sensible change in the quantities of interest.

Equations (1) and (2) require setting the boundary conditions, while Eq. (2) also requires specifying the initial conditions. The equilibrium values (Eq. (10)) were used as the initial values of concentration in all models. Boundary conditions will be discussed separately before considering the results of each simulation but the surfaces of the electrodes have universal conditions—the ion concentrations have equilibrium values. These conditions result in the electroneutrality of the liquid near the electrode, there is no near-electrode charged layers in the model (but they do exist in reality). However, this has no impact on the processes of primary interest—on those at the surfaces of solid dielectrics placed far from the electrodes.

3. Diffuse layer of accumulated charge

To begin with, it is worthwhile to consider an analytical solution for the diffuse layer. Although calculations of this kind or comments on their results were presented earlier ([4,6,16–18] and others), the layer structure in the case of liquid dielectric and strong electric field remains insufficiently analyzed when represented in terms of the electric field strength rather than the electric potential. It is the electric field strength

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