Simulation of delay reaction-drift-diffusion system applied to charging effects in electron-irradiated dielectrics

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Abstract. The paper is devoted to reaction-convection-diffusion system modeling with time delay and its application for simulation of charging process in dielectrics irradiated by electron bunches. The model of dielectric charging is expressed by mixed initial-boundary value problem for locally instantaneous Poisson equation and time-dependent transport equation as reaction-convection-diffusion equation with time delay. An advanced finite difference scheme was proposed to numerically solve the delay parabolic partial differential equation. Monte-Carlo simulation of electron transport in electron irradiated target was also used to estimate a source function. The special program application was designed using Matlab. The computing experiments were performed to estimate the charge density distribution, potential distribution and electron beam-induced field intensity in electron irradiated ferroelectrics.

1. Introduction
Nowadays “reaction-convection-diffusion” systems have become increasingly relevant for various physical applications. Among numerous models one can specify a class of the systems formalized by partial differential equations (PDE) with time-delay or hereditary effect [1-3]. In particular, functional differential equations can be also referred to delay PDE. When solving applied problems physical sense of delay is often attributed to the finite velocity of distribution of perturbations or the inertial nature of the systems. These systems can not generate a response instantly under external exposure and some time lag can be found. The results of mathematical modelling dynamic systems with time delay have been discussed elsewhere [1-6]. Note also that analytical solution can be obtained for rather limited class of parabolic PDE with delay [4]. Specifically previous studies have been reported the analysis of solution properties such as stability, asymptomaticity, frequency, oscillations et al. In addition, in order to study the model object using computational experiment we need to construct flexible and effective calculation scheme. For this reason numerical methods (especially finite difference methods) play a major role in practice to simulate dynamic “reaction-convection-diffusion” system with time delay [5-6].

The model of dynamic charging process in dielectrics under nonequilibrium conditions can be expressed with use of drift-diffusion approach, which allows one formulates this model mathematically as the system of Poisson equation and time-dependent transport equation [7-10]. The series of the our previous works [9-10] has been devoted to development of mathematical models describing stationary and evolutionary charging processes of ferroelectrics exposed to electron beam irradiation in scanning electron microscope (SEM). Ferroelectrics as a subclass of polar dielectrics are
one of the most promising research objects used in practical applications. SEM enables creating new modes of image formation and modification of the ferroelectric properties. In this case one of the relevant effect of external impact is the charging of ferroelectrics by electrons injected into the sample surface. The great importance of charging effect is due to practical problems of using SEM techniques for formation of controlled submicro- and nanodimensional domain structures in ferroelectrics [11].

Development of mathematical models permits us to simulate the maps of attendant effects, analyze the observed injection, thermal and polarizing processes in an irradiated samples and also predict the conditions of the control polarization switching and domain structure formation. The modification of dynamic charging model has been proposed in view of importance of intrinsic radiation-stimulated conductivity of a sample and also with use of preliminary Monte-Carlo simulation of electron transport in irradiated target [12]. The mathematical model of dynamic charging has been defined as an initial-boundary problem for the system of the PDE, where the basic equation represents by nonlinear reaction-drift-diffusion equation. Considering physical mechanisms which underline diffusive character of charging process we may assume that the delay effect as a reaction to external exposure can be introduced in the corresponding equation of mathematical problem definition. Therefore the current study was designed to develop mathematical model of “reaction-convection (drift)-diffusion” system with time delay and its application for numerical simulation of time-dependent charging processes in polar dielectrics irradiated with SEM.

2. Problem formulation and governing equations

In general case, mathematical model of charging process in dielectrics is defined by locally instantaneous Poisson equation as well as continuity equation. Since high-voltage SEM mode is considered we can use only two independent spatial coordinates: the radius \( r \) and the height \( z \). In addition the internal source has semispherical or semiellipsoid approximation. Therefore we assume that problem is characterized by cylindrical symmetry as it is shown in Fig. 1.

![Figure 1. Geometrical scheme of the irradiated sample.](image)

Further, suppose inductively that the action of focused electron beam starts at the time moment \( t=0 \) perpendicularly to a plane of a sample surface \( z=0 \). After some transformations [9-10] we can express the nonstationary model of charging process taking into account intrinsic radiation-stimulated conductivity of a sample as initial-boundary value problem for the following system of equations:

\[
\begin{align*}
\frac{\partial \rho}{\partial t} &= D \Delta \rho - \mu_n \left( E_r \frac{\partial \rho}{\partial r} + E_z \frac{\partial \rho}{\partial z} \right) - \frac{\mu_n}{\varepsilon \varepsilon_0} \rho^2 + G(r,z), \\
\Delta \phi &= -\frac{\rho}{\varepsilon \varepsilon_0}, \\
E &= -\text{grad} \phi,
\end{align*}
\]

(1)

where \( \rho(r,z,t) \) is charge density distribution, C/m\(^3\); \( 0 \leq r \leq R \), \( 0 \leq z \leq Z \), \( 0 < t \leq T \); \( R, Z \) are geometrical parameters of sample, m; \( E \) is field intensity, V/m; \( \phi \) is potential, V; \( \varepsilon \) is dielectric permittivity; \( \varepsilon_0 \) is dielectric constant, F/m; \( G(r,z) \) is term related to internal source, C/(m\(^3\)-s); \( D = \mu_n kT / e \) is diffusion coefficient, m\(^2\)/s; \( k \) is Boltzmann constant, J/K; \( T \) is temperature, K; \( \mu_n \) is
drift mobility of electrons, m^2/(V-s); e is electron charge, C; \( \Delta = \begin{cases} \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \frac{\partial^2}{\partial z^2}, & \text{if } r \neq 0 \\ 2 \frac{\partial^2}{\partial r^2} + \frac{\partial^2}{\partial z^2}, & \text{if } r = 0 \end{cases} \) is Laplace operator.

The reaction-drift-diffusion equation and Poisson equation need to be complemented by initial conditions at time moment \( t_0 = 0 \) for \( 0 \leq r \leq R, \ 0 \leq z \leq Z \):

\[
\rho(r,z,t_0) = \rho_0(r,z), \quad E(r,z,t_0) = E_0(r,z), \quad \varphi(r,z,t_0) = \varphi_0(r,z)
\]
and boundary conditions corresponding to physical sense for \( t \in (0,T] \):

\[
\left. \frac{\partial \rho}{\partial r} \right|_{r=0, 0 \leq z \leq Z} = 0, \quad \left. \frac{\partial \rho}{\partial z} \right|_{z=0, 0 \leq r \leq R} = 0, \quad \left. \frac{\partial \varphi}{\partial r} \right|_{r=0, 0 \leq z \leq Z} = 0, \quad \left. \frac{\partial \varphi}{\partial z} \right|_{z=0, 0 \leq r \leq R} = 0,
\]

\[
\rho|_{r=R, 0 \leq z \leq Z} = 0, \quad \rho|_{z=Z, 0 \leq r \leq R} = 0, \quad \varphi|_{r=R, 0 \leq z \leq Z} = 0, \quad \varphi|_{z=Z, 0 \leq r \leq R} = 0.
\]

Furthermore, let us suppose that changes in gradient of the injected charges density lead to following effect. The system responds by changes in diffusion current component not instantly, but with some time lag \( t^* \). Using the concept of “single-phase-lag” model \([13-14]\) the expression of diffusion current density \( J_{diff} \) can be modified taking into account time delay effect:

\[
J_{diff}(r,z,t^*) = -D \cdot \nabla \rho(r,z,t).
\]

Here the latter is transformed to generalized relation of diffusion current density with respect to time moment \( t \). Whence we specify the contribution of diffusion component into current density change at instant time moment as well as at time moment “in the past”:

\[
J_{diff}(r,z,t) = -D \cdot \nabla \rho(r,z,t) - \bar{D} \cdot \nabla \rho(r,z,t-t^*),
\]

where \( \bar{D} = \gamma \cdot D \) is corresponding diffusion coefficient and \( 0 \leq \gamma \leq 1 \) is constant of delay.

This allow us to write the expression for divergence of diffusion current density in consideration of the time delay \( t^* \). The continuity equation is given by following equation as superposition of diffusion current and drift current contributions respectively:

\[
\frac{\partial \rho(r,z,t)}{\partial t} = D \Delta \rho(r,z,t) + \bar{D} \Delta \rho(r,z,t-t^*) - \frac{\mu_e}{\varepsilon_0} \rho^2(r,z,t) - \mu_n \left( \frac{\partial \rho(r,z,t)}{\partial r} + E_z \frac{\partial \rho(r,z,t)}{\partial z} \right) + G(r,z).
\]

Here initial condition (2) is modified using prehistory of process:

\[
\rho(r,z,t) = \rho_0(r,z,t) \text{ for } -t^* \leq t \leq 0, \ 0 \leq r \leq R, \ 0 \leq z \leq Z.
\]

This implies that problem definition is given by continuity equation (6), the Poisson equation, the expression for field intensity in system (1), prehistory (7) and boundary conditions (3)-(4).

3. Numerical method

The equation (6) represents the so-called “reaction-convection-diffusion” equation. This equation is defined as parabolic PDE with constant time delay and it can be solved by finite difference method \([5-6]\). Note that the relevant issue of numerical solving such equation is approximation of convection term. In order to solve the reaction-convection-diffusion equation we proposed the modification of computational splitting scheme based on alternating direction method of Peacan-Rachford \([15]\) for approximation of “diffusion” part of equation. Also the Roberts-Weiss scheme \([16]\) was used for approximation of “convection” part. In order to approximate the “reaction” part of PDE the “frozen coefficient” method was also applied. Let us consider space-time grid with steps \( h_1, h_2, \tau \) related to...
variables \( r, z, t \) respectively: \( \omega_{i,j}^k = \{ t_i = h_t, i = 1, N, z_j = jh_z, j = 1, M, t^k = k\tau, k = 1, 2, \ldots \} \). The computational subshemes on \( k + 1/2 \) and \( k + 1 \) time layers can be written as:

\[
\rho_{y,j}^{k+1/2} - \rho_{y,j}^k = \frac{1}{2} G_{y,j}^{k+1/2} - \frac{\mu_m}{2\varepsilon_0} \left( \rho_{y,j}^{k+1/2} - \rho_{y,j}^k \right)^2 - \frac{\mu_m}{2} \left( E_y \frac{\rho_{y,j}^{k+1/2} - \rho_{y,j+1}^k + \rho_{y,j-1}^k - \rho_y^j}{2h_t} + E_z \frac{\rho_{y+1,j}^{k+1/2} - \rho_{y-1,j}^k}{2h_z} \right) + \frac{D}{2} \left( \frac{\rho_{y+1,j}^{k+1/2} - 2\rho_{y,j}^{k+1/2} + \rho_{y-1,j}^{k+1/2}}{h_z^2} + \frac{1}{2h_t} \frac{\rho_{y+1,j}^{k+1/2} - \rho_{y-1,j}^{k+1/2} + \rho_{y,j}^{k+1/2} - 2\rho_{y,j}^k + \rho_{y,j-1}^k}{h_z^2} \right),
\]

\[ \tag{8} \]

\[
\rho_{y,j}^{k+1} - \rho_{y,j}^{k+1/2} = \frac{1}{2} G_{y,j}^{k+1} - \frac{\mu_m}{2\varepsilon_0} \left( \rho_{y,j}^{k+1} - \rho_{y,j}^{k+1/2} \right)^2 - \frac{\mu_m}{2} \left( E_y \frac{\rho_{y,j}^{k+1} - \rho_{y,j+1}^{k+1/2} + \rho_{y,j-1}^{k+1/2} - \rho_{y,j}^{k+1/2}}{2h_t} + E_z \frac{\rho_{y+1,j}^{k+1} - \rho_{y-1,j}^{k+1/2} + \rho_{y,j}^{k+1/2} - 2\rho_{y,j}^{k+1/2} + \rho_{y,j-1}^{k+1/2}}{2h_z} \right) + \frac{D}{2} \left( \frac{\rho_{y+1,j}^{k+1} - 2\rho_{y,j}^{k+1} + \rho_{y-1,j}^{k+1}}{h_z^2} + \frac{1}{2h_t} \frac{\rho_{y+1,j}^{k+1} - \rho_{y-1,j}^{k+1} + \rho_{y,j}^{k+1} - 2\rho_{y,j}^{k+1/2} + \rho_{y,j-1}^{k+1/2}}{h_z^2} \right),
\]

\[ \tag{9} \]

Subschemes (8) and (9) are completed by approximations of Derichlet boundary conditions: \( \rho_{N,j}^{k+1/2} = 0, \rho_{L,j}^{k+1/2} = 0 \) and \( \rho_{N,j}^{k+1} = 0, \rho_{L,j}^{k+1} = 0 \). Neumann boundary conditions are automatically included in the modified differential equations with use of additional layers on coordinates with fictitious points and approximations: \( \frac{\rho_{0,j}^{k+1/2} - \rho_{2,j}^{k+1/2}}{2h_t} = 0 \) and \( \frac{\rho_{i,0}^{k+1} - \rho_{i,2}^{k+1}}{2h_z} = 0 \). The constructed numerical scheme was modified taking into account delay effect. According to approximation for diffusion component with time delay the subschemes (8) and (9) are complemented by expression:

\[
\frac{D}{2} \left( \frac{\rho_{i+1,j}^{k,q} - 2\rho_{i,j}^{k,q} + \rho_{i-1,j}^{k,q}}{h_z^2} + \frac{1}{2h_t} \frac{\rho_{i+1,j}^{k,q} - \rho_{i-1,j}^{k,q} + \rho_{i,j}^{k,q} - 2\rho_{i,j}^{k,q} + \rho_{i,j-1}^{k,q}}{h_z^2} \right),
\]

where \( q = t^* / \tau \) is time lag index. Note that numerical simulation of delay system requires controlling memory-stored structure of “window of delay” (values of variables on each time layer included to delay period). This aspect leads to resource-intensive computational process. The obtained systems are solved by sweep method on each time layer. The Poisson equation on each time layer is solved using alternating direction method (relaxation scheme). The procedure of approximation analysis results in second order accuracy for space and time variables \( O(\tau^2 + h_t^2 + h_z^2) \). The stability analysis by means of maximum principle suggests the absolute stability of the scheme. Also, the constructed scheme is monotone.

The absolute value of field intensity is evaluated using field components \( (E_r, E_z) \). In addition, Monte-Carlo simulation of electron trajectories in irradiated sample allows us to specify the initial charge distribution \( \rho_0(r,z,t) \) (for the model of relaxition charging process) and the source function \( G(r,z) \) (for the problem of dynamic charge accumulation).

4. Simulation results and discussion
The special program application developed with Matlab package was designed to simulate dynamic electron beam-stimulated charging processes in dielectrics. Three modes of charging process were implemented in the program: the stationary mode, the dynamic mode of charge relaxation and the dynamic mode of charge accumulation. The relation of deposits of “drift” (convection) and “diffusion” parts in the model for dynamic modes can be evaluated due to analysis of value of Peclet number \( Pe = \nu_d \cdot L / D \), where \( \nu_d = \mu_r E \) is the drift velocity in m/s and \( L \) is characteristic scale of distance, for instance, linear size of a gradient zone in m. Note that \( Pe << 1 \) and diffusion dominates in the mode of charge accumulation. Whereas \( Pe >> 1 \) and drift dominates in the mode of charge relaxation. Using the analog of Fourier number \( Fo = D \cdot \tau / L^2 \) enables the scales ratio of time and distance to be specified. The computing experiments were performed by the example of promising ferroelectric materials lithium niobate (LiNbO_3) and lithium tantalate (LiTaO_3).
The mode of charge relaxation. The Fig. 2 demonstrates the simulation results for spatial distribution of potential and field intensity distribution induced by the injected charges in LiTaO₃ in the mode of charge relaxation at time of observation t=1 ps (calculation parameters are h₁=0.25 µm, τ=0.5·h₁ µs at start beam energy £₀=25 keV and surface charge density σsurf=10 C/m²). The estimation of Peclet number gives 5·10⁵ (more than unity), which means drift dominates and we have weak diffusion. The computing experiment allows us to observe charge relaxation dynamics and identify a relaxation time in this mode. Here the relaxation time is going to be more than 100 ps.

![Figure 2](image1.png)

**Figure 2.** The simulation results: potential distribution – (a); distribution of field intensity absolute value and its vector field of (in the insert figure) – (b).

Mode of charge accumulation dynamics. The Fig. 3 shows the computational results for charging characteristics of LiNbO₃ taking into account delay effect.

![Figure 3](image2.png)

**Figure 3.** The absolute value profile of field intensity at the fixed time moments (curves correspond to the modes: 1, 2, 3 – without delay, 1’, 2’, 3’ – with delay, time lag is t*=4 ns) – (a); changes in potential minimum value at various time lag: 1 – t*=20 ns, 2 – t*=2 ns, 3 – without delay – (b).

The computing parameters are h₁=0.25 µm, τ=1 ns, γ=1. Calculated data are displayed in comparison with simulation data provided nonregistering delay at start beam energy £₀=10 keV, irradiation time 10 s and surface charge density σsurf=20 C/m². In this mode Peclet number equals to 0.04 (less than unity). This means that diffusion process dominates. The computing experiment indicates significant difference in values of estimated characteristics both of maximum value level and value at a point of source impact in a gradient zone.
The mode of stationary charging. The estimation of characteristic value of dynamic time by Fourier criterion yields 50 µs. The exposure time is approximately changes in the range from 1 s to 10 s when SEM is used to control polarization switching in ferroelectrics. This allows us to conclude that stationary mode is realized. The analysis of simulation data suggests that field intensity in nonirradiated part of a sample exceeds the coercive field (2.1-10^7 V/m for LiTaO₃). This can result to polarization switching in oppositely orientated domains. Provided start beam energy E₀ equals to 15 keV and surface charge density σsurf equals to 5 C/m² the maximum value of field intensity is 2.1-10⁹ V/m at the distance of 0.05 µm from the source and it corresponds to 3-10⁷ V/m at the distance of 5 µm. At the same time the use of σsurf=1 C/m² does not allow switching process to be stimulated in nonirradiated part of crystal at different values of start beam energy. Also the use of reduced accelerating voltages (~5 kV) can not create the condition for polarization switching when the surface charge density varies over the range 5-20 C/m². This confirms that surface charge density and accelerating voltages require to be optimally controlled.

5. Conclusions
The reaction-drift-diffusion model with time delay was proposed to simulate dynamic charging process in ferroelectric electron beam-irradiated with the SEM. An advanced splitting scheme was constructed to solve the delay parabolic PDE numerically. The analysis indicates the second order accuracy for space and time variables, monotony and absolute stability of the scheme. The main peculiarity of delay system numerical simulation is necessity of memory-stored control for delay “window” structure. This aspect leads to resource-intensive computational process. The relation of “drift” and “diffusion” deposits for dynamic modes of the model was also discussed using analysis of value of Peclet number. The computing experiments by the example of LiNbO₃ and LiTaO₃ ferroelectrics were performed to estimate the charging characteristics in the stationary mode, the dynamic mode of charge relaxation and the dynamic mode of charge accumulation.

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