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Numerical simulation of electron beam-induced dielectric charging using advanced computational scheme for solving semilinear reaction-diffusion equation *

Anna Pavelchuk[1] , Anna Maslovskaya[1†]

Department of Mathematics and Computer Sciences, Amur State University, 21 Ignatyevskoe Shosse, Blagoveshchensk 675000, Russia

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Abstract. This paper presents the numerical simulation of characteristics of dynamic charging process in dielectrics irradiated by electron beam at diagnostics and modification with use of scanning electron microscope techniques. The mathematical model involves locally instantaneous Poisson equation and time-dependent transport equation assuming drift-diffusion transport mechanism. An advanced numerical scheme based on implicit alternating direction method is suggested for solving semilinear reaction-diffusion-type equation in several space dimensions with acceptable accuracy. Monte-Carlo simulation of electron transport in irradiated target is also included into total hybrid computational scheme to estimate a source function. The special program application developed with Matlab package is proposed to simulate dynamic charging processes electron beam-stimulated in polar dielectrics. The computing experiment by the example of ferroelectric TGS crystal is performed to estimate the charge density distribution, potential distribution as well as field intensity electron beam-induced.

Keywords: simulation, charging process of dielectrics, semilinear reaction-diffusion equation, implicit alternating direction method, computing experiment

1 Introduction

Scanning electron microscopy techniques are actively used in coming years as methods of analysis and treatment of different functional materials. The electron probe of scanning electron microscope (SEM) affects as a source producing ionizing, electric and thermal action on the sample. The investigation of polar dielectrics with SEM-techniques requires possessing verified information concerning attendant effects, which arise in materials under the electron beam exposure. One of the constitutive effect of electron beam irradiation consists in charging dielectrics due to electron bombardment of dielectric target[2]. Numerous studies have shown that the charging-up effect occurs at any magnifications and any actual values of probe current[2, 5]. Generally, the dielectric charging phenomena have been considered as negative effect, which needs to be prevented. As an example, low-accelerating voltages have been used to visualize ferroelectric domain structure with SEM[13]. On the contrary, the methods of SEM-direct polarization switching in ferroelectrics based on charging effect have been reported elsewhere[7, 11]. Methods of mathematical and computer simulation can be successfully applied to describe electron beam-induced charging process in dielectrics. As well-known Monte-Carlo method is widespread approach for electron transport simulation in irradiated target[1, 8]. In addition, deterministic models of charging process in dielectrics are extensively developed in recent years[3, 9, 12, 14, 15]. We have previously suggested stationary model[12] as well as nonstationary model[14] describing charging phenomena in polar dielectrics taking into account intrinsic radiation-induced conductivity as well as charge distribution calculated

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† Corresponding author. *E-mail address*: maslovskayaag@mail.ru

by Monte-Carlo method. The solutions of the problems have been based on numerical net point methods such as finite difference and finite element methods. In [14] simple explicit fractional step splitting method has been employed to integrate semilinear evolution partial differential equation using a very small time step to keep numerical stability and accuracy. This makes computation very slow. In the same time, this approach allows us to simulate charge relaxation process, but not dynamics of charging. More in-depth analysis of numerical experiments requires computational scheme to be improved. Numerous studies have been devoted to design effective numerical method for solving fully nonlinear[16] as well as semilinear[6] parabolic partial differential equation (PDE). Actually, the use of fractional step-splitting methods[10, 17] allows us to construct stable computational scheme without losing accuracy.

The present study was undertaken to provide the advanced computational scheme for semilinear reaction-diffusion PDE to simulate charging processes in polar dielectric materials arising under electron beam exposure. To specify the problem statement it should be mentioned that charging effects induced by electron beam irradiation in polar dielectrics would be under consideration. The used basic technique was finite difference method. In particular, to solve the problem we employed implicit alternating direction method.

2 Mathematical model

2.1 Governing equations

Let us consider the well-known PDE system, which includes continuity equation and locally instantaneous Poisson equation. This system models charging phenomena in dielectric materials:

$$\begin{cases} \frac{\partial \rho}{\partial t} = G - div\mathbf{j}, \\ div\mathbf{E} = \frac{\rho}{\varepsilon\varepsilon_0}, \end{cases} \tag{1}$$

where ρ is charge density distribution, C/m^3; $\mathbf{j} = \sigma\mathbf{E}$ is current density, A/m^2; E is field intensity, V/m; ε is dielectric permittivity of the material; ε_0 is dielectric constant, $C/(V \cdot m^2)$; $G-$ generation component, $C/(m^3 \cdot s)$.

We assumed that the intrinsic radiation-induced conductivity occurs in the irradiated polar dielectric. So long as diffusive current reduces conduction current, the current density can be given as sum of diffusion $\mathbf{j}^{dif}$ and drift current $\mathbf{j}^{dif}$ components:

$$\mathbf{j} = \mathbf{j}^{dif} + \mathbf{j}^{dr}, \mathbf{j}^{dr} = \sigma\mathbf{E}, \mathbf{j}^{dif} = -Dgrad\rho \tag{2}$$

where $\sigma = \mu_n\rho$ is intrinsic radiation-induced conductivity, $C/(V \cdot s \cdot m)$; $D = \mu_n kT/e$ is diffusion coefficient, m^2/s; k is Boltzmann constant, J/K; T is temperature, K; $\mu_n = \mathbf{v}_{dr}/\mathbf{E}$ or $\mu_n = e \cdot \bar{l}/\left(m \cdot \bar{v}\right)$ is drift mobility of electrons, $m^2/(V \cdot s)$; $\mathbf{v}_{dr}$ is drift velocity of electron, m/s; $\bar{l}$ is average free path, m; $\bar{v}$ is average thermal velocity, m/s; m is electron mass, kg; e is electron charge, C.

The Fig. 1 shows the geometry of model sample as well as primary beam position.

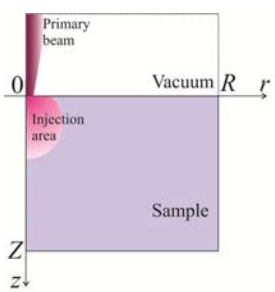


Fig. 1: General geometrical scheme of the problem

The final system of equations was modified in terms of cylindrical symmetry of the problem. To be definite, assume that the action of focused electron beam starts at the time moment $t = 0$ perpendicularly to a plane of a sample surface $z = 0$. Also it is necessary to take into account, that $div\mathbf{j}$ can be specified as follows $\sigma div\mathbf{E} + \mathbf{E}grad\sigma - D\Delta\rho$.

Whence the several-dimensional nonstationary problem of charging process simulation can be expressed as initial-boundary problem for PDE system:

$$\begin{array}{l} \frac{\partial\rho}{\partial t} = G\left(r,z\right) - \frac{\mu_n}{\varepsilon\varepsilon_0}\cdot\rho^2 - \mu_n\cdot\left(E_r\frac{\partial\rho}{\partial r} + E_z\frac{\partial\rho}{\partial z}\right) + D\left(\frac{\partial^2\rho}{\partial r^2} + \frac{1}{r}\frac{\partial\rho}{\partial r} + \frac{\partial^2\rho}{\partial z^2}\right), \\ \frac{\partial^2\phi}{\partial r^2} + \frac{1}{r}\frac{\partial\phi}{\partial r} + \frac{\partial^2\phi}{\partial z^2} = -\frac{\rho}{\varepsilon\varepsilon_0}, \\ \mathbf{E} = -grad\phi, \quad 0 \le r \le R, \quad 0 \le z \le Z, \quad 0 \le t \le T. \end{array} \tag{3}$$

The addition of initial $\rho\left(r,z,t_0\right) = \rho^0$ and boundary conditions:

$$\left.\frac{\partial\rho}{\partial r}\right|_{r=0} = 0, \quad \left.\frac{\partial\rho}{\partial z}\right|_{z=0} = 0, \quad \left.\frac{\partial\phi}{\partial r}\right|_{r=0} = 0, \quad \left.\frac{\partial\phi}{\partial z}\right|_{z=0} = 0 \tag{4}$$

$$\left.\rho\right|_{r=R} = 0, \quad \left.\rho\right|_{z=Z} = 0, \quad \left.\phi\right|_{r=R} = 0, \quad \left.\phi\right|_{z=Z} = 0 \tag{5}$$

allows us to obtain closed-form expression of the problem (3).

2.2 Outline of computational scheme

In order to evaluate electron penetration depth as well as distribution of the electron energy losses we performed the simulation of electron trajectories with use of Monte-Carlo method. Energy distribution in the sample volume was approximated by Gaussian function:

$$I = I_0\cdot\exp\left(-\frac{\left(\bar{r}-\delta_1\right)^2}{2\cdot\delta_2^2}\right) \tag{6}$$

$$\bar{r} = \sqrt{r^2+z^2} \tag{7}$$

where I_0 is normalization constant; δ_1 and δ_2 are parameters numerically calculated in m. The latter result can be included into the problem (3)-(5) as initial charge density distribution ρ^0 (when analyzing process of charge relaxation) or as generation component G (when analyzing accumulating charging process) with corresponding normalized dimensional parameter I_0.

The solution of the problem (3)-(5) is based on numerical methods designed for PDE. Notice that first equation (continuity equation) of the system (3) can be specified as semilinear reaction-diffusion equation. To construct the algorithm for solving continuity equation we used implicit alternating direction method. Let us consider space-time grid with steps h_1, h_2, τ related to variables r, z, t respectively:

$$\omega^{\tau}_{h_1,h_2} = \left\{r_i = ih_1, \quad i = \overline{0,n}; \quad z_j = jh_2, \quad j = \overline{0,m}; \quad t^k = k\tau, \quad k = 0,1,2,\cdots\right\}.$$

Hence a typical point in the domain is given by $\left(r_i, z_j, t^k\right)$. An implicit alternating direction scheme for continuity equation gives the following discretized system of equations:

$$\begin{array}{l} \frac{\rho_{ij}^{k+1/2}-\rho_{ij}^{k}}{\tau} = \frac{1}{2}G_{ij}^{k+1/2}\left(r_i,z_j\right) - \frac{1}{2}\mu_n\cdot\left(E_r\frac{\rho_{i+1j}^{k+1/2}-\rho_{i-1j}^{k+1/2}}{2h_1} + E_z\frac{\rho_{ij+1}^{k}-\rho_{ij-1}^{k}}{2h_2}\right) \\ \quad + \frac{1}{2}D\left(\frac{\rho_{i+1j}^{k+1/2}-2\rho_{ij}^{k+1/2}+\rho_{i-1j}^{k+1/2}}{h_1^2} + \frac{1}{r_i}\frac{\rho_{i+1j}^{k+1/2}-\rho_{i-1j}^{k+1/2}}{2h_1} + \frac{\rho_{ij+1}^{k}-2\rho_{ij}^{k}+\rho_{ij-1}^{k}}{h_2^2}\right), \end{array} \tag{8}$$

$$\begin{array}{l} \frac{\rho_{ij}^{k+1}-\rho_{ij}^{k+1/2}}{\tau} = \frac{1}{2}G^{k+1/2}\left(r_i,z_j\right) - \frac{\mu_n}{\varepsilon\varepsilon_0}\cdot\rho_{ij}^{k+1/2}\cdot\rho_{ij}^{k+1} \\ \quad - \frac{1}{2}\mu_n\cdot\left(E_r\frac{\rho_{i+1j}^{k+1/2}-\rho_{i-1j}^{k+1/2}}{2h_1} + E_z\frac{\rho_{ij+1}^{k+1}-\rho_{ij-1}^{k+1}}{2h_2}\right) \\ \quad + \frac{1}{2}D\left(\frac{\rho_{i+1j}^{k+1/2}-2\rho_{ij}^{k+1/2}+\rho_{i-1j}^{k+1/2}}{h_1^2} + \frac{1}{r_i}\frac{\rho_{i+1j}^{k+1/2}-\rho_{i-1j}^{k+1/2}}{2h_1} + \frac{\rho_{ij+1}^{k+1}-2\rho_{ij}^{k+1}+\rho_{ij-1}^{k+1}}{h_2^2}\right). \end{array} \tag{9}$$

Note, that Eqs. (8) and (9) are specified for internal nodes of computational grid. Then initial condition is also discretized:

$$\rho\left(r_i, z_j, t_0\right) = \rho_{i,j}^0.$$

The first-order derivative for Neumann boundary condition has second order approximation with $\mathrm{O}\left(h_1^2\right)$ accuracy. This implies that we have for boundary conditions at the first splitting step

$$\frac{\rho_{1,j}^{k+1/2} - \rho_{-1,j}^{k+1/2}}{2h_1} = 0, \quad \rho_{n,j}^{k+1/2} = 0, \quad \rho_{i,m}^{k+1/2} = 0. \tag{10}$$

The former condition includes fictitious nodes, which can be taken into account when constructing approximation for $\rho_{0,j}$, $j = \overline{0, m-1}$ with the general subschema (8).

Also for the second splitting step we can obtain

$$\frac{\rho_{i,1}^{k+1} - \rho_{i,-1}^{k+1}}{2h_2} = 0, \quad \rho_{i,m}^{k+1} = 0, \quad \rho_{m,j}^{k+1} = 0 \tag{11}$$

where the Neumann boundary condition can be similarly used for $\rho_{i,0}$ approximation, $i = \overline{0, n-1}$, with the general subschema (9).

The total systems on the each time layer $k + 1/2$ and $k + 1$ were solved numerically by marching method. Analyzing coefficients we can show that the condition of a marching method correctness is satisfied. The convergence control is due to approximation as well as stability analysis. The conventional procedure of approximation analys[16] results in $\mathrm{O}\left(h_1^2, h_2^2, \tau^2\right)$ accuracy for the scheme (8)-(11). In order to explore stability of implicit scheme (6)-(10) we used harmonic analysis method[8]. According to this approach solution is expressed as

$$\rho_{ij}^k = \alpha_k \exp\left[i\left(\lambda_n r_i + \lambda_m z_j\right)\right].$$

As commonly known the necessary condition for stability of differential problem with respect to initial data is given by the following equation:

$$\left|\frac{\alpha_{k+1}}{\alpha_k}\right| < 1. \tag{12}$$

As the variable $|\alpha_{k+1}/\alpha_k| = |\alpha|$ generally can be complex one can use the $\left|\alpha^2\right|$. In this case, the condition (12) will stay correct. Then after some transformations we can obtain

$$\left|\frac{\alpha_{k+1/2}}{\alpha_k}\right| = \left|\frac{1 - \sigma_2 P_2 - \gamma_3 2i \sin\left(\lambda_m h_2\right)}{1 + \sigma_1 P_1 + \gamma_1 2i \sin\left(\lambda_n h_1\right) - \gamma_2 2i \sin\left(\lambda_n h_1\right)}\right|$$

where $P_1 = 4\sin^2\left(\frac{\lambda_n h_1}{2}\right)$, $P_2 = 4\sin^2\left(\frac{\lambda_m h_2}{2}\right)$, $\sigma_1 = \frac{\tau}{2}\frac{D}{h_1^2}$, $\sigma_2 = \frac{\tau}{2}\frac{D}{h_2^2}$, $\gamma_1 = \frac{\mu_n E_r}{4h_1}$, $\gamma_2 = \frac{\tau D}{4r_i h_1}$ and $\gamma_3 = \frac{\mu_n E_z}{4h_2}$.

To analyze the second subschema (9) one needs to cast out term approximated generation component $G\left(r, z\right)$, which does not impact on stability of the scheme. Besides, frozen coefficients method[4] allows us to analyze scheme stability for equation with nonlinear component. Note that frozen coefficients method for Neumann stability analysis is based on more heuristic than rigorous approach. Nonlinear term is specified as $k\rho^2 = (k\rho)\cdot\rho$, where $k = \mu_n/(\varepsilon\varepsilon_0)$, where expression in the brackets plays the role of a constant C depending on variables r, z, t. The constant C is calculated explicitly from the first fractional step $\left(k\cdot\rho^{k+1/2}\right)\cdot\rho^{k+1}$. So that we can similarly obtain

$$\left|\frac{\alpha_{k+1}}{\alpha_{k+1/2}}\right| = \left|\frac{1 - \sigma_1 P_1 + \left(\gamma_2 - \gamma_1\right) 2i \sin\left(\lambda_n h_1\right)}{1 + \sigma_2 P_2 + C\tau + \gamma_3 2i \sin\left(\lambda_m h_2\right)}\right|.$$

In fact, we have that the harmonic amplitude ratio (12) is satisfied so long as the condition take place:

$$\left|\frac{\alpha_{k+1}^2}{\alpha_k^2}\right| < 1.$$

It is obvious that as constant C is limited and positive definite, the latter equation will be true permanently. This means that implicit alternating direction scheme (8)-(9) is absolutely stable. Stability and approximations are followed by convergence of the constructed numerical scheme.

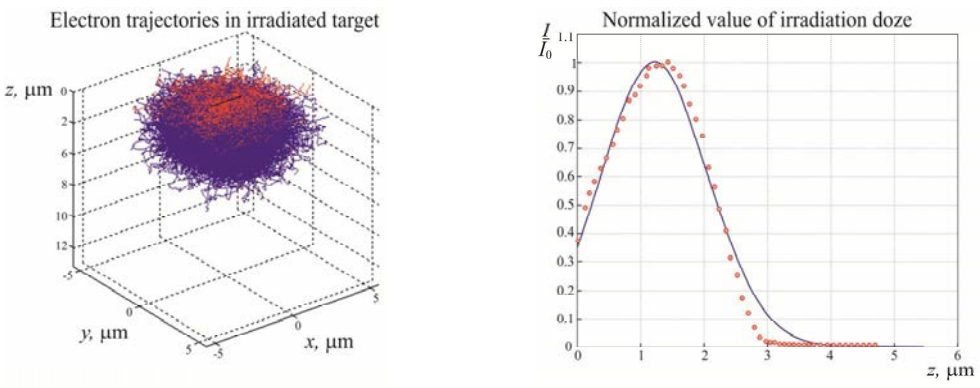
The Poison equation was solved by an iterative finite difference method. Specifically the implicit alternating direction method was used. This permits one to obtain a system of equations numerically solved. The formula $\mathbf{E} = -grad\phi$ in the system (3) was also used to estimate the vector and absolute value of field intensity $|\mathbf{E}| = \sqrt{(E_r)^2 + (E_z)^2}$ as well as electron beam-induced polarization component $\mathbf{P} = (\varepsilon - 1)\,\varepsilon_0\mathbf{E}$ on each time layer.

3 Numerical experiment and simulation results

The proposed calculation scheme was realized as program application "SIMULATION OF CHARGING PROCESSES IN POLAR DIELECTRICS" with use of Matlab programming. For example, we performed a numerical experiment to demonstrate results of charging characteristics computing for a typical ferroelectric crystal such as triglycine sulfate TGS $((NH_2CH_2COOH)_3 \cdot H_2SO_4)$.

Let us consider, that sample is irradiated by electron beam of the SEM with average energies (for instance accelerating voltage U is set to be 15 kV, start energy of electrons E_0 is respectively equal to 15 keV). The sample is a TGS disk of Z=1 mm thick and $R = 2.5$ mm in radius. The simulation parameters are defined as follows: irradiated area $S = 1.54 \cdot 10^{-10} m^2$, probe current $I = 1$ nA, surface density of injected charges $\sigma_{surf} = 0.15 C/m^2$. The dielectric permittivity ε equals 50 and diffusion coefficient D is estimated to be $0.01547 cm^2/s$ for TGS crystal.

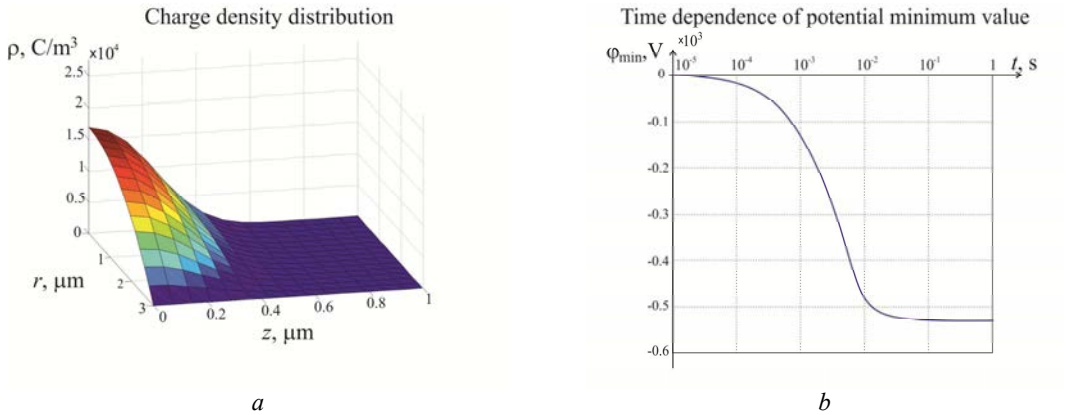
The result of simulation of electron trajectories in irradiated TGS is demonstrated in the Fig. 2a. The electron transport was computed by means of Monte-Carlo method. According to this procedure one needs to determine the sample geometrical size, the type of irradiated target (element composition), number of electrons N and the initial parameters of experimental observation (start energy of electrons E_0). The Fig. 2 b shows normalized accumulated irradiation doze distribution. Circular markers are mean values estimated by Monte-Carlo simulation and the solid line is approximation obtained with Eq. 6 by least-squares fitting procedure. Further let us assume half-elliptic approximation of the internal source. Parameters δ_1 and δ_2 were estimated to be $1.211 \mu m$ and $0.832 \mu m$ respectively.



a *b*

Fig. 2: The 3D-simulated electron trajectories $-a$, the accumulated irradiation dose in normalized units $-b$ in TGS (at start beam energy $E_0 = 15keV$, number of electrons $N = 5000$, blue trajectories correspond to electrons injected into the sample, whereas red trajectories indicate backscattering electrons)

In order to simulate charging process in polar dielectric we suppose, that initial value of charge density equals zero and component G gives accumulation of charge with time. This component was also defined using formula (6). In this case normalization constant G_0 was going to be $3.68 \cdot 10^4 C/(m^3 \cdot s)$. Fig. 3 a displays the simulation result of spatial charge density distribution caused by accumulated electrons in TGS at time moment t^*. Also absolutely stable computational scheme (8)-(11) allows performing calculations with sufficiently great time step. For this reason, we can simulate not only the potential distribution on each time layer, but also observe potential dynamics during irradiation time. The calculation result of time dependent variation of potential minimum value is shown in Fig. 3 b.



a *b*

Fig. 3: The spatial distribution of charge density at time moment $t^* = 5\ ms - a$; time dependent variation of potential minimum value - b

Here we can conclude that the potential distribution reaches the stationary state over the time period $\Delta t \sim [0, 0.1]$ s at fixed above parameters. In addition, we can visualize vector fields and absolute values of the field intensity and the polarization electron beam-induced.

This result indicates that electron injection into the ferroelectrics can lead to accumulation of charges, which causes electric fields arising in irradiated and unirradiated parts of the sample. Such phenomena can induce polarization reversal processes in ferroelectrics experimentally observed. The values of field intensity in the injection area is approximately evaluated to be $2.45 \cdot 10^7 V/m$, which is attributed to depolarizing fields for TGS crystals. At the same time the field intensity in unirradiated part of sample stays rather significant $|E|_r \approx 10^6 V/m$, which is more than coercive field $E_c = 2 \cdot 10^4 V/m$ for this material.

4 Conclusion

Thus, the paper presents a simulation approach to specify the dynamic characteristics of charging process in polar dielectrics irradiated by electron bunches of average energies when analyzing and modifying materials with the SEM techniques. The study shows that mathematical statement of the model can be successfully described by the hybrid computational scheme, which includes Monte-Carlo computations of electron dose distribution, as well as PDE system involving continuity equation and Poisson equation. The present study documents numerical solutions of continuity equation and Poisson equation based on finite difference method. Specifically implicit alternating direction method was used providing $O(h^2 + \tau^2)$ approximation order, verified absolute stability and, as a consequence, convergence of constructed numerical scheme.

We proposed the special program application developed with Matlab programming to completely simulate dynamic charging processes electron beam-stimulated in polar dielectrics at variation modelling parameters: start beam energy, beam diameter, number of calculated electrons, probe current, irradiation time, geometrical and physical parameters of the irradiated sample. The output data, which we can observe at given experimental

parameters, were the follows: charge density distribution, potential distribution, field intensity and polarization electron beam-induced.

So long as ferroelectrics are great promise materials for microelectronic devices and SEM modes can be applied to modify the polar structure of these crystals the results of numerical experiments were demonstrated by the example of typical ferroelectric crystal TGS. The Monte-Carlo simulation of electron trajectories allowed us to estimate the form, geometrical size of electron distribution injected into the sample and as a result, parameters of internal source function. It was found that potential distribution reaches the stationary state over the time period $\Delta t \sim [0, 0.1]$ s at specified model parameters. Our data indicate that field intensity in the injection area is comparable with depolarizing fields for these crystals. Moreover, the field intensity on injection zone border remains rather significant which supports the experimental capability using electron beam in order to inverse the orientation of domain structure. Additional studies are required to improvement the model taking into account the time-delay effects of response formation under electron irradiation. The simulation of electron beam-stimulated effects in typical ferroelectrics provides a set of tools for building new levels of understanding about mechanisms of electron interactions with active dielectrics. This underlines a new ways for SEM modification and application the polar materials.

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