ONE-DIMENSIONAL MODELLING OF A CARBON NANOTUBE-BASED BIOSENSOR

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KEYWORDS

Modelling, reaction-diffusion, biosensor, carbon nanotube.

ABSTRACT

This paper presents a one-dimensional-in-space mathematical model of an amperometric biosensor based on a carbon nanotube electrode deposited on a perforated membrane. The developed model is based on nonlinear reaction-diffusion equations. The conditions at which the one-dimensional mathematical model can be applied to an accurate simulation of the biosensor response are investigated. The accuracy of the response simulated by using one-dimensional model is evaluated by the response simulated by the corresponding two-dimensional model. The mathematical model and the numerical solution are also validated by an experimental data. The obtained agreement between the simulation results and experimental data is admissible for different configurations of the biosensor operation. The numerical simulation was carried out using the finite difference technique.

INTRODUCTION

Biosensors are analytical devices mainly used to measure concentrations of substances (Turner et al., 1987; Scheller and Schubert, 1992). They are relatively cheap and widely used in environment monitoring, drug detection and food industry (Wollenberger et al., 1997). Main parts composing a biosensor are: a biochemically active material, usually an enzyme, selectively detecting a target analyte (substrate), and a transducer. The transducer converts the biological recognition event into an electrical signal. The latter is then processed and displayed to an end user of the device.

Since carbon nanotubes (CNT) were discovered (Iijima, 1991), they were used in various applications. Because of their unique properties, carbon nanotubes are also used to build highly sensitive biosensors (Ahammad et al., 2009; Balasubramanian and Burghard, 2006).

A development of new biosensors is a rather expensive process requiring to perform a lot of experiments. In order to reduce the cost, a part of the experiments can be replaced by a computer simulation (Amatore et al., 2006). The action of practical biosensors is often modelled by a non-linear reaction-diffusion system (Schulmeister, 1990; Baronas et al., 2010). A CNT-based biosensor was mathematically modelled by Lyons (Lyons, 2009). A two-compartment mathematical model involving the mass transport and the enzyme kinetics was formulated in a one-dimensional domain, and the corresponding problem was solved analytically assuming the steady state conditions. More advanced biosensors than the modelled one are usually covered by outer porous or perforated membranes (Turner et al., 1987; Scheller and Schubert, 1992; Wollenberger et al., 1997).

Recently, a novel biosensor based on a carbon nanotube enzyme-loaded electrode deposited on a perforated polycarbonate membrane was developed (Razumienė et al., 2009), and a mathematical model of the biosensor was proposed (Baronas et al., 2011). The model has been formulated in a two-dimensional domain, and therefore the simulation was time and resource consuming. This is especially important when investigating numerically peculiarities of the biosensor response in wide ranges of the model parameters. The multifold numerical simulation of the biosensor response based on the one-dimensional model is much more efficient than the simulation based on the corresponding two-dimensional model.

In this paper, a corresponding one-dimensional-in-space model for the biosensor based on a CNT enzyme-loaded electrode deposited on a perforated membrane is proposed. The mathematical model and the numerical solution are validated by the experimental data (Razumienė et al., 2009; Baronas et al., 2011). The obtained agreement between the simulation results and experimental data is admissible for different configurations of the biosensor operation.

The conditions at which the one-dimensional mathematical model can be applied to accurate simulation of the biosensor action are investigated in this paper. The accuracy of the biosensor response simulated by using one-dimensional model is evaluated by the response simulated by the corresponding two-dimensional model. The numerical simulation was carried out using the finite difference technique.

PRINCIPAL STRUCTURE OF THE BIOSENSOR

The investigated biosensor has a layered structure (Razumienė et al., 2009). The biosensor was built by bind-

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ing the carbon nanotubes to the perforated membrane. Some of the carbon nanotubes are sinked into holes of the membrane during the preparation process. The membrane-CNT film was loaded with an enzyme and covered with an insulating film. Part of the enzyme was left outside of the CNT mesh and formed a layer between the CNTs and a insulating film. The principal structure of the active surface of the biosensor is shown in figure 1.

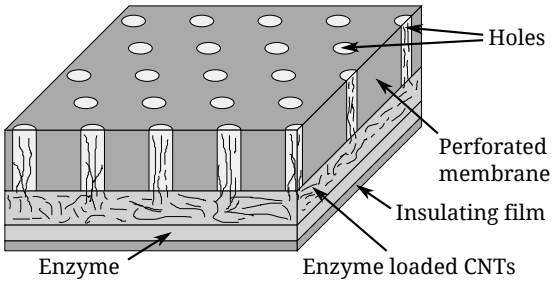

Figure 1: Principal structure of the active surface of the biosensor. The figure is not to scale

The following enzymatic reactions take place in the enzyme-loaded layers of the biosensor:

$$\mathrm{E_o} + \mathrm{S} \xrightarrow{k_1} \mathrm{E_r} + \mathrm{P}, \tag{1}$$

$$\mathrm{E_r} + \mathrm{M_o} \xrightarrow{k_2} \mathrm{E_o} + \mathrm{M_r}, \tag{2}$$

where k_1 and k_2 are the rate coefficients of the corresponding reactions. In reaction (1) the enzyme in the oxidized form ($\mathrm{E_o}$) detects the target substrate (S) and is reduced ($\mathrm{E_r}$). The other product (P) of the reaction has no impact on the biosensor response, and therefore is omitted in the following model. The reduced enzyme is re-oxidized in reaction (2) by converting a mediator from its oxidized form ($\mathrm{M_o}$) to the reduced one ($\mathrm{M_r}$). In the layer of enzyme-loaded carbon nanotubes, the reduced mediator participates in the following electrochemical reaction:

$$\mathrm{M_r} \longrightarrow \mathrm{M_o} + n_\mathrm{e}\mathrm{e}^-. \tag{3}$$

In reaction (3) the mediator is re-oxidized and electrons forming the response current are emitted. Here n_e stands for a number of electrons emitted per one reaction event. Reaction (3) is considered to be very fast. Such assumption is a common when modelling amperometric biosensors (Scheller and Schubert, 1992).

MATHEMATICAL MODEL

The model is formulated as a system of non-linear reaction-diffusion equations with initial and bounary conditions. Applying the homogenization process to the perforated membrane allows to formulate the mathematical model of the considered biosensor in a one-dimensional domain, on a line perpendicular to the surface of the biosensor (Bakhvalov and Panasenko, 1989). The surface of the insulating film is assumed to be zero point in the space interval. The model of the biosensor involves four layers (Ω_i) with different properties,

$$\begin{aligned} &\Omega_i \equiv (x_{i-1}, x_i), \quad x_0 = 0,\ x_i = x_{i-1} + d_i, \\ &\qquad \Gamma_0 \equiv \{0\},\ \Gamma_i \equiv \{x_i\}, \quad i = 1,2,3,4, \end{aligned} \tag{4}$$

where d_1 is the thickness of the enzyme layer, d_2 – the thickness of the enzyme-loaded CNT mesh, d_3 – the thickness of the perforated membrane, and d_4 stands for the thickness of the Nernst diffusion layer forming on the surface of the perforated membrane.

Governing Equations

Due to the enzyme immobilization in the CNT mesh, no mass transport of the enzyme is considered. The dynamics of the enzyme concentration is only impacted by biochemical reactions (1) and (2),

$$\begin{aligned} \frac{\partial E_{\mathrm{o},i}}{\partial t} &= -k_1 E_{\mathrm{o},i} S_i + k_2 M_{\mathrm{o},i} E_{\mathrm{r},i}, \\ \frac{\partial E_{\mathrm{r},i}}{\partial t} &= k_1 E_{\mathrm{o},i} S_i - k_2 M_{\mathrm{o},i} E_{\mathrm{r},i}, \\ &\qquad x \in \Omega_i,\ i = 1,2,\ t > 0, \end{aligned} \tag{5}$$

where x and t stand for space and time, respectively. $U_i = U_i(x,t),\ U_i \in \{E_\mathrm{o}, E_\mathrm{r}, S, M_\mathrm{o}\}$ are concentrations of the corresponding species in i^{th} layer (Ω_i) of the biosensor.

The substrate is affected by the diffusion in all layers of the biosensor. Additionally, in the layers filled with the enzyme the substrate participates in the reaction (1),

$$\begin{aligned} \frac{\partial S_i}{\partial t} &= D_{\mathrm{S}_i} \frac{\partial^2 S_i}{\partial x^2} - k_1 E_{\mathrm{o},i} S_i, \quad i = 1,2, \\ \frac{\partial S_i}{\partial t} &= D_{\mathrm{S}_i} \frac{\partial^2 S_i}{\partial x^2}, \qquad\qquad\quad i = 3,4, \\ &\qquad\qquad x \in \Omega_i,\ t > 0, \end{aligned} \tag{6}$$

where D_{S_i} ($i = 1,4$) is the diffusion coefficient of the substrate in the corresponding layer, and $D_{\mathrm{S},i}$ ($i = 2,3$) is the effective diffusion coefficient of the substrate in the CNTs and the perforated membrane.

The mediator in the oxidized form is affected by the diffusion in the entire biosensor and the Nernst diffusion layer. In the layer of the enzyme-loaded CNTs (Ω_2), the mediator is consumed in the reaction (2) and immediately regenerated in the reaction (3). The reactions compensate each other in terms of the mediator concentration. No electrochemical reaction occurs in the layer entirely filled with the enzyme Ω_1. Therefore, the mediator is consumed in reaction (2) without regeneration. The dynamics of the concentration of the oxidized mediator is described by the following equations ($t > 0$):

$$\begin{aligned} \frac{\partial M_{\mathrm{o},i}}{\partial t} &= D_{\mathrm{M}_{\mathrm{o},i}} \frac{\partial^2 M_{\mathrm{o},i}}{\partial x^2} - k_2 M_{\mathrm{o},i} E_{\mathrm{r},i}, \quad i = 1, \\ \frac{\partial M_{\mathrm{o},i}}{\partial t} &= D_{\mathrm{M}_{\mathrm{o},i}} \frac{\partial^2 M_{\mathrm{o},i}}{\partial x^2}, \quad i = 2,3,4, \quad x \in \Omega_i, \end{aligned} \tag{7}$$

where $D_{\mathrm{M}_{\mathrm{o},i}}$ is the diffusion coefficient of the oxidized mediator in the corresponding region, $i = 1,4$. $D_{\mathrm{M}_{\mathrm{o},2}}$ is

the effective diffusion coefficient of $\mathrm{M_o}$ in the enzyme-loaded CNTs (Ω_2), and $D_{\mathrm{M_{o,3}}}$ is the corresponding coefficient for the perforated membrane.

Because of very high rate of the electrochemical reaction (3), the reduced mediator resides only in the enzyme layer (Ω_1), where it diffuses and is generated in the reaction (3),

$$\frac{\partial M_{\mathrm{r},1}}{\partial t} = D_{\mathrm{M_{r,1}}} \frac{\partial^2 M_{\mathrm{r},1}}{\partial x^2} + k_2 M_{\mathrm{o},1} E_{\mathrm{r},1}, \tag{8}$$

where $M_{\mathrm{r},1} = M_{\mathrm{r},1}(x,t)$ is the concentration of the reduced mediator in Ω_1, and $D_{\mathrm{M_{r,1}}}$ is the diffusion coefficient of $\mathrm{M_r}$ in the enzyme layer.

Boundary Conditions

The modelled experiments were performed by intensively mixing the solution to be analysed (Razumienė et al., 2009). This leads to constant concentrations of the species above the Nernst diffusion layer,

$$S_4(x_4,t) = S_0, \quad M_{\mathrm{o},4}(x_4,t) = M_0, \tag{9}$$

where S_0 is the substrate and M_0 is the mediator concentrations in the bulk solution.

At the surface of the insulating film ($x = 0$), the non-leakage boundary conditions are applied for all the diffusive substances,

$$\left.\frac{\partial U}{\partial x}\right|_{\Gamma_0} = 0, \quad U \in \{S_1, M_{\mathrm{o},1}, M_{\mathrm{r},1}\},\ t > 0. \tag{10}$$

The matching conditions are applied on all the boundaries between adjacent regions,

$$D_{U_i} \left.\frac{\partial U_i}{\partial x}\right|_{\Gamma_i} = D_{U_{i+1}} \left.\frac{\partial U_{i+1}}{\partial x}\right|_{\Gamma_i}, \qquad U_i|_{\Gamma_i} = U_{i+1}|_{\Gamma_i}, \tag{11}$$

for $U_i = S_i,\ i = 1,2,3$ and $U_i = M_{\mathrm{o},i},\ i = 2,3$.

Due to the electrochemical reaction (3) taking place on the boundary Γ_1 between the enzyme layer and the enzyme-loaded CNTs, the concentration of $\mathrm{M_r}$ is permanently reduced to zero,

$$M_{\mathrm{r},1}|_{\Gamma_1} = 0, \quad t > 0. \tag{12}$$

All the reduced mediator is immediately re-oxidized on this boundary,

$$D_{\mathrm{M_{o,2}}} \left.\frac{\partial M_{\mathrm{o},2}}{\partial x}\right|_{\Gamma_1} = D_{\mathrm{M_{o,1}}} \left.\frac{\partial M_{\mathrm{o},1}}{\partial x}\right|_{\Gamma_1} + D_{\mathrm{M_{r,1}}} \left.\frac{\partial M_{\mathrm{r},1}}{\partial x}\right|_{\Gamma_1}, \qquad M_{\mathrm{o},1}|_{\Gamma_1} = M_{\mathrm{o},2}|_{\Gamma_1}. \tag{13}$$

Initial Conditions

The numerical simulation of an experiment starts at the moment $t = 0$, when the substrate and the mediator are poured into the buffer solution and reaches the external boundary of the Nernst diffusion layer,

$$S_4|_{\Gamma_4} = S_0, \quad M_{\mathrm{o},4}|_{\Gamma_4} = M_0. \tag{14}$$

Simultaneously ($t = 0$), the substrate and the mediator are absent elsewhere,

$$S_i = M_{\mathrm{o},i} = 0, \quad x \in \overline{\Omega}_i \setminus \Gamma_4,\ i = 1,2,3,4. \tag{15}$$

All the enzyme is assumed to be in the oxidized form at the beginning of the experiment,

$$E_{\mathrm{o},1} = E_0, \quad E_{\mathrm{r},1} = 0, \quad x \in \Omega_1,\ t = 0, \tag{16}$$

where E_0 is the total concentration of the enzyme in the enzyme layer (Ω_1). The layer of the carbon nanotubes is assumed to be only partially filled with the enzyme,

$$E_{\mathrm{o},2} = \eta E_0, \quad x \in \Omega_2,\ t = 0, \tag{17}$$

where $0 \le \eta < 1$ is the ratio of enzyme concentration in CNTs to its concentration in Ω_1.

Output of the Biosensor

The output current of the biosensor is generated due to the electrochemical reaction (3) taking place in the region Ω_2 of the CNT electrode. The reaction (3) was assumed so fast that all the mediator in the reduced form is immediately oxidized. The reduced mediator $\mathrm{M_r}$ arises in the region Ω_2 due to the enzymatic reaction (2) as well as the diffusion from the adjacent region Ω_1 through the boundary Γ_1. The latter part of the reduced mediator is completely oxidized in (3). Taking into account these two sources of $\mathrm{M_r}$ for the electrochemical reaction (3) leads the total output current $j(t)$ of the biosensor,

$$j(t) = n_{\mathrm{e}} F \times \left(k_2 \int_{x_1}^{x_2} E_{\mathrm{r},2} M_{\mathrm{o},2}\, dx - D_{\mathrm{M_{r,1}}} \left.\frac{\partial M_{\mathrm{r},1}}{\partial x}\right|_{\Gamma_1} \right), \tag{18}$$

where F is the Faraday constant.

When using practical biosensors the saturated output current is commonly used as a response of the biosensor. The steady state current density J of the considered biosensor is defined as follows:

$$J = \lim_{t \to \infty} j(t). \tag{19}$$

Effective Diffusion Coefficients

In order to reduce the number of the model parameters, the species S, $\mathrm{M_o}$ and $\mathrm{M_r}$ are assumed to have the same diffusion coefficients in a certain medium. The enzyme

and Nernst diffusion layers are assumed to be homogeneous, therefore diffusion coefficients for the species are expressed as

$$\begin{aligned} D_U &= D_\mathrm{e}, \qquad U \in \{\mathrm{S}_1, \mathrm{M}_{\mathrm{o},1}, \mathrm{M}_{\mathrm{r},1}\}, \\ D_U &= D_\mathrm{n}, \qquad U \in \{\mathrm{S}_4, \mathrm{M}_{\mathrm{o},4}\}, \end{aligned} \tag{20}$$

where D_e is the diffusion coefficient of the species in the enzyme, and D_n is the corresponding diffusion coefficient in the diffusion layer.

The CNT layer as well as the perforated membrane are really non-homogeneous mediums. Assuming these layers as the periodic media and applying the volume averaging approach to them lead to the effective diffusion coefficients for these mediums (Whitaker, 1999; Bakhvalov and Panasenko, 1989). The CNT layer (Ω_2) is considered as a composite of three compartments: the carbon nanotubes, the enzyme and the bulk solution. The perforated membrane (Ω_3) was treated as a composite of non-permeable material, and the bulk solution together with carbon nanotubes in the holes of the membrane. Assuming low volume of the CNTs the effective diffusion coefficients can be expressed in terms of the volume fractions, the tortuosity and the diffusion coefficients for the corresponding compartments,

$$\begin{aligned} D_U &= \theta_2 \left(\eta D_\mathrm{e} + (1-\eta) D_\mathrm{n}\right), \quad U \in \{\mathrm{S}_2, \mathrm{M}_{\mathrm{o},2}\}, \\ D_U &= \theta_3 \rho D_\mathrm{n}, \qquad\qquad\qquad\quad U \in \{\mathrm{S}_3, \mathrm{M}_{\mathrm{o},3}\}, \end{aligned} \tag{21}$$

where ρ stands for the perforation level of the membrane expressed as the volume fraction of the holes, θ_2 and θ_3 are the tortuosities defining the structural properties of the corresponding media.

Similar approach for estimating the effective diffusion coefficients was also applied to formulating the two-dimensional model of the considered biosensor (Baronas et al., 2011) and to modelling the biosensor with the perforated membrane in the one-dimensional space (Petrauskas and Baronas, 2009).

NUMERICAL SIMULATION

The proposed mathematical model was formulated as an initial boundary value problem with PDEs containing non-linear terms representing reactions. Analytical solutions of this type of systems are known in very special cases only, therefore numerical methods are usually used to get approximate solutions in wide ranges of the model parameters (Schulmeister, 1990; Baronas et al., 2010). The method of finite differences was employed to solve numerically the equation system of the proposed model (Samarskii, 2001). The domain of the model was discretized by applying a regular mesh to each region Ω_i, $i = 1, 2, 3, 4$, and constant step was used to discretize the time during the simulations.

In simulation, the steady state was assumed already reached if the decay of the biosensor current over time becomes small enough,

$$T_\mathrm{R} = \min_{j(t)>0} \left\{ t : \frac{d\, j(t)}{d\, t} \times \frac{t}{j(t)} < \varepsilon \right\}. \tag{22}$$

The dimensionless decay rate $\varepsilon = 0.01$ was used in the simulations presented in this paper. The current density $j(T_\mathrm{R})$ was assumed as the steady state biosensor response.

The following values of the model parameters were used as a basic configuration of the considered biosensor and were kept constant in all the simulations:

$$\begin{gathered} d_1 = 10^{-7}\,\mathrm{m}, \;\; d_2 = 4 \times 10^{-7}\,\mathrm{m}, \;\; d_4 = 1.5 \times 10^{-4}\,\mathrm{m}, \\ E_0 = 4.55 \times 10^{-2}\,\mathrm{mol\,m^{-3}}, \;\; n_\mathrm{e} = 2, \;\; \eta = 0.5, \\ D_\mathrm{e} = 3 \times 10^{-10}\,\mathrm{m^2 s^{-1}}, \;\; D_\mathrm{n} = 2D_\mathrm{e}, \;\; \theta_2 = 1/3, \\ k_1 = 6.9 \times 10^{2}\,\mathrm{m^3 mol^{-1} s^{-1}}, \\ k_2 = 6.9 \times 10^{4}\,\mathrm{m^3 mol^{-1} s^{-1}}. \end{gathered} \tag{23}$$

The adequateness of the proposed model was evaluated by comparing the simulated responses with the responses of the corresponding physical experiments. The results of the comparison are shown in figure 2.

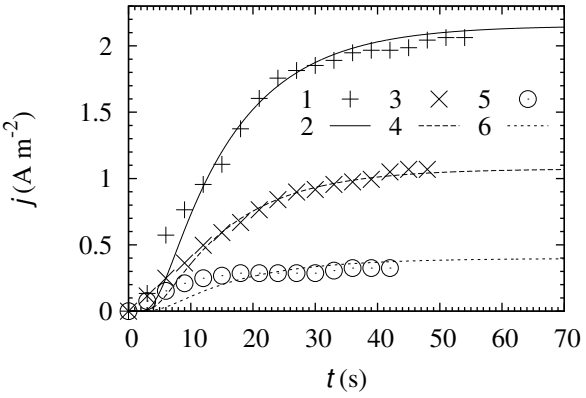

Figure 2: Biosensor current densities obtained experimentally (1, 3, 5) and numerically (2, 4, 6) at $\rho = 0.0625$, $\theta_3 = 0.5$, $d_3 = 10^{-5}$ m and the following concentrations of the substrate and the mediator: $M_0 = 0.2$, $S_0 = 9.9$ (1, 2), $M_0 = 0.05$, $S_0 = 4.98$ (3, 4), $M_0 = 0.005$, $S_0 = 1.99\,\mathrm{mol\,m^{-3}}$ (5, 6). The other parameters were as defined in (23)

As one can see in figure 2, the simulated responses of the biosensor are close to the results of the corresponding physical experiments. When approaching the steady state, the corresponding current densities differ not more than 10%. Similar accuracy of the simulation was observed when simulating operation of the considered biosensor using the two-dimensional model (Baronas et al., 2011). However, the two-dimensional model is much more computational resource consuming. An individual numerical simulation runs approximately 25 hours using the two-dimensional model and approximately 5 minutes using the corresponding one-dimensional model for the biosensor configurations employed in the experiments presented in figure 2. The simulations were performed on a computer with Intel® Core™ i5-540M (2.53 GHz) CPUs, each simulation running on one CPU only.

The two-dimensional model takes into consideration the geometry of the perforated membrane, while the perforation topology is approximated by introducing the effective diffusion coefficients in the one-dimensional model. Therefore, the one-dimensional model can be considered as an approximation of the corresponding two-dimensional one. The two-dimensional model was reduced by introducing two perforation parameters: the perforation ratio ρ and the tortuosity θ_3.

In order to investigate the conditions under which the one-dimensional mathematical model can be applied to an accurate simulation of the biosensor response, a number of simulations was performed and the modelling error of the one-dimensional model was calculated, assuming the two-dimensional model as a precise one,

$$\nu = \frac{J - J_{2\mathrm{D}}}{J_{2\mathrm{D}}}, \tag{24}$$

where $J_{2\mathrm{D}}$ is the density of the steady state current obtained by using the two-dimensional model formulated in (Baronas et al., 2011). A similar approach to a comparison of one and two-dimensional models was also used in (Petrauskas and Baronas, 2009).

RESULTS AND DISCUSSION

The accuracy of the proposed one-dimensional model (6)–(19) was evaluated for different values of the parameters of the geometry of the perforated membrane. The biosensor response was simulated and the modelling error ν was calculated at different values of the perforation ratio ρ, the tortuosity θ_3 and the thickness d_3.

Impact of the Perforation Level

The dimensionless perforation level ρ depends on the topology and the form of the holes in the membrane. In the case of two-dimensional modelling, the holes were modelled by right cylinders of uniform diameter and spacing, forming a regular hexagonal pattern. The corresponding one-dimensional modelling treats the membrane as a homogeneous medium with the corresponding effective (averaged) diffusion coefficients. In order to investigate the impact of the perforation geometry on the modelling error ν, the numerical simulations were performed using the one and two-dimensional models at various levels of the membrane perforation. The simulations of the biosensor response were repeated for the same concentrations of the substrate and the mediator as in the physical experiments depicted in figure 2. Figure 3 shows the calculated values of the one-dimensional modelling error ν.

As one can see in figure 3, the absolute value $|\nu|$ of the modelling error is less than 10% when the perforation level ρ is greater than 0.002. The real biosensor was modelled at the perforation level $\rho = 0.0625$ (see figure 2). At this value of the level ρ the modelling error ν is approximately equal to 0.075. Similar modelling errors

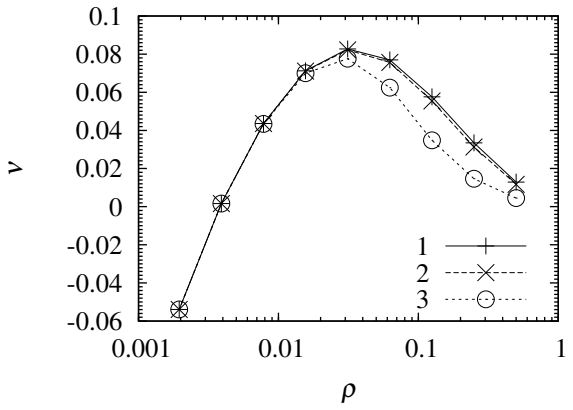

Figure 3: The dependence of the one-dimensional modelling error ν on the membrane perforation level ρ at the tortuosity $\theta_3 = 0.5$, the thickness $d_3 = 10^{-5}$ m and the following concentrations of the substrate and the mediator: $M_0 = 0.2,\ S_0 = 9.9$ (1), $M_0 = 0.05,\ S_0 = 4.98$ (2), $M_0 = 0.005,\ S_0 = 1.99\,\mathrm{mol\,m^{-3}}$ (3). Values of other parameters are defined in (23)

are usually admissible for an investigation of the peculiarities of the biosensor response (Baronas et al., 2010). When the perforation level ρ decreases below 0.002, the error escalates.

Impact of the Tortuosity in the Perforated Membrane

The tortuosity θ_3 of the perforated membrane varies due to the structural properties of the carbon nanotubes sinked into the holes. The tortuosity impacts the effective diffusion coefficient of the substances in the perforated membrane. In order to investigate the impact of the tortuosity on the accuracy of the one-dimensional model, multiple numerical experiments were performed using both models, and the relative error ν of the one-dimensional modelling was calculated. The simulation results are shown in figure 4.

As one can see in figure 4, the error ν of the one-dimensional modelling decreases with decreasing the tortuosity θ_3. The maximum error reaches at the theoretical maximum tortuosity $\theta_3 = 1$. Although the error ν is quite high ($\nu \approx 0.12$) at $\theta_3 = 1$, it is still at the level allowing one to use the one-dimensional model for approximate estimations of the biosensor behaviour.

Impact of the Perforated Membrane Thickness

The thickness of the perforated membrane influences the flux of the substrate and the mediator from the bulk to the active region of the biosensor. The role of the effective diffusion coefficients in the perforated membrane increases with increasing the thickness d_3. In order to investigate the impact of the thickness d_3 of the perforated membrane on the one-dimensional modelling error ν, multiple numerical simulations were performed by changing the thickness d_3 from $0.1\,\mu\mathrm{m}$ up to $0.1\,\mathrm{mm}$. Figure 5 presents the calculated values of the error ν.

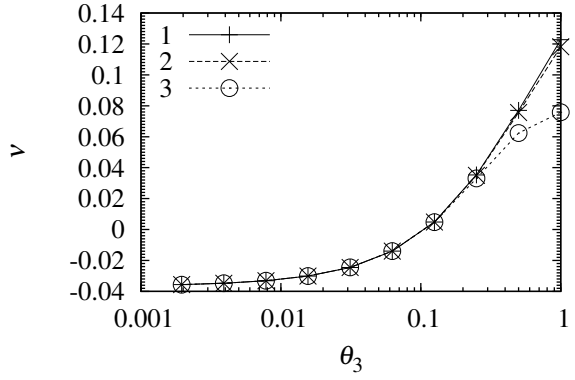

Figure 4: The modelling error ν versus the membrane tortuosity θ_3 calculated at the perforation level $\rho = 0.0625$ and the thickness $d_3 = 10^{-5}\,\mathrm{m}$. The other parameters and the notation are the same as in figure 3

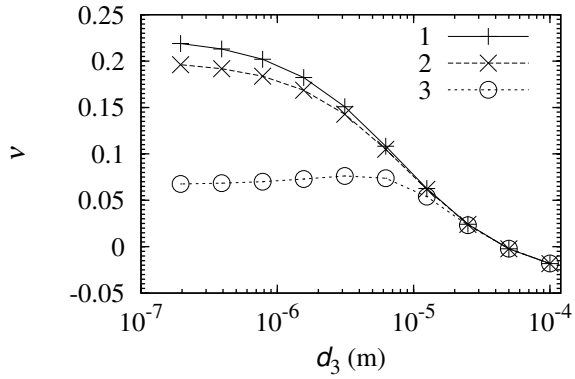

Figure 5: The modelling error ν versus the membrane thickness d_3 calculated at the perforation level $\rho = 0.0625$ and the tortuosity $\theta_3 = 0.5$. The other parameters and the notation are the same as in figure 3

One can see in figure 5 that the modelling error ν is a monotonous decreasing function of thickness d_3 for the higher concentrations of the substrate and the mediator (curves 1 and 2) and is a non-monotonous function in the case of the low concentrations (curve 3). However, the corresponding absolute value $|\nu|$ is a non-monotonous function of d_3 for all the considered concentrations. Only when the perforated membranes is relatively tick ($d_3 > 5\,\mu\mathrm{m}$), the proposed one-dimensional model can be successfully applied to predicting the biosensor response.

CONCLUSIONS

The proposed one-dimensional mathematical model (6)–(19) can be successfully used to simulate the response of the biosensor based on the carbon nanotube electrode at practical configurations of the biosensor operation (figure 2).

The accuracy of the simulated response considerably depends of the geometry of the outer perforated membrane. The mathematical model (6)–(19) accurately describes the biosensor action for relatively thick ($d_3 > 5\,\mu\mathrm{m}$, figure 5) perforated membranes of moderate as well as high perforation level ρ ($\rho > 0.002$, figure 3). Otherwise, the corresponding two-dimensional mathematical model should be used (Baronas et al., 2011), though computational resource consuming.

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