Transient Simulation of Field-Effect Biosensors : How to Avoid Charge Screening Effect

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***Abstract*—We developed a numerical simulator to model the operation of a bio-FET in transient state. The simulator takes a realistic device structure as a simulation domain, and it employs the drift-diffusion equation for ion / channel transport, and the Ramo-Shockley theorem for accurate calculation of non-faradaic current. For efficient transient simulation, the implicit time integration scheme is employed where the solution at each time step is obtained from the coupled Newton-Raphson method. Using the simulator, we found that the sensitivity of bio-FET can be improved with transient measurement by redistribution of the mobile ions by an external electric field.**

Keywords—bio-FET, charge screening effect, transient simulation)

I. INTRODUCTION

Field-effect transistor-based biosensors (bio-FET) have been used to detect various biomolecules [1], [2]. In this type of biosensor, the probe molecules are attached to the FET gate, and the probe-target binding event occurs changing conductance of the channel. It is usually measured in the steady state. However, there are research results that higher sensitivity can be obtained by measuring in the transient state after applying the step pulse bias [1], [2]. About this, only one-dimensional conceptual simulation and experimental results were reported; lacking in detailed theoretical study about what happens in the transient state [1]. Therefore, in this study, we have developed a physics-based 3-dimentsional simulator to analyze the terminal current of the realistic device structure including detailed surface physics. Since the buffer solution is used as an electrolyte in many practical experiments, a numerically efficient transient simulation method has been developed to simultaneously include various kinds of ion movements and reactions and couple with the model for the channel transport. Also, we considered the biomolecule layer as an ion-permeable membrane including the partitioning effect from the Born equation. We applied the developed simulator to the actual device structure and showed that the movement of ions in the electrolyte in the transient state affects the channel conductance, which improves the sensitivity.

II. PHYSICS MODELS

The device structure used in the simulation is shown in Fig.1. This structure is based on the fabricated reference-electrode free device structure. The potential of the electrolyte is close to that of the source electrode due to the large capacitive coupling between the source electrode and the electrolyte solution. Therefore, the source electrode acts as a reference electrode and no additional reference electrode is required.

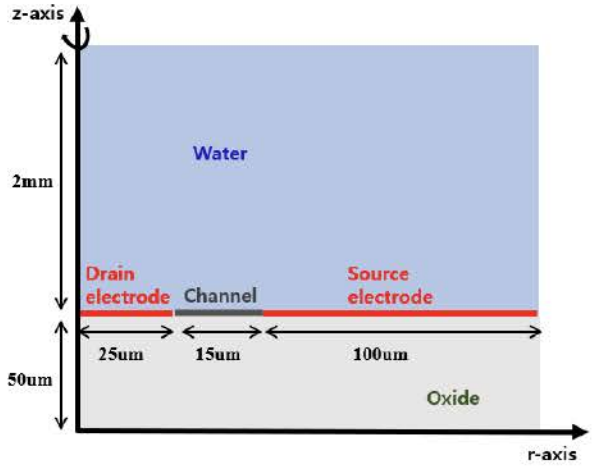

Fig. 1. Schematic of the simulation domain. The cylindrical coordinates are chosen to take advantage of the concentric feature of the electrode

A. The Poisson Equation

The electric potential in the device is governed by the Poisson equation.

$$\nabla^2 V = -\rho/\varepsilon_0 \qquad (1)$$

B. Transport of ions

We considered the phosphate-buffered saline (PBS) buffer solution usually used in experiments [3]. For the charge transport in the solution, we used the drift diffusion equation for each ions ($Na^+, K^+, Cl^-, H^+, OH^-, HPO_4^{2-}, H_2PO_4^-$):

$$J = -zu[n]\nabla V - D\nabla[n] \qquad (2)$$

Where u, D, z and [n] are the mobility, diffusivity, charge state and density of the corresponding ion. We assume a constant mobility and the diffusivity is obtained from the Einstein relation. Since the biomolecules generally react with the

978-1-7281-0940-4/19/$31.00 ©2019 IEEE

hydrogen ion, the pH value should be carefully modeled according to the buffer reactions near the metal electrode and solution interface. It is considered as the net generation and recombination terms of and we express the continuity equation for each ion as follows:

$$d[n]/dt = -\nabla \cdot \mathrm{J} + \mathrm{G} - \mathrm{R} \quad (3)$$

The generation and recombination terms for each ion can be calculated using the detailed balance condition in equilibrium [3].

$$[H^+] : G = a_1 + b_1[H_2PO_4^-],$$
$$R = a_2[H^+][OH^-] + b_2[H^+][HPO_4^{2-}] \quad (4a)$$
$$[OH^-] : G = a_1, \mathrm{R} = a_2[H^+][OH^-] \quad (4b)$$
$$[HPO_4^{2-}] : \mathrm{G} = b_1[H_2PO_4^-], R = b_2[H^+][\mathrm{HPO_4}^{2-}] \quad (4c)$$
$$[H_2PO_4^-] : \mathrm{G} = b_2[H^+][\mathrm{HPO_4}^{2-}], \mathrm{R} = b_1[H_2PO_4^-] \quad (4d)$$

These are expressed in terms of the concentration of several ions. Therefore, if there is buffer reaction (generation and recombination), the continuity equations for each of the seven ions are coupled together and should be considered in the Jacobian matrix.

C. Channel

By default, the drift-diffusion equation and the continuity equation are used for the channel as well as the ions. Also, we assume that the channel is neutral. Ohmic contact was used as boundary condition between metal and semiconductor. To reduce numerical complexity, we assumed that channel is thin film. So thin that the charges in the film are considered as interface (surface) charges and the current in it as surface current.

D. Biomolecule layer

The biomolecule layer is treated as an ion-permeable membrane layer considering the Gibbs free energy barrier using the Born equation [3]. The thickness of this layer is 4nm and the charge is assumed to be uniformly distributed. We assume that the probe molecule is electrically neutral and the concentration of the target molecule after binding event is $10^{14}/cm^2$. When a biomolecule is present in an electrolyte, it causes modulation of permittivity. Therefore, when the mobile ions pass through the membrane layer, they feel a free energy barrier and the drift-diffusion equation in the membrane region is modified as

$$\mathrm{J} = -\mathrm{zu[n]}(\nabla \mathrm{V} + \Delta \mathrm{G}) - \mathrm{D}\nabla[\mathrm{n}]. \quad (5)$$

where G is Gibbs free energy difference:

$$\Delta \mathrm{G} = G_{membrain} - G_{water} = \frac{N_A z^2 e^2}{8\pi\varepsilon_0 r_0}\left(\frac{1}{\varepsilon_{membrain}} - \frac{1}{\varepsilon_{water}}\right) \quad (6)$$

E. Terminal current

The terminal current consists of direct current flowing through the channel and non-faradaic current due to the movement of the ions in the electrolyte solution. To calculate the non-faradaic current in the numerical simulation, we employed the extended Ramo-Shockley theorem [4]:

$$I_j(t) = -\sum_i^N q_i v_i(t) \cdot \nabla f_j(r_i) \quad (7)$$

where $f_j(r)$ is the electric potential at position r when the j-th electrode is kept at unit potential while all the other electrodes are grounded and all the charges are removed from the simulation domain.

TABLE I. SIMULATION DEFAULT PARAMETERS

Parameter	*Symbol*	*Default value*
Biomolecule layer density	Np	$10^{14}/cm^2$
Bulk concentration of ions	$[Na^+]$	0.1M
	$[K^+]$	0.022M
	$[Cl^-]$	0.1M
	$[HPO_4^{2-}]$	0.01M
	$[H_2PO_4^-]$	0.22M
Ion mobilites	u_{Na^+}	$5.9 \times 10^{-4} cm^2/V \cdot s$
	u_{K^+}	$8.6 \times 10^{-4} cm^2/V \cdot s$
	u_{Cl^-}	$7.0 \times 10^{-4} cm^2/V \cdot s$
	$u_{HPO_4^{2-}}$	$3.83 \times 10^{-4} cm^2/V \cdot s$
	$u_{H_2PO_4^-}$	$3.03 \times 10^{-4} cm^2/V \cdot s$
	u_{H^+}	$33.3 \times 10^{-4} cm^2/V \cdot s$
	u_{OH^-}	$18.8 \times 10^{-4} cm^2/V \cdot s$
Channel mobility	u_n	$20 cm^2/V \cdot s$
	u_p	$20 cm^2/V \cdot s$
Temperature	T	300K
Stern layer thickness	d_{stern}	5A
Water permittivity	ε_{water}	80
Stern layer permittivity	ε_{stern}	40
Biomolecule layer permittivity	ε_{bio}	3.9
Oxide permittivity	ε_{ox}	3.9

III. NUMERICAL METHOD

Since the size of the region of interest near the electrode is very small, a careful generation of the mesh size is necessary to achieve the numerical efficiency. Since the electrical double layer (EDL) length is usually only a few nanometers and we need to calculate the potential near the channel-electrolyte surface, we used a very small mesh spacing of 2.5A near the surface and a gradually increasing mesh size to bulk. The simulation is based on a cylindrical coordinate and the finite volume (box integration) method was used. We used the Scharfetter-Gummel scheme as the optimal way to discretize the drift-diffusion equation. In transient simulation, the explicit method requires very small time step for numerical stability. Since the required simulation time period is on the order of hundreds of us, the explicit approach would require

prohibitively large number of time steps and is therefore not suitable for efficient simulation. Therefore, to freely use the size of the time step, we used the implicit method (Backward Euler method). When using this approach, we also need to minimize the truncation errors due to the large time step. For example, for a step pulse bias, at the moment when the applied voltage is changed, the ions are rapidly redistributed. Thus, the time step of about 10 ns is used at this moment. Then, the time step can be gradually increased as the ion redistribution rate is reduced after EDL charging. At each time step, we need to find the solutions for the ten coupled equations, but the convergence is not good for the Gummel iteration method. Therefore, the fast and robust convergence was obtained through the coupled Newton scheme.

IV. Results

Fig.2 shows the probe-target binding event. When the target material is negatively charged, this negative charge affects the channel conductivity. Therefore, the channel current change and the target material can be measured. However, there is a space charge layer called the electric double layer at the interface between the electrolyte and the solid surface. In typical experimental and simulation environments, this EDL is only a few nanometers thick. Therefore, the charge of a biomolecule is significantly screened by ions and therefore can not achieve high sensitivity. However, as shown in Fig. 3, after a step pulse is applied to the drain electrode, ions are redistributed in the transient state and the EDL is extended. Therefore, the channel can better sense the charge of the target material, and it is confirmed that higher sensitivity was observed at the time of ion redistribution as Fig. 4.

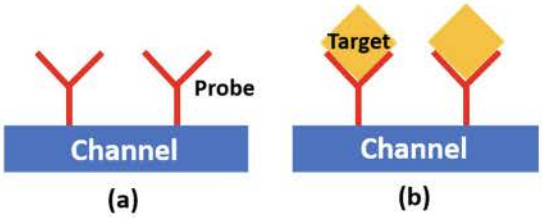

Fig. 2. Schematic of (a) befer and (b) after of the probe-target binding event. .

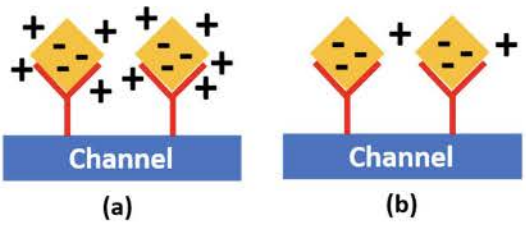

Fig. 3. Distribution of the ions around bio molecule at (a) steady state and (b) transient state.

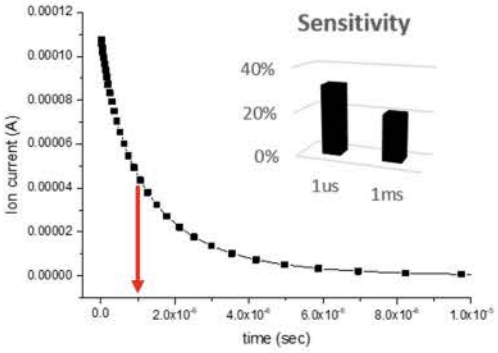

Fig. 4. Non-faradaic current and sensitivity after unit pulse is applied to the drain electrode.

V. Conclusion

We developed 3-dimensional physics-based model for simulation for transient simulation of bio-FET. It is confirmed that the sensitivity can be improved by measuring the transient current, which is due to the field effect and redistribution of the mobile ions around the channel.

Acknowledgment

This work was supported by the Brain Korea 21 Plus Project in 2019.

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