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Simulation of ionic current through the nanopore in a double-layered semiconductor membrane

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Abstract
We study the effects of different nanopore geometries (double-conical, single-conical, cylindrical) on the electrostatic potential distribution and ionic conductivity in a double-layered semiconductor nanopore device as functions of the applied membrane bias. Ionic current–voltage characteristics as well as their rectification ratios are calculated using a simple ion transport model. Based on our calculations, we find that the double-layered semiconductor membrane with a single-conical nanopore with a narrow opening in the n-Si layer exhibits the largest range of available potential variations in the pore and, thus, may be better suited for control of polymer translocation through the nanopore.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Due to their large surface area and small volume, nanopores are an excellent example of nanoscale devices capable of efficient interaction with ions and biological molecules in electrolyte solution. In these devices, molecules, while permeating through the pore, interact with its surface. Because most biological molecules in ionic solution are charged, electrostatic interaction between these molecules and ions in the solution, as well as charges in the membrane and on the nanopore surface, is the crucial factor affecting the behavior of such systems.

The most vivid example of the nanopore’s effectiveness can be found in living cells where concentrations of ions in the cell and the extracellular environment are regulated by numerous ion channels and where the transport of biomolecules in and out of the cell nucleus occurs. In some of these biological nanopores, permeability can be changed by chemical or electrical signals. Presently, biological nanopores are used for biomolecule sensing and characterization [1–3], and the technique of ionic current blockage is particularly widespread [4–6].

Advances in the manufacture of semiconductor devices provide the possibility to design artificial nanopores that replicate or enhance characteristics of biological ion channels. Being more robust and variable than their biological counterparts, artificial nanopores are found to be effective in single molecule detection, protein filtering and analysis of biomolecule properties [7–11]. Ionic diodes [12, 13], voltage-tunable ionic field-effect transistors [14–19] and pores in multilayered membranes [20, 21] have also been proposed. These membranes can find applications in electronic single molecule sensing, manipulation and characterization, as well as in lab-on-a-chip devices and nanofluidics [1].

Static surface charge in the pore creates a predominance of either anions or cations in the channel, leading to selectivity of the pore (ionic filter). More complicated spatial charge distributions can lead to diode-like and transistor-like ionic devices, while non-cylindrical geometries often result in non-Ohmic current–voltage curves [19, 22].

In this work we are concerned with nanopores in semiconductor membranes that allow one to configure the electrostatic potential across the pore using voltage applied to layers of doped silicon. Although various configurations have been proposed by us for such devices [20, 21], in this work we focus on membranes with two adjacent layers of heavily doped p- and n-Si (see figure 1), which we have previously introduced. Thus, the membrane studied in this work has two silicon layers with opposite doping which form a p–n junction with large built-in potential that can be further
enhanced by applying a reverse bias. Since the p–n junction is a very narrow region in a heavily doped Si membrane (~4 nm for the considered doping concentration), it creates a high-gradient electric potential in the pore which may be useful for certain applications. Applied forward bias decreases the built-in potential as well as the variation of the potential in the pore, which indicates that the electrostatic properties of the pore may be easily varied by manipulating the bias on the membrane.

Specifically, in this work we consider semiconductor membranes with a nanopore and analyze the electrostatic potential distribution and ionic currents in the pore. A double-conical and two single-conical geometries of the nanopore are investigated at various applied biases to provide a systematic comparison of membrane characteristics for different nanopore geometries and evaluate our ability to electrically control the nanopore conductance. We find that besides the cylindrical nanopore, a single-conical nanopore with a smaller opening in the n-Si exhibits the widest range of potential variation in the pore and, thus, is better suited for ion filtering applications, as well as for control over biomolecule translocation through the pore.

2. Model

2.1. Electrostatic membrane model

The modeled system is a semiconductor membrane, consisting of two adjacent layers of n and p heavily doped silicon (Si) with density $2 \times 10^{20} \text{ cm}^{-3}$. The membrane, which is 26 nm thick, is covered by a 0.8 nm thick layer of silicon dioxide (SiO$_2$) and is immersed in an electrolyte solution. The electrolyte is fully dissociated KCl with concentration [KCl]$_0$ = 0.1 M.

Our model, similar to previous publications [15, 16, 20, 21], is based on the self-consistent solution of Poisson’s equation:

$$\nabla \cdot [\varepsilon(\vec{r}) \nabla \varphi(\vec{r})] = -\rho(\vec{r}),$$  
(1)

where $\varphi(\vec{r})$ is the electric potential, $\varepsilon(\vec{r}) = \varepsilon_r(\vec{r}) \varepsilon_0$ is the dielectric permittivity, $\varepsilon_r(\vec{r})$ is the relative permittivity of the material and $\varepsilon_0$ is the permittivity of free space.

The charge density $\rho(\vec{r})$ in the membrane is

$$\rho(\vec{r}) = q \{ p(\vec{r}) - n(\vec{r}) + N_{d}^{+}(\vec{r}) - N_{d}^{-}(\vec{r}) + N_{surf}(\vec{r}) \},$$  
(2)

where $q$ is the elementary charge, $p(\vec{r})$ and $n(\vec{r})$ are the concentrations of electrons and holes, $N_{d}^{+}(\vec{r})$ and $N_{d}^{-}(\vec{r})$ are the acceptor and donor densities in the semiconductor regions, and $N_{surf}(\vec{r})$ represents fixed surface charge on the membrane, specifically in the oxide.

The charge density in the electrolyte is

$$\rho(\vec{r}) = q \{ [K^{+}](\vec{r}) - [Cl^{-}](\vec{r}) \},$$  
(3)

where $[K^{+}](\vec{r})$ is the local concentration of potassium ions and $[Cl^{-}](\vec{r})$ is the local concentration of chloride ions.

The electrons and holes in the semiconductor regions follow Fermi–Dirac statistics:

$$n(\vec{r}) = N_c \mathcal{F}_{1/2} \left( \frac{E_i - E_c}{kT} \right),$$  
(4)

$$p(\vec{r}) = N_v \mathcal{F}_{1/2} \left( \frac{E_i - E_v}{kT} \right),$$  
(5)

where $N_c$ and $N_v$ are the effective densities of states in the conduction and valence bands, respectively [23], $\mathcal{F}_{1/2}$ is the Fermi integral [24] of order 1/2, $E_i$ is the Fermi energy level. The conductance and valence band edges in Si are calculated as

$$E_c = -q \varphi(\vec{r}) + E_{g}^{Si} / 2,$$  
(6)

$$E_v = -q \varphi(\vec{r}) - E_{g}^{Si} / 2,$$  
(7)

where $E_{g}^{Si}$ is the energy band gap of Si. We set $E_i$ to zero if no voltage bias is applied to the semiconductor material.

The conductance and valence band edges in SiO$_2$ are given as

$$E_{c}^{SiO_2} = E_c^g + q \chi^g - q \chi^{Si},$$  
(8)

$$E_{v}^{SiO_2} = E_v^g - E_{g}^{SiO_2},$$  
(9)

where $q \chi^{SiO_2}$ and $q \chi^g$ are the electron affinities of the corresponding materials, and $E_{g}^{SiO_2}$ is the band gap of SiO$_2$.

Since the membrane is a p–n junction, an applied reverse bias, or a forward bias that is smaller than the built-in potential

![Figure 1](Nanotechnology_22_165202_Fig1.png)

Figure 1. The schematics of the semiconductor membrane with the three modeled nanopore geometries. Left: symmetric double-conical; center and right: single-conical with a narrow opening in the p-Si layer and n-Si layer, respectively.
of the junction, does not produce significant current through the junction. Therefore, the applied bias can be modeled by shifting the Fermi levels in the corresponding regions:

\[ E_f = -qV_n, \quad \text{in } n \text{-Si}, \]  
\[ E_f = -qV_p, \quad \text{in } p \text{-Si}, \]  

where \( V_n \) and \( V_p \) are the voltages applied to the corresponding semiconductor layers.

The ion concentrations in the electrolyte (KCl) obey Boltzmann statistics:

\[ [\text{K}^+] = \left[ \text{KCl} \right]_0 \exp \left( -\frac{q\Phi(\vec{r})}{kT} \right), \]  
\[ [\text{Cl}^-] = \left[ \text{KCl} \right]_0 \exp \left( \frac{q\Phi(\vec{r})}{kT} \right), \]

where \( \left[ \text{KCl} \right]_0 \) is the concentration in the electrolyte bulk.

For simulations, the coordinate system is chosen so that the \( y \)- and \( z \)-axes are parallel to the membrane and the \( x \)-axis is perpendicular to it. The size of the system is taken to be large enough to ensure that both the concentrations and the potential reach corresponding asymptotic values at the boundaries. In particular, the concentrations of electrons and holes must reach their bulk values within the modeled membrane, while the concentrations of ions become equal to the bulk electrolyte values \( \left[ \text{KCl} \right]_0 \) far from the membrane, since the influence of the membrane and the nanopore is minimized at large distances.

To solve equation (1), we set the electrostatic potential \( \Phi(\vec{r}) \) to zero on boundaries in the bulk electrolyte that are above and below the membrane (\( x = 0 \) and \( L_x \)), and its normal derivative is set to zero on other boundaries, i.e.,

\[ \Phi|_{x=0,L_x} = 0, \quad \frac{\partial \Phi}{\partial y}|_{y=0,L_y} = 0, \quad \frac{\partial \Phi}{\partial z}|_{z=0,L_z} = 0. \]  

2.2. Computational scheme

Numerical simulations are performed on a uniform, rectangular, three-dimensional grid consisting of 161 \( \times \) 81 \( \times \) 81 grid points with the size of the grid spacing equal to 0.4 nm. Thus, the size of the simulated volume is 64.4 \( \times \) 32.4 \( \times \) 32.4 nm\(^3\). The reduction of the grid spacing to 0.2 nm leads to the difference of \( \sim 2 \text{ mV} \) for the average potential in the nanopore vicinity, which is about 2% of the total potential variation in the pore. Our choice of grid spacing allows for fast computation without significantly sacrificing the precision. Various material constants and simulation parameters are listed in table 1.

Poisson’s equation (1) is solved iteratively until self-consistent solution is reached. Gummel’s method [25] is
Figure 3. Profiles of the electrostatic potential \( \phi(\vec{r}) \) through the center of the pore for two single-conical pores. In (A) and (C), the membrane bias is \( V_n = -0.8 \text{ V} \); in (B) and (D), \( V_n = 0.8 \text{ V} \). \( V_p = 0 \text{ V} \) in both cases. In (A) and (C) ((B) and (D)), the potentials are shown for the SCp (SCn) geometry.

Table 1. Material parameters and constants.

<table>
<thead>
<tr>
<th>Material</th>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si</td>
<td>( E_{Si}^{g} )</td>
<td>1.124 eV</td>
</tr>
<tr>
<td>SiO(_2)</td>
<td>( E_{SiO(_2)}^{g} )</td>
<td>9.0 eV</td>
</tr>
<tr>
<td>Si</td>
<td>( q_{\chi_{Si}} )</td>
<td>4.05 eV</td>
</tr>
<tr>
<td>SiO(_2)</td>
<td>( q_{\chi_{SiO(_2)}} )</td>
<td>0.95 eV</td>
</tr>
<tr>
<td>Si</td>
<td>( \varepsilon_{Si} )</td>
<td>11.70</td>
</tr>
<tr>
<td>SiO(_2)</td>
<td>( \varepsilon_{SiO(_2)} )</td>
<td>3.9</td>
</tr>
<tr>
<td>Si</td>
<td>( N_{i_{Si}} )</td>
<td>( 2.0 \times 10^{20} \text{ cm}^{-3} )</td>
</tr>
<tr>
<td>SiO(_2)</td>
<td>( N_{i_{SiO(<em>2)}}, N</em>{surf} )</td>
<td>( 2.0 \times 10^{20} \text{ cm}^{-3}, -4.0 \times 10^{20} \text{ cm}^{-3} )</td>
</tr>
<tr>
<td>KCl</td>
<td>( T )</td>
<td>300 K</td>
</tr>
<tr>
<td>KCl</td>
<td>( \varepsilon_{KCl} )</td>
<td>80.0</td>
</tr>
<tr>
<td>KCl</td>
<td>( [\text{KCl}]_0 )</td>
<td>0.1 M</td>
</tr>
</tbody>
</table>

\(*\) This \( N_{surf} \) corresponds to the surface charge density \( \sigma = -0.16 \text{ electrons nm}^{-2} \).

2.3. Nanopore geometries and membrane biases

We consider three nanopore geometries illustrated in figure 1: symmetric double-conical (DC), single-conical with a narrow opening in the p-Si layer (SCp), and single-conical with a narrow opening in the n-Si layer (SCn).

In single-conical geometries, the larger opening of the pore has the diameter denoted by \( D \) that we vary between 2 and 10 nm. The smaller diameter of the pore is always fixed at 2 nm. In the double-conical geometry, both outer diameters are equal to \( D \) and are varied while the center diameter fixed at 2 nm. The value of \( D = 2 \text{ nm} \) makes all three geometries (DC, SCp and SCn) cylindrical and identical.

We study a range of membrane voltage biases between \(-0.8 \text{ and } 0.8 \text{ V}\) applied to the n-Si and p-Si layers, and denoted \( V_n \) and \( V_p \), respectively.

Throughout the work, we follow a color scheme in which data pertaining to the DC pore with \( D = 10 \text{ nm} \) are drawn in blue, data for the SCp pore with the same \( D \) are in green, and data for the SCn pore are in red. The common cylindrical case \((D = 2 \text{ nm})\) is shown in black. The data for all other shapes with intermediate diameters are shown in color variations between black and blue/green/red for DC, SCp, and SCn geometries, respectively.

used to accelerate convergence. To solve for the potential \( \phi(\vec{r}) \), equation (1) is first linearized. The linearized differential equation is solved using an implicit finite difference method \([26]\) that gives a system of linear equations. The iteration process is repeated until the relative difference in the potential values between two consecutive iterations does not exceed \( 10^{-7} \).
3. Results and discussion

3.1. Electrostatic potential and ionic concentration

The calculated electric potential through the center of the double-conical pore (DC) is shown in figure 2. Two distinct membrane biases with $V_n = -0.8$ V (figure 2(A)) and $V_n = 0.8$ V (figure 2(B)) are considered, with $V_p = 0$ V in both situations. The former produces negative potential in the pore on both sides of the membrane and the latter results in positive potential on the n-Si side of the membrane and negative potential on the p-Si side of the membrane inside the pore. Due to the presence of negative static charge on the surface of the nanopore, $V_n$ has a stronger effect on the potential compared to $V_p$ (results are not shown). In general, we find that large negative biases ($V_n \lesssim -0.8$ V) applied to the n-Si layer have similar effects on the potential in the pore as described above for the specific case of $V_n = -0.8$ V.

According to equations (12) and (13), negatively charged Cl$^-$ ions follow an opposite trend with $[\text{K}^+] < [\text{KCl}]_0$ for positive potential, and $[\text{K}^+] > [\text{KCl}]_0$ for negative potential. This is demonstrated in figures 2(B) and (D) where $[\text{K}^+]$ and $[\text{Cl}^-]$ concentrations are shown for the DC geometry ($D = 6$ nm). The resulting variation in ionic concentrations is between $0.02 \times [\text{KCl}]_0$ and $50 \times [\text{KCl}]_0$.

The potential profiles for single-conical pores are shown in figures 3(A)–(D). The membrane bias voltages $V_n$ and $V_p$ are the same as for the double-conical nanopore shown in figure 2. In figures 2 and 3, the common case of cylindrical nanopore geometry is shown by black curves. We observe that for large pore diameters $D$, $\phi(\vec{r}) \approx 0$ at the wide end of the pore, and $|\phi(\vec{r})|$ is largest at its narrow end. This is why $V_n$, when applied to the narrow end of the SCn pore, affects the potential much more strongly than $V_p$ in the SCP nanopore, where it is applied to the wide end (cf curves for diameter $D = 10$ nm in figures 3(B) and (D) (red color) with curves that correspond to the same diameter in figures 3(A) and (C) (green color)). This effect is described in more detail below, see the discussion for figure 5. Comparison of $V_n$ and $V_p$ in terms of their ability to control potential distribution is also provided there.
3.2. Characterization of the electrostatic potential in the nanopore

We now perform a general characterization of the potential distribution in the nanopore. For this purpose we record two values of the potential $\phi_i$, $i = n, p$, inside the nanopore near the centers of the n-Si and p-Si layers, $\phi_n = \psi(x = 26 \text{ nm})$ (marked by a solid square) and $\phi_p = \psi(x = 38 \text{ nm})$ (marked by an open square), respectively, see figures 4(A) and (B). Thus, the value of $\phi_n$ is mostly affected by $V_n$, while the influence of $V_p$ is minimized. Similarly, $\phi_p$ is controlled by $V_p$ and it depends weakly on $V_n$. Also, we found that the $\phi_n$ and $\phi_p$ potentials are close to the average potentials on the n-Si and p-Si sides of the pore, respectively.

Cylindrical nanopore. Due to the presence of the dopant ions, the n-Si side of the membrane has a positively charged depletion layer, whereas the p-Si side of the membrane has a negatively charged depletion layer near the membrane surface. Since the surface oxide layer (SiO$_2$) is negatively charged, it effectively negates the positively charged depletion layer on the n-Si side, while enhancing the negatively charged depletion layer on the p-Si side. Thus, when no membrane bias is applied, there is an excess of K$^+$ ions in the pore on the p-Si side of the membrane, while no excess ions are accumulated on the n-Si side of the membrane. The high concentration of potassium ions screens the electrostatic potential in the pore and makes it difficult to control the p-Si side of the pore with applied membrane bias, as shown below. To demonstrate this, we plot $\phi_n$ as a function of the applied membrane bias voltage $V_n$ (keeping $V_p = 0$) and $\phi_p$ as a function of $V_p$ ($V_n = 0$), see figure 4(C). We observe that the value of $\phi_n$ on the n-Si side rises quickly by about 80 mV while $\phi_p$ on the p-Si side grows more slowly by less than 25 mV over the same range of applied membrane biases.

Non-cylindrical nanopores. In figures 5(A)–(C) the potentials $\phi_n$ versus $V_n$ (with $V_p = 0$) and $\phi_p$ versus $V_p$ (with $V_n = 0$) for the non-cylindrical nanopore geometries with $D = 4$ and 6 nm are shown. We also show the potentials $\phi_p$ and $\phi_n$ calculated for the cylindrical nanopore, for reference purposes (black curves). We summarize the features found in figure 5 as follows.

First we notice that the increase in the nanopore diameter $D$ always leads to the shrinkage of the available potential range for all three nanopore geometries, cf curves with triangles ($D = 6 \text{ nm}$) and curves with squares ($D = 2 \text{ nm}$) in figures 5(A)–(C). This is because the increase in $D$ effectively positions the membrane further from the nanopore’s center axis and therefore allows for greater screening by the electrolyte solution. As a result, the electrostatic potential become less sensitive to the applied membrane bias. In addition, due to the negative surface charge, the potential $\phi_n$ on the n-Si side of the membrane is more sensitive to the applied bias voltage for all three geometries. At the same time, the variation of $\phi_p$ is restricted to about 10–15 mV, similar to what was found for the cylindrical nanopore (figure 4(C)), see the three bottom curves in figures 5(A)–(C).

Furthermore, out of all considered nanopore geometries besides the cylindrical case, it is the SCn nanopore, which has the smaller opening on the n-Si side of the membrane, that exhibits a wider range of accessible potentials and, thus, it is better suited for tunable control over the ionic current flowing through the nanopore, which is further demonstrated in section 3.3. Finally, it is possible that this nanopore (SCn) will also be better suited to control of the translocation of a large molecule permeating through the nanopore.
If we vary both membrane bias voltages ($V_n$ and $V_p$), we obtain a continuous range of potential differences $\Delta \varphi = \varphi_n - \varphi_p$ available for each structure, see figure 6. We can tune the applied membrane biases to obtain any specific potential difference $\Delta \varphi$ in the pore, within the available range. A particular value of $\Delta \varphi$ is closely related to a particular regime of ion filtering through the pore. For example, a large potential difference $\Delta \varphi$ results in a diode-like ionic current–voltage characteristic with a large rectification ratio, as shown in section 3.3. Similarly, $\Delta \varphi$ is also important for controlling biomolecule translocation through the pore. Larger $\Delta \varphi$ means that there is a larger electric field and stronger force applied to a charged molecule. As such, we are interested in the nanopore membranes with the largest range of $\Delta \varphi$ values and, according to figure 6, these are the cylindrical nanopore and the single-conical nanopore with the smaller opening on the n-Si side of the membrane (SCn).

3.3. Ionic current–voltage characteristics and rectification ratios

We use a previously described model [20, 21, 27] to estimate the ionic current $I$ through the pore versus external electrolyte bias $V$, at constant membrane voltage bias. In this model we assume that the ionic fluxes are governed by the Nernst–Planck equation with contributions from ion diffusion and drift. The total ionic current through the nanopore is given by the sum of the fluxes for all ionic species. In calculations of ionic fluxes, we use the electrostatic potential, which is the sum of the equilibrium self-consistent potential obtained from the solution of the Poisson equation (1) and the potential due to the electrolyte bias $V$ ($V$ is assumed to drop linearly across the membrane).

The ionic current–voltage characteristics that we obtained using this approach are shown in figure 7 for different nanopore geometries. All subplots in figure 7 contain two current–voltage characteristics calculated for (1) $(V_n, V_p) = (-0.8, 0.4) \text{ V}$ (green squares), and (2) $(V_n, V_p) = (0.8, -0.8) \text{ V}$ (red circles).
We calculated the rectification ratio \( RR \) for each current–voltage curve as a ratio of ionic currents at \( |V_0| = 0.12 \) V: \( RR = I_+ (V_0) / I_- (V_0) \). In figure 8, the rectification ratios as a function of the potential variation in the pore are shown for all considered nanopore geometries. We find that the rectification ratio depends on the variation of the electrostatic potential in the nanopore, \( \Delta \phi = \phi_n - \phi_p \), and for all nanopore geometries this dependence is close to linear. Thus, high values of \( RR \) are found for membranes with large \( \Delta \phi \) with diode-like \( I-V \) characteristics while small \( RR \)s are obtained for membranes with small \( \Delta \phi \) and linear (Ohmic) current dependences. We see that the degree of current rectification varies among the nanopore geometries. For cylindrical nanopores (figure 7(A)) the rectification ratios are the greatest (maximum \( RR \sim 12 \)). For non-cylindrical nanopores with the same \( D \), the rectification ratio is the largest for the SCn nanopore, as seen in figures 7(B)–(D). The data shown in the plots also indicate a wider range of regimes accessible via variations in membrane biases for the cylindrical and SCn nanopores. This also corroborates our earlier assessment that single-conical nanopores with narrow openings in n-Si are better suited for control of a large molecule translocation.

4. Conclusion

In this work we performed a general study of nanopores in a semiconductor membrane made of two layers of n-doped and p-doped Si material. In particular, we assessed our ability to change the nanopore potential by applying voltage to the semiconductor membrane layers for different nanopore geometries: cylindrical, double-conical and single-conical. We considered only small pores with diameters of 2–10 nm, such that the electric field induced by the n-Si and p-Si layers significantly affects the potential distribution in the pore, and consequently the pore’s ionic conductivity. We demonstrate how variable voltages at the semiconductor layers, \( V_n \) and \( V_p \), allow us to manipulate the potential, changing the pore’s potential landscape, which gives rise to Ohmic (linear) or diode-like ionic current–voltage characteristics.

The data we obtained show a clear difference between current–voltage characteristics for pores with different diameters \( D \). At the same time, the effect of the charge situated in the membrane as well as due to the applied potential bias, doping, surface, etc can also be significant. We find that the single-conical membrane with a narrow pore opening on the n-Si side allows for a wider range of potential variation in the pore and, as such, can be better suited to exert control over a single molecule translocating through the pore. This is the case for our relatively small pores, the diameter of which is comparable to the Debye length for physiological solution strengths (1 mM–1 M).

In this work we focused on how the nanopore shape affects the \( I-V \) curves and potentials; however, as a next step, we also plan to study the effects on the ionic nanopore currents due to the membrane thickness, doping value, solution concentration and membrane surface charge, all of which can be easily incorporated in our model.

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References