FET pH Sensor Model Version 1.0.1

Piyush Dak and Muhammad Ashraful Alam Purdue University West Lafayette, IN 47907 Last Updated: Sep 11, 2014

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1. Introduction

The potential of hydrogen (pH) of a solution is a measure of the concentration of hydronium ions (H⁺) of a solution and is an important marker for many biological and chemical systems. The FET pH sensor model describes operation a field-effect transistor (FET) that can monitor the pH of a solution. The model is versatile and can be used for different classes of FET pH-sensors. In this manual, we will discuss the general features of this model, and describe how to use the model to characterize the response of the FET based pH sensors, and provide a scheme for extracting the parameters from the measurements. Finally, we will conclude with examples of circuit level simulation using the model. To test all the simulations described in the manual, the user should run "*perform_analysis.m*" file in Matlab (see Appendix B, C & D). The circuit simulations have been tested using HSPICE version E-2010.12-SP2. We use different FET models based on the device geometry (e.g. single gate or double gated devices). The optimization routine has been tested on Matlab version R2013a.

2. Terminal Voltages and Parameter List



2.1. Definition of Terminal Voltages

Fig. 1 (a) Equivalent circuit representation of electrolyte and surface groups on the oxide/functionalized layer. The pH acts as a terminal which controls the voltage difference between fg and geff *i.e.* $\psi_0 = V_{geff} - V_{fg}$. (b) Representation of a pH sensor. The pH controlled voltage source and the transistor are treated as separate circuit elements which are connected as shown.

The FET based pH sensor is modeled as two decoupled circuit elements *i.e.* a pH dependent nonlinear voltage source and a transistor, see Fig. 1.

- 1. **pH dependent voltage source**: The potential across the electrolyte i.e. the difference between the fluid gate voltage (V_{fg}) and effective gate voltage (V_{geff}) is a function of the pH, see Fig. 1a. Therefore, the pH dependence of the sensor can be captured using a pH dependent voltage source (ψ_0). For circuit simulation, pH can be represented as an external voltage source (V_{pH}). Physically, pH is not a voltage source in the same sense a battery is, but its effect on the transistor I-V characteristics can be viewed as a voltage source in the compact model. The definition of pH as a voltage source enables transient and small signal analysis with respect to pH in the circuit simulator.
- II. **Transistor**: The second element in Fig. 1 (b) involves a classical MOSFET transistor with gate, source, and drain terminals. It can be any MOS transistor which is supported by the circuit-simulator such as long-channel MOSFET and a double gated FET, etc. or more generally, an externally defined Verilog-A MOSFET model, such as MIT Virtual Source

Model[1]. The gate of the transistor is actuated by V_{geff} which is responsive to the pH of the solution. In Fig. 1 (b), the transistor is depicted with 4 terminals, source (V_s), drain (V_d), gate (V_{geff}) and bulk/back gate (V_{bg}).

We will define the functional relationship between pH of a solution and the voltage source (ψ_0) in Sec. 3; but first we need to define the parameters and physical quantities of interest. Nodes and corresponding node voltages are as follows:

| Node | Description | Voltage |
|--------|------------------------|-----------|
| pHnode | $-\log_{10}[H^+]$ | V(pHnode) |
| fg | Floating gate voltage | V(fg) |
| geff | Effective gate voltage | V(geff) |

2.2. Fundamental/Material Constants:

Following fundamental or material constants are used for in the code.

| Math | Verilog-A | Definition | Description | Value | Unit |
|------------------|-----------|--------------|---------------------------------|------------------------|------------------|
| Symbol | Symbol | Туре | | | |
| N _{avg} | `Navg | User defined | Avogadro's constant | 6×10^{26} | #/m ³ |
| ϵ_r | `RP_EPSw | User defined | Relative permittivity of buffer | 80 | - |
| | | | solution (water) | | |
| q | `P_Q | Inbuilt | Electronic Charge | 1.6×10^{-19} | С |
| ϵ_0 | `P_EPSO | Inbuilt | Permittivity in vaccum | 8.85×10^{-12} | F/m |
| v _t | \$vt | Inbuilt | Thermal voltage at current | ~0.026 | V |
| | | Function | temperature | | |

2.3. Parameter List

Parameters used in the pH-FET sensor model are listed below. Parameter highlighted in bold need to be optimized. Table also lists the physical meaning of each parameter.

| Math | Verilog-A | Description | Default | Unit |
|------------------------|-----------|-----------------------------------------------------------------------|--------------------|------------------|
| Symbol | Symbol | | | |
| <i>pK</i> _a | рКа | $-\log_{10}[K_a]$ | | - |
| | | where $K_a = \frac{[AOH][H_s^+]}{[AOH_2^+]}$ is the acid dissociation | -2.0 | |
| | | constant for the surface group | | |
| <i>pK</i> _b | рКb | $-\log_{10}[K_b]$ | | - |
| | | where $K_a = \frac{[AO^-][H_s^+]}{[AOH]}$ is the base dissociation | -6.0 | |
| | | constant for the surface group | | |
| N _{OH} | NOH | Density of the surface groups | 5×10^{14} | cm ⁻² |
| i ₀ | i0 | Ionic concentration of the buffer solution | 0.1 | Molar |
| C _{stern} | Cstern | Stern layer capacitance | 0.2 | F/m ² |
| _ | sternmod | sternmod=0 for using GC model, sternmod=1 for using GCS model | 1 | - |

2.4. Derived Variables

| Math | Verilog-A | Formula | Description | Unit |
|--------------|-----------|---------------------------------------|----------------------------------|------------------|
| Symbol | Symbol | | | |
| pH_{pzc} | pzc | $(pK_a + pK_b)/2$ | pH value at which the surface | - |
| £ | | | charge becomes zero | |
| ΔpK | deltapK | $pK_b - pK_a$ | Difference between the pK | - |
| | | | values | |
| $Q_{0,dl}$ | Q0dl | $\sqrt{8\epsilon_w v_t q n_0}$ | The double layer charge density | F/m ² |
| | | | at zero surface potential | |
| ΔpH | deltapH | (pH - pzc) | pH relative to pzc <i>i.e.</i> | - |
| | | | deltapH=pH-pzc | |
| - | NOH_SI | $N_{OH} 	imes 10^4$ | Density of surface groups in SI | #/m ² |
| | | | units | |
| n_0 | n0 | $N_{AVG} \times i0$ | Ionic concentration | #/m ³ |
| ψ_0 | psi0 | See Section 3 | Potential difference between the | V |
| | | | surface and the reference | |
| | | | electrode | |
| C_{dl} | Cdl | $\sqrt{8\epsilon_w v_t q n_0}/(2v_t)$ | Double layer capacitance at zero | F/m ² |
| | | • | surface potential | |
| δ | delta | $2 \times 10^{-\frac{\Delta pK}{2}}$ | - | - |
| β | beta | See Section 3 | Buffer capacity of the surface | - |
| α | alpha | $\beta/(1+\beta)$ | - | - |
| ϵ_w | ew | $\epsilon_r \times \epsilon_w$ | Permittivity of buffer solution | F/m |
| | | | (water) | |

Table below lists the derived variables used in the Verilog-A implementation of the model.

3. Model Equations

As stated earlier, the electrolyte combined with the interface charges is equivalently modeled as a pH dependent voltage source. Two different versions of model are introduced: First, a simplified pH sensor model [2] which relates the surface potential of the MOSFET as a linear function of the change in pH above its point of zero charge (pzc). The second model is more accurate and relates the surface potential as an implicit function of pH of the buffer solution (see, Appendix A for derivation). Further, each of these models have two different options, namely to use the Guoy-Chapman (GC) model or Guoy-Chapman-Stern (GCS) model [3] for computation of the surface potential. In GC theory, the ions are considered as point charges that can approach arbitrary close to the surface. This may causes unrealistic high concentrations of ions near interface for high surface potentials. The GCS model corrects this by introducing a dielectric layer between interface and diffuse layer to limits the point of nearest approach of ions. The equations describing each of these models are as follows:

3.1. Simplified pH sensor model:

The effective gate potential (V_{geff}) for the transistor is calculated by using the following set of equations. Here, pH is the input parameter; other parameters characterize the bulk fluid and the interface between the gate oxide and the fluid. The parameters are defined in Sec. 2.

$$V_{geff} \equiv V_{fg} - \psi_0$$

$$\psi_0 \equiv -\log_e 10 \times v_t \times \alpha \times (pH - pH_{pzc})$$

where,

$$\alpha \equiv \frac{\beta}{1+\beta}$$

with β defined as follows for 2 different models:

3.1.1. With Guoy-Chapmen Model:

$$\beta = q \, N_{OH} \times \frac{\delta}{C_{dl} \times v_t}$$

3.1.2. With Guoy-Chapmen Stern Model:

$$\beta = q \; N_{OH} \times \frac{\delta}{C_{eq} \times v_t}$$

with,

$$C_{eq} = \frac{C_{dl} \times C_{stern}}{(C_{dl} + C_{stern})}$$

and,

$$\delta = 2 \times 10^{-\frac{\Delta pK}{2}}$$

This model works well only in a limited range of ψ_0 such that $|\psi_0| \ll \frac{kT}{q}\beta$ [2]. In order to get more accurate result, the robust pH sensor model described next must be used.

3.2. Robust pH sensor model

3.2.1. With Guoy-Chapmen Model:

$$\psi_{0} = 2 v_{t} \operatorname{asinh}\left(-\frac{2qN_{OH}}{Q_{0,dl}}\frac{\left(\tanh\left(\frac{\psi_{0}}{v_{t}} + \log_{e}10 \times \Delta pH\right)\right)}{\left(2 + c \times \sqrt{1 - \left(\tanh\left(\frac{\psi_{0}}{v_{t}} + \log_{e}10 \times \Delta pH\right)\right)^{2}\right)}\right)\right)$$

ere $c = exp\left(\log_{e}10 \times \frac{\Delta pK}{2}\right)$, $Q_{0,dl} = \sqrt{8\epsilon_{e}v_{t}qn_{0}}$, and $\Delta pH = pH - pH_{res}$

where $c = exp\left(\log_e 10 \times \frac{\Delta pK}{2}\right)$, $Q_{0,dl} = \sqrt{8\epsilon_w v_t q n_0}$, and $\Delta pH = pH - pH_{pzc}$

3.2.2. With Guoy-Chapmen Stern Model:

$$\psi_0 = 2 v_t \operatorname{asinh}\left(\frac{Q_{OH}}{Q_{0,dl}}\right) + \frac{Q_{OH}}{C_{stern}}$$

where,

$$Q_{OH} = -2qN_{OH} \frac{\left(\tanh\left(\frac{\psi_0}{v_t} + \log_e 10 \times \Delta pH\right)\right)}{\left(2 + c \times \sqrt{1 - \left(\tanh\left(\frac{\psi_0}{v_t} + \log_e 10 \times \Delta pH\right)\right)^2}\right)}$$

4. Parameter Extraction Procedure

A key challenge of any compact model development is to characterize the parameters of the model (see, Sec. 3) by interpreting the experimental data. We suggest two different approaches:

4.1. Approximate Extraction Method

When the available experimental data is limited (the response of the pH sensor is available only for few pH values), the best way to extract parameters is to simulate the response of the sensor for different sets of input parameters and compare the results with experimental data. A reasonably good match between simulation and experiment identifies the desired parameter set. By this method, parameters roughly close to their accurate value can be obtained. Fig. 2 shows the match of the experimental data obtained from Go *et. al.* [4] with the compact model.



Fig. 2 Match of experimental data with the robust Verilog-A pH sensor and BSIM SOI model. The model gives consistent results with experimental data.

The parameters used for the fit are listed in Table below.

| Parameter | Value | Ref |
|------------|------------------------------------|------------|
| u0 * (W/L) | 16 | Calibrated |
| vth0 | 1.475 | Calibrated |
| рКа | 6 | [5] |
| pKb | 10 | [5] |
| NOH | $8 \times 10^{14} \text{ cm}^{-2}$ | [5] |

4.2. Least Square-Fit Extraction method

This is more robust and accurate way of extracting the parameters. The process of parameter extraction is divided into two parts:

Step1: Extraction of FET specific parameters:

In order to determine the FET specific parameters, the FET device with metal gate is to be fabricated within the same chip as the pH sensing device. The device parameters can be obtained using the extraction procedure provided in the user manual for NEEDs Silicon MIT Virtual Source Model by Rakheja *et al.* [1]. For the current work, it is assumed that these device parameters have already been obtained.

Step2: Extraction of pH specific parameters:

Three pH specific parameters need to be optimized i.e. pK_a , pK_b and N_{OH} . These parameters depend on the type of the oxide or the functionalized metal layer used to do pH sensing. For a known oxide (functionalized surface), these parameters are more or less constant and can be obtained from literature. However for accurately representing the device characteristics, it is good to optimize these parameters. The lower and upper bounds of the parameters and their initial guesses are mentioned in the Table below.

| Parameter | Lower Bound | Upper Bound | Initial Guess |
|-----------|-------------|-------------|---------------|
| рКа | -15 | 15 | -1 |
| pKb | -15 | 15 | 4 |
| NOH | 2e14 | 8e14 | 3e14 |

a) Description of the parameter extraction routine

The following files are required to extract the pH specific parameters.

1. run_optimizer.m

This file runs the optimizer for extraction of pH dependent parameters. It takes a set of $I_{ds} - V_{fg}$ characteristics for different pH and ionic concentration values and runs the *determine_threshold_voltage.m* file to extract threshold voltage for different curves. The change in surface potential $(\Delta \psi_0) = -\Delta V_T$. This change along with the pH and i0 values are fed into the *optimize_pHparameters.m* file to obtain the optimized parameters. Once the optimized parameters are obtained, these are manually updated into Verilog-A file "*experiment_mvs.sp*". Then HSPICE simulation is run to update the output files.

2. optimize_pHparameters.m:

Input Parameters: coeff_init (Initial guess of the pH parameters in order pKa, pKb, NOH), xdata (with column 1 containing pH values and column 2 containing i0 values at which the experiment is performed).

Output Parameters: coeff_deltaVt (optimized pH parameters)

3. surface_potential_shift.m: This file determines the change in surface potential relative to the minimum pH and maximum i0 value.

Input Parameters: x (array containing the pH parameters in order pH, pKb and log10(NOH)), pH_i0 (array with column 1 containing pH values and column 2 containing corresponding i0 values)

Output Parameters: deltapsi0 (change in surface potential relative to minimum pH and maximum i0 for the input array provided).

4. **pH_ robust_model_1_0_1.m:** This is the Matlab implementation of the Verilog-A pH sensor model and solves the model equations described in "Robust pH sensor model" to determine the output parameters.

Input Parameters: pH, pKa, pKb, NOH, i0, Cstern, sternmod

Output Parameters: psi0 i.e. V(fg,geff)

5. **determine_threshold_voltage.m:** This Matlab function determines the threshold voltage of the MOSFET. type=1 to determine constant current threshold voltage with Icons being the value of the constant current. Use type=2 to determine the linear threshold voltage.

Input Parameters: Vgs, Ids, type, Icons, Vrange (optional)

Output Parameter: Vt

Input file format:

Input file must be provided in the following format:

Transfer Characteristics $(I_{ds} - V_{fg})$ with respect to the fluid gate bias

| V_{fg} (V) | $I_{ds}(A)$ (at low V_{ds}) | $I_{ds}(A)$ (at high V_{ds}) |
|--------------|--------------------------------|---------------------------------|
| | | |
| | | |
| | | |
| | | |
| | | |

Since, the threshold voltage change is independent of the gate bias as the pH changes, therefore, the third column which contains the $I_{ds} - V_{fg}$ characteristics at high V_{ds} can be eliminated. Sample experimental (synthetic) data is provided in folder:

"pH_model_1_0_1_experimental_data/wet_measurement/"

b) Parameter Optimization Results

In order to optimize the pH sensing parameters, it is important to have experimental data for a wide range of pH values (pH=1 to 13). The optimizer can work with limited range as well, albeit with reduced accuracy for the back extracted parameters. For the following illustration, we use a synthetic experimental dataset for transfer characteristics (with respect to fluid gate). The difference in threshold voltage give the change in the surface potential relative to a reference pH value. This change in surface potential is optimized using optimize_pHparameters.m as shown in Fig. 3 (a). The optimized parameters are extracted to a file "optimized_parameters_pH.txt".

Once the pH parameters are optimized and FET parameters are known (through pre-optimization of dry measurement data), pH sensor model is complete. Fig. 3 (a) shows the match of the pH sensor model with the experimental data.



Fig. 3 (a) Match of change in threshold voltage as a function of pH. (b), (c) Match of transfer characteristics for different pH values with the pH sensor model.

5. Circuit Simulation

5.1. Sensitivity enhancement in Double-Gated FET pH sensor

The sensitivity of a single gated FET pH sensor is limited by Nernst Limit (59 mV/dec at room temperature). This limit can be overcome by using a double gated FET as reported in literature [4]. In DGFET pH sensor, one sweeps the metal gate (poly-silicon gate), instead of the fluid gate, to obtain the transistor characteristics $(I_{DS} - V_{MG})$. A fixed bias is applied to the fluid gate (V_{LG}), and the pH sensitivity is measured in terms of the threshold voltage shift of the metal gate ($\Delta V_{T,MG}/\Delta pH$). To enable higher sensitivity, the metal gate capacitance (C_{MG}) should be larger than the fluid gate capacitance (C_{LC}). The maximum sensitivity that can be achieved with metal gate operation is given by, S = $\frac{\Delta V_{T,MG}}{\Delta pH} = \frac{C_{MG}}{C_{LG}} \frac{\Delta V_{T,LG}}{\Delta pH}$, where $\frac{\Delta V_{T,LG}}{\Delta pH}$ is the pH sensitivity (limited by Nernst Limit) with fluid gate operation and a constant metal gate voltage. This high sensitivity occurs when both the fluid gate channel and metal gate channel are inverted. Fig. 4 (a) and (b) shows the comparison of $I_{ds} - V_{gs}$ characteristics for 2 different fluid gate voltages. Fig. 4 (c) and (d) show the corresponding change in threshold voltage as a function of pH. The device sensitivity (S) increases by almost 5x as the fluid gate voltage changes from -0.8V (accumulation-inversion) to 1.0V (depletion-inversion mode). The sensitivitv amplification was tested using BSIM SOI FD model in HSPICE.



Fig. 4 Sensitivity enhancement in a double gated n-FET pH sensor. (a), (b) Transfer characteristics with respect to metal gate voltage for two different fluid gate voltages. (c), (d) Constant current (1 nA) threshold voltage as a function of pH for the two cases. Sensitivity increase by 5X as V_{LG} is increased from -0.8 V to 1V.

5.2. Response of ISFET to stepwise change in pH

As an example of the robustness of the model, we simulated the response of n-channel, partially depleted (PD) SOI MOSFET in response to a stepwise change in pH value. Fig. 5 (a) shows the netlist, the input pH value as a function of time and output current as a function of time. When pH is high (pH=8), the proton ion concentration in bulk is small; therefore, the surface is deprotonated giving a more negative charge on the surface (with respect to point-of-zero charge). This leads to lesser channel conductance and hence small current (see, Fig. 5 (c)). When pH decreases to a smaller value (pH=3), the surface of the oxide becomes positively charged, and hence results in a higher drain current. For the transient simulation, BSIM SOI PD model was used.



Fig. 5 (a) Circuit diagram. (b) Input pH as a function of time (c) Output drain current as a function of time.

6. Summary

The manual describes the generalized FET based pH sensor model. The implementation of the model in Verilog-A as well as the parameter extraction procedures are discussed. When the available experimental data are limited, then the parameters can be obtained by directly comparing the simulation data (for different sets of parameters) with the experimental data. However, to obtain precise value of model parameters it is recommended to obtain characteristics for both dry measurement (without fluid gate but same transistor) and wet measurement (with different pH and i0 values) and do a least-square minimization to obtain the best model fit parameters. The model also illustrates the sensitivity amplification in DGFET pH sensor. Future work involves inclusion of CV analysis for pH sensing model, inclusion of finite ion size effects in the model and inclusion of DNA sensing model. Please contact Piyush Dak (piyushjdak@gmail.com) regarding any questions/comments about the FET pH sensor model.

References:

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Appendix

A. Derivation of the surface potential across the electrolyte:

$$AOH^{2+} \xrightarrow{K_a} AOH + H_s^+$$
 [1]

$$K_{a} = \frac{[AOH][H_{s}^{+}]}{[AOH^{2+}]}$$
[2]

$$AOH \xrightarrow{K_b} AO^- + H_s^+$$
 [3]

$$K_{b} = \frac{[AO^{-}][H_{s}^{+}]}{[AOH]}$$
^[4]

$$[H_s^+] = [H^+] \exp\left(-\frac{q\psi_s}{kT}\right)$$
^[5]

$$\sigma_{\rm dl} = -\sigma_{\rm dl,0} \sinh\left(\frac{q\psi_0}{2kT}\right)$$
[6]

$$\sigma_{OH} = q([AOH^{2+}] - [AO^{-}])$$
^[7]

$$[AOH2+] + [AOH] + [AO-] = NOH$$
 [8]

By using (2), (4), (7) and (8), we get

$$[AOH] = \frac{N_{OH}}{\left(\frac{[H_s^+]}{K_a} + \frac{K_b}{[H_s^+]} + 1\right)}$$
[9]

This gives,

$$\sigma_{OH} = q N_{OH} \frac{\left(\frac{[H_s^+]}{K_a} - \frac{K_b}{[H_s^+]}\right)}{\left(\frac{[H_s^+]}{K_a} + \frac{K_b}{[H_s^+]} + 1\right)}$$
[10]

$$= -2qN_{OH}\frac{(\tanh(\beta\psi_s + \log 10 \times \Delta pH))}{(10^{\Delta pK/2}\operatorname{sech}(\beta\psi_s + \log 10 \times \Delta pH) + 2)}$$
[11]

$$\psi_s = \psi_0 + \frac{\sigma_{OH}}{C_{stern}}$$

 $\sigma_{OH} + \sigma_{dl} + \sigma_{FET} = 0$

Since,

$$\sigma_{FET} \ll \sigma_{dl}$$

$$\psi_0 = -\frac{2kT}{q} \operatorname{asinh}\left(\frac{\sigma_{dl}}{\sigma_{dl,0}}\right)$$
$$\Rightarrow \psi_s = \frac{2kT}{q} \operatorname{asinh}\left(\frac{\sigma_{OH}}{\sigma_{dl,0}}\right) + \frac{\sigma_{OH}}{C_{stern}}$$

B. HSPICE toolkit

In order to analyze the HSPICE output data, HSPICE toolkit available in Matlab is used. This can be downloaded from <u>http://www.cppsim.com/InstallFiles/hspice_toolbox.tar.gz</u>. Make sure to add it in the Matlab path. You can use the following Matlab command to add this to the default path: addpath(*location_of_hspice_toolbox_folder*)

C. Simulating HSPICE netlist:

HSPICE code can be run on file "filename.sp" using following command: % hspice filename.sp

D. Checklist for model analysis:

- 1. Install the HSPICE toolkit to enable data extraction from HSPICE files.
- 2. Download the complete package in folder FETpH. Compile the following HSPICE files (in case the user needs to change any parameters)
 - a. Approximate parameter extraction: FETpH\pH_model_1_0_1_HSPICE_netlists\go_match\ experiment_go.sp
 - b. **Transient Analysis:** FETpH\pH_model_1_0_1_HSPICE_netlist\transient\pHtransient.sp
 - c. Least-square fit parameter extraction: FETpH\pH_model_1_0_1_HSPICE_netlists\mvs_match\experiment_mvs.sp
 - d. DGFET pH sensor: FETpH\pH-model_1_0_1_HSPICE_netlist\dgfet_amplification\backgate_dgfet.sp

Compilation of these files will generate the FILENAME.sw* files which will be analyzed using the perform_analysis.m file.

3. Go to the folder containing the "perform_analysis.m" file and run it. Make sure that the utility file determine_threshold_voltage.m is within Matlab path.