

Two-Phase Flow Modeling of Copper Electrowinning Using Bubbly Flow

Introduction

Copper electrowinning is the process of copper extraction from an electrolyte solution, and its subsequent deposition at the cathode surface, by passing an external current through the electrolytic cell. In the cell, an insoluble anode is typically used. During the process, oxygen bubbles are generated at the anode surface leading to a large recirculation zone between the anode and cathode surfaces.

In the model example presented here, charge and mass transports of ionic species are modeled using the Tertiary Current Distribution, Nernst-Planck interface and the twophase flow generated due to oxygen gas evolution at the anode surface is modeled using the Bubbly Flow, Laminar Flow interface.

The model example is based on a scientific paper (Ref. 1).

Model Definition

The electrowinning cell model geometry is shown in Figure 1. The two vertical boundaries represent the anode and the cathode surfaces, respectively. Since the oxygen gas evolves at the anode surface, this boundary also acts a gas inlet. The top boundary

serves as an inlet for the liquid phase and an outlet for the gas phase. The outlet boundary for both the liquid and gas phases is as shown Figure 1.

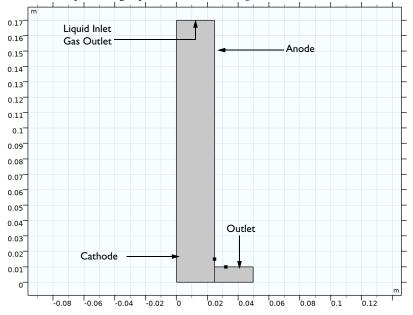


Figure 1: Model domain with boundaries corresponding to the anode, cathode, and vertical symmetry walls.

Mass transport by convection, diffusion and migration for three ionic species (copper, Cu²⁺, hydrogen, H⁺and bisulfate, HSO₄⁻) is solved using the Tertiary Current Distribution, Nernst-Planck physics interface:

$$\frac{\partial \varepsilon_l c_i}{\partial t} + \nabla \cdot \mathbf{N}_i + \mathbf{u} \cdot \nabla c_i = 0$$

where ε_l is the electrolyte volume fraction. ε_l is defined in terms of the liquid phase fraction which is solved for in the Bubbly Flow, Laminar Flow interface, as discussed later in this section.

The flux for each of the ions in the electrolyte is given by the Nernst–Planck equation:

$$\mathbf{N}_{i} = -D_{i, \text{eff}} \nabla c_{i} - z_{i} u_{i, \text{eff}} F c_{i} \nabla \phi_{l}$$

where \mathbf{N}_i denotes the transport vector (mol/(m²·s)), c_i the concentration in the electrolyte (mol/m³), z_i the charge for the ionic species, $u_{i,\text{eff}}$ the mobility of the charged

species (m²/(s·J·mole)), F Faraday's constant (As/mole), and ϕ_l the potential in the electrolyte (V).

The transport properties such as diffusion coefficients are defined using the electrolyte volume fraction:

$$D_{i, \text{ eff}} = \varepsilon_l^{1.5} D_i$$

The velocity field used in the mass transport equation comes from the Bubbly Flow, Laminar Flow interface and is corrected for the electrolyte volume fraction.

The electroneutrality condition is given by the following expression:

$$\sum_{i} z_{i} c_{i} = 0$$

ELECTROCHEMICAL REACTIONS

The copper reduction and the oxygen evolution due to water decomposition are the two electrochemical reactions considered in the copper electrowinning model presented here.

The copper reduction at the cathode surface proceeds as:

$$Cu^{2+} + 2e^{-} \rightarrow Cu$$

The oxygen evolution at the anode surface proceeds as:

$$2H_2O \rightarrow 4H^+ + O_2 + 4e^-$$

Concentration dependent kinetics is used to model the copper reduction and oxygen evolution oxidation reactions, which will set the local current density according to:

$$i_{\rm loc,\,m} = i_{0,\,\rm m} \Big(C_{\rm R,m} \exp \Big(\frac{\alpha_{\rm a,m} F \eta_{\rm m}}{RT} \Big) - C_{\rm O,m} \exp \Big(-\frac{\alpha_{\rm c,m} F \eta_{\rm m}}{RT} \Big) \Big)$$

where $i_{0,m}$ is the exchange current density, $C_{R,m}$ is the reduced species expression, $C_{O,m}$ is the oxidized species expression, $\alpha_{a,m}$ is the anodic transfer coefficient, $\alpha_{c,m}$ is the cathodic transfer coefficient, and η_m is the overpotential for species m (Cu and H, respectively).

The overpotential $\eta_m(V)$ is calculated from:

$$\eta_{\rm m} = \phi_{\rm s, ext} - \phi_l - E_{\rm eq, m}$$

The total current of 5.14 A is applied at the anode surface which is used to calculate the external electric potential, $\phi_{s, ext}$.

The equilibrium potentials for the copper reduction and oxygen evolution oxidation reactions are calculated using the Nernst equation:

$$E_{\text{eq,Cu}} = E_{\text{eq, Cu}}^0 + \frac{RT}{nF} \ln \left(\frac{c_{\text{Cu}}}{c_{\text{Cu,ref}}} \right)$$

$$E_{\text{eq,H}} = E_{\text{eq,H}}^0 + \frac{RT}{nF} \ln \left(\frac{c_{\text{H}}}{c_{\text{H,ref}}}\right)^4$$

where $E_{
m eq,\,Cu}^0$ and $E_{
m eq,H}^0$ are the standard equilibrium potentials for the copper reduction and oxygen evolution oxidation reactions and are considered to be 0.34 V and 1.23 V, respectively. Moreover, $c_{\mathrm{Cu,ref}}$ and $c_{\mathrm{H,ref}}$ are the reference concentrations for copper and hydrogen ions, respectively.

At the anode and cathode surface boundaries, fluxes of ionic species are defined in terms of the electrochemical reactions as:

$$\mathbf{n} \cdot \mathbf{N}_{\mathrm{m}} = \frac{v_{\mathrm{m}} i_{\mathrm{loc, m}}}{n_{\mathrm{m}} F}$$

where v_{m} is the stoichiometric coefficient, $i_{\mathrm{loc,m}}$ is the local current density, n_{m} is the number of electrons, and F is Faraday's constant (96,485 C/mol). This will set the flux to be proportional to the electrode current density according to Faraday's law.

The electrode kinetics parameters: $i_{0 \text{ Cu}}=100 \text{ A/m}^2$ and $i_{0 \text{ H}}=3\times10^7 \text{ A/m}^2$ are taken from Ref. 1.

BUBBLY FLOW INTERFACE

The Bubbly Flow interface sets up a two-phase flow model for oxygen gas bubbles in a liquid electrolyte. The physics interface tracks the averaged gas-phase concentration rather than each bubble in detail. The physics interface solves for the liquid velocity, the pressure, and the volume fraction of the gas phase. Details of the governing equations are presented in the theory section for the Bubbly Flow interfaces in the CFD Module User's Guide.

The Physical Model settings for the Bubbly Flow interface provides a low gas concentration option which is active per default. This option is applicable if the gas concentration is about 1%, in which case the transport equations can be simplified compared to cases with higher gas concentrations. Considering the possibility of higher gas-phase fraction, particularly near the anode surface, the low gas concentration option is disabled in this model.

For demonstration purpose, density and viscosity of the liquid phase are assumed to be uniform and are considered to be 1200 kg/m³ and 0.835×10^{-3} kg/m/s, respectively. However, they can be set to be dependent upon the copper concentration.

For laminar flow the gas velocity \mathbf{u}_{g} is calculated from:

$$\mathbf{u}_{g} = \mathbf{u}_{l} + \mathbf{u}_{slip}$$

where \mathbf{u}_{l} stands for the liquid-phase velocity, and \mathbf{u}_{slip} stands for the relative velocity between gas and liquid, the so-called slip velocity.

The slip velocity is calculated from a slip model. The Bubbly Flow interface provides several slip models. The most appropriate slip model for this electrowinning cell is a pressure-drag balance slip model with a drag coefficient tuned for small spherical bubbles using Hadamard–Rybczynski model. The gas bubbles diameter is considered to be 50 μm.

The top boundary of the electrowinning cell acts as an inlet for the liquid phase and as an outlet for the gas phase. The Inlet boundary feature is used here to set the normal inflow velocity to 0.0001 m/s and to set the gas outlet boundary condition.

The Outlet boundary feature is used to set zero pressure for the liquid phase and to set gas outlet boundary condition for the gas phase for the boundary shown in Figure 1.

The Wall boundary feature is used to set no slip boundary condition for the liquid phase and to set the gas mass flux at the anode surface as

$$-\mathbf{n} \cdot \mathbf{N}_{\mathcal{O}_2} = \frac{M w_{\mathcal{O}_2} i_{\text{loc}, H}}{4F}$$

where N_{O_2} is the gas mass flux for oxygen bubbles; Mw_{O_2} is the molar mass of oxygen; $i_{loc,H}$ is the local current density of oxygen evolution reaction evaluated at the Tertiary Current Distribution, Nernst-Planck physics interface; and F is Faraday's constant.

Results and Discussion

Figure 2 shows surface plot of the liquid phase velocity magnitude along with arrow plot of the liquid phase velocity field after 60 s of deposition operation. A large vortex can be seen at the top of the electrowinning cell in Figure 2. The higher magnitude of liquid phase velocity at the anode surface indicates the generation of oxygen bubbles due to water decomposition electrochemical reaction and their rise to the top of the cell due to buoyancy effects.

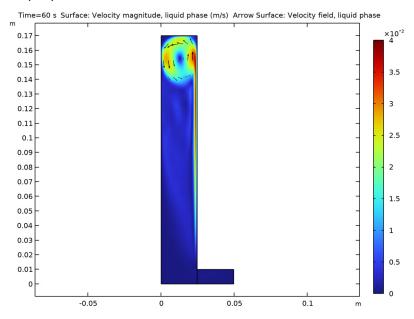


Figure 2: Surface plot of the liquid phase velocity magnitude along with arrow plot of the liquid phase velocity field after 60 seconds of deposition operation.

Figure 3 shows surface plot of the gas phase volume fraction along with streamline plot of the liquid phase velocity field after 60 s of deposition operation. The gas phase volume fraction is seen to be higher near anode surface in Figure 3, similar to the liquid phase velocity shown in Figure 2. The buoyancy force of oxygen bubbles generated at the anode

surface is a strong driving force for the fluid flow in the electrowinning cell. The highest magnitude of the gas phase volume fraction is found to be just over 0.02.

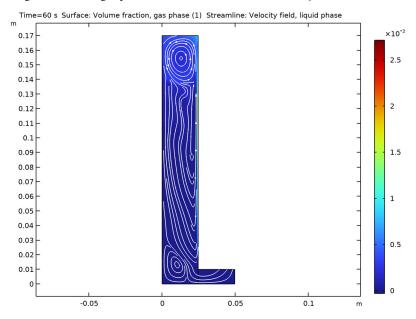


Figure 3: Surface plot of the gas phase volume fraction along with streamline plot of the liquid phase velocity field after 60 seconds of deposition operation.

Figure 4 shows surface plot of the copper concentration along with streamline plot of the total flux after 60 s of deposition operation. It can be seen that the copper concentration is quite uniform throughout the electrowinning cell due to a strong stirring effect of oxygen bubbles. The lowest copper concentration is observed at the cathode surface

(which is not clearly visible in Figure 4) since copper ions are consumed here during deposition.

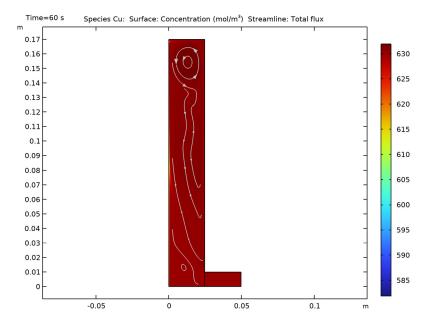


Figure 4: Surface plot of the copper concentration along with streamline plot of the total flux after 60 seconds of deposition operation.

To bring out the depletion of copper ions at the cathode surface, several line plots are plotted next for the copper concentration.

Figure 5 shows line plot of the copper concentration along the cathode surface after 60 s of deposition operation. It can be seen that the copper concentration varies considerably with distance from the bottom of the cathode surface. The peak in the copper

concentration at the top of the cathode surface corresponds to a vortex in the liquid phase velocity magnitude observed in Figure 2.

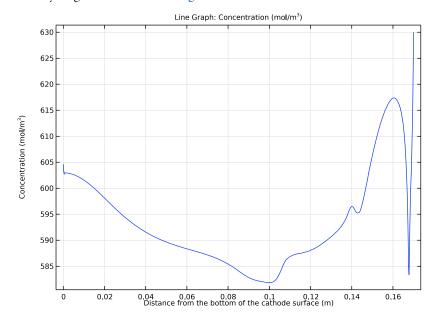


Figure 5: Line plot of the copper concentration along the cathode surface after 60 seconds of deposition operation.

Figure 6 shows line plot of the change in copper concentration with time for 60 s of deposition operation at three different locations on the cathode surface. It can be seen that the copper concentration attained a steady state at the top and middle of the cathode surface whereas it decreased continuously at the bottom of the cathode surface.

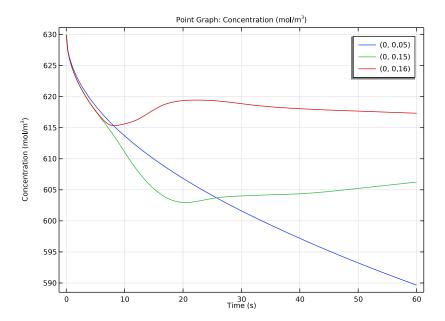


Figure 6: Line plot of the change in copper concentration with time at different locations on the cathode surface.

Figure 7 shows line plot of the change in copper concentration across the boundary layer thickness at a specific position on the cathode surface after 60 s of deposition operation. It can be seen that the copper concentration varies considerably across boundary layer thickness and it remains uniform toward the bulk of the electrolyte solution. This result

confirms that the mesh used in the model is fine enough to resolve the change in copper concentration across the boundary layer at the cathode surface.

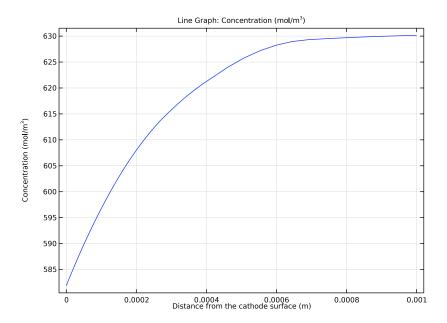


Figure 7: Line plot of the change in copper concentration within the boundary layer at a specific position on the cathode surface after 60 seconds of deposition operation.

Figure 8 shows line plot of the change in copper deposition thickness along the cathode surface after 60 s of deposition operation. It can be seen that the copper deposition is considerably higher at the top half of the cathode surface when compared to the bottom half. The peak in the copper deposition thickness at the top of the cathode surface corresponds to a vortex in the liquid phase velocity magnitude observed in Figure 2.

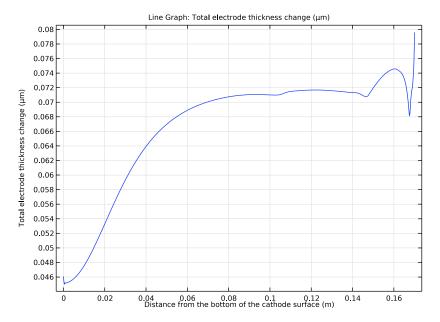


Figure 8: Line plot of the copper deposition thickness along the cathode surface after 60 seconds of deposition operation.

Reference

1. J.M. Werner, W. Zeng, M.L. Free, Z. Zhang, and J. Cho, "Modeling and Validation of Local Electrowinning Electrode Current Density Using Two Phase Flow and Nernst-Planck Equations," Journal of The Electrochemical Society, vol. 165, p. E190, 2018.

Application Library path: Electrodeposition Module/Tutorials/ cu_electrowinning_bubbly_flow

Modeling Instructions

From the File menu, choose New.



In the New window, click Model Wizard.

MODEL WIZARD

- I In the Model Wizard window, click **2** 2D.
- 2 In the Select Physics tree, select Electrochemistry>Tertiary Current Distribution, Nernst-Planck>Tertiary, Electroneutrality (tcd).
- 3 Click Add.
- 4 In the Number of species text field, type 3.
- 5 In the Concentrations (mol/m³) table, enter the following settings:

cCu cH cHS04

- 6 In the Select Physics tree, select Fluid Flow>Multiphase Flow>Bubbly Flow>Bubbly Flow, Laminar Flow (bf).
- 7 Click Add.
- 8 Click Study.
- 9 In the Select Study tree, select Preset Studies for Selected Physics Interfaces> Tertiary Current Distribution, Nernst-Planck>Time Dependent with Initialization.
- 10 Click Done.

GEOMETRY I

Draw the geometry by making a union of two rectangles.

Rectangle I (rI)

- I In the Geometry toolbar, click Rectangle.
- 2 In the Settings window for Rectangle, locate the Size and Shape section.
- 3 In the Width text field, type 0.02467.
- 4 In the Height text field, type 0.17.

Rectangle 2 (r2)

- I In the Geometry toolbar, click Rectangle.
- 2 In the Settings window for Rectangle, locate the Size and Shape section.
- 3 In the Width text field, type 0.025.
- 4 In the Height text field, type 0.01.

5 Locate the **Position** section. In the **x** text field, type 0.02467.

Point I (pt I)

- I In the Geometry toolbar, click Point.
- 2 In the Settings window for Point, locate the Point section.
- 3 In the x text field, type 0.032.
- 4 In the y text field, type 0.01.

Point 2 (pt2)

- I In the Geometry toolbar, click Point.
- 2 In the Settings window for Point, locate the Point section.
- 3 In the x text field, type 0.02467.
- 4 In the y text field, type 0.015.
- 5 Click | Build Selected.

GLOBAL DEFINITIONS

Load the model parameters from a text file.

Parameters 1

- I In the Model Builder window, under Global Definitions click Parameters I.
- 2 In the Settings window for Parameters, locate the Parameters section.
- 3 Click Load from File.
- 4 Browse to the model's Application Libraries folder and double-click the file cu electrowinning bubbly flow parameters.txt.

TERTIARY CURRENT DISTRIBUTION, NERNST-PLANCK (TCD)

First set the species for the electroneutrality condition and then set up the electrochemical model, consisting of species properties and separator domains, and boundaries comprising of an anode surface, a cathode surface, an inflow and an outflow.

- I In the Model Builder window, under Component I (compl) click Tertiary Current Distribution, Nernst-Planck (tcd).
- 2 In the Settings window for Tertiary Current Distribution, Nernst-Planck, locate the **Electrolyte Charge Conservation** section.
- 3 From the From electroneutrality list, choose cHSO4.

Species Charges 1

- I In the Model Builder window, under Component I (compl)>Tertiary Current Distribution, Nernst-Planck (tcd) click Species Charges 1.
- 2 In the Settings window for Species Charges, locate the Charge section.
- 3 In the z_{cCu} text field, type 2.
- 4 In the z_{cH} text field, type 1.
- 5 In the z_{cHSO4} text field, type -1.

Separator I

- I In the Physics toolbar, click **Domains** and choose **Separator**.
- 2 In the Settings window for Separator, locate the Domain Selection section.
- 3 From the Selection list, choose All domains.
- **4** Locate the **Convection** section. Specify the **u** vector as

- **5** Locate the **Diffusion** section. In the $D_{\rm cCu}$ text field, type DCu.
- **6** In the D_{cH} text field, type DH.
- **7** In the D_{cHSO4} text field, type DHSO4.
- 8 Locate the Porous Matrix Properties section. In the ϵ_l text field, type bf.phil.

Anode Surface

Set the electrode kinetics at the anode surface.

- I In the Physics toolbar, click Boundaries and choose Electrode Surface.
- 2 In the Settings window for Electrode Surface, type Anode Surface in the Label text field.
- 3 Select Boundary 8 only.
- 4 Locate the Electrode Phase Potential Condition section. From the Electrode phase potential condition list, choose Total current.
- **5** In the $I_{1,total}$ text field, type Itot.

Oxygen Evolution Reaction

- I In the Model Builder window, under Component I (compl)>Tertiary Current Distribution, Nernst-Planck (tcd)>Anode Surface click Electrode Reaction I.
- 2 In the Settings window for Electrode Reaction, type Oxygen Evolution Reaction in the Label text field.

- **3** Locate the **Stoichiometric Coefficients** section. In the n text field, type 4.
- 4 In the v_{cH} text field, type -4.
- **5** Locate the **Equilibrium Potential** section. In the $E_{\text{eq.ref}}(T)$ text field, type Eeq0_02.
- **6** Locate the **Electrode Kinetics** section. In the $i_{0,ref}(T)$ text field, type i0a_02.

Cathode Surface

Next, define the electrode kinetics at the cathode surface.

- I In the Physics toolbar, click Boundaries and choose Electrode Surface.
- 2 Select Boundary 1 only.
- 3 In the Settings window for Electrode Surface, type Cathode Surface in the Label text field.
- 4 Click to expand the **Dissolving-Depositing Species** section. Click + Add.
- **5** In the table, enter the following settings:

Species	Density (kg/m^3)	Molar mass (kg/mol)
Cu	rho_Cu	Mw_Cu

Copper Deposition Reaction

- I In the Model Builder window, under Component I (compl)>Tertiary Current Distribution, Nernst-Planck (tcd)>Cathode Surface click Electrode Reaction I.
- 2 In the Settings window for Electrode Reaction, type Copper Deposition Reaction in the Label text field.
- **3** Locate the **Stoichiometric Coefficients** section. In the n text field, type 2.
- 4 In the v_{eCu} text field, type -1.
- 5 In the Stoichiometric coefficients for dissolving-depositing species: table, enter the following settings:

Species	Stoichiometric coefficient (I)
Cu	1

- **6** Locate the **Equilibrium Potential** section. In the $E_{\text{eq.ref}}(T)$ text field, type Eeq0_Cu.
- **7** Locate the **Electrode Kinetics** section. In the $i_{0,ref}(T)$ text field, type i0c_Cu.
- **8** In the α_a text field, type alphaa_Cu.

Inflow I

Now, set the concentration of species at the Inflow boundary.

- I In the Physics toolbar, click Boundaries and choose Inflow.
- 2 Select Boundary 3 only.
- 3 In the Settings window for Inflow, locate the Concentration section.
- **4** In the $c_{0 \text{ cCu}}$ text field, type cocu.
- **5** In the $c_{0,cH}$ text field, type coh.

Outflow I

Set the Outflow boundary.

- I In the Physics toolbar, click Boundaries and choose Outflow.
- 2 Select Boundary 9 only.

Initial Values 1

Set the initial values for concentration.

- I In the Model Builder window, click Initial Values I.
- 2 In the Settings window for Initial Values, locate the Initial Values section.
- 3 In the cCu text field, type c0Cu.
- **4** In the cH text field, type coH.

BUBBLY FLOW, LAMINAR FLOW (BF)

Next, set up physics for the Bubbly Flow. The low gas concentration formulation is typically valid for gas phase fraction of 0.01. Considering that we may get higher gas phase fraction, particularly near the anode surface, clear the Low gas concentration check box under Physical Model.

- I In the Model Builder window, under Component I (compl) click Bubbly Flow, Laminar Flow (bf).
- 2 In the Settings window for Bubbly Flow, Laminar Flow, locate the Physical Model section.
- 3 Clear the Low gas concentration check box.

Fluid Properties 1

Now, set the physical properties for the liquid and gas phases. For simplicity, density and viscosity of the liquid phase are assumed to be constant. Set Calculate from ideal gas law option for density of the gas phase and Pressure-drag balance option for slip model.

- I In the Model Builder window, under Component I (compl)>Bubbly Flow, Laminar Flow (bf) click Fluid Properties 1.
- 2 In the Settings window for Fluid Properties, locate the Liquid Properties section.

- **3** From the ρ_1 list, choose **User defined**. In the associated text field, type rho.
- **4** From the μ_l list, choose **User defined**. In the associated text field, type nu.
- 5 Locate the Gas Properties section. From the ρ_g list, choose Calculate from ideal gas law.
- **6** In the M_g text field, type Mw_02.
- **7** In the d_b text field, type d_b .
- 8 Locate the Slip Model section. From the Slip model list, choose Pressure-drag balance.

Initial Values 1

Set the initial value for pressure.

- I In the Model Builder window, click Initial Values I.
- 2 In the **Settings** window for **Initial Values**, type rho*g_const*(0.17-y) in the p text field.

Gravity I

Next, add gravitation force using Gravity feature.

- I In the Physics toolbar, click **Domains** and choose **Gravity**.
- 2 In the Settings window for Gravity, locate the Domain Selection section.
- 3 From the Selection list, choose All domains.

Wall (Gas Inlet)

Next, set the gas mass influx at the anode surface.

- I In the Physics toolbar, click Boundaries and choose Wall.
- 2 In the Settings window for Wall, type Wall (Gas Inlet) in the Label text field.
- 3 Select Boundary 8 only.
- 4 Locate the Gas Boundary Condition section. From the Gas boundary condition list, choose Gas flux.
- **5** In the $N_{\rm og\phi g}$ text field, type (tcd.iloc_er1)*Mw_02/(4*F_const).

Liquid Inlet and Gas Outlet

Set the liquid inlet velocity and gas outlet using Inlet feature.

- I In the Physics toolbar, click Boundaries and choose Inlet.
- 2 In the Settings window for Inlet, type Liquid Inlet and Gas Outlet in the Label text field.
- 3 Select Boundary 3 only.
- **4** Locate the **Velocity** section. In the U_0 text field, type Vb.

5 Locate the Gas Boundary Condition section. From the Gas boundary condition list, choose Gas outlet.

Outlet I

Set the liquid and gas outlet using Outlet feature.

- I In the Physics toolbar, click Boundaries and choose Outlet.
- 2 Select Boundary 9 only.

GLOBAL DEFINITIONS

Default Model Inputs

Set up the temperature value used in the entire model.

- I In the Model Builder window, under Global Definitions click Default Model Inputs.
- 2 In the Settings window for Default Model Inputs, locate the Browse Model Inputs section.
- 3 In the tree, select General>Temperature (K) minput.T.
- 4 Find the Expression for remaining selection subsection. In the Temperature text field, type Τ.

MESH I

Set the extra fine mesh near the cathode surface and set the boundary layers at the Wall features.

- I In the Model Builder window, under Component I (compl) click Mesh I.
- 2 In the Settings window for Mesh, locate the Physics-Controlled Mesh section.
- **3** From the **Element size** list, choose **Finer**.
- 4 Locate the Sequence Type section. From the list, choose User-controlled mesh.

Size

- I In the Model Builder window, under Component I (compl)>Mesh I click Size.
- 2 In the Settings window for Size, locate the Element Size section.
- 3 From the Predefined list, choose Finer.

Size 1

- I In the Model Builder window, click Size I.
- 2 In the Settings window for Size, locate the Geometric Entity Selection section.
- 3 In the list, choose 2, 5, 6, 7, 8, and 10.
- 4 Click Remove from Selection.

- **5** Select Boundary 1 only.
- **6** In the list, select **1**.

Size 2

- I In the Model Builder window, right-click Mesh I and choose Size.
- 2 In the Settings window for Size, locate the Geometric Entity Selection section.
- 3 From the Geometric entity level list, choose Point.
- **4** Select Point 6 only.
- 5 Locate the Element Size section. From the Calibrate for list, choose Fluid dynamics.
- 6 From the Predefined list, choose Extra fine.
- 7 Drag and drop below Size 1.

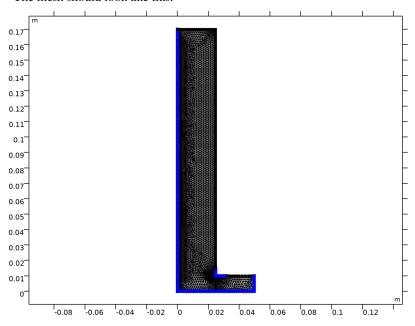
Boundary Layer Properties 1

Since it is usually a good idea to not have boundary layers on a boundary through which a significant gas flux is expected, anode boundary is excluded from boundary layers.

- I In the Model Builder window, expand the Boundary Layers I node, then click Boundary Layer Properties 1.
- **2** Select Boundaries 1, 2, 5–7, and 10 only.
- 3 In the Settings window for Boundary Layer Properties, locate the Layers section.
- 4 In the Number of layers text field, type 4.
- 5 From the Thickness specification list, choose First layer.
- 6 In the Thickness text field, type 0.00005.

7 Click III Build All.

The mesh should look like this:



STUDY I

Finally, update the study settings. Since we have not set the initial values for electrolyte potential, we will make use of Current Distribution Initialization study nodes. Add another Current Distribution Initialization study node and set current distribution type to Secondary. Disable Bubbly Flow interface from Current Distribution Initialization study nodes. Set the time range at the Time Dependent study node. Update the default solver setting by adding a Fully Coupled study node and then the model is ready to be solved.

Step 3: Current Distribution Initialization 2

- I In the Study toolbar, click Study Steps and choose Other> **Current Distribution Initialization.**
- 2 In the Settings window for Current Distribution Initialization, locate the Study Settings section.
- 3 From the Current distribution type list, choose Secondary.
- 4 Right-click Step 3: Current Distribution Initialization 2 and choose Move Up.

Step 3: Time Dependent

- I In the Model Builder window, click Step 3: Time Dependent.
- 2 In the Settings window for Time Dependent, locate the Study Settings section.
- 3 In the Output times text field, type range (0,0.25,60).

Solution I (soll)

- I In the Study toolbar, click Show Default Solver.
- 2 In the Model Builder window, expand the Solution I (soll) node.
- 3 Right-click Study I>Solver Configurations>Solution I (soll)>Time-Dependent Solver I and choose Fully Coupled.
- 4 In the Study toolbar, click **Compute**.

RESULTS

Liquid (bf)

Update the default plot for the liquid velocity by adding arrow surface.

In the Model Builder window, under Results click Liquid (bf).

Arrow Surface 1

- I In the Liquid (bf) toolbar, click Arrow Surface.
- 2 In the Settings window for Arrow Surface, click Replace Expression in the upper-right corner of the Expression section. From the menu, choose Component I (compl)> Bubbly Flow, Laminar Flow>Velocity and pressure>u,v - Velocity field, liquid phase.
- 3 Locate the Coloring and Style section. From the Color list, choose Black.
- 4 In the Liquid (bf) toolbar, click Plot. Surface plot for the liquid phase velocity should look like Figure 2.

Gas Phase (bf)

Next, update surface plot of the gas phase fraction by adding velocity streamlines.

In the Model Builder window, under Results click Gas Phase (bf).

Streamline I

- I In the Gas Phase (bf) toolbar, click Streamline.
- 2 In the Settings window for Streamline, click Replace Expression in the upper-right corner of the Expression section. From the menu, choose Component I (compl)>Bubbly Flow, Laminar Flow>Velocity and pressure>u,v - Velocity field, liquid phase.

- 3 Locate the Streamline Positioning section. From the Positioning list, choose Uniform density.
- 4 In the Separating distance text field, type 0.02.
- 5 Locate the Coloring and Style section. Find the Point style subsection. From the Type list, choose Arrow.
- 6 From the Arrow length list, choose Logarithmic.
- 7 From the Color list, choose White.
- 8 In the Gas Phase (bf) toolbar, click on Plot. Surface plot of the gas phase fraction should look like Figure 3.

Concentration, Cu (tcd)

The copper concentration at time t = 60 s should look like Figure 4.

Copper Concentration along Cathode Surface

Next, plot the copper concentration line plot along the cathode surface.

- I In the Home toolbar, click Add Plot Group and choose ID Plot Group.
- 2 In the Settings window for ID Plot Group, type Copper Concentration along Cathode Surface in the Label text field.
- 3 Locate the Data section. From the Time selection list, choose Last.

Line Grabh I

- I In the Copper Concentration along Cathode Surface toolbar, click Line Graph.
- 2 Select Boundary 1 only.
- 3 In the Settings window for Line Graph, click Replace Expression in the upper-right corner of the y-Axis Data section. From the menu, choose Component I (compl)> Tertiary Current Distribution, Nernst-Planck>Species cCu>cCu - Concentration - mol/m3.
- 4 Locate the x-Axis Data section. From the Parameter list, choose Expression.
- **5** In the **Expression** text field, type **y**.
- 7 Select the **Description** check box. In the associated text field, type **Distance** from the bottom of the cathode surface.
- Line plot of the copper concentration along the cathode surface at time t = 60 s should look like Figure 5.

Now, to plot the change in copper concentration with time at three different locations of the cathode surface, follow the below instructions.

Cut Point 2D I

- I In the Results toolbar, click Cut Point 2D.
- 2 In the Settings window for Cut Point 2D, locate the Point Data section.
- 3 In the \mathbf{x} text field, type 0 0 0.
- 4 In the y text field, type 0.05 0.15 0.16.

Copper Concentration (Point Graph)

- I In the Results toolbar, click \(\subseteq \text{ID Plot Group.} \)
- 2 In the Settings window for ID Plot Group, type Copper Concentration (Point Graph) in the Label text field.

Point Grabh 1

- I In the Copper Concentration (Point Graph) toolbar, click V Point Graph.
- 2 In the Settings window for Point Graph, locate the Data section.
- 3 From the Dataset list, choose Cut Point 2D 1.
- 4 Click Replace Expression in the upper-right corner of the y-Axis Data section. From the menu, choose Component I (compl)>Tertiary Current Distribution, Nernst-Planck> Species cCu>cCu - Concentration - mol/m3.
- 5 In the Copper Concentration (Point Graph) toolbar, click Plot.
- 6 Click to expand the **Legends** section. Select the **Show legends** check box. Point graph of the copper concentration should look like Figure 6.

Now, to plot concentration variation within the boundary layer at a specific position on the cathode surface, follow the below instructions.

Cut Line 2D I

- I In the Results toolbar, click Cut Line 2D.
- 2 In the Settings window for Cut Line 2D, locate the Line Data section.
- 3 In row Point 1, set y to 0.1.
- 4 In row **Point 2**, set **y** to **0.1**.
- 5 In row Point 2, set x to 0.001.

Copper Concentration (Boundary Layer)

I In the Results toolbar, click \to ID Plot Group.

2 In the Settings window for ID Plot Group, type Copper Concentration (Boundary Layer) in the Label text field.

Line Graph 1

- I In the Copper Concentration (Boundary Layer) toolbar, click \(\frac{1}{2} \) Line Graph.
- 2 In the Settings window for Line Graph, locate the Data section.
- 3 From the Dataset list, choose Cut Line 2D 1.
- 4 From the Time selection list, choose Last.
- 5 Click Replace Expression in the upper-right corner of the y-Axis Data section. From the menu, choose Component I (compl)>Tertiary Current Distribution, Nernst-Planck> Species cCu>cCu - Concentration - mol/m3.
- 6 Locate the x-Axis Data section. From the Parameter list, choose Expression.
- 7 In the Expression text field, type x.
- 8 Select the **Description** check box. In the associated text field, type Distance from the cathode surface.
- 9 In the Copper Concentration (Boundary Layer) toolbar, click **Plot**. The copper concentration plot in boundary layer should look like Figure 7.

Copper Deposition Thickness

Finally, plot the copper deposition thickness along the cathode surface at time t = 60 s.

- I In the Home toolbar, click Add Plot Group and choose ID Plot Group.
- 2 In the Settings window for ID Plot Group, type Copper Deposition Thickness in the **Label** text field.
- 3 Locate the Data section. From the Time selection list, choose Last.

Line Graph 1

- I In the Copper Deposition Thickness toolbar, click \(\subseteq \) Line Graph.
- 2 Select Boundary 1 only.
- 3 In the Settings window for Line Graph, locate the Selection section.
- **4** Click to select the **Activate Selection** toggle button.
- **5** Select Boundary 1 only.
- 6 Click Replace Expression in the upper-right corner of the y-Axis Data section. From the menu, choose Component I (compl)>Tertiary Current Distribution, Nernst-Planck> Dissolving-depositing species>tcd.sbtot - Total electrode thickness change - m.
- 7 Locate the y-Axis Data section. From the Unit list, choose µm.

- 8 Locate the x-Axis Data section. From the Parameter list, choose Expression.
- **9** In the **Expression** text field, type **y**.
- 10 Select the Description check box. In the associated text field, type Distance from the bottom of the cathode surface.
- II In the Copper Deposition Thickness toolbar, click **Plot**.

The copper deposition thickness plot should look like Figure 8.