

Lithium Plating with Deformation

In a lithium metal battery, lithium metal is deposited during charging on the negative electrode according to

$$\text{Li}^+ + e^- \rightarrow \text{Li}(s)$$

Due to mass transport and ohmic effects in the electrolyte, small initial protrusions on the metal surface will be subjected to accelerated growth during charging, which in the worst case may lead to the formation of dendrites, subsequent internal short circuits, and thermal runaway. This example explores the method of reverse pulse charging for mitigating the formation of dendrites.

The model uses the Lithium-Ion Battery, Deformed Geometry model wizard entry that adds a Lithium-Ion Battery interface along with Deformed Geometry formulation. Additionally, an Events interface is used to set up the forward and reverse current duty cycles.

Model Definition

Figure 1 shows the model geometry of the lithium-ion battery that consists of two domains (positive porous electrode and separator). The negative lithium metal electrode surface is located at y = 0 mm, with a small protrusion with a height of 40 mm, centered around x = 0. The materials considered are NMC 622 and LiPF₆ 3:7 EC:EMC for the positive electrode and electrolyte, respectively.

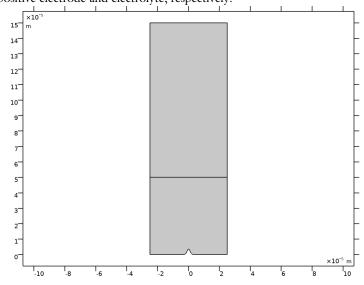


Figure 1: Model geometry.

In the first part of the tutorial, a forward current plating model is set up to simulate how a small protrusion of lithium grows during the plating process. A constant electrode current density of 1 C is applied on the top boundary of the positive porous electrode, and the resulting growth velocity along the negative lithium electrode surface is used as a boundary condition for the deformed geometry (ALE) time-dependent simulation.

In the second part of the tutorial, a forward and reverse current duty cycle is set up to examine how the protrusion of lithium is attenuated depending on the length of the reverse pulse. The current density for the reverse pulse $i_{\rm rev}$ on the top boundary of the positive porous electrode is set to -15 C. Denoting the forward duty cycle (the relative time spent in forward mode) as $t_{\rm fwd}$ (dimensionless), the forward pulse current density $i_{\rm fwd}$ can be calculated as

$$i_{\text{fwd}} = \frac{(i_{\text{avg}} - (1 - t_{\text{fwd}})i_{\text{rev}})}{t_{\text{fwd}}}$$
 (1)

The total simulated plating time is 0.75 h. The cycle time for a single forward and reverse duty cycle is 180 s. The Events interface is used to set up an Event Sequence consisting of the forward and reverse current duty cycles. The applied electrode current density on the top boundary of the positive porous electrode is appropriately calculated based on the

forward and reverse states and respective current densities. A parametric sweep is set up in the second part of the tutorial to simulate different lengths of the forward plating duty cycle t_{fwd} and examine the lithium electrode surface evolution during the forward and reverse current duty cycle.

This model uses linear elements for all the dependent variables in the Lithium-Ion Battery interface for faster computation times.

Results and Discussion

Figure 2 shows the lithium electrode surface profile evolution during the forward plating cycle. The initial protrusion grows in size and will result in an uneven surface.

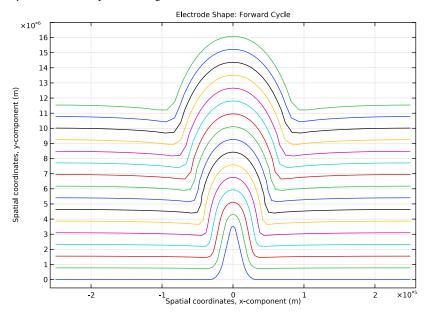


Figure 2: Electrode shape evolution for forward current duty cycle ($t_{\rm fwd}$ = 1).

Figure 3 shows the lithium electrode surface profile evolution during the forward and reverse current duty cycle, for $t_{\rm fwd}$ = 0.85. The initial protrusion is now attenuated as the plating proceeds.

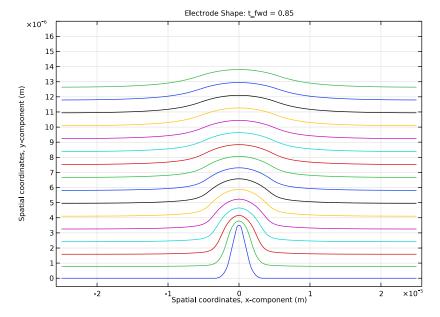


Figure 3: Electrode shape evolution for forward and reverse current duty cycle ($t_{\rm fwd}$ = 0.85).

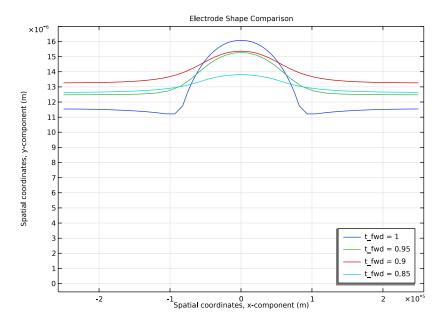


Figure 4: Comparison of final electrode surface profiles for different values of t_{fwd}

Finally, Figure 4 shows a comparison of the lithium electrode surface profiles at the last time step for different values of the forward plating duty cycle t_{fwd} .

Application Library path: Battery_Design_Module/Batteries,_Lithium-Ion/ li_plating_with_deformation

Modeling Instructions

This tutorial consists of two parts. In the first part you will set up a forward current plating model to simulate how a small protrusion of lithium grows during the plating process.

In the second part you will set up a forward and reverse current duty cycle, and examine how the protrusion of lithium is attenuated depending on the length of the reverse pulse.

From the File menu, choose New.

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>Batteries>Lithium-Ion Battery, **Deformed Geometry**.
- 3 Click Add.
- 4 Click Study.
- 5 In the Select Study tree, select Preset Studies for Selected Physics Interfaces> Time Dependent with Initialization.
- 6 Click M Done.

GLOBAL DEFINITIONS

Parameters 1

Load the model parameters from a text file.

- 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 li_plating_with_deformation_parameters.txt.

GEOMETRY I

Set up the geometry that consists of two domains (positive porous electrode and separator). The negative lithium electrode surface is located at y = 0 mm, with a small protrusion with a height of 40 mm, centered around x = 0.

Polygon I (boll)

- I In the Geometry toolbar, click / Polygon.
- 2 In the Settings window for Polygon, locate the Coordinates section.
- **3** In the table, enter the following settings:

x (m)	y (m)
0	H_prot
-W_prot/2	0
-W_cel1/2	0

x (m)	y (m)
-W_cel1/2	H_sep
W_cel1/2	H_sep
W_cell/2	0
W_prot/2	0

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 W cell.
- 4 In the **Height** text field, type H pos.
- 5 Locate the **Position** section. In the x text field, type -W cell/2.
- 6 In the y text field, type H sep.

Fillet I (fill)

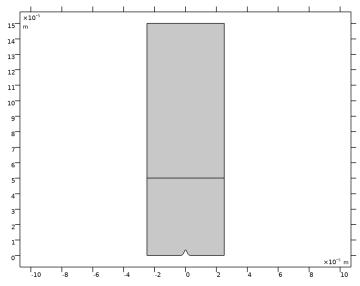
Round the corners by adding fillets.

- I In the **Geometry** toolbar, click **Fillet**.
- 2 On the object poll, select Point 1 only.
- 3 In the Settings window for Fillet, locate the Radius section.
- 4 In the Radius text field, type H_prot/10.

Fillet 2 (fil2)

- I In the **Geometry** toolbar, click **Fillet**.
- 2 On the object fill, select Points 3 and 6 only.
- 3 In the Settings window for Fillet, locate the Radius section.
- 4 In the Radius text field, type W prot/2.
- 5 In the Geometry toolbar, click **Build All**.
- **6** Click the **Zoom Extents** button in the **Graphics** toolbar.

7 In the Model Builder window, click Geometry 1.



ADD MATERIAL

The model has a lithium metal negative electrode, a NMC 622 positive electrode, and a LiPF6 3:7 EC:EMC electrolyte. Import the materials from the Battery Material Library.

- I In the Home toolbar, click 👯 Add Material to open the Add Material window.
- 2 Go to the Add Material window.
- 3 In the tree, select Battery>Electrolytes>LiPF6 in 3:7 EC:EMC (Liquid, Li-ion Battery).
- 4 Click Add to Component in the window toolbar.

ADD MATERIAL

- I Go to the Add Material window.
- 2 In the tree, select Battery>Electrodes>NMC 622, LiNi0.6Mn0.2Co0.202 (Positive, Liion Battery).
- 3 Click Add to Component in the window toolbar.

MATERIALS

NMC 622, LiNi0.6Mn0.2Co0.2O2 (Positive, Li-ion Battery) (mat2) Select Domain 2 only.

ADD MATERIAL

- I Go to the Add Material window.
- 2 In the tree, select Battery>Electrodes>Lithium Metal, Li (Negative, Li-ion Battery).
- 3 Click Add to Component in the window toolbar.
- 4 In the Home toolbar, click Radd Material to close the Add Material window.

MATERIALS

Lithium Metal, Li (Negative, Li-ion Battery) (mat3)

- I In the Settings window for Material, locate the Geometric Entity Selection section.
- 2 From the Geometric entity level list, choose Boundary.

Use a box selection to select all the (lower) boundaries pertaining to the lithium electrode surface, including the protrusion.

- **3** Select Boundaries 2, 6–8, and 11–13 only.
- 4 Click **Create Selection**.

By creating a selection in this way you can conveniently select the same set of boundaries again later on.

- 5 In the Create Selection dialog box, type Lithium Electrode Surface in the Selection name text field.
- 6 Click OK.

LITHIUM-ION BATTERY (LIION)

Now start setting up the forward cycle lithium-ion battery model.

Porous Electrode I

- I In the Model Builder window, under Component I (compl) right-click Lithium-Ion Battery (liion) and choose Porous Electrode.
- 2 Select Domain 2 only.
- 3 In the Settings window for Porous Electrode, locate the Electrolyte Properties section.
- 4 From the Electrolyte material list, choose LiPF6 in 3:7 EC:EMC (Liquid, Liion Battery) (mat I).
- **5** Locate the **Electrode Properties** section. In the σ_s text field, type sigmas_pos.
- 6 Locate the Porous Matrix Properties section. In the ε_s text field, type epss_pos.

Particle Intercalation I

I In the Model Builder window, click Particle Intercalation I.

- 2 In the Settings window for Particle Intercalation, locate the Species Settings section.
- **3** In the $c_{s,init}$ text field, type cs_init_pos.
- **4** Locate the **Particle Transport Properties** section. In the r_p text field, type rp_pos .

Electrode Surface I

- I In the Physics toolbar, click Boundaries and choose Electrode Surface.
- 2 In the Settings window for Electrode Surface, locate the Boundary Selection section.
- 3 From the Selection list, choose Lithium Electrode Surface.
- 4 Click to expand the Dissolving-Depositing Species section. Use a Dissolving-Depositing **Species** to define the growth velocity of the lithium electrode surface.
- 5 Click + Add.
- **6** In the table, enter the following settings:

Species	Density (kg/m^3)	Molar mass (kg/mol)
Li	rho_Li	M_Li

7 Clear the Solve for surface concentration variables check box.

Electrode Reaction 1

- I In the Model Builder window, click Electrode Reaction I.
- 2 In the Settings window for Electrode Reaction, locate the Stoichiometric Coefficients section.
- 3 In the Stoichiometric coefficients for dissolving-depositing species: table, enter the following settings:

Species	Stoichiometric coefficient (I)
Li	1

Electrode Current: Forward Cycle

- I In the Physics toolbar, click Boundaries and choose Electrode Current.
- 2 In the Settings window for Electrode Current, type Electrode Current: Forward Cycle in the Label text field.
- **3** Select Boundary 5 only.
- 4 Locate the Electrode Current section. From the list, choose Average current density.
- **5** In the $i_{s,average}$ text field, type i_app_fwd.

DEFORMED GEOMETRY

Select the separator domain as the **Deforming Domain**.

Deforming Domain I

- I In the Model Builder window, expand the Deformed Geometry node, then click Deforming Domain 1.
- 2 Select Domain 1 only.

MULTIPHYSICS

Nondeforming Boundary I (ndbdg1)

Set zero normal displacement at the vertical nondeforming boundaries.

- I In the Model Builder window, expand the Multiphysics node, then click Nondeforming Boundary I (ndbdgI).
- 2 In the Settings window for Nondeforming Boundary, locate the Nondeforming Boundary section.
- 3 From the Boundary condition list, choose Zero normal displacement.

LITHIUM-ION BATTERY (LIION)

Set linear elements for all the dependent variables in the **Lithium-Ion Battery** interface to speed up computation time.

- I In the Model Builder window, under Component I (compl) click Lithium-Ion Battery (liion).
- 2 In the Settings window for Lithium-lon Battery, click to expand the Discretization section.
- 3 From the Electrolyte potential list, choose Linear.
- 4 From the Electrolyte salt concentration list, choose Linear.
- 5 From the Electric potential list, choose Linear.

MESH I

Modify the mesh as follows to get more mesh elements along the lithium electrode surface.

Free Triangular 1

In the Mesh toolbar, click Free Triangular.

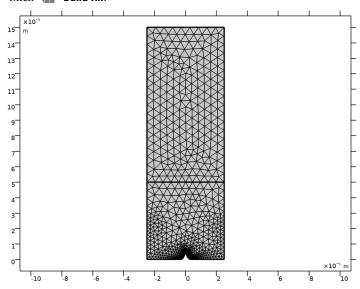
Size 1

- I Right-click Free Triangular 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 Boundary.
- 4 From the Selection list, choose Lithium Electrode Surface.
- 5 Locate the Element Size section. From the Predefined list, choose Finer.
- 6 Click the Custom button.
- 7 Locate the Element Size Parameters section.
- 8 Select the Maximum element growth rate check box. In the associated text field, type 1.1.

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**.
- 4 Click Build All.



STUDY: FORWARD CYCLE

The forward cycle problem is now ready for solving. Update the time unit and output times before solving.

- I In the Model Builder window, click Study I.
- 2 In the Settings window for Study, type Study: Forward Cycle in the Label text field.

Step 2: Time Dependent

- I In the Model Builder window, expand the Study: Forward Cycle node, then click Step 2: Time Dependent.
- 2 In the Settings window for Time Dependent, locate the Study Settings section.
- **3** From the **Time unit** list, choose **h**.
- 4 In the Output times text field, type range (0,0.05/C rate avg,0.75/C rate avg).
- 5 In the Home toolbar, click **Compute**.

RESULTS

Create a line plot for the shape of the lithium electrode surface at different times during the forward cycle (Figure 2) as follows.

Electrode Shape: Forward Cycle

- I In the Home toolbar, click Add Plot Group and choose ID Plot Group.
- 2 In the Settings window for ID Plot Group, type Electrode Shape: Forward Cycle in the Label text field.
- 3 Click to expand the Title section. From the Title type list, choose Manual.
- 4 In the Title text area, type Electrode Shape: Forward Cycle.

Line Grabh I

- I Right-click Electrode Shape: Forward Cycle and choose Line Graph.
- 2 In the Settings window for Line Graph, locate the Selection section.
- 3 From the Selection list, choose Lithium Electrode Surface.
- **4** Locate the **y-Axis Data** section. In the **Expression** text field, type y.
- 5 Locate the x-Axis Data section. From the Parameter list, choose Expression.
- 6 In the Expression text field, type x.

Electrode Shape: Forward Cycle

- I In the Model Builder window, click Electrode Shape: Forward Cycle.
- 2 In the Settings window for ID Plot Group, locate the Axis section.
- 3 Select the Manual axis limits check box.
- 4 In the y minimum text field, type -5.5e-7.
- 5 In the y maximum text field, type 1.7e-5.

ADD PHYSICS

You have now completed the first part of this tutorial where the applied current included only the forward current duty cycle. We will now set up the second part of the tutorial where the applied current includes both the forward and reverse current duty cycles.

Start by adding an **Events** interface and set up an **Event Sequence** consisting of the forward and reverse current duty cycles.

- I In the Home toolbar, click Add Physics to open the Add Physics window.
- 2 Go to the Add Physics window.
- 3 In the tree, select Mathematics>ODE and DAE Interfaces>Events (ev).
- 4 Click Add to Component I in the window toolbar.
- 5 In the Home toolbar, click Add Physics to close the Add Physics window.

EVENTS (EV)

Event Sequence 1

- I Right-click Component I (compl)>Events (ev) and choose Event Sequence.
- 2 In the Settings window for Event Sequence, locate the Sequence Control section.
- **3** Select the **Loop** check box.

Sequence Member 1

- I In the Model Builder window, expand the Event Sequence I node, then click Sequence Member 1.
- 2 In the Settings window for Sequence Member, locate the Sequence Member section.
- 3 In the Discrete state name text field, type state fwd.
- 4 From the End condition list, choose Duration.
- 5 In the Duration text field, type T cycle*t fwd.

Event Sequence 1

In the Model Builder window, click Event Sequence 1.

Sequence Member 2

- I In the Physics toolbar, click Attributes and choose Sequence Member.
- 2 In the Settings window for Sequence Member, locate the Sequence Member section.
- 3 In the Discrete state name text field, type state_rev.
- 4 From the End condition list, choose Duration.
- 5 In the Duration text field, type T cycle*t rev.

DEFINITIONS (COMPI)

Next, set up the variable for the applied current density for the forward and reverse cycle and use this variable to set the electrode current for the battery.

Variables 1

- I In the Model Builder window, under Component I (compl) right-click Definitions and choose Variables.
- 2 In the Settings window for Variables, locate the Variables section.
- **3** In the table, enter the following settings:

Name	Expression	Unit	Description
i_app	state_fwd*i_fwd+ state_rev*i_rev	A/m²	Applied current density (forward and reverse cycle)

LITHIUM-ION BATTERY (LIION)

Electrode Current: Forward Cycle

In the Model Builder window, under Component I (compl)>Lithium-lon Battery (liion) right-click Electrode Current: Forward Cycle and choose Duplicate.

Electrode Current: Forward and Reverse Cycle

- I In the Model Builder window, under Component I (compl)>Lithium-lon Battery (liion) click Electrode Current: Forward Cycle 1.
- 2 In the Settings window for Electrode Current, type Electrode Current: Forward and Reverse Cycle in the Label text field.
- **3** Locate the **Electrode Current** section. In the $i_{s,average}$ text field, type i_app.

ADD STUDY

The forward and reverse cycle model is now ready for solving. Use a Parametric Sweep to simulate different lengths of the forward plating duty cycle.

- I In the Home toolbar, click Add Study to open the Add Study window.
- 2 Go to the Add Study window.
- 3 Find the Studies subsection. In the Select Study tree, select Preset Studies for Selected Physics Interfaces>Lithium-Ion Battery> Time Dependent with Initialization.
- 4 Click Add Study in the window toolbar.
- 5 In the Home toolbar, click Add Study to close the Add Study window.

STUDY: FORWARD AND REVERSE CYCLE

- I In the Model Builder window, click Study 2.
- 2 In the Settings window for Study, type Study: Forward and Reverse Cycle in the Label text field.

Parametric Sweep

- I In the Study toolbar, click Parametric Sweep.
- 2 In the Settings window for Parametric Sweep, locate the Study Settings section.
- 3 Click + Add.
- **4** In the table, enter the following settings:

Parameter name	Parameter value list	Parameter unit
t_fwd (Forward current duty cycle)	0.95 0.9 0.85	

Step 1: Current Distribution Initialization

Also, disable Electrode Current: Forward Cycle node in this study.

- I In the Model Builder window, click Step I: Current Distribution Initialization.
- 2 In the Settings window for Current Distribution Initialization, locate the Physics and Variables Selection section.
- 3 Select the Modify model configuration for study step check box.
- 4 In the tree, select Component I (compl)>Lithium-lon Battery (liion)> Electrode Current: Forward Cycle.
- 5 Click O Disable.

Step 2: Time Dependent

- I In the Model Builder window, click Step 2: Time Dependent.
- 2 In the Settings window for Time Dependent, locate the Study Settings section.
- **3** From the **Time unit** list, choose **h**.
- 4 In the Output times text field, type range(0,0.05/C_rate_avg,0.75/C_rate_avg).
- 5 Locate the Physics and Variables Selection section. Select the Modify model configuration for study step check box.
- 6 In the tree, select Component I (comp1)>Lithium-Ion Battery (liion)> Electrode Current: Forward Cycle.
- 7 Click O Disable.
- 8 Click to expand the Study Extensions section. Select the Automatic remeshing check box.

STUDY: FORWARD CYCLE

Before solving the forward and reverse cycle study, disable appropriate nodes in the forward cycle study for completeness.

Step 1: Current Distribution Initialization

- I In the Model Builder window, under Study: Forward Cycle click Step 1: Current Distribution Initialization.
- 2 In the Settings window for Current Distribution Initialization, locate the Physics and Variables Selection section.
- 3 Select the Modify model configuration for study step check box.
- 4 In the tree, select Component I (compl)>Lithium-lon Battery (liion)> Electrode Current: Forward and Reverse Cycle.
- 5 Click / Disable.

Step 2: Time Dependent

- I In the Model Builder window, click Step 2: Time Dependent.
- 2 In the Settings window for Time Dependent, locate the Physics and Variables Selection section.
- **3** In the table, enter the following settings:

Physics interface	Solve for	Equation form
Events (ev)		Automatic (Time dependent)

- 4 Select the Modify model configuration for study step check box.
- 5 In the tree, select Component I (compl)>Lithium-lon Battery (liion)> Electrode Current: Forward and Reverse Cycle.
- 6 Click / Disable.

STUDY: FORWARD AND REVERSE CYCLE

Finally, solve the forward and reverse cycle model.

- I In the Model Builder window, click Study: Forward and Reverse Cycle.
- 2 In the Settings window for Study, locate the Study Settings section.
- 3 Clear the Generate default plots check box.
- 4 In the Study toolbar, click **Compute**.

RESULTS

As before, create a line plot for the shape of the lithium electrode surface at different times during the forward and reverse cycle (Figure 3).

Electrode Shape: Forward Cycle

In the Model Builder window, under Results right-click Electrode Shape: Forward Cycle and choose **Duplicate**.

Electrode Shape: Forward and Reverse Cycle

- I In the Model Builder window, under Results click Electrode Shape: Forward Cycle I.
- 2 In the Settings window for ID Plot Group, type Electrode Shape: Forward and Reverse Cycle in the Label text field.
- 3 Locate the Data section. From the Dataset list, choose Study: Forward and Reverse Cycle/ Parametric Solutions I (sol6).
- 4 From the Parameter selection (t_fwd) list, choose From list.
- 5 In the Parameter values (t_fwd) list, select 0.85.
- 6 Click to expand the Title section. In the Title text area, type Electrode Shape: t fwd = eval(t_fwd).
- 7 Locate the Data section. From the Time selection list, choose Interpolated.
- 8 In the Times (h) text field, type range (0,0.05,0.75).
- 9 In the Electrode Shape: Forward and Reverse Cycle toolbar, click **Plot**.

Electrode Shape Comparison

Finally, create a line plot for the shape of the lithium electrode surface at the last time step for different values of the forward plating duty cycle (Figure 4).

- I In the Home toolbar, click **Add Plot Group** and choose ID Plot Group.
- 2 In the Settings window for ID Plot Group, type Electrode Shape Comparison in the **Label** text field.
- 3 Locate the Data section. From the Dataset list, choose None.
- 4 Click to expand the **Title** section. From the **Title type** list, choose **Manual**.
- 5 In the Title text area, type Electrode Shape Comparison.

Line Graph 1

- I Right-click Electrode Shape Comparison and choose Line Graph.
- 2 In the Settings window for Line Graph, locate the Data section.
- 3 From the Dataset list, choose Study: Forward Cycle/Solution I (soll).

- 4 From the Time selection list, choose Last.
- 5 Locate the Selection section. From the Selection list, choose Lithium Electrode Surface.
- **6** Locate the **y-Axis Data** section. In the **Expression** text field, type y.
- 7 Locate the x-Axis Data section. From the Parameter list, choose Expression.
- **8** In the **Expression** text field, type x.
- **9** Click to expand the **Legends** section. Select the **Show legends** check box.
- 10 From the Legends list, choose Evaluated.
- II In the **Legend** text field, type t fwd = 1.
- 12 Right-click Line Graph I and choose Duplicate.

Line Graph 2

- I In the Model Builder window, click Line Graph 2.
- 2 In the Settings window for Line Graph, locate the Data section.
- 3 From the Dataset list, choose Study: Forward and Reverse Cycle/ Parametric Solutions I (sol6).
- **4** From the **Time selection** list, choose **Last**.
- **5** Locate the **Legends** section. In the **Legend** text field, type t_fwd = eval(t_fwd).

Electrode Shape Comparison

- I In the Model Builder window, click Electrode Shape Comparison.
- 2 In the Settings window for ID Plot Group, locate the Axis section.
- 3 Select the Manual axis limits check box.
- 4 In the y minimum text field, type -5.5e-7.
- 5 In the y maximum text field, type 1.7e-5.
- 6 Locate the Legend section. From the Position list, choose Lower right.
- 7 In the Electrode Shape Comparison toolbar, click **Plot**.