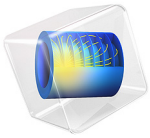


Battery Electrode with a Particle Size Distribution

Introduction

In many lithium-ion battery models, an extra (pseudo) dimension is introduced in the porous electrodes to define the diffusion of intercalated lithium on the electrode particle scale. Mathematically this is achieved by associating every point in the porous electrode of the base dimension with a local geometric entity, representing an electrode particle, in which the concentration field of intercalated lithium is defined and solved for. In the classical Newman (P2D) model, all electrode particles are assumed spherical, and the intercalated lithium concentration field is hence defined on a one-dimensional extra dimension, assuming spherical symmetry. In the **Lithium-Ion Battery Interface**, concentration fields on 1D extra dimensions using either spherical, cylindrical or planar symmetry can be defined automatically in any space dimension of the base geometry by the **Particle Intercalation** node, which is active by default as a subnode to the **Porous Electrode**, **Highly Conductive Porous Electrode** or **Additional Electrode Material** nodes.

Battery electrodes featuring large heterogeneities in terms of particle sizes may sometimes not be adequately described by homogenized models using one single particle size only. The most straight-forward way to model the effect of multiple particle sizes, is to add multiple **Additional Porous Electrode** material nodes to a model. This approach may however become tedious, and computationally inefficient when modeling a large ensemble of different particle sizes. As an alternative to adding multiple instances of the **Additional Porous Electrode** material node, this tutorial demonstrates how to instead deploy a user-defined **Extra Dimension** to define the solid phase diffusion of intercalated lithium for a range of particle sizes.

The distribution of particle sizes in the electrode is added to the model in the form of a histogram.

Model Definition

BASE DIMENSION MODEL

Figure 1 shows the base geometry of the battery cell model. The base geometry is defined in 1D as a half cell consisting of one positive porous nickel–manganese–cobalt (NMC) electrode and one separator domain, with a lithium metal counter electrode defined on the exterior boundary of the separator domain.

The charge and mass balances in the base dimension are defined and solved for the following three dependent variables:

- The electrode phase potential, ϕ_s (V) (only in the porous electrode domain)

- The electrolyte phase potential, ϕ_l (V)
- The electrolyte concentration, c_l (mol/m^3)

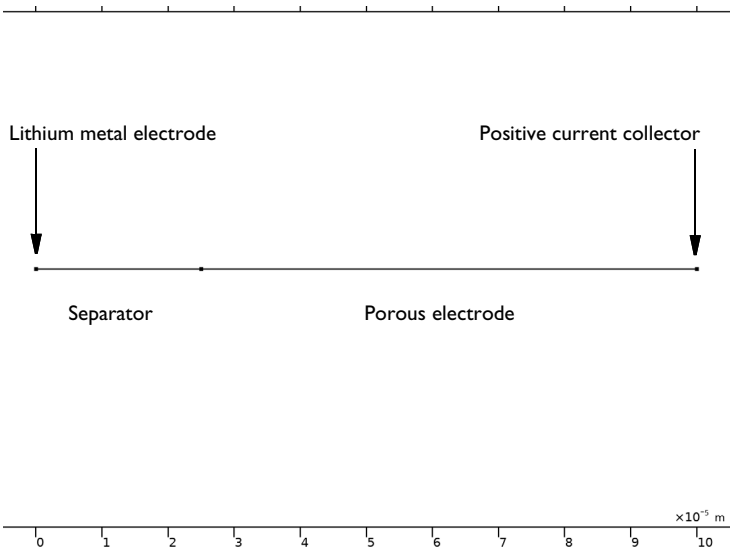


Figure 1: Model base geometry.

EXTRA DIMENSION MODEL

Figure 2 shows the extra dimension model, which is attached to the porous electrode domain in the base model geometry. The extra dimension model is formulated to solve for the concentration c_s (mol/m^3) on a 2D unit square (x_s, y_s), where x_s represents a dimensionless radial coordinate and y_s is used to define a y_s-dependent particle radius, R_p, ranging between the minimum and maximum particle radii, $R_{p,\min}$ and $R_{p,\max}$, respectively.

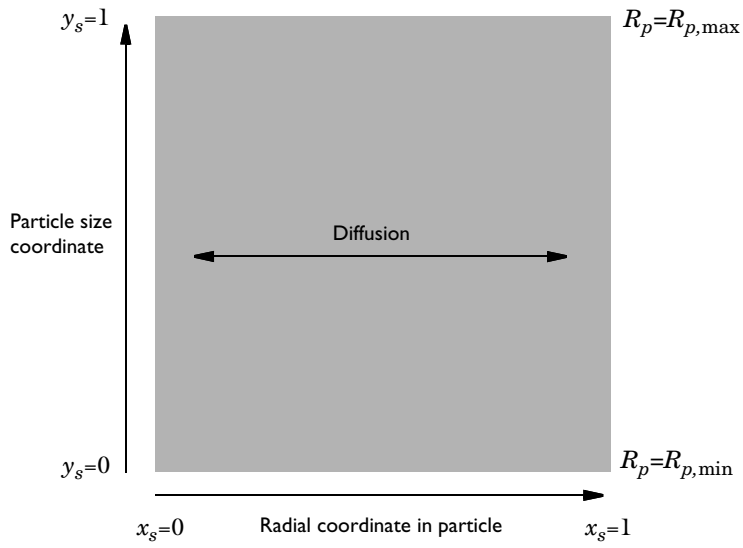


Figure 2: Extra dimension geometry.

A mass balance is defined, based on diffusion in the x_s direction, according to

$$\frac{\partial c_s}{\partial t} + \frac{1}{x_s}\frac{\partial}{\partial x_s}\left(-x_s^2 \frac{D_s}{R_p^2}\frac{\partial c_s}{\partial x_s}\right) = 0$$

where D_s is the diffusion coefficient for intercalated lithium, with the boundary conditions

$$-D_s \frac{\partial c_s}{\partial x_s} = 0$$

at the center of the particles ($x_s = 0$) and

$$-D_s \frac{\partial c_s}{\partial x_s} = R_p \frac{i_{\text{loc}}}{F}$$

at the surface of the particles ($x_s = 1$).

The local current density, i_{loc}, is typically a function both of the concentration in the extra dimension (at $x_s = 1$) as well as the three base dimension variables.

$$i_{\text{loc}} = i_{\text{loc}}(c_s, \phi_s, \phi_l, c_l)$$

Note that for a non-particle-size-distribution case, the above equations would be identical. The only difference would be that the extra dimension domain would be formulated in 1D with the spatial coordinate x_s only, instead of the (x_s, y_s) unit square.

The y_s coordinate is used to define the range of particle sizes. Using a linear mapping relation, the particle radius is defined as

$$R_p(y_s) = R_{p,\,\min} + y_s \times (R_{p,\,\max} - R_{p,\,\min})$$

(For the non-particle-size-distribution case, R_p is a constant).

As a result of R_p varying with y_s, the concentration along the particle surface will become nonuniform in the y_s direction as the particles are dynamically charged or discharged, which in turn means that the local current density will also vary along the particle surface $(x_s = 1)$:

$$i_{\text{loc}} = i_{\text{loc}}(c_s(y_s), \phi_s, \phi_l, c_l)$$

PARTICLE SIZE HISTOGRAM AND COUPLING TO THE BASE DIMENSION

The extra dimension model described in the previous section defines the dynamic behavior of the intercalated concentration of lithium for a range of particle sizes. A frequency distribution function, $f_{\text{hist}}(R_p)$, defines the statistical occurrences of the different particle sizes in the porous electrode. The function couples the local current density, i_{loc}, on the individual particle surfaces to the volumetric current density, i_{v}, which is used for defining the mass and charge source and sink terms in the base model.

Figure 3 shows the frequency distribution function between the minimum and maximum radius values $R_{p,\min} = 2$ µm and $R_{p,\max} = 14$ µm. The data for the frequency function is taken from Ref. 1.

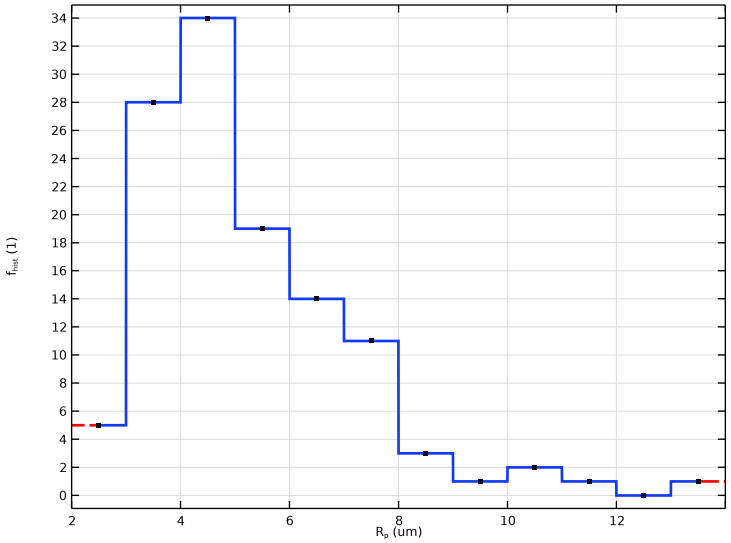


Figure 3: Particle size histogram defining the frequency distribution function $f_{\text{hist}}(R_p)$ in the model.

The volumetric current density is calculated by multiplying the active volume fraction of the electrode, ε_s, with the integral of the total current of the particle ensemble as defined by f_{hist}, divided by the corresponding total volume of the ensemble:

$$i_v = \varepsilon_s \frac{\int_{R_{p,\min}}^{R_{p,\max}} f_{\text{hist}} i_{\text{loc}} 4\pi R_p^2 \mathrm{d}R_p}{\int_{R_{p,\min}}^{R_{p,\max}} f_{\text{hist}} 4\pi R_p^3/3 \mathrm{d}R_p}$$

SIMULATIONS

The tutorial model runs two simulations for a 30 min discharge at 10 A/m^2, followed by a 2.5 h relaxation period. The first simulation uses a non-particle-distribution model using the built-in extra dimension formulation in the **Particle Intercalation** node. The second simulation runs the same charge-relaxation pulse, but with a particle size distribution according to Figure 3.

Figure 4 shows the cell voltages as well as the surface and center particle concentrations at the separator-electrode boundary for the non-particle-size-distribution case during the current pulse and the following relaxation. Figure 5 shows the voltage dynamics for the particle-size-distribution case, and the corresponding surface and center concentrations for the largest and smallest particles in the ensemble.

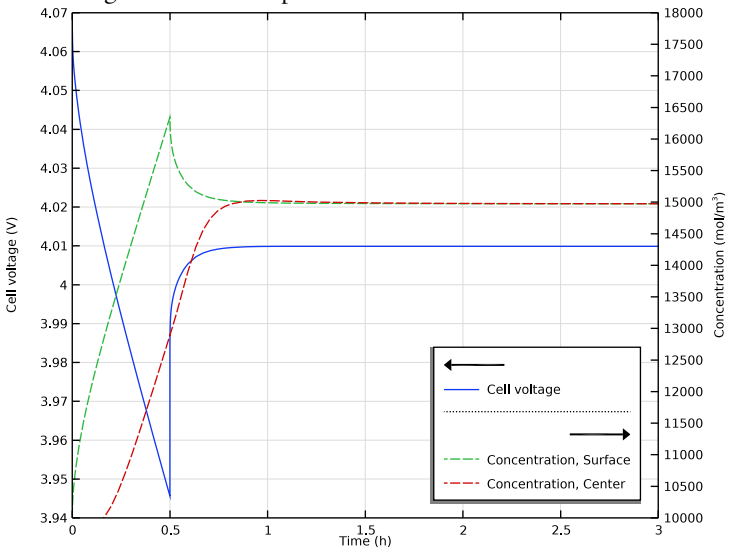


Figure 4: Cell voltage (left hand axis) and surface and center concentrations in the electrode particles at the separator-porous electrode boundary (right hand axis) for the non-particle-size-distribution case.

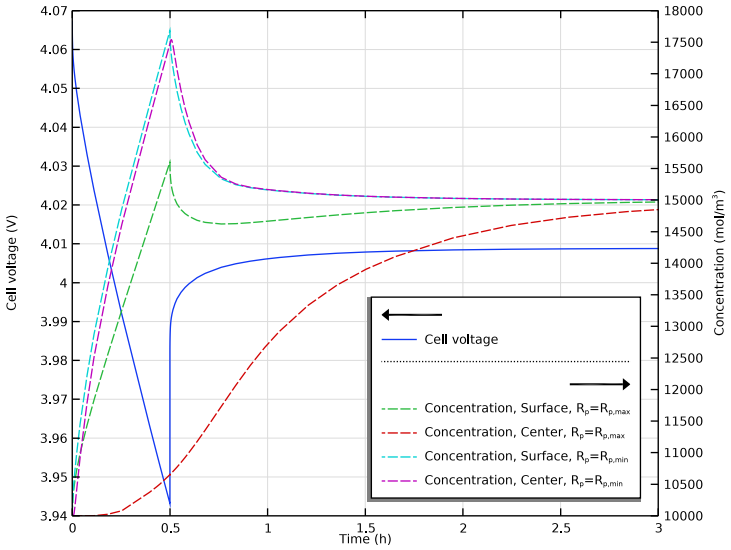


Figure 5: Cell voltage (left hand axis) and surface and center concentrations in the electrode particles at the separator-porous electrode boundary for both the largest and smallest particles (right hand axis) when including a particle size distribution.

Figure 6 compares the voltage dynamics of the two cases. As can be seen, the voltage curves are fairly similar during the 30 min load, but differ significantly during the following relaxation period.

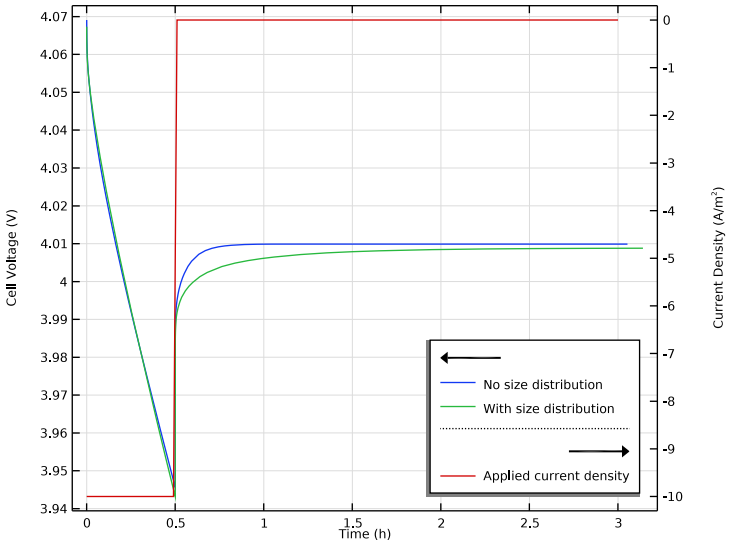


Figure 6: Cell current density and corresponding voltage response.

Finally, the concentration field in the extra dimension at the end of the current pulse at the electrode-separator and the electrode current-collector boundaries is displayed in Figure 7 and Figure 8. A more uniform concentration profile is seen in the x_s direction for the smallest particles ($y_s = 0$) than for the largest particles ($y_s = 1$).

Only small differences can be seen between Figure 7 and Figure 8. This indicates that most mass transport limitations are associated to the transport within the particles (especially the larger ones) rather than along the thickness of the porous electrode.

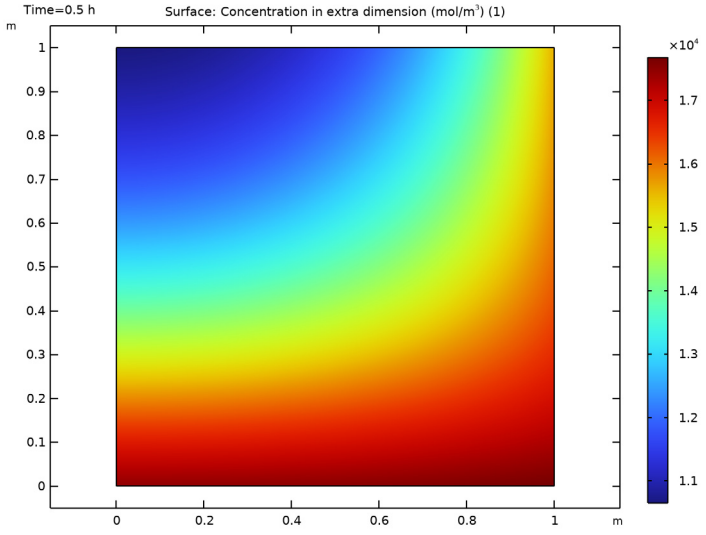


Figure 7: Concentration field in the extra dimension (x_s, y_s) at the separator-porous electrode boundary at the end of the current pulse.

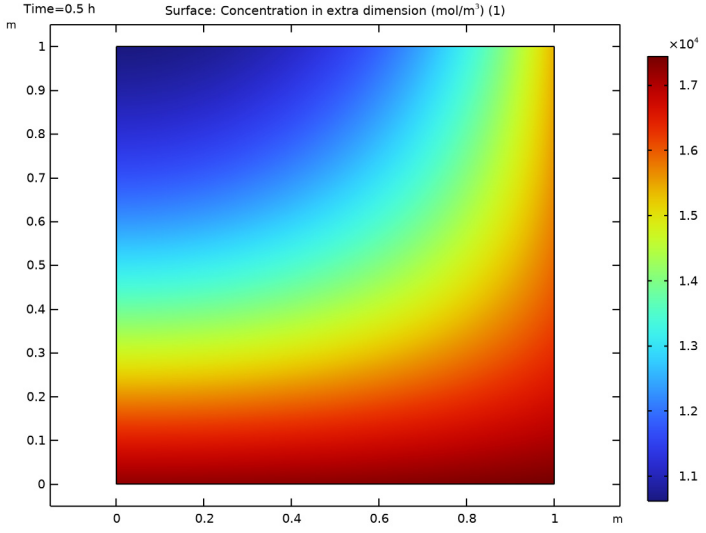


Figure 8: Concentration field in the extra dimension (x_s, y_s) at the external current collector-porous electrode boundary at the end of the current pulse.

Notes About the COMSOL Implementation

A **State Variables** node is used to compute some help variables integrated from the particle histogram. By setting **Update** to **Only initialization**, this computation only needs to be performed once during the simulation.

Reference

1. C-H. Chen, F.B. Planella, K. O'Regan, D. Gastol, W.D. Widanage, and E. Kendrick, "Development of Experimental Techniques for Parameterization of Multi-scale Lithium-ion Battery Models," *J. Electrochem. Soc.*, vol. 167, 080534, 2020.

Application Library path: `Battery_Design_Module/Batteries,_Lithium-Ion/particle_size_distribution`

Modeling Instructions

From the **File** menu, choose **New**.

NEW

In the **New** window, click **Model Wizard**.

MODEL WIZARD

1 In the **Model Wizard** window, click **1D**.

2 In the **Select Physics** tree, select **Electrochemistry>Batteries>Lithium-Ion Battery (liion)**.

3 Click **Add**.

4 Click **Study**.

5 In the **Select Study** tree, select **General Studies>Time Dependent**.

6 Click **Done**.

GLOBAL DEFINITIONS

Parameters 1

1 In the **Model Builder** window, under **Global Definitions** click **Parameters 1**.

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 `particle_size_distribution_parameters.txt`.

Interpolation - Particle Radius Histogram

Import the particle size histogram from a text file into an interpolation polynomial as follows:

1 In the **Home** toolbar, click f(x) **Functions** and choose **Global>Interpolation**.

2 In the **Settings** window for **Interpolation**, type `Interpolation - Particle Radius Histogram` in the **Label** text field.

3 Locate the **Definition** section. In the **Function name** text field, type `f_hist`.

4 Click **Load from File**.

5 Browse to the model's Application Libraries folder and double-click the file `particle_size_distribution_histogram.txt`.

6 Locate the **Interpolation and Extrapolation** section. From the **Interpolation** list, choose **Nearest neighbor**.

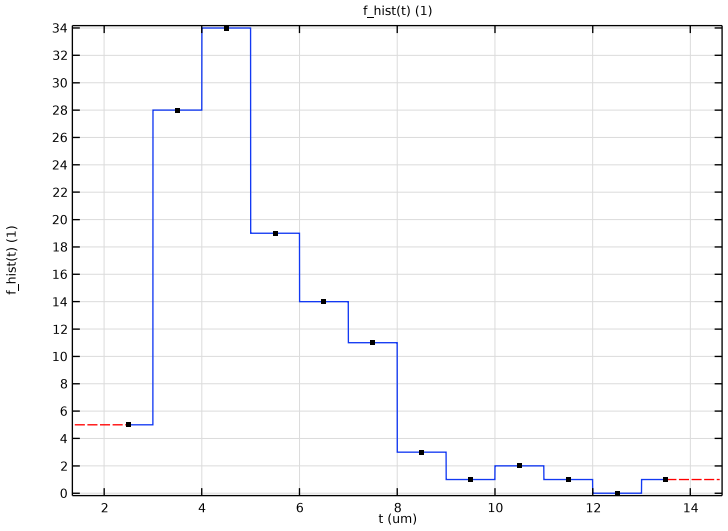
7 Locate the **Units** section. In the **Function** table, enter the following settings:

Function	Unit
f_hist	1

8 In the **Argument** table, enter the following settings:

Argument	Unit
t	um

9 Click **Plot**.



Add some state variables that calculate certain integrated properties from the histogram data as follows:

10 Click the **Show More Options** button in the **Model Builder** toolbar.

11 In the **Show More Options** dialog box, select **General>Variable Utilities** in the tree.

12 In the tree, select the check box for the node **General>Variable Utilities**.

13 Click **OK**.

State Variables - Particle Measures From Histogram

1 In the **Home** toolbar, click **Variable Utilities** and choose **Global>State Variables**.

2 In the **Settings** window for **State Variables**, type `State Variables - Particle Measures From Histogram` in the **Label** text field.

3 Locate the **State Components** section. In the table, enter the following settings:

State	Initial value	Update expression	Description
vol_particles	integrate(f_hist(Rp_arg)*4*pi*Rp_arg^3/3,Rp_arg,Rp_min,Rp_max)		Volume of all particles
area_particles	integrate(f_hist(Rp_arg)*4*pi*Rp_arg^2,Rp_arg,Rp_min,Rp_max)		Area of all particles
mass_averaged_Rp_squared	integrate(f_hist(Rp_arg)*Rp_arg^2*4*pi*Rp_arg^3/3,Rp_arg,Rp_min,Rp_max)/integrate(f_hist(Rp_arg)*4*pi*Rp_arg^3/3,Rp_arg,Rp_min,Rp_max)		Mass-averaged squared particle radius

4 From the **Update** list, choose **Only initialization**.

Step 1 (step1)

Add a step function to define the battery current pulse as follows:

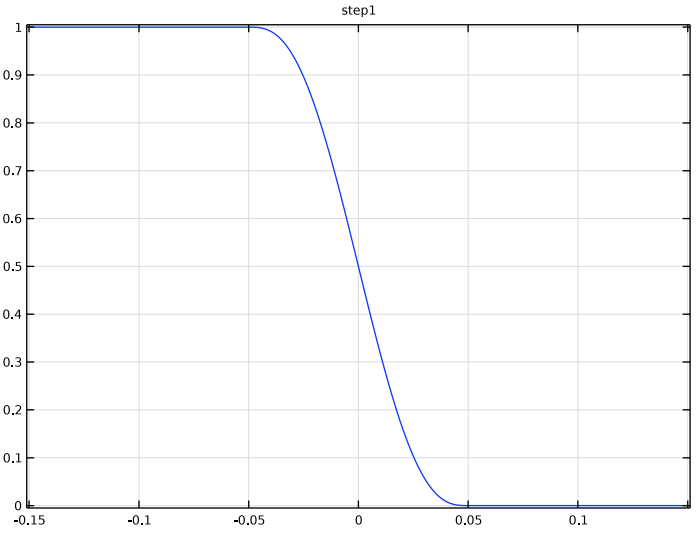
1 In the **Home** toolbar, click f(x) **Functions** and choose **Global>Step**.

2 In the **Settings** window for **Step**, locate the **Parameters** section.

3 In the **From** text field, type 1.

4 In the **To** text field, type 0.

5 Click **Plot**.



Variables 1

Add a number of global variables as follows.

1 In the **Model Builder** window, right-click **Global Definitions** and choose **Variables**.

2 In the **Settings** window for **Variables**, locate the **Variables** section.

3 Click **Load from File**.

4 Browse to the model's Application Libraries folder and double-click the file `particle_size_distribution_variables.txt`.

After import, make sure all variable expressions are colored black, otherwise some state variable or function definition is missing.

GEOMETRY 1

Now proceed to set up the battery model for the non-particle-size-distribution case.

Interval 1 (i1)

1 In the **Model Builder** window, under **Component 1 (comp1)** right-click **Geometry 1** and choose **Interval**.

2 In the **Settings** window for **Interval**, locate the **Interval** section.

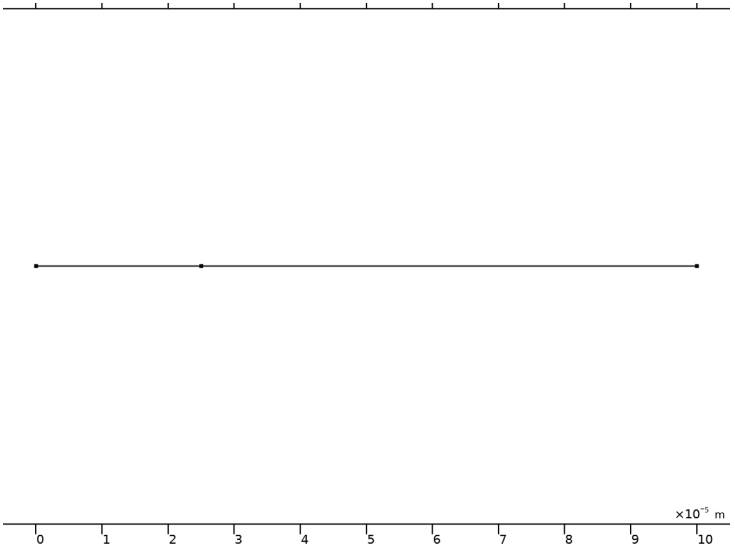
3 From the **Specify** list, choose **Interval lengths**.

4 In the table, enter the following settings:

Lengths (m)
L_sep
L_pos

5 In the **Home** toolbar, click **Build All**.

6 In the **Model Builder** window, click **Geometry 1**.



ADD MATERIAL FROM LIBRARY

In the **Home** toolbar, click **Windows** and choose **Add Material from Library**.

ADD MATERIAL

1 Go to the **Add Material** window.

2 In the tree, select **Battery>Electrodes>Lithium Metal, Li (Negative, Li-ion Battery)**.

3 Right-click and choose **Add to Component 1 (comp1)**.

4 In the tree, select **Battery>Electrodes>NMC 111, LiNi0.33Mn0.33Co0.33O2 (Positive, Li-ion Battery)**.

5 Right-click and choose **Add to Component 1 (comp1)**.

6 In the tree, select **Battery>Electrolytes>LiPF6 in 3:7 EC:EMC (Liquid, Li-ion Battery)**.

7 Right-click and choose **Add to Component 1 (comp1)**.

8 In the **Home** toolbar, click **Add Material** to close the **Add Material** window.

MATERIALS

Lithium Metal, Li (Negative, Li-ion Battery) (mat1)

1 In the **Settings** window for **Material**, locate the **Geometric Entity Selection** section.

2 From the **Geometric entity level** list, choose **Boundary**.

3 Select Boundary 1 only.

NMC 111, LiNi0.33Mn0.33Co0.33O2 (Positive, Li-ion Battery) (mat2)

1 In the **Model Builder** window, click **NMC 111, LiNi0.33Mn0.33Co0.33O2 (Positive, Li-ion Battery) (mat2)**.

2 Select Domain 2 only.

LiPF6 in 3:7 EC:EMC (Liquid, Li-ion Battery) (mat3)

1 In the **Model Builder** window, click **LiPF6 in 3:7 EC:EMC (Liquid, Li-ion Battery) (mat3)**.

2 Select Domain 1 only.

LITHIUM-ION BATTERY (LIION)

Separator 1

1 In the **Model Builder** window, under **Component 1 (comp1)>Lithium-Ion Battery (liion)** click **Separator 1**.

2 In the **Settings** window for **Separator**, locate the **Porous Matrix Properties** section.

3 In the ε_l text field, type `epsl_sep`.

Electrode Surface - Lithium Metal

1 In the **Physics** toolbar, click **Boundaries** and choose **Electrode Surface**.

2 In the **Settings** window for **Electrode Surface**, type `Electrode Surface - Lithium Metal` in the **Label** text field.

3 Select Boundary 1 only.

Porous Electrode - No Particle Size Distribution

1 In the **Physics** toolbar, click **Domains** and choose **Porous Electrode**.

2 In the **Settings** window for **Porous Electrode**, type `Porous Electrode - No Particle Size Distribution` in the **Label** text field.

3 Select Domain 2 only.

4 Locate the **Electrolyte Properties** section. From the **Electrolyte material** list, choose **LiPF6 in 3:7 EC:EMC (Liquid, Li-ion Battery) (mat3)**.

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`.

7 In the ε_l text field, type `epsl_pos`.

Particle Intercalation 1

1 In the **Model Builder** window, click **Particle Intercalation 1**.

2 In the **Settings** window for **Particle Intercalation**, locate the **Species Settings** section.

3 In the $c_{s,init}$ text field, type `cs_init`.

4 Locate the **Particle Transport Properties** section. In the r_p text field, type `Rp_no_distr`.

Porous Electrode Reaction 1

1 In the **Model Builder** window, click **Porous Electrode Reaction 1**.

2 In the **Settings** window for **Porous Electrode Reaction**, locate the **Electrode Kinetics** section.

3 In the $i_{0,ref}(T)$ text field, type `i0_ref_no_distr`.

4 Locate the **Active Specific Surface Area** section. From the **Active specific surface area** list, choose **User defined**. In the a_v text field, type `Av_no_distr`.

Electrode Current Density 1

1 In the **Physics** toolbar, click **Boundaries** and choose **Electrode Current Density**.

2 Select Boundary 3 only.

3 In the **Settings** window for **Electrode Current Density**, locate the **Electrode Current Density** section.

4 In the $i_{n,s}$ text field, type `i_app`.

Initial Values 1

1 In the **Model Builder** window, click **Initial Values 1**.

2 In the **Settings** window for **Initial Values**, locate the **Initial Values** section.

3 In the *phis* text field, type `4[V]`.

STUDY 1 - NO PARTICLE SIZE DISTRIBUTION

1 In the **Model Builder** window, click **Study 1**.

2 In the **Settings** window for **Study**, type `Study 1 - No Particle Size Distribution` in the **Label** text field.

3 Locate the **Study Settings** section. Clear the **Generate default plots** check box.

Step 1: Time Dependent

1 In the **Model Builder** window, under **Study 1 - No Particle Size Distribution** click **Step 1: 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(0,0.5,3)`.

DEFINITIONS (COMP1)

Before solving, add some probes as follows:

Point Probe 1 (point1)

1 In the **Definitions** toolbar, click  **Probes** and choose **Point Probe**.

2 In the **Settings** window for **Point Probe**, type `E_cell_no_distr` in the **Variable name** text field.

3 Locate the **Source Selection** section. Click  **Clear Selection**.

4 Select Boundary 3 only.

5 Click **Replace Expression** in the upper-right corner of the **Expression** section. From the menu, choose **Component 1 (comp1)>Lithium-Ion Battery>phis - Electric potential - V**.

6 Locate the **Expression** section.

7 Select the **Description** check box. In the associated text field, type `Cell voltage`.

8 Click to expand the **Table and Window Settings** section. Click + **Add Table**.

9 Click + **Add Plot Window**.

Point Probe 2 (point2)

1 In the **Definitions** toolbar, click  **Probes** and choose **Point Probe**.

2 In the **Settings** window for **Point Probe**, type `cs_surface_no_distr` in the **Variable name** text field.

3 Locate the **Source Selection** section. Click  **Clear Selection**.

4 Select Boundary 2 only.

5 Click **Replace Expression** in the upper-right corner of the **Expression** section. From the menu, choose **Component 1 (comp1)>Lithium-Ion Battery>Particle intercalation> liion.cs_surface - Insertion particle concentration, surface - mol/m³**.

6 Locate the **Expression** section.

7 Select the **Description** check box. In the associated text field, type `Concentration, Surface`.

8 Locate the **Table and Window Settings** section. Click + **Add Table**.

9 From the **Plot window** list, choose **Probe Plot 2**.

Point Probe 2 (cs_surface_no_distr)

Right-click **Point Probe 2 (cs_surface_no_distr)** and choose **Duplicate**.

Point Probe 3 (point3)

1 In the **Model Builder** window, under **Component 1 (comp1)>Definitions** click **Point Probe 3 (point3)**.

2 In the **Settings** window for **Point Probe**, type `cs_center_no_distr` in the **Variable name** text field.

3 Click **Replace Expression** in the upper-right corner of the **Expression** section. From the menu, choose **Component 1 (comp1)>Lithium-Ion Battery>Particle intercalation> liion.cs_center - Insertion particle concentration, center - mol/m³**.

4 Locate the **Expression** section. In the **Description** text field, type `Concentration, Center`.

STUDY 1 - NO PARTICLE SIZE DISTRIBUTION

In the **Home** toolbar, click = **Compute**.

RESULTS

Probe Plot - Study 1

Polish the probe plot as follows:

1 In the **Model Builder** window, under **Results** click **Probe Plot Group 1**.

2 In the **Settings** window for **1D Plot Group**, type `Probe Plot - Study 1` in the **Label** text field.

3 Locate the **Title** section. From the **Title type** list, choose **None**.

4 Locate the **Plot Settings** section. Select the **Two y-axes** check box.

5 In the table, select the **Plot on secondary y-axis** check box for **Probe Table Graph 2**.

6 Select the **y-axis label** check box. In the associated text field, type `Cell voltage (V)`.

7 Select the **Secondary y-axis label** check box. In the associated text field, type `Concentration (mol/m<sup>3</sup>)`.

8 Locate the **Axis** section. Select the **Manual axis limits** check box.

9 In the **x minimum** text field, type `0`.

10 In the **x maximum** text field, type `3`.

11 In the **y minimum** text field, type `3.94`.

12 In the **y maximum** text field, type `4.07`.

13 In the **Secondary y minimum** text field, type `10000`.

14 In the **Secondary y maximum** text field, type `18000`.

15 Locate the **Legend** section. From the **Position** list, choose **Lower right**.

Probe Table Graph 1

1 In the **Model Builder** window, expand the **Probe Plot - Study 1** node, then click **Probe Table Graph 1**.

2 In the **Settings** window for **Table Graph**, click to expand the **Legends** section.

3 From the **Legends** list, choose **Manual**.

4 In the table, enter the following settings:

Legends
Cell voltage

Probe Table Graph 2

1 In the **Model Builder** window, click **Probe Table Graph 2**.

2 In the **Settings** window for **Table Graph**, locate the **Coloring and Style** section.

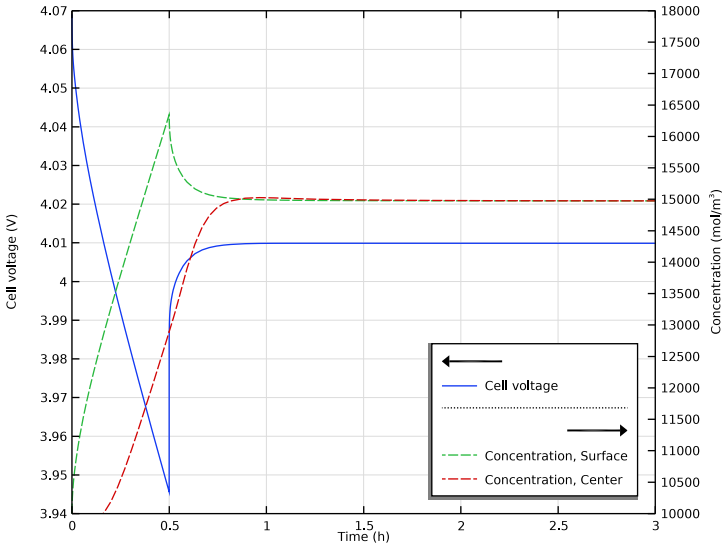
3 Find the **Line style** subsection. From the **Line** list, choose **Dashed**.

4 Locate the **Legends** section. From the **Legends** list, choose **Manual**.

5 In the table, enter the following settings:

Legends
Concentration, Surface
Concentration, Center

6 In the **Probe Plot - Study 1** toolbar, click **Plot**.



GLOBAL DEFINITIONS

We now proceed the second part of the tutorial, where an **Extra Dimension** is added to handle the particle-size distribution.

ADD COMPONENT

In the **Model Builder** window, right-click **Global Definitions** and choose **2D**.

EXTRA DIMENSION 1 (XDIM1)

Name the spatial independent coordinates in the extra dimension `xs` and `ys` as follows:

1 In the **Settings** window for **Extra Dimension**, locate the **Frames** section.

2 Find the **Spatial frame coordinates** subsection. In the table, enter the following settings:

First	Second	Third
xs	ys	z1

GEOMETRY 2

Now draw the geometry in the extra dimension.

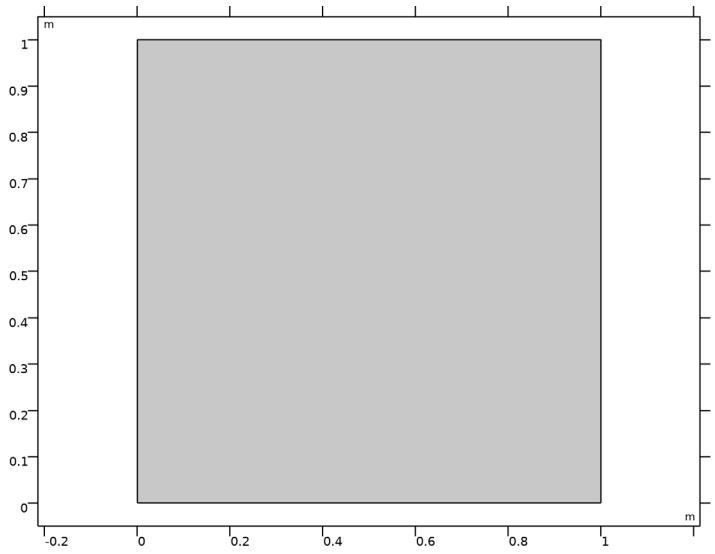
Square 1 (sq1)

1 In the **Geometry** toolbar, click **Square**.

2 Click **Build All**.

3 Click the **Zoom Extents** button in the **Graphics** toolbar.

4 In the **Model Builder** window, click **Geometry 2**.



MESH 2

Define the mesh in the extra dimension as follows:

Mapped 1

In the **Mesh** toolbar, click **Mapped**.

Distribution 1

1 Right-click **Mapped 1** and choose **Distribution**.

2 Select Boundaries 1 and 4 only.

3 In the **Settings** window for **Distribution**, locate the **Distribution** section.

4 In the **Number of elements** text field, type 12.

Distribution 2

Set up the mesh to have a finer resolution toward the surface of the particles as follows:

1 In the **Model Builder** window, right-click **Mapped 1** and choose **Distribution**.

2 Select Boundaries 2 and 3 only.

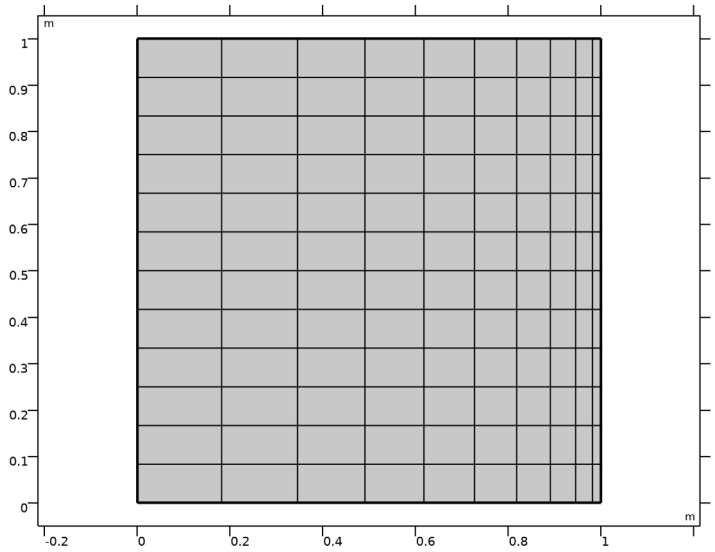
3 In the **Settings** window for **Distribution**, locate the **Distribution** section.

4 From the **Distribution type** list, choose **Predefined**.

5 In the **Number of elements** text field, type 10.

6 In the **Element ratio** text field, type 10.

7 In the **Model Builder** window, right-click **Mesh 2** and choose **Build All**.



DEFINITIONS (XDIM1)

Add a number of integration operators on the extra dimension. These will be used to compute the total current of the whole particle ensemble, and to evaluate the solution in some specific points.

Integration over Extra Dimension - xdint_surf

1 In the **Model Builder** window, expand the **Global Definitions>Extra Dimension 1 (xdim1)> Definitions** node.

2 Right-click **Global Definitions>Extra Dimension 1 (xdim1)>Definitions>Extra Dimensions** and choose **Integration over Extra Dimension**.

3 In the **Settings** window for **Integration over Extra Dimension**, type `Integration over Extra Dimension - xdint_surf` in the **Label** text field.

4 Locate the **Operator Name** section. In the **Operator name** text field, type `xdint_surf`.

5 Locate the **Source Selection** section. From the **Geometric entity level** list, choose **Boundary**.

6 Select Boundary 4 only.

Integration over Extra Dimension - xdint_surf_Rmax

1. Right-click **Extra Dimensions** and choose **Integration over Extra Dimension**.
2. In the **Settings** window for **Integration over Extra Dimension**, type `Integration over Extra Dimension - xdint_surf_Rmax` in the **Label** text field.
3. Locate the **Operator Name** section. In the **Operator name** text field, type `xdint_surf_Rmax`.
4. Locate the **Source Selection** section. From the **Geometric entity level** list, choose **Point**.
5. Select Point 4 only.

Integration over Extra Dimension - xdint_surf_Rmin

1. Right-click **Extra Dimensions** and choose **Integration over Extra Dimension**.
2. In the **Settings** window for **Integration over Extra Dimension**, type `Integration over Extra Dimension - xdint_surf_Rmin` in the **Label** text field.
3. Locate the **Operator Name** section. In the **Operator name** text field, type `xdint_surf_Rmin`.
4. Locate the **Source Selection** section. From the **Geometric entity level** list, choose **Point**.
5. Select Point 3 only.

Integration over Extra Dimension - xdint_center_Rmax

1. Right-click **Extra Dimensions** and choose **Integration over Extra Dimension**.
2. In the **Settings** window for **Integration over Extra Dimension**, type `Integration over Extra Dimension - xdint_center_Rmax` in the **Label** text field.
3. Locate the **Operator Name** section. In the **Operator name** text field, type `xdint_center_Rmax`.
4. Locate the **Source Selection** section. From the **Geometric entity level** list, choose **Point**.
5. Select Point 2 only.

Integration over Extra Dimension - xdint_center_Rmin

1. Right-click **Extra Dimensions** and choose **Integration over Extra Dimension**.
2. In the **Settings** window for **Integration over Extra Dimension**, type `Integration over Extra Dimension - xdint_center_Rmin` in the **Label** text field.
3. Locate the **Operator Name** section. In the **Operator name** text field, type `xdint_center_Rmin`.
4. Locate the **Source Selection** section. From the **Geometric entity level** list, choose **Point**.
5. Select Point 1 only.

EXTRA DIMENSION 1 (XDIM1)

In the **Model Builder** window, collapse the **Global Definitions>Extra Dimension 1 (xdim1)** node.

DEFINITIONS (COMP1)

Before an extra dimension can be used in physics, it must be attached on a selection in the base geometry.

Attached Dimensions 1

1. In the **Model Builder** window, under **Component 1 (comp1)** right-click **Definitions** and choose **Extra Dimensions>Attached Dimensions**.
2. In the **Settings** window for **Attached Dimensions**, locate the **Geometric Entity Selection** section.
3. From the **Geometric entity level** list, choose **Domain**.
4. Select Domain 2 only.
5. Locate the **Attached Dimensions** section. Under **Extra dimensions to attach**, click **+ Add**.
6. In the **Add** dialog box, select **Extra Dimension 1 (xdim1)** in the **Extra dimensions to attach** list.
7. Click **OK**.

Variables - Particle Domain in Extra Dimension

Now add some variable expressions related to the extra dimension.

1. Right-click **Definitions** and choose **Variables**.
2. In the **Settings** window for **Variables**, type `Variables - Particle Domain in Extra Dimension` in the **Label** text field.
3. Locate the **Geometric Entity Selection** section. From the **Geometric entity level** list, choose **Domain**.
4. Select Domain 2 only.
5. From the **Extra dimension attachment** list, choose **Attached Dimensions 1**.

   The particle radius depends linearly on the `ys` spatial variable as follows:
6. Locate the **Variables** section. In the table, enter the following settings:

Name	Expression	Unit	Description
`Rp`	`Rp_min+(Rp_max-Rp_min)*ys[1/m]`	m	`Particle radius`

Variables - Particle Surface in Extra Dimension

1 Right-click **Definitions** and choose **Variables**.

2 In the **Settings** window for **Variables**, type `Variables - Particle Surface in Extra Dimension` in the **Label** text field.

3 Locate the **Geometric Entity Selection** section. From the **Geometric entity level** list, choose **Domain**.

4 Select Domain 2 only.

5 From the **Extra dimension attachment** list, choose **Attached Dimensions 1**.

6 From the **Geometric entity level** list, choose **Boundary**.

7 Select Boundary 4 only.

8 Locate the **Variables** section. Click **Load from File**.

9 Browse to the model's Application Libraries folder and double-click the file `particle_size_distribution_xdim_variables.txt`.

Some expressions will render warnings, indicating missing variable definitions. This is expected at this stage.

Variables - Porous Electrode Domain

1 Right-click **Definitions** and choose **Variables**.

2 In the **Settings** window for **Variables**, type `Variables - Porous Electrode Domain` in the **Label** text field.

3 Locate the **Variables** section. In the table, enter the following settings:

Name	Expression	Unit	Description
`iloc_distr`	`root.xdim1.xdint_surf(iloc* f_hist(Rp)*4*pi*Rp^2)*(Rp_max-Rp_min)[m^-2]/ (area_particles[m^2])`		`Particle-averaged local current density`

4 Click the **Show More Options** button in the **Model Builder** toolbar.

5 In the **Show More Options** dialog box, select **Physics>Equation-Based Contributions** in the tree.

6 In the tree, select the check box for the node **Physics>Equation-Based Contributions**.

7 Click **OK**.

LITHIUM-ION BATTERY (LIION)

Define the particle concentration diffusion in the particles by the use of two weak contribution nodes. The first contribution defines the mass balance on the extra dimension domain. The second contribution defines the lithium flux on the particle surface boundaries.

Weak Contribution - Domain Equation in Extra Dimension

1 In the **Physics** toolbar, click **Domains** and choose **Weak Contribution**.

2 In the **Settings** window for **Weak Contribution**, type `Weak Contribution - Domain Equation in Extra Dimension` in the **Label** text field.

3 Select Domain 2 only.

4 Locate the **Domain Selection** section. From the **Extra dimension attachment** list, choose **Attached Dimensions 1**.

5 From the **Selection** list, choose **All domains**.

6 Locate the **Weak Contribution** section. In the **Weak expression** text field, type `xs^2*(-Rp^2*test(cs)*d(cs,TIME)-d(cs,xs)*Ds*test(d(cs,xs)[m^2]))`.

The expression you just typed in indicates an unknown variable `cs`. Define the dependent variable `cs`, representing the solid lithium concentration in the particles, as follows:

Auxiliary Dependent Variable - cs

1 In the **Physics** toolbar, click **Attributes** and choose **Auxiliary Dependent Variable**.

2 In the **Settings** window for **Auxiliary Dependent Variable**, type `Auxiliary Dependent Variable - cs` in the **Label** text field.

3 Locate the **Domain Selection** section. From the **Extra dimension attachment** list, choose **Attached Dimensions 1**.

4 From the **Selection** list, choose **All domains**.

5 Locate the **Auxiliary Dependent Variable** section. In the **Field variable name** text field, type `cs`.

6 In the **Initial value** text field, type `cs_init`.

Weak Contribution - Domain Equation in Extra Dimension

The expression in the **Weak expression**edit field should now be colored black. You may also go back to the three **Variables** nodes you defined before in **Component 1** to check that these are now colored black, not indicating any missing variables or operators.

Weak Contribution - Boundary Condition in Extra Dimension

1 In the **Physics** toolbar, click **Domains** and choose **Weak Contribution**.

2 In the **Settings** window for **Weak Contribution**, type `Weak Contribution - Boundary Condition in Extra Dimension` in the **Label** text field.

3 Select Domain 2 only.

4 Locate the **Domain Selection** section. From the **Extra dimension attachment** list, choose **Attached Dimensions 1**.

5 From the **Geometric entity level** list, choose **Boundary**.

6 Select Boundary 4 only.

7 Locate the **Weak Contribution** section. In the **Weak expression** text field, type `xs^2*(-iloc/F_const)*test(cs)*Rp`.

Porous Electrode - No Particle Size Distribution

Define the porous electrode model for the particle size distribution as follows:

1 In the **Model Builder** window, right-click **Porous Electrode - No Particle Size Distribution** and choose **Duplicate**.

Porous Electrode - With Particle Size Distribution

1 In the **Model Builder** window, under **Component 1 (comp1)>Lithium-Ion Battery (liion)** click **Porous Electrode - No Particle Size Distribution 1**.

2 In the **Settings** window for **Porous Electrode**, type `Porous Electrode - With Particle Size Distribution` in the **Label** text field.

By selecting **Nonintercalating particles**, the built-in framework for defining particle diffusion in particles of a single size is turned off.

3 Locate the **Particle Properties** section. From the list, choose **Nonintercalating particles**.

Porous Electrode Reaction 1

1 In the **Model Builder** window, expand the **Porous Electrode - With Particle Size Distribution** node, then click **Porous Electrode Reaction 1**.

2 In the **Settings** window for **Porous Electrode Reaction**, locate the **Electrode Kinetics** section.

3 From the $i_{\text{loc,expr}}$ list, choose **User defined**. In the associated text field, type `iloc_distr`.

4 Locate the **Active Specific Surface Area** section. In the a_{v} text field, type `Av_distr`.

DEFINITIONS (COMP1)

Before solving, add a series of new probes related to the particle size distribution model.

Point Probe 4 (point4)

1 In the **Definitions** toolbar, click **Probes** and choose **Point Probe**.

2 In the **Settings** window for **Point Probe**, type `E_cell_distr` in the **Variable name** text field.

3 Locate the **Source Selection** section. Click **Clear Selection**.

4 Select Boundary 3 only.

5 Click **Replace Expression** in the upper-right corner of the **Expression** section. From the menu, choose **Component 1 (comp1)>Lithium-Ion Battery>phis - Electric potential - V**.

6 Locate the **Expression** section.

7 Select the **Description** check box. In the associated text field, type `Cell voltage`.

8 Locate the **Table and Window Settings** section. Click **Add Table**.

9 Click **Add Plot Window**.

Point Probe 5 (point5)

1 In the **Definitions** toolbar, click **Probes** and choose **Point Probe**.

2 In the **Settings** window for **Point Probe**, type `cs_surface_Rmax` in the **Variable name** text field.

3 Locate the **Source Selection** section. Click **Clear Selection**.

4 Select Boundary 2 only.

5 Locate the **Expression** section. In the **Expression** text field, type `root.xdim1.xdint_surf_Rmax(cs)`.

6 Select the **Description** check box. In the associated text field, type `Concentration, Surface, Largest Particles`.

7 Locate the **Table and Window Settings** section. Click **Add Table**.

8 From the **Plot window** list, choose **Probe Plot 3**.

Point Probe 5 (cs_surface_Rmax)

Right-click **Point Probe 5 (cs_surface_Rmax)** and choose **Duplicate**.

Point Probe 6 (point6)

1 In the **Model Builder** window, under **Component 1 (comp1)>Definitions** click **Point Probe 6 (point6)**.

2 In the **Settings** window for **Point Probe**, type `cs_center_Rmax` in the **Variable name** text field.

3 Locate the **Expression** section. In the **Expression** text field, type `root.xdim1.xdint_center_Rmax(cs)`.

4 In the **Description** text field, type `Concentration, Center, Largest Particles`.

Point Probe 6 (cs_center_Rmax)

Right-click **Point Probe 6 (cs_center_Rmax)** and choose **Duplicate**.

Point Probe 7 (point7)

1 In the **Model Builder** window, under **Component 1 (comp1)>Definitions** click **Point Probe 7 (point7)**.

2 In the **Settings** window for **Point Probe**, type `cs_surface_Rmin` in the **Variable name** text field.

3 Locate the **Expression** section. In the **Expression** text field, type `root.xdim1.xdint_surf_Rmin(cs)`.

4 In the **Description** text field, type `Concentration, Surface, Smallest Particles`.

Point Probe 7 (cs_surface_Rmin)

Right-click **Point Probe 7 (cs_surface_Rmin)** and choose **Duplicate**.

Point Probe 8 (point8)

1 In the **Model Builder** window, under **Component 1 (comp1)>Definitions** click **Point Probe 8 (point8)**.

2 In the **Settings** window for **Point Probe**, type `cs_center_Rmin` in the **Variable name** text field.

3 Locate the **Expression** section. In the **Expression** text field, type `root.xdim1.xdint_center_Rmin(cs)`.

4 In the **Description** text field, type `Concentration, Center, Smallest Particles`.

STUDY 1 - NO PARTICLE SIZE DISTRIBUTION

In the **Model Builder** window, collapse the **Study 1 - No Particle Size Distribution** node.

ADD STUDY

1 In the **Home** toolbar, click **Windows** and choose **Add Study**.

2 Go to the **Add Study** window.

3 Find the **Studies** subsection. In the **Select Study** tree, select **General Studies> Time Dependent**.

4 Click **Add Study** in the window toolbar.

5 In the **Home** toolbar, click **Add Study** to close the **Add Study** window.

STUDY 2 - WITH PARTICLE SIZE DISTRIBUTION

1 In the **Model Builder** window, click **Study 2**.

2 In the **Settings** window for **Study**, type `Study 2 - With Particle Size Distribution` in the **Label** text field.

Step 1: Time Dependent

1 In the **Model Builder** window, under **Study 2 - With Particle Size Distribution** click **Step 1: 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(0,0.5,3)`.

5 Click to expand the **Results While Solving** section. From the **Probes** list, choose **Manual**.

6 In the **Probes** list, choose **Point Probe 1 (E_cell_no_distr)**, **Point Probe 2 (cs_surface_no_distr)**, and **Point Probe 3 (cs_center_no_distr)**.

7 Under **Probes**, click  **Delete**.

8 Locate the **Physics and Variables Selection** section. Select the **Modify model configuration for study step** check box.

9 In the tree, select **Component 1 (comp1)>Lithium-Ion Battery (liion)>Porous Electrode - No Particle Size Distribution**.

10 Right-click and choose **Disable**.

11 In the **Model Builder** window, click **Study 2 - With Particle Size Distribution**.

12 In the **Settings** window for **Study**, locate the **Study Settings** section.

13 Clear the **Generate default plots** check box.

14 In the **Home** toolbar, click  **Compute**.

RESULTS

Probe Plot - Study 2

1 In the **Model Builder** window, under **Results** click **Probe Plot Group 2**.

2 In the **Settings** window for **1D Plot Group**, type `Probe Plot - Study 2` in the **Label** text field.

3 Locate the **Title** section. From the **Title type** list, choose **None**.

4 Locate the **Plot Settings** section. Select the **Two y-axes** check box.

5 In the table, select the **Plot on secondary y-axis** check box for **Probe Table Graph 2**.

6 Select the **y-axis label** check box. In the associated text field, type `Cell voltage (V)`.

7 Select the **Secondary y-axis label** check box. In the associated text field, type `Concentration (mol/m<sup>3</sup>)`.

8 Locate the **Axis** section. Select the **Manual axis limits** check box.

9 In the **x minimum** text field, type `0`.

10 In the **x maximum** text field, type `3`.

11 In the **y minimum** text field, type `3.94`.

12 In the **y maximum** text field, type `4.07`.

13 In the **Secondary y minimum** text field, type `10000`.

14 In the **Secondary y maximum** text field, type `18000`.

15 Locate the **Legend** section. From the **Position** list, choose **Lower right**.

Probe Table Graph 1

1 In the **Model Builder** window, expand the **Probe Plot - Study 2** node, then click **Probe Table Graph 1**.

2 In the **Settings** window for **Table Graph**, locate the **Legends** section.

3 From the **Legends** list, choose **Manual**.

4 In the table, enter the following settings:

Legends
Cell voltage

Probe Table Graph 2

1 In the **Model Builder** window, click **Probe Table Graph 2**.

2 In the **Settings** window for **Table Graph**, locate the **Coloring and Style** section.

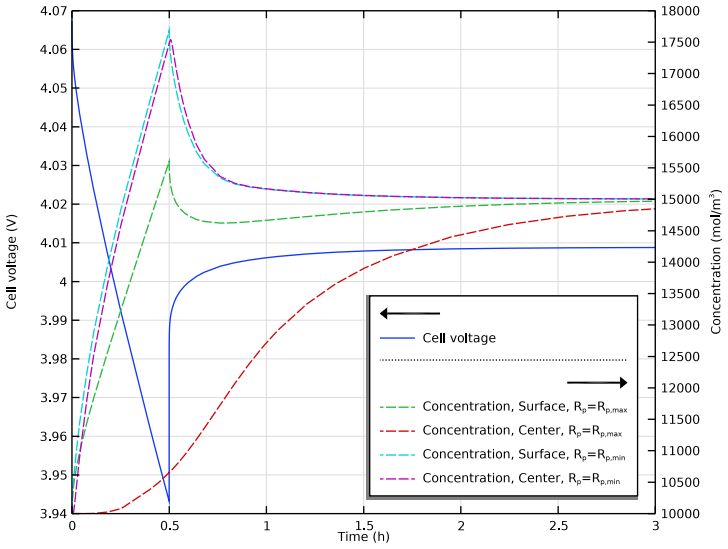
3 Find the **Line style** subsection. From the **Line** list, choose **Dashed**.

4 Locate the **Legends** section. From the **Legends** list, choose **Manual**.

5 In the table, enter the following settings:

Legends
Concentration, Surface, R_p=R_{p,max}
Concentration, Center, R_p=R_{p,max}
Concentration, Surface, R_p=R_{p,min}
Concentration, Center, R_p=R_{p,min}

6 In the **Probe Plot - Study 2** toolbar, click **Plot**.



Potential Comparison

1 In the **Home** toolbar, click **Add Plot Group** and choose **1D Plot Group**.

2 In the **Settings** window for **1D Plot Group**, type `Potential Comparison` in the **Label** text field.

Probe Table Graph 1

In the **Model Builder** window, under **Results>Probe Plot - Study 1** right-click **Probe Table Graph 1** and choose **Copy**.

Potential Comparison

In the **Model Builder** window, under **Results** right-click **Potential Comparison** and choose **Paste Table Graph**.

Probe Table Graph 1

In the **Model Builder** window, under **Results>Probe Plot - Study 2** right-click **Probe Table Graph 1** and choose **Copy**.

Potential Comparison

In the **Model Builder** window, under **Results** right-click **Potential Comparison** and choose **Paste Table Graph**.

Probe Table Graph 1

1 In the **Model Builder** window, click **Probe Table Graph 1**.

2 In the **Settings** window for **Table Graph**, locate the **Legends** section.

3 In the table, enter the following settings:

Legends
No size distribution

Probe Table Graph 1.1

1 In the **Model Builder** window, click **Probe Table Graph 1.1**.

2 In the **Settings** window for **Table Graph**, locate the **Legends** section.

3 In the table, enter the following settings:

Legends
With size distribution

Global 1

1 In the **Model Builder** window, right-click **Potential Comparison** and choose **Global**.

2 In the **Settings** window for **Global**, locate the **Data** section.

3 From the **Dataset** list, choose **Study 2 - With Particle Size Distribution/Solution 2 (sol2)**.

4 From the **Time selection** list, choose **Interpolated**.

5 In the **Times (h)** text field, type `range(0,0.01,3)`.

6 Locate the **y-Axis Data** section. In the table, enter the following settings:

Expression	Unit	Description
i_app	A/m^2	Applied current density

Potential Comparison

1 In the **Model Builder** window, click **Potential Comparison**.

2 In the **Settings** window for **1D Plot Group**, locate the **Title** section.

3 From the **Title type** list, choose **None**.

4 Locate the **Plot Settings** section. Select the **Two y-axes** check box.

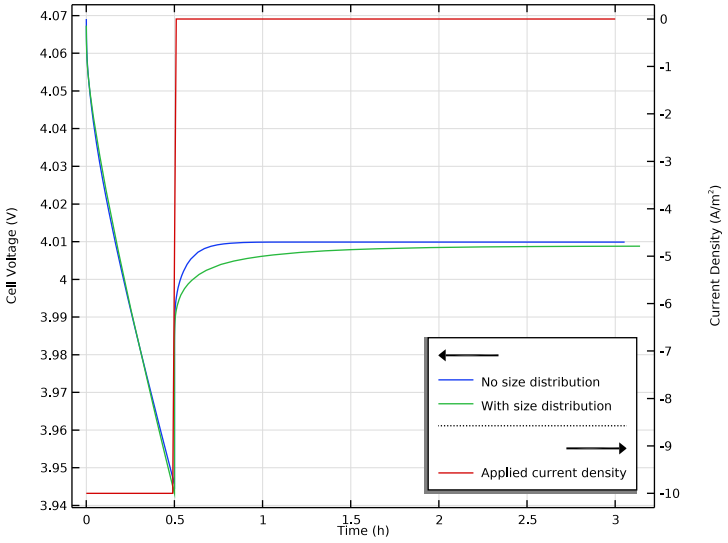
5 In the table, select the **Plot on secondary y-axis** check box for **Global 1**.

6 Select the **y-axis label** check box. In the associated text field, type `Cell Voltage (V)`.

7 Select the **Secondary y-axis label** check box. In the associated text field, type `Current Density (A/m<sup>2</sup>)`.

8 Locate the **Legend** section. From the **Position** list, choose **Lower right**.

9 In the **Potential Comparison** toolbar, click **Plot**.



Study 2 - With Particle Size Distribution/Solution 2 (sol2)

To create plots on the extra dimension, a new dataset needs to be created.

In the **Model Builder** window, under **Results>Datasets** right-click **Study 2 - With Particle Size Distribution/Solution 2 (sol2)** and choose **Duplicate**.

Study 2 - With Particle Size Distribution/Solution - xdim

1 In the **Model Builder** window, under **Results>Datasets** click **Study 2 - With Particle Size Distribution/Solution 2 (4) (sol2)**.

2 In the **Settings** window for **Solution**, type `Study 2 - With Particle Size Distribution/Solution - xdim` in the **Label** text field.

3 Locate the **Solution** section. From the **Component** list, choose **Extra Dimension 1 (xdim1)**.

Concentration Distribution in Particles Adjacent to Separator

1 In the **Home** toolbar, click **Add Plot Group** and choose **2D Plot Group**.

2 In the **Settings** window for **2D Plot Group**, type `Concentration Distribution in Particles Adjacent to Separator` in the **Label** text field.

3 Locate the **Data** section. From the **Dataset** list, choose **Study 2 - With Particle Size Distribution/Solution - xdim (sol2)**.

4 From the **Time (h)** list, choose **0.5**.

Surface 1

1 Right-click **Concentration Distribution in Particles Adjacent to Separator** and choose **Surface**.

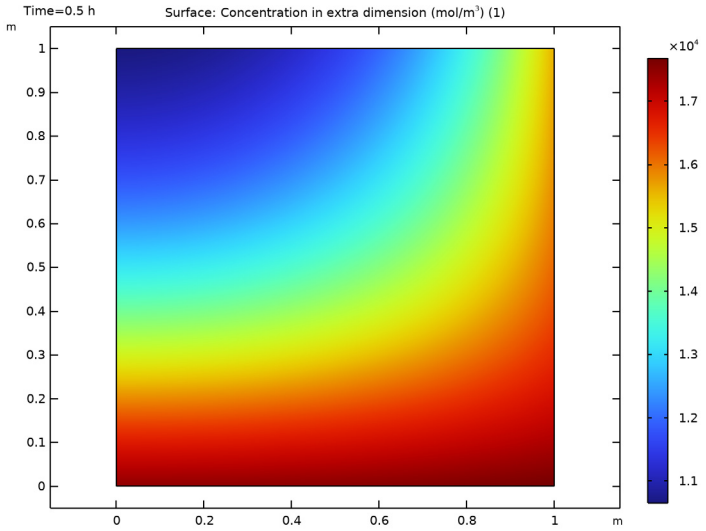
2 In the **Settings** window for **Surface**, locate the **Expression** section.

3 In the **Expression** text field, type `comp1.atxd1(L_sep+L_pos/1000,cs)`.

In the above expression, the `atxd1` operator defines where in the base dimension the evaluation should be made.

4 Select the **Description** check box. In the associated text field, type `Concentration in extra dimension (mol/m<sup>3</sup>)`.

5 In the **Concentration Distribution in Particles Adjacent to Separator** toolbar, click **Plot**.



Concentration Distribution in Particles Adjacent to Separator

In the **Model Builder** window, right-click **Concentration Distribution in Particles Adjacent to Separator** and choose **Duplicate**.

Concentration Distribution in Particles Adjacent to Current Collector

1 In the **Model Builder** window, under **Results** click **Concentration Distribution in Particles Adjacent to Separator 1**.

2 In the **Settings** window for **2D Plot Group**, type `Concentration Distribution in Particles Adjacent to Current Collector` in the **Label** text field.

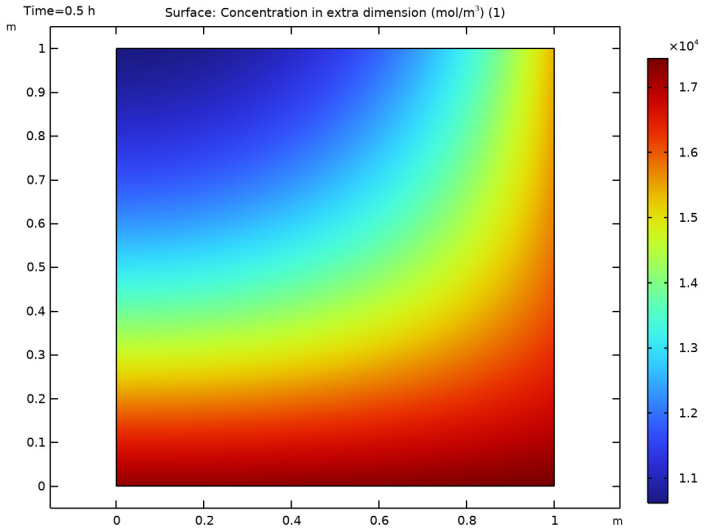
Surface 1

1 In the **Model Builder** window, expand the **Concentration Distribution in Particles Adjacent to Current Collector** node, then click **Surface 1**.

2 In the **Settings** window for **Surface**, locate the **Expression** section.

3 In the **Expression** text field, type `comp1.atxd1(L_sep+L_pos*0.999,cs)`.

4 In the **Concentration Distribution in Particles Adjacent to Current Collector** toolbar, click **Plot**.



STUDY 1 - NO PARTICLE SIZE DISTRIBUTION

Go back to the study for the no-particle-size-distribution case and turn off features related to the particle distribution. In this way, the old study can be recomputed at any time for the no-distribution case.

Step 1: Time Dependent

1 In the **Model Builder** window, under **Study 1 - No Particle Size Distribution** click **Step 1: Time Dependent**.

2 In the **Settings** window for **Time Dependent**, click to expand the **Results While Solving** section.

3 From the **Probes** list, choose **Manual**.

4 In the **Probes** list, choose **Point Probe 4 (E_cell_distr)**, **Point Probe 5 (cs_surface_Rmax)**, **Point Probe 6 (cs_center_Rmax)**, **Point Probe 7 (cs_surface_Rmin)**, and **Point Probe 8 (cs_center_Rmin)**.

5 Under **Probes**, click **Delete**.

6 Locate the **Physics and Variables Selection** section. Select the **Modify model configuration for study step** check box.

7 In the tree, select **Component 1 (comp1)>Lithium-Ion Battery (liion)>Weak Contribution - Domain Equation in Extra Dimension**.

8 Right-click and choose **Disable**.

9 In the tree, select **Component 1 (comp1)>Lithium-Ion Battery (liion)>Weak Contribution - Boundary Condition in Extra Dimension**.

10 Right-click and choose **Disable**.

11 In the tree, select **Component 1 (comp1)>Lithium-Ion Battery (liion)>Porous Electrode - With Particle Size Distribution**.

12 Right-click and choose **Disable**.