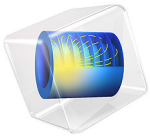

Optimization of a Catalytic Microreactor

Introduction

In this model, a solution is pumped through a catalytic bed, where a reactant undergoes chemical reaction as it gets in contact with the catalyst. The purpose of the example is to maximize the total reaction rate for a given total pressure difference across the bed. This is achieved by finding an optimal catalyst distribution. The distribution of the porous catalyst determines the total reaction rate in the bed. A large amount of catalyst results in a low flow rate through the bed, while less catalyst gives a high flow rate but low conversion of the reactant.

This modeling example is based on Ref. 1.

Note: This application requires the Optimization Module.

Model Definition

The model geometry is shown in Figure 1. The reactor consists of an inlet channel, a fixed catalytic bed, and an outlet channel.

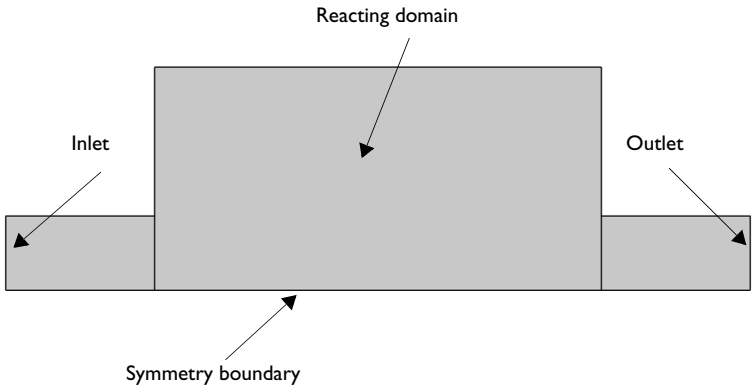

Figure 1: Model geometry.

The optimal catalyst distribution should maximize the average reaction rate, which is expressed as the integral of the local reaction rate, r (SI unit: mol/(m^3·s)), over the domain, Ω. This is equivalent to minimizing the negative of this average reaction rate:

$$\min\left\{-\frac{1}{\mathrm{vol}(\Omega)}\int_{\Omega} r\,d\Omega\right\}$$

Assuming a first-order catalytic reaction with respect to the reactant species, the local reaction rate is determined by

$$r = k_a(1-\varepsilon)c \tag{1}$$

where ε denotes the volume fraction of solid catalyst, c refers to the concentration (SI unit: mol/m^3), and k_a is the rate constant (SI unit: 1/s).

The mass transport is described by the convection and diffusion equation

$$\nabla\cdot(-D\nabla c) = r - \mathbf{u}\cdot\nabla c$$

where $\mathbf{u}$ denotes the velocity vector (SI unit: m/s) and D is the diffusion coefficient (SI unit: m^2/s). The Navier-Stokes equations describe the fluid flow:

$$\begin{aligned}\rho(\mathbf{u}\cdot\nabla)\mathbf{u} &= -\nabla p + \nabla\cdot\mu(\nabla\mathbf{u} + (\nabla\mathbf{u})^T) - \alpha(\varepsilon)\mathbf{u}\\ \nabla\cdot\mathbf{u} &= 0\end{aligned} \tag{2}$$

The coefficient $\alpha(\varepsilon)$ depends on the distribution of the porous catalyst as

$$\alpha(\varepsilon) = \frac{\mu}{\mathrm{Da}\cdot L^2}\cdot\frac{q(1-\varepsilon)}{q+\varepsilon} \tag{3}$$

where Da is the Darcy number; L is the length scale (SI unit: m); and q is a dimensionless parameter, the interpretation of which is discussed in the next section.

From Equation 3, the direct conclusion is that when ε equals 1, α equals zero and Equation 2 reduces to the ordinary Navier-Stokes equations. In this case the reaction rate is zero; see Equation 1.

To summarize, the optimization problem is

$$\min_{\varepsilon}\left\{-\frac{1}{\mathrm{vol}(\Omega)}\int_{\Omega}(k_a(1-\varepsilon)c)\,d\Omega\right\} \tag{4}$$

where

$$\rho(\mathbf{u}\cdot\nabla)\mathbf{u} = -\nabla p + \nabla\cdot\mu(\nabla\mathbf{u} + (\nabla\mathbf{u})^T) - \alpha(\varepsilon)\mathbf{u}$$
$$\nabla\cdot\mathbf{u} = 0$$

$$\nabla\cdot(-D\nabla c) = r - \mathbf{u}\cdot\nabla c$$

$$0 \le \varepsilon \le 1$$

and physical boundary conditions apply.

CONVEX OPTIMIZATION PROBLEMS

One of the most important characteristics of an optimization problem is whether or not the problem is *convex*. This section therefore briefly describes this property. For a more general discussion of the subject, see for example Ref. 2.

A set C is said to be convex if for any two members x, y of C, the following relation holds:

$$tx + (1-t)y \in C \quad \text{for every } t \in [0, 1]$$

that is, the straight line between x and y is fully contained in C. A convex function is a mapping f from a convex set C such that for every two members x, y of C

$$f(tx + (1-t)y) \le t\,f(x) + (1-t)f(y) \quad \text{for every } t \in [0, 1] \tag{5}$$

An optimization problem is said to be convex if the following conditions are met:

- the design domain is convex
- the objective and constraints are convex functions

The importance of convexity follows simply from the result that *if x^* is a local minimum to a convex optimization problem, then x^* is also a global minimum*. This is easily proven by simply assuming that there is a y such that $f(y) < f(x^*)$, and then using Equation 5.

This particular optimization problem is nonlinear, because a change in ε implies a change in the concentration, c. Because of this implicit dependence, it is very difficult to determine whether or not the objective is convex. There is therefore no guarantee that the optimal solution you obtain is globally optimal or unique. In the best of cases, running the optimization gives a good local optimum.

The parameter q can be used to smoothen the interfaces between the catalyst and the open channel. To see the effect of this parameter, rewrite Equation 3 as

$$\alpha(\varepsilon) = \frac{\mu}{\mathrm{Da}\cdot L^2}\cdot\frac{1-\varepsilon}{1+\dfrac{\varepsilon}{q}}$$

It follows that when q approaches infinity, α is the (inverse) porosity. On the other hand, lowering the value of q decreases the magnitude of α.

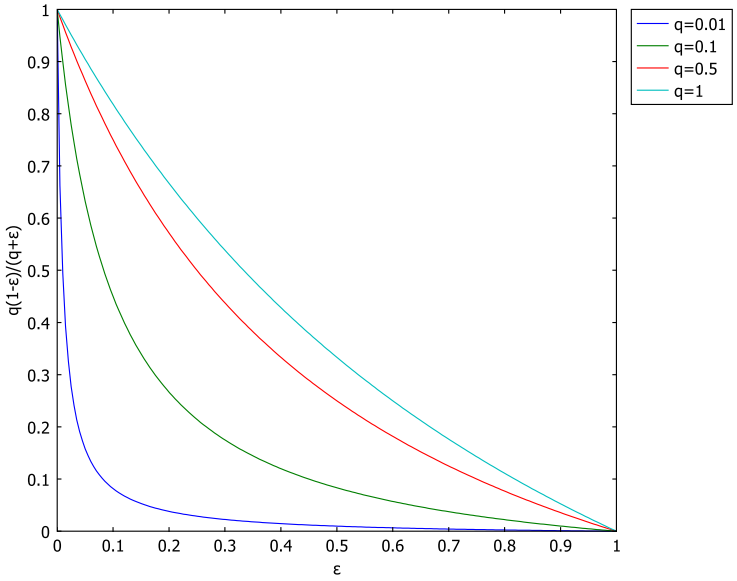

Figure 2: $q(1-\varepsilon)/(q-\varepsilon)$ plotted as a function of ε for different values of q.

Figure 2 shows $q(1-\varepsilon)/(q-\varepsilon)$ plotted as a function of ε for different values of q. This plot shows that lowering the value of q, increases the convexity of the force coefficient. For a low q value, an increase in ε around 0.5, imposes a small increase of the force coefficient, while for a higher value of q, a change in ε imposes an almost equal change for the whole range. Therefore, for a lower q value, the solution is not sharp at the interfaces. On the other hand, for small values of ε, the force term decreases rapidly when q is small, and thus affects the flow field to a much wider extent. In the limit when q approaches infinity, α as a function of ε is a straight line.

Results and Discussion

Figure 3 shows the velocity field in the empty channel. This is the starting point for the optimization.

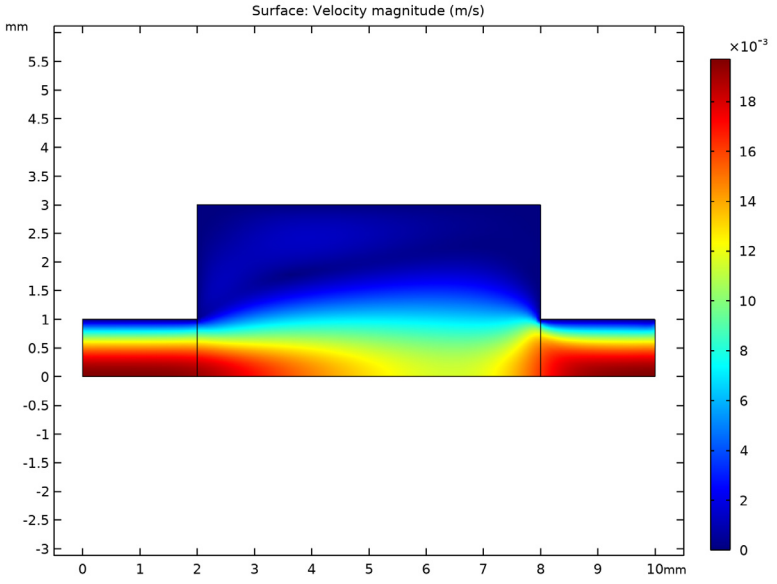

Figure 3: Velocity field in the open channel.

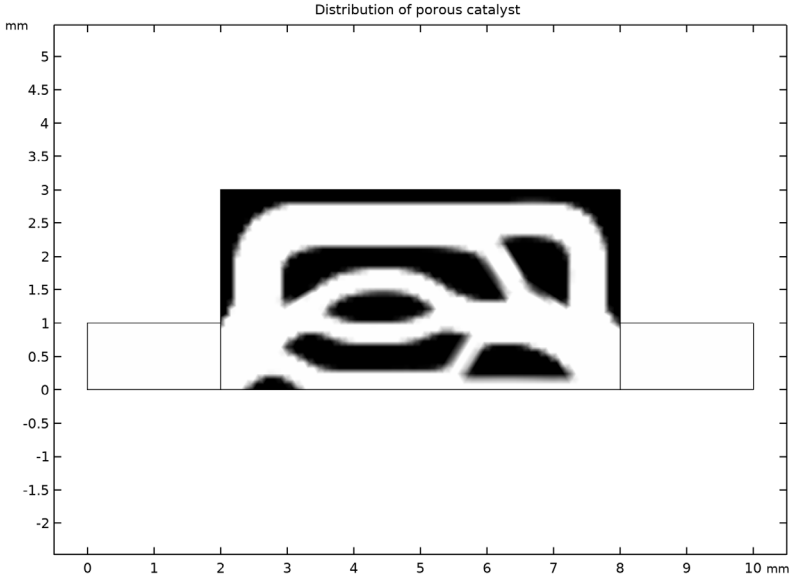

Figure 4: Distribution of the porous catalyst seen in black and open channel in white.

Figure 4 shows the distribution of the porous catalyst in black and the open channels in white. This result shows that, optimally, the supply of the reactant should be distributed over a large area of the reactor. Note also that the amount of open channel volume is significant.

Figure 5 shows the concentration distribution in the reactor. This plot shows how the porous catalyst is fed with the reactant through the open channels. The plot naturally resembles that of Figure 4.

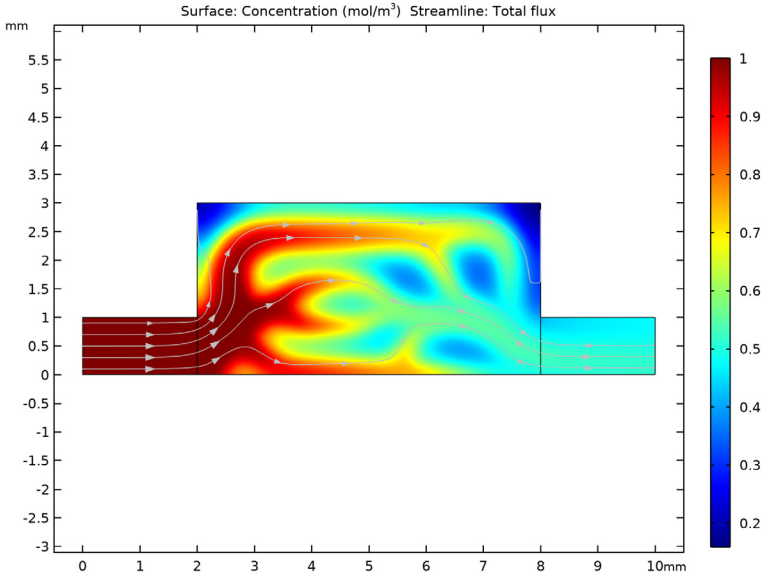

Figure 5: Concentration distribution in the reactor after optimization.

Let

$$F_i = \int_{\partial\Omega_i} \mathbf{n}_{\text{flow}} \cdot (-D\nabla c + c\mathbf{u}) ds \,,$$

where $\mathbf{n}_{\text{flow}}$ refers to the normal to the boundary $\partial\Omega_i$ in the flow direction (that is, pointing in to the domain at the inlet and out from the domain at the outlet). Then F_i is a measurement of the flow of the species with concentration c through the boundary $\partial\Omega_i$ per unit length in the transverse dimension. The conversion, X, of the reactant is defined as

$$X = \frac{F_{\text{in}} - F_{\text{out}}}{F_{\text{in}}}$$

In this case, the conversion of the reactant is around 50%.

Figure 6 shows the velocity field in the reactor. The porous catalyst slows down the flow significantly compared to Figure 3.

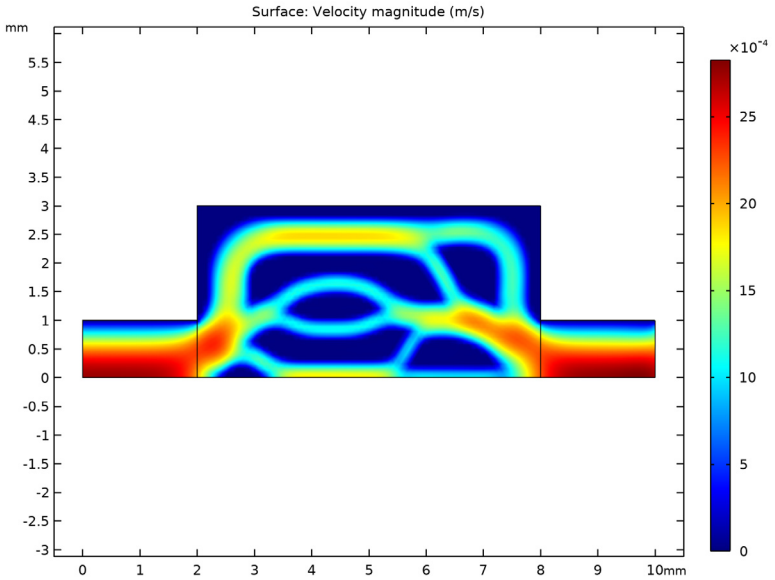

Figure 6: Velocity field in the reactor after optimization.

References

1. F. Okkels and H. Bruus, "Scaling Behavior of Optimally Structured Catalytic Microfluidic Reactors," *Phys. Rev. E*, vol. 75, pp. 016301 1–4, 2007.

2. S.G. Nash and A. Sofer, *Linear and Nonlinear Programming*, McGraw-Hill, 1995.

Application Library path: `Chemical_Reaction_Engineering_Module/Reactors_with_Mass_Transfer/microreactor_optimization`

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 **2D**.

2 In the **Select Physics** tree, select **Fluid Flow>Single-Phase Flow>Laminar Flow (spf)**.

3 Click **Add**.

4 In the **Select Physics** tree, select **Chemical Species Transport>Transport of Diluted Species (tds)**.

5 Click **Add**.

6 Click **Study**.

7 In the **Select Study** tree, select **General Studies>Stationary**.

8 Click **Done**.

ROOT

Load parameters from a text file.

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 `microreactor_optimization_parameters.txt`.

GEOMETRY 1

Next, create the geometry. The reactor consists of three domains: the inlet channel, the reacting domain, and the outlet channel (see Figure 1).

1 In the **Model Builder** window, under **Component 1 (comp1)** click **Geometry 1**.

2 In the **Settings** window for **Geometry**, locate the **Units** section.

3 From the **Length unit** list, choose **mm**.

Rectangle 1 (r1)

1 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 `2*L`.

4 In the **Height** text field, type `L`.

Rectangle 2 (r2)

1 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 `6*L`.

4 In the **Height** text field, type `3*L`.

5 Locate the **Position** section. In the **x** text field, type `2*L`.

Rectangle 3 (r3)

1 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 `2*L`.

4 In the **Height** text field, type `L`.

5 Locate the **Position** section. In the **x** text field, type `8*L`.

6 Click **Build Selected**.

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

The geometry should now look like that in Figure 1.

DEFINITIONS

Define integration couplings to use for calculating the conversion of the reactant.

Integration 1 (intop1)

1 In the **Definitions** toolbar, click **Nonlocal Couplings** and choose **Integration**.

2 In the **Settings** window for **Integration**, locate the **Source Selection** section.

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

4 Select Boundary 1 only.

Integration 2 (intop2)

1 In the **Definitions** toolbar, click **Nonlocal Couplings** and choose **Integration**.

2 In the **Settings** window for **Integration**, locate the **Source Selection** section.

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

4 Select Boundary 12 only.

Integration 3 (intop3)

1 In the **Definitions** toolbar, click **Nonlocal Couplings** and choose **Integration**.

2 Select Domain 2 only.

Add a density topology feature, which can be used to distinguish between free flow and solid regions. This variable will be coupled back to the **Laminar Flow** interfaces later.

Density Model 1 (dtopo1)

1 In the **Definitions** toolbar, click **Optimization** and choose **Density Model**.

Only the center part of the channel geometry is needed in the optimization, so you only have to define the feature there.

2 Select Domain 2 only.

3 In the **Settings** window for **Density Model**, locate the **Filtering** section.

4 From the **Filter type** list, choose **None**.

5 Locate the **Interpolation** section. From the **Interpolation type** list, choose **Darcy**.

6 In the q_{Darcy} text field, type `q`.

7 Locate the **Control Variable Initial Value** section. In the θ_0 text field, type `1`.

Now the design variable used in the optimization is defined. The initial value 1 corresponds to a channel free from porous material.

Variables 1

1 In the **Definitions** toolbar, click **Local Variables**.

Use the integration operators to define the conversion of reactant and the scaled objective function.

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

3 In the table, enter the following settings:

Name	Expression	Unit	Description
`F_in`	`intop1(tds.tflux_cx)`	mol/(m·s)	`Molar flow at inlet`
`F_out`	`intop2(tds.tflux_cx)`	mol/(m·s)	`Molar flow at outlet`
`X_conv`	`(F_in-F_out)/F_in`		`Conversion of reactant`
`obj`	`intop3(-phi/vol)`		`Objective function`

Here, `tds.tfluxx_c` is the COMSOL Multiphysics variable for the x-component of the total flux.

Variables 2

1 In the **Definitions** toolbar, click **Local Variables**.

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

3 From the **Geometric entity level** list, choose **Domain**.

4 Select Domain 2 only.

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

Name	Expression	Unit	Description
phi	k_a*(1-dtopo1.theta)*c	mol/(m³·s)	Local reaction rate
alpha	(eta/(Da*L^2))*dtopo1.theta_p	Pa·s/m²	Drag-force coefficient

ADD MATERIAL

1 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 **Built-in>Water, liquid**.

4 Click **Add to Component** in the window toolbar.

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

LAMINAR FLOW (SPF)

Volume Force 1

1 In the **Model Builder** window, under **Component 1 (comp1)** right-click **Laminar Flow (spf)** and choose **Volume Force**.

2 Select Domain 2 only.

3 In the **Settings** window for **Volume Force**, locate the **Volume Force** section.

4 Specify the $\mathbf{F}$ vector as

-alpha*u	x
-alpha*v	y

Inlet 1

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

2 Select Boundary 1 only.

3 In the **Settings** window for **Inlet**, locate the **Boundary Condition** section.

4 From the list, choose **Pressure**.

5 Locate the **Pressure Conditions** section. In the p_0 text field, type `delta_p`.

Symmetry 1

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

2 Select Boundaries 2, 5, and 9 only.

Outlet 1

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

2 Select Boundary 12 only.

TRANSPORT OF DILUTED SPECIES (TDS)

Transport Properties 1

1 In the **Model Builder** window, under **Component 1 (comp1)>** **Transport of Diluted Species (tds)** click **Transport Properties 1**.

2 In the **Settings** window for **Transport Properties**, locate the **Diffusion** section.

3 In the D_c text field, type `D`.

Reactions 1

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

2 Select Domain 2 only.

3 In the **Settings** window for **Reactions**, locate the **Reaction Rates** section.

4 In the R_c text field, type `-phi`.

Concentration 1

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

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

3 Select the **Species c** check box.

4 In the $c_{0,c}$ text field, type `c_in`.

5 Select Boundary 1 only.

Outflow 1

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

2 Select Boundary 12 only.

This example requires a fine mesh, both to solve the physics problem and to resolve the topology optimization problem.

MULTIPHYSICS

Couple the interfaces with the **Reacting Flow** multiphysics node.

Reacting Flow, Diluted Species 1 (rfd1)

In the **Physics** toolbar, click **Multiphysics Couplings** and choose **Domain>Reacting Flow, Diluted Species**.

MESH 1

1 In the **Model Builder** window, under **Component 1 (comp1)** click **Mesh 1**.

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

3 From the **Sequence type** list, choose **User-controlled mesh**.

Size

1 In the **Model Builder** window, under **Component 1 (comp1)>Mesh 1** click **Size**.

2 In the **Settings** window for **Size**, locate the **Element Size** section.

3 From the **Calibrate for** list, choose **General physics**.

4 From the **Predefined** list, choose **Extremely fine**.

Corner Refinement 1, Size 1

1 In the **Model Builder** window, under **Component 1 (comp1)>Mesh 1**, Ctrl-click to select **Size 1** and **Corner Refinement 1**.

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

Boundary Layers 1

1 In the **Model Builder** window, right-click **Boundary Layers 1** and choose **Disable**.

2 In the **Settings** window for **Boundary Layers**, click **Build All**.

STUDY 1

Although you can choose to solve the optimization problem directly, it can be useful to check that the solution for the PDE problem looks sound before starting the optimization.

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

RESULTS

Velocity (spf)

The first default plot (see Figure 3) shows the velocity field in the reactor.

Now solve the optimization problem.

STUDY 1

Optimization

1 In the **Study** toolbar, click **Optimization**.

Choose the `SNOPT` solver rather than `MMA` since the objective function contains an implicit tradeoff between a high flow rate and a high active catalyst area. This makes the objective function severely nonlinear.

2 In the **Settings** window for **Optimization**, locate the **Optimization Solver** section.

3 From the **Method** list, choose **SNOPT**.

4 In the **Optimality tolerance** text field, type `1.0E-6`.

5 Click **Add Expression** in the upper-right corner of the **Objective Function** section. From the menu, choose **Component 1 (comp1)>Definitions>Variables>comp1.obj - Objective function - mol/(m³·s)**.

6 Locate the **Output While Solving** section. Select the **Plot** check box.

This setting gives a plot of the evolving velocity distribution in the **Graphics** window.

Solution 1 (sol1)

1 In the **Study** toolbar, click **Show Default Solver**.

2 In the **Model Builder** window, expand the **Solution 1 (sol1)** node.

3 In the **Model Builder** window, expand the **Study 1>Solver Configurations>Solution 1 (sol1)>Optimization Solver 1** node, then click **Stationary 1**.

4 In the **Settings** window for **Stationary**, locate the **General** section.

5 In the **Relative tolerance** text field, type `1e-6`.

6 In the **Study** toolbar, click **Compute**.

RESULTS

Velocity (spf)

The velocity field in the reactor after optimization should resemble that in Figure 6.

Concentration (tds)

The third default plot shows the concentration distribution in the reactor after optimization (Figure 5).

Streamline 1

1 In the **Model Builder** window, expand the **Concentration (tds)** node, then click **Streamline 1**.

2 In the **Settings** window for **Streamline**, locate the **Streamline Positioning** section.

3 From the **Positioning** list, choose **On selected boundaries**.

4 In the **Number** text field, type `5`.

5 Locate the **Selection** section. Select the **Activate Selection** toggle button.

6 Select Boundary 1 only.

7 In the **Concentration (tds)** toolbar, click **Plot**.

To reproduce the plot in Figure 4, modify the default plot with the following steps.

Distribution of porous catalyst

1 In the **Model Builder** window, expand the **Results>Topology Optimization** node, then click **Output material volume factor**.

2 In the **Settings** window for **2D Plot Group**, type `Distribution of porous catalyst` 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 `Distribution of porous catalyst`.

Surface 1

1 In the **Model Builder** window, expand the **Distribution of porous catalyst** node, then click **Surface 1**.

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

3 From the **Color table** list, choose **GrayScale**.

4 Clear the **Color legend** check box.

5 In the **Distribution of porous catalyst** toolbar, click **Plot**.

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

To display the result for the conversion rate, continue as follows.

Global Evaluation 1

1 In the **Results** toolbar, click **Global Evaluation**.

2 In the **Settings** window for **Global Evaluation**, click **Add Expression** in the upper-right corner of the **Expressions** section. From the menu, choose **Solver>Objective functions> opt.obj1 - Objective function - mol/(m³·s)**.

3 Click **Evaluate**.

TABLE

1 Go to the **Table** window.

The value appears in the **Table** window below the **Graphics** window.