

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.

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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/(m³·s)), over the domain, Ω . This is equivalent to minimizing the negative of this average reaction rate:

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

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/m³), 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 **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:

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

$$\nabla \cdot \mathbf{u} = 0$$
(2)

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

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

$$\min_{\varepsilon} \left\{ -\frac{1}{\operatorname{vol}(\Omega)} \int_{\Omega} (k_a(1-\varepsilon)c) d\Omega \right\}$$
(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:

 $tx + (1-t)y \in C$ 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

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

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

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: $q(1-\epsilon)/(q-\epsilon)$ 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.

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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}_{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

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

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

- I 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 \bigcirc Study.
- 7 In the Select Study tree, select General Studies>Stationary.
- 8 Click **M** Done.

ROOT

Load parameters from a text file.

GLOBAL DEFINITIONS

Parameters 1

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

GEOMETRY I

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

- I In the Model Builder window, under Component I (compl) click Geometry I.
- 2 In the Settings window for Geometry, locate the Units section.
- 3 From the Length unit list, choose mm.

Rectangle 1 (r1)

- 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 2*L.
- 4 In the **Height** text field, type L.

Rectangle 2 (r2)

- I In the Geometry toolbar, click 📃 Rectangle.
- 2 In the Settings window for Rectangle, locate the Size and Shape section.
- **3** In the **Width** text field, type 6*L.
- 4 In the **Height** text field, type 3*L.
- 5 Locate the Position section. In the x text field, type 2*L.
- **6** Locate the **Selections of Resulting Entities** section. Select the **Resulting objects selection** check box.

Rectangle 3 (r3)

- 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 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 틤 Build Selected.
- 7 Click the **Zoom Extents** button in the **Graphics** toolbar.

The geometry should now look like that in Figure 1.

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

DEFINITIONS

Integration 1 (intop1)

- I 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)

- I 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.

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.

5 In the Definitions toolbar, click 🕐 Optimization and choose Topology Optimization> Density Model.

TOPOLOGY OPTIMIZATION

Density Model I (dtopol)

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

- I In the Settings window for Density Model, locate the Geometric Entity Selection section.
- 2 From the Selection list, choose Rectangle 2.
- 3 Locate the Filtering section. From the Filter type list, choose None.
- 4 Locate the Interpolation section. From the Interpolation type list, choose Darcy.
- **5** In the q_{Darcy} text field, type q.
- 6 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.

DEFINITIONS

Variables I

I In the **Definitions** toolbar, click a =**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 |

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

Variables 2

- I In the **Definitions** toolbar, click $\partial =$ **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 |

Objective Function

- I In the Definitions toolbar, click probes and choose Domain Probe.
- 2 In the Settings window for Domain Probe, type Objective Function in the Label text field.
- **3** In the **Variable name** text field, type obj.
- 4 Locate the Source Selection section. From the Selection list, choose Rectangle 2.
- 5 Click Replace Expression in the upper-right corner of the Expression section. From the menu, choose Component I (compl)>Definitions>Variables>phi Local reaction rate mol/(m³·s).

ADD MATERIAL

- 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 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

- I In the Model Builder window, under Component I (compl) 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 **F** vector as

-alpha*u x

-alpha*v y

Inlet 1

- I 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 I

- I In the Physics toolbar, click Boundaries and choose Symmetry.
- 2 Select Boundaries 2, 5, and 9 only.

Outlet I

- I In the Physics toolbar, click Boundaries and choose Outlet.
- **2** Select Boundary 12 only.

TRANSPORT OF DILUTED SPECIES (TDS)

Transport Properties 1

- I In the Model Builder window, under Component I (compl)> Transport of Diluted Species (tds) click Transport Properties I.
- 2 In the Settings window for Transport Properties, locate the Diffusion section.
- **3** In the D_c text field, type D.

Reactions I

- I 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 $R_{\rm c}$ text field, type phi.

Concentration 1

- I 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 I

- I 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 An Multiphysics Couplings and choose Domain>Reacting Flow, Diluted Species.

MESH I

- I In the Model Builder window, under Component I (compl) click Mesh I.
- 2 In the Settings window for Mesh, locate the Sequence Type section.
- 3 From the list, choose User-controlled mesh.

Size

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

Corner Refinement I, Size I

- I In the Model Builder window, under Component I (compl)>Mesh I, Ctrl-click to select Size I and Corner Refinement I.
- 2 Right-click and choose Disable.

Boundary Layers 1

- I In the Model Builder window, right-click Boundary Layers I and choose Disable.
- 2 In the Settings window for Boundary Layers, click 📗 Build All.

STUDY I

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.

I 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 I

Topology Optimization

- I In the Study toolbar, click of Optimization and choose Topology Optimization.
- 2 In the Settings window for Topology Optimization, locate the Optimization Solver section.
- 3 In the Maximum number of iterations text field, type 50.
- 4 Click Add Expression in the upper-right corner of the Objective Function section. From the menu, choose Component I (compl)>Definitions>compl.obj - Objective Function mol/(m³·s).
- 5 Locate the Objective Function section. From the Type list, choose Maximization.
- 6 From the Objective scaling list, choose Manual.
- 7 In the Scale text field, type 0.1.
- 8 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 (soll)

I In the Study toolbar, click **here** Show Default Solver.

The original MMA (1987) implementation has worse final convergence than GCMMA, but it can also be less prone to local minima, which is more important for many topology optimization problems.

- 2 In the Model Builder window, expand the Solution I (soll) node, then click Optimization Solver I.
- 3 In the Settings window for Optimization Solver, locate the Optimization Solver section.
- 4 Clear the Globally Convergent MMA check box.

- **5** Click to expand the **Advanced** section. From the **Compensate for nojac terms** list, choose **Off** to avoid warnings in the log.
- 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

- I In the Model Builder window, expand the Concentration (tds) node, then click Streamline I.
- 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. Click to select the **Delivered 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

- I 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

- I In the Model Builder window, expand the Distribution of porous catalyst node, then click Surface I.
- 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 on 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

- I In the Results toolbar, click (8.5) 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.objl Objective Function mol/(m³·s).
- 3 Click **=** Evaluate.

TABLE

I Go to the Table window.

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

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