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A Computationally Efficient Method for Optimization of Transport-Reaction Processes

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Abstract. This paper proposes a computationally efficient method for the solution of optimization problems arising in the context of transport-reaction processes. The method is applied to a typical diffusion-reaction process and is shown to outperform conventional approaches.

Key Words. Partial differential equations, Karhunen-Loéve expansion, Galerkin's method, successive quadratic programming, diffusion-reaction processes

1. Introduction

Optimization problems arising in the context of transport-reaction processes typically involve maximization/minimization of a nonlinear functional (e.g., integral of a nonlinear function over the domain of definition of the process) subject to a finite set of partial differential equation (PDE) equality constraints and a finite set of spatially dependent inequality constraints. As an example of such an optimization problem, consider the maximization of the integral of the reaction rate of an exothermic catalytic reaction over the surface of the catalyst by choosing properly the temperature of the cooling medium used to remove heat from the catalyst and satisfying mass and energy balances for reactant species and catalyst, respectively, and constraining the catalyst and cooling medium temperatures to be lower that certain values.

The conventional approach to solve optimization problems including PDEs is to directly apply standard spatial discretization techniques (e.g., finite differences, finite elements) to transform the nonlinear functional into a nonlinear function and the PDE constraints into a large set of algebraic constraints. Then, the resulting finite dimensional nonlinear programming problem is solved by using standard methods (Biegler *et al.*, 1997; Bertsekas, 1995; Floudas, 1995; Biegler *et al.*, 1995; Lagnese *et al.*, 1995; Floudas and Panos, 1992; Vasantharajan *et al.*, 1990; Edgar and Himmelbau, 1988). Unfortunately, even though this approach is conceptually straightforward, it may require using a very large number of discretization points/elements in order to compute the optimal solution with the desired accuracy, and thus, it may be computationally expensive. The reason for which this approach may be computationally inefficient is that a brute force discretization with finite differences/elements does not account for the inherent characteristics of the PDE equality constraints.

One approach to solve optimization problems including PDEs, while accounting for the inherent characteristics of the PDEs in the discretization process, is to use Galerkin's method with the eigenfunctions of the linear spatial differential operator as basis functions for the discretization (Yu and Seinfeld, 1973). Even though, this approach may significantly reduce the dimension of the optimization problem which results from the spatial discretization, it cannot be applied to problems that involve nonlinear spatial differential operators (e.g., nonlinear dependence of the diffusion coefficient and thermal conductivity on temperature).

In this work, we propose a computationally efficient method for the solution of optimization problems in transport-reaction processes governed by nonlinear PDEs. We initially construct an ensemble of solutions of the PDE equality constraints for different values of the design variables, which is then used to compute a set of empirical eigenfunctions for the optimization problem by employing Karhunen–Loéve expansion (Park and Lee, 1998; Park and Cho, 1996; Sirovich, 1987*a*; Sirovich, 1987*b*). The empirical eigenfunctions are subsequently used as basis functions in a Galerkin's model reduction procedure employed to transform the original infinite-dimensional optimization problem to a low-dimensional nonlinear program. The resulting nonlinear program is solved by using successive quadratic programming (SQP). The proposed method is used to maximize the integral of the reaction rate over the length of a catalytic rod, where an exothermic reaction takes place and diffusive phenomena are very important, by selecting coolant temperature. Comparisons with conventional approaches show the promise of the proposed

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method.

2. Formulation of the optimization problem

We consider a system of nonlinear PDEs with the following description:

$$0 = L(x) + f(x,d) \tag{1}$$

subject to the boundary condition:

$$Cx + D\frac{dx}{d\eta}\Big|_{\Gamma} = R \qquad \text{on } \Gamma \tag{2}$$

where $x(z)$ denotes the vector of state variables whose dimension is assumed to be n, $z = [z_1, z_2, z_3] \in \Omega \subset \mathbb{R}^3$ is the vector of spatial coordinates, Ω is the domain of definition of the process and Γ its boundary. $L(x)$ is a possibly nonlinear spatial differential operator which includes first-and second-order spatial derivatives, $f(x,d)$ is a nonlinear vector function, d is the vector of design variables which are assumed to be constant, C, D are constant matrices, $\frac{dx}{d\eta}\Big|_{\Gamma}$ denotes the derivative in the direction perpendicular to the boundary and R is a constant vector.

A typical optimization problem for the system of Eqs. 1-2 can be formulated as follows:

$$\begin{aligned} &min \int_{\Omega} G(x,d)dz \\ &s.t. \\ &0 = L(x) + f(x,d) \\ &Cx + D\frac{dx}{d\eta}\Big|_{\Gamma} = R \\ &g(x,d) \le 0 \end{aligned} \tag{P}$$

where $\int_{\Omega} G(x,d)dz$ is the objective function and $g(x,d)$ is the vector of inequality constraints. Both $G(x,d)$ and $g(x,d)$ are assumed to be continuous differentiable.

3. Methodological framework for solution

Owing to the presence of the nonlinear PDE equality constraint of Eq.1, the optimization problem (P) cannot be solved directly, and a numerical spatial discretization scheme should be employed to reduce the PDE system of Eq.1 into a set of algebraic equations. The standard approach to address this problem is to utilize finite differences or finite elements to perform the spatial discretization, and then solve the resulting finite-dimensional nonlinear program using standard techniques. The main disadvantage of this approach is that the number of nonlinear algebraic inequality constraints resulting from the spatial discretization may be very large, thereby leading to a computationally expensive optimization problem. To overcome the above problems, we employ the following methodology for solving (P).

1. Initially, we form an ensemble of solutions of the PDE system of Eq.1 for different values of the design variables which satisfy the inequality constraints.
2. Then, we apply Karhunen-Loéve expansion to this ensemble to derive a set of empirical eigenfunctions (dominant spatial patterns that minimize the mean square error over all the ensemble elements).
3. These empirical eigenfunctions are then used as basis functions within a Galerkin's model reduction framework to transform the original infinite dimensional nonlinear program into a low-dimensional nonlinear program.
4. Finally, the resulting nonlinear program is solved by using SQP.

Due to space limitations, the details of the theoretical development will not be presented here.

4. Illustrative example

We consider a catalytic rod where an exothermic reaction of the form $A \rightarrow B$ takes place. Due to the exothermicity of the reaction, the rod is placed on the top of a cooling system that uses five independent cooling zones to remove heat (see Figure 1 for a schematic of the process).

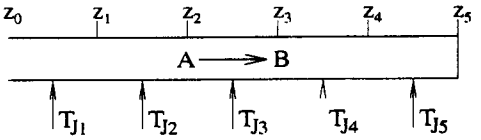

Fig. 1. Catalytic rod with five cooling zones

Under standard modeling assumptions, the steady-state distribution of the concentration of species A, C_A, and temperature of the rod, T, is described by the following two ODEs:

$$\begin{aligned} D\frac{d^2C_A}{dz^2} &= k_1 C_A e^{\left(\frac{-E}{RT}\right)} - H(C_A^{\infty} - C_A) \\ K\frac{d^2T}{dz^2} &= \triangle H_R C_A k_1 e^{\left(\frac{-E}{RT}\right)} - U(T_J - T) \end{aligned} \tag{3}$$

subject to the boundary conditions:

$$\begin{aligned} @z &= 0, l \qquad C_A = C_A^{\infty} \\ @z &= 0, l \qquad T = T^{\infty} \end{aligned} \tag{4}$$

where D, K, l are the diffusivity, conductivity and length of the rod, k_1, E, $\triangle H_R$ are the reaction rate constant, activation energy and enthalpy of reaction, C_A^{∞} is the concentration of A in the bulk, T_J is the temperature of the cooling system, U is the heat transfer coefficient and R is the universal gas constant.

The optimization problem is to maximize the rate of production of species B throughout the rod by choosing the temperature of the five independent cooling zones. The rod temperature and the temperatures of the heating zones at all positions are required to be lower than certain maximum values. Mathematically, this optimization problem is formulated as follows:

$$\begin{aligned} &max \int_0^l k_1 C_A e^{\left(\frac{-E}{RT}\right)} dz \\ &\text{s.t.} \end{aligned}$$

$$D\frac{d^2C_A}{dz^2} = k_1 C_A e^{\left(\frac{-E}{RT}\right)} - h(C_A^{\infty} - C_A)$$

$$K\frac{d^2T}{dz^2} = \triangle H_R k_1 C_A e^{\left(\frac{-E}{RT}\right)} - U(T_J - T)$$

$$T_J = \sum_{j=1}^{5} [H(z_j - z_{j-1}) - H(z_{j+1} - z_j)]\, T_{Jj}$$

$$@z = 0, l \qquad C_A = C_A^{\infty}$$

$$@z = 0, l \qquad T = T^{\infty}$$

$$T_{min} \leq T(z) \leq T_{max} \qquad \forall z$$

$$T_{Jmin} \leq T_{Jj} \leq T_{Jmax} \qquad (j = 1, ..., 5)$$

where $H(\cdot)$ is the standard Heaviside function.

Using the following dimensionless variables,

$$x_1 = \frac{C_A - C_A^{\infty}}{C_A^{\infty}} \quad x_2 = \frac{T - T^{\infty}}{T_{max}} \quad z^* = \frac{z}{l}$$

$$\alpha = \frac{k_1}{h} \quad \beta = \frac{E}{RT_{max}} \quad \lambda_1 = \frac{hl^2}{D} \quad u_j = \frac{T_{Jj}}{T_{max}}$$

$$\gamma = \frac{-\triangle H_R k_1 C_A^{\infty}}{T_{max} U} \quad \lambda_2 = \frac{Ul^2}{K} \quad \nu = \frac{T^{\infty}}{T_{max}}$$

the above optimization problem can be written as:

$$min \left(-\int_0^1 x_1 e^{\left(\frac{-\beta}{x_2 + \nu}\right)} dz^* \right)$$

s.t.

$$\frac{d^2x_1}{dz^{*2}} = [\alpha(x_1 + 1)e^{\left(\frac{-\beta}{x_2 + \nu}\right)} + x_1]\lambda_1 \qquad (PE)$$

$$\frac{d^2x_2}{dz^{*2}} = [-\gamma(x_1 + 1)e^{\left(\frac{-\beta}{x_2 + \nu}\right)} + x_2 + \nu - u]\lambda_2$$

$$u(z^*) = \sum_{j=1}^{5} \left[H(z_j^* - z_{j-1}^*) - H(z_{j+1}^* - z_j^*)\right] u_j$$

$$@z^* = 0, 1 \qquad x_1 = 0$$

$$@z^* = 0, 1 \qquad x_2 = 0$$

$$x_{1min} \leq x_1(z^*) \leq x_{1max} \qquad \forall z^*$$

$$x_{2min} \leq x_2(z^*) \leq x_{2max} \qquad \forall z^*$$

$$u_{min} \leq u_j \leq u_{max} \qquad (j = 1, ..., 5)$$

The values of the dimensionless constants were chosen as: $\alpha = 2.0$, $\beta = 0.3$, $\gamma = 0.5$, $\lambda_1 = 1.0$, $\lambda_2 = 100.0$ and $\nu = 0.5$, while the values of the constraints were chosen as: $x_{1min} = -1.0$, $x_{1max} = 0$, $x_{2min} = -0.5$, $x_{2max} = 0.5$, $u_{min} = 0.5$ and $u_{max} = 1.0$.

Since our objective is to test the efficiency and accuracy of the proposed optimization method, we initially solve the optimization problem (PE) by directly applying a second-order accurate finite difference scheme to reduce it to a finite-dimensional problem, which we then solve using SQP. The quadratic problems in the SQP method were solved by using MINOS. Specifically, we used 206 discretization points to derive a finite-dimensional program which includes 412 state and 5 design variables, 412 equality constraints and 834 inequality constraints. We verified that further increase in the number of discretization points leads to identical results. The solution to this problem required 22 minutes and led to $u_1 = 0.8478$, $u_2 = 0.6718$, $u_3 = 0.6864$, $u_4 = 0.6718$, $u_5 = 0.8478$, and $J_{opt} = 0.6505$ (note that, as expected, owing to the symmetry of the process with respect to the center of the rod $u_1 = u_5$ and $u_2 = u_4$.)

We now use the proposed method to solve (PE). We initially construct an ensemble of solutions (snapshots) of the process model (equality constraints of (PE)) by taking a snapshot for each design variable at its maximum and minimum value; this leads to $2^3 = 8$ snapshots which give us 8 sets of values for x_1 and x_2 (further increase on the number of snapshots led to identical results). We apply Karhunen-Loéve expansion to this set of snapshots to compute 4 empirical eigenfunctions for concentration ($\phi_{1i}(z^*)$) and 4 for temperature ($\phi_{2i}(z^*)$). Both sets of eigenfunctions are orthogonal (Sirovich, 1987*a*). The time required to compute the snapshots and the empirical eigenfunctions was 30 seconds. Expanding $x_1(z^*)$ and $x_2(z^*)$ as linear combinations of the empirical eigenfunctions

$$x_1(z^*) = \sum_{i=1}^{4} a_{1i}\phi_{1i}(z^*), \quad x_2(z^*) = \sum_{i=1}^{4} a_{2i}\phi_{2i}(z^*) \quad (5)$$

where a_{1i} and a_{2i} are constant coefficients, substituting into the optimization problem and taking the inner product of the PDE equality constraints with the empirical eigenfunctions, we obtain:

$$min \left(-\int_0^1 e^{\tau} \sum_{i=1}^{4} a_{1i}\phi_{1i}(z^*) dz^* \right)$$

s.t.

$$0 = \int_0^1 A_1(z^*)\phi_{1i}(z^*)dz^* - a_{1i}\lambda_1 \int_0^1 \phi_{1i}^2(z^*)dz^* - \lambda_1 \alpha \int_0^1 Q(z^*)\phi_{1i}(z^*)dz^* \qquad (i = 1, ..., 4)$$

$$0 = \int_0^1 A_2(z^*)\phi_{2i}(z^*)dz^* - a_{2i}\lambda_2 \int_0^1 \phi_{2i}^2(z^*)dz^* + \lambda_2 \gamma \int_0^1 Q(z^*)\phi_{2i}(z^*)dz^* - 0.5\lambda_2 \int_0^1 \phi_{2i}(z^*)dz^* + \lambda_2 \int_0^1 u(z^*)\phi_{2i}(z^*)dz^* \qquad (i = 1, ..., 4)$$

$$-1.0 \leq \sum_{i=1}^{4} \alpha_{1i}\phi_{1i}(z^*) \leq 0 \qquad \forall z^*$$

$$-0.5 \leq \sum_{i=1}^{4} \alpha_{2i}\phi_{2i}(z^*) \leq 0.5 \qquad \forall z^*$$

$$0.5 \leq u_j \leq 1.0 \qquad (j = 1, ..., 5)$$

where:

$$A_1(z^*) = \sum_{i=1}^{4} (\alpha_{1i} \frac{d^2\phi_{1i}(z^*)}{dz^{*2}})$$

$$A_2(z^*) = \sum_{i=1}^{4} (\alpha_{2i} \frac{d^2\phi_{2i}(z^*)}{dz^{*2}})$$

$$Q(z^*) = (\sum_{i=1}^{4} \alpha_{1i}\phi_{1i}(z^*) + 1)e^{\tau}$$

$$\tau = \frac{-\beta}{\sum_{i=1}^{4} \alpha_{2i}\phi_{2i}(z^*) + 0.5}$$

The above problem includes 8 state and 5 design variables, 8 equality constraints and 834 inequality constraints and was solved by SQP in 15 seconds. The results are $u_1 = 0.8600$, $u_2 = 0.6572$, $u_3 = 6930$. Using these values in the detailed finite difference model, we obtained $J_{emp} = 0.6504$. It is clear that the proposed approach yields an accurate solution to (PE), while requiring a significantly smaller computational time compared to the conventional approach (spatial discretization with finite differences).

For the sake of comparison, we also solved (PE) by using Galerkin's method with *sine* functions as basis functions in the series expansion of $x_1(z^*)$ and $x_2(z^*)$ (note that *sines* constitute a natural choice for the problem in question because they are the eigenfunctions of the linear second-order operator subject to Dirichlet boundary conditions). For N=30, we derived a nonlinear program with 65 variables, 60 equality constraints and 834 inequality constraints. This program was solved by SQP in 3 minutes and 10 seconds and the optimal values are: $u_1 = 0.8525$, $u_2 = 0.6618$, $u_3 = 0.6895$. Substituting these values in the finite difference model we obtained J_{anl}=0.6502.

The optimal rod temperature, concentration of A and reaction rate profiles obtained by using the three different approaches are given in Figures 2, 3, and 4, respectively. In all cases, the maximum temperature temperature constraints are satisfied and the results are almost identical.

From the result of this case study, it is clear that the proposed method provides a computationally efficient approach for solving optimization problems in transport-reaction processes governed by PDEs.

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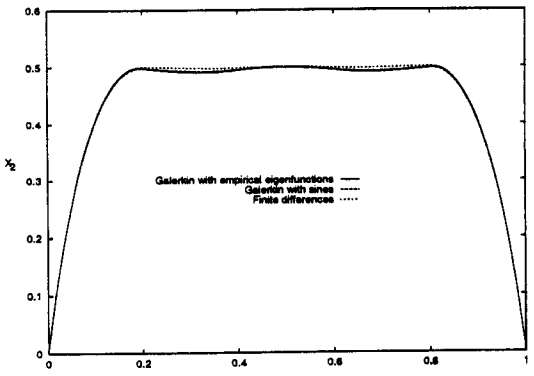

Fig. 2. Optimal temperature profiles

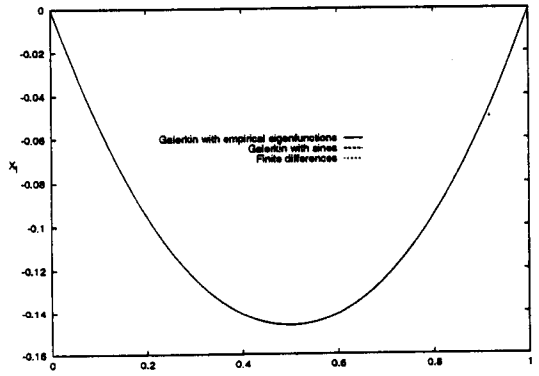

Fig. 3. Optimal concentration profiles

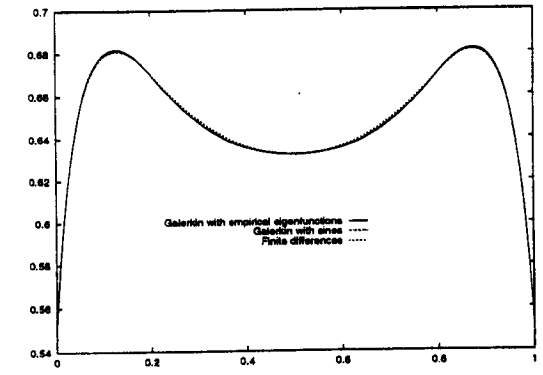

Fig. 4. Optimal reaction rate profiles