Optimal Control Laws for Batch and Semi-batch Reactors Using the Concept of Extents

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In the context of dynamic optimization of batch and semi-batch reactors, this contribution presents a method that uses the concept of extents to generate solutions that satisfy the necessary conditions of optimality given by Pontryagin's maximum principle. The method is divided in two parts. In the first part, the reactor model is written in terms of decoupled extents, and adjoint-free optimal control laws are generated for all possible types of arcs that may occur in the optimal solution. In the second part, the correct sequence of arcs is determined, and, for each sequence, the optimal switching times and initial conditions are computed numerically.

The optimal control problems (OCPs) are formulated in Mayer form, with n_u piecewise-continuous inputs $\mathbf{u}(t)$, n_x states $\mathbf{x}(t)$ described by the differential equations $\dot{\mathbf{x}}(t) = \mathbf{f}(\mathbf{x}(t),\mathbf{u}(t))$, $\mathbf{x}(t_0) = \mathbf{x}_0$, the cost function $\varphi(t_f,\mathbf{x}(t_f))$, the n_t terminal constraints $\boldsymbol{\psi}(t_f,\mathbf{x}(t_f)) \leq \mathbf{0}$, the n_g mixed path constraints $\mathbf{g}(\mathbf{x}(t),\mathbf{u}(t)) \leq \mathbf{0}$ and the n_h first-order pure-state constraints $\mathbf{h}(\mathbf{x}(t)) \leq \mathbf{0}$. This class of OCPs is not restrictive in most dynamic optimization problems dealing with reactors.

The optimal input trajectories are composed of a (typically finite) number of arcs. For each arc and for each input, the optimal input is determined by either an active path constraint or a condition that expresses a physical compromise that depends exclusively on the dynamics of the system [1].

Let the input u_j be one element of $\mathbf{u}$. The goal is to find an expression that relates the optimal input u_j, or one of its time derivatives, to the states, the inputs or the time derivatives of the inputs, thus resulting in an *adjoint-free* optimal control law. For each arc, one of the following cases is possible:

1. The optimal input u_j is determined by the active path constraint $g_k(\mathbf{x},\mathbf{u}) = 0$.
2. The optimal input u_j is determined by the active path constraint $h_k(\mathbf{x}) \leq 0$, and u_j is obtained such that $\partial h_k/\partial \mathbf{x}(\mathbf{x})\ \mathbf{f}(\mathbf{x},\mathbf{u}) = 0$.
3. Otherwise, the optimal input u_j is determined by the condition $\det(\mathcal{M}_{u_j}) = 0$, where

$$\mathcal{M}_{u_j} := [\partial \mathbf{f}^{u_j}/\partial u_j(\mathbf{x},\mathbf{u}) \quad \Delta_j\, \partial \mathbf{f}^{u_j}/\partial u_j(\mathbf{x},\mathbf{u}) \quad \cdots \quad \Delta_j{}^{\rho_j-1}\, \partial \mathbf{f}^{u_j}/\partial u_j(\mathbf{x},\mathbf{u})],$$

and the operators $\Delta_j,\ldots,\Delta_j{}^{\rho_j-1}$ are defined as

$$\Delta_j\, \mathbf{v} := \partial \mathbf{v}/\partial \mathbf{x}\ \mathbf{f}(\mathbf{x},\mathbf{u}) - \partial \mathbf{f}^{u_j}/\partial \mathbf{x}^{u_j}(\mathbf{x},\mathbf{u})\ \mathbf{v} + \textstyle\sum_{n\geq 0} \partial \mathbf{v}/\partial \mathbf{u}^{(n)}\ \mathbf{u}^{(n+1)},$$

$$\Delta_j{}^{l}\, \mathbf{v} := \Delta_j\, (\Delta_j{}^{l-1}\, \mathbf{v}), \quad \text{if } l = 2,\ldots,\rho_j-1,$$

for any vector field $\mathbf{v}$ of dimension ρ_j, with $\mathbf{x}^{u_j}$ being the ρ_j-dimensional vector of states that can be influenced by manipulating u_j, and $\mathbf{f}^{u_j}(\mathbf{x},\mathbf{u})$ such that $\dot{\mathbf{x}}^{u_j} = \mathbf{f}^{u_j}(\mathbf{x},\mathbf{u})$.
However, the input u_j and its time derivatives may not appear explicitly in the function $\det(\mathcal{M}_{u_j})$. Hence, as a general approach to find the optimal input u_j when it is not determined by an active path constraint, the function $\det(\mathcal{M}_{u_j})$ is subject to time differentiation until u_j or one of its time derivatives appears in $\mathrm{d}^{r_j}(\det(\mathcal{M}_{u_j}))/\mathrm{d}t^{r_j}$, for some r_j. Let $u_j^{(\xi_j)}$ be the highest-order time derivative of u_j that appears in $\mathrm{d}^{r_j}(\det(\mathcal{M}_{u_j}))/\mathrm{d}t^{r_j}$. Then, the optimal input $u_j^{(\xi_j)}$ is obtained such that $\mathrm{d}^{r_j}(\det(\mathcal{M}_{u_j}))/\mathrm{d}t^{r_j} = 0$.

The mass and heat balances for batch and semi-batch reactors can be written using the concept of extents [2]. Let us consider a homogeneous batch or semi-batch reactor with R independent reactions and p independent inlets ($p = 0$ for batch reactors), where $\mathbf{u}_{in}(t)$ is the p-dimensional vector of inlet flowrates, and $q_{ex}(t)$ is the exchanged heat power. The numbers of moles $\mathbf{n}(t)$ and the heat $Q(t)$ can be expressed as a linear combination of extents, according to

$$\mathbf{n}(t) = \mathbf{N}^{\mathrm{T}}\,\mathbf{x}_r(t) + \mathbf{W}_{in}\,\mathbf{x}_{in}(t) + \mathbf{n}_0,$$
$$Q(t) = (-\Delta\mathbf{H})^{\mathrm{T}}\,\mathbf{x}_r(t) + \check{\mathbf{T}}_{in}{}^{\mathrm{T}}\,\mathbf{x}_{in}(t) + x_{ex}(t) + Q_0,$$

where $\mathbf{n}_0$ is the S-dimensional vector of initial numbers of moles, Q_0 is the initial heat, $\mathbf{N}$ is the $R\times S$ stoichiometric matrix, $\Delta\mathbf{H}$ is the R-dimensional vector of heats of reaction, $\mathbf{W}_{in}$ is the $S\times p$ inlet-composition matrix, $\check{\mathbf{T}}_{in}$ is the p-dimensional vector of inlet specific enthalpies, $\mathbf{x}_r(t)$ is the R-dimensional vector of extents of reaction, $\mathbf{x}_{in}(t)$ is the p-dimensional vector of extents of inlet, and $x_{ex}(t)$ is the extent of heat exchange.

The state vector of dimension $n_x := R + p + 1$ is

$$\mathbf{x}(t) := [\mathbf{x}_r(t)^{\mathrm{T}} \quad \mathbf{x}_{in}(t)^{\mathrm{T}} \quad x_{ex}(t)]^{\mathrm{T}},$$

while the input vector of dimension $n_u := p + 1$ is

$$\mathbf{u}(t) := [\mathbf{u}_{in}(t)^{\mathrm{T}} \quad q_{ex}(t)]^{\mathrm{T}}.$$

The dynamic equations can be written compactly as

$$\dot{\mathbf{x}}(t) = \mathbf{f}(\mathbf{x}(t),\mathbf{u}(t)), \quad \mathbf{x}(t_0) = \mathbf{0}_{R+p+1},$$

by defining

$$\mathbf{f}(\mathbf{x}(t),\mathbf{u}(t)) := [\mathbf{r}_v(\mathbf{x}(t))^{\mathrm{T}} \quad \mathbf{u}(t)^{\mathrm{T}}]^{\mathrm{T}},$$

where $\mathbf{r}_v(\mathbf{x}(t))$ is the R-dimensional vector of reaction rates. In batch and semi-batch reactors, with $\dot{x}_j = f_j(\mathbf{x},\mathbf{u}) := u_j$, it is possible to define the following vectors of dimension $\rho_j := R+1$:

$$\mathbf{x}^{u_j} := [\mathbf{x}_r{}^{\mathrm{T}} \quad x_j]^{\mathrm{T}},$$
$$\mathbf{f}^{u_j}(\mathbf{x},\mathbf{u}) := [\mathbf{r}_v(\mathbf{x})^{\mathrm{T}} \quad u_j]^{\mathrm{T}}.$$

Note that, since this system is input-affine, $\det(\mathcal{M}_{u_j})$ and its time derivatives are polynomial functions of u_j and its time derivatives, thus resulting in a finite number of solutions, and typically a single solution that satisfies the condition in Case 3.

Let us define the state vector $\check{\mathbf{x}}_j$ as the complement of the state x_j (all states $\mathbf{x}$ except x_j), and the vector $\check{\mathbf{f}}_j(\mathbf{x},\mathbf{u})$ as the corresponding complement of $f_j(\mathbf{x},\mathbf{u})$. Then, one can prove that, when the optimal input u_j is not determined by an active path constraint:

1. For reactors with a single independent reaction, the input u_j is determined by

$$\mathrm{d}(\det(\mathcal{M}_{u_j}))/\mathrm{d}t = \partial(\partial \mathbf{r}_v/\partial x_j(\mathbf{x}))/\partial x_j \; u_j + \partial(\partial \mathbf{r}_v/\partial x_j(\mathbf{x}))/\partial \check{\mathbf{x}}_j \; \check{\mathbf{f}}_j(\mathbf{x},\mathbf{u}),$$

since u_j and its time derivatives do not appear in

$$\det(\mathcal{M}_{u_j}) = \partial \mathbf{r}_v/\partial x_j(\mathbf{x}).$$

2. For reactors with 2 independent reactions, the input u_j is determined by

$$\det(\mathcal{M}_{u_j}) = \det([\partial \mathbf{r}_v/\partial x_j(\mathbf{x}) \quad \partial(\partial \mathbf{r}_v/\partial x_j(\mathbf{x}))/\partial x_j]) \; u_j$$
$$+ \det([\partial \mathbf{r}_v/\partial x_j(\mathbf{x}) \quad -\partial \mathbf{r}_v/\partial \mathbf{x}_r(\mathbf{x}) \; \partial \mathbf{r}_v/\partial x_j(\mathbf{x}) + \partial(\partial \mathbf{r}_v/\partial x_j(\mathbf{x}))/\partial \check{\mathbf{x}}_j \; \check{\mathbf{f}}_j(\mathbf{x},\mathbf{u})])$$

One can use symbolic computation software to evaluate the function $\det(\mathcal{M}_{u_j})$ and its time derivatives and obtain the optimal input u_j or one of its time derivatives when it is not determined by an active path constraint.

The advantage of the proposed approach is that it reduces the set of possible arcs to a finite number of possibilities. This, in turn, results in a finite number of arc sequences if one assumes an upper bound on the number of arcs present in the optimal solution. Hence, instead of solving the original infinite-dimensional problem, one can simply perform numerical optimization for each arc sequence, using the switching times between arcs and the initial conditions as decision variables.

Note that the dynamic state equations can be integrated forward in time, since it is possible to evaluate the corresponding inputs without knowledge of the adjoint variables. Once the forward integration is complete, one can integrate backward in time to obtain the corresponding adjoint variables, which enables the computation of the gradients with respect to the switching times and initial conditions of the arcs.

The proposed approach will be illustrated to maximize the final quantity of product in an acetoacetylation reaction, subject to constraints on the final concentration of reactants and by-products [3].

[1] B. Srinivasan, S. Palanki, and D. Bonvin. Dynamic optimization of batch processes: I. Characterization of the nominal solution, *Comput. Chem. Eng.*, 27:1-26, 2003.
[2] D. Rodrigues, S. Srinivasan, J. Billeter, and D. Bonvin. Variant and invariant states for chemical reaction systems, *Comput. Chem. Eng.*, 73:23-33, 2015.
[3] B. Chachuat, B. Srinivasan, and D. Bonvin. Adaptation strategies for real-time optimization, *Comput. Chem. Eng.*, 33:1557-1567, 2009.