

Solution of Chemical Dynamic Optimization Using the Simultaneous Strategies^{*}

LIU Xinggao (刘兴高)^{**}, CHEN Long (陈珑) and HU Yunqing (胡云卿)

State Key Laboratory of Industry Control Technology, Department of Control Science & Engineering, Zhejiang University, Hangzhou 310027, China

Abstract An approach of simultaneous strategies with two novel techniques is proposed to improve the solution accuracy of chemical dynamic optimization problems. The first technique is to handle constraints on control variables based on the finite-element collocation so as to control the approximation error for discrete optimal problems, where a set of control constraints at element knots are integrated with the procedure for optimization leading to a significant gain in the accuracy of the simultaneous strategies. The second technique is to make the mesh refinement more feasible and reliable by introducing length constraints and guideline in designing appropriate element length boundaries, so that the proposed approach becomes more efficient in adjusting elements to track optimal control profile breakpoints and ensure accurate state and control profiles. Four classic benchmarks of dynamic optimization problems are used as illustrations, and the proposed approach is compared with literature reports. The research results reveal that the proposed approach is preferable in improving the solution accuracy of chemical dynamic optimization problem.

Keywords dynamic optimization, simultaneous strategy, control constraints, mesh refinement, solution accuracy

1 INTRODUCTION

With the growing appreciation for dynamic simulation in computer aided chemical process engineering, accurate chemical dynamic optimization tools have become more and more important [1, 2]. Dynamic optimization has been used in a number of off-line and on-line problems in process industry [3–9], including transitions between specified operating conditions, operating profiles for batch process operation, parameter estimation, model development and discrimination for dynamic systems, design of control systems, and the solution of optimization problems for control and identification, especially non-linear model predictive control (NMPC) [10–13].

A wide variety of approaches have been developed to solve chemical dynamic optimization problems and could be classified into direct methods and indirect methods. Indirect methods, based on Pontryagin's maximum principle, formulate the problem in variational form and bring up the necessary conditions for optimality as a two-point boundary value problem (TPBVP), then use various iterative methods to obtain the solution of the resulting TPBVP [14, 15]. Direct methods are characterized by transforming the original infinite dimensional dynamic optimization problem into a finite dimensional nonlinear programming (NLP). In contrast with indirect methods, direct methods do not require an analytic expression for the necessary conditions, nor initial guesses for the adjoint variables. Instead, the dynamic (state and control) variables are adjusted to optimize the objective function

directly [16–18].

The most widely used direct methods include control parameterization (CP) techniques and simultaneous strategies. In CP techniques, also known as the sequential strategies, only the control variables are discretized over finite elements by polynomials. The coefficients of the polynomial and the mesh size become the decision variables in the resulting NLP problem [19–21]. The gradient of the objective function with respect to the control coefficients and parameters can be calculated either from direct sensitivity equations of the differential algebraic equation (DAE) system or by integration of the adjoint equations [22, 23]. However, the requirement of repeated numerical integration of the DAE model makes the solution procedure time consuming for large scale problems, and dealing with path constraints is another difficulty. Furthermore, sequential approaches cannot handle open loop unstable problems. More information on these approaches can be found in literature [24–26].

Simultaneous strategies fully discretize both control and state variables by piecewise polynomials on time intervals (called finite elements), leading to larger-scale NLP problems, where the coefficients and interval lengths become the decision variables, and special optimization strategies [27, 28] are usually required. Corresponding to a particular implicit Runge-Kutta method, simultaneous strategies directly couple the solution of the DAE system with the optimization problem. The DAE system is solved only once at the optimal point, avoiding excessive computational effort.

The simultaneous strategies have a lot of

Received 2011-07-23, accepted 2012-03-16.

^{*} Supported by the Joint Funds of NSFC-CNPC of China (U1162130), the International Cooperation and Exchange Project of Science and Technology Department of Zhejiang Province (2009C34008), the National High Technology Research and Development Program of China (2006AA05Z226) and the Zhejiang Provincial Natural Science Foundation for Distinguished Young Scientists (R4100133).

^{**} To whom correspondence should be addressed. E-mail: liuxg@iipc.zju.edu.cn

advantages [29–33], and the main shortcoming may be the large dimensionality of the resulting NLP, but this can be solved efficiently using large-scale NLP solvers, such as IPOPT (a primal-dual barrier NLP method) and by exploiting the structure of the collocation equations [33–35]. Recently, more attention has been paid to three basic issues associated with the simultaneous strategies: adjoint estimation, convergence and convergence rates, which are closely related and constitute an interesting and active area of research [36–39].

The purpose of this study is to explore a simultaneous approach with two novel techniques for improving the solution accuracy of chemical dynamic optimization problem. A set of control constraints at finite-element boundary points is proposed to enforce the accuracy requirement. In many cases, lacking of these constraints may lead to insufficiency for getting a precise optimal trajectory. By extrapolating the orthogonal polynomials of the control variables, the control constraints are extended to boundary points of each finite element. Another technique for refining finite elements in simultaneous strategies is proposed. Adjusting and adding finite elements is very important for accurate representation of state profiles and optimal breakpoint locations in control profiles. The first improvement ensures the control profile within the desired range at the collocation points, and the second improvement decreases the degree of freedom in the NLP optimization process, so that the accuracy is increased. The effect of the second improvement is similar in numerical integration: the accuracy increases from Newton-Cotes integration method to Gauss integration, while the degree of freedom is decreased. Tanartkit and Biegler [40] proposed a nested strategy with elements adjusted in outer loop while the control and state variables optimized in inner loop. When successful, the strategy requires repeated nonlinear programming solutions as well as safeguards to deal with non-differentiable functions. In this study, a novel simple and reliable length constraints and a guideline for adjusting the element lengths are proposed to ensure that the discrete problem is an optimal approximation to the original one.

2 FORMULATION OF PROBLEM

We consider a general chemical dynamic optimization problem (P1) with constraints

$$\min_{z(t), y(t), u(t), v} J = \psi[z(t_f), t_f]$$

s.t. DAE model:

$$\dot{z}(t) = f[z(t), y(t), u(t), v] \quad (1)$$

$$c[z(t), y(t), u(t), v] = 0 \quad (2)$$

$$c_f[z(t_f)] = 0 \quad (3)$$

Initial conditions:

$$z(t_0) = z_0 \quad (4)$$

Boundary constraints:

$$\begin{aligned} z^L &\leq z(t) \leq z^U, \quad y^L \leq y(t) \leq y^U, \\ u^L &\leq u(t) \leq u^U, \quad v^L \leq v \leq v^U \end{aligned} \quad (5)$$

The unknowns in this optimization problem are the differential state vector $z(t) \in R^n$, algebraic state vector $y(t) \in R^k$, control vector $u(t) \in R^m$ for $t \in [t_0, t_f]$, as well as time-independent parameter vector v . $z(t)^T = [z^{(1)}(t), z^{(2)}(t), \dots, z^{(n)}(t)]$.

The vector function f is assumed to be continuous differentiable with respect to z and u . Without loss of generality, the chemical dynamic process model is assumed to be a DAE system with index ≤ 1 , because a high-index DAE model can be transformed into an equivalent DAE with index = 1 as using the dummy derivative method described in [41, 42]. Eq. (2) are path constraints, while Eq. (3) are end-point constraints.

3 IMPROVED SIMULTANEOUS APPROACH

Simultaneous strategies fully discretize the DAE system by piecewise polynomial functions over finite elements, $t_0 < t_1 < \dots < t_{NE} = t_f$, $h_i = t_i - t_{i-1}$ (NE is the number of finite elements). The state and control profiles are described by the Lagrange interpolation polynomials in each element:

$$\left. \begin{aligned} t &= t_{i-1} + h_i \tau \\ z(t) &= \sum_{j=0}^K \ell_j(\tau) z_{ij} \end{aligned} \right\} t \in [t_{i-1}, t_i], \quad \tau \in [0, 1] \quad (6)$$

where $\ell_j(\tau) = \prod_{k=0, k \neq j}^K \frac{\tau - \tau_{ik}}{\tau_{ij} - \tau_{ik}}$, K is the number of collocation points, and z_{ij} ($i = 1, \dots, NE$; $j = 1, \dots, K$) represent orthogonal collocation (OC) coefficients produced by Lagrange polynomials and are convenient to provide variable bounds, initialize state profiles, or interpret solution profiles. Alternatively, a monomial basis representation [43] can be used to apply collocation on finite elements, which leads to smaller condition numbers and rounding errors. The control variables and algebraic state variables are approximated over the interval $t \in [t_{i-1}, t_i]$ in the following form

$$u(t) = \sum_{j=1}^K \bar{\ell}_j(\tau) u_{ij} \quad (7)$$

$$y(t) = \sum_{j=1}^K \bar{\ell}_j(\tau) y_{ij} \quad (8)$$

where $\bar{\ell}_j(\tau) = \prod_{k=1, k \neq j}^K \frac{\tau - \tau_{ik}}{\tau_{ij} - \tau_{ik}}$, $t \in [t_{i-1}, t_i]$, $\tau \in [0, 1]$, u_{ij}

and y_{ij} are collocation coefficients.

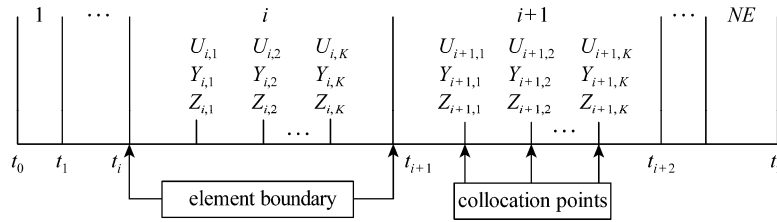


Figure 1 Collocation on finite elements

Figure 1 illustrates the collocation method on finite elements. As most simultaneous strategy studies, discontinuity is only allowed in $u(t)$ and $y(t)$ at element boundaries, although continuity of $z(t)$ is retained, which will be further discussed in the next section.

After discretization, the differential-algebraic equations of problem P1 can be expressed as

$$\sum_{j=0}^K \dot{\ell}_j(\tau_k) z_{ij} - h_i f(z_{ij}, y_{ij}, u_{ij}, v) = 0; \quad i=1, \dots, NE; \quad j=1, \dots, K \quad (9)$$

$$c(z_{ij}, y_{ij}, u_{ij}, v) = 0; \quad i=1, \dots, NE; \quad j=1, \dots, K \quad (10)$$

where the expression $\dot{\ell}_j(\tau) = d\ell_j(\tau)/d\tau$ can be calculated offline since it depends only on the location of Legendre roots, $\tau_j \in (0,1)$ are Gauss points and $\tau_0 = 0$, we have

$$z_{i0} = \sum_{j=0}^K \ell_j(\tau=1) z_{(i-1)j}, \quad i=2, \dots, NE \quad (11)$$

In order to control the approximation error, two novel effective modifications are proposed as follows.

Firstly, the control variables $u(t)$ in Eq. (7) are defined as K th-order polynomials, one order lower than state variables in each element. u_{ij} ($i=1, \dots, NE$; $j=1, \dots, K$) interpret the control profiles and the bounds of control variables are given by

$$u^L \leq u_{ij} \leq u^U, \quad i=1, \dots, NE, \quad j=1, \dots, K$$

Obviously, the constraints of control variables are enforced only at the collocation points, excluding the boundary points. An implicit assumption is that discontinuity of control variables is allowed at boundaries of elements. As a result, the solution is sought with a fixed number of non-smooth points.

To enforce the accuracy requirement, the control constraints at the boundaries of each element are added to the above NLP problem (P1), so as to ensure the constraints not being violated at the finite element points in time:

$$u^L \leq u(t_i) \leq u^U, \quad i=1, \dots, NE \quad (12)$$

$$u^L \leq u(t_{i+1}) \leq u^U, \quad i=1, \dots, NE \quad (13)$$

where $u(t_i)$ and $u(t_{i+1})$ are calculated by extrapolating each piecewise polynomial of control variables to

the end point of the element

$$u(t_i) = \sum_{j=1}^K u_{ij} \cdot \bar{\ell}_j(\tau=0), \quad i=1, \dots, NE \quad (14)$$

$$u(t_{i+1}) = \sum_{j=1}^K u_{ij} \cdot \bar{\ell}_j(\tau=1), \quad i=1, \dots, NE \quad (15)$$

where $\tau=0$ and $\tau=1$ indicate the location of the collocation points (or the initial and end point of each element).

Secondly, another crucial factor for the quality of final optimal solution is the lengths of finite elements. Adjusting and adding finite element is essential for the representation of accurate state profiles and also to optimal breakpoint locations in control profiles [44]. However, the treatment of the element size determination also is one of the serious difficulties in the solution procedure, which is concerned with stability, accuracy and optimality conditions of the solution.

By using simultaneous strategies, both element lengths and control variables can be determined via sequential quadratic programming (SQP) as degrees of freedom in the problem.

An efficient approach of incorporating element placement constraints in the NLP problem (P1) is proposed to provide an effective refine mesh on the finite element lengths:

$$h_i^L \leq h_i \leq h_i^U, \quad i=1, \dots, NE \quad (16)$$

where h_i^L and h_i^U are specially selected depending on the practical optimization problems. The choice of h_i^L is similar in magnitude to the size of the expected original performance criterion if the states are well-behaved, and h_i^U is similar in magnitude to the size of expected end-point time t_f .

The guide for obtaining h_i^L and h_i^U is to solve the problem by first enlarging the feasible area and relaxing the constraints on controls and then using these initial guesses to design desirable parameters according to the optimal performance index.

These embedded boundaries can make the tailoring to elements more adaptive, and therefore provide a number of opportunities for minimizing the approximate error between the discrete optimal solution and the continuous one.

With the proposed control constraints and element placement constraint, problem P1 is transformed

to a nonlinear program problem P2 as follows

$$\begin{aligned}
 & \min_{z_{ij}, y_{ij}, u_{ij}} J = \psi[z_{t_i}, t_f] \\
 & \text{s.t.} \quad \sum_{j=0}^K \ell_j(\tau_{ik}) z_{ij} - h_i f(z_{ij}, y_{ij}, u_{ij}, v) = 0; \\
 & \quad i = 1, \dots, NE, \quad j = 1, \dots, K \\
 & c(z_{ij}, y_{ij}, u_{ij}, v) = 0; \quad i = 1, \dots, NE, \quad j = 1, \dots, K \\
 & \quad c_f[z_{t_i}] = 0 \\
 & z_{(i+1)0} = \sum_{j=0}^K \ell_j(\tau = 1) z_{ij}, \quad i = 1, \dots, NE - 1 \\
 & z_{i,0} = z(t_0) = z_0, \quad z_{t_i} = \sum_{j=0}^K \ell_j(\tau = 1) z_{NEj} \\
 & z^L \leq z_{ij} \leq z^U, \quad y^L \leq y_{ij} \leq y^U, \\
 & u^L \leq u_{ij} \leq u^U, \quad v^L \leq v \leq v^U; \\
 & \quad i = 1, \dots, NE, \quad j = 1, \dots, K \\
 & u^L \leq u(t_i) = \sum_{j=1}^K u_{ij} \cdot \bar{\ell}_j(\tau = 0) \leq u^U; \\
 & \quad i = 1, \dots, NE, \quad j = 1, \dots, K \\
 & u^L \leq u(t_{i+1}) = \sum_{j=1}^K u_{ij} \cdot \bar{\ell}_j(\tau = 0) \leq u^U; \\
 & \quad i = 1, \dots, NE, \quad j = 1, \dots, K \\
 & h_i^L \leq h_i \leq h_i^U; \quad i = 1, \dots, NE
 \end{aligned}$$

Consider the solution of the NLP (P3) resulted from the simultaneous strategies of problem P2:

$$\begin{aligned}
 & \min_x g(x) \\
 & \text{s.t.} \quad F(x) = 0 \\
 & \quad x^L \leq x \leq x^U
 \end{aligned}$$

where $x = [(dz/dt)_{i,j}; z_{ij}; y_{ij}; u_{ij}; u(t_i); u(t_{i+1}); t; v]$.

Considering the large problem size, large number of inequalities and a potentially large number of degrees of freedom, the IPOPT algorithm [45, 46] is employed to solve the above NLP problem. The algorithm follows a barrier approach, where the bound constraints are replaced with logarithmic barrier terms added to the objective function (P4):

$$\begin{aligned}
 \min \varphi(x) &= g(x) - \mu \sum_{i=1}^n \ln[x_i - x_i^L] - \mu \sum_{i=1}^n \ln[x_i^U - x_i] \\
 \text{s.t.} \quad & F(x) = 0
 \end{aligned}$$

where μ is a barrier parameter, $\mu > 0$; and x_i denotes the i th component of vector x . Since the objective function of this barrier problem becomes arbitrarily large as x_i approaches either of its bounds, a local solution

$x^*(\mu)$ of this problem lies in the interior of this bounds, i.e. $x^U > x^*(\mu) > x^L$. The degree of influence of the barrier is determined by the size of μ , and under mild conditions [47], $x^*(\mu)$ converges to a local solution x^* of the original problem (P3) as $\mu \rightarrow 0$.

Consequently, the strategy for solving the original NLP is to solve a sequence of barrier problem (P4) for decreasing barrier parameters μ_l , where l is the counter for the sequence of sub-problems. IPOPT follows a primal-dual approach and applies a Newton method to the resulting Karush-Kuhn-Tucker (KKT) conditions for each sub-problem l . This leads to a solution of the following linear system at each Newton iteration k

$$\begin{bmatrix} H_k + \Sigma_k & A_k^T \\ A_k & 0 \end{bmatrix} \begin{bmatrix} \Delta x \\ \lambda^+ \end{bmatrix} = - \begin{bmatrix} \nabla \varphi(x_k) \\ F(x_k) \end{bmatrix}$$

where H_k is the Hessian matrix of the Lagrangian function $\nabla[g(x_k) + F(x_k)^T \lambda_k]$, $A_k = \nabla F(x_k)$, and $\Sigma_k = V_k^L (x_k - X^L)^{-1} + V_k^U (X^U - x_k)^{-1}$ is the barrier term, with $X = \text{diag}(x)$. The exact first and second derivatives for this method can be evaluated in a number of ways, such as automatically through the AMPL (a modeling language for mathematical programming) interface [48].

4 NUMERICAL EXAMPLES

In order to evaluate the performance of the proposed simultaneous approach, a few benchmark problems are used, which present various challenges such as bang-bang behavior, singular arc, discontinuities in optimal profiles, etc. The problems have been solved by different numerical techniques, so the available data are used for comparison. Although comparing CPU time on different platforms or iterative times for different approaches is unreasonable, the available data of them are also given as open literature reports in this field to provide more information, in addition, their low values of the proposed approach reveal its efficiency.

4.1 Case 1: Catalyst mixing problem

This example was introduced by Gunn and Thomas [49] and considered in a number of studies. It was solved analytically [50] and numerically [21, 40, 51, 52]. The problem involves a plug-flow isothermal reactor with two catalysts, which activate three reactions: $A \longrightarrow B \longrightarrow C$, and the rates of activation are varied for each pair of reaction and catalyst agents. Different catalyst mixing policies will lead to a wide range of product compositions. The goal is to find the optimal profile for catalyst blend along the length of reactor such that the production of component C is

maximized. Detailed model is as follows

$$\begin{aligned} \max_u J &= x_3(t_f) \\ \text{s.t. } \dot{x}_1(t) &= u(t) \cdot [10x_2(t) - x_1(t)] \\ \dot{x}_2(t) &= u(t) \cdot [x_1(t) - 10x_2(t)] - [1 - u(t)] \cdot x_2(t) \\ x_3(t) &= 1 - x_1(t) - x_2(t) \\ x_1(0) &= 1.0, \quad x_2(0) = 0 \\ 0 &\leq u(t) \leq 1.0 \\ 0 &\leq t \leq t_f \end{aligned}$$

where x_1 , x_2 , and x_3 are the mole fractions of substances A, B and C respectively, u is the fraction of catalyst (associated with the reversible reaction), and t_f is the reactor length.

The example poses a severe test for optimization methods because of the index problem. The Hamiltonian function H of the problem is a linear function of u and the partial derivative of H with respect to u has to be differentiated three times before an explicit expression of the derivative of the manipulated variable can be obtained. Therefore, the index of the system along with the singular arc is three.

Logsdon and Biegler [51] showed that because the singular segment is of index 3, it is necessary to apply a higher order collocation method to solve this problem. They converted this problem from index 3 to index 0 by parameterizing the control variables, using variable length piecewise constant sections, and obtained control profiles that matched the analytical solutions. Vassiliadis [21] solved this problem for the reactor length of 1.0 with control parameterization techniques and obtained an analytical optimal value of 0.0480557 after 38.1 CPU seconds as shown in Table 6. Tanartkit and Biegler [40] employed a nested simultaneous strategy and achieved a value of 0.04807 for $t_f = 1.0$, using 90.0–112.5 CPU seconds. Dadebo and McAuley [52] applied iterative dynamic programming (IDP) technique to this problem and reported 0.04805 for $t_f = 1.0$ and 0.476946 (for $t_f = 12$), where 0.476946 is exactly the same as the analytical result demonstrated by Jackson [50]. Using ant colony approach, Rajesh *et al.* [53] reached an optimal value of 0.47615. Banga *et al.* [54] adopted integrated-controlled-random-search method (ICRS) and reached a final value of 0.0479 in 29500 iterations, while Irizarry [55] reached 0.04805 after 1697 iterations using LARES method (a global stochastic optimization approach).

In this study, the problem is solved for reactor lengths 12.0 and 1.0 using the proposed algorithm. For $t_f = 12.0$, we start with the initial guess $u = 0.5$ and set element length constraints by $0.2 \leq h_i \leq 12$. When the number of elements is 2 and the number of collocation points is 15, the analytical optimum value of 0.476946 is achieved with only 17.797 CPU seconds in 400 iterations, shown in Tables 1 and 6. For $t_f = 1.0$, the initialization $u = 0.5$ is retained while the element bounds are changed to be $0.00006 \leq h_i \leq 1$. When the number

Table 1 Computational results for catalyst mixing problem ($t_f = 12.0$)

No. of elements	No. of points	Objective value	No. of iterations
2	3	0.476946	400
4	4	0.476950	400
6	5	0.476941	400
8	6	0.476945	400

Table 2 Computational results for catalyst mixing problem ($t_f = 1.0$)

No. of elements	No. of points	Objective value	No. of iterations
3	3	0.0480523	400
3	4	0.0480580	400
4	5	0.0480557	400
9	6	0.0480542	400

Table 3 Car problem—solution statistics

No. of elements	No. of points	Objective value	No. of iterations
2	2	30.000000	10
2	3	30.000000	16
3	2	30.000000	20
4	2	30.000000	23

Table 4 Computational results for parallel reaction in tubular reactor

No. of elements	No. of points	Objective value	No. of iterations
2	5	0.5738923	47
3	5	0.5738923	47
7	2	0.5738015	400

Table 5 Computational results for unconstrained mathematical system

No. of elements	No. of points	Objective value	No. of iterations
2	3	0.761599	152
2	4	0.761594	400
4	4	0.761594	373
6	2	0.761593	156

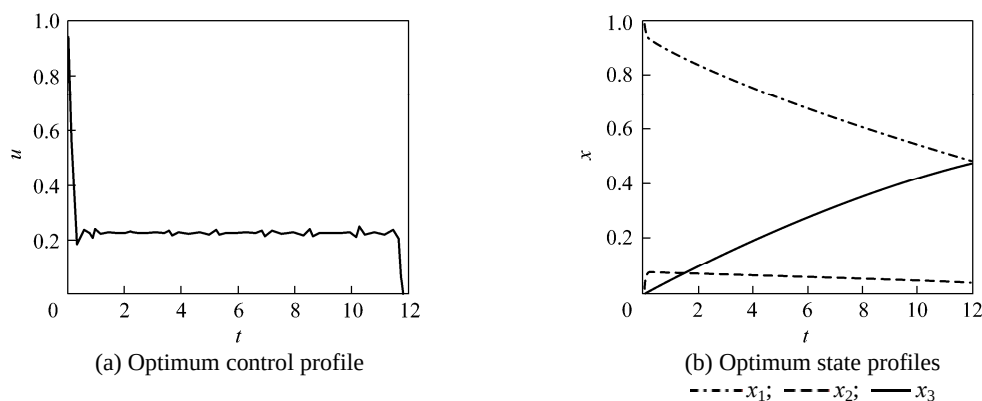
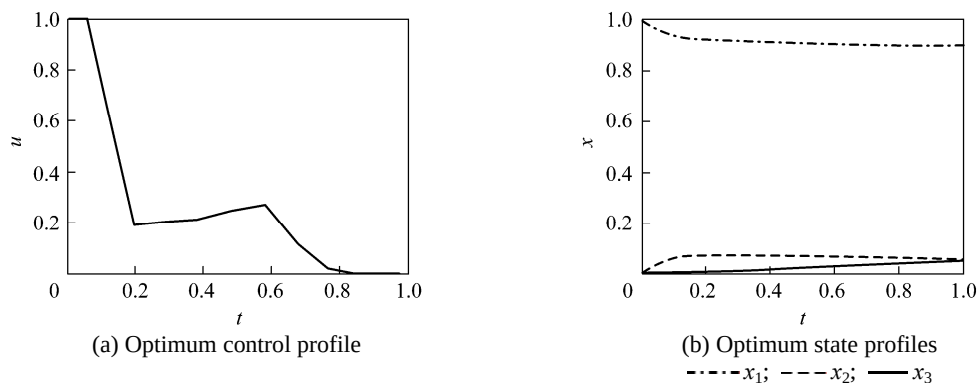
of elements is 4 and the number of collocation points is 16, an optimal solution of 0.0480557 is obtained after a short computational time of 17.906 s in 400 iterations, which is exactly the same as the analytical optimal value as shown in Tables 2 and 6.

In order to examine the robustness of the proposed algorithm, different segmentation and collocation parameters are used. The relevant statistics are given in Tables 1 and 2, in which all optimal objective results are very close to the analytical optimal value 0.0480557 or 0.476946, so that the proposed approach

Table 6 Computational performance comparison of different algorithms for the test problems

Problem		Previous study		Present study
		Method [literature]	Global optimum	Global optimum
catalyst mixing problem	$t_f = 12.0$	ant colony [53]	0.47615	0.476946^①
		iterative dynamic programming [52]	0.476946 ^①	
	$t_f = 1.0$	nested simultaneous strategy [40]	0.04807	0.0480557^①
		ICRS method [54]	0.0479	
		LARES method [55]	0.04805	
		iterative dynamic programming [52]	0.04805	
car problem		control parameterization [21]	0.0480557 ^① , 0.0480553	
		nested simultaneous strategy [40]	30.0	30.000000^①
		control parameterization [21]	30.000000 ^①	
parallel reaction in tubular reactor		control parameterization [21]	0.57353	0.5738923
		iterative dynamic programming [52]	0.57353	
		modified collocation-based NLP [51]	0.57353	
		CVP/CVI [56]	0.56910/ 0.57349	
		nested simultaneous strategy [40]	0.5738 ^②	
unconstrained mathematical system		ant colony [53]	0.76238	0.761594^①
		iterative dynamic programming [57]	0.76159 ^②	

① Analytical results. ② The best result from literature.

**Figure 2** Catalyst mixing problem results ($t_f = 12.0$)**Figure 3** Catalyst mixing problem results ($t_f = 1.0$)

offers not only accurate but also robust numerical results.

Figures 2 and 3 show the optimal control policy and corresponding state profiles for $t_f = 12.0$ and

$t_f = 1.0$, all tendencies of which agree well with the results given in literature [50–53], proving the validity of the proposed approach.

4.2 Case 2: Car problem

As a demonstration of the ability of algorithm to locate the discontinuity, the classical optimal control example, car problem, is considered. The objective is to find an optimal acceleration strategy for a car to travel a predetermined distance in minimum time. The car starts and stops at zero velocity and it is assumed that there is no traveling speed limit. The problem is defined as follows

$$\begin{aligned} \min_{u(\cdot), t_f} J &= t_f \\ \text{s.t.} \quad \dot{y}_1 &= u \\ \dot{y}_2 &= y_1 \\ y(0) &= [0, 0]^T, \quad y(t_f) = [0, 300]^T \\ -2.0 &\leq u(t) \leq 1.0 \\ 0 &\leq t \leq t_f, \quad 0 \leq t_f \leq 50.0 \end{aligned}$$

where y_1 is the velocity, y_2 is the distance, and u is the acceleration. The latter is treated as the control variable. The analytical optimal control profile to this problem is a bang-bang control strategy with a discontinuity at 20 s.

Vassiliadis [21] studied this example by using control parameterization techniques and obtained an optimal objective value of 30.000000, which is exactly the same as the analytical result and is the best result up to now reported in the open literature, with a time expense of 2.5–2.9 CPU seconds as shown in Table 6. Tanartkit and Biegler [40] employed a nested simultaneous strategy and obtained a value of 30.0 using 5.0–5.2 CPU seconds.

In this study, we apply the proposed collocation method to this problem. The initial control profile is assumed a uniform value of 0.5. The element lengths are bounded by $0.1 \leq h_i \leq 20$. As shown in Table 3, we set different numbers of finite elements and collocation points, and obtain the analytical optimum value of 30.000000 in all conditions using CPU time from 0.156 s to 0.671 s, as shown in Tables 3 and 6, without any difficulty, revealing the accuracy and robustness of the proposed approach.

4.3 Case 3: Parallel reaction in tubular reactor

This is an interesting optimal control problem with the saturate control profile originally from Ray [19]. It was studied by several researchers [51, 52, 56, 58], where a tubular reactor makes two products according to the parallel reaction: $A \longrightarrow B$, $A \longrightarrow C$. The objective is to maximize the yield of B at the final time. The dimensionless model describing the system is (with x_1 as dimensionless concentration of A and x_2 as dimensionless concentration of B)

$$\max_u J = x_2(t_f)$$

$$\text{s.t.} \quad \dot{x}_1 = -(u + 0.5u^2) \cdot x_1$$

$$\dot{x}_2 = u \cdot x_1$$

$$x(0) = [1, 0]^T$$

$$0 \leq u \leq 5, \quad t_f = 1$$

Biegler [56] studied this example by using control vector iteration (CVI) and control vector parameterization (CVP), and obtained $J = 0.57349$ and $J = 0.56910$ respectively for the case of a starting profile of $u = 1.0$ as shown in Table 6. He concluded that the optimum with CVP could never reach that with CVI because the functional choice for $u(t)$ is incomplete. Logsdon and Biegler [51] reported a better value of 0.57353 after 53 iterations using a modified collocation-based NLP formulation. Applying the control parameterization techniques, Vassiliadis [21] obtained an objective value of 0.57353, with 93.2 CPU seconds based on SUN SPARC 2. Dadebo and McAuley [52] adopted iteration dynamic programming and achieved the same value. Later, Tanartkit and Biegler [40] obtained a better objective value of 0.5738, which is the best result, up to now, reported in the open literature, using nested simultaneous strategy and requiring 164 CPU seconds.

In this study, the problem is solved by the improved simultaneous algorithm. We start at the initial guess value of $u = 0.5$ and confine element lengths to $1.0 \times 10^{-6} \leq h_i \leq 1.0$. With two finite elements and five collocation points, the maximum of 0.5738923 are obtained using 0.625 CPU seconds based on Intel Celeron 430 (1.7G/512M) as shown in Tables 4 and 6, which is better than the best literature result, 0.5738, reported by Tanartkit and Biegler [40], further revealing the accuracy of the proposed approach.

Table 4 gives the optimal yields of B for different parameters of elements and collocation points. All results are around the optimal value, revealing the robustness and accuracy of the proposed algorithm. Fig. 4 gives the optimal control profile, which has saturation at the end and is consistent with the theoretical analysis. The tendency agrees quite well with the results from previous research [51, 52, 56].

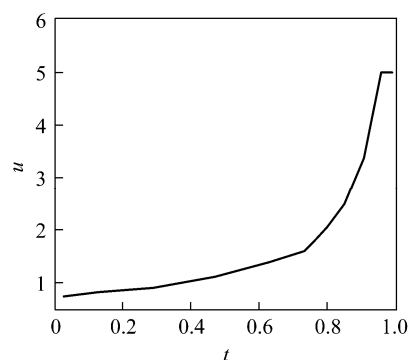


Figure 4 Optimal control profile for parallel reaction in tubular reactor

4.4 Case 4: Unconstrained mathematical system

This case is from Luss [57] and Rajesh *et al* [53]. The detailed mathematical model of the problem is formulated as

$$\min_u J = x_2(t_f)$$

$$\text{s.t. } \dot{x}_1 = u$$

$$\dot{x}_2 = x_1^2 + u^2$$

$$x(0) = [1, 0]^T, \quad t_f = 1$$

The problem has an analytical solution

$$u^* = -\frac{2e}{1+e^2} \sinh(1-t)$$

and a global minimum value

$$J_{\min} = \frac{e^2 - 1}{1 + e^2} \approx 0.761594$$

Luss [57] used iterative dynamic programming technique to solve this problem and obtained an optimal result of 0.76159 as shown in Table 6, which is the best result in literature and very close to analytical value (0.761594). Rajesh *et al.* [53] reported a minimum objective value of 0.76238 by using ant colony algorithm.

The simultaneous approach proposed in this study is applied to this case. Starting with the initialization of $u=0$ and mesh bounds of $0 \leq h_i \leq 1.0$, the global optimum value 0.761594 is achieved, which is exactly the same as the analytical value, and it is the first time to obtain the analytical solution result by numerical method, proving the accuracy of the proposed approach.

In order to examine the robustness of the proposed algorithm, different segmentation and collocation parameters are considered. Table 5 presents the optimal results for different segmentation and collocation parameters, which are very close to the analytical value, revealing the robustness of the proposed approach.

Figure 5 show the optimal control policy. The optimal control profile obtained by our proposed approach

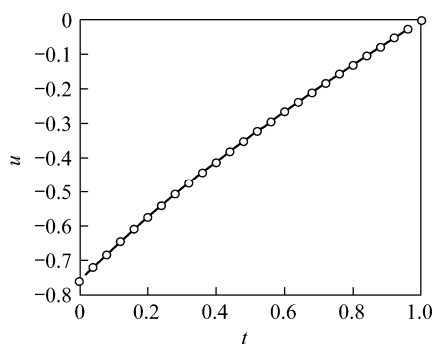


Figure 5 Comparison of control profile for unconstrained mathematical system
— OC; ○ analytical

match the analytical profile exactly. The tendency agrees quite well with those reported by Luss [57] and Rajesh *et al* [53].

5 CONCLUSIONS

In this work, we present an improved simultaneous approach for solving chemical dynamic optimization problems. Control constraints are extended to the finite-element boundary points so that the accuracy of the optimal control profiles is improved. Moreover, an effective mesh refinement technique is proposed to further improve the accuracy of chemical dynamic optimization algorithm. Several classic dynamic optimization examples are carried out as illustrations.

The results with the proposed approach are compared with those in literature. For the cases with analytical result, the proposed numerical approach can achieve the analytical result; for the case with only the best literature result, the better global optimum is achieved by the proposed approach. These reveal the accuracy of the proposed approach.

To examine the robustness of the proposed approach, different segmentation and collocation parameters are explored. The optimal results are very close to the global optimum in all cases, revealing the robustness of the proposed approach.

REFERENCES

- Barton, P.I., Pantelides, C.C., "Modeling of combined discrete/continuous processes", *AIChE J.*, **40** (6), 966–979 (1994).
- MacRosty, R.D.M., Swartz, C.L.E., "Dynamic optimization of electric arc furnace operation", *AIChE J.*, **53** (3), 640–653 (2007).
- Zondervan, E., Roffel, B., "Dynamic optimization of chemical cleaning in dead-end ultra filtration", *J. Memb. Sci.*, **307** (2), 309–313 (2008).
- Parvasi, P., Khosravanipour, M.A., Rahimpour, M.R., "Dynamic modeling and optimization of a novel methanol synthesis loop with hydrogen-permselective membrane reactor", *Int. J. Hydrogen Energy*, **34** (9), 3717–3733 (2009).
- Demiray, T., Andersson, G., "Optimization of numerical integration methods for the simulation of dynamic phasor models in power systems", *Int J. Electr. Power Energy Syst.*, **31** (9), 512–521 (2009).
- Fikar, M., Kovács, Z., Czermak, P., "Dynamic optimization of batch diafiltration processes", *J. Memb. Sci.*, **355** (1–2), 168–174 (2010).
- Egea, J.A., Balsa-Canto, E., García, M.S.G., Banga, J.R., "Dynamic optimization of nonlinear processes with an enhanced scatter search method", *Ind. Eng. Chem. Res.*, **48** (9), 4388–4401 (2009).
- Salau, N.P.G., Tonel, G., Trierweiler, J.O., Secchi, A.R., "Data treatment and analysis for on-line dynamic process optimization", *Comput. Aid. Chem. Eng.*, **25**, 519–524 (2008).
- Panos, C., Kouramas, K.I., Georgiadis, M.C., Pistikopoulos, E.N., "Dynamic optimization and robust explicit model predictive control of hydrogen storage tank", *Comput. Chem. Eng.*, **34** (9), 1341–1347 (2010).
- Robertson, D.G., Lee, J.H., Rawling, J.B., "A moving horizon-based approach for least-squares estimation", *AIChE J.*, **42** (8), 2209–2224 (1996).
- Xiao, J., Li, J., Xu, Q., Huang, Y.L., "ACS-based dynamic optimization for curing of polymeric coating", *AIChE J.*, **52** (4), 1410–1422 (2006).
- Xiao, J., Huang, Y.L., Lou, H.H., "A probability distribution estimation based method for dynamic optimization", *AIChE J.*, **53** (7), 1805–1816 (2007).
- Kadam, J.V., Marquardt, W., Srinivasan, B., Bonvin, D., "Optimal grade transition in industrial polymerization processes via NCO

- tracking", *AIChE J.*, **53** (3), 627–639 (2007).
- 14 Denham, W., Bryson, A., "Optimal programming problems with inequality constraints (II) Solution by steepest ascent", *AIAA J.*, **2** (1), 25–34 (1964).
- 15 Dixon, L.C.W., Bartholomew-Biggs, M., "Adjoint-control transformations for solving practical optimal control problems", *Optim. Contr. App. Met.*, **2** (4), 365–381 (1981).
- 16 Abel, O., Marquardt, W., "Scenario-integrated modeling and optimization of dynamic systems", *AIChE J.*, **46** (4), 803–823 (2000).
- 17 Cruz, P., Mendes, A., Magalhaes, F.D., "Wavelet-based adaptive grid method for the resolution of nonlinear PDEs", *AIChE J.*, **48** (4), 774–785 (2002).
- 18 Hong, W., Wang, S., Li, P., Wozny, G., Biegler, L.T., "A quasi-sequential approach to large-scale dynamic optimization problems", *AIChE J.*, **52** (1), 255–268 (2006).
- 19 Ray, W.H., *Advanced Process Control*, McGraw-Hill, New York (1981).
- 20 Morison, K.R., "Optimal control of processes described by systems of differential-algebraic equations", Ph.D. thesis, Univ. of London, London, UK (1984).
- 21 Vassiliadis, V.S., "Computational solution of dynamic optimization problems with general differential-algebraic constraints", Ph.D. thesis, Univ. of London, London, UK (1993).
- 22 Caracotsios, M., Stewart, W.E., "Sensitivity analysis of initial value problems with mixed ODEs and algebraic equations", *Comput. Chem. Eng.*, **9** (4), 359–365 (1985).
- 23 Ozyurt, D.B., Barton, P.I., "Cheap second order directional derivatives of stiff ODE embedded functions", *SIAM J. Sci. Comput.*, **26** (5), 1725–1743 (2005).
- 24 Cervantes, A.M., Biegler, L.T., "Optimization strategies for dynamic systems", In: *Encyclopedia of Optimization*, Floudas, C., Pardalos, P., eds., Kluwer, USA (2000).
- 25 Tao, H., Liu, Xinggao, "An improved control parameterization method for chemical dynamic optimization problems", In: *Proceedings of the 6th world congress on intelligent control and automation*, IEEE Press, Dalian, China, 1650–1653 (2006).
- 26 Lin, Y.D., Stadtherr, M.A., "Deterministic global optimization of nonlinear dynamic systems", *AIChE J.*, **53** (4), 866–875 (2007).
- 27 Cervantes, A., Biegler, L.T., "Large-scale DAE optimization using simultaneous nonlinear programming formulations", *AIChE J.*, **44** (5), 1038–1050 (1998).
- 28 Cervantes, A.M., Wächter, A., Tütüncü, R., Biegler, L.T., "A reduced space interior point strategy for optimization of differential algebraic systems", *Comput. Chem. Eng.*, **24** (1), 39–51 (2000).
- 29 Cuthrell, J.E., Biegler, L.T., "Simultaneous optimization and solution methods for batch reactor control profiles", *Comput. Chem. Eng.*, **13** (11), 49–62 (1989).
- 30 Hager, W.W., "Runge-Kutta methods in optimal control and the transformed adjoint system", *Numerische Mathematik*, **87** (2), 247–282 (2000).
- 31 Bock, H.G., "Numerical treatment of inverse problem in differential and integral equations", In: *Recent Advances in Parameter Identification Techniques for ODE*, Deuffhard, P., Hairer, E., eds., Pergamon Press, Germany (1983).
- 32 Kameswaran, S., Biegler, L.T., "Simultaneous dynamic optimization strategies: Recent advances and challenges", *Comput. Chem. Eng.*, **30** (10–12), 1560–1575 (2006).
- 33 Biegler, L.T., Cervantes, A.M., Wächter, A., "Advances in simultaneous strategies for dynamic process optimization", *Chem. Eng. Sci.*, **57** (4), 575–593 (2002).
- 34 Barz, T., Kuntsche, S., Wozny, G., Arellano-Garcia, H., "An efficient sparse approach to sensitivity generation for large-scale dynamic optimization", *Comput. Chem. Eng.*, **35** (10), 2053–2065 (2011).
- 35 Bartl, M., Li, P., Biegler, L.T., "Improvement of state profile accuracy in nonlinear dynamic optimization with the quasi-sequential approach", *AIChE J.*, **57** (8), 2185–2197 (2011).
- 36 Schwartz, A.L., Polak, E., "Consistent approximations for optimal control problems based on Runge-Kutta", *SIAM. J. Control. Optim.*, **34** (4), 1235–1269 (1996).
- 37 Pietz, J.A., "Pseudospectral collocation methods for the direct transcription of optimal control problems", Master thesis, Rice Univ., Houston, TX, USA (2003).
- 38 Kameswaran, S., Biegler, L.T., "Convergence rates for direct transcription of optimal control problems using collocation at Radau points", *Comput. Optim. Appl.*, **41** (1), 81–126 (2008).
- 39 Biegler, L.T., "An overview of simultaneous strategies for dynamic optimization", *Chem. Eng. Proc.*, **46** (11), 1043–1053 (2007).
- 40 Tanartkit, P., Biegler, L.T., "A nested simultaneous approach for dynamic optimization problems (II) The outer problem", *Comput. Chem. Eng.*, **21** (12), 1365–1388 (1997).
- 41 Feehery, W.F., "Dynamic optimization with path constraints", Ph.D. thesis, Massachusetts Institute of Technology, USA (1998).
- 42 Mattsson, S. E., Söderlind, G., "Index reduction in differential-algebraic equations using dummy derivatives", *SIAM J. Sci. Comput.*, **14** (3), 677–692 (1993).
- 43 Bader, G., Ascher, U., "A new basis implementation for a mixed order boundary value ODE solver", *SIAM J. Sci. Stat. Comput.*, **8** (4), 483–500 (1987).
- 44 Vasantharajan, S., Biegler, L.T., "Simultaneous strategies for optimization of differential algebraic systems with enforcement of error criteria", *Comput. Chem. Eng.*, **14** (10), 1083–1100 (1990).
- 45 Wächter, A., Biegler, L.T., "On the implementation of a primal-dual interior point filter line search algorithm for large-scale nonlinear programming", *Math. Program.*, **106** (1), 25–57 (2006).
- 46 Rivera-Toledo, M., Flores-Tlacuahuac, A., Vilchis-Ramirez, L., "Dynamic optimization of the methylmethacrylate cell-cast process of plastic sheet production", *AIChE J.*, **55** (6), 1464–1486 (2009).
- 47 Forsgren, A., Gill, P.E., Wright, M.H., "Interior methods for nonlinear optimization", *SIAM Review.*, **44** (4), 525–597 (2002).
- 48 Fourer, R., Gay, D.M., Kernighan, B.W., *A Modeling Language for Mathematical Programming AMPL*, The Scientific Press, South San Francisco, USA (1993).
- 49 Gunn, D.J., Thomas, W.J., "Mass transport and chemical reaction in multifunctional catalyst systems", *Chem. Eng. Sci.*, **20** (2), 89–100 (1965).
- 50 Jackson, R., "Optimal use of mixed catalysts for two successive chemical reactions", *J. Optim. Theory Appl.*, **2** (1), 27–39 (1968).
- 51 Logsdon, J.S., Biegler, L.T., "Accurate solution of differential-algebraic optimization problems", *Ind. Eng. Chem. Res.*, **28** (11), 1628–1639 (1989).
- 52 Dadebo, S.A., McAuley, K.B., "Dynamic optimization of constrained chemical engineering problems using dynamic programming", *Comput. Chem. Eng.*, **19** (5), 513–525 (1995).
- 53 Rajesh, J., Gupta, K., Kusumakar, H.S., Jayaraman, V.K., Kulkarni, B.D., "Dynamic optimization of chemical processes using ant colony framework", *Comput. Chem. Eng.*, **25** (6), 583–595 (2001).
- 54 Banga, J.R., Irizarry-Rivera, R., Seider, W.D., "Stochastic optimization for optimal and model-predictive control", *Comput. Chem. Eng.*, **22** (4–5), 603–612 (1998).
- 55 Irizarry, R., "A generalized framework for solving dynamic optimization problems using the artificial chemical process paradigm: Applications to particulate processes and discrete dynamic systems", *Chem. Eng. Sci.*, **60** (21), 5663–5681 (2005).
- 56 Biegler, L.T., "Solution of dynamic optimization problems by successive quadratic programming and orthogonal collocation", *Comput. Chem. Eng.*, **8** (3–4), 243–248 (1984).
- 57 Luss, R., "Application of iterative dynamic programming to state constrained optimal control problems", *Hung. J. Ind. Chem.*, **19** (4), 245–254 (1991).
- 58 Eaton, J.W., Rawlings, J.B., "Feedback control of chemical processes using on-line optimization techniques", *Comput. Chem. Eng.*, **14** (44), 469–479 (1990).