On an optimization method of chemical reactors

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Abstract. The class of model problems of optimal process control of chemical reactors is considered. A new method of constructing successive approximations to the optimal problem solution is proposed, based on the implementation of the constructed conditions for improving control in the form of fixed-point problems. This form allows to apply and modify the theory and methods of searching for fixed points to optimize control. A comparative analysis of the effectiveness of the proposed optimization method with well-known computational algorithms in the calculation of specific model problems of optimization of chemical reactors is carried out.

1. Introduction

The theory and methods of chemical reactors optimization based on mathematical models are of considerable interest in the field of chemical materials science and have an extensive bibliography [1-3]. The optimization problems under consideration are characterized by various features, such as the structure of the system of equations and constraints, the dimension of the state space, types of nonlinearities, multicriteria etc. At present, there is no universal computational procedure that is sufficiently effective for calculating various optimization problems. Therefore, the development of effective optimization methods focused on taking into account the features of classes of applied problems is relevant.

The theory of optimal control was developed in various schools of thoughts, including the science of our country that made a significant contribution to its formation (A. A. Feldbaum, L. S. Pontryagin, A. A. Milyutin, and their followers). A number of numerical and approximate methods were developed for solving optimal control problems that continue to develop in different directions (R. Gabasov, F. M. Kirillova, V. F. Krotov, V. I. Gurman, F. L. Chernousko, R. P. Fedorenko [4], et al.).

In recent years, the works of V. A. Srochko [5] and A. S. Buldaev [6-8] have actively developed nonlocal methods for improving control as opposed to local methods (for example, gradient methods) do not use a laborious operation varying control at each iteration of methods in a sufficiently small neighborhood of control.

2. Optimization task and method

The class of optimization problems for chemical reactors, reduced to the following general form, is considered:

\[ \dot{x}(t) = f(x(t), u(t), t), \quad x(t_0) = x_0, \] (1)

\[ u(t) \in U, \quad t \in T = [t_0, t_1], \] (2)
\[ \Phi(u) = \phi(x(t_1)) + \int_{t_0}^{t_1} F(x(t), u(t), t) \, dt \to \min_{u \in V} , \]  

(3)

where \( x(t) = (x_1(t), \ldots, x_n(t)) \) is the state vector of the system, \( u(t) = (u_1(t), \ldots, u_m(t)) \) is the vector of controlled variables. As admissible controls, we consider the set \( V \) of piecewise continuous functions on \([t_0, t_1]\) with values in a convex closed set \( U \subset R^m \). The time interval \( T \) and the initial state \( x_0 \) are set.

Using the Lagrange method or the penalty method, problems with terminal, phase, and mixed constraints can be reduced to the form \((1)-(3)\).

In the class of problems under consideration, it is assumed that the function \( \phi(x) \) is continuously differentiable on the set \( R^n \), the vector functions \( F(x, u, t) \), \( f(x, u, t) \) and their derivatives \( F_x(x, u, t) \), \( F_u(x, u, t) \) are continuous in the aggregate of arguments \( (x, u, t) \) on the set \( R^n \times U \times T \).

Consider the Pontryagin function with an adjoint variable \( p \)

\[ H(p,x,u,t) = \langle p, f(x,u,t) \rangle - F(x,u,t) , \]

and in accordance with \([7]\) introduce a modified differential-algebraic conjugate system:

\[ \dot{p}(t) = -H_x(p(t), x(t), w(t), t) - r(t) , \]

(4)

\[ p(t_1) = -\phi_x(x(t_1)) - q , \]

(5)

\[ H(p(t), y(t), w(t), t) - H(p(t), x(t), w(t), t) = \{H_x(p(t), x(t), w(t), t) + r(t), y(t) - x(t)\} , \]

(6)

\[ \phi(y(t_1)) - \phi(x(t_1)) = \{\phi_x(x(t_1)) + q, y(t_1) - x(t_1)\} , \]

(7)

in which, by definition, \( r(t) \equiv 0 \), \( q = 0 \) is assumed in the case of the \( \phi(x) \), \( F(x, u, t) \), \( f(x, u, t) \) functions linear in the variable \( x \) (state-linear problem \((1)-(3))\), as well as in the case of \( x(t) = y(t) \) with the corresponding \( t \in T \).

In the state-linear problem \((1)-(3))\), the modified conjugate system \((4)-(7))\) by definition coincides with the standard conjugate system.

In the state nonlinear problem \((1)-(3))\), the algebraic equations \((6))\) and \((7))\) can always be analytically solved with respect to the values of \( r(t) \) and \( q \) in the form of explicit or conditional formulas (possibly not the only way) \([7]\).

Thus, the differential-algebraic conjugate system \((4)-(7))\) can always be reduced (possibly not in a unique way) to a differential conjugate system with uniquely defined values of \( r(t) \) and \( q \).

To control \( v \in V \), we denote \( x(t, v) \), \( t \in T \) is the solution of system \((1), (2))\) for \( u(t) = v(t) \). For a pair of controls \( u \in V, v \in V \), we denote \( p(t, u, v) \), \( t \in T \) is the solution of the conjugate system \((4)-(7))\) for \( w(t) = u(t) \), \( x(t) = x(t, u) \), \( y(t) = x(t, v) \).

Consider the problem of improving control in the following form: for a given control \( u \in V \), it is required to find a control \( v \in V \) with the condition of \( \Phi(v) \leq \Phi(u) \).

Let us denote the \( R_U \) is the operator of projection onto the set \( U \) in the Euclidean norm:
\[ P_U(z) = \arg \min_{w \in U} \left( \| w - z \| \right). \]

In accordance with [7], the projection conditions for improving the control \( u \in V \) with the projection parameter \( \alpha > 0 \) in problem (1), (2) take the form:

\[
v(t) = P_U \left( u(t) + \alpha \left( H_u \left( p(t,u,v),x(t,v),u(t),t \right) + s(t) \right) \right), \quad t \in T. \tag{8}
\]

\[
H(p(t,u,v),x(t,v),v(t),t) - H(p(t,u,v),x(t,v),u(t),t) = H_u \left( p(t,u,v),x(t,v),u(t),t \right)
+ s(t), v(t) - u(t)), \tag{9}
\]

where in equation (9), by definition, \( s(t) = 0 \) is assumed in the case of the \( F(x,u,t) \), \( f(x,u,t) \) functions linear in the variable \( u \) (linear control problem (1)-(3)), as well as in the case of \( u(t) = v(t) \) with the corresponding \( t \in T \).

For the linear control problem (1)-(3), system (8), (9) by definition reduces to equation (8) with \( s(t) = 0 \), \( t \in T \).

For a non-linear control problem, equation (9) can always be analytically solved respectively with the value of \( s(t) \) (perhaps not the only way).

Thus, system (8), (9) can always be reduced to an equation of the form (8) with a uniquely determined quantity \( s(t) \).

According to [7], the solution of system (8), (9) provides improved control \( u \in V \) for any parameter \( \alpha > 0 \) with an estimate of the improvement:

\[
\Phi(v) - \Phi(u) \leq \frac{1}{\alpha T} \int_0^T \| v(t) - u(t) \|^2 dt.
\]

Moreover, the improvement of control is guaranteed not only in a sufficiently small neighborhood of control \( u \in V \). Thus the improvement procedure based on the solution of system (8), (9) has the property of nonlocality, in contrast to gradient and other local methods for improving control.

Conditions (8), (9) for a given method of unambiguous resolving equations (6), (7) and (9), respectively, quantities \( r(t) \), \( q \) and \( s(t) \) can be considered as a fixed point problem for the control operator uniquely determined by the right-hand side of equation (8).

To solve fixed point problems, one can use well-known simple iteration methods and their modifications [9]. As applied to the problem (8), (9), a simple iteration algorithm for \( k \geq 0 \) has the form:

\[
v^{k+1}(t) = P_U \left( u(t) + \alpha \left( H_u \left( p(t,u,v^k),x(t,v^k),u(t),t \right) + s(t) \right) \right), \tag{10}
\]

\[
H(p(t,u,v^k),x(t,v^k),v^k(t),t) - H(p(t,u,v^k),x(t,v^k),u(t),t) = H_u \left( p(t,u,v^k),x(t,v^k),u(t),t \right)
+ s(t), v^k(t) - u(t)), \quad t \in T. \tag{11}
\]

For \( k = 0 \) the initial approximation \( v^0 \in V \) is specified for the iterative process, which, in the practice of calculations, is usually chosen to control \( u \in V \).
Iterating for the index $k \geq 0$ is conducted until the first strictly improvement in control $u \in V$. Next, a new fixed-point problem is constructed to improve the obtained projection control and the iterative process is repeated.

If a strict control improvement does not occur, then the iterative process is carried out until the condition:

$$|\Phi(v^{k+1}) - \Phi(v^k)| \leq |\Phi(v^k)| \cdot \epsilon,$$

where $\epsilon > 0$ is the specified accuracy of the calculation. At this point, the construction and calculation of successive problems for improving control ends.

3. Examples

A comparative analysis of the effectiveness of the proposed optimization method is carried out on specific model problems of reactor optimization known in the literature. The computational implementation of the optimization method has the following features.

The numerical solution of phase and conjugate Cauchy problems is carried out by the fifth or sixth order accuracy Runge–Kutta–Werner method using IMSL library of the Fortran PowerStation 4.0 language [10]. Values of controlled, phase and conjugate variables are stored in nodes of a fixed uniform grid $hT$ on the interval $T$ with a given sampling step $h > 0$. In the intervals between adjacent nodes of the grid $hT$, the value of the control function was assumed to be constant and equal to the value in the left node.

3.1. Example 1

The problem of a chemical reactor optimization is considered [4,11]:

$$\begin{align*}
\dot{x}_1(t) &= -(k_1(u) + k_2(u) + k_3(u))x_1, \quad x_1(0) = 1, \\
\dot{x}_2(t) &= k_1(u)x_1 - k_4(u)x_2, \quad x_2(0) = 0, \\
\dot{x}_3(t) &= k_4(u)x_2 - k_5(u)x_3, \quad x_3(0) = 0.
\end{align*}$$

$u(t) \in U = [0, 0.823], \quad t \in T = [0, 1]; \quad \Phi(u) = -x_3(1) \rightarrow \text{min}.$

The system of differential equations describes the reactions that occur in a mixture of three substances, $x_i(t), i = 1,2,3$ is their concentration. The first substance is raw materials, the second is an intermediate product, and the third is the final product. The temperature, on which the reaction intensities depend, is considered as a control $u(t)$.

The functions $k_i(u)$ have the characteristic form of chemical kinetics

$$k_i(u) = C_i \exp \left[ \frac{E_i}{R} \left( \frac{1}{658} - \frac{1}{u} \right) \right], \quad i = 1,\ldots,5.$$

The values of the constants are taken from the book [12]: $C_1 = 1.02, \quad C_2 = 0.93, \quad C_3 = 0.386, \quad C_4 = 3.28, \quad C_5 = 0.084, \quad R = 1.9865, \quad E_1 = 16000, \quad E_2 = 14000, \quad E_3 = 15000, \quad E_4 = 10000, \quad E_5 = 15000.$

In [4], this problem was solved using the gradient projection method (MPG), in [11] it was done with the conditional gradient method (MCG) and the conditional quasigradient method of the first order (MCG-1).

To solve the problem with the proposed optimization method, we determine the necessary structures.
The Pontryagin function has the form:
\[ H(p,x,u,t) = -p_1[k_1(u) + k_2(u) + k_3(u)]x_1 + p_2[k_1(u)x_1 - k_4(u)x_2] + p_3[k_4(u)x_2 - k_5(u)x_3] . \]

Since the problem is linear in the state, the differential-algebraic conjugate system coincides with the standard conjugate system and has the form:
\[ \dot{p}_1(t) = p_1[k_1(u) + k_2(u) + k_3(u)] - p_2k_1(u), \quad p_1(1) = 0, \quad \dot{p}_2(t) = p_2k_4(u) - p_3k_4(u), \quad p_2(1) = 0, \]
\[ \dot{p}_3(t) = p_3k_5(u), \quad p_3(1) = 1. \]

To solve the fixed point problem, having the form (8), (9), an iterative process in the form of (10), (11) was used. In this case, the value \( s(t) \) is determined by the following rule:
\[
\begin{aligned}
  s(t) = & \left( H\left(p^k(t) , x^k(t), v^k(t), t\right) - H\left(p^k(t) , x^k(t), u(t), t\right) \right. \\
  & - H_u\left(p^k(t) , x^k(t), u(t), t\right) \cdot v^k(t) - u(t), \\
  0, & \left. v^k(t) = u(t) \right).
\end{aligned}
\]

Here we introduce the notation \( x^k(t) = x(t,v^k) \), \( p^k(t) = p(t,u,v^k) \).

In the initial problem of improving control, \( u(t) \equiv 600 \) was chosen as the initial (starting) control.

Table 1 shows comparative results with the known methods of calculating the problem by the proposed fixed-point method (FPM) for various values of the parameter \( \alpha \). \( \Phi^* \) is the estimated value of the functional. \( N \) is the total number of calculated phase and conjugate Cauchy problems.

| Method      | \( \Phi^* \) | \( N \) |
|-------------|--------------|--------|
| MCG         | -0.43620     | 876    |
| MCG–1       | -0.43682     | 215    |
| MPG         | -0.435       |        |
| FPM \((\alpha = 10^4)\) | -0.43735     | 47     |
| FPM \((\alpha = 10^3)\) | -0.43558     | 155    |

As the parameter decreases from \( \alpha = 10^3 \), the convergence of the iterative process slows down and the total number of Cauchy computational problems increases. As the parameter increases to \( \alpha = 10^5 \), the iterative process of the FPM ceases to converge.

Figures 1 and 2 illustrate the calculated control and the corresponding phase trajectories obtained by the fixed-point method for the parameter \( \alpha = 10^4 \).

3.2. Example 2
The stabilization of a chemical reactor is considered; it is an apparatus with a stirrer and a supply channel for refrigerant [13]:
\[
\Phi(u) = \int_0^{0.78} \left( x_1^2(t) + x_2^2(t) \right) dt \rightarrow \min, \quad \dot{x}_1(t) = -2(x_1 + 0.25) + (x_2 + 0.5) e^{a_1t^2} - (x_1 + 0.25)u,
\]
\[
25x_1 \]
\[ x_1(0) = 0.05, \quad \dot{x}_2(t) = 0.5 - x_2 - (x_2 + 0.5)e^{-x_1^2}, \quad x_2(0) = 0, \quad x_1(0.78) = 0, \]
\[ x_2(0.78) = 0, \quad u(t) \in [-1, 1], \quad t \in [0, 0.78]. \]

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig1}
\caption{Control.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig2}
\caption{Phase trajectories.}
\end{figure}

The functions \( x_1(t) \) and \( x_2(t) \) respectively the deviations of the temperature and concentration of the reacting mixture. The control \( u(t) \) characterizes the flow of refrigerant that regulates the irreversible exothermic reaction.

In [13], the problem reduces to the finite-dimensional optimization problem using discretization by state variables and control and replacing derivatives with finite differences according to the Euler scheme.
We apply the proposed method to solve the continuous problem of optimal control under consideration. We reduce the problem to a problem of the form (1)-(3) without terminal restrictions by introducing the penalty functional \( \Phi^M(u) \) with a sufficiently large penalty parameter \( M > 0 \):

\[
\Phi^M(u) = x(0.78) + M \left( x^2(0.78) + x_2^2(0.78) \right) \to \min,
\]

\[
\dot{x}_1(t) = -2(x_1 + 0.25) + (x_2 + 0.5)e^{x_1+2} - (x_1 + 0.25)u, \quad x_1(0) = 0.05,
\]

\[
\dot{x}_2(t) = 0.5 - x_2 - (x_2 + 0.5)e^{x_1+2}, \quad x_2(0) = 0, \quad \dot{x}_3(t) = x_1^2 + x_2^2, \quad x_3(0) = 0,
\]

\[
u(t) \in [-1, 1], \quad t \in [0, 0.78].
\]

The Pontryagin function for the penalty problem has the form:

\[
H(p, x, u, t) = p_1 \left[ -2(x_1 + 0.25) + (x_2 + 0.5)e^{x_1+2} - (x_1 + 0.25)u \right] + p_2 \left[ 0.5 - x_2 - (x_2 + 0.5)e^{x_1+2} \right] + p_3 \left[ x_1^2 + x_2^2 \right].
\]

The modified conjugated system (4)-(7) by the rules of the unambiguous choice of the quantities \( r(t) \) and \( q \) is reduced to the system:

\[
p_1 = -H_{x_1} - \eta(1), \quad p_1(0.78) = -Mx_1(0.78) - My_1(0.78) , \quad \dot{p}_2 = -H_{x_2} - r_2(t),
\]

\[
p_2(0.78) = -Mx_2(0.78) - My_2(0.78), \quad p_3 = -1,
\]

wherein

- if \( y_1(t) \neq x_1(t) \), then \( \eta(t) = \frac{\Delta_p H}{y_1(t) - x_1(t)} - H_{x_1} - H_{x_2} \frac{y_2(t) - x_2(t)}{y_1(t) - x_1(t)} , r_2(t) = 0; \)
- if \( y_1(t) = x_1(t) \), then
  - if \( y_2(t) \neq x_2(t) \), then \( \eta(t) = 0 , r_2(t) = \frac{\Delta_p H}{y_2(t) - x_2(t)} - H_{x_2}; \)
  - if \( y_2(t) = x_2(t) \), then \( \eta(t) = 0 , r_2(t) = 0. \)

The notation \( \Delta_p H = H(p, y, w, t) - H(p, x, w, t) \) is introduced here.

Due to the linearity of the optimal control problem for control, system (8), (9) reduces to the fixed point problem (8) with \( s(t) \equiv 0 \), to solve which the simple iteration algorithm was used for \( k \geq 0 \):

\[
y^{k+1}(t) = P_1 \left[ u(t) + \alpha H_u \left( p(t, u, v^k), x(t, v^k), u(t), t \right) \right], \quad t \in T.
\]

In the initial problem of improving control, \( u(t) \equiv 1 \) was chosen as the starting control.

The calculation by the proposed method of fixed points (FPM) was carried out for various values of the parameter \( \alpha > 0 \) and the penalty coefficient \( M > 0 \). The convergence of the FPM for various values of \( M > 0 \) depended on the choice of the parameter \( \alpha > 0 \). For example, for \( M = 2 \), the FPM
converged at $\alpha < 0.4$. Moreover, for small $\alpha > 0$ the convergence rate was slow. The best speed, estimated by the number of solved Cauchy problems, was observed for $\alpha \in (0.10; 0.14)$. With an increase in the penalty parameter for convergence, we had to decrease $\alpha$. Moreover, the convergence rate also slowed down. The best-calculated value of the target functional $\Phi^*$ was achieved for the parameters $\alpha = 0.13$, $M = 2$.

Table 2 provides a comparative analysis of the calculation results by the proposed FPM method and the gradient projection method (MPG) used in [13] to calculate a finite-dimensional analog of the problem.

| Method  | $\Phi^*$   | $x_1^* (0.78)$ | $x_2^* (0.78)$ | $N$ |
|---------|------------|----------------|----------------|-----|
| FPM     | 0.00200    | -9.62·10^{-4}  | 1.24·10^{-3}   | 90  |
| MPG     | 0.00220    | -6.16·10^{-6}  | -0.63·10^{-6}  | -   |

Figures 3 and 4 show the calculated control $u(t)$ and the corresponding phase trajectories $x_1(t)$, $x_2(t)$, obtained by the FPM method.

4. Conclusion

The conducted comparative computational experiments on model optimization problems of chemical reactors demonstrate the computational and qualitative efficiency of the proposed fixed point method acceptable for practice in comparison with known methods. The developed method for finding approximately optimal solutions has a fairly wide convergence region in the initial approximation and is characterized by the convenience and simplicity of the experimental setup of a scalar projection parameter that regulates the quality and convergence rate of the iterative process under consideration. In general, the optimization method based on the calculation of the constructed fixed-point problems is reduced to alternating solutions of Cauchy problems for phase and conjugate variables of the problem.

The indicated properties of the proposed fixed-point method are important factors in improving the computational and qualitative efficiency of solving optimization problems of chemical reactors.

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References

[1] Boyarintsev A I and Kafarov V V 1969 Optimization Methods in Chemical Technology (Moscow: Chemistry) p 563
[2] Ostrovsky G M and Berezhinsky T A 1984 Optimization of Chemical-Technological Processes. Theory and Practice (Moscow: Chemistry) p 240
[3] Bochkarev V V 2014 Optimization of Chemical-Technological Processes (Tomsk: Tomsk Polytechnic University Press) p 264
[4] Fedorenko R P 1978 Approximate Solution of Optimal Control Problems (Moscow: Nauka) p 487
[5] Srochko V A 2000 Iterative Methods for Solving Optimal Control Problems (Moscow: Nauka) p 160
[6] Buldaev A S 2008 Perturbation Methods in Problem of the Improvement and Optimization of the Controlled Systems (Ulan-Ude: Buryat State University Press) p 260
[7] Buldaev A S 2017 Fixed-point methods based on the projection operations in problems of optimization of control functions and parameters of dynamical systems Buryat State University Bulletin. Mathematics, Informatics 1 38–54
[8] Buldaev A S and Burlakov I D 2018 About one approach to the numerical solution of nonlinear optimal speed problems Bulletin of the South Ural State University. Series “Mathematical Modelling, Programming and Computer Software” 11(4) 55–66
[9] Samarsky A A and Gulin A V 1989 Numerical Methods (Moscow: Nauka) p 432
[10] Bartenev O V 2001 Fortran for Professionals. Mathematical Library IMSL. Part 2 (Moscow: Dialog-MIFI) p 320
[11] Srochko V A, Antonik V G and Mamonova N V 2007 Computational comparison of gradient type methods in optimal control problems Bulletin of Irkutsk State University. Series “Mathematics” 1 275–90
[12] Rosenbrock H H and Storey C 1966 Computational Techniques for Chemical Engineers (Oxford: Pergamon Press) p 346
[13] Kirk D E 2004 Optimal Control Theory. An Introduction (New York: Dover Publications) p 452