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Evolutionary Algorithm for Solving the Optimal Speed Problem for Catalytic Processes

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Abstract

The paper presents a numerical algorithm for calculating the control that converts a catalytic process from an initial state to a given final state in minimum time. The formulation of the problem of optimal speed for the chemical process on the basis of its mathematical model is performed. The temperature of the reaction mixture is considered as a control. A numerical algorithm based on the method of differential evolution for solving the problem is given. The advantage of the proposed approach is that in its application there is no need to set the initial approximation of the solution based on the physical and chemical sense of the problem. The work of the algorithm is tested on an industrially significant process of synthesis of phthalic anhydride. The optimum temperature regime and the shortest duration of the process at a given value of phthalic anhydride concentration, at a fixed content of naphthoquinone in the reaction mixture, under the condition of complete consumption of naphthalene have been calculated. The efficiency of application

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of the developed algorithm is demonstrated by comparing the found solutions of rapidity problems with the solutions calculated using other methods.

1 Introduction

In the study of chemical technology processes, it is often necessary to solve optimization problems. One of such problems is the optimal speed problem. In this problem it is required to choose the values of control parameters at each moment of time in order to move the process from a given initial state to a given final state in minimum time. This class of problems includes problems of search for optimal control of periodic chemical-technological processes, problems of the fastest transfer of a process from one operating mode to another, problems of choosing the optimal temperature profile in an ideal displacement reactor, and others.

One of the difficulties arising in solving problems based on the mathematical description of chemical processes is the nonlinearity of their dynamics. Most often, a mathematical model of a chemical process is represented as a system of ordinary differential equations containing an exponential dependence of the reaction rate constant on temperature [1–3]. Nonlinearity of mathematical models of chemical processes complicates their investigation and control and leads to the need for special approaches. Therefore, the development of methods and algorithms for finding the optimal control of catalytic processes in terms of speed is an urgent task.

The Pontryagin maximum principle is widely used to solve nonlinear optimal speed problems [4–6]. Using the maximum principle, the initial problem is reduced to a boundary value problem for a system of ordinary differential equations. The main difficulty in applying this method to solve the problems of optimal control of catalytic processes is the need to determine the initial conditions for the solution of the coupled system taking into account the physical and chemical sense of the problem.

In [7], a method for determining the optimal control in terms of speed for nonlinear controllable systems in which the linear occurrence of phase variables and control can be distinguished is proposed. However, the method is inapplicable for systems with nonlinear control and nonlinear input of state variables.

In most cases, it is sufficient to have an approximate solution of the optimization problem when controlling chemical processes. This is due to the need for prompt response to rapidly changing process conditions. Therefore, in the study of chemical processes it is possible to apply the method of finite-dimensional approximation of the control problem, reducing the original problem of infinite-dimensional optimization to the problem of mathematical programming. When solving finite-dimensional problems, most optimization methods effectively find the global optimum at a well-chosen initial approximation, and are applied only for smooth and convex problems (for example, gradient methods [8]). A nonlinear fast problem may have a non-convex reachability region and, as a consequence, local extrema. This creates additional difficulties in developing a search procedure for determining the global extremum.

Therefore, evolutionary computational methods can be applied to solve the optimal speed problem for catalytic processes. Evolutionary methods have successfully proved themselves in solving practical problems when traditional methods are ineffective [9–13]. One such method is the differential evolution method [14–16]. This method is direct and does not depend on the initial approximation of the problem solution. The lack of sensitivity of the differential evolution method to the initial approximation is achieved due to the fact that at each iteration the set of possible solutions is simultaneously optimized. This allows increasing the search area. The method of differential evolution has a smaller number of adjustable parameters, unlike other evolutionary search methods, such as genetic algorithms, the method of artificial immune systems and others. The aim of this work is to develop a numerical algorithm for solving the optimal speed problem for a catalytic process based on the differential evolution method.

2 Materials and methods

Let the mathematical description of the catalytic process be a system of ordinary differential equations

$$\frac{dx}{dt} = f(x(t), u(t), t) \tag{1}$$

with initial conditions

$$x(0) = x^0, \tag{2}$$

where $x(t) = (x_1(t), x_2(t), \ldots, x_n(t))^T$ – vector of substance concentrations (phase variables), $u(t)$ – control parameter, $t \in [0, T]$ – time, $f(x(t), u(t), t)$ $= (f_1(x(t), u(t), t), f_2(x(t), u(t), t), \ldots, f_n(x(t), u(t), t))^T$ – vector function continuous together with its partial derivatives.

Let the values of concentrations of substances at the final moment of time $t = T$ be given:

$$x(T) = x^1. \tag{3}$$

The control parameter $u(t)$ can be the temperature of the reaction mixture, pressure, feed rate of reagents, etc. Let us consider as a control $u(t)$ the reaction temperature, the values of which are limited:

$$u_{\min} \le u(t) \le u_{\max}, \quad t \in [0, T]. \tag{4}$$

We will look for the reaction temperature $u(t)$ in the class of piecewise constant functions. Let us divide the segment $[0, T]$ by points $t_1, t_2, \ldots, t_s$, so that $t_0 < t_1 < \ldots < t_{s+1}$, $t_0 = 0$, $t_{s+1} = T$. At each of the segments $t \in [t_j, t_{j+1}]$ temperature takes a constant value $u(t) = u_j$.

It is required to determine the temperature regime $u(t)$ ($t \in [0, T]$), which transfers the catalytic process from the initial state (2) to the final state (3) for the minimum time T^*. That is, it is necessary to minimize the target functional:

$$G(u) = T \to \min. \tag{5}$$

Let us formulate an algorithm for finding a solution to problem (1)–(5) based on the method of differential evolution. The operation of the method is based on the simulation of evolutionary processes to which vectors-indivisibles are subjected [17, 18]. The target functional (5) is the fitness function that determines the suitability of the individuated vector as a solution to the problem. Usually, the condition of computation termination is the achievement of the maximum number of iterations specified by the user.

Let us introduce individuated vectors p_i $(i = \overline{1, m})$, which form a population and are potential solutions to the speed problem. As components of each vector-indivisible, we set the control $u_j = u(t_j)$, $t_j \in [0, T]$, $j = \overline{0, s}$, $t_0 = 0$, $t_{s+1} = T$, and the duration of the chemical reaction:

$$p_{ij} = \begin{cases} u_{ij}, & j = \overline{0, s}, \\ T, & j = s + 1, \end{cases} \tag{6}$$

where $i = \overline{1, m}$.

Let the fitness function be not the objective functional (5), but a function of the form

$$Q(p_k) = \sqrt{\sum_{i=1}^{n} (x_i^k - x_i^1)^2}, \tag{7}$$

where $x_1^k, x_2^k, \ldots, x_n^k$ are the values of substance concentrations calculated by solving the system of differential equations (1) with initial conditions (2) for the vector-individual p_k at the final time $t = p_{k\ s+1}$ and piecewise constant control with nodes $(p_{k0}, p_{k1}, \ldots, p_{ks})$. The smaller the value of $Q(p_k)$, is, the more accurately the terminal conditions (3) are satisfied, so the vector-individual p_k is more adaptable and more suitable as a solution to the speed problem than others.

Let us set as a stopping criterion the fulfillment of the inequality for some vector-individual p_k of the current population:

$$Q(p_k) < \varepsilon, \tag{8}$$

where ε – user defined parameter.

Condition (8) can be fulfilled at several possible temperature regimes. Therefore, we will enter possible solutions to the speed problem into a separate array of individual vectors *solutions*. At the end of the algorithm, from the *solutions* array we select the vector p_k, whose last component $p_{k\ s+1}$ corresponding to the duration of the chemical reaction, takes the smallest value. The selected vector-individual will be an approximate solution to the optimal speed problem (1)–(5).

The differential evolution algorithm for solving problem (1)–(5) consists of the following steps.

Step 1. Set the parameters of the algorithm: m – number of vectors-individuals in the population, $\overline{T}$ – maximum duration of the chemical reaction, s – number of points of division of the segment $[0, \overline{T}]$, $kross \in [0,1]$ – crossover parameter, $mut \in [0,5;1]$ – mutation parameter [14], ε – parameter for ending the calculations.

Step 2. Generate a population of individual vectors $p_i = (p_{i0}, \dots, p_{i\ s+1})$, where

$$p_{ij} = \begin{cases} u_{\min} + \gamma_{ij}(u_{\max} - u_{\min}), & j = \overline{0, s}, \\ \gamma_{ij}\overline{T}, & j = s+1. \end{cases} \tag{9}$$

Here $\gamma_{ij} \in [0,1]$ is a random number, $i = \overline{1, m}$.

Step 3. Calculate the fitness of each individual vector p_i ($i = \overline{1, m}$). To do this, find a numerical solution to the system of differential equations (1) with a time interval division step $[0, p_{i\ s+1}]$, equal to $p_{i\ s+1}/s$.

Step 4. Check condition (8) for each individual vector p_i ($i = \overline{1, m}$). If it is satisfied, then place vector p_i into array *solutions*.

Step 5. Set the first individual vector as the target vector: $p_{mishen} = p_1$.

Step 6. Find the most adapted vector-individual in the population p_{prisp}.

Step 7. Perform the mutation operation. Randomly select two different individual vectors p_a, p_b, where $p_a \neq p_{mishen}$, $p_b \neq p_{mishen}$, $p_a \neq p_{prisp}$, $p_b \neq p_{prisp}$. Create a mutant vector p_{mutant}:

$$p_{mutant} = p_{prisp} + mut(p_a - p_b).$$

Step 8. Perform the crossover operation on the mutant vector and the target vector. Create a trial vector p_{prob}:

$$p_{prob\ j} = \begin{cases} p_{mutant\ j}, & \alpha_j \leq kross, \\ p_{mishen\ j}, & \alpha_j > kross, \end{cases}$$

where $\alpha_j \in [0,1]$ – random number, $j = \overline{0, s}$.

Step 9. Calculate the fitness function value for the trial vector $Q(p_{prob})$.

Step 10. Check condition (8) for the trial vector p_{prob}. If it is satisfied, then place the trial vector into the array *solutions*.

Step 11. Update the population. If $Q(p_{prob}) < Q(p_{mishen})$, then place the trial vector into the population instead of the target vector, i.e. $p_{mishen} = p_{prob}$. Otherwise, leave the target vector p_{mishen} in the population.

Step 12. If all individual vectors of the current population are considered as the target vector, i.e. $p_{mishen} = p_m$, then go to step 13. Otherwise, go to the next target vector $p_{mishen} = p_{mishen+1}$ in the population and go to step 6.

Step 13. Check condition (8) for the individual vectors of the current population. If condition (8) is satisfied for the vector p_i $(i = \overline{1, m})$, then place it in the *solutions* array and go to step 14. Otherwise, go to step 5.

Step 14. Find in the array *solutions* the individual vector p_k, whose last component takes the smallest value. Take $T^* = p_{k\ s+1}$ as the shortest duration of a chemical reaction. Assign the values of the elements p_{kj} $(j = \overline{0, s})$ to the values of the temperature $u(t)$ at discrete moments of time according to formula (6).

Thus, the authors have made the following modifications to the classical differential evolution algorithm:

1) the structure of the individual vector includes discrete values of the control parameter and time;

2) the fitness function of the individual vector is not the target functional in the form (5), but the deviation between the given final state of the process and the one calculated using the algorithm;

3) a special array *solutions* is introduced to store the most adapted

vectors-individuals, from which the solution with the smallest time value is selected at the end of the algorithm;

4) the condition for the end of the computational process is a deviation from the terminal state of the controlled process.

3 Research results

Let's apply the developed algorithm to solve the problems of speed on the example of the process of phthalic anhydride production. Phthalic anhydride is used to produce various dyes, medicines, insecticides, additives for lubricating oils, additives for aviation fuel, and others.

Let us introduce the following designations: X_1 – naphthalene, X_2 – naphthoquinone, X_3 – phthalic anhydride, X_4 – carbon dioxide, X_5 – maleic anhydride. Taking into account the introduced designations, the set of chemical transformations of the process of phthalic anhydride production can be described by a sequence of stages [19]:

$$\begin{aligned} &X_1 \to X_2,\\ &X_2 \to X_4,\\ &X_1 \to X_3,\\ &X_1 \to X_4,\\ &X_2 \to X_3,\\ &X_3 \to X_5. \end{aligned} \tag{10}$$

The kinetics of the process can be described by a system of ordinary differential equations

$$\frac{dx_i}{dt} = f(x(t), u(t), t) \tag{11}$$

with initial conditions

$$x_i(0) = x_i^0, \quad i = \overline{1,5}, \tag{12}$$

where $x_i(t)$ – concentration of the i-th substance ($i = \overline{1,5}$) (mole fraction), $u(t)$ – reaction temperature (K), $t \in [0, T]$ – reaction time (h).

According to the law of acting masses, the right parts of the system of differential equations (11) are the functions

$$
\begin{aligned}
f_1(x,u,t) &= -k_1(u)x_1 - k_3(u)x_1 - k_4(u)x_1,\\
f_2(x,u,t) &= k_1(u)x_1 - k_2(u)x_2 - k_5(u)x_2,\\
f_3(x,u,t) &= k_3(u)x_1 + k_5(u)x_2 - k_6(u)x_3,\\
f_4(x,u,t) &= k_2(u)x_2 + k_4(u)x_1,\\
f_5(x,u,t) &= k_6(u)x_3,
\end{aligned}
$$

where $k_j(u)$ – rate constant of the j-th stage ($1/h$, $j = \overline{1,6}$), depending on the temperature u based on the Arrhenius equation

$$k_j(u) = k_{0j} \exp\left(-\frac{E_j}{Ru}\right),$$

where k_{0j} – pre-exponential multiplier (l/h), E_j – activation energy of the j-th stage (J/mol), R – universal gas constant (8,31 J/(mol·K)).

Numerical values of kinetic parameters of the reaction of phthalic anhydride synthesis are given in [19].

Let us consider as a control the reaction temperature $u(t)$, the values of which are subject to constraints:

$$620K \leq u(t) \leq 644K, \quad t \in [0,T]. \tag{13}$$

Let the initial concentrations be given by the values (mole fraction)

$$x_1(0) = 1, \quad x_i(0) = 0, \quad i = \overline{2,5}. \tag{14}$$

The target product of reaction (10) is phthalic anhydride X_3. Let it be required that its concentration at the end of the reaction is 0,77 mole fractions:

$$x_3(T) = 0,77. \tag{15}$$

It is necessary to find the control $u(t)$, taking into account the constraints (13), which transfers the process described by the system (11) from state (14) to state (15) in the minimum time T^*.

To solve the problem (10)–(15), a software package has been developed in the Delphi programming language that implements the algorithm of the differential evolution method. The algorithm is applied with the following parameters: population size $m = 50$, maximum duration of chemical reaction $\overline{T} = 5$ h, number of time interval partition points $s = 50$, crossover parameter $kross = 0,7$, mutation parameter $mut = 0,8$, calculation termination parameter $\varepsilon = 0,01$.

As a result of calculations, it was found that the highest concentration of phthalic anhydride, equal to 0,763 mole fractions (Fig. 1), can be obtained in the shortest time $T^* = 1,45$ h. At the same time, the minimum allowable temperature $u(t) = 620$ K should be maintained throughout the process. The relative error of fulfilling the terminal condition was 0,91 %.

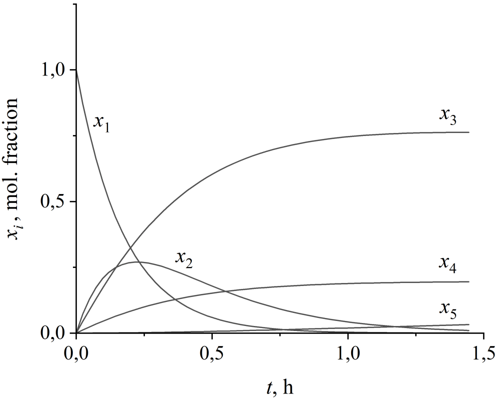

Figure 1. Dynamics of substance concentrations in problem (10)–(15)

Table 1 presents the results of solving the system of differential equations (11) with initial conditions (14) at some permissible values of temperature and reaction time. The table shows that the developed algorithm was used to calculate the shortest process time for the synthesis of phthalic anhydride at temperature $u = 620$ K, with $x_3(T) = 0,763$ mole fractions.

The developed algorithm is compared with the algorithm of differential evolution without modification. The algorithm parameters are set to the same values that were used to solve the problem using a modified genetic algorithm. Possible solutions were not recorded in a separate array, and the first individual satisfying condition (8) was accepted as an approximate solution to the problem. The estimated performance indicators of the algorithms and their obtained values are shown in Table 1. The values of the indicators were evaluated based on the results of 20 runs of each algorithm at different values of accuracy ε. Reliability was defined as the ratio of the number of successful runs of the algorithm, in which the best T^* value was achieved, to the total number of runs.

Table 1. Results of testing evolutionary algorithms

Differential evolution algorithm	Best T^*, value	Average T^* value	Reliability	Number of iterations
		$\varepsilon = 0,1$		
Without modification	1,565	1,584	0,80	2231
With modification	1,458	1,460	0,85	2214
		$\varepsilon = 0,01$		
Without modification	1,531	1,538	0,90	4397
With modification	1,450	1,452	0,95	3830
		$\varepsilon = 0,001$		
Without modification	1,481	1,487	0,95	5783
With modification	1,450	1,451	0,95	4879

The table shows that at low computational accuracy ($\varepsilon = 0,1$) the number of iterations for which the solution is found by both algorithms differs insignificantly with a small difference in the reliability level. At the same time, the value of T^*, calculated with the modified algorithm is smaller compared to the algorithm without modification. As the value of the parameter ε increases, the smallest time T^* found by both methods is almost comparable. However, the time to compute the solution using the algorithm without modification increases. Therefore, we can conclude that at low computational accuracy the running time of the algorithms is approximately the same, but the modified algorithm calculates the best value of T^*. The modified algorithm requires less time to obtain a more accurate solution compared to the algorithm without modification.

Let us consider the speed problem for the synthesis of phthalic anhydride, in which the concentration of intermediate X_2 at the end of the reaction is set to 0,3 mole fractions:

$$x_2(T) = 0,3. \tag{16}$$

Using the developed algorithm, the optimal temperature regime and the corresponding concentrations of substances were calculated at $\varepsilon = 0,1$ (Fig. 2, 3). The process should begin at a temperature of 620 K. After 0,075 h, the temperature should be increased to the maximum permissible value (644 K) and maintained at this level until the end of the process. The shortest process time was 0,186 h. In this case, the concentration of naphthoquinone was 0,286 mole fractions, which is 4,7% lower than the specified value (16).

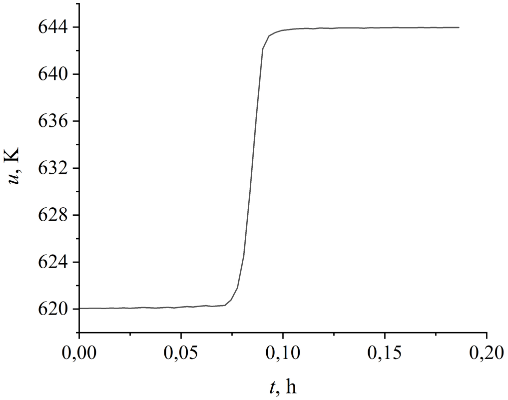

Figure 2. Optimal temperature regime in problem (10)–(14), (16)

Table 2 shows the values of naphthoquinone concentration calculated by varying the process duration for different isothermal regimes. It follows from the data of the table that the closest value of naphthoquinone concentration to the value of 0,3 mole fractions (0,272 mole fractions) is provided at a constant temperature of 628 K and the process duration equal to 0,2 h. However, the error of fulfillment of condition (16) (9,3 %) is greater than its error for the solution of the problem obtained using the differential evolution algorithm (4,7 %). At the same time, the process

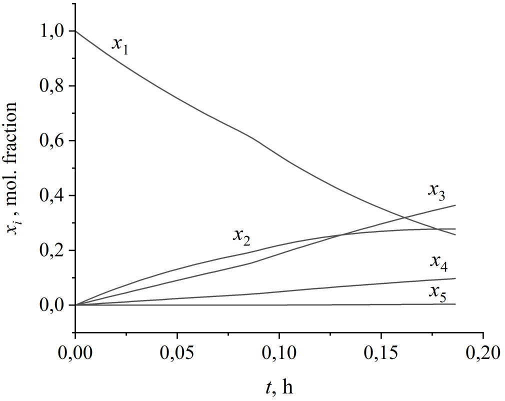

Figure 3. Dynamics of substance concentrations in problem (10)–(14), (16)

time exceeds the calculated smallest value of the process duration equal to 0,186 h.

Now let us determine the control $u(t)$, at which the initial substance, naphthalene, is completely consumed in the shortest time, that is

$$x_1(T) = 0. \tag{17}$$

The calculations were carried out with an accuracy of $\varepsilon = 10^{-3}$. The calculations showed that the reaction should be carried out at the maximum permissible temperature $u(t) = 644$ K for 1 hour. In this case, $1,61 \cdot 10^{-4}$ mole fractions of the initial substance will remain in the reaction system (Fig. 4).

The speed problem (10)–(14), (17) is also solved using the method of variations in the control space [20]. The solution search was carried out with a step for the control equal to 0,01 and the number of time interval partition points equal to 50. At the initial approximation $u^0(t) = 632$ K and $T^0 = 5$ h, $T^* = 1,52$ h was obtained. The structure of the temperature curve is close to the temperature profile calculated using the evolutionary algorithm. The relative error of the temperature vector is 4,11 %.

However, at the initial approximation $u^0(t) = 620$ K, $T^0 = 5$ h, the

Table 2. The value of the concentration of the intermediate substance X_2, calculated under different reaction conditions

u, K	T, h	$x_2(T)$ mole fraction
620	0,1	0,210
	0,2	0,268
	0,3	0,258
	0,5	0,179
	0,1	0,043
628	0,1	0,225
	0,2	0,272
	0,3	0,247
	0,5	0,155
	1	0,030
636	0,1	0,239
	0,2	0,271
	0,3	0,233
	0,5	0,130
	1	0,020
644	0,1	0,252
	0,2	0,266
	0,3	0,214
	0,5	0,107
	1	0,013

local variation method reached a local optimum. The shortest process time $T^* = 1,5$ h, $x_1(T^*) = 2,24 \cdot 10^{-4}$ mole fractions.

Hence, we can see that the method of local variations finds a solution to the problem with well-chosen initial values of the desired parameters. The operation of the evolutionary algorithm does not depend on the choice of the initial approximation, since the initial values of the control parameters are set randomly at step 2.

Based on the results of numerical experiments, we can conclude that the algorithm for finding a solution to the optimal speed problem works correctly. Therefore, it can be applied in the study of regularities of chemical processes on the basis of their mathematical models.

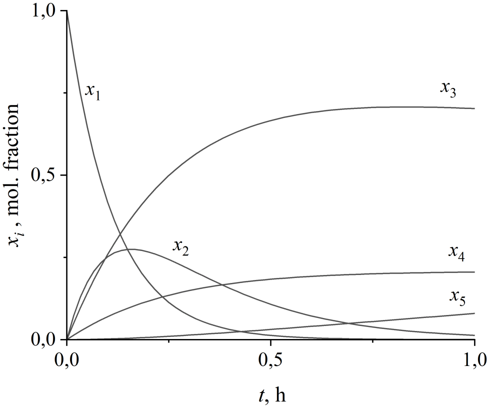

Figure 4. Dynamics of substance concentrations in problem (10)–(14), (17)

4 Conclusion

The search for the solution of optimal speed problems for chemical processes described by nonlinear systems of differential equations can be carried out using the developed algorithm of differential evolution. The peculiarity of the proposed approach is that it can be applied to obtain solutions to optimal speed problems at an unknown initial approximation, which is usually set by the researcher based on the meaning of the problem.

The algorithm has been tested in the process of obtaining phthalic anhydride. The optimal temperature regime and the shortest duration of the process are calculated for a given concentration of the target reaction product, phthalic anhydride, with a fixed content of the intermediate, naphthoquinone, in the reaction mixture, provided that the initial substance, naphthalene, is completely consumed. A comparison of the obtained calculation results with the results of solving problems using the method of differential evolution without modification and the method of variations in the control space showed the advantage of the proposed approach.

The algorithm can be applied to solve problems of speed in chemical engineering with several control actions by modifying the structure of the individual vector.

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