Information-computational system for optimizing the conditions for catalytic reactions

K.F. Koledina[1,2], S.N. Koledin[2]

[1]Institute of Petrochemistry and Catalysis, Russian Academy of Sciences, prospect Oktyabrya 141, Ufa, Russia 450075
[2]Ufa State Petroleum Technological University, Kosmonavtov str. 1, Ufa, Russia, 450062

Abstract. An information-computational system for multi-criteria optimization of catalytic reactions has been developed. Information and computing system combines subsystems for solving individual computing problems. Each subsystem is designed for separate calculations - direct kinetic task, calculation of the inverse kinetic problem, multi-criteria optimization of the reaction conditions. For the catalytic reaction of the benzylalkyl ethers synthesis, using the developed information- computational system, the problem of optimizing the conditions has been solved. Based on the kinetic model, the optimal values of temperature and reaction time are determined.

1. Introduction

An information-computational system is a system for scientific research: systematic data storage, storage, and selection of mathematical models, implementation of numerical algorithms for solving problems, complex and voluminous calculations.

Unified methodology for the analysis of complex catalytic reactions based on kinetic models allows the study of a wide class of catalytic reactions. Methods of multicriteria optimization in the form of an information-computational system will allow analyzing catalytic reactions with a view to their subsequent implementation in production

2. Information system structure

The information-computational system for multicriteria optimization of catalytic reactions based on the kinetic model consists of the following groups and modules:

Group I - Modules for the development of a catalytic reaction kinetic model:
1) The module for calculating kinetic curves with the choice of a mathematical description [1, 2] and a solution algorithm is a direct kinetic problem [3, 4].

2) The module for calculating kinetic parameters with the choice of the type of residual functional and the solution algorithm (inverse kinetic problem [5-7]).

II group. Database of kinetic models and optimality criteria:

3) Database module of kinetic models and optimality criteria.

III group. Modules of optimization and optimal control of the catalytic reaction conditions:
4) A single-criteria optimization module with a choice of variable parameters, an optimality criterion [8], a solution algorithm [9].

5) Multicriteria optimization module with a choice of variable parameters, optimality criteria, solution algorithm [10].

6) The module for optimal control of the reaction conditions with the choice of control parameters, optimality criterion, solution algorithm [11].

When the processing system, each module interacts with others. Such a decomposition of the original problem flexibly develops and modifies each of the modules. The whole system continues to work stably.

3. Optimization algorithms in the information-computational system

It is necessary to use single-criterion optimization algorithms to solve the inverse kinetic problem and the problem of determining the optimal conditions for a catalytic reaction with one optimality criterion. The inverse kinetic problem is incorrectly posed. To solve this problem, it is necessary to apply global and local optimization methods. In these methods, a global optimum and a local refinement are sought. For single-criterion optimization, the following algorithms are implemented in the module of the information-computational system: genetic algorithm and Hook Jeeves algorithm.

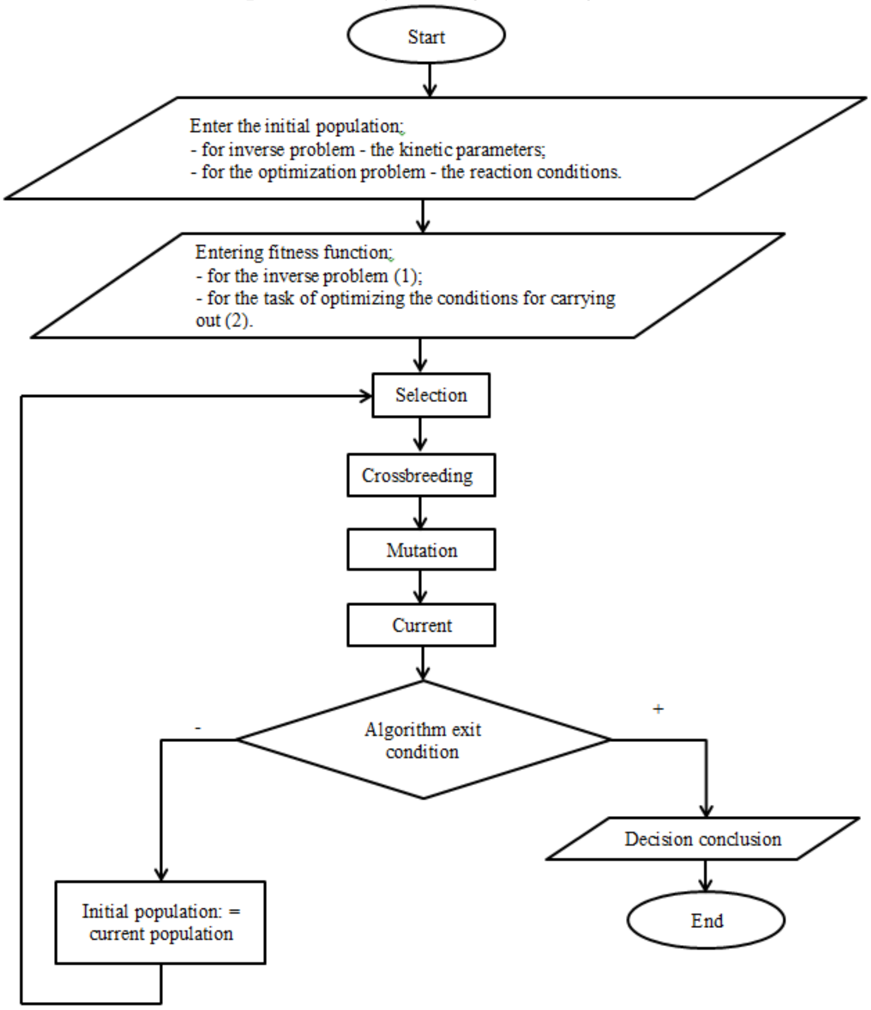

Figure 1. The scheme of the genetic algorithm.

The genetic algorithm solves the optimization problem by imitating the principles of biological evolution, repeatedly changing the population of individual points using the rules on the model of combinations of genes in biological reproduction. Due to its random nature, the genetic algorithm increases the chances of finding a global solution. It allows solving problems without restrictions, with restrictions on variables, and general optimization problems. The solver does not require differentiability or continuity of the objective function [12].

In figure 1 shows the scheme of the genetic algorithm. At the first stage, the initial population is formed (introduced or determined) - the first approximation of the desired parameter value. For the inverse kinetic problem, the parameters are the rate constants of the stages and activation energy, for the task of optimizing the conditions - temperature, reaction time, etc. Then, at the Selection level, the population develops with the exception of solutions with a lower fitness function. The sample contains the values of the population with the best values of the fitness function.

For the inverse kinetic problem, the fitness function is the residual functional (1).

$$Z_Y = \sum_{i=1}^{N}\sum_{j=1}^{M} \frac{\left|y_{ij}^e - y_{ij}^c\right|}{y_{ij}^e} \to \min; \; Z_T = \sum_{i=1}^{N}\sum_{j=1}^{M} \frac{\left|T_{ij}^e - T_{ij}^c\right|}{T_{ij}^e} \to \min. \tag{1}$$

Where Z_Y is a concentration component of a residual functional; Z_T is a temperature component of a residual functional; y_{ij}^e и y_{ij}^c are experimental and calculated values of component concentrations, N is a number of substances, M is a number of measuring points over time of the observed substances during the reaction.

For the task of optimizing the conditions for carrying out a catalytic reaction, the fitness function is an optimality criterion (2).

$$f(X) = f(t^*, T, \mathbf{y^0}) \to \max_{X \in D_X}. \tag{2}$$

Where f is a optimality criterion function; t^* is a reaction time, min; T is a temperature, K; $\mathbf{y^0}$ is a vector of initial component concentrations, *mol / l*; X is a vector of variable parameters, D_X is a range of permissible values of the vector X.

At the Crossing stage, the population sparse at the previous stage is restored due to the grouping (crossing) of the genes (values) of existing solutions. At this stage, a local refinement of the extremum occurs. At the Mutation stage, random changes occur in the genes of existing solutions. This allows us to diversify the population and determines the global nature of optimization.

To refine the global optimum found by the genetic algorithm in the information-computing system, the Hook-Jeeves method is implemented.

The Hook-Jeeves method is a direct search method or a zero-order method. It is divided into two phases: exploratory search and pattern matching [13]. The general algorithm for the Hook-Jeeves method is presented in Figure 2.

In the exploratory search is selected a certain initial vector of values of the required parameters $U=\{u_1,u_2,..., u_{|U|}\}$. The step value h is set. Then, the value of the optimization optimization function is calculated at three points: $U=\{u_1, u_2,..., u_{|U|}\}$, $U=\{u_1+h, u_2,..., u_{|U|}\}$, $U=\{u_1-h, u_2,..., u_{|U|}\}$, and the transition to the point with the smallest value of the objective function is performed. For the inverse kinetic problem, the optimization target is the residual functional (1), and for the optimization of the conditions, is the optimality criterion (2). The exploratory search ends after iterating over all $|U|$ coordinates. The resulting point is called the base.

In pattern matching search, a step is performed from the obtained base point (the coordinates of the base point are the desired parameters: for the inverse kinetic problem - the rate constants of the stages and activation energy, for the task of optimizing the conditions - temperature, reaction time, etc.) along the straight line connecting this point from the previous base. The step is equal to the distance between the base point and the previous base point. The result is a point for which an exploratory search is conducted.

4. Modeling and optimization of the catalytic reaction conditions in the synthesis of benzylalkyl ethers

In [14], a kinetic model was developed for the reaction of benzylbutyl ether synthesis in the presence of a metal complex catalyst. The reaction produces the desired product. $PhCH_2OBu$ benzyl butyl ether and by-product $PhCH_2OCH_2Ph$ dibenzyl ether. The introduction of the process into production requires determining the optimal reaction conditions in order to obtain the highest yield of the target product and the smallest by-product. Based on the kinetic reaction model, the formulation of the multicriteria optimization problem is possible.

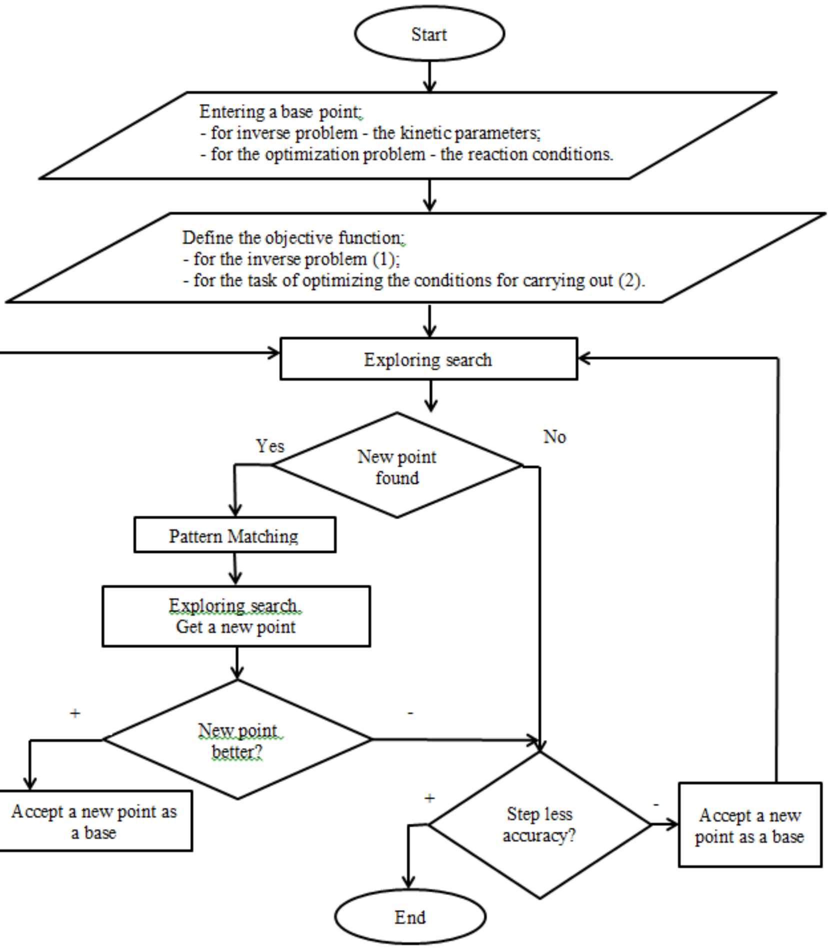

Figure 2. The algorithm of the Hook-Jeeves method.

Chemical experiments were carried out at several temperatures from 140 to 175°C. In this temperature range is necessary to determine the extrema values to achieve the optimality criteria of the reaction. Then the variable parameters are temperature and reaction time, with corresponding physicochemical limitations.

The main parameters of the multicriteria optimization problem for the synthesis of benzylbutyl ether have the form [15-17]:

- Vector of variable parameters

$$X=(x_1, x_2), \tag{3}$$

where x_1 – reaction temperature, T; x_2 – reaction time, t^*.

- Vector function of optimality criteria

F(X)=(f$_1$(X), f$_2$(X)):

$$f_1(X) = y_{PhCH_2OBu(Y_6)}(t^*,T,N) \to \max\ ;\ f_2(X) = y_{PhCH_2OCH_2Ph(Y_9)}(t^*,T,N) \to \min\ . \tag{4}$$

- $F(X)$ with values in the target space $\{F\}= R^{(F)}=R^2$ defined in area

$$D_X \subset \{X\}=R^{|X|}=R^2\text{: } T \in [T^-; T^+],\ t^* \in [t^{*-}; t^{*+}]. \tag{5}$$

Task multiobjective optimization conditions of the catalytic reaction of benzyl butyl synthesis have the form (3)-(5).

The solution to the problem of a multicriteria optimization algorithm was conducted Pareto-approximation NSGA-II [18-20] in Matlab using parallelization [21, 22]. A condition for exiting the algorithm was a minimal change in the value of the optimality criterion (less 10-6), in accordance with the experimental values.

Figures 3 and 4 show the results of solving the multicriteria optimization problem for conditions of the synthesis in benzyl butyl ether in the presence of a metal complex catalyst.

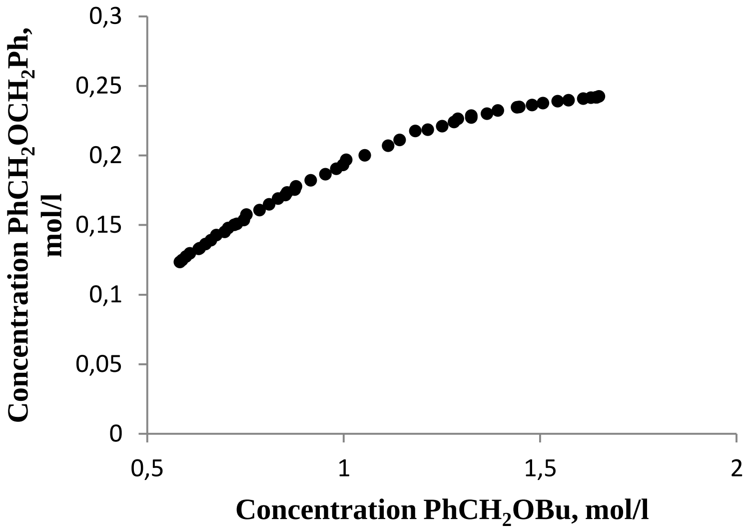

Figure 3. Approximation of the Pareto front in the MCO problem for the synthesis reaction of benzylbutyl ether in the presence of a metal complex catalyst.

According to the approximation of the Pareto front (Figure 3) with the yield of the target benzyl butyl ether, the output of the secondary dibenzyl ether also increases. The corresponding values of the varied parameters, i.e. the Pareto set is shown in Figure 4.

If the reaction is carried out up to 600 min (as in a chemical experiment [14]), the optimum temperature is 140°C (figure 4). However, a longer reaction time leads to an increase in the yield of the target and by-products. The increase in the duration of the process up to 800 minutes requires heating the mixture to 170°C to maintain an upward trend in product yields, as calculated.

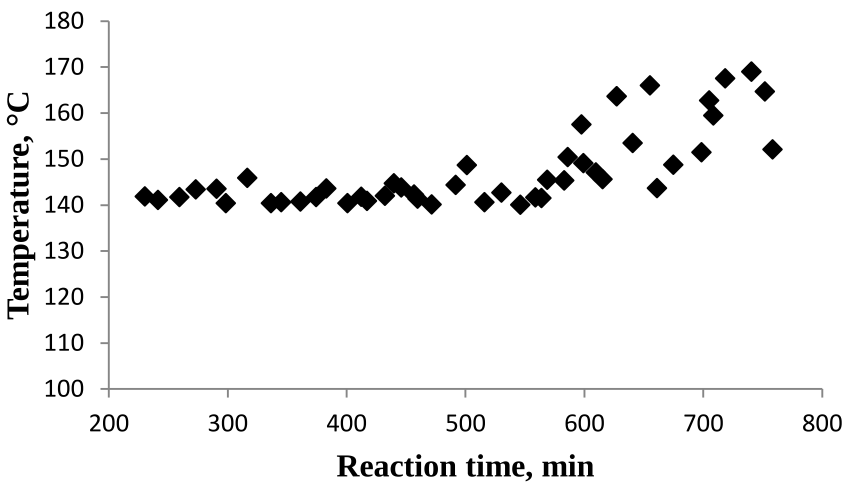

Figure 4. Approximation of the Pareto set of MCO problems for the synthesis of benzylbutyl ether in the presence of a metal complex catalyst.

The adequacy of the calculated values is confirmed by experimental data and assessing the quality of the approximation Pareto [23-26]:

Uniformity of distribution of solutions (average distance between solutions).

$$\bar{d}(A)=\frac{1}{|C_1|*|C_2|}\sum_{i\in C_1,j\in C_2} d(i,j) \quad (6)$$

where $i \in C_1$, $j \in C_2$ - solutions, C_1, C_2 – clusters, $d(i, j)$ - distance between solutions i, j.

Average dispersion is a measure of uniformity of distribution of solutions.

$$I_S(A) = \sqrt{\frac{1}{|A|-1}\sum_{j=1}^{|A|}(\bar{d} - d_j)^2}, \qquad (7)$$

where $|A| = |C_1| + |C_2|$ - solution set of capacity.

The values of evaluation criteria Pareto approximation (6), (7) are shown in Table. 1. These values correspond to the error in the experimental data.

Table 1. Quality assessment of Pareto approximations**.**

	Phenotype	Genotype
The average distance between solutions $\bar{d}(A)$	0,008	0,006
Medium dispersion $I_S(A)$	0,153	0,13
Cardinality of the set of solutions $\|A\|$	50	50

5. Conclusion

Thus, an information-computational system for multi-criteria optimization of catalytic reactions has been developed. Information and computing system combines subsystems for solving individual computing problems. Each subsystem is designed for separate calculations - direct kinetic problem, calculation of the inverse kinetic problem, multi-criteria optimization of the reaction conditions. For the catalytic reaction of the synthesis of benzylalkyl ethers, using the developed information-computing system, the problem of optimizing conditions for the solution has been solved. Based on the kinetic model, the optimal values of temperature and reaction time are determined.

6. Acknowledgments

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7. References

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