Parallelizing a solution of multicriteria optimization problem for the conditions of a chemical reaction based on CUDA technology

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Abstract. In this work, the task of multicriteria optimization for the conditions of the reaction of alcohols with dimethyl carbonate in the presence of a catalyst for cobalt carbonyl or tungsten carbonyl was solved. The criteria of optimality re considered - the output of the target product and the profitability of the process. Implemented calculation of attainable program in the space of optimality criteria and approximating the Pareto front, the Pareto set. The results of the parallelization efficiency of the computing process are presented.

1. Introduction

At present, given the almost universal use of electronic computing systems, the efficient use of computing power is one of the most pressing scientific problems.

The product range of the chemical, petrochemical and oil refining industries is constantly expanding. Product quality is improving. The increased technical level of chemical production requires a reduction in the development time of new catalytic processes and improvement of existing processes. The task is also to establish the true mechanism of the chemical process to optimally control the entire technological system. An important stage in the mathematical modeling of chemical-technological processes is the development of adequate kinetic models (KM) of the chemical process [1,2]. KM includes kinetic equations, the pattern of changes in the concentration of substances over time, the values of kinetic parameters. The optimization problem statement assumes the existence of criteria, variable parameters and some restrictions on variable parameters [3,4]. A task with several mutually independent optimization criteria is a multicriteria optimization task. The study of dynamic (time-varying) variable parameters leads to the optimal control problem. Thus, the identification of optimal conditions for carrying out complex catalytic reactions leads to the problems of single-criterion optimization, multicriteria optimization, and optimal control.

2. Reaction of dimethyl carbonate with alcohols in the presence of metal catalysts

The homogeneous liquid-phase catalytic reaction of dimethyl carbonate (DMC) with alcohols in the presence of a metal complex catalyst $Co_2(CO)_8$ is new and belongs to the field of "green chemistry". The reactivity of DMC is lower than that of commonly used methylating agents: methyl halides (MeX; X = I, Br, Cl) and dimethyl sulfate. But it has less toxicity. DMC is preferable for methylation of alcohols, acids, and amines [5,6]. For a successful reaction, it is necessary to use a large excess of DMC (1:40) and “harsh conditions” (more 200^{o}C), which is a serious obstacle to the widespread use of DMC as a methylating agent in synthetic practice. It was previously shown [7] that the interaction of alcohols with dimethyl carbonate (DMC) in the presence of W-, Mn-, V-, and Co-containing metal

complex catalysts allows one to obtain the corresponding simple and complex ethers under milder conditions with high selectivity. The most active metal complex catalyst $Co_2(CO)_8$. As a result of this reaction, alkyl methyl esters and alkyl methyl carbonates are formed. Alkyl methyl ethers are used as an additive to engine oil, and are used as intermediates for the manufacture of medicines, dyes, and aromatic additives. Carbonates are used as organic solvents, as useful protecting groups for alcohols and phenols in organic synthesis, as an electrolyte solvent in lithium-ion batteries, and as monomers for the phosgene-free synthesis of polyurethanes and polycarbonates.

For a new, industrially significant, catalytic reaction of DMC with alcohols, it is necessary to develop a kinetic model. Based on this model, identify the optimal process conditions.

3. Kinetic model of the catalytic reaction of dimethyl carbonate with alcohols

The mathematical model of chemical kinetics problems is a system of ordinary nonlinear differential equations (SONDE) with initial data, i.e., the Cauchy problem (1) [8-11].

$$\frac{dy_i}{dt} = \sum_{j=1}^{J} v_{ij} \left(k_j^0 \cdot \exp\left(-\frac{E_j}{RT} \right) \cdot \prod_{i=1}^{I} (y_i)^{|\alpha_{ij}|} - k_{-j}^0 \cdot \exp\left(-\frac{E_{-j}}{RT} \right) \cdot \prod_{i=1}^{I} (y_i)^{\beta_{ij}} \right), \; j = 1,\dots J; t \in [0, t^*];$$
$$k_j = k_j^0 \cdot \exp\left(-\frac{E_j}{RT} \right); \quad (1)$$

starting conditions: $y_i(t = 0) = y_i^0$.

Where v_{ij} are stoichiometric coefficients; J is a number of steps; y_i is a concentration of a reactant, mol/L; I is a number of compounds; E_j^+, E_j^- – activation energy of direct and reverse reactions, kcal/mol; R – gas constant 8,31 J/(mol*K); T is a temperature, K; α_{ij} – negative elements of the matrix v_{ij}, β_{ij}– positive elements v_{ij}, k_j^0, k_{-j}^0 – preexponential factors, $k_j – k_j$ is a rate constant of steps; 1/min; t^* is a reaction time, min.

In [12,13], a mathematical reaction model was constructed. Based on the proposed reaction scheme, a kinetic reaction model is constructed. The values of preexponential factors and activation energies of the stages are given in table 1.

Table 1. Kinetic parameters of the catalytic reaction of DMC with alcohols in the presence of $Co_2(CO)_8$.

N	Stages	$\ln k_j^0$, L/(mol*min)	E_j, kcal/mol
k_1	$Co_2(CO)_8 + ROH \rightarrow Co^{2+}(ROH) + Co(CO)_4^- + 4CO$	22,00±0,01	24,10±0,01
k_2	$Co(CO)_4^- + (MeO)_2CO \rightarrow Me^+[Co(CO)_4^-] + CO_2 + MeO^-$	11,9±0,8	8,1±0,8
k_3, k_8	$Co(CO)_4^- + (MeO)_2CO \leftrightarrow Co(CO)_4CO_2Me + MeO^-$	20,70±0,01; 10,90±0,01	13,40±0,01; 9,00±0,01
k_4	$Co(CO)_4CO_2Me + ROH \rightarrow HCo(CO)_4 + CO_2 + ROMe$	18,20±0,01	15,00±0,01
k_5	$Co(CO)_4CO_2Me + ROH \rightarrow ROCO_2Me + HCo(CO)_4$	12,0±0,1	5,6±0,1
k_6	$Me^+[Co(CO)_4^-] + ROH \rightarrow ROMe + HCo(CO)_4$	20,1±0,4	21,5±1,5
k_7	$HCo(CO)_4 + MeO^- \rightarrow MeOH + Co(CO)_4^-$	15,90±0,01	10,60±0,01

Uncertainty in the values of kinetic parameters (table 1) arises because the inverse kinetic problem has a non-unique solution. To determine the parameters, a large number of experimental data were processed at various temperatures and initial amounts of the catalyst [12]. The obtained values of the kinetic parameters vary in the indicated interval.

4. Formulation of multi-objective optimization problem

The initial value of the amount of catalyst ykt and the reaction time were considered as variable parameters t_1:

$$X=X(y_{kt}, t_1). \quad (2)$$

For the reaction of alcohols with DMC, the output of the target product (alkyl methyl carbonate) will be considered as optimality criteria $ROCO_2Me$). Then the criterion will take the form:

$$f_1(X)=y_{ROCO2Me}(X)\rightarrow max \quad (3)$$

The second criterion considered is the profitability of the reaction depending on the costs of the reagents, some variables and fixed costs, as well as on a certain specific weight (price) of the target products.

$$f_2(X) = \frac{\sum_{prod=1}^{Pr} y_{prod}(X)\cdot\eta_{prod}}{\sum_{source=1}^{Sr} y_{source}(X)\cdot\eta_{source} + \psi(X) + A} \rightarrow \max \quad (4)$$

Here y_{prod} is the concentration of the reaction products; y_{source} is the reactant concentration; η is the vector of a specific weight of the costs of components (normalized by the sum of the component costs and expenses); ψ refers to the variable expenses (normalized by the sum of the component costs and expenses); A describes the constant expenses (normalized by the sum of the component costs and expenses); Pr is the number of products; Sr is the number of reactants.

The problem of optimizing the conditions of the catalytic reaction of alcohols with dimethyl carbonate is:

- Functionality of optimization criteria $F=(f_1, f_2)$ according to (3), (4).
- Variable (free) parameters X according to (2).
- Mathematical model in the form of a system SONDE (1).
- Direct limits on variable parameters D_U.

Then

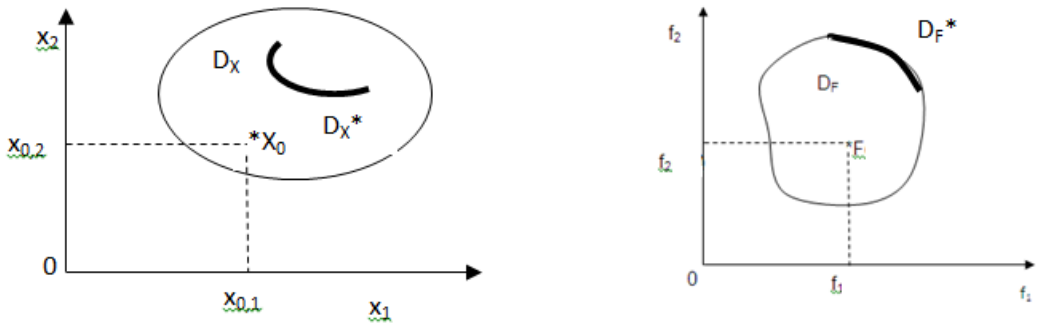
$$\max_{X\in D_X} F(X) = F(X^*) = X^* \quad (5)$$

where X^* - optimal values of variable parameters.

5. Multicriteria optimization of catalytic reaction conditions

The solution to the problem of multicriteria optimization (MCO) is a compromise solution according to several independent criteria. It is also called the Pareto solution or the Pareto approximation. The solution to the MCO problem in the range of optimization criteria will be called the Pareto front. The solution to the problem in the range of values of variable parameters is by the Pareto set.

The numerical definition of the Pareto set and front can be expressed as follows [14-18]. Let $X=(x_1, x_2, ..., x_{|X|})$ - vector of variable parameters. D_X - set of valid vector values X (fig.1a). Then, $F(X)=(f_1(X), f_2(X),..., f_{|F|}(X))$ – optimization criteria vector, $F(X)$ – display the set D_X into some set D_F, which is called the reachability domain. From the set of D_F one can distinguish a subset of D_F^* points that are not dominated by other points (fig. 1a). The set D_F^* is called the Pareto front. The subset of points of variable parameters $D_X^* D_X$, corresponding to the set D_F^* is called the Pareto set (fig. 1b).



a) b)

Figure 1. Multicriteria optimization conditions of the catalytic reaction: a) the range of variable parameters and the Pareto set; b) reachability area and Pareto front.

6. Parallelization computing process

The solution to the problem of multicriteria optimization of complex catalytic processes is computationally complex and time-consuming. In this regard, work is actively ongoing in the field of methods and algorithms for organizing parallelization computing systems and their functioning. One of the directions in this area is the distribution of core fluxes of various types. The advantages of this approach: since all the cores are a central part of the operating system and provide coordinated access to computer resources, such as processor time, memory, external hardware, an external input and output device, it is possible to allocate and use each stream as an independent computing system that functions parallel and independent of other flows. In addition, this approach allows you to optimize existing devices at the program level, avoiding their replacement or conversion. One of the key tasks of parallelization by cores is the organization of the distribution of computing power on threads, that is, the redistribution of sub-tasks of the main task across the kernels. Such redistribution is, in essence, scheduling the sub-tasks of this task with the simultaneous formation of the necessary resources in a certain period [19].

In tasks with a small dimension of the range values of variable parameters, a solution is possible by a grid algorithm. A set of D_X covered by a mesh with the desired measurement accuracy. Then, at each grid node, the values of the objective functions are calculated (Fig. 2). Also, grid nodes can be randomly generated. The advantage of this algorithm is the efficient parallelization of the solution.

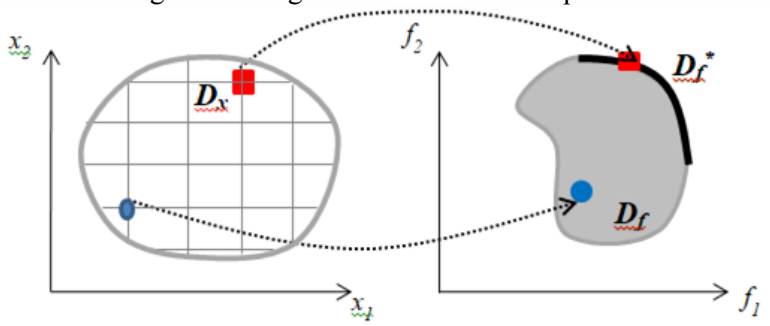


Figure 2. The grid algorithm for solving MCO problem.

The task of multicriteria optimization conditions for the reaction of alcohols with dimethyl carbonate (1) - (5) was solved using in-line parallelization methods in a computer system: central processor Intel Core i7-9750H, video card: Nvidia Geforce RTX2060RAM: 16GB.

In the solution set different intervals for each flow: catalyst amount of from 1 to 5 mM in steps of 0.01 mM. The interval is divided by the number of threads.

A program was developed in the Python programming environment using libraries: Numpy (array generation), Math (calculation of mathematical functions), Scypi (numerical methods for solving the system of differential equations (1)), Threading - library for parallelization on a CPU (Central Processing Unit), Datetime - the module needed to find the program run time, Numba - library used for parallelization on GPU [20].

When solving, the island model of parallelism was used. Parallel population algorithms based on the island model of parallelism are called migration algorithms or commutation multipopulation algorithms in some publications. The essence of the algorithms of this class is as follows. Create a multipopulation $S = S_1 \cup S_2 \ldots \cup S_P$, consisting of the number of subpopulations (islands) equal to the number of working processors used P parallel computer. Each island processes its own processor system. Data exchange between the islands is performed after the independent iterations (seasons) c used in accordance with island neighborhood topology. In MCO tasks, each of the islands S_i has its own local archive sets, and the host processor supports global archives. The island model uses static load balancing of a parallel computer and provides, as a rule, high performance under the following conditions: the computing system is homogeneous; the sizes of the subpopulations are the same and quite large; condition for the completion of iterations of each of the subpopulations S_i is achieving the same number of iterations [20].

Parallelization of the computational process was carried out on a central processing unit (CPU) and a graphic processor (GPU), using CUDA technology. Figures 3 and 4 show the results on acceleration and efficiency of parallelization application in the MCO task for the CPU.

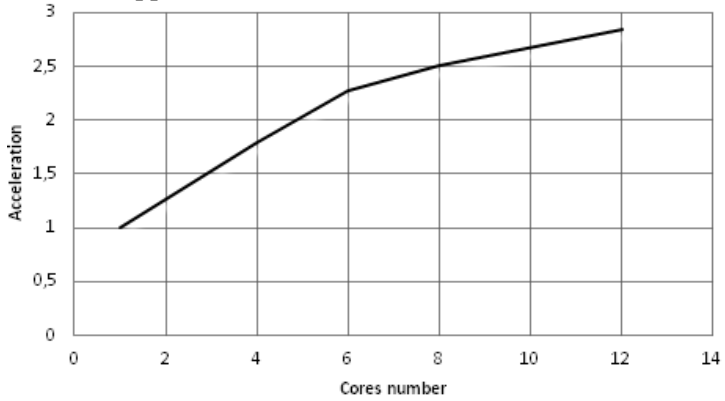


Figure 3. CPU parallelization acceleration.

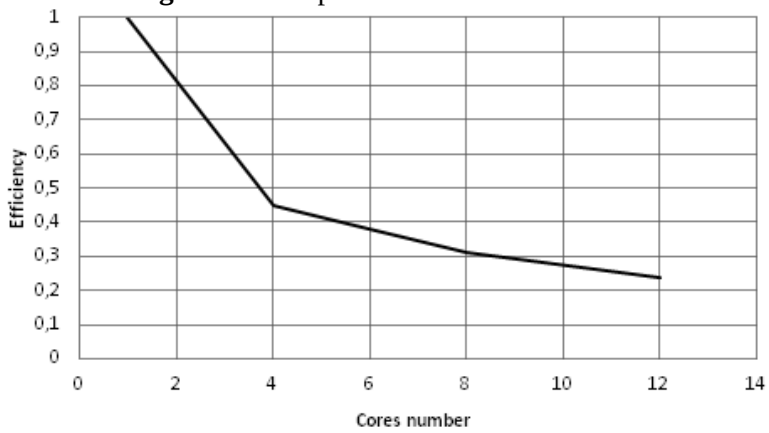


Figure 4. CPU parallelization efficiency.

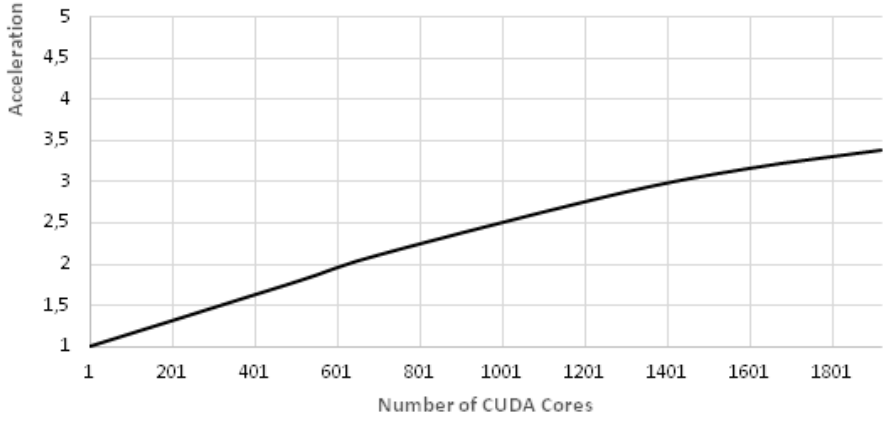


Figure 5. Acceleration of parallelization on the GPU by CUDA.

Acceleration is non-linear, because synchronization time of threads and operating systems is necessary. From fig. 3.4 it follows that the parallelization efficiency, when using 1 to 4 threads decreases faster than when changing the number of cores from 4 to 12 threads. Regardless of how many threads are parallelized, the cores of the central processor will distribute the computational load on all available threads. The processor cannot evenly distribute the load on the threads, since when using a small number of threads, the threads used are loaded as much as possible.

Figures 5 and 6 show the results on acceleration and efficiency of parallelization application for GPU with CUDA technology.

The parallelization efficiency decreases more rapidly with an increase in the number of CUDA cores from 1 to ~ 582 than with a change in the number of CUDA cores from ~ 582 to 1920 (Fig. 6). Thus, with a significant increase in the number of CUDA cores, the efficiency will decrease slightly. This behavior of the graph is caused by the fact that CUDA technology regulates the number of cores involved, and the greatest performance gain is achieved when using 600 or more cores.

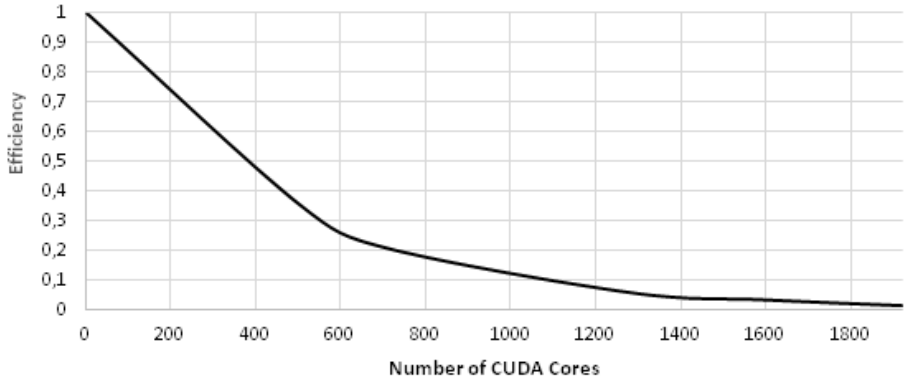


Figure 6. CUDA GPU Parallelization Efficiency

7. Results

When solving the Pareto approximation problem, the following results were obtained: Reachability area D_F objective functions (3) (4) for the reaction of dimethyl carbonate (DMC) with alcohols in the presence of a catalyst $Co_2(CO)_8$ (fig. 7).

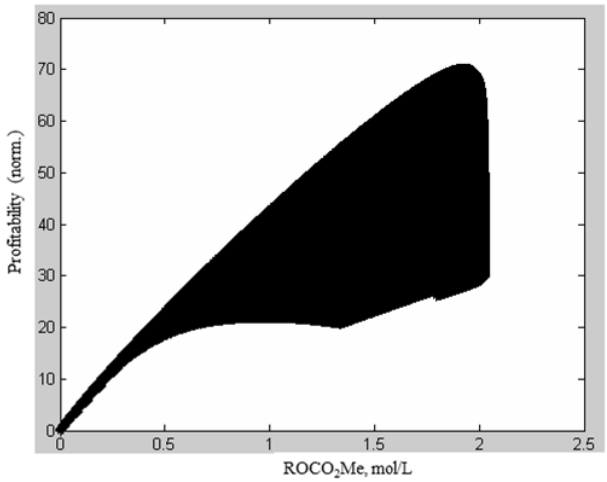


Figure 7. The value of the target functions reachability in catalytic reaction DMC with alcohols.

According to the results of solving the MCO problem and based on the obtained values, we can talk about a set of points on the basis of which the decision maker (DM) can take practical values according to the reaction conditions.

8. Conclusion

Thus, by the methods of the grid algorithm, the front and Pareto set for the MCO problem of the catalytic reaction of alcohols with DMC are determined. For decision-makers, variation areas of the parameters along the Pareto set are provided, with the corresponding extrema along the Pareto frontier. In a practical, technological sense, the application of the above methods makes it possible to obtain exhaustive options for the conditions of chemical processes. And at the same time, take into account economic and production requirements in any production cycle.

Parallelization on a video card significantly reduces the time spent on a computational experiment (from days to minutes). Acceleration efficiency is non-linear. This is due to the difficulty of

parallelizing the solution of a system of differential equations of large dimension. In the future, it is planned to use and parallelize evolutionary algorithms to solve multi-task optimization problems.

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