

Article

The System of Models and Optimization of Operating Modes of a Catalytic Reforming Unit Using Initial Fuzzy Information [†]

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Abstract: The study aims to develop a system of models and a method for optimizing the operating modes of a catalytic reforming unit using fuzzy information, which makes it possible to effectively control the reforming process of the object under study. The object of study of this work is a catalytic reforming unit that has been operating for more than half a century and is characterized by the lack of clarity of some part of the initial information. The research methods are methods of system analysis, mathematical modeling, multicriteria optimization, and expert assessments, as well as methods of theories of fuzzy set theories, which allows formalizing and using fuzzy information, as well as experimental-statistical methods. As a result of the conducted research, the following main results were obtained. Based on a systematic approach, an effective methodology has been developed for developing a system of models of interconnected plant units using various types of available information, including fuzzy information. Using the proposed method, hybrid models have been developed to determine the volume of the produced catalyzate and its quality indicators. A scheme has been constructed for combining the developed models of the main units of the catalytic reforming unit into a single package of models. The built system of models makes it possible to systematically simulate the operation of the plant under study and improve the efficiency of the facility by increasing the volume of target products produced and improving its quality indicators. A statement of the problem of multicriteria optimization is obtained, taking into account the partial fuzziness of the initial information, and a heuristic method for its solution is developed, which is based on the use of knowledge, experience, and intuition of the decision-maker. The results of modeling and optimization show the effectiveness of the proposed fuzzy approach.

Keywords: installation of catalytic reforming; hydrogenated; catalysis; mathematical model; multicriteria optimization in a fuzzy environment; fuzzy information; methods of fuzzy set theories; heuristic method; decision-makers

1. Introduction

In refineries for the production of high-quality motor fuels that meet modern environmental requirements, thermocatalytic processes are currently used, which take place in

catalytic cracking and reforming units in the presence of catalysts [1,2]. Such technological installations are related to economic and ecological systems [3].

This is because these installations in the course of work produce petroleum products that are in demand on the world market (higher quality motor gasoline, household gas, and raw materials for petrochemical synthesis), which determines the economic attractiveness of production. In addition, various wastes are thrown away and this negatively affects the natural environment. Since the technological units of catalytic cracking and reforming include various interconnected technological units, they belong to complex chemical engineering systems (CES).

In addition, many technological units of cracking and reforming operate under conditions of scarcity and unclearness of initial information about their states.

The shortage or absence of initial quantitative information on the operation of complex chemical engineering systems is associated with the difficulty of measuring some technological parameters, and can also be caused by the shortage, low reliability, or absence of measuring instruments at production facilities. All this greatly complicates the research and development of mathematical models and the optimization and control of the CES operating modes using traditional mathematical methods [4,5].

Often in practice, such hard-to-quantify CES are quite effectively controlled by experienced technologists, human operators, due to their knowledge, experience, and intuition, which are expressed in natural language, i.e., fuzzy information [6]. In this regard, the tasks of formalizing and using fuzzy information, which is the experience and judgment of a human operator, who in practice is a decision-maker (DM), an expert in the subject area, have become very relevant for optimizing and managing the operating modes of such CES.

This led to the emergence and development of a promising direction in science and technology—a fuzzy approach to modeling, optimization, and control of complex CES based on the methods of expert assessments and theories of fuzzy sets [7–12].

It should be noted that one of the urgent tasks of oil refining and the economy of the Republic of Kazakhstan is to increase the depth of oil refining and increase the production of high-quality motor fuels. The importance of this task is also justified by the fact that at present there is a great demand for high-quality and environmentally friendly motor fuels in the domestic and world markets.

In connection with the increased environmental requirements for motor fuels, the relevance of the production of high-quality gasoline that meets the requirements of environmental standards and regulations is growing even higher. The most effective approach to solving the above problem under the conditions of catalytic cracking and reforming process units operated at the refineries of Kazakhstan is the use of modern methods of mathematical modeling in a fuzzy environment [13,14].

The developed mathematical models of technological objects for the production of petroleum products in a fuzzy environment and modern methods of multicriteria optimization make it possible to create a computer system for efficient control of the operating modes of the cold storage system and the production of high-quality motor fuel [15–18].

To conduct a comparative analysis, we analyze the works [6,9,11,17], in which approaches to the development of mathematical models of objects are studied in conditions of fuzzy initial information. In the analyzed works, the issues of production control with fuzzy initial information [6], the application of fuzzy set theories [9], as well as the problems of modeling a catalytic cracking reactor [11] and controlling the reforming process [17] were studied. However, in these and other well-known works on modeling complex objects in a fuzzy environment, the issues of developing a system of mathematical models of interconnected technological units have not been sufficiently studied. In this regard, the issues of developing a system of models of interconnected CES units, which are characterized by fuzziness, and methods of multi-criteria optimization of their operating modes can be attributed to problems that have not been fully resolved, and therefore require additional research.

Since many CES consist of interconnected technological objects, it is necessary to investigate and solve the problems of constructing a complex of interconnected models of individual CES units and its system modeling. In this regard, this work is devoted to the development of a methodology for constructing a package of mathematical models of interconnected CES aggregates, as system modeling of complex systems and optimization of their operating modes in a fuzzy environment are of theoretical and practical importance.

In this regard, the development of an approach to the construction of mathematical models of interconnected technological objects of a catalytic reforming unit and optimization of its operation modes using the initial fuzzy information is determined as the main goal of this work.

To achieve the formulated goal, the following research tasks are solved in the work:

1. To create a methodology for the development of models of interconnected units for systemic modeling of CES on the example of a catalytic reforming process unit;
2. To build models of reforming reactors, taking into account the fuzziness of the initial information on the basis of the proposed methodology;
3. To formalize the statement of the problem of multicriteria optimization of the operating modes of reforming reactors in a fuzzy environment and the development of a heuristic method for its solution.

A schematic illustration that makes it easier to understand the general essence of the work is shown in Figure 1.

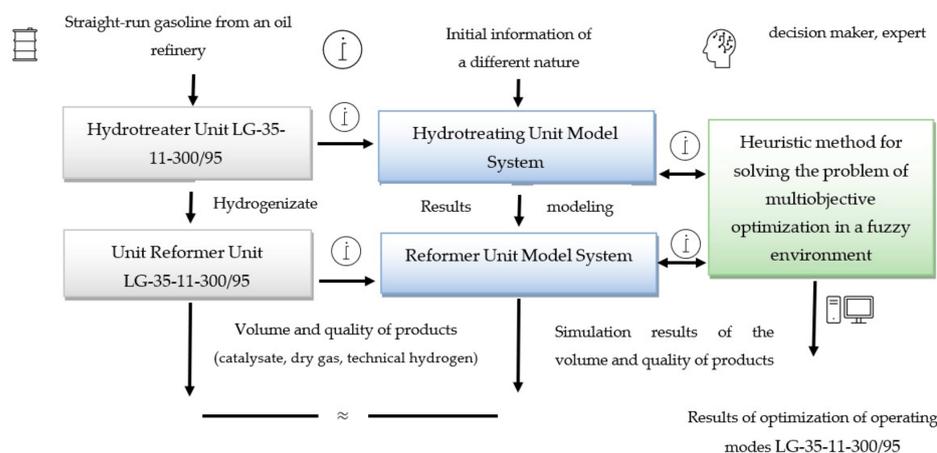


Figure 1. A general scheme showing the structure of the present work.

The following conclusions can be drawn:

- in conditions of scarcity, fuzzy initial information for the development of mathematical models and optimization of operating modes of the catalytic reforming unit, hybrid methods should be used to effectively use various available and fuzzy information to solve these problems;
- as a result of computer simulation of various operating modes of the catalytic reforming unit, it is possible to determine such ratios of input and operating parameters that provide an efficient operating mode of the unit;
- based on the formulation and solution of the problem of multicriteria optimization of regime parameters, taking into account the fuzziness of the initial information, it is possible to ensure the optimal mode of operation of the object in a fuzzy environment due to the knowledge, experience, and intuition of decision makers and experts;
- the optimal operating mode of the catalytic reforming unit is ensured by the maximum volume and high quality of the produced motor fuel, i.e., high-octane and environmentally friendly motor gasoline

Subsequent sections of this article are organized as follows: Section 2 outlines the materials and methods used in the study; Section 3 presents the results of the study—the proposed methodology for developing models of interconnected units for systemic modeling of CES on the example of a catalytic reforming process unit; developed models of reforming reactors taking into account the fuzziness of the initial information based on the proposed methodology; statement of the problem of multicriteria optimization of the operating modes of reforming reactors in a fuzzy environment and a heuristic method for its solution, as well as the results of practical application of the obtained research results; Section 4 discusses the main results of the study; Section 5 shows the main contribution of the work; Section 6 presents conclusions.

2. Materials and Methods

As a specific technological unit for the production of high-quality mother fuel, this work considers a catalytic reforming unit of the LG-35-11/300-95 model operating at the Atyrau refinery. The issues of increasing the efficiency of the work of the investigated by ensuring the optimal operating mode of the catalytic reforming unit are investigated and solved using modern mathematical methods and methods of fuzzy mathematics [6–13,17–19].

Here is the basic information and a brief description of the research object. The catalytic reforming unit LG-35-11/300-95 of the Atyrau Refinery was commissioned in 1971 with a design feedstock capacity of 300 thousand tons per year. As a result of the reconstructions carried out, the productivity was increased to 450 thousand tons per year [20].

The main target product of the LG-35-11/300-95 unit is high-octane motor gasolines with an octane rating of up to 95 points (according to the research method). The plant also produces liquefied domestic gas and hydrogen-containing gas.

Currently, the LG-35-11/300-95 unit at the Atyrau Refinery consists of the following units:

1. Block of preliminary hydrotreating of straight-run gasoline (naphtha), intended for hydrotreating of straight-run gasoline from the installation of primary oil refining from sulfurous and other harmful organic compounds and obtaining a hydrogenated product;
2. Block of reforming of hydrotreated gasoline, hydrogenated product intended for converting naphthenes and paraffins into aromatic hydrocarbons in the presence of a UOP-R-56 catalyst and obtaining catalyzate;
3. Block for stabilization of platformate, intended for de-ethnization and stabilization of unstable catalyzate in the C-6 fractionating absorber of the unit.

The process flow diagram of the catalytic reforming unit with the listed units is shown in Figure 2.

Hydrotreating unit. In this block, the hydrotreating process takes place with the participation of technical hydrogen (TH) and a catalyst of the S-12T type, developed by the American company UOP. The mixture of raw materials and TH is fed through the P-16 pump to the series-connected heat exchangers H-1/1-3, where, due to the counterflow of carbonated raw materials from the R-1 reactor and the H-3 reboiler, it is heated to a temperature of 260 °C.

Further, the raw material is fed to the F-101 hydrotreating furnace. From the F-101 furnace, a mixture of raw materials and gas with a temperature of 300–343 °C is fed to the R-1 hydrotreating reactor. In the reactor R-1 with the participation of the catalyst S-12T, the reaction of hydrotreating the feedstock proceeds, and the raw material is preliminarily hydrotreated to remove sulfur, nitrogen, and oxygen-containing compounds. The heat of the mixture of unstable hydrogenate, circulating gas from the outlet of the reactor, and the heat of reaction of gases with a temperature of 340–420 °C is used to heat the mixture of raw materials and gas, first in the heat exchanger H-3 of the stripping column C-1, then in the heat exchangers H-1/1-3 [20].

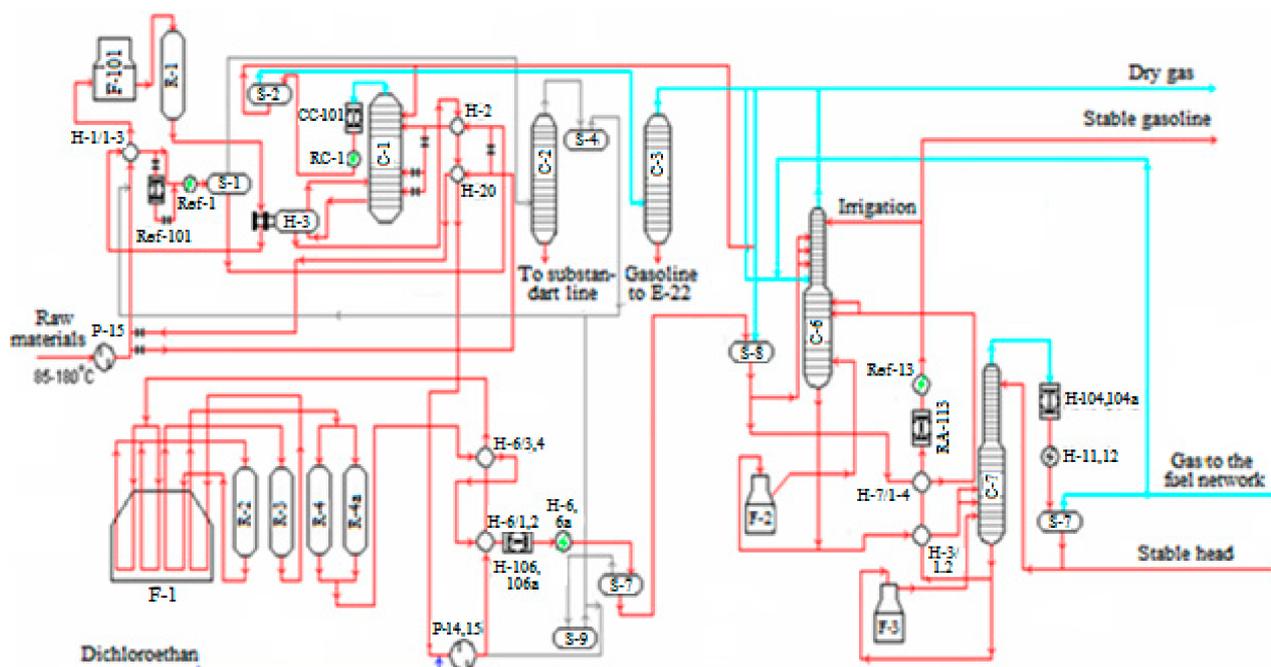


Figure 2. Process flow diagram of the LG-35-11/300-95 catalytic reforming unit. Technological lines: red—Petrol; grey—Technical hydrogen; Birch—Process gas; Blue—Dichloroethane.

After cooling to a temperature of 35 °C in the Ref-101 and Ref-1 refrigerators, the product enters the S-1 separator, where the TH is separated from the liquid and is fed to the C-2 absorber for purification from hydrogen. The gas from the outlet of the absorber C-2, after passing through the separator S-4, is divided into two streams:

- (1) the circulating gas, after being compressed in the compressors, is fed back to the feedstock hydrotreating system;
- (2) excess TH from the outlet of the installation, the liquid phase of the separator S-1 passes through the heat exchanger H-2, here it is heated to a temperature of 150 °C and floats on 7, 9, 23 trays of the stripping column C-1. In column C-1, sulfuric hydrogen and water are stripped from the hydrogenate with a temperature of up to 270 °C and a pressure of up to 15 atm. In addition, light hydrocarbons are removed from the top of the column.

After the stripping column C-1, the total composition of sulfur compounds in the hydrogenation product should not exceed 0.0005% of the mass. Gases in the state of vapor from the top of the column C-1 come out with a temperature of 135 °C, pass through the CC-101 and RC-1 condensers, and with a temperature of 35–40 °C are fed to the separator S-2. From the separator S-2, the liquid phase is returned to the stripping column C-1. Hydrocarbon gas from the S-2 separator for hydrogen sulfide removal enters the K-3 absorber. Hydrocarbon gas from the top of the C-3 absorber is fed to the fractionation absorber of another unit or to the refinery's fuel network.

In the process of hydrotreating, a chemical transformation of a substance occurs under the influence of hydrogen gas with high pressure and high temperature [4,21]. In the process of hydrotreating in the composition of petroleum products, fuels and sulfur compounds are reduced, additional unsaturated hydrocarbons are saturated, the composition of the tar, oxygenated compounds decreases, as well as hydrocracking of hydrocarbon molecules.

The reforming unit is designed to carry out the processes of converting naphthenes and paraffins into aromatic hydrocarbons using the UOP-R-56 catalyst. Due to their high-octane numbers, aromatic hydrocarbons are used as commercial quality gasoline.

In the catalytic reforming unit, the hydrogenate, i.e., the product of the hydrotreating unit, through heat exchangers H-2 and H-20, enters the intake of pumps P-14, 15 and is

mixed under pressure with the circulating gas. Next, the resulting mixture is heated by the heat of the reforming reactors R-2, R-3, R-4.4a is heated in H-6. Then it enters the F-1 furnace for heating to the required temperature. To increase the activity of the catalyst, dichloroethane is injected onto the heated stream (before R-2, R-3, R-4.4a).

Reactors R-2, R-3, and R-4.4a undergo the reforming process. At the same time, the intermediate heating of these reactors is performed by the F-1 multi-chamber reforming furnace. Since the aromatization process has a negative thermal effect, the temperature in R-2, R-3, R-4.4a decreases. The decrease in temperature is restored to 490–530 °C due to heating in the second and third stages of the reforming furnace.

From the parallel-connected reactors R-4 and R-4a, the product mixture, which has a temperature of 490–530 °C, enters the heat exchangers H-6/3-4. After cooling in these heat exchangers to a temperature of 250–300 °C, the gas-product mixture is fed to the S-7 separator, in which it is separated into target products for liquid catalysis and hydrogen-containing gas. The resulting unstable catalysis from the high pressure separator S-7 is fed to the separator S-8 (low pressure separator) for further separation.

The separated hydrocarbon gas together with the gases from the C-3 absorber is fed to the C-6 column (fractionation absorber). Heated to 156 °C in H-7, the remainder of the catalysis is fed down column C-6 for fractionation. Hydrogen-containing gas from the top of the S-7 separator is fed back to the reforming system through the S-9 separator (circulating gas separator).

Since the unit under study is characterized by a lack of quantitative information and the vagueness of some important parameters that affect the efficiency of the facility and the quality of products, problems arise in the development of mathematical models and control of the processes of improving the quality of gasoline, using traditional mathematical methods. However, at this facility, experienced operator-technologists have been working for many years who are decision-makers, expert specialists, and can effectively manage the hydrotreating process based on their intuitions, knowledge, and experience, using fuzzy information.

It follows from the above that the object under study operates under conditions of uncertainty due to the lack and fuzziness of the initial information. Therefore, it will be necessary to solve the problems of developing a system of mathematical models of interconnected technological objects of the catalytic reforming unit using the initial fuzzy information in the form of knowledge, experience, and intuitions of decision makers and experts. The resulting system (package of models), with the help of decision-making methods, allows one to select the optimal operating modes of a technological unit in a fuzzy environment, providing a significant economic effect [22,23].

Therefore, in order to optimize and effectively control a catalytic reforming process unit, it is necessary to develop a system of adequate mathematical models of interconnected units of these units using available information of a different nature, including fuzzy information. For this, the work uses experimental statistical methods [24,25], methods of system analysis [26,27], methods of expert assessments, and the mathematical apparatus of theories of fuzzy sets, as well as hybrid methods for the development of mathematical models [7–10,28].

Informal methods of systems analysis and expert methods during research allow collecting, processing, and the use of formalized fuzzy information to build mathematical models. Research in work is carried out in the following sequence:

- study and description of the object—the investigated technological unit; collection processing of available experimental and statistical information about the functioning of the object;
- organizing and conducting an expert assessment to collect the missing part of the initial information about the operation of the facility in the form of fuzzy information;
- processing of the collected fuzzy information, definition of term-sets, and construction of the function of accessories of fuzzy parameters of the research object;

- creation of a methodology for constructing a package of mathematical models of interconnected objects of a technological unit based on available information of an experimental-statistical, fuzzy nature, and theoretical information about the object;
- on the basis of the proposed methodology, the development of a package of mathematical models of the technological unit;
- using the developed models and methods of decision-making in a fuzzy environment, select the optimal operating mode of the technological unit for the production of high-quality gasoline, which ensures high efficiency and environmental safety of the research object.

3. Results

Catalytic reforming unit LG-35-11/300-95 is a complex of chemical engineering system, consisting of interconnected blocks and units, which are simultaneously affected by a large number of different parameters. The main interconnected units of the plant include reactor (hydrotreating R-1; reforming R-2, R-3, R-4.4a), columns (C-1, C-2, C-3), hydrotreating and reforming furnaces F-101, F-1, separators, heat exchangers, etc. (see Figure 2).

For systemic modeling of the catalytic reforming unit operation, it is necessary to develop a package of mathematical models of these main units. Since the technological units of the catalytic reforming unit are interconnected, changes in the operating parameters of one of them lead to a change in the parameters of others, which affects the processes of hydrotreating and reforming.

In this regard, in order to optimize and control the process of hydrotreating and reforming in the optimal mode, it is necessary to have a package of related mathematical models of these main units, which are compiled on the basis of a systematic approach [26]. In practice, under production conditions, some important parameters characterizing the quality of products of hydrotreating and reforming processes are difficult to measure quantitatively or not measured, but can be imprecisely assessed by decision makers and experts. In practice, due to the experience, knowledge, and intuition of the operator-technologist and decision maker, formalized in the form of fuzzy information, the problem of choosing effective operating modes and managing these objects is often successfully solved.

Therefore, based on the methodology of system analysis, we propose a methodology for determining the optimal types and developing a package of models of interconnected units of complex CES in a fuzzy environment, such as a catalytic reformer.

3.1. Methodology for the Development of Models of Interconnected Units for Systemic Modeling of CES (on the Example of a Catalytic Reforming Process Unit)

As a result of the analysis of various methods for developing mathematical models of complex CES, it was revealed that research papers do not cover the issues of system modeling of such systems, which consist of interconnected technological units in conditions of uncertainty and fuzziness of the initial information. In conditions of uncertainty associated with a lack of initial information, it is usually proposed to apply probabilistic modeling methods or simulation methods [24,29,30].

However, in cases of uncertainty due to the fuzziness of the initial information, which often arise in the study and management of real CES, the use of probabilistic methods is unjustified. This is because under these conditions statistical information is insufficient or absent, and the axioms of the probability theory (statistical stability of an object, repeatability of experiments under the same conditions, etc.) are not fulfilled. Sometimes the available initial information is only fuzzy information, which is the knowledge, experience, and judgments of decision makers and experts.

With the competence of these sources of information, and with the correct organization of their interrogation, collection, and processing of fuzzy information on its basis, it is possible to build models that take into account all the complex and non-formalizable relationships of various parameters of the studied CES. The resulting models based on fuzzy information can be more meaningful than the models developed by traditional

methods, and most importantly, adequately describe real CES in conditions of fuzzy initial information.

Consider the main idea of the proposed methodology for the development of a package of mathematical models of interconnected CES units using the example of the development of a package of models for the main technological units of a catalytic reforming unit.

Mathematical models of each unit (element) of the CES can be constructed using various methods, depending on the availability of one or another type of initial information and on other factors. Thus, for individual elements of the chemical engineering system, it is possible to build various types of models, for example, statistical, fuzzy, or combined. Therefore, to combine such models into a single package (system) of models, on the basis of which system modeling and optimization of the installation as a whole are carried out, it is necessary:

- analyze the advantages and disadvantages of each type of model, the construction of which is possible;
- develop criteria for comparison and selection of models by cost, by purpose, by accuracy, etc.;
- to define the principles of combining the developed models into a single package.
- Based on the results of the study, the analysis of various types of models of the main units of the catalytic reforming unit LG-35-11/300-95 was carried out. As a result of studies of the specifics of the main units of the catalytic reforming unit [14,20,31], experimental data and expert demand, and analysis of approaches to modeling such or similar units, an assessment of possible types of models of the main units of the catalytic reforming unit was carried out. The result of the system analysis carried out on the evaluation of the models is presented in the form of Table 1. For evaluation and ranking the types of models, a five-point scale was used.

Table 1. Analysis and assessment of the types of models of the main units of the reforming unit LG-35-11/300-95.

Aggregates Catalytic Reforming Units (Main)	Criterion for Comparison and Choice of Model Type	Types of Models			
		Deterministic	Statistical	Fuzzy	Combined
Reactors: R-1; R-2; R-3; R-4, R-4a	Availability of necessary information	2	4	4	5
	Development cost	1	4	3	3
	Degree of adequacy	4	3	4	4
	Applicability for solving the problem of optimization and control	3	3	4	5
	Possibility of combining the model into a package	4	3	3	3
	<i>Integrated criterion</i>	14	17	18	20
Furnaces: F-101, F-1	Availability of necessary information	3	5	4	5
	Development cost	2	4	4	4
	Degree of adequacy	5	4	4	4
	Applicability for solving the problem of optimization and control	4	5	4	4
	Possibility of combining the model into a package	4	4	4	4
	<i>Integrated criterion</i>	18	22	20	21

Table 1. Cont.

Aggregates Catalytic Reforming Units (Main)	Criterion for Comparison and Choice of Model Type	Types of Models			
		Deterministic	Statistical	Fuzzy	Combined
Separators: S-1; S-2; S-7; S-8; S-9	Availability of necessary information	4	5	4	5
	Development cost	3	4	4	4
	Degree of adequacy	5	4	4	4
	Applicability for solving the problem of optimization and control	4	5	4	4
	Possibility of combining the model into a package	4	4	4	4
	<i>Integrated criterion</i>	20	22	20	21
Heatxc-hangers: H-1; H-2; H-3; H-6, refrigerators: Ref-1; Ref-6	Availability of necessary information	5	4	4	5
	Development cost	5	3	4	3
	Degree of adequacy	5	5	4	5
	Applicability for solving the problem of optimization and control	4	5	5	5
	Possibility of combining the model into a package	4	5	4	4
	<i>Integrated criterion</i>	23	22	21	22

Note: Assessment (ranking) on a point scale (from 1 to 5), where 1 is the lowest grade; 5th highest score. The estimates can be fuzzy, such as fuzzy numbers or sets.

As the main criteria for comparing and choosing the optimal type of model, by which they are evaluated, the following are highlighted: the availability of the necessary information for building a model of the corresponding type; the cost of developing a model; the degree of model adequacy; applicability of the model for solving the problem of optimization and control in a fuzzy environment; the possibility of combining the developed model into a single package for the purpose of systemic modeling of the operation of the unit as a whole.

Table 1 reflects the results of the expert evaluation according to the given criteria for comparison and choice of the type of model of the main units of the catalytic reforming unit, obtained on the basis of processing the results of the expert analysis by the Delphi method [7]. Based on the information given in the above table, it is possible to make a multicriteria choice of the optimal type of the model of the catalytic reforming units according to the specified comparison criteria and the choice of the model type.

The optimal type of model is determined by the maximum value of the integrated criterion. Since the local criterion is scoring, the integrated criterion for choosing the optimal type of model is defined as the sum of the scores of the local criteria.

As the results of the analysis and evaluation for complex units of the LG-35-11/300-95 installation such as reactors and furnaces show, due to the difficulty of studying the processes occurring in them and the impossibility of obtaining reliable data, the construction of deterministic models for them is practically impossible or economically unreasonable.

For heat exchangers, the construction of deterministic models is possible, since there is sufficient theoretical information necessary to build a model of this type and the integrated criterion takes the maximum value (23) (see Table 1).

Statistical (stochastic) models of furnaces F-101, F-1, and separators S-1; S-2; S-7; S-8; S-9 catalytic reforming units are relatively easy to build, convenient for combining into a single system of models, and are suitable for solving problems of optimization and plant control. Based on the results of the study, it can be concluded that for furnaces and separators, the most optimal is the development of statistical models, since the integrated criterion takes the maximum value—22 points each.

At the catalytic reforming unit LG-35-11/300-95 of the Atyrau Refinery, the collection of reliable experimental and statistical information for the construction of statistical mathematical models of hydrotreating reactors R-1 and reforming reactors R-2, R-3, R-4.4a is

hampered by the lack of special industrial devices and the low reliability of the available measuring instruments. This is confirmed by the results of the expert assessment, since the integrated criterion for these types of models is low.

In this regard, as a more effective mathematical apparatus that supplements the missing data on the basis of fuzzy information, the methods of fuzzy set theories have been chosen [9,10]. At the same time, to collect fuzzy information around expert opinions in natural language, methods of expert assessments are used, for example, the Delphi method. To formalize, process, and use fuzzy information in the construction of models, methods of fuzzy set theories and combined (hybrid) methods of developing models are used [9–11,22,28]. As can be seen from Table 1, the optimal type of model for reactors is a combined model, which is built using available information of a different nature, for example, statistical data and fuzzy information. The adequacy of such hybrid models with the correct and full use of available data and fuzzy information is high enough, and they can also be effectively used to optimize reforming processes.

In this regard, in practice, in order to build models with a shortage of initial information, it is necessary to use any available information. Models of technological units obtained on the basis of such data will be called combined. They can be obtained using various combinations of the available data. However, the construction of combined models may be impractical due to the need for a stage of organization, research, and experiments of various nature, as well as preliminary processing of the collected data.

The combination of individual models of units into a package is carried out in accordance with the course of the technological process in the chemical engineering system. In this case, for the outputs of one model, the results of modeling one unit are input parameters for the model of another unit. For example, in the catalytic reforming unit, the simulation results of the R-2 reactor are the initial data for modeling the operation of the 2nd stage of the F-1 multi-chamber furnace; the simulation results of this furnace stage are the input data for the R-3 reactor models.

The results of modeling the operation of the R-3 reactor are the initial data for the 3rd stage of the F-1 furnace and the output results, which are the initial data for the models of the R-4.4a reactors. Thus, in addition to the adequacy and effectiveness of their application in a computer system for modeling and optimizing the operating modes of the installation, the main criteria for choosing types of unit models, in addition to the adequacy and effectiveness of their use in the computer system for modeling and optimizing the operating modes of the installation, also include the ease of combining them into a single package of models, mutual correspondence of output, and input variables of related models. Based on the results of research and analysis of the operation of interconnected units of hydrotreating and reforming units of the catalytic reforming unit, and in accordance with the flow of hydrotreating and reforming processes, the developed models are combined into a single package of models. The scheme of combining individual models of the main units into a package of models is shown in Figure 3.

In the above diagram, combining individual models of units into a single package of models (Figure 3) through F-101, R-1, H-3, S-1, S-2, C-1, C-2, C-3, F-1, R-2, R-3, R-4, R-4a, H-6, S-7 designates the software-implemented models of the main units of the hydrotreating and reforms units of the LG-35-11/300-95 unit. For convenience, the designations of the programs are taken in accordance with the designations of the modeled main units of the catalytic reforming unit discussed above.

With the help of this software package on a computer, it is possible to systematically simulate various operating modes of the catalytic mode installation and determine the optimal operating modes that provide a significant economic effect and environmental safety of production. In addition, the system modeling of the operation of the technological unit allows to identify the “bottlenecks” of the plant, the solution of which will allow increasing the power and productivity of the technological unit.

On the basis of the system analysis methodology, we propose a methodology for the development of mathematical models of interconnected units of chemical engineering sys-

tems, for example, the LG-35-11/300-95 installation based on fuzzy information and other available data. In this case, the available initial information can be statistical, experimental data, theoretical information, and a fuzzy description of the object's operation.

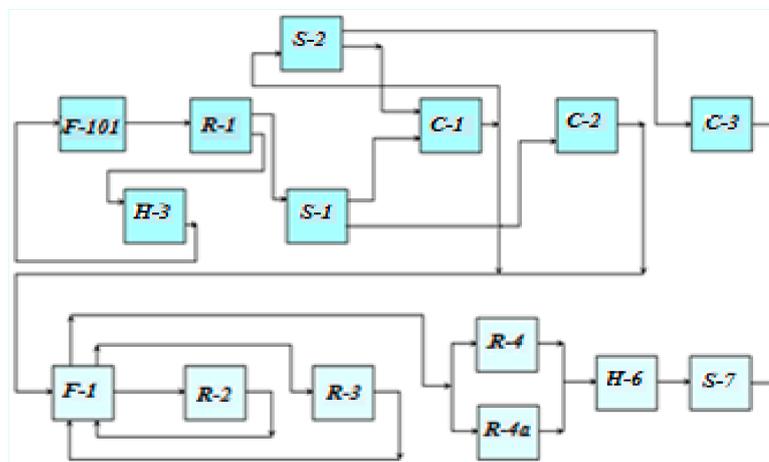


Figure 3. Scheme of combining models of the main units of hydrotreating and reforming units of the LG-35-11/300-95 catalytic reforming unit of the Atyrau Refinery into a package of models.

We offer the pseudocode of the proposed methodology for developing models of interrelated CES units:

BEGINNING of the development of models of interconnected CES units

- ** 1. Research of CES by methods of system analysis, collection and processing of available information about the state and operation of CES units and their relationships, determination of the purpose of modeling;
- ** 2. Taking into account the purpose of modeling, generating criteria for comparing and evaluating the types of models of CES elements that are possible to construct for an element;
- ** 3. Organize and conduct an expert assessment of each type of model for CES units according to the selected criteria and determine the effective type of model for each unit using the integrated criterion.
- ** 4. Taking into account the results of system analysis and expert evaluation and, depending on the characteristics of the available initial information, build various types of models. To determine the optimal type of model and build the appropriate model, go to the next paragraphs.
- ** 5. If there is sufficient theoretical information describing the operation of the CES element under study and the deterministic model has the maximum value according to the integrated criterion, then for it, using analytical methods, construct a deterministic model. Otherwise, go to the next item.
- ** 6. If the statistical data that describe the operation of the investigated element (unit) of the CES are sufficient, or they can be collected on the basis of experiments, according to the integrated criterion, the statistical model is optimal, takes the maximum value, then takes the basis of experimental and statistical methods to construct a statistical model of the unit. Otherwise, go to the next item.
- ** 7. If both theoretical and statistical information describing the operation of the investigated unit is insufficient, and the collection of such information is impossible or economically inexpedient but there is unclear information about the object, then the following sittings are possible. In the case of the maximum value of the integrated criterion of the fuzzy model, if the input parameters of the unit are clear and the output parameters are fuzzy, then go to step 9 to build the fuzzy model. Otherwise, if

both input and output parameters of the unit are unclear, go to the next item to build a linguistic model.

- ** 8. Determine and select informative input $\tilde{x}_i \in \tilde{A}_i, i = \overline{1, n}$ and output $\tilde{y}_j \in \tilde{B}_j, j = \overline{1, m}$ fuzzy parameters (variables) of the unit, describing, respectively, the input, operating parameters, and the quality of the unit. These parameters are necessary for building a model and are linguistic variables: $\tilde{A}_i \in X, \tilde{B}_j \in Y$ —fuzzy subsets, X, Y —universal sets, input and output parameters.

- *** 8.1. Based on the methods of expert assessment with the involvement of decision makers, determine the term-sets $T(X_i, Y_j)$ describing the parameters of the modeled object and construct the membership functions of the fuzzy parameters of the object: $\mu_A(\tilde{x}_i), \mu_B(\tilde{y}_j)$. Based on the experience of modeling technological objects of oil refining production in a fuzzy environment, the following adaptable structure of the membership function can be recommended:

$$\mu_{B_j}^p(\tilde{y}_j) = \exp(Q_{B_j}^p \left| (y_j - y_{mdj})^{N_{B_j}^p} \right|),$$

where $\mu_{B_j}^p(\tilde{y}_j)$ —membership function describing the output fuzzy parameters to a fuzzy set \tilde{B}_j ; p —quantum number (sampling interval); $Q_{B_j}^p$ —a parameter (coefficient) that determines the level of fuzziness, which is determined when identifying the membership function; $N_{B_j}^p$ —coefficients defining the domain of definition of terms of the membership function of fuzzy parameters and allowing to change the shape of the membership function graph; y_{mdj}^p —fuzzy variable that most closely matches a given term on a quantum p . this variable is determined from the following condition

$$\mu_{B_j}(y_{mdj}^p) = \max_j \mu_{B_j}(y_j).$$

- *** 8.2. Determine connections between input and output linguistic variables, build fuzzy mappings between R_{ij} and \tilde{x}_i . For the convenience of using fuzzy mapping in the calculation, the matrix of connections with membership functions is determined:

$$\mu_{R_{ij}}(\tilde{x}_i, \tilde{y}_j) = \min[\mu_{A_i}(\tilde{x}_i), \mu_{B_{ji}}(\tilde{y}_j), i = \overline{1, n}, j = \overline{1, m}].$$

Then a linguistic model is built with a general structure:

$$\text{If } \tilde{x}_1 \in \tilde{A}_1 \left(\tilde{x}_2 \in \tilde{A}_2 \left(\dots, \left(\tilde{x}_n \in \tilde{A}_n \right) \right) \right) \text{ then } \tilde{y}_j^M \in \tilde{B}_j, j = \overline{1, m}$$

- *** 8.3. Based on compositional inference rule $\tilde{B}_j = A_i \circ R_{ij}$ to determine the fuzzy values of the output parameters of the investigated element of the CES, then the numerical values of the output parameters are determined from the fuzzy solutions. In this case, using the compositional inference rule, the output parameters of the object are determined, which determine the quality of its work, for example, using the maximin product. Let be \tilde{x}_i , which means the values of the input fuzzy parameters of the object assessed by experts. In this case, the set of current values of the input parameters is defined as a fuzzy set in which the membership functions of the input parameters will be maximal:

$$\mu_{A_i}(\tilde{h}) = \max(\mu_{A_i}(\tilde{x}_i^*)).$$

Then the fuzzy values of the output variables are determined in the form of membership functions, expressing the maximum product:

$$\mu_{B_j}(\tilde{y}_j^*) = \max \left\{ \min_{x_i \in \tilde{X}_i} [\mu_{A_i}(\tilde{x}_i^*), \mu_{R_{ij}}((x_i^*, \tilde{y}_j^*))] \right\}.$$

The quantitative values of the output parameters can be determined using the following expression: $y_j^c = \operatorname{argmax}_{\tilde{y}_j^*} \mu_{B_j}(\tilde{y}_j^*)$, selects the values of the output parameters in which the membership function reaches its maximum values. In order to check and ensure the adequacy of the model, go to item 11.

- ** 9. Identify and select clear input $x_i \in A_i$, $i = \overline{1, n}$ and fuzzy output $\tilde{y}_j \in \tilde{B}_j$, $j = \overline{1, m}$ the parameters of the unit describing the input operating parameters and the quality of the unit. These parameters are necessary to build a fuzzy model of the investigated unit of the chemical–technological system.

- *** 9.1. Determine the structure of fuzzy models $\tilde{y}_j = \tilde{f}_j(x_1, \dots, x_n, \tilde{a}_0, \tilde{a}_1, \dots, \tilde{a}_n)$, $j = \overline{1, m}$ (structural identification of models). For example, the structure of the model can be defined in the form of multiple regression fuzzy equations:

$$\tilde{y}_j = \tilde{a}_{0j} + \sum_{i=1}^n a_{ij} x_{ij} + \sum_{i=1}^n \sum_{k=i}^n a_{ikj} x_{ij} x_{kj}, j = \overline{1, m}.$$

- *** 9.2. Solve the problem of parametric identification of fuzzy parameters, regression coefficients $(\tilde{a}_0, \tilde{a}_1, \dots, \tilde{a}_n)$, for example, using the method of least squares modified on the basis of the set of level α , and go to item 11.

- ** 10. If both theoretical and statistical data and fuzzy information describing the operation of a technological system unit are insufficient or their collection is not economically feasible, and the integrated criterion for the combined model has the maximum estimate using the hybrid method, build a combined (hybrid) model [28]. In this case, the combined model is developed on the basis of available information of a different nature (theoretical, statistical, fuzzy). To do this, to describe a specific parameter of the object, various combinations of the above-described points of the methodology 6, 8, 9 are used.

- ** 11. Checking the adequacy of the model by condition:

$$R = \min \sum_{j=1}^m (y_j^M - y_j^E)^2 \leq R_D,$$

where y_j^M —calculated (model), y_j^E —experimental (real) values of the output parameters of the object, R_D —is the permissible deviation value. If the adequacy condition is met, then the model is recommended for modeling and determining the optimal operation modes of the object, providing an increase in the efficiency of the object—in our case, a catalytic reforming unit. Otherwise, the reason for the inadequacy of the model is determined and the transition is processed to the corresponding points of the described methodology to eliminate the causes of inadequacy and increase it. In this case, the reason for the inadequacy of the model can be not including some parameters in the model that significantly affect the process, or incorrect structural and/or parametric identification of the model, etc.

Display Results

The proposed methodology is based on the application of the methodology of system analysis, methods of expert assessments, and the apparatus of fuzzy set theory. Due to the systematic use of these methods, the proposed method allows developing a package of models of interrelated technological units of CES in conditions of scarcity and fuzziness of

the initial information. Since various data and fuzzy information, which is the experience, knowledge, and intuition of decision makers, experts, are used to build models, a wide range of non-formalizable relationships between various CES parameters is taken into account, which ensures high adequacy of the developed models.

3.2. Building Models of Reforming Reactors Taking into Account the Fuzzy Initial Information Based on the Proposed Methodology

As a result of the application, models of interconnected reforming reactors and catalytic reforming units were built. Mathematical models of interconnected reforming reactors of the LG-35-11/300-95 unit are built on the basis of the methodology proposed above in Section 3.1 for developing models of interconnected CES units using experimental statistical and expert data. Applying the idea of the method of sequential switching on of regressors [6,22,32,33] identified the following structures of combined models of reforming reactors R-2, R-3, R-4, R-4a:

$$y_1^{R_2} = a_0 + \sum_{i=1}^5 a_i x_i + \sum_{i=1}^5 \sum_{k=i}^5 a_{ik} x_i x_k, \quad (1)$$

$$y_1^{R_3} = a_0 + \sum_{i=1}^5 a_i x_i + \sum_{i=1}^5 \sum_{k=i}^5 a_{ik} x_i x_k, \quad (2)$$

$$y_1^{R_{4,4a}} = a_0 + \sum_{i=1}^5 a_i x_i + \sum_{i=1}^5 \sum_{k=i}^5 a_{ik} x_i x_k, \quad (3)$$

$$y_j = a_{0j} + \sum_{i=1}^5 a_{ij} x_{ij} + \sum_{i=1}^5 \sum_{k=i}^5 a_{ikj} x_{ij} x_{kj}, \quad j = 2, 3, \quad (4)$$

$$\tilde{y}_j = \tilde{a}_{0j} + \sum_{i=1}^5 \tilde{a}_{ij} x_{ij} + \sum_{i=1}^5 \sum_{k=i}^5 \tilde{a}_{ikj} x_{ij} x_{kj}, \quad j = \overline{4, 6}. \quad (5)$$

where $y_1^{R_2}, y_1^{R_3}, y_1^{R_{4,4a}}$ —output parameters, production volumes from reforming reactors R-2, R-3, R-4, R-4a; $\tilde{y}_j, j = \overline{4, 6}$ —respectively, the output of dry gas and WGH at the outlet of the reactor block; $\tilde{y}_j, j = \overline{4, 6}$ —main quality indicators of catalyzate: octane number \tilde{y}_4 ; fractional composition: 10% distillation (\tilde{y}_5) and 50% distillation (\tilde{y}_6).

The given quality indicators of catalyzate are determined with the participation of a person, operators, specialists of the central plant laboratory, and are expressed in indistinctness. For example, at the request of the relevant standards, the octane number should be at least $\tilde{\geq} 86$ according to the motor method, and the fractional composition should not be more than $\tilde{\leq} 70$ °C (for 10% distillation) and $\tilde{\leq} 115$ °C (for 50% distillation).

In the resulting structures of models (1)–(5), the input and operating parameters of the reforming reactors ($x_i, i = \overline{1, 5}$) are designated as follows: x_1 —Flow rate of the supplied raw material (50–80 m³/h); x_2 —space velocity in reforming reactors (1.0–1.5 h^{−1}); x_3 is the temperature in the reforming reactors (470–510 °C in R-2, 480–520 °C in R-3, and 490–525 °C in R-4.4a); x_4 —pressure in reforming reactors (25–39 kg/cm² in R-2, 22–35 kg/cm² in R-3, and 20–30 kg/cm² in R-4.4a); $x_5 = \frac{H_2}{x_1}$ the hydrogen/feed ratio takes values in the range from 300 to 500 nm³; a_{0j}, a_{ij}, a_{ikj} and $\tilde{a}_{0j}, \tilde{a}_{ij}, \tilde{a}_{ikj}$ —respectively, clear and fuzzy parameters (regression coefficients) of models (1)–(5), which are subject to identification.

From the results of the structural identification of the reforming reactor models, it can be seen that the models describing the yields of products (catalysis, dry gas, and hydrogen-containing gas) (1)–(4) were constructed by experimental-statistical methods in the form of multiple regression equations. The models that assess the quality of catalyzate are identified in the form of fuzzy multiple regression equations based on methods for synthesizing fuzzy models using fuzzy information from decision makers and experts.

The identification of the parameters of the models (1)–(4) was carried out using the well-known method of parametric identification using the REGRESS software package in which the least squares method is implemented. As a result of parametric identifications and the dependence of the yield of catalyzate and hydrogen-containing gas on the input, regime parameters $x_i, i = \overline{1,5}$ were determined in the form of multiple regression equations, which, due to the large volume, are given in Appendix A.

Based on the simulation results, on the basis of the obtained models, a graph of the dependence of the volume of catalyzate from the outlet of the R-4.4a reactors on the temperature in these reactors was built with fixed values of the remaining input and operating parameters. The plotted graph of the dependence of the volume of catalyzate from the outlet of the reactor block on the temperature of the last reactors is shown in Figure 4.

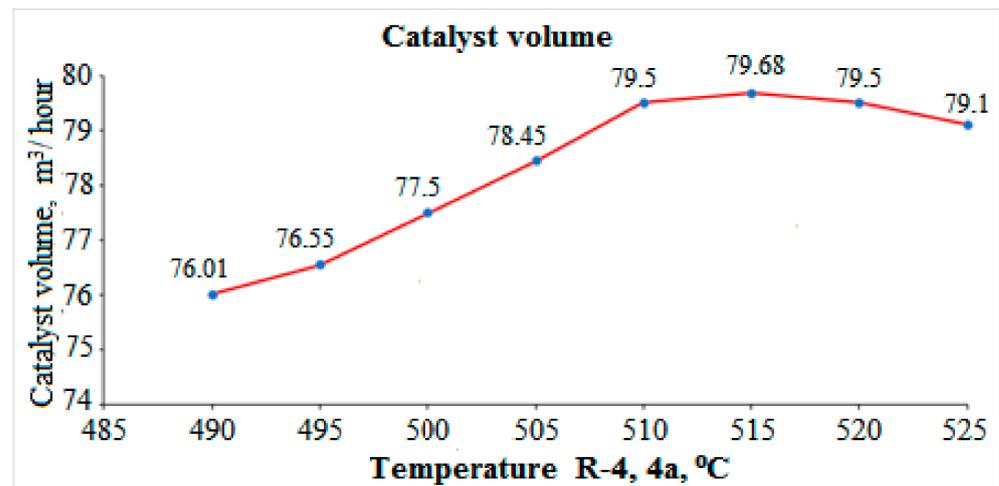


Figure 4. Scheme $y_1^{R_{4,4a}} = f(x_3, x_1, x_2, x_4, x_5)$ —fixed: x_1 —Input of raw materials—80 m³/h; x_2 —Volume velocity—1.3 h⁻¹; x_4 —Pressure in reactors P-4.4a 24 kg/cm²; x_5 —Ratio H₂/raw materials—400 nm³.

For the purpose of parametric identification of fuzzy regression coefficients $\tilde{a}_{ij}, i = \overline{0,5}$ and $\tilde{a}_{ikj}, i, k = \overline{0,5}, j = \overline{4,6}$ models (5) fuzzy sets describing the quality indicators of products are divided into the following sets of level $\alpha = 0.5; 0.75; 1$. Then, based on the methods of fuzzy set theories, the values of the quality indicators of the catalyzate are determined \tilde{y}_4, \tilde{y}_5 and \tilde{y}_6 for each level α . Then, for each level $\alpha = 0.5; 0.75; 1$ models (5) describing the quality indicators of catalyzate, we obtain systems of multiple regression equations. Thus, the identification problem for the resulting system of coefficient models $a_{ij}^{\alpha,q}, i = \overline{0,5}, j = \overline{4,6}, q = \overline{1,3}$ can be solved as well-known problems of estimating the parameters of multiple regression, for example, the least squares method. In this case, you can use the appropriate software packages.

We used the REGRESS software package, which allows us to identify the coefficients of nonlinear regression based on the modified least squares method. Identified Regression Coefficients $a_{ij}^{\alpha,q}$ fuzzy models then, in order to estimate the values of fuzzy coefficients, are combined on the basis of the following rule of fuzzy set theories:

$$\tilde{a}_{ij} = \bigvee_{\alpha \in [0.5,1]} a_{ij}^{\alpha,q} \text{ or } \mu_{\tilde{a}_{ij}}(a_{ij}) = \sup_{\alpha \in [0.5,1]} \min \left\{ \alpha, \mu_{a_{ij}^{\alpha,q}}(a_{ij}) \right\}, \text{ где } a_{ij}^{\alpha,q} = \left\{ a_i \mid \mu_{\tilde{a}_{ij}}(a_{ij}) \geq \alpha \right\}. \quad (6)$$

The results of parametric identification at the selected α levels (0.5; 0.75; 1; 0.75; 0.5) of fuzzy models (5) describing the dependence of the qualitative indicators of the catalyzate \tilde{y}_j (on the input parameters x_1, x_2, x_3, x_4, x_5 , for example, for the octane number of the

catalyzate (\tilde{y}_4) and 10% distillation (\tilde{y}_5) were also transferred to Appendix B due to the large volume.

Similarly, it is possible to identify parametric and estimation models of 50% distillation (\tilde{y}_6), also characterizing the qualities the results of modeling the operation of reforming reactors and comparing them with the results of modeling other known reforming models, as well as real data obtained from the LG-35-11/300-95 unit of the Atyrau refinery, are shown in Table 2.

Table 2. Results of modelling based on known models [34], developed models taking into account fuzzy information and real data from the LG-35-11/300-95 unit of the Atyrau oil refinery factory.

Output Parameters Evaluating the Quantity and Quality of Products	Known, Deterministic Models	Developed Models with Fuzzy Information	Real Data
Output targetproduct, % (mass)	94.7	95.3	95.0
The content of aromatic hydrocarbons y_A , % (mass.)	68.9	-	-
Output catalyst (volume), m ³ /h	77.2	77.9	77.7
The octane number of catalyzate by the motor method		87	(86) ^L

Note: the input and operating parameters of the process are taken approximately the same; (.)^L means that the data was determined with human participation by laboratory.

3.3. Formalization and Formulation of the Problem of Multicriteria Optimization of the Operating Modes of Reforming Reactors in a Fuzzy Environment and the Development of a Heuristic Method for Its Solution

The main criteria for optimizing the operating modes of reforming reactors are maximizing the volume of production. y_1 and y_3 , in addition to improving the quality indicators of products, take into account the imposed restrictions. In practice, these criteria (quantity and quality of gasoline) in the field of effective solutions are contradictory. Since the quality indicators of gasoline: octane number (\tilde{y}_4); fractional composition of gasoline 10% distillation (\tilde{y}_5) and 50% distillation (\tilde{y}_6) not directly measured but determined with the participation of a person (decision maker), expressed indistinctly, for example, using the term “not less” (\lesseqgtr) or “no more” (\gtrless). In this regard, the problem of optimizing the operating modes of reforming reactors must be formulated considering the fuzziness and it is necessary to develop a heuristic method for its solution based on taking into account the experience and knowledge of the decision maker, and their preferences when choosing solutions.

Formalize and present the formulation of the problem of optimizing the operating modes of the reforming reactors based on the models of the reactor block of the reforming unit in conditions of multi-criteria and fuzziness.

Let $F(x) = (f_1(x), f_2(x))$ be the vector of criteria assessing the quality of work, the yield of gasoline and hydrogen-containing gas; $\varphi_q(x) \gtrless b_q$, $q = \overline{1,3}$ —Unclear restrictions imposed on the quality indicators of gasoline: on the octane number “not less than $\lesseqgtr 92$ ”; fractional composition according to GOST 2177-82—10% distillation “no more than $\gtrless 75$ ”; and 50% distillation “no more than $\gtrless 115$ ” [35]. Sign \sim means that the restrictions are not clear.

Each of the local criteria depend on the control vector $x = (x_1, x_2, x_3, x_4, x_5)$ (x_1 —raw material consumption; x_2, x_3 and x_4 —respectively, the space velocity, temperature, and pressure in the reforming reactor R-4.4a; x_5 —H₂/raw material ratio). It should be noted that some of the criteria and limitations may not be clear. Operating and control parameters also have their own boundary values, set by the technological regulations of the installation: $x_j \in \Omega \supset X$, $X = [x_j^{min}, x_j^{max}]$, $j = \overline{1,5}$, where x_j^{min} , x_j^{max} —lower and upper limits of parameter change x_j , $j = \overline{1,5}$. These limits and change intervals may not be clear. (\gtrless , \lesseqgtr , \cong).

It is required to determine and select such a mode of operation of the reforming unit that provides the optimal value of the criteria vector while meeting the specified constraints and the vagueness of some initial data, as well as taking into account the preferences of the decision maker.

A formalized optimization problem under multi-criteria and fuzzy conditions can be written as the following decision-making problem:

$$\max_{x \in X} f_i(x), \quad i = 1, 2 \quad (7)$$

$$X = \{x \in \Omega, \varphi_q(x) \tilde{\geq} b_q, q = \overline{1,3}\} \quad (8)$$

The solution to this problem is the value of the control vector $x^* = (x_1^*, x_2^*, x_3^*, x_4^*, x_5^*)$, providing such values of local criteria that satisfy the decision maker and the conditions of fuzzy constraints are met.

Using the idea of the principles of absolute (relative) assignment and Pareto optimality under fuzzy conditions, it is possible to concretize the mathematical formulation of problem (7) and (8):

$$\max_{x \in X} \mu_0(x), \quad \mu_0(x) = \sum_{i=1}^2 \gamma_i \mu_0^i(x) \text{ or } \mu_0(x) = \sum_{i=1}^2 \gamma_i \log \mu_0^i(x), \quad (9)$$

$$X = \left\{ x : x \in \Omega \wedge \max_{x \in \Omega} \sum_{q=1}^3 \beta_q \mu_q(x) \wedge \sum_{q=1}^3 \beta_q = 1 \wedge \beta_q \geq 0, q = \overline{1,3} \right\}, \quad (10)$$

where \wedge —means a logical “and” that requires that all statements it associates to be true, $\gamma = (\gamma_1, \gamma_2)$ and $\beta = (\beta_1, \beta_2, \beta_3)$ —accordingly, weight vectors reflecting the mutual importance of criteria and constraints, $\mu_q(x)$, $q = \overline{1,3}$ —membership functions (FP) describing the degree of fulfillment of fuzzy constraints. Thus, to formulate and solve the problem of decision-making on the management of the reforming process in a fuzzy environment, the methods of decision-making theories [17], fuzzy sets and expert assessments [2,4,14,17–19] are used.

Let us briefly describe the main steps of the algorithmization of the proposed method based on compromise schemes using the principles of absolute or relative concession (for criteria) and Pareto optimality (for constraints), used to solve the problem of multi-criteria optimization of the operating modes of reforming reactors.

Heuristic method ARA + PO is based on the use of the principles of absolute or relative assignment (ARA) and Pareto optimality (PO) for solving the problem of multicriteria optimization with m criteria and L fuzzy constraints:

Step 1. The values of the vector of weight coefficients are set $\gamma = (\gamma_1, \dots, \gamma_m)$, $\sum_{i=1}^m \gamma_i = 1$, $\gamma_i \geq 0$, $i = \overline{1, m}$, which reflects the mutual importance of local criteria.

Step 2. If $\mu_0(x)$ or $\gamma = (\gamma_1, \dots, \gamma_m)$ then the terms of the term-set are determined for them, and the membership functions are constructed.

Step 3. The decision maker, the experts, choose the terms of the term set $T(X, Y)$, which describe the fuzzy parameters of the object.

Step 4. With the involvement of decision makers and experts, membership functions are constructed that estimate the degree of fulfillment of fuzzy constraints $\mu_q(x)$, $q = \overline{1, L}$.

Step 5. The values of the weight coefficients are set in the form of a vector $\beta = (\beta_1, \dots, \beta_L)$, reflecting the mutual importance of fuzzy constraints.

Step 6. Based on the mathematical model, the integrated criterion is maximized $\mu_0(x) = \sum_{i=1}^m \gamma_i \mu_0^i(x)$ in the event of an absolute assignment or $\mu_0(x) = \sum_{i=1}^m \gamma_i \log \mu_0^i(x)$ in the case of relative concession (9) on the admissible set X , which is determined based on the Pareto principle of optimality (10). Current decisions are determined: $x(\gamma, \beta)$; $\mu_0^1(x(\gamma, \beta))$, $\mu_0^2(x(\gamma, \beta))$, \dots , $\mu_0^m(x(\gamma, \beta))$, $\mu_1(x(\gamma, \beta))$, \dots , $\mu_L(x(\gamma, \beta))$.

Step 7. The results obtained are presented to the decision maker for analysis and final decision making. If the current solutions satisfy the DM, then go to step 8. If the current results do not satisfy the DM, then they are assigned new values of the vectors $\gamma = (\gamma_1, \dots, \gamma_m)$, or $\beta = (\beta_1, \dots, \beta_L)$ and we return to step 6.

Step 8. The best results are displayed, which are selected by the decision maker, namely: vector $x^*(\gamma, \beta)$ and $\mu_0^1(x^*(\gamma, \beta))$, $\mu_0^2(x^*(\gamma, \beta))$, \dots , $\mu_0^m(x^*(\gamma, \beta))$, which pro-

vides the maximum value of the integrated criterion $\mu_0(x^*(\gamma, \beta))$, and maximum degrees of fulfillment of fuzzy constraints $\mu_1(x^*(\gamma, \beta)), \dots, \mu_L(x^*(\gamma, \beta))$.

Let us further assume that $f_1(x) = \mu_0^1(x)$ —normalized main criterion, the volume of gasoline (catalyzate) produced. Since there is no special requirement for the output of the hydrogen-containing gas, in order to simplify the task, we do not consider it. Suppose that for each fuzzy constraint describing the quality indicators of gasoline $\varphi_q(x) \gtrsim b_q, q = \overline{1,3}$ the membership function of its execution is constructed $\mu_q(x), q = \overline{1,3}$. Either a number of priorities are known for the restrictions $I_R = \{1, 2, 3\}$ or a weight vector reflects the mutual importance of these constraints $\beta = (\beta_1, \beta_2, \beta_3)$.

As already noted, the criterion and restrictions depend on the parameters $x_j, j = \overline{1,5}$. These dependences describe mathematical models of the reforming reactor [31].

Then, by modifying the principles of ARA and PO for the case of one criterion and fuzziness, it is possible to write the following statement of the problem of the operating modes of the reforming reactor unit in the form of a decision-making problem with fuzzy constraints:

$$\max_{x \in X} \mu_0^1(x), \quad (11)$$

$$X = \left\{ x : x \in \Omega \wedge \max_{x \in \Omega} \sum_{q=1}^3 \beta_q \mu_q(x) \wedge \sum_{q=1}^3 \beta_q = 1 \wedge \beta_q \geq 0, q = \overline{1,3} \right\} \quad (12)$$

To solve the problem (11), (12), we use the developed heuristic method ARA + PO, based on the involvement of a decision maker and their knowledge and experience in choosing a solution. Let us present the results of solving problem (11), (12) using the proposed method ARA + PO. Since in our case one criterion is considered, its weight is equal to 1 and there is no need to determine the weight coefficients of local criteria.

In the task at hand, the criterion $\mu_0^1(x)$ clearly, therefore it is not defined for its T(X.Y)—Term-set and membership functions are not built.

The term-set is determined, describing fuzzy restrictions. As a result of expert judgment, when describing the limitations, decision makers and experts have chosen the term “not less” (for the octane number) and “not more” (for the fractional composition) and their derivatives, which are obtained using various modifiers.

The membership functions of the fulfillment of fuzzy constraints are constructed $\mu_q(x), q = \overline{1, L}$. Based on clause 8.1 of the above methodology for developing models of interconnected aggregates and research results, the following membership functions for fulfilling constraints are built:

$$\mu_1(x) = \exp\left(82.0|y_2 - 95|^{0.78}\right); \mu_2(x) = \exp\left(72.0|y_3 - 70|^{0.85}\right); \mu_3(x) = \exp\left(110.0|y_4 - 115|^{0.50}\right)$$

y_2, y_3, y_4 —numerical values of fuzzy indicators of the quality of catalyzate obtained using multiple levels $\alpha = 1; 82.0, 72.0, 110.0$ —parameters that are determined when identified and determine the level of fuzziness at a level of $\alpha = 0.5; 95, 70, 115$ —parameters that determine the fuzzy variable that most closely matches the selected term, for which the membership function takes the maximum value; 0.78, 0.85, 0.50—coefficients for changing the domain of definition of terms and the shape of the graph of the membership function of fuzzy parameters.

The decision maker introduces the value of the weight vector of restrictions $\beta = (\beta_1, \beta_2, \beta_3)$, taking into account the importance of local constraints. In our problem, decision makers entered the following values $\beta_1 = 0.6, \beta_2 = 0.2, \beta_3 = 0.2$, i.e., vector $\beta = (0.6; 0.2; 0.2)$.

The problem of maximizing the criterion is solved, yield of catalyzate $\max_{x \in X} \mu_0(x)$ subject to the imposed fuzzy constraints. Current decisions are determined: $x(\beta); \mu_0^1(x(\beta))$ and $\mu_1(x(\beta)), \mu_2(x(\beta)), \mu_3(x(\beta))$. The decision is presented to the decision maker. If the current results do not satisfy the decision maker, then the values of the vector $\beta = (\beta_1, \beta_2, \beta_3)$ and returns to point 2. Otherwise, go to point 8.

The search for a solution stops, the results of the final choice of the decision maker are displayed: the values of the control vector $x^*(\beta)$ best criterion value $\mu_0^1(x^*(\beta))$ and the degree of fulfillment of fuzzy constraints $\mu_1(x^*(\beta))$, $\mu_2(x^*(\beta))$, $\mu_3(x^*(\beta))$. When solving problem (11),(12), the decision maker chose the final solution after the 5th cycle. The results obtained are shown in the form of a table (see Table 3).

Table 3. Comparison of optimization results according to the proposed algorithm, according to the deterministic method [34] and real data.

Values of Criteria and Constraints	Deterministic Bath Method	The Proposed Method (ARA + PO)	Real Data (Atyrau Refinery)
Target product yield (gasoline)—riterion $y_1 = f_1(x) = \mu_0^1(x)$, m ³ /h	77.0	79.0	78.5
The octane number of the product $(\tilde{y}_2 = \varphi_1(x))$;	86	87	(86) ^L
Fractional composition of catalyzate; 10% distillation, oC, $(\tilde{y}_3 = \varphi_2(x))$; 50% distillation, oC, $(\tilde{y}_4 = \varphi_3(x))$.	70 115	70 114	(70) ^L (114) ^L
MF of constraint fulfillment $\tilde{y}_2 = \mu_1(x^*(\beta))$;	-	1.0	-
MF of constraint fulfillment $\tilde{y}_3 = \mu_2(x^*(\beta))$;	-	1.0	-
MF of constraint fulfillment $\tilde{y}_4 = \mu_3(x^*(\beta))$.	-	0.98	-
Optimal values of input and operating parameters $x^* = (x_1^*, x_2^*, x_3^*, x_4^*, x_5^*)$: x_1^* —loading of raw materials; m ³ /h	80	80	80
x_2^* —volume velocity in reactors; hour ⁻¹	1.7	1.3	1.5
x_3^* —temperature in reactors P-4.4a; °C	500	493	495
x_4^* —pressure in the reactors P-4.4a; kg/cm ²	26	25	25
x_5^* —the ratio of hydrogen/hydrocarbons	415	400	400

Note: MF—membership function; (L) means that the corresponding quality indicators are determined by laboratory methods with the participation of a person and require sufficient time; (-) means that the corresponding indicators are not determined by this method. The search time for a solution in the compared methods is almost the same: about one minute, taking into account the time of entering or correcting the required data.

4. Discussion

In order to assess and select the most effective type of model for each unit of the cold-weather system, according to Table 1, a system analysis is carried out according to the selected criteria of possible types of models for each unit based on the methods of expert evaluation. The assessments of each type of model for each unit are carried out on a point scale. Therefore, by simply summing the assessments of each type of model, it is possible to determine the values of the integrated criterion. The most efficient type of model for each CES unit is determined by the maximum value of the integrated criterion.

If it is impossible to assess on a point scale, the assessments can be made unclear, using the terms “less”, “more”, etc. In that case, it is necessary to construct the membership functions of these terms and, on the basis of the rules of fuzzy set theories, to choose the most effective type of model.

As can be seen from clauses 5–9 of the methodology for developing models of interconnected CES units proposed in Section 3.1, models of each unit can be built using various initial information and appropriate methods for developing models. As a result, various types of models can be built, for example, statistical, fuzzy, or combined. Then these models, in accordance with the process flow scheme, are combined into a single system (package) of models, simulation results of one aggregate are taken as input data for models of other aggregates. For example, the models of the main units of the catalytic reforming

unit LG-35-11/300-95 of the Atyrau refinery are combined according to the scheme shown in Figure 3.

The main advantages of the results of the study include:

- The created methodology for developing models of interrelated technological CES units allows developing more efficient models of individual units in conditions of deficiency and fuzziness of initial information based on available information of a different nature, combining them into a single package of models and systematically modeling and optimizing the operating modes of CES units. The proposed method for developing models of interrelated technological CES units can be used in the development of mathematical models of CES of various industries under conditions of uncertainty and fuzziness of the initial information;
- since in the developed models of reforming reactors, in addition to experimental and statistical data, theoretical information, fuzzy information is also used, representing the knowledge, experience, and intuition of decision makers and experts, the resulting models take into account deep causal relationships, and they are more adequate and efficient;
- The formulated mathematical formulation of the fuzzy problem of multi-objective optimization of the operating modes of reforming reactors and the developed heuristic method for its solution are based on the modification and adaptation of various optimality principles for operation in a fuzzy environment. The originality and novelty of such a fuzzy approach to solving a fuzzy problem of multicriteria optimization from known methods for solving fuzzy problems by converting them to a set of clear problems based on a set of level α , which leads to the loss of a significant part of the original fuzzy information, is that the problem is posed and solved in fuzzy environment without converting it to clear tasks. Thus, in the proposed fuzzy approach to solving the problem of multicriteria optimization in a fuzzy environment, the collected initial fuzzy information, i.e., the knowledge, experience, and intuition of experts, is used to the maximum. This makes it possible to obtain more adequate and efficient solutions for production problems that are characterized by fuzziness. The results obtained confirm the effectiveness of the fuzzy approach to solving problems of optimizing the operating modes of a CES with fuzzy initial information.

The simulation results based on known models and developed models, taking into account fuzzy information and real data from the LG-35-11/300-95 unit of the Atyrau Oil Refinery presented in Table 2, show that the simulation results based on the developed models they are compared with are the best. This can be explained by the fact that when building models using fuzzy information, knowledge, wholesale and intuition of a human operator, decision maker, experts, deep causal relationships between various non-formalized input, and output parameters of the object are taken into account.

The advantages of the developed reactor models are that they make it possible to determine a more efficient operating mode of the reactors. The effectiveness of such a regime is that the percentage of the target product yield, the volume of catalyzate from the reactors increases, and this more accurately corresponds to the real data. Thus, we can conclude that the developed models are more adequate and efficient as compared to deterministic models in the conditions of a deficit and indistinctness of the initial information. In addition, the developed models make it possible to determine quality indicators, the octane number of the catalyzate and its fractional composition, which are characterized by indistinctness.

The formulated mathematical formulation of the problem of optimizing the operating modes of reforming reactors based on the models of a block of reactors of a reforming unit in conditions of multicriteria and fuzziness (7), (8) is concretized for the case of one criterion (gasoline yield) and three fuzzy restrictions. Fuzzy constraint $\varphi_q(x) \gtrsim b_q$, $q = \overline{1,3}$ allows one to take into account the quality of gasoline (octane number and fractional composition) in the process of maximizing the volume of gasoline produced. In the case of ambiguity of criteria or weighting factors, it is necessary to define a term-set and build membership functions that allow taking into account their fuzziness. In a concrete formulation of the

optimization problem (11), (12) in the form of a decision-making problem, the criterion is presented in a normalized form.

This makes it possible to ensure the convenience of applying the methods of fuzzy sets, since the criterion, like the membership function, takes on a value in the interval $[0,1]$. The value of the criterion is determined on the basis of mathematical models of the reforming reactor, constructed in [31], taking into account the vagueness of the initial information.

As a result of the analysis and discussion of the results given in Table 1, the following advantages of the results obtained can be noted:

- the proposed heuristic method is more efficient than the deterministic method;
- when solving a control problem based on the proposed method, the adequacy of the solution of the production problem increases, since additional qualitative information (experience, knowledge of the decision maker, experts) is taken into account, which more fully describes the real situation without idealization;
- The proposed and used heuristic method allows determining and taking into account the degree of fulfillment of fuzzy constraints.

The proposed heuristic method based on the modification of the principles of absolute (relative) concession and Pareto optimality is more efficiently applied in the case of the possibility of determining the concession and the Pareto optimality principle (the number of objects in the interval 7 ± 2). In other situations, it is recommended to set and solve the problem on the basis of other optimality principles that are more suitable for the situation in production.

As the main directions of future research, one can single out expanding the base of methods for solving decision-making problems for optimizing operating modes in a fuzzy environment of a technological unit based on a modification and a combination of other optimality principles, and also creation on the basis of the developed models and methods for solving the problem of multicriteria optimization in a fuzzy environment of an intellectualized decision support system for controlling the operating modes of the research object.

5. The Major Contributions of the Work

Consider the main results and the main contribution of this work.

1. A technique has been created for development of models of interconnected technological units for system modeling of CES based on initial information of a different nature. The proposed technique is described on the example of models of the main technological units of a catalytic reforming unit;
2. Models of reforming reactors were developed based on the use of available experimental and statistical data and fuzzy information from a human operator, decision maker, subject matter experts, and using the proposed methodology;
3. A mathematical statement of the problem of multicriteria optimization of the operating modes of reforming reactors in a fuzzy environment is formalized and obtained. Based on the modification of the principles of absolute or relative concession and Pareto optimality, a heuristic method for solving the formulated problem of multicriteria optimization in a fuzzy environment is developed. An example of the practical application of the proposed heuristic method for solving the problem of optimizing the operating modes of the LG-35-11/300-95 catalytic reforming unit of the Atyrau Refinery is given. The results obtained confirm the effectiveness of the fuzzy approach to solving the problems of optimizing the operating modes of the CES with fuzzy initial information.

6. Conclusions

Based on the results obtained, the following conclusions can be drawn:

1. A methodology is proposed for developing models of interrelated technological units of CES in conditions of deficiency and fuzziness of initial information, based on the use of available information of a different nature;
2. Based on the proposed methodology, a system of reforming reactor models was built, taking into account the fuzziness of the initial information;
3. A mathematical statement of the problem of multicriteria optimization of the operating modes of reforming reactors in a fuzzy environment is formulated and a heuristic method for its solution based on the modification of various optimality principles is developed.

The developed models of reforming reactors and optimization methods make it possible to optimize the operating modes of the reforming unit in conditions of fuzzy initial information. This is an important consequence of the main results of the study.

The main contribution of the research to science lies in the development of methods for modeling interrelated technological objects of CES and multicriteria optimization in a fuzzy environment. The proposed methods make it possible to develop more efficient models of complex, fuzzy objects and optimize their operation modes based on the maximum use of the initial fuzzy information.

The novelty of the proposed methodology for developing mathematical models of interconnected units of a technological system lies in the application of the methodology of system analysis, methods of expert assessments, and fuzzy set theories, which allow solving problems of uncertainty. The novelty of the developed heuristic method lies in the modification of the principles of optimality used, which allows the maximum use of the knowledge, experience, and intuitions of decision makers, the experts in the decision-making process in a fuzzy environment.

The limitation of the research results is that in order to apply the proposed methodology for developing models of interrelated CES units and the heuristic method for solving the problem of multi-criteria optimization of their operating modes, it is necessary to have experienced decision makers, subject matter experts, organization, and conduct expert evaluation, which requires time.

The direction of future research based on the results obtained is to create an intelligent decision support system to control the operating modes of the objects under study.

The highlight of the work is that in the proposed fuzzy approach to solving the optimization problem in a fuzzy environment, unlike the known approaches, the original fuzzy problem is not replaced by deterministic ones, which allows maximum use of fuzzy information. This allows the user to significantly increase the adequacy and efficiency of the decision in a fuzzy environment, which often occurs in production.

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Appendix A. Results of Parametric Identification of Reformer Models (1)–(4)

$$y_1^{R_2} = 0.398481x_1 + 12.153846x_2 - 0.032113x_3 - 0.983750x_4 + 0.019750x_5 + 0.004937x_1^2 + 9.349112x_2^2 - 0.000065x_3^2 - 0.03792x_4^2 + 0.000493x_5^2 + 0.227885x_1x_2 + 0.00010x_1x_3 + 0.001975x_1x_4 + 0.000495x_1x_5 + 0.003754x_2x_3 - 0.486154x_2x_4 - 0.000643x_3x_4;$$

$$y_1^{R_3} = 0.3950x_1 + 12.1076923x_2 - 0.0186235x_3 - 0.975880x_4 + 0.0196750x_5 + 0.0050441x_1^2 + 8.9547088x_2^2 - 0.0000645x_3^2 - 0.0498958x_4^2 + 0.0000492x_5^2 + 0.2388874x_1x_2 + 0.0000101x_1x_3 + 0.0020835x_1x_4 + 0.0005172x_1x_5 + 0.0358732x_2x_3 - 0.5044872x_2x_4 - 0.0006638x_3x_4;$$

$$y_1^{R_{4,4a}} = 0.3989835x_1 + 11.1869231x_2 - 0.0315895x_3 - 1.0239130x_4 + 0.0253700x_5 + 0.0050697x_1^2 + 9.2899408x_2^2 - 0.0000585x_3^2 - 0.044518x_4^2 + 0.0000491x_5^2 + 0.2301828x_1x_2 + 0.0001003x_1x_3 + 0.0021684x_1x_4 + 0.0004988x_1x_5 + 0.0364498x_2x_3 - 0.5250836x_2x_4 - 0.0006867x_3x_4.$$

$$y_3 = 500.0000x_1 + 7142.8571x_2 + 10.10100x_3 - 1458.3333x_4 + 25.000x_5 + 6.2500x_1^2 + 5102.0408x_2^2 + 0.0204x_3^2 - 60.7639x_4^2 + 0.0625x_5^2 + 178.5714x_1x_2 + 0.2525x_1x_3 - 15.6250x_1x_4 + 15.6345x_1x_5 - 297.6190x_2x_4 - 2.5252x_3x_4 - 0.05051x_3x_5 - 1.0417x_4x_5$$

Appendix B. Results of Parametric Identification of Fuzzy Models (5) That Evaluate the Quality Indicators of Catalyzate on Selected Sets of Level α

$$y_3 = 500.0000x_1 + 7142.8571x_2 + 10.10100x_3 - 1458.3333x_4 + 25.000x_5 + 6.2500x_1^2 + 5102.0408x_2^2 + 0.0204x_3^2 - 60.7639x_4^2 + 0.0625x_5^2 + 178.5714x_1x_2 + 0.2525x_1x_3 - 15.6250x_1x_4 + 15.6345x_1x_5 - 297.6190x_2x_4 - 2.5252x_3x_4 - 0.05051x_3x_5 - 1.0417x_4x_5$$

$$\begin{aligned} \tilde{y}_4 = & \left(\frac{0.5}{0.430000} + \frac{0.75}{0.433000} + \frac{1}{0.435000} + \frac{0.75}{0.437000} + \frac{0.5}{0.440000} \right) x_{14} \left(\frac{0.5}{20.076906} + \frac{0.75}{20.076916} + \frac{1}{20.076923} \right. \\ & + \left. \frac{0.75}{20.076930} + \frac{0.5}{20.076938} \right) x_{24} - \left(\frac{0.5}{0.052810} + \frac{0.75}{0.052824} + \frac{1}{0.052834} + \frac{0.75}{0.052844} + \frac{0.5}{0.052858} \right) x_{34} - \\ & - \left(\frac{0.5}{0.724870} + \frac{0.75}{0.724950} + \frac{1}{0.720000} + \frac{0.75}{0.725050} + \frac{0.5}{0.725130} \right) x_{44} + \left(\frac{0.5}{0.042209} + \frac{0.75}{0.042339} + \frac{1}{0.042439} + \right. \\ & + \left. \frac{0.75}{0.042539} + \frac{0.5}{0.042669} \right) x_{54} + \left(\frac{0.5}{0.005198} + \frac{0.75}{0.005328} + \frac{1}{0.005438} + \frac{0.75}{0.005548} + \frac{0.5}{0.005688} \right) x_{14}^2 - \\ & - \left(\frac{0.5}{15.443467} + \frac{0.75}{15.443637} + \frac{1}{15.446787} + \frac{0.75}{15.443937} + \frac{0.5}{15.443112} \right) x_{24}^2 + \left(\frac{0.5}{0.030058} + \frac{0.75}{0.030138} + \frac{1}{0.030138} + \right. \\ & + \left. \frac{0.75}{0.030278} + \frac{0.5}{0.030358} \right) x_{44}^2 + \left(\frac{0.5}{0.000004} + \frac{0.75}{0.000054} + \frac{1}{0.000104} + \frac{0.75}{0.000154} + \frac{0.5}{0.000224} \right) x_{54}^2 + \\ & + \left(\frac{0.5}{0.000100} + \frac{0.75}{0.000170} + \frac{1}{0.000220} + \frac{0.75}{0.000270} + \frac{0.5}{0.000340} \right) x_{14}x_{34} + \left(\frac{0.5}{0.000125} + \frac{0.75}{0.000205} + \frac{1}{0.000265} + \right. \\ & + \left. \frac{0.75}{0.000325} + \frac{0.5}{0.000405} \right) x_{14}x_{54} - \left(\frac{0.5}{0.557242} + \frac{0.75}{0.557492} + \frac{1}{0.557692} + \frac{0.75}{0.557892} + \frac{0.5}{0.558142} \right) x_{24}x_{44} + \\ & + \left(\frac{0.5}{0.000006} + \frac{0.75}{0.000046} + \frac{1}{0.000086} + \frac{0.75}{0.000126} + \frac{0.5}{0.000166} \right) x_{34}x_{54}; \end{aligned}$$

$$\begin{aligned} \tilde{y}_5 = & \left(\frac{0.5}{0.406050} + \frac{0.75}{0.406150} + \frac{1}{0.406250} + \frac{0.75}{0.406400} + \frac{0.5}{0.406600} \right) x_{15} - \left(\frac{0.5}{9.285214} + \frac{0.75}{9.285514} + \frac{1}{9.285814} + \right. \\ & + \left. \frac{0.75}{9.285914} + \frac{0.5}{9.286214} \right) x_{25} - \left(\frac{0.5}{0.065793} + \frac{0.75}{0.065873} + \frac{1}{0.065923} + \frac{0.75}{0.065973} + \frac{0.5}{0.066053} \right) x_{35} - \\ & - \left(\frac{0.5}{0.541417} + \frac{0.75}{0.541567} + \frac{1}{0.541667} + \frac{0.75}{0.541767} + \frac{0.5}{0.541917} \right) x_{45} - \left(\frac{0.5}{0.015849} + \frac{0.75}{0.015979} + \frac{1}{0.016049} + \right. \\ & + \left. \frac{0.75}{0.016119} + \frac{0.5}{0.016249} \right) x_{55} + \left(\frac{0.5}{0.004978} + \frac{0.75}{0.005048} + \frac{1}{0.005108} + \frac{0.75}{0.005178} + \frac{0.5}{0.005078} \right) x_{15}^2 - \\ & - \left(\frac{0.5}{6.6325961} + \frac{0.75}{6.6326331} + \frac{1}{6.6326531} + \frac{0.75}{6.6326731} + \frac{0.5}{6.6327101} \right) x_{25}^2 + \left(\frac{0.5}{0.000053} + \frac{0.75}{0.000103} + \frac{1}{0.000133} + \right. \\ & + \left. \frac{0.75}{0.000163} + \frac{0.5}{0.000313} \right) x_{35}^2 - \left(\frac{0.5}{0.022179} + \frac{0.75}{0.022449} + \frac{1}{0.022569} + \frac{0.75}{0.022689} + \frac{0.5}{0.022959} \right) x_{45}^2 - \\ & - \left(\frac{0.5}{0.000009} + \frac{0.75}{0.000029} + \frac{1}{0.000039} + \frac{0.75}{0.000049} + \frac{0.5}{0.000069} \right) x_{55}^2 + \left(\frac{0.5}{0.000428} + \frac{0.75}{0.000589} + \frac{1}{0.000659} + \right. \\ & + \left. \frac{0.75}{0.000729} + \frac{0.5}{0.000878} \right) x_{15}x_{35} - \left(\frac{0.5}{0.386185} + \frac{0.75}{0.386655} + \frac{1}{0.386905} + \frac{0.75}{0.387155} + \frac{0.5}{0.387625} \right) x_{25}x_{45} - \\ & - \left(\frac{0.5}{0.011015} + \frac{0.75}{0.011314} + \frac{1}{0.011464} + \frac{0.75}{0.011614} + \frac{0.5}{0.011915} \right) x_{25}x_{55} - \left(\frac{0.5}{0.000477} + \frac{0.75}{0.000599} + \right. \\ & + \left. \frac{1}{0.000669} + \frac{0.75}{0.000739} + \frac{0.5}{0.000857} \right) x_{45}x_{55}. \end{aligned}$$

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