

Hydrotreating unit models based on statistical and fuzzy information

Alua Tanirbergenova^{1*}, Batyr Orazbayev¹, Yerbol Ospanov², Sholpan Omarova³, Ildar Kurmashev⁴

¹ Department of System Analysis and Control, L.N. Gumilyov Eurasian National University

² Department of Automation and Information Technology, Shakarim University

³ Department of Digital Engineering and IT Analytics, Karaganda University of Kazpotreboyozyuz

⁴ Department of Informatization of Education, Manash Kozybayev North Kazakhstan University

ABSTRACT

This paper presents the results of mathematical models' development for hydrotreating reactor, stripping column, absorbers and hydrotreating furnace which are basic units of hydrotreating block in catalytic reforming unit. Since these objects of modeling in reforming unit of Atyrau refinery operate in conditions of insufficiency and fuzziness of the initial information, their mathematical models are developed on the basis of a systematic approach, using available information of different nature (experimental-statistical data, fuzzy information from the experts), with appropriate methods of construction for mathematical models. Mathematical models, describing the dependence of the production output from the hydrotreating reactor, columns and furnace, are designed as a nonlinear regression models based on experimental-statistical data. Whereas models, evaluating the quality indicators of generated products from the hydrotreating reactor and columns, i.e., hydrogenation, hydrogen-containing and hydrocarbon-containing gases are built based on fuzzy information from specialists-experts in the form of fuzzy multiple regression equations. We have plotted the graph for the dependence of hydrogenation products output on the temperature in the hydrotreating reactor. To describe the dependence of the optimum temperature value in hydrotreating process on raw material quality, a linguistic model is designed based on compositional rules of inference and fuzzy information. Membership functions of fuzzy parameters are constructed for linguistic models.

Keywords: Hydrotreating, Hydrotreater, Hydrogenation, Hydrotreating reactor, Mathematical model, Fuzzy information, Membership function, Fuzzy model.

Corresponding Author:

Alua Tanirbergenova
Department of System Analysis and Control
L.N. Gumilyov Eurasian National University
010008, 2 Satpayev Str., Nur-Sultan, Republic of Kazakhstan
E-mail: a.tanirbergenova7069@nuos.pro

1. Introduction

In this study, authors discuss in more detail the first direction of improvement for hydrotreating processes, based on hydrotreating unit of catalytic reforming installation in Atyrau refinery. There are known research works on mathematical modeling and optimization of technological objects and processes of oil refining, including hydrotreating process [1-18]. However, in practice, production situations can often be associated with the of insufficiency and fuzziness of the initial information, therefore the problems of modeling and optimization of their operation modes, formulation and solutions using traditional methods do not provide adequate solutions These objects include hydrotreating unit LH in catalytic reforming of Atyrau refinery, basic units of which operate in the conditions of uncertainty associated with the randomness and fuzziness of initial information [19-26].

In order to create a mode optimization and control system of the hydrotreating unit in catalytic reforming

facility, it is necessary to develop a system of mathematical models for main interconnected units of the facility, namely hydrotreating reactor R-1, steam column C-1, absorbers C-2 and C-3, and hydrotreating furnace F-101. In the development of mathematical models of these basic units of hydrotreating facility there may be a problem related to insufficiency, uncertainty and fuzziness of the initial information. The uncertainty can be caused due to deficiency, randomness and fuzziness of the available information, which is necessary for the development of mathematical models of objects. In these cases, one has to use an appropriate approach, such as a hybrid method proposed in [27-38], which allows constructing mathematical models of objects based on the available information of various character.

Let be $\{\bar{x}_i, i = \overline{1, l}\}$ and $\{\bar{x}_i, i = \overline{1, m}\}$ – a set of available input parameters of the object, which is characterized by the probability – \bar{x} and fuzziness \bar{x} – values $\{\bar{x}_i, i = \overline{1, l}\}$ are determined using instrumentation, but are characterized by randomness. Values $\{\bar{x}_i, i = \overline{1, m}\}$ are fuzzy and estimated by human, i.e., specialists-experts based on their knowledge, experience and intuition fuzzily, i.e., by words and phrases. Based on such initial information of the different nature, it is necessary to identify the structure and parameters of models for main units of hydrotreating facility. Consequently, it is required to use the methods of probability theory, experimental and statistical modeling techniques, as well as known methods modified and adapted for operation in the fuzzy environment, for example the method of forward selection in regression, and the method of least squares [39-50], as well as a hybrid method of building models [51,52].

Thus, based on the aforementioned methods, it is necessary to develop a system of mathematical models for hydrotreating reactor R-1, columns C-1, C-2 and C-3, and hydrotreating furnace F-101. For R-1 it is needed to identify equations that enable calculating the values of the output parameters of the reactor: y_1 hydrogenate volume; and evaluating the values of quality indicators of the product: $\bar{y}_2, \bar{y}_3, \bar{y}_4$ – respectively, unsaturated hydrocarbons, sulfur and water-soluble acid and alkali in the composition of hydrogenate. For columns C-1, C-2 and C-3 it is necessary to identify models that enable determining the hydrogenation output from C-1, HCG from C-2 and hydrocarbon-containing gas from C-3, as well as evaluating the basic quality indicators of column products from input parameters of columns. Mathematical models of the hydrotreating furnace F-101 must determine the amount of feed with gas and flow temperature from the exit of furnace depending on values of F-101 input parameters.

The aim of this work is to develop a system of mathematical models for basic units of hydrotreating block in Atyrau refinery catalytic reforming based on the available information of different nature, which can be used in optimizing the process parameters and modes control of hydrotreating unit.

2. Material and methods

Hydrotreating unit of catalytic reforming installation is one of the main units of the installation [53-60]. In the hydrotreating unit, hydrotreating process of straight-run gasoline from crude oil distillation occurs. Accordingly, there is increase in oil quality by removing the sulfur from their composition and as well as other harmful compounds and impurities that degrade the performance characteristics of the process equipment and metal units [61-75]. Thus, the process of hydrotreating reduces corrosion of metal equipment and pollution of environment and atmosphere. Hence, an actual task of technology science and petroleum production is the study and improvement of hydrotreating processes in refinery based on research-based methods, such as methods of mathematical modeling and optimization.

Consider the description of the technological diagram for hydrotreating unit in catalytic reforming of Atyrau refinery (AR) shown in Figure 1. The catalytic reforming process is designed to produce high octane motor gasoline, feedstock for petrochemical synthesis (aromatic hydrocarbons) and for hydrogen-containing gas (HCG), which are used in hydrogenation processes in refining. The target product of the catalytic reforming LH-35-11 / 300-95 in Atyrau refinery are high-octane commercial gasoline and liquefied natural gas.

The installation LH consists of 4 units: preliminary hydrotreating unit of feedstock, i.e., straight-run gasoline, produced at the unit of primary oil refining; catalytic reforming unit, which increases the octane number of the hydrotreated gasoline; deethanizer and catalyzate stabilization unit; purification unit of circulating hydrocarbon-containing gases by monoethanolamine (MEA) and regeneration of MEA [76-83].

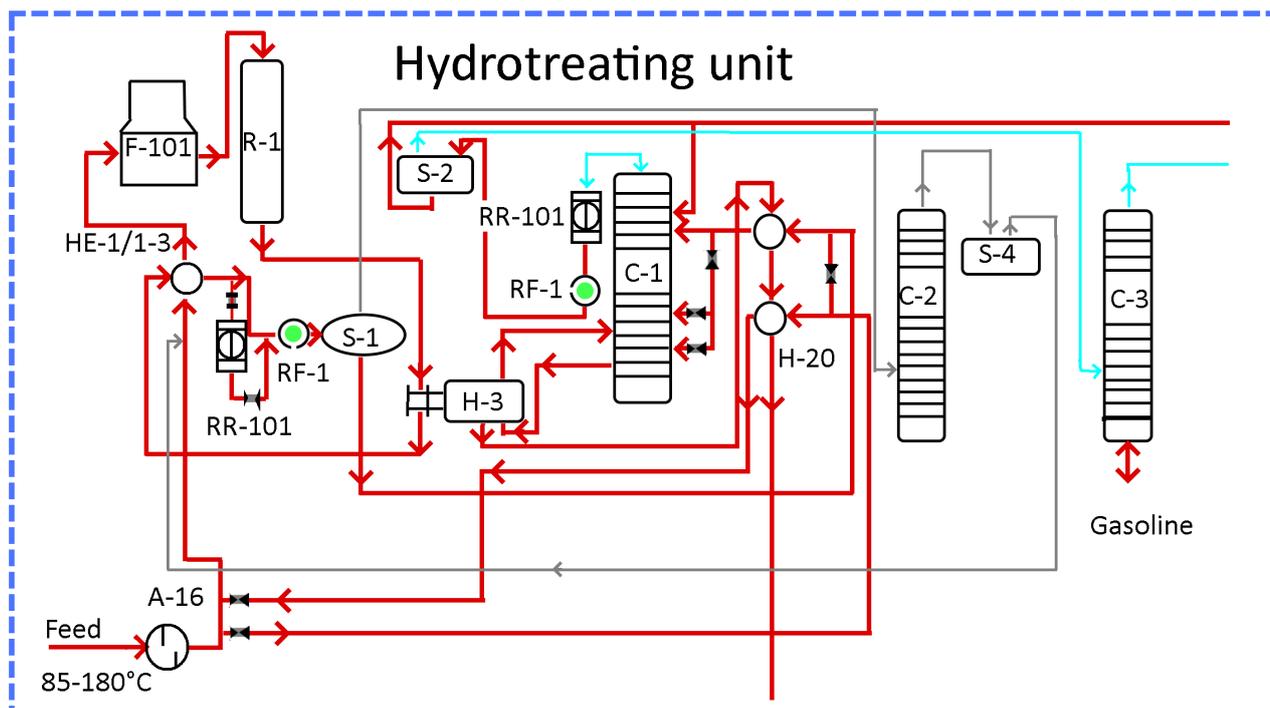


Figure 1. Technological diagram for hydrotreating unit in catalytic reforming of Atyrau refinery

Feedstock for hydrotreating unit LH in Atyrau refinery is straight-run gasoline, produced in primary refining unit AVT ET-3. Hydrotreating process proceeds in HCG environment and it relates to a catalytic process, designed for purification and removal of organic compounds of sulfur, oxygen and nitrogen, which are poison for the catalyst, from the feedstock and from straight-run gasoline. Feed from reservoir tank is supplied with HCG by A16 pump for mixing. The mixture of feed and HCG is supplied to serially connected 3 heat exchangers HE-1/1-3, here due to the counterflow of aerated feed from the reactor R-1 and HE-3 reboiler is heated to a temperature of 260 °C, then further supplied to the hydrotreating furnace F-101. From furnace F-101, the mixture of feed and gas with a temperature of 300-343 °C is fed to hydrotreating reactor R-1. Hydrotreating reaction proceeds in the reactor R-1 with use of S-12 catalyst, i.e., feed is preliminarily hydrorefined from sulfur, nitrogen and oxygen containing compounds. Heat of the mixture of unstable hydrogenate and circulating gas from the reactor output and heat of the gas reaction at a temperature 340-420 °C are used for heating the mixture of feed and gas firstly in heat exchanger HE-3 of stripping column SC, then in heat exchangers HE-1/1-3 [84-90].

Products in the form of gas, after cooling to 35 °C temperature in refrigerators RR-101 and RF-1 are supplied to the separator S-1. HCG is separated from the liquid in S-1 and is fed to the absorber C-2 for removal of hydrogen. Gas from the exit of the absorber C-2 after passing through the separator S-4 is divided into two streams:

- 1) circulating gas, after compression in the compressor is fed back to the system of feedstock hydrotreating;
- 2) excess HCG from the exit of unit, the liquid phase of separator S-1 passes through the heat exchanger HE-2, where it is heated to a temperature of 150 °C and fed to 7, 9, 23 tray of stripping column C-1. Hydrogen sulfide and water are stripped from C-1 column, which has 30 trays, from hydrogenate at temperatures up to 270 °C and pressures up to 15 atm, in addition to that light hydrocarbon are discharged through the top of column.

After stripping column C-1, overall composition of sulfur compounds in hydrogenate must not exceed 0.0005 wt%. Gases in the vapor state from the top of column C-1 are out at a temperature of 135 °C, they pass through refrigerators RR-101 and RF-1 and fed into the separator S-2 with a temperature of 35-40 °C. From the separator S-2, it is fed back to stripping column C-1 in liquid phase. Precipitated water in the separator S-2 is discharged into the sewers. Hydrocarbon gas from the separator S-2 enters the absorber C-3 for purifying from hydrogen sulfide. Hydrocarbon gas from the top of the absorber C-3 is fed into the fractionating absorber C-6 or the fuel tank of factory. Thus, during hydrotreating process, chemical transformation of substances occurs under the influence of hydrogen gas at high pressure and high temperature [91]. During the

hydrotreating process, sulfur compounds are reduced in petroleum products, additional unsaturated hydrocarbons are saturated, resin composition and oxygenate-containing are lowered and hydrocracking of hydrocarbon molecules are done. Improvement of hydrotreating processes in refinery using modeling techniques allows the following [92]: conducting hydrotreating processes in the optimal mode; improving the quality indicators of generated products.

3. Results and discussion

To solve this problem, i.e., to develop a system of mathematical models for the main units of the hydrotreating facility, which determine the dependence of the output parameters of the unit (product, quality) on the input mode parameters using available information of various character. For example, to develop mathematical models of hydrotreating reactor that enable determining the amount of hydrogenate from the output of the hydrotreating reactor R-1, authors use experimental-statistical data, characterized by probability and expert information, having fuzzy nature [93; 94; 95-110]. For structural identification of reactor R-1 models, the idea of forward selection in regression, whereas identification of the model parameters is based on a modified method of least squares. Thus, the mathematical models for the reactor R-1 of hydrotreating unit are designed using statistical data and fuzzy information processed by methods of mathematical statistics and the theory of fuzzy sets. Fuzzy information is collected and formalized by the methods of expert assessment and fuzzy set theory. Based on processing of experimental-statistical data and expert information, as well as by a method for constructing the fuzzy model [111; 112-121], authors conduct structural identification of models that describe the quality of products from hydrotreating reactor R-1 in the form of the following fuzzy equations of multiple regression:

$$\hat{y}_j = a_{0j}x_{ij} + \sum_{i=1}^5 a_{ij}x_{ij} + \sum_{i=1}^5 \sum_{k=i}^5 \bar{a}_{ijk}x_{ij}x_{kj}, \quad j = \overline{2,4}, \quad (1)$$

where: \hat{y}_2 – unsaturated hydrocarbons in the composition of the product, i.e. hydrogenate (not more than, i.e. $\leq 1\%$); \hat{y}_3 – sulfur in hydrogenate content ($\leq 0.00005\%$); \hat{y}_4 – water-soluble acids and alkali in hydrogenate content ($\cong 0\%$); x_1 – feed, in this case, straight-run gasoline (45-80 m³/hr.); x_2 – reactor pressure (20-35 kg/cm²); x_3 – reactor temperature (300-343 °C); x_4 – rate of feed supply (0.5-5 h⁻¹); x_5 – circulating hydrogen-containing gases (HCG) – ratio hydrogen/ hydrocarbons (200-500 nm³); $\bar{a}_{0j}, \bar{a}_{ij}, \bar{a}_{ijk}$, $i = \overline{1,5}$ – identified fuzzy coefficients, free term; linear effect; quadratic effect and mutual effect, respectively. The allowable values of the output fuzzy parameters, and intervals of changing input and operating parameters are denoted in the parenthesis [122].

To identify the unknown parameters (regression coefficients) of the model (1): \bar{a}_{ij} ($i = \overline{0,5}$), $j = \overline{2,5}$ and \bar{a}_{ijk} ($i, k = \overline{1,5}$), $j = \overline{2,5}$ – membership functions of fuzzy sets describing the quality of hydrogenate are distributed to the following level set α : $\alpha = 0.5; 0.85; 1$. Since in this case the membership function has the bell-shaped form, values of fuzzy parameters are obtained at 5 points $\alpha = 0.5; 0.85$ (left); $1; 0.85; 0.5$ (right). Values of the input, regime $x_{ij}, i, j = \overline{1,5}$ and output $\hat{y}_2, \hat{y}_3, \hat{y}_4$ parameters are observed for each selected α level. Thus, the obtained models, describing the product quality from the exit of reactor R-1 in the form of multiple regression for each α level. Since the obtained equations have a form of regression equations, the problem of identification of the unknown coefficients $\alpha_{ij}^{\alpha q}, i = \overline{0,5}, j = \overline{2,4}, q = \overline{1,3}$ can be resolved using known parametric identification methods, for example by the method of least squares. In this work to identify the regression coefficients authors use REGRESS program package, which is based on modified least squares method to determine the regression coefficients of the linear and nonlinear regression models with any number of input parameters $x_i, i = \overline{1, n}$.

In this manner, after parametric identification, mathematical models, describing the effect of input, mode parameters $x_i, i = \overline{1, n}$ on the quality of the hydrogenate, i.e., on the content of unsaturated hydrocarbons (\hat{y}_2), sulfur (\hat{y}_3) and water-soluble acids and alkalis (\hat{y}_4) for each α level have the following form:

$$\begin{aligned}
 y_2 = & \left(\frac{0.5}{0.05} + \frac{0.85}{0.07} + \frac{1}{0.08} + \frac{0.85}{0.09} + \frac{0.5}{0.095} \right) - \left(\frac{0.5}{0.00215} + \frac{0.85}{0.0029} + \frac{1}{0.00324} + \frac{0.85}{0.00375} + \frac{0.5}{0.00425} \right) x_1 + \\
 & \left(\frac{0.5}{0.00591} + \frac{0.85}{0.00592} + \frac{1}{0.00593} + \frac{0.85}{0.00594} + \frac{0.5}{0.00595} \right) x_2 + \left(\frac{0.5}{0.0002} + \frac{0.85}{0.0005} + \frac{1}{0.0007} + \frac{0.85}{0.00095} + \frac{0.5}{0.0013} \right) x_3 + \\
 & \left(\frac{0.03125}{0.5} + \frac{0.04333}{0.85} + \frac{0.05333}{1} + \frac{0.06333}{0.85} + \frac{0.07333}{0.5} \right) x_4 + \left(\frac{0.0004}{0.5} + \frac{0.0005}{0.85} + \frac{0.0006}{1} + \frac{0.0007}{0.85} + \frac{0.0008}{0.5} \right) x_5 - \\
 & \left(\frac{0.00002}{0.5} + \frac{0.00003}{0.85} + \frac{0.00004}{1} + \frac{0.00005}{0.85} + \frac{0.00007}{0.5} \right) x_1^2 + \left(\frac{0.00021}{0.5} + \frac{0.00022}{0.85} + \frac{0.00023}{1} + \frac{0.00024}{0.85} + \frac{0.00025}{0.5} \right) x_2^2 + \\
 & \left(\frac{0.00012}{0.5} + \frac{0.00018}{0.85} + \frac{0.00023}{1} + \frac{0.00028}{0.85} + \frac{0.00033}{0.5} \right) x_3^2 - \left(\frac{0.01675}{0.5} + \frac{0.01727}{0.85} + \frac{0.01777}{1} + \frac{0.01713}{0.85} + \frac{0.01818}{0.5} \right) x_4^2 + \\
 & \left(\frac{0.000008}{0.5} + \frac{0.000014}{0.85} + \frac{0.00002}{1} + \frac{0.00003}{0.85} + \frac{0.00005}{0.5} \right) x_5^2 - \left(\frac{0.0003}{0.5} + \frac{0.00035}{0.85} + \frac{0.0004}{1} + \frac{0.00045}{0.85} + \frac{0.0005}{0.5} \right) x_1 x_2 + \\
 & \left(\frac{0.000024}{0.5} + \frac{0.00003}{0.85} + \frac{0.000033}{1} + \frac{0.00004}{0.85} + \frac{0.000047}{0.5} \right) x_1 x_3 - \left(\frac{0.00068}{0.5} + \frac{0.0007}{0.85} + \frac{0.00073}{1} + \frac{0.00075}{0.85} + \frac{1}{0.5} \right) + \\
 & \frac{0.00077}{0.85} + \frac{0.5}{0.5} x_1 x_4 + \left(\frac{0.000012}{0.5} + \frac{0.000019}{0.85} + \frac{0.000027}{1} + \frac{0.000035}{0.85} + \frac{0.000043}{0.5} \right) x_1 x_5 - \left(\frac{0.00083}{0.5} + \frac{0.0009}{0.85} + \frac{0.00098}{1} + \right. \\
 & \left. \frac{0.0001}{0.85} + \frac{0.0015}{0.5} \right) x_2 x_4 + \left(\frac{0.000005}{0.5} + \frac{0.000006}{0.85} + \frac{0.000007}{1} + \frac{0.000008}{0.85} + \frac{0.000009}{0.5} \right) x_2 x_5 - \left(\frac{0.0001}{0.5} + \frac{0.00015}{0.85} + \right. \\
 & \left. \frac{0.00012}{1} + \frac{0.00015}{0.85} + \frac{0.00018}{0.5} \right) x_3 x_5;
 \end{aligned}$$

(2)

$$\begin{aligned}
 y_3 = & \left(\frac{0.5}{0.002} + \frac{0.85}{0.003} + \frac{1}{0.004} + \frac{0.85}{0.005} + \frac{0.5}{0.006} \right) - \left(\frac{0.5}{0.00014} + \frac{0.85}{0.00015} + \frac{1}{0.00016} + \frac{0.85}{0.00017} + \frac{0.5}{0.00018} \right) x_1 + \\
 & \left(\frac{0.5}{0.00027} + \frac{0.85}{0.00028} + \frac{1}{0.00029} + \frac{0.85}{0.0003} + \frac{0.5}{0.00031} \right) x_2 + \left(\frac{0.5}{0.00002} + \frac{0.85}{0.00003} + \frac{1}{0.00004} + \frac{0.85}{0.000045} + \frac{0.5}{0.00005} \right) x_3 + \\
 & \left(\frac{0.00044}{0.5} + \frac{0.0005}{0.85} + \frac{0.00053}{1} + \frac{0.00054}{0.85} + \frac{0.00055}{0.5} \right) x_4 + \left(\frac{0.000002}{0.5} + \frac{0.0000025}{0.85} + \frac{0.000003}{1} + \frac{0.0000035}{0.85} + \frac{1}{0.5} \right) + \\
 & \frac{0.000004}{0.85} + \frac{0.5}{0.5} x_5 - \left(\frac{0.000001}{0.5} + \frac{0.0000015}{0.85} + \frac{0.000002}{1} + \frac{0.0000025}{0.85} + \frac{0.000003}{0.5} \right) x_1^2 + \left(\frac{0.00001}{0.5} + \frac{0.000015}{0.85} + \frac{0.00002}{1} + \right. \\
 & \left. \frac{0.000025}{0.85} + \frac{0.00003}{0.5} \right) x_2^2 + \left(\frac{0.00015}{0.5} + \frac{0.00017}{0.85} + \frac{0.00018}{1} + \frac{0.00019}{0.85} + \frac{0.0002}{0.5} \right) x_4^2 + \left(\frac{0.00002}{0.5} + \frac{0.00003}{0.85} + \frac{0.00004}{1} + \right. \\
 & \left. \frac{0.00005}{0.85} + \frac{0.00006}{0.5} \right) x_1 x_2 + \left(\frac{0.000001}{0.5} + \frac{0.000009}{0.85} + \frac{0.00001}{1} + \frac{0.00002}{0.85} + \frac{0.00003}{0.5} \right) x_1 x_3 - \left(\frac{0.5}{0.00007} + \frac{0.85}{0.00013} + \right. \\
 & \left. \frac{0.00018}{1} + \frac{0.00023}{0.85} + \frac{0.00030}{0.5} \right) x_1 x_4 + \left(\frac{0.00001}{0.5} + \frac{0.00009}{0.85} + \frac{0.00010}{1} + \frac{0.00020}{0.85} + \frac{0.00030}{0.5} \right) x_2 x_3 - \\
 & \left(\frac{0.00038}{0.5} + \frac{0.00044}{0.85} + \frac{0.00049}{1} + \frac{0.00054}{0.85} + \frac{0.00064}{0.5} \right) x_2 x_4 + \left(\frac{0.000002}{0.5} + \frac{0.000003}{0.85} + \frac{0.000004}{1} + \frac{0.85}{0.000005} + \right. \\
 & \left. \frac{0.000006}{0.5} \right) x_3 x_4;
 \end{aligned}$$

(3)

$$\begin{aligned}
 y_4 = & \left(\frac{0.5}{0.00023} + \frac{0.85}{0.00024} + \frac{1}{0.00025} + \frac{0.85}{0.00026} + \frac{0.5}{0.00027} \right) - \left(\frac{0.5}{0.001} + \frac{0.85}{0.0015} + \frac{1}{0.002} + \frac{0.85}{0.0025} + \frac{0.5}{0.003} \right) x_1 + \\
 & \left(\frac{0.5}{0.00024} + \frac{0.85}{0.00032} + \frac{1}{0.00037} + \frac{0.85}{0.00042} + \frac{0.005}{0.5} \right) x_2 - \left(\frac{0.5}{0.00003} + \frac{0.85}{0.00004} + \frac{1}{0.00005} + \frac{0.85}{0.00006} + \frac{0.00007}{0.5} \right) x_3 + \\
 & \left(\frac{0.00659}{0.5} + \frac{0.00664}{0.85} + \frac{0.00667}{1} + \frac{0.00670}{0.85} + \frac{0.00675}{0.5} \right) x_4 + \left(\frac{0.000002}{0.5} + \frac{0.000003}{0.85} + \frac{0.000004}{1} + \frac{0.000005}{0.85} + \frac{1}{0.5} \right) + \\
 & \frac{0.000006}{0.85} + \frac{0.5}{0.5} x_5 - \left(\frac{0.000001}{0.5} + \frac{0.000005}{0.85} + \frac{0.00001}{1} + \frac{0.000015}{0.85} + \frac{0.000020}{0.5} \right) x_1^2 + \left(\frac{0.000207}{0.5} + \frac{0.000215}{0.85} + \frac{0.000222}{1} + \right. \\
 & \left. \frac{0.000230}{0.85} + \frac{0.000330}{0.5} \right) x_4^2 + \left(\frac{0.000001}{0.5} + \frac{0.000005}{0.85} + \frac{0.00001}{1} + \frac{0.000015}{0.85} + \frac{0.000020}{0.5} \right) x_1 x_2 - \left(\frac{0.000005}{0.5} + \frac{0.00001}{0.85} + \right. \\
 & \left. \frac{0.00002}{1} + \frac{0.00003}{0.85} + \frac{0.00004}{0.5} \right) x_1 x_4 + \left(\frac{0.000004}{0.5} + \frac{0.000005}{0.85} + \frac{0.000006}{1} + \frac{0.000007}{0.85} + \frac{0.000008}{0.5} \right) x_2 x_4 - \\
 & \left(\frac{0.000001}{0.5} + \frac{0.000005}{0.85} + \frac{0.000001}{1} + \frac{0.0000015}{0.85} + \frac{0.00002}{0.5} \right) x_3 x_4 + \left(\frac{0.5}{0.000001} + \frac{0.85}{0.000005} + \frac{1}{0.000010} + \frac{0.85}{0.000015} + \right. \\
 & \left. \frac{0.0000020}{0.5} \right) x_4 x_5;
 \end{aligned}$$

(4)

The identified values of the coefficients $\alpha_{ij}^{\alpha q}, i = \overline{0.5}, j = \overline{2.4}, q = \overline{1.3}$ are combined together with the help of the expression below from the fuzzy set's theory [122]:

$$a_{ij} = \bigvee_{\alpha \in [0.5, 1]} \alpha_{ij}^{\alpha q} \text{ or } \mu \tilde{a}_{ij}(a_{ij}) = \text{SUP}_{\alpha \in [0.5, 1]} \min \{ \alpha, \mu \alpha_{ij}^{\alpha q}(a_{ij}) \},$$

where:

$$a_{ij}^{\alpha q} = \{a_i | \mu_{\tilde{a}_{ij}}(a_{ij})\} \tag{6}$$

In the derived models, regressors which have no effect on \tilde{y}_2 , \tilde{y}_3 and \tilde{y}_4 or very little influence are set to zero, i.e., not shown. As a result of conducted study and data processing data it is determined that for finding the volume of product output from the reactor R-1, i.e., the amount of hydrogenate y_1 – on the basis of experimental-statistical data, one can construct a statistical model, which allows estimating the value y_1 from the input and mode parameters $x_i, i = 1,5$ using non-linear regression equations. After identifying the structure and parameters of this model, in the same way to the above approach, a mathematical model that allows calculating the volume of hydrogenate from the exit of reactor R-1 has the following form:

$$y_1 = 7.00 + 0.233x_1 + 0.130x_2 + 0.011x_3 + 2.333x_4 - 0.0175x_5 + \\ +0.0031x_1^2 + 0.0048x_2^2 + 0.00003x_3^2 + 0.7778x_4^2 - 0.00004x_5^2 + \\ +0.0017x_1x_2 + 0.00015x_1x_3 + 0.03111x_1x_4 + 0.00023x_1x_5 + \\ +0.08642x_2x_4 - 0.00065x_2x_5 + 0.00730x_3x_4 \tag{7}$$

Authors have constructed the plot describing the dependence of hydrogenate output on temperature in the reactor x_3 for the fixed input values of feed input and the other mode parameters: x_1, x_2, x_4 and x_5 . (Figure 2). The influence of the other mode parameters x_1, x_2, x_4 and x_5 and their mutual influence on the output parameters of the reactor are considered.

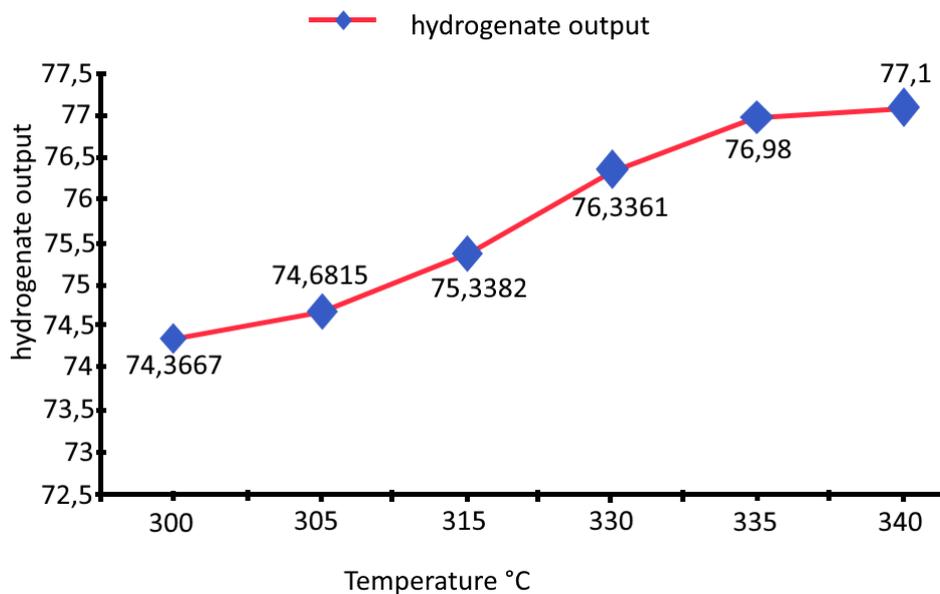


Figure 2. Graph of relationship $y_1 = f_1(x_3)$ for fixed $x_1=80$ m³/hour, $x_2= 30$ kg/cm², $x_4= 3$ hour⁻¹ and $x_5=400$ nm³

The linguistic model is built to determine the optimum temperature of hydrotreating process on the basis of expert information, compositional rules of logical inference and base of rules. The resulting linguistic model has a logic relationship: “If the feed is heavy, then the temperature of process is low, otherwise, if the feedstock is light, the temperature of process is high”. On the basis of expert assessment and using methods of fuzzy sets theory in the form of an exponential dependence, the membership functions are constructed that describe the fuzzy parameters of linguistic model: heavy (low thermal resistance) feed – straight-run gasoline:

$$\mu_A(m) = \exp(-|(ts - 185)^{0.5}|); \tag{8}$$

light feed:

$$\mu_A(ts) = \exp(|(ts - 165)^{0.5}|); \tag{9}$$

low temperature:

$$\mu_B(nt) = \exp(|(nt - 300)^{0.5}|); \tag{10}$$

high temperature:

$$\mu_B(vt) = \exp(|(vt - 400)^{0.5}|). \tag{11}$$

Thus, the structure of linguistic model, evaluating the optimum temperature in dependence on quality of feed:

$$\text{If } \tilde{x}_1 \in \tilde{A}_1 \wedge \tilde{x}_2 \in \tilde{A}_2 \wedge \dots \wedge \tilde{x}_n \in \tilde{A}_n, \text{ then } \tilde{y}_j \in \tilde{B}_j, j = \overline{1, m}, \tag{12}$$

is determined by compositional rules of logical inference:

$$\text{If } \tilde{x}_1 \in \tilde{A}(ts), \text{ then } \tilde{y} \in \tilde{B}(nt), \text{ else If } \tilde{x} \in \tilde{A}(ls), \text{ then } \tilde{y} \in \tilde{B}(vt). \tag{13}$$

In the obtained linguistic models, the following notations are introduced: *ts*, *ls*, *nt*, *vt* – “heavy feed”; “light feed”; “low temperature” and “high temperature”, respectively; \tilde{x} , \tilde{y} – input and output linguistic variables, which describe the quality of feed and the optimum temperature of hydrotreating process, respectively; \tilde{A}, \tilde{B} – fuzzy subsets that describe \tilde{x} and \tilde{y} .

Let us provide the results of the development of mathematical models of columns C-1, C-2 and C-3, which are also the main units of hydrotreating facility. Stripping column C-1 of hydrotreating unit is intended to separate water vapor and hydrogen sulfide from the product, i.e., of hydrogenate. Content of sulfur compounds in the product must not exceed 0,0005 wt%. Column C-2 of hydrotreating unit is absorber, which is intended for purification from hydrocarbon gases. Whereas, column C-3 of hydrotreating unit is also absorber, where purification of hydrogen sulfide from hydrocarbon gases occurs.

Based on study results of operating modes of columns C-1, C-2, C-3, and data analysis, as well as taking into account additional fuzzy information provided by the specialists-experts to develop mathematical models of these columns, it is decided to use a hybrid method of modeling [123; 124]. Based on research work results of columns C-1, C-2 and C-3 and the results of expert assessment, authors identify the following input (*x*) and output (*y*) parameters describing the mode of operation: x_1 – volume of feed at the inlet of the columns; x_2 – inlet temperature; x_3 – pressure in the columns; x_4 – supply rate in column C-1; y_1 – volume of hydrogenate from the exit of column C-1; y_2 – volume of HCG from the exit of column C-2; y_3 – volume of hydrocarbon-containing gases from the exit of column C-3; y_4 – content of sulfur compounds in hydrogenate, quality of C-1 product; y_5 – content of HCG, quality of C-2 product; y_6 – content of hydrocarbon-containing gases, quality of C-3 product.

Input parameters for columns x_1, x_2, x_3, x_4 , i.e., feed consumption, inlet temperature, pressure in the columns and the supply rate for column C-1 and the output parameters y_1, y_2, y_3 describing the volume of the product at the output of columns are measurable parameters. This means that it is possible to collect statistical data of these parameters: $x_i, i = \overline{1, 4}; y_j, j = \overline{1, 3}$. Whereas, quality indicators of products generated in columns K-1, K-2 and K-3 $y_j, j = \overline{4, 6}$ are described fuzzily. Therefore, the fuzzy quality indicators are estimated by experts, formalized and processed using methods of fuzzy set theory.

Thus, the structure of mathematical models of stripping column C-1, absorbers C-2 and C-3 in hydrotreating facility is identified by the method of forward selection in regression and its modifications in the form of the following nonlinear regression and fuzzy regression equations:

$$y_j = a_{0j} + \sum_{i=1}^4 a_{ij}x_{ij} + \sum_{i=1}^4 \sum_{k=i}^4 a_{ikj}x_{ij}x_{kj}, j = \overline{1, 3}; \tag{14}$$

$$\tilde{y}_j = \tilde{a}_{0j} + \sum_{i=1}^4 \tilde{a}_{ij}x_{ij} + \sum_{i=1}^4 \sum_{k=i}^4 \tilde{a}_{ikj}x_{ij}x_{kj}, j = \overline{4, 6}; \tag{15}$$

Identification of parameters, i.e., regression coefficients $a_{0j}, a_{ij}, a_{ikj}, i = \overline{1,4}; k = \overline{i,4}; j = \overline{1,3}$ of regression models (14) is performed based on the method of least squares using statistical data of object. To identify the fuzzy parameters $\tilde{a}_{0j}, \tilde{a}_{ij}, \tilde{a}_{ikj}, i = \overline{1,4}; k = \overline{i,4}; j = \overline{1,3}$ of fuzzy regression models (15) based on level set α , fuzzy equations are converted to a system of non-fuzzy equations, which are equivalent to the original fuzzy equations. Then similarly to the procedure of identification of fuzzy parameters for reactor R-1 model (1), fuzzy coefficients for different levels of α are identified through the method of least squares criterion to minimize the error between the model and experimental (real) data. To determine the volume of target product from column C-1, i.e., hydrogenate after the parametric identification of the model (14) for $J = 1$, the following results are obtained:

$$y_1 = f_1(x_{11}, x_{21}, x_{31}, x_{41}) = -3.65 + 0.2433x_{11} + 0.0365x_{21} + 1.8250x_{31} - 1.3272x_{41} + 0.0018x_{11}^2 + 0.00009x_{21}^2 - 0.1141x_{31}^2 - 0.0603x_{41}^2 + 0.00041x_{11}x_{21} + 0.02027x_{11}x_{31} + 0.00456x_{21}x_{31} \quad (16)$$

Also, with method of forward selection in regression and least squares, structures (14) and parameters are identified for absorber columns C-2 and C-3, i.e., volumes of HCG (y_2) and hydrocarbon-containing gas (y_3) are determined from absorber C-2 and from the column C-3, respectively:

$$y_2 = f_2(x_{12}, x_{22}, x_{32}) = 84.9999 + 0.2982x_{12} + 2.8333x_{22} - 2.4285x_{32} + 0.0001x_{12}^2 + 0.0944x_{22}^2 - 0.0694x_{32}^2 - 0.0066x_{12}x_{22} - 0.0028x_{12}x_{32} + 0.2428x_{22}x_{32}; \quad (17)$$

$$y_3 = f_3(x_{13}, x_{23}, x_{33}) = 83.4999 + 0.2973x_{13} + 2.7833x_{23} - 7.5909x_{33} - 0.0001x_{13}^2 + 0.0927x_{23}^2 - 0.6901x_{33}^2 - 0.0065x_{13}x_{23} + 0.0090x_{13}x_{33} + 0.7591x_{23}x_{33}. \quad (18)$$

Similarly, to the model (1) for the assessment of quality indicators of hydrogenate $\tilde{y}_2, \tilde{y}_3, \tilde{y}_4$, it is possible to identify the fuzzy parameters of model (15) for evaluation of the product quality from columns K-1, K-2 and K-3: $\tilde{y}_j, j = \overline{1,3}$.

Mathematical models for hydrotreating furnace F-101 of hydrotreating unit. The cylindrical hydrotreating furnace F-101 is intended for heating the hydrotreating product, i.e., hydrogenate to the required temperature. According to the study and analysis results, the following main parameters are identified that influence the operation of the furnace F-101 and hydrotreating process: x_1 – flow rate, volume of feed at the inlet of the furnace F-101 in the range of 60-80 m³ / hour; x_2 – temperature at the inlet of furnace F-101, in the range of 170-190 °C; x_3 – pressure in furnace F-101, in the interval 40-43 kg/cm². As a result of analysis for the collected data, and operation modes study of the hydrotreating furnace, the experimental-statistical method has been selected for the development of its model. Optimum operation mode of furnace can be selected on the basis of the mathematical model, which describes the effect of input variables on the output parameters, i.e., provides information on the thermal operation of the furnace. Mathematical description, which is the basis of the mathematical model, must determine the thermal operation parameters of furnace [125; 126].

The main disadvantage of hitherto used methods for calculating the furnace characteristics is that these methods only determine the integral indices of the heat exchange process, and do not regulate the possibility of the furnace tube heating. In the recent time, the researchers have proposed modeling methods based on theoretical studies to determine local heat transfer values, for example, the zone method. In mathematical terms the idea of the zone method calculation is the replacement of integral-differential equations describing the process of heat exchange with the algebraic equations approximating them with the limited system. By solving the obtained algebraic equations, energy characteristics of heat transfer is defined, i.e., temperature and flows of local zones. For this purpose, furnace research is divided into the limited number of area and volume zones, which have the same radiation properties. This approach to the calculation of the furnace characteristics can provide sufficient accuracy, to improve the accuracy it is necessary to increase the number of zones. However, this process is quite complex and the collection of the necessary information for its application in practice is also difficult.

Simple models are required to model the operation of industrial furnaces in interactive mode and to enable fast acquisition of necessary information and results. For this reason, the analytical method of N.I. Belokon can be used as a basis for modeling algorithm, this method is based on the simultaneous solution of heat transfer and heat balance equations [126]. Regression models are identified to calculate the output parameters of the hydrotreating furnace F-101 based on experimental and statistical data. The law of distribution of random measurements is taken close to the normal distribution, i.e., $M[\varepsilon_j] = 0, D[\varepsilon_j] = G^2 = const, j = \overline{1, m}$.

Thus, the structure of the model, evaluating output of hydrotreating furnace: y_1 – volume of mixture of feedstock and gas, and y_2 – temperature of the outlet flow from the furnace, is identified by the following nonlinear regression equations:

$$y_j = a_{0j} + a_{1j}x_1 + a_{2j}x_2 + a_{3j}x_3 + a_{4j}x_1^2 + a_{5j}x_2^2 + a_{6j}x_3^2 + a_{7j}x_1x_2 + a_{8j}x_1x_3 + a_{9j}x_2x_3 + \varepsilon_j, j = \overline{1, 2} \quad (19)$$

The following notation in the model (19) can be adopted: $a_{ij}, i = \overline{0, 3}, j = \overline{1, 2}$ model parameters, which will be identified, the known method of least squares can be used for their evaluation; x_1, x_2, x_3 – furnace F-101 mode parameters, volume of feed (x_1); inlet temperature of the furnace (x_2) And pressure in the furnace (x_3), respectively. The following are the results of identification of regression coefficients in the model (19) using processed statistical data and using REGRESS program:

$$y_1 = 3.7500 + 0.2922x_1 + 0.0208x_2 - 0.0893x_3 + 0.0025x_1^2 + 0.0001x_2^2 + 0.0021x_3^2 + 0.0011x_1x_2 + 0.0023x_1x_3 + 0.0045x_2x_3; \quad (20)$$

$$y_2 = 17.0000 - 0.2208x_1 + 0.7555x_2 + 0.4047x_3 - 0.0028x_1^2 + 0.0016x_2^2 - 0.0096x_3^2 + 0.0037x_1x_2 + 0.0157x_1x_3 + 0.0045x_2x_3. \quad (21)$$

As a result of system analysis and expert assessment, it has been determined that development of statistical and deterministic models using appropriate methods is the most effective for heat exchangers and separators in hydrotreating unit, respectively. The proposed approach to the development of mathematical models of interrelated technological objects on the basis of the available information allows the development of various models of real technological objects in the conditions of insufficiency and fuzziness of the initial information. The structure of developed mathematical models for main units of hydrotreating facility: reforming reactor R-1, columns C-1, C-2 and C-3, reforming furnace F-101 is identified as non-linear regression equations. The equations, describing the volume of production from units, have the form of conventional multiple regression equations. Whereas, the equations, describing the quality of products from the main units (content of unsaturated hydrocarbons – \bar{y}_2 , sulfur – \bar{y}_3 and water-soluble acids and alkalis – \bar{y}_4 ; quality of products from columns C-1, C-2 and C-3: $\bar{y}_j, j = \overline{4, 6}$) have the form of fuzzy multiple regression equations.

In conditions of fuzziness in both input and output parameters, i.e., when the input and output of hydrotreating reactor are described using linguistic variables, it is proposed to construct the linguistic models based on compositional rules of logical inference. This approach is implemented in constructing the linguistic model describing the dependence of the optimum temperature of the hydrofining process on quality indicators of feedstock. In this case, for the construction of the membership function of fuzzy parameters, it is necessary to select the exponential dependence, which has adjusting coefficients to more adequately describe the function. The generated models can be used when optimizing the process parameters, selecting optimal operation modes and controlling hydrotreating process.

4. Conclusion

This paper provides the research results of hydrotreating process fundamentals, the primary directions of modernization and improvement in refinery hydrotreating are highlighted. The main results are demonstrated in the direction of the hydrotreating processes in the optimal mode based on design and modelling of operation modes of hydrotreating reactor. The mathematical models of the main units in hydrotreating facility of reactor R-1 are obtained; stripping column and absorbers C-1, C-2, C-3; reforming furnace F-101 of catalytic cracking facility in Atyrau refinery are characterized by insufficiency and fuzziness of the original information. To solve the problems with fuzziness of initial information and the development of mathematical

models, it is proposed to use the available information of different nature using the hybrid method of model development. The mathematical models of the main units in hydrotreating facility are based on experimental-statistical data and fuzzy information from specialists-experts. The mathematical models for determining the volume of product from the exit of units are identified as statistical models of regression type. Whereas models, evaluating fuzzily described quality indicators of generated product, are defined in the form of fuzzy equations. Structural identification of developed models is conducted on the basis of the idea of forward selection in regression, whereas parametric identification is implemented by the modified method of least squares using REGRESS software package.

The graph is plotted for dependence of the hydrogenate output on temperature in the hydrotreating reactor R-1 for fixed values of the other mode parameters. Using linguistic compositional rules of inference rules, the linguistic model is built that describes the dependence of the optimum temperature value on the feedstock quality.

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