1. Introduction

The relevance of the problem of sustainable manufacturing in our time is not in doubt. Ensuring the sustainability of chemical production is possible through the implementation of the twelve principles of green chemistry, among which the ninth principle reads: «Catalytic processes (whenever possible the most selective) should always be preferred». Currently, approximately 90 % of industrial chemical and petrochemical industries use catalysts. Finding the best catalyst for a particular process is no easy task. Therefore, the presence of intelligent computer systems which can help in finding the best solution for choosing a catalyst is always relevant. That is why a study is conducted, the result of which is a decision support system for choosing a catalyst for the methane oxidative condensation [1].

2. The object of research and its technological audit

The object of research is the catalytic process of oxidative condensation of methane (OCM) and intelligent analysis and decision-making technologies to select the best catalyst option. Many studies are devoted to the problem of choosing catalysts that are most suitable for use in this particular process. First of all, the catalysts must have high activity and selectivity for the desired products. However, the catalysts must exhibit good stability and maintain their own activity and selectivity over a long period of time. Finally, the catalysts should be characterized by such physical properties that allow them to work in production with minimal losses. Often, combinations of catalysts must be used to achieve chemical process goals.
Despite the high interest in the process of methane oxidative condensation, as well as the fact that the mechanism of the reaction and catalysts for the reaction of OCM are considered in many studies, it still has not reached an effective industrial implementation. It is clear that a good choice of catalyst contributes to a better course of the OCM process, and therefore contributes to obtaining high-quality products. Given the complexity and multivariance of the task of choosing a catalyst for the methane oxidative condensation, it would be appropriate to develop a decision support system capable of issuing recommendations for determining the most effective catalyst.

3. The aim and objectives of research

The aim of research is the design and development of an intelligent decision support system to select the most effective catalyst for the methane oxidative condensation.

To achieve this aim it is necessary to solve the following tasks:
1. To study the process of methane oxidative condensation and determine the characteristics by which the catalysts should be selected for the process.
2. To develop appropriate software to solve the problem of choosing the optimal catalyst, which should be part of the decision support system (DSS).

4. Research of existing solutions of the problem

Many publications were studied by the authors of the presented work which highlighted the general problems of the choice of catalysts and the problems of the choice of catalysts for the OCM process itself. For example, in [2], criteria are given that can be used to select the best catalyst for a particular oil refining process. At the same time, the author notes that the price of the catalyst may be a secondary factor. But other factors, such as possible raw materials, reactor parameters and operating conditions, market demand for products are much more important factors when choosing the optimal catalyst. These factors should be transformed into quantitative requirements for the characteristics of the catalyst, such as activity, selectivity and stability. In the end, the selected catalysts must be checked for compliance with these objective selection criteria.

One can cite another work [3] devoted to the main aspects of this subject; but it also addresses other important issues, among which the following are of particular interest. The first aspect is how to make catalytic cracking more environmentally friendly. The second aspect related to the emergence of monatomic catalysts for the implementation of important chemical reactions. The authors note that recently the goal of many developments is the desire to use raw materials such as pyrolysis oil, microalgae and such biological waste, sawdust and various non-food products from lignocellulose. An important task, however, is to ensure that CO₂ can be converted into fuel or useful materials and to reduce its concentration in the atmosphere. Therefore, the authors noted significant progress in the development of new catalysts, especially for the CO₂ treatment.

Many publications related to the subject of catalytic processes can be cited. In the post-Soviet space, the well-known Institute of Catalysis named after G. Boreskov of the Siberian Branch of the Russian Academy of Sciences, which publishes the «Catalysis in Industry» journal, on the pages of which there are many publications devoted to problems of catalytic processes [4]. Similar publications are constantly appearing on the MIT website [5]. There are more specific sites, with reference information, give recommendations on the choice of catalysts, for example, the site [6] provides in tabular form information on how to choose gas purification catalysts.

The problems of catalytic processes are the subject of many works both in well-known magazines and in information resources on various sites. All of them are intended to help technologists in deciding which catalyst is best for a given process. But still, finding the best solution remains a complex, multivariate, and often multi-step process. Therefore, the development of a computer system that will help in finding the best solution will always be a promising task.

In the last quarter of the 20th century after the oil crisis, the view of petrochemists turned to natural gas as an alternative source of hydrocarbons. Proven reserves of natural gas exceed similar reserves of oil. In addition, significant quantities of the main components of natural gas-methane and ethane are formed during the processing of biomass of plant and animal origin, as well as in the processing of organic raw materials. As noted in [7], the often repeated mantra that it is necessary to look for new renewable energy sources is simply not true. It is currently believed that there is enough natural gas in the world to meet the needs of the planet for about 230 years! For example, in January 2016, it was announced that Australia would increase natural gas production by about 150 % over the next 4 years. Until recently, the desire to discover and use renewable raw materials for energy production and for the production of materials was extremely strong. However, now, the understanding that non-renewable reserves (such as gas) are still sufficient, another emphasis appears. This is the use of both non-renewable and renewable raw materials in an environmentally efficient manner. Back and for the processing of light hydrocarbons, the challenge remains the creation of new eco-efficient technologies. One of such technologies can be methane oxidative condensation (OCM), which is being actively studied, as in the territory of the former Soviet Union. For example, the work [8] describes the OCM in the presence of new mesoporous amorphous catalysts containing lanthanum, cerium ions and their mixtures. It was shown that the previously discovered effect of the non-additive action of lanthanum and cerium ions in the OCM process is a fundamental regularity that is characteristic of both a mixture of oxides of these rare-earth elements (REE) and these elements introduced into the nanosstructured silicate matrix of ions. In another work [9], the OCM process was studied, which uses Li-W-Mn-O-SiO₂ composite materials as catalysts, which can be obtained using various synthesis methods: solid-phase, sol-gel synthesis and silica impregnation (SI), as well as reagents of various chemical nature. The authors of the work presented the results and carried out a comparative analysis of the OCM process taking into account data on the phase composition of the Li-W-Mn-O-SiO₂ composites, which is formed using various synthesis methods. The phase composition of the composites after their participation in the OCM reaction was determined. This assessment of the features of the phase composition of Li-W-Mn-O-SiO₂ composites, which affects their performance in the OCM. As it is possible to see, even from these two works, the problem of choosing
a catalyst has many options and almost every such publication adds a new layer of information that must be taken into account when solving this problem.

It is possible to cite many publications of different researchers on this topic. So, work [10] provides a whole report on the design of an OCM installation in Pennsylvania (USA). The resulting ethylene and ethane intended for sale to the olefin plant for further processing into polymers and plastics. All installation design data is provided. The OCM process consumes 9.10 billion pounds of methane and 46.2 billion pounds of oxygen per year. Methane and oxygen are converted in four fixed-bed isothermal catalytic reactors. The catalyst is LiMgO in the form of spherical granules with a diameter of 50 mm. The project for the specific OCM production presented in this work can be useful in comparison with similar industries and in terms of choosing the most effective catalyst. The authors envisage further studies of catalysts that will increase methane conversion in combination with increased selectivity for C₂ hydrocarbons and provide large profits.

A new concept for the OCM process with subsequent oligomerization to liquid was developed as part of the EU OCMOL project [11]. This technology is based on the principles of process intensification using advanced structured microreactor technology. It is also a fully integrated industrial process through the reuse and recycling of by-products, in particular CO₂, at every stage of the process. The focus is on the engineering aspects of the main reaction steps, i.e., catalysts, kinetics and equipment. An analysis of this, as in the previous project, can devote many pages.

Quite a lot of useful information on the choice of catalysts for the OCM process can be found in [12]. In this paper, let’s consider a complex microkinetic model that includes the characteristics of a catalyst and takes into account the homogeneous as well as heterogeneous catalyzed reaction steps in the OCM process. The model was used to evaluate large kinetic data sets obtained on 5 different catalytic materials. Application models have been extended from alkaline-magnesian catalysts represented by Li/MgO and Sn-Li/MgO and alkaline-earth lanthanum catalysts represented by Sr/La₂O₃, to rare-earth alkaline-earth calcium oxides provided by LaSr/ CaO, and to a Na-Mn-W/SiO₂. The authors of this work were able to adequately simulate the performance of all 5 investigated catalysts in terms of conversion of reactants and selectivity of products in the entire range of experimental conditions. It was found that the activity of Sr/La₂O₃, in terms of methane conversion, is about 2, 5, 30, and 33 times higher than over La-Sr/CaO, Sn-La/MgO, Na-Mn-W/SiO₂ and Li/MgO catalysts, respectively, under the same operating conditions. This was mainly due to the high stability of adsorbed hydroxyl, the high stability of adsorbed oxygen, and the high concentration of Sr/La₂O₃ active sites. It was revealed that the selectivity with respect to C₂ products depends on the coefficient of adhesion of the methyl radical and the stability of adsorbed oxygen and was the highest for the Na-W-Mn/SiO₂ catalyst. Subsequently, the database created in the DSS being developed was filled with this information.

A new version of the OCM process is considered in [13]. The new OCM process is based on adsorption. A low fraction of ethylene in OCM is considered appropriate. The proposed process is more reliable and flexible with a high reactor capacity. Another advantage of the process is the ability to use all the heat generated by the reaction. This energy saving, combined with the high selective performance of the reactor, can reduce carbon should to a value where it is lower than for ethane cracking. That is, from the point of view of continuous production, this is a significant advantage in places with strict emissions controls.

As can be seen from the reviewed works, the OCM process always uses various catalysts. Typically, each of them considers only a certain type or set of catalysts. Therefore, when designing and exploring options for the OCM process, computer support in the form of an intelligent DSS system, in which there will be aggregated information from disparate sources, will always be useful. Based on the collected information, databases (DB) and knowledge bases (KB) of decision-making systems for choosing a catalyst for OCM can be formed.

5. Methods of research

It should be noted that the process of methane oxidative condensation has some advantages among other methods:
- to convert methane to ethylene, only one process step is needed;
- ethylene is the main raw material for petrochemical synthesis, from which it is possible to get a wide range of products;
- OCM reaction proceeds at atmospheric pressure (in contrast to the Fischer–Tropsch synthesis [14]).

At the moment, researchers of the OCM reaction have come to a consensus that this reaction proceeds according to a homogeneous-heterogeneous mechanism [8]. This means that methane activation occurs on the surface of a solid oxide catalyst with a detachment of one hydrogen atom from a methane molecule with the formation of CH₃ radicals (1). The recombination of propellant radicals occurs in the gas phase with the formation of an ethane molecule (2).

\[
\begin{align*}
\text{CH}_4 + [O] & \rightarrow \text{CH}_3^* + [OH], \\
\text{CH}_3^* + \text{CH}_3^* & \rightarrow \text{C}_2\text{H}_6
\end{align*}
\]

where [O] – the active oxygen center on the surface of the catalyst; [OH] – adsorbed on the surface of the hydroxyl group catalyst.

Further, the process proceeds according to the following scheme: a water molecule and an oxygen vacancy on the catalyst surface are formed from two adsorbed hydroxyl groups [OH]:

\[
2[OH] \rightarrow \text{H}_2\text{O} + [O] + [...].
\]

where [...] is an oxygen vacancy.

Now it is necessary to restore the active oxygen center. This occurs by reoxidizing the surface of the catalyst with oxygen in the gas phase:

\[
\text{O}_2(\text{gas}) + 2[...] \rightarrow 2[O].
\]

Ethane dehydrogenation with the formation of ethylene can occur both on the surface of the catalyst and in the gas phase (at temperatures above 700 °C):

\[
\text{C}_2\text{H}_6 \rightarrow \text{C}_2\text{H}_4 + \text{H}_2.
\]
The catalysts for the OCM reaction can be divided into two large groups: metal oxides, it is difficult to recover and metal oxides are reduced. As it is known, complex multiphase catalysts consist of a carrier (substrate) and a promoter – a substance that is added in small quantities to the carrier. Metal oxides are a carrier (basic substance) of the catalyst. The preparation of the catalyst is carried out in various ways: by impregnating the carrier with a solution containing the promoter, coprecipitation from solutions, solid-phase synthesis, etc. The promoter on the carrier is designated as Me/carrier. Bi- and multifunctional catalysts are also distinguished in which two or more substances are active components of the catalyst. In such catalysts, all active phases, for example PbO/Al2O3, are indicated.

The best catalysts make it possible to obtain selectivity for C2 products in the range of 60–80 %, with a methane conversion of 20–30 %. According to economic calculations, the OCM process becomes profitable with a methane conversion of 25 %.

In early 2015, Siluria Technologies in Texas, USA launched a commercial pilot plant for the direct conversion of natural gas to ethylene (OCM) [15]. During the year since the launch, 18 test tests were successfully completed, during which various working conditions were checked, including different temperatures, pressures, and gas flow rates at the inlet. Siluria’s plant is flexible for incoming raw materials, it works both in air and in oxygen, and it also provides the ability to change the amount of ethane by creating a recycle. The basic approach of Siluria Technologies was to test a huge number of catalysts [16]. The company built an automated system that could simultaneously quickly synthesize hundreds of different catalysts, and then check how well they convert methane to ethylene. During test trials, more than 30,000 catalysts were developed and tested.

It is clear that a good choice of catalyst contributes to a better course of the OCM process, and therefore contributes to obtaining high-quality products. When choosing a catalyst, it is necessary to take into account the parameters of the process itself, such as: the temperature of the OCM reaction, the conversion of CH4, the selectivity of C2 hydrocarbons, the yield of C2 hydrocarbons and the reaction rate, etc. [1, 16].

As mentioned earlier, catalysts for the reaction of OCM are divided into two large groups. The first group includes oxides of alkaline-earth elements promoted by alkali metals, as well as oxides of trivalent rare-earth earth elements (REE), both by themselves and promoted by alkaline or alkaline-earth elements (Table 1). Promotion is carried out by the method of impregnation with hydroxides or salts of alkali or alkaline earth metals, followed by drying and calcination to decompose salts and remove water. The amount of promoter, as a rule, does not exceed 5–7 atm %. In terms of metal. When the concentration of the promoter is more than 10 atm % the properties of the catalyst are actually determined by the properties of the phase formed on the surface, in particular the properties of alkaline metal carbonates. Lithium, sodium, strontium and barium are in the composition of the catalysts in the form of the corresponding ions.

![Fig. 1. The scheme of the reaction of methane oxidative condensation](Image)

The first group of catalysts most effectively operates in a continuous mode, that is, with the simultaneous supply of reactants—methane and oxygen. The second group of catalysts includes metal oxides, are reduced, and, accordingly, they work better in the oxidation-reduction mode, that is, in a batch mode. These catalysts conduct the process due to the oxygen lattice of the catalyst, followed by regeneration of the latter by the oxygen of the gas phase. However, the promotion of these alkali and alkaline earth metal catalysts also improves the efficiency of the process.

The effectiveness of the catalyst is determined by the ability of the catalyst to activate oxygen (4), that is, to create active oxygen centers for breaking the C–H bond in the methane molecule according to expression (1). When a monovalent additive is added to the oxide of a divalent or trivalent metal (or a divalent additive to the oxide of a trivalent or tetravalent metal), excess oxygen vacancies are created that serve as oxygen activation centers in accordance with (4). As can be seen from the Table 1, in almost all catalysts, except for trivalent REE oxides, additional active centers are created by introducing low-valent additives. REE oxides have the structure of defective fluorite, in which 25 % of the oxygen positions are vacant. Therefore, they are able to activate additional oxygen due to the nature of their structure.

Based on the complexity of the task of choosing a catalyst for the considered process of methane oxidative condensation, it would be appropriate to develop a decision support system capable of issuing recommendations on a catalyst that is most effective by all criteria. The decision support system (DSS) is a computerized system that, based on the accumulated information in the form of data and knowledge, can help in the process of making managerial decisions in any area of human activity [17]. DSS is implemented as an interactive automated computer...
system that helps the decision maker (DM) to use DSS data and models for identification, computer experiments, analysis and decision making in this problem area. In the developed DSS, this is the choice of the best catalyst.

Thus, the intellectual DSS developed by the authors of the article should help in choosing the catalyst that is most effective under the given conditions of the OCM process. The system has two main intellectual components. The first is based on the application of the hierarchy analysis method (HAM) and requires the participation of a human expert, the second on the methods of Data Mining) without the participation of a specialist.

After solving the problem of choosing a catalyst, the system provides for further modeling of the process in a ChemCad environment.

The DSS structure for the choice of catalyst is shown in Fig. 2.

- A program for selecting a catalyst by the method of hierarchy analysis, designed to decompose the problem and stage-by-stage prioritization based on pairwise comparisons.
- Sec5 program is designed for intelligent processing of experimentally obtained data, based on a decision tree.
- ChemCad program is designed for computer simulation of chemical-technological processes.

For the development of DSS software, an object-oriented C++ programming language and a Microsoft Visual Studio development environment were chosen.

6. Research results

6.1. The use of DSS to solve the problem of choosing a catalyst for the OCM process. An analysis of the literature [1, 15] shows that the catalyst for the OCM process is characterized by the following five criteria: temperature, CH4 conversion, selectivity of C2 hydrocarbons, yield of C2 hydrocarbons, and reaction rate. The task of choosing a catalyst, which is a necessary component of the OCM process, can be considered as a decision making task. Accordingly, for making decisions it is possible to use HAM [18].

The general structure of the catalyst selection process can be presented in hierarchical form (Fig. 3), which contains three levels:

1) goal – a brief description of the task;
2) criteria – a quantitative or qualitative characteristic that is essential for judgments about the object;
3) alternatives – objects between which it is necessary to make a choice.

Table 2 shows the most widely studied catalysts for the methane oxidative condensation and their indicators.

If there are authoritative experts on catalytic processes, it is possible to use the DSS component, implements a method for analyzing hierarchies. With this approach, to compare two objects by any criterion, the expert uses a nine-point scale predefined in the HAM. Based on the judgments of the DM expert, matrices of pairwise comparisons are constructed that allow at the end of this procedure to determine the most acceptable catalyst.

In the absence of such experts, it is possible to use data on the catalysts of the OCM process stored in the DSS database (DB). Further, the Sec5 software component based on decision trees allows one to determine the catalyst for the methane oxidative condensation, which best suits the given process parameters.

After solving the problem of choosing the optimal catalyst, in DSS the user is invited to conduct computer simulation of the OCM process in the ChemCad environment.

After that, it is possible to choose the best version of the technological scheme, the input streams of which are natural gas and oxygen, and the output streams are ethylene and a mixture of hydrogen, carbon dioxide and water. The implementation of the technological scheme in the ChemCad program is shown in Fig. 4 [19].
6.2. Evaluation of the results. Next, a computer simulation of the OCM process was conducted in a ChemCad environment to verify the correctness of the solutions proposed by the developed DSS for various catalysts.

Five catalysts with specific characteristics are considered. A comparison of the performance of the technological scheme of the OCM process using various catalysts is shown in Fig. 5.

As can be seen from Fig. 5, the most effective is the LiCl/MnO₂ catalyst, which provides the highest performance of the technological scheme for the obtained ethylene.

From this it can be seen that the solutions proposed by DSS can be implemented in the implementation of the corresponding production.

### Table 2

| Catalyst             | Temperature, °C | Conversion CH₄, % | Selectivity of C₂ hydrocarbons, % | Yield of C₂ hydrocarbons, % | Reaction rate, molec/m²/s |
|----------------------|-----------------|------------------|----------------------------------|-----------------------------|---------------------------|
| PbO/MgO              | 700             | 13.1             | 56.2                             | 7.2                         | 3·10¹⁹                    |
| Bi₂O₃                | 800             | 21               | 68.8                             | 14                          | 6·10¹⁷                    |
| La₂O₃                | 750             | 12.2             | 67.4                             | 8.3                         | 5.5·10¹⁸                  |
| Na₂O₃/MgO            | 770             | 26               | 46                               | 11.2                        | 1·2·1⁰¹⁸                  |
| Ca-ore               | 870             | 28               | 77.5                             | 22.2                        | 2·7·1⁰¹⁷                  |
| MgO                  | 750             | 29               | 50                               | 14.5                        | 5·10¹⁶                    |
| Li/MgO               | 700             | 34               | 58                               | 20                          | 4·10¹⁷                    |
| LiCl/MgO₂            | 750             | 47.2             | 64.7                             | 30.8                        | 7·10¹⁹                    |
| BaPbO₃               | 800             | 22               | 62.7                             | 14                          | 4·5·10¹⁷                  |
| SrCe₀.₉Yb₀.₁         | 750             | 53               | 60                               | 31.6                        | 2·5·10¹⁹                  |
| Ba₂Sb(LaBi)O₆        | 850             | 41.6             | 43.5                             | 18.1                        | 1·10¹⁹                    |
| HfO₂(Al₂O₃)         | 750             | 23.8             | 82.3                             | 19.5                        | 6·10¹⁷                    |
| LiCa₂Bi₃O₄Cl₆       | 720             | 41.7             | 46.5                             | 19.3                        | 7·10¹⁷                    |

**Fig. 4.** The technological scheme of the methane oxidative condensation (OCM) with recycling in a ChemCad medium: 1 – heat exchanger; 2 – mixer; 3 – evaporator; 4 – demethanizer; 5 – heater; 6 – mixer; 7 – OCM reactor; 8 – mixer; 9 – ethane dehydrogenation reactor; 10 – heater; 11 – separator; 12 – ethylene recovery column; 13 – heater; circles are devices of the circuit, and squares – the numbering of flows.

**Fig. 5.** Comparative diagram of the effectiveness of the catalysts for the methane oxidative condensation (OCM).
7. SWOT analysis of research results

Strengths. The positive effect of the object of research in the possible information support and automation of decision-making on the choice of a catalyst for the process of methane oxidative condensation. Since it is possible to combine the knowledge and experience of experts and the accumulated information in the databases, it becomes possible to choose the best catalyst option for this implementation of the technological scheme of the OCM process. This includes both data mining procedures and modeling methods, as part of an integrated computer DSS.

Weaknesses. The weaknesses of the proposed system include the relatively small size of the database on possible catalysts and the constant need to replenish it, as new catalysts appear and the criteria for choosing the best one change.

Opportunities. The proposed DSS will help facilitate decision-making on the choice of a catalyst for OCM and improve the choice itself. Interaction with the WAR algorithm as part of ChemCad gives additional opportunities for assessing the stability of the proposed scheme of the OCM process under the conditions of a selected catalyst.

Threats. From the design organization that is engaged in the creation of production or the improvement of the existing one that implements the OCM process, it is required, in addition to the ability to install the appropriate DSS software, also its qualified use. This is especially true for the use of hierarchy analysis in the DSS structure. An engineering qualification for this may not be enough and there is a problem in attracting another specialist in knowledge.

8. Conclusions

1. The possibilities of implementing the process of methane oxidative condensation are considered. It is proved that the process is promising, however, despite the fact that the reaction mechanism has long been known, there is still no effective industrial implementation. Since the process is catalytic, particular attention should be paid to the choice of catalyst. It is determined that the main parameters of the OCM reaction itself. These are temperature, CH₄ conversion, selectivity of C₂ hydrocarbons, yield of C₂ hydrocarbons, reaction rate, etc.

2. Intelligent software decision-making system for the choice of catalyst is developed, which contains two intelligent components. The first of them is based on the application of the hierarchy analysis method and requires the participation of a human expert, the second – on the methods of intelligent data processing in the See5 environment without the participation of a specialist. The operation of the system was tested on examples of the selection of a catalyst for the OCM process.

After evaluating the effectiveness of the selected catalyst, a computer simulation of technological process variants using various catalysts in a ChemCad medium is carried out. Based on the results, the best version of the scheme is chosen.

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