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ЭКСЕРГЕТИЧЕСКИЙ АНАЛИЗ СИСТЕМЫ ТЕРМОХИМИЧЕСКОЙ РЕГЕНЕРАЦИИ ТЕПЛОТЫ НА БАЗЕ ПАРОВОЙ КОНВЕРСИИ МЕТАНА

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Вопросы изучения процессов паровой конверсии метана являются важными, поскольку они могут применяться не только с целью получения водорода, но и при реализации принципов термохимической регенерации теплоты теплоэнергетических, теплотехнических и в принципе любых топливоиспользующих установок, поскольку они позволяют снижать затраты материальных и энергетических ресурсов в расчете на единицу получаемой продукции с одновременным улучшением экологических показателей. В последнее время для исследования систем, в которых происходит преобразование одних видов энергии в другие, применяется эксергетическая методология, позволяющая наиболее адекватно оценивать степень термодинамического совершенства. Такая методология ранее была применена к установкам по преобразованию химической энергии органического топлива в электрическую и тепловую энергию – электростанции, котельные, теплоэлектроцентрали; преобразованию электроэнергию в теплоту различного потенциала – холодильные установки и тепловые насосы и ряду других установок и систем. В рамках данной работы проведено исследование по определению эксергетического КПД процесса паровой конверсии метана. Установлено, что в условиях использования термохимической регенерации возможно достижение высокой степени регенерации тепла отходящих дымовых газов при умеренной температуре. Определен равновесный состав продуктов реакции паровой конверсии метана для различных параметров протекания процесса (температуры, давления, отношения водяного пара к метану 1 и 2). Выполнен термодинамический анализ процесса паровой конверсии метана для определения возможных диапазонов изменения технологических параметров реакционной смеси в зависимости от предназначения установки – обеспечение химических процессов сырьем, проведение регенерации теплоты или комбинированные энерго-химические комплексы с комплексным использованием потенциала органического топлива.

Ключевые слова: метан, водород, паровая конверсия, ресурсосбережение, материальный баланс, эксергия

EXERGIC ANALYSIS OF THERMOCHEMICAL HEAT RECOVERY SYSTEM BASED ON METHANE STEAM CONVERSION

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The issues of studying the processes of steam reforming of methane are important, since they can be used not only to produce hydrogen, but also in the implementation of the principles of thermochemical heat recovery of thermal power, heat engineering and, in principle, any fuel-using installations, since it allows reducing the cost of material and energy resources per unit of received production with simultaneous improvement of ecological indicators. Recently, to study systems in which some types of energy are converted into others, exergetic methodology has been used, which makes it possible to most adequately assess the degree of thermodynamic perfection. This methodology was previously applied to installations for converting the chemical energy of organic fuel into electrical and thermal energy – power plants, boiler houses, thermal power plants; converting electricity into heat of various potentials – refrigeration units and heat pumps and a number of other installations and systems. Within the framework of this work, a study was carried out to determine the exergy efficiency of the methane steam reforming process. It has been established that under the conditions of using thermochemical regeneration, it is possible to achieve a high degree of heat recovery from flue gases at a moderate temperature. The equilibrium composition of the reaction products of steam reforming of methane was determined for various parameters of the process (temperature, pressure, ratio of water vapor to methane 1 and 2). A thermodynamic analysis of the methane steam reforming process was performed to determine the possible ranges of changes in the technological parameters of the reaction mixture, depending on the purpose of the installation – providing chemical processes with raw materials, performing heat recovery, or combined energy-chemical complexes with the integrated use of the fossil fuel potential.

Key words: methane, hydrogen, steam reforming, resource saving, material balance, exergy

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INTRODUCTION

The steam conversion of methane is a basic process of the chemical industry in the technology of obtaining ammonia, methanol, formaldehyde, acetic acid, ethylene glycol, etc., and is also used in the production of pure hydrogen [1-4], which is especially relevant in the light of hydrogen energy. Methane processing is carried out by catalytic method [5] with proceeding of basic steam reforming reaction (1), as well as water gas reaction (2) in case of excessive water vapor content in the initial mixture:

$$CH_4 + H_2O = CO + 3H_2 – 206{,}3 \text{ MJ/kmol}; \qquad (1)$$

$$CO + H_2O = CO_2 + H_2 + 41{,}2 \text{ MJ/kmol}. \qquad (2)$$

Therefore, the reaction products or as they are called synthesis gas will have in their composition in addition to the remains of unreacted methane and water vapor, carbon monoxide and hydrogen, and carbon dioxide, the content of which will depend on technological parameters such as temperature, pressure, composition of the initial reaction mixture, and may vary from 2% to 16%.

There are various technological arrangements for methane conversion reactions into synthesis gas. In this case, from the chosen technology will depend on the efficiency of the process, which can be divided into economic, obviously associated with minimum costs, and hence the maximum integral effect and technical efficiency - determining the perfection of the conversion process and the cost of all types of energy to implement the technological process.

The technology of steam conversion of methane can be realized in units of thermochemical regeneration (THR), which allow to solve the problem of increasing the thermal efficiency of industrial fire-technical devices [6-8]. The essence of THR consists in using the physical heat of waste flue gases for preliminary endothermic processing of the original fuel (methane), which in this case receives a larger reserve of chemically bound heat.

Application of THR technology for heat recovery of flue gases in high-temperature furnaces was investigated by various authors. In [9] the author considered steam conversion using different initial fuels: methanol, ethanol, n-butanol and glycerol. It was found that with increasing temperature and decreasing pressure, the degree of fuel conversion increased, but only to a certain limit. The maximum degree of conversion of methanol was observed at 600 K, ethanol at 730 K, n-butanol at 860 K and glycerol at 890 K. As a result, recommendations on the use of different fuels for thermochemical regeneration at different flue gas temperatures were formed.

Thermochemical heat regeneration increases the gross heat of combustion of the new synthetic fuel in terms of 1 kg of the original fuel. Thus, when using the reaction products it is possible to significantly reduce the amount of fuel consumed by the plant. D.I. Paschenko in [10] investigated the degree of increase in the lower heating value of fuel combustion as a result of THR. Using thermodynamic analysis, the author evaluated the efficiency of using ethanol, methanol, glycerol, propane and methane as fuel for THR. As a result of the studies, the author found that the steam conversion of methane was the most thermodynamically advantageous reaction due to the best opportunity to transform the heat of the waste flue gases.

The flue gases of gas turbines with a rather high temperature can also serve as a source of thermal energy for THR reactions. Application of methane steam conversion reactions for THR of flue gas heat on an example of Siemens SGT6-8000H turbine was investigated in the article [11]. The author determined the limiting degree of flue gas heat recovery, which is more than 69% for a flue gas temperature of 900 K at a reactor pressure of 5 bar and a methane/steam ratio of 1/2. It was also found that with increasing pressure in the reactor, the efficiency of THR decreases, so the optimal pressure in the reactor should be at 5-10 bar.

RESEARCH METHODOLOGY

In the present work presented thermodynamic analysis of methane steam conversion, the results of which are to assess the maximum efficiency of methane conversion depending on technological parameters and conditions of the process.

Thermodynamic analysis must necessarily have data on the composition of gases formed during the implementation of processes of steam conversion of methane. In this article, as a tool for thermodynamic analysis, the Gibbs free energy minimization method is used, which consists in the fact that the state of the system is the most favorable for the flow of chemical reactions, if the Gibbs free energy value is minimal and its change at various parameters of the thermodynamic system tends to zero:

$$dG_t = 0, \qquad (3)$$

where G^t is the Gibbs free energy, kJ.

To calculate the composition of synthesis gas in equilibrium reactions of methane steam conversion, the following assumptions were made:

1. the thermodynamic system is close to ideal (which is true at relatively low pressures, up to 5-10 bar, and high reactor temperatures) [10];
2. the process is stationary, so the change in potential and kinetic energy is negligible;

3. the reactor wall is adiabatic, so there are no heat losses through the outer walls of the reactor;

4. only equilibrium chemical reactions at the selected fuel composition and reactor operating conditions were considered.

The free Gibbs energy of methane steam conversion reactions can be determined through the total chemical potential of each component:

$$G_t = \sum n_i \cdot \mu_i\ , \qquad (4)$$

where n_i is the number of moles of the i-th component, mol; μ_i is the chemical potential of the i-th component, kJ/mol.

The chemical potential of a component determines the amount of energy for one mole of a substance required to convert it from its standard state to the system in question at constant pressure, temperature, and concentration of the other reaction components. Since the value of the chemical potential does not remain constant, an extensive database is needed to calculate the Gibbs free energy. The equation for determining the chemical potential of each reaction component is:

$$\mu_i = \Delta G^0_{f_i} + R \cdot T \cdot \ln(\frac{f_i}{f_i^0}), \qquad (5)$$

where $\Delta G^0_{f_i}$ is the Gibbs energy for the chemical conversion of the i-th component under normal conditions, kJ; R is the universal gas constant, kJ/(mol·K); T is the temperature in the reactor, K; f_i is the fugacity (volatility) of the i-th component, Pa; f_i^0 is the fugacity of the i-th component under normal conditions, Pa.

If we combine expressions (4) and (5), the equations for determining the Gibbs free energy will take the following expression:

$$G^t = \sum n_i \cdot \Delta G^0_{f_i} + \sum n_i \cdot R \cdot T \cdot \ln(\frac{f_i}{f_i^0}). \qquad (6)$$

The thermodynamic analysis of methane vapor conversion reactions was performed in a specialized program Aspen Hysys. This program, due to its extensive database, allows to calculate of equilibrium chemical reactions at various thermodynamic parameters of the system. The calculation was performed for the scheme shown in Fig. 1.

The diagram presents a steam boiler, a mixer and a chemical reactor (Gibbs reactor). In the steam boiler water steam is produced, which is necessary for the steam conversion reaction of methane. Residual heat of flue gases can be used as a source of heat for steam formation. Further, the obtained steam and methane are evenly mixed in a mixer, and the temperature of methane increases due to heat exchange with water vapor. In the Gibbs reactor, the composition of methane steam conversion reaction products (synthesis gas) is calculated by the Gibbs energy minimization method.

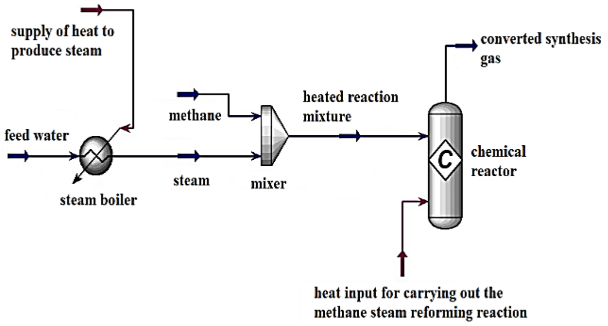

Fig. 1. Calculation scheme of THR based on methane steam conversion

Рис. 1. Расчетная схема ТХР на базе паровой конверсии метана

During thermodynamic analysis it is necessary to know the maximum degree of methane conversion, which at known (given) process parameters, such as temperature, pressure and composition of the initial mixture, can be achieved in the THR reactor. The degree of methane conversion is determined by the expression:

$$\beta = \frac{CH_4^{in} - CH_4^{out}}{CH_4^{in}} \cdot 100\%, \qquad (7)$$

where CH_4^{in} and CH_4^{out} are the amount of methane at the reactor inlet and outlet, respectively.

The maximum degree of methane conversion corresponds to the equilibrium degree of methane conversion for given conditions of the process. In modern reactors at a volume rate of about 6000 h^{-1} using nickel-containing catalysts, a methane conversion degree close to the equilibrium is achieved.

It is known that the most reasonable indicator of efficiency with for processes similar to those occurring in the plants under consideration is the exergy balance [12-19]. The exergy balance is built on the basis of material and thermal balances of the process or plant, taking into account the quality or technical operability of the substance flow, i.e., the maximum amount of useful mechanical work that can be obtained at the transition from the current state of the system to a state of equilibrium with the environment. The equation of material balance by chemical elements that make up the chemical engineering system for the stationary mode of operation is as follows:

$$\sum_{j=1}^{l} g_{ij} = 0, \qquad (8)$$

where g_{ij} is the flow of the i-th element through the j-th input or output; l is the total number of inputs and outputs.

When performing calculations of material, energy (thermal) and exergy balances, it is assumed that the flows entering the system are positive, and the flows leaving the system are negative.

When performing evaluation calculations, when the so-called external problem is solved, it is very convenient to apply the "black box" principle, in which functional models reflect only the externally perceived behavior of the object and the interaction of individual constituents within the object is not considered [20, 21].

In such a formulation of the research problem as a criterion for assessing the thermodynamic efficiency of the considered variants of THR schemes with methane vapor conversion, the exergy efficiency was taken, which according to the recommendations [22] is defined by the expression:

$$\eta_{ex} = \frac{\sum Ex_{use}}{\sum Ex_{sup}}, \qquad (9)$$

where $\sum Ex_{use}$ is the sum of exergies determining the useful effect of the installation, kW; $\sum Ex_{sup}$ is the total value of all types of exergies supplied to the system (initial mixture, heat and electricity to compensate for the pressure drop during the passage of the mixture through the elements of the scheme), kW.

In practical calculations, the physical exergy of the substance flow is determined through two components: isothermal Ex_T, which is related to the difference in pressure, and isobaric Ex_p, which is a consequence of the difference in temperature of the substance from the state of the environment [22]:

$$E_x = E_{x_T} + E_{x_P} = R \cdot T_0 \cdot \ln\left(\frac{p}{p_0}\right) + $$
$$+ c_p \cdot \left(T - T_0 - T_0 \cdot \ln\left(\frac{T}{T_0}\right)\right), \qquad (10)$$

where T_0 and p_0 are the ambient temperature and pressure, K and Pa, respectively; T and p are the temperature and pressure of the considered substance flow, K and Pa, respectively; c_p is the average value of isobaric heat capacity in the considered process, kJ/(kg·mol·K).

RESULTS AND DISCUSSION

In determining the compositions of the resulting synthesis gas, it was assumed that the temperature in the reaction zone is constant and the composition is close to the equilibrium. Under the accepted assumptions, the composition of steam conversion products can be unambiguously determined by the temperature and pressure of the process and the initial composition of the steam-gas mixture. The results of calculations using the developed methodology for the scheme in Fig. 1 are presented in Table 1 and Table 2.

The methodology for calculating the composition of the resulting products of the methane steam conversion reaction was verified with the data from work [23]. The verification was performed for a pressure range from 1 to 10 MPa at a reaction temperature of 773 K and CH_4/H_2O = 1/2 ratio. Fig. 2 shows the calculated compositions according to our proposed methodology for the above conditions. The analysis of Fig. 2 shows a high convergence of the calculation results.

Table 1

Gas composition of methane steam conversion at 5 bar

***Таблица 1*. Состав газов паровой конверсии метана при давлении 5 бар**

Component of reaction products	Process temperature, °C				
	600	700	800	900	1000
CH_4	0.3044	0.1957	0.0984	0.0431	0.0189
	0.1667	0.0864	0.0256	0.0045	0.0008
H_2	0.3569	0.5054	0.6255	0.6939	0.7248
	0.3672	0.5070	0.5977	0.6235	0.6237
H_2O	0.2409	0.1467	0.0753	0.0346	0.0158
	0.3677	0.2598	0.1933	0.1759	0.1772
CO	0.0343	0.1032	0.1776	0.2199	0.2375
	0.0263	0.0799	0.1358	0.1608	0.1697
CO_2	0.0635	0.0489	0.0232	0.0085	0.0031
	0.0721	0.0668	0.0476	0.0353	0.0286

Note: The numerator shows values for the ratio in the initial mixture of steam/methane at 1/1, and the denominator is 2/1
Примечание. В числителе указаны значения соотношения в исходной смеси пар/метан 1/1, а в знаменателе 2/1

Table 2

Gas composition of methane steam conversion at 10 bar

***Таблица 2.* Состав газов паровой конверсии метана при давлении 10 бар**

Component of reaction products	Process temperature, °C				
	600	700	800	900	1000
CH_4	0.3439	0.2497	0.1483	0.0745	0.0352
	0.1987	0.1263	0.0554	0.0147	0.0029
H_2	0.2911	0.4290	0.5597	0.6526	0.7029
	0.3001	0.4347	0.5481	0.6067	0.6200
H_2O	0.2869	0.1962	0.1161	0.0602	0.0295
	0.4221	0.3163	0.2311	0.1886	0.1800
CO	0.0211	0.0717	0.1436	0.1985	0.2268
	0.0163	0.0561	0.1136	0.1530	0.1681
CO_2	0.0569	0.0535	0.0322	0.0142	0.0057
	0.0628	0.0666	0.0518	0.0370	0.0290

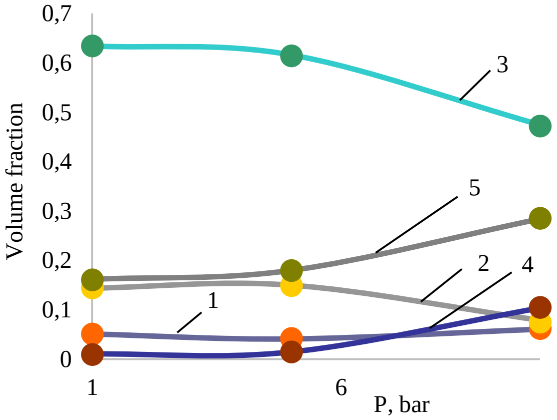

Fig. 2. Comparison of the calculated compositions (line) of methane steam conversion products with the data of [23] (markers): 1 – CO_2; 2 – CO; 3 – H_2; 4 – CH_4; 5 – H_2O
Рис. 2. Сопоставление расчетных составов (линия) продуктов паровой конверсии метана с данными работы [23] (маркеры): 1 – CO_2; 2 – CO; 3 – H_2; 4 – CH_4; 5 – H_2O

The thermodynamic calculations were performed using tables of thermodynamic properties of methane, water and water vapor [24] and the following dependencies to determine the gross exergy values of the flux of matter, heat and electricity, respectively:

$$Ex_{nom,i} = G_i \cdot [(h - h_0) - T_0 \cdot (s - s_0)]; \qquad (11)$$

$$Ex_Q = Q \cdot [1 - (T_0/T)]; \qquad (12)$$

$$Ex_N = N, \qquad (13)$$

where G_i is the flow rate of the i-th substance, kg/s; h and h_0 are the specific enthalpies of the substance flow respectively at the initial state of the system and at the calculated environmental parameters, kJ/kg, respectively; s and s_0 are the similar specific values for entropy, kJ/(kg·K), respectively; Q is the heat supplied to the system, kW; T is the temperature of supplied heat, K; N is the electric power consumed to conduct the THR process, kW.

Fig. 3 shows the results of calculations by expression (7) of methane conversion coefficient depending on process parameters. The concentration of methane at the inlet depends on the ratio of H_2O to CH_4, and at the outlet was determined according to [25].

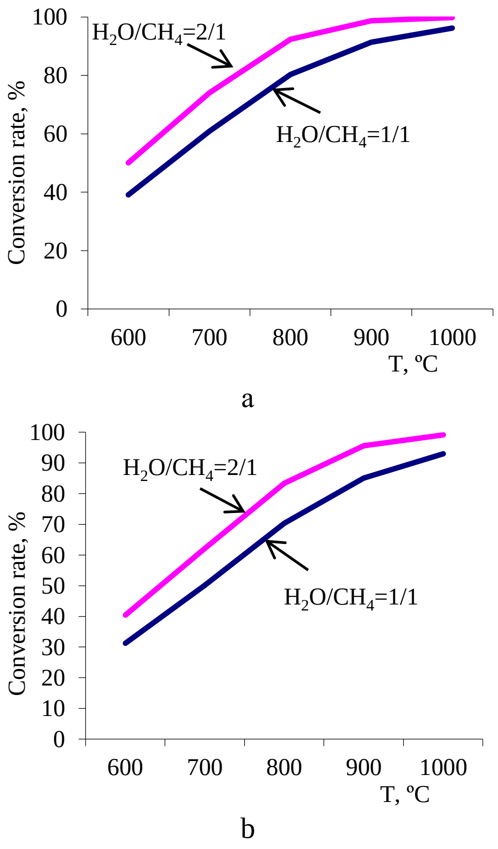

Fig. 3. Dependence of methane conversion rate on process parameters (temperature and H_2O/CH_4 ratio) at process pressure: a) 5 bar b) 10 bar

Рис. 3. Зависимость степени конверсии метана от технологических параметров (температуры и отношения H_2O/CH_4) при давлении процесса: а) 5 бар и б) 10 бар

As can be seen from the presented figures, with an increase in temperature, a decrease in pressure, and an increase in the relative amount of water vapor, the degree of methane conversion increases, which is confirmed by the data [26]. It may also be noted that an increase in pressure slows down the process of methane conversion the stronger the lower the temperature. And if in the chemical industry, despite an equilibrium shift in the undesirable direction, higher process pressures are used to reduce energy and capital costs, in the conditions of THR it is more preferable to use low pressures.

Fig. 4 shows the results of calculations of exergic efficiency by expression (9). In calculations ambient parameters T_0=273 K and p_0=0.1 MPa, as well as the condition of isothermicity in the reactor by heating medium and temperature of reaction mixture were taken.

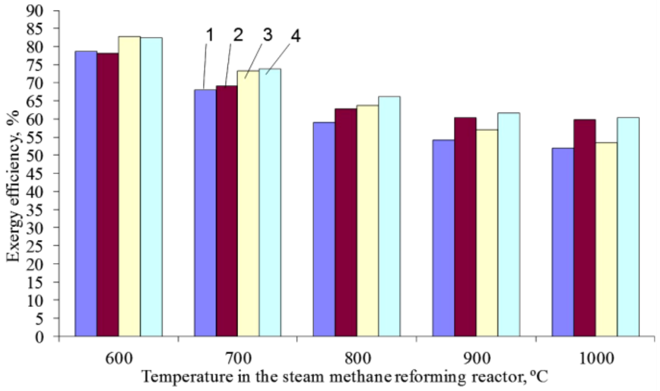

Fig. 4. Dependence of the exergy efficiency of the THR unit with steam conversion of methane on process parameters:
1 – H_2O/CH_4=1/1, p=5 bar; 2 – H_2O/CH_4=2/1, p=5 bar;
3 – H_2O/CH_4=1/1, p=10 bar; 4 – H_2O/CH_4=2/1, p=10 bar

Рис. 4. Зависимость эксергетического КПД установки ТХР с паровой конверсией метана от технологических параметров:
1 – H_2O/CH_4=1/1, 5 бар; 2 – H_2O/CH_4=2/1, 5 бар;
3 – H_2O/CH_4=1/1, 10 бар; 4 – H_2O/CH_4=2/1, 10 бар

When analyzing the data obtained, we can conclude that an increase of temperature of steam conversion of methane reduces thermodynamic efficiency of conversion process, for example for H_2O/CH_4=1/1 and p=5 bar from 78.7% to 51.9%, and for H_2O/CH_4=2/1 and p=10 bar - from 82.4% to 60.4%. This fact can be explained by the fact that the expenditure of exergy heat to provide the conversion process increases. And after 900 °C the exergy efficiency decreases slightly, on average by 1.3-3.5%, it is associated with almost complete conversion of the initial methane. Increasing the amount of supplied water steam according to the presented calculation scheme increases the value of exergy efficiency, except for the variant with the process temperature of 600 °C.

CONCLUSION

1. Increasing demand for hydrogen and hydrogen-bearing gas in the near future will be satisfied by organic fuel and, first of all, by natural gas. At the same time, use of natural gas in power and heating units will be associated with increased requirements for thermal efficiency, which can be improved by using thermochemical heat recovery units based on steam conversion reactions.

2. A methodical approach to estimate thermodynamic (exergic) efficiency of methane steam conversion process has been formed. This methodology makes it possible to establish the most favorable technological parameters of the THR process based on steam conversion of methane.

3. On the basis of the performed studies it was established that to reduce the concentration of residual methane in the converted gas at given temperatures and pressures of the conversion process the steam consumption must be increased, which for the case of thermochemical heat recovery is not a priority, because then the converted gas for the most part will be used as fuel [27-29].

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