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# STUDY OF THE KINETICS OF THE PROPANE-BUTANE FRACTION PYROLYSIS PROCESS

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**Abstract**

This study investigates the kinetics of the pyrolysis process of a propane-butane fraction. The thermal decomposition reaction was conducted in a quartz reactor packed with 0.3–0.5 mm quartz chips (hereinafter referred to as "quartz") under an oxygen-free environment and elevated temperatures. The research focused on analyzing the decomposition process, which involves breaking C-C and C-H bonds in the absence of air at high temperatures. Before propane and butane undergo physical adsorption on the quartz surface, they first decompose into radicals. The subsequent thermal decomposition of the propane-butane fraction, primarily driven by C-C and C-H bond cleavage, is hypothesized to occur predominantly on the quartz surface within the reactor specifically designed for this process. At temperatures ranging from 500 to 800 °C, the catalytic decomposition of primary hydrocarbons on the quartz surface was examined under helium conditions, both with and without quartz. The results demonstrated the suppression of coke formation under these conditions, highlighting the significant catalytic role of the quartz surface in facilitating hydrocarbon decomposition.

**Keywords** Propane-butane fraction, pyrolysis, empty reactor, ethylene, propylene, kinetic equation, Arrhenius equation.

## INTRODUCTION

In the global petrochemical industry, there is a tendency to increase the demand for lower alkenes. Thermal pyrolysis processes with "water vapour" in tube furnaces are the main sources of ethylene and propylene production [1-3], they are used in various branches of the national economy for the production of polyethene, polypropylene, phenol, acetone, alcohols, varnishes, solvents, as well as synthesis of other substances used as raw materials as mediators to make.

Another urgent task in the production technology of lower alkenes is the selection of raw materials for the pyrolysis process.

To date, there are no industrial enterprises producing lower alkenes by catalytic pyrolysis and catalytic conversion of C3-C4 alkanes in Uzbekistan. Lack of highly efficient catalysts is a limiting factor[4-8]. In most cases, the proposed catalysts for pyrolysis processes consist of various individual and complex oxides, which are part of zeolites and ceramics.

Research on catalytic pyrolysis of individual hydrocarbons and technical mixtures made it possible to form several laws that are important for determining the mechanism of the process [9-15]. It was found that hydrocarbons of the same class (alkanes, alkenes) of these compounds do not interact, and the composition of catalytic pyrolysis products obeys the rule of additivity [16-23]. Aromatic hydrocarbons accelerate the process, ethylene has no effect and propylene inhibits the decomposition of alkanes [24-26]. The study of the kinetics of catalytic pyrolysis of alkanes and their mixtures showed that the order of the reaction is one [27-31] or one and a half [32-36], depending on whether the chains are broken by the recombination of different or the same radicals. The process of thermal pyrolysis of alkanes is characterized by a first-order reaction [37-43]. The activation energy of catalytic pyrolysis of alkanes varies widely depending on the type of catalyst. For example, the pyrolysis activation energy of various hydrocarbons in the presence of potassium vanadate and stannate is in the range of 167-230 kJ/mol [44-48].

The laws of the catalytic pyrolysis process were studied on the example of propane, n-butane, ethylene, propylene and their mixtures [49-53].

Since there are many publications on catalysts for the pyrolysis of hydrocarbons, we will only consider catalysts for the pyrolysis of lower alkanes [$C_2$-C<]. The proposed catalysts are composed of various individual and complex oxides [54-58], ceramics [59-63], and cement [64-68], which are part of zeolites.

## Experimental part

In the calculations, it was assumed that the dependence of the amount of moles of the component on the peak surface is linear. The composition of the products of the decomposition reaction of the propane-butane mixture in the absence of air and under the influence of heat and breaking the C-C and C-H bonds was calculated according to the following scheme:

a) The surface area (height) was recalculated to the amount of substance according to the following formula:

$$n_{ef} = \frac{S}{k_i}, \qquad n_{ef} = \frac{H}{k_i} \qquad (1)$$

Here, $k_i$ - which determines the sensitivity of the detector to the same component.

b) then, the molar percentages were converted to mass. To recalculate the mole fractions of the components, they are multiplied by the corresponding values of molecular masses:

$$m_i = M_i \cdot \chi_i \qquad (2)$$

c) according to the following equation the mass concentration of the i-component was determined:

$$\omega_i = \frac{m_i}{\sum_i m_i} \times 100\% \qquad (3)$$

g) the degree of change (X) was determined according to the following formula:

$$X = \frac{\omega_{\text{int}} - \omega_{\text{prod}}}{\omega_{\text{int}}} \times 100\% \qquad (4)$$

$\omega_{\text{prod}}$ - the total concentration of propane and butane in the reaction products.

d) the selectivity of the component was found according to the following equation:

$$S_i = \frac{\omega_i}{X} \times 100\% \qquad (5)$$

In addition to the composition of products in the gas mixture, the formation of coke is the process of breaking the S-S and S-N bonds in the propane-butane mixture under the influence of high temperature in an airless place was also conducted in the reactor.

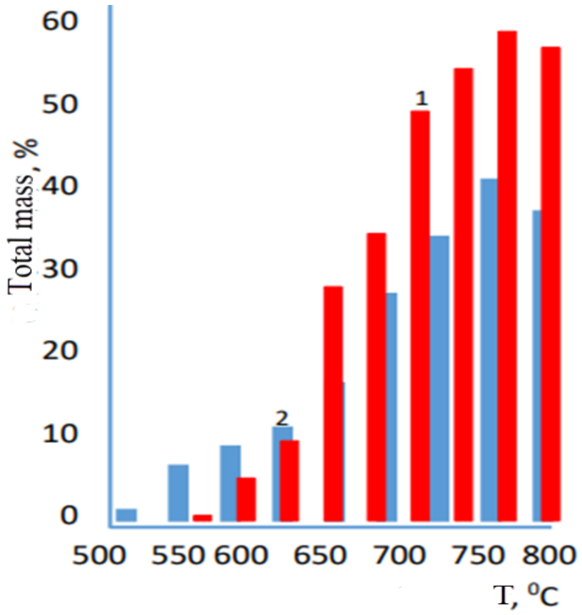
## RESULTS AND DISCUSSION

Kinetic laws of thermal decomposition of propane-butane hydrocarbon mixture. Decomposing the propane-butane mixture in the absence of air at high temperatures in the absence of air in a reactor designed for the process of breaking C-C and C-H bonds has a significant effect on the process of breaking C-C and C-H bonds. The surface of the reactor designed to carry out the process is never inert during the reaction. Therefore, in the pulse and flow systems of the reactor surface designed for the process, the propane-butane mixture is broken under the influence of high temperature in the absence of air, with the breaking of C-C and C-H bonds as a result of decomposition at high temperature without the presence of air in the reactor designed for the process of breaking the C-C and C-H bonds influence on the decomposition process was studied.

From the data in Table 1 and Figures 1, and 2, it can be seen that increasing the process temperature from 500 to 800 °C helped to increase the yield of ethylene and reached the maximum at 780 °C.

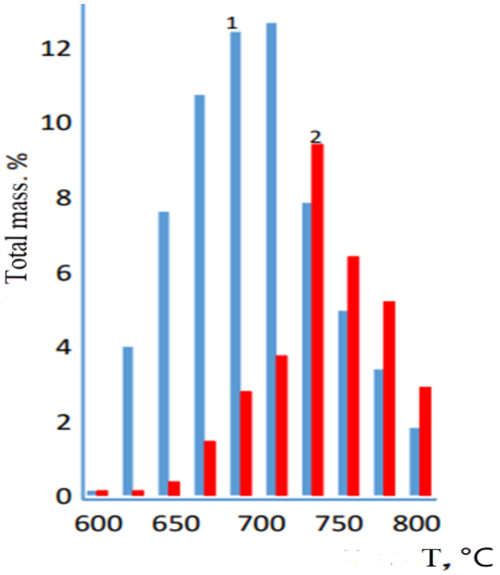
**Table 1. Results of the decomposition reaction with the breaking of C-C and C-H bonds as a result of decomposing a propane-butane mixture at high temperature without the presence of air**

| Contact | T, °C | Propane-butane fraction conversion, % | Productivity, wt.% in relation to transferred raw materials | | | | | Selectivity on $C_2H_4$, % |
|---|---|---|---|---|---|---|---|---|
| | | | $CH_4$ | $C_2H_6$ | $C_2H_4$ | $C_3H_6$ | $\sum C_2$-$C_4$ unsaturated ethylenic series hydrocarbons | |
| An empty reactor designed to carry out the process (τ=6,6s) | 600 | 6.6 | 2.9 | 2.9 | 0.8 | 0 | 0.8 | 12.1 |
| | 700 | 40.1 | 9.5 | 3.6 | 15.9 | 11.1 | 27.0 | 39.7 |
| | 750 | 82.1 | 21.2 | 5.4 | 47.8 | 7.7 | 55.5 | 58.2 |
| | 780 | 98.6 | 28.8 | 5.4 | 60.9 | 3.5 | 64.4 | 61.8 |
| | 800 | 98.5 | 31.2 | 7.6 | 57.9 | 1.8 | 59.7 | 58.8 |
| Reactor filled with metal fragments (τ=4,6 s) | 600 | 6.6 | 2.9 | 2.9 | 0.8 | 0 | 0.8 | 12.1 |
| | 700 | 40.1 | 9.5 | 3.6 | 15.9 | 11.1 | 27.0 | 39.7 |
| | 750 | 82.1 | 21.2 | 5.4 | 47.8 | 7.7 | 55.5 | 58.2 |
| | 780 | 98.6 | 28.8 | 5.4 | 60.9 | 3.5 | 64.4 | 61.8 |
| | 800 | 98.5 | 31.2 | 7.6 | 57.9 | 1.8 | 59.7 | 58.8 |



1 – reactor designed for empty quartz process, 2 – quartz reactor designed for process filled with metal particles

**Figure 1. Ethylene yield in the thermal decomposition of a propane-butane mixture in the absence of air in a quartz reactor designed for the process in the absence and presence of metal fragments**



1 – an empty quartz reactor designed for the implementation of the process, 2 – a quartz reactor designed for the implementation of the process filled with metal particles

**Figure 2. The yield of propylene in the thermal decomposition of a propane-butane mixture in the absence of air in a reactor designed for the implementation of a quartz process in the absence of metal fragments and in its presence**

Previously, the propane-butane mixture in the quartz reactor designed for the process was decomposed at high temperatures without the presence of air in the reactor designed for the C-C and C-H bond breaking under the influence of high temperatures in the absence of air, it is believed that it allows to exclude the effect. A detailed study of the decomposition reaction leading to the breaking of C-C and C-H bonds as a result of decomposing a propane-propane-butane mixture under the influence of high temperature in the absence of air in a quartz reactor designed to carry out the process of breaking C-C and C-H bonds at high temperature in the absence of air, allowed us to assume that it is carried out conditionally in the following two directions through the chain free radical mechanism in the gas phase:

As a sample reaction of the thermal transformation of the propane-butane fraction in an air-free space in a quartz reactor designed for a process filled with chips of 0 = 0.3-0.5 mm (hereafter referred to as "quartz") and as a result of decomposition at high temperature in the reactor without the presence of air under the influence of heat The process of cleavage with the breaking off of C-C and C-H bonds was studied. Table 2 shows the typical experimental results of the decomposition reaction with the breaking of C-C and C-H bonds as a result of decomposing a propane-butane mixture in a helium environment at high temperature in the absence of air in a reactor designed for the process of breaking C-C and C-H bonds at high temperature without the presence of air.

**Table 2. The results of the decomposition reaction of propane-butane mixture in a pulsed system in a helium environment with the breaking of CC and CH bonds under the influence of high temperature in the absence of air**

| Contact | T, °C | Propane-butane fraction conversion, % | Productivity, wt.% in relation to transferred raw materials | | | |
|---|---|---|---|---|---|---|
| | | | $CH_4$ | $C_2H_6$ | $C_2H_4$ | $C_3H_6$ |
| An empty reactor designed to carry out the process (τ=6,6 s) | 600 | 43.0 | 13.7 | 0 | 20.0 | 9.3 |
| | 700 | 69.6 | 22.9 | 0 | 37.5 | 9.2 |
| | 750 | 81.2 | 28.0 | 0 | 45.5 | 7.7 |
| | 780 | 87.7 | 34.9 | 0 | 50.9 | 1.9 |
| | 800 | 87.2 | 37.5 | 1.1 | 47.6 | 1.0 |
| A reactor designed for the implementation of the process filled with metal fragments (τ=9.0 s) | 600 | 43.5 | 23.4 | 2.6 | 17.5 | 0 |
| | 700 | 64.6 | 31.5 | 4.1 | 29.0 | 0 |
| | 750 | 82.3 | 37.4 | 5.2 | 39.7 | 0 |
| | 780 | 94.7 | 41.1 | 5.3 | 46.3 | 2.0 |
| | 800 | 95.7 | 41.9 | 5.5 | 46.5 | 1.8 |
| A quartz reactor designed for the process (τ=0,75 s) | 600 | 31.2 | 6.9 | 1.0 | 12.7 | 10.6 |
| | 700 | 50.0 | 11.2 | 3.0 | 22.1 | 13.7 |
| | 750 | 78.5 | 37.9 | 3.0 | 27.9 | 9.7 |
| | 780 | 89.5 | 43.0 | 4.0 | 34.4 | 8.1 |
| | 800 | 98.8 | 38.3 | 5.0 | 48.0 | 7.5 |

From the data in Table 2, it follows that the yield and ratio of the products of the decomposition reaction of the propane-butane mixture under the influence of high temperature in the absence of air by breaking the C-C and C-H bonds depends on both the contact time and the volume of the contact surface of the propane-butane fraction with quartz (SiO2, SiO2-x (OH)x).

Based on the experimental data on the effect of temperature on the yield of the products of the decomposition reaction of the propane-butane mixture under the influence of high temperature in the absence of air, the activation energies of the gross decomposition processes of the initially saturated hydrocarbons and the formation of reaction products are calculated (Table 3).

**Table 3. Values of the activation energies of the gross processes of the consumption of initial components (propane, butanes) and the assembly of their products in a reactor designed for the process of breaking C-C and C-H bonds under the influence of high temperature in a helium atmosphere in an air-free place of propane-butane mixture**

| Contact | τ, c | $E^{efa}$, kJ/mol | | | |
|---|---|---|---|---|---|
| | | $CH_4$ | $C_2H_4$ | $C_3H_8$ | $\sum C_4H_{10}$ |
| An empty reactor | 6.6 | 62.6 ±3.9 | 85.1 ± 16.5 | 211.9 ± 9.2 | 302.9 ± 19.2 |
| An empty reactor | 9.0 | 37.8 ±3.2 | 77.0 ±8.6 | 179.0 ± 14.9 | 287.6 ± 16.3 |
| Quartz | 0.75 | 35.9 ±2.7 | 109.4 ± 14.8 | 211.6 ±20.7 | 196.6 ±23.0 |

Before the physical adsorption of propane and butane on quartz, it can be assumed that their decomposition into radicals, and then the decomposition process of the propane-butane fraction in a reactor designed to break the S-S and S-N bonds in a general airless space and under the influence of heat, mainly develops on the surface of quartz. The latter is consistent with the results of other studies.

The temperature dependence of the propane-butane fraction decomposition rate constant has the following form:

An empty reactor designed to carry out the process (τ=6.6, k=1.20·1014·exp(-257.40/RT), c–1 (923-1053 K);

An empty reactor designed to carry out the process (τ=9.0 k=3.19·1012·exp(-233.30/RT), c–1 (973-1063 K);

The quartz reactor designed for the process (τ=6.6,

k=1.97·1011·exp(-204.10/RT), c–1 (903-1083 K);

Thus, in the reactor designed for the process of breaking the C-C and C-H bond under the influence of high temperature of the propane-butane mixture in an airless place at 500-800 °C, the formation of coke in the presence and absence of quartz in the helium environment is "suppressed" by the initial hydrocarbons on the quartz surface is shown to represent the catalytic decamp n.

**The effect of the reactor material designed for the process in a reactor designed for the process of breaking the S-S and S-N bonds under the influence of high temperature in an air-free space of a propane-butane mixture in a flow system.** The results of the study are presented in Figures 3, and 4.

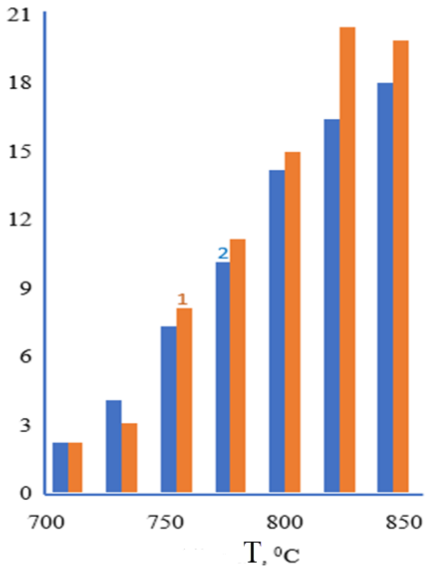


**Figure 3. Propylene yield during thermal decomposition of the propane-butane fraction in the absence of air in quartz (1) and steel (2) reactors**

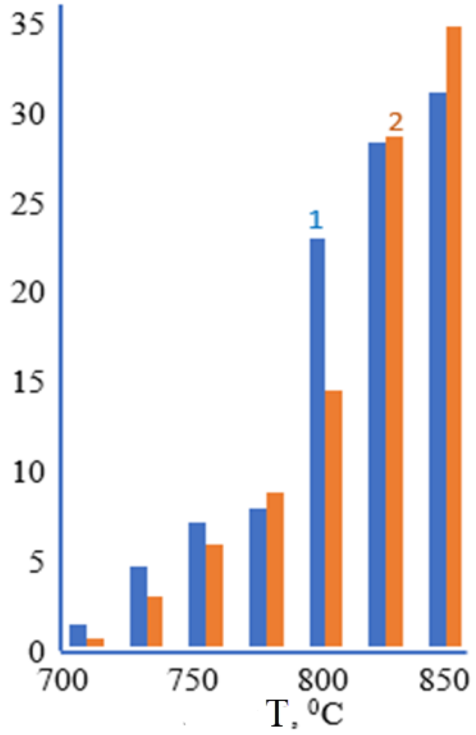


**Figure 4. Ethylene yield during thermal decomposition of the propane-butane fraction in the absence of air in quartz (1) and steel (2) reactors**

The amount of carbon deposits formed on the walls of the steel process reactor is five times higher than that of the quartz process reactor (Table 4).

**Table 4. The selectivity of the decomposition reaction (T=850 ⍰) in the reactor designed for the process of breaking the C-C and C-H bond under the influence of high temperature in the absence of air in the presence of "water vapour" in the flow system**

| Contact | $C_2H_4/CH_4$ | $C_3H_6/CH_4$ | $C_2H_4/C_3H_6$ | a* | cox, weight % |
|---|---|---|---|---|---|
| A quartz reactor designed to carry out the process | 1.70 | 1.04 | 1.63 | 0.71 | 0.23 |
| A steel reactor designed to carry out the process | 2.02 | 0.99 | 2.03 | 0.75 | 1.12 |

Note: a* is the percentage of consumption of propane and butanes involved in the targeted routes.

$\alpha = \frac{n_э}{n_э + n_п}$, $n_э$ **and** $n_п$-- the amount of moles of ethylene and propylene in the reaction mixture at the exit from the reactor designed for the process. Experimental results continue with the formation of ethylene and propylene, in the absence of air and under the influence of heat, and the propane-butane mixture in the absence of air under the influence of high-temperature C-C and C-H bond allows to determine changes in the ratio of the main directions of the decomposition reaction in the reactor designed for the process of breaking the.

The process of breaking the C-C and C-H bonds of the propane-butane mixture in the absence of air under the influence of high temperature was also carried out in a quartz reactor designed for the implementation of the process filled with quartz-quartz pieces Ø = 0.3-0.5 mm (Table 5).

The experimental results presented in Table 5 show that during the decomposition reaction in quartz in the absence of air and under the influence of heat and propane-butane mixture under the influence of high temperature in the absence of air, the contact time increases, the conversion of raw materials and the yield of ethylene increase at the same temperature. , the propylene yield passes through a maximum (~820 °C).

**Table 5. Results of the decomposition reaction of the propane-butane mixture in a flow system with the presence of "water vapour" under the influence of high temperature in the absence of air with the breaking of C-C and C-H bonds**

| Contact | T, °C | Propane-butane fraction conversion, % | Productivity, wt.% in relation to transferred raw materials | | | | | Selectivity on $C_2H_4$, % |
|---|---|---|---|---|---|---|---|---|
| | | | $CH_4$ | $C_2H_6$ | $C_2H_4$ | $C_3H_6$ | $\sum C_2$-$C_4$ unsaturated ethylenic series hydrocarbons | |
| An empty reactor designed to | 730 | 10.5 | 2.7 | 1.5 | 3.3 | 3.0 | 6.3 | 31.4 |
| | 770 | 25.2 | 4.2 | 3.7 | 6.6 | 10.7 | 17.3 | 26.2 |
| | 820 | 53.4 | 10.4 | 3.8 | 22.6 | 16.6 | 39.2 | 42.3 |
| | 840 | 63.3 | 13.5 | 4.8 | 25.6 | 19.4 | 45.0 | 40.4 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| carry out the process (τ=1.5 s) | 880 | 87.5 | 20.7 | 5.2 | 43.5 | 18.1 | 61.6 | 49.7 |
| Quartz reactor designed for process implementation (τ=0.7 s) | 730 | 10.7 | 1.9 | 0.9 | 2.6 | 5.3 | 7.9 | 24.3 |
| | 770 | 31.0 | 5.9 | 3.2 | 8.8 | 13.1 | 21.9 | 28.4 |
| | 820 | 79.4 | 19.6 | 5.3 | 29.4 | 25.1 | 54.5 | 37.0 |
| | 850 | 92.6 | 25.2 | 5.7 | 40.1 | 21.6 | 61.7 | 43.3 |
| | 870 | 97.9 | 32.3 | 7.4 | 46.8 | 12.4 | 59.2 | 47.8 |
| A quartz reactor (τ=6.6 s) designed for the implementation of the process | 730 | 23.9 | 4.2 | 1.8 | 6.3 | 11.6 | 17.9 | 26.4 |
| | 770 | 48.5 | 9.9 | 3.1 | 15.4 | 20.1 | 35.5 | 31.8 |
| | 825 | 85.4 | 19.7 | 5.6 | 39.4 | 20.7 | 60.1 | 46.1 |
| | 845 | 97.7 | 30.1 | 7.0 | 46.6 | 14.0 | 60.6 | 47.7 |

The difference in ethylene production can be explained by the presence of a catalytic effect, as well as the fact that the presence of quartz significantly increases the contact surface (heterogeneous factor S/V (S is the internal surface area of the reactor designed for the implementation of the process, V is the volume of the reactor designed for the implementation of the process) from 10 times increases more). Based on the radical chain mechanism, it can be assumed that the rate of heterogeneous decomposition of saturated hydrocarbons increases with increasing S/V at low temperatures and the rate of heterogeneous chain termination increases at high temperatures.

Based on the obtained results, activation energies were determined (Table 6). It should be noted that these activation energies are effective quantities associated with the entire process (decomposition of propane and butanes, formation of methane and ethylene). Calculation of the effective activation energy, on the one hand, the process of breaking the S-S and S-N bond under the influence of high temperature in the airless place of propane-butane mixture was also carried out in a quartz reactor filled with quartz-quartz fragments Ø = 0.3-0.5 mm ng homogeneous- allows to clarify the heterogeneous mechanism, on the other hand, to obtain comparative information about the efficiency of catalytic systems.

**Table 6. Values of the activation energies of the gross processes of the consumption of the main initial components (propane, butanes) and the assembly of their products in the decomposition reaction with the breaking of C-C and C-H bonds as a result of the decomposition of the propane-butane mixture at high temperature in the flow system with the presence of "water vapour"**

| Contact | t, s | $E^{efa}$, kJ/mol | | | |
|---|---|---|---|---|---|
| | | $CH_4$ | $C_2H_4$ | $C_3H_8$ | $\sum C_4H_{10}$ |
| An empty reactor | 1.5 | 147.8 ±5.5 | 140.3 ± 4.6 | 168.5 ±6.1 | 140.4 ± 4.2 |
| An empty reactor | 0.7 | 175.9 ±7.0 | 185.7 ±6.2 | 230.6 ± 7.2 | 273.4 ± 9.4 |
| Quartz | 6.6 | 139.3 ± 4.2 | 172.5 ±3.6 | 178.9 ±3.9 | 205.9 ±6.1 |

The temperature dependence of the propane-butane fraction decomposition rate constant is expressed by the following equations:

An empty reactor designed to carry out the process (t=1.5s)k=1.20·1014·exp(-257.40/RT), c–1 (923-1053 K);

An empty reactor designed to carry out the process (t=0.7s)k=3.19·1012·exp(-233.30/RT), c–1 (973-1063 K);

Quartz reactor designed for the process implementation (t=6.6s)k=1.97·1011·exp(-204.10/RT), c–1 (903-1083 K);

Kinetic laws of the decomposition reaction in a reactor designed to break the S-S and S-N bonds under the influence of high temperature in a vacuum of propane-butane mixture. From the results of the kinetic study, it follows that the decomposition reaction of lower molecular saturated hydrocarbons at atmospheric pressure, at moderate temperatures (up to 750 °C) in airless space and under the influence of heat proceeds to the first order, since the values of ln(1/1-X) increased linearly with increasing X ( Figure 5).

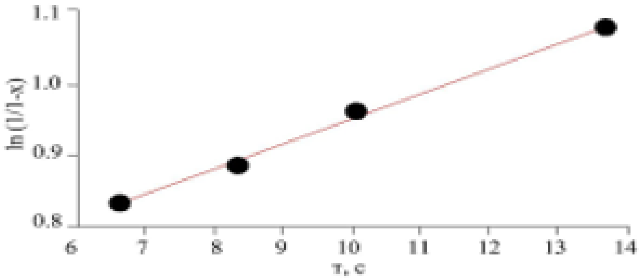


**Figure 5. In an empty quartz reactor at T=700°C, the propane-butane mixture is decomposed by breaking the C-C and C-H bonds under the influence of high temperature in an airless place.ln(1/1-X) as a function of contact time**

Based on the dependence of lnk and 1/T in Arrhenius coordinates (Figs. 6, 7), CC and The dependence of the effective activation energies of the decomposition reaction with the breaking of C-H bonds and their decomposition rate constant was determined:

Propane k=8.81·1015·exp(-243.70/RT), c–1 (913-1013 K);

Butane k=2.00·1014·exp(-268.60/RT), c–1 (973-1013 K).

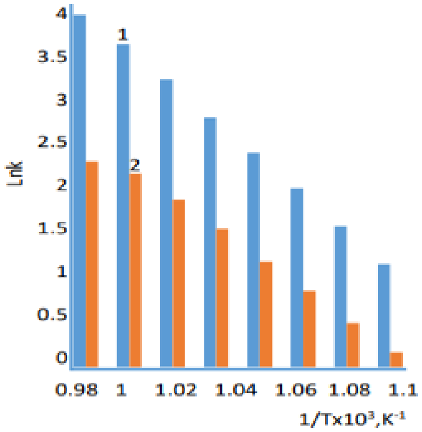


**Figure 6. Semi-logarithmic anamorphism of temperature dependences of effective rate constants (c–1) of propane decomposition reactions, coupled reactions of ethylene and methane formation in an empty reactor designed to implement the process**

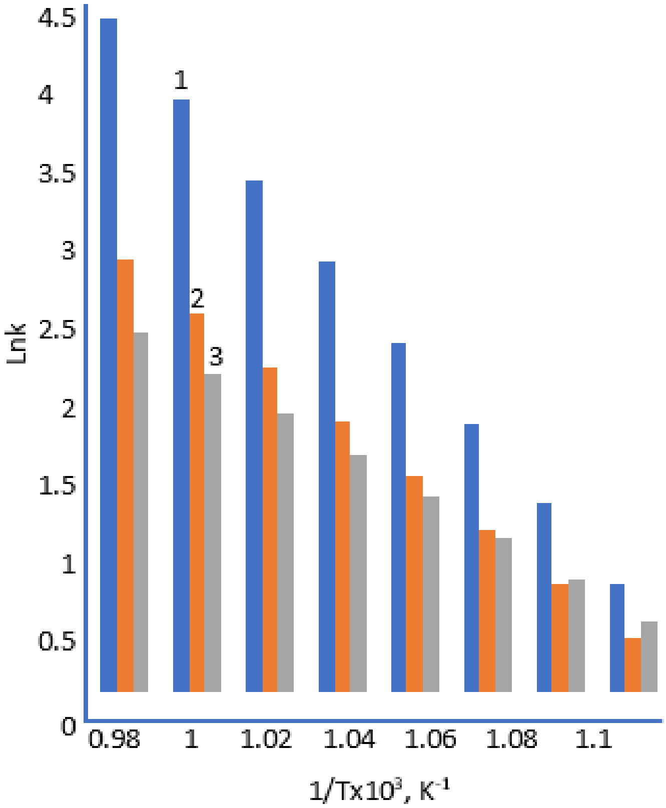


**Figure 7. Semi-logarithmic anamorphism of effective rate constants (c–1) of temperature dependences of n-butane decomposition reactions, adjacent reactions of ethylene and methane formation in an empty reactor designed to implement the process**

**Table 7. Values of the activation energies of the gross processes of the consumption of the main initial components (propane, butanes) and the accumulation of their products in the decomposition reaction with the breaking of C-C and C-H bonds in an empty quartz reactor**

| Hydrocarbon | t, s | $E^{efa}$, kJ/mol | | | |
|---|---|---|---|---|---|
| | | SN4 | S2N4 | S3N8 | ∑S4N10 |
| Propane | 6.6 | 185.7 ±6.5 | 205.5 ± 10.0 | 243.7 ± 11.4 | - |
| Bhutan | 6.6 | 132.5 ±5.6 | 176.1 ± 8.1 | - | 268.6 ± 13.7 |

The values of activation energies obtained from experimental data were compared with those found theoretically using reference values of binding energies based on the assumption of transition state formation in quartz.

## CONCLUSIONS

Thus, it was shown that the catalytic

decomposition of primary hydrocarbons on the surface of quartz in the presence and absence of quartz in a reactor designed for the cleavage of C-C and C-H bonds in the absence of air at 500-800 ° C represents the formation of coke in the presence and absence of quartz in a helium environment. Based on the relationship between lnk and 1/T in Arrhenius coordinates, the relationship between the effective activation energies of the decomposition reaction with the cleavage of C-C and C-H bonds and their decomposition rate constants were determined as a result of the decomposition of propane-butane mixtures in a reactor designed for the cleavage of C-C and C-H bonds in the absence of air at high temperatures.

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