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Research Article

MECHANISM OF PROPANE-BUTANE FRACTION DECOMPOSITION PROCESS INTO LOW MOLECULAR SATURATED AND UNSATURATED HYDROCARBONS

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ABSTRACT

The decomposition of propane-butane fractions into low molecular weight saturated and unsaturated hydrocarbons is a key process in petrochemical production. This study explores the kinetic parameters and reaction mechanisms using reactors designed for impulse and continuous flow operations. The results indicate that the decomposition reactions follow first-order kinetics under specific experimental conditions. Conducted at temperatures ranging from 400°C to 700°C with catalysts, the experiments reveal that the process occurs at two distinct active catalytic sites (Z and Z). The mechanism involves the formation of surface radical complexes (C-Z), which govern the reaction pathways. Depending on the stability of these complexes, the process proceeds through either sequential hydrogen detachment, leading to carbon formation on the catalyst surface, or hydrogen recombination from the gas phase with the C-Z complex, resulting in methane production. This study provides valuable insights into the decomposition mechanisms of propane-butane fractions and lays the groundwork for optimizing catalytic processes to enhance the yield of desired low molecular weight hydrocarbons.

KEYWORDS

Propane-butane fractions, hydrocarbon decomposition, catalytic process, kinetic parameters, active catalytic sites, surface radical complexes, first-order kinetics, methane formation, carbon deposition, impulse reactors, continuous flow reactors, petrochemical production.

INTRODUCTION

Unsaturated hydrocarbons of the ethylene series are among the most critical products in primary organic synthesis. They serve as key intermediates in numerous organic synthesis processes and form the foundation for producing high-tonnage polymers [1–7]. Products derived from these hydrocarbons are widely utilized across various branches of the chemical industry. One of the primary industrial applications of light unsaturated ethylene hydrocarbons, such as ethylene and propylene, is the production of polyethene and polypropylene, as well as their oxides. Due to the rapid growth in the demand for ethylene and propylene, there is a constant search for reliable sources of inexpensive raw materials and more efficient production technologies [8–14].

The primary industrial method for producing propylene involves the thermal decomposition of liquid hydrocarbon raw materials, such as naphtha. However, the predominant product of naphtha thermal cracking is ethylene. Additionally, a growing share of global ethylene production comes from the thermal cracking of ethane, which is derived from natural gas. This shift highlights the increasing urgency to develop targeted processes for producing propylene from more affordable and accessible gas-based raw materials [15–19].

Breaking carbon-carbon bonds in light alkanes through homogeneous selective oxidation is considered a promising direction for the development of targeted propylene production processes [20–27]. Catalytic oxidation of carbon-carbon bonds offers significant advantages over non-catalytic methods [28–31]. Interestingly, the potential for enhancing propane's thermal decomposition efficiency by adding unsaturated compounds like ethylene has not been extensively studied [32–38]. Experimental results show that during the co-oxidation of propane and ethylene, the yield of propylene significantly increases compared to the oxidation of propane alone [39–43].

This process can be organized to enable a highly efficient direct production of propylene from propane. These findings suggest a promising potential method for producing propylene and other unsaturated hydrocarbons of the ethylene series [44–47].

Experimental part

According to the theory of thermal decomposition of hydrocarbons, the decomposition of saturated hydrocarbons under the influence of heat in the absence of air and propane-butane mixture under the influence of high temperature in the absence of air with the presence of a reactor designed to carry out the process of breaking the C–C and C–H bonds corresponds to the process.

For kinetic studies, the use of reactors designed for the implementation of a pulsed process determines that the equation for determining the kinetic parameters is applicable only for first-order reactions (valid for the ideal displacement regime).

When calculating the abstract rate constant of the decomposition of hydrocarbon raw materials in the reactor, the following formula (1) was used for the ideal compression mode of products into the reaction zone in the reactor designed for the process with the pulsed introduction of raw materials and the reactor designed for the flow process.

$$k_{ef} = \frac{1}{\tau} ln \frac{1}{1-x_i}, c^{-1} \qquad (1)$$

Here, x_i – is the degree of modification of the initial hydrocarbon,

τ – is the contact time determined by the ratio of the volume of the reactor designed for the implementation of the process and the volume velocity of the flow passing through the reactor designed for the implementation of the process.

The thermal effect of the reaction of the decomposition reaction with the breaking of C–C and C–H bonds in the absence of air and under the influence of heat and the propane-butane mixture under the influence of high temperature in the absence of air was determined according to the following formula (2):

$$\Delta H = 290.5(1-x_i)+263.8_i \qquad (2)$$

Here, x_i is the degree of modification of the initial hydrocarbon.

The activation energy was estimated from the experimental data using the least squares method (3). For this, the Arrhenius equation was used in logarithmic form:

$$lnk_{ef} = lnA - \frac{E_a^{ef}}{RT} \qquad (3)$$

where A is an old-exponential multiplier;

E_a^{ef} - effective activation energy;

R is the universal gas constant (8.31 J/mol·K).

Solving the problem of linear regression in coordinates - $lnk_{ef} - \frac{1}{T}$, A and $\frac{E_a^{ef}}{R}$ parameters found.

RESULTS AND DISCUSSION

Development of a physicochemical model of the pyrolysis process. To explain the observed changes in hydrogen and methane concentrations in the reaction products, it is necessary to have an idea about its intermediate stages, which take place in the presence of active centres in a catalyst with high catalytic activity and productivity. Based on the obtained experimental data, it can be assumed that the decomposition of hydrocarbons in the presence of catalysts in the temperature range of 400-700°C occurs in two types (Z and Z) of active catalytic centres. This process is observed by the formation of surface radical complexes C-Z. Depending on the dominance of the resulting C-Z (active centre - carbon), either the sequential separation of hydrogen atoms through the formation of carbon on the surface of the catalytic particle or the joining of active hydrogen atoms from the gas phase to C-Z with the formation of methane can occur. Decomposition of hydrocarbons can occur due to the breaking of C-C bonds with the formation of CH_x complexes on the surface of catalyst particles. In the same work, it was shown that subsequent dehydrogenation of surface CH_x complexes can lead to the formation of hydrogen and carbon. The hypothesis of two different types of active sites present on the surface of a catalyst with high catalytic activity and productivity is stated for the first time in the present thesis work.

Depending on the strength of C-Z (active centre - carbon) bonds, two parallel (competitive) processes can occur in C-Z complexes. With the formation of carbon on the surface of the catalytic particles in the Z centres with stronger C-Z bonds, sequential separation of hydrogen atoms occurs in the gas phase:

$$Z\bullet CH_3 \rightarrow Z\bullet CH_2 + \bullet H$$
$$Z\bullet CH_2 \rightarrow Z\bullet CH + \bullet H \; Z\bullet C \rightarrow Z + C$$
$$Z\bullet CH \rightarrow Z\bullet C + \bullet H$$

The carbon formed on the surface of the catalytic particle dissolves in its volume and separates on the opposite side according to the carbide cycle mechanism. Molecular hydrogen is formed by the interaction of active hydrogen atoms with each other or with the initial hydrocarbon molecules:

$$\bullet H + C_3H_8 \rightarrow H_2 + \bullet C_3H_7 \bullet H + \bullet H \rightarrow H_2$$

At Z centres with less strong C-Z bonds, hydrogen atoms can bond to form methane from the gas phase:

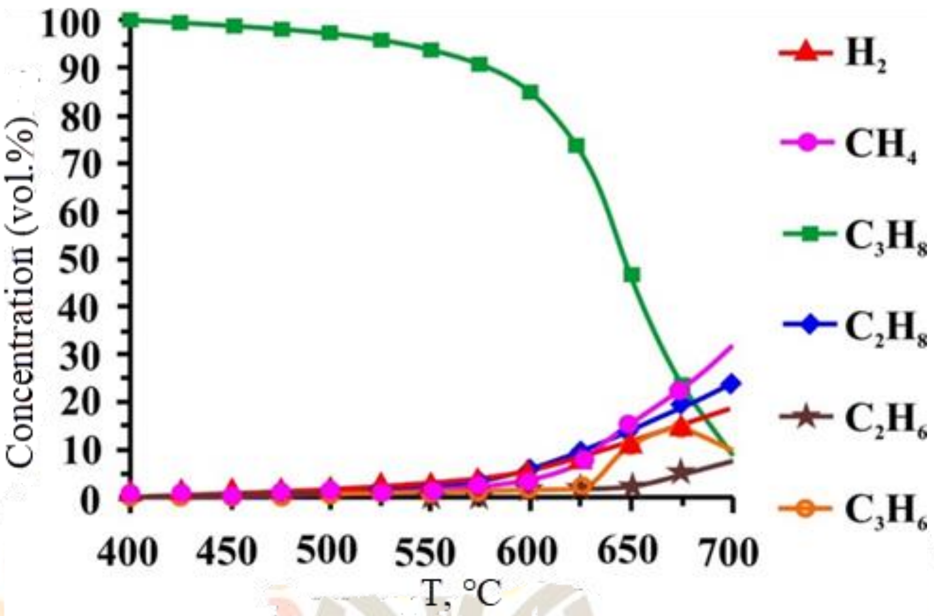
$$Z \bullet CH + \bullet H \rightarrow Z \bullet CH_2$$
$$Z \bullet CH_2 + \bullet H \rightarrow Z \bullet CH_3 \quad Z \bullet CH_3 + C_3H_8 \rightarrow Z + CH_4 + \bullet C_3H_7$$
$$Z \bullet CH_3 + \bullet H \rightarrow Z + CH_4$$

So, the total reaction looks like this:

$$C_3H_8 \rightarrow 2C + CH_4 + 2H_2$$

The proposed scheme of the catalytic reaction is confirmed by the evidence that nickel catalysts with high catalytic activity and productivity are the most active for methanation reactions. It is for this reason that the selectivity of the process of propane to hydrogen conversion is strongly dependent on the degree of deactivation of the catalyst with high catalytic activity and productivity during the reaction.

Decay of propane into lower molecular saturated and unsaturated hydrocarbons with the breaking of C–C and C–H bonds. Figure 1 shows the experimental results obtained in a laboratory reactor with a volumetric consumption of hydrocarbons of 1 l/h on the composition of propane decomposition reaction products without the presence of a catalyst with high catalytic activity and productivity. The temperature must be much higher than 700°C to obtain much higher concentrations of hydrogen by breaking down the thermal C–C and C–H bonds of propane into lower molecular saturated and unsaturated hydrocarbons.



Picture 1. Temperature dependence of the concentrations of the decomposition products into lower molecular saturated and unsaturated hydrocarbons with the breaking of S3N8 thermal C–C and C–H bonds in a laboratory reactor

When alkanes break down into lower molecular saturated and unsaturated hydrocarbons with the breaking of C–C and C–H bonds, the initial formation of the chain occurs due to the breaking of CC bonds, which are more blunt than S-N bonds in the gas phase.

The process of thermal decomposition of propane (Fig. 1) can be described by the following simplified scheme. Initially, under the influence of temperature, with the formation of primary free radicals (chain initiation), the breaking of the CC bond occurs:

$$C_3H_8 \rightarrow \bullet CH_3 + \bullet C_2H_5$$

Resistant to decomposition, but extremely reactive methyl (-CH_3) and ethyl (-C_2H_5) radicals react with the initial molecules of propane, removing a hydrogen atom from them:

$$\bullet C_2H_5 + C_3H_8 \rightarrow C_2H_6 + \bullet C_3H_7$$
$$\bullet CH_3 + C_3H_8 \rightarrow CH_4 + \bullet C_3H_7$$

As a result, methane, ethylene and propyl radicals (-C_3H_7) are formed. Large, relatively unstable radicals spontaneously form more stable methyl radicals (-CH_3) and ethylene molecules (when C-C bonds are broken) or hydrogen atoms (-N) and propylene molecules (when C-H bonds are broken) according to the β-rule decomposes with:

$$\bullet C_3H_7 \rightarrow C_2H_4 + \bullet CH_3$$
$$\bullet C_3H_7 \rightarrow C_3H_6 + \bullet H$$

For the ethyl radical, decomposition according to the β-rule is impossible, this radical can decompose with the separation of a hydrogen atom and the formation of ethylene:

$$\bullet C_2H_5 \rightarrow C_2H_4 + \bullet H$$

Atomic hydrogen, in turn, is reduced to molecular hydrogen by removing another hydrogen atom from the original propane molecule:

$$\bullet H + C_3H_8 \rightarrow H_2 + \bullet C_3H_7$$

There is also the possibility of interaction of hydrogen atoms with each other or with methyl radicals (radical recombination) with the formation of molecular hydrogen and methane:

$$\bullet H + \bullet H \rightarrow H_2$$
$$\bullet H + \bullet CH_3 \rightarrow CH_4$$

In the disproportionation reaction of methyl and ethyl radicals, the formation of methane along with ethylene is not an exception:

$$\bullet CH_3 + \bullet C_2H_5 \rightarrow CH_4 + C_2H_4$$

Thus, at the time of low feedstock in the reactor, with the breaking of thermal C–C and C–H bonds of propane, decomposition into lower molecular saturated and unsaturated hydrocarbons occurs with the formation of hydrogen, methane, ethylene and small amounts of ethane and ethylene in the gas phase. The mechanism of hydrogen formation includes several stages. The process of methane formation goes parallel to the hydrogen formation process. It should be noted that at temperatures above 600°C, with the breaking of the thermal C–C and C–H bonds of propane, during decomposition into lower molecular saturated and unsaturated hydrocarbons, pyro-carbons begin to separate from the gas phase, which settles in the form of solids on the walls of the reactor. After the experiments, the inner surface of the reactor is completely covered with pyro-carbon.

Degradation of normal butane into lower molecular saturated and unsaturated hydrocarbons with the breaking of C–C and C–H bonds. Similar results were obtained in the study of the thermal decomposition of butane under the same conditions. As in the case of propane, the products of the decomposition reaction of butane are hydrogen, methane, ethylene, ethylene and propylene. The curve of the change of propylene concentration will have a similar appearance as for the decomposition of propane into lower molecular saturated and unsaturated hydrocarbons with the breaking of C–C and C–H bonds. The difference is that the decomposition of the butane molecule begins at a lower temperature (450 °C) with the formation of hydrogen and propylene. The concentration of ethylene also exceeds the concentration of ethane in all considered ranges of temperatures. The concentration, however, is low and does not exceed 11 vol.% even at 750°C.

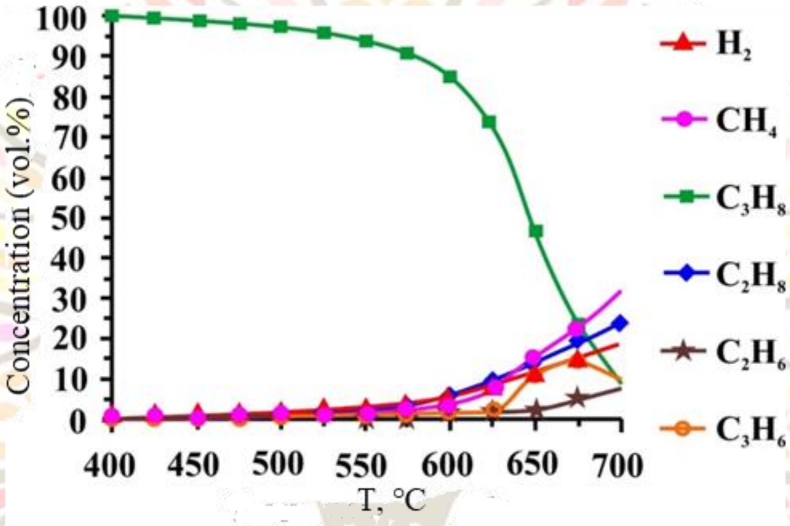


Figure 2. Temperature dependence of the concentrations of the decomposition products of n-butane into lower molecular saturated and unsaturated hydrocarbons with the breaking of thermal C–C and C–H bonds in a laboratory reactor

The distribution of butane cracking reaction products depicted in Fig. 2 can also be explained following the rules of the theory of the radical-chain mechanism of hydrocarbon decomposition mentioned above. Initially, primarily free radicals are formed due to the breaking of C-C bonds in the weakest place (methyl, ethyl and propyl):

$$C_4H_{10} \rightarrow \bullet C_2H_5 + \bullet C_2H_5$$
$$C_4H_{10} \rightarrow \bullet CH_3 + \bullet C_3H_7$$

Then the process develops in two possible directions. Unstable propyl radicals decompose to form more stable methyl and ethyl radicals or hydrogen atoms and the corresponding molecules of alkenes according to the reactions described above. Free methyl and ethyl radicals and active hydrogen atoms interact with the original butane molecules and remove hydrogen atoms from them, resulting in the formation of hydrogen, methane, ethane and secondary butyl (-C_4H_9) radicals:

$$\bullet CH_3 + C_4H_{10} \rightarrow CH_4 + \bullet C_4H_9$$
$$\bullet C_2H_5 + C_4H_{10} \rightarrow C_2H_6 + \bullet C_4H_9$$
$$\bullet H + C_4H_{10} \rightarrow H_2 + \bullet C_4H_9$$

Butyl radicals, being more unstable than propyl radicals, can cleave both through C-C and through C-H bonds. In the first case, methyl and ethyl radicals are formed together with ethylene and propylene, and in the second case - hydrogen atoms and butylene molecules:

$$\bullet C_4H_9 \rightarrow C_2H_4 + \bullet C_2H_5$$
$$\bullet C_4H_9 \rightarrow C_3H_6 + \bullet CH_3$$
$$\bullet C_4H_9 \rightarrow C_4H_8 + \bullet H$$

Since butylene was not detected in the reaction products (Fig. 2), it can be suggested that the $\bullet C_4H_9 \rightarrow C_4H_8 + \bullet H$ reaction almost does not occur, because the butyl radicals were mainly decomposed by the much weaker C-C bond under these conditions. Indeed, in butyl radicals, the homolysis energy of the C-H bond (364 kJ/mol) significantly exceeds the homolysis energy of the C-C bond (264 kJ/mol).

CONCLUSIONS

Thus, at the time of low feedstock in the reactor, with the breaking of thermal C–C and C–H bonds of propane, decomposition into lower molecular saturated and unsaturated hydrocarbons occurs with the formation of hydrogen, methane, ethylene and small amounts of ethane and ethylene in the gas phase. The mechanism of hydrogen formation includes several stages. The process of methane formation goes parallel to the hydrogen formation process. It should be noted that at temperatures above 600 °C, with the breaking of the thermal C–C and C–H bonds of propane, during decomposition into lower molecular saturated and unsaturated hydrocarbons, pyro-carbons begin to separate from the gas phase, which settles in the form of solids on the walls of the reactor. After the experiments, the inner surface of the reactor is completely covered with pyro-carbon.

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