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Evaluation of The Efficiency of Selected Catalysts for The Pyrolysis of a Propane–Butane Fraction

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Abstract: This study evaluates the efficiency of selected catalysts for the pyrolysis of a propane–butane fraction under inert, oxygen-free conditions. Experiments were conducted in both pulse and flow reactors within a temperature range of 650–860 °C. Catalyst performance was assessed based on conversion and selectivity. In a quartz reactor, thermal cracking led to high yields of unsaturated hydrocarbons, especially ethylene and propylene, with ethylene selectivity reaching 55% at 730 °C. Flow reactors with Zn-, Cu-, Mn-, and Fe-based coatings showed that the Zn-coated system provided the highest olefin yield (62.2 wt.%) and minimal coke formation (0.24%). Activation energy analysis indicated that Zn required less energy (150–200 kJ/mol), supporting a radical reaction mechanism. The catalytic activity followed the order: Zn > Cu > Mn > Fe. The enhanced performance of Zn is attributed to its low oxidation state and hydrogenation capability, which suppresses coke formation. These results demonstrate the critical role of catalyst material and reaction environment in maximising olefin production during propane–butane pyrolysis.

Keywords: Propane–butane, catalytic activity, ethylene, coke, quartz reactor.

Introduction:

Currently, the efficient processing of hydrocarbon feedstocks is of critical importance in both the industrial and energy sectors. Pyrolysis of propane–butane fractions under oxygen-free, high-temperature conditions enables the production of valuable compounds such as ethylene, propylene, butadiene, and aromatic hydrocarbons. This process plays a key role not only in the petrochemical industry but also in the manufacturing of polymers, fuels, and organic intermediates [1–3].

Globally, various catalysts are being developed to optimise pyrolysis processes and reduce energy consumption. Catalysts based on nickel, chromium, aluminosilicates, and zinc have been widely applied to improve process efficiency. These catalysts contribute to lowering the required reaction temperature, increasing the yield of target products, and suppressing undesired side reactions [4–8].

In Uzbekistan, research is also being conducted in this direction. The Bukhara Institute of Engineering and Technology is engaged in improving pyrolysis technologies for liquid petroleum products and performing calculations for key processing equipment [9–15]. In addition, studies are being carried out in the field of petroleum and gas refining technologies, focusing on evaluating catalyst performance and implementing innovative technological solutions.

METHODS

Two types of reactor systems were used in this study to carry out the pyrolysis of a propane–butane mixture: a pulse microreactor and a flow reactor. All experiments were conducted in an inert (oxygen-free) atmosphere within the temperature range of 650–860 °C. Reaction conditions such as temperature, residence time, and type of coating were varied to assess their impact on process efficiency.

In the initial stage, thermal decomposition of propane and butane was studied in a blank quartz tube using the pulse reactor, without any catalytic influence. Under these conditions, only thermal cracking occurred due to the high temperature. The results showed that increasing the temperature up to 730 °C significantly improved the conversion of the propane–butane fraction and the selectivity toward olefins—particularly ethylene (C_2H_4) and propylene (C_3H_6), with selectivity reaching up to 55%.

In the next stage, experiments were conducted in a flow reactor equipped with wall coatings of different metals: Zn, Cu, Mn, and Fe. Each metal coating was evaluated based on overall cracking efficiency, combined olefin yield ($C_2H_4 + C_3H_6$), coke formation, and activation energy characteristics.

According to the experimental results, the Zn-coated reactor achieved the highest olefin yield (62.2 wt.%), the lowest coke formation (0.24%), and the lowest activation energy (150–200 kJ/mol). This behaviour is attributed to the low oxidation state and high hydrogenation ability of the Zn surface, which helps suppress coke formation. The overall catalytic performance and inhibitory effect of the metal coatings followed the order: Zn > Cu > Mn > Fe.

RESULTS AND DISCUSSION

The kinetic behaviour of thermal decomposition of a propane–butane mixture under oxygen-free and high-temperature conditions was studied with respect to the reactor wall material. The efficiency of high-temperature pyrolysis in an inert environment is largely influenced by the characteristics of the selected catalyst. Catalyst performance is primarily assessed by its catalytic activity, i.e., the amount of feedstock converted per unit time, and its selectivity, i.e., the ability to produce target products such as ethylene, propylene, and other unsaturated C_2–C_4 hydrocarbons.

The following criteria are commonly used to evaluate catalytic activity:

- Conversion rate (X) – the percentage of the hydrocarbon feedstock that undergoes transformation relative to the initial amount;
- Rate constant (k) – the kinetic parameter characterising the speed of the reaction;
- Yield (Y) – the amount of target product (e.g., g/m^3) such as ethylene or propylene produced;
- Selectivity (S) – the ratio of target product yield to the total amount of products formed.

To determine these parameters, the pulse microreactor method is widely used. In this approach, a short pulse of the propane–butane mixture is introduced into a preheated reactor, and the decomposition results are analysed. This method enables rapid and reliable evaluation of the relative activity and selectivity of catalysts.

During pulse reactions, the propane–butane mixture is introduced at a defined flow rate through a specially designed reactor using a carrier gas. The temperature within the catalyst bed is raised to 500–800 °C, allowing the feedstock to undergo pyrolysis. The reaction products are separated and quantified using gas chromatography at the reactor outlet.

Gas chromatography allows identification of the following components:

- Unsaturated hydrocarbons: ethylene, propylene, butenes;
- Saturated hydrocarbons: ethane, propane, butane;
- By-products: methane, hydrogen, coke, and others.

An essential aspect of catalyst performance is its operational stability, or its ability to retain catalytic properties over an extended period. This parameter is usually assessed through multi-day experiments. A decline in catalyst activity over time is typically attributed to deactivation, fouling of active sites, or coking. In such cases, a layer of graphite-like carbon may form inside the reactor, blocking the active surface of the catalyst.

Optimal Conditions and Coke Formation

To maximise the yield of the target product—ethylene—while minimising coke formation, it is essential to determine the optimal reaction temperature, residence time, and carrier gas flow rate. According to experimental results:

- At 600–700 °C, propane and butane undergo high conversion rates;
- Above 700 °C, selectivity tends to decrease, and coke formation increases.
- A residence time of 2–5 seconds results in the

highest ethylene yields.

In addition, the regeneration capacity of the catalyst was evaluated. A catalyst that can restore its activity after several operating cycles is considered reliable and effective.

Influence of Reactor Material on Thermal Cracking

During the pyrolysis of a propane–butane mixture at high temperatures in an inert atmosphere, the chemical and physical properties of the reactor material play a critical role in determining the reaction efficiency. Specifically, the interaction between the reactor walls or internal inserts and the reaction medium—i.e., heterogeneous surface effects—can influence the mechanism of thermal transformation and the selectivity of the products.

Two reactor configurations were compared experimentally:

1. Blank quartz reactor, made entirely of quartz glass without any internal filler;
2. Quartz reactor filled with metal (steel) inserts, where pieces of industrial-grade steel were placed inside the reactor.

Experiments conducted within the temperature range of 500–800 °C showed that the conversion of C_1–C_4 hydrocarbons increased with rising temperature. For instance, at 750 °C:

- The blank quartz reactor exhibited a conversion rate of 82.1%;
- The steel-filled reactor showed a slightly lower conversion rate of 78.5%.

Significant differences were also observed in the yield of unsaturated ethylene series hydrocarbons (C_2–C_3 fractions):

- In the quartz reactor, the total mass fraction reached 55.5 wt.%;
- In the steel-filled reactor, this figure decreased to 43.5 wt.%.

These results highlight the influence of reactor materials on catalytic activity and reaction pathways. Construction materials should not induce unintended catalytic effects. The quartz reactor, being chemically inert, serves as a neutral medium, ensuring high selectivity and reactivity.

Activation Energy Analysis

In experiments carried out in a quartz reactor without any catalyst, the calculated effective activation energies for the thermal decomposition of propane and butane hydrocarbons were as follows:

- Propane: ~197 kJ/mol
- Butane: ~303 kJ/mol

These values, when compared to the C–C bond dissociation energy and the heat of physical sorption on silica gel (~326 kJ/mol), suggest that both propane and butane molecules are relatively prone to thermal transformation under the given conditions.

Effect of Water in the Reaction Medium

The presence of water vapour in the reaction environment was identified as one of the factors reducing the activity of quartz materials. This effect is primarily attributed to chemisorption of water on the quartz surface, which blocks active surface sites and decreases the overall activity of the reactor medium intended for hydrocarbon conversion.

Kinetics of Individual Saturated Hydrocarbons (Propane and Butane)

Studying the thermal decomposition kinetics of the individual components of the propane–butane feedstock—namely propane (C_3H_8) and n-butane (C_4H_{10})—is essential for understanding their respective reaction mechanisms and optimising reactor parameters.

In this section, the temperature-dependent behaviour of propane and butane was examined separately. Experiments were conducted in a blank quartz reactor to eliminate heterogeneous surface effects and ensure an inert environment. The objective was to assess the individual kinetic behaviour of each component in the mixture by isolating their contributions.

Experimental Conditions and Methodology

The pyrolysis of propane and n-butane was carried out at high temperatures (650–730 °C) under oxygen-free conditions. The reaction products were identified and quantified using gas chromatography, with both qualitative and quantitative analyses performed.

Table 1 presents the major products formed at various temperatures and their corresponding concentrations (in mass %):

Table 1. Thermal Cracking Results of Propane and n-Butane (in a Blank Quartz Reactor)

Hydrocarbons (wt.%)	Temperature (°C)							
		650	680	700	730	650	680	700
		C_3H_8				н- C_4H_{10}		

CH_4		2.0	3.4	9.3	18.8	3.8	7.8	15.9
C_2H_6		0	0.2	0.7	2.1	0.2	0.6	1.2
C_2H_4		2.9	6.6	15.8	40.4	6.7	17.1	34.6
C_3H_6		2.5	4.8	15.1	11.5	8.4	17.0	18.2
Unsaturated C_2–C_3		5.4	11.4	30.9	51.9	15.1	34.7	52.8
Conversion (%)		10.3	15.0	40.9	72.8	19.1	43.1	69.9
C_2H_4 selectivity (%)		28.2	44.0	38.6	55.5	35.1	41.1	49.5

Conversion, Product Distribution, and Reaction Selectivity

Conversion: As the reaction temperature increased, the conversion rates of both propane and butane significantly rose. At 730 °C, propane reached a conversion of 72.8%, while n-butane achieved as high as 97.8%, indicating the greater susceptibility of the n-butane molecule to thermal decomposition.

Formation of Unsaturated Hydrocarbons: Both propane and butane, at 700–730 °C, led to the formation of unsaturated hydrocarbons such as ethylene (C_2H_4) and propylene (C_3H_6). The mass fraction of unsaturated hydrocarbons from butane reached 68.2 wt.%, which is higher than that from propane (51.9 wt.%).

Selectivity: The selectivity toward C_2H_4 in both cases ranged between 40% and 55%, peaking at 730 °C. While butane's C_2H_4 selectivity was slightly lower (51.4%) than propane's, its higher conversion rate compensated for this difference.

These experimental results clearly demonstrate that the pyrolysis of a propane–butane mixture under high-temperature and oxygen-free conditions proceeds via a radical chain mechanism. The process exhibits both homogeneous and heterogeneous characteristics, where the reactor environment, especially the material of the reactor walls, significantly affects the kinetics and reaction mechanisms.

Key Observations:

- Influence of Reactor Wall Material: In an inert quartz reactor, the reaction occurs predominantly in the gas phase, minimising the formation of undesirable products such as polymers, soot, and coke.
- Reduced Coke Formation: In pulse-mode quartz reactors, radical fragments are rapidly swept out by the carrier gas, lowering the probability of olefin polymerisation and coke deposition.
- Product Selectivity: Quartz reactors enabled the efficient formation of lower unsaturated hydrocarbons (C_2–C_4 olefins) with high yield and selectivity from both propane and butane.

Table 2. Pyrolysis results of a propane–butane mixture (τ = 1.5 s) in a flow system under oxygen-free conditions using selected catalysts at high temperature.

Interaction	T, °C	Propane-butane hydrocarbon mixture 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 molecularly unsaturated ethylene series hydrocarbons	
Zn-retaining coating	740	13.4	1.8	1.6	3.8	6.2	10.0	28.4
	760	20.9	3.0	1.5	6.5	9.9	16.4	31.1
	775	27.9	4.9	1.0	10.2	11.8	22.0	36.6
	800	28.9	4.6	0.9	10.9	12.5	23.4	37.7
	820	42.9	8.7	1.2	17.4	15.6	33.0	40.6
	845	69.9	15.2	1.5	33.8	19.4	53.2	48.4
	855	79.5	14.8	2.5	46.2	16.0	62.2	58.1
Cu-retaining coating	730	8.7	1.5	0	2.8	4.4	7.2	32.2
	760	17.7	3.9	0.6	6.8	6.4	13.2	38.4
	785	34.3	7.4	2.5	12.4	12.0	24.4	36.2
	800	40.3	8.7	2.4	15.1	14.1	29.2	37.5
	840	68.4	17.0	3.5	29.7	18.2	47.9	43.4
	855	81.4	21.8	4.9	36.9	17.8	54.7	45.3
	750	18.6	2.1	0.3	6.0	10.2	16.2	32.3

Mn-sparing coating	770	28.2	4.6	0.4	8.9	14.3	23.2	31.6
	790	37.9	7.2	1.2	13.0	16.5	29.5	34.3
	810	48.2	10.5	2.0	18.6	17.1	35.7	38.6
	830	57.7	11.6	3.5	24.6	18.0	42.6	42.6
	850	74.3	17.6	4.8	31.8	20.1	51.9	42.8
Fe-retaining coating	730	5.1	0.5	2.1	0.7	1.8	2.5	13.7
	760	11.0	1.5	2.1	2.9	4.5	7.4	26.4
	790	17.0	3.2	2.3	4.7	6.8	11.5	27.6
	810	29.9	5.3	2.5	9.7	12.4	22.1	32.4
	845	45.6	9.4	3.1	17.3	15.8	33.1	37.9
	860	68.7	15.5	3.7	29.5	20.0	49.5	42.9
Uncoated steel reactor	730	8.6	1.6	0.5	2.7	3.8	6.5	31.4
	750	21.9	4.1	1.5	7.1	9.2	16.3	32.4
	765	24.5	4.9	2.0	8.4	9.2	17.6	34.3
	790	39.7	8.0	3.9	14.3	13.5	27.8	36.0
	830	63.9	13.2	7.0	28.2	15.5	43.7	44.1
	850	76.0	17.1	7.4	34.5	17.0	51.5	45.4

Influence of Catalyst Coating Composition and Selectivity Analysis

The results above indicate that the nature of the metal in the catalytic film coating has a significant impact on the overall conversion and the efficiency of target product formation, even without fundamentally altering the reaction mechanism. The highest yield of light unsaturated hydrocarbons in the C_2–C_3 range (62.2 wt.%), along with the lowest coke formation rate, was obtained using a zinc-based coating during the high-temperature pyrolysis of a propane–butane mixture under oxygen-free conditions (see Tables 1 and 2).

Additionally, coatings containing Cu and Zn demonstrated increasing propylene yields with rising temperature, followed by a decline due to secondary reactions of the formed products.

As shown in Table 2, the Zn-coated reactor exhibited a higher conversion efficiency for propane and butane into ethylene (conversion factor = 0.81) compared to thermal pyrolysis (conversion factor = 0.75), indicating the catalytic promotion effect of Zn. The overall yield of ethylene and propylene was also higher in the Zn-coated system relative to non-catalytic thermal cracking.

The lowest amount of coke deposition was recorded for the Zn-containing coating, comparable to results obtained in an inert quartz reactor during the pyrolysis of a propane–butane fraction (see Table 3).

Table 3. Selectivity of propane–butane hydrocarbon mixture pyrolysis in a flow reactor under oxygen-free conditions at T = 850 °C.

Interaction	C_2H_4/CH_4		C_3H_6/CH_4	C_2H_4/C_3H_6	a*	Coke, wt.%
The steel reactor is unlined	2.02		0.99	2.03	0.75	1.12
	A steel jacketed reactor designed to decompose the propane and butane mixture fraction by heating it in a vacuum and at high temperature					
Zn-retaining coating	3.12		1.08	2.89	0.81	0.24
Cu-retaining coating	1.69		0.82	2.07	0.76	1.20
Mn-sparing coating	1.81		1.14	1.58	0.70	1.25
Fe-sparing coating	1.90		1.68	1.48	0.70	1.05

Note and Selectivity Coefficient α*: The coefficient **α*** represents the fractional consumption of propane and butane involved in target-oriented reaction pathways (3.1–3.4). It is defined by the equation:

$$\alpha * = \frac{n_e}{n_e + n_p}$$

Where:

- n_e = number of moles of ethylene in the reactor outlet mixture,

- n_p = number of moles of propylene in the reactor outlet mixture.

As seen from Table 3, a key factor influencing catalytic activity is the nature of the metal cation in the film coating.

Based on the combined ethylene and propylene yield, the catalytic activity of metal-containing coatings follows the trend:

Zn > Cu > Mn > Fe

The same sequence is observed when considering the inhibitory effect on coke formation.

The superior catalytic activity of the Zn-containing coating is most likely attributed to zinc's low oxidation potential and high hydrogenation ability, which reduces the formation of polycondensation products. In contrast, other metal oxides (e.g., CuO, MnO_2, Fe_2O_3) may accelerate hydrocarbon aromatisation, leading to increased coke accumulation on the catalytic surface.

Analysis of Kinetic Parameters Based on Table 4

To gain deeper insight into the reaction mechanisms involved in the high-temperature pyrolysis of a propane–butane hydrocarbon mixture under inert (oxygen-free) conditions, the activation energy of the gross process was calculated.

Experiments were performed using reactor walls coated with different metallic films (Zn, Cu, Mn, Fe), and the calculated kinetic data are summarised in Table 4.

Table 4. Activation energies for the pyrolysis of a propane–butane hydrocarbon mixture ($\tau = 1.5$ s)

Reactor coating	CH_4 (kJ/mol)	C_2H_4 (kJ/mol)	C_2H_4 (kJ/mol)	$\sum C_4H_{10}$ (kJ/mol)
Uncoated (steel)	130.0 ± 3.6	164.0 ± 6.5	205.5 ± 8.2	175.9 ± 6.2
Zn-coated	154.8 ± 4.2	149.9 ± 4.2	150.0 ± 5.3	200.8 ± 6.1
Cu-coated	164.8 ± 6.1	158.3 ± 4.9	237.3 ± 8.4	200.1 ± 7.8
Mn-coated	173.7 ± 5.6	154.5 ± 4.7	178.2 ± 8.8	172.8 ± 6.3
Fe-coated	179.2 ± 6.7	188.2 ± 7.1	198.0 ± 7.8	175.9 ± 7.9

Analysis and explanation of activation energies

The activation energies (E_a-eff) presented in this table serve to study the effect of various metal coatings on the reaction rate. It is apparent that metal elements have a heterogeneous effect on the homogeneous-gas phase processes inside the reactor. This effect is characterised by a high energy requirement, especially for Cu and Fe coatings, which indicates a strong inhibition of the reaction.

The lower E_a-eff values achieved with Zn coatings are due to the property of this metal to facilitate radical formation and dissociation reactions. At the same time, Mn coatings also provide moderate activity and form a stable reaction environment.

CONCLUSION

Based on the conducted research, the following conclusions were drawn:

1. The thermal pyrolysis of a propane–butane mixture at elevated temperatures (650–860 °C) under inert conditions proceeds via a radical chain mechanism. In this process, the reactor material and its internal coating play a critical role.

2. Experiments conducted in an empty quartz reactor demonstrated high conversion and selectivity. The inertness of the reactor wall material positively influenced the reaction efficiency.

3. When steel reactors were coated with Zn, Cu, Mn, and Fe, the catalytic activity and selectivity significantly varied. The highest total yield of ethylene and propylene (62.2 wt.%) was observed with the Zn-containing coating.

4. The Zn coating exhibited the lowest activation energies (150.0 kJ/mol for propane), enabling efficient control over the formation of target products. Additionally, it demonstrated the lowest coke formation (0.24%), highlighting its effectiveness.

5. Maximum yield of ethylene and propylene was achieved within the temperature range of 700–730 °C and a residence time of 1.5–5 seconds. Temperatures exceeding 750 °C led to reduced selectivity and an increased likelihood of coke formation.

6. The long-term operational stability and regeneration potential of the catalysts were also evaluated. Zn and Cu coatings retained their catalytic activity well over multiple cycles.

In summary, reactors coated with Zn-based films showed high activity, selectivity, and minimal coke deposition during the pyrolysis of propane–butane mixtures. These results support their application as a promising technological solution for the efficient production of olefins.

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