ENHANCING THE EFFICIENCY OF INDUSTRIAL PYROLYSIS OF STRAIGHT-RUN GASOLINE

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Abstract: *A methodology has been proposed to improve the efficiency of the industrial straight-run gasoline pyrolysis process, in which part of the gasoline deficit is compensated by propane. The procedure consists of two stages. In the first stage, a theoretically substantiated mathematical model of gasoline pyrolysis is applied to determine the operating parameters that maximize the total yield of ethylene and propylene. In the second stage, a mathematical model of propane thermal cracking is used to define the propane feed that provides the maximum yield of target products at the optimal temperature obtained in the first stage (*T_{opt} *= 1123 K).*
Keywords: *gasoline pyrolysis, propane pyrolysis, co-pyrolysis, mathematical model, ethylene, propylene.*

Introduction

Currently, the primary industrial method for large-scale production of ethylene and propylene is steam pyrolysis of straight-run gasoline. However, rising gasoline demand in the national economy and the extensive formation of by-products diminish the efficiency of this method. Therefore, more efficient approaches for the production of ethylene and propylene are required.

In our country, Azerbaijan, two large-capacity units are in operation: a fluid catalytic cracking unit for petroleum fractions and the EP-300 straight-run gasoline pyrolysis unit. Analysis of the composition and quantities of by-products formed in these units – hydrogen, methane, ethane, propane, butane, etc. [1] – as well as literature data on their use as additional additives to the reactor feed to enhance the efficiency of straight-run gasoline pyrolysis [2–5], show that the available quantities of these by-products are sufficient for developing efficient industrial methods for producing ethylene and propylene through the pyrolysis of straight-run gasoline.

The industrial application of this method requires the development of a specialized procedure for its optimal implementation. The purpose of the methodology proposed in this work is to carry out the following sequential steps:

1. Selection and verification of the adequacy of the mathematical model for the straight-run gasoline pyrolysis process carried out at the EP-300 unit of the Ethylene–Polyethylene Plant.
2. Optimization of the process based on a theoretically substantiated mathematical model of straight-run gasoline pyrolysis, taking the total yield of target products (ethylene + propylene) as the optimization criterion.
3. Development of a mathematical model for the propane pyrolysis process based on the combined overall stoichiometric equation and the gross-kinetic equation, together with the heat balance and pressure drop equations.
4. Optimization of the propane pyrolysis process using its mathematical model. The task is to determine the optimal reactor feed to achieve the maximum yield of target products (ethylene + propylene).

5. Calculation of the yield of target products resulting from the combined pyrolysis of gasoline and propane based on their complete mathematical models. Gasoline compensation and savings are achieved through the additional supply of propane, the optimal amount of which was determined at the previous stage.

Study of the pyrolysis process of straight-run gasoline using its mathematical model. The main objective of this stage is to determine the operating parameters of the pyrolysis process of straight-run gasoline that ensure the maximum overall yield of target products (ethylene + propylene). To address the tasks of modeling and optimizing the considered process, it is necessary to employ theoretically substantiated kinetic models that retain accuracy when extrapolating operating parameters. An analysis of the literature on the mathematical description of the pyrolysis of straight-run gasoline [6–9] has shown that, for this process, it is advisable to use a model developed by dividing the feedstock composition into four principal groups of individual hydrocarbons [4].

The composition of the gasoline feed fraction was characterized by the following groups:

1. Paraffin hydrocarbons (P^i) – 38.4 wt.%;
2. Isoparaffin hydrocarbons (monomethyl-substituted, polymethyl-substituted) – 33.5 wt.%;
3. Naphthenic hydrocarbons (N) – 19.8 wt.%;
4. Aromatic hydrocarbons (A) – 8.3 wt.%.

Taking into account the chemical generalization of the initial substances, reaction products, and the most significant chemical transformations, an extended chemical scheme of gasoline pyrolysis was proposed in [3,4]. It includes 4 primary and 3 secondary reactions of the pyrolytic conversion of the feedstock with 14 current components.

$$
\begin{aligned}
&P^1 \xrightarrow{W_1} \nu_1^1 H_2 + \nu_2^1 CH_4 + \nu_3^1 C_2H_4 + \nu_4^1 P_{2-4} + \nu_5^1 C_3H_6 + \nu_6^1 C_4H_6 + \nu_7^1 C_4H_8\\
&P^2 \xrightarrow{W_2} \nu_1^2 H_2 + \nu_2^2 CH_4 + \nu_3^2 C_2H_4 + \nu_4^2 P_{2-4} + \nu_5^2 C_3H_6 + \nu_6^2 C_4H_6 + \nu_7^2 C_4H_8\\
&P^3 \xrightarrow{W_3} \nu_1^3 H_2 + \nu_2^3 CH_4 + \nu_3^3 C_2H_4 + \nu_4^3 P_{2-4} + \nu_5^3 C_3H_6 + \nu_6^3 C_4H_6 + \nu_7^3 C_4H_8\\
&H \xrightarrow{W_4} \nu_1^4 H_2 + \nu_2^4 CH_4 + \nu_3^4 C_2H_4 + \nu_4^4 P_{2-4} + \nu_5^4 C_3H_6 + \nu_6^4 C_4H_6 + \nu_7^4 C_4H_8 + \nu_8^4 A \qquad (1)\\
&C_2H_4 \xrightarrow{W_5} \nu_1^5 C_2H_2 + \nu_2^5 H_2\\
&\nu_1^6 C_2H_4 + \nu_2^6 C_3H_6 + \nu_3^6 C_4H_8 \xrightarrow{W_6} \nu_4^6 A + \nu_5^6 H_2\\
&\nu_1^7 C_4H_6 + \nu_2^7 A \xrightarrow{W_7} C
\end{aligned}
$$

Decomposition processes at low pressures usually proceed as first-order reactions; therefore, the expression for the reaction rate will be:

$$W_j = k_j \cdot P_i \qquad (2)$$

The temperature dependence of the rate constants is represented by the Arrhenius equation:

$$k_j = k_{0j} \exp(-E_j/RT),\ (j=1\div7) \qquad (3)$$

In general, the kinetic model of gasoline pyrolysis for an elementary section of the pyrolyzer can be represented by a set of differential material balance equations

$$\frac{G_s dg_i}{dl} = F\sum_{j=1}^{7} \nu_i^j W_j = F\sum_{j=1}^{7} \nu_i^j k_{0j} \exp\left(-E_j RT\right) \times P \times \left(\frac{G_s g_i}{M_i} \Big/ \sum_{i=1}^{14} \frac{G_s g_i}{M_i} \right) \quad i = 1 \div 14 \qquad (4)$$

Table 1 presents the mass stoichiometric coefficients, activation energy values, and pre-exponential factors of the rate constants [3].

Table 1. The mass stoichiometric coefficients, activation energy values, and pre-exponential factors of the rate constants

Reaction number, j	**Pre-exp. factors.**	**Act. energy**	**The mass stoichiometric coefficients**							
	k_{0j}	E_j, **Dj/mol**	ν_1^j	ν_2^j	ν_3^j	ν_4^j	ν_5^j	ν_6^j	ν_7^j	ν_8^j
1	$0.30 \cdot 10^{14}$	$25.12 \cdot 10^4$	0.01	0.18	0.43	0.08	0.19	0.06	0.05	-
2	$0.20 \cdot 10^{14}$	$25.12 \cdot 10^4$	0.01	0.21	0.29	0.08	0.25	0.08	0.08	-
3	$0.30 \cdot 10^{13}$	$23.02 \cdot 10^4$	0.01	0.27	0.17	0.03	0.30	0.03	0.19	-
4	$0.30 \cdot 10^{16}$	$29.31 \cdot 10^4$	0.01	0.18	0.22	0.06	0.13	0.07	0.04	0.29
5	$0.30 \cdot 10^{12}$	$27.21 \cdot 10^4$	0.80	0.20	-	-	-	-	-	-
6	$0.58 \cdot 10^{8}$	$16.75 \cdot 10^4$	0.25	0.55	0.20	-	-	-	-	-
7	$0.40 \cdot 10^{9}$	$16.75 \cdot 10^4$	0.40	0.60	-	-	-	-	-	-

To obtain a complete mathematical description of the, differential equations of the heat balance and pressure drop, accounting for operational parameters such as the temperature and pressure of an elementary section of the reactor coil, should be added to the kinetic model. The heat balance equation for an elementary pyrolyzer section is expressed as:

$$\frac{dT}{dl} = \frac{4\pi d_n q - \sum_j \Delta H_{Rj} r_j}{\sum_{i=1}^{15} n_i C_{pi}} \qquad (5)$$

The pressure drop for an elementary section of the pyrolyzer was evaluated according to the Darcy–Weisbach equation:

$$\frac{dP}{dl} = -\frac{2\lambda_{tr} R \sum_{i=1}^{15} n_i TG_S}{\pi d_v^3 \cdot g \cdot P} \qquad (6)$$

Term 15 in the summation of equations (5) and (6) represents water vapor, which does not participate in chemical reactions but absorbs part of the system's heat for heating and influences the pressure drop along the pipe.

Equations (4), (5), and (6) represent the complete mathematical model for gasoline pyrolysis.

The model was validated on the EP-300 unit of the Sumgait Ethylene-Polyethylene Plant ('Azerikimya'). The conditions were chosen to reflect the operating modes of the SRT-II industrial gasoline furnace. The results are shown in Table 2.

Table 2. Results of validation testing of the mathematical model of the gasoline pyrolysis process at an industrial facility.

№	**1**		**2**		**3**	
	Gasoline, 14000 kg/h	**Water vapor, 10800 kg/h**	**Gasoline, 15000 kg/h**	**Water vapor, 10800 kg/h**	**Gasoline, 12000 kg/h**	**Water vapor, 12000 kg/h**
	EXP %, mass	**MOD %, mass**	**EXP %, mass**	**MOD %, mass**	**EXP %, mass**	**MOD %, mass**
C_2H_6	3.34	3.49	3.24	3.07	3.89	3.75
C_2H_4	24.43	23.43	26.06	27.62	25.51	23.97
C_3H_8	0.43	0.45	0.48	0.45	0.55	0.58
C_3H_6	10.69	11.23	11.89	11.29	12.53	13.40

ΣC_4	3.59	3.41	3.81	4.03	4.15	3.91

The data show that the relative error in the component-by-component composition of the pyrolysis products does not exceed 4–5%. This demonstrates the adequacy of the proposed model for straight-run gasoline pyrolytic decomposition and confirms its applicability in calculations.

During the optimization of the gasoline pyrolysis process using its mathematical model, the optimal operating parameters were identified, ensuring maximum productivity of the pyrolysis furnace in terms of ethylene and propylene yields. The optimization criterion was expressed as follows:

$$\max Q = \left\{T, P, g_B^0, \theta\right\} \tag{7}$$

under the following constraints applied to the process parameters:

$$\begin{gathered} (1048 \le T \le 1148)\,K; \\ (0.4 \le P \le 0.5)\,MPa; \\ \theta = 1:1;\ 1.1:1; 1.2:1; 1.3:1; 1.4:1; 1.5:1;\ 2:1 \end{gathered} \tag{8}$$

The optimization problem was solved using the sliding tolerance method and Powell's method with the help of the "Poisk" software system [10]. The obtained optimal values were as follows: reaction zone temperature, 1123 K; inlet pressure, 0.42 MPa; and gasoline-to-steam mass ratio, 1.5:1. The process was carried out in a four-inch plug-flow tubular reactor with a total coil length of 138 m. For a feed rate of 16,000 kg/h, the yields were: ethylene – 24.89%, propylene – 13.74%, methane – 15.76%, acetylene – 0.53%, propane – 0.55%, ethane – 3.6%, butadiene – 3.9%, butenes – 4.91%, and hydrogen – 0.93%. Under these conditions, the productivity was Q = 6180 kg/h.

Study of the propane thermal cracking process using its mathematical model. The objective of this stage of the research is to identify the optimal reactor loading that ensures the maximum overall yield of the target products (ethylene and propylene). Using data obtained from the Ethylene-Polyethylene Plant in Sumgait, kinetic studies of the process were carried out under the following constraints on the varying parameters:

$$\begin{gathered} (800 \le T \le 1123)K \\ \left(3000 \le g^0_{C_3H_8} \le 4200\right) kg/h \\ \theta = 1:1,\ 1.5:1,\ 2:1 \\ P_0 = 0.42\ MPa;\ P_f \le 0.195\ MPa \end{gathered} \tag{9}$$

The reactor is a four-inch tubular furnace with a coil length of 180 m and an outer tube diameter of 0.149 m.

Based on the conducted research, a kinetic model of the process was developed. The stoichiometric equations are presented below [11]:

$$
\begin{aligned}
&C_3H_8 \xrightarrow{k_1} CH_4 + C_2H_4\\
&C_3H_8 \xrightarrow{k_2} C_3H_6 + H_2\\
&C_3H_8 + H_2 \xrightarrow{k_3} C_2H_6 + CH_4\\
&C_2H_6 \xrightarrow{k_4} C_2H_4 + H_2\\
&C_3H_8 + CH_4 \xrightarrow{k_5} C_2H_6 + C_2H_4 + H_2\\
&C_3H_6 + H_2 \xrightarrow{k_6} C_2H_4 + CH_4\\
&C_2H_4 + 2H_2 \xrightarrow{k_7} 2CH_4
\end{aligned}
\qquad (10)
$$

The complete kinetic model of the process, corresponding to this stoichiometric scheme, is represented by the following kinetic equations [11]:

$$
\frac{dn_{C_3H_8}}{dl} = -u^{-1} n_{C_3H_8}\left(k_1 + k_2 + k_3\frac{n_{H_2}P}{\left(\sum_i n_i\right)RT} + k_5\frac{n_{CH_4}P}{\left(\sum_i n_i\right)RT}\right)
$$

$$
\frac{dn_{C_3H_6}}{dl} = u^{-1}\left(k_2 n_{C_3H_8} - k_6 n_{C_3H_6}\frac{n_{H_2}P}{\left(\sum_i n_i\right)RT}\right)
$$

$$
\frac{dn_{C_2H_6}}{dl} = u^{-1}\left(k_3 n_{C_3H_8}\frac{n_{H_2}P}{\left(\sum_i n_i\right)RT} - k_4 n_{C_2H_6} + k_5 n_{C_3H_8}\frac{n_{CH_4}P}{\left(\sum_i n_i\right)RT}\right)
$$

$$
\begin{aligned}
\frac{dn_{C_2H_4}}{dl} = u^{-1}\Bigg[&k_1 n_{C_3H_8} + k_4 n_{C_2H_6} + k_5 n_{C_3H_8}\frac{n_{CH_4}P}{\left(\sum_i n_i\right)RT} + k_6 n_{C_3H_6}\frac{n_{H_2}P}{\left(\sum_i n_i\right)RT} -\\
&- k_7 n_{C_2H_4}\left(\frac{n_{H_2}P}{\left(\sum_i n_i\right)RT}\right)^2\Bigg]
\end{aligned}
\qquad (11)
$$

$$
\begin{aligned}
\frac{dn_{CH_4}}{dl} = u^{-1}\Bigg[&k_1 n_{C_3H_8} + k_3 n_{C_3H_8}\frac{n_{H_2}P}{\left(\sum_i n_i\right)RT} + k_6 n_{C_3H_6}\frac{n_{H_2}P}{\left(\sum_i n_i\right)RT} -\\
&- k_5 n_{C_3H_8}\frac{n_{CH_4}P}{\left(\sum_i n_i\right)RT} + 2k_7 n_{C_2H_4}\left(\frac{n_{H_2}P}{\left(\sum_i n_i\right)RT}\right)^2\Bigg]
\end{aligned}
$$

$$\frac{dn_{H_2}}{dl} = u^{-1}\left[k_2 n_{C_3H_8} - k_3 n_{C_3H_8} \frac{n_{H_2} P}{\left(\sum_i n_i\right) RT} + k_4 n_{C_2H_6} \frac{n_{H_2} P}{\left(\sum_i n_i\right) RT} + \right.$$

$$\left. + k_5 n_{C_3H_8} \frac{n_{CH_4} P}{\left(\sum_i n_i\right) RT} - k_6 n_{C_3H_6} \frac{n_{H_2} P}{\left(\sum_i n_i\right) RT} - 2k_7 n_{C_2H_4} \left(\frac{n_{H_2} P}{\left(\sum_i n_i\right) RT} \right)^2 \right],$$

The kinetic constants of the model k_j and E_j were determined using the combined nonlinear programming methods of Rosenbrock, Powell, and McCormick [12, 13]. The resulting constants of the kinetic model are presented in Table 3.

Table 3. Kinetic parameters of the model

Reaction number, j	**Pre-exponential factor, k_{0j}**	**Activation energy, E_j Dj/mol**
1	64260.16	48488
2	25033.76	47234
3	1162.027	34485
4	1.15599	23575
5	1220.584	41716
6	6.0671	17138
7	13086.02	63954

In the pyrolysis process, temperature and pressure change continuously along the reactor, which must be reflected in the full mathematical model. The heat balance equation is expressed as:

$$\frac{dT}{dl} = \frac{\frac{dQ}{dl} - \sum_j r_j \Delta H_{Rj}}{\sum_i n_i C_{pi}} \qquad (12)$$

The pressure change along the length of a tubular reactor was derived using the Darcy–Weisbach equation for a straight circular pipe. The resulting expression is shown below [11]:

$$\frac{dP}{dl} = -0.50962 \cdot 10^{-13} \left(1 + \frac{\psi d_v}{L_0}\right) \frac{\lambda_{tr} \left(\sum_i n_i\right) T}{P d_v^5} \qquad (13)$$

Equations (11), (12), and (13) – representing the kinetic model, heat balance, and pressure loss, respectively – constitute the complete mathematical model of the propane pyrolysis process.

In the study of the process using the developed model, the total yield of the target products (propylene and ethylene), as well as their relative yields with respect to the total reactor feed, were employed as the optimization criterion. The process was analyzed under the aforementioned constraints (9) on the decision variables.

By varying the total reactor load within the specified range at a fixed inlet temperature, we determined the optimal load values corresponding to the maximum yields of propylene and ethylene. The results, presented in the figure, indicate that at T = 800 K, the combined yield of C_3H_6 and C_2H_4

decreases monotonically with increasing fresh feed load $g^0_{C_3H_8}$ and, consequently, with decreasing conversion, with the maximum yield observed at lower load values.

For each subsequent temperature T, the dependence of $g_{(C_3H_6 + C_2H_4)}$ on the fresh load $g^0_{C_3H_8}$ shows a maximum, which shifts toward higher total charge values with increasing temperature. The maximum yield increases with temperature and reaches 4000 kg/h at the optimal gasoline pyrolysis temperature of 1123 K.

Further continuation of the process in the region of high temperatures and total loads proved impossible due to the violation of the constraint on the outlet pressure of the reactor. The shaded area in the figure illustrates this statement. The dashed curve connects the points of maximum product yields for the corresponding temperatures, which are taken as optimal (Fig.).

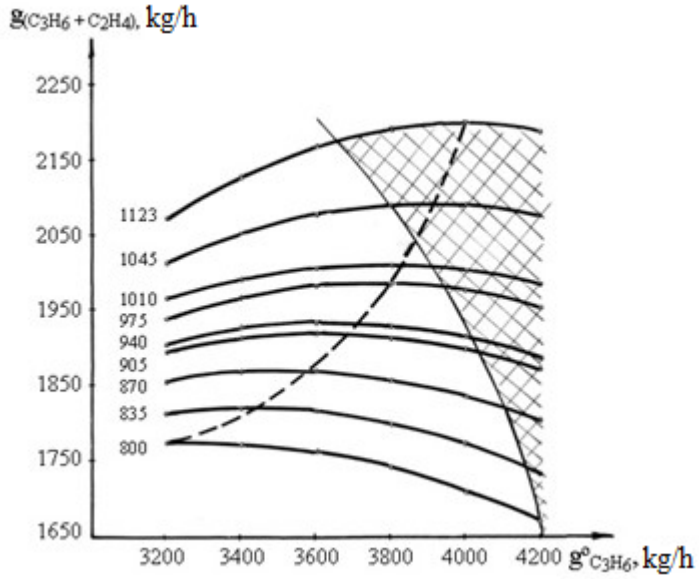

Figure. Determination of optimal inlet temperatures T=(800-1123)^{0}K depending on the total reactor load ($g^0_{C_3H_8}$ =3200-4200 kg/h).

Results of the study on joint pyrolysis of gasoline and propane. Using comprehensive mathematical models for both gasoline and propane pyrolysis, the yields of the main target products from their combined pyrolysis were calculated. The results, obtained under the optimal operating parameters established for gasoline pyrolysis, are presented in Table 4.

Table 4. Comparative analysis of the results of gasoline pyrolysis and combined pyrolysis of gasoline with additional propane supply.

Temperature, ^{0}C	Residence time, sec.	Feedstock, kg/h			Product yield, kg/h				
		Gasoline	Water vapor	Propane	Ethylene	Propylene	Ethane	Propane	ΣC$_4$
850	1	16000	10500	–	3980	2200	570	90	780
850	1	12000	10500	4000	4320	2440	610	730	490

Comparative analysis of the results in Table 4 indicates that, under these conditions, ethylene productivity increases by 340 kg/h and propylene by 240 kg/h when 4.000 kg/h of propane is

substituted for straight-run gasoline. This approach enhances the yield of target products while saving 4.000 kg/h of gasoline.

CONCLUSION

The study explores the co-pyrolysis of straight-run gasoline with propane as a more economical route to ethylene and propylene production. Partial replacement of costly naphtha by propane, supplied from the G-43-107M and EP-300 units in our republic, improves the yield of target olefins and significantly increases profitability through gasoline savings. A method for improving the efficiency of industrial pyrolysis of straight-run gasoline is proposed. Its effectiveness was demonstrated through experiments involving additional propane supply. The study identified an optimal process temperature of 1123 K and an optimal propane feed of 4000 kg/h. Under these conditions, the overall yield of target products increases by about 10%, the gasoline deficit of 4000 kg/h is compensated, and byproduct formation is reduced.

NOTATION

A – aromatic hydrocarbons;
C – high-molecular-weight compounds;
C_{pi} – heat capacity of the i-th component, J/(mol·K);
d_n – outer diameter of the pyrocoil, m;
d_v – inner diameter of the pyrocoil, m;
E_j – activation energy of the j-th reaction, J/mol;
F – cross-sectional area of the coil, m²;
G_s – total feedstock (gasoline), kg/h;
g_i=1–14 – weight fractions of components, dim.less: hydrogen (1), methane (2), ethylene (3), normal paraffinic hydrocarbons C_2–C_4 (4), propylene (5), divinyl (6), butenes (7), aromatic hydrocarbons (8), acetylene (9), high-molecular compounds (10), paraffinic compounds of the feedstock (11), monomethyl-substituted paraffinic compounds of the feedstock (12), polymethyl-substituted paraffinic compounds of the feedstock (13), naphthenic hydrocarbons of the feedstock (14);
$g_{C_2H_4}$, $g_{C_3H_6}$ – yields of ethylene and propylene, kg/h;
$g^0_{C_3H_8}$ – total propane feed, kg/h;
g^0_B – total gasoline feed, kg/h;
ΔH_{Rj} – heat effect of the j-th reaction, J/mol;
k_j – rate constant of the j-th reaction;
k_{0j} – pre-exponential factor of the rate constant of the j-th reaction;
L_0 – length of the straight section of the pipe, m;
l – current reactor length, m;
M_i – molecular weight of the i-th component, kg/kmol
N – naphthenes;
n_i – current molar flow rate of the i-th component in pyro-gas, mol/h;
n_{15} –current molar flow rate of water vapor, mol/h
P – current pressure in the reactor, Pa
P_i – partial pressure of the component;
P^1, P^2, P^3 – normal, monomethyl-substituted, and polymethyl-substituted paraffins;
$P_{2\text{-}4}$ – paraffins with 2–4 carbon atoms;
P_0, P_k – pressure at the inlet and outlet of the reactor, Pa;
Q – reactor capacity for ethylene and propylene, kg/h
q – heat flux on the heating surface, J/(m²·h);
R – universal gas constant, J/(mol·K);
r_j – rate of the j-th reaction, mol/(h·m);

T – temperature, K;
u – linear velocity of the gas flow, m/h;
W_j – rate of the j-th reaction;
θ – mass ratio of feedstock to steam;
λ_{tr} – friction coefficient;
$\nu_{i,j}$ – mass coefficients equal to the weight fraction of the product formed during complete decomposition of the feedstock;
ψ – dimensionless coefficient accounting for local resistances.

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