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Thermal cracking of recycled hydrocarbon gas-mixtures for re-pyrolysis: operational analysis of some industrial furnaces

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Thermal cracking of recycled hydrocarbon gas-mixtures for re-pyrolysis: operational analysis of industrial furnaces

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Abstract
Thermal decomposition process of recycled hydrocarbon gas mixtures in industrial furnaces is analyzed by computer simulation. The detailed kinetic and mathematical model developed was validated by using the process control laboratory cracked gas analysis of an industrially operated furnace. The effects of feed compositions and operational conditions are examined to select the favorable operating parameters and to achieve the possibly highest online operation period of the furnace. The effect of deposited coke on the lifetime of radiant coils is examined by a heat-transfer model. The simulation study confirmed that temporal variations of the feedstock composition could be harmonized well with the operating parameters of furnaces with the purpose of achieving maximum effectiveness.

Keywords: Steam cracking, recycled hydrocarbon gas-mixtures, modeling, simulation, coke formation

1. Introduction
Raw materials for production of most largely used plastics, i.e. polyethylene, polypropylene and PVC are industrially produced by thermal cracking of different individual hydrocarbons such as ethane, propane, n-butane, or mixtures such as LPG, virgin naphtha and gas oil. The product yields, as well as the extent of secondary reactions, i.e. the expected online operation period of furnaces can vary significantly as a function of the feed composition and operational conditions. This is especially so in the case of recycled cracked gases for re-pyrolysis, which can be mixed also with fresh hydrocarbons and may contain a large quantity of coke precursors such as olefins and di-olefins, depending on the effectiveness of hydrogenation process upstream the cracking furnace. When modeling tubular furnaces which are widely

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used also for cracking of recycled gases special attention has to be paid on the secondary reactions leading to production of coke since the coke formation increases the pressure drop and reduces the furnace selectivity.

Investigating this process, Renjun et al. [1], Plehiers et al. [2] and Kopinke et al. [3,4] presented relative rates of coke formation for different hydrocarbons. Heynderickx and Nozawa [5] investigated the influence of high-emissivity coatings on the reactor tubes and furnace walls, Stefanidis et al. [6] presented CFD simulations of steam cracking furnaces, while Heynderickx et al. [7] studied decoking of an ethane cracker by simulation. Jegla et al. [8] developed a method for systematic retrofit of tubular furnaces. Niaei et al. [9] applied the zone method for combined simulation of the furnace and the radiant box, while Sadrameli and Green [10] presented an analytical semi-empirical model to apply as a practical method for predicting conversion. An optimal temperature profile along the furnace was computed by Masoumi et al. [11] by using an objective function taking into account the costs of the decoking period. However, there have not been reported detailed studies concerning steam cracking of recycled gas mixtures to examine interdependency of feed compositions and operational conditions.

The aim of the present work is to study the effects of operating parameters on the product yields and on the furnace on-stream period taking into account the special compositions of the feed available using a kinetic and mathematical model. Numerical experiments are performed with different feed compositions and operational conditions to find the favorable set of operating parameters, and to achieve the possibly highest online operation period of the furnace. The simulation results are compared with the process control laboratory cracked gas analysis of an industrially operated furnace as well as with performance data obtained from daily operational practice.

2. Mathematical model

2.1. Kinetic model

A mechanistic kinetic model was developed on the basis of a detailed reaction network containing 233 individual reactions including those leading to formation of coke and carbon oxides [12]. Kinetic parameters were assigned to each reaction from published and arranged literature data. Validation of the model was performed by comparing its results with those obtained experimentally from cracked gas analysis of an industrially operated furnace, fitting the kinetic parameters to experimentally measured
yield data. Reactions leading to coke formation were considered with their rate constants found in the published literature data [1, 4]. This type of arrangement, adaptation and fitting of frequency factors and activation energies has been presented recently by Gál and Lakatos [14].

2.2. Reactor model

The geometry of the furnace coil and high Reynolds-numbers used in thermal decomposition process enable tubular reactor and plug-flow assumptions. As a consequence, mass, energy and momentum balances can be written as follows [12, 13].

Mass balance:

$$\frac{\partial c_i(x,t)}{\partial t} = \sum_{k=1}^{N_r} \alpha_{ki} r_k(e, T) - v \frac{\partial c_i(x,t)}{\partial x}, \quad i = 1 \rightarrow N_c, \quad k = 1 \rightarrow N_r, \quad (1)$$

where $c_i$ is the concentration of reactant $i$, $x$ is the axial distance along the reactor, $v$ denotes the cracked gas convective velocity, $r_k$ is the rate of the reaction $k$, and $\alpha_{ki}$ stands for the stoichiometric coefficient of component $i$ in the reaction $k$. $N_c$ denotes the number of species, while $N_r$ stands for the number of reactions.

Enthalpy balance:

$$\left( \sum_{i=1}^{N_c} C_{pi} c_i \right) \frac{\partial T(x,t)}{\partial t} = \sum_{k=1}^{N_r} (-\Delta H_k) r_k(e, T) - v \left( \sum_{i=1}^{N_c} C_{pi} c_i \right) \frac{\partial T(x,t)}{\partial x} + AU \left[ T_{fb} - T(x,t) \right] \quad (2)$$

where $T$ is the cracked gas temperature, $C_{pi}$ is the heat capacity of species $i$, $\Delta H_k$ denotes the heat of reaction $k$, $U$ denotes the overall heat transfer coefficient from the firebox to the cracked gas, $d_i$ is the inner diameter of the reactor tube, $A$ denotes the surface area in a unit distance of the tube, and $T_{fb}$ stands for the temperature of firebox.

The balance equations (1) and (2) were solved subject to the following boundary and initial conditions:

$$c_i(0,t) = c_{i,in}(t) \quad i = 1 \rightarrow N_c, \quad T(0,t) = T_{in}(t) \quad (3)$$

$$c_i(x,0) = c_{i,0}(x) \quad i = 1 \rightarrow N_c, \quad T(x,0) = T_{0}(0) \quad (4)$$

Based on the mathematical model (1)-(4), a computer model was developed using the CHEMCAD simulator [15, 16]. On the basis of the large thermo-chemical and kinetic database, CHEMCAD can also be used for mechanistic kinetic modeling of hydrocarbons thermal decomposition. In order to achieve a complete yield structure at the exit of radiant coil the following input data are required for the simulation:
- Operating parameters of the furnace: feed flow rate, steam to hydrocarbon ratio, pressure and temperature at the inlet of radiant section. These data were provided according to real operating conditions of the reference furnace.

- Parameters of the rate expressions of reactions, frequency factors and activation energies, included into the reaction network. These data were applied on the basis of the kinetic model developed [12, 14].

2.3. Heat transfer model

Heat transfer from the firebox of the furnace to process gas-mixture mainly depends on the coke deposited on the inner surface of radiant coil when the operating parameters are stabilized and controlled. For this reason, special attention was paid on coke formation rates from different coke precursors. Due to the rising temperature profile and to the increasing concentration of coke precursors (ex. ethylene and propylene) along the radiant coil a rising coke layer thickness was supposed along the coil in spite of continuously increasing diameter of the coil as it is shown in Fig.1. To check this, the radiant box was divided into five isothermal zones, and the coil was also divided into five sections. This method is a simplified form of that presented earlier by Niaei et al. [9] and Masoumi et al. [11].

As the inner diameter of the radiant pipe and the coke layer thickness are varying, the pressure drop along the radiant pipe also varies that was described using the equation

\[
dp = \left( f \frac{L}{144d_d^2g} + \xi(x) \right) \frac{\rho v^2}{2} \tag{5}
\]

where \( p \) denotes the pressure, \( \rho \) is the density of gas mixture, \( L \) the equivalent pipe length, \( g \) is acceleration due to gravity, \( \xi(x) \) is the local resistance coefficient of reactor tube junctions or bends, and \( f \) denotes the friction factor which is calculated using the expression for smooth pipes:

\[
f = \frac{0.3164}{Re^{0.25}} \tag{6}
\]

Having applied this method, the local coking rates as well as modification of the mean tube skin temperatures and the overall heat transfer coefficient (HTC) could also be followed section by section whilst the reaction heat per tube in each section was accurately calculated by the simulator.
3. Experimental section

The reference furnace, denoted as CF1, consists of a radiant and a convection section, as well as six transfer line exchangers (TLE) with one steam drum. The furnace contains 48 radiant coils connected to one transfer line exchanger (TLE) by eight as it is shown in Fig.1. Firing in the radiant section is performed by sidewall and floor burners, where natural gas is burnt, which is also mixed with some methane fraction formed in the cracking process. Preheating of the feed and dilution steam, as well as preheating and superheating of the high-pressure steam takes place in the convection section. Between the feed pre-heater and steam super-heater a boiler-feed-water pre-heater is placed. Before a transfer line exchanger two small-diameter coils, having diameters 39 mm are connected to a single coil of diameter 57 mm. The mean residence time of the reaction mixture in the radiant section is very short: it takes only 0.3 s.

Figure 1

The process parameters of the furnace, such as temperatures, pressures, flow-rates are monitored by an Advanced Process Control system (APC) and can be registered in accordance with the feed- and cracked gas analysis [14].

The reference furnace processes a hydrogenated C4-C5 mixture that generally contains 60-65% n-butane, 10-15% pentanes and 5-15% unsaturated C4 and C5 compounds. The remaining species are saturated C6 and some propylene. Of course, the feed composition was always analyzed by components. The sampling of the cracked gases at the furnace outlet was performed manually, by cooling the sample down to ambient temperature in order to make the liquid phase condensed. Both the liquid and gas phase were analyzed by lab chromatographs, and the compositions were calculated on the basis of these analyses. At least three samples were taken under similar operational conditions. Comparing the results obtained, the procedure was repeated till the yields of the main components, i.e. methane, ethylene, propylene and residual n-butane agreed with precision ±0.1% in three subsequent analyses. The model was validated by comparing the yields of species resulted in simulation with those obtained from cracked gas analysis under different operational conditions.

Table 1 shows a comparison of the measured and simulated product yields obtained by the validated model involving kinetic parameters adopted and then fitted to the measurement data. It is seen well, that simulated yield data are in good agreement with the measured ones after fitting the kinetic parameters.
Numbers 1, 2 and 3 refer to diverse operational conditions, i.e. different feed compositions and different on-line operation time.

4. Simulation results and discussion

The flow rate and composition of the feed, the coil outlet temperature (COT) and the dilution steam to hydrocarbon ratio (ST/HC) can be considered as independent variables in industrial operation of cracking furnaces. The coil inlet pressure (CIP) and the cross-over temperature (XOT) depend on the status of convection tubes where solid particles can be deposited on both the inner and outer surfaces of the pipes. Since the feed flow rates are always intended to be kept at maximum, simulations were carried out at the highest possible feed rate.

4.1. Effects of the coil outlet temperature (COT)

The purpose of examining the effect of COT was to quantify its influence on product yields and their ratios and variation of coke formation rate. Simulations were executed at five different COT values while three series of results could also be compared with experimentally measured data. Variation of the product yields obtained in simulation runs and that of coke formation rate at different coil outlet temperatures is shown in Fig. 2.

It can be concluded that when cracking such mixtures COT is strongly limited by the exponentially increasing coke formation rate, i.e. by the expected online operation period of the furnace. The trend of ethylene yield also shows that increase of COT above 845ºC is not reasonable either for technological or economical considerations.

4.2. Influence of the dilution steam/hydrocarbon ratio (ST/HC)

It is a question of profitability to operate the furnace at the lowest possible steam/hydrocarbon ratio since a lower steam rate reduces specific energy consumption of the production unit. Basically, on one thing we should decide: up to what extent this ratio can be reduced without having a significant negative effect on product yields and on furnace online period. Simulations were carried out at four ST/HC ratios comparing their influence at two different COT values. Table 2 shows that reducing the ST/HC ratio by 10%,
from the basic design equal to 0.5 has only a small influence on product yields but a shorter runtime can be expected because of higher coke formation rate.

→ Table 2

As Table 2 also shows, there is a significant loss of ethylene by reducing ST/HC ratio by more than 20%, together with a steeply increasing coke formation rate. Neither propylene yield nor the n-butane conversion changes, while the methane formation has a rising trend. It should be noted that effects of the dilution steam reduction cannot be simulated with a high accuracy since the catalytic effect of tube metals are not described by any known kinetics. Nevertheless, practical experiences also confirm the data shown in Table 2.

4.3. Variation of the feed composition

Product yields were examined at three ethane concentration levels while COT was kept constant, as well as at three COT levels with the same ethane concentration. It was stated that applying a relatively small increase of COT and that of ethane concentration, much higher ethylene yield can be reached without seriously affecting the furnace performance. The propylene yield will be lower both at higher ethane concentration and increased COT. Coke formation rate can also be reduced by increasing the ethane ratio, while COT is kept at the same level. In conclusion, it looks attractive to add an amount of 10% ethane to increase the ethylene yield and to reduce the coke formation rate. Note that the coke formation rate is of tolerable value even at COT of 845°C.

The results of simulation runs carried out at five different unsaturated concentrations in such a way that concentration of each component in the feed was varied proportionally are presented in Fig.3. The remaining independent variables (COT, ST/HC) were kept constant. As it is shown, the trend of methane yield and that of propylene show a linear decline with increasing unsaturated rate. The increase of conversion of n-butane is only apparent since its relative concentration in the feed also declines when the unsaturated concentration is higher. Nevertheless, variation of ethylene yield shows a slightly rising trend what is not surprising taking into account that the activation energies of 1-buthene and 2-buthene decomposition rates are about 40-45 kJ/mol lower than those of n-butane, while 1-buthene is formed by radical-recombination at negligible energy level representing an additional ethylene source.

→ Figure 3
4.4. Examination of heat transfer

As it was mentioned, the coke deposited onto the inner surface of radiant coil has a high influence on the online operation period of the furnace. Industrial furnaces are decoked after 30-60 days of operation depending mainly on the type of processed feed. Fig. 4 shows the variation of reaction heat, local tube skin temperature and local coking rate as well as the variation of the overall heat transfer coefficient along the radiant coil. It demonstrates well how increasing coke layer reduces the efficiency of heat transfer and induces increased tube metal temperature. The observed changes in the reaction heat are explained by variation of the dominant reactions of hydrocarbons thermal decomposition along the axial distance. This means that while the highly endothermic chain-initiation reactions are dominant in the first part of the coil, the chain-termination ones are more accented in the last part that are proceeded with negligible activation energy.

Nevertheless, this coke layer has negative impact also on the lifetime of radiant pipes. As it is shown in Fig.4, the coke layer thickness increases drastically in the last third part of the coil therefore this part, in order to achieve sufficient heat transfer rate to the cracked gas is exposed to a much higher thermal load due to the decreased overall heat transfer coefficient induced by the thick coke layer. This is the reason why this piece of coils is worth to replace more frequently than the remaining part of the furnace. Since in this way the complete replacement of furnaces can be made scarcer the maintenance costs of steam crackers are reduced significantly.

→ Figure 4

4.4. Influence of the residence time of furnaces

Results of simulation runs carried out by varying the radiant coil geometry, i.e. the mean residence time of furnaces are shown in Fig.5. Here, yield data and coke formation rate of the primarily investigated furnace, denoted as CF1 are compared with those computed for two other furnaces having different processing times. Geometrical data of two existing furnaces, denoted as CF2 and CF3 and having residence times 0.65 sec and 1.1 sec, respectively, were taken as reference to keep the residence times used in simulation, in real domain. Simulation results obtained with similar feed compositions and total olefin content, which was equal 15.27% in each run, carried out with three values of COT, and yield data
were compared with those obtained from the reference furnace CF1. The ST/HC ratio was adjusted similarly in the case of all three furnaces.

As it was expected, a higher conversion of n-butane could be achieved both in CF2 and CF3 furnace than in the reference CF1 furnace. The yield of ethylene is 7% higher in CF2 and 11.1% higher in CF3 at the same COT. Though the coke formation rate is increased significantly in both furnaces, by 21% and 32% respectively, this amount of coke will be deposed on a five times higher surface in CF2 and on a ten times higher one in CF3. Taking into consideration also the differences in residence times, it can be concluded that 35-40% longer online operation periods can be expected in the case of furnaces CF2 and CF3. As it was shown, coke formation rate can be reduced significantly by decreasing the olefin content of the feed. This can be done by mixing fresh hydrocarbons like ethane and/or n-butane into the recycled streams. According to industrial practice, recycled ethane is always available and n-butane is also worth to use for this purpose. In conclusion, recycled stream cracking in furnaces with longer residence times appears to have some advantages. Naturally, the hydrogenation reactor upstream the cracking furnace as well as the properties of its catalyst plays a substantial role in this complex process as well.

5. Conclusions
The simulation study, carried out by means of a computer model developed for examining thermal cracking of recycled gas mixtures and implemented in CHEMCAD environment, revealed that variations originated in diverse feed compositions can be harmonized well with the operating parameters of furnaces for the purpose of achieving maximum profitability.

Having examined re-pyrolysis of these gas mixtures it was shown that increasing the COT is strongly limited by the coke formation rate, which increases steeply above 840ºC while the ethylene yield becomes increased just slightly. The coke layer thickness and the main local tube skin temperature continuously increases along the radiant coil, especially in the last third part, which has a negative impact on the lifetime of the coils and requires special maintenance techniques. It was also shown as an energy-saving opportunity that reduction of the dilution steam ratio up to 20%, compared with the basic design, causes no significant increase in the coke formation rate.
Acknowledgement

Support provided by TVK Olefin Unit operational and laboratory staff is gratefully acknowledged.

Nomenclature

\( \Delta H_k \) – heat of reaction \( k \) [J/mol]
\( A \) – surface area per unit axial distance [m²]
\( c_i \) – concentration of reactant \( i \) [mol/m³]
\( C_{pi} \) – heat capacity of species \( i \) [J/kg/K]
\( d \) – diameter of pipe/fitting [m]
\( f \) – friction factor [-]
\( g_c \) – acceleration of gravity [m/s²]
\( L \) – equivalent pipe length [m]
\( N_c \) – number of species
\( N_r \) – number of reactions
\( r_k \) – rate of the reaction \( k \) [mol/m³/s]
\( T \) – temperature of cracked gas [K]
\( t \) – time [s]
\( \bar{t} \) – mean residence time [s]
\( T_{fb} \) – temperature of firebox [K]
\( U \) – overall heat transfer coefficient from the firebox to cracked gases [W/m²/K]
\( v \) – convective velocity of cracked gas [m/s]
\( x \) – axial distance along the reactor [m]
\( \alpha_{ki} \) – stoichiometric coefficient of component \( i \) in the reaction \( k \)
\( \Delta p \) – pressure drop [MPa]
\( \rho \) – density of cracked gas [kg/m³]

References


Capture of figures

Figure 1: Schematic diagram of the radiant coils of the simulated furnace connected to one transfer line exchanger

Figure 2: Variation of product yields as a function of coil outlet temperature (COT)

Figure 3: Variation of product yields as a function of olefin concentration in the feed

Figure 4: Variation of heat transfer conditions along the axial distance of radiant coil

Figure 5: Comparison of product yields by cracking recycled mixture in furnaces having different residence times: CF1 - \( \tau = 0.3 \) s; CF2 - \( \tau = 0.65 \) s; CF3 - \( \tau = 1.1 \) s;
Figures

Figure 1: Schematic diagram of the radiant coils of reference furnace CF1 connected to one transfer line exchanger

Radiant coil

$d_1=39 \text{ mm}$

$d_2=57 \text{ mm}$
Figure 2: Variation of product yields as a function of coil outlet temperature (COT)

Figure 3: Variation of product yields as a function of olefin concentration in the feed
Figure 4: Variation of heat transfer conditions along the axial distance of radiant coil

Figure 5: Comparison of product yields by cracking the recycled mixture in furnaces having different residence times: CF1 - \( t = 0.3 \) s; CF2 - \( t = 0.65 \) s; CF3 - \( t = 1.1 \) s;
Tables

Table 1 Comparison of measured and simulated yield data, expressed in mass percentage, of the reference furnace CF1 at different operational conditions

<table>
<thead>
<tr>
<th>Concentration</th>
<th>Case no</th>
<th>Measured mass %</th>
<th>Fitted mass %</th>
<th>Measured mass %</th>
<th>Fitted mass %</th>
<th>Measured mass %</th>
<th>Fitted mass %</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1</td>
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<td>3</td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>1</td>
</tr>
<tr>
<td>Methane</td>
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<td>18.67</td>
<td>18.53</td>
<td>18.64</td>
<td>18.27</td>
<td>18.52</td>
<td></td>
</tr>
<tr>
<td>Ethylene</td>
<td>30.64</td>
<td>30.58</td>
<td>31.13</td>
<td>31.08</td>
<td>32.30</td>
<td>32.17</td>
<td></td>
</tr>
<tr>
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<td>19.51</td>
<td>19.64</td>
<td>19.54</td>
<td>19.61</td>
<td>19.26</td>
<td>19.37</td>
<td></td>
</tr>
<tr>
<td>n-butane</td>
<td>8.50</td>
<td>8.58</td>
<td>8.57</td>
<td>8.63</td>
<td>10.08</td>
<td>9.87</td>
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</tr>
<tr>
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<td>1.34</td>
<td>1.36</td>
<td>1.34</td>
<td>1.36</td>
<td>1.33</td>
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<td></td>
</tr>
<tr>
<td>Toluene</td>
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<td>0.25</td>
<td>0.22</td>
<td>0.25</td>
<td>0.22</td>
<td>0.25</td>
<td></td>
</tr>
</tbody>
</table>

Table 2 Variation of product yields, given in mass percentage, as a function of reduction of the steam/hydrocarbon (ST/HC) ratio, related to the basic design, for two values of COT

<table>
<thead>
<tr>
<th>Reduction of steam</th>
<th>5 %</th>
<th>10 %</th>
<th>20 %</th>
<th>30 %</th>
</tr>
</thead>
<tbody>
<tr>
<td>COT</td>
<td>835 °C</td>
<td>840 °C</td>
<td>835 °C</td>
<td>840 °C</td>
</tr>
<tr>
<td>Methane</td>
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<td>18.75</td>
<td>18.32</td>
<td>18.72</td>
</tr>
<tr>
<td>Ethylene</td>
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<td>33.73</td>
<td>32.07</td>
<td>33.18</td>
</tr>
<tr>
<td>Propylene</td>
<td>20.63</td>
<td>20.15</td>
<td>20.68</td>
<td>20.34</td>
</tr>
<tr>
<td>Butadiene</td>
<td>3.97</td>
<td>3.85</td>
<td>3.97</td>
<td>3.89</td>
</tr>
<tr>
<td>n-butane(residual)</td>
<td>8.74</td>
<td>7.58</td>
<td>8.72</td>
<td>7.88</td>
</tr>
<tr>
<td>Benzene + toluene</td>
<td>1.61</td>
<td>1.63</td>
<td>1.71</td>
<td>1.74</td>
</tr>
<tr>
<td>Coke</td>
<td>0.0084</td>
<td>0.0089</td>
<td>0.0091</td>
<td>0.0097</td>
</tr>
</tbody>
</table>