Reduction of Coke Accumulation and Energy Resources by Adding Steam and Carbon Dioxide in Naphtha Based Ethylene Production

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Abstract

In this study, the production of ethylene and hydrogen is studied via the thermal cracking of ethylene in an ethylene plant based in Libya. During the process of thermal cracking, a mix of naphtha and steam is input into tubes that are directed to the naphtha main line. The utilization of steam is generally used because of the partial removal of coke which has undesirable effects on the process. The coke accumulation on the coils, or tubes, result in a decrease in pressure and also reduction in the yields produced. In this work, the naphtha thermal cracking process is both designed and solved numerically. A thorough comparison of the design results and the data extracted from the experiment reveal that the design may predict the overall process precisely. Also, the direct effects of CO2 are studied with regard to the accumulation of coke. Based on the results of two separate scenarios, the process of thermal cracking with the CO2 is beneficial to the overall process due to the higher yield of ethylene and propylene, and less accumulation of coke, and, in turn, less thickness on the coils inside the furnace. The results from the simulation show that the run time, or run length, of the furnace with the addition of CO2 becomes almost two times as the run time with adding steam. Based on these results, this study has proven to be worthy to explore, and the addition of CO2 has been observed to have noticeably positive results on the thermal cracking process.

Keywords: naphtha, ethylene, hydrogen, coke accumulation

1. Introduction

This section presents an introduction for this study. The background is described, along with all the key terms used in this study.

1.1 Hydrogen

Generally, hydrogen is a suitable alternative for traditional fuels used in the ethylene plant, due to the fact that it may be produced easily in different ways. The production of hydrogen is very important, especially with the increasing demands for it in several industries, especially the petrochemical industry. Hydrogen may be produced by thermal cracking of naphtha and other hydrocarbons, according to researchers. Pyrolysis has a byproduct, which is hydrogen, and is produced in a relatively large amount, almost as large as the end products of the process. As a result, this method is strongly suggested by researchers for the production of hydrogen.

1.2 Ethylene

Ethylene is used as the main feedstock for the production of several plastics, with the majority being polyethylene. It is an important element within the petrochemical industry. It is relatively cheap when compared to other elements, and at the same time, has a high purity. Because of this, it is used as a core component in numerous petrochemical industries. It may also produce beneficial chemicals when reacting with other chemicals that are found abundantly. During the 1990's, there has been a rapid increase in ethylene production, from about 30 to 200 billion pounds (lb). Hence, simple improvements in the production process of ethylene may mean huge profit for industries. In modern times, critical approaches for the production of ethylene are the thermal cracking of alkanes, including ethane, propane, butane, naphtha and finally, gas oil.

1.3 Thermal Cracking

The process of thermal cracking is an important one, especially in terms of olefin production. During this process, a mix of hydrocarbons and steam are input into furnaces, with olefins produced as an output of this process. There are several parameters that affect the product distribution, which are the feed composition, the furnace temperature, and the steam ration. A great amount of research has been done in an attempt to enhance the modeling of the thermal cracking process. Lee and Aitani have studied the feed composition and how it affects ethylene production. Keyvanloo et al. has investigated several parameters, which are the effects of steam ratio, temperature, and run time on the production during the steam cracking of naphtha.

1.4 Coking

Coking results from the thermal cracking of hydrocarbons, and its intensity and rate of accumulation rely on the parameters that are input to the furnace. Coking has bad effects on the pyrolysis process. Among these bad effects is the reduction of the temperature of the furnace. In order to maintain the heat transfer at the level required, the temperature must be raised at certain intervals. Coil metallurgy is a core limitation for temperature increase on the tube walls. Also, the rise in temperature leads to a higher rate of coke accumulation. Additionally, the cross section reduction of coils, as a result of coke accumulation, creates a decrease in the pressure in the tubes. Coking results in higher maintenance expenses by the ethylene plant.

During decoking, the furnace is stopped completely. The average run time of the furnace is about 30-90 days, and is based on several factors including type of furnace, operating conditions, and type of feedstock. The removal of coke, or decoking, is done with the use of both air and steam. Because of thermal cycling, and changes in temperature, the lifetime of the coils is reduced.

2. Literature Review

There are a limited amount of studies conducted on the attempt to carry out the process of thermal cracking with the addition of CO2. This is beneficial to the overall process due to the predicted higher yields of ethylene and propylene, and less accumulation of coke, and, consequently, less thickness on the coils inside the furnace.

Numerous studies have revealed that coke is formed because of either reactants of products (Virk et al.; Froment et al.; Kumar and Kunzru). In this study, the simulation of a thermal cracking furnace of an ethylene plant in Libya is conducted, followed by a thorough comparison of the findings of the simulation and industrial data is carried out. After this, carbon dioxide, CO2, is used instead of steam, and the impact of the use of CO2 are studied on the production of the end products and the coke accumulation in the furnace.

The primary parameters that influence and impact the product distribution are the feed composition, the furnace gas temperature and the overall steam ratio. Over the past couple of years, numerous investigations had been conducted on the modeling of thermal cracking operation for several hydrocarbons. Lee and Aitani studied the impact of feed composition for ethylene production. Also, Keyvanloo et al. investigated the impact of steam ratio, temperature and the residence time on the yields in steam cracking operation of naphtha. Additionally, Belohlav et al. introduced a model to describe thermal cracking of ethane, petroleum gases, and naphtha.

Ghassabzadeh et al. studied the process of ethylene and propylene with use of kerosene thermal cracking in a practical setup. They made use of several simplification considerations to show a suitable kinetic model that estimates product yields effectively. Impacts of temperature, time of residence and overall steam ratio on the products in gas and oil thermal cracking operations were investigated by Depeyre et al. They implemented a system for product distributions based on radical mechanisms. Towfighi et al. showed a reaction network system for simulation of LPG cracking plants. Also, many other scholars introduced mechanistic systems for the atmospheric gas and oil cracking operations based on experiments.

3. Methodology

The production of olefins utilizes both gas and liquid furnaces. Table 1 presents information on the products of liquid furnaces from a particular petrochemical company. It has been proved that using a gas furnace instead of a liquid furnace produces about one and a half times more ethylene.

component	Mass (dry %)	Mass (wet %)	flow(ton/h)
Water	0.00	34.01	22.19
Hydrogen	1.13	0.72	0.499
methane	20.01	13.01	8.840
Carbon monoxide	0.131	0.089	0,051
Carbon dioxide	0.02	0.02	0.005
Sum c1	21.03	48.01	32.03
acetylene	0.87	0.56	0.376
ethylene	33.13	21.75	15.19
Ethane	3.51	2.30	15.01
C2,s	39.02	25.070	17.01
C3H4,s	1.17	0.81	0.532
propylene	16.53	11.23	7.01
propane	0.91	0.65	0.387
C3,s	17.53	11.62	7.83
butadiene	4.66	3.11	2.069
butanes	4.00	2.66	1.777
butanes	1.56	1.04	0.687
Sum C4,s	10.34	6.89	4.587
C5,s	2.41	1.62	1.07
benzene	5.19	3.47	2.31
C6 non-aromatics	0.49	0.32	0.224
C6,s	5.70	3.77	2.54
toluene	1.56	1.03	0.656
C7 non-aromatics	0.12	0.06	0.399
C7,s	1.76	1.22	0.734
xylenes	0.29	0.17	0.111
C8 non-aromatics	0.001	0.001	.003
C8,s	0.91	0.61	0.39
C9,s	0.26	0.18	0.785
C10,s	1.78	1.21	0.786
	100.00	100.00	66560

Table 1. Products of liquid furnaces from petrochemical company in Libya

Naphtha is one of the most beneficial feedstock for the process of thermal cracking due to its high ethylene selectivity when compared with other paraffin. The pyrolysis is easy and less costly than other hydrocarbons. In the ethylene plant being studied, a mix of naphtha and steam move through two rounds in the gas furnaces. Table 2 presents the general flow rates of naphtha and steam for every round in a common gas furnace during the runtime of the furnace.

Time/day	Flow rate of naphtha	Flow rate of steam	Coil outlet temp °c	Coil outlet
·	(MT/hr)	(MT/hr)	•	pressure Kg/cm ²
2	150	90	810.2	1.31
4	148.6	89.16	810.2	1.30
6	150.01	90	810.8	1.33
8	151.2	90.72	809.9	1.31
10	147.3	88.06	810.8	1
12	150	90	810.00	1.31
14	148.8	89.28	810.9	1.31
16	148.9	89.28	810.4	1.31
18	148.2	88.92	810.6	1.31
20	151.0	90.6	810.6	1.31
22	149.7	89.25	810.7	1.32
24	148.96	89.29	810.5	1.31
26	147.98	89.29	810.7	1.31
28	149.98	89.29	810.3	1.33
30	148.78	89.27	810	1.32
32	146.9	8814	810	1.31
34	145.9	87.54	808.9	1.31
36	149.9	89.30	810	1.31
38	149.2	89.24	810	1.31
40	151.1	90.70	811,1	1.32
42	150.9	90.68	811.2	1.33
44	150.3	90.1	810.6	1.33
48	150.1	90.12	810.3	1.33
50	150.6	90.23	810	1.32
52	150.4	90.21	810.9	1.33
54	150.21	90.10	810	1.34
56	150.23	90.11	811	1.4
58	150.2	90.11	811.5	1.39
60	151	90.68	810.4	1.34

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The naphtha cracking furnace consists of convection and radiation sections. This is shown in Figure 1. The feed is input into the convection section at 121C and 5.9 bars. The stream is diluted with steam, at 0.3 kg/kg naphtha, at a temperature of 175C and 5.7 bars. Next, the mix is heated to 810 C. It later enters the radiation section, where the naphtha molecules are broken apart, and, in turn, the end products are produced (ethylene, propylene, methane, butadiene, among others). This entire process occurs in radiant tubes that are in the furnace at 900-1200 C.



Figure 1. Furnace in thermal cracking of naphtha

During this process, the temperature role is critical, and therefore the coil temperature (COT) of these furnaces is maintained at a stable temperature of 800C. Table 4 presents information about the COTs and pressures. After that, the end products exit the furnace at 775 - 885C, and then the temperature decreases at a fast rate in the transfer line exchanger (TLE), to avoid other reactions. Lastly, all of the end products are then compressed and moved to the separation stages. This is presented in Figure 2.



Figure 2. Cracking plants and separation units

Steam was input to reduce the pressure of mass aromatics. As a result, the number of condensation reactions is also lowered, and also removes some of the coke that has been accumulated along the tubes. According to, Choudhary et al., carbon dioxide acts similar to steam during the process of pyrolysis. In this model CO2 is used instead of steam, and the effects it has on the removal of coke are studied. In this work, the model used was run under normal operating conditions, with the addition of 2 kg CO2/kg ethylene input rather than steam.

Numerous molecular reaction schemes were proposed for the pyrolysis of ethylene. For this work, Forment's molecular scheme was used. Based on this model, several reactions are taken into consideration:

$$C_2H_6 \leftrightarrow C_2H_4 + H_2 \quad \Delta H = 136,330,000 (I/K \text{ mol})$$
 (1)

$$2C_2H_6 \rightarrow C_3H_8 + CH_4 \quad \Delta H = -11,560,000 \text{ (J/K mol)}$$
 (2)

$$C_3H_8 \rightarrow C_3H_6 + H_2 \quad \Delta H = 124,910,000 \text{ (J/K mol)}$$
 (3)

$$C_3H_8 \rightarrow C_2H_4 + CH_4 \quad \Delta H = 82,670,000 \text{ (J/K mol)}$$
 (4)

$$C_3H_8 \leftrightarrow C_2H_2 + CH_4 \quad \Delta H = 133,450,000 \text{ (J/K mol)}$$
 (5)

$$C_2H_2 + C_2H_4 \rightarrow C_4H_6$$
 $\Delta H = -171,470,000 \text{ (J/Kmol)}$ (6)

$$2C_{2}H_{6} \rightarrow C_{2}H_{4} + 2CH_{4} \quad \Delta H = 71,102,000 \text{ (J/K mol)}$$
(7)
$$C_{2}H_{6} + C_{2}H_{4} \rightarrow C_{3}H_{6} + CH_{4} \quad \Delta H = -22,980,000 \text{ (J/K mol)}$$
(8)

After that, the rate equations for the reactions mentioned above are the following:

$$r_{1} = k_{1} \left[\frac{F_{C_{2}H_{6}}}{F_{t}} \left(\frac{P_{t}}{RT} \right) - \frac{F_{C_{2}H_{4}}F_{H_{2}}}{F_{t}^{2}Kc_{1}} \left(\frac{P_{t}}{RT} \right)^{2} \right]$$
(9)

$$r_2 = k_2 \left[\frac{F_{C_2 H_6}}{F_t} \left(\frac{P_t}{RT} \right) \right]$$
(10)

$$r_3 = k_3 \left[\frac{F_{C_3 H_8}}{F_t} \left(\frac{P_t}{RT} \right) \right]$$
(11)

$$r_4 = k_4 \left[\frac{F_{C_3 H_8}}{F_t} \left(\frac{P_t}{RT} \right) \right]$$
(12)

$$r_5 = k_5 \left[\frac{F_{C_3H_6}}{F_t} \left(\frac{P_t}{RT} \right) - \frac{F_{C_2H_2}F_{CH_4}}{F_t^2Kc_5} \left(\frac{P_t}{RT} \right)^2 \right]$$
(13)

$$r_{6} = k_{6} \left[\frac{F_{C_{2}H_{2}}F_{C_{2}H_{4}}}{F_{t}^{2}} \left(\frac{P_{t}}{RT} \right)^{2} \right]$$
(14)

$$r_7 = k_7 \left[\frac{F_{c_2 H_6}}{F_t} \left(\frac{P_t}{RT} \right) \right]$$
(15)

$$r_8 = k_8 \left[\frac{F_{C_2 H_6} F_{C_2 H_4}}{F_t^2} \left(\frac{P_t}{RT} \right)^2 \right]$$
(16)

Also, for this work, propylene, ethylene, and butadiene are the coke precursors:

$$C_2H_4 \rightarrow coke$$
 (17)

$$C_3H_6 \rightarrow coke$$
 (18)

$$C_4H_6 \rightarrow coke$$
 (19)

For reactions 17, 18 and 19, the rate equations are shown to be:

$$r_{17} = k_{17} \left[\left(\frac{F_{C_2 H_4} P_t}{F_t R T_l} \right)^{1.34} \right]$$
(20)

$$r_{18} = k_{18} \left[\left(\frac{F_{C_3 H_6} P_t}{F_t R T} \right)^{1.34} \right]$$
(21)

$$r_{19} = k_{19} \left[\left(\frac{F_{C_4 H_6} P_t}{F_t R T} \right)^{1.37} \right]$$
(22)

Equations 23 and 24 show the partial removal of coke reactions.

$C+H_2O\rightarrow CO+H_2$	$\Delta H = 131,000,000 \text{ (J/Kmol)}$	(23)

$$C + CO_2 \rightarrow 2CO \quad \Delta H = 172,459,000 \text{ (J/Kmol)}$$
 (24)

For these reactions, the rate equations are:

$$r_{23} = k_{23} \left[\frac{F_{H_2 O} P_t}{F_t} \right]$$
(25)
$$r_{24} = k_{24} \left[\left(\frac{F_{CO_2} P_t}{F_t} \right)^{0.31} \right]$$
(26)

Table 6 and Table 7 provide the kinetic constants of the equations that were previously mentioned.

Rate coefficient	$A(s^{-1} \text{ or } l \text{ mole}^{-1} s^{-1})$	E (J/mole)
k1	4.65E+13	2.73E+05
k ₂	3.85E+11	2.73E+05
k3	5.89E+10	2.15E+05
k4	4.69E+10	2.12E+05
k ₅	6.81E+08	1.54E+05
k ₆	1.03E+09	1.73E+05
k7	6.37E+23	5.30E+05
k ₈	7.08E+10	2.53E+05
k17	5.00E+14	2.24E+05
k18	2.77E+09	1.16E+05
k19	5.61E+18	2.74E+05
k23	5.09E+04	2.38E+05
k24	1.12E+08	2.45E+05

Table 6	Ethane	thermal	cracking	narameters
	Lunane	uncimai	Clacking	parameters

Equilibrium constants	based o	on concent	tration
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Temperature (°C)	775	800	825
K _{c1}	8.90E-03	1.28E-02	1.80E-02
K _{c5}	9.85E-03	1.38E-02	1.89E-02

Mathematical Modeling

Figure 3 illustrates a cross section of the cracking coils. In this work, a simple model with one dimension was taken into consideration. Also, a collection of equations that include mass, momentum, and energy were utilized. The following notes were made to make the model easier:

- The plug flow pattern was employed
 The mixture of gas was assumed a suitable gas
 Axial dispersions of mass and heat, and radial concentration gradients are not considered
 Thermal entrance region effects are not considered
 The scenario is studied as a quasi-steady data to simulate the rate of coke accumulation.



Figure 3. Cross section of cracking coil

Based on those considerations, the derived equations are:

$$\frac{dF_j}{dz} = \left(\sum_i s_{ij} r_{ri}\right) \frac{\pi d_t^2}{4} \tag{27}$$

$$\sum_{j} F_{j} c_{pj} \frac{dT}{dz} \equiv Q(z) \pi d_{t} + \frac{\pi d_{t}^{2}}{4} \sum_{i} r_{ri} (-\Delta H)_{i}$$
⁽²⁸⁾

$$\left(\frac{1}{M_m P_t} - \frac{P_t}{\eta G^2 RT}\right) \frac{dP_t}{dz} = \frac{d}{dz} \left(\frac{1}{M_m}\right) + \frac{1}{M_m} \left(\frac{1}{T} \frac{dT}{dz} + Fr\right)$$
(29)

The friction factor for the straight units of the coils and tube bends are the following:

$$Fr = 0.092 \frac{\text{Re}^{-0.2}}{d_t}$$
(30)

$$Fr = 0.092 \frac{\text{Re}^{-0.2}}{d_t} + \frac{\zeta}{\pi R_b}$$
(31)

The parameter of the tube bend is the following:

$$\zeta = \left(0.7 + 0.\beta 5 \frac{\Lambda}{90^{\circ}}\right) \left(0.051 + 0.19 \frac{d_t}{R_b}\right)$$
(32)

The rate of coke formulation may be computed like the following:

$$\frac{\partial C}{\partial t} = (d_t - 2t_c) \frac{\alpha M_c r_c}{4\rho_c}$$
(33)

Numerical solution

Backward finite difference approximation is used in order to convert the collection of ordinary differential equations (ODEs) to nonlinear algebraic equations. All three of Mass, momentum balance, and energy are ODEs, and to avoid this, backwards finite difference is utilized.

Model validation

Three different kinetic mechanisms for ethane pyrolysis were proposed by Sundaram and Froment. They claimed that a model type 5 reaction system could accurately predict experimental data. However, in this work, an 8 reaction system was used, so that the data is very accurately predicted. Table 8 gives a comparison of the models with data taken from the ethylene plant, and also shows how the model used in this work is more accurate. Table 8. Comparison of models using data taken from ethylene plant

4. Results

The general behavior of the cracking coils with the steam and CO_2 are explained. Also, the effects of both diluents on the ethylene and hydrogen yields, coke thickness, and temperature profiles in the furnaces are studied during the process.

Table 8. Model comparison				
Product	MT/hr	Eight furnaces modeling	Eight furnaces % error	
		results		
ethane	7.6035	6.501	6.1234	
methane	4.66	3.547	2.718	
ethylene	24.87	28.871	7.052	
propylene	21.77	23.14	7.05043	
Mixed c4	14.56	13.23	3.454	
Pay gas	25.80	23.437	3.019	
acetylene	0.67	0.8245	4.896	
temperature	810	811	2.098	



Figure 4. Molar flowrate of methane, propylene, hydrogen and ethylene with cracking coils length

This unit of the plant must be stopped when the thickness of the coke in the furnace is about 10 - 20% of the tube diameter. The cracking process with the use of steam may run for about 70 days before this unit of the plant is stopped for decoking. The run time for the model will be over 140 days. Because of this, the process will result in the production of higher ethylene yields with lower operating and maintenance expenses.

The tube wall temperature is plotted as a function of the length of the reactor and the time. The run time of the furnace in a normal thermal cracking process is about 70 days, and the temperature of the tube wall reaches about 1370 K, which is just less than the maximum temperature that is allowed. However, with the presence of CO_2 , the temperature reaches the maximum allowed temperature after about 140 days.

The experimental tube wall temperatures during a run time of the furnaces at the ethylene plant involved in the study are presented in Figure 5. According to the figure, it is clear that, considering the time (in days) and the furnace length, the tube wall temperature is higher when using CO_2 instead of steam. Due to no coke accumulation for the first couple of days, the tube wall temperatures are less than 1220K. Eventually, the temperature increases to about 1280 K. The temperature in the tube wall changes dramatically as a function of furnace length and time. The reason for this is that the coke accumulation is different in different parts of the coils, and therefore, the heat transfer is not stable across the furnace.



Figure 5. Wall temperature of tube for cracking coils length and time in days using a) steam and b) CO2

5. Conclusion

In this work, a gas thermal cracking process at an ethylene production plant in Libya was taken into consideration for a simulation. Ethylene was used for the gas pyrolysis process and as a result, ethylene and hydrogen, among other important end products, were produced. A model was utilized to study the thermal cracking process of ethylene in the existence of steam. The results of the model and real world data from the plant were compared, and were almost identical. Next, the model was processed with CO2 to study its effects on the furnace operation and rate of production of the end products. Several parameters were compared in both of the scenarios, including the ethylene flow rate, hydrogen flow rate, gas temperature, temperature of tube wall, and coke thickness, among others. Based on this comparison, the benefits of the use of CO2 instead of steam in the process of ethylene cracking were obtained. The benefits of the use of CO2 are the following:

- The yields of ethylene and hydrogen increase.
- The run time of the furnace is doubled when using CO2 instead of steam.
- The rate of accumulation of coke is decreased.
- Adding CO2 would decrease fuel consumption and as a result be safer for the environment.

This work acts as a foundation for future related studies on the investigation of benefits of adding Co2 instead of steam to the ethylene process cracking. It gives useful insights for future researchers that aim to increase effectiveness and efficiency of ethylene cracking process.

In terms of future directions, more experiments may be carried out with CO2 addition during the thermal cracking process. This study had considered results from a petroleum company based in Libya with data recorded for a period of 60 days. The same experiment may be carried out at a different plant, and with data recorded for longer periods of time. Also, the same experiment may be repeated multiple times to get the average of the outcome of all of the experiments for more accurate results, and, in turn, more accurate evaluation of the results.

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