

Thermodynamic Analysis of Fuel Oil Blended Stock (FOBS) to Light Hydrocarbon

Mei Lian Teo, Mazura Jusoh, Zaki Yamani Zakaria*

School of Chemical and Energy Engineering, Universiti Teknologi Malaysia 81310 UTM Skudai, Johor, Malaysia
 zakiyamani@utm.my

The depletion of fossil fuels and the increasing demand for fuels worldwide have urged researchers to search for alternatives to traditional fossil fuels. Recently, different researchers introduced various technologies to upgrade the Heavy Fuel Oil (HFO), a Fuel Oil Blended Stock (FOBS) residual from the petroleum refinery. The cracking of HFO into a lighter fuel with higher efficiency on burning and economic value was introduced. In this study, n-eicosane was chosen as a surrogate fuel for the multicomponent HFO due to the high percentages of hydrocarbon in HFO. The purpose of this study was to investigate thermodynamic modelling of FOBS cracking into light hydrocarbon in the presence of oxygen based on the total Gibbs energy minimisation method. The effect of different reaction conditions on the cracking of n-eicosane was studied. Equilibrium product compositions of n-eicosane at temperature range of 573 K - 1273 K, pressure of 1 bar, n-eicosane/oxygen ratios (EO) (0.5:0.5, 0.7:0.3, 0.8:0.2, 0.9:0.1, 0.95:0.05) were studied. The results revealed that the main products contain hydrogen gas, carbon, n-alkanes and 1-alkenes. The results showed that C-C bond homolytic scission through free radical mechanism prevailed in the reactions. Simulation results demonstrated that a small amount of induced oxygen in the cracking process increased the production of lighter hydrocarbons. Production rate of desired hydrocarbons can be controlled under optimum conditions. Current research on oxygen-induced cracking is still limited. This research proved that the presence of oxygen in the cracking process is cost-saving and could be introduced in the refinery as a solution to FOBS issue.

1. Introduction

Fuel oil blended stock (FOBS) is one of the crude oil derivatives that still pique worldwide attention to world energy market demand. Crude oil is still responsible for 32 % of the world energy supply, and it is expected to hold its position in the next decade. According to the International Energy Outlook 2016, the demand for fossil fuels in generating energy will still be prominent compared to other renewable energy in 2040 (Conti et al., 2016). The utilisation of renewable energy is still limited, especially in low-income countries due to the high-cost technology. In this case, heavy fuel oil (HFO), as one of the most abundant FOBS component from petroleum refinery, continue to grab the attention. In this issue, the abundant resources of heavy oil had brought to the need to upgrade the HFO to produce lighter fuel that produce cleaner energy on burning (Kar et al., 2018). The upgrading process of fuel oil also offers a modern approach to dispose of the HFO in a sustainable method and reduces the environmental problem caused by HFO. The increase of world fuel prices due to the scarcity of natural oil and gas and the strengthening control in-laws has grabbed the attention of researchers in upgrading HFO.

The main components of HFO are saturates, aromatics, resins, and asphaltenes. The sources of fuel oil affect the compositions of each component in HFO. HFO are heavy hydrocarbons with high density and viscosity. The presence of asphaltenes in HFO make it a dense compound. The thermochemical conversion processes used in upgrading heavy fuel oil can be categorised into thermal cracking, hydrocracking, catalytic cracking and pyrolysis (Demirbas et al., 2016). Since the 1900s, numerous studies had been conducted to investigate the cracking of various hydrocarbons. The thermolysis of higher molecular weight of n-paraffins, including n-eicosane in the vapour-phase under atmospheric pressure and at a temperature range of 623 K to 893 K, proved that 1-alkenes formed as the main products (Zhou et al., 1987). Thermal cracking of hydrocarbons produces a wide range of liquid and gases products. In most past researches, the result of cracking is only

valid for a low conversion of feedstocks (Jiang et al., 2019). The cracking temperature was controlled at a suitable range to ensure only primary and secondary cracking occurred. Thermal cracking at lower temperatures encourages the production of liquid fuels. A higher cracking temperature promotes higher quantities of olefins in the products (Ghashghaee and Shirvani, 2018). Catalytic cracking with the use of catalysts catches researchers' attention in recent years. The cracking process can be achieved conversion in a lower temperature range and shorter retention time with the help of a catalyst. One of the common catalysts used is zeolite ZSM-5 (Safari et al., 2020). The effect of different reactions conditions to improve the catalytic process are widely studied. By increasing the amount of catalyst, it had been proven that the compositions of desired products formed at the end of the process increase.

Heavy fuel oil with a lower commercial value undergoes the cracking process to produce smaller hydrocarbon molecules with a higher retail value that meet the market demand. Due to the differences in compositions of HFO from different resources, the upgrading process of the HFO might produce various products. Pure hydrocarbons were chosen to represent HFO in multiple studies for simplification due to the high percentages of hydrocarbon in fuel oil (Shimada et al., 2018). The n-eicosane was selected as a surrogate fuel for the HFO in the present study. Limited research was conducted to investigate heavy fuel oil cracking with oxygen to initiate the upgrading process. To the best of our knowledge, no similar research had been conducted on oxygen-induced cracking with n-eicosane as the surrogate fuel. In this research, the major products formed under cracking are identified by applying the total Gibbs energy minimisation method. Simulation of thermodynamic analysis is applied to investigate the effect of reaction parameters, including temperature and n-eicosane/oxygen ratios (EO), on the yield of products at equilibrium. The usage of oxygen aims to open the chances of the low-cost reactant in enhancing the cracking process.

2. Methodology

The thermodynamic analysis of cracking of n-eicosane with oxygen was performed using HSC Chemistry version 11.0 software based on the minimisation method of the total Gibbs energy. All chemical species involved were considered when the system reaches equilibrium at constant temperature and pressure. The equilibrium rate constants for all possible reactions were determined and the equilibrium compositions of each species were calculated when the Gibbs energy of the system reaches its minimum. The species considered in this study were n-eicosane and oxygen as the reaction reactants. The reaction products were hydrogen, coke, methane, water, carbon monoxide, carbon dioxide, ethane, ethene and acetylene. The outlet composition of the products was assumed to be in equilibrium at the exit of the reactor. The formation of minor byproducts was not considered in this study. For gas-phase reaction equilibrium, a mixture of ideal gases was assumed. Solid carbon is the only species that exist in the solid phase. The total number of mol of reactant input was kept at 1 kmol. The operating temperature was kept in the range of 573 K to 1,273 K while the ratios of n-eicosane to oxygen were 0.5:0.5, 0.7:0.3, 0.8:0.2, 0.9:0.1 and 0.95:0.05. In all conditions, the pressure was kept at 1 bar. Complete conversion of n-eicosane was recorded in all considered reaction parameters, indicating the feasibility of cracking n-eicosane with oxygen.

3. Result and Discussion

In this research, the main reactions in the cracking of n-eicosane with oxygen are listed in Table 1. There are a total number of 20 reactions proposed in this process. The reaction network based on the main products formed is shown in Figure 1. Both exothermic and endothermic reactions are summarised in the figure to provide a better overview of the process. The cracking process begins with the main reaction that required high energy to crack the large hydrocarbon (Zhuman et al., 2020), n-eicosane into smaller hydrocarbon. A free radical mechanism that involves the C-C bond homolytic scission occurs. With the use of oxygen, it can be observed that products that contain oxygen atoms are formed. As shown in the first step of Figure 1, other than the production of smaller alkanes and alkenes, the reactions between n-eicosane and oxygen also produce CO, CO₂ and hydrogen as the first products from both cracking and partial oxidation processes. Each of the first products undergoes secondary reactions until an equilibrium condition is achieved. Cracking of smaller components and side reactions occur subsequently. Methanation occurs when CO and CO₂ react with hydrogen. The conversion of CO to CO₂ and solid carbon happens through the Boudouard Reaction (Hervy et al., 2018). The hydrogen will also react with CO₂ in the water gas shift reaction. Ethane, ethylene and acetylene react with oxygen in partial oxidation to produce hydrogen and CO₂. The exothermic oxidative dehydrogenation that involves the hydrogen abstraction from hydrocarbon occurs in both ethane and ethene. The endothermic dehydrogenation of ethane and ethylene produce ethene and acetylene. In this study, there are not aromatic or branched olefins recorded in the product equilibrium composition. Only linear alpha-olefin is found in the outlet composition. This is due to the main reactions that occurred are C-C bond homolytic

scission and β -scission. The formation of aromatic compounds only occurred under the severe cracking region with high reaction temperatures (Zhang et al., 2018). Current reaction parameters are insufficient for the formation of the aromatic compounds. In short, a series of chain reactions occur during the cracking process. In the cracking process, the main products formed are $\text{H}_2(\text{g})$, $\text{C}(\text{s})$, $\text{CH}_4(\text{g})$, $\text{H}_2\text{O}(\text{g})$, $\text{CO}(\text{g})$, $\text{CO}_2(\text{g})$, $\text{C}_2\text{H}_6(\text{g})$, $\text{C}_2\text{H}_4(\text{g})$ and $\text{C}_2\text{H}_2(\text{g})$ while the minor products formed are $\text{C}_3\text{H}_4(\text{g})$, $\text{C}_3\text{H}_8(\text{g})$, $\text{C}_3\text{H}_6(\text{g})$, $\text{C}_4\text{H}_6(\text{g})$, $\text{C}_4\text{H}_8(\text{g})$ and $\text{C}_4\text{H}_{10}(\text{g})$. Among the main products, hydrogen and carbon recorded the highest value of production compared to others. Besides, it is found that the percentage of oxygen conversion is 100 % in all EO ratios.

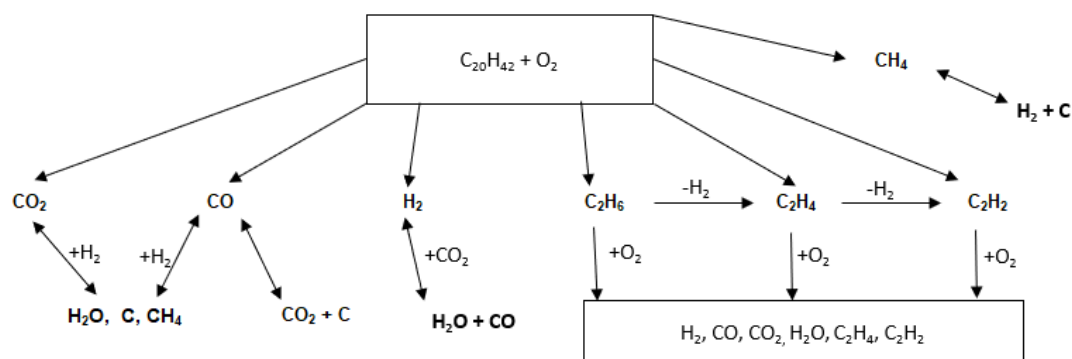


Figure 1 Reaction Network based on main products formed in the cracking of *n*-eicosane with oxygen at 1 bar

Table 1: Main Reactions in the Cracking of *n*-Eicosane with Oxygen

Reaction	Type of Reaction	Equation
R1	Cracking of <i>n</i> -eicosane with O_2	$x \text{C}_{20}\text{H}_{42}(\text{g}) + y \text{O}_2(\text{g}) \rightarrow m \text{H}_2(\text{g}) + n \text{CO}(\text{g}) + q \text{CH}_4(\text{g}) + s \text{C}_2\text{H}_6(\text{g}) + t \text{C}_2\text{H}_4(\text{g})$
R2	Partial oxidation of <i>n</i> -eicosane with O_2	$\text{C}_{20}\text{H}_{42}(\text{g}) + 10 \text{O}_2(\text{g}) \rightarrow 21 \text{H}_2(\text{g}) + 20 \text{CO}(\text{g})$
R3	Partial oxidation of <i>n</i> -eicosane with O_2	$\text{C}_{20}\text{H}_{42}(\text{g}) + 20 \text{O}_2(\text{g}) \rightarrow 21 \text{H}_2(\text{g}) + 20 \text{CO}_2(\text{g})$
R4	Partial oxidation of methane with O_2	$2 \text{CH}_4(\text{g}) + \text{O}_2(\text{g}) \rightarrow 4 \text{H}_2(\text{g}) + 2 \text{CO}(\text{g})$
R5	Partial oxidation of methane with O_2	$\text{CH}_4(\text{g}) + \text{O}_2(\text{g}) \rightarrow 2 \text{H}_2(\text{g}) + \text{CO}_2(\text{g})$
R6	Partial oxidation of ethane with O_2	$\text{C}_2\text{H}_6(\text{g}) + \text{O}_2(\text{g}) \rightarrow 3 \text{H}_2(\text{g}) + 2 \text{CO}(\text{g})$
R7	Partial oxidation of ethane with O_2	$\text{C}_2\text{H}_6(\text{g}) + 2 \text{O}_2(\text{g}) \rightarrow 3 \text{H}_2(\text{g}) + 2 \text{CO}_2(\text{g})$
R8	Partial oxidation of ethene with O_2	$\text{C}_2\text{H}_4(\text{g}) + \text{O}_2(\text{g}) \rightarrow 2 \text{H}_2(\text{g}) + 2 \text{CO}(\text{g})$
R9	Partial oxidation of ethene with O_2	$\text{C}_2\text{H}_4(\text{g}) + 2 \text{O}_2(\text{g}) \rightarrow 2 \text{H}_2(\text{g}) + 2 \text{CO}_2(\text{g})$
R10	Methane decomposition	$\text{CH}_4(\text{g}) \leftrightarrow 2\text{H}_2(\text{g}) + \text{C}(\text{s})$
R11	Dehydrogenation of ethane	$\text{C}_2\text{H}_6(\text{g}) \leftrightarrow \text{C}_2\text{H}_4(\text{g}) + \text{H}_2(\text{g})$
R12	Dehydrogenation of ethene	$\text{C}_2\text{H}_4(\text{g}) \leftrightarrow \text{C}_2\text{H}_2(\text{g}) + \text{H}_2(\text{g})$
R13	Oxidative dehydrogenation of ethane	$2 \text{C}_2\text{H}_6(\text{g}) + \text{O}_2(\text{g}) \rightarrow 2 \text{C}_2\text{H}_4(\text{g}) + 2 \text{H}_2\text{O}(\text{g})$
R14	Oxidative dehydrogenation of ethene	$2 \text{C}_2\text{H}_4(\text{g}) + \text{O}_2(\text{g}) \rightarrow 2 \text{C}_2\text{H}_2(\text{g}) + 2 \text{H}_2\text{O}(\text{g})$
R15	Water gas shift reaction	$\text{CO}(\text{g}) + \text{H}_2\text{O}(\text{g}) \leftrightarrow \text{H}_2(\text{g}) + \text{CO}_2(\text{g})$
R16	Boudouard Reaction	$2\text{CO}(\text{g}) \leftrightarrow \text{CO}_2(\text{g}) + \text{C}(\text{s})$
R17	Methanation	$\text{CO}(\text{g}) + 3 \text{H}_2(\text{g}) \leftrightarrow \text{CH}_4(\text{g}) + \text{H}_2\text{O}(\text{g})$
R18	Methanation	$\text{CO}_2(\text{g}) + 4 \text{H}_2(\text{g}) \leftrightarrow \text{CH}_4(\text{g}) + 2 \text{H}_2\text{O}(\text{g})$
R19	Reduction of CO	$\text{H}_2(\text{g}) + \text{CO}(\text{g}) \leftrightarrow \text{H}_2\text{O}(\text{g}) + \text{C}(\text{s})$
R20	Reduction of CO_2	$\text{CO}_2(\text{g}) + 2\text{H}_2(\text{g}) \leftrightarrow 2\text{H}_2\text{O}(\text{g}) + \text{C}(\text{s})$

Figure 2a illustrates the hydrogen production in the cracking of *n*-eicosane with oxygen at 1 bar. The number of moles of hydrogen produced increases with the increase in the reaction temperature. The highest number of moles of hydrogen produced can be observed in the highest EO ratio, which is 0.95:0.05. The lowest EO ratio 0.5:0.5 produced the lowest amount of hydrogen. At all EO ratios, hydrogen production showed a rapid increase from 573 K to 853 K. After 853 K, the increment in hydrogen production almost remains stable, showing that high-temperature conditions will no longer promote the high production of hydrogen. R1, R2 and R3 mainly cause the rapid increases in hydrogen production in the early stage. The high yield of hydrogen in

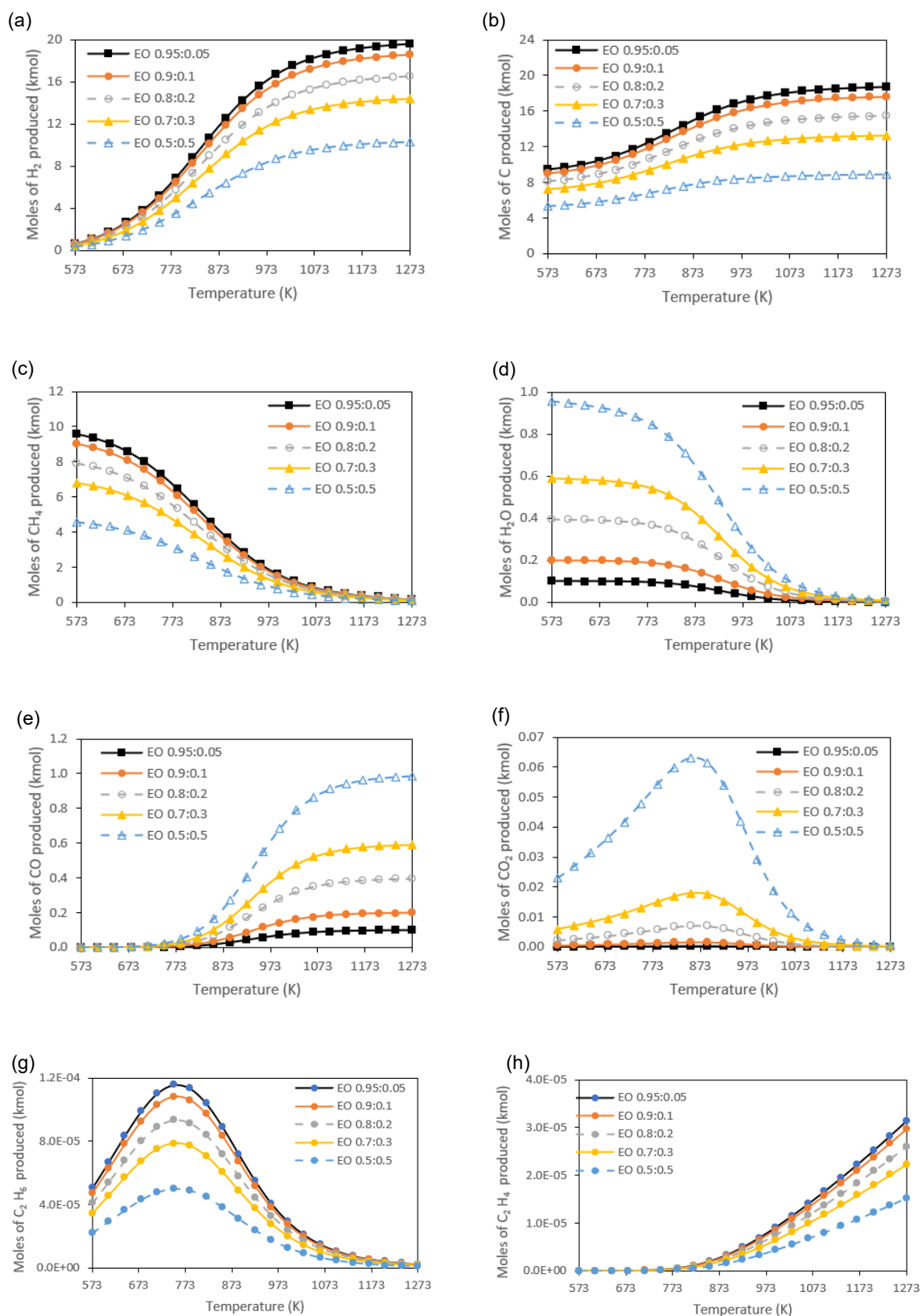


Figure 2: Moles of (a) H_2 (b) C (c) CH_4 (d) H_2O (e) CO and (f) CO_2 (g) C_2H_6 (h) C_2H_4 produced in the cracking of *n*-icosane with oxygen at 1 bar

this process is beneficial as it has a variety of usage, primarily used as a clean fuel that produces zero greenhouse gases (Abdalla et al., 2018).

Figure 2b illustrates the carbon production in n-eicosane cracking with oxygen at 1 bar. The high EO ratio and high temperature enhances the production of carbon. More carbon is produced in the highest EO ratio 0.95:0.05 due to the availability of more carbon in the reactant, but the lowest carbon recorded in the EO ratio 0.5:0.5. The production of carbon could be explained via R10, R16, R19 and R20. Carbon production seems to be undesired in a catalytic reaction as it may reduce the catalyst activity (Eshraghian and Husein, 2018). However, no catalyst is applied in the current study. Carbon formation is beneficial to be used in various fields. The methane production is shown in Figure 2c. Methane has the highest yield among the light hydrocarbon formation in the cracking process (Shvets et al., 2017). The formation of methane is higher at lower temperatures and it decreases gradually with the increase in the reaction temperature. Similar to the carbon formation, more methane is produced in a higher EO ratio due to the presence of more carbon in the reactant. The water production in the cracking of n-eicosane with oxygen at 1 bar is illustrated in Figure 2d. Water formation decreases as temperature rises. A low EO ratio promotes higher water formation due to more oxygen in the feed, especially in low temperatures. The formation of water in high EO is smaller and steadier. Even though water is an unwanted product in this process, the methanation of CO_x in R17 and R18 has contributed to a similar production trend in methane and water.

Figure 2e illustrates the carbon monoxide production in the cracking of n-eicosane with oxygen at 1 bar. When the temperature of reaction increases, the moles of CO produced increase. The rising trend of CO production at higher temperatures is due to oxygen that promotes the partial oxidation of n-eicosane. This situation is similar to the oxidative cracking of naphthenic and aromatic hydrocarbons (Zhu et al., 2006). Therefore, it is observed that at the lowest EO ratio 0.5:0.5, the presence of more oxygen promotes the highest production of CO. The addition of oxygen induces the cracking of n-eicosane by enhancing the selectivity of CO_x in products formed compared to traditional cracking. The production of CO is desired as CO is a valuable petrochemical feedstock that can generate other compounds. For instance, CO can produce synthetic fuels via Fischer–Tropsch synthesis (Mehariya et al., 2020).

Figure 2f illustrates the carbon dioxide production in the oxygen-induced cracking. The formation of CO₂ is steadier at a high EO ratio. A lower EO ratio promotes the formation of CO₂ compared to a high EO. The amount of CO₂ produced at a low EO ratio increase slowly until it achieves the maximum range of around 800 K to 900 K. The yield of CO₂ produced in oxygen-induced cracking is lower than CO (Liu et al., 2004). A lower yield of CO₂ is advantageous as it helps to reduce the release of greenhouse gases into the atmosphere. Therefore, the addition of oxygen to facilitate the cracking could create a more environmentally friendly process than the conventional cracking process.

The production of ethane is illustrated in Figure 2g. The R1 mainly triggers the production of ethane. The number of moles of ethane increases from 573 K to a maximum point around 750 K. At higher temperatures, the amount of ethane decreases due to the R11. The availability of carbon in the reactants promotes higher ethane synthesis.

Figure 2h illustrates the ethylene production in the cracking of n-eicosane with oxygen at 1 bar. It has been observed that ethylene conversion increase with the increase in temperature in all EO ratio. Higher EO ratio recorded higher ethylene yield due to the higher number of carbons in reactant at R1. Cracking stated in the R1 is mainly responsible for ethylene production before side reactions occur.

By comparing Figures 2b and 2c, it can be observed that the formation of methane as the primary carbon product at a lower temperature than pure carbon. The analysis can deduce that methane formation is most prevailing before reaching a certain cracking point at a higher temperature (Henderson et al., 2020).

When the yield of ethane starts to drop from its maximum point around 750 K, it is recorded that there is an increasing trend in ethylene yield as shown in Figure 2h. This situation is mainly attributed to the R11 that converts ethane to ethylene. In other words, with the increase in temperature, the production of 1-olefin is higher than n-paraffins with more than one carbon.

4. Conclusions

Based on the total Gibbs energy minimisation method, the thermodynamics analysis of FOBS to light hydrocarbon favours the production of hydrogen, carbon and methane. Under the studied temperature, a higher temperature favoured hydrogen production while a lower temperature favoured methane production. The maximum yield of main products is achieved at an EO ratio 0.95:0.05 under the investigated temperature at 1 bar. The Optimum condition for hydrogen production is achieved at temperature 1273 K at 1 bar while for hydrocarbon production, methane production is optimum at temperature 573 K at 1 bar pressure. The atmospheric pressure operating condition is cost-saving compared to a high-pressure condition that needs extra care, especially safety. The addition of oxygen in the cracking process is beneficial as only a small

modification is needed to supply an oxygen stream into the reactor. Further investigations on the detailed mechanism of each reaction can be performed to provide a better understanding of the process.

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