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THE INFLUENCE OF REFLUX TEMPERATURE ON THE YIELD OF TRANSPORTATION FUELS DURING PLASTIC WASTE PYROLYSIS

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Abstract: The operational parameters of the reflux play an important role during a plastic waste pyrolysis process as it recirculates the heavy hydrocarbons into the reactor for further molecule scissoring. The impact of the reflux temperature on the pyrolysis oil composition was investigated and the results are summarized in this paper. A plastic waste blend containing HDPE, LDPE, PP and PS was pyrolyzed in a laboratory scale batch reactor in the reflux temperature range of 150–300 °C, while the obtained pyrolysis oil was further processed by atmospheric distillation. Results show that by decreasing the reflux temperature the yield of gasoline range hydrocarbons can be significantly enhanced: 285 $g/kg_{solid\ waste}$ gasoline yield was measured at 300 °C reflux temperature, while 150 °C resulted 446 $g/kg_{solid\ waste}$. Additionally, the pyrolysis gas yield containing mostly C_1-C_4 hydrocarbons is also enhanced by lowering the reflux temperature. Results show, that producing gasoline and diesel range hydrocarbons from plastic wastes through pyrolysis is a viable method to reduce the accumulation of plastic wastes and could provide value added transportation fuels.

Keywords: Plastic waste, pyrolysis, reflux temperature

INTRODUCTION

Plastic waste generation and accumulation became a serious problem in the world. The global plastic waste generation reached 302 Mt in 2015, while the total plastic waste ever generated from primary plastics had reached 5,800 Mt [1]. By projecting current global waste management trends to 2050, it is assumed that the primary plastic waste generated will be around 26,000 Mt [1]. 29.1 Mt of plastic ended up in waste stream in the EU during the year of 2018 [2]. About 24.9% of this plastic waste was landfilled, 32.5% was recycled while 42.6% was utilized for energy recovery [2]. As the amount of plastic waste continuously increases, some alternative methods are being developed

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including pyrolysis, which is a promising method to generate value-added liquid fuel suitable for transportation as it can reduce the carbon footprint of the transportation industry and optimize waste management towards zero landfilling [3].

The pyrolytic products can be divided into a gas, liquid, and solid fraction. Based on the process parameters the gas/liquid ratio can be changed. The influencing factors of plastic waste pyrolysis were investigated by several researchers such as temperature [4–6], pressure [7, 8], time [5, 9], type of reactor [10, 11], catalyst [12–14] and plastic waste material used [15–20]. Very limited information can be found in the literature about the reflux temperature influence on the pyrolysis oil composition [21]. Therefore, the main goal of this study is to investigate the impact of the reflux temperature change on the plastic waste pyrolysis process. A batch reactor equipped with a reflux condenser was utilized for pyrolysis runs, while the pyrolysis oils were further processed by atmospheric distillation to determine the transportation fuel yields.

1. Materials and Methods

The pyrolysis runs were performed in a laboratory scale batch reactor equipped with a temperature controlled reflux. The reflux is a simple shell-and-tube heat exchanger, where the temperature and the flow rate of the cooling air can be adjusted in order to control the temperature of the pyrolysis vapors exiting the reflux. The vapors exiting the reflux are condensed in a secondary, water cooled heat exchanger and the liquid product (pyrolysis oil) is collected in a product container at room temperature. The remaining gases were collected in a sample bag and flared after the measurements. *Figure 1* shows the schematic illustration of the measurement system.

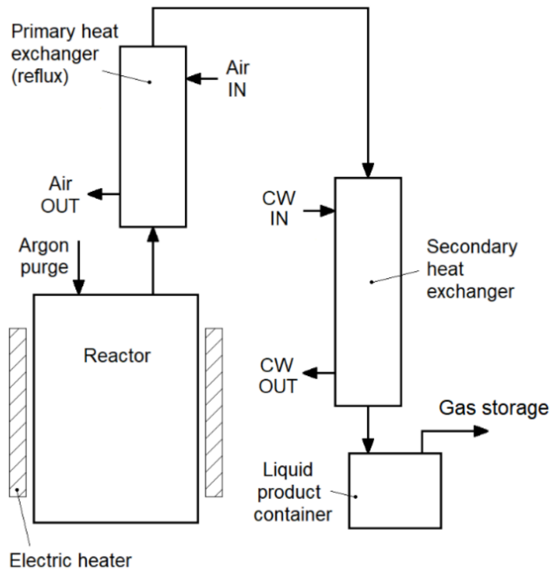

Figure 1
Schematic illustration of the measurement system

A mixture of PP, LDPE, HDPE and PS was used representing the typical plastic demand in the European Union in 2018 [2]. Based on the data found in [2] the solid plastic waste blend used in this study contained 34.8% PP, 31.6% LDPE, 22.0% HDPE and 11.6% PS. 200 g solid waste blend was loaded into the reactor in each case, while the temperature of the reactor heater was set to 700 °C. Four pyrolysis measurements were performed while the reflux temperature was gradually increased from 150 °C to 300 °C with 50 °C increments. The obtained pyrolysis oils were further processed by atmospheric distillation to separate the gasoline and diesel range hydrocarbons. The 20–200 °C range was considered as gasoline, while the 200–350 °C was considered as diesel range hydrocarbons.

2. RESULTS

The mass distribution of the products at different reflux temperatures is summarized in *Table 1*.

Table 1

The distribution of products at different reflux temperatures

Reflux temperature, °C	Gas, %	Liquid (oil), %	Solid, %
150	32.2	62.5	5.3
200	27.9	66.7	5.4
250	17.8	76.7	5.5
300	15.4	80.7	3.9

Based on the obtained results it can be stated that the temperature of the reflux has a significant impact on the liquid and gas yield. The influence is not evident in the case of solid residues. The main function of the reflux is to capture heavy hydrocarbon molecules and return them into the reactor for further molecule scissoring. Typically, the heavier hydrocarbon molecules have higher boiling temperatures as well, thus, it is supposed that by decreasing the reflux temperature the amount of lighter hydrocarbon molecules is increased in the final pyrolytic liquid product. It follows that the gaseous product is intensified as the reflux temperature decreases. This effect lowers the total liquid product during the pyrolysis process. Additionally, the more evaporation-condensation cycle increases the time necessary to complete the pyrolysis process as seen in *Figure 2*, where the pyrolysis vapor temperatures exiting the reflux are depicted during the pyrolysis runs. Based on these results it can be stated that by increasing the reflux temperature the pyrolysis oil yield can be increased while the time necessary to finish the pyrolysis can be decreased.

The results of the atmospheric distillation is shown in *Figure 3*. It can be seen that by lowering the reflux temperature the amount of the gasoline range hydrocarbons in pyrolysis oil can be significantly intensified. The presence of hydrocarbons with boiling temperatures higher than 350 °C even at 150 °C reflux temperature indicates that the efficiency of the reflux is low.

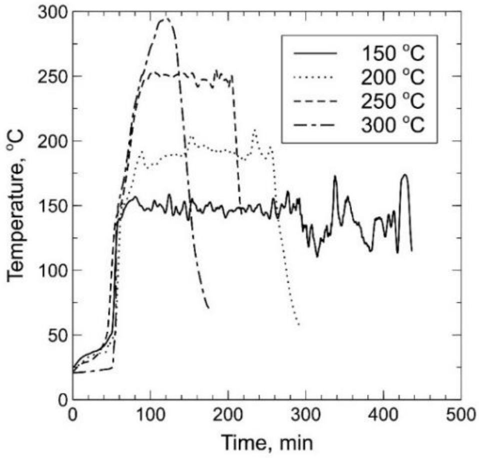

Figure 2
Typical temperature stability of vapors exiting the reflux.
The heating procedure of the reactor started at zero minute

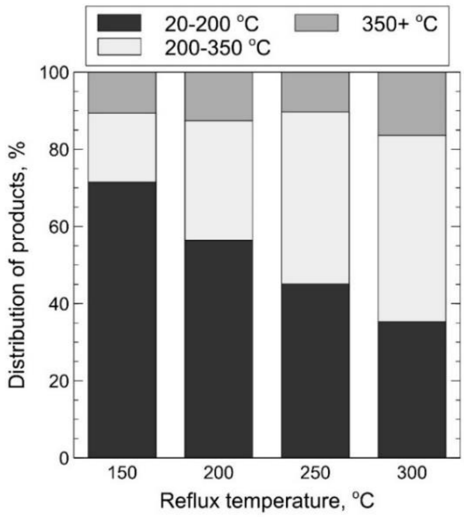

Figure 3
Distribution of different products in pyrolysis oil

Thus, an important conclusion is that reflux design might also play an important role in the pyrolysis process. Although the amount of pyrolysis oil decreases by decreasing the reflux temperature, the gasoline yield increases as shown in *Figure 4*. The highest gasoline yield of 446 $g/kg_{solid\ waste}$ was measured at 150 °C reflux temperature, which is roughly 0.6 $L/kg_{solid\ waste}$ by considering 0.75 g/ml density. On the other side, the highest diesel yield of 390 g/kg$_{solid\ waste}$ was measured at 300 °C reflux temperature. These results show, that lowering the reflux temperature is a viable method in cases when maximizing gasoline production is desirable.

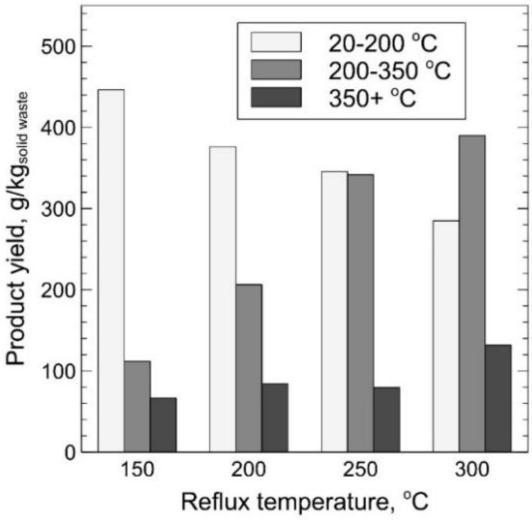

Figure 4

Yields of gasoline (20–200 °C) and diesel (200–350 °C) range hydrocarbons

Based on these results, both the gasoline yield and the flammable gaseous pyrolysis product yield can be significantly enhanced by lowering the reflux temperature. The gaseous product contains mainly C_1-C_4 hydrocarbons, thus, the heating value and the flow rate of the gas might be high enough to cover the heat requirement of the reactor. Investigating this assumption is one possible direction of the research. Additionally, eliminating the distillation residue (350+ °C portion), i.e. converting it into fuel product would be an important requirement.

CONCLUSIONS

The influence of the reflux temperature on the gasoline and diesel yield during plastic waste pyrolysis was experimentally investigated in an electrically heated batch reactor. The plastic waste blend contained PP, LDPE, HDPE and PS in a ratio representing the typical plastic demand in the EU in 2018. Results show, that although the pyrolysis oil yield is decreasing by lowering the reflux temperature, the gasoline yield increases as the reflux recirculates the heavy hydrocarbon molecules into the reactor for further molecule scissoring. The highest gasoline yield of 446 g/kg$_{solid\ waste}$ was measured at 150 °C reflux temperature while the highest diesel yield of 390 g/kg$_{solid\ waste}$ was measured at 300 °C reflux temperature.

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