Thermal Pyrolysis of Used Lubricant and Cooking Oil Mixtures

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Abstract: Pyrolysis is the one solution to recycle hydrocarbon-based waste material such as used lubricant and cooking oil. The aim of this research was to determine the effect of temperature and sample ratio on the liquid yields of a mixture of used lubricant and cooking oil. The semi batch reactor was used with a constant nitrogen flow rate of 5 mL/min. Three different ratios of sample mixture were applied in this experiment: 0.5:1, 1:1, and 1.5:1, and three different temperatures: 400°C, 450°C, and 500°C. The thermal pyrolysis of a mixture of used lubricant and cooking oil was deemed as the most effective pyrolysis to produce liquid fraction was obtained from reaction condition with the sample mixture ratio of 0.5:1 at 500°C. At this reaction condition, the liquid yield was 58.90% which consist of 64.12% were C1-C3 and 29.54% were C5-C15. Liquid fraction is predicted to increase as the temperature increase and the ratio of used lubricant to cooking oil decrease. When the ratio is increased, more gas fraction is produced.

Keywords: Used oils; and plastic waste; thermal pyrolysis

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Introduction

Pyrolysis is the one solution to recycle waste materials like used cooking and lubricant oil into useful material such as fuel (Alfernando, Sarip, Anggraini, Nazarudin, 2019; Prabasari, Sarip, Rahmayani, Nazarudin, 2019; Fitriyanti, 2020), reduced sulphur content of fuel (Bhaskar, Uddin, Muto, Sakata, Omura, Kimura, Kawakami, 2004), surfactants (Sharma, Toor, Brandão, Pedersen, Rosendahl, 2021), plastisizer (Cai, Yue, Hao, & Ma, 2020), and etc. At this method, waste material is heated at a high temperature and then break into new material. Pyrolysis can be carried out using catalyst (Alfernando et al., 2019; Prabasari et al., 2019; Fitriyanti, 2020) or without catalyst (Ayodeji and Oni, 2018; Alfernando, Nugraha, Prabasari, Haviz, Nazarudin, 2020). Due to a quite expensive catalyst, thermal cracking is more preferred despite its limitation in selectivity (Gaur, Mishra, Chowdhury, Baredar, Verma, 2020).

Santhoshkumar and Ramanathan (2010) reprocess used lubricant oil or waste engine oil (WEO) with pyrolysis method. Pyrolysis was done by heating the used lubricant oil at varilated temperatures above the saturation temperature in the reactor with no oxygen. The result showed that the optimal temperature was at 500°C. The compositions in a liquid product were 62.74% alkanes (paraffins), and the rests were ketones, alcohol, acids and others.

Bio-oil from thermal pyrolysis of used cooking oil have been produced at the laboratory scale. The result showed that the best condition to produce bio-oil (52.34%) was at 550°C (Nazarudin, Prabasari, Ulyarti, Susilawati, Oktadio, 2020). Thermal pyrolysis of polyethylene terephthalate (PET) plastic waste and palm fibre mixtures were carried out at 400°C, 425°C, 450°C in 10, 20, 30 minutes reaction time in which the
highest oil liquid product (17%) was produced at 450°C for 10 minutes reaction (Nazarudin, Jayanti, Alfernando, Prabasari, Ulyarti, Sarpì, 2020).

Method

The study was conducted by thermal pyrolysis in the semi-batch reactor (Figure 1). The samples and the liquid products of pyrolysis were analyzed by gas chromatography-mass spectrometry (GC-MS). There were three different ratios of samples 0.5:1, 1:1, and 1.5:1, and three different temperatures: 400°C, 450°C, and 500°C. The used cooking and lubricant oil were mixed and reacted with ratio and temperature as can be seen in Table 1. The nitrogen was flowed constantly 5 ml/min during 30 minutes reaction. The liquid products were taken every 5 minutes during the reaction.

![Figure 1. Schematic of Semi batch reactor (R-01: batch reactor, T-02: Oil Liquid Product storage tank, IE-01: Ice Trap)](image)

| Reaction number | Code | Ratio | Code | Temperature (°C) |
|-----------------|------|-------|------|-----------------|
| 1               | -1   | 0.5:1 | -1   | 400             |
| 2               | -1   | 0.5:1 | 1    | 500             |
| 3               | 1    | 1.5:1 | -1   | 400             |
| 4               | 1    | 1.5:1 | 1    | 500             |
| 5               | 0    | 1:1   | 0    | 450             |
| 6               | 0    | 1:1   | 0    | 450             |
| 7               | 0    | 1:1   | 0    | 450             |

Information:

\[X_1 = \text{ratio of used lubricant and cooking oil (0.5:1, 1:1, 1.5:1).}\]
\[0.5 = 5\text{ gram} ; 1 = 10\text{ gram} ; 1.5 = 15\text{ gram}\]
\[X_2 = \text{temperature (°C) (400, 450, 500)}\]

Result and Discussion

Composition of waste material

The composition of waste material were examined using GC-MS and the results are presented in Table 2 and Table 3.

| No. | RT  | % Area | SI | Comounds                              | Formula     | MW  |
|-----|-----|--------|----|---------------------------------------|-------------|-----|
| 1   | 1.98| 2.45   | 97 | Carboxylic acid                       | CH₃NO₂      | 61  |
|     |     |        |    | Nitrous oxide                         | N₂O         | 44  |
|     |     |        |    | 1-Propene, 2-methyl                   | C₃H₈         | 56  |
| 2   | 2.05| 60.59  | 96 | 3,5-Diisopropyl-1,2,4 trithiolane     | C₈H₁₆S₃     | 208 |
| 3   | 23.61| 5.72  | 80 | Eicosane                              | C₂₀H₄₂      | 282 |
| 4   | 32.57| 1.80  | 95 | Heneicosane                           | C₂₁H₄₄      | 296 |
|     |     |        |    | Tricosane                             | C₂₃H₄₄      | 324 |
| 5   | 34.70| 2.63  | 96 | Heneicosane                           | C₂₁H₄₄      | 296 |
| 6   | 36.73| 3.58  | 97 | Eicosane                              | C₂₀H₄₂      | 282 |
|     |     |        |    | Tricosane                             | C₂₁H₄₄      | 324 |
| 7   | 38.67| 5.02  | 97 | Tricosane                             | C₂₀H₄₂      | 282 |
|     |     |        |    | Heptadecane                           | C₁₇H₃₆      | 240 |
| 8   | 40.54| 5.52  | 97 | Octadecane                            | C₁₈H₃₈      | 254 |
|     |     |        |    | Eicosane                              | C₂₀H₄₂      | 282 |
|     |     |        |    | Tricosane                             | C₂₁H₄₄      | 324 |
| 9   | 42.32| 5.22  | 97 | Tricosane                             | C₂₀H₄₂      | 282 |
|     |     |        |    | Pentacosane                           | C₂₅H₅₂      | 352 |
Thermal pyrolysis of mixture of used cooking and lubricant oil

There were three types of products produced by thermal pyrolysis of a mixture of used cooking oil and used lubricant oil. The products were liquid, gas, and coke with the liquid was the main product in this reaction.

The liquid yields are the mass ratio of liquid fraction and the total sample mixture (used lubricant and cooking oil). Figure 2 shows that the highest liquid yields for thermal pyrolysis of a mixture of used cooking oil and used lubricant oil was at reaction number 2 (ratio 0.5:1, temperature 500°C). The liquid yield at this condition was 58.9%. The yield for liquid fraction in this experiment is much lower than previously reported (Trabelsi, Zaafouri, Baghdadi, Naoui, Ouerghi, 2018) who reported 80% liquid yields over thermal pyrolysis of used cooking oil. Besides the difference in the feed, the high amount of liquid yields is due to a much higher temperature used in the pyrolysis 800°C (Trabelsi et al., 2018).

The GC-MS analysis shows that the liquid fraction (reaction number 2) of thermal pyrolysis of the mixture consist of 19 components as shown in Table 4. Thermal pyrolysis has cracked the C₁ to C₃₀ components in the used lubricant and cooking oil mixtures to produce C₂ - C₃₀ hydrocarbon compounds in the liquid fraction which is categorized as diesel-like fuel. Among these products in the liquid fraction, 64.12% were C₁-C₅ and 29.54% were C₅-C₁₅. The cumulative of liquid yields for thermal pyrolysis used cooking and lubricant oil mixtures can be seen in Figure 4. This graphs shows the progress of liquid production during pyrolysis and that the liquid production at reaction number 2 is increasing in quite constant rate.

The gas conversion for thermal pyrolysis of used lubricant and cooking oil mixture is shown in Figure 4. This result is higher than thermal pyrolysis of waste lubricant oil at similar temperature reported previously (Fuentes, Font, Gómez-Rico, Martín-Gullón, 2007). Controlling the ratio of used oil may help researcher to obtain what product is more preferred, either liquid or gas. The other study of co-pyrolysis was reported by Phetyim, Pivsa-Art. where used lubricant oil and

### Table 3. The composition of used cooking oil as shown by GC-MS

| No. | RT  | % Area | SI  | Compounds                                           | Formula Molecules | Molecular Weight |
|-----|-----|--------|-----|----------------------------------------------------|-------------------|------------------|
| 1.  | 39.92 | 20.95  | 90  | 13-Oxabicyclo[10.1.0]tridecane                    | C₁₃H₂₂O           | 182              |
|     |      |        |     | Oleic Acid                                        | C₁₃H₂₄O₂          | 282              |
|     |      |        |     | cis-7-tetradecene-1-ol                            | C₁₃H₂₂O           | 212              |
| 2.  | 41.75 | 37.00  | 86  | 1,3-Dipalmitoylglycerol                            | C₁₃H₂₆O₃          | 569              |
|     |      |        |     | 3-[(2-Aminoethoxy)(hydroxy)phosphoryl]oxy]-2-      | C₁₃H₂₆NO₃P        | 691              |
|     |      |        |     | (palmitityloxy)propyl palmitate                    |                   |                  |
|     |      |        |     | Glyceryl 1,3-diesterate                            | C₁₃H₂₆O₃          | 624              |
|     |      |        |     | Docosanoic acid                                   | C₂₂H₄₄O₂          | 340              |
| 3.  | 44.57 | 7.88   | 90  | (Z,6),(Z,9)-Pentadecadien-1-ol                    | C₁₅H₃₀O           | 224              |
|     |      |        |     | Cyclooctadecyne                                   | C₁₅H₃₂O           | 224              |
|     |      |        |     | 1,6,6,11-Hexadecatriene                            | C₁₅H₃₂O           | 224              |
|     |      |        |     | 1,6,11,13-Octadecatriene                           | C₁₅H₃₂O           | 224              |
| 4.  | 44.72 | 34.17  | 88  | 1,3-Didein                                        | C₁₃H₂₆O₂          | 621              |
|     |      |        |     | 9-Octadecen-1-ol,(Z)                               | C₁₃H₂₆O₂          | 268              |
|     |      |        |     | 13-octadecen,(Z)                                   | C₁₃H₂₆O₂          | 266              |

![Figure 2](image-url)

**Figure 2** The liquid yields for thermal pyrolysis of mixture of used lubricant and cooking oil mixture
mixed plastic waste were cracked to produce a diesel-like fuel (Phetyim & Pivsa-Art, 2018).

The coke conversion for thermal pyrolysis of used lubricant and cooking oil mixture for every reaction number is shown in Figure 5. Temperature plays an important role in the completeness of the thermal pyrolysis reaction. The lowest temperatures applied in this experiment produced the highest coke production. The higher the temperature, the lower the Cokes conversion.

**Response surface analysis for thermal pyrolysis of used lubricant and cooking oil**

The response surface analysis was applied to obtain the optimum reaction condition for thermal pyrolysis of used lubricant and cooking oil. As seen in Figure 5, all graphs in the surface plots are flats without any indication can reach a maximum or minimum peak. This is due to the remote experimental design region. From these graphs, it can be seen that the temperature should be higher than 500°C and the ratio should be lower than 0.5:1 in order to obtain maximum liquid fraction (Figure 6). In other words, if liquid fraction is preferred, used lubricant oil oil should be used far less than used cooking oil. However, more used lubricant oil should be used more if gas fraction is preferred (Figure 6). Since coke is the product that should be minimised, lower ratio is more preferred while no agreement on temperature can be made (Figure 7).

**Table 4. The composition of liquid fraction (reaction number 2) from thermal pyrolysis of used lubricant and cooking oil mixture as shown by GC-MS**

| No. | RT  | % Area | SI | Compounds                              | Formula Molecules | Molecular Weight |
|-----|-----|--------|----|----------------------------------------|-------------------|------------------|
| 1.  | 1.97| 49.65  | 92 | Oxalid acid                            | C₂H₂O₄             | 90               |
|     |     |        |    | Carbamic acid                          | CH₂NO₂             | 61               |
|     |     |        |    | 1,1-dibromo-2-chloro-2-fluoro          | C₂H₂BrClF          | 250              |
|     | 2.03| 2.17   | 25 | 4-phenoxy-trimethylsilyl ester         | C₁₃H₂₆O₃Si         | 252              |
|     | 2.24| 6.51   | 94 | Acetaldehyde                           | C₃H₄O               | 74               |
|     | 3.11| 0.94   | 81 | 2-Propenoic Acid                       | C₃H₆O₂              | 44               |
| 4.  | 2.38| 4.10   | 80 | Furan, 2-methyl                        | C₃H₆O              | 82               |
| 5.  | 2.53| 11.20  | 96 | Acetic Acid                            | C₃H₄O₂             | 60               |
| 6.  | 2.68| 3.27   | 95 | 2-Propylene oxide, 1-hydroxy           | C₃H₆O₂              | 44               |
| 7.  | 3.11| 0.94   | 81 | 2-Propenoic Acid                       | C₃H₆O₂              | 72               |
| 8.  | 4.22| 1.46   | 85 | 1,2-butanediene, 3-methoxy            | C₄H₁₀O              | 84               |
| 9.  | 6.51| 0.25   | 96 | 2,5-Hexanedione                        | C₅H₁₀O₂             | 114              |
| 10. | 7.93| 0.28   | 85 | 2-Pentanone, 3-methyl                  | C₅H₁₀O              | 100              |
| 11. | 36.64| 0.39  | 92 | Heneicosane                            | C₂₁H₄₄              | 296              |
|     |     |        |    | Pentacosane                            | C₂₁H₴₂              | 352              |
|     |     |        |    | Docosane                               | C₂₁H₴₆              | 310              |
|     |     |        |    | Tricosane                              | C₂₁H₴₈              | 324              |
|     |     |        |    | Pentacosane                            | C₂₁H₴₂              | 352              |
| 12. | 38.58| 0.45  | 94 | Tetracosane                            | C₂₂H₵₂              | 282              |
|     |     |        |    | Octadecane, 2-methyl                  | C₁₈H₳₂              | 268              |
|     |     |        |    | Triacontane                           | C₂₃H₵₂              | 422              |
|     |     |        |    | Germacrane                            | C₂₃H₳₂O             | 210              |
| 14. | 41.35| 0.23  | 83 | Tridecan                              | C₂₃H₳₂O₂            | 268              |
|     |     |        |    | Cyclopentane -heneicosyl              | C₂₅H₴₈              | 422              |
|     | 41.65| 0.44  | 84 | Docosanoic Acid                       | C₂₅H₴₸₂             | 340              |
| 15. | 42.21| 0.33  | 92 | Tricosane                              | C₂₃H₴₈              | 324              |
|     |     |        |    | Eicosane                               | C₂₃H₴₂              | 282              |
| 16. | 42.84| 0.24  | 80 | Octadecane, 1-chloro                  | C₁₈H₳₇Cl            | 288              |
|     |     |        |    | 1-Hexacosanol                         | C₁₈H₳₆O            | 382              |
|     |     |        |    | Hexadecane, 1-chloro                  | C₁₈H₳₆Cl            | 260              |
| 17. | 44.64| 0.36  | 82 | 9-octadeceanal                        | C₁₈H₳₆O            | 266              |
| 18. | 45.71| 0.25  | 81 | Di-octyl phthalate                    | C₁₈H₳₆O₄            | 390              |
Conclusion

Thermal pyrolysis of a mixture of used lubricant and cooking oil mixture produce diesel like fuel. The most effective pyrolysis of a mixture of used cooking oil: used lubricant oil is in ratio 0.5:1 and temperature 500°C. The surface plots of thermal pyrolysis of used lubricant and cooking oil mixture showed that the optimum condition for liquid and gas yields was beyond the experimental design applied in this experiment.

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