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Thermal Pyrolysis of Used Lubricant and Cooking Oil Mixtures

Nazarudin [1,2,4*], S.P.Amalia[2], Afrida[2], Ulyarti[3,4]

[1] Chemical Engineering Department, Faculty of Sain and Technology, Universitas Jambi, Jambi, Indonesia.
[2] Chemsitry Education Department, FKIP, Universitas Jambi, Jambi, Indonesia
[3] Department of Agriculture Product Technology, Faculty of Agriculture, University of Jambi, Indonesia
[4] Centre of Excellent in Bio-Geo Material, University of Jambi, Indonesia

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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: 400⁰C, 450⁰C, and 500⁰C. 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 500⁰C. At this reaction condition, the liquid yield was 58.90% which consist of 64.12% were C_1-C_3 and 29.54% were C_5-C_{15}. 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 despites 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 variated temperatures above the saturation temperature in the reactor with no oxygen. The result showed that the optimal temperature was at 500ᵒC. 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 550ᵒC (Nazarudin, Prabasari, Ulyarti, Susilawati, Oktadio, 2020). Thermal pyrolysis of polyethylene terephthalate (PET) plastic waste and palm fibre mixtures were carried out at 400ᵒC, 425ᵒC, 450ᵒC in 10, 20, 30 minutes reaction time in which the

Email: nazarudin@unja.ac.id

highest oil liquid product (17%) was produced at 450°C for 10 minutes reaction (Nazarudin, Jayanti, Alfernando, Prabasari, Ulyarti, Sarip, 2020).

Method

The study was conducted by thermal pyrolysis in the semi-batch reactor (**Figure 1)**. The samples and the liquid products of pyrolisis 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 bee 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.

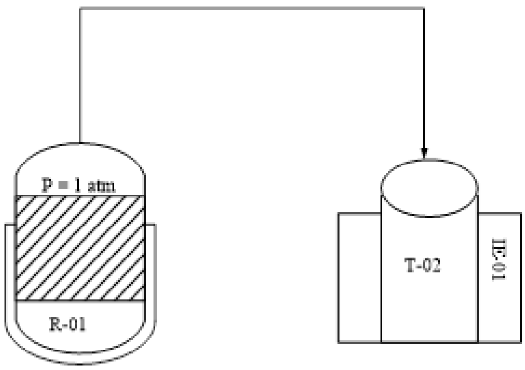

Figure 1. Schematic of Semi batch reactor (R-01: batch reactor, T-02: Oil Liquid Product storage tank, IE-01: Ice Trap)

Table 1. Thermal pyrolysis design (Gaspers, 1995)

Reaction number	X_1		X_2	
	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 = ratio of used lubricant and cooking oil (0.5:1, 1:1, 1.5:1).
0.5 = 5 gram; 1 = 10 gram; 1.5 = 15 gram
X_2 = 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.

Table 2. The composition of used lubrican oil as shown by GC-MS

No.	RT	% Area	SI	Compounds	Formula	MW
1.	1.98	2.45	97	*Carbamic acid*	CH_3NO_2	61
				Nitrous oxide	N_2O	44
2.	2.05	60.59	96	*1-Propene, 2-methyl*	C_4H_8	56
3.	23.61	5.72	80	*3,5- Diisopropyl-1,2,4 trithiolane*	$C_8H_{16}S_3$	208
4.	32.57	1.80	95	*Eicosane*	$C_{20}H_{42}$	282
				Heneicosane	$C_{21}H_{44}$	296
				Tricosane	$C_{23}H_{48}$	324
5.	34.70	2.63	96	*Heneicosane*	$C_{21}H_{44}$	296
				Eicosane	$C_{20}H_{42}$	282
				Tricosane	$C_{23}H_{48}$	324
6.	36.73	3.58	97	*Tricosane*	$C_{23}H_{48}$	324
				Eicosane	$C_{20}H_{42}$	282
				Heptadecane	$C_{17}H_{36}$	240
7.	38.67	5.02	97	*Octadecane*	$C_{18}H_{38}$	254
				Eicosane	$C_{20}H_{42}$	282
				Tricosane	$C_{23}H_{48}$	324
8.	40.54	5.52	97	*Tricosane*	$C_{23}H_{48}$	324
				Eicosane	$C_{20}H_{42}$	282
				Octadecane	$C_{18}H_{38}$	254
9.	42.32	5.22	97	*Tricosane*	$C_{23}H_{48}$	324
				Eicosane	$C_{20}H_{42}$	282
				Pentacosane	$C_{25}H_{52}$	352

No.	RT	% Area	SI	Compounds	Formula	MW
				Octadecane	$C_{18}H_{38}$	254
10.	44.03	4.42	97	*Tricosane*	$C_{23}H_{48}$	324
				Octadecane	$C_{18}H_{38}$	254
11.	45.68	3.05	95	*Tricosane*	$C_{23}H_{48}$	324
				Octadecane	$C_{18}H_{38}$	254
				Pentacosane	$C_{25}H_{52}$	352

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*	$C_{12}H_{22}O$	182
				Oleic Acid	$C_{18}H_{34}O_2$	282
			89	*Cis-7-tetradecene-1-ol*	$C_{14}H_{28}O$	212
2.	41.75	37.00	86	*1,3-Dipalmitoylglycerol*	$C_{35}H_{68}O_5$	569
			85	*3-{[(2-Aminoethoxy)(hydroxy)phosphoryl]oxy}-2-(palmitoyloxy)propyl palmitate*	$C_{37}H_{74}NO_8P$	691
			84	*Glyceryl 1,3-distearate*	$C_{39}H_{76}O_5$	624
				Docosanoic acid	$C_{22}H_{44}O_2$	340
3.	44.57	7.88	90	*(Z)6,(Z)9-Pentadecadien-1-ol*	$C_{15}H_{28}O$	224
			87	*Cyclododecyne*	$C_{12}H_{20}$	164
				1,E-6,Z-11-Hexadecatriene	$C_{16}H_{28}$	220
				1,E-11,Z-13-Octadecatriene	$C_{18}H_{32}$	248
4.	44.72	34.17	88	*1,3-Diolein*	$C_{39}H_{72}O_5$	621
			87	*9-Octadecen-1-ol,(Z)*	$C_{18}H_{36}O$	268
			86	*13-octadecenal,(Z)*	$C_{18}H_{34}O$	266

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.

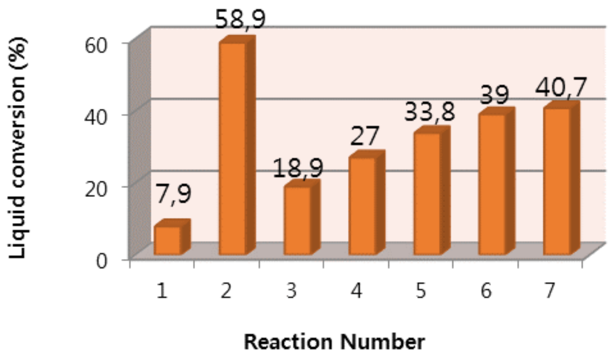

Figure 2 The liquid yields for thermal pyrolysis of mixture of used lubricant and cooking oil mixture

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^{o}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^{o}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 C_1 to C_{39} components in the used lubricant and cooking oil mixtures to produce C_2 - C_{30} hydrocarbon compounds in the liquid fraction which is categorized as diesel-like fuel. Among these products in the liquid fraction, 64.12% were C_1-C_3 and 29.54% were C_5-C_{15}. The cumulative of liquid yields for thermal pyrolysis used cooking and lubricant oil mixtures can bee seen in **Figure 4**. This graphs shows the progress of liquid production during pyrolisis 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 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 ttemperatures applied in this experiment produced the highest coke production. The higher the temperature, the lower the Cokes conversion.

Respons surface analysis for thermal pyrolysis of used lubricant and cooking oil

The respons surface analysis was applied to obtain the optimum reaction condition for thermal pyrolysis of used lubricant and cooking oil. As seen in **Figure 5-7**, 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^{o}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 pyrolisis 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*	$C_2H_2O_4$	90
				Carbamic acid	CH_3NO_2	61
				1,1-dibromo-2-chloro-2-fluoro	$C_3H_2Br_2ClF$	250
2.	2.03	2.17	25	*4-phenoxy-,trimethylsily ester*	$C_{13}H_{20}O_3Si$	252
			24	*3,4 –Dimethoxyphenylpentanoic acid*	$C_{16}H_{26}O_4Si$	310
			23	*4,4-Dinitro-6,6-ethylenedioxy-4,5,6,7-benzofuroxane*	$C_8H_8N_4O_8$	288
				Benzo cazepin,7,8,9-trimethoxy	$C_{13}H_{19}NO_3$	237
			22	*bixindial*	$C_{24}H_{28}O_2$	348
3.	2.15	23.45	67	*Hexaborane-12*	B_6H_{12}	78
			60	*1,2-tetramethylenediborane*	$C_4H_{12}B_2$	82
			59	*Nickel 1-amino-1,9-diisothiocipno-4,8-di-azaudecamine*	$C_{11}H_{22}N_6NIS_2$	360
			58	*Trans-2,3-Epoxyoctane*	$C_8H_{16}O$	128
4.	2.38	4.10	80	*Furan,2-methyl*	C_5H_6O	82
5.	2.53	11.20	96	*Acetic Acid*	$C_2H_4O_2$	60
6.	2.68	3.27	95	*2-Propanone, 1-hydroxy*	$C_3H_6O_2$	74
			94	*Acetaldehyde*	C_2H_4O	44
7.	3.11	0.94	81	*2-Propenoic Acid*	$C_3H_4O_2$	72
8.	4.22	1.46	85	*1,2-butadiene,3-methoxy*	C_5H_8O	84
9.	6.51	0.25	95	*2,5-Hexanedione*	$C_6H_{10}O_2$	114
10.	7.93	0.28	85	*2-Pentanone, 3-methyl*	$C_6H_{12}O$	100
11.	36.64	0.39	92	*Heneicosane*	$C_{21}H_{44}$	296
				Pentacosane	$C_{25}H_{52}$	352
				Docosane	$C_{22}H_{46}$	310
12.	38.58	0.45	94	*Tricosane*	$C_{23}H_{48}$	324
				Pentacosane	$C_{25}H_{52}$	352
				Eicosane	$C_{20}H_{42}$	282
13.	40.44	0.55	94	*Tricosane*	$C_{23}H_{48}$	324
				Octadecane, 2-methyl	$C_{19}H_{40}$	268
				triacontane	$C_{30}H_{62}$	422
14.	41.35	0.23	83	*Germacrane*	$C_{15}H_{30}$	210
				Tridecanol	$C_{13}H_{28}O$	200
			82	*Cyclopentane -heneicosyl*	$C_{26}H_{52}$	364
15.	41.65	0.44	84	*Docosanoic Acid*	$C_{22}H_{44}O_2$	340
16.	42.21	0.33	92	*Tricosane*	$C_{23}H_{48}$	324
				Eicosane	$C_{20}H_{42}$	282
17.	42.84	0.24	80	*Octadecane, 1-chloro*	$C_{18}H_{37}Cl$	288
				1-Hexacosanol	$C_{26}H_{54}O$	382
				Hexadecane,1-chloro	$C_{16}H_{33}Cl$	260
18.	44.64	0.36	82	*9-octadecenal*	$C_{18}H_{34}O$	266
19.	45.71	0.25	81	*Di –n-octyl phthalate*	$C_{24}H_{38}O_4$	390

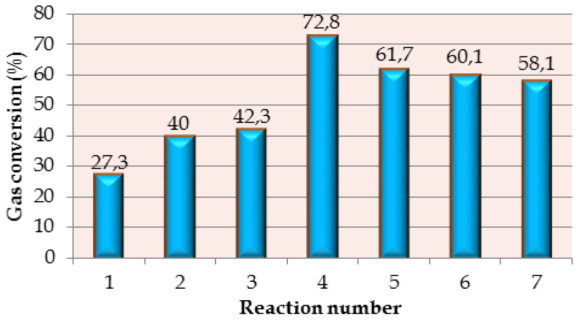

Figure 3. Gas yields for thermal pyrolysis of used lubricant and cooking oil mixture

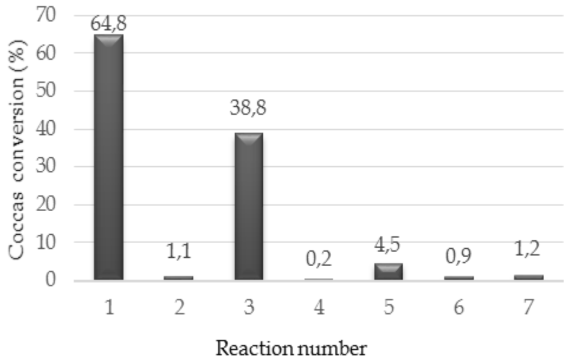

Figure 4. Coke yields for thermal pyrolysis of used lubricant and cooking oil mixture

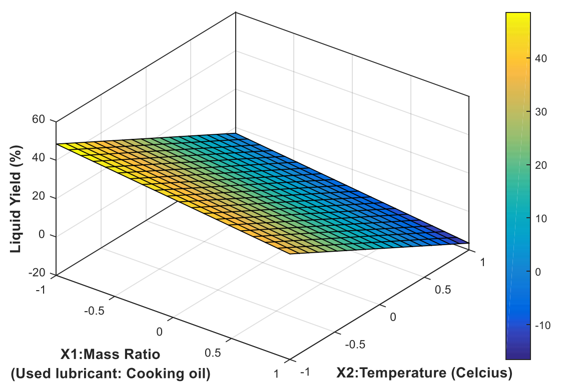

Figure 5. Surface plots of liquid yields on thermal pyrolysis of used lubricant and cooking oil mixture

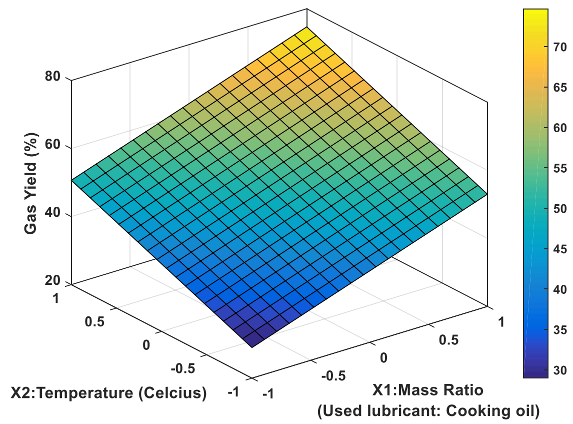

Figure 6. Surface plots of gas yields on thermal pyrolysis of used lubricant and cooking oil mixture

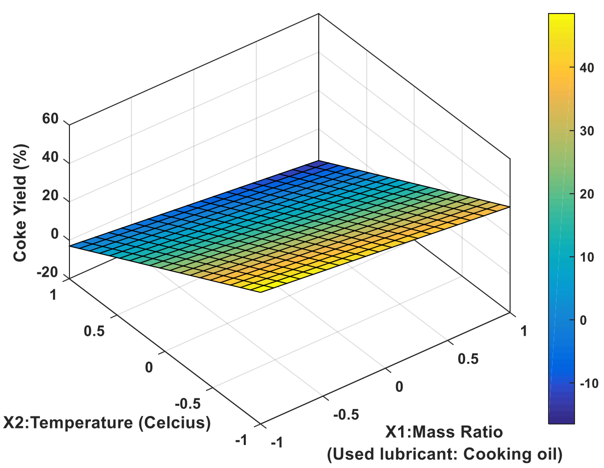

Figure 7. Surface plots of coke yields on thermal pyrolysis of used lubricant and cooking oil mixture

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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