Purification of Used Engine Oil by Pyrolysis– Adsorption Method Assisted Coal Activated Carbon

Alwathan^{1,2*,a}, Siti Hamidah Mohd-Setapar^{2,3,b}, Muh. Irwan^{1,c}, Ramli Thahir^{1,d}, Harjanto^{1,e}, Wahyudi^{1,f}, Sirajuddin^{1,g}, Damianus Samosir^{1,h}, Sarifudin Okko^{1,i}, Sekar Ayu^{1,k}, Rahmat Budi Toyo^{1,1} dan Muhammad Reza Fadilah^{1,m}

¹ Department of Chemical Engineering, Politeknik Negeri Samarinda, Jalan Dr. Ciptomangunkusumo Kampus Gunung Lipan Samarinda, 75131 Kalimantan Timur Province Indonesia

² Razak Faculty of Technology and Informatic, Universiti Teknologi Malaysia, Jalan Sultan Yahya Petra 54100 UTM Kuala Lumpur, Malaysia

³ Malaysia-Japan International Institute of Technology (MJIIT), Universiti Teknologi Malaysia, Jalan Sultan Yahya Petra

54100 UTM Kuala Lumpur, Malaysia

*,a alwathan@polnes.ac.id

Abstract—The increase in the use of engine vehicles every year is directly proportional to the amount of engine oil waste produced. Used engine oil is a class of hazardous waste that is harmful to the environment. This study aims to purify the base fuel from pyrolysis of waste engine oil by comparing integrated and non-integrated pyrolysis adsorption purification methods using activated low-rank coal as an adsorbent medium. The method used in this study is Pyrolysis using activated carbon from low-rank coal that has been carbonized at a temperature of 600 ° C and activated using H₃PO₄-NaHCO₃. Pyrolysis is carried out by adding activated carbon with a ratio of 10% (w / w) of 11 grams from the volume of base fuel samples and Pyrolysis temperature of 350°C, then carried out by a variety of methods, namely the adsorption method followed by Pyrolysis, Pyrolysis followed by adsorption, and Pyrolysis adsorption integration. From this study, the optimum value of purification was obtained in the variation of integration adsorption pyrolysis with a density of 0.7631gr / ml and a specific gravity value of 0.7639 and an API degree of 53.7318, while for viscosity value of 0.5542 cP. The results of the product analysis have entered the standard specification value of gasoline or gasoline.

Keywords—Pyrolysis; adsorption; activated carbon; base fuel; used engine oil

I. Introduction

Currently, the need for oil as one of the needs of engine vehicles always increases every year. With the large number of uses of engine oil in motorized vehicles in East Kalimantan Province, it will certainly have an impact on environmental pollution because the engine oil that has been used will be discarded because it no longer meets the standards as a lubricant. The presence of contaminants in lubricant waste, one of which is heavy metals which if discharged into the environment without recycling will be harmful to the ecosystem, both soil and water due to their non-biodegradable nature. Therefore, to reduce the impact of waste oil, it is necessary to purify to remove impurities contained in waste oil, purification can also increase the added value of waste oil itself by processing waste oil waste into liquid fuel.

There are three commonly used methods of purifying used oil into liquid fuel, namely thermal pyrolysis, catalytic pyrolysis, and microwave pyrolysis. The catalytic method uses a catalyst to assist the pyrolysis process, the thermal pyrolysis method does not use a catalyst relying only on heat in the process of breaking carbon bonds, and the microwave method utilizes electromagnetic microwaves for the breakdown of carbon bonds. However, in principle, the three methods still rely on heat in the pyrolysis process, the pyrolysis method that uses heat treatment is carried out from the initial temperature or room temperature to the intended temperature. The raw materials in the process of p pyrolysis are entered at the beginning of the process, then the reaction time of pyrolysis begins to be calculated until the reaction state is reached, which is the desired amount

of yield. Thermal energy is applied externally to the reactor in terms of conventional electrical heating by heating all substances in the reactor including the evolving pyrolysis volatiles, the surrounding gases, and the reactor chamber itself. Electric heaters have resistance heaters, and voltage controls are used to regulate the heating rate. Heating control is carried out by the control panel [3].

In the pyrolysis process, additional sorbents are needed which can add the purification value of the pyrolysis results (base fuel). Some adsorbents that can be used in absorbing metals are activated carbon, silica gel, alumina, zeolite, and polymers. The higher the addition of adsorbents, the higher the metal removal because the more surface area [4], coal has these criteria but its use in the field is still minimal. Abundant low-rank coal types are still rarely used as non-fuel, making activated carbon from coal will encourage the use of low-rank coal which is currently felt to be limited. Making activated carbon from low-rank coal to help the adsorption process is carried out by the carbonization process, then continued with the activation process. Activation is the process of treating carbon to open the pores of carbon. Adsorbents can be improved by surface capability by activation process first, processing activation can be done through physical activation and chemical activation [5]. The adsorption. process that occurs in activated charcoal has an active group located on the surface of activated charcoal interacting with the adsorbate. The influence of Van der Waals forces between the surface of activated charcoal and adsorbate causes adsorbate to be adsorbed into the pores of activated charcoal [6].

Several previous studies are references to this study, including research that aims to reduce sulfur levels in used engine oil assisted by activated carbon from activated low-rank coal using the adsorption distillation pyrolysis method, obtained the results of % sulfur removal of 82.86% with sulfur value levels of 659 ppm at an optimum temperature of 350 ° C with 10% W/W activated coal. For density analysis, the results were obtained at 0.8083 gr/mL with a %yield value of 75%, the resulting base fuel was clear yellow-brown [7]. Mardyaningsih and Leki recycle used engine oil by adsorption and pyrolysis

methods to find out some of the physical properties of base oil products. The research produced base oil with the following physical properties: clear yellow color, pungent odor, flammable, specific gravity 0.8 ml/g, viscosity 5.14 g / cm seconds and 5.45 g/cm seconds, calorific value 16,800 J/g and flash point 80-98 °C [8].

Low-rank coal or Brown Coal can be used as an adsorbent by using a combination solution of H_3PO_4 – NaHCO₃ is 2.5M on chemical activation [9]. In this study will use the same adsorbent derived from Brown Coal. with a calorific value of 4503 J/g. However, this study will focus on the purification method of pyrolysis – Adsorption, namely base fuel or raw materials which is the result of pyrolysis purified again by pyrolysis and adsorption techniques, as the main test parameter is the yield of the results obtained, then the yield of adsorption results and, or pyrolysis obtained will be measured and calculated, then it will be analyzed density, viscosity, specific gravity, color, yield, and °API (API, which is an abbreviation for the American Petroleum Institute, is a standard code that determines oil quality).

II. Research Methodology

A. Activation of coal into activated carbon

Low - rank coal is crushed to reduce its size which ranges from 100-200 mesh, then carbonized to increase the carbon value. Carbonization is carried out through a pyrolysis process at a temperature of 600° C for 3 hours. The coal that has been carbonized is then soaked using a mixed acid H₃PO₄-NaHCO₃ with a concentration of 2.5 M for 6 hours. Next, it is filtered and neutralized until the pH is constant using distilled water, then the final activation step is drying to remove the water content at a temperature of 105° C in an oven until the weight is constant.

B. Purification method

After the coal activation process has been completed, the used oil is then prepared by filtering it first as raw material in the purification process which is carried out under predetermined conditions of time, temperature and adsorbent mass ratio. The process is carried out using a Vol. 11, No. 1, pp. 26-30, April 2024

Pyrolysis distillation tool with three types of variations in the purification process as follows:

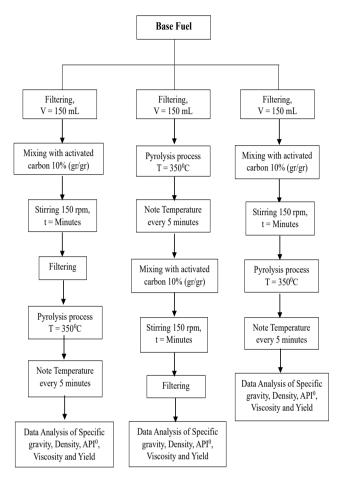


Figure 1. The variations in the purification process

III. Results and Discussion

Activated carbon from activated low-rank coal (Lowrank Coal) is used in the adsorption process as an adsorbent medium in the purification process of engine oil waste, in this case the purification process used is the adsorption process – pyrolytic distillation. Pyrolysis is a thermal decomposition process that can break long-chain hydrocarbon bonds into short-chain hydrocarbons. Before the adsorption process of pyrolysis of used oil is carried out, preparations must be made for low-rank coal to become activated carbon so that it can be used as an additive that is expected to function as an absorbent medium.

Low-rank coal is used as an absorbent medium that has gone through an activation process tested for its absorption ability by analyzing iodine number or activated carbon iodine trapping power with results obtained of 1229.62 mg / g, which value has met SNI 06-3730 - 1995 which states the value for iodine trapping power min 750 mg / g. Used oil as raw material will be pyrolyzed first using a distillation-pyrolysis device, the top result in the form of condensate is referred to as base fuel, where the results are still low in terms of quality obtained results as in Table 1 below.

Table 1. Preliminary	data on fuel	base and	gasoline standards
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Parameters	Initial base fuel data	Standard Parameters	Source
Density	0.7835 g/ml	0.715 – 0.770 g/ml	Decree of the Director General of Oil and Gas 0177.K/10/DJM. T/2018
Viscosity	0.9638 cP	$1.3 - 2.4 \text{ mm}^2/\text{s}$	ASTM D 445
Specific Gravity	0,7835	0,70 - 0,77	ASTM D 1298
API Degrees	48,9298	50 - 85	ASTM D 1250

ASTM is a measurement method standard from the American Society for Testing and Materials.

The data obtained will be used as a reference or comparison to the advanced purification process where the observational data obtained from purification through pyrolysis - adsorption with three variations of the method the first is the purification process by integration where coal that has been activated as activated carbon will be mixed simultaneously with base fuel (I), the second way is the gradual purification process, namely the base fuel is adsorption first then The results obtained are then pyrolyzed again using a pyrolysis distillation device (II), and the third is purification which is carried out in stages, where the base fuel pyrolysis process is carried out first in a pyrolysis distillation device then continued with the adsorption process (III) from the three methods obtained a comparison of results as follows.

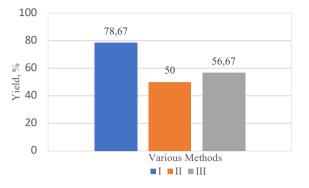


Figure 2. Comparison yields of different methods

Figure 2 shows that the integration adsorption pyrolysis method has an optimal value of % yield, which is 78.6667% higher than 20% of the other yields. This is because activated carbon acts as a catalyst that accelerates and increases vield, this also shows that the addition of adsorbents can increase the cracking reaction (decomposing) that occurs in the pyrolysis process. A cracking reaction is an endothermic reaction where this reaction involves the process of breaking the hydrocarbon chain, the same thing does not happen in methods (II) and (III) activated carbon uses more functions as an adsorbent and does not act as an catalyst as applies to a method (I).

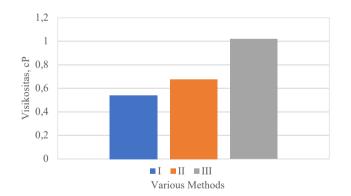


Figure 3. Comparison yields of different methods

From Figure 3. The visibility value that is closest to the value of gasoline is the pyrolysis method which is carried out in an integrated manner with the adsorption process (I), which is 0.5542 cP. The adsorption method followed by the pyrolysis process and the pyrolysis method followed by the adsorption process have higher analysis results, namely with values of 0.6758 cP method (II) and 1.0190 cP method (III) respectively. High viscosity indicates that the base fuel is slightly thicker than the method (I), high viscosity can indicate that there are some impurities, especially metals or minerals that are still dissolved in the base fuel.

In terms of determining the quality of petroleum products, other important parameters are API Degree, Specific gravity and Density. These three parameters are interrelated with each other, but the most important and basic basis for knowing specific gravity is density, where the density results obtained from each method are method (I) which is 0.7631 g / ml, method (II) is 0.7702 g / ml. Based on this density we can determine the specific gravity as shown in the image below.

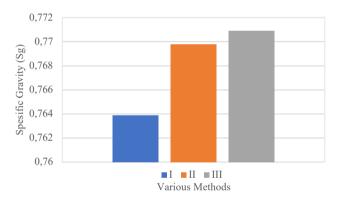


Figure 4. Comparison Specific Gravity of different methods

From Figure 4. The same thing is also found with the specific value of gravity that is closest to the value of gasoline is the pyrolysis method which is carried out integrated with an adsorption process, which is 0.7639. While the adsorption method followed by the pyrolysis process and the pyrolysis method followed by the adsorption process.

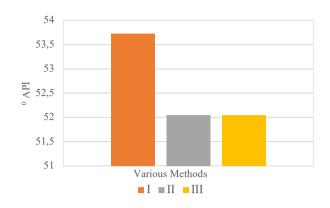


Figure 4. Comparison 0API of different methods

Based on Figure 2-5 above, it shows that the integrated pyrolysis-adsorption method, namely the process of mixing activated carbon with used oil base fuel that is pyrolyzed simultaneously using a distillation device. Pyrolysis can play a very good role in purifying used oil to be used as an alternative liquid fuel with low impurities, found API gravity or the best Fire Degree at method (I), which is 53.50 API, the value is higher than the other two methods and falls within the ASTM D 1250 standard.

Conclusion

- 1. In this research, the best method is method (I), in the purification process, which is carried out in an integrated manner between pyrolysis and adsorption, where this method produces base fuel with a higher oil quality standard than the other two methods.
- The best parameter results in method (I) are as follows: a density of 0.7631 g. /ml, Specific Gravity 0.7639, API Degree 53.7318 and kinematic viscosity 0.5542 cP; with this purification method, a more optimal yield was also obtained, namely 78.6667%

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References

- S. Jodeh et al., "The study of mobility of Pb and Zn from New and Used Lubricant Engine Oil in Soil Using PVC Columns .," vol. 02, no. 1, pp. 22– 28, 2015.
- [2] Y.Pratomo, A.S. Sani and Azharuddin,"Rancang bangun alat pengolahan oli bekas menjadi bahan bakar cair diesel dengan perlakuan panas." Agustus 2020. 1(1),2723–3359 https://doi.org/10.5281/zenodo.4540
- [3] C. M. Ahire and S. M. Lawankar, "Waste Automotive oil as alternative fuel for IC Engine," Int. Res. J. Eng. Technol., vol. 4, no. 6, pp. 934–939, 2017, [Online]. Available: https://irjet.net/archives/V4/i6/IRJET-V4I6174.pdf
- [4] U. H. Hasyim, "Review: Kajian Adsorbsi Logam Dalam Pelumas Bekas Dan Prospek Pemanfaatannya Sebagai Bahan Bakar," J. Konversi, vol. 5, no. 1, p. 11, 2016, doi: 10.24853/konversi.5.1.11-16.
- [5] E. Kusdarini, A. Budianto, and D. Ghafarunnisa, "Produksi Karbon Aktif Dari Batubara Bituminus Dengan Aktivasi Tunggal H₃PO₄, Kombinasi H₃PO₄-NH₄HCO₃, Dan Termal," Reaktor, vol. 17, no. 2, pp. 74–80, 2017, doi: 10.14710/reaktor.17.2.74-80.
- [6] A. Candra, T. Sulastry, M. Anwar, "Pengaruh Konsentrasi dan Waktu Kontak pada Adsorpsi Arang Aktif Terhadap Viskositas Oli (Minyak Pelumas) Bekas, Kimia, J., Universitas Negeri Makassar, F., & Dg Tata Parang Tambung, J. (2016).
- [7] Alwathan and Hasan, Mengatasi sulfur pada limbah oli mesin menggunakan batu bara. CV. Literasi Nusantara Abadi, 2022. [Online]. Available: https://penerbitlitnus.co.id/portfolio/mengatasi-sulfur-padalimbah-oli-mesin
- [8] M. Mardyaningsih and A. Leki, "Analisis Base Oil Hasil Proses Adsorpsi dan Pyrolisis pada Oli Mesin Bekas," J. Tek. Mesin, vol. 1, no. 1, p. 3, 2018, https://doaj.org/article/a0fb320f10604ba582a989dcdc4c8457
- [9] Y. Patmawati and Alwathan, "Penggunaan Aktivator H3PO4 dan Kombinasi H₃PO₄- NaHCO₃ Pada Aktivasi Low-rank Coal Kalimantan Timur," Indones. J. Fundam. Sci., vol. 5, no. 1, pp. 26–32, 2019.10] R. Thahir, A. Altway, S. R. Juliastuti, and Susianto, "Production of liquid fuel from plastic waste using integrated Pyrolysis method with refinery distillation bubble cap plate column," Energy Reports, vol. 5, pp. 70–77, 2019, doi: 10.1016/j.egyr.2018.11.004.