Reduction of Total Sulfur Content in BiosolarTM with Catalytic Oxidative Desulfurization Method Using Hydrogen Peroxide as Oxidizing Agent and Acetic Acid as a Catalyst

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Abstract: Many researchers are currently developing the best method to reduce the total sulfur content in fuels, such as the Catalytic Oxidative Desulfurization (Cat-ODS) method. The Cat-ODS oxidizes sulfur compounds in diesel oil to sulfur oxides, which have more polar and reactive characteristics. In this study, the Cat-ODS method conducted by oxidation process followed by liquid-liquid extraction using H_2O_2 (oxidizer), CH_3COOH (catalyst), and CH_3OH (solvent). FTIR is used to analyze the total sulfur content. The condition with oxidation temperature of 50°C, extraction stirring time of 40 minutes, and solvent/Biosolar volume ratio of 1:4 provides the highest desulfurization rate, 26.1%.

Keywords: Cat-ODS; sulfur; desulfurization; liquid-liquid extraction

1. Introduction

The increasing number of vehicles in Indonesia impacts the emergence of global climate change which require engineers to think ahead to improve the quality of the engine or the fuel itself[16)]. Moreover, the production and combustion process of fuel in vehicle's engine contributes to the environmental damage[11, 23)]. The diesel fuel commercially sold in Indonesia has been mandated to be blended with biodiesel up to 30%-volume since 2020[12)]. Biodiesel is an environmentally friendly, non-toxic and free of harmful sulfur biofuel. However, in fact, the use of diesel fuel with a mixture of biodiesel in Indonesia still causes serious problems which affects engine performance[23)].

Before the EPA (United States Environmental Protection Agency) began to regulate sulfur in diesel, diesel contained 5,000 parts per million (ppm) of sulfur. The EPA began to regulate the sulfur content of diesel fuel in 1993. Starting in 2006, the EPA began implementing more stringent regulations to reduce the amount of sulfur in diesel fuel to 15 ppm. This fuel is known as ultra-low sulfur diesel (ULSD). Ultralow sulfur diesel fuel, with sulfur less than 15 parts per million (ppm), enables the required DPF (Diesel Particular Filter) and SCR (Selective Catalytic Reduction) systems to meet the extremely low emission limits of Euro VI standards.

Every step towards lower emission limits has been adjusted and made possible by the lower sulfur content of the fuel. For example, 50 ppm sulfur diesel enables the required DOC (Diesel Oxidation Catalyst) for PM (Particulate Matter) and HC (Hydrocarbon) control and a vanadium-based SCR system for NOX control meets Euro IV emission limits (EPA Diesel Fuel Standards and Rulemaking., 2019). The content of diesel in Indonesia also varies; Biodiesel (the lowest variant) is subsidized fuel which has a sulfur content of 2,500 ppm, Dexlite has a sulfur content of 1,200 ppm, and the most expensive Dex variant has a sulfur content of 300 ppm. (SNI 7182:2015)

High sulfur content in diesel will cause engine components to be damaged more quickly and can cause corrosion in the combustion system (fuel line, nozzle, and combustion chamber) which results in the engine being inefficient and causing high maintenance costs. In terms of pollution, the residual gas from engine combustion when the sulfur content is high, the formation of sulfur dioxide (SO_2) gas will also increase. SO_2 is harmful in high concentrations, long-term exposure at low concentrations is also problematic. A systematic review concluded that sulfur dioxide exposure is associated with increased mortality from cardiorespiratory disease[15)].

Petroleum distillates, including gasoline, jet and diesel, are mixtures of hydrocarbons. These mixtures are classified according to the boiling ranges during which different extracts are obtained. In general, light distillates contain mercaptans, sulfides, and disulfides while heavier distillates contain more heterocyclic aromatic sulfur species. The middle distillate contains mostly benzothiophene and alkylated derivatives, while the main contaminants of the diesel stream are dibenzothiophene and dibenzothiophene derivatives. Diesel fuel is another commonly used transportation fuel but is used more often for heavy trucks and buses. Diesel fuel is the middle distillate stream in the refinery, with a boiling point range of 160–380°C . Diesel fuel ($C_{10}H_{22}$ - $C_{15}H_{32}$) consists of 75% aliphatic hydrocarbons [paraffins (alkanes) and naphthalene (cycloalkanes)] and a low percentage of aromatic hydrocarbons [alkylbenzenes and olefins (styrene)][5)]. Benzothiophene (BT) and dibenzothiophene (DBT) derivatives are the most common compounds found in diesel[19)].

This ODS method shows that the sulfur content in heavy oil can be reduced from 2.75 wt% to 1.14 wt%. The ODS (Oxidative Desulfurization) process through the H_2O_2-CH_3COOH organic acid system is a mild reaction condition system, where the peroxide-organic acid mixture has a strong oxidizing ability to achieve high sulfur removal efficiency[6)].

From all of many available desulfurization methods, the ODS method is able to fulfill the shortcomings of the hydro-desulfurization (HDS) method without reducing the quality of the desulfurization results carried out. The main advantage of the ODS method is that it can be operated in the liquid phase and at standard operating conditions (temperature and pressure) without the need for hydrogen and special tools. Thus, the energy required is also minimal and can minimize capital costs and process operations. Then, the results of the desulfurization process using the ODS method can also remove complex sulfur compounds, such as benzothiophene (BT), dibenzothiophene (DBT), and their alkylated derivatives, which are difficult to remove by the hydro-desulfurization (HDS) method[6,18, 25)].

This study carried out a desulfurization process using the Cat-ODS (Catalytic Oxidative Desulfurization) method that uses an oxidizing agent to oxidize sulfur compounds in diesel fuel (benzothiophene) into more polar sulfones so that the oxidation products can be easily removed by the next process to be carried out by extraction[10)].

The oxidizing agent used in this research is H_2O_2 with acetic acid as a catalyst and methanol will be used as a solvent because of its polarity[10)]. Several criteria can be used as a selection guide to choose a solvent for the liquid-liquid extraction process. It should also be noted that all the desired properties cannot be met by one type of solvent. Therefore, a midpoint should be made based on the crucial factors that affect the performance of the extraction process. The main factors in the choice of solvent to be considered are selectivity, percentage recovery, density, surface tension, solubility with the solution to be extracted, corrosive properties, viscosity, vapor pressure, boiling point, freezing point, toxicity and cost of solvent. Selectivity is the first property to be examined for the application of solvents in the separation process[4,18,21)].

Acetic acid is known to provide an acidic medium for the Cat-ODS process and stabilize hydrogen peroxide[24)]. Furthermore, the oxidation product of the Cat-ODS process is sulfone which is polar and then extracted by liquid-liquid extraction.

Previous researcher conducted the investigation of oxidation temperature and the solvent/diesel oil volume ratio for the extraction process[10)]. In this study, an increase in the oxidation temperature from 30°C to 70°C led to a remarkable increase in the reaction rate and the extraction efficiency is reported to increase with increasing volume of solvent[10)]. The extraction time must reach equilibrium for the best results[2)].

In the Cat-ODS research on diesel oil available in Indonesia, especially Biosolar, the variables to be varied are the operating conditions of the Cat-ODS in the form of a temperature, solvent/Biosolar ratio, and solvent-in-Biosolar stirring time. Farshi, Amir, and Payam Shiralizadeh have optimized the Cat-ODS process, but the oil used is HFO (heavy fuel oil)[8)]. Therefore, in this research, optimization was carried out to determine the best operating conditions of the Cat-ODS biodiesel process with acetic acid catalyst and methanol as solvent and to determine the sulfur content, researchers used ASTM-FTIR absorbance correlation. The results of this study are expected to be a reference for the Cat-ODS process to improve the quality of diesel fuel in Indonesia by reducing sulfur levels and can be applied as an effort to reduce pollutant substances while maintaining environmental quality.

2. Experimental Procedure

A common process of oxidation in Cat-ODS involves three substances, which are high sulfur content oil, oxidant, and catalyst. In this study, the high sulfur content oil is Biosolar, a commercial diesel product with highest sulfur content in Indonesia. The Biosolar used in this study contains 378 ppm sulfur. The oxidizing agent that will convert the sulfur compound to be more polar in this method varies. But hydrogen peroxide solution 30%-w was chosen as the oxidizing agent used for this experiment. Many studies have conducted experiments with various types of oxidizing agents, such as NO_2[20)], nitric acid[20)], air[17)], organic hydroperoxides[15)], potassium ferrate[15)], and hydrogen peroxide[13)]. With the advantages and disadvantages of each oxidizing agent, hydrogen peroxide is believed to be the superior oxidizing agent because it is considered the most beneficial. Starting from a meager price, it does not cause pollution, is environmentally friendly, not too corrosive, and its availability is easy to find[18)]. Catalyst used was glacial acetic acid. Oxidation process converts sulfur compounds to their sulfone forms and polarize them. The remaining sulfone compounds were extracted through liquid-liquid extraction using analytical grade methanol as a polar solvent. Before going through ODS process, pure Biosolar was analyzed to obtain initial sulfur content and FTIR spectrum of untreated Biosolar.

2.1 Oxidation experiments

Erlenmeyer flask was used as a reactor chamber for ODS process. Initially, 100 mL of Biosolar was inserted into Erlenmeyer flasks and labeled according to the treatment given (temperatures: 30ºC, 50ºC, or 70ºC; solvent/Biosolar volume ratio: 1:1, 1:2, or 1:4; extraction time: 10 minutes, 20 minutes, or 40 minutes). Hydrogen peroxide with a molar ratio of 3:1 (H_2O_2 : sulfur) followed by acetic acid with a molar ratio of 2:1 (CH_3COOH : sulfur) were added into the sample. The Erlenmeyer flask was then placed on a hot-plate stirrer and a magnetic bar was put inside the flask. A thermometer was installed, and the mouth of Erlenmeyer flask was covered with aluminum foil. The sample was then mixed vigorously and heated according to the label (30ºC, 50ºC, or 70ºC). Each sample was oxidized for 30 minutes.

2.2 Extraction experiments

Before going through the extraction process, every high-temperature sample (50ºC and 70ºC) was rested at room temperature until the thermometer indicates a temperature of 30ºC. Methanol was then added according to the label (1:1, 1:2, or 1:4 solvent/Biosolar volume ratio) and a mixing process was conducted at a temperature of 30ºC. Run the extraction process for 10 minutes, 20 minutes, or 40 minutes according to the label. Once the extraction process was finished, the final sample must be separated between organic and polar phases. It was settled in a separator funnel for 2 hours. Two phases were formed, where the bottom phase was the treated Biosolar.

3. Method of Analysis

The total sulfur content of untreated and treated Biosolar is determined by using Fourier Transform Infrared Spectrometer (FTIR). According to recent studies[3, 7, 9)], the IR region resulted from FTIR can identify and measure total sulfur content in diesel oil quantitative and qualitatively with 62% accuracy towards ASTM D-4294, the international standard test method to measure sulfur in petroleum products. The use of FTIR spectroscopy as a sulfur detector does not require complicated sample preparation and expensive cost. The spectroscopy used in FTIR is absorption, which works based on differences in the absorption of IR radiation by molecules. The peak of 1458 cm^{-1} wavelength recorded on the IR spectrum shows the presence of aromatic range[1)], which shows the characteristic of Biosolar since 70% of Biosolar contains diesel oil that formed of aromatic range. Meanwhile, the sulfur content of Biosolar shows strong absorption at 1169 cm^{-1}. The absorption of 1458 cm^{-1} and 1169 cm^{-1} resulted on IR spectrum is defined as W_{1458} and W_{1169}. To get the total sulfur content in ppm using W_{1169} and W_{1458} , the calculation is performed with the following equation:

$$Total\ Sulfur\ Content\ (ppm) = \frac{\left(\frac{W_{1169}}{W_{1458,}}\right)+0.0499}{0.0018} \qquad (1)$$

Thus, both the total sulfur content of untreated Biosolar (TSC_U) and treated Biosolar (TSC_T) in ppm are gained. Furthermore, the desulfurization percentage can be calculated with:

$$Desulfurization\ (\%) = \frac{TSC_U - TSC_T}{TSC_U} \times 100\% \qquad (2)$$

4. Results and Discussion

Overall data for each sample resulted from the data processing carried out can be seen in Table 1. It was shown that given the Cat-ODS treatment and liquid-liquid extraction can reduce the total sulfur content in Biosolar from 378 ppm to the lower content. As shown in Table 1, all samples in this study were varied with three independent variables: oxidation temperature, extraction stirring time, and solvent/Biosolar volume ratio.

Table 1. Overall Result of the Research with The Variation of Oxidation Temperature, Pre-extraction Stirring Time, and Solvent/Biosolar Volume Ratio.

Sample Name	**Oxidation Temperature (ºC)**	**Extraction Stirring Time (minute)**	**Solvent/Biosolar Volume Ratio**	**Desulfurization (%)**
1	30	10	1:1	11.4
2	30	10	1:2	4.0
3	30	10	1:4	6.0
4	30	20	1:1	8.0
5	30	20	1:2	10.5
6	30	20	1:4	12.2
7	30	40	1:1	4.5
8	30	40	1:2	12.4
9	30	40	1:4	7.1
10	50	10	1:1	23.0
11	50	10	1:2	13.0
12	50	10	1:4	8.5
13	50	20	1:1	11.4

Sample Name	Oxidation Temperature (°C)	Extraction Stirring Time (minute)	Solvent/Biosolar Volume Ratio	Desulfurization (%)
14	50	20	1:2	18.7
15	50	20	1:4	21.4
16	50	40	1:1	17.3
17	50	40	1:2	12.2
18	50	40	1:4	26.1
19	70	10	1:1	1.8
20	70	10	1:2	16.9
21	70	10	1:4	4.2
22	70	20	1:1	4.4
23	70	20	1:2	23.4
24	70	20	1:4	7.4
25	70	40	1:1	10.6
26	70	40	1:2	7.1
27	70	40	1:4	11.9

4.1 Effect of Oxidation Temperature

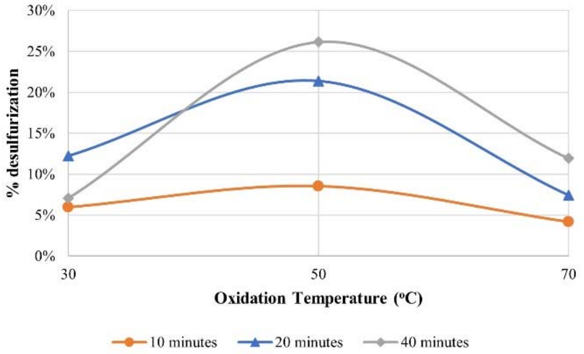

Fig. 1: Effect of Oxidation Temperature on Desulfurization at solvent/Biosolar ratio of 1:4(v/v)

To visualize the characteristic effect of oxidation temperature on desulfurization rate, results were plotted for the respective extraction times. In Figure 1, it can be seen that the desulfurization rate increased from 30°C to 50°C, but gradually decreased until 70°C. Has been obtained 8.5%, 16.8%, and 9.8% average desulfurization for oxidation temperatures of 30°C, 50°C, and 70°C, respectively, by calculating the average result for each oxidation temperature, which confirmed that 50°C showed better performance of desulfurization relative to 30°C and 70°C. Another study stated that oxidation at higher temperature was unfavorable due to the decomposition of hydrogen peroxide to undesirable side products other than hydroxyl radicals which decreased the efficiency of desulfurization process and affects the quality of diesel fuel[10]. That is why we see a downrate of desulfurization performance from oxidation temperature of 50°C to 70°C. Besides that, reaction at temperature higher than 70°C may lead to the oxidation of useful components in the fuel.

4.2 Effect of Extraction Stirring Time

The desulfurization can be between 1.8% to 26.1%. The largest desulfurization was 26.1% which was obtained in sample 18 with a temperature of 50°C, a solvent/Biosolar volume ratio of 1:4, and an extraction time of 40 minutes. To visualize the characteristic effect of extraction time on desulfurization, the effect of extraction time can be plotted for the respective temperature and solvent volume ratios (see Figure 2).

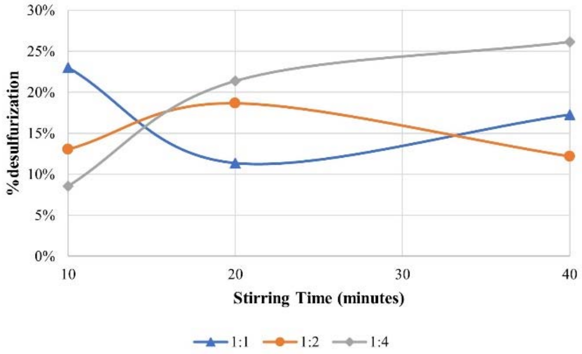

Fig. 2: Effect of Extraction Time on Desulfurization; at 50°C

It can be seen in Figure 2 in general, the best desulfurization rate is at 40 minutes. This pattern is consistent with 4 of 9 sample classes. In this study, the best desulfurization rate was achieved at 26.1% with the extraction time of 40 minutes, temperature of 50°C, and solvent/Biosolar volume ratio of 1:4. Abro et al. (2016) reported that extraction time must reach equilibrium for the best result, if it is longer than the equilibrium time then there is no significant increase in desulfurization performance. As shown in this research, the time needed to reach equilibrium in sulfone extraction at 50°C was 40 minutes.

4.3 Effect of Solvent/Biosolar™ Volume Ratio

The solvent/Biosolar volume ratio is an essential parameter in the extraction of oxidized sulfur from the Cat-ODS process. The graph in Figure 4(a) shows the effect of the solvent/Biosolar volume ratio on the desulfurization produced at the same oxidation reaction temperature, 70°C. As can be observed from Figure 4(a), the behavior of solvent/Biosolar volume ratio is uncertain and affected by other variables: oxidation temperature and extraction stirring time. However, for the oxidation temperature at 70°C, the solvent volume ratio of 1:1 and 1:4 shows the same behavior. The 10 and 20 minutes extraction stirring time has the same pattern, starting from 1:1, and the desulfurization increases at solvent/Biosolar volume ratio of 1:2. Nevertheless, the desulfurization became lower when the solvent/Biosolar is decreasing to 1:4. Except for extraction stirring time of 40 minutes, the desulfurization reached the lowest point for solvent/Biosolar volume ratio of 1:2 and increases due to decreasing the solvent/Biosolar volume ratio.

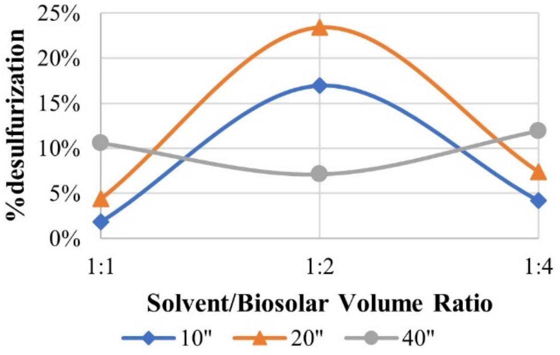

Fig. 3: Effect of Solvent/Biosolar Volume on Desulfurization with Oxidation Temperature of 70ºC

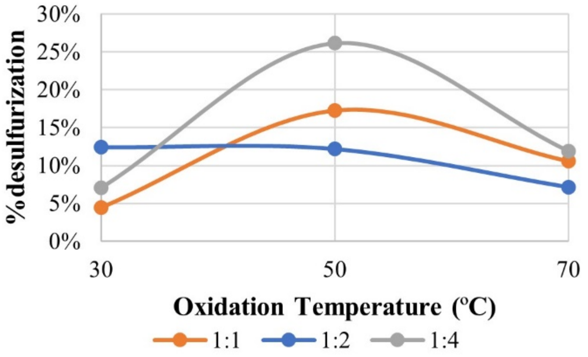

Fig. 4: Effect of Solvent/Biosolar Volume on Desulfurization with Extraction Stirring Time of 40 Minutes

As shown in Figure 4(b), the desulfurization resulted from solvent/Biosolar volume ratio of 1:4 that carried out at an oxidation temperature of 50°C and an extraction stirring time of 40 minutes is 26.1%. The solvent/Biosolar volume ratio of 1:2 gave a desulfurization of 23%. The result is not much different from the results of 1:4. However, the desulfurization value produced by the solvent/Biosolar volume ratio of 1:1 and 1:2 with the same oxidation reaction temperature and stirring time (samples 16 and 17) is only 17.3% and 12.2%. These numbers are quite small compared to sample 18, with a solvent/Biosolar volume ratio of 1:4. It is expected that the increase in the solvent/Biosolar volume ratio will result in a high desulfurization value as the volume of the solvent increases which can extract more sulfur from diesel oil. The pattern shown by oxidation temperature of 70°C in Figure 4(b) is the same as 50°C. The solvent/Biosolar ratio of 1:4 has the highest desulfurization at high temperature, whereas the 1:1 solvent/Biosolar has the lowest desulfurization at room temperature (30°C).

The resulting data supports the research conducted by Sobati, Dehkordi, and Shahrokhi in 2010. The desulfurization is affected by the solvent/Biosolar volume ratio. The increasing of the solvent volume also reduces the recovery value of the Biosolar itself. In other words, increasing the solvent/Biosolar ratio cannot be expected to increase the desulfurization. The increasing ratio of the volume ratio between the solvent and Biosolar is also followed by a continuous increase in Biosolar dissolved in the extraction solvent. So, besides losing sulfur at the end of the extraction, there has been a yield loss due to the decrease amount of Biosolar.

In summary, the highest desulfurization occurred in sample 18, with desulfurization up to 26.1%, from 378 ppm to 279 ppm. The operating conditions in sample 18 were able to reduce up to 99 ppm, which made the quality of Biosolar much better. Sample 18 used an oxidation reaction temperature of 50°C, an extraction stirring time of 40 minutes, and a solvent/Biosolar volume ratio of 1:4.

5. Conclusion

The research can be concluded as below:

- The desulfurization rate increased from oxidation temperature of 30°C to 50°C, but gradually decreased until 70°C.
- The extraction time in the Cat-ODS extraction process affected the reduction in sulfur content, which was indicated by various desulfurization values. Extraction time of 40 minutes was the best extraction time in 4 of 9 sample classes.
- The solvent/Biosolar volume ratio affected the result of desulfurization of Cat-ODS and liquid-liquid extraction. The increasing of the solvent/Biosolar ratio cannot be expected to increase the desulfurization. It takes the right amount of solvent/Biosolar to get the highest desulfurization.
- The best desulfurization was 26.1% at an oxidation temperature of 50°C, solvent/Biosolar volume ratio of 1:4, and extraction time at 40 minutes.

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Nomenclature

W_{1169}	Absorption at wavelength 1169 cm^{-1}
W_{1458}	Absorption at wavelength 1458 cm^{-1}
TSC_U	Sulfur Content of Untreated Biosolar (ppm)
TSC_T	Sulfur Content of Treated Biosolar (ppm)

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