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Lead Adsorption in Lubricant Waste using Zeolite

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ABSTRACT

Lubricant waste is one of the hazardous refuses which are regulated on the limit of lead content according to Government Regulations (Kep-51/MenLH/10/1995). Therefore, it is necessary to research for reducing the lead content. The purpose of this study is to understand the effect of adding adsorbents to decrease lead content in waste of lubricants taken from ships. The waste lubricant was recycled by adsorption using zeolite. Lubricant waste samples of 200 mL each were physically and chemically identified subjected to adsorption process using zeolite adsorbent whose concentrations (%w/w) were varied as follows: 7.5%, 13.25%, 14.25%, 15.75% and 17.5% with stirring speed of 150 rpm and contact time for 60 minutes. The best results were obtained at the adsorbent amount of 26.5 grams (concentration of 13.25%), for which lead content reduction reached 83%. The ANOVA F was obtained to be 13.42, and hence the study concluded that the amount of the adsorbent was related to the decrease in lead content.

Keywords: adsorption; lead; lubricant waste; zeolite

ABSTRAK

Pelumas bekas adalah salah satu limbah berbahaya yang dibatasi kadar timbalnya dalam Peraturan Pemerintah, sehingga perlu penelitian untuk menurunkan kadar timbal. Tujuan penelitian ini adalah untuk memahami pengaruh penambahan adsorben untuk menurunkan kadar timbal dalam pelumas bekas yang berasal dari mesin kapal. Pelumas bekas didaur ulang dengan adsorpsi menggunakan zeolit. Setiap sampel berisi 200 mL pelumas bekas yang telah diidentifikasi sifat fisika dan kimia-nya diadsorpsi menggunakan adsorben zeolit. Variasi konsentrasi zeolit (%w/w) yang digunakan yaitu: 7,5%; 13,25%; 14,25%; 15,75% dan 17,5% dengan kecepatan pengadukan 150 rpm dan waktu kontak 60 menit. Hasil terbaik diperoleh pada jumlah adsorben 26,5 g (konsentrasi 13,25%) dengan penurunan kadar timbal mencapai 83%. Faktor F pada hasil ANOVA adalah 13,42. Dengan demikian dapat

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disimpulkan bahwa terdapat korelasi antara konsentrasi adsorben dan penurunan kadar timbal.

Kata kunci: *adsorpsi; pelumas bekas; timbal; zeolit*

1. Introduction

Environmental pollution is one of the most significant issues in public health and safety that we are currently facing. Pollutants come from various sources and media; one of them is categorized as hazardous waste. One of the dangerous pollutants is lead (Pb). It is a heavy metal that can be dangerous to the environment and toxic to human health. Annually, Palembang seaport in Indonesia collects 28,680.21 m^3 waste of lubricants containing 18 ppm Pb (Ramadhaniar, 2019), accumulating around 0.52 m^3 lead per year. This Pb content in industrial waste exceeds the maximum limit set by the government of 1 mg/L effluent (Kep-51/MenLH/10/1995). In the event of lead absorption in the body, the accumulation of this metal can lead to the disruptions of many bodily functions and illnesses (Irianti, 2017). Lead accumulation in the body can cause kidney failures, neuronal disturbances, disruption of red-blood-cell formation, childhood developmental challenges, and blindness (Cope, 2004).

Various efforts have been taken by the government to prevent environmental pollutions that have harmful effects on human health by issuing regulations to curb pollutions. One of these is the Ministry of Environment Regulation number 5/2009. In the Article 7 Point 1 of this regulation, the government requires all general and special maritime ports to have treatment facilities for wastes originating from the businesses and activities of water vessels. Every naval port in Indonesia, therefore, must have waste treatment facilities for domestic and industrial wastes called Recipient Facilities (RF). This regulation is following that of the International Maritime Organization (IMO).

One of these maritime wastes is lubricant oil waste from ships. These waste of lubricant oil must be treated not only to prevent pollution but also for reuses. Studies have been undertaken to achieve these goals, and some of them employed clay-type adsorbents to reduce the heavy metal contents and recover lubricant oil (Pratiwi, 2013). A study also conducted by Kemala et al. (2019) for the adsorption of heavy metals in lubricant oil waste using activated acid clay and calcination.

Adsorption is a process involving adhesions of atoms, ions, or molecules of adsorbates to the surface of adsorbents (Fatimah, 2014). The adsorbates can be in the form of gases, liquids, or dissolved solids. One of the adsorbents commonly used for this process is zeolite, which is an aluminosilicate mineral with a tetrahedral structure connected with oxygen bonds. With the tendencies of Al ions positively charged and Si ions negatively charged, there are cations in the pores that stabilize the structure. These cations are impurities to be replaced during the ion-exchange processes (Trisunaryanti, 2016). Zeolite was selected for this study because the ratio of silicate and alumina is 68.26 to 12.69 to ensure complete adsorption of Pb by the silicate compounds (Yunus, 2018). Zeolite also gives optimum results compared to other types of adsorbent (Murachman et al., 2014).

Several factors influence the adsorption process, e.g. (i) specific surface area of adsorbent, (ii) stirring speed during adsorption process, and (iii) solubility of adsorbate in the solution (Asip et al., 2008). Furthermore, according to Le Chatelier's principle, the adsorption process is exothermic. Thus, to increase the number of adsorbates being adsorbed, the temperature must be lowered, and the pressure must be increased (Ginting, 2008).

This study is intended to understand the effect of increasing adsorbent concentrations to reduce lead content in lubricant waste collected from ship engines. The increase in adsorbent concentration will provide an increase in surface area to improve the adsorption process.

2. Research Methodology

2.1 Materials

The research materials consisted of lubricant oil waste samples (from the IPC group, see Table 1 for composition), natural zeolite from Lampung (for composition based on Arita et al. (2015), see Table 2), hydrochloric acid (HCl) (37%, Merck) and distilled water. The zeolite has a surface area of 22.38 m^3/g and pore volume of 5.35 cc/g, as characterized using Quantachrome Surface Area Analyzer (SAA) employing Multi-Point BET method. Lampung natural zeolite was chosen as it is well known as good Pb adsorbent with a firm structure suitable for heavy metal adsorption (Ginting et al., 2017).

Table 1. Chemical composition of lubricant waste

Chemical Content	Unit	Concentration
Total Base Number	mg KOH/g	10.24
Fuel	% vol	5.90
Oxidant	A/0.1 mm	0.07
Soot	A/0.1 mm	0.00
Sulfation	A/0.1 mm	0.03
Water	% vol	0.00
B	ppm	0
Na	ppm	1
Si	ppm	4
V	ppm	-
Ag	ppm	0
Al	ppm	3
Cr	ppm	1
Cu	ppm	4
Fe	ppm	18
Mo	ppm	0
Ni	ppm	0
Pb	ppm	18
Sn	ppm	0

Table 2. Chemical composition of zeolite

Chemical content	Concentration (%wt.)
SiO_2	68.26
Al_2O_3	12.99
Fe_2O_3	1.37
CaO	2.09
MgO	0.83
Na_2O	0.64
K_2O	4.11
MnO	0.06
TiO_2	0.23
H_2O	-

2.2 Procedures

The zeolite was firstly activated using acid to optimize the adsorption condition of Pb from lubricant waste. The mixing was set at 150 rpm and a contact time of 60 minutes with samples in the amount of 200 mL per data point. The lubricant oil waste samples had been previously analyzed for physical properties and chemical contents before the experiments. The average initial Pb content of lubricant oil waste was 6 ppm. The independent variable the concentrations of zeolite comprised 7.5, 13.25, 14.25, 15.75, and 17.5 (%w/w).

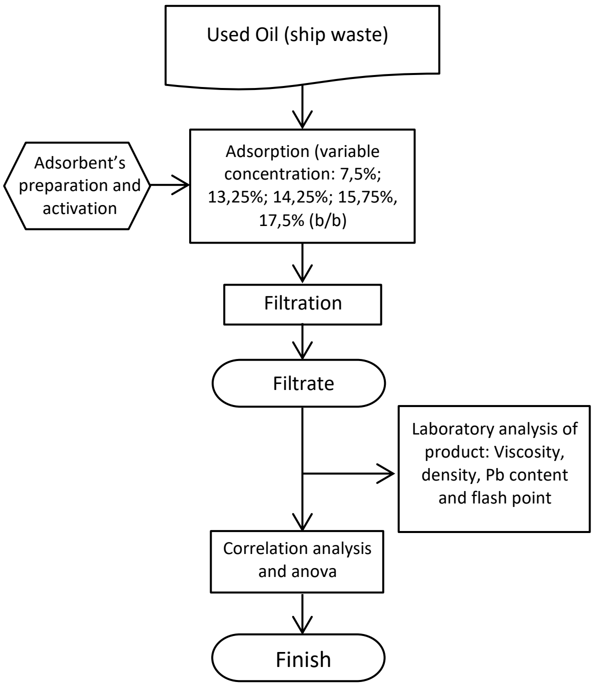


Figure 1. Experimental procedure

The first step involved the activation of zeolite with 3M HCl for 30 minutes, followed by neutralization and drying at 140–150 °C for two hours. The Pb adsorption process started with the addition of 2M HCl into 200 mL of samples of lubricant oil waste. The mixture was then stirred for fifteen minutes. Mixture samples in the amount of 150 mL were blended with zeolites at the previously determined concentrations and stirred for 60 minutes at room temperature using a jar test. The next steps included separation and filtration using Buhner funnels and vacuum pump, resulting in the final samples to be analyzed for Pb-content. This experimental procedure is summarized in Figure 1.

2.3 Instrument and data analysis

Inductively-Coupled Plasma (ICP) instrument (Petrolab Services) was used to measure Pb-content, according to ASTM D5185-13e1. Scanning Electron Microscope (SEM) instrument (PPF-LIPI) emitting electrons at 4.515 keV was employed to confirm the adsorption of Pb on the surface of zeolite adsorbent. Research data were analyzed using ANOVA (analysis of variance).

3. Results and Discussion

The results from the adsorption tests to reduce the Pb-content in lubricant oil waste using various amounts of zeolite are presented and plotted in Figure 2.

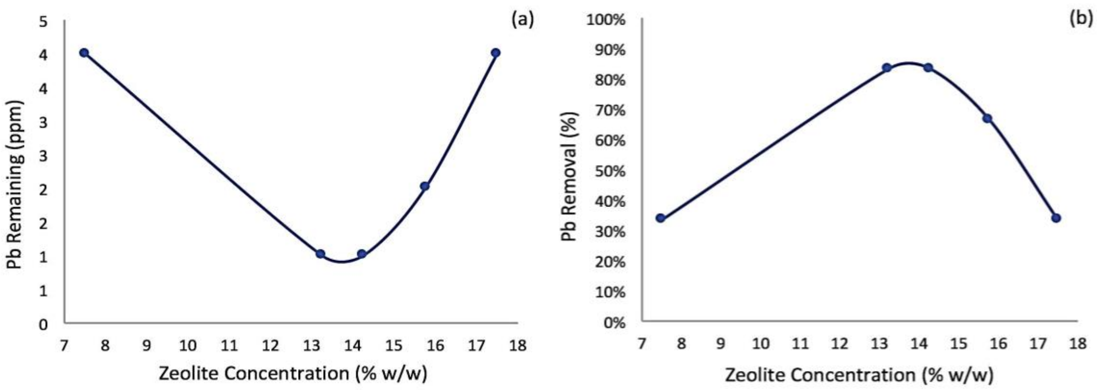
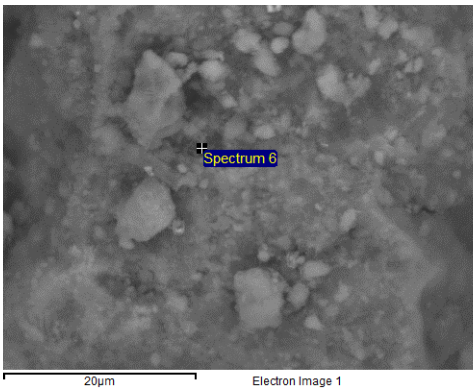


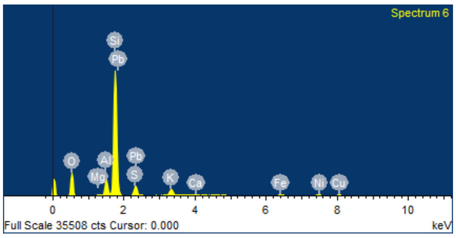
Figure 2. Results of Pb-content adsorption tests using various concentrations of zeolite adsorbent at mixing speed of 150 rpm and contact time of 60 minutes: (a) Concentration of remaining Pb (ppm); and (b) Percentage of Pb removal (%)



(a)

Element	Weight%	Atomic%
C	7.66	13.43
O	36.18	47.61
Mg	0.14	0.12
Al	4.15	3.24
Si	38.67	28.99
S	4.64	3.05
K	3.11	1.68
Ca	0.39	0.20
Fe	0.83	0.31
Ni	1.78	0.64
Cu	2.10	0.70
Pb	0.34	0.03
Totals	100.00	100.00

(b)



(c)

Figure 3. (a) SEM images of Pb-adsorbing zeolite; (b and c) elemental analysis result

The results displayed in Figure 2 show that Pb-contents keep decreasing with an increased amount of zeolite up to 26.5 gram (13.25%), where there was an 83% reduction of Pb-content. Further increased zeolite content, however, did not result in a further decline. This phenomenon can be explained by the possibility that adsorption reaches an equilibrium at 26.5 grams of zeolite addition. In contrast, the further increase in the volume fraction of adsorbent might interfere with the Pb adsorption process. There is still a need, however, to determine the adsorbent amount where Pb adsorption is optimum. The current result shows a more significant reduction than that of a previous study (Pratiwi, 2013), which employed acid clay adsorbents and yielded a maximum decrease of 56%.

Further analyses of correlation and Analysis of Variance (ANOVA) were conducted on the adsorbent concentrations and resulting Pb-contents. ANOVA was performed for determining the effect of adsorbent concentration on the results of Pb-content. F-table for df (1;10) at a confidence level of 0.95 is 4.92. The ANOVA yielded an F-value of 13.42, which indicates that this study supports the hypothesis, i.e., there is a significant correlation between the amount of zeolite and reduction of Pb-content.

The adsorption of Pb on the surface of zeolite adsorbent is also confirmed by the results of SEM characterization (Figure 3). The adsorption of Pb was evident, especially at spectrum 6, showing the concentration of adsorbed Pb at 0.34 %wt.

4. Conclusions

This study successfully performed a reduction of Pb-content in lubricant oil waste by adsorption using zeolite adsorbent. With an initial Pb-content of 6 ppm, the additions of zeolite were able to reduce the Pb-content to 1 ppm at the adsorbent amount of 26.5 grams, the contact time was 60 minutes and mixing speed was 150 rpm. The calculated F-value is 13.42, which is higher than 4.92 obtained from the F-table for df (1;10) at a confidence level of 0.95. This result indicates that the regression is significant, and the null hypothesis (H_0) of no correlation between adsorbent additions and Pb-content reduction is rejected. In conclusion, this study supports the alternative hypothesis (H_1) of a significant relationship between the zeolite addition amounts and Pb-content reduction.

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