

Solid Concentration Effect for Solvent Extraction Process of Oily Contaminated Soil

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Abstract

Solvent extraction has been used as a method to wash oil content of oily contaminated soil in industry for years. Some solvents and temperature ranges has been chosen to increase the oil recovery rate of extraction process, however only few studies reported that it has been able to reach remaining Total Petroleum Hydrocarbon (TPH) less than 0.5% in less than 30 minutes. During the experiments, acetone and toluene chosen to extract oil content from contaminated soil by using solvent extraction process. Temperature selected were between 24°C up to 70°C. Mixing apparatus which has been utilized was centrifugation machine with 1000 rpm (1570 g) operational speed. Duration of treatment process was 10 minutes with some variations of solid to solvent ratio. During the experiments, it was observed that by using toluene and acetone as solvents, the optimum Total Petroleum Hydrocarbon (TPH) removal obtained at temperature 50°C. In the other hand, optimum solid to solvent ratio toluene ratio was 1:6. As a solvent acetone observed capable to reduce TPH content until below 0.5% as threshold limit for TPH of contaminated soil regulated by environmental regulation in Indonesia. During the experiments it was also observed the dependency of solid concentration (C_s) with dissociation coefficient (K_D). In the other hand, heavy metal at the remaining extracted soil after soil washing was observed available in safe concentration to be discharged to the environment base on regulation in Indonesia.

Keywords: solvent extraction, soil washing, contaminated soil, TPH, centrifugation, oil sludge, acetone, toluene, solid treatment.

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INTRODUCTION

Solvent extraction process essentially is an adsorption-desorption process of oil between the solid and liquid phases (Liang et. al., 2014). Biceroglu in 1994 has been used intermediate hydrocarbon source to be able to extract oil sludge from refinery storage to become lighter hydrocarbon at several stages of

temperature. Temperature that has been selected was from 20°C to 50°C with ratio between solvent and oil sludge is 1:2 respectively and duration of extraction was 1 hour. It was reported that 23-32% hydrocarbon content has been recovered. Solvent and hydrocarbon that has been separated with the solid phase will be fed to crude oil tank for further refinery treatment. Solid

phase residue then utilized as alternative energy or being landfilled.

El Naggat et. al., in 2010 studied solvent extraction process to recover oil content of oil sludge. It was observed that the usage of some solvents for such as naphthalene, kerosene cut, n-heptane, toluene and some other solvents. From several experiments, it was reported that toluene has the highest extraction efficiency, which is 75.94%. Zubaidy and Abuelnasr in 2010 made comparative study related to the effect of some organic solvents such as methyl ethyl keton (MEK) and liquefied petroleum gas condensate (LPGC). They observed that the most efficient comparison between MK and Sludge 4:1. Implementation of that comparison obtain the highest oil recovery was 39% for MEK and 32% for LPGC. It was also reported that the usage of MEK has been also able to separate most of ashes, asphaltene, and some other contaminants. Nevertheless, it was found that high sulphur content presence in the carbon residue of recovered oil. Solvent extraction basically conducted with pouring oil sludge with some solvents with certain proportion to separate water, solids, and extracted solvents. The extracted solvents would then to be sent to distillation unit for separating oil and solvent. (Al-Zahrani et.al, 2013).

Guangji Hu. Et.al, 2013 reported that solvent extraction process has been conducted by mixing solvent and oil sludge in a reactor with certain retention time so that solvent and oil sludge mixed homogeneously and then settled for certain time to make sludge residue settled in the bottom. Liang, J et. Al (2014) reported the kinetics of solvent extraction process. He reported solid effect (Cs) in solvent extraction treatment of pre-treated oily sludge can be explained by using the Surface Component Activity (SCA). Using SCA model, the surface site (fs) was proposed as a function of solid concentration (Cs) other than in unity form.

This experiment, other than having perspective in oil recovery, was more focus on the remaining solid that has been left over the solvent extraction process identified by TPH content. To examine the metal content of remaining soil after the solvent extraction process was also significant to be conducted. Referring applied environmental regulation shall give contribution for further implementation at larger scale of application.

MATERIALS AND METHODS

Oily contaminated soil has been transported from Kalimantan-Indonesia to be tested and treated in Laboratory at Bogor-Indonesia. Two organic solvents were selected for experiments including Toluene and Acetone with pro-analysis specification produced by Merck. A Simple random sampling method as recommended in USEPA SW-846 guideline was used for collecting sludge, while its TPH was determined as the target contaminant that is of concern in statistical analysis.

The sample was stored in a jar at 25°C. Aliquot of 1000 grams of contaminated soil were collected in a tray. Sample were dried by using open air for three days to minimize moisture content for further experiments. After drying the solid sample is mix evenly so to ensure the homogeneity.

The following table 1. will describe the arrangement of sample preparation to be further treated by using solvent extraction method. TPH measured utilizing USEPA 8440 method about total recoverable petroleum hydrocarbons by using infrared spectrophotometry. Analysis has been conducted through 2 grams of dry soil sample extracted by using 10 ml of tetrachloroethylene then mixed by horizontal shaker with 150 rpm velocity and 15 minutes duration. After decanted, extract passed through silica gel and glass wool column. Silica gel removes polar compounds so that the remaining would only non-polar total petroleum hydrocarbon presence in the extract. Absorbance of extract measured by using infrared (IR) spectrometer after sufficient dilution factor has been implemented. TPH content calculated based on the following equation:

$$TPH\text{concentration} = \frac{R \times D \times V}{W} \quad (1)$$

Where:

R = spectrometer read (mg/ml)

V = extract volume (ml)

D = dilution factor

W = sample weight (kg)

Metal elements for solid were measured using inductively Coupled Plasma (ICP) analysis base on USEPA 3050 B Method by conducting acid digestion method using HNO_3 and H_2O_2 .

Solvent extraction method conducted by running centrifugation SETA Oil Test Centrifuge at 1000 rpm (1570 g) and 10 minutes duration of extraction. Prior being centrifuged, sample was shaken by using horizontal shaker for two minutes. Extract of aliquot and solid was separated by using vacuum filtration method. Type of filter that has been used during the filtration process was *Whatman series* with specification CAT-1825-047 that has 47 mm of diameter. Each sample has been weighted as listed in the table 2 for obtaining several comparison of soil to solvent.

Test has been conducted to observe in which ratio soil to solvent is giving most efficient performance during the solvent extraction process for removing oil content which is calculated as Total

Table 1. List of Contaminated Soil Properties

Parameter	Value	Unit
pH	8.3	-
TPH	6.9	%
Moisture	11.6	%
Density	1.2	g/m^3

Table 2. List of Comparison soil to Solvent

Solvent	Test	Soil : Solvent
Toluene	t-1	1:3
	t-2	1:4
	t-3	1:5
	t-4	1:6
	t-5	1:7
Acetone	t-6	1:3
	t-7	1:4
	t-8	1:5
	t-9	1:6
	t-10	1:7

Petroleum Hydrocarbon (TPH) from the soil matrix. Ratio for those two solvents for better performance would also be conducted, moreover comparative study of temperature effect also has been examined. Temperature that has been chosen during the test was 24°C, 50°C and 70°C.

Further test for examining how far the TPH remaining in the soil was conducted for acetone. Test was conducted to see the significance of soil to acetone in three variance of temperature, which are 24°C, 50°C and 70°C as shown in table 3.

TPH Removal

Oil recovery efficiency (Ro) from the oily contaminated soil and the oil concentration in the solvent (Co) were calculated from:

$$Ro = \frac{(r_0 - r_{10})}{r_0} \quad (2)$$

$$Co = \frac{(r_0 - r_{10})}{m/V} \quad (3)$$

Where r_0 and r_{10} are the initial and residue oil contents that measured as Total Petroleum Hydrocarbon (TPH) in the contaminated soil (g/g), respectively, m is the mass of soil (g) and V is the solvent volume (mL). For solvent extraction equilibrium system as reported by J. Liang et.al in 2014 dissociation coefficient (K_D) can be represented as:

$$K_D = Co / r_o \quad (4)$$

As observed by Zubaidy and Abouelnasser (2010) and also followed by Liang, J et.al (2014) the fact that K_D changes with solid concentration (Cs) indicates that real extraction systems are not ideal (or thermodynamic). In the other word, it reveals that solid particle-particle interactions exist in a real system. It is including a deviation of a real system. With changing Cs, the strength of the particle-particle interactions will vary, resulting the change in K_D . It would be conducted similarly to the solution system.

The result of this experiment would be shown in Figure 6.

RESULTS AND DISCUSSIONS

For the data of each solvent the ANOVA single factor test has been conducted to see the significance variance of the data. Base on the following calculation it was found that from group of data for duplo test that has been performed, F values are $> F_{crit}$ so that the null hypothesis stating the data are equal was rejected. Each group of data also has p value below 0.05.

Figure 1. below would show the representative data of test result as mean of each duplo test for temperature 24°C operation condition. It also showed that within the same comparison between toluene and acetone at 24°C giving between 0.33 up to 0.8.

The order of Ro for selected solvents are Acetone>Toluene. In general at this temperature, acetone as a solvent giving best performance to remove TPH of contaminated soil.

The following Figure 2. Solvent extraction at 50°C shown the same indication for working temperature 24°C where acetone gave better performance. However, in general performance at this temperature increase significantly compare to performance at room temperature.

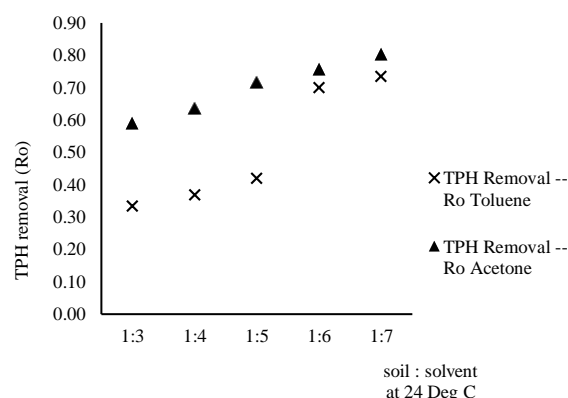


Figure 1. TPH Removal at 24°C

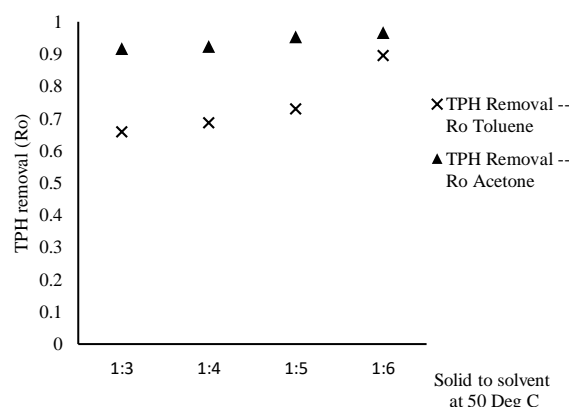


Figure 2. TPH removal at 50°C

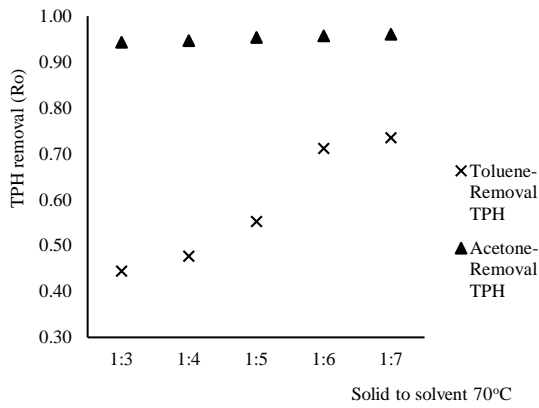


Figure 3. TPH removal at 70°C

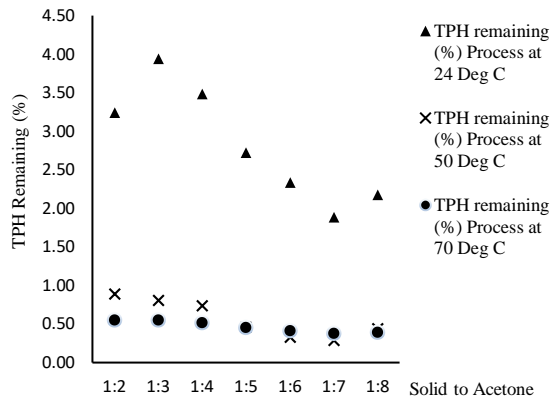


Figure 4. Optimization of Acetone Extraction

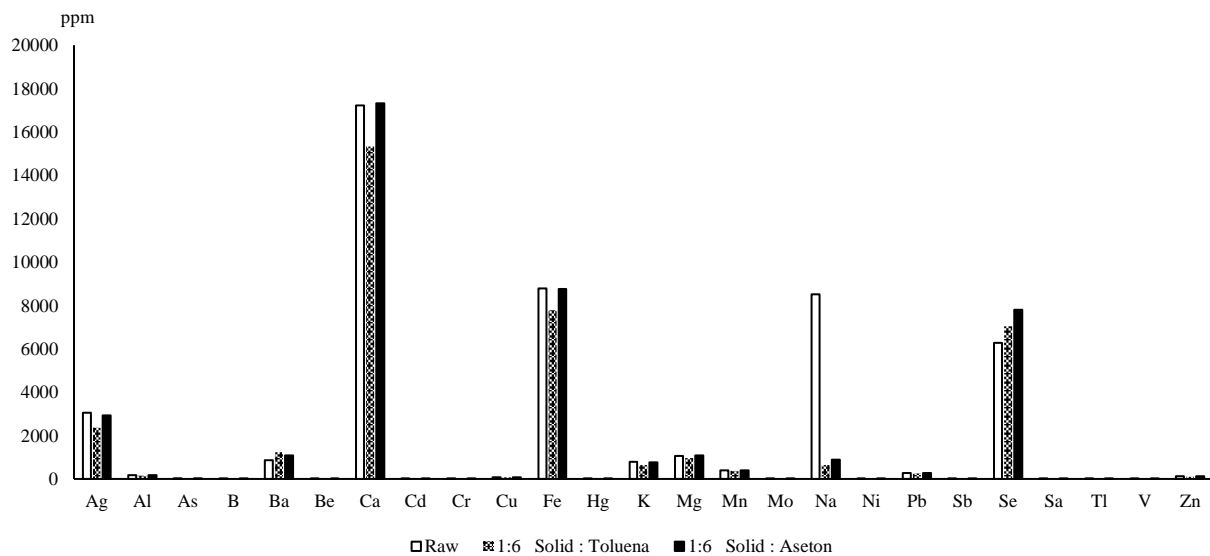


Figure 5. Metal content of remaining solid pre- and post- solvent extraction process

The following Figure shall show TPH removal between toluene and acetone at 70°C. The picture below indicates that acetone at 70°C working very well to reduce TPH content. However further study related to the evaporated acetone occurred during thermal process that exceeding its boiling point.

Seeing the following Figure 4, even though the acetone working more efficient in removing TPH 70°C, optimum working temperature by using acetone is 50°C. As boiling point of acetone is 56.3°C, it would be beneficial if working temperature would be lower than boiling point so the evaporation of solvent that may give some impacts could be avoided.

In Figure 4, it shall also be seen that the TPH content of the soil can reach under 0.5% in one stage of solvent extraction. Compare with the applied regulation that has been implemented in Indonesia, this TPH content meet the threshold limit value to be able to discharge to the environment. The other components that shall be examined are heavy metals and other toxicity parameters to ensure the soil would be safe to be discharge to the environment.

In Figure 5 below, Availability of metal concentration for remaining solid after solvent extraction process indicated that all metal concentration were not increased exceeding threshold limit value that has been regulated in Indonesia, therefore it shall be safe to be discharged to the environment base on TPH and metal content parameter.

The previous research to study solid concentration that has been conducted by Zubaidy and Abouelnasser (2010) and Liang, J. Et al. (2014) for respectively LPGC, Methyl Ethyl Keton and cyclohexane, n-butanol, kerosene, and n-butanol showing the similarity of observation result that Cs is not independent to the K_p .

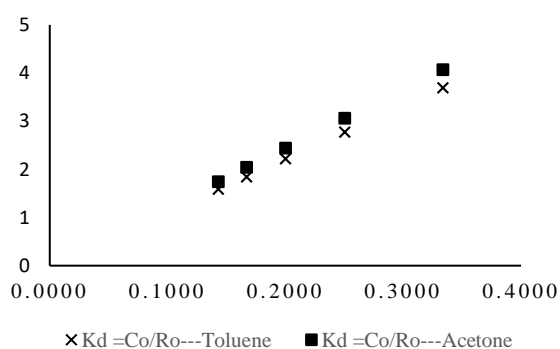


Figure 6. Cs dependency to K_D

CONCLUSIONS

The experiments that has been conducted gave information that using acetone gave higher significant result compare to toluene regarding the remaining TPH content at post-treatment contaminated soil. Treatment by using acetone may reduce the TPH content lower than 0.5 %. It was also shown that the changing of solid concentration effect gave variance linear change of K_D of the extraction system. The dependency of solid concentration also observed during the study giving confirmation for the same phenomenon that has been observed by the previous related experiments.

Optimum temperature that has been selected during the optimization process for toluene and acetone was 50°C. The operational process lower than solvent's boiling point was also a consideration to choose the optimum temperature.

This study shows that it is essential to optimize systematically so that the experiment shall give good description that solvent extraction process is a promising process that shall be considered to treat oily contaminated soil which currently considered available in large number in Indonesia. Further test that related to specific related parameter such as lethal dosage 50 and other related parameters shall be treated to verify the confidence level to discharge it directly to the environment.

REFERENCES

Al-Futaisi, A., Jamrah, A., Yaghi, B., Taha, R., (2007), *Assessment of Alternative Management techniques of Tank Bottom Petroleum Sludge in Oman*. Journal of Hazardous Material, 141, pp. 557-564.

Al-Otoom, A., Alawzi, M., Al-Omari, N., Al-Hsienat, E., (2010), *Bitumen Recovery from Jordanian Oil Sand by Froth Flotation using Petroleum Cycles Oil Cuts*, Energy, 35, pp. 4217-4225.

Al-Zahrani, S.M. and Putra, M.D., (2013), *Used lubricating oil generation by various solvent extraction technique*. J. Ind. Eng. Chem., 19, pp. 536-539.

Bicoreglu, O. (1994), *Rendering Oily waste land treatable or usable*. US Patent 5, pp. 288,391.

Elektrowicz, M dan Habibi, S., (2005), *Sustainable Waste Management: recovery of fuels from Petroleum Sludge*. Can. J Civil Engineering, 32, pp. 164-169.

El Naggar, A.Y., Saad, E.A., Kandil, A.T., Elmoher, H.O., (2010), *Petroleum cuts as solvent extractor for oil Recovery from Petroleum sludge*, Journal Petroleum and Technology Alternative Fuels.

Hu, G., Li, J., Thring, R.W., Arocena, J. (2014), *Ultrasonic oil recovery and salt removal from refinery tank bottom sludge*, Journal of Hazardous Materials.

Hu, G., Li, Ji., Zeng, G., (2013), *Recent development in the treatment of oily sludge from Petroleum Industry: A Review*, Journal of Hazardous Materials, 261, pp. 470-490.

Hu, G., Li, J., Huang, S., Li, Y. (2016), *Oil recovery from petroleum sludge through ultrasonic assisted solvent extraction*. Journal of Environmental and Health, Part A, 51, pp. 921-929.

Hu, G., Li, J., Hou, H. (2015), *A combination of solvent extraction and freeze thaw for oil recovery from petroleum refinery wastewater treatment pond sludge*. Journal of Hazardous Materials, 283, pp. 832-840.

Liang, J., Zhao, L., Du, Na., Li, H., Hou, W., (2014), *Solid effect in solvent extraction treatment of pre-treated oily sludge*. Separation and Purification Technology.

Qin, H., Ma, J., Qing, W., Liu, H., Chi, M., Bai, J., Zhang, L (2015), *Shale Oil Recovery from Oil Shale Sludge Using Solvent Extraction and Surfactant Washing*. Estonian Academy Publisher, 32, pp. 269-287.

Taiwo, E.A. dan Otolorin, J.A., (2009), *Oil Recovery from Petroleum Sludge by solvent extraction*, Petroleum and Science Technology., 27, pp. 836-844.

Trowbridge, T.D. and Holcombe, T.C. (1995), *Refinery sludge treatment/hazardous waste minimization via dehydration and solvent extraction*. Journal of the Air and Waste Management Association, 45, pp. 782-788.

USEPA 8440., (1996), revision 0.

USEPA 3050B, (1996), revision 2.

Zhang, J., Li, J.B., Thring, R.W., Hu, X., Song, X.Y., (2012), *Oil Recovery from Refinery Oily Sludge via Ultrasound and Freeze/Thaw*. Journal of Hazardous Material, 203-204, pp. 195-103.

Zubaidy, E.A.H., and Abouelnasr, D.M., (2010), *Fuel Recovery from waste oily sludge using solvent extraction*, Process Safety Environment Journal