Stabilization/Solidification of used lubricating oil containing Fe and Pb with Portland cement and bentonite

Ayu Nindyapuspa^{1*}, Taty Alfiah¹, and Virgo Indrawan Toda¹

¹Department of Environmental Engineering, Institut Teknologi Adhi Tama, Surabaya, Indonesia

Abstract. Used lubricating oil belong hazardous waste based on the Government Regulation Number 101 in 2014 about Hazardous Waste Management. Used lubricating oil contain hydrocarbon and heavy metals such as Fe and Pb. One of method to immobilize used lubricating oil is stabilization/solidification (S/S). S/S method always uses Portland cement to immobilize the heavy metals, but Portland cement can not immobilize the hydrocarbon. In this research, S/S method used Portland cement and bentonite are binding the heavy metals and hydrocarbon in the used lubricating oil. The composition used was cement bentonite 25:75, cement bentonite 75:25, cement 100%, and bentonite 100%. Used lubricating oil was added in 5%, 10%, and 15% weights. The S/S products were examined by measuring the TCLP of the Fe and Pb. The TCLP test was compared with the Fe and Pb concentration in the used lubricating oil. The results showed that the TCLP test of the Fe on the S/S product containing 5%, 10%, 15% used lubricating oil with cement 100% was 371.14 mg/L, 403.66 mg/L, and 417.89 mg/L respectively. Meanwhile, the Fe concentration in the used lubricating oil was 47.78 mg/L. The adding of bentonite to the S/S product decreased the Fe concentration that was leached. The TCLP test of the Fe in the S/S products that contained 5%, 10%, and 15% used lubricating oil with bentonite 100% were 15.24 mg/L, 0.085 mg/L, and 0.428 mg/L respectively. The S/S method using Portland cement and bentonite decreased the Pb concentration leached into the environment.

1 Introduction

Lubricating oils consist of complex mixtures of hydrocarbon molecules, mostly isoalkanes. Lubricating oil is used to maintain the machines in factories, vehicles, servicing shops, and more. Lubricating oils are used to maintain the machines that produce used lubricating oil. The used lubricating oil in the machines is hazardous waste.

Based on Government Regulation Number 101 the Year 2014 about Hazardous Waste Management, used lubricating oil is hazardous waste from a non-specific resource. Used lubricating oil contains organic matters such as petroleum hydrocarbon (PHC) and heavy metals. The heavy metals found in used lubricating oil are iron, copper, and lead.

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^{*} Corresponding author: <u>ayunindyapuspa@itats.ac.id</u>

The leaching of used lubricating oil can be dangerous to the environment. The heavy metals that are contained in used lubricating oil are toxic, mutagenic, and carcinogenic. As the concentration of heavy metals in wastes varies across a wide range and may exceed the acceptance limit of the environment, heavy metal-bearing waste poses a serious threat to human and animal health when it spilled into the environment [1]. Used lubricating oil is a flammable liquid and has spreading potential to blanket environmental media, such as surface water. Used lubricating oil contains toxic material which, when inhaled, ingested or absorbed into the environment, then it will cause immediate or delayed adverse impacts on the environment using bioaccumulation upon the existing biotic systems. One of the methods that can be used to treat used lubricating oil is stabilization/solidification (S/S).

Stabilization/solidification is a hazardous waste treatment method using a mixture of cement and pozzolan to immobilize the waste so then the hazardous waste is less leachable [2]. The stabilization/solidification method is an effective method to use to reduce the toxicity of hazardous waste and the United States Environmental Protection Agency has approved its use for the clean-up of certain sites [1]. The S/S method is effective to use to immobilize inorganic waste, but it is not suitable for organic waste. The S/S product, with the addition of natural pozzolan, decreases the leaching result of heavy metal. The composition of natural pozzolan cement and waste (60:10:30) decreases the leaching concentration by up to 90% [3].

The S/S method usually uses cement, lime, and silicate to immobilize the inorganic components of the hazardous waste. The S/S method is based on the interaction between cement and the hazardous waste to form solid matter, both physically and chemically. Portland cement is widely used in the S/S method because it is purchasable and easy to use [4]. About 25 billion tonnes are produced each year globally. Most of all, Portland cement is used for concrete construction projects [5]. Cement is not able to solidify waste containing organic compounds, such as used lubricating oil. The adding of bentonite can absorb organic compounds. This research aims to determine the optimum composition of cement and bentonite to solidify used lubricating oil and to determine the results of the leaching test of the Fe and Pb after solidification.

2 Material and methods

The used lubricating oil that was solidified was from a motor vehicle repair shop. Before the used lubricating oil was solidified, the Fe and Pb concentration of the used lubricating oil was measured. The materials prepared were pulverized Portland Cement (PPC), bentonite, and used lubricating oil. All of the materials of the S/S product were mixed to have certain compositions. The compositions used in this research were cement 100%, cement-bentonite 25:75, cement-bentonite 75:25, and bentonite 75:25. Each composition was added to the used lubricating oil with a composition of 5%, 10%, and 15% weight. The total S/S products used in this research was 12. The S/S product compositions using Portland cement, bentonite, and used lubricating oil have been shown in Table 1.

The compositions of the S/S product were mixed, and the mixture was poured into the mold with 5x5x5cm sized. After the mixture of S/S product was poured, the S/S product must be settled in the mold in 24 hours. After 24 hours, the S/S products were released from the mould, and they were covered by a wet cloth for curing for 28 days. The wet cloth as a cover of S/S product should always wet. After 28 days, S/S products were analyzed their compressive strength and the TCLP of Fe and Pb. The results were compared with the Fe and Pb concentration in the used lubricating oil before stabilization/solidification was conducted.

Portland cement and bentonite were sieved with a 100 mesh sieve. The compositions used in this research were cement:bentonite 25:75, cement:bentonite 75:25, cement 100%, and bentonite 100%. Each composition was used to solidify the used lubricating oil with 5%, 10%, and 15% weight percentage. The compositions of Portland cement, bentonite, and used lubricating oil have been shown in Table 1.

Cement bentonite composition	Percentage weight of used lubricating oil		
	5%	10%	15%
cement 100%	A1	A2	A3
cement bentonite 25:75	A4	A5	A6
cement bentonite 75:25	A7	A8	A9
bentonite 100%	A10	A11	A12

Table 1. Composition of S/S product.

The method was conducted without specialized equipment. The Portland cement, bentonite, and used lubricating oil that had a determined composition were mixed to make them homogeneous, before being poured into a mold that 5x5x5cm in size. The mixture poured into the mold weighed 300 g. The mold that contained the mixture was covered with a wet cloth to control the moisture of the S/S products. After 24 hours, the S/S products were released from the mold, and the S/S products were covered by a wet cloth for 28 days. After 28 days, the S/S products were analyzed using the TCLP test to study the Fe and Pb. The TCLP test results were compared with the standard in the Government Regulation Number 101 the Year 2014 on Hazardous Waste Management (Peraturan Pemerintah Nomor 101 Tahun, 2014) and with the concentration of Fe and Pb in the used lubricating oil stated as well.

3 Results and discussion

In this research, the materials that were used to solidify the used lubricating oil were Portland cement and bentonite. Before solidification was conducted, the analysis of the concentration of Fe and Pb in the used lubricating oil was conducted first. Analyzing the concentration of Fe and Pb in the used lubricating oil was conducted to compare the decreasing rate of the Fe and Pb concentration before and after solidification. The concentration of Fe and Pb in the used lubricating oil has been shown in Table 2.

Table 2. The concentration of Fe and Pb in	the used lubricating oil.
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Heavy metal	Concentration (mg/L)
Fe	47.78
Рb	0.79

Based on the TCLP test results, the addition of Portland cement to the S/S product increased the Fe concentration. The TCLP test of Fe in the S/S product was higher than the Fe concentration in the used lubricating oil. The S/S product with the composition that was 100% cement had the highest TCLP value of Fe. The S/S product with a composition of

100% bentonite had the lowest TCLP value of Fe. The highest TCLP value of Fe was in the S/S product with a composition of cement 100% containing 15% used lubricating oil (417.89 mg/L). The lowest TCLP value of Fe was in the S/S product with a composition of 100% bentonite containing 10% used lubricating oil (0.085 mg/L). The TCLP value of Fe is not regulated in Government Regulation Number 101 the Year 2014 on Hazardous Waste Management (Peraturan Pemerintah Nomor 101 Tahun 2014). The adding of bentonite to the S/S product decreased the TCLP value of Fe. The TCLP values of the Fe results have been shown in Fig. 1.

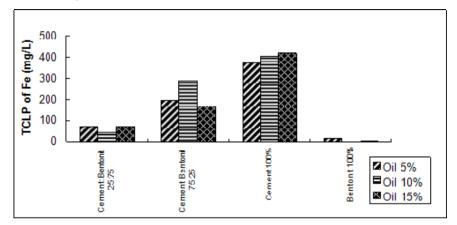


Fig. 1. The TCLP of Fe results in the S/S Product.

Portland cement contains 67% CaO, 22% SiO₂, 5% Al₂O₃, 3% Fe₂O₃, and 3% other components. Normally, it has four major phases: alite (50-70% Ca₃SiO₅ or C₃S), belite (15-30% Ca₂SiO₄ or C₂S), aluminate (5-10% Ca₃Al₂O₆ or C₃A), and ferrite (5-15% Ca₄Al₂Fe₂O₁₀ or C₄AF). Generally, organic compounds such as used lubricating oil affect the hydration between Portland cement and water [5]. Almost all organic compounds retard the hydration of cement, such as used lubricating oil as in this research. Therefore, to absorb the organic compounds, bentonite was added to the S/S product composition. The Fe that was contained in the cement was leached and caused the increase of the Fe value. In this research, bentonite absorbed the used lubricating oil well, so the TCLP value of Fe was low as a result. The hydration reactions have been shown in Eq. 1 and 2.

 $2C_{3}S + (3 - x + n) H \to C_{x}SH_{n} - (3 - x) CH$ (1)

$$2C_2S + 4.3H \rightarrow C_{1.7}SH_4 + 0.3CH$$
 (2)

The Pb concentration in the used lubricating oil was 0.79 mg/l. After the used lubricating oil was solidified, there was seen to be a decrease in the TCLP value of Pb. This happened because the Pb was trapped in the matrix of cement and bentonite. The addition of bentonite to the S/S product decreased the TCLP value of Pb. The S/S product with a composition of 100% bentonite was able to solidify the used lubricating oil 15% better than the S/S product with a composition of 100% cement. The used lubricating oil with a 5%, 10%, and 15% weight solidified with 100% cement had the highest TCLP values of Pb. The TCLP value of the Pb results has been shown in Fig. 2.

The standard of TCLP A and TCLP B related to the value of Pb are regulated in Government Regulation Number 101 the Year 2014 on Hazardous Waste Management (Peraturan Pemerintah Nomor 101 Tahun 2014). The Pb concentration for TCLP A is higher than 3 mg/L, and the Pb concentration for TCLP B is between 0.5-3 mg/L. If an S/S product contains Pb with a concentration higher than 3 mg/L after solidification, then the

S/S product must be disposed of the secure landfill. If the S/S product contains Pb between 0.5-3 mg/L after solidification, then the S/S product can be reused as a paving block. In this research study, all of the S/S products had a TCLP value of lead below 0.5 mg/L. Therefore all of the S/S product can be reused as paving blocks. The S/S product with a composition of cement and bentonite solidified the Pb well, although the waste contained used lubricating oil up to 15% in weight. The efficiency of the S/S method when solidifying the heavy metals depends on the solubility value of each heavy metal. The solubility of Pb hydroxides decreases with an increasing pH up to a value of about 10. In particular, the Pb concentration in leachate decreases with an increasing pH. When the pH reaches 9-11, Pb forms an insoluble hydroxide [6].

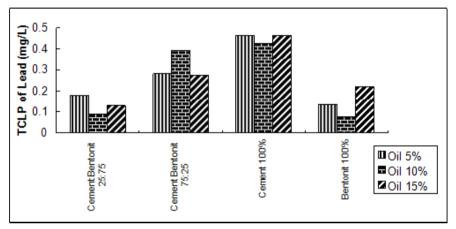


Fig. 2. The TCLP of Pb results in the S/S Product.

The efficiency of the S/S method to solidify contaminants may be improved using absorbents to absorb organic compounds specifically. The absorbents that can be used are metal oxide, clays, natural materials (zeolite, fly ash, polymers), and activated carbon. Bentonite is effective at solidifying used lubricating oil because bentonite is an organophilic clay that can absorb organic compounds. There were absorption processes both physically and chemically. Bentonite absorbed the organic matter while cement absorbed the inorganic matter. The physical adsorption phenomena occurred when the contaminants in the solution (pore water) were attracted to the surface of the particles because of the unsatisfied charges of the particles themselves. Chemical absorption refers to high affinity and specific absorption, which generally occurs through covalent bonding. The surface charges, chemical reactions involving surface functional groups and specifically absorbed ions greatly modify the binding capacity of the hydration products. The heavy metal will be precipitated as hydroxide, carbonates, sulfates, and silicates at the pH maximum [7].

4 Conclusions

An S/S product with a cement and bentonite composition cannot solidify the Fe contained in the used lubricating oil up to 15% weight. An S/S product with a cement and bentonite composition can solidify the Pb contained in the used lubricating oil up to 15% weight. The addition of bentonite can solidify the used lubricating oil more efficiently because bentonite absorbs the used lubricating oil. The optimum composition to use to solidify used lubricating oil is cement and bentonite 25:75. To improve the efficiency of the S/S method, the addition of activated carbon can be conducted in advanced research. This research was funded by Direktorat Riset dan Pengabdian Masyarakat Direktorat Jendral Penguatan Riset dan Pengembangan Kementerian Riset, Teknologi dan Pendidikan Tinggi Tahun Anggaran 2018.

References

- 1. A.K. Minocha, N. Jain, C.L. Verma, Con. Build. Mat. 17 (2003)
- 2. E.F. Barth, J. of Hazard. Mat. 24 (1990)
- 3. A. Gailius, B. Vacenovska, R. Drochytka, Materials Sci. (Medziagotyra) 16 (2010)
- 4. M.P. Utomo, E.W. Laksono, Pros. Seminar Nasional Penelitian, Pendidikan, dan Penerapan MIPA (2007)
- 5. K. Celik, M.D. Jackson, M. Mancio, Cem. Con. Comp. 45 (2014)
- 6. S. Paria, P.K. Yuet, Environ. Rev. 14 (2006)
- 7. Q.Y. Chen, M. Tyrer, C.D. Hills, X.M. Yang, P. Carey, Waste Man. 29 (2009)