The experimental test of the best abasorbents for controlling poly aromatic hydrocarbon on sea water around graving dock and hhip loading terminal

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Abstract. Bentonite and zeolite adsorbents have the ability to adsorb PAH content around the shipyard and ship loading and unloading areas. The factors that potentially affect the level are: a. The types of bentonite and zeolite adsorbents, b. Sampling time is 0, 2, 4, 6 and 8 hours. Adsorption of poly aromatic hydrocarbon by zeolite adsorbent as much as 150 gram done at certain place with volume of 3 liters of sea water taken from ship loading and unloading terminal area and at time variable 0, 7, 14, 28 and 42 days. Measurement of PAH on day 0 by way of direct examination of sea water concentration and PAH type. Determination of the best adsorbent, seen from the ability of the absorption of PAH both in the high concentration and the number of PAH types adsorbed, the results showed that zeolite adsorbents have better adsorption capacity because it has sufficient surface area and does not clot when exposed to water. The concentration of sea water PAH at ship loading and unloading terminal is very high, that is 65,45ppb, especially for PAH type of naphthalene and 2-Bromonaphthalene ie 28,3255 and 27,2821ppb respectively. The ability of zeolite adsorbent and maximum adsorption of PAH at 28 days adsorption, ie 19,75ppb is dominated by Benzo(a)pyrene type PAH with concentration of 19,2771ppb. Based on Linear Regression test to total PAH, it can be concluded that the value of R Square 0,190511343 which shows that the absorption time by zeolite absorbent and absorption power of poly aromatic hydrocarbons compound of the whole types has no significant effect. Therefore, polynomial testing is done through the equation \( y = -8,959x^3+62,858x^2-125,49x+72,379 \) yield value \( R^2= 1 \) which means absorption of 100% polyaromatic hydrocarbons compound in time.

Keyword: Adsorben, Bentonite, Zeolite, PAH

1. Introduction

Oil spills often occur on the high seas that come from ships, and often the occurrence of disposal by foreign ships in the water of Indonesia then there was a shift to the sea lips due to the air pressure and sea water waves. Operational ship ports are often packed by large vehicles fueled diesel, among others container vehicles and trucks loading goods for shipping by ship. The exhaust gases released by the large vehicle exhausts are the largest contributors to the formation of pollutants in the form of particulate poly aromatic hydrocarbon, which will then mix with sea water through rain water. In the activities of docking repair (tank cleaning) fuel oil on the graveing dock and floating repair around the port of loading and unloading of ships often occur oil spills mixed with sea water. Oil spills after mixing with sea water will happen chemical reactions that cause the formation PolyAromatic Hydrocarbon (PAH) compound.
and the reaction is accelerated of with the sun and free air, so that in a certain period can cause sea pollution[1]

With the inclusion of PAH as one of the parameters determining the quality of seawater, it is necessary to conduct research to obtain a description of how many types and concentrations of PAH content in sea water graving dock and sediment. Through the control of contamination of Poly Aromatic Hydrocarbon content of the waters around graving dock and sediment, it will get the waters of healthy sea water for marine biota. The experiment of PAH absorption by using the best adsorbent and determining the optimum policy of neutralizing the waste process is done with the classical optimization approach which yields the minimum process cost. This technique is done after the best known adsorbent type and can only achieve optimum process cost, but at the same time can not enter the environmental management as one of the goals that must also be achieved. Multiple objective goal programming, which is a procedure for solving multi purpose problems can be an alternative to not only economic objectives being calculated but also environmental interests[2]

The results showed that, Bentonite Adsorbents and natural Zeolites made of clay can be used for the absorption of Poly Aromatic Hydrocarbon (PAH) in seawater by the amount of 5 types of PAH that can be absorbed and variable time 2 to 8 hours. There was a significant correlation between the activated/not activated treatment with HCL solution on absorption capacity of PAH adsorbent bentonite and zeolite. There was a significant correlation between the sampling time variable and the absorption capacity of the PAH compound by bentonite and zeolite adsorbents. The longer the sampling time the more the number of adsorbents and the greater the concentration of PAH absorbed by the adsorbent.

This study aims to measure the adsorbent capacity of the zeolite type and adsorb the content of PAH in seawater from the ship loading and unloading area, determining the absorption rate by the adsorbent with the time and volume variables of sea water. The result of this research activity is expected to produce the model of seawater contamination control using MultipleObjective Goal Programming method, which is a form of development model for determining the policy of concentration of adsorbent for optimum in cost and environment and can be followed up to government policy for all shipyard doing docking repair in graving dock and floating repair around the port of loading and unloading vessels to neutralize oil spills by using appropriate PAH to avoid (marine pollution) in accordance with IMO provisions set forth in protocol marpoll 1973/1978 (Annex 1)[3]

2. Adsorben and Poly AromaticHydrocarbon
2.1 Adsorbent zeolite
Zeolite natural mineral which is an alumino-silicate compound with a caged structure is widely available in the area of Indonesia scattered in various locations. Zeolite minerals in Indonesia in general in the form of pure zeolite minerals with impurities in the form of calcium carbonate, complex silicate compounds and others. Until now, the utilization of zeolite is limited only for agricultural and fishery purposes as absorbent material of ammonia in waters and nutrient binders in plants. So zeolite has a very cheap selling value if only limited to be used as a material of fishery and agricultural purposes. For example, green zeolites are commonly found in Cidadap area, Tasikmalaya regency that has been utilized for agriculture and fisheries with low selling value. Therefore, to increase the selling point of green zeolite, a series of green adsorption activities were conducted on chromium metal ions. Through this paper will be described the adsorption power of zeolite by using Langmuir equation and Freundlich equation. There are three adsorption adsorption isotherms, namely Freundlich adsorption isotherm, Langmuir, and BET (Brunauer, Emmet and Teller). However, since the adsorption of molecules or ions on the surface of the solid is generally limited to a single molecular layer (monolayer), the adsorption follows the Freundlich and or Langmuir adsorption equations[4]

2.2 Poly AromaticHydrocarbon
The compound PAH (Polycyclic Aromatic Hydrocarbine) is a widespread organic compound in nature, its shape composed of several cyclicaromatic chain is hydrophobic. PAH compounds contain two or more benzene chains, derived from pyrolysis, incomplete combustion (forest burning, motor discharges, volcanoes) and combustion processes that use high temperatures in petroleum processing.
According to NEFF the combustion process greatly affects the type and amount of PAH produced. Source PAH dialam is burning fossil fuel. According to the high concentrations of PAHs were obtained in marine sediments close to urban areas. This may be a common pattern because PAH tend to congregate in marine sediments close to urban areas. PAH compounds easily settle down the waters, and are very toxic to aquatic organisms. PAH dissolved in water, at levels between 0.1 and 0.5 ppm can already cause poisoning against all larvae of aquatic biota. According to PAH compounds will accumulate to a high level in low-animal bodies, because these compounds are difficult to digest in their bodies, PAHs will accumulate in the tissues of marine organisms. get the most flat PAH levels in green shell size 1.0-1.5 cm in Jakarta Bay equal to 167,253ppb. get levels of PAH in the body Nomeifish(Horpodon nehereud) in Tarakan waters ranged from 1.582-2.747 ng/g, and get levels of PAH in the fish body from the southern sea, Baron are, Yogyakarta is petek fish (Chlorinomus lyson) ranged from 0.9-310.9 ng/g. And Kunikran fish (Uppeneus moluccensis) ranges from 1.9-1072.5 ng/g. When the shellfish and fish are eaten by humans, the PAH will accumulate in the human body, and to some extent can cause disruption to human health.[5]

This aromatic hydrocarbon compound is toxic, one of which is the PAH which is an aromatic compound with two or more benzene rings. PAHs dissolved at concentrations of 0.1 to 0.5 ppm can cause toxicity to living things, whereas low-level PAHs can decrease the rate of growth, development, and eating of aquatic creatures. This situation has been disclosed by for fish, crustaceans and mollusces. In addition, petroleum hydrocarbons absorbed into the body of the biota cause a stinging taste and require a certain time to be lost. At 10 ppm content of aromatic hydrocarbon compounds can cause changes in behavior patterns in marine biota and at levels > 1000ppm can cause death. This situation is harmful to aquatic organisms living and feeding in marine sediments. The threshold value (NAB) of aromatic hydrocarbons for marine biota is 0.003 ppm

2.3. The type and content of polyaromatic hydrocarbon

Previous research (Zulaihah, 2015) on seawater and sediment on the graving dock floor and around the shipyard's shipping area, as well as on ship loading and unloading terminals. The result data of the six samples consist of 4 samples of sea water and 2 samples from sediment. From the results of the test showed that there are 7 types of PAH with low molecular weight which is called LMW and 9 types of PAH with high molecular weight commonly called HMW. The results of the study (Zulaihah 2015) showed that PAH content in sea water that has a light molecular weight (LMW) on the graving dock floor of 0.543ppm, outside the graving dock 25.191ppm, and loading and unloading port area 3.345ppm. While the adsorbent adsorption result of bentonite in activation for 8 hours which have been obtained in this research equal to 0.422ppm, hence if adsorbent is used to adsorb PAH content above outside graving dock 25.191ppm needed time 19.9 day. The spread of high molecular weight (HMW) PAAH content in sea water outside the graving dock and in the port area of loading and unloading vessels is very high at 44.681ppb and 217.047ppb. While the result of bentonite absorption in activation at 8 hours for PAH content having high molecular weight of 0.71ppm, if the adsorbent is used to adsorb the largest PAH at loading and unloading port 217.047 it takes 101.87 days. The result of monitoring of research center of oceanography of LIPI year 2011 is 29.054ppb and in year 2013 is 35.193ppb, where sample is taken at sea. So to absorb if using bentonite adsorbent in activation takes 13.6 days and for monitoring in 2011 and 16.5 days with the assumption of adsorbent is installed every day 24 hours[6]

3. Material test

3.1 Annova Testing 2 Factors

Annova test is done with 2 factors, namely heat treatment and test duration. Omactivated bentolit compounds and not heated by 2 hours (A2) and activated bentolites with 8 hours (A8) and bentolite and heated with 2 hours (S1) and activated bentolite concluded that the duration of testing and heat treatment did not significantly influence the absorption capacity of PAH compound by bentonite adsorbent. Its adsorption abilty is limited so it needs to be activated with strong acids to produce bentonite with higher adsorption capabitilily. This research in addition to activation also do the heating to increase the absorption. Considering that PAH is a non-soluble compound. There was a significant relation ship between treatment the activated or not activated with the absorption of bentonite and zeolite. The results
of the study according so that the unactivated bentonite adsorbent in the 2 hour time variable can absorb PAH content with the type of Naphthalene, anthrasne, and fluoren with total absorbed PAH of 0.674 and time 8 hours of Naphthalene, anthrasne, fluoren, pyrene and chrysene total 1.132ppm. where as bentonite adsorbent was activated at 2 hours absorption time there were 4 types of adsorbent absorbed with total 0.9102 at 8 hours into 5 types of adsorbent absorbed with total 1.131ppm[6].

3.2 The effect of zeolite adsobene being activated and not dictated to PAH levels
Effect of PAH absorption in loading and unloading area of ship and unloading ex-presiden terminal, zeolite absorbent is activated first by using HCL, then inserted into seawater for absorption process of PAH pollutant in seawater with time variable 2, 4, 6 and 8 hours, as shown in table 1 and figure 1.

### Table 1. The content of PAH absorbed by zeolite adsorben which are activated and not activated

| PAH  | B2  | B8  | D2  | D8  |
|------|-----|-----|-----|-----|
| Naph | 0.2313 | 0.2339 | 0.23680 | 0.2361 |
| Anth | 0.0000 | 0.1892 | 0.1893 | 0.1898 |
| Fluo | 0.0000 | 0.0000 | 0.2471 | 0.2480 |
| Py   | 0.2315 | 0.2320 | 0.2320 | 0.2276 |
| Ch   | 0.0000 | 0.0000 | 0.0000 | 0.2276 |
| Total| 0.4627 | 0.6546 | 0.6684 | 1.1335 |

**Figure 1.** The effect of zeolite absorbent is activated and not activated to the type and concentration of PAH

The result of the research on table 1 showes that the unactivated zeolite adsorbent in the 2 hour time variable can absorb PAH content with Naphthalene and pyrene type with total PAH 0.463 while at 8 hours of PAH type is absorbed by Naphthalene, Anthracen and Pyrene with total 0.6546ppm. Activated zeolite adsorbent at 2 hours, there are 4 adsorbent types absorbed such as Naphthalen, Anthracene, Fluoranthe and Pyrene with total 0.9148 and at 8 hours into 5 types of adsorbents absorbed such as Naphthalen, Anthrasene, Fluoranthe, Pyrenechryse with total 1.133ppm[7].

3.3 The effect of sampling time on zeolite adsorben
The influence of time in the absorption of PAH in the area of ship reparation is because the area is suspected to contain PAH Pollutan from waste disposal at the time of reparation of ship according to table 2. And figure 2.
Table 2. PAH content absorbed by Zeolite adsorbent with time variable 2, 4, 6 and 8 hours

| PAH   | B2   | B4   | B6   | B8   |
|-------|------|------|------|------|
| Naph  | 0.2313 | 0.2314 | 0.2319 | 0.2339 |
| Anth  | 0.0000 | 0.1894 | 0.1893 | 0.1892 |
| Fluo  | 0.0000 | 0.2317 | 0.0000 | 0.0000 |
| Py    | 0.2314 | 0.0000 | 0.2316 | 0.2315 |
| Ch    | 0.0000 | 0.0000 | 0.0000 | 0.0000 |
| Total HMW | 0.4627 | 0.6525 | 0.6528 | 0.6546 |
| Total HMW | 0.4630 | 0.6530 | 0.6528 | 0.6546 |

Figure 2. Level of absorbent zeolite absorption PAH with time 2, 4, 6 dan 8 hours

Table 2 shows the zeolite adsorbents has the ability to absorb with two PAH types on 2 hour variable, such as Naphthalene and Pyrene. While in the time variables 4, 6 and 8 only 3 types of PAH can be absorbed with a maximum total concentration of 0.6546 ppb. Zeolite Adsorbent the longer the absorption time the greater the concentration of PAH and the more adsorbent absorbed, as well as for activated adsorbent, the more type and concentrations of PAH are adsorbed. The result of annova calculation with 3 variables shows that there is a significant relationship between the treatment is activated or not activated with the absorption of bentonite and zeolite with the indicated that \( F \) arithmetic \( 179.5922 \) is bigger than the \( F \) table 161. So with variable time, there is a significant relationship between the duration of immersion with the absorption of compounds by bentonite and zeolite premises showed that \( F \) arithmetic 162.3714 is greater than \( F \) table amount 161[8]

3.4 Effect of absorption time by Zeolite adsorbent on PAH concentration and the number of PAH types, on the amount of 150 gram sacrificial adsorbent in 3 liters of water sea.

The use zeolite absorbent for the absorption treatment at a given volume, 150 grams of zeolite were fed into 3 liters of seawater with time variable of 1, 2, 4 and 6 weeks. Number and type of PAH absorbed in accordance with table 3. And figure 3.
Table 3. Concentrations and PAH types absorbed by Zeolite adsorbents with variable time 0 to 42 days.

| Type of PAH        | Concentration of PAH on the seawater (day 0) | Concentration of PAH on the zeolite adsorbents (day 7th) | Concentration of PAH on the zeolite adsorbents (day 14th) | Concentration of PAH on the zeolite adsorbents (day 28th) | Concentration of PAH on the zeolite adsorbents (day 42nd) |
|--------------------|---------------------------------------------|----------------------------------------------------------|----------------------------------------------------------|----------------------------------------------------------|----------------------------------------------------------|
| Naphthalene        | 28.326                                      | 0.021                                                    | 0.680                                                    | 0.348                                                    | 0.120                                                    |
| 2-Bromonaphthalene | 27.282                                      | 0.046                                                    | 0.400                                                    | 0.121                                                    | 0.827                                                    |
| Anthracene         | 4.538                                       | 0.267                                                    | 0.041                                                    | 0.000                                                    | 0.113                                                    |
| Fluoranthen        | 2.209                                       | 0.257                                                    | 0.029                                                    | 0.000                                                    | 0.072                                                    |
| Pyrene             | 3.096                                       | 0.090                                                    | 0.017                                                    | 0.000                                                    | 0.041                                                    |
| Benzo(a)anthracene | 0.000                                       | 0.110                                                    | 0.000                                                    | 0.004                                                    | 0.176                                                    |
| Benzo(a) pyrene    | 0.000                                       | 0.000                                                    | 0.000                                                    | 19.277                                                   | 0.758                                                    |
| Total              | 65.451                                      | 0.791                                                    | 1.166                                                    | 19.750                                                   | 2.107                                                    |

Figure 3. Concentrations and types of PAHs absorbed by absorbent zeolite with a range of 0 to 42 hours

From table 3, show that the concentration of PAH on sea water in the very high ship loading and unloading area is 65.45ppb, especially for PAH type of Naphthalene and 2-Bromonaphthalene is 28.3255ppb and 27.2282ppb. With absorption, concentration of PAH which can be absorbed as much as 1,166ppb. The ability of zeolite adsorbent in maximum absorption of PAH at 28 days, that is 19.75ppb. Which is dominated by Benzo(a)Pyrene type PAH with concentration equal to 19.2771ppb. This indicates that the PAH type of Naphthalene and 2-Bromonaphthalene which is PAH type consisting of 2 aromatic plicyclic of low molecular compounds, which undergoes oxidation reaction due to the presence of oxygen and heat, into a Benzo (a) Pyrene PAH type which has more than 3 aromatic compounds ring and has high specific gravity. At the time of 42 days absorption ability of zeolite absorption decreased, that is become 2,789ppb. [9]

Based on the result of Linear Regression test to total PAH, it can be concluded that the value of R square 0.19051343 which shows that the absorption time by zeolite adsorbent on absorption power of
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hydrocarbon hydrocarbon compound of whole type does not significantly influence. Therefore, polynomial testing is done through the equation: \( y = -8.959x^3 + 62.858x^2 - 125.49x + 72.379 \) yields \( R^2 = 1 \) which means the absorption state of 100% hydrocarbon polymeric compound as a whole influences by time, indicating that the concentration of PAH in sea water diarea ship loading and unloading terminal is very high, that is 65.45 ppb, especially for PAH type of naphthalene and 2-Bromonaphthalen that is respectively 28.3255 and 27.2821 ppb. With absorption of zeolite adsorbent for 7 days, PAH can be absorbed as much as 0.791 ppb, at 14 days of absorption, PAH concentration can be absorbed as much as 1.166 ppb. The ability of zeolite adsorbent in maximum absorption of PAH at 28 days, is 19.75 ppb which is dominated by Benzo (a) pyrene type PAH with concentration of 19.2771 ppb. This indicates that the PAH type of naphthalene and 2-Bromonaphthalen which is a PAH type consisting of 2 low molecular weight aromatic polycyclic compounds undergoes an oxidation reaction. Due to the presence of oxygen and heat, it becomes a Benzo PAH (a) Pyren having more than 3 aromatic compounds cicin and has a high specific gravity. At the time of 42 days absorption ability of zeolite absorption decreased, that is become 2.789 ppb. [10]

Based on ANOVA test result on naphthalene compound, it can be concluded that the absorption time by zeolite adsorbent on absorption of polymer hydrocarbon naphthalene does not significantly influence. This is because the compound of naphthalene found in contaminated sea water PAH of 28.3255 ppb can be absorbed by zeolite adsorbent, but with the heat of the environment and absorption time for 28 days. Then the naphthalene undergoes oxidation and reduction reactions to form Benzo (a) pyren.

Naphthalene is a crystalline aromatic hydrocarbon shaped white solid with a formula C10H8 molecule and is in the form of two unified benzene rings. This compound is volatile, volatile despite formation of solids. The resulting vapor is flammable. Naphthalene is one component that includes benzene aromatic hydrocarbons, one molecule of naphthalene is a fusion of a pair of benzene rings. Naphthalene is one type of polycyclic aromatic hydrocarbon. The polycyclic compound can be hydrogenated (reduced) annually at room pressure and temperature. The partially reduced ring system still contains benzene rings. Most of the aromatic properties of the ring system still exist and are preserved. To hydrogenate all aromatic rings in naphthalene can be carried out at high temperature and pressure. PAHs are formed when complex organic substances are exposed to high temperatures or high pressures [11]

Based on ANOVA test of bromopnaphthalenen, it can be concluded that the absorption time by zeolite adsorbent on absorption of poliaromatik hydrocarbon type 2-Bromonaphthalene has no significant effect. This is because the 2-Bromonaphthalene compound contained in seawater contaminated with PAH waste of 27.2821 ppb can be absorbed by zeolite adsorbent, but in the presence of environmental heat and absorption time for 28 days, the 2-Bromonaphthalene undergoes oxidation and reduction reactions so that Benzo forms (a) pyren and partly evaporates or decomposes in view of its volatile nature. The naphthalene bromination reaction can occur at room temperature using the FeBr3 catalyst. The reaction which is happened to use reaction mecanism as shown in figure 5. Naturally occurring compounds in food or processed products that are toxic to humans. Compound bioactive compounds that have physiological effects in the body that affect positive to human health.

4. Conclusion
4.1 From the calculation of anova either 2 or 3 variable then that the factors that influence the absorption of bentonit and zeolit compounds is the treatment of adsorbents are activated and not activated and variable sampling time from 2 to 8 hours. Moderate heating at the time of sampling did not show any significant relationship.
4.2 The results of inspection in the laboratory of Oceanography Institute LIPI showed that Zeolite adsorbents have better absorption capability due to zeolite has sufficient surface area and not clot when contacted by water.
4.3 The concentration of PAH in seawater at ship loading and unloading terminal is very high, that is 65.45 ppb, especially for PAH type of naphthalene 28.3255 and 2-Bromonaphthalen 27.2821 ppb. Ability of zeolite in maximum absorption of PAH at 28 days, that is 19.75 ppb which is dominated by Benzo (a)
pyren type PAHren with concentration equal to 19.2771ppb this shows that PAH type of naphthalene and 2-Bromonaphthalen which is PAH type consist of 2 aromatic polycyclic compounds with low molecular weight, undergo oxidation reactions due to the presence of oxygen and heat, into Benzo PAH type (a) Pyren which is an aromatic compound more than 3 cicin and has a high specific gravity.

4.4 Based on the results of Linear Regression test to total PAH, it can be concluded that the value of R^2 0.190511343 which indicates that the absorption time by zeolite adsorbent on the absorption of hydrocarbon hydrocarbon compounds of the whole type does not significantly influence. Therefore, polynomial testing is done through the equation: 

\[ y = -8.959x^3 + 62,858x^2 - 125,49x + 72,379 \]

yield value R^2 = 1 which means the absorption state of 100% hydrocarbon poliaromatics as a whole is influenced by time.

4.5 Based on the results of the ANOVA test showed that the absorption time by zeolite adsorbent on absorption of naphthalene and 2-bromo naphthalene did not significantly influence. In the presence of environmental heat and absorption time for 28 days, the compound undergoes oxidation and reduction reactions to form Benzo (a) pyren.

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