The Effect of Activated Carbon on Biodegradation of Soil and Ground Water Contaminated with Petroleum

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ABSTRACT

The soil and ground water contaminated with petroleum research increases in many observation. This will become important since the soil and ground water contaminated petroleum was hazardous waste and toxic for human life, aquatic biota and not environmental friendly. Biological Remediation process as called Bioremediation is one of popular treatment for Total Petroleum Hydrocarbon (TPH) removal, which is in slurry phase system (contaminated soil contacted with make up water). In this research was hold on slurry phase system. The purpose of this research was to evaluate the biodegradation of hydrocarbon degradating bacteria in the presence of activated carbon hold on slurry phase system. In this research Total petroleum hydrocarbon (TPH) removal will become measurement parameter since activated carbon and bacteria simultaneously in 250 mL batch reactor. From this research was found that with 10 gr activated carbon and bacteria revealed that there was a significant TPH reduction in the level 11,12 % to 0,94 % during 18 hour observation. The percentage reduction was found 91,55% for reactor with activated carbon contacted bacteria and 85,5 % for reactor with bacteria only. The pattern of bacterial growth in activated carbon added to the reactor which is also better than the reactor without the addition of activated carbon.

Keywords: activated carbon, biodegradation, slurry, TPH.

INTRODUCTION

The process of exploration and refining of petroleum and natural gas primarily petroleum cause environmental degradation, this was due to contamination by petroleum from the beginning of the process until the final stages of distribution as well as spills or leaks occur when the exploration, refining and distribution. This condition can cause soil contamination on groundwater and even dangerous for human health, aquatic biota and other living creatures if not handled properly.

These contaminants can be in liquid phase of oil, gas and solid. The pollutants from hazardous wastes categorized as having physical and chemical characteristics that can cause illness to humans and other living creatures, especially type cyclic aromatic hydrocarbon chains that are difficult to be degradated by microorganisms that exist in the soil. Several studies were conducted to overcome the problem of soil or groundwater contaminated with petroleum, such biological processes.

Remediation process of soil and ground water contaminated oil by microorganisms (bioremediation) has been progressing quite rapidly, both in scale or applied research. Biological processes will depend on the character of pollutants and microorganisms are used, and also the bioavailability of pollutants. The study has been developed using activated carbon adsorption process as a combination of physical and biological degradation. In this study a combination of physical and biological processes are examined from various aspects of the study, and expect to get the CONCLUSIONS of the effect of addition of activated carbon on the biodegradation happens.



METHODS

Research conducted in several stages, namely the preparation phase materials and tools, experimental phase using a variation of carbon physically active, proceed to the stage of research using a combination of activated carbon and a consortium of oil degrading bacteria, followed by data processing. Processing data is done using the equations associated with mechanism of processes that occurred in each experiment, also conducted observations by using GC/MS.

In the preparation stage included the preparation of activated carbon materials (in this case using a commercial activated carbon). Activated carbon is used type of *Granular Activated Carbon* (with commercial name Karbosorb type ANK-103) with surface areas of 700-1400 m²/g and 12-40 mesh particle size produced from *coconut shell*. The Soil contaminated with crude oil derived from artificial wastewater by adding crude oil originating from the Environmental Biotechnology Laboratory collection PPAU-ITB in the soil. The soil is sterilized before using Autoclaf to avoid the influence of indigenous microorganisms in the process of biodegradation process of trial and so that the effect is expected only from the bacteria were added into the experimental process. While the bacteria used in the process of biodegradation is a consortium of oil degrading bacteria Environmental Biotechnology Laboratory collection PPAU-ITB which have been isolated from different regions and have been tested biodegradation, with maximum specific growth rate μ_{max} of 0.018 hour ⁻¹ and k_s 0.224% (v/v) (Wardah, 2003).

Tools required in this study include 500 mL erlenmeyer as a reactor for the observation of physical processes and a combination of physical and biological processes, shakers, oven, waterbath and spectrophotometer. Observations allowance for TPH using activated carbon and a consortium of bacteria carried in the slurry phase, by adding 75 g of petroleum-contaminated soil in 250 mL of distilled water (30% solid), because the percentage of solid in slurry phase bioremediation were in the range 20-50% (Eweis & Ergas, 1998), it is mentioned that bioremediation processes run optimally. On experiments using activated carbon TPH concentration in the slurry is 2%, and 4.7%, while the variation of active carbon is added into the reactor is the observation time 5,10,15,20 grams with 0,1,2,4, and 24 hours. This experiment aimed to obtain the amount of addition of activated carbon that provides the largest removal efficiency TPH at concentrations that have been varied, the activated carbon can be set aside specific hydrocarbon compounds poliaromatik anthracene in the soil matrix and water with the reaction rate constant 0018-0051 day^{-1,} with the adsorption on activated carbon is able to improve provision for anthracene in the matrix (Owabor & Aluyor, 2008).

In the physical-biological experiments, using activated carbon and a consortium of oil degrading bacteria, the bacteria previously adapted and acclimatization in the minimum liquid SBS medium so as to speed the phase lag, so that when inserted into the reactor, bacteria in a state close to logarithmic phase. As a source of nitrogen and bacteria needed Phosphat then added KH $_2$ PO $_4$ and (NH $_4$) $_2$ SO $_4$, while the carbon sources derived from crude oil. The experiment was conducted at ranges up to 18 hours, against four variations of the treatment, which uses bacteria and carbon active, only bacteria, only the activated carbon and without any treatment. The parameters used were: TPH (gravimetrically), the viability of bacterial cells (total plate count method, and spectrophotometry (optical density), pH, temperature and Dissolved oxygen (DO).

RESULTS AND DISCUSSIONS

TPH Removal and Efficiency in Experiments Using Activated Carbon. This experiment was done by adding 75 g soil and 250 mL of distilled water in 500 mL erlenmeyer at each concentration of TPH and the variation of activated carbon used. By using the shaker 120 rpm for observation time 0,1,2,4



and 24 hours to see the provision for TPH. For 2% concentration slurry highest TPH removal efficiency occurred at 1 hour observation time ie 96.8% on the addition of 20 grams of activated carbon, whereas at 24 hours into the highest removal efficiency occurred in the addition of five grams of activated carbon with 87.5% efficiency. While the slurry at a concentration of 4.7% highest TPH removal efficiency occurred at 1 hour observation time ie 98.51% on the addition of 10 grams of activated carbon. The highest removal efficiency occurred at the first observation time 1 hour. This indicates that at a longer contact time resulted in desorption from the surface of activated carbon into the slurry solution, so the addition of activated carbon for the removal of TPH in the slurry phase will be effective in a relatively short time. The same thing happened at a concentration of 4.7% TPH, the second image can be seen that the loading slurry concentration 4.7% TPH, the highest removal efficiency occurred at 1 hour observation time. This also indicates that the contact time and amount of activated carbon is added will affect TPH removal efficiency, so need to get the time and the optimum amount of activated carbon were added.

On the physical removal of TPH using activated carbon for each variation of TPH concentration, occurred subsequent desorption of contaminants from the soil occurs in the adsorption of contaminants in the slurry solution on the surface of activated carbon.

The adsorption is physical adsorption occurs so relatively weak bond formed, and therefore at a longer contact time is expected to cause contaminants desorption back into the slurry solution. This situation is evidenced in the increase again TPH slurry on a large observation time of two hours at a concentration Slurry 4.7%, but does not occur at a concentration of 2%. From these results it is known that the allowance for TPH in the slurry phase was relatively short time needed for a greater concentration when compared with smaller concentrations. But this state has a weakness how to reduce or even overcome the re-occurrence of desorption of contaminants that have been adsorbed on the surface of activated carbon, which lasted TPH optimal provision. This includes the underlying future experiments that combine the physical removal of TPH using activated carbon with biological processes that use bacteria consortium. In experiments in physics allowance TPH using activated carbon is found that the largest TPH removal efficiency occurred at a concentration of 4.7% TPH slurry with the addition of 10 g of activated carbon with the adsorption of contaminants is not more than two hours. These results will be used as basis for selection of the concentration of slurry and addition of activated carbon to be used on subsequent biodegradation experiment.

Identify Characteristics of domestic Bacterial Growth with Addition of Activated Carbon. To better understand the profile of bacterial growth with the addition of activated carbon, the bacterial growth observed in the SBS medium *(base salt solution)* in each of the TPH concentration of 2% and 4.7%, as was done in experiments using activated carbon. The growth of bacteria in the medium was evaluated by *optical density* using a spectrophotometer level. The observation is presented in Figure 1.

From the first picture, can be seen that the growth of bacteria at 2% concentration of TPH in the presence of activated carbon begins to experience a growth in the first hour of observation time, but fluctuate in the third hour and thereafter at each variation of the addition of activated carbon. While on a larger concentration (4.7% TPH) growth of bacteria tended to increase since the first hour to four variations on each of the activated carbon used.



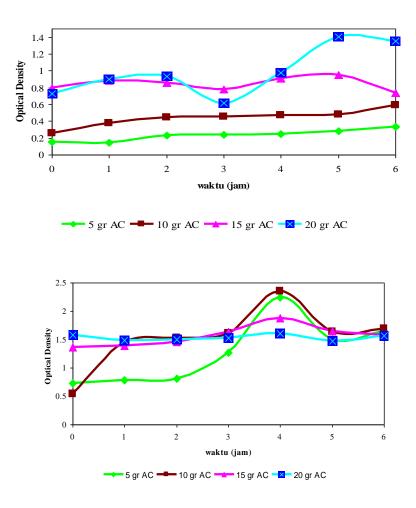
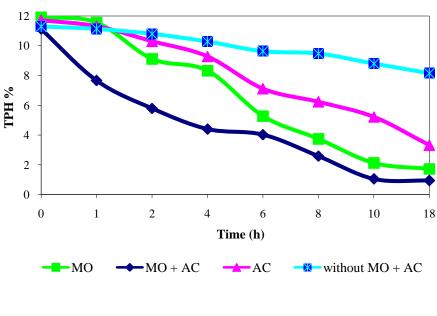


Figure 1. (a) Growth of bacteria at 2% concentration of TPH (b)Growth of bacteria at concentrations of 4.7% TPH

Determination of TPH in Slurry Phase by Addition of Activated Carbon and Petroleum Degrading Bacterial Consortium. At this stage of research carried out by using a simple batch reactor (erlenmeyer, 500 mL) with stirring 120 rpm on shaker in the slurry phase of the fourth reactor, namely: reactor 1 (MO + AC) with the addition of a consortium of bacteria and activated carbon, reactor 2 (MO) only the addition of a consortium of bacteria, reactor 3 (AC), only the addition of activated carbon while the reactor 4 (Without MO + AC) without the addition of a consortium of bacteria and activated carbon (control). The removal of TPH can be seen in the Figure 2.

From the images could be seen that there is the largest decline in TPH occurred in the reactor using activated carbon and bacterial consortium with TPH from 11.12% to 0.94% during 18 hours. While in the reactor MO TPH reduction of 11.91% to 1.722%, while in the AC reactor TPH reduction occurred from 11.71% to 3.314%, different case with the reactor without any treatment TPH suffered only distilled water until the dispersion due to addition of TPH were in the range 11.29 to 8.15%. The addition of 10 grams of saturated activated carbon was achieved at the large time span of four hours, so analyzed that the adsorption occurs on the range of 1-4 hours and biodegradation occurs at a larger time span, as has been tested on the growth characteristics of bacterial consortia, where the bacteria continued to grow since 1-4 hours on the SBS medium is contacted with activated carbon.



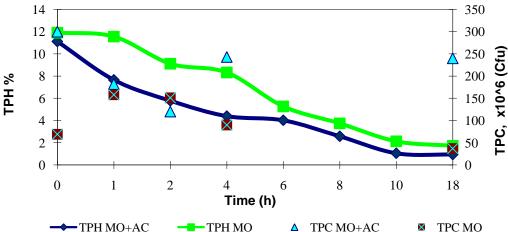


Figure 2. TPH Removal and viability of bacterial cells in Each Reactor

When sorption sites already saturated or reduced the ability of bacterial consortia are more dominant role in the provision TPH up to 18 hours of contact time, so the provision TPH tend more slowly and more sloping curve is formed. But the interesting thing here that time of bacteria in the reactor stasionery MO + AC longer than the reactor MO, where up to 18 hours to six days of bacteria was nearing decay, this can be seen from the profile found in the number of bacterial colonies. This situation causes biodegradation ability decreases. Survival of the bacteria in MO + AC reactor is caused by the existence of activated carbon, because in addition to used as media of bacterial adherence on the surface of activated carbon, also because of the contaminant (crude oil) which has adsorped by activated carbon can be used as a carbon source for bacteria, the presence of carbon also able to enhance the ability of active compounds mineralization poliaromatic specific hydrocarbon phenanthrene by bacteria (Leglize, et al., 2008), because of the adhesion on the surface of activated carbon by bacteria.

Slurry Phase Biodegradation Kinetics in a Batch. Kinetics of biodegradation was observed at the reactor MO + AC, MO reactor and AC reactor. Biodegradation dominated by reaction kinetics of zero



order and first order (Doran, 1995; Fogler, 1999), the same is also evident in the bioremediation of contaminated soil in the reactor bioslurry anthracene followed zero order kinetics and first order evidenced by the coefficient of determination (r²) The greatest (Prasanna et al, 2008).

Thing to do is create Ct vs t plots in each reactor with the approach of TPH data: Order zero (n = 0): plot of Ct vs t, where the slope = -k, and the Order of 1 (n = 1): plot of ln (Ct / Co.) vs. t, where the slope = k, while the half-life (t $_{\frac{1}{2}}$) = C/2k at zero order, the order of 1 = 0.693 / k.It was found that the MO + AC reactor on the order of a coefficient of determination is greater than zero order so that MO + AC reactor is said to have first-order reaction in the mechanism of biodegradation, the price reaction rate constant is 0.3541 hour $^{-1}$ with half-an, 96 hours. While in the reactor, MO and AC tend to follow zero order in the mechanism of degradation, with the price of 16 386 and 12 340 k g/kg soil / h and 3.4 and 4.5 half hours.

The higher the reaction order indicates the more rapid reaction takes place, this is consistent with the data allowance for TPH that occurred in the reactor when compared with the MO and AC reactors. In the reactor MO + AC biggest TPH removal efficiency when compared with the MO and AC reactors. This situation is supported by data showing the first order reaction kinetics in a larger reactor (n = 1) when compared with MO reactor and AC (n = 0).

The observation by using GC/MS also showed significant changes to the hydrocarbon compounds before and after treatment.

CONCLUSIONSS

From this research can be concluded that the addition of activated carbon and a consortium of bacteria in the slurry phase reactor batch of crude oil contaminated soil can increase TPH removal efficiency without inhibiting bacterial growth, when compared to just using a consortium of bacteria or activated carbon alone, so it will be able to enhance soil and ground water bioremediation process optimization in the *slurry treatment*.

REFERENCES

Doran, PM, 1995, Bioprocess Engineering Principles, Academic Press, UK

Eweis, Ergas, 1998, Bioremediation Principles, McGraw Hill, USA

Fogler, HS, 1999. Elements of Chemical Reaction Engineering, Third Edition, Prentice Hall Inc. USA. USA.

Leglize.P, Alain.S, Jacques.B, Corinne.L, Adsorption of phenanthrene on activated carbon increases mineralization rate by specific bacteria, Journal of Hazardous materials, 151 (2008): 339-347

Notodarmojo, S, 2005, Soil and Groundwater Pollution, Publisher files. Bandung. Bandung.

Owabor, CN, Aluyor, EO, Application of adsorbents as a novel technique During biodegradation of polycyclic aromatic hydrocarbons (anthracene), African Journal of biotechnology, 7 (18) (2008) :3321-3325.

Prasanna, Mohan.S, Reddy BP, Sarma.P, N, *Bioremediation of Contaminated Soil in Anthracene Bio-slurry phase reactor Operated in Periodic discontinuous batch mode, Journal of Hazardous materials,* 153 (2008): pp 244-251

Wardah, A, 2003, Effect of Compost on soil Bioremediation Lubricating Oils contaminated by bacteria on the scale of Commercial Products Laboratory, ITB. Bandung.

