IJCPE
Iraqi Journal of Chemical and Petroleum Engineering

Iraqi Journal of Chemical and Petroleum Engineering
Vol.9 No.2 (June 2008) 69-74
ISSN: 1997-4884

University of Baghdad
College of Engineering

Biotreatment of Oil Wells Drilling Waste in an Agricultural Soil

Muna H. Al-Joubori* and Ayad A. A. H. Abd Al-Razaq*

* *Department of Biology, College of Science, University of Baghdad*

*** Department of Geology, College of Science, University of Baghdad*

Abstract

One of the most important environmental issues is the responsible effective and economic treatment of drilling waste especially oily waste.

In this research two fungal isolates named Pleurotus ostreatus and Trichoderma harzianum were chosen for the first time to treat biologically the oily drilled cuttings contaminated with diesel which resulted from drilling oil wells use oil based muds (OBMs).

The results showed that the fungi under study utilized the hydrocarbon of contaminated soil as a source of nutrient and growth and that both fungi can be considered hydrocarbon degrading microorganisms. The used biotreatment is cost effective process since most of the materials used in the cultivation and growth of the present fungi were available and cheap agricultural waste.

The best hydrocarbon degradation was observed in case of using both fungi together with 8 % by weight microorganisms concentration ratio and with the same ratio of nutrients components expressed as carbon/nitrogen/phosphorus equal to 100/60/10 with 10/1 carbon/potassium which gave average total petroleum hydrocarbon degradation of about 395 ppm per day. Good results were obtained using the new nutrients components ratio(C/N/P=100/60/10 with C/K=10) compared to other studies applied different nutrients ratios for the same type of diesel contamination.

Key words: oil wells, drilling waste, biotreatment, fungal

Introduction

Generally, drilling wastes include formation cuttings and drilling fluids with different kinds of chemical additives depending on the type of the drilling fluid that have been used. When oil-based or synthetic-base drilling fluids are used, the discharged cuttings typically have a hydrocarbon content of 10 to 15 % along with residual weighting material. These cuttings require specialized treatment prior to the appropriate disposal.

In situations where the cuttings need to be treated, there are several possible options among of them cuttings re-injection, stabilization-solidification, landfill and bioremediation [1].

Biotreatment is a well-proven environmentally acceptable technology that uses microorganisms to biologically degrade hydrocarbon waste into nontoxic residues and reduce contaminates concentrations to acceptable levels [2].

Many microorganisms involving bacteria, fungi and yeasts have been isolated from soil contaminated with oil and studied for their ability of utilizing or producing hydrocarbons [3, 4, 5]. Many studies proved that the most of the component in oils are prone to be degraded by microorganisms, as shown in Table 1[6].

Table 1 Biodegradability of hydrocarbons in optimal conditions [6]

Crude and refined oils	Biodegradability (%)
Gasoline	100
Jet fuel	90-100
Naphtha, Condensates	90-100
Diesel oil	80-90
Fuel oil	80-85
Heavy fuel oil	10-50
Crude oil	40-70
Lubricating oil	30-75

Microorganisms need nutrients for the buildup of their cellular biomass. The favorable nutrients conditions for hydrocarbons bioremediation are often expressed as C/N/P (carbon/nitrogen/phosphorous) ratio with recommended values 100/10/1 to 100/50/10 [7]. These values with C/K (carbon/potassium) equal to 17 allow an optimal microbial growth and biodegradation of hydrocarbons [8].

In this study, the selected fungi named *Pleurotus ostreatus (Po)* (white type) and *Trichoderma harizanum (Th)* are screened for their ability in hydrocarbon degradation of drill cuttings contaminated with diesel. Moreover, these fungi have been used usefully in hydrocarbon degradation of drilled cuttings contaminated with crude oil [9].This biotreatment gave best average total petroleum hydrocarbon degradation of about 205 ppm/day in case of both fungi with 5% MCR and 5% NCR expressed as C/N/P=100/50/10.The production of different extra-cellular enzymes by fungus Po have been reported by many studies [10-13], which expected to be capable of degrading diesel infested soil. Similarly, the fungi Th producing extra enzymes and examined for its activity in such degradation.

The selected fungi can be utilized commercially for conversion of various agriculture products and wastes into fungal culture containing valuable protein. Many agriculture byproducts like cereal, straw, rice hulls, reed residues and cattail residues are available in Iraq with a dense association form occupy hundred square kilometers of land. *Pleurotus* mushroom can directly grow on these agriculture wastes involves preferential lignin and cellulose degradation due to its enzymatic activity [9].

Experimental Work

The selected drill cuttings are in the range of 40 to 60 percent by volume petroleum hydrocarbons in the diesel range (C_{12} to C_{18}). The contaminated cuttings were crushed and mixed with soil at a ratio of 1:1 (weight by weight). The sample with total petroleum hydrocarbon concentration (TPHC) equal to 19500 ppm was selected to be the base to measure the biodegradability of the present fungi.

Biotreatments of soil samples consisted of the following:

1. Un-amended soil (waste + soil),
2. Soil supplemented with fertilizers to provide a carbon/nitrogen phosphorous ratio of 100/60/10 with C/K=10, and
3. Soil supplemented with fertilizers to provide a carbon/nitrogen/ phosphorus ratio of 35/10/1 with C/K=10.

Plastic containers with a capacity of 500 gm with eight replicates for each treatment were prepared to carry out the biotreatment processes. Different concentrations of microorganisms weight ratio (MCR) were used which expressed as 6, 7 and 8 % (weight to dry weight of total mixture). Two values of nutrients weight ratio (NWR) 6 % and 8 % (weight to dry weight of total mixture) are considered at cases of nutrients addition.

Each strain of fungi was processed with the above treatments as well as a combination of both.

Moisture was maintained at approximately 20 % and the temperature ranged from 20 to 40° C. Fifty cm^3 grab samples were taken at time zero then at weekly intervals (0, 7, 14, 21, 28, 35, 42 and 49 days). Samples were analyzed for their TPHC using high performance liquid-chromatography (HPLC type Shimadzu LC-4A).

Fungi were grown on potato dextrose agar (PDA) media and maintained in slants at 4° C [13]. Preparation of inoculums and substrates had done according to many studies [9, 14] which confirmed an optimum processing condition of growth.

RESULTS AND DISCUSION

Fig. 1 illustrates the result of biotreatment technology of un-amended soil. This figure shows that TPHC decreased when MCR increased especially in the case of using both strains of fungi together. This noticeable degradation will be observed clearly with the rest treatments and the best hydrocarbon degradation was obtained with the combinations of both fungi strains at 8 % MCR which gave about 231 ppm per day. Also, it was shown that the results of fungi Th are relatively close to those of Po.

Figs.2 and 3 show the effect of nutrients addition on the present biotreatment with the selected NCR (C/N/P=35/10/1 and 100/60/10) respectively and with 6 % NWR. These results demonstrated the necessity of nutrients for the bio-process. For example, at the case of combination of both stains with 8 % MCR, an average reduction in TPHC of about 302 and 322 ppm per day was shown. This is a good result in comparison with a value of 231 ppm per day that obtained with the same conditions in the case of un-amended sample.

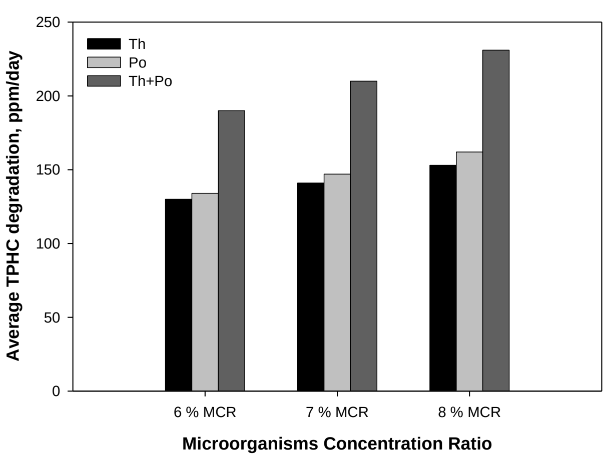


Fig. 1 Average hydrocarbon degradation response to both fungi strains and ratios without nutrients

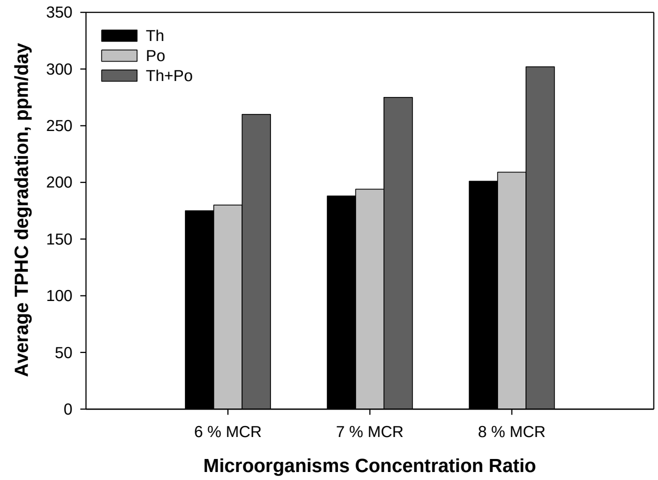
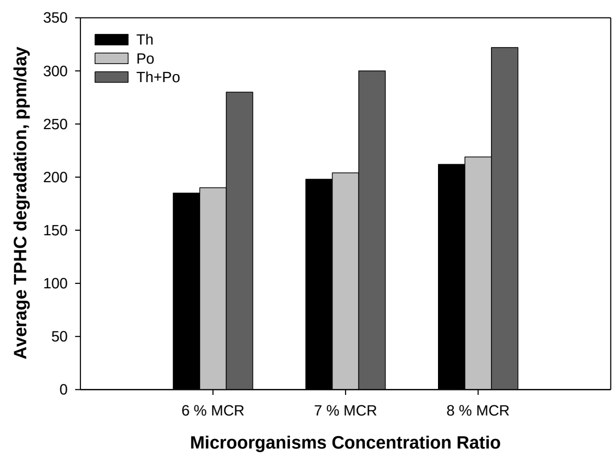


Fig. 2 Average hydrocarbon degradation response to both fungi stains and ratios with 6 % NWR and C/N/P=



35/10/1, C/K=10

Fig. 3 Average hydrocarbon degradation response to both fungi strains and ratios with 6 % NWR and C/N/P= 100/60/10, C/K=10

Fig. 4 and 5 indicate the effect of another ratio equal to 8 % NWR. Here, a significant increasing in whole degradation was shown, which explains how the increment in NWR will lead to progress in current biotreatment. The case of using both fungi together at 8 % MCR with 8 % NWR and C/N/P = 100/60/10 with C/K=10 illustrates the observed enhancement. Moreover, it can be seen that TPHC declined from 19500 ppm to less than 150 ppm during 49 days with less than 1000 ppm remaining after 30 days which appears to be a fairly typical exponential type degradation curve as shown in Fig. 6.

Fig. 7 represents a relationship between TPHC with time for selected values of MCR, NWR and NWC. The purpose of this figure is to show the significant difference in the microbial treatment between the un-amended and the nutrient amended soils. According to these results with a limit of 10000 ppm as an example, the addition of nutrients to the diesel contaminated soil could reach this value in approximately 35, 26, 21 and 14 days respectively to 44 days for an un-amended soil.

This biotreatment is an environmentally acceptable process which converts agricultural byproducts into water and carbon dioxide and results in the generation of metabolic energy and biomass.

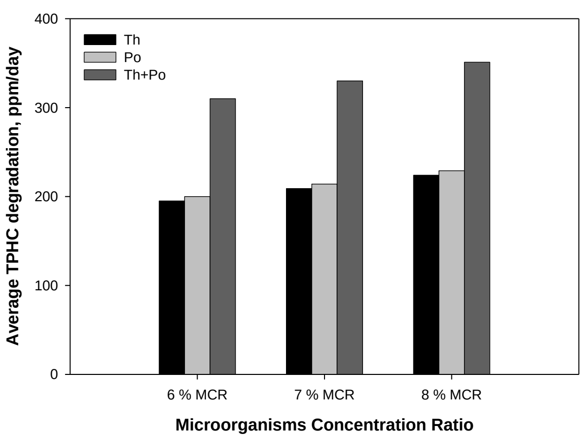


Fig. 4 Average hydrocarbon degradation response to both fungi stains and ratios with 8 % NWR and C/N/P= 35/10/1, C/K=10

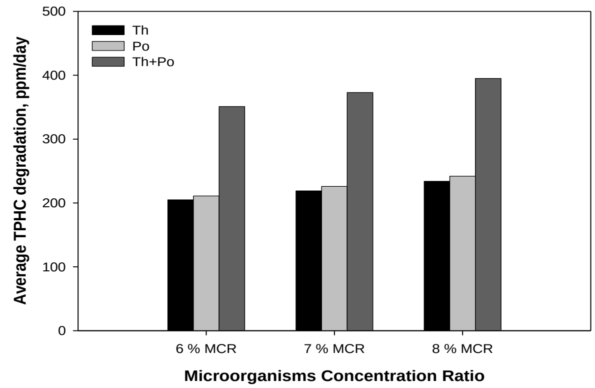


Fig. 5 Average hydrocarbon degradation response to both fungi stains and ratios with 8 % NWR and C/N/P= 100/60/10, C/K=10

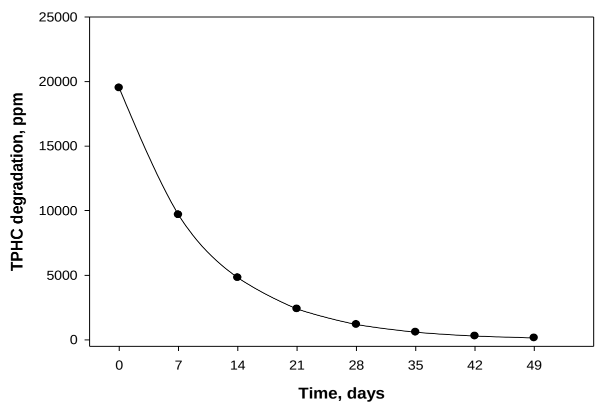


Fig. 6 Hydrocarbon degradation over time during the biotreatment of diesel contaminated soil at 8 % MCR, 8 % NWR and C/N/P=100/60/10, C/K=10

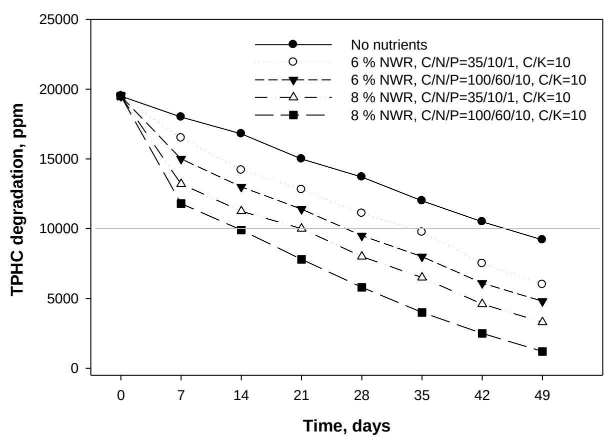


Fig. 7 Hydrocarbon concentrations in soils over time with 7 % MCR of both fungi together

CONCLUSIONS

The following conclusions were observed from this study:

1. The biotreatment results of *Trichoderma harizanum* are relatively close to those of *Pleurotus ostreatus*. It can be noticed that at the same value of MCR and with each strain of fungi, the reduction in TPHC is mostly identical.
2. Using both strains together gave better hydrocarbon degradation than alone usage.
3. Degradation in TPHC increases with an increase in MCR.
4. Degradation in TPHC increases with nutrients addition.
5. The nutrient ratios are added to make a progress in biotreatment of petroleum compounds that are difficult to degrade by the site's indigenous biota. In other words, adding nutrient amendments to diesel contaminated soil can enhance biotreatment rates rather than the case of un-amended soil.
6. The enhancing in hydrocarbon degradation with both fungi strains together is reasonable-expected matter due to the double action in producing extra-enzyme that showed the ability to treat the present contamination.
7. This study proved that the selected fungi can be considered as hydrocarbon degradation microorganisms. Also the new ratio of nutrients components which expressed as C/N/P=100/60/10 with C/K=10 can be recommended to give best microbial growth and diesel degradation.
8. The suitability of using different cheep and available agricultural waste substrate in the cultivation of the microorganisms understudy made this kind of treatment to be considered an effective treatment option for petroleum hydrocarbon contaminated soils.
9. If the limiting factors such as temperature, moisture, nutrients and C/N ratio of the compost are not adequately addressed, the resulting is a long treatment period.
10. The best reduction in TPHC (within this study) can be observed at the case of using the both fungi together, amended soil with C/N/P= 100/60/10, C/K=10, 8 % NWR and 8 % MCR, which gave an average of 395 ppm per day. In comparison with biodegradation rates reported in the literatures for other sites, this is good rate for biodegradation.

REFERENCE

1. International Association of Oil and Gas Producers "Environmental Aspects of the Use and Disposal of Non-Aqueous Drilling Fluids Associated with Offshore Oil and Gas Operations", Report 342, (2003).
2. McMillen, S.J. and Gary, N. R., "Biotreatment of Exploration and Production Wastes", SPE 27135, presented at the Second International Conference on Health, Safety and Environment in Oil and Gas Exploration and Production, Jakarta, Indonesia, January 25-27 (1994).
3. ZoBell, C. E.: "Assimilation of hydrocarbons by microorganisms", Adv. Enzymol, no. 10, pp. 443-486, (1950).
4. Beerstecher, E.: "Petroleum Microbiology," Elsevier Publishing Company, Amsterdam, pp. 375, (1954).
5. Davis, J. B.: "Petroleum Microbiology," Elsevier Publishing Company, Amsterdam, London, New York, (1967).
6. Chaineau, C. H., Jean, C. S. and Anne, M., "Is Biotreatment a Solution for The Treatment of Oily Waste?, " SPE 78548, presented for presentation at the 10th Abu Dhabi International Petroleum Exhibition and Conference, (2002).
7. Atlas, R. M., and Bartha, R.: "Hydrocarbon Biodegradation and Oil Spill Bioremediation," Advances in Microbial Ecology, 12, pp. 287-388, (1992).
8. Chaineau, C. H., Morel, J. L., and Oudot, J.: "Biodegradation of Fuel Oil Hydrocarbons in The Rhizophere of Maize, J. Environ. Qual., 29, pp. 569-578, (2000).
9. Abd Al-Razaq, A. A. Haleem, "Different Techniques to Minimize Oil Wells Drilling Wastes", Ph. D. Thesis, University of Baghdad, (2007).
10. Hassan, A. A. K.: "Production of Fibrinolytic Proteas from Pleurotus Ostreatus by Solid State Fermentation," Ph. D. Thesis University of Baghdad, (2005).
11. Bourbonnais, R. and Paice, M. G., "Oxidation Enzymes for the Lignin Degrading Fungus Pleurotus Sajor-Caju in Plant Cell Wall Polymers Biogenesis and Biodegradation", American Chemical Society, Symposium Series, 399, 472-481 (1988a).

12. Bourbonnais, R., Paice, M. G., "Veratryl Alcohol Oxidaze from the Lignin Degrading Basidion Yeasts Pleurotus Sajor-Caju", Biochemical Journal, 255, 445-450 (1988b).
13. Al-Jaboury, M. H., Hassen, A. A. and Alani, S. R., "Production of Fibrinolytic Protease from Various Fungi Isolates and Species", (in press).
14. Quimio, T. H., Chang, S. and Royer, D. J., "Technical Guidelines for Mushroom Growing in the Tropics", FAO Plant production and Protection, paper 106, Rome, Italy, (1990).