Removal Of Petroleum Hydrocarbons From The Oil-Contaminated Marine Environment For Sustainable Aquaculture And Hatchery Uses

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Introduction

Malaysia is situated on the international maritime trade route (Chua et al., 2000). Busy navigational traffic and rapid modernization activities in the four territorial states of the Straits of Malacca have risked the marine environment to the threat of oil pollution. Hydrocarbons are extremely toxic to a wide spectrum of aquatic organisms (Law et al., 1997; Law et al., 2002). Law (1997) suggested a safety level of 50 μg L^{-1} water soluble fraction for protecting *P. monodon* post larvae in the marine environment. The notorious impacts of hydrocarbons in fact triggered the research and development of oil combating techniques. Ever since late 60s', techniques have been actively developed to combat oil pollution; however there is still no sufficient technique (Clark, 1997). Most of the techniques are either destructive to the environment or it is too costly for implementation especially for the aquaculture farmers in the coastal areas. Therefore, this project was undertaken to develop efficient and affordable oil removal techniques as alternate treatments for removing hydrocarbons from the water and sediment of the marine environment for sustainable usages of coastal aquaculture. A water filtering system with an integrated water-filtration cartridge that consisted of selected local materials was developed to remove water-soluble fraction of crude oil in the oil-contaminated seawater. Excellent adsorption possessed by the local materials enable a novel cartridge to be produced in this study. As for the sediment, biodegradation was studied for degrading hydrocarbons contain in the oil-contaminated marine sediment. Where, active oil-degrading bacteria were isolated from the coastal environment and their ability to degrade petroleum hydrocarbons was evaluated. Results revealed that, application of the inoculation alone may not be efficient enough because biodegradation may be retarded by interspecies competition and other inhibiting compounds produced during bacterial metabolisms. A dual-stage treatment is studied to improve biodegradation in the sediment. Results of the study show that both water filtration and dual stage biodegradation techniques are capable of reducing hydrocarbons in the water and sediment down to their respective safety level. Efficiency of the techniques was further confirmed by bioassay assessment conducted on *P. monodon* post larvae.

Materials and Methods

Water-filtering system. Treated local materials, coconut husk, shrimp waste, sand and granular activated carbon were examined for their hydrocarbons adsorption. Compounds adsorbed by different materials were determined by using GCMS. Where as, hydrocarbons adsorption was quantified by using a portable fluorometer (Parsons et al., 1984; Law et al., 2002). Based on the adsorption of the hydrocarbons compounds, an integrated water-filtering cartridge consisted of the local materials were developed based on their noble ability to adsorb hydrocarbon by the materials. Later, filtering capacity, hydrocarbons compounds adhesions and optimal filtration rate of the integrated cartridges were determined. Freundlich equation was utilized for assessing the filtration rate and the maximum capacity of the filtration cartridges. Bioassay of the filtrate was conducted on *P. monodon* post larvae to assess efficiency of the integrated cartridges. Bioassay technique as described in Law and Yeo (1997) was used to verify filtrate safety. *Dual stage biodegradation.* A microbial consortium that consisted of three isolated bacteria was employed in this study. AR3, a *Pseudomonas* bacterium is used to remove aromatic hydrocarbon. Where as, OG1 and OG2 were used to remove straight chain hydrocarbons and organic matters respectively. The active oil degrading bacteria were isolated from the coastal sediment of Port Dickson and Marang respectively. The study was conducted on shrimp pond sediment collected from shrimp farms located in Setiu and Marang, Terengganu. The organic rich sediment was then contaminated by using Tapis A crude oil at 1500 mg kg^{-1} and it was treated by using bacterial consortium isolated from Malaysia coastal environment. Efficiency of oil degradation by using single stage, dual-stage and three-stage were conducted. Three-stage treatment is an attempt to improve performance of the dual stage treatment. Hydrocarbon content in the sediment was monitored quantitatively by using fluorescence-spectrometric technique where as, qualitative analysis was conducted by using GC-FID with MSD confirmation (UNEP, 1992). Later, bioassay was conducted on *P. monodon* post larvae to assess safety of the treated sediment. Dual-stage is a combination of two conventional treatments with a refreshing stage in between of the treatments.

Results and Discussion

*Water filtering system*All absorbents used in this study demonstrated high ability in adsorbing aromatic hydrocarbons in seawater at salinity 30 ppt, pH 8 and temperature 30°C. They follow the Freundlich absorption isotherm equation closely with a first order reaction. The maximum adsorption of aromatic hydrocarbons by coconut husks, palm press fiber, granular activated carbon, shrimp shell and marine silts are 1.81 mg/g, 1.57 mg/g, 1.35 mg/g, 1.21 mg/g and 1.19 mg/g respectively. In fact, all of them have the ability to reduce aromatic hydrocarbons in seawater from 10 mg/L to 0.05 mg/L which is safe for hatchery operation. Chromatography results revealed that most of the USEPA listed priority aromatic hydrocarbons are absence in the filtrate. Furthermore, results of the bioassay revealed that, the filtrate is safe for hatchery and aquaculture uses as *P. monodon* post larvae showed good survival in the experiment.

Dual stage biodegradation: The most common post-harvest cleaning procedure practiced in Malaysian aquaculture is washing, flushing and exposed the pond bottom for air drying. Aquaculture sediment becomes more complicated when hydrocarbons contaminated the pond sediment. Results revealed that, the techniques are insufficient to remove hydrocarbons content in organic rich pond sediment. High organic substances content in the pond sediment acted as alternate carbon sources for oil bacteria. Masking effect of the organic substances on hydrocarbons in the pond sediment has retarded degradation of hydrocarbons by oil bacteria. Results of the study revealed that, only 102 mg kg^{-1} Tapis A crude oil (6.7% of hydrocarbons) was removed from the sediment compared to 637.3 mg kg^{-1} of Tapis A crude oil removal in sediment which without masking effect of organic substances in sediment. Bacteria utilize organic and hydrocarbons as their carbon source for growth. As organic carbon and hydrocarbons are degraded by the microbes, biomass in the pond sediment increased rapidly and this leads to intense inter-species and intra-species competition. Besides, accumulation of secondary metabolites in the sediment may cause retardation on organic carbon and hydrocarbon degradation. When these toxic substances were accumulated up to a certain level, it will retard and even shut down biodegradation processes. Thus, a multiple stages treatment is innovated to overcome the problems. Multiple stages treatment is basically a combination of two or more conventional biodegradation (single stage). Dual stages biodegradation is sufficient in degrading organic carbon and hydrocarbons in the pond sediment. Dual stage treatment removed 966.5 mg kg-1 Tapis A crude oil and 25.18 mg C g^{-1} organic carbon in sediment containing 1500 mg Tapis A crude oil kg-1 and 25 mg C g^{-1} organic carbon. The conventional single stage treatment only degraded 637.3 mg kg^{-1} Tapis A crude oil from the oil contaminated sediment for a period of 20 days. Three stages treatment was attempted to further improve organic carbon and hydrocarbons degradation, however the amount of organic carbon and hydrocarbons degraded by three stages treatment did not reveal significant improvement. Certain recalcitrant compounds; benzo(a)pyrene, anthracene, chrysene and pyrene were removed from the sediment by the dual-stage treatment. Efficiency of the dual stage treatment in removing adverse effect of organic carbon and hydrocarbons in the pond sediment was further affirmed by the bioassay assessments on P. monodon post larvae. Results revealed that, dual stage treatment was able to remove adverse effect of hydrocarbons in pond sediment. Maximum growth rate of P. monodon post larvae in water in contact with dual stage treated sediment for a period of 45 days was 0.743±0.015 g day^{-1} and survival rate of 77%. These results are similar and not significantly different from that of the organic and hydrocarbons free sediments. These results are much higher than the untreated hydrocarbons contaminated pond sediment. Hydrocarbon pollution revealed a serious effect on P. monodon post larvae. Results of the bioassay shows that there is no survival of tiger shrimp post larvae in the 1500 mg kg^{-1} crude oil contaminated pond sediment. Dual stage treatment is able to remove the adverse effects of oil on *P. monodon* post larvae. Based on the growth rate and survival of tiger shrimp post larvae on the oil polluted pond sediment, Dual stage treatment is able to increase the yield in the oil polluted pond sediment.

Conclusions

Local materials such as coconut husk, shrimp waste and palm press fiber exhibited excellent hydrocarbons adsorption. An efficient and cost-effective filtration system could be developed by using these materials which are agricultural wastes. This filtering system not only recycling the waste, it also capable of removing major toxic hydrocarbons contained in the water soluble fraction of crude oil. Safety of the filtrate was proven by positive results of the bioassay on *P. monodon* post larvae. This filter cartridge could be applied on coastal aquaculture and marine hatchery. Biodegradation of hydrocarbons in pond sediment is a complicated process as high organic matter in the sediment revealed another carbon sources for oil-degrading bacteria. Application of the dual stage biodegradation is able to improve biodegradation of hydrocarbons in the sediment and hence improve yield in the pond sediment. It is estimated that a gross profit of RM 152,378 will be obtained. It is believed that, the filtration system and the dual stage treatment is able to safe-guard safeties of pond water and sediment from oil-pollution.

Benefits from the study

This study provided a solid background for the substrate selection in the development of an efficient and cost-effective cartridge by using local materials to remove hydrocarbons from the seawater. The local materials could act as add-on substrates in present filtration cartridges. In fact, the filtering system could also applied on the waste and effluent discharged from the petrol-chemical industry. It is believed that, the filtration system could further reduce the level of COD discharged from the industry which is one of the priority concerns of DOE. The dual-stage treatment is able to remove both organic and hydrocarbons level in pond sediment. This help in reducing the occurrence of disease outbreak in aquaculture pond. Good sediment condition after treatment could promote growth and survival of cultures in the pond. Besides, the dual-stage treatment is able to reduce the long term effects of oil on the culture in the oil-contaminated sediment. As the dual-stage treatment is able to boost bacterial degradation in the sediment, it could be applied on treating domestic waste which is rich in organic content and other hazardous compounds such as PCBs in the treatment plants.

Patent(s), if applicable:

Nil

Stage of Commercialization, if applicable:

Nil

Project Publications in Refereed Journals

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Graduate Research

Name of Graduate	Research Topic	Field of Expertise	Degree Awarded	Graduation Year
Hii Yii Siang	Biodegradation of organic carbon and hydrocarbons in aquaculture sediment: a dual stage treatment system	Marine pollution	Ph. D.	Pending
Mok Mun Loong	Treatment Of Petroleum Hydrocarbon Residues In Seawater Using Local Materials.	Marine pollution	M. Sc.	2003

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