Treatment of Diesel Oil Contaminated Soil by Ex-situ Bioremediation

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Abstract—**Treatment of oil-polluted soil is a challenging problem faced by all refineries and petrochemical industries. In this research study, bioremediation of diesel oil contaminated soil was conducted for diesel concentration ranging from 5% to 20%. The physicochemical characteristics of diesel oil contaminated soil were studied. The effects of soil amendments, namely coconut ash powder, biofilter activated sludge, and NPK fertilizer, on total petroleum hydrocarbon removal efficiency were studied. The maximum total petroleum hydrocarbon removal efficiency achieved was 94.5% when 4g NPK, 40g of activated sludge and 40g of coconut ash powder per 1000g of contaminated soil were used. The studies on the effect of temperature confirmed the optimal temperature as 35°C. The parametric studies confirmed that the degradation efficiency decreased with increase in diesel oil concentration.**

Keywords-bioremediation; fertilizer; sludge;diesel;efficiency

I. INTRODUCTION

Pollution caused by oil spills is a critical problem occurring near petrochemical plant sites. Treatment of oil contaminated soil can be handled by physical, chemical or combined techniques. The effectiveness of these techniques highly depends on the hydrocarbon fractions present and their solubility properties. Bioremediation of soils contaminated by hydrocarbons is recognized as an alternative method having advantageous characteristics. Bioremediation technologies either in-situ or ex-situ utilize the natural potential of microorganisms to degrade complex contaminants [1-3]. Different organisms are employed using various techniques of bioremediation according to the hydrocarbons present in the contaminated soil. As the composition of each type of oil is unique, there are different ways to deal with them through microbes and flora. Bioremediation is operated naturally or with addition of microbes and fertilizers [4-6]. The microorganisms which can decompose or transform the chemical substances present in petroleum and petroleum derivatives are enriched though the optimal supply of nutrients. Spilled crude oil affects physicochemical properties of the soil such as temperature, structure, nutrient status and pH. The successful implementation of a bioremediation process usually needs the application of strategies customized for the specific environmental conditions of the contaminated site [7, 8]. Crude oil forms a layer on the surface of the soil and this oil film acts as a physical barrier between air and soil thereby causing a breakdown of soil texture followed by soil dispersion [4]. The light hydrocarbon fractions of the crude oil evaporate while the greasy fractions permeate slowly into the soil and are slowly biodegraded by microbes which naturally inhabit the soil [5]. Crude oil spills of diversified composition are considered as a difficult substrate mix as the microorganisms could not have resistance for all the pollutants. The objectives of this study are diesel oil contaminated soil treatment and the study of the effects of relevant variables on the bioremediation efficiency.

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II. MATERIALS AND METHODS

A. Chemicals

The components used in the bioremediation of crude oil contaminated soil are organic manure composed of activated sludge, coconut powder shell and inorganic fertilizer. The inorganic fertilizer (NPK-15-15-15) which is composed of nitrogen, phosphorus and potassium (K) was purchased from a local agro-chemical shop.

B. Preparation of Contaminated Soil

Soil samples were collected from a site near Sohar port and were screened by sieving to 2mm to remove heavier fractions. Twenty kg of sieved soil were weighted and distributed equally in 20 plastic boxes, each containing 1kg of soil. Subsequently, the 20 boxes were divided into 4 groups of 5 boxes each. Samples of known quantity of diesel oil were weighed and well mixed with the soil in the first box to prepare the 5% desired concentration. Similarly, the other samples of desired concentrations were prepared. A sample of activated sludge from the biofilter used for treating benzene was taken and used as a microbial source. The isolated sludge was dried in an oven to evaporate the remaining water and crushed well to fine powder. The coconut shells were collected from a nearby farm and dried for 3 weeks in sunlight. The dried coconut shells were burned until they were totally converted to ash. The coconut shells ash was sieved to obtain fine particles of 2mm size.

C. Ex-Situ Bioremediation Procedure

The soil samples were dug to a depth of 0-15cm. The excavated top soil, unlike the activated sludge and coconut powder shell ash was sun dried for three weeks and ground using a mortar and a pestle. The samples were sieved using a 2mm standard mesh sieve and were measured using by an electronic weighing balance. This remediation experiment was carried out in a well perforated 1.5lt plastic container with an estimated depth of 6cm. Crude oil concentration was prepared at levels of 5, 10, 15 and 20% (by mass). The individual cells were mixed with the aid of a stirrer. Stirring was conducted every four days for eight weeks. During this period, the samples from the 20 labeled cells were taken to the laboratory for analysis of total hydrocarbon content, once in a week. The physicochemical properties and total petroleum hydrocarbon (TPH) were analyzed using standard methods [9, 10].

III. Results and Discussion

A. *Physicochemical Properties of Contaminated Soil*

The physical and chemical properties of the contaminated soil are presented in Table I. The soil samples were analyzed and found to have acidic properties with moderate moisture content.

TABLE I. CONTAMINATED SOIL PHYSICOCHEMICAL PROPERTIES

Parameter	Value
pH (water/soil=5/1)	5.80
Moisture content (%)	17.40
TOC (%)	14.20
TPH (mg/kg)	38425.25

B. *Effect of Coconut Shell Powder on TPH Removal Efficiency*

In this set of experiments, the influence of coconut shell powder addition on TPH removal efficiency was studied by adding coconut shell powder in the ratio of 40g/1000g contaminated soil. The experiments were repeated with samples with 5% to 20 % diesel oil concentration. The experiments were conducted at ambient temperature of 34°C. The results obtained from the experiments are presented in Figure 1 and indicated that TPH removal efficiency decreased as diesel oil concentration increased. The microbial culture present in the soil degraded 5% diesel oil polluted soil faster and the maximum removal efficiency achieved was 49.1%. The TPH removal efficiency of the experiments conducted with 10% diesel oil polluted sample was 35.6%. The removal efficiencies achieved with 15% and 20% samples were very low due to the excessive pollutant load on the microbial community. Above a certain concentration, diesel oil is highly resistant to biodegradation. The importance of soil amendments is described in [12].

C. *Effect of Activated Sludge and Coconut Shell Powder on TPH Removal Efficiency*

In this set of experiments, the effect of the addition of two soil amendments, namely biofilter activated sludge and coconut shell powder, on diesel oil removal efficiency was studied by adding equal quantities of the two components in the contaminated soil samples. The experiments were conducted with 1000g soil samples polluted with 5% to 20% diesel oil concentration. The data from Figure 2 describe that the TPH removal efficiency increased due to the addition of biofilter activated sludge. An improvement of approximately 10% was observed due to the increased microbial population present in the activated sludge. The biofilter activated sludge contained microorganisms acclimated to hydrocarbons and contributed to better removal percentages. But, the efficiency decreased when the diesel oil percentages increased. The TPH removal efficiency was around 50% in the diesel oil concentration range 5%-10% and later on decreased to 36.9% which could be due to the excessive pollutant loading resistance to microbial degradation.

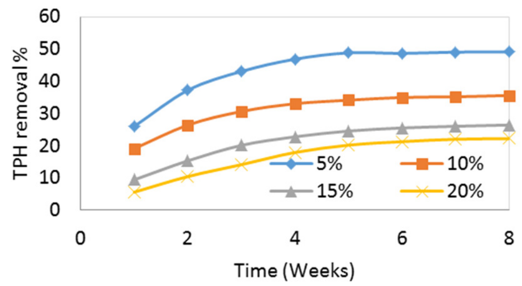

Fig. 1. Effect of coconut shell powder addition on TPH removal

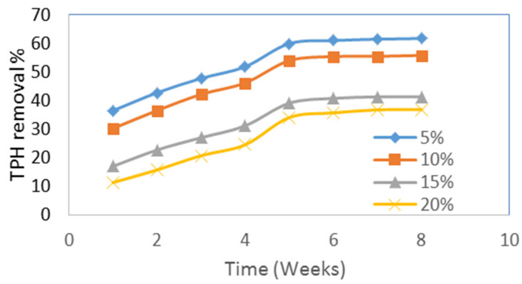

Fig. 2. Effect of activated sludge and coconut shell powder addition on TPH removal

D. *Effect of NPK Fertilizer on TPH Removal Efficiency*

The influence of addition of NPK fertilizer on TPH removal efficiency was studied by adding 4g of NPK fertilizer to the contaminated soil samples. The results shown in Figure 3 indicate that the addition of NPK fertilizer to the contaminated soil samples enhanced the removal efficiency compared to the previous experiments on coconut powder and sludge addition. The maximum removal efficiency achieved with 5% and 10% diesel oil contaminated soil samples was 74.9% and 51% respectively. The TPD removal efficiency achieved with 15% and 20% diesel oil samples was 27.2% and 14.2%, respectively. The lack of organic feeding material was reported to limit the oil degradation [13, 14].

E. *Effect of Activated Sludge, NPK and Coconut Shell Powder on TPH Removal Efficiency*

In this set of experiments, the influence of addition of three additives on total petroleum hydrocarbon removal efficiency was studied by adding 40g of activated sludge, 40g of coconut shell powder and 4g of NPK fertilizer to 1000g of contaminated soil in 4 different samples which had 5% to 20% diesel oil concentration. The experiments were conducted under an average ambient temperature of 34°C. The obtained results, shown in Figure 4, inferred that the addition of these 3 amendments resulted in high TPH removal efficiency for the 5% and 10% diesel oil concentration samples. The percentage of removal efficiency decreased as diesel oil concentration increased. The 5% sample showed the highest removal efficiency of 94.5%, followed by 79.5% by the 10% sample. However, the removal efficiency achieved with the 15% and 20% samples was less than 50% due to higher diesel oil concentration. Studies on bioremediation of diesel oil-contaminated soil using bio stimulation reported similar results [15].

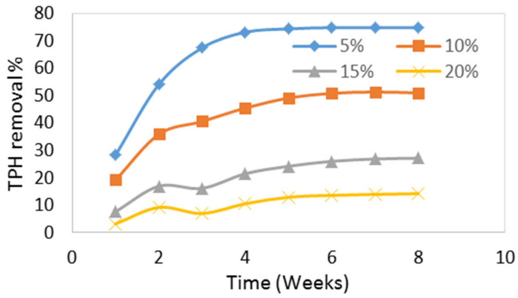

Fig. 3. Effect of NPK fertilizer on TPH removal

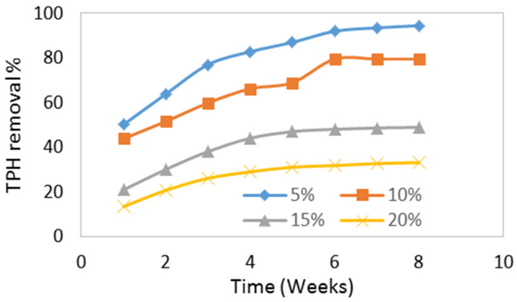

Fig. 4. Effect of activated sludge, NPK fertilizer and coconut shell powder on TPH removal

F. Temperature Effect on TPH Removal Efficiency

Temperature is reported as a controlling factor for biodegradation of petroleum hydrocarbons. Extreme temperatures inhibit the enzymatic functioning of most microorganisms and will also reduce the action of enzymes in a diesel oil spilled environment. With decrease in temperature, the rates of degradation decrease probably because of the decreased rates of enzymatic activity and increased oil viscosity. Temperature also affects the solubility of hydrocarbons. Although hydrocarbon biodegradation can occur over a wide range of temperatures, the rate of biodegradation generally decreases with decreasing temperature. The effect of temperature on TPH removal was studied with 10% diesel oil contaminated soil over a period of 8 weeks. The highest removal efficiency achieved was 89.5% when the temperature was 38°C and the TPH removal efficiency decreased to 79.5% at 35°C. At the highest temperature of 40°C, the TPH removal efficiency decreased to 68.5%. The decrease in efficiency at higher temperatures was due to the thermal resistance and inhibition to microbial action. The effect of mesophilic temperature on TPH removal was studied and 70% removal was reported after 12 weeks [12].

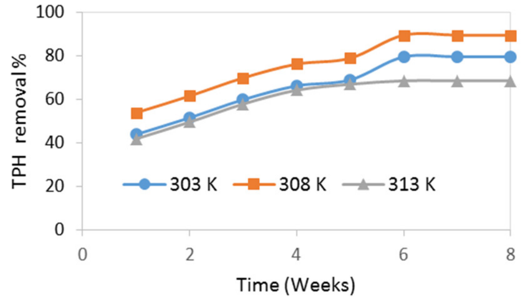

Fig. 5. Effect of operating temperature on TPH removal.

IV. Conclusions

This study investigated and proved that bioremediation of diesel oil contaminated soil is feasible with nutrient addition. The contaminated soil amended with activated sludge, NPK fertilizer and coconut shell powder showed the highest percentage (99%) of total petroleum hydrocarbon degradation when 5% diesel oil contaminated soil was used. With increase in diesel oil concentration, the degradation efficiency decreased. The findings of this study showed that the rate of biodegradation depends majorly on soil nutrient availability. Both organic and inorganic fertilizers proved to enhance the multiplication of indigenous microbes thus enabling rapid biodegradation of contaminants in the soil.

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