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A Study on Combination of a Chemical and Biological Method for Oily Sludge Biodegradation

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Abstract:

Oily sludge is a waste product of petroleum industries or oil refineries and poses hazardous effect on the environment if released untreated. Treating and disposing such waste product is of major concern and created much difficulties. The several mechanical, chemical and biological ways of treating the wastes are incineration, stabilization/ solidification, oxidation and bio-degradation (bioremediation), etc. These methods have their own advantages and disadvantages. This review is based on co-treatment of the oily-sludge using a chemical method(oxidation) and a biological method(bioremediation). These two methods when combined, considerably increases the percentage of degradation of the oily sludge and decreases the time taken for bioremediation.

Keywords: Oily sludge, oxidation, bioremediation, co-treatment.

1. Introduction:

When oil, solid and water mixes together and precipitates out, it is known as the oily sludge. It is an emulsion of water, solids, petroleum hydrocarbons and metals [1]. Huge amount of oily sludge are produced by petroleum industries and causes hazardous effects on the environments such as: physical and chemical changes of natural habitats, harmful effects on marine habitat and ecosystem etc [2]. The contaminant oil from the oily sludge dumped on the soil may percolate through the soil and leach out thus contaminating the ground water [3]. Oily sludge contains many components which are carcinogenic and toxic to immune system [3]. The components of oily sludge includes Polycyclic aromatic hydrocarbon(PAH), aliphatic hydrocarbon, NSO(Nitrogen, sulphur, oxygen) compounds and asphaltenes[3]. Almost 75% of the petroleum hydrocarbons are aliphatic and aromatic[1]. Polycyclic aromatic hydrocarbons present in the oily sludge, when comes in contact with humans may cause skin problems such as erythema and cancer, bladder, gastrointestinal and sinonasal cancer,etc. [2]. Oil contamination also contains substances like toluene, xylene, benzene, ethyl benzene which cause severe effects on human such as mutations, birth defects, liver disease, depression etc. [1]. The soil

that are contaminated by oil becomes infertile [2]. The general methods which were used earlier for oily sludge disposal like landfill, incineration and bioremediation have some limitations which cannot be ignored[4]. Landfill associates foul odour, soil and ground water contamination, incineration produces toxic gases in huge amount and bioremediation takes long time [4]. However, the biodegradability could be enhanced by a method called oxidation [4]. The organic components of the oily sludge is degraded by the process of oxidation with the help of either a chemical or a oxidizing agent[5]. The oxidizing agent is added to the oily sludge which oxidizes the organic components present in the sludge to form carbon-dioxide, water or non-hazardous materials [5]. Oxidation degrades the organic part of the oily sludge by cleavage of large organic compounds to smaller ones in a short period of time and makes the waste easily biodegradable which takes less time than usually required for bioremediation[6]. In a study by Veronica C. Mora et al. [1], bioremediation of oily sludge contaminated soil was studied preceded by oxidation step using persulfate and permanganate as the oxidizing agents. These two oxidizing agents has certain desirable characteristics like high solubility in aqueous solution, high temperature stability at room temperature and comparitively cost effective[1]. Bajagain et al.,2018a [7], Lemaire et al., 2019 [8], and Peluffo et al.,2018[9] in their study revealed that persulfate usage as an oxidizing agent showed excellent petroleum hydrocarbon removal from soil. According to Bajagain et al., 2019[10], several types of organic components of the oily sludge can be degraded using permanganate as the oxidizing agent owing to its high redox potential and slow rate of decomposition. Also due to presence of unmanageable components of oily sludge which cannot be degraded by bioremediation due to several factors like low hydrocarbon availability, microbial community reduction due to changed soil condition subject to oily sludge dumping, efficiency of bioremediation decreases [11].

So, combination of oxidation and bioremediation can be used to overcome the problems arising due to the the individual methods.

In the work of Veronica C. Mora et al.,2020 [1], they combined oxidation with bioremediation to increase the degradation of the waste sludge. The study is mainly done to establish the effectiveness of the usability of aged contaminated soil earlier used for farming to degrade components of oily sludge. Indigeneous bacterial population are expected to be present in the weathered soil used for oily sludge bioremediation. The method employed in this work are as follows

2. Materials And Methods

2.1. Collection of oily sludge

Oily sludge was collected from the bottom of tank storing crude oil, from a petroleum company and was stored at 5^{o}C.

2.2. Sampling of weathered (Aged contaminated) soil

Soil samples were collected from a site contaminated with hydrocarbons and is located near a petrochemical plant. In this site, landfarming treatment was done some years ago, contaminated with

petrochemical waste. The sample was sieved and kept at 4^{o}C.

2.3. Characterization of Physical and Chemical properties

The physical and chemical properties of the oily sludge and the contaminated soil was characterized and listed. Methods are elaborated in the paper [1]. The parameters are moisture content, pH, redox potential(Eh), total organic carbon, culturable heterotrophic bacteria, Polyaromatic hydrocarbon–degrading bacteria(PAH-DB), Aliphatic hydrocarbon-degrading bacteria(AH-DB), PAH and AH.

2.4. Oxidation Treatment

10 g of oily sludge was mixed with 20ml of aqueous phase containing

1) Persulfate (Sodium persulfate, $FeSO_4.7H_2O$ and EDTA)
2) Potassium permanganate 1
3) Potassium permanganate 2

All these reactions were carried in a closed glass container kept in dark, shaken continuously and incubated for 7 days at room temperature. Water without oxidant was kept as control.

2.5. Co-treatment with Oxidation and Bioremediation

a) Biological degradation of oxidized oily sludge (Box):- Oily sludge mixed with aqueous permanganate solution was kept in dark, incubated for 7 days at room temperature and shaken continuously. After that, it was mixed with the soil contaminated with hydrocarbons.

b) Biological degradation of Oily Sludge (BOS):- Oily sludge was mixed with hydrocarbon contaminated soil.

These were incubated at 30^{o}C for 127 days in the dark, aerated regularly and moisture content was maintained between 25% and 30% w/w by adding ultrapure water.

After treatment, the samples were collected after 1 day, then every 7 days of 1st month of treatmentand then after 43, 57, 92 and 127 days.

2.6. Enzymatic Assay

Lipase and dehydrogenase assays were performed because activity of lipase indicates hydrocarbon biodegradation and activity of dehydrogenase indicates total soil microbial activity.

3. Results

3.1. Oxidation Treatment

Degradtion of the PAH and TPH present in the oily sludge using three types of oxidants (PS,PM1,PM2) were compared and according to the previous studies, it was found that Permenganate 1 was able to degrade the most and hence it was choosen for co-treatment.

3.2. Co-treatment with oxidation and bioremediation

The three main PAH found in oily sludge were phenanthrene(PHE), chrysene(Chry) and benzo(a)pyrene (BaP) and their initial concentration was measured one day after mixing oily sludge and contaminated soil is Box and BOS. Initial concentration of PHE, Chry and BaP in Box was lower than BOS as in Box permanganate pretreatment was done. Box showed almost zero values for PAH after 24th day whereas BOS showed 25±4 mg/$Kg_{Dry\ soil}$ concentration

after the treatment ends. Not much difference in values was observed for aromatic hydrocarbons(AH) for BOx and BOS at the end of the treatment. For TPH, at the end of the treatment, final concentration with BOx was much lower than the final concentration with BOS. For BOx and BOS, there is not much difference in the heterotrophic bacterial count. An increase in the PAH- degrading bacteria for both BOx and BOS was seen for first was seen for first 22 days and then it increased significantly for BOx till the end of the treatment. The AH- degrading bacterial count was same for BOx and BOS for the first 8 days of incubation but after that the count decreased for BOS maintaining significant difference from BOx till the end.

3.3. Enzymatic Analysis

For lipase activity, the value for BOx kept on increasing till day 8 and then it lessened and couldnot be detected at day 29. The value for BOS decreases between 15 to 22nd day, then increases till day 43 and then again decreases till the end of the treatment.

For dehydrogenase activity, the value for both BOx and BOS reached maximum during 15th to 22nd day. This made clear that adding oily sludge resulted in an increase in the biological activity after an initial lag phase. BOx showed better bioremediation efficiency than BOS due to formation of intermediate substances as a result of oxidation which made them water soluble and increasing their bioavailability. Also organic matter oxidation helped in nutrient mobilization in soil.

4. Conclusion

From this study it was evident that oxidation treatment with permanganate prior to bioremediation improved the biodegradability of hydrocarbons present in oily sludge and contaminated soil due to oxidation of organic components. Microbial increment in BOx also suggested that oxidation pretreatment supported the growth of microbial communities needed for bioremediation. Two months of incubation was sufficient to decrease the contamination level for both oily sludge and contaminated soil.

All these results suggests that combination of oxidation and bioremediation for treatment of petroleum wastes, is a very promising way of waste treatment and can be employed by industries to lower the environmental impact of petroleum production.

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