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The Remediation of Waste Drilling Muds by a Combined Plant-microbe System

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In the present study, waste drilling muds from an active drilling operation were used to evaluate the feasibility and efficiency of bioremediation in the laboratory during a one-year trial and following a phytoremediation treatment. The results showed that total organic carbon (TOC) content in the muds decreased from 89800 to 21700 mg/kg after 48 h of biotreatment in a slurry reactor, and the chroma removal efficiency reached 81.5%. TOC contamination level was decreased by 84.6% after 240 days of phytoremediation.

Keywords: bioremediation, drilling muds, petroleum, phytoremediation, total organic carbon

INTRODUCTION

Drilling muds are used to lubricate and cool down the drill bit during petroleum drilling operations and also to carry the drill cuttings to the surface for further screening and disposal. The drilling muds are broadly classified into three groups: (a) water-based muds, principally aqueous solutions of polymers and clays in brines with different types of solids and additives; (b) oil-based muds, invert emulsions of brine into an oil phase stabilized by surfactants; and (c) synthetic-based muds (SBM), synthetic material like polyesters and vegetable esters are used as its continuous phase (Coussot et al., 2004).

Drilling muds are very complex fluids and contain numerous components such as mineral, oil, organics, and additives (viscosifiers such as clays, polymers, cellulose, xanthan gum, and guar; weighting agents such as barytine and carbonate; filtrate reducers such as starch, carboxy methyl cellulose, and resins; and clays swelling inhibitors such as KCl and glycol; Coussot et al., 2004). Wastes associated with SBM contain a certain amount of pollutants and heavy metals, which may pose risk when discharged into the environment. Disposal of drilling wastes constitutes one of the most significant waste discharges associated with oil well drilling.

Bioremediation is currently one of the most widely used and cost-effective treatment technologies for oil-contaminated water and soil (Huesemann et al., 2002; Lu et al., 2009; Lu et al., 2010a). Phytoremediation is defined as the use of plants to remove, contain, or render harmless environmental contaminants (Cunningham and Berti, 1993). Phytoremediation has some advantages over other...
commonly used remediation techniques such as less disruptive of the environment than other methods, no need for disposal sites, stimulating microbial activity, and the establishment of deep-rooted vegetation helps to stabilize soil (Mackova et al., 1997; Lu et al., 2010b). Further, plants may survive higher concentrations of hazardous wastes than many microorganisms used for bioremediation.

As SBM consists of numerous different components, it is very difficult to define the mechanisms involved in their degradation. It has also been suggested that the pollutants rather than the geographical origin of the soil sample were more important in determining the functional or species diversity within bacterial communities (Maila et al., 2006). Even though the positive effects of combining plants with microorganisms have been displayed in many other studies, there are few studies on combined remediation of drilling wastes. It is uncertain if microorganisms and plants will work well on the specific environment such as drilling wastes. Therefore the objective of this study is to test the feasibility for remediation of waste drilling muds by using a combination of green plants and different microorganisms under laboratory conditions over a two-year experiment, and to realize a better understanding of mechanism of bioremediation process of drilling muds.

**EXPERIMENTAL**

**Drilling Muds Collection and Characterization**

Used drilling muds to be tested were obtained from an active drilling operation in Sichuan, China. The base fluid used in the original drilling mud was polysulfonate potassium drilling fluid system. The drilling muds on site were stored in a storage pool. A little water was precipitated from the surface of muds. The muds have the characteristics of sticky dark-brown fluid and offensive odor. Samples were stored in uncontaminated polyethylene containers and immediately transported back to the laboratory and maintained at 4°C until used.

The main physical, chemical, and biological characteristics of the studied sample are shown in Table 1. The pH and electrical conductivity of drilling muds were measured in 1/10 (dry muds/water) suspensions using a digital pH meter and an electric conductivity meter, respectively. Total carbon and total organic carbon (TOC) were quantified by using the Walkley Black method (Nelson and Sommers, 1982). Total nitrogen was extracted by Kjeldhal digestion, and determined in the form of NH₄-N by the indophenol blue method (Lu, 2000). Total petroleum hydrocarbon (TPH) was determined by USEPA Method 1664A (modified from EPA 821/B-94–004b) using n-hexane as the extraction solvent (Juteau et al., 2003). The extracts were rotary dried and dehydrated with

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>9</td>
</tr>
<tr>
<td>Density, g/cm³</td>
<td>1.71</td>
</tr>
<tr>
<td>Electrical conductivity, μS/cm</td>
<td>1050</td>
</tr>
<tr>
<td>Moisture content,%</td>
<td>31.3</td>
</tr>
<tr>
<td>TPH, mg/kg</td>
<td>15300</td>
</tr>
<tr>
<td>Cl⁻, mg/kg</td>
<td>1980</td>
</tr>
<tr>
<td>Total carbon, mg/kg</td>
<td>106200</td>
</tr>
<tr>
<td>Total organic carbon, mg/kg</td>
<td>89800</td>
</tr>
<tr>
<td>Total nitrogen,%</td>
<td>0.05</td>
</tr>
<tr>
<td>THBC, CFU</td>
<td>5.24 × 10⁶</td>
</tr>
</tbody>
</table>
anhydrous sodium sulfate. Total heterotrophic bacterial counts (THBC) were determined by spread plating method (Zubber, 1994). All measurements were done in triplicate.

Bioremediation Experiment

All bioremediation experiments in this case were carried out in a series of identical stainless steel-made vessels, which have a total volume of 100 L. Each reactor received 50 kg of drilling muds (oven-dried basis). A mechanical mixer set at 600 rpm provided mixing. Aeration was via a diffuser stone situated at the bottom of the reactor. Together with the water, NH₄NO₃, and KH₂PO₄ were added to achieve a C:N:P ratio of 100:1:1 (Cookson, 1995; Yan et al., 2011). Water was supplied to the microcosm to increase the water content to 40%. The batch cultures were incubated at 28°C on a rotary shaker at 150 rpm. The treatments included two cases: (a) drilling muds supplied with nutrients (named biotreatment) and (b) drilling muds supplied with nutrients and 1% HgCl₂ to account for abiotic mineralization (named control).

Phytoremediation Experiment

After biotreatment, the drilling muds were mixed with an agriculture soil with no known contamination history at a mass ratio of 1:2 (oven-dried basis). Each 2500 g dry weight of mixed soil was placed in a 20 cm diameter plastic pot with a drainage hole in the bottom. Several seeds of ryegrass were sown in the soil in each pot. Three replicates were performed for each treatment. Evolution of soil and leachate contamination with time was studied.

RESULTS AND DISCUSSION

TOC Removal During Bioremediation

The TOC was selected as the principal parameter for analysis in the present study, because the time required for the determination of the organic content in a sample is only 3–4 h. In addition, TOC can reflect overall organic contamination degree of water, soil, and sediment. As such, the results of TOC removal are particularly useful in determining the performance of the bioremediation system. The TOC removal efficiency of the bioremediation system is presented in Figure 1. The results showed that, after 48 h, the initial TOC contamination level, i.e., 89800 mg/kg drilling muds, was reduced to 21700 mg/kg in the biotreated sample, corresponding to decontamination percentage of 75.8%. In contrast, only 8.6% of the initial TOC was eliminated in the abiotic control. In addition, TOC was reduced to 26400 mg/kg in the biotreated sample after 24 h. However, the TOC reduction was only 4700 mg/kg from the 24th to 48th h. Therefore, the waste drilling muds are not suitable to be remediated only by the microbial method.

Chroma Removal During Bioremediation

The chroma removal during bioremediation is shown in Figure 2. As can be seen, chroma removal efficiency reached 81.5% in the biotreated sample after 48 h, compared to 8.4% removal in the control. This result indicated that the biostimulation approach was an effective process for chroma removal of drilling muds.
FIGURE 1  Time course of TOC removal during bioremediation.

FIGURE 2  Chroma removal during bioremediation.
Polysulfonate potassium drilling fluids contain quite complex components; therefore it is very difficult to determine concentration changes of different components. In this study, concentration changes of Cl$^-$, NO$_3^-$, and SO$_4^{2-}$ were monitored during treatment because these anions were the metabolic products of corresponding organic groups. During bioremediation, a certain amount of slurry was centrifuged at 8000×g for 5 min, and the released Cl$^-$, NO$_3^-$, and SO$_4^{2-}$ ions were measured by ion chromatography with a Shim-pack IC-A3 chromatographic column. As shown in Table 2, the concentrations of Cl$^-$, NO$_3^-$, and SO$_4^{2-}$ ions increased gradually with time. It can be inferred that the microflora in the drilling muds could degrade various pollutant components. In addition, the sharp increase in the concentration of SO$_4^{2-}$ indicated that the microorganisms could utilize sulphonate groups well.

Bioremediation is widely used in the treatment of oil-contaminated water and soil, and microorganisms may adapt to extreme environments. Zekri et al. (2009) isolated an active strain of anaerobic thermophilic bacteria from the environment of the United Arab Emirates. The maximum oil degradation by the strain was observed at 70°C at 10000 ppm NaCl. The degradation rate can be maximized by lowering the salinity and increasing the temperature of the studied systems. Xu et al. (2011) used vegetable oil to extract residual contaminants, and then the extracted soil containing non-recoverable oil was bioremediated under aerobic slurry-phase conditions. After seven weeks of treatment, total extractable organics was reduced from 23100 to 4030 mg/kg soil.

Phytoremediation

Ryegrass performed well in reducing TOC content in soil. After 240 days, the initial TOC contamination level (i.e., 8850 mg/kg soil) was reduced to 1360 mg/kg, corresponding to decontamination percentage of 84.6% (data not shown). The chroma of the leachate in each pot was determined. As shown in Figure 3, the chroma of the leachate was significantly reduced after 240 days of phytoremediation, which agreed well with the changes in TOC content. Overall, the plant-microbe system could continually decompose complex pollutants, which were difficult to degrade only by microorganisms.

Plant roots exude a range of organic compounds that stimulate the activity of microorganisms in the rhizosphere. Root exudates might act as structural analogs to pollutant molecules and qualitatively change the microbial community by encouraging microbial populations. The successful application of plants to clean up contamination of soils and sediments has been well documented in the published literature. Mature reed wetland was utilized by Ji et al. (2004) to remediate heavy oil-based drill cuttings. The results showed that the reed wetland treatment system was effective in degrading the extra heavy oils contained in the drill cuttings without extra fertilization, and that only 4.2% of the
initial hydrocarbon amount persisted in the surface soil layer after two years. Ezzatian et al. (2009) found that *Puccinellia distans* and tall fescue could increase microbial population significantly, and Hydrogel could significantly improve capability of *Puccinellia distans* for hydrocarbon degradation.

**CONCLUSIONS**

We aimed to evaluate the combined effect of microflora and plant growth on the enhancement of bioremediation of waste drilling muds. TOC concentration in the treated drilling muds can be continuously decreased to a very low level. These results indicate that the combined plant-microbe system is an effective option for the treatment of waste drilling muds.

**REFERENCES**


