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Short communication

Application of a two-liquid-phase system for the remediation of diesel oil-contaminated soil

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ABSTRACT

A study was conducted to evaluate preliminarily the feasibility of two-liquid-phase system for the remediation of diesel oil-contaminated soil. As mixing speed and temperature increased, the removal of diesel oil increased above 40% relatively because the rapid mixing and high temperature caused to enhance the mass transfer of diesel oil from soil to silicone oil. As the amount of silicone oil increased, the removal of aromatics in diesel oil was enhanced by 90%, which was larger than aliphatics. These results presented a potential of two-liquid-phase system as an alternative method for the remediation of diesel oil-contaminated soil.

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Introduction

Soil contamination by diesel oil is one of serious environmental problems [1–5]. Diesel oil consists of straight and branched alkanes, cycloalkanes, monoaromatics, and polyaromatics [1–5]. Although the content of aromatics in diesel oil is low, they are potentially carcinogenic and genotoxic. Because diesel oil is a complex and low-volatile mixture, many researches are attempted to remove efficiently diesel oil from the contaminated soil [1–5]. Generally, bioremediation is the most widely applied remediation option for diesel oil-contaminated soil [2–5]. However, this method is a time-consuming process because a long chain length and the increase in branching degree among the components of diesel oil decrease the degradation rate of microorganisms for contaminants [2]. In order to improve biodegradation rate of diesel oil, researchers introduced additives such as surfactants, biosurfactants and water-miscible solvents [4–15]. Surfactants can be adsorbed onto soil, which can cause a secondary contamination [8–10]. Because of their toxicity to microorganism or sequestration of contaminants within their micelle, the biodegradation of contaminants can be inhibited. Biosurfactants are another well-studied additives due to their low toxicity [4,5]. While biosurfactants showed good results for the remediation of diesel oil-contaminated soil, it is still necessary to

investigate on a universal biosurfactant with high oil removal efficiency [11]. Water-miscible solvents used mainly ethanol and acetone. However, the field application is difficult due to safety problem of these solvents [12–15]. Recently, water-immiscible liquid (WIL) has been used as an alternative additive for the remediation of PAHs-contaminated soil. Because this solvent is separated with water, a two-liquid-phase (TLP) system is called. In the TLP system, the contaminants desorbed from soil are transferred to WIL [12,13]. The used WIL has a high capacity which can dissolve hydrophobic organic pollutants. In addition, the solubility of each component in WIL is not almost influenced by the co-existence of other components [12,13]. The focus of the present work was to evaluate the feasibility of TLP system preliminarily as a new method to remove diesel oil from contaminated soil. Several tests in a laboratory-scale reactor have been performed to investigate the effects of operation parameters such as temperature, mixing speed, the content of silicone oil, and the viscosity of silicone oil. This is the first report about the application of TLP system for the remediation of diesel oil-contaminated soil.

Materials and methods

TLP system

Before the preparation of soil contaminated artificially by diesel oil, the uncontaminated soil was pre-sterilized by repeating three times to autoclave for 60 min at 121 °C and sequentially to set for

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1 day at the atmosphere temperature in the clean room. Soil was kaolinite (San-Cheong, Korea, organic content 0.52%) screened below 150 μm [16]. Diesel oil was obtained from L gas station (Korea). The contaminated soil was prepared by mixing between 240 g of soil and acetone dissolved 4 mL of diesel oil. And then, acetone was volatilized from the contaminated soil at atmosphere. Initial diesel oil concentration was approximately from 12,000 to 15,000 mg diesel oil/kg soil. The used water-immiscible liquid was silicone oil (dimethylpolysiloxane, 5 cS, 20 cS, 200 cS, and 500 cS, Sigma). Silicone oil is a representative additive in the TLP system [12–15]. Previous study was reported about the performance of silicone oil to remove hydrophobic compounds such as PAHs from the contaminated soil [17]. For the application of TLP system, 240 g of diesel oil-contaminated soil was mixed with 800 mL of water (30% w/w slurry) and the different volumes of silicone oil (2.5%, 7.5%, and 15% v/v). The slurry and silicone oil were agitated at mixing speed of 150 rpm and 300 rpm and temperature of 25 °C and 30 °C, respectively. After mixing, silicone oil was separated definitely with soil phase including water due to its water-immiscible property and was almost not adsorbed on the soil [12–15]. In order to analyze the removal efficiency of diesel oil from the contaminated soil in the TLP system, 5 mL of slurry and 1 mL of silicone oil were sampled periodically.

Analysis

In order to measure residual diesel oil concentration in soil and the concentration of diesel oil transferred to silicone oil, 1 g of dried soil and 1 mL of silicone oil were mixed with some amount of hexane (Merck) in an end-over shaker at 180 rpm for 24 h, respectively. After stagnation, the solvent fractions were analyzed at 210 nm and 254 nm by HPLC (Waters, USA) with Petro XL column (250 \times 4.6 mm, Genesis, UK). Elution was performed using 100% hexane at the flow rate of 1.0 mL/min.

Results and discussions

The HPLC chromatographic profiles of diesel oil at 210 nm and 254 nm are shown in Fig. 1a and b. Although most of diesel oil consists of aliphatics, aromatics were mainly detected at two wavelengths. As shown in Fig. 1, aliphatics were detected prior to aromatics. The peaks of aliphatics were sharp and were not separated accurately compared with those of aromatics. However, the differences of chromatograms at 210 nm and 254 nm during the operation of TLP system were observed (Fig. 2). Compared with chromatograms of diesel oil in a soil phase and a WIL phase at 210 nm, the peak of aliphatics in a soil phase was diminished and in a WIL phase was dominated significantly. On the other hand, at 254 nm, the peak area of aromatics in a soil phase was reduced and in a WIL phase became broader.

Effect of mixing speed and temperature

In the TLP system, the mass transfer rate of diesel oil from soil to WIL phase was controlled by mixing speed and temperature. Fig. 3 describes the change of diesel oil concentration in a soil phase and a WIL phase at 210 nm and 254 nm. The used mixing speed was 150 rpm and 300 rpm, and the temperature was 25 °C and 30 °C, respectively. Although the volatility of diesel oil was enhanced slightly in the increase of mixing speed and temperature, silicone oil was known to treat efficiently volatile compounds [15]. So, we assumed that the removal efficiency of diesel oil was not affected largely by its volatility. At 25 °C, when mixing speed was fastened from 150 rpm to 300 rpm, the removal efficiency of diesel oil increased about 20% at 210 nm with mainly the peak of aliphatics. At 254 nm with broadly the peak of aromatics, the removal efficiency of diesel oil was enhanced above 40%. Also, at 150 rpm,

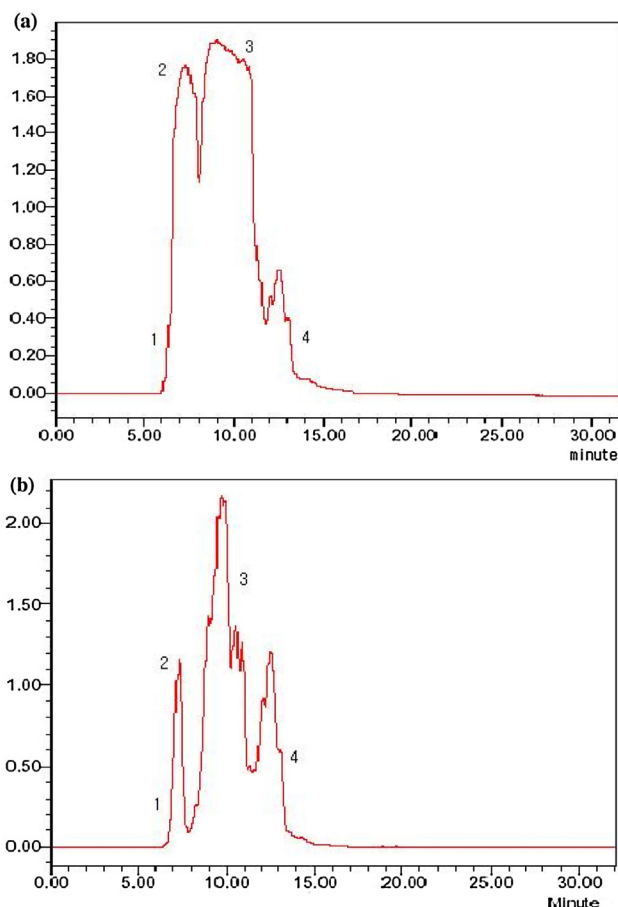


Fig. 1. Chromatograms of diesel oil made by HPLC at 210 nm (a) and 254 nm (b); 1 aliphatics, 2 mono-aromatics, 3 di-aromatics, and 4 poly-aromatics.

when temperature increased from 25 °C to 30 °C, the removal efficiencies of diesel oil increased above 20% and 40% at two wavelengths, respectively. The main reason was because the mobility of components in diesel oil increased due to high mixing speed or temperature. On the other hand, at 30 °C, the removal efficiency of diesel oil was almost not influenced by the increase of mixing speed. At 300 rpm, although temperature increased, the removal efficiencies of diesel oil also were not changed largely. The removal efficiency of aromatics was higher than that of aliphatics because aromatics had higher hydrophobicity than aliphatics. Consequently, when the TLP system was mixed with 300 rpm at 30 °C, the removal efficiency of each component in diesel oil was not influenced by controlling other parameter.

Effect on the amount of silicone oil

The amount of silicone oil affected the mass transfer extent of diesel oil from soil to WIL. Fig. 4 shows profiles on the change of diesel oil concentration in a soil phase and a WIL phase when the amount of silicone oil increased from 2.5% (v/v) to 15% (v/v). The overall removal efficiency of diesel oil from soil was not influenced by the amount of silicone oil. However, the mass transfer extent of diesel oil to a WIL phase had different trends with the amount of silicone oil. At 210 nm, the mass transfer extent of diesel oil was about 70%, while was above 90% at 254 nm. As mentioned previously, these trends were observed due to higher hydrophobicity of aromatics. When the amount of silicone oil increased, the mass transfer extent of diesel oil was enhanced at 210 nm. However, at 254 nm, the mass transfer extent of diesel oil decreased in only 2.5% (v/v) silicone oil. Because the solubility

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