### BIOREMEDIATION OF SOIL CONTAMINATED WITH POLYCYCLIC AROMATIC HYDROCARBONS USING A DRUM BIOREACTOR

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#### Introduction

Soils contaminated with polycyclic aromatic hydrocarbons (PAHs) are often found in the sites of coke production and gas manufacturing plants<sup>1</sup>. Remediation costs of traditional treatment technologies such as incineration, solidification and chemical treatment are high<sup>2</sup>. For the clean-up of PAH-contaminated site, the bioremediation is often limited by mass transfer and low aqueous solubilities of the PAHs. The limitation in mass transfer results in low bioavailability and subsequent low biodegradation rates<sup>3</sup>.

Attempts to increase the contact between degrading microorganisms (bacteria) and contaminants to uniform process control have been made using bioreactors which operate in slurry-phase or dry-phase<sup>4</sup>. A recent study by Woo (1997)<sup>5</sup> investigated the effect of using drum bioreactor to evaluate the potential applicability of the bioreactor to treat two and three ring PAHs-polluted soil.

In this study, a novel rotating drum bioreactor system was developed and extensively validated for the remediation of sites contaminated with two to four ring-PAHs such as naphthalene, anthracene, fluoranthene, phenanthrene and pyrene. *Pseudomonas putida* and *Pseudomonas fluorescence* known as microorganisms of high PAHs degrading capacities were selected for the experiment.

#### **Materials and Methods**

A schematic diagram is illustrated in Fig.1. To prepare the experiment, two microorganisms; *Pseudomonas putida* and *Pseudomonas fluorescence* were grown on the agar plate coated with PAHs.

The rotating drum bioreactor with baffle containing 7.02 L of volume was employed in bench scale. The bioreactor of slope type, which was similar to the conventional rotary kiln, was used to increase effective reactor capacity compared with that of horizontal type and material handling.

The objective of this research was to evaluate the applicability of the bioreactor treating PAHs contaminated soil and to develop the compact bioreactor with least screening and watering process. Therefore, the experiment was carried out to optimize microbial growth in the bioreactor and biodegradation rates of five PAHs under various conditions.

Silt loam soil was used for this research. The diameter of the grain was in the range of 0.074 mm to 2 mm and the five PAH-contaminated soil was prepared at the pollutant levels of 50, 100, 200, 300 and 500 mg/kg-soil, respectively.

Soil using the bioreactor was treated without separating coarse sand from the sampled soil mixed with sand, silt and clay. This will be realized developing compact bioreactor by the engineering that coarse sand is mixed by the rotating drum reactor.

The moisture content of soil was 40 wt %. All experiments were conducted at 25 °C. pH was maintained at 7.5 and air was supplied at 0, 0.5, 1.5 or 3 L/min.

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**Figure 1.** Schematic diagram of experimental apparatus. 1. Drum bioreactor 2. Prehumidifier 3. Compressor 4. Water trap 5. Activated carbon 6. Hood 7. Flowmeter 8. Motor. 9. rpm controller 10. Temperature controller 11. Heater. 12. Thermocouple 13. Valve 14. Regulator 15. Sampling ports.

#### **Results and Discussions**

The PAH-contaminated soil with different initial concentration was treated under various operating conditions to evaluate the effect of initial concentration on the removal rate of PAHs. Based on the first-order kinetic model, half-life, degradation rate, k (day<sup>-1</sup>) and over 95% of degradation of PAHs were determined by analyzing experimental data obtained.

#### Influence of air flow rate and moisture content

Fig. 2. shows the influence of the air flow rate on biodegradation rate by mixed culture. As the air flow increased, biodegradation of PAHs were increased.

It was clearly observed that the influence was gradually higher from 0, 0.5, 1.5 to 3 L/min on air flow rate the period degradation of PAHs was shorter from in around 21 to 16 in days. During early five days, physical loss due to volatilization and biological degradation was occurred, especially for naphthalene.

The influence of soil moisture content mixed along with the addition of adequate amount of water was 40% at the initial operating condition. The changes for the degradation of PAHs and microbial population with time were separately analyzed in the soil phases. The number of microorganisms was dramatically decreased during the early stage for 25 days due to the deficiency of carbon substrate. However, the population slightly increased for easily degradable PAHs (naphthalene, fluoranthene and phenanthrene) for short time. After the period for the recalcitrant PAHs (anthracene and pyrene), the number of microorganisms increased with PAHs degradation. This result indicated that microbial activity was subjected to the amount of water content in soil. However, the degradation rate was not decreased at over 30 % of water content in the soil of the reactor. In other words, the influence of moisture content on adsorption was comparatively small at over 30 % of water content in the soil.

#### Influence of soil grain size on adsorption capacity and biodegradation rate

The bioreactor system was operated to treat the various grain soils by mixing with an adequate amount of water. Therefore, it was observed that the degradation of PAHs according to the soil grain size was evaluated in flasks. In the case of coarse soil, the degradation rate of PAHs was 1.6 times higher than that of fine soil as shown in Fig.3. The uncontrolled soil was also evaluated to verify pure biodegradation. The major physical loss was due to volatilization, especially for naphthalene.



Figure 2. PAHs degradation by mixed culture with different air flow rate (25 °C, pH 7.5)



\* Soil Grain Size

1: 0.5 mm~2 mm, 2: 0.42 mm~0.5 mm, 3: 0.25 mm~0.42 mm, 4: 0.149 mm~0.25 mm, 5: 0.074 mm~0.149 mm 6: 0.074 mm below

- \* individual bar
- left: initial conc.
- middle: without microorganism 10days later
- right: with mixed culture 10days later

Figure 3. PAHs concentrations(mg/kg) with different soil grain size (25 °C, pH 7.5)





(b) degradation rate  $k(day^{-1})$  of PAHs



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#### Biodegradation characteristics of microorganism for the bioreactor

In the batch system, PAHs were disappearing at the different degradation rates according to their hydrophobicity. Over 95 % of PAHs (500 mg/kg-soil) with 2 to 4 rings (naphthalene, anthracene, fluoranthene, phenanthrene and pyrene) was degraded within 27 days as shown in Fig. 4(a). The difference in the bioavailability of PAHs might be explained by the difference in the solubility of PAHs in water and the number of aromatic rings.

Therefore degradation rate increased proportionally to the solubility of PAHs and decreased to the log Kow (log octanol-water partition coefficient). The removal rate of PAHs was faster in mixed culture system than in single species (*Pseudomonas putida* or *Pseudomonas fluorescence*). The degradation rate  $k(day^{-1})$  of total PAHs was 0.07940, 0.08856 and 0.10835 day^{-1} of mixed culture, *Pseudomonas putida* and *Pseudomonas fluorescence* alone and mixed culture as shown in Fig. 4(b), respectively.

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#### References

- 1. IISI ENCO, "Report on Remediation Status In Steel Site", ENCO-39 (2001)
- 2. Nyer, E.K, In-situ treatment technology, Lewis Publisher, (1997)
- 3. Bekins, B.A., Warren, E., and Godsy, E.M., "A comparison of zero-order, first-order, and monod biotransformation models", Groundwater, Vol. 36, No. 2, pp.261-268, (1998)
- 4. Banerjee, D.K., "Monitoring the biological treatment of anthracene-contaminated soil in a rotatingdrum bioreactor", Applied Microbiology and Biotechnology, Vol 43, (1995)
- Woo, S. H., Park, J. M., Song, K. S., Song, C. S. and Lee, K. P., "Bioremediation of soil contaminated with polycyclic aromatic hydrocarbons using a drum bioreactor system", In Situ and On-Site Bioremediation: Volume 2, Fourth International In-Situ and On-Site Bioremediation Symposium, Battelle Press, pp. 233-238 (1997)