PRE-TREATMENT METHOD IN A BIO-DEGRADATION SYSTEM FOR REMOVING POLYCHLORINATED DIOXINS FROM CONTAMINATED SOIL AND SEDIMENTS

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Introduction

The purpose of this study is to build a low cost and ecological bio-degradation method compared with all current thermal or chemical methods for treating polychlorinated dioxins in contaminated soil and sediments. Up to now, we have established the following points; (1) The SH2B-J2 strain, a gram-positive bacterium belonging to the genus *Geobacillus* is an aerobic thermopile whose optimal cultivation temperature is 65C[1,2,4,8)]. (2) The strain is a microorganism capable of directly cleaving ether-bound of dioxins at the first step of the reaction[2,3,4)]. (3) The strain is able to decompose polychlorinated dioxins without re-synthesizing any of the most potent toxic compounds such as 2,3,7,8-TCDD, etc[1,4)]. (4) The crude cellular membrane enzyme produced by the strain is the group *glutathione-s-transferase*[3,4,5,8)]. Each machine was designed and manufactured using the above points and is a wet type mill machine with a 0.04m^3 capacity and a wet type vibration strainer with a 0.002m^3 capacity for dividing silt and clay fractions from contaminated soil and sediments as slurry[4)] and enriching chlorinated dioxins within contaminated soil[1,4)]. In this paper, we discuss the validity of methods for pre-treating polychlorinated dioxins within contaminated soil and sediments prior to bio-degradation.

Material and Methods

Contaminated soil and sediments are heterogeneous systems containing heavy metals and dilute concentrations of dioxins. Dividing each fraction of the silt/clay and sand/gravel prior to bio-degradation is quite effective as a form of pre-treatment for reducing the mass intended for bio-degradation enriching dioxins and removing obstructive heavy metals from heterogeneous systems by mill machine and vibrating strainer. We verify the validity of these pre-treatment methods based on the following experimental evidence and offer an effective hypothesis; (1) The reaction-rate were evaluated from the correlation between a lower dioxin concentration (pg-TEQ) and the cultivation-time by co-cultivating with the SH2B-J2 strain and using a mixed solution of polychlorinated dioxins at a temperature of 65C[4,7,9)]. We found that the enzyme reaction-rate is proportional to the dioxin concentration. Pre-treatment therefore enhances the reaction-rate in dioxin-contaminated soil and sediments. (2) The model is actually a multilayer quartz-protein-silicate model of silt and clay that includes both dioxins and heavy metals. Breaking down this silt and clay structure during pre-treatment is therefore quite effective boosting the enzyme reaction-rate and removing heavy metals that obstruct the enzyme reaction. Table 1 shows the procedure for enriching dioxins and removing heavy metals within contaminated soil.

Results and Discussion:

Properties of test specimens on chlorinated dioxins in contaminated soil and sediments

We used a mixture of fly ash and ordinary soil in the model as the specimen. The real contaminated was gathered from the vacant lot of a hospital building in Tokyo. Each specimen contained more Polychlorodibenzofurans (PCDFs) than polychlorodibenzo-*p*-dioxins (PCDDs) as shown in Table 2, from analytical results by GC/MS. These facts show that ash from wastes burnt by incinerators was a source of these dioxins within the contaminated soil.

Pre-treatment method to enrich dioxins from contaminated soil and sediments

Soil is the mixture of a silt/clay fraction and a sand/gravel fraction. Assuming that there is no interaction between the silt/clay fraction and sand/gravel fraction, we can define the dioxins enrichment efficiency from contaminated soil by using the following equations[10].

[***z***: Mass of soil] = [***x***: Mass of sand/gravel] + [***y***: Mass of silt/clay] (g)
[***c***: Dioxins within soil] = [***a***: Dioxins on sand/gravel] + [***b***: Dioxins within silt/clay] (pg-TEQ/g)
[***r***: Ratio of silt/clay and sand/gravel] = [***y***: Mass of silt/clay] / [***x***: Mass of sand/gravel] = (***c***-***a***) / (***b***-***c***)
[***s***: Ratio of silt/clay] = [***y***: Mass of silt/clay] / [***z***: Mass of soil]×100 = {***r***/(1+***r***)}×100 (%)
[γ: Dioxins enrichment efficiency]
= [***b***: Dioxins within silt/clay]×[***y***: Mass of silt/clay] / ([***c***: Dioxins within soil]×[***z***: Mass of soil])×100
= (***b***/***c***)×s (%)

We found that the dioxins enrichment efficiency: **γ** was 99.8 % as a result of the pre-treatment method using the mill machine under the condition of a contaminated soil to pure water ratio of 1:1 as shown in Figure 1[9]. On the other hand, **γ** was 50%～80% as a result of the pre-treatment method using the vibrating strainer under the condition of contaminated soil to pure water ration of 1:15 as shown in Figure 2[10] and Figure 3[10]. Only a silt/clay fraction can attach or adsorb dioxins within contaminated soil. The collision among silt/clay, stress and shear forces on the silt/clay fraction caused by the mill machine breaking down the multi-layer structure makes the mill machine pre-treatment more efficient for dioxins enrichment than the vibrating strainer. Therefore, effective pre-treatment to enrich dioxins from contaminated soil not only requires dividing the silt/clay fraction but also applying stress and shear forces on the silt/clay fraction.

Pre-treatment method to remove obstructive heavy metals from contaminated soil and sediments
We first tested the respective changes in the dioxins concentration from adding water, acid-treatment and alkaline-treatment. The dioxins concentration was enriched by weakly dividing a silt/clay fraction of contaminated soil using the vibrating strainer under the condition of a contaminated soil to pure water ratio of 1:15 as shown in Figure 4[10]. However, the dioxins concentration was not affected by either adding water or alkaline-treatment. The alkaline-treatment in particular extremely slowed the sedimentation velocity due to the formation of colloidal hydroxides. The acid-treatment on the other hand had absolutely no effect on the sedimentation velocity. The acid-treatment moreover can break down the multi-layer structure of the silt/clay so, this treatment can enrich dioxins by the elution from a silt/clay fraction. We then tested changes in the lead (Pb) metal concentration by respectively adding water, acid-treatment and alkaline-treatment. We assumed that Pb metal attaches to the silt/clay fraction. The Pb metal concentration within contaminated soil was enriched by weakly dividing a silt/clay fraction from contaminated soil by using vibrating strainer under the condition of a contaminated soil to pure water ratio of 1:15 as shown in Figure 5[10].

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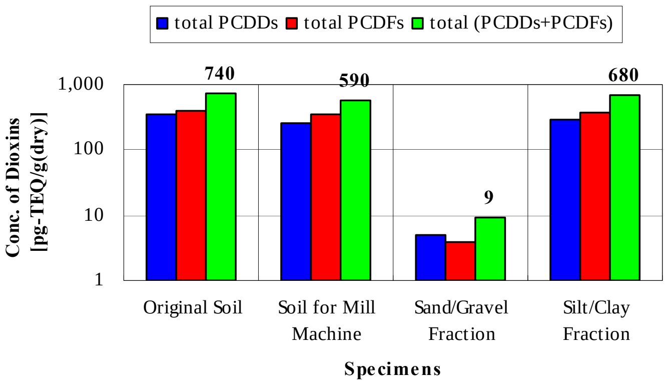
Table 1 Procedure for enriching dioxins and removing heavy metals within contaminated soil

Acid-treatment:	**Alkaline-treatment:**
①Divided 100g of silt/clay from contaminated soil	①Divided 100g of silt/clay from contaminated soil
②Added 150g of pure water	②Added 150g of pure water
③Slurry	③Slurry
④Regulated pH of 3.5 by HNO_3	④Regulated pH of 12 by NaOH
⑤Let stand for 7days	⑤Let stand for 7days
⑥Dioxin conc. detected by GC/MS	⑥Dioxin conc. detected by GC/MS
⑦Pb conc. detected by IPC/MS	⑦Pb conc. detected by IPC/MS

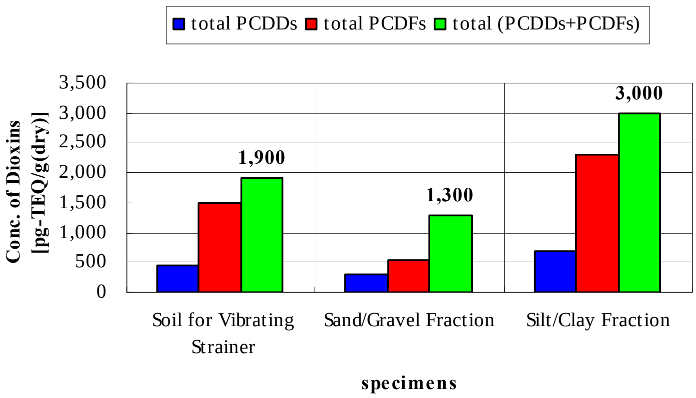
Table 2 Dioxins concentration within each contaminated soil as a test specimen

Specimens	*Gathered contaminated soil*	*total PDDDs*	*total PDDFs*
MC	Model contaminated soil by fly ash mixed with soil	340 pg-TEQ/g	400 pg-TEQ/g
H18	Soil scraped from surface to depth of 300mm	570 pg-TEQ/g	2,000 g-TEQ/g
A1	Soil filtered under strainer of within 40mm mesh	92 g-TEQ/g	260 pg-TEQ/g



Mill Machine

Figure 1 Dioxin enrichment of specimen named MC by mill machine: γ = 99.8 %



Vibrating Strainer

Figure 2 Dioxin enrichment of specimen named H18 by the vibrating strainer: γ = 55.7 %

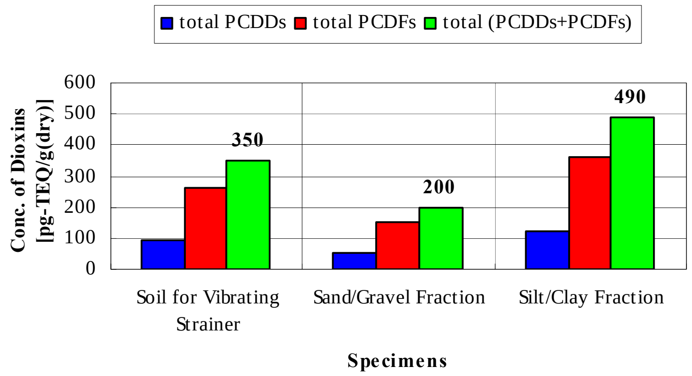


Figure 3 Dioxin enrichment of specimen named A1 by vibrating strainer: γ = 72.3 %

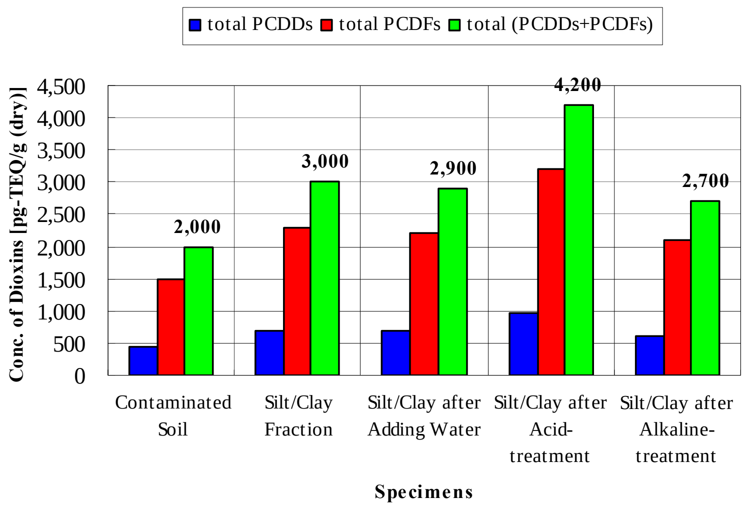


Figure 4 Dioxin enrichment of specimen named H18 by vibrating strainer

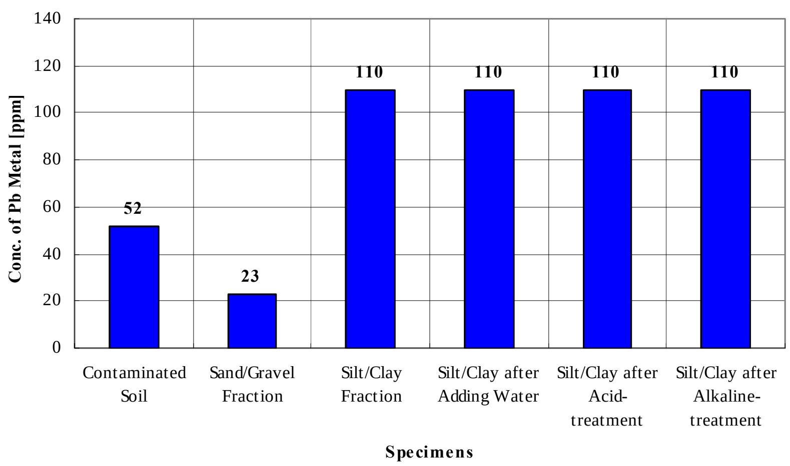


Figure 5 Dioxin enrichment of specimen named A1 by vibrating strainer