# NUMERICAL SIMULATION ON THE BIOREMEDIATION OF OIL CONTAMINATED SOIL USING A MICROBIOLOGICAL REACTION MODEL

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***ABSTRACT*:** Degradation of oil using microbials, known as bioremediation, is a promising method to remedy contaminated soil land. In the study, experiment is first conduct for oil mixed soil. It is found that oil evaporation dominates in the initial days, and then biodegradation plays a role. Also, temperature and water content influence the biodegradation rate. Based on the experiment, a microbiological model is constructed to simulate microbial growth and oil decomposition. Biodegradation rate is influenced by oil concentration, temperature, water content and oxygen concentration. Those factors are simulated using transport models considering mass and heat conservations. The microbiological model and the transport models are coupled in an analytical system. Using the system, the overall bioremediation process can be reasonably reproduced.
***KEYWORDS***: Oil contaminated soil, Biodegradation, Modeling

## 1. INTRODUCTION

In Japan, with the decline of overall vehicle sale and increase of eco-cars, in recent years the demand of gasoline oil is decreasing and some gas stations have been closed. To reutilize the land, the premise is to remedy the soil contaminated by gasoline. Several methods can be used, such as combustion, distillation, washing and biodegradation. The biodegradation, also known as bioremediation, is to decompose oil using microbials. Its cost is relatively low and easy to be conduct. On the other hand, bioremediation is a complicated bio-chemical process. Aided with oxygen and enzymes, oil is decomposed into water and $CO_2$, and microbial population grows due to biosynthesis. Such a process is greatly influenced by factors such as temperature, the contents of water, oil and oxygen. Those factors change according to environment and time history. Therefore, to promote the efficient application of biodegradation, it is necessary to evaluate its effect by coupling the environment conditions and microbial activity.

In Concrete Laboratory, the University of Tokyo, a multi-scale analytical system has been established to simulate substance and heat transports in porous media under environmental conditions. This system was first designed for concrete materials (Maekawa et al 2003). Recently, it was extended for compositing process of organic waste by coupling it with a microbiological model (Bongochgetsakul and Ishida 2008). The composting rate depends on microbial growth rate, which is affected by temperature, the contents of water, oxygen, waste substrate and ammonia. Those factors were simulated based on mass or heat conservation equations in waste. Although microbial species and the reaction in oil biodegradation are different from those in composting, the authors consider that similar approach coupling the reaction and environmental factors can be applied. Therefore, in

this study, the purpose is to establish such an analytical system which is capable of simulating the oil biodegradation process in soil. First, to understand the main characteristics experimental study is conduct.

## 2. EXPERIMENTAL STUDY

### 2.1 Experiment Program

Soil were prepared by mixing clay loam with 10% seed compost by weight. Soil porosity is 0.53. The seed compost is a commercial product in which microbials with oil degradation ability were cultivated and sufficient mineral ions were added preliminarily. The water contents were adjusted to 23.1% and 33.3% by weight ratio, respectively. Then, standard gasoline was added into specimens using atomizer and mixed repeatedly. After that, soil was laid on mesh wires in a plastic container (Fig. 1). In the side-wall below and upper side wall, holes were opened to ventilate. The containers were reserved under constant environment for 1 month and oil concentration change was measured. The temperatures were 20°C and 40°C, and RHs were 60% and 90%. Every time 10g soil was sampled and oil concentration was measured using TPH method (Ministry of the Environment, Government of Japan 2006). The specimen groups are shown in Table 1. For each group, a sterilized specimen was prepared as the comparison. At the beginning, those specimens were heated at 120°C for 20 minutes in a closed container to eliminate all the microbials.

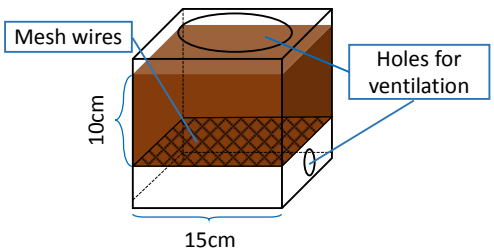


Fig. 1 Soil specimens in container

Table 1 Experiment groups

| Group | Microbial or sterilized | Temperature & RH | Water content (%) |
|---|---|---|---|
| T40 | Microbial | 40°C & 90% | 23.1 |
| T40-S | Sterilized | | |
| T20 | Microbial | 20°C & 60% | 23.1 |
| T20-S | Sterilized | | |
| T40W | Microbial | 40°C & 90% | 33.3 |
| T40W-S | Sterilized | | |

### 2.2 Result and Discussion

The experiment results are shown in Fig. 2. The initial measured oil concentrations fluctuate between 4000~7000 mg/(kg-dry soil). Such a fluctuation may be from the ununiform oil distribution. The added oil amount is very small, so oil is in the form of thin layer attaching at soil grain surface rather than liquid drops. Even with carefully mixing, it is difficult to uniformly spread the oil over the whole bulk soil. When 10g soil was sampled in the measurement, it reflects the local concentration and may deviation from the average concentration exits.

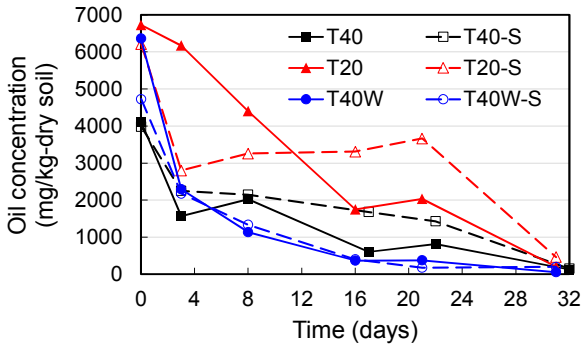


Fig. 2 Experiment results of oil concentration change

In spite of the deviations, for all the groups the oil concentration declines from the beginning. This even occurs for the sterilized ones, so can be mainly attributed to evaporation. Oil contains varieties of hydrocarbons, and those with light molecular weights tend to evaporate in the first several days. Past researches have pointed out such an evaporation (Takahata and Taki 2005; Prince et al 2007). In this

study it is confirmed again that oil evaporation into the atmosphere is unavoidable when evaluating the oil degradation.

If we neglect the first week and focus on the subsequent period, it can be seen that the concentration continues to decline. Furthermore, if we compare the microbial one with the sterilized one, for example T20 and T20-S, the former one declines faster. Normalizing by concentration at 8 day, this trends becomes more obvious (Fig. 3). This implies that expect for the evaporation, microbials became active and biodegradation played a role. Also, biodegradation effect of T20 appears more remarkable than that of T40. This implies that at 20°C microbials have higher activity than that at 40°C. Finally, for T40W with higher water content (33.3%), little degradation is observed. This implies that water may be excessive and such an aquatic environment is no longer appropriate for microbial activity.

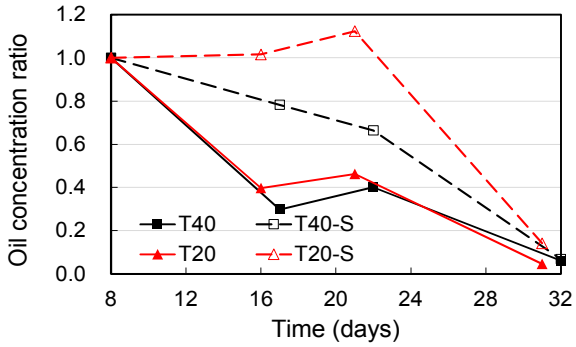


Fig. 3 Oil concentration ratio normalized by 8th day

As a summary, at current stage the experiment results cannot provide quantitative evaluation due to the deviations. Measurement method needs to be improved. However, principal characteristics can be summarized as follows: (a) Oil evaporation dominates in the initial days, and after that biodegradation becomes remarkable; (b) for low temperature (20°C), biodegradation rate appears higher than that at high temperature (40°C); (c) Low water content (23.1%) is more suitable for biodegradation than high water content (33.3%).

## 3. ANALYTICAL SYSTEM

### 3.1 Framework of the analytical system

Biodegradation and environmental factors are coupled physically. On the one hand, biodegradation rate depends on temperature, contents of water, oil and oxygen. Also, during the process, oxygen and oil are consumed, and water and heat are generated. Those consumed or released substances or heat change the local environment, and finally affect the subsequent biodegradation. In other words, an interaction exists.

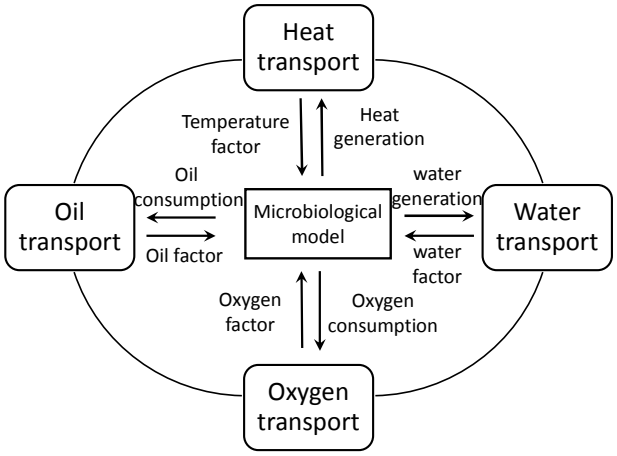


Fig.4 Framework of the analytical system

Based on the discussion, an analytical system for biodegradation is established. The scheme is shown in Fig. 4. It consists of two parts: transport sub-system, and the microbiological model. The transport sub-system includes four models: heat transport model, water transport model, oxygen transport model and oil transport model. Soil is assumed as porous continuum in space. In each model, continuous flows are used to describe the transports of substances or heat. They obey mass or energy conservation law. The solutions are temperature, water content, oxygen concentration, and oil concentration, respectively. The microbiological model is to kinetically describe the microbial growth and the corresponding oil

biodegradation. The microbial growth rate is a function of the current microbial population and the factors delivered from the transport sub-system. The consumed amounts of oxygen and oil as well as the generated water and heat in time intervals are calculated. Then, they are delivered to the transport sub-system for subsequent transport calculations.

### 3.2 Microbiological model

The chemical compositions of oil, which are hydrocarbons, are decomposed by microbials. According to research by Das and Chandran 2007, they are first attacked by oxygenases in an oxidative process aided by enzymes. In this process, oxygen is incorporated. After that, through peripheral pathways hydrocarbons are converted step by step into metabolic intermediates in a tricarboxylic acid cycle. Then, those metabolic intermediates are used for biosynthesis, with the incorporation of mineral ions in soil such as ammonium cations. As a result, microbial population grows, and water and $CO_2$ are generated as respiration products. Although the overall process is very complicated, if neglecting intermediate reactions and focusing on the reactants and final products, stoichiometrically the chemical reaction equation can be written as

$$\begin{aligned} &C_aH_b + (a + 0.25b + 0.5yn + 0.75zn - 0.25xn - wn)O_2 \\ &+ znNH_4^+ \rightarrow nC_wH_xO_yN_z + (a - wn)CO_2 \\ &\qquad + (0.5b + 1.5zn - 0.5xn)H_2O + znH^+ \end{aligned} \tag{1}$$

$C_aH_b$ is the molecular formula of hydrocarbon, whereas $C_wH_xO_yN$ is the molecular formula of biomass. $n$ is the conversion ratio. Varieties of microbial species live in soil and oil contains various hydrocarbons, so $C_aH_b$ and $C_wH_xO_yN$ should be regarded as their average formula. Also, Eq. 1 should be only noted as a representative equation for all the reactions. An ideal approach is to treat the reactions of each hydrocarbon and microbial species separately, but at current stage it is overly complex and difficult to operate. Usually, a typical gasoline oil consists of hydrocarbons with between 4 and 12 carbon atoms per molecule. Herein, octane with the molecular formula $C_8H_{18}$ is used as the representative composition. Besides, past researches indicated that $C_5H_7O_2N$ is a typical molecular formula of microbials (Edwards and Nirmalakhandan 1999; Lamy et al 2013; Komilis et al 2010), so Eq. 1 becomes

$$\begin{aligned} &C_8H_{18} + (12.5 - 5n)O_2 + nNH_4^+ \rightarrow \\ &nC_5H_7O_2N + (8.0 - 5n)CO_2 + (9.0 - 2n)H_2O + nH^+ \end{aligned} \tag{2}$$

Eq. 1 is stoichiometric equations, but the kinetics is not involved. The kinetics depends on the microbial population change, which can be generally divided into four phases (Fig. 5).

1) Lag phase. When oil is given, microbials are not active immediately but after a period of time, which is called lag phase. In the lag phase, microbial population hardly increases. The lag phase depends on the culture history and growth condition.

2) Exponential phase. Each microbial cell divides to form two cells, resulting in an exponential increase of microbial population. In this phase microbials are in the healthiest state and oil can be degraded continuously. The growth rate depends on environment as well as microbial characteristics.

3) Stationary phase. Microbial growth rate slows down and stop due to loss of bio-activity. In this phase, the microbial population is almost constant. Although oxygen is intaken to sustain the living, the overall consumption is much lower than that in the exponential phase, so oil degradation is negligible.

4) Death phase. The oil is almost depleted, so the microbials start to die. Some may sustain themselves by consuming dead ones to survive for longer time, but the overall population will decline exponentially.

To simulate the growth, stagnation and death of

microbials, a first-order rate equation is expressed as

$$\frac{dX}{dt} = \alpha\mu X - \beta\mu_d X \tag{3}$$

where, $X$: microbial weight density (kg/m$^3$), which represents microbial population in unit soil volume; μ: environmental-dependent growth-rate (/day); $\mu_d$: death-rate(/day); α: parameter indicating the growth phase; β: parameter indicating the death phase.

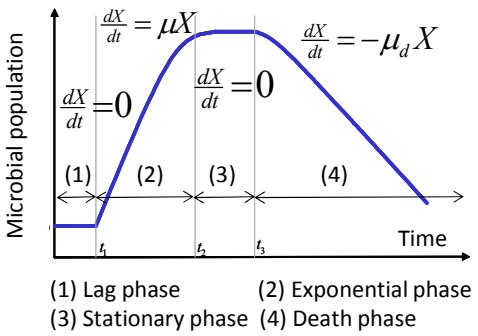


Fig.5 Typical curve of microbial population change

The growth-rate μ is crucial to the microbial growth and oil degradation. It is expressed using the following equation

$$\mu = \mu_T(T) \cdot f_{O2}(\theta_{O2g}) \cdot f_w(\theta_w) \cdot f_{ol}(m_{ol}) \tag{4}$$

where, $\mu_T$: temperature factor; $f_{O2}$: oxygen factor; $f_w$: water factor; $f_{ol}$: Oil factor; T: temperature (°C); $\theta_w$: volumetric water content (m$^3$/m$^3$); $\theta_{O2g}$: Gaseous oxygen volumetric concentration (m$^3$/m$^3$); $m_{ol}$: oil concentration (kg/m$^3$).

Temperature effect on chemical reaction rate usually follows the Arrhenius law. With increased temperature the reaction rate increases. However, for organism reaction, Arrhenius law is not applicable. There is an optimum temperature at which enzyme activity reaches maximum. Upon the optimum temperature, enzyme loses the activity gradually, so reaction rate decreases. Therefore, the temperature factor is given as the following equation

$$\mu_T(T) = \frac{A_T \cdot e^{-\frac{E_1}{RT}}}{1 + K_T \cdot e^{-\frac{E_2}{RT}}} \tag{5}$$

where, $A_T$, $K_T$: exponential factors (1/day); $E_1$, $E_2$: activation energies (J/mol); $R$: universal gas constant (J/mol/K); $T$: absolute temperature (K). Referring to the experiment results of T20 and T40, tentative values are given to the exponential factors and activation energies, and the function $\mu_T(T)$ is plotted in Fig. 6. It can be seen than $\mu_T$ first increase with temperature until upon 30°C, and then decreases. Its value at 20°C is higher than that at 40°C, which implies higher activity.

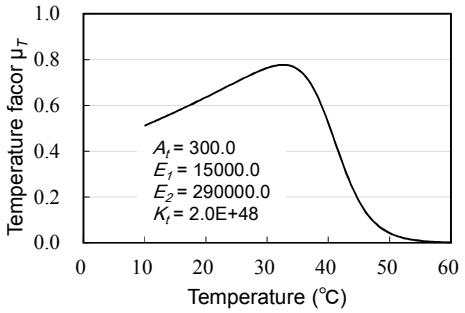


Fig.6 Temperature influential factor

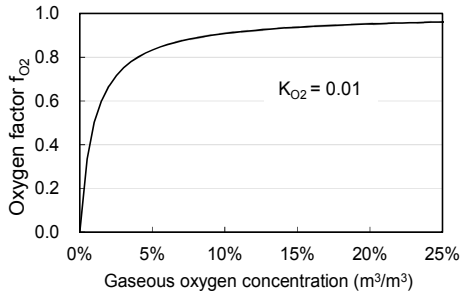


Fig.7 Oxygen influential factor

Since oil degradation is an aerobic reaction, the oxygen amount is important. Only dissolved oxygen can be intaken by microbials, so oxygen factor should be a function of dissolve oxygen. In this system, it is assumed that the equilibrium of gaseous and dissolved oxygen follows Henry's law. With this assumption, dissolved and gaseous oxygen have one-to-one positive relationship. This will be explained in Section 3.3. Here, equivalently oxygen factor is set as a function of gaseous oxygen as

$$f_{O2}(\theta_{O2g}) = \frac{\theta_{O2g}}{K_{o2} + \theta_{O2g}} \quad (6)$$

where, $K_{o2}$: half-saturation coefficient. $K_{o2}$ represents the gaseous oxygen concentration at which the oxygen factor becomes half of its maximum value. The function of oxygen factor is shown in Fig. 7. It can be seen that oxygen factor increase with gaseous oxygen concentration. This trend is more obvious at low concentration range.

Microbials require water as a living environment for mineral ions and dissolved oxygen intake. On the other hand, over aquatic environment is harmful for microbial growth. In the experiment, that little biodegradation is observed in T40W may be due to the high water content. Herein, the water factor is expressed as the following equation

$$f_w(\theta_w) = \begin{cases} \dfrac{1}{1+(K_\theta(\theta_{opt}-\theta_w))^m}, & (0 \le \theta_w \le \theta_{opt}) \\ \dfrac{\theta_{\max}-\theta_w}{\theta_{\max}-\theta_{opt}}, & (\theta_{opt} < \theta_w \le \theta_{\max}) \\ 0.0\ , & (else) \end{cases} \quad (7)$$

where, $\theta_{max}$: the maximum water content above which microbial activity becomes zero ($m^3/m^3$); $\theta_{opt}$: the optimum water content at which microbial activity reaches maximum ($m^3/m^3$); $K_\theta$, $m$: water content parameters. The function of water factor is shown in Fig. 8. Clearly, the factor increases as water content increases until the optimum content, and then decreases sharply. When it is upon the maximum content, microbial activity becomes zero.

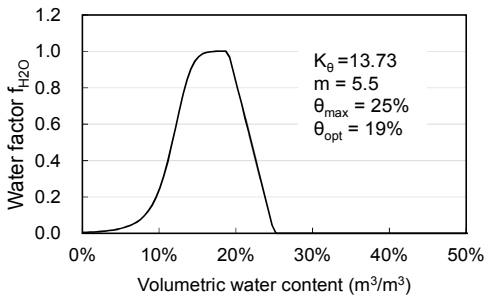


Fig.8 Water influential factor

Oil, as the reactant, its concentration also influences the grow rate. Without sufficient oil microbials cannot sustain their growth. Oil factor is represented as the following equation

$$f_{ol}(m_{ol}) = \frac{m_{ol}}{K_{ol}X + m_{ol}} \quad (8)$$

where, $K_{ol}$: coefficient related to half-saturation of oil concentration effect. Since $X$ is microbial weight density in Eq. 3, $K_{ol} \cdot X$ represents oil concentration at which oil factor becomes half of its maximum value. With constant $K_{ol}$ value and various $X$ values, the function of oil factor is plotted in Fig. 9. Obviously, similar with oxygen factor, with constant microbial population, microbial activity decreases as oil concentration decreases. Furthermore, with the same oil concentration, the oil factor value of large population is lower than that of small population. This is reasonable, because the oil amount shared for an individual microbial decreases, which will affect the activity of the overall community.

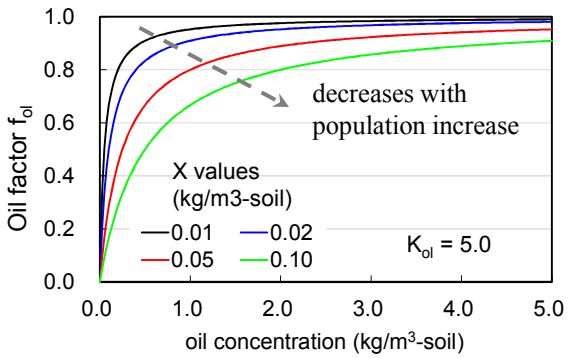


Fig.9 Oil influential factor

With the microbial grow rate, according to the stoichiometric relationship in Eq. 2, oil degradation rate, oxygen consumption rate, water generation rate are obtained as the following equations, respectively

Oil degradation rate: (mass/time)

$$\frac{dCH}{dt} = \frac{1}{n} \cdot \frac{M_{CH}}{M_{CHON}} \cdot \frac{dX}{dt} \quad (9)$$

Oxygen consumption rate: (mass/time)

$$\frac{dO_2}{dt} = \frac{12.5-5n}{n} \cdot \frac{M_{O2}}{M_{CHON}} \cdot \frac{dX}{dt} \quad (10)$$

Water generation rate: (mass/time)

$$\frac{dH_2O}{dt} = \frac{9.0-2n}{n} \cdot \frac{M_{H2O}}{M_{CHON}} \cdot \frac{dX}{dt} \quad (11)$$

Besides, heat generation rate can be calculated from the change in Gibb's free energy $\Delta G^0$ (kJ/model)

$$\frac{dQ_h}{dt} = \frac{\Delta G^0}{M_{CHON}} \cdot \frac{dX}{dt} \quad (12)$$

where, $M_{CHON}$, $M_{CH}$, $M_{O2}$, $M_{H2O}$: molecular mass of microbial, oil, oxygen and water, respectively; $Q_h$: heat generation. In the analysis in this paper, the conversion ratio $n$ is assumed to be 0.4.

### 3.3 Transport sub-system

Heat transport model

The heat transport in soil is described using the heat conservation equations as follows:

$$C\frac{\partial T}{\partial t} = div(D_h \nabla T) + Q_h \quad (13)$$

where, $C$: average oil heat capacity based on volumetric fractions of soil grain, water and vapor (kJ/m$^3$/K); $T$: temperature (°C); $t$: time (day); $D_h$: thermal conductivity (kJ/m/day/K); $Q_h$: heat source term due to oil degradation (kJ/m$^3$/day).

In the boundary, the transferred heat amount is proportional to amount of temperature difference between soil surface and that of the environment as

$$J_{h,sur} = -\gamma_h (T_s - T_{env}) \quad (14)$$

where, $T_s$: surface temperature (°C); $T_{env}$: ambient temperature (°C); $\gamma_h$: heat transmission coefficient (kJ/m$^2$/day/K).

Water transport model

Water transport involves both vapor and liquid water in soil. Considering mass conservation, the transport equation is described using the equation

$$\frac{\partial \theta_w}{\partial t} = -div(J_{wl} + J_{wv}) + Q_w \quad (15)$$

where, $\theta_w$: volumetric water content in soil (m$^3$/ m$^3$); $J_{wl}$: liquid water flux (m$^3$/m$^2$/day); $J_{wv}$: vapor flux (m$^3$/m$^2$/day); $Q_w$: water source term due to oil degradation (m$^3$/m$^3$/day).

Water flux is principally driven by both water content gradient and temperature gradient. $J_{wl}$ and $J_{wv}$ in Eq. 15 are expressed as

$$J_{wl} = -\left( k_w \frac{\partial \psi_w}{\partial \theta_w} \nabla \theta_w + k_w \frac{\partial \psi_w}{\partial T} \nabla T \right) \quad (16)$$

$$J_{wv} = -(k_{wv} \nabla \theta_w + k_{wT} \nabla T) \quad (17)$$

where, $\psi_m$: matrix potential of pore pressure due to capillary tension (m); $k_w$: unsaturated hydraulic conductivity (m/day); $k_{wv}$: vapor diffusivity due to moisture gradient (m$^3$/m/day); $k_{wT}$: vapor diffusivity due to the temperature gradient (m$^3$/m/day/K).

In the boundary, vapor exchange amount between soil surface and environment depends on RH difference, so the flux is given as

$$J_{w,sur} = -\gamma_w (h_s - h_{env}) \quad (18)$$

where, $h_s$: soil surface RH; $h_{env}$: ambient RH; $\gamma_w$: moisture transmission coefficient (kg/m$^2$/day);

Oxygen transport model

Considering both dissolved and gaseous oxygen mass conservation, the transport is expressed as

$$\frac{\partial}{\partial t}\left[(\phi - \theta_w)\rho_g + \theta_w \cdot \rho_d\right] = -div(J_{o2}) + Q_{O2} \quad (19)$$

where, $\rho_g$: concentration of gaseous oxygen (kg/m$^3$); $\rho_d$: concentration of dissolved oxygen (kg/m$^3$); $J_{o2}$: total oxygen flux inside the soil (kg/m$^3$/day); $Q_{O2}$: oxygen sink term due to oil degradation (kg/m$^3$/day).

The total flux consists of those of dissolved and gaseous oxygen, so is expressed as

$$J_{O2} = J_{O2g} + J_{O2d} = \tau_g a D_0^g \cdot \nabla \rho_g + \tau_d a D_0^d \cdot \nabla \rho_d \quad (20)$$

where, $J_{O2g}$, $J_{O2d}$: flux of gaseous and dissolved oxygen, respectively; $\tau_g$, $\tau_d$: tortuosity coefficients of gaseous and dissolved oxygen transport in soil pores, respectively; $D_0^g$: oxygen diffusion coefficient in free atmosphere ($m^2$/day); $D_0^d$: oxygen diffusion coefficient in bulk water ($m^2$/day); $a$: volumetric fraction of free-air-space in soil.

Under isothermal equilibrium condition, dissolved oxygen amount in water is determined by partial pressure of gaseous oxygen, and Henry's law is used to describe such an equilibrium. Herein, oxygen is assumed as quasi-equilibrium state, so with Henry's law the relationship between dissolved and gaseous oxygen is expressed as

$$\rho_g = \frac{M_{O2}}{R \cdot T} H_{O2} \rho_d \quad (21)$$

where, $M_{O2}$: oxygen molecular weight (kg/mol); $R$: universal gas constant (J/mol/K); $T$: absolute temperature (K); $H_{O2}$: Henry's constant for oxygen (Pa/(kg/m3)).

Substituting Eq. 21 into Eq. 19 and Eq. 20, the final equation with only the gaseous oxygen concentration as the variable can be obtained. It is omitted here. Besides, in the boundary, the surface transport of oxygen is expressed as

$$J_{O2,sur} = -\gamma_{O2}(\rho_{g,s} - \rho_{env}) \quad (22)$$

where, $J_{O2,sur}$: oxygen flux at the surface boundary (kg/$m^3$/day); $\gamma_{O2}$: oxygen transmission coefficient ($m^3$/$m^3$/day); $\rho_{g,s}$: gaseous oxygen concentration at the surface ($m^3$/$m^3$), $\rho_{env}$: gaseous oxygen concentration of ambient ($m^3$/$m^3$).

Oil transport model

Caused by capillary tension and temperature gradient, the mechanism of oil transport in soil is similar with water transport. Hence, transport equation and fluxes are similar with water transport. Because the oil content is much smaller than water content, herein not the volumetric soil content but oil weight in unit soil volume is used. The mass conservation is

$$\frac{\partial m_{ol}}{\partial t} = -div(J_{oll} + J_{olv}) + Q_{ol} \quad (23)$$

where, $m_{ol}$: oil weight in unit soil volume (kg/ $m^3$); $J_{oll}$: liquid oil flux (kg/$m^2$/day); $J_{olv}$: vapor oil flux (kg/$m^2$/day); $Q_{ol}$: oil sink term due to degradation ($m^3$/$m^3$/day);

In the boundary, oil evaporation rate depends on temperature and RH. High temperature cause fast evaporation, while high RH will decreases the evaporation (Fingas 1998). Thus the flux $J_{ol,sur}$ (kg/$m^3$/day) is given as

$$J_{ol,sur} = -\gamma_{ol} \frac{T_{env}}{h_{env}} \quad (24)$$

Oil evaporation ability depends on its chemical compositions and temperature (Zhou and Crawford 1995). Hydrocarbons with light molecular weight are easy to evaporate, whereas those with heavy molecular weight hardly evaporate. Herein, a maximum evaporation ratio $k_{eva}$ is defined. Tentatively, $k_{eva}$ is assumed the value 0.5 at 40°C, and it increases with temperature as

$$k_{eva} = \frac{m_{eva}}{m_{ol\_0}} = 0.5 \cdot \left( \frac{T}{T_0} \right)^{\frac{1}{3}} \quad (25)$$

where, $m_{eva}$: maximum evaporable oil content (kg/$m^3$); $m_{ol\_0}$: initial oil content (kg/$m^3$). When the evaporated oil content reaches $m_{eva}$ in the boundary, oil evaporation will stagnate.

### 3.4 Numerical method and calculation flow

The transport sub-system and the microbiological model are integrated, and FEM method is used in the numerical analysis. In this paper, the geometry

model is established based on the soil specimens in the experiment. Due to the geometry symmetry of the specimens, one-eighth is used in the analysis. Because in the symmetry planes there are no substance and heat transport perpendicular to the plane, no transfer elements there are set. In the side face, the soil is surrounded by the plastic walls, so only heat transfer elements are set to allow heat exchange. In the surfaces which contact air, elements are set to all the transport models in order to simulate heat, water, oxygen, and oil exchanges with the atmosphere. The meshing is shown in Fig. 10.

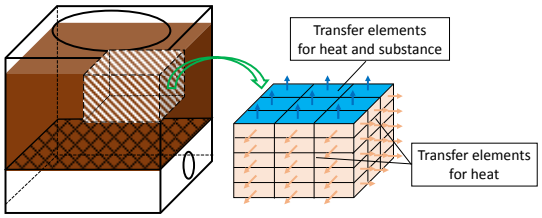


Fig. 10 Mesh of the FEM model

The calculation flow is shown in Fig. 11. First of all, the material properties of soil, oil and microbials as input as initial conditions. Also, environment conditions such as atmosphere temperature, RH, and oxygen concentration are input. After that, the transport equations are discretized in space and time fields according to the mesh and input time steps. In each time step, heat transport is calculated first. Then the temperature is delivered to the microbiological model together with the instant values of water content, oxygen concentration, and oil concentration. With those factors, the biomass increase during the time interval is calculated, and generation (heat, water) or consumption (oxygen, oil) amounts are calculated according to the stoichiometric relationships. Those generation and consumption amounts are delivered back to the transport models as source or sink terms. Then, the transports of water, oxygen and oil are calculated sequentially. The generated heat is used for heat transport in the next time step. In each time step, solution convergence of each model is checked. To ensure precision and convergence, time steps are set to be very fine during the whole analysis. The oil concentration obtained in the analysis in space field is averaged and shown in Section 4.

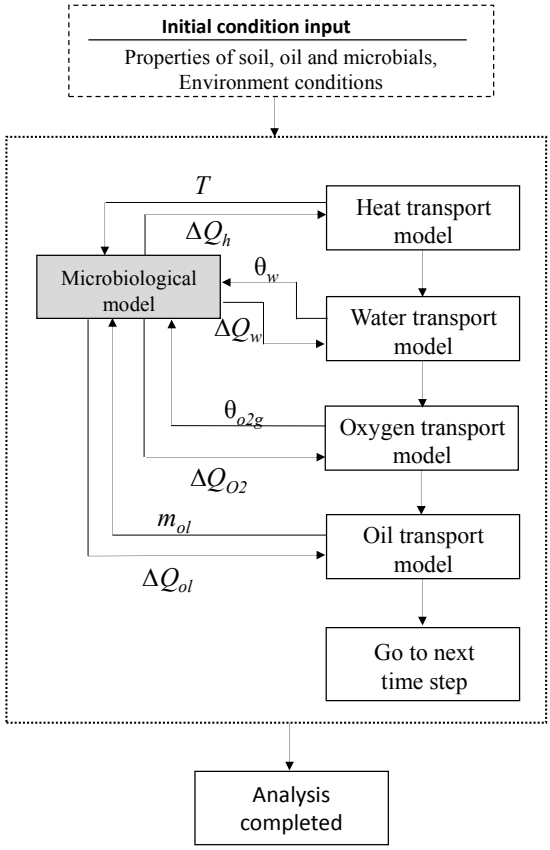


Fig. 11 Calculation flow of oil biodegradation

## 4. ANALYSIS RESULT AND DISCUSSION

### 4.1 Oil evaporation

The analysis results of oil evaporation are shown in Fig. 12. In those cases, microbials are set inactive, which are equivalent to the sterilized cases in the experiment. The influence of temperature and RH can be clearly shown. With the same temperature 20°C, oil evaporates slower at high RH (90%) than that at low RH (60%), whereas the ultimate evaporation ratios are the same. On the other hand, with the same RH (90%), high temperature (40°C) causes fast evaporation rate and high ultimate evaporation ratio. In all the cases, the evaporation

mainly occurs in the initial days and then tend to be stable. This trend coincides with the experiment results and past researches. In the future, further experimental study should be done quantitatively to elucidate the influence of chemical composition and temperature on oil evaporation.

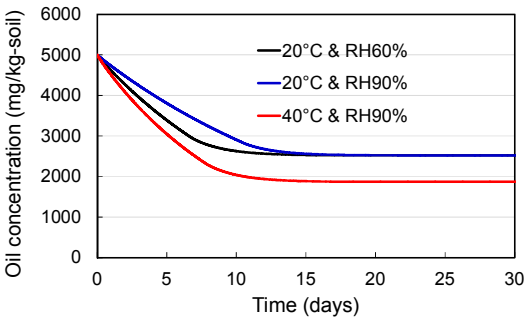


Fig. 12 Analysis of oil evaporation

### 4.2 Influence of temperature on biodegradation

The analysis results with biodegradation effect are shown in Fig. 13. In those cases, RH is 90% and initial water content is 23.1%, while environment temperatures are different. It can be seen that evaporation dominates at early time, while biodegradation becomes dominant at later time. In addition, when the temperature is high as 40°C, although the evaporated oil portion increases, biodegradation effect decreases adversely, resulting in a less effect of total degradation. This can be explained using temperature factor in Fig. 6. At 40°C the temperature factor has decreased from its peak, and microbials are losing their activity because of the temperature. This trends agrees with the experiment results.

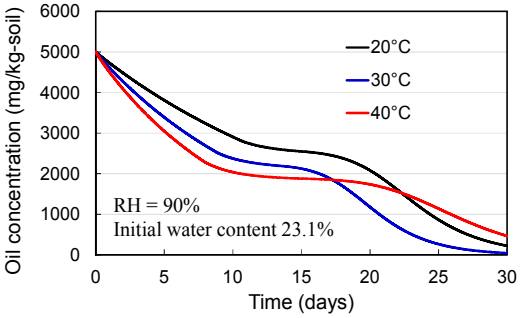


Fig. 13 Oil degradation under varied temperature

### 4.3 Influence of RH and initial water content on oil degradation

The analysis of oil degradation with changed RH and initial water contents are shown in Fig. 14. The temperatures are all 40°C. The one with initial water weight content 33.3% shows no biodegradation effect, which roughly agrees with the experiment case. Also, with the same water content 23.1%, the biodegradation effects of the cases with RH 60% and 90% are different. This can be attributed to that RH affect the water evaporation rate to environment, so influence water environment where microbials are living. The volumetric water content development of those cases are shown in Fig. 15. It can be seen that the water contents are different according to their initial values and RH. For initial water content 33.3%, although water loss occurs from the beginning, within the first two weeks the volumetric water content is upon 25%. According to Fig. 8, the water factor remains zero. It implies that the early activity of microbials is inhibited by the excessive water. Although after that water factor increases because water content decreases, the overall potential of microbial growth and corresponding oil degradation has been postponed. For real site soil, such an inhabitation or postpone may be caused by underground water.

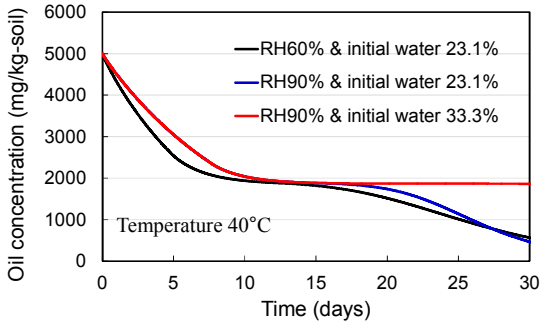


Fig. 14 Oil degradation with varied RH and initial water content

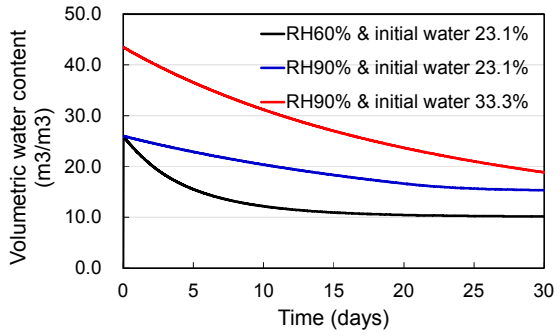


Fig. 15 Volumetric water content change

### 4.4 Influence of oxygen concentration on oil degradation

In the experiment, holes were opened for ventilation, so oxygen from the atmosphere can be regarded to be sufficient. In the above analysis cases, the initial gaseous oxygen concentration in soil, and that of the atmosphere, are assumed to be 21%. Therefore, although oxygen is consumed due to the microbial growth, sufficient supply is ensured. Herein, another case with initial and environment oxygen concentrations 2.1% is analyzed. The analysis results are shown in Fig. 16. It can be found that for the case with environment oxygen 2.1 %, the biodegradation speed is much slower than that with oxygen concentration 21%. The insufficiency of oxygen will cause the declined microbial activity. This may happen in real site for large scale underground soil. In the soil from deep underground, it is difficult for oxygen consumption to be supplied from the atmosphere. Ventilation facilities may be necessary to promote the biodegradation efficiency.

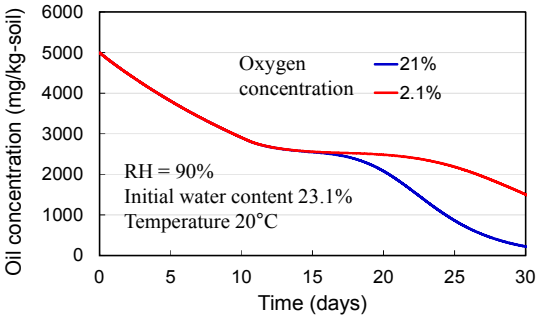


Fig. 16 Oil degradation under changed oxygen

## 5. CONCLUSIONS

In this research, in order to evaluate biodegradation effect of oil contaminated soil under environment conditions, experimental study is first conduct. In the experiment, gasoline oil is added into soil specimens made of clay loam and seed compost. Oil degradation effect with different temperature and water content is investigated. It is found that in the initial several days, oil evaporation is the dominant reason for oil concentration decline. After that, biodegradation effect plays an important role. Moreover, the specimen at 20°C shows higher biodegradation effect than that at 40°C. Initial water is another factor, and the specimen with low water content 23.1% shows higher biodegradation effect than that with 33.3%.

Based on the experiment results, an analytical analysis coupling microbial population growth and oil biodegradation with environmental conditions is established. A microbiological model is constructed, considering the growth and death rate of microbial population at different phases. The growth rate is quantified according to environmental factors, including temperature, water content, oxygen concentration, and oil concentration. The amounts of heat and water generation, and oxygen and oil consumption during microbial growth are quantified according to microbiological reaction equation. On the other hand, the transports of heat, water, oxygen and oil in soil as well as their exchanges with atmosphere are simulated using a transport sub-system, on the basis of mass or energy conservation equations. With such an analytical system, oil degradation in soil specimens are simulated using FEM method. The results are generally consistent with the experiment. Oil evaporation at early time and biodegradation are later time can be reproduced. The influences of

temperature, RH, water content and oxygen concentration on the oil degradation effect are discussed based on the analytical results.

At current stage, only soil specimens in laboratory conditions are studied experimentally and simulated using the analytical system. The final objective is to simulate and evaluate biodegradation effect of large-scale contaminated soil ground in real site. This system provide a foundation for such an evaluation, so large-scale analysis with this system will be attempted in the future research. On the other hand, still problems exist in the measurement of oil concentration in the experiment. The accuracy of measurement needs to be improved. In addition, more foundational research, such as the influences of microbial species and oil chemical composition on the degradation effect should be carried out.

**REFERENCES**


Bongochgetsakul, N. and Ishida, T., 2008. A new analytical approach to optimizing the design of large-scale composting systems, *Bioresource Technology*, 99 (6): 1630-1641.

Das, N. and Chandran, P., 2011. Microbial degradation of petroleum hydrocarbon contaminants: an overview, *Biotechnology Research International*, vol. 2011, Article ID 941810, 13 pages.

Edwards, F. and Nirmalakhandan, N., 1999. Modeling an airlift bioscrubber for removal of airphase BTEX, *Journal of Environmental Engineering*, 125(11), 1062-1070.

Fingas, M., 1998. Studies on the evaporation of crude oil and petroleum products, Journal of Hazardous Materials, 57(1-3), 41-58.

Komilis, D., Vrohidou, A., Voudrias, E., 2010. Kinetics of aerobic bioremediation of a diesel-contaminated sandy soil: effect of nitrogen addition, *Water, Air, and Soil Pollution*, 208(1-4), 193-208.

Lamy, E., Tran, T., Mottelet, S., Pauss, A. and Schoefs, O., 2013. Relationships of respiratory quotient to microbial biomass and hydrocarbon contaminant degradation during soil bioremediation, *International Biodeterioration & Biodegradation*, 83(2013), 85-91

Maekawa, K., Ishida, T. and Kishi, T., 2003. Multi-scale modeling of concrete performance: integrated material and structural mechanics, *Journal of Advanced Concrete Technology*, 1(2): 91-126.

Ministry of the Environment, Government of Japan, 2006. *Countermeasure Guideline for Oil Contamination*, Document B. (In Japanese)

Prince, R., Parkerton, T. and Lee, C, 2007. The primary aerobic biodegradation of gasoline hydrocarbons, *Environmental Science & Technology*, 41(9), 3316-3321.

Takahata, Y. and Taki, H., 2005. The current state and future development of bioremediation for the petroleum-contaminated soil. *Journal of environmental conservation engineering*, 34(4): 242-248. (In Japanese)

Zhou, E. and Crawford, R., 1995. Effects of oxygen, nitrogen, and temperature on gasoline biodegradation in soil, *Biodegradation*, 6(2), 127-140.