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EFFECT OF MOISTURE CONTENT AND CHEMICAL NATURE ON METHANE FERMENTATION CHARACTERISTICS OF MUNICIPAL SOLID WASTES

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Effects of the moisture content and the chemical nature of solid wastes on their methane production in landfill were investigated using a solid bed incubated at 41 °C. The solid-beds were filled with different moisture contents of meat, cabbage, sewage sludge cake, carrot, rice and potato. A simple model contained methane production potential, P, maximum methane production rate, R_m, and lagphase time, λ, was developed to fit the cumulative methane production curve in the batch experiment; and all the parameters in the model for various solid beds were estimated. For each proper solid waste, the lagphase time decreased, but the methanogenic activity increased with the increase of moisture content. The results reflect that the moisture content threshold limit (MS_L) for the methane production in the solid bed was about 60%.

Key Words: *anaerobic degradation, lagphase time, mathematical model, methane production, moisture content, methanogenic activity, municipal solid waste*

1. INTRODUCTION

Methane is the most abundant and stable hydrocarbon gas in the atmosphere and plays an important role in "Green house" effect[1),2)]. Methane is released into the atmosphere mainly by a variety of biogenic processes[3)]. Biogenic sources of methane production have been complied by several authors[1),2),3)]. The landfill is an important source for methane production by artificiality. In addition, because municipal solid waste is low in nitrogen and phosphorus, the methane produced in the sanitary landfill represents a usable form of energy[4),5)]. It is, therefore, necessary to clarify and control the methane production in the landfill. Several studies have been reported on parameters which may enhance methane production[6),7)] in the sanitary landfill. The moisture content is one of an important environmental factor[8)]. Barlaz[6)] evaluated the effect of moisture content on methane production in 55 gallon drums filled with shredded refuse by leachate recycle. He found that leachate recycle without neutralization stimulated the accumulation of carboxylic acids. Daily leachate neutralization stimulated methane production and the consumption of carboxylic acids. Barlaz[11)] suggested that moisture contents of 55% or higher could lead to more rapid production of methane from the landfill, and he did not always observe methane production from the refuse in which the moisture contents were less than 35%. Ghosh[8)] reported that the feeds of solid-phase fermentation process with moisture contents between about 40% and 75% could be fermented, but a moisture content of 60% or higher is preferred. However, the work of Ghosh was focused on the methane-phase fermentation.

Refuse conversion to methane is assumed to proceed on the pathway similar to that described for anaerobic sludge digestion[5]. Three major groups of bacteria are involved in methane production from refuse: (i) the hydrolytic and fermentative bacteria; (ii) the acetogenic bacteria; (iii) the methanogenic bacteria. The hydrolytic and fermentative bacteria can convert biological polymer such as proteins, lipids and carbohydrates to amino acids, long-chain fatty acids and sugars, respectively, which are then fermented to carboxylic acids, alcohols, carbon dioxide and hydrogen. According to the studies on the anaerobic digestion of wastewater and sludge, the kinetics of methane fermentation strongly depends on the chemical nature of organic matters. It has been elucidated the rate-limiting step of the overall methane fermentation for sludge is hydrolysis, but for soluble sugars, methanogenesis is the rate-limiting step[9),10)].

Municipal refuse contains many kinds of organic matters, including proteins, lipids, and carbohydrates. A typical municipal refuse in Japan is composed of 44% cellulose plus hemicellulose, 13% lignin, 3% protein and 2% lipid[12]. It is necessary to clarify the degradation behavior of each component in landfill. However, the characteristics of methane fermentation for various substance at a wide range of moisture contents have not been well understood yet.

The objective of this study was to investigate the effect of moisture content and chemical nature on methane fermentation characteristics of the organic fraction of municipal solid wastes. For this purpose, a total of six kinds typical solid wastes including meat, cabbage, sewage sludge cake, carrot, rice and potato were selected as experimental materials, and their moisture contents were ranged from 86 to 98%. Furthermore, a simple model including the parameters of methane production lagphase time, rate and potential was developed to assess the capacity and the characteristics of the organic fraction of municipal solid wastes converting to methane.

2. MATERIALS AND METHODS

(1) Solid bed bottle

Municipal solid wastes (MSW) have a complex composition including various organic and inorganic compounds. In order to investigate the effects of the organic fraction of municipal solid wastes on the methane production, a total of six different organic matters, including meat, cabbage, carrot, rice, potato, and sludge cake were used as the main components.

In this study, the solid-bed, which was a vial of 120 filled with the solid wastes and seeded with digestion sludge, was used to measure the methane production. Except for rice, the size of each individual solid waste was about 2 cm. The seed sludge used for this study was taken from a ten-liter laboratory digester, operated at the temperature of 37±1 °C and the HRT of 20 days. This digester has been run over one year by feeding sludge cake. The solids concentration in the digester was maintained at 3 - 4 *wt. %*. The sludge cake was conditioned with lime and obtained from a municipal sewage treatment plant at Sendai City, Japan. For each vial, 40 grams of the same seed sludge and a proper quantity of organic matters, as listed in **Table 1**, were added, and then filled up to 80 grams with deoxygen incubation media. The composition of incubation media was listed in **Table 2**[13], where resazurin was used to detect oxygen contamination, sodium sulfide and L-cysteine were added to provide the reducing environment. A mixture gas containing 80% N_2 and 20% CO_2 was used as a headspace gas. Since the temperature in the decomposition of refuse in landfills generally range from 35 °C to 42 °C[12] and the optimum temperature for methane production is 41 °C[14], the 41 °C of incubation temperature was used in this study. The solid-beds were then incubated at a rotary cell culture and rotated at 1.5 rpm for expecting to stimulate both the hydrolytic and methanogenic bacteria by providing better contact among samples, nutrients and microorganisms. The volumetric measurement of biogas was conducted with glass syringes (5 - 50 mL depending on gas volume) as Wen's approach[15]. The sample syringe was initially flushed with the

Table 1 The fraction of organic matters and seed sludge employed.

Composition	Moisture contents of solid-beds (%)						
Sludge cake	97.2	94.4	91.6	88.8	86.0	83.2	77.7
Meat	96.0	94.0	92.2	90.0	88.0	86.0	-
Cabbage	98.1	97.8	97.3	97.0	96.6	96.3	95.5
Carrot	98.0	97.6	97.2	-	96.3	95.9	95.0
Rice	97.1	95.8	94.4	93.0	91.6	90.3	87.5
Potato	88.6	87.3	85.9	84.5	83.1	87.8	79.0
Weight of solid wastes (g)	5	10	15	20	25	30	40
Seed sludge (g)	40	40	40	40	40	40	40
Media (g)	35	30	25	20	15	10	0

Table 2 The composition of media.

Components	Concentration	
KH_2PO_4	0.4	$g{\cdot}L^{-1}$
K_2HPO_4	0.4	$g{\cdot}L^{-1}$
NH_4Cl	1.0	$g{\cdot}L^{-1}$
Mineral solution[(a)]	10.0	mL
Vitamin solution[(b)]	10.0	mL
$NaHCO_3$	4.0	$g{\cdot}L^{-1}$
$MgCl_2{\cdot}6H_2O$	0.21	$g{\cdot}L^{-1}$
Cysteine $HCl{\cdot}H_2O$	0.5	$g{\cdot}L^{-1}$
$Na_2S{\cdot}9H_2O$	0.25	$g{\cdot}L^{-1}$
Resazurine	0.002	$g{\cdot}L^{-1}$
pH	7.0-7.2	

(a) Contains, in grams per liter of distilled water: nitrotriacetic acid, 4.5; $FeCl_2{\cdot}4H_2O$; $CoCl_2{\cdot}6H_2O$, 0.12; $Alk(SO_4)_2$, 0.01; NaCl, 1.0; $CaCl_2$, 0.02; Na_2MoO_4, 0.01; $MnCl_2{\cdot}4H_2O$, 0.10; H_3BO_3, 0.01; $CuSO_4{\cdot}5H_2O$, 0.01; $NiCl_2{\cdot}6H_2O$, 0.02.
(b) Contains, in milligrams per liter of distilled water: biotin, 2; folic acid, 2; pyridoxine HCl, 10; thiamine HCl, 5; riboflavin, 5; nicotinic acid, 5; DL-calcium pantothenate, 5; vitamin B_{12}, 0.1; *p*-aminobenzoic acid.

mixture gas. Syringes were horizontally held for gas measurements and all the readings were taken at the incubation temperature. Volume measurements were made by allowing the syringe plunger to move and equilibrate between the vial and atmospheric pressures. Readings were verified by drawing the plunger to the equilibrium point. On releasing, the plunger should be returned to the original equilibrium volume. In order to continue the assay, the gas was removed for wasting.

(2) Analytical method

The percentage of methane and carbon dioxide in biogas were analyzed by a gas chromatograph (Shimazu 8A) equipped with a thermal conductivity detector and a 2 m stainless column packed with activated carbon (60/80 mesh). The operational temperatures of the injection port, the oven and the detector were 140, 120 and 140 °C, respectively. Helium was used as the carrier gas at a flow rate of 30 $mL{\cdot}min^{-1}$. Concentration of volatile fatty acids (VFAs) was determined by another gas chromatograph of the same model equipped with a FID detector and a 2 m glass column packed with KOCL-FM (60/80 mesh). The operational temperatures for the injection port, the oven and the FID detector were 160, 140 and 160 °C, respectively. Before the analysis of VFA, the phosphoric acid was added to control the pH of samples.

(3) Development of model fitting a cumulative methane production curve

To describe a bacterial growth curve in a batch culture, many mathematical models have been suggested[16), 17)]. Among them, the Gompertz equation was recently found to be the most suitable model[18)], which is written as:

$$y = \int_0^t r_g = A \cdot \exp\{-\exp[\frac{\mu_m \cdot e}{A}(\lambda - t) + 1]\} \qquad (1)$$

where, y is population size,
r_g is bacterial growth rate,
A is asymptotic phase,
μ_m is maximum growth rate,
λ is lagphase time, and
e is *exp (1).*

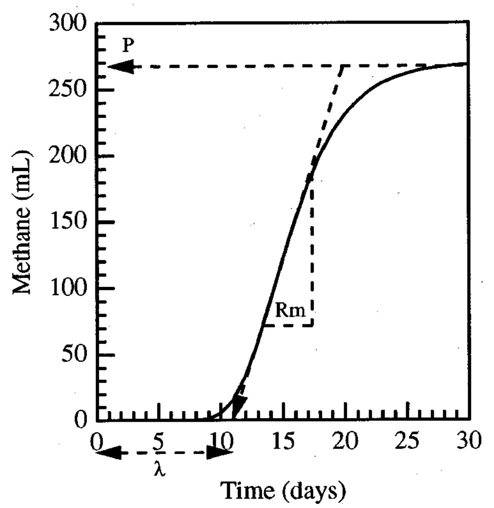

Fig.1 The schematic methane production curve was described by the modified Gompertz equation.

The relationship between the bacterial growth rate and the substrate utilization rate can be defined by the following equation.

$$r_g = Y_1(-r_{su}) \qquad (2)$$

where, Y_1 is maximum yield coefficient,
r_{su} is substrate utilization rate.

On the other hand, the relationship between the substrate utilization rate and the methane production rate is:

$$-r_{su} = Y_2 \cdot r_m \qquad (3)$$

where, r_m is methane production rate,
Y_2 is maximum yield coefficient.

According to Eqs. (2) and (3), the rate of methane production can be defined as follows:

$$r_m = \frac{r_g}{Y_1 \cdot Y_2} \qquad (4)$$

Therefore, the cumulative methane production, M, can be defined as:

$$M = \int_0^t r_m = \int_0^t \frac{r_g}{Y_1 \cdot Y_2} = \frac{1}{Y_1 \cdot Y_2}\int_0^t r_g \qquad (5)$$

If the term $\int_0^t r_g$ is substituted in Eq. (1), the resulting expression for the cumulative methane production is:

$$M = \frac{A}{Y_1 \cdot Y_2} \cdot \exp\{-\exp[\frac{(\mu_m / Y_1 \cdot Y_2) \cdot e}{(A / Y_1 \cdot Y_2)}(\lambda - t) + 1]\} \qquad (6)$$

The term A/Y_1Y_2 can be replaced by the term P, defined as methane production potential, while μ_m/Y_1Y_2 can be defined as maximum methane production rate. Therefore, the resulting final expressions is:

$$M = P \cdot \exp\{-\exp[\frac{R_m \cdot e}{P}(\lambda - t) + 1]\} \qquad (7)$$

In Eq. (7), P, R_m and λ are the important parameters affecting the characteristics of methane production. **Fig.1** illustrates the typical curve of cumulative methane production with P, R_m and λ.

3. RESULTS AND DISCUSSION

(1) Model analysis

Based on the experimental data on the cumulative

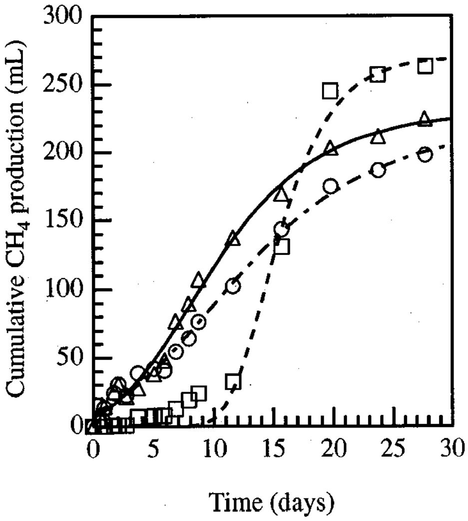

Fig.2 The nonlinear regression fit of the modified Gompertz equation on the cumulative methane productions obtained from the solid beds.

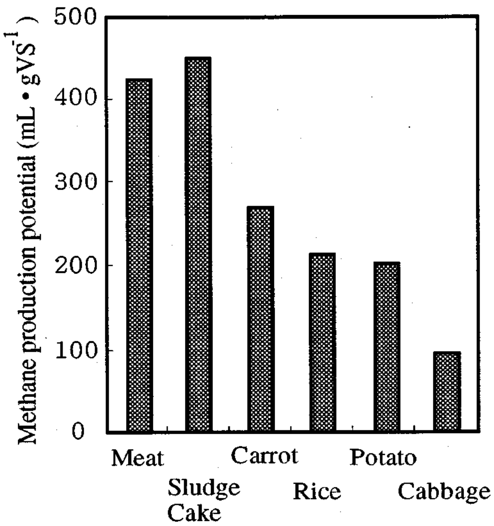

Fig.3 Specific methane production potential of the organic fraction of municipal solid waste.

methane production, all parameters in Eq. (7), P, R_m and λ, were evaluated by the Newton method and judged by the diagnosis procedure including the *Student-t* test, *D.W.* test and *K.S.* test according to the approach reported by Wen *et al.*[15]. The best values of the parameters were used to fit the model of Eq. (7). As an example, **Fig.2** shows the fitting curves using the best values of the parameters and Eq. (7). Both the curve fitting and statistical analysis demonstrated that Eq. (7) was suitable to describe the progress of cumulative methane production on the anaerobic digestion.

(2) Methane production potential of municipal solid wastes

Fig.3 shows the average methane production potential of each municipal solid waste under various levels of moisture contents used in this study. The results reveal that the methane production potentials of meat, cabbage, carrot, rice, potato and sludge cake are 424, 96, 269, 214, 203 and 450 mL•gVS^{-1}, respectively. Based on the methane production potential, the municipal solid waste can be divided into three groups: (i) meat and sludge cake; (ii) carrot, rice and potato; (iii) cabbage. The main chemical components of the group (i) are proteins and lipids, while the main chemical components for the group (ii) and (iii) are starch and cellulose, respectively[19),20)]. These results demonstrate that the methane production potential of the organic fraction of municipal solid waste depended on their chemical nature.

(3) Effect of moisture content on lagphase time

Due to its highly uncertain characteristics, the lagphase time is usually neglected in kinetics analysis of a batch experiment. In this study, the lagphase times at various moisture contents of different solid wastes were exactly evaluated using Eq. (7). The effect of moisture content on the lagphase time is shown in **Fig.4**. Overall, for each individual solid waste, the lagphase time decreased with the increase in the moisture content. It has been reported that the period of lagphase was influenced by the initial concentrations of microorganisms and substances stimulating the growth of bacteria[21)]. In this study, since the same quantity of seed sludge and the same size of the samples were used in each batch experiment, the change of lagphase time for the same kinds of solid wastes at different moisture contents was mainly caused by the change of moisture content. For expample, the lagphase time for sludge cake was 6 days at the moisture content of 97%, but was more

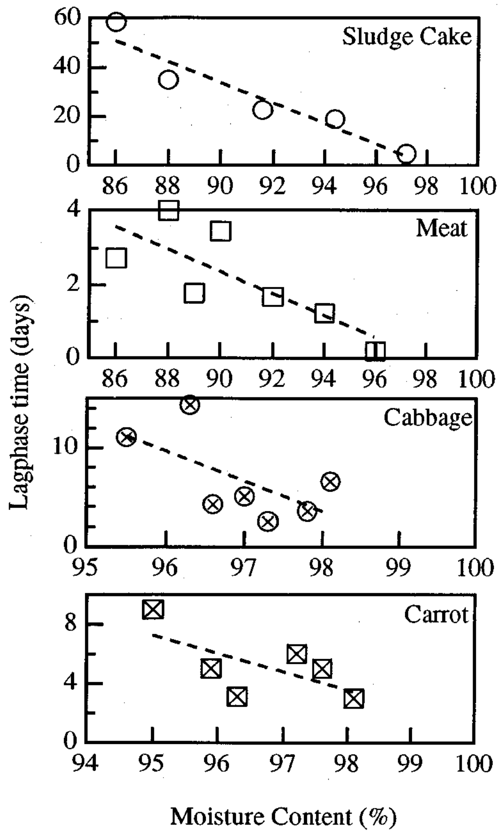

Fig.4 Effects of lagphase time on the moisture content of the solid wastes.

than one month at the moisture content of 88%. These results are similar to the studies of Ghosh[8] and Barlaz[11] on the high-solids bed and the sanitary landfill, respectively, indicating the moisture content is able to stimulate methane production. Apparently, moisture content is an important factor in affecting the degradation characteristics of sludge cake.

On the other hand, the solid-beds contained with rice and potato had not any methane production during the incubation period. The experimental results show that the total VFA level in the solid-beds of rice and potato was higher than 4000 mg•L^{-1}, and as a result, the pH was lower than 3. Based on the study of Cheng *et al.*[22], the concentration of organic acids higher than 2,000 mgCOD•L^{-1} could inhibit the activity of methanogens. It means that the growth of methanogens in the solid-beds of rice and potato were inhibited by the high concentration of organic acids.

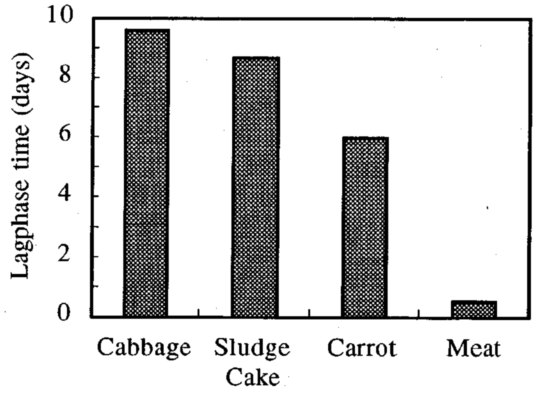

Fig.5 The lagphase time relationship among cabbage, sludge cake, carrot and meat at the moisture content of 96%.

Fig.5 shows that the lagphase times of meat, carrot, sludge cake and cabbage at the same moisture content of 96% were respectively 0.6, 6.0, 8.7 and 9.6 days, indicating that the lagphase time was also influenced by the chemical nature of solid wastes at the same moisture content.

(4) Effect of moisture content on maximum methane production rate

Fig.6 shows the effects of the moisture contents on the maximum specific methane production rates, R_m, in the batch experiment for various kinds of solid wastes. Because the same quantity of proper seeding was used in each batch experiment, the maximum specific methane production rate at each condition represents a methanogenic activity at that condition. Overall, the R_m increased with the increase in the moisture content in the range conducted by this study. This may indicate that the moisture content threshold limit, MS_L, on the methane production of sludge cake, meat, carrot and cabbage were 57%, 80%, 88% and 95% respectively. The MS_L of sludge cake was 57% which is similar to the value of 60% for methane fermentation in a solid bed and a landfill reported by Ghosh[8] and Barlaz[6], respectively. However, in this study, the MS_L for meat, carrot and cabbage were around 80%, 88% and 95 %, respectively, which are significantly higher than 60%. This apparent difference was caused by the chemical na-

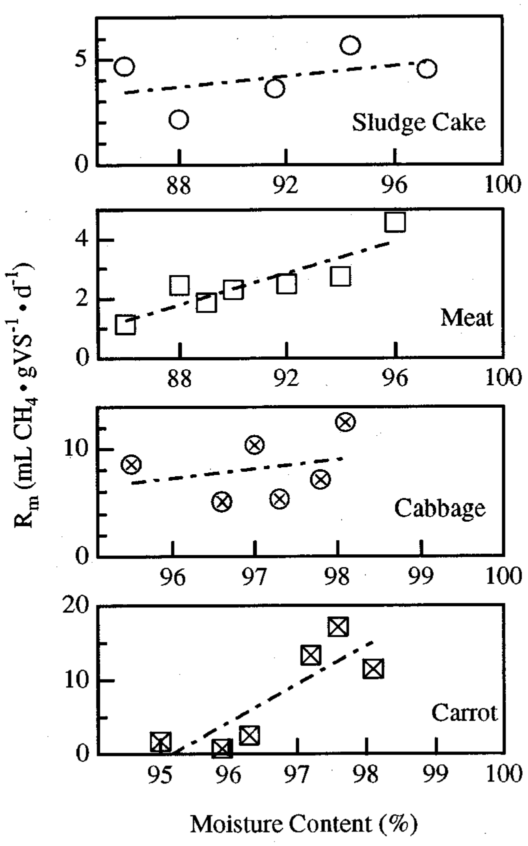

Fig.6 Effects of specific methane production rate on the moisture content of the solid wastes.

ture, such as the degradation characteristics and pH buffer ability of solid wastes. Because lime was used as the coagulant aid for sludge dewatering, the solid-beds of sludge cake had a high capacity to buffer organic acids and reduce the inhibition of VFA. Thus, the MS_L on the solid-bed of sludge cake was caused by moisture content. However, the pH values of the solid-beds for meat, carrot and cabbage were respectively 5.0, 4.8 and 5.2. At such condition, methane production was inhibited by pH and high level of organic acids. For these cases, the MS_L was mainly caused by organic acids, but not moisture content.

4. CONCLUSIONS

The principle conclusions derived from this investigation are as follows:

(1) A simple model was developed to describe the cumulative methane production curve in the batch culture. By using this model, key parameters in a cumulative methane production, lagphase time, maximum methane production rate, and methane production potential can be exactly estimated based on the experimental data.

(2) The methane production potential of the organic fraction of municipal solid wastes depended upon their chemical nature. Each gram VS of the solid wastes of sludge cake, meat, carrot, rice, potato and cabbage had methane production potential of 450, 424, 269, 214, 203 and 96 mL, respectively.

(3) For each solid waste, the lagphase time of methane fermentation decreased with the increase in the moisture content, and a linear relationship between lagphase time and moisture content was obtained in the moisture contents ranged from 88 - 98%.

(4) The methanogenic activity of the solid-bed decreased with the decrease in the moisture content. The moisture content threshold limit, at which activity dropped to zero, was found to be 56.6% for sludge cake, but rather high more than 80% for meat, carrot and cabbage. In latter cases, the methanogenic activity was inhibited by the high level of organic acids rather than moisture content.

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都市廃棄物のメタン発酵特性に及ぼす有機物の種類と含水率の影響

頼　俊吉・李　玉友・野池達也

本論文は，都市ごみのメタン発酵に及ぼす有機物の種類とその含水率の影響を解明することを目的として，肉，キャベツ，下水汚泥脱水ケーキ，ニンジン，ライスおよびジャガイモの6種類の物質を選択し，それぞれの含水率を変化させてメタン発酵の回分実験を行い，解析を加えたものである．モデル実験装置としてのバイアルびんは41℃で培養した．回分培養における累積メタンガス生成曲線を表す数学モデルを開発した．同モデルを用いることで，各実験条件における累積メタンガス生成曲線をよく表現でき，またそれぞれの条件における最大メタン生成量，誘導期及び最大メタン生成速度の三つのパラメーターをに求めることができた．これらの動力学パラメーターに対する有機物の種類とその含水率の影響について考察した．

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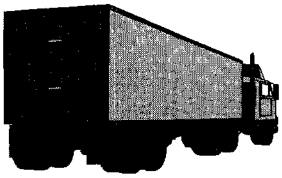

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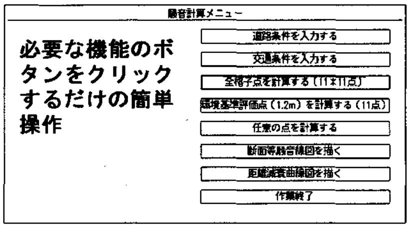
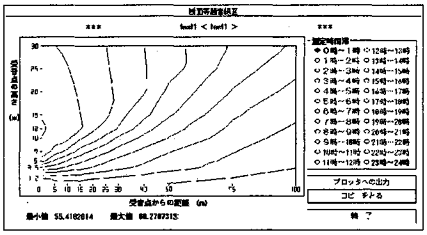
■道路条件・交通条件を独立して設定・保存可能なので多くのケースを計算する場合に効率的です。

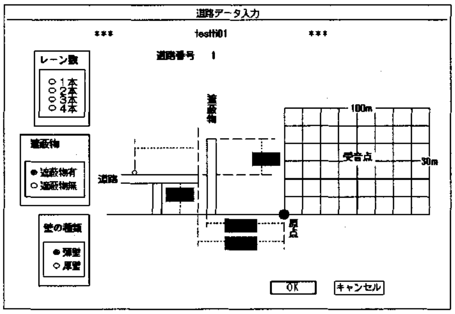
■計算結果はＡ４版の美しい帳票形式で印刷されるので、そのまま報告書などに用いる事ができます。

■等騒音値線図（コンター）・距離減衰図はプリンターの他ペンプロッターにも出力可能（ＨＰＧＬ対応プロッターが必要）

■購入しやすい低価格を実現

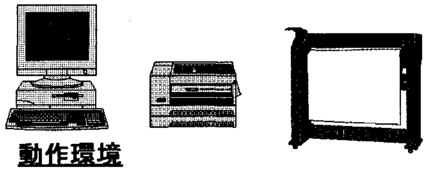






時間帯別断面コンター（上）

道路条件の入力画面（左）



動作環境

Windows3.1/95の動作するパーソナルコンピューター
メモリー８ＭＢ以上、ディスク空き容量１０ＭＢ以上を推奨
Windows3.1/95に対応するプリンター
オプションとして、
ＨＰＧＬ対応プロッターに出力可能

□お問合せ・資料請求は

Windowsは米国マイクロソフト社の登録商標です。

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TEL 06-241-4126

電子メールによるお問合せ・資料請求も受付けております。（mabe@c

CIVAS;Civil Engineering Analysis Service
CRC総研がお届けする土木・建設分野向けサービス

地盤解析には　3次元地盤FEM解析プログラム　for Windows®

Mr.SOIL-3D 95

[Mr.SOIL-3D for Windows 95]
[トンネル掘削による主応力コンター図]

450本を超える販売実績をもつ地盤FEM解析の定番ソフトです。3D解析を可能にしており、FEM解析初心者でも、使い易い3Dプリポストプロセッサも完備。もちろん2D解析も可能で非線形弾性解析(電中研法、Duncan-Chang法)、弾塑性解析(歪み軟化、硬化)等の多岐にわたる解析が選択できます。ステップ解析機能を有し、施工順序に従って地盤の挙動を把握できます。

'97.1 リリース

'96.9 リリース

地下水解析には　広域地下水変動解析プログラム　for Windows®

PC/UNISSF 95

[UNISSF for Windows 95]
[掘削に伴う水位低下コンターと流速ベクトル図]

データ作成、結果の表示等のFEM解析に必要な諸作業がすべてマウスで行える強力なプリポスト機能を備えた浸透流解析ソフトです。準3次元解析版と断面2次元解析版が有ります。工事施工時の周辺地下水影響解析等の定常／非定常解析が可能です。降雨・揚水井・浸出面が取り扱え、水位・流量の経時変化に加え、浸出点の位置、流量を求めることができます。

実力の解析ツール群 続々登場!

連成解析には　応力・浸透・熱連成解析プログラム　for Windows®

CONHEAT 95

[CONHEAT for Windows 95]
[地中温排水パイプ周りの温度と流速分布図]

圧密解析から、応力・浸透・熱の連成問題まで解析が可能な2次元FEM解析ソフトです。多段掘削・盛土や降雨条件が扱え、経時観測記録より、変形・透水係数を逆解析で求めることができます。弾性・非線形弾性・弾塑性・弾粘塑性を示す地盤に適用でき、凍結についても適用可能です。プリポスト完備で優れたユーザーインターフェースを実現しています。

'96.11 リリース

'96.11 リリース

地質解析には　3次元地質解析プログラム　for Windows®

GEORAMA 95

[GEORAMA for Windows 95]
[3次元地質モデル図]

EWS版で好評を得た3D地質解析ソフトのWindows95対応版です。地形データやボーリングデータ等の情報から地質面を3次元的に推定後、3D地質モデルを構築し、任意断面及び、3Dの各種地質関連図面を出力表示します。人手に頼っていた地質図作成、地質モデルデータベース管理等に大幅なコストダウンをもたらします。計画立案等のプレゼンテーションを強力にサポートします。

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新製品リリース時に発売キャンペーンを実施!
キャンペーン期間中、新規ユーザー向け特別価格&旧バージョンユーザー向け優待価格を設定。
また、上記プログラムを用いての受託解析サービスも展開。詳しくは左記までお問合わせください。

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