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Optimization of Conditions of Digested Sludge Using Landfill Biocover Material for Enhance Methane Oxidation

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Abstract. The modified digested sludge (MDS) can be used as landfill biocover that serve to minimize CH_4 emissions by optimizing CH_4 oxidation. MDS composited by coal ash and digested sludge was studied in this paper. The permeability coefficient and the compressive strength of MDS were tested in full factorial $L_9(3^4)$ experiment using simulation column. Results show that when the mix ratio of coal ash and digested sludge is about 1:1-1.5:1, the permeability coefficient < 10^{-4} cm/s and compressive strength ≥ 50 kPa, MDS can meet the engineering requirements for landfill operation. The optimal conditions of MDS were as follow: mix ratio of coal ash/digested sludge = 1:1, nutrient solution = 0.05 ml/g, thickness of biocover = 400mm, and moisture content of MDS = 40%. The maximum CH_4 oxidation efficiency was 88.6% and the volume flow rate of CH_4 was 0.75 mmol/(kg•d) under the optimal conditions.

Introduction

Landfill gas is generated by anaerobic decomposition of organic waste. It is mainly composited by methane (CH_4 50-65%) and carbon dioxide (CO_2 35-50%) [1, 2]. CO_2 formed inside landfills and released into the atmosphere produces a negligible effect on the environment due to the global warming potential (GWP). However, the effect of CH_4 is 23 times that of an equal mass of CO_2 over a period of 100 years. Concentration of CH_4 in atmosphere has more than two times over the last 150 years. It is estimated that CH_4 will become No.1 green gas which get the heel of CO_2 in 2030 [3, 4]. Therefore, even a small reduction in anthropogenic CH_4 emissions would be significant.

A great quantity of literature demonstrated that biocover with rich microorganism has high CH_4 oxidation capacity [5-7]. Such as composts, digested sludge and aged refuse all have the capable of oxidizing CH_4 [8-11].

The aim of this study is to screen out an effective, economical and environmental biocover for landfill operation. A group of $L_9(3^4)$ orthogonal array experiments were used to find out the optimum conditions of MDS. Mix ratio of digested sludge and coal ash, permeability coefficient and compressive strength of MDS were tested through simulation column experiment.

Methods and materials

Materials. Digested sludge was obtained from a sewerage treatment plant in Guangzhou, coal ash was provided by Trading Co.Ltd. in Guangzhou. The chemical compositions of slag were listed in Table 1.

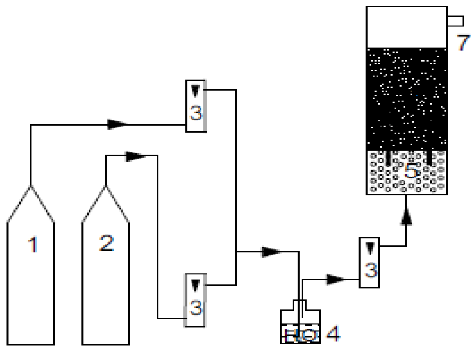
Table 1. The chemical and physical properties of each material

Project	Moisture content (%)	Organic content (%)	pH	Electric conductivity (μs/cm)
Coal ash	0.37	4.13	8.81	591.0
Sludge	73.32	24.72	7.05	1046.0

Methane gas (99.9%) and carbon dioxide (99.5%) were purchased from a chemical reagent factory in Guangzhou. The nutrient solution is the nutrient of methanotrophic bacteria. A liter nutrient solution includes 0.85 g $NaNO_3$, 0.53 g KH_2PO_4, 2.17 g Na_2HPO_4, 0.037 g $MgSO_4•7H_2O$, 0.17 g K_2SO_4, 0.007 g $CaCl_2•2H_2O$, 0.5 ml 1 mol/L H_2SO_4, 11.2 mg $FeSO_4•7H_2O$, 2.5 mg $CuSO_4•5H_2O$ and 2 ml microelement solution. A liter microelement solution includes 0.204 g $ZnSO_4•7H_2O$, 0.223 g $MnSO_4•4H_2O$, 0.062 g H_3BO_3, 0.048 g $Na_2MoO_4•2H_2O$, 0.048 g $CoCl_2•6H_2O$ and 0.083 g KI.

The initial moisture content was tested by electrothermal blowing dry box (101A-2E, from Shanghai AnTing). Organic content was tested at 600℃ for constant weight by muffle (SX3-3-10, Hangzhou ZhuoChi). The pH were test by pH meter (Phs-25, Shanghai YingGe), electric conductivity was determined by conductivity meter (DDB-303A, Shanghai LeiCi).

Device of the Experiment. The experiment setup is shown in Fig. 1. The PVC column was 1000 mm in length and 150 mm in diameter. Each PVC column had two layers: a lower layer which consisted of 200 mm of crushed gravel (20 mm in diameter) as a gas dispersion layer, in turn, an upper layer which consisted of biocover. The perforated plate (10 mm in aperture) and geotextile placed over the crushed gravel layer which prevents biocover to jam gas dispersion.



1. CH_4 2. CO_2 3. rotor flow meter 4. humidifier 5. crushed gravel layer 6. biocover 7. outlet

Fig. 1. Apparatus for the experiments

MDS was packed in each column. CH_4 and CO_2 were mixed in a ratio of 1:1 using rotor flow meter. After increasing their humidity by a humidifier, the gas mixture is injected in each column at a rate of 20 ml/min for 60 min, and then the column inlet was tightly closed. The concentration of headspace gas was monitored over time by taking samples from the outlet, using a gas tight 1.0 cm^3 syringe. CH_4 and CO_2 concentration was direct determined using GC-6890N gas chromatograph with hydrogen flame ionization detector. The gasification chamber temperature is 100℃ and the detector temperature is 150℃.The oven temperature is initially kept at 70℃ and gradually increased to 80℃ at the speed of 2.5 ℃/min. The runtime is 5 min. Standard gas mixtures having known concentration of CH_4 and CO_2 are used for standard curve. Maximum CH_4 oxidation efficiency and V(CH_4) were used to evaluate the efficiency of methane oxidation:

$$\text{Maximum } CH_4 \text{ oxidation efficiency} = \frac{(C_{CH_4})_{t,0} - (C_{CH_4})_{t,i}}{(C_{CH_4})_{t,0}}; \tag{1}$$

$$V(CH_4) = \frac{n_{CH_4}}{d \cdot m}; \tag{2}$$

[a] $(C_{CH_4})_{t,0}$ is CH_4 concentration (% V/V) at the beginning of the experiment, $(C_{CH_4})_{t,i}$ is CH_4 concentration (% V/V) at the end of the experiment.

[b] n_{CH_4} is the amount of substance of CH_4 consumpition, d is the days of experiment, m is biocover weight.

Detecting of compressive strength and permeability coefficient. The compressive strength of samples was measured by a machine (YS-500 type). The permeability coefficient of samples was measured by simulated rainfall experiment. The permeability was calculated by the following formula:

$$\text{permeating coefficient}(\text{cm/s}) = \frac{\text{total infiltration quantity (mL)}}{\text{barre Cross - Section (cm}^2)} \cdot \frac{1}{\text{penetration time (s)}} \quad (3)$$

Coverage optimization of material conditions. The oxidation capacity of CH_4 depends on both the physical and the chemical properties of the biocover. A group of $L_9(3^4)$ orthogonal array experiments were used to find out the optimum conditions of MDS (Table 3).

Results and discussion

Compressive strength and permeability coefficient. The engineering requirement for compressive strength of biocover was stronger than 50 kPa. Fig.2 shows that when the mix ratio is in the range of 1:1-1.5:1, MDS compressive strength increase with the addition of coal ash. When the mix ratio of coal ash/digested sludge >1.5, the compressive strength presents descend. It is because the coal ash addition changes the water content of digested sludge, and indirect changes its compressive strength.

Table 2. Osmotic coefficient of coal ash and sludge in various mixing ratios

Mix ratio（coal ash/digested sludge）	1:0	0.5:1	1:1	1.5:1	1.75:1	2:1	2.5:1
Permeability Coefficient (cm/s)	6.65-2.78 $\times 10^{-8}$	5.94 $\times 10^{-6}$	3.71 $\times 10^{-5}$	1.01 $\times 10^{-4}$	4.29 $\times 10^{-4}$	6.30 $\times 10^{-4}$	1.53 $\times 10^{-3}$

The engineering requirements for osmotic coefficient of biocover was less than 10^{-4} cm/s. Table 2 shows that the permeability coefficient of MDS improved with the addition of coal ash. The permeability coefficient of MDS is 1.01×10^{-4} cm/s when the coal ash is mixed with digested sludge ratio is 1.5:1. MDS can meet with the demands of the permeability requirements of the biocover. So MDS mix ratio of coal ash and digested sludge was studied at the levels of 1:1, 1.25:1 and 1.50:1. Three other significant factors (moisture, nutrient addition and thickness of covers) and their levels were selected based on the experimental of other authors [7-11].

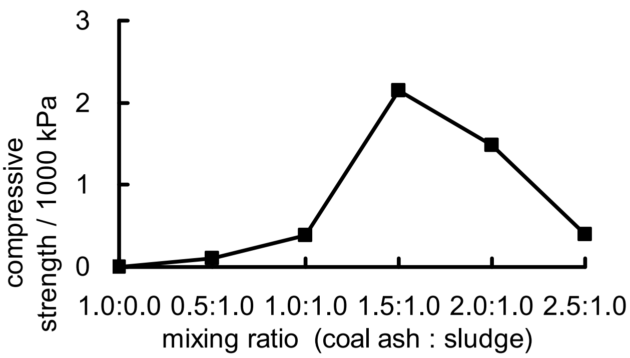


Fig. 2. Bearing capacity of coal ash and sludge in various mixing ratios

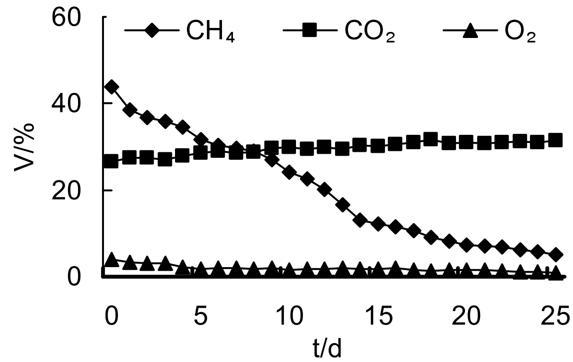


Fig. 3. Changes in gas concentration under optimat conditions

The result of orthogonal method and the range analysis. Levels of each operating factor and the design matrix of the $L_9(3^4)$ orthogonal array were summarized in Table 3. It indicated the different methane oxidation efficiency varying from different chemical and physical properties of materials. The maximum CH_4 oxidation efficiency was 81.88% in No.3 experiment, where mix ratio of coal ash/digested sludge was 1:1, 0.10 ml/g nutrients were added to 400 mm thickness landfill biocover that contained 40% moisture. However, the max V(CH_4) was 1.02 mmol/(kg·d) in No.9 experiment, where mix ratio was 1.5:1, 0.05 ml/g nutrients were added to 200 mm thickness landfill biocover that contained 40% moisture. All factors definitely influence methane oxidation ability but the degree of influence is the issue. By comparing R of maximum CH_4 oxidation efficiency, the degree of influence is mixing ratio > nutrient addition > cover thickness > moisture content. The optimal conditions were $A_1C_2D_3B_3$ where mixing ratio was 1:1, 0.05 ml/g nutrients were added to 400 mm thickness landfill biocover that contained 40% moisture. However, by comparing R of $V(CH_4)$, the degree of influence is cover thickness > mixing ratio > nutrient addition > moisture content. The optimal conditions were $D_1A_1C_2B_3$ where mixing ratio was 1:1, 0.05 ml/g nutrients were added to 200 mm thickness landfill biocover that contained 40% moisture. Because the importance of each target was different, each parameter had various effects on each target, accordingly optimal condition was different.

The effect of each factor on CH_4 oxidation ability. In Table 3, R(E) of mixing ratio is 31.1, which indicated that the mix ratio of coal ash/digested sludge was a critical physical parameter affecting CH_4 oxidation ability. The best mixing ratio of coal ash and digested sludge is 1:1.

The larger grain diameter of material, the faster vertical speed of gas, then it may be enhancing methane oxidation efficiency. And the digested sludge is rich in microorganism but coal ash is only inert substance, the more digested sludge contained in biocover, the more microorganisms in material. It was clear that increasing microbial activity can improve methane oxidation efficiency.

Table 3. L_9 (3^4) orthogonal experiment and the resulting amylase measurements

Experiment No.		A [Mixing ratio]	B [Moisture]	C [Nutrient addition (ml/g)]	D [(Cover thickness (mm)]	E[Maximum CH_4 oxidation efficiency (%)]	F[$V(CH_4)$b mmol/(kg•d)]
		Operating factors and their levels				Mean responses	
1		1:1	20	0.00	200	68.1	1.08
2		1:1	30	0.05	300	77.3	0.79
3		1:1	40	0.10	400	81.9	0.46
4		1.25:1	20	0.05	400	56.4	0.40
5		1.25:1	30	0.10	200	37.6	0.79
6		1.25:1	40	0.00	300	40.2	0.39
7		1.5:1	20	0.10	300	49.4	0.54
8		1.5:1	30	0.00	400	56.2	0.36
9		1.5:1	40	0.05	200	63.0	1.22
A	k_1	75.8	58.0	54.8	56.2		
	k_2	44.7	57.0	65.6	55.6		
	k_3	56.2	61.7	56.3	64.8		
R(E)		31.1	4.7	10.8	9.2		
B	k_1	0.78	0.67	0.61	1.03		
	k_2	0.53	0.65	0.80	0.57		
	k_3	0.71	0.69	0.60	0.41		
R(F)		0.25	0.04	0.20	0.62		

Results from the R(E) of moisture content (Table 3) shows that a minor effect on biocover at various moisture contents, the wetter the material the higher methane oxidation. It was hypothesized that when the moisture content was low, which led to water from the bacterial cells to flow out and eventually the death of the microbial population [12]. But higher moisture affects the movement of gas through the landfill and the microbial growth and made the compressive strength of material was low. Based on an overall consideration of various factors, best moisture content is 40% in this study.

The effect of nutrient on CH_4 oxidation ability was presented in Table 3, it was clear that adding nutrients positively affected CH_4 oxidation ability, but it was not the more the better. Considering the largest CH_4 oxidation ability and the lowest cost, the best nutrient addition was 0.05 ml/g in this study.

The effect of layer thickness is also shown in Table 3. It indicated that the effect of cover thickness on the maximum CH_4 oxidation efficiency and $V(CH_4)$ followed the different trend. The 400mm layer thickness had the higher maximum oxidation than 300mm and 200mm layer thickness, but it had crosscurrent on $V(CH_4)$. Zhao [13] reported that the landfill had a larger volume of biocover, larger population of methanotrophic bacteria and thus greater bacterial activities, resulting in higher CH_4 oxidation efficiency values. The cover thickness should meet economic benefit and engineering requirements. This study suggests selecting 200 mm as daily cover and 400 mm as intermediate cover.

Methane oxidation efficiency in the landfill biocover under optimal conditions. Methane oxidation efficiency in the optimal conditions is shown in Fig. 3. CH_4 concentration in the headspace gas dropped from around 43.9% to 5.0% in 25d, the CO_2 concentration increased from 26.6% to 31.4% and CH_4 oxidation efficiency was up to 88.6%. This indicates that the conditions from $L_9(3^4)$ orthogonal experimental were optimal for methane oxidation. Hilger and Humer [5] reported that methanotrophs consume CH_4 and oxidize it to CO_2 and water for energy yield. The decrease in CH_4 concentration and the increase in CO_2 concentration in the headspace gas were clear indications that there was CH_4 oxidation process.

Conclusions

The amount of methane released at the top of the landfill can be reduced on its way through biocover. In this study, CH_4 oxidation efficiency was different under different physical and chemical of material and environmental factors. The moisture content is a minor physical parameter affecting the methane oxidation ability, but mix ratio of coal ash / digested sludge, nutrient solution addition and cover thickness play a significant role in methane oxidation. The maximum CH_4 oxidation efficiency and V (CH_4) were 88.6% and 0.75 mmol/(kg•d) under the optimal conditions. This study demonstrates that MDS can be use as an effective, economical and environmental biocover material for landfill operation.

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