

IMPACT OF HYDROPHOBIC BIOCHAR LANDFILL COVER SOIL ON METHANE OXIDATION

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Abstract: To achieve the carbon neutrality goal, the reduction of CH₄ from landfills needs to be considered. Bio soil landfill cover can achieve efficient CH₄ removal by adsorption and microbial oxidation, but in the engineering application stage, rainwater entry was found to significantly reduce CH₄ and O₂ diffusion. The air permeability and hydraulic conductivity of the landfill cover soils with the addition of hydrophobic biochar were improved and the methane reduction potential was reduced. hydrophobic biochar-amended soil cover simulation column (RH) was prepared and the biochar-amended soil cover simulation column (RB) was used as a control to study their CH₄ emission reduction characteristics. CH₄ oxidation simulation column tests manifest CH₄ oxidation enhancement on the capacity of RB and RH showed that the optimal CH₄ influx gas concentration was 35%, the optimal simulated landfill gas influx rate was 10 mL/min, and the optimal temperature was 30°C. The methane removal rate of RB can reach 99.96%. Overall, the use of hydrophobic biochar as a cover soil amendment to reduce methane emissions from landfills appears to be a promising alternative to conventional soil covers.

INTRODUCTION

Landfill disposal of municipal solid waste represents one of the largest anthropogenic global methane emission sources^[1-7]. Open landfills were found to represent 91% of all landfill methane emissions^[2]. These results demonstrate that open landfills need to be targeted to achieve significant near-term methane emission reductions. The main technologies for methane reduction in landfills are resource utilization, end-of-pipe control and in-situ reduction^[8]. The release of methane from landfills is continuous, dispersed and unstable. Landfill gas collection systems of China have low collection efficiency (30%)^[9] and methane concentrations that fail to meet resource utilization requirements. In-situ abatement technology uses

adsorption and biochemical oxidation in the landfill cover system to achieve methane reduction. In-situ abatement technology is a cheaper and more effective option for older and open landfills with low CH₄ production^[10].

Clay is a commonly used landfill cover material currently^[11-13]. Clay has the advantages of low price, wide source, non-toxic and non-hazardous as well as easy construction, with the disadvantages of easy cracking, restriction of CH₄ diffusion and lack of nutrition^[10, 14-16]. To address the defects of clay, researchers used biological mulching materials such as sludge^[17-20], compost^[10], and mineralized waste^[4, 21, 22], among which abundant microorganisms can improve the CH₄ oxidation capacity of the mulch and further promote CH₄ emission reduction. Despite the high removal

efficiency of biological mulch in laboratory studies, the formation of exopolymeric material within the mulch system clogs the pores and can impede gas diffusion and substrate availability to microorganisms, resulting in reduced methanogenic activity^[23]. Therefore, the key to enhancing methane reduction is to increase the porosity of the landfill cover. It has been found that biochar amended landfill soil cover increases the porosity, permeability, specific surface area and high bioaffinity of the soil, which is conducive to microbial growth and attachment, and ultimately CH₄ adsorption and methane oxidation capacity is improved^[24, 25]. Therefore, biochar has a catalytic effect on methane reduction. Biochar is a complex organic carbon solid produced by the cracking of waste biomass under anoxic or zero oxygen conditions. On the one hand, the water retained by biochar provides favorable conditions for the proliferation of methanotrophs, and the more biochar content in the cover layer, the stronger the diffusion of CH₄ and O₂, and eventually the CH₄ adsorption efficiency and oxidation reaction rate increase. On the other hand, the more biochar content in the cover layer, the higher the hydraulic conductivity of the cover material, the rainfall penetrates the cover layer into the waste pile, the rainwater occupies the pores to reduce the diffusion of CH₄ and O₂, and the CH₄ oxidation capacity decreases. The permeability of the landfill overlay is required to be less than 10⁻⁷cm/s^[26]. The soil permeability coefficient of 10% biochar was greater than 10⁻⁷cm/s^[27], and the reduction of biochar ratio would lead to the reduction of methane adsorption and oxidation performance^[28]. The key, therefore, is not only to promote the diffusion of oxygen and methane, but also to prevent rainwater from entering the mulch. Therefore, a new economical landfill covering material with high porosity, high air permeability and low hydraulic conductivity should be designed to promote methane oxidation and achieve methane emission reduction in landfills.

Previously, the hydrophobic and porous structure of hydrophobic biochar has been shown to be able to account for both air permeability and hydraulic

conductivity. This structure not only has a high potential for CH₄ adsorption, but also can enhance gas transport characteristics and reduce the adverse effects of pore plugging^[8, 27]. In this study, the CH₄ oxidation performance of hydrophobic biochar-amended soil cover simulation column (RH) and the biochar-amended soil cover simulation column (RB) were studied in comparison. The purpose of this study is to investigate the CH₄ transport properties and optimum reaction conditions of RH and RB as landfill cover materials. The specific objectives of this study are to: (1) quantify the CH₄ oxidation capacity of RB and RH at different CH₄ Influx gas concentrations (25% and 35%), different simulated landfill gas (LFG) inflow rates (10 mL/min, 15 mL/min, 20 mL/min), and different temperatures (20°C, 25°C, 30°C, 35°C, and 40°C). (2) To investigate the effect of the presence of moisture on CH₄ transport and diffusion in RB and RH treated soils, simulating natural conditions under continuous rainfall weather conditions. The results from this study can help identify the potential use of Hydrophobic biochar as amendments to landfill soil with the aim of achieving cost-effective, sustained methane mitigation.

MATERIALS AND METHODS

BC and HBC

BC was obtained from Desheng Activated Carbon Company in Liyang, China. The BC was produced by burning rice straw at 500°C under limited oxygen environments. HBC was obtained from modification at 60°C which the above biochar as raw material and KH-570 as modifier. The contact angle with water of HBC was 143.99°. The soil was obtained from the long-term contact with biogas in the film cover of the Guilin Mountain Pass landfill (about 1 year of exposure), and it was retrieved, dried and then passed through a #30 screen to remove mixed garbage and large stones. The BC and HBC were mixed with the soil uniformly according to the volume ratio of 1:5 to obtain two different covering materials, namely, biochar modified

soil and hydrophobic biochar modified soil. Their physical and chemical properties were shown in Table 1.

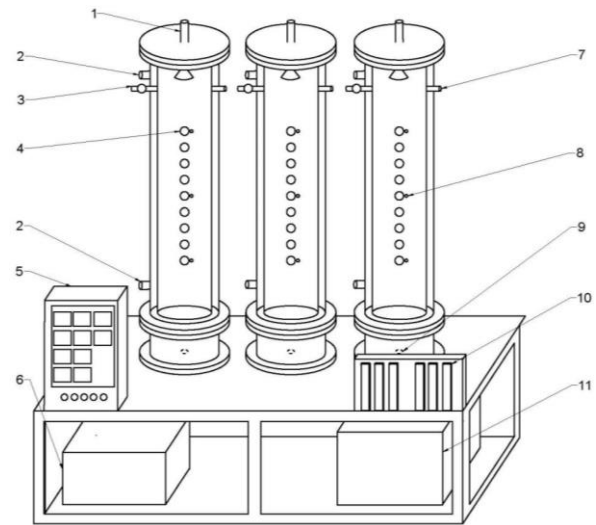
TABLE 1
hysicochemical properties of three covering materials

Indicators	Unit	Material	
		Biochar modified soil	Hydrophobic biochar modified soil
Organic content	g/kg	59.2	63.3
TN	g/kg	2.71	2.16
TP	g/kg	1.5	1.26
TK	g/kg	16.3	15.2
pH	-	7.64	7.74
Specific gravity	g/cm ³	2.12	2.20
Volume density	g/cm ³	1.14	1.18
Porosity	%	46.12	46.31
Water holding capacity	g/kg	530.41	410.96
Water content	%	10	10

Column test device

Figure 4.1 shows the CH₄ simulated column test device for the landfill cover. The column oxidation tests were conducted to compare the oxidation characteristics of CH₄ through BC and HBC. These tests were conducted using two identical PVC columns measuring 15 cm in diameter and 100 cm in length. RB was the column filled with BC and RH was the column filled with HBC. The columns were composed of a permeable layer, a gravel layer, a cover layer and an air layer from bottom to top. The permeable layer with a height of 15 cm was used to drain excess water from the system. The gravel layer with a height of 10 cm was used to support and drain the cover layer, which was composed of pebbles with a diameter of about 1cm. The cover layer with a height of 60 cm was used to fill the cover layer material which is slightly tamped. The air layer with a height of 10 cm was mainly used to simulate atmospheric

air flow. There was a rainfall simulation device at the top of the column, which could quantitatively simulate precipitation. The simulated landfill gas was introduced into the rotameter and humidifier through the PTFE tubing and then was passed into the column bottom at a pre-selected constant flow rate. A large number of small holes of the same size and uniform distribution were arranged on the annular pipe at the bottom of the column, which enabled the simulated landfill gas to enter the covering layer evenly through the buffer of the gravel layer. Air was blown into the PTFE tubing by the blower and was introduced into the top of the column through the rotameter and air humidifier. There was a water bath circulating temperature control device outside the column to control the external ambient temperature of the column. The No. 0 sampling port was set at the top air outlet. In addition, there were 9 other sampling ports with 5 cm intervals on the outer surface of the cylinder, which were numbered 1 to 9 from top to bottom. Used to collect gas and soil samples. The physical map is shown in Figure 4.2. These sampling ports were used to collect gas and soil samples.



1. shower nozzle 2. Water bath circulation port 3. Air outlet 4. Sampling port 5. Electric control box 6. Water box 7. Air inlet 8. Temperature probe 9. CH₄ inlet 10. Glass rotameter 11. Water bath

FIG. 1. Schematic diagram of CH₄ oxidation simulation device for landfill cover

Column operation and sampling

The simulated landfill gas was composed of CO₂, CH₄, and N₂, where CO₂ and CH₄ had the same volume. Before the test, the device for the simulation experiment had been operating stably for one month. In order to analyze the amount of CH₄ oxidized by methane-oxidizing bacteria, it was assumed that all adsorption behaviors had reached their adsorption capacity and that any reduction in CH₄ content was the result of microbial oxidation. The air inflow rate was 50 mL/min to simulate the natural flow of the atmosphere. The inflow rate of simulated landfill gas was controlled by rotameter to be 10–20 mL/min. The initial ambient temperature of the columns were set to 25 °C, and the initial moisture content were set to 10%. The entire test process was divided into four stages. In the first stage, the CH₄ inlet concentration of the column was 25%, and the aeration rate was 15 mL/min. In the second stage, the CH₄ inlet concentration of the column was 35%, and the aeration rate were 10 mL/min, 15 mL/min, and 20 mL/min, respectively. In the third stage, the CH₄ inlet concentration of 35% and the aeration rate of 20 mL/min were kept unchanged, and the ambient temperature of the columns were set to 20 °C, 25 °C, 30 °C, 35 °C and 40 °C, respectively. The fourth stage was the simulated precipitation test. Under simulated landfill gas conditions at a concentration of 35% and a rate of 20 mL/min. The column was uniformly poured with 5 L of water through the top shower for 5 consecutive days until the cover layer was completely saturated. When the seepage layer at the bottom of the column is no longer dripping, the soil sample in the column is taken to measure the water content by the weight method, and the permeability coefficient of the material in the column is measured by the ring knife method. Every two days, gas samples were collected from the #0—#9 sampling ports of the column with gas sampling bags, which were analyzed by GC-7890 gas chromatograph.

RESULTS AND DISCUSSION

CH₄ oxidation performance of columns

The simulation test device had been operating stably for one month beforehand. The adsorption of biochar on CH₄ can reach the adsorption capacity in a short time, so the reduction of CH₄ content in the experiment was assumed to be the result of microbial oxidation. When the CH₄ concentration in the simulated landfill gas was 25% and 35%, CH₄ content and removal rates of RB and RH varied with column depth as shown in Figure 2.

As shown in Fig 2(a) and (b), when the CH₄ concentration in the simulated landfill gas was 25%, the CH₄ content of the two columns increases as the column depth increases. The top CH₄ content of RB and RH were 0.74% and 0.22%, respectively, and the total methane removal rate were 97.05% and 99.13%, respectively. When the CH₄ concentration in the simulated landfill gas was increased to 35%, the top CH₄ content of RB and RH were 0.96% and 0.17%, respectively, the total methane removal rate was 95.80% and 99.50% (Figure 2(c)(d)), and the total methane removal rate of RH was the lowest. The CH₄ removal of RB and RH were mainly concentrated in the 50–60 cm. It could be seen that CH₄ oxidation activities mainly occurred at the bottom of RB and RH. It might be that the high concentration of CH₄ at the bottom was more suitable for the self-growth and reproduction of methane oxidizing bacteria, which promoted the occurrence of methane oxidation behavior. It may be that the large specific surface area and large porosity of biochar were conducive to the diffusion of CH₄ and O₂, and the nutrients it carries led to the high rate of methane oxidation^[29, 30]. After being hydrophobically modified, the modified biochar retained its original performance, and at the same time, its agglomeration was reduced and air permeability was improved, which was more suitable for the growth and reproduction of methanotrophs.

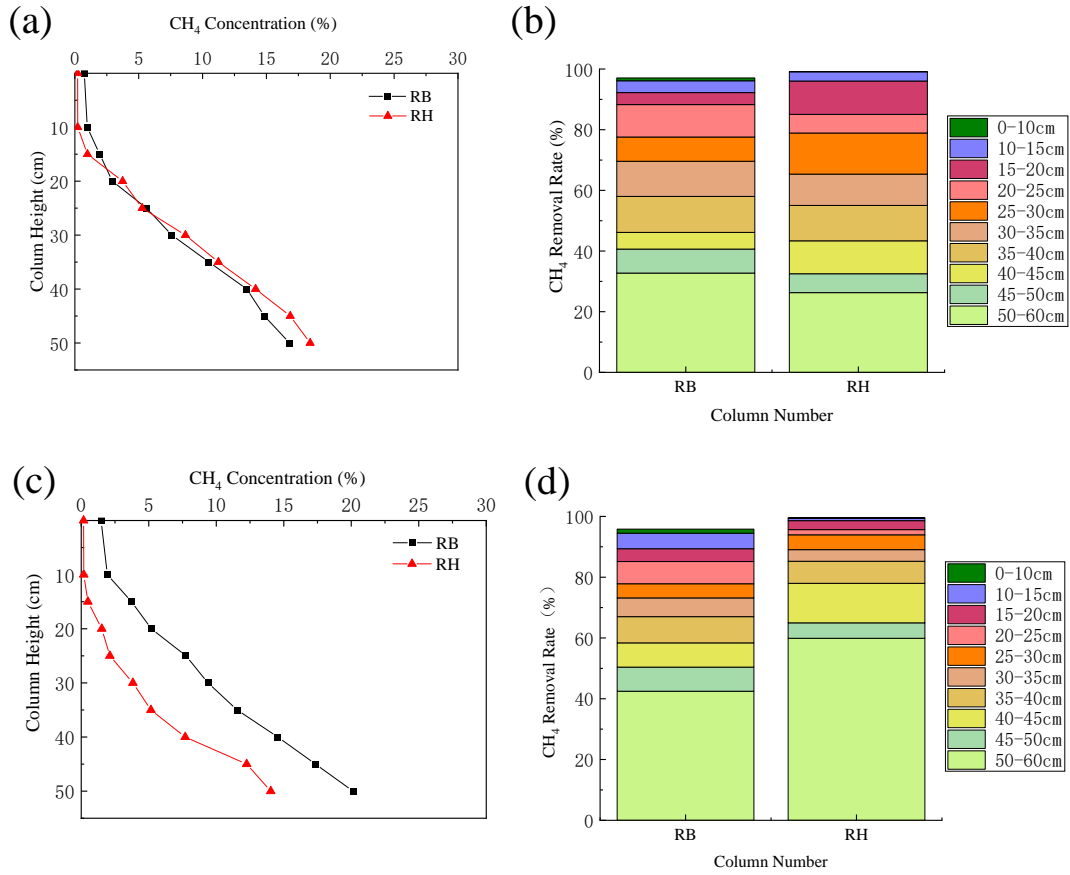
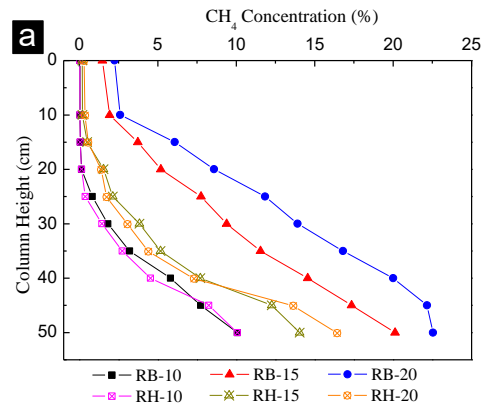


FIG. 2. Changes of CH₄ concentration and removal rate in columns with depth when CH₄ concentration is 25% (a) and (b). Changes of CH₄ concentration and removal rate in columns with depth when CH₄ concentration is 35% (c) and (d). Reaction conditions: 15 mL/min of inflow rate, 25 °C of ambient temperature.

Effect of CH₄ influx rate

The CH₄ influx rate affected the CH₄ oxidation. To explore the effect of influx on the change of the CH₄ oxidation, the CH₄ concentration was set to 35%, the ambient temperature was set to 25 °C, and RB and RH were operated at three different aeration rates simultaneously. As shown in Fig. 3, as the CH₄ influx rate increased, the CH₄ content of RB and RH increased. When the CH₄ influx rate were 10, 15 and 20 mL/min, the CH₄ content on the top of RB were 0.04%, 0.96% and 2.27%, respectively, the total removal rate of CH₄ were 99.89%, 95.80% and 93.52%, respectively. The CH₄ content on the top of the RH were 0.02%, 0.17% and 0.29%, respectively, the total removal rate of CH₄

were 99.94%, 99.50% and 99.18%, respectively. In general, both RB and RH had good removal efficiency of CH₄, but RH was better, which removal rate of CH₄ had reach more than 99%.



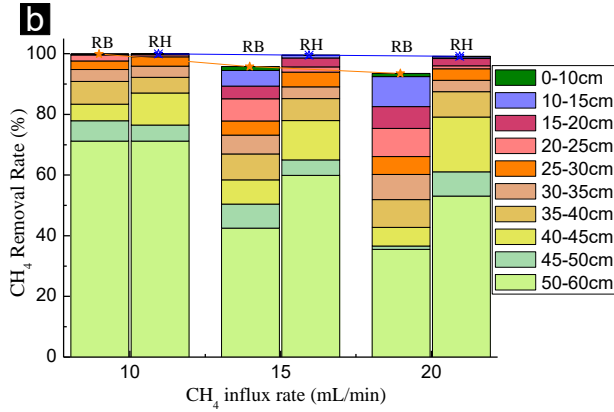


FIG. 3. Changes of CH₄ concentration in columns with depth at different CH₄ influx rate (a).
CH₄ removal rate in columns with depth at different CH₄ influx rate (b).

Effect of temperature

The ambient temperature offers significant influence in CH₄ oxidation process^[10]. The CH₄ content and removal rate of RB and RH at different temperatures when CH₄ was influxed at a concentration of 35% and a rate of 20 mL/min were shown in Fig. 4. When the temperature were 20 °C, 25 °C, 30 °C, 35 °C and 40 °C, the top CH₄ content of RB were 2.43%, 2.27%, 1.44%, 1.58% and 2.84%, respectively, and the removal rate of CH₄ were 93.05%, 93.52%, 95.89%, 95.49% and 91.89%, respectively. The top CH₄ content of RH were 0.57%, 0.29%, 0.01%, 0.05% and 0.50%, respectively, and the removal rate of CH₄ were 98.38%, 99.18%, 99.96%, 99.86% and 98.58%, respectively. The CH₄ removal rate of RH and RB all reached the highest at 30 °C. Some previous studies have shown that the optimum temperature for growth of methanophilic bacteria is about 30 °C^[31-34]. When the tmperature was lower or higher than the optimum temperature, the microbial activity gradually decreases as the temperature changes, and the CH₄ removal rate also decreases. Compared with RB, RH had a better removal effect on CH₄ (99.96%), it may due to the hydrophobic modification of biochar reduced the agglomeration between particles and

increased the porosity^[35]. Hence, the diffusion of CH₄ and O₂ in the cover layer and the adhesion of microorganisms were promoted.

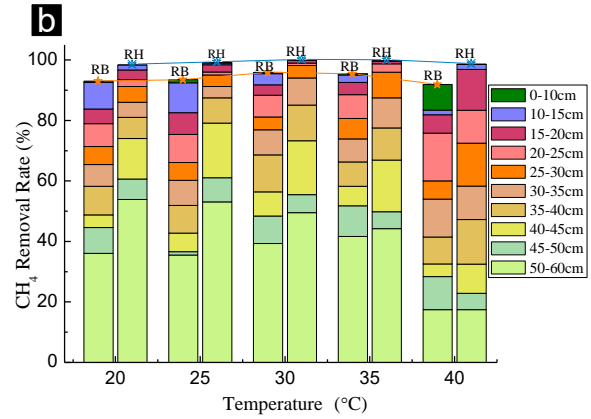
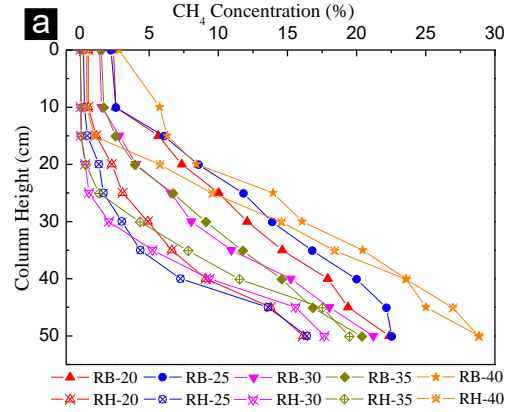


FIG. 4. Changes of CH₄ concentration in columns with depth at different temperatures (a).
CH₄ removal rate in columns with depth at different temperatures (b).

CH₄ oxidation under simulate rainfall

To study the waterproof performance and permeability of hydrophobic biochar-amended soil mulch simulated column RH by simulated rainfall experiments. The outlet CH₄ content was 9.94% and 8.05% for RB and RH, respectively, and the total CH₄ removal rate was 71.59% and 77.01%, respectively. The highest methane removal rate of biochar-amended soil cover simulation column

RB was found at column depth 15-20 cm. The hydrophobic biochar-amended soil cover simulation column RH showed the highest methane removal rate at column depths of 10-15 cm. Compared to RB, moisture has less effect on the methane removal efficiency of RH. After testing, the water contents of RB and RH were 31.22% and 22.68%, respectively, and the permeability coefficients were 1.15×10^{-7} and 7.68×10^{-8} , respectively. It shows that the hydrophobic biochar-amended soil cover simulation column RH is more waterproof than the biochar-amended soil cover simulation column RB.

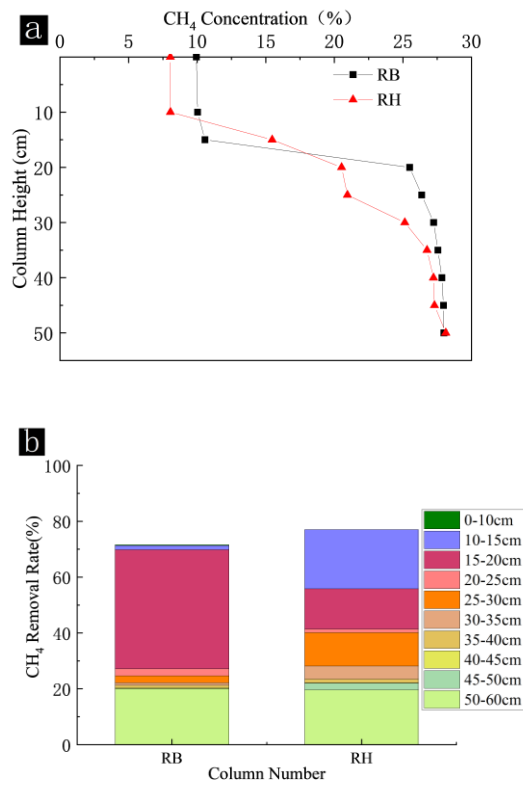
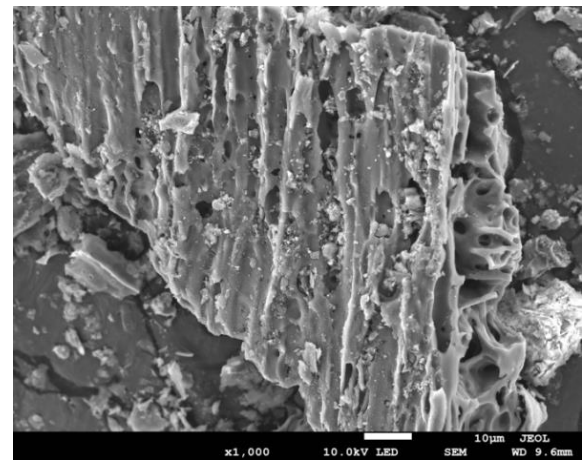


FIG. 5 Changes of CH₄ content and removal rate in columns with depth at different temperatures

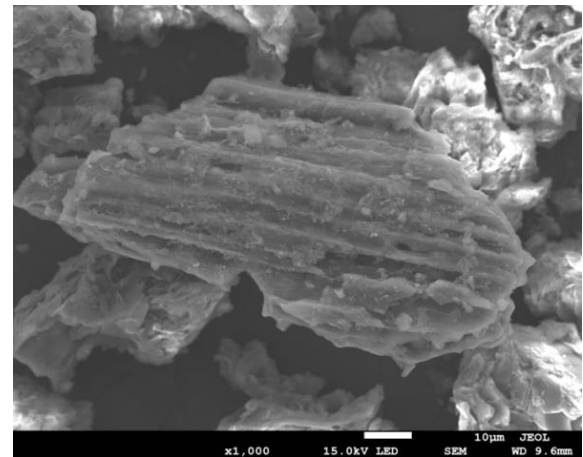
SEM image analysis before and after simulated column reaction

Figure 6 (a) and (b) shows SEM images of biochar amended soil cover before and after the reaction of simulated column RB. The surface of the biochar

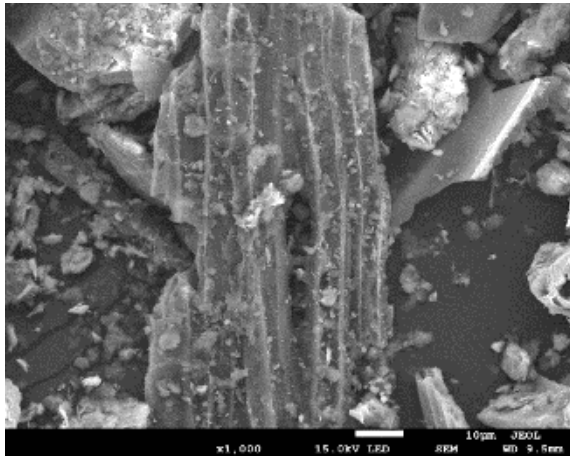
amended soil material is clearly granular and loosely structured before the reaction. However, some mineralization and microbial residues appeared on the surface of RB after the reaction. Figure 6(c)(d) shows the SEM images of the simulated column RH before and after the reaction of the hydrophobic biochar amended soil cover. Similar to biochar, the surface of hydrophobic biochar after the reaction has numerous more agglomerated substances than before the reaction, which probably are mineralized substances and microbial residues.



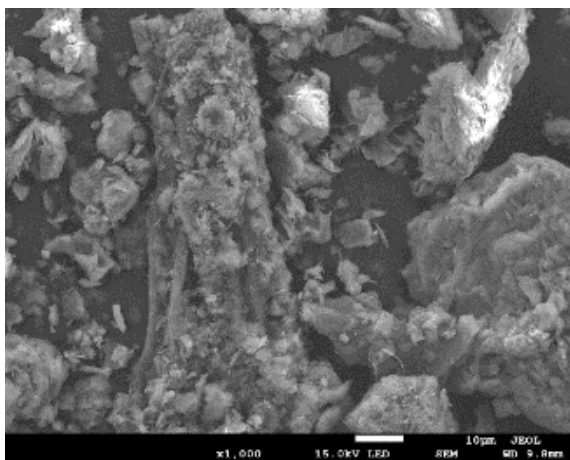
(a) Before RB reaction (10 mm)



(b) After RB reaction (10 μm)



(c) Before RH reaction (10 µm)



(d) After RH reaction (10 µm)

FIG. 6 SEM images before and after RB and RH reactions

CONCLUSION

The oxidative removal of CH_4 by RH was very satisfactory, with a removal rate of 99.13%, which was higher than that of 97.05% by RB. Methane removal by both RB and RH occurred mainly at depths of 50-60 cm. The removal rate of CH_4 for both RB and RH decreased with increasing simulated landfill gas influx rate. However, the buffering capacity of RH was better than that of RB when the simulated landfill gas influx rate was increased. The optimum temperature for methane removal was 30°C for both RB and RH, at which the removal rates were 95.89% and 99.96%, respectively.

Under simulated precipitation conditions, RH had better water repellency and better removal of CH_4 than RB. SEM image characterization indicates that mineralized and microbial residues were observed in both RB and RH after the simulated landfill reaction. Therefore, the oxidation of CH_4 is due to the combined effect of the porous structure of hydrophobic biochar and nutrients.

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