Water Retention and Unsaturated Hydraulic Behaviors of a Biochar-modified Silt

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ABSTRACT: Biochar has been used to modify the soil cover of municipal solid waste (MSW) landfill to mitigate methane emission. To model the coupled water and reactive gas transport in unsaturated soil cover, water retention curve and unsaturated permeability function of biochar-modified soil cover are required. A laboratory study is presented to investigate the effects of biochar content (*BC*) and void ratio on the water retention and unsaturated hydraulic behaviors of a biochar-modified silt. Biochar contains high internal porosities and it exhibits negative surface charge. Adding biochar to the silt alters the microstructures of modified soil. The pore size distributions measured by the mercury intrusion porosimetry indicate that the modified silt contains more micro-porosities than the untreated silt. Results of modified evaporation test showed that water retention capacity increases with increasing *BC*. In other words, the modified silt can hold more water for a given suction. On the other hand, the modified silt exhibits a lower saturated permeability and also a lower rate of change in permeability with respect to suction. Despite the modified silt is less permeable than the untreated silt in the low suction regime, it becomes more permeable after drying to the high suction regime.

KEYWORDS: Biochar, Silt, Water retention curve, Unsaturated permeability, Pore size distribution

1. INTRODUCTION

According to IPCC, methane emissions from municipal solid waste (MSW) landfills are about 7% of global emissions (IPCC, 2007). As methane is a more harmful greenhouse gas than carbon dioxide, how to mitigate methane emissions is an important research topic. A bio-cover is a novel soil cover system that mitigates methane emissions from MSW landfills using the methanotrophic bacteria to oxidize methane (Huber-Humer et al. 2009, Stern et al. 2007, Humer and Lechner 1999). Methanotrophic bacteria can consume methane as a carbon and energy source. The microbial process is aerobic and the main products of microbial oxidation are carbon dioxide and water. The design of bio-cover mainly involves the optimization of environmental conditions for methanotrophic bacteria such that biotic methane consumption is enhanced. Scheutz et al. (2009) presented a review of microbial methane oxidation in different bio-cover systems and pointed out that soil texture, water content, gas supply (methane and oxygen), temperature and nutrients are key influencing factors for methane oxidation in landfill cover soils. Sufficient amount of water is required to support the growth and activity of methanotrophic bacteria. However, too much water can reduce significantly gas transport in the soil resulting in limiting microbial methane oxidation. Thus, an optimum range of water content is required to achieve the maximum gas transport and microbial activity. The amount of water available in the soil cover is controlled by the water retention capacity and permeability. Water can be drained easily in a high permeable soil leading to more air-filled pores available for gas transport. On the other hand, a high water retention capacity can support microbial growth. Hence, these two hydraulic properties are crucial in the design of bio-cover.

Comparing with conventional landfill cover soils, vegetation and organic rich materials such as compost and sewage sludge possess higher microbial methane consumption because they have more nutrients, higher water retention capacity and increased porosities (Bohn et al. 2011, Huber-Humer et al. 2008). Biochar is an organic material produced from biomass through pyrolysis. It is the most stable form of carbon in soil (Pessenda et al. 2001). In agriculture, agricultural waste (e.g. rice and maize stalks) can be turned into biochar, which is used to increase soil carbon sequestration and soil productivity. Its highly porous structure and high organic matter content can favor the microbial activity, which has been recently considered as a potential soil amendment material to enhance methane oxidation in the bio-cover (Sadasivam and Reddy 2015, Reddy et al. 2014). Yargicoglu et al. (2015) showed that a high variability in the physical and chemical properties of different biochars was observed due to different feedstocks and production processes. Modification of landfill cover soil by biochar can change its physical and chemical properties which affect methane oxidation, gas transportation, adsorption and water retention behaviors.

Past studies on biochar-modified soil cover have mainly focused on the physical and chemical properties, methane oxidation and gas absorption. Microbial methane oxidation is an aerobic process where oxygen is required. Besides, water is needed for microbial activity. Moreover, landfill cover soils are unsaturated in nature. Thus, the water retention curve and unsaturated permeability function are required to model the coupled transport process of water and reactive gas in unsaturated soil cover. Previous studies have shown that methane oxidation can be related to the field capacity and wilting point of soil cover. However, water retention curves of biochar-modified soils reported in the literature are limited to narrow range of suction, either suction below 1.5 MPa or above 10 MPa (Wong et al. 2017, Ojeda et al. 2015, Or et al. 2007). Hence, either air entry value (*AEV*) or residual water content may be missing from the reported water retention curves. Water flow in unsaturated soil cover is influenced by water content or soil suction. Thus, water permeability of unsaturated soil is not a constant. However, most of previous studies on biochar-modified soils presented only saturated hydraulic behavior. The lack of published data for unsaturated permeability may be due to the fact that its direct measurement is time consuming. Hence, transient flow methods (multi-step inflow/outflow test, evaporation test, etc.) have merits over steady state methods because of reduced testing time. In this study, two series of modified evaporation tests were conducted on a biochar-modified silt to investigate the effects of biochar content (*BC*) and void ratio on its water retention curve and unsaturated permeability function. To measure water retention curve over a wide range of suction, filter paper method was also adopted to estimate indirectly the suction of modified soil samples in the high suction regime. Furthermore, mercury intrusion porosimetry was also conducted to measure the pore size distribution from which microstructural analysis was used to interpret the impact of biochar on the water retention and hydraulic behaviors of unsaturated soil cover.

2. MATERIALS AND METHODOLOGY

2.1 Materials

The tested soil is a fine-grained soil. The basic physical properties were determined in accordance with the procedures given in GB/T 50123-1999 (Ministry of Construction P.R. China, 1999). The specific gravity is 2.62. The grain size distribution curve shown in Figure 1 reveals that the soil consists of 60% of silt and 40% of clay.

The liquid limit and plasticity index are 27% and 9, respectively. The soil is classified as silt of low plasticity (ML) according to the Unified Soil Classification System (USCS). The maximum dry density and optimum water content obtained from the standard compaction test are 1760 kg/m^3 and 16%, respectively.

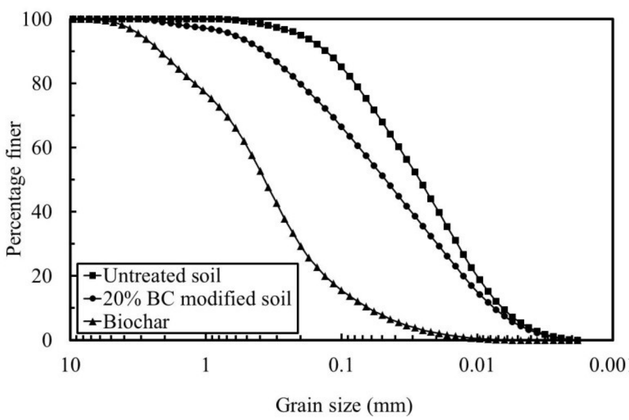

Figure 1 Grain size distribution curves of untreated soil, 20% *BC* modified soil and biochar

Biochar used in this study was derived from the rice straw, which was pyrolyzed at a temperature of 500°C. The sample has been grinded mechanically by the manufacturer and the maximum grain size is below 2 mm. The specific gravity of the biochar is 0.69. The specific area per unit weight is 118 m^2/g. The organic matter content is 61%. The modified soil samples were prepared by mixing thoroughly the air-dried biochar and soil to achieve a *BC* of 10% and 20% of dry mass of soil. The basic physical properties of the modified soil are summarized in Table 1. The properties of untreated soil are also presented in the table for comparison. The specific gravity of modified soil decreases with increasing *BC* content because biochar has a relatively low specific gravity of 0.7 resulting from its high internal porosities. Figure 1 depicts the grain size distribution curves of untreated soil, 20% *BC* modified soil and biochar. It should be noted that the curves are the average of three samples for each soil. As the grain sizes of biochar are coarser than those of untreated soil, the percentages of sand and silt particles found in the modified soil are higher than those in the untreated soil. On the other hand, addition of biochar increases significantly the liquid limit and plasticity index of modified soil. By adding a *BC* of 20%, the modified soil changes from silt of low plasticity (ML) to silt of high plasticity (MH). Past studies (Liang et al. 2006, Mukherjee et al. 2011, Yargicoglu et al. 2015) have reported negative Zeta potential values in biochars that reflect their property of negative surface charge. This negative charge may affect the chemistry of clay/water leading to the change in plasticity of modified soil. Furthermore, the maximum dry density of modified soil decreases, but the optimum water content increases with increasing *BC*. This is consistent with the compaction test results obtained from low and high plasticity clays. Hence, maximum dry density decreases but optimum water content increases with increasing plasticity for the modified soil. In summary, the modified soil consists of higher percentages of coarser grains, but it exhibits higher plasticity and are less compactable (or low dry density with the same compaction effort) than the untreated soil.

2.2 Test programme and methodology

2.2.1 Test programme and specimen preparation

Water retention curve and unsaturated permeability function of biochar-modified soils were measured in this study. The effects of void ratio and *BC* on these properties were investigated. Two initial void ratios of 1.0 and 1.16 and two *BC* of 10% and 20% were tested. Furthermore, the pore-size distribution was measured and used to interpret the change in the water retention and unsaturated hydraulic behaviors of modified soil. Compacted specimens were prepared by wet tamping method. The specimens were compacted to the target void ratios at the optimum water contents specified in Table 1. Four different tests were conducted. First, saturated permeability was measured by the constant head test using a flexible wall triaxial permeameter. Second, modified evaporation test was used to measure water retention curve and unsaturated permeability function for suction below 400 kPa. To measure water retention curve for suction above 400 kPa, contact filter paper method was adopted to estimate indirectly the matric suction of unsaturated soil. Finally Mercury Intrusion Porosimetry (MIP) was used to measure pore size distribution.

Table 1 Basic physical properties of biochar-modified soil

	Untreated soil	**10% BC**	**20% BC**
Specific gravity	2.62	2.53	2.46
Liquid limit (%)	27	42	54
Plasticity index	9	14	19
Maximum dry density (kg/m^3)	1760	1370	1220
Optimum water content (%)	16	27	33

2.2.2 Constant head permeability test

Saturated permeability was determined by the constant head permeability test using a flexible triaxial permeameter. The specimens were 39 mm in diameter and 80 mm in height. Before the permeability tests, the specimens were saturated for a period of 24 hours inside a vacuum chamber filled with de-aired water. During the test, a cell pressure of 100 kPa was applied firstly to confine the specimen. After completion of consolidation, a constant hydraulic gradient was applied between the top and bottom of a specimen. Then, the water flow was measured by a digital pressure/volume controller. For each specimen the permeability tests were conducted under three different hydraulic gradients ranging from 25 to 75.

2.2.3 Modified evaporation test

Water retention curve and unsaturated permeability function for suction below 400 kPa were measured by the modified evaporation test (Schindler 1980, Wind 1968). The test involves simultaneous measurements of weight of water loss and suctions at two different heights in a soil column of 60 mm in diameter and 80 mm in height. The schematic setup of the soil column is depicted in Figure 2. Before the test, the soil column was saturated for a period of 24 hours inside a vacuum chamber filled with de-aired water. Thereafter, two miniature high suction tensiometers were installed at 10 mm and 70 mm below the top of soil column to measure the suctions. The miniature tensiometer was 14 mm in diameter and 20 mm in length. It consisted of a strain gauge type pore pressure transducer (PPT), a water reservoir (a volume of 2.8 mm^3) and a filter made of a 5 bar high air entry value ceramic disk (a thickness of 6 mm). The tensiometer would measure suction up to 500 kPa if the filter was saturated properly. Besides, the volume of water reservoir behind the filter should be as small as possible to reduce the possibility of cavitation (Ridley and Burland 1993). The strain gauge type PPT had a maximum strain value of 1% and a sensitivity of 0.02%. A 5 V input voltage was required and the output voltage was measured by a compact digitial millivoltmeter. The millivolmeter had an accuracy of 0.001 mV, corresponding to a water pressure of 1 kPa. Details of calibration and saturation procedures of the tensiometer can be found in Chen et al. (2015). The soil column was placed on the top of a balance (with a resolution of 0.1 g) to monitor the amount of water loss (i.e. evaporation rate) during the test. The test was terminated when the top tensiometer has reached its maximum suction limit (500 kPa) or the weight of water loss became negligible.

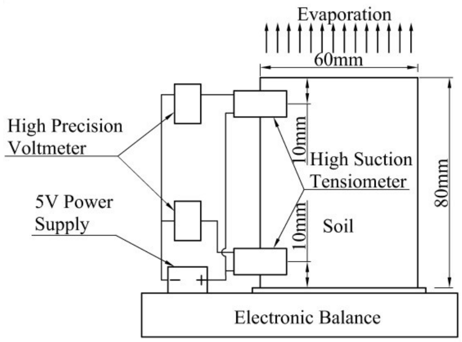

Figure 2 Schematic setup of modified evaporation test

2.2.4 Filter paper method

Contact filter paper method was adopted to determine indirectly the matric suction of unsaturated soil for suction above 400 kPa. Whatman No.42 filter paper was used in this study. The matric suction measurement was conducted in accordance with the procedures recommended in ASTM D5298-16 (ASTM, 2016). A filter paper was first placed between two protective filter papers to minimize contamination. Then the sandwiched filter papers were inserted between two soil specimens of 61.8 mm in diameter and 20 mm in height. The two specimens were sealed by plastic warp to maintain a good contact between the filter papers and soil specimens, and stored inside a plastic jar. The whole setup was kept inside a temperature controlled chamber. Based on a control test, a period of one week was sufficient for reaching equilibrium of water content between the filter papers and soil specimens.

Bicalho et al. (2010) have compared several existing calibration curves for the Whatman 42 filter paper for estimating soil suction. Despite some discrepancies, consistent agreement can be found in the suctions estimated from the calibration curves suggested by Chandler et al. (1992) and ASTM D5298-16 (ASTM 2016). It should be noted that ASTM D5298-16 only recommends one calibration curve for both total suction and matric suction. However, the calibration curve proposed by Chandler et al. (1992) is specified for matric suction, which was adopted in this study to infer the matric suction (s in kPa) from the water content of filter paper (w_f in %):

$$log_{10}(s) = 4.842 - 0.0622 \cdot w_f \quad \text{for } w_f < 47\% \qquad (1)$$

Furthermore, for each suction measurement, the shrinkage of specimen was determined by measuring its dimensions using a caliper.

2.2.5 Mercury intrusion porosimetry

Freeze drying technique was adopted to dehydrate MIP specimens to minimize the shrinkage induced by drying that may change the microstructure of specimens. To maximize the heat transfer, the specimens were cut into cubes of volume less than 1 cm^3 and were freeze-dried under a temperature of -80 ^{o}C. Pore master GT-60 (Quantachrome, US) was used to measure pore size distribution and the applied mercury intrusion pressure ranging from 8 kPa to 138 MPa.

3. RESULTS OF MODIFIED EVAPORATION TEST

For estimating the water retention curve and unsaturated permeability function from the modified evaporation test, it is assumed that both water content and suction vary linearly with depth in the soil column (Schindler 1980). As an illustration, the results of modified evaporation test for the 20% *BC* modified soil compacted at an initial void ratio of 1.16 are depicted in Figures 3 and 4. Figure 3 shows the values of suction measured by the two high suction tensiometers during the evaporation test. In the first 40 hours of the test, the difference in suction between the two tensiometers is less than 5 kPa, i.e., the hydraulic gradient is small. During the next 20 hours of test, the difference in suction becomes substantial leading to a higher hydraulic gradient between the two tensiometers. Figure 4 indicates that the weight of water loss increases linearly with time. In other words, the evaporation rate remains almost constant during the test. It is because the increase in the hydraulic gradient is compensated by the decrease in unsaturated permeability as a result of water loss. Hence, the evaporation test was conducted under a nearly constant flux condition.

For determining water retention curve, the average value of suctions measured by the two tensiometers at each time interval is assumed as the suction in the middle of soil column. It is shown that it takes around a duration of 60 hours for reaching an average suction of 420 kPa in the middle of soil column. The corresponding average water content is calculated by subtracting the weight of water loss measured by the balance from the initial weight of soil column. Then the volumetric water content was calculated from the initial volume of test specimen because negligible volume change was observed during the test.

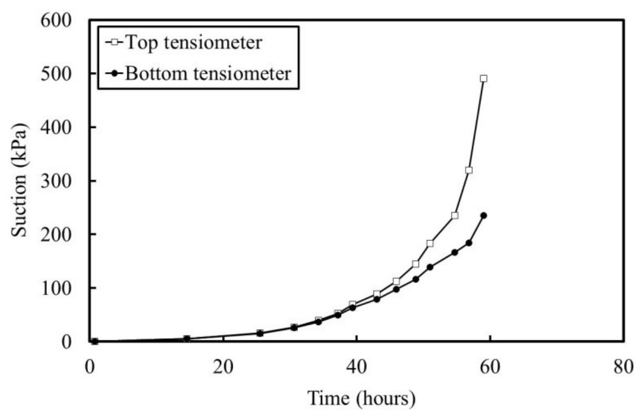

Figure 3 Suction measurements from two tensiometers during evaporation test

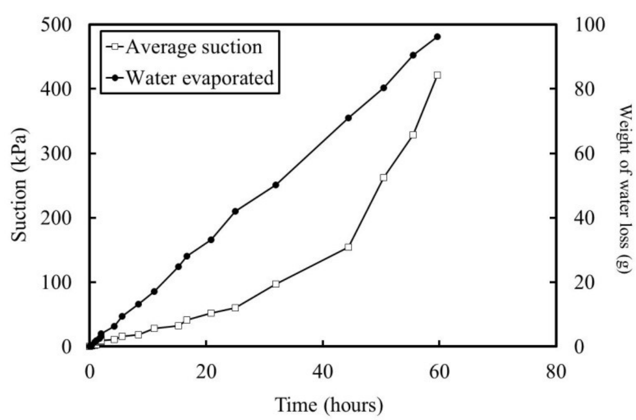

Figure 4 Change in average suction and weight of water loss during evaporation test

For determining unsaturated permeability, the water flow (q_i) through a plane located in the middle of soil column for a given time interval between time steps i and i + 1is related by Darcy's law.

$$\mathrm{k}(s_i) = -\frac{q_i}{\Delta s_i/_{z} + 1} \qquad (2)$$

where $k(s_\mathrm{i})$ = coefficient of permeability at s_i, s_i = average suction at time step i, Δs_i = difference in suction between tensiometers at time step i, z = distance between tensiometers. q_i can be evaluated by the difference of volumetric water content measured between time steps i and i+1. Thus, the only unknown in Eqn.(2) is $k(s_\mathrm{i})$. Consider the data measured for each time step, the unsaturated permeability function can be derived for a suction up to around 400 kPa.

4. WATER RETENTION CURVES

4.1 Effects of *BC*

Figure 5 shows the main drying water retention curves for untreated soil, 10% and 20% *BC* modified soil. All specimens were compacted to an initial void ratio of 1.0. As expected, the untreated soil has a low air entry value (*AEV*) of around 1 kPa because of a high initial void ratio. It is found that *AEV* increases with increasing *BC*. Despite compacted to a high void ratio of 1.0, *AEV* increases from 1 kPa to 15 kPa by adding 20% biochar by weight. The figure also indicates that the residual water contents are reached around a suction of 10 MPa for the three measured water retention curves. The residual water content also follows a trend similar to that of *AEV*, i.e. it increases with increasing *BC* content. In other words, addition of biochar improves the water retention capacity of modified soil by holding more water in the pores for a given suction. Based on the basic physical properties of modified soil, it consists of higher percentages of coarser grains but it also exhibits higher plasticity. Previous microstructural studies using scanning electronic microscopy (Wong et al. 2017) revealed that biochar particles contained many internal micro-porosities which can be filled up by clay particles. Besides, it seems that more flocculated structures may be formed in the modified soil due to the negative surface charge of biochar. Hence, the intra-aggregate pores should be increased substantially in the modified soil. It is postulated the high internal micro-porosities of biochar itself and substantial intra-aggregate pores in the modified soil matrix contribute to its higher water retention capacity.

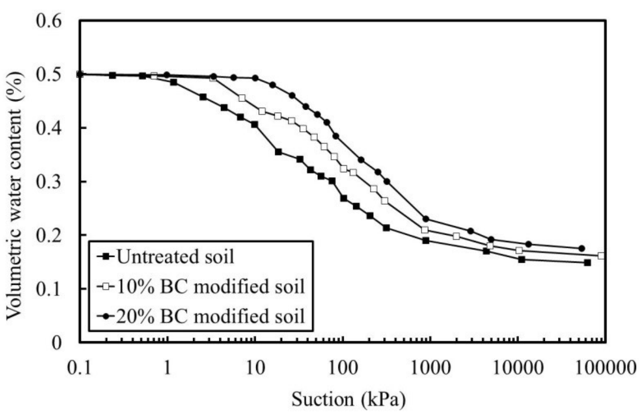

Figure 5 Effects of *BC* on water retention curve of modified soil compacted at an initial void ratio of 1.0

4.2 Effects of void ratio

Figure 6 shows the water retention curves for 20% *BC* modified soil compacted at an initial void ratio of 1.0 and 1.16. Similar to other soil types, *AEV* increases with decreasing void ratio. However, the residual water content does not change significantly for the range of void ratio studied. It should be noted that an initial void ratio of 1.0 corresponds to 85% of maximum dry density. Thus, the applied compaction effort is low. It seems that this low compaction effort can only reduce the size of inter-aggregate pores, but it does not affect significantly the intra-aggregate pores. As a result, only *AEV* increases, but not the residual water content.

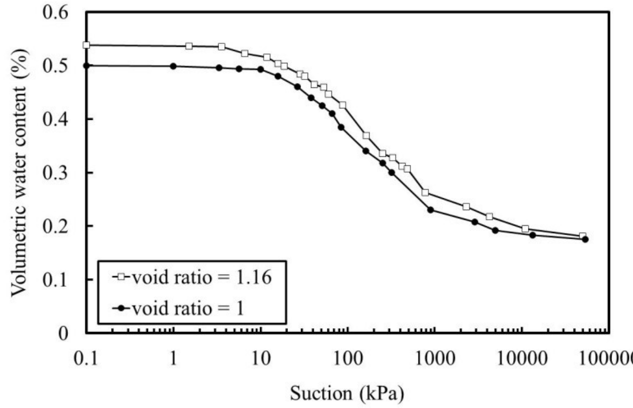

Figure 6 Effects of void ratio on a 20% BC modified soil

5. PERMEABILITY

5.1 Saturated permeability

Figure 7 depicts the effects of *BC* on saturated permeability of modified soil. The measured permeability is in the order of magnitude of 10^{-7} m/s. Such high permeability is attributed to the high initial void ratio of 1.0 of tested specimens. In practice, the biocover is placed on the top 0.3 m of landfill final cover and a high permeability can improve gas flow leading to greater biotic methane consumption. It is also shown that the saturated coefficient of permeability decreases slightly with increasing *BC*. Adding a *BC* of 20% can decrease the saturated permeability of modified soil to one third of its original value. Table 1 indicates that the 20% BC modified soil is a high plasticity fine-grained soil. Its lower permeability may be the results of a more flocculated structure due to the negative surface charge of biochar. The effects of different pore sizes on the permeability of modified soil will be discussed later.

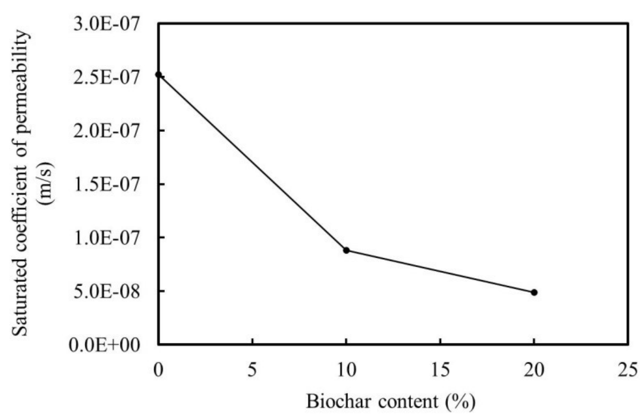

Figure 7 Effects of *BC* on saturated permeability of modified soil compacted at an initial void ratio of 1.0

5.2 Unsaturated permeability

Figure 8 shows the effects of *BC* on unsaturated permeability function for the modified soil. Two suction regimes are considered. For suction below 5 kPa, untreated soil is more permeable. However, modified soil is more permeable for suction above 5 kPa. Adding biochar increases the water retention capacity of modified soil resulting in a lower reduction rate in permeability with respect to suction during the drying process. Hence, unsaturated permeability of modified soil is higher than that of untreated soil in the high suction regime. This trend is similar to that between coarse-grained and fine-grained soils. Similar to water retention behaviour, the high internal micro-porosities of biochar itself and substantial intra-aggregate pores in the modified soil matrix are also dominant factors to control the permeability of modified soil. Figure 9 shows the unsaturated permeability function for 20% *BC* modified soil compacted at an initial void ratio of 1.0 and 1.16. As expected, the permeability decreases with decreasing void ratio.

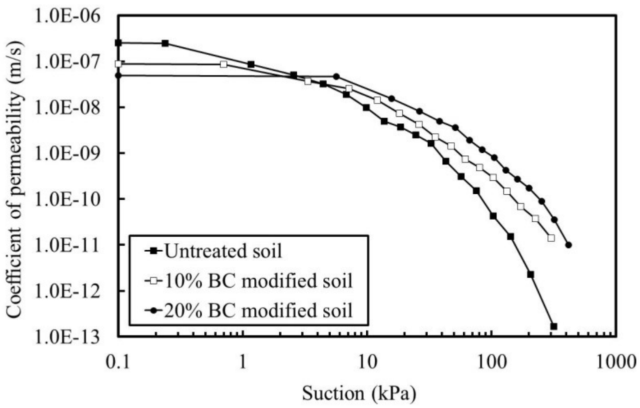

Figure 8 Effects of *BC* on unsaturated permeability of modified soil compacted at an initial void ratio of 1.0

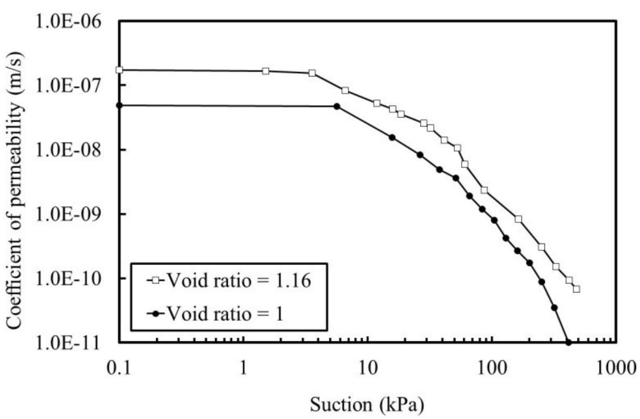

Figure 9 Effects of void ratio on a 20% BC modified soil

6. MICRO-STRUCTURES

Figure 10 depicts the pore-size distributions (PSD) measured by MIP for the modified soil compacted to an initial void ratio of 1.0. It is evident that both modified and untreated soils exhibit unimodal distribution. The *PSD* is discussed by considering the following four different ranges of pore diameter (d): (i) $d > 100$ μm, (ii) 10 μm $< d < 100$ μm, (iii) 1 μm $< d < 10$ μm, (iv) 0.1 μm $< d < 1$ μm and (v) $d < 0.1$ μm. Table 2 summarises the percentages of pore volumes in different range of d for modified and untreated soils.

The air entry value (AEV) of the water retention curve is the air-water pressure difference (or soil suction, s) above which the air commences to enter the pores initially filled with water. In general air invades the largest pore first on a soil specimen. If the pores are considered as a series of capillary tubes of diameter d, the Young-Laplace equation can be used to relate s and d:

$$s = \frac{4Tc}{d} \quad (3)$$

where T = surface tension between water and air (72.8 mN/m at 20°C) and θ = contact angle. If a zero contact angle is assumed, the pore diameter corresponding to an AEV of 1 kPa (untreated soil) and 15 kPa (20% BC modified soil) shall be around the order of 300 μm and 20 μm, respectively. It should be noted that the pore diameters measured by the MIP are limited in the range between 0.01 μm and 180 μm. Hence, a pore diameter of 300 μm could not be detected as shown in Figure 10. However, around 18% of pore diameters between 10 μm and 100 μm were observed for the two biochar-modified soil specimens. Thus, the range of largest pore observed in the PSD is consistent with the AEV obtained from the water retention curve for the biochar-modified soil. If the pore diameter is within the measurable range of MIP, the measured PSD may provide qualitative information on the AEV of the water retention curve. Table 2 also shows that the percentages of macro-pores ($d > 10$ μm) and micro-pores ($d < 0.1$ μm) are similar for both modified and untreated soils. Hence, it is anticipated that the pore diameters between 0.1 μm and 10 μm account for the differences in the water retention curve and permeability between the modified and untreated soils. Within this range of pore sizes, the modified soil has higher pore volumes for pore sizes between 0.1 μm and 1 μm, which is the range of internal pore sizes within the biochar resulting in a higher water retention capacity. On the other hand, the untreated soil has higher pore volumes for pore sizes between 1 μm and 10 μm leading to a higher saturated coefficient of permeability.

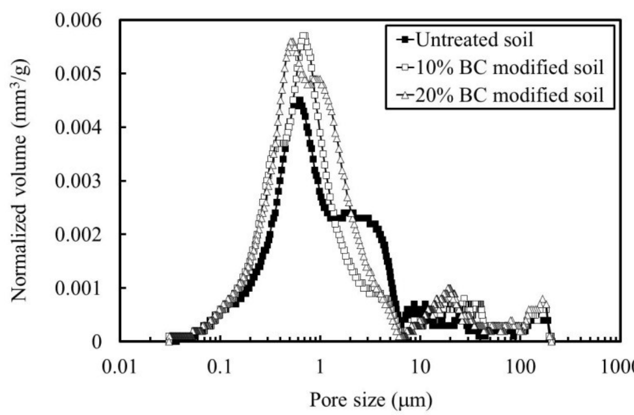

Figure 10 Pore-size distributions of modified soil compacted at an initial void ratio of 1.0

Table 2 Percentages of pore volumes in different range of pore diameters

	Percentage		
	Untreated soil	10% *BC*	20% *BC*
$d > 100$ μm	1.5	1.7	1.7
10 μm $< d < 100$ μm	13.1	17.8	17.3
1 μm $< d < 10$ μm	40.4	22.5	26.8
0.1 μm $< d < 1$ μm	40.0	53.0	49.6
$d < 0.1$ μm	5.0	5.0	4.6

7. CONCLUSIONS

This paper presents two series of laboratory study to investigate the effects of biochar content (*BC*) and void ratio on water retention and unsaturated hydraulic behaviors of a compacted silt modified by a rice straw derived biochar. The test results indicated that despite the modified soil consists of more coarse grains than the untreated soil, it exhibits higher plasticity because of the negative surface charge of biochar. Compared to the untreated soil, the internal porosities of biochar alter the microstructures of the modified soil leading to a higher pore volume for pore sizes ranging between 0.1 μm and 1 μm. The change in microstructures contributes to its higher water retention capacity and higher unsaturated permeability in the high suction regime.

8. ACKNOWLEDGEMENT

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