

# Effect of microplastics on swelling behaviour of geosynthetic clay liners (GCLs)

## Les effets des microplastiques sur le gonflement des géosynthétiques bentonitiques (GSB)

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**ABSTRACT:** Microplastics (MPs) are emerging persistent pollutants and landfills have been evidenced as one of the main sinks of MPs. Landfill-based lining systems are employed to prevent contaminated leachate from migrating to underlying aquifers. Geosynthetic clay liners (GCLs) composed of bentonite clay are widely used in lining systems due to their high swelling capacity and low hydraulic conductivity. However, bentonite clay is chemically reactive, and its engineering properties are significantly affected when exposed to ion-concentrated solutions. No studies have investigated the impacts of MPs on the engineering properties of GCLs despite the well documented presence of MPs in landfill leachate. In this paper, results of an experiment during which a GCL was exposed to spherical 1  $\mu\text{m}$  polystyrene MPs in the laboratory are reported. The effects of the exposure on the swelling potential of the GCL and the interaction between MP and bentonite, using scanning electron microscopy (SEM), are discussed. Swelling index (SI) and oedometer swelling strain tests were conducted to characterize swelling behaviour. While the preliminary results reported here are limited to a single type, size and shape of MP, no evidence of an effect of polystyrene MPs on swelling of GCL's bentonite has been found.

**RÉSUMÉ:** Les microplastiques (MP) sont des polluants pérennes, et les centres de décharges de déchets (dépotoirs) ont été identifiées comme l'un des principaux sites de contamination par les MP. Des systèmes de revêtement, à la base des dépotoirs, sont utilisés pour empêcher le lixiviat contaminé de migrer vers les aquifères sous-jacents. Les revêtements à base de géosynthétiques bentonitiques (GSB) composés d'argile bentonite sont largement utilisés en raison de leur grande capacité de gonflement et de leur faible conductivité hydraulique. Cependant, l'argile bentonite est chimiquement réactive, et ses propriétés hydrauliques and mécaniques sont modifiées lorsqu'elle est exposée à des solutions concentrées en ions. Aucune étude n'a examiné les impacts des MP sur les propriétés des GSB malgré la présence bien documentée des MP dans le lixiviat des décharges. Dans cet article, les résultats de plusieurs essais de laboratoire en cours, durant lesquelles un GSB a été exposé à des MP de polystyrène sphérique de 1  $\mu\text{m}$  de taille, sont rapportés. Les effets de MP sur le potentiel de gonflement du GSB et l'interaction entre le bentonite et les MP, par l'intermédiaire de microscopie électronique à balayage (MEB) sont discutés. Des tests d'indice de gonflement (SI) et de contrainte de gonflement à l'odomètre ont été réalisés pour caractériser le comportement de gonflement. Bien que les résultats préliminaires rapportés ici se limitent à un type, une forme et une taille de MP, aucune preuve d'un effet des MP en polystyrène sur le gonflement de la bentonite des GSB n'a été trouvée.

**Keywords:** Polystyrene microplastics; GCLs; swell; clay microstructure.

## 1 INTRODUCTION

The accumulation of plastic waste in the environment may pose a risk to human health and environmental sustainability. Weathering can accelerate the breakdown of large-sized plastics and generate tiny fragments smaller than 5 mm, termed microplastics (MPs) (Li et al., 2021). Microplastics have been

categorised as a global emerging contaminant by the World Health Organization (Alan et al., 2022).

In the past few years, progressive efforts have been made to study MPs in terrestrial environments. Many studies have investigated the transport and retention of MPs in saturated porous media and explored the effects of co-contaminants on MP transport and vice versa (Li et al., 2021; Rong et al., 2022). A few studies

have reported changes in the properties of MP-contaminated soils. Microplastics may alter the pore structures of soil due to their small particle sizes and may influence soil properties. Jannesarahmadi et al. (2023) found that 30~200  $\mu\text{m}$  polyethylene (PE) and polyvinylchloride (PVC) influence evaporation characteristics, porosity and water-retention of sand-bentonite mixtures. Yu et al. (2023) investigated the effects of MPs on the saturated hydraulic conductivity, water holding capacity and wet-aggregate size distribution of agricultural soils. They found no significant changes in these properties when soils were exposed to polyester and polypropylene (PP) at environmentally relevant MP concentrations. Wang et al. (2023) reported similar findings that small amounts of polyethylene (PE) MPs did not vastly alter water-holding capacity, whereas a high mass ratio can reduce it significantly.

The above studies have experimentally studied the properties of MP-contaminated soils by mixing soil samples with MP particles in the dry state. However, soils may be contaminated by hydration with MP-rich leachate, especially in landfills. Geosynthetic clay liners (GCLs) are used to prevent contaminated leachate from migrating to underlying aquifers. They need to be sufficiently hydrated by the subsoil and to achieve significant swelling, before exposure to leachate, in order to achieve the required low hydraulic conductivity. This is because bentonite is chemically reactive and premature exposure to leachate can inhibit its swelling potential (Tian et al., 2016). To the best of the authors' knowledge, no study has explored the effects of exposure to MPs on the swelling capacity of GCLs used in landfills. To address this gap, this study reports two sets of tests: swell index (SI) tests on sodium-bentonite extracted from the GCLs and oedometer swell strain tests of entire GCLs exposed to polystyrene MPs. In addition, scanning electron microscopy (SEM) tests on bentonite extracted from the GCL after the oedometer tests were carried out.

## 2 MATERIALS AND METHODS

### 2.1 Tested materials

An X2000 GCL with powdered sodium bentonite, manufactured by Geofabrics Australia was used in this study. The bentonite was sandwiched between a needle-punched nonwoven (NW) cover geotextile and a scrim-reinforced (woven + nonwoven, W/NW) carrier geotextile. The GCL samples were stored in a storage box at 20~25°C. The initial thickness of the GCL (including both bentonite and geotextiles) was approximately 10~11 mm, with an initial gravimetric

water content of the bentonite of 9.7%. Other basic properties of the GCL are shown in Table 1.

Polystyrene microplastics (PSMP) were chosen as the MP in this study because of their reported presence in landfill leachates and wide availability (Kabir et al., 2023). PSMP with a mean diameter of 1  $\mu\text{m}$  were purchased from Bioscientific Australia. This size was chosen on the assumption that the smaller the MP particle, the more likely it is to penetrate into the GCL. PSMP were provided in 100% dry powders and aqueous suspensions with 2.5% solids (w/v) concentration. The dry powders and aqueous suspensions were stored under 20~25°C and 3~5°C, respectively. PSMP solutions used in swell index (SI) and oedometer swell strain tests were prepared by diluting the stock solution to target concentrations (e.g., 50 mg/L and 100 mg/L). While reported concentrations of MPs reported in studies of landfill leachate are generally much lower, many of these studies are limited to larger-sized MPs. The conservatively high concentrations of PSMP were chosen to increase the likelihood of detecting any effects on GCL swelling.

Table 1. X2000 GCL basic properties.

Properties	Value	Data source
Bentonite mass per unit area ( $\text{g}/\text{m}^2$ )	4250	Supplier
GCL total mass per unit area ( $\text{g}/\text{m}^2$ )	4900	Supplier
Hydraulic conductivity (m/s)	$2.4 \times 10^{-11}$	Supplier
Swell index (mL/2g)	$\geq 24$	Supplier
Gravimetric water content as stored	9.3%	Measured by oven-dried method

### 2.2 Swell index (SI) test

Bentonite clay is the major component in engineering GCLs, contributing to their swelling and barrier performance. The swell index (SI) has been widely used to evaluate the swelling characteristics of bentonite used in GCLs. A series of SI tests were conducted in this study based on the ASTM D 5890 standard, modified to allow measurement of the effect of MPs. Four scenarios of bentonite exposed to MPs were considered, as shown in Table 2. In tests 1, 3 and 4, 2 g of bentonite powder extracted from GCLs was placed at the bottom of a 100 mL volumetric flask. Working solutions (i.e., distilled water and PSMP solutions) were slowly added to the flask along its sidewall to 100 mL. In test 2, 2 g of bentonite was mixed with 10 mg of PSMP dry powder, and the mixture was carefully poured into the bottom of the volumetric flask. Distilled water was slowly added to reach 100 mL. The swelling volume of bentonite was

monitored. Tests 1 and 2 were repeated to confirm the reliability of results. Given the unevenness of the swelling surface of the sample, two readings were taken in each case, corresponding to lowest and highest levels of the surface.

Table 2. Test plan for SI tests.

Test No.	Sample	Working Solution	PSMP /Bentonite Mass Ratio	Repeat Tests
1	2 g of bentonite	Distilled water	/	Yes
2	2 g of bentonite + 10 mg PSMP powder	Distilled water	1:200	Yes
3	2 g of bentonite	Distilled water with 100 mg/L PSMP	1:200	No
4	2 g of bentonite	Distilled water with 50 mg/L PSMP	1:400	No

### 2.3 Oedometer swell strain test

The GCL sheets were cut into 50 mm diameter specimens, and the initial height  $H_0$  of specimens was measured by a vernier calliper. An oedometer test apparatus was employed to measure the vertical swell deformation  $\Delta H$ , as shown in Figure 1. The apparatus consisted of a stainless steel oedometer cell (50 mm inner diameter and 53 mm height), a stainless-steel container and a dial gauge (25.4 mm capacity and 0.0254 mm (1 mil) graduation).

A GCL specimen, a quartz porous disc and a top cap were placed in the oedometer cell. Extra care was needed during the installation to minimise bentonite loss from the edge of the GCL specimen. No porous disc and filter paper were used between the bottom of the container and the GCL specimen to prevent MPs from being entrapped by instrument accessories during hydration. The top cap provided a 2 kPa seating pressure to simulate a near-free swell scenario.

Four types of permeating solutions, loosely approximating onsite scenarios of GCL hydration were considered: distilled water (baseline), PSMP in distilled water,  $CaCl_2$ , and PSMP in  $CaCl_2$ . The permeating solutions were added to the container once the dial gauge reading became stable after applying the seating pressure to the samples in the oedometer setup. The experimental procedures were based on the ASTM D 4546 standard. Dial gauge readings were recorded at eleven different time on the first day (i.e., 15 seconds, 30 seconds, 1 minute, 2 minutes, 4 minutes, 8 minutes, 15 minutes, 30 minutes, 1 hour, 2 hours, 3 hours), and then once a day (i.e., 24 hours, 48

hours, etc) until the swelling rate slowed down to less than 0.01 mm/hour. The swell strains at different time were calculated as  $\Delta H/H_0 * 100\%$ . Duplicate tests were conducted to assess the reliability of results. The different scenarios considered for this test are shown in Table 3.

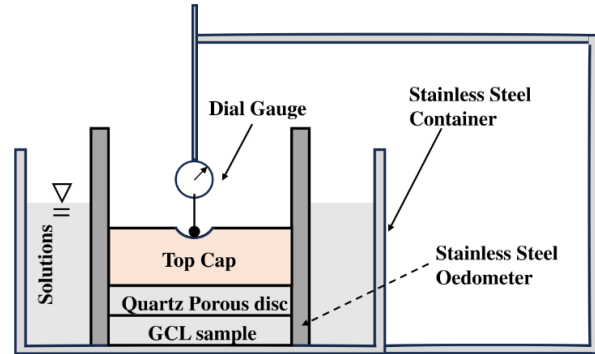


Figure 1. Oedometer test apparatus (not in scale).

Table 3. Test plan for oedometer swell strain tests.

Test No.	Specimen information		Permeating solution
	Initial thickness	Initial GCL mass	
1	10.7 mm	12.2 g	Distilled water
2	10.6 mm	12.5 g	Distilled water
3	10.8 mm	11.0 g	Distilled water with 100 mg/ L PSMP
4	10.5 mm	10.3 g	Distilled water with 100 mg/L PSMP
5	10.7 mm	11.3 g	10 mM $CaCl_2$
6	10.4 mm	11.5 g	10 mM $CaCl_2$
7	10.1 mm	11.5 g	10 mM $CaCl_2$ with 100 mg/L PSMP
8	10.2 mm	10.6 g	10 mM $CaCl_2$ with 100 mg/L PSMP

### 2.4 Scanning electron microscopy (SEM)

Scanning electron microscopy (SEM) imaging of bentonite specimens taken from the sample at the end of the oedometer swell strain tests was further conducted. The aim was to investigate any interactions between bentonite and PSMP, and any changes in the former as a result of exposure to the latter.

A Sigma VP HD (Zeiss) was used for high-resolution imaging. The oedometer-test GCL samples were freeze-dried under  $-50\text{ }^\circ\text{C}$  vacuum conditions. Small amounts of bentonite were extracted from the edge and centre of the freeze-dried sample. In addition, a specimen was cut from the carrier geotextile by a pair of scissors. These specimens were glued on the SEM stub by a piece of carbon tape. Specimens on the stub were gold-coated to minimize charging effects on SEM imaging. This study used an EHT of 5kV and aperture of 30  $\mu\text{m}$ .

### 3 RESULTS AND DISCUSSION

#### 3.1 Swell index (SI) test

The results of the swell index (SI) test are shown in Figure 2. The ASTM D 5890 standard requires 16~24 hours before bentonite volume reading. In the experiments conducted here, it is found that around ten days are needed before swelling ceases.

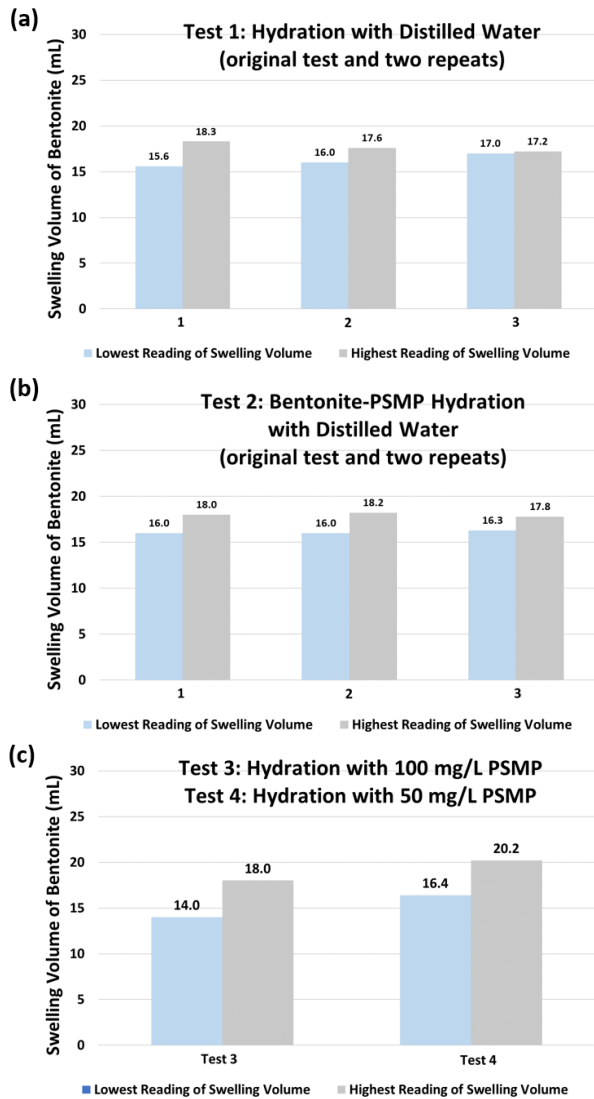


Figure 2. Swell index (SI) results (a) Test 1; (b) Test 2; (c) Tests 3 and 4.

Figures 2a and 2b confirm reasonable repeatability of the SI tests. The standard deviations of the highest volume reading in the set tests 1 and 2 are 0.45 and 0.16 mL, respectively, and fall within the readability of the volumetric flask of 0.5 mL.

No differences can be found in the SI values between pure bentonite and bentonite-PSMP mixture hydrating with distilled water. The average of the highest volume readings in tests 1 and 2 are 17.7 mL and 18.0 mL, respectively. Hence, it appears that, for

a mass ratio of PSMP to bentonite of up to 0.5%, the presence of PSMP does not affect the swell index of sodium bentonite. Tests 2 and 3 studied the SI of bentonite under the same mass ratio between bentonite and PSMP but in different PSMP-bentonite mixing conditions. No differences can be observed in the SI values in these two cases. The SI of bentonite in 100 mg/L PSMP solutions, based on lowest reading, is slightly smaller than that in 50 mg/L PSMP, as seen in Figure 2c, but more data is required before determining whether this difference is significant.

#### 3.2 Oedometer swell strain test

Changes in the vertical swell strain of GCL specimens exposed to PSMP solutions are shown in Figure 3. The rate of swelling initially increases to 0.9 mm/hour then slowly declines after 4 minutes. The swell strains reported below correspond to the time  $t_{limit}$  at which swell rate first reaches 0.01 mm/hour. The value of  $t_{limit}$  was determined through interpolation. Table 4 and Figure 3 show generally good repeatability of experiments, with the possible exception of tests 7 and 8. The swell strain of GCL hydrating in 10 mM  $CaCl_2$  with 100 mg/L PSMP are 34.8% and 39.7% in tests 7 and 8, respectively. In addition, in test 3, the rate of 0.01 mm/hour has been reached after 65 hours, compared to 35 hours in the repeated test 4.

As expected, samples exposed to  $CaCl_2$  experienced reduced swelling (swell strains of 38.3% and 38.9% in tests 5 and 6, compared to 43.0% and 43.1% in tests 1 and 2). This is due to the increase in ion concentration in the pore water which inhibits osmotic swelling of Na-bentonite.

Table 4. Elapsed time and swell strain at 0.01 mm/hour swell rate of oedometer swell strain tests.

Test No.	Hydration solution	$t_{limit}$ (hour)	Swell strain (%)
1	DW	83	43.0
2	DW (repeat)	95	43.1
3	PSMP 100 mg/L in DW	65	42.4
4	PSMP 100 mg/L in DW (repeat)	35	38.7
5	10 mM $CaCl_2$	51	38.3
6	10 mM $CaCl_2$ (repeat)	61	38.9
7	PSMP 100 mg/L in 10 mM $CaCl_2$	45	34.8
8	PSMP 100 mg/L in 10 mM $CaCl_2$ (repeat)	59	39.7

\* DW is distilled water;  $t_{limit}$  is the time it takes to reach 0.01 mm/hour swell rate

The swell strains of tests 3 and 4 are 42.4% and 38.7%, respectively. A small decrease in the swell

strain is observed in GCL hydrated in distilled water with 100 mg/L PSMP compared to that in distilled water. In addition, a decrease in swell strain is found in test 7, compared to tests 4 and 5. However, no such decrease is recorded in repeat test 6 and, therefore, the results are inconclusive. Further tests are needed to identify any co-effects of PSMP and CaCl<sub>2</sub> on the swell characteristics of GCLs.

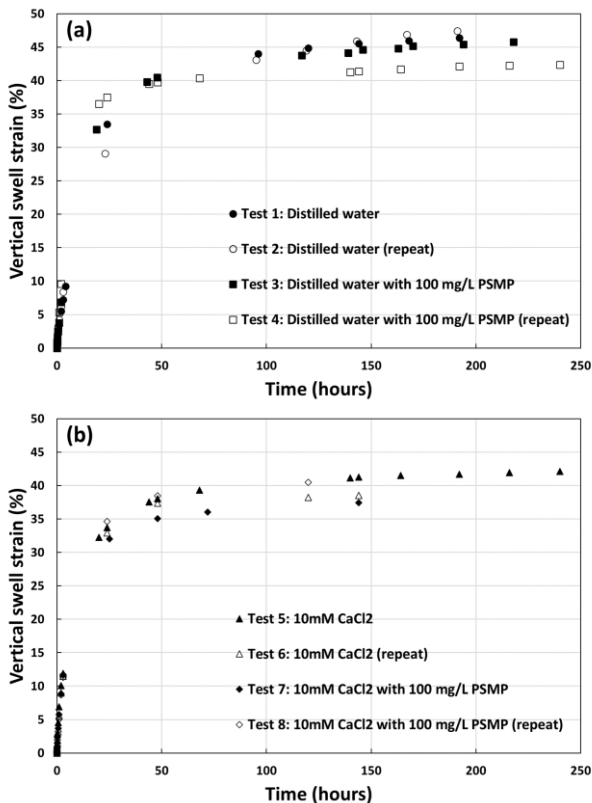


Figure 3. Oedometer swell strain test results (a) Tests 1-4 in distilled water with/without PSMP; (b) Tests 5-8 in 10 mM CaCl<sub>2</sub> with/without PSMP.

### 3.3 SEM of PSMP and clay microstructure

The morphology and size of PSMP are shown in Figure 4. The PSMP particles appear to be regular microspheres with 1 μm size as described by the supplier. The highly regular spherical shape of the particles makes it easier to distinguish them from bentonite and geotextiles in SEM images.

Figure 5 shows SEM images of GCL components of oedometer swell strain test 3. PSMP particles are observed on the bentonite specimen taken at the edge of GCL, as seen in Figure 5a. PSMP particles attach to the clay surface. Some large pores (around 1 μm in size) appear in the clay microstructure although it is unclear whether they are due to interaction with PSMP or handling of the specimen. Figure 5b shows that, on the carrier geotextile specimen, many PSMP particles aggregate and attach to the retained bentonite but not, it appears, to the geotextile itself, possibly indicating a

preference for the clay. It is not clear whether this is accidental or indicates selective attachment to the bentonite. Finally, in Figure 5c, no PSMP can be seen on the bentonite extracted from the centre of GCL. It seems that the PSMP has not penetrated through the bentonite under the conditions of the test reported in this paper.

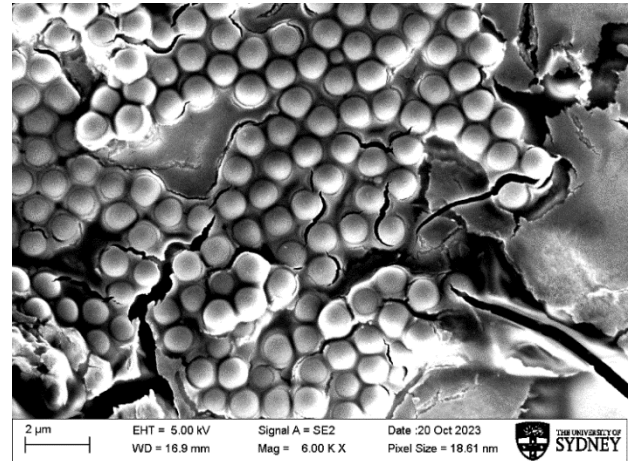


Figure 4. SEM image of PSMP.

## 4 CONCLUSIONS

This study investigated the effects of PSMP on the swelling characteristics of GCLs. The swelling behaviour has been quantified by the swell index and oedometer vertical strain tests. Small decreases in swelling of GCL exposed to 100 mg/L of 1 μm spherical PSMP, relative to control experiments, have been observed. However, further testing is required to establish whether these differences are significant or fall within the range of experimental variability. SEM imaging shows that individual PSMP are retained on the bentonite attached to the carrier geotextile during hydration.

The study has been limited to a single size, single type MP. Further tests are ongoing by the authors to evaluate the effects of higher ionic strength and PSMP concentrations of hydration solutions, as well as the effects of other types, sizes and concentrations of MPs.

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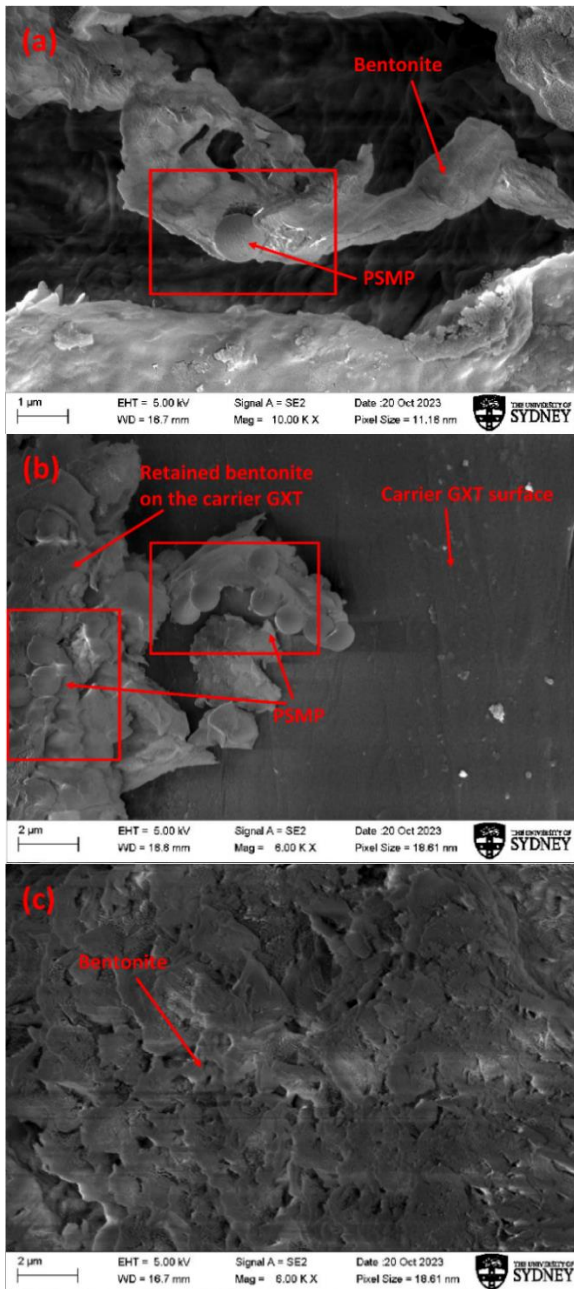


Figure 5. SEM images of the GCL specimen in test 3 (a) bentonite taken from the edge of GCL; (b) carrier geotextile (GXT) specimen with bentonite attached; (c) bentonite taken from the centre of GCL.

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