

Hydraulic conductivity to Jet-A1 of GCLs after up to 100 freeze–thaw cycles

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Needle-punched geosynthetic clay liner (GCL) specimens subjected to up to 100 freeze–thaw cycles in the laboratory, and GCL samples recovered from the field after 3 years, are examined to assess their permeability with respect to both water and Jet A-1 fuel using flexible wall permeameters. The mean GCL hydraulic conductivity with respect to water is shown to be 3.3×10^{-11} m/s before freeze–thaw. The mean hydraulic conductivities after up to 100 freeze–thaw cycles did not change significantly, and were in the range 2.2 – 5.3×10^{-11} m/s. Freeze–thaw cycles did reduce the entry pressure required for Jet A-1 to begin to permeate through the GCL from about 27 to 55 kPa with no freeze–thaw cycles to 13.8–20.7 kPa for 3 to 50 freeze–thaw cycles, and to 0–13.8 kPa after 100 freeze–thaw cycles. For the GCL specimens subjected to 5, 12 and 50 freeze–thaw cycles in the laboratory, the hydraulic conductivity with respect to Jet A-1 is less than 3×10^{-11} m/s at a pressure just above the entry pressure, compared with less than 3×10^{-12} m/s for GCL samples recovered from the field. The combined effect of many freeze–thaw cycles and permeation with Jet A-1 did result in an increase in hydraulic conductivity; however, the effect was small, and the GCL performed well with a maximum hydraulic conductivity of 1×10^{-10} m/s after 100 freeze–thaw cycles.

Des spécimens de chemisages en argile géosynthétique aiguilletés (GCL), soumis à près de 100 cycles de congélation – décongélation en laboratoire, ainsi que des échantillons de GCL prélevés sur le champ au bout de trois ans, sont examinés afin d'établir leur perméabilité à l'eau et au carburéacteur A-1 en utilisant des perméamètres à paroi souple (FWP). On démontre que la conductivité hydraulique moyenne des GCL en présence de l'eau est égale à $3,3 \times 10^{-11}$ m/s avant la congélation – décongélation. Les conductivités hydrauliques moyennes au bout de près de 100 cycles de congélation – décongélation ne changeaient pas de façon significative, et étaient comprises dans la plage $2,2$ – $5,3 \times 10^{-11}$ m/s. Les cycles de congélation – décongélation réduisaient la pression d'entrée nécessaire pour permettre au Jet A-1 de commencer à s'infiltrer dans les GCL d'environ 27 à 55 kPa sans cycles de congélation – décongélation jusqu'à 13,8 – 20,7 kPa pour 3 à 50 cycles de congélation – décongélation, et de 0 à 13,8 kPa au bout de 100 cycles de congélation – décongélation. Pour les spécimens de GCL soumis à 5, 12 et 50 cycles de congélation – décongélation en laboratoire, la conductivité hydraulique pour le Jet A-1 est inférieure à 3×10^{-11} m/s avec une pression tout juste supérieure à la pression d'entrée, contre moins de 3×10^{-12} m/s pour des échantillons de GCL prélevés sur le champ. L'effet cumulé de nombreux cycles de congélation – décongélation et de perméation avec le Jet A-1 a donné lieu à une augmentation de la conductivité hydraulique; toutefois, l'ampleur de cet effet était limité, et le GCL a présenté de bonnes performances avec une conductivité hydraulique maximale de 1×10^{-10} m/s au bout de 100 cycles de congélation – décongélation.

KEYWORDS: geosynthetics; pollution/migration control

INTRODUCTION

A composite liner composed of a fluorine-surface-treated high-density polyethylene (f-HDPE) geomembrane (GM) and a needle-punched geosynthetic clay liner (GCL) was used as temporary barrier to the migration of a hydrocarbon spill from fuel tanks on Brevoort Island in the Canadian eastern Arctic (Li *et al.*, 2002). Following the construction of the barrier system in the summer of 2001, a programme of site monitoring (Bathurst *et al.*, 2006) and laboratory tests was initiated to evaluate the likely performance of both the f-HDPE GM and the GCL in this challenging environment. There has been considerable recent research relating to the use of GCLs in geoenvironmental applications (e.g. Lake & Rowe, 2004; Southen & Rowe, 2005; Rowe *et al.*, 2005;

Barroso *et al.*, 2006; Bouazza & Vangpaisal, 2006; Dickinson & Brachman, 2006; Hurst & Rowe, 2006; Touze-Foltz *et al.*, 2006; Podgorney & Bennett, 2006; Saidi *et al.*, 2006; Bouazza *et al.*, 2007; Katsumi *et al.*, 2008; Vukelić *et al.*, 2008). The present paper is concerned with the potential effect of freeze–thaw cycles on the hydraulic conductivity with respect to Jet A-1 (a common fuel in the Canadian Arctic) of the GCL employed at Brevoort Island, and the implications for the length of time for which this temporary barrier can be expected to contain the hydrocarbon in the subsurface prior to clean-up.

The barrier system was required only down to permafrost at a depth of 2–3 m (Li *et al.*, 2002; Bathurst *et al.*, 2006). Consequently, the barrier is subject to (a) freeze–thaw, (b) low hydraulic head, and (c) low overburden pressure. It is important to emphasise that in this spill-containment application the barrier can come into contact with neat hydrocarbon floating on the water, but that the head difference across the barrier due to the hydrocarbon is small. This is quite different from typical municipal waste landfill base liner applications where: (a) freeze–thaw is unlikely, owing to the insulation provided by the waste, as well as heat generated by the waste; (b) the hydraulic head is normally low, but could be significant if there was a failure of the

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leachate collection system; and (c) hydrocarbons in contact with the barrier are likely to be dissolved in the leachate at low concentrations (typically ppm or lower) rather than neat hydrocarbon experienced in spill-containment applications such as that at Brevoort Island.

Previous research using flexible wall permeameters (FWP) by Hewitt *et al.* (1997), Kraus *et al.* (1997) and Podgorney & Bennett (2006) has shown that needle-punched GCLs with sodium bentonite can withstand at least 150 freeze–thaw cycles without experiencing a significant increase in hydraulic conductivity with respect to water. Using rigid wall permeameters (RWP), Rowe *et al.* (2006) showed that 12 freeze–thaw cycles did result in an increase in the hydraulic conductivity with respect to jet fuel. However, it can be argued that, owing to the high gradients used in these tests, the results may overestimate the effect of freeze–thaw. Thus the objective of the present study is to extend the previous work by Rowe *et al.* (2006) by evaluating the permeability of GCL subjected to freeze–thaw cycles with respect to Jet A-1 using FWPs. Particular attention will be focused on the effect of freeze–thaw on the entry pressure at which permeation by Jet A-1 begins, which could not be evaluated in the previous RWP tests, and the effect of substantially more freeze–thaw cycles on the hydraulic performance of GCLs with respect to a hydrocarbon permeant such as Jet A-1. Consideration will be given to the effect of 0, 5, 12, 50 and 100 laboratory freeze–thaw cycles. In addition, the effect of 3 years of natural freeze–thaw cycles and interaction with the local soil pore water on GCL samples exhumed from the field at Brevoort Island will be examined. In each case the permeability (hydraulic conductivity) will be assessed, first with respect to deaired water and subsequently with respect to Jet A-1 fuel.

PREVIOUS STUDIES

The permeation of GCLs with hydrocarbons has received limited past attention. The limited tests that have been performed have used both FWPs and RWPs. Each have advantages and disadvantages (Petrov *et al.*, 1997a). FWPs allow tests to be conducted at low gradients more representative of field conditions, but it can take an excessively long period of time to conduct compatibility tests at these low gradients. RWPs involve unrealistically high gradients, but allow compatibility tests to be conducted to chemical equilibrium in a reasonable period of time. Daniel *et al.* (1993) permeated unsaturated GCLs with hydrocarbons (benzene, gasoline, methanol, tert-butylethylether, trichloroethylene) using an FWP, and found that for gravimetric water contents less than 50% the GCL had a very high hydraulic conductivity ($k \approx 10^{-7}$ m/s). In contrast, for gravimetric water contents greater than 100%, the hydraulic conductivity was very low ($k < 10^{-11}$ m/s). Petrov *et al.* (1997b) used an RWP to examine the hydraulic conductivity of hydrated GCLs permeated with ethanol/water mixtures at hydraulic gradients of 300–900. They showed that for high concentrations (>75% ethanol) the hydraulic conductivity was higher than that of water owing to diffuse double-layer contractions caused by the ethanol.

Rowe *et al.* (2005) used an RWP to examine the hydraulic conductivity of GCLs with respect to Jet A-1 Fuel for saturated and unsaturated conditions and a range of temperatures. They found that GCLs with a low degree of saturation ($S_r = 60\%$) prior to permeation with hydrocarbon did not perform as well as a hydraulic barrier to jet fuel as specimens with higher (90% and 100%) degrees of saturation, either before or after freezing. This finding is consistent with that of Daniel *et al.* (1993). When frozen, the intrinsic permeability of the unsaturated GCLs decreased, and was lower at -20°C than at -5°C .

Very few studies have examined GCLs subjected to freeze–thaw cycles and then permeated with hydrocarbons. Rowe *et al.* (2004) reported the results of flexible-wall hydraulic conductivity tests using water and Jet A-1 on the GCL used at Brevoort Island and subjected to 0, 6 and 13 freeze–thaw cycles. Like Shan & Lai (2002), who found that there was no flow of gasoline through a GCL under a hydraulic gradient of 150, Rowe *et al.* (2004) noted that there was no flow of Jet A-1 into the GCL until the pressure head above the GCL exceeded a threshold value corresponding to a gradient of 90–160 or more.

Rowe *et al.* (2006) evaluated the hydraulic conductivity (with respect to both water and Jet A-1) of GCLs subjected to 0, 5 and 12 freeze–thaw cycles in the laboratory and after 1 and 3 years in situ at Brevoort Island using an RWP at confining stress levels of 15 ± 3 kPa. It was demonstrated that, because of the interfacial tension between pore water and Jet A-1, the hydraulic conductivity with respect to Jet A-1 was initially very low, but that it increased once Jet A-1 had been forced to permeate through the GCL specimen. It was also demonstrated that the hydraulic conductivity with respect to Jet A-1 increased with the number of freeze–thaw cycles. This increase was attributed to an expansion of the macropores in the GCL due to freezing, and an expansion of free-pore space due to double-layer contraction as a result of interaction with the permeating Jet A-1. Using Olsen's (1961) cluster model it was estimated that the double layer contracted by 20–40% after permeating with Jet A-1, while the free space expanded to 1.2–2.5 times that before Jet A-1 permeation. SEM images showed that the bentonite pore size for GCLs subjected to up to 12 freeze–thaw cycles is two to three times larger than that of the bentonite in the virgin GCL.

Both the SEM images and the cluster model suggested that the increase in hydraulic conductivity with respect to Jet A-1 was predominantly due to the development of macropores during freezing, which gave rise to an increase in free-pore space. However, owing to the nature of the RWP tests it was not possible to establish the entry pressures or the effect of hydraulic gradient on the hydraulic conductivity. The results also suggested that further freeze–thaw cycles might lead to a decrease in the entry pressure and a further increase in hydraulic conductivity, but this hypothesis could not be confirmed since only 12 freeze–thaw cycles were examined. On the other hand, it was acknowledged that the RWP tests were conducted at gradients substantially higher than would be expected in the field, and it was hypothesised that they would overestimate the effects of freeze–thaw. These findings have prompted the current study, which is directed at testing these hypotheses.

MATERIALS AND TEST METHODS

Permeants

The Jet A-1 used in this study had a measured density of 0.82 g/ml (standard deviation ± 0.005) based on ASTM D 1480-02 (ASTM, 2004), and the interfacial tension of de-ionised water and Jet A-1 at 20 (± 1) $^\circ\text{C}$ was 32.3 (± 2) mN/m based on ASTM D 971-91 (ASTM, 1994).

Sample preparation

The thermally treated, needle-punched GCL (Bentofix NWL) tested in the investigation was composed of a layer of granular sodium bentonite sandwiched between a non-woven cover geotextile and a scrim-reinforced carrier geotextile. The GCL had a specified minimum average roll value of mass per unit area of 4060 g/m², but the actual roll of GCL had substantially greater mass per unit area than

specified, with an average value of 5010 g/m² (standard deviation of 510 g/m²). The GCL obtained from the field was nominally the same as that tested in the laboratory, with the same initial properties, although because of the variability that can occur in a roll of GCL the mass per unit area of the samples from the field was at the upper end of the range measured for this product, as indicated in Table 1. GCL specimens were prepared in 70 mm diameter hydration chambers. In each case, the virgin GCL specimens were prepared as described below. The specimens were hydrated for 5 days with water under a confining pressure of about 15 (±3) kPa and a hydraulic gradient of 20 (Step 1). The hydration chambers were placed in a freezer at –15°C for about 24 h and then removed and placed in a room with a regulated temperature of 21 (±1) °C for about 24 h (ASTM D 6035-96; ASTM, 1997). This cycle was repeated up to the number of freeze–thaw cycles of interest (i.e. 5, 12, 50 and 100). In the last cycle, the frozen GCL specimen was removed from the hydration chamber and installed into the FWP to thaw for the last time. This procedure minimised the effect of stress release when the specimen was transferred from the hydration chamber to the FWP, because the frozen specimen did not experience any significant volume change.

At the time of installation of the barrier at Brevoort Island (Bathurst *et al.*, 2006), samples of the geomembrane and GCL were buried at a range of depths similar to that of the materials used in the barrier, to allow subsequent exhumation and testing. After 3 years some of these samples were carefully excavated, placed in sealed plastic containers to prevent moisture loss, and brought back to the laboratory for testing. Owing to the potential for additional free swell there was probably some increase in bulk void ratio between exhumation and testing. To the extent that this would impact on the findings it could be expected to result in worse behaviour than for samples in situ; however, as will be seen in the subsequent sections, the behaviour of the exhumed samples was still very good. The GCL samples recovered from the field were installed into an FWP at the field water content without the hydration process in the laboratory

Flexible wall chemical compatibility test

FWP hydraulic conductivity tests were performed using a Tri-Flex 2 permeability test system (Hoskin Scientific Ltd) with 195 ml pressure interface chambers (model K-790) to

control and monitor volumes of Jet A-1 inflow (bottom) and outflow (top). Because of chemical interaction between Jet A-1 and conventional membrane sleeves (Mukunoki *et al.*, 2003), a special Viton membrane sleeve was used in these tests. There was no evidence of chemical interaction in tests with Jet A-1 permeation over a period exceeding 1 year.

Freeze–thaw cycle tests

The initial fluid pressure difference across a GCL specimen was 6.9 kPa. The average effective stress at the middle of the GCL was 13.1 kPa. Permeants flowed from the cover geotextile to the carrier geotextile side of the GCL. Each GCL specimen was permeated with water to establish hydraulic conductivity to water at 21 (±1) °C until the ratio of inflow and outflow volume equalled unity. It was then permeated with Jet A-1 at 21 (±1) °C.

Owing to the interfacial tension between Jet A-1 and pore water in the GCL, Jet A-1 will not flow through the GCL until applied pressure exceeds the entry pressure. In this study, the inflow fluid pressure was increased in steps. Table 2 summarises the applied fluid pressure difference Δu across the GCL, and the effective stress at the mid point in the GCL for the tests conducted. The applied hydraulic gradient is given by

$$i = \frac{\Delta u / \gamma_L}{H_f} \quad (1)$$

where γ_L is the unit weight of the permeant at 21°C and H_f is the thickness of the GCL (Table 1).

RESULTS

GCL subjected to up to 100 freeze–thaw cycles

Figure 1(a) shows a side view of virgin GCL (0 freeze–thaw cycles) before permeating with water. The side view of a GCL specimen subjected to 12 freeze–thaw cycles was very similar to that with no freeze–thaw cycles. However, after 50 freeze–thaw cycles some macrostructure had developed, and this increased with the number of cycles. An example specimen after 100 freeze–thaw cycles is shown in Fig. 1(b).

The bulk void ratio of the GCLs was calculated in accordance with Rowe *et al.* (2004, Appendix D, p. 551). Table 1 summarises the bulk void ratio of each GCL speci-

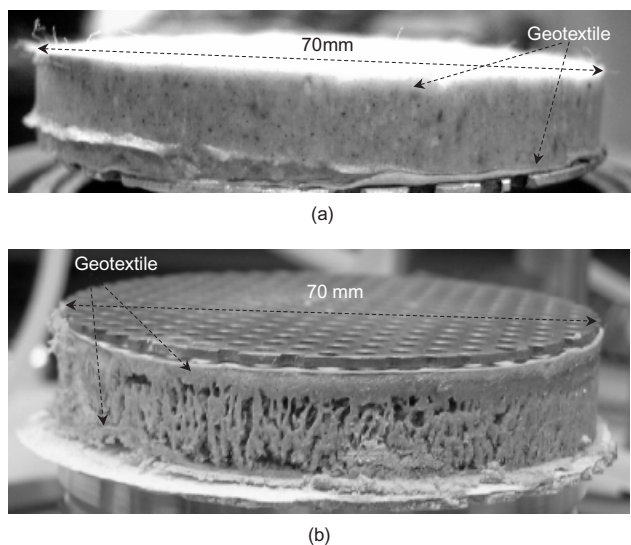
Table 1. Mass per unit area, bulk void ratio before Jet A-1 permeation, and liquid content and specimen thickness after termination of testing

Test number	Mass per unit area of GCL: g/m ²	Number of freeze–thaw cycles	Bulk void ratio before Jet A-1 permeation	Mean bulk void ratio before Jet A-1 permeation	Liquid content after termination: %	Mean liquid content after termination: %	GCL thickness after termination, H_f : mm
1	5389	0	3.0	2.7	143.9	148.3	9.2
2	4049		2.4		152.7		7.7
3	5644	5	3.7	4.4	189.7	190.4	12.6
4	5298		5.0		191.1		14.1
5	4771	12	4.3	4.6	225.7	218.1	11.2
6	4649		4.8		210.5		11.2
7	4986	50	5.5	4.7	257.0	252.0	17.1
8	4407		3.9		247.0		11.8
9	4628	100	4.5	4.6	325.0	286.8	13.5
10	5205		4.7		248.5		13.0
11	5493	3*	5.3	5.5	242.0	229.0	20.0
12	5631		5.7		216.0		16.4

*Samples removed from the field after 3 years; number of freeze–thaw cycles based on field monitoring of subsurface temperatures using buried thermocouples (Bathurst *et al.*, 2006).

Table 2. Applied fluid pressure difference across GCL (Δu) and effective stress (σ') at mid point of GCL specimen

Permeant	Effective stress, σ' : kPa	Pressure difference across GCL, Δu : kPa
Water and Jet A-1	13.1	6.9
	16.5	13.8
	20.0	20.7
	23.4	27.6
	26.9	34.5
Jet A-1	30.3	41.3
	33.8	48.2
	37.3	55.1
	40.7	62.0

**Fig. 1. Side view of GCL specimens: (a) without freeze-thaw cycles; (b) after 100 freeze-thaw cycles**

men before Jet A-1 permeation and the liquid content after termination of the testing. The liquid content, which is defined as fluid mass (i.e. pore water and Jet A-1) divided by dry mass of bentonite in the GCL, was measured in the same way as moisture content. As shown in Table 1, the liquid content of the bentonite increased with the number of freeze-thaw cycles. The mean liquid content after 100 freeze-thaw cycles was 1.9 times greater than that for no freeze-thaw cycles. The mean bulk void ratio of 4.6 (standard deviation ± 0.56) after 5, 12, 50 and 100 freeze-thaw cycles was 1.7 times greater than that with no freeze-thaw cycles. Since the freeze-thaw cycles were conducted without allowing additional water to enter the specimen, the initial moisture content (before permeation) was essentially the same for all cases. The increase in liquid content with freeze-thaw cycles is attributed to filling of the increased voids formed during freeze-thaw with either water or Jet A-1 during permeation. It is noted that GCL specimens taken from field samples had the greatest bulk void ratio of all the samples examined. This was in part because the mass per unit area of the samples from the field was, coincidentally, at the upper end of the normal range of mass per unit area encountered in a roll of GCL (see Table 1), as noted earlier, and in part because of free swell that probably occurred between sampling in the field and testing in the laboratory. Apart from this minor difference, the GCL from the field was identical to that tested in the laboratory. The

liquid content was typical of that for laboratory-prepared specimens after 12–50 freeze-thaw cycles.

Hydraulic conductivity and permittivity of GCL with respect to water

Table 3 summarises the hydraulic conductivity and permittivity with respect to water for all the GCL specimens examined. The permittivity was calculated from the measured hydraulic conductivity and the thickness of the GCL at the termination of the test (Table 1). Although the GCL specimens recovered from the field after 3 years have the greatest bulk void ratio of all the cases examined (Table 1), the mean hydraulic conductivity (i.e. 2.3×10^{-11} m/s; Table 3) and permittivity (1.25×10^{-9} s $^{-1}$) was the smallest. The permittivity ratio, defined as the ratio of the permittivity at a given number of freeze-thaw cycles to that with no freeze-thaw cycles, typically decreased by 20–30% owing to freeze-thaw cycles, as shown in Table 3. The permittivity ratio was the smallest for the GCL recovered from the field in both the FWP tests reported herein (0.3) and in the RWP tests (0.2) reported by Rowe *et al.* (2006). Although there is some variability from specimen to specimen, it can be concluded that, from a practical perspective, there is insignificant difference between the hydraulic conductivity or permittivity of samples subjected to up to 100 freeze-thaw cycles compared with the virgin sample when permeated with water at an average effective stress of 13 kPa. The findings from the FWP and RWP tests were also quite consistent with respect to the effect of freeze-thaw on permittivity for the 12 cycles examined in both test series.

Hydraulic conductivity with respect to Jet A-1

Figures 2 and 3 show the cumulative inflow volume plotted against elapsed time for test specimens initially subjected to 0 and 100 freeze-thaw cycles. Given the design of the apparatus, after replacement of water with Jet A-1 in the supply chamber there is about 12.4 ml of water in the tube, porous disc and geotextile above the bentonite layer that must be displaced before the Jet A-1 reaches the bentonite layer of the GCL. Thus there is flow until this occurs. However, once the jet fuel reaches the bentonite, the flow stops until the applied pressure is increased to exceed the entry pressure. Except for Test 9, Jet A-1 did not permeate the GCL at a fluid pressure difference across the GCL specimen of 6.9 kPa. The applied fluid pressure was increased in steps every 7–10 days until a pressure was reached at which Jet A-1 began to flow through the specimen. This pressure is recorded in Table 4, together with the bulk void ratio at the end of the test. Since the pressure was increased in 6.9 kPa steps, the actual entry pressure lies within the range between the last pressure difference shown with no flow (NF) in Table 4 and the value where the first hydraulic conductivity with respect to Jet A-1 is recorded.

The entry pressure is sensitive to the structure of the GCL sample (i.e. the distribution and size of the larger macropores) and hence varies from sample to sample. This effect is exacerbated by freeze-thaw owing to the effects of different ice-lensing that occurs in each specimen on freezing (see Fig. 1(b) for an example of ice-lensing). For no freeze-thaw cycles the entry pressure ranged from about 27 kPa to about 55 kPa. Freeze-thaw appears to have increased the pore size and consequently reduced the entry pressure. For 50 or fewer freeze-thaw cycles the entry pressure exceeded 6.9 kPa. After 100 cycles the entry pressure was less than 6.9 kPa for Test 9 and between 6.9 kPa and 13.8 kPa for Test 10. The GCL specimen recovered from the field after 3 years of

Table 3. Hydraulic conductivity, permittivity with respect to water and permittivity ratio for FWP and RWP tests

Test number	Number of freeze–thaw cycles	Hydraulic conductivity: m/s	Mean hydraulic conductivity: m/s	Permittivity, ϕ : s^{-1}	Average permittivity: s^{-1}	Permittivity ratio in FWP tests, ϕ_{nF-TCs}/ϕ_0	Permittivity ratio in RWP tests, ϕ_{nF-TCs}/ϕ_0 (Rowe <i>et al.</i> , 2006)
1	0	3.0×10^{-11}	3.3×10^{-11}	3.1×10^{-9}	3.8×10^{-9}	1.0	1.0
2		3.6×10^{-11}		4.5×10^{-9}			
3	5	3.2×10^{-11}	4.3×10^{-11}	2.3×10^{-9}	3.0×10^{-9}	0.8	0.8
4		5.3×10^{-11}		3.7×10^{-9}			
5	12	4.2×10^{-11}	3.1×10^{-11}	3.5×10^{-9}	2.6×10^{-9}	0.7	1.0
6		2.0×10^{-11}		1.6×10^{-9}			
7	50	7.8×10^{-11}	5.3×10^{-11}	5.2×10^{-9}	3.8×10^{-9}	1.0	–
8		2.7×10^{-11}		2.4×10^{-9}			
9	100	4.2×10^{-11}	3.6×10^{-11}	3.3×10^{-9}	2.8×10^{-9}	0.7	–
10		3.0×10^{-11}		2.2×10^{-9}			
11	3*	2.5×10^{-11}	2.3×10^{-11}	1.2×10^{-9}	1.25×10^{-9}	0.3	0.2
12		2.2×10^{-11}		1.3×10^{-9}			

*Samples removed from the field after 3 years; number of freeze–thaw cycles based on field monitoring of subsurface temperatures using buried thermocouples (Bathurst *et al.*, 2006).

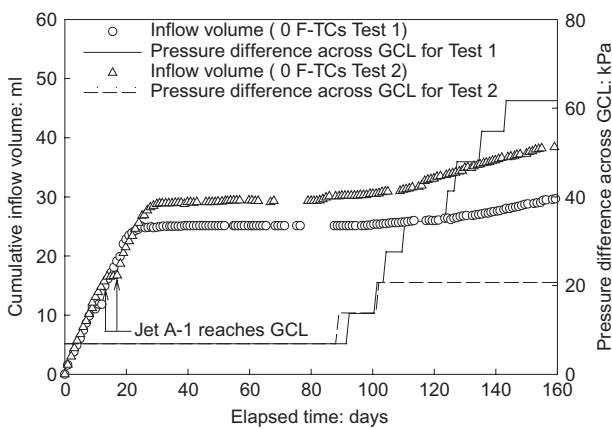


Fig. 2. Cumulative flow volume and pressure difference across GCL against elapsed time for specimen without freeze–thaw cycles

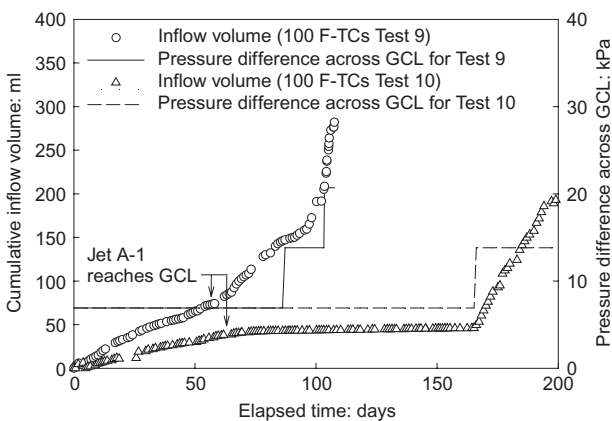


Fig. 3. Cumulative flow volume and pressure difference across GCL against elapsed time for specimen subjected to 100 freeze–thaw cycles

natural freeze–thaw cycles had entry pressures exceeding 6–9 kPa.

After a period of permeation at just above the entry pressure, the fluid pressure was increased and the hydraulic conductivity measured at this increased pressure. The results are given in Table 4. As shown for Test 2 in Fig. 4, even with no freeze–thaw cycles the hydraulic conductivity with

respect to Jet A-1 is a function of the fluid pressure, and hence of the hydraulic gradient. For this test there was no flow of Jet A-1 until the pressure difference reached 27.6 kPa, at which point the hydraulic conductivity was 0.14×10^{-11} m/s. When the pressure increased from 34.5 kPa and 41.3 kPa the hydraulic conductivity increased from 0.65×10^{-11} m/s to 7.8×10^{-11} m/s (Table 4). At any given gradient the hydraulic conductivity remained constant. For example, Fig. 5 shows that for Test 10, after 100 freeze–thaw cycles, the hydraulic conductivity with respect to Jet A-1 remained constant at 1.1×10^{-10} m/s up to a permeation of 4.1 pore volumes of Jet A-1 through the specimen.

Figure 6 shows plots of hydraulic conductivity against the number of freeze–thaw cycles for five pressure differences across the GCL. Because of the stepwise manner with which the pressure was increased, the entry pressure will have been exceeded by different amounts for the different samples. Notwithstanding this, there is a trend of increasing hydraulic conductivity with respect to Jet A-1, with an increase in (a) the number of freeze–thaw cycles, and (b) the pressure difference across the specimen (i.e. gradient). It is also clear that for situations relevant to the field application, where the fluid pressure difference across the GCL would be less than 7 kPa, there was no flow for the field-recovered samples (where there is typically only one freeze–thaw cycle per year) or any of the virgin samples, except one of the two subjected to 100 freeze–thaw cycles, and even for this case the hydraulic conductivity was still only 1.1×10^{-10} m/s. Given the typically low number of freeze–thaw cycles per year observed in the field, and the low gradient across the GCL, this suggests that, with respect to the effects of freeze–thaw, the GCL investigated in this study is likely to perform well in the field for many decades.

The hydraulic conductivity with respect to Jet A-1 obtained from RWP tests by Rowe *et al.* (2006) for 0, 5 and 12 freeze–thaw cycles and on samples recovered after 3 years in the field is also reported in Table 4. Comparing the results with those from the FWP reported herein for similar numbers of freeze–thaw cycles it can be seen that, as hypothesised earlier, the RWP tests give substantially higher k values. This arises because of the very large gradients that are typically developed in RWP tests. Since these gradients far exceed those likely to be experienced in field applications such as that considered here, it is apparent that RWP tests will overestimate the effect of freeze–thaw on the hydraulic conductivity of GCLs in contact with hydrocarbons such as Jet A-1.

Table 4. Hydraulic conductivity k with respect to Jet A-1 from RWP tests and FWP tests for fluid pressure difference (Δu) across GCL specimen, and bulk void ratio at end of test

Test number	Number of freeze-thaw cycles	k from RWP tests: m/s (Rowe <i>et al.</i> , 2006)	k with respect to Jet A-1 from FWP tests (this paper): m/s								Bulk void ratio after Jet A-1 permeation	Mean bulk void ratio after Jet A-1 permeation
			Δu : kPa									
			6.9	13.8	20.7	27.6	34.5	41.3	55.2	68.6		
1	0	2×10^{-11}	NF	NF	NF	NF	NF	NF	NF	NF	3.7	3.2
2	5	8×10^{-11}	NF	NF	NF	NF	NF	NF	NF	NF	2.7	5.0
3	12	1.45×10^{-10}	NF	NF	NF	NF	NF	NF	NF	NF	4.4	5.0
4	50	ND	2.1×10^{-11}	NF	NF	NF	NF	NF	NF	NF	5.5	5.0
5	100	ND	NF	NF	NF	NF	NF	NF	NF	NF	5.2	6.3
6	3*	5.3×10^{-11}	NF	NF	NF	NF	NF	NF	NF	NF	4.8	5.6
7			7.8×10^{-11}	NF	NF	NF	NF	NF	NF	NF	7.5	7.1
8				9.2×10^{-12}	2.6×10^{-11}	1.4×10^{-12}	6.5×10^{-12}	7.8×10^{-11}	4.1×10^{-11}	9.1×10^{-12}	5.9	
9				7.8×10^{-13}	8.4×10^{-13}	2.6×10^{-12}	7.9×10^{-11}	1.6×10^{-11}	1.5×10^{-10}	2.5×10^{-12}	5.2	
10				2.0×10^{-11}	8.4×10^{-12}	2.6×10^{-12}	7.9×10^{-11}	1.6×10^{-11}	1.5×10^{-10}	9.1×10^{-12}	8.2	
11				1.1×10^{-12}	2.6×10^{-12}	1.4×10^{-12}	6.5×10^{-12}	7.8×10^{-11}	4.1×10^{-11}	2.5×10^{-12}	6.0	
12				3.4×10^{-10}	1.1×10^{-12}	3.0×10^{-11}	1.5×10^{-10}	9.1×10^{-12}	2.5×10^{-12}	3.3×10^{-11}		

*Samples removed from the field after 3 years; number of freeze-thaw cycles based on field monitoring of subsurface temperatures using buried thermocouples (Bathurst *et al.*, 2006).

[†]Hydraulic conductivity in the range of entry pressure.

NF = no flow of influent. ND = no data.

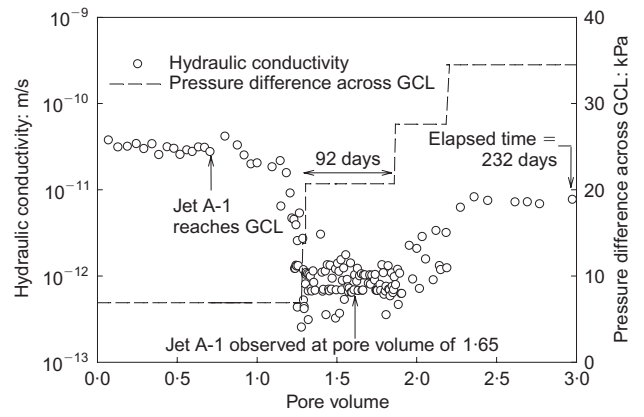


Fig. 4. Variation in hydraulic conductivity and fluid pressure difference across GCL against pore volumes permeated through specimen for Test 2 (no freeze-thaw cycles)

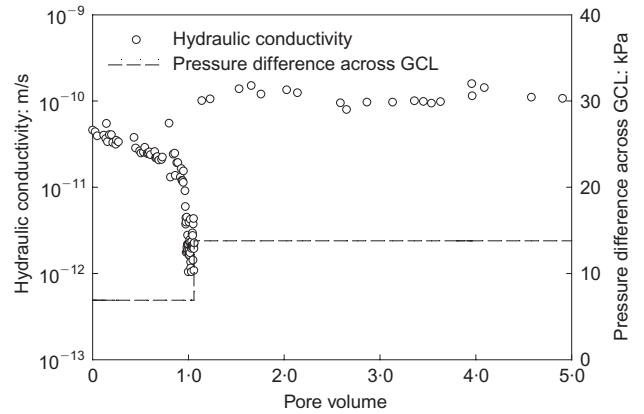


Fig. 5. Variation in hydraulic conductivity and fluid pressure difference across GCL against pore volumes permeated through specimen for Test 10 (100 freeze-thaw cycles)

Intrinsic permeability

The intrinsic permeability K_L is calculated from

$$K_L = \frac{k_L \eta_L}{\gamma_L} \tag{2}$$

where k_L is a hydraulic conductivity for the permeant [LT^{-1}], η_L is a dynamic viscosity of the permeant [$ML^{-1}T^{-1}$], and γ_L is a unit weight of the permeant [$ML^{-2}T^{-2}$]. For a given permeant, a change in intrinsic permeability indicates a change in pore structure. Pore structure is affected by the change of effective stress, so it is important to recognise this when considering the intrinsic permeability obtained from different tests. Table 5 gives the intrinsic permeability based on permeation with water at a 6.9 kPa fluid pressure difference across the specimens, and for Jet A-1 at a 20.7 kPa pressure difference. The intrinsic permeability of the GCL permeated with de-aired water at $\Delta u = 6.9$ kPa did not vary significantly with the number of freeze-thaw cycles (see Fig. 7).

Average values of intrinsic permeability with respect to Jet A-1 for a given number of freeze-thaw cycles are listed in Table 5. There is a trend of increasing intrinsic permeability with increasing number of freeze-thaw cycles when these data are plotted as shown in Fig. 7. The samples recovered from the field after 3 years still had quite a low average intrinsic permeability, similar to that of samples with 0 to 12 freeze-thaw cycles. Hence the laboratory results are consistent with field specimens that experienced three freeze-thaw cycles over a 3-year period.

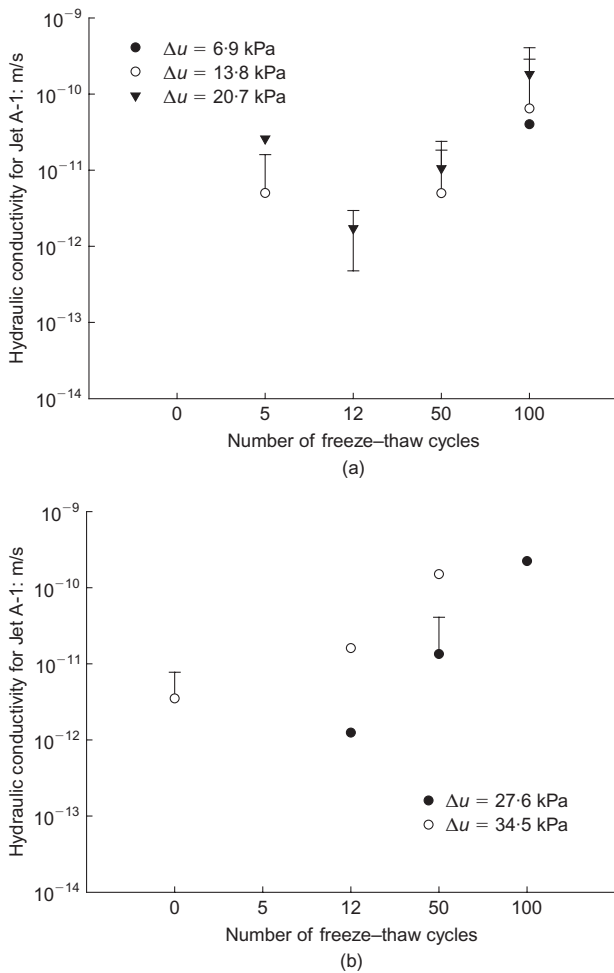


Fig. 6. Hydraulic conductivity of GCLs permeated with Jet A-1 at fluid pressure differences across GCL of: (a) 6.9, 13.8 and 20.7 kPa; (b) 27.6 and 34.5 kPa

Permittivity

Figure 8 shows the variation in permittivity of the GCL with respect to the number of freeze–thaw cycles. It can be seen that there was really no visually consistent trend in the

effect of freeze–thaw on permittivity with respect to de-aired water. However, for specimens permeated with Jet A-1 there is a trend of increasing permittivity with the number of freeze–thaw cycles, although the permittivity remained low (typically below that with respect to water as given in Table 3) for 50 or fewer freeze–thaw cycles.

Table 5 also reports the permittivity values obtained from RWP tests by Rowe *et al.* (2006). Comparison of these values with those from the FWP tests reported herein indicates that the RWP tests give substantially higher permittivities for a given number of freeze–thaw cycles, and emphasises the point made earlier that the RWP tests are likely to overestimate the effect of freeze–thaw on the performance of GCLs when permeated by hydrocarbons.

DISCUSSION

For 50 or fewer freeze–thaw cycles the entry pressure was greater than 6.9 kPa, and no flow of Jet A-1 would be expected through the GCL under field conditions such as those similar to the barrier application at Brevoort Island. However, as indicated by the intrinsic permeability values shown in Fig. 7, freeze–thaw did alter the pore structure of the GCL (see also Fig. 1). This change had two effects. First, it tended to reduce the entry pressure. This is probably due to the development of large macropores, which more readily permit entry of Jet A-1, as identified by Rowe *et al.* (2006). Second, it gave rise to an increase in hydraulic conductivity at a given pressure difference above the entry pressure. This is in sharp contrast to the findings with respect to permeation by de-aired water, where there was no evident increase in hydraulic conductivity, intrinsic permeability or permittivity with up to 100 freeze–thaw cycles. This indicates that although there may be a change in pore structure due to freeze–thaw, it had no significant effect on the performance of the GCL as a barrier to water at an effective stress of 13 kPa.

The sensitivity of the hydraulic conductivity with respect to Jet A-1 to the pressure difference across the GCL means that, to get values relevant to actual field conditions, tests should be performed at an effective stress and gradient representative of the field conditions. Comparisons of the results reported herein using FWP tests with those obtained

Table 5. Intrinsic permeability of GCLs tested with respect to water and Jet A-1, and permittivity with Jet A-1 from FWP tests and RWP tests

Test number	Number of freeze–thaw cycles	Intrinsic permeability after permeation with water at $\Delta u = 6.9$ kPa: m^2	Intrinsic permeability after permeation with Jet A-1 at $\Delta u = 20.7$ kPa: m^2	Average intrinsic permeability after permeation with Jet A-1: m^2	Permittivity after permeation with Jet A-1, ϕ : s^{-1}	Average permittivity after permeation with Jet A-1 in FWP: s^{-1}	Average permittivity after permeation with Jet A-1 in RWP: s^{-1} (Rowe <i>et al.</i> , 2006)
1	0	3.1×10^{-18}	NF	NF	NF	NF	2.75×10^{-9}
2		3.7×10^{-18}	NF		NF		
3	5	3.3×10^{-18}	2.2×10^{-18}	2.2×10^{-18}	1.4×10^{-9}	1.40×10^{-9}	7.8×10^{-9}
4		5.4×10^{-18}	N/A		N/A		
5	12	4.3×10^{-18}	2.2×10^{-19}	1.4×10^{-19}	2.0×10^{-10}	1.2×10^{-10}	1.42×10^{-8}
6		2.0×10^{-18}	7.0×10^{-20}		1.0×10^{-10}		
7	50	7.9×10^{-18}	1.67×10^{-18}	8.8×10^{-19}	9.0×10^{-10}	4.9×10^{-10}	ND
8		2.8×10^{-18}	9.0×10^{-20}		1.0×10^{-10}		
9	100	4.3×10^{-18}	2.85×10^{-17}	2.43×10^{-17}	2.14×10^{-8}	1.76×10^{-8}	ND
10		3.1×10^{-18}	2.01×10^{-17}		1.38×10^{-8}		
11	3*	2.6×10^{-18}	2.3×10^{-19}	1.2×10^{-19}	1.0×10^{-10}	7.0×10^{-11}	7.5×10^{-9}
12		2.2×10^{-18}	1.0×10^{-20}		1.0×10^{-10}		

*Samples removed from the field after 3 years; number of freeze–thaw cycles based on field monitoring of subsurface temperatures using buried thermocouples (Bathurst *et al.*, 2006). NF: No flow of influent. ND = no data.

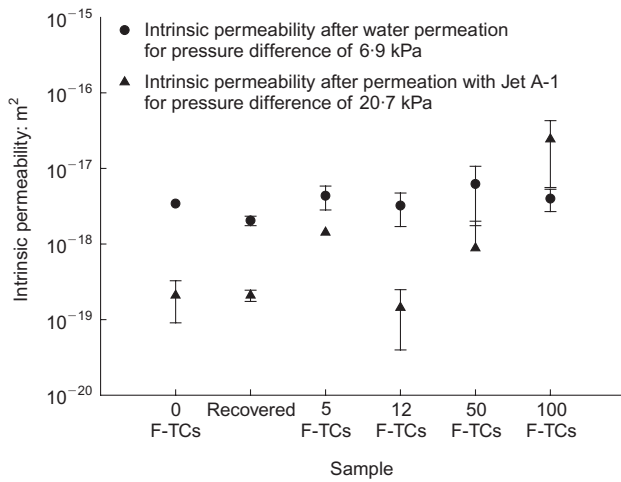


Fig. 7. Intrinsic permeability of GCLs permeated with water and Jet A-1

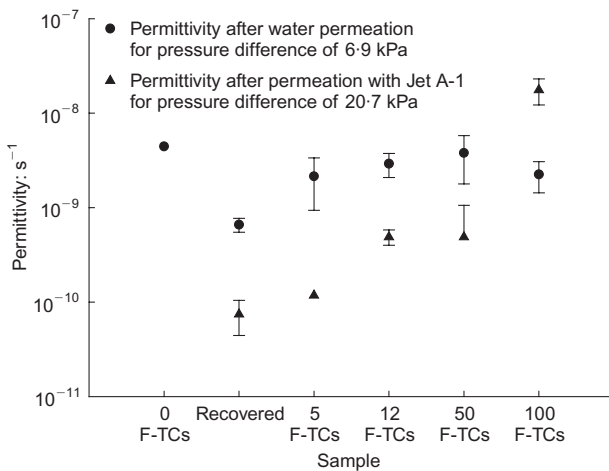


Fig. 8. Permittivity of GCLs permeated with water and Jet A-1

by Rowe *et al.* (2006) using RWP (Tables 4 and 5) demonstrate that tests conducted at high gradients, as is commonly the case when an RWP is used, will overestimate the effect of clay–hydrocarbon interaction and hence the hydraulic conductivity of the GCL.

The GCL samples recovered from the field after 3 years performed very well, with no flow at a 6.9 kPa pressure difference, and a low hydraulic conductivity at higher pressure differences. Thus it appears that the three freeze–thaw cycles experienced by the GCL in the field had not significantly impacted on its performance as a barrier to Jet A-1.

Despite the effect of freeze–thaw on the GCL, the data suggest that the GCL would remain an excellent barrier to Jet A-1 under the field conditions anticipated at Brevoort Island for at least 100 freeze–thaw cycles, and the hydraulic conductivity of the GCL with respect to Jet A-1 is expected to be less than 8.0×10^{-11} m/s.

All tests reported herein were conducted after permeation with water, and hence were saturated prior to permeation by Jet A-1. This is considered appropriate for the conditions at Brevoort Island, where the samples recovered from the field had a high degree of saturation (> 90%). However, it should be emphasised that the hydraulic conductivity of the GCL with respect to hydrocarbons will be sensitive to the degree of saturation (water content) of the bentonite in the GCL at the time of contact with hydrocarbons, as has been demonstrated by Daniel *et al.* (1993) and Rowe *et al.* (2005). Thus the findings reported herein may not be applicable to GCLs

that do not have a high degree of saturation prior to contact with hydrocarbons.

CONCLUSIONS

The hydraulic conductivity with respect to Jet A-1 fuel was examined for a thermally locked, needle-punched GCL used to construct a trial subsurface barrier against hydrocarbon-contaminated groundwater at Brevoort Island in the Canadian Arctic (Li *et al.*, 2002; Bathurst *et al.*, 2006). Hydraulic conductivity tests were performed on saturated virgin GCL samples subjected to 0, 5, 12, 50 and 100 freeze–thaw cycles, and also on specimens from sample coupons recovered from the field at Brevoort Island after 3 years in place. The tests reported herein were performed on the same GCL as that studied by Rowe *et al.* (2006); however, these previous tests were performed with an RWP for only up to 12 freeze–thaw cycles. In contrast, the present paper examined the results obtained using an FWP for up to 100 freeze–thaw cycles and hence provided insights into the effect of freeze–thaw on entry pressure and the hydraulic conductivity of GCLs to a hydrocarbon at low gradients representative of the field application at Brevoort Island. The results from the present test were compared with those obtained by Rowe *et al.* (2006) using an RWP.

The results from the current tests, performed using an FWP at $21 (\pm 1)$ °C, indicate the following conclusions for the GCL investigated and conditions examined.

- The mean hydraulic conductivity with respect to water was 3.3×10^{-11} m/s before freeze–thaw. The range of mean hydraulic conductivities after up to 100 freeze–thaw cycles was $2.2\text{--}5.3 \times 10^{-11}$ m/s at an effective stress of 13 kPa and a pressure difference 6.9 kPa across the GCL specimens (from virgin and field-recovered samples).
- The entry pressure of Jet A-1 for GCL with no freeze–thaw cycles was between about 27 and 55 kPa. The range of entry pressure for GCL subjected to 3 (field samples), 5, 12 and 50 freeze–thaw cycles was 13.8–20.7 kPa and that for 100 freeze–thaw cycles was 0–13.8 kPa. Thus freeze–thaw did reduce the entry pressure, and this is attributed to an increase in the size of the bentonite macropores, confirming the hypothesis based on the findings of Rowe *et al.* (2006).
- For the GCL specimens subjected to 5, 12, and 50 freeze–thaw cycles, the hydraulic conductivity with respect to Jet A-1 was less than 3×10^{-11} m/s at the pressure required to initiate flow (i.e. just above the entry pressure).
- The hydraulic conductivity of GCL recovered from the field with respect to Jet A-1 was less than 0.3×10^{-11} m/s at the low pressure heads expected in this field application.
- The combined effect of freeze–thaw cycles and permeation with Jet A-1 did result in some increase in hydraulic conductivity at low effective stress (13–16.5 kPa) and applied fluid pressure differences across the specimens in the range 6.9–13.8 kPa. However, the effect was small, and the GCL performed well after up to 100 freeze–thaw cycles (maximum hydraulic conductivity of 1×10^{-10} m/s after 100 cycles).
- The RWP tests of Rowe *et al.* (2006) and FWP tests reported herein gave rise to similar findings with respect to the effect of freeze–thaw on samples permeated with water, but it was shown that because the RWP involves gradients substantially higher than would be expected in the field application considered

here, they overestimated the effects of freeze–thaw with respect to permeation by Jet A-1.

Based on the above results, and provided the GCL remains saturated or near saturated, it appears that the GCL used as a component of a composite liner in the spill-containment application at Brevoort Island can be expected to perform well as a barrier with respect to the combined effects of freeze–thaw cycles and Jet A-1 permeation for decades.

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