

Saline – Sodic Resistant Bentonite in Geosynthetic Clay Liners: Potential for Coal Seam Gas Applications

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ABSTRACT

The handling, intermittent storage and disposal of saline-sodic brines, such as from coal seam gas (CSG) production, can adversely impact the hydraulic performance of clay-based barrier systems. The scarcity of suitable and economical clayey soils for traditional compacted clay liners throughout CSG localities in Australia has resulted in an increased use of geosynthetic clay liners (GCLs) to ensure environmental containment of process waters because of their technical equivalence, ready availability and ease of deployment compared to traditional clayey liners. Two new bentonite products were evaluated for their hydraulic performance to saline and saline-sodic brines for inclusion in GCLs. Standard GCL test methods applied to two Arumpo bentonite products that had been specifically beneficiated and/or modified for resistance to saline-sodic brines, were revealed to have remarkable hydraulic barrier performance. Results for fluid loss and saturated hydraulic conductivity were 2.5–5x better in seawater, 1 M and 2 M sodium chloride (NaCl), and a 20% saline-sodic brine, compared to bentonites contained in typical GCL products available in Australia. In particular, pre-hydration had minimal, but positive, impact on hydraulic conductivity. These preliminary test results indicate the suitability of these two bentonite products for inclusion in GCLs for environmental protection in coal seam gas applications. Longer term compatibility testing has been initiated based on favourable results to date.

Key Words: Coal seam gas, geosynthetic clay liners, hydraulic barriers, hydraulic conductivity, fluid loss, swell index

1 INTRODUCTION

Australian mineral and energy exploration industries produce enormous volumes of highly saline liquid wastes annually – in 2012 the gas industry contributed to the extraction of nearly 300 GL of water (Australian Bureau of Statistics, 2012). With the advent of coal seam gas (CSG) exploration and production, this value is expected to rise significantly in the near future. Due to the high saline-sodic chemistries of many CSG process waters, their above ground temporary containment poses long-term threats to Australian ground and surface waters. Regulatory authorities increasingly require that impoundments be lined with composite liner systems comprised of a geomembrane (GM) primary barrier and one or more low permeability mineral layers to minimise transport of aqueous leachates into the surrounding groundwater. The composite liner system has proven to be the most effective way of minimising leakage rates to values several orders of magnitude less than the individual components alone (Bonaparte et. al., 1989). The mineral liners must maintain a hydraulic conductivity lower than the design value for the projected contaminating lifespan of the waste containment facility. While design criteria exist, for example the Best Practice Environmental Management of EPA Victoria (BP EM 2010), for application in municipal solid waste landfills (i.e. less aggressive environment), clayey soil based liner lifetime expectancies are unknown in aggressive saline and saline-sodic environments and estimates are usually unsubstantiated by rigorous investigations (Bouazza and Gates, 2013).

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An increased use of geosynthetic clay liners (GCLs) throughout the mining industries in Australia (Hornsey et al. 2010), in part is due to the scarcity of suitable and economical clayey soils for traditional liners. GCLs are readily available, have good technical equivalence and are easy to deploy compared to traditional clayey liners (Bouazza 2002). GCLs are manufactured products composed of ~5-10 mm of processed sodium bentonite sealed between two synthetic geotextiles held together by needle-punching or stitch bonding. Bentonite is processed (e.g., sodium beneficiated) to impart favourable sealing characteristics and GCLs are usually combined with a GM to form a composite liner. While providing suitable protection against transport of low salinity waters (Kolstad et al. 2004, Jo et al. 2004, 2005), interaction of GCLs with saline leachates (generally >0.3 M) results in increased potential for hydraulic incompatibility (Guyonet et al. 2005; Lange et al. 2009), enabling leachates to contaminate underlying sediments and groundwater if they are used as single liners or where primary physical barriers (GM) have failed. One projected use of GCLs is in the environmental containment of CSG process waters as they constitute a significant threat to Australian groundwater and ecosystems, due to their saline and saline-sodic chemistries.

These issues require hydraulic barrier systems capable of functioning under geochemical conditions that are generally adverse to containment. The purpose of this study therefore, was to develop a suitably modified bentonite which could withstand excessive high saline-sodic conditions typical of CSG applications. Two new bentonite products were evaluated for their hydraulic performance to saline and saline-sodic brines and found to have remarkable fluid loss and permeability characteristics.

2 MATERIALS AND METHODS

2.1 Bentonites

Three bentonites from Australia and one from overseas were tested in their as-received form for their ability to withstand saline-sodic leachates. One Australian bentonite underwent two different non-disclosed beneficiations and modifications (ABM1 and ABM2).

2.2 Test Methods

ASTM standard methods for fluid loss (ASTM D5891), swell index (ASTM D5890) and saturated hydraulic conductivity (ASTM D5887 & ASTM D6766) were used. All tests were carried out using potentially incompatible saline leachates. Swell index tests were modified from the standard by dropping the clay powders directly into 90 mL of the saline leachate, and then topped-up with the appropriate leachate. Fluid loss tests were modified (Liu et al. 2014) from the standard in that (i) bentonites were reacted with the saline leachates and (ii) all measurements were made gravimetrically and converted to volume based on the specific gravity of the leachate (determined with pycnometers).

Saturated hydraulic conductivity tests, using flexible-walled triaxial permeameters, were performed on ABM1 in synthetic seawater (~0.6 M) and 1 M NaCl, and on ABM2 in 2 M NaCl and 20% saline-sodic brine. Some tests were conducted with the sample initially pre-hydrated with deionised water and other tests were conducted by directly hydrating the dry bentonite product with the leachate. Dry clay powder was lightly packed into the triaxial cell with carrier and cover geotextiles to mimic a GCL. All tests were conducted for at least 30 days, which resulted in only ~1-3 pore volumes of flow.

2.3 Test Leachates

Leachates tested included artificial seawater created following ASTM D1141, 0.3 M, 1 M and 2 M sodium chloride (NaCl), as well as CSG surrogate brine consisting of 20% soluble sodium salts (2:1:1 NaCl: sodium bicarbonate (NaHCO₃): sodium carbonate (Na₂CO₃)). The brine typically had pH in the range 9.1 – 9.2 and electrical conductivity ~145-155 mS/cm. All leachates were either prepared in-house (for fluid loss and swell index) or purchased as certified products (hydraulic conductivity tests).

3 RESULTS AND DISCUSSION

3.1 Swell Index and Fluid Loss

The results from reaction of four unmodified bentonite products currently used in GCLs in Australia with 20% saline-sodic brine are compared in Table 1. All samples had low swell index (SI) and high fluid loss (FL) when reacted with the 20% saline-sodic brine. The resulting cake thickness combined with the filtrate flux returned calculated (Liu et al. 2014) saturated hydraulic conductivity (k_{calc}) values of 8×10^{-10} m/s or greater, which is equivalent to a constant head of water moving 25 mm or more per year. From these results, the expected service life of un-modified bentonites used in GCLs against CSG brines is not expected to exceed one year.

Table 1: Bentonite sources and testing results on as-received bentonites with 20% saline-sodic CSG brine surrogate. k_{calc} calculated hydraulic conductivity from the FL tests.

Bentonite	Source	SI (mL/2g)	FL (mL)	Filter cake (mm)	k_{calc}^a (m/s)
CSK	Asia	4.0	103	4.0	8.2×10^{-10}
TAU	Australia	4.0	118	3.7	8.2×10^{-10}
EAU	Australia	5.0	97	5.9	1.3×10^{-9}
ESA	South Africa	5.5	106	4.3	8.7×10^{-10}

^a SI, FL, cake thickness and k_{calc} based on single measurements only.

The results of the ABM products, reacted with the same 20% saline-sodic brines, as well as with 0.3 M and 2.0 M NaCl are given in Table 2. Note that SI values for both ABM1 and ABM2 in 20% brine (Table 2) are double that of the other (un-modified) bentonites (Table 1), but that the FL values are ~2.5x and 5x less. The resulting k_{calc} values are of a similar magnitude lower as well for the ABM products in 20% saline-sodic brine. ABM1 performed exceptionally well in 0.3 M NaCl (FL=17) and ABM2 performed comparably in 2 M NaCl (FL=23). These latter solutions were trialed to determine the effect of elevated ionic strength without excessively elevated pH (2 M NaCl pH≈7.6).

Results indicate that ABM1 is somewhat limited in its ability to withstand strongly saline-sodic leachates compared to ABM2 (e.g. FL is 2x), but should perform well at lower ionic strengths, even at near neutral pH. ABM2 performs exceptionally well in strongly saline and saline-sodic leachates (compare results in Table 2 with those in Table 1).

Table 2: ABM sample results on 0.3M, 2 M NaCl and 20% saline-sodic CSG brine surrogate. k_{calc} calculated hydraulic conductivity from the FL tests.

ABM Products	Leachate	SI (mL/2g)	FL (mL)	Filter cake (mm)	k_{calc} (m/s)
ABM1	0.3 M NaCl	8	17	2.4	7.6×10^{-11}
s.d. (n=3) ^a		-	0.5	-	2×10^{-12}
ABM1	20% brine	8.5	40	4.7	3.2×10^{-10}
s.d. (n=8)		-	3	0.3	1.4×10^{-11}
ABM2	2 M NaCl	8	23	4.6	2.1×10^{-10}
s.d. (n=4)		-	1	0.6	3.7×10^{-11}
ABM2	20% brine	9	22	4.5	1.7×10^{-10}
s.d. (n=4)		-	2	0.4	2.3×10^{-11}

^a s.d. is the standard deviation.

3.2 Triaxial Saturated Hydraulic Conductivity

The results of the triaxial hydraulic conductivity ($k_{triaxial}$) tests are shown in Table 3 and also in Figures 1 and 2. Pore volumes of flow (PVF) were determined using a hydrated particle density of 1.5 g/cm^3 (Gibbs and Gates, 2014). Comparisons indicate that the $k_{triaxial}$ values are 2 to 25 x lower than corresponding k_{calc} determined from the fluid loss measurements, which confirm earlier FL tests showing it is a useful conservative compatibility test for bentonites in high ionic strength or aggressive

leachates (Gates et al. 2012; Liu et al. 2014). Note also that pre-hydration has a weakly positive effect on the k_{triaxial} values returned. This last result indicates that the ABM products are able to hydrate effectively even in high ionic strength leachates. It must be noted here that the pH and EC measurements (Table 3) were taken on the effluent at the end of the tests, so in the case of samples prehydrated with deionised water, the EC values reflect contribution of the initial pore water.

Table 3: Flexible wall triaxial hydraulic conductivity tests on ABM products in various saline and saline-sodic leachates and under conditions of pre-hydration (with deionised water) and no pre-hydration.

Product	leachate	pre-hydration	pH In/Out	EC In / Out (mS/cm)	PVF (cm ³)	k_{triaxial} (m/s)
ABM1	Seawater	none	8.1 / 8.5	59 / 54	2.87	3.1×10^{-11}
		DI water	8.1 / 8.1	59 / n.m. ^a	1.29	1.1×10^{-11}
	1 M NaCl	none	8.5 / 8.5	88 / 90	3.36	2.2×10^{-11}
		DI water	8.5 / 8.5	88 / 58	1.38	1.1×10^{-11}
ABM2	2 M NaCl	none	7.6 / 7.8	153 / 146	7.19	3.4×10^{-11}
		DI water	7.6 / 7.8	153 / 107	3.54	2.0×10^{-11}
	20% brine	none	9.4 / 9.2	101 / 99	1.33	8×10^{-12}
		DI water	9.4 / 9.3	101 / 62	0.91	5×10^{-12}

^a n.m. not measured.

For the tests in 2M NaCl (Figure 1) the k_{triaxial} values for AMB2 prehydrated in deionised water were approximately $\sim 1 \times 10^{-11}$ m/s, but by the end of the test they had increased to 2×10^{-11} ms after 3.5 pVF (Table 3), whereas the k_{triaxial} values for AMB2 contacted directly with 2 M NaCl increased from about 2×10^{-11} to 3.4×10^{-11} m/s after 7.19 PVF.

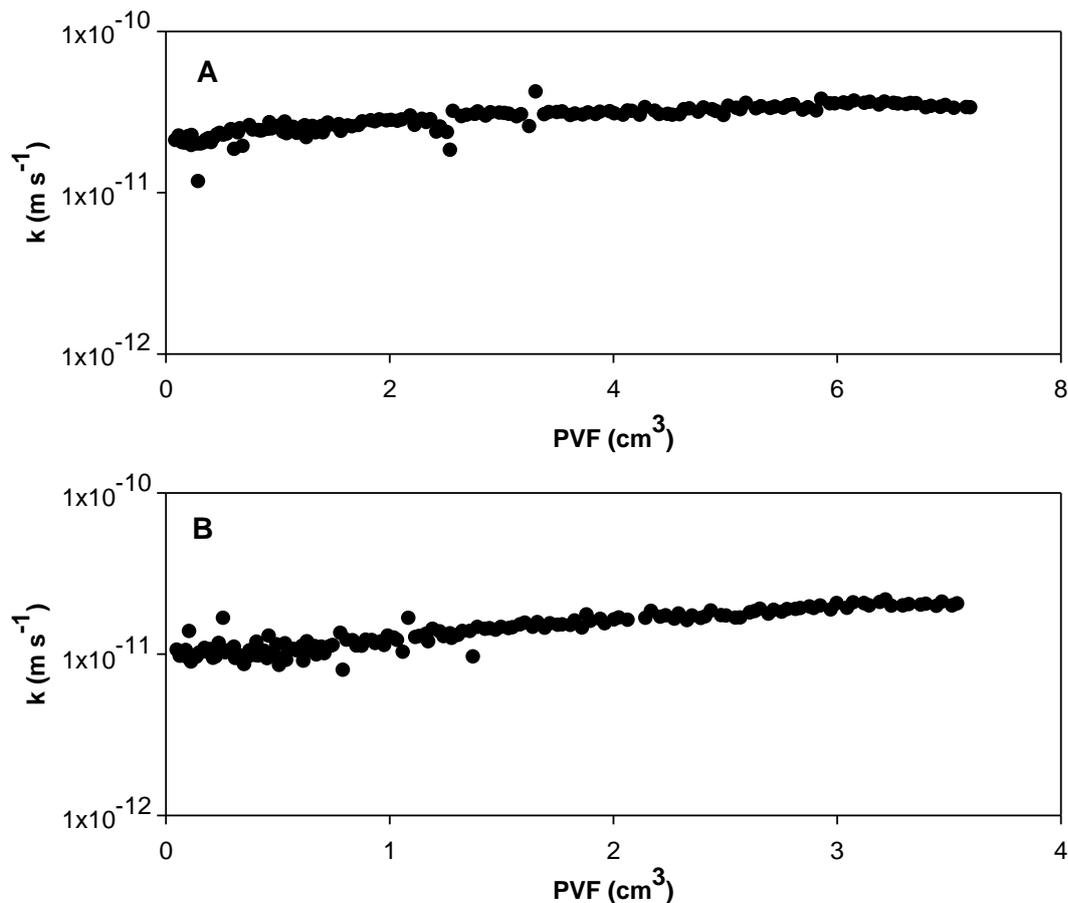


Figure 1. Saturated hydraulic conductivity (k_{triaxial}) of ABM2 in 2M NaCl; A) non pre-hydrated, B) pre-hydrated in deionised water.

The k_{Triaxial} values for ABM2 directly contacted with the 20% saline-sodic brine remained constant at about 0.8×10^{-11} m/s, and that of ABM2 prehydrated in water remained constant near 0.5×10^{-11} m/s, but only 1.33 and 0.91 PVF had penetrated these samples (Table 3, Figure 2). As can be seen in Table 3, the EC of the effluent was not yet at equilibrium with the influent for the pre-hydrated samples. However, given the dilution due to the initial pore water (deionised) present, it appears that breakthrough of the saline waters had occurred. Thus, the preliminary results indicate that both products have low k_{Triaxial} values in saline and saline-sodic leachates, at least in the short term of the study presented here.

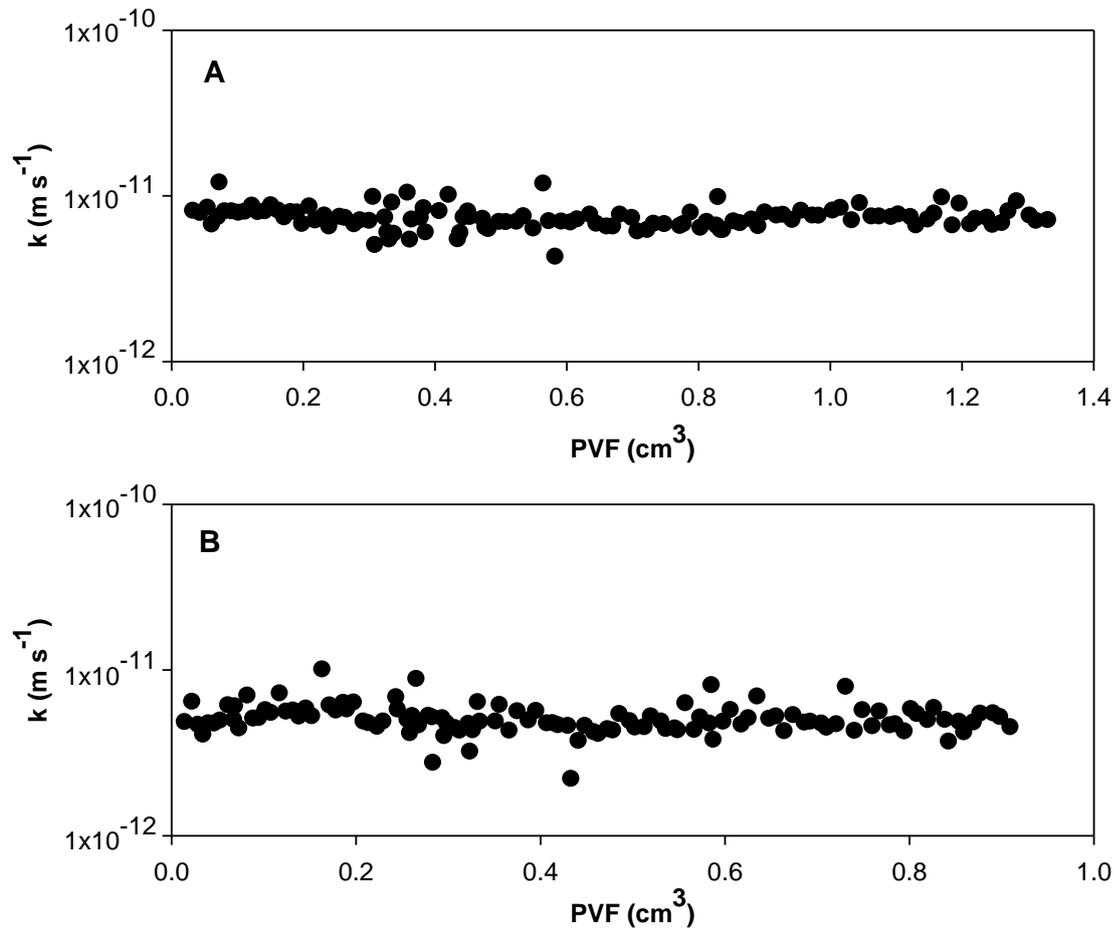


Figure 2. Saturated hydraulic conductivity (k_{Triaxial}) of ABM2 in 20% saline-sodic brine; A) non pre-hydrated, B) pre-hydrated in deionised water.

4 Conclusions

Two new Arumpo bentonite products were observed to have remarkable hydraulic performance properties to saline-sodic leachates compared to bentonites typically used in GCLs in Australia. The swell index and the fluid loss values of the ABM1 and ABM2 products, measured directly in the saline or saline-sodic leachates, respectively, are double and as much as 5x lower, than comparable results on traditional (unmodified) bentonite products. Saturated hydraulic conductivity values, calculated from fluid loss fluxes, were a similar magnitude lower for the ABM1 and ABM2 products. Pre-hydration of the ABM products in de-ionised water had a small, but positive, effect on the saturated hydraulic conductivity, as measured using flexible wall triaxial permeameters. The results indicate that the ABM products are resistant, and provide very low saturated hydraulic conductivities, to the detrimental effects of the high ionic strength of saline-sodic leachates. Further tests are required to validate these results over longer time periods and larger flow volumes and to influent-effluent chemical equilibrium.

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