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Hydraulic and Volume Change Behaviour of Biopolymer Amended Granular Bentonite for Containment Applications

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ABSTRACT

The macro-voids present in granular bentonite (GB) in Geosynthetic clay liners (GCLs) does not seals in presence of high concentration salt solutions due to reduction in the osmotic potential. The maximum allowed hydraulic permeation rate 1×10^{-9} m/sec would not be achieved under these circumstances. Further, a volumetric collapse of the barrier material induces distress to the leachate collection system and lead to the failure of the pipes. The ability of second-generation (2G) GCLs that contain polymer-amended GBs are currently explored by several researchers for containing the high ionic strength salt solutions. Guar gum is a sustainable biopolymer and has been successfully utilized for ground improvement applications. The guar gum forms a thick and viscous gel in the presence of water and salt solutions, which might help in clogging the macro-voids present in the compacted GB. The application of guar gum for improving the sealing ability of GB is not explored yet. The hydraulic and volumetric performance of guar gum-amended GB under the influence of self-weight of waste, salts present in the leachate was explored in this work. In this study, the results of hydraulic and volume change behaviour of guar gum-amended GB with high strength salt solutions and loading conditions were discussed to evaluate the ability of 2G liners to prevent the migration of the leachate to the environment. The experiment results with different percentages of the guar-gum with high concentration of three different ionic solutions were presented. The results were compared with the utilization of different synthetic polymers at present for containment of the waste having high concentration leachate.

Keywords: Granular Bentonite, Guar Gum, Geosynthetic Clay Liners, Sealing Ability, Engineered Barrier, Chemo-Mechanical Loadings.

1 INTRODUCTION

The leachate from the municipal solid waste (MSW) landfills contains high concentrations of ionic salts, heavy metals, and other emerging contaminants. Engineered barriers are used to encapsulate the harmful contaminants (Kaufhold et al., 2015; Bharat and Das, 2017; Shackelford and Sample-Lord, 2014). Bentonite based geosynthetic clay liners (GCLs) are used as barrier in landfills. Granular bentonites (GB) are preferably used in GCLs due to its workability and it is primary product during the bentonite processing (Kutlić et al., 2012; Yadav and Tadikonda 2022). GB swells upon hydration and fills the macro-voids present between the bentonite granules. The ability of breaking granules of GB into individual bentonite particles upon hydration with water is known as sealing ability of GB. The process of disintegration of GB granules takes place due to the development of diffused double layers (DDLs) around the bentonite particles, which further lead to the reduction in the hydraulic conductivity of bentonite ($< 10^{-9}$ m/s). The saturated GB containing GCL acts as hydraulic barrier due very low advective flow, and diffusion becomes the predominant contaminant transport mechanism in such cases (Bharat 2014; Shackelford et al., 2000; Das and Bharat, 2017; Yadav and Bharat, 2022b). Thus, fluid permeation rate of 10^{-9} m/s acts as limiting value for the advection flow of contaminants and it is achieved when complete sealing of the macro-voids takes place. The time require to achieve the limiting value of fluid permeation rate is known as sealing time. MSW landfill leachate contains high concentrations of inorganic cations like sodium, potassium, and calcium. The hydraulic permeation rates of bentonites increase in presence of these cations due to reduction in DDL thickness (Lee and Shackelford 2005). Moreover, the GB loses its swelling ability in presence of high concentration salt solutions, which leads

to poor sealing ability of GB in such environment (Yadav and Tadikonda 2022). The macro voids present in GB can cause the migration of heavy metals, and other harmful contaminants to pass through the GCL. In addition to the chemical loading, the GCL is subjected to the mechanical loading conditions due to the self-weight of the waste in landfills. The GCLs are subjected to the wide range of the mechanical loadings from 20 kPa in initial stage to the 400 kPa at closure of landfill. The hydraulic performance of the GCLs is more critical towards the lower mechanical loadings i.e., 20 and 50 kPa (Das and Bharat, 2021; Yadav and Tadikonda, 2022). This is attributed to the disintegration of the granules of GB into individual particles due to the application of higher mechanical loads. The hydraulic performance of polymer amended GB at low mechanical loadings are of great interest among the researchers. The hydraulic performance of GCL is more critical due initial hydration due to lower mechanical loading on the GCL. Further, application of mechanical loads due to the weight of waste helps the GB to attain the sealing ability. Further, the volume change behaviour of the GCLs is also important because any collapse of the GCL can cause the failure of the pipes in leachate collection layer and it can cause a distress in cover system of the landfills (Yadav and Tadikonda, 2022).

Second generation (2G) GCLs are explored currently for containment applications. These GCLs contains chemically modified bentonites either with organic materials or with polymers (Yang et al. 2022). Onikata et al., (1996) has developed the multi-swellable bentonite by mixing dry bentonite with propylene carbonate (PC). Sodium carboxymethylcellulose (Na-CMC) was mixed with dry bentonite to develop HYPER clay (Di Emidio et al., 2010; 2011). PC and Na-CMC intercalated the montmorillonite particle and activated the osmotic swelling of the bentonite particles. Mechanisms controlling the hydraulic performance as per recent researchers are pore clogging (Scalia et al., 2014; Tian et al., 2019), increase in swelling capacity (Onikata et al., 1996; Katsumi et al., 2008), and reduction in cation exchange (Deng et al., 2006; Ashmawy et al., 2002). However, mechanism controlling the hydraulic performance of the polymer amended bentonite is not completely understood. Moreover, the elusion of polymers and non-sealing ability of some of the polymer amended GB limits the utilization of these polymers for amendment of GB in GCLs.

Sustainable polymers such as guar-gum are commonly used nowadays to improve the shear strength and hydraulic performance of soils (Khatami & O'Kelly 2013; Moghal & Vydehi, 2021; Sujatha & Saisree, 2019; Zhang et al., 2020). There is no study available on the hydraulic and volume change performance of the guar-gum amended GB under hydro-chemo-mechanical loading conditions. Such studies are of great environmental interest as it aims to develop guar-gum amended GB as noble material for the waste containment applications. The objective of the present study is to evaluate the permeation and volume change of guar gum amended GB under hydro-chemo-mechanical loading conditions. The study evaluated the influence of guar gum content on permeation rates and volume change for different cationic pore fluids. Further, the permeation rates and volume change of guar gum amended GB were compared with synthetic polymer amended GB.

2 MATERIALS AND METHODOLOGY

2.1 Materials

The granular bentonite (GB) exhumed from commercially available GCL were utilized in this study. The index and surface properties of GB were reported from the earlier work of the authors (Yadav and Tadikonda, 2022) and produced in Table 1.

Table 1. *Index and surface properties of studied GB*

Property	Value
Specific gravity	2.78
Liquid limit	658
Plastic limit	48
Specific surface area (m^2/g)	648
Cation exchange capacity (meq/100g)	152.6
Na^+	86.1
K^+	0.8
Ca^{2+}	42.2
Mg^{2+}	23.5

Guar gum, a biopolymer, procured was supplied commercially by Urban Platter. Commercially available synthetic polymer, sodium polyacrylate, was procured from Sigma-Aldrich. Required amount of guar gum and polymer were weighed and mixed with air-dried GB, separately. The chloride salts of sodium, potassium, and calcium were precured from Spectrochem (India) with 99% purity. The required amounts of respective salts were weighed and mixed with distilled water to get 0.5 M concentrations.

2.2 Methodology

The hydraulic and volume change of the guar-gum amended GBs were studied using modified set-up as described in earlier works of the Authors (Yadav and Tadikonda, 2022; Yadav and Bharat, 2022) shown in Figure 1. Modified set-up was fabricated to accommodate 10 mm thick sample with 53 mm diameter bentonite sample and 5 mm thick porous stones were placed on either end of the compacted GB at a dry density of 1.2 Mg/m^3. A filter paper was used in between the GB and porous stones to prevent the clogging of pore of the porous stones. The setup was used to evaluate the hydraulic, sealing, and volume change behaviour of compacted GB, GBG and GBP samples under chemo-mechanical loading conditions. The test setup was placed on loading assembly and a dial-gauge was attached to measure the volume change of sample with least count of 0.002 mm. The burette with desired pore-fluid was connected to the sample and valve of burette was closed until the start of the experiment. A mechanical load of 50 kPa was applied on the sample to simulate the weight of the waste. The permeation experiment was started by opening the valve of the burette. The pore-fluid was continuously changed to maintain the head and the concentration during the experiment. The experiment was continued until the readings of the fluid permeation and volume change was found to be constant over a period of 24 hours. The required amount of dry guar gum or polymer was mixed with air-dried GB and then compacted at required density. GBG5 represents the combination of 5% guar gum with 95% GB by weight. A fluid permeation rate lower than 1×10^{-9} m/s represents the sealing of the macro-voids present in the compacted GB (Das and Bharat, 2021; Yadav and Tadikonda, 2022).

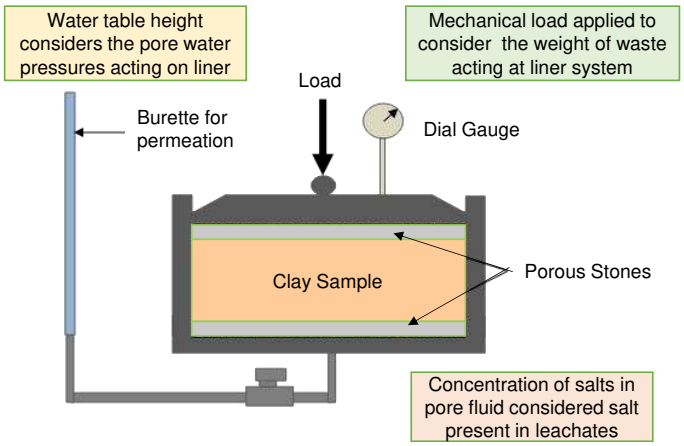

Figure 1. *Schematic diagram for permeation set-up under loading conditions.*

3 RESULTS AND DISCUSSION

3.1 Influence of percentage of guar gum

The temporal variations of fluid permeation rates of guar gum amended GB were presented in Figure 2(a) for 0.5M NaCl pore fluid under 50 kPa mechanical loading. The permeation rates were found to be higher initially for both GB and guar gum amended GB (GBG), which reduces with time upon hydration due to development of DDLs. GBG sample attains the limiting value of hydraulic permeation due to the formation of gel upon hydration of guar gum. This gel clogs the pore present between the granules of the GB. Further, hydraulic permeation started increasing and attains the permeation rate higher than liming value. Final hydraulic permeation of both GB and GBG samples were found to be in order of 10^{-8} – 10^{-9} m/s. This increase in the hydraulic permeation is probably due to the elution of guar gum in salt environment.

The temporal variations of the volume change in terms of normalized thickness for 0.5 M NaCl as pore fluid and 50 kPa mechanical loading were presented in Figure 2b. Both GB and GBG samples showed

an initial collapse due to the application of 50 kPa mechanical loading. The normalized thickness started increasing upon hydration due to the formation of DDLs. Both GB and GBG samples attained a normalized thickness of 1.08-1.13. Higher normalized thickness was observed for GB as compared to the GBG samples, due to development of higher repulsive forces among the GB as compared to the GBG samples. Swelling rate of GB sample was found to be higher than GBG samples due to lower permeation of pore fluid in GBG samples.

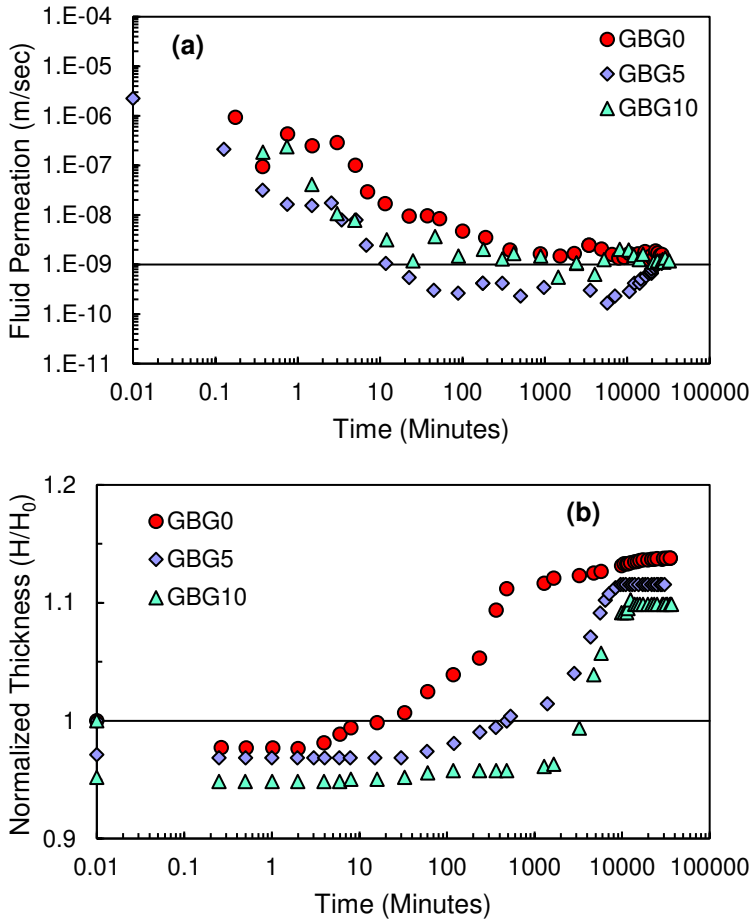

Figure 2. *Temporal variations of (a) fluid permeation; and (b) normalized thickness with guar gum amended GB using 0.5M NaCl as pore-fluid under 50 kPa mechanical loading.*

The temporal variations of hydraulic permeation rates of GB and GBG samples in 0.5M KCl environment were plotted in Figure 3(a). The hydraulic permeation rates were found to be higher initially, which decreases with time upon hydration due to development of DDLs. The equilibrium hydraulic permeation of GB sample was found to be in order of 10^{-8} m/s. The GBG samples also showed decrease in the fluid permeation and sample with 10 % guar gum content achieved the sealing after 30 minutes from the start of the experiment. Further, the permeation rate of GBG samples increased and achieved equilibrium hydraulic permeation higher than the limiting value. Both GBG samples showed equilibrium hydraulic permeation slightly higher than liming value and in order of 10^{-9} m/s. The lower hydraulic permeation of GBG samples were attributed to the formation of the gel and this gel clogs the pore present in GB. The increase in permeation rates is possibly due to the biopolymer elusion in high concentration potassium environment.

The temporal variations of the volume change in terms of the normalized thickness were plotted in Figure 3(b) under KCl environment. Both GB and GBG samples showed an initial collapse due to the application of 50 kPa mechanical loading. These samples started swelling upon hydration with pore fluid due to development of DDLs. GB sample achieved equilibrium normalized thickness of 0.99 with KCl as pore fluid. GBG5 achieved a maximum normalized thickness of 1.1 and started decreasing the after achieving a maximum value, this decrease in normalized thickness is possibly due to biopolymer elusion and its degradation in high concentration salt environment. Moreover, GBG10 sample was not able to achieve original thickness and swelling was also found to be delayed in this sample. This is attributed

to the lower permeation of fluid due to high percentage of the guar-gum. GBG10 sample achieved an equilibrium normalized thickness of 0.98.

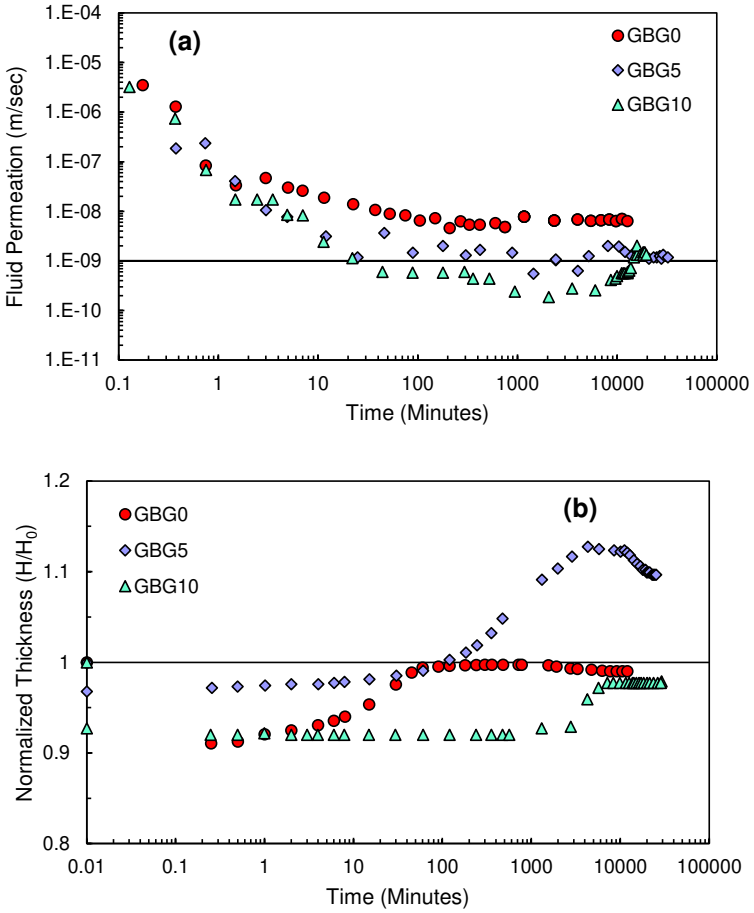

Figure 3. *Temporal variations of (a) fluid permeation; and (b) normalized thickness with guar gum amended GB using 0.5M KCl as pore-fluid under 50 kPa mechanical loading.*

Temporal variations of fluid permeation of GB and GBG samples for high concentration of $CaCl_2$ pore-fluid under 50 kPa mechanical loading were plotted in Figure 4(a). Fluid permeation was found to be higher initially for both GB and GBG samples, which decreases with time upon hydration due to formation of DDLs. GB sample achieved an equilibrium fluid permeation in order of $10^{-7} – 10^{-8}$ m/s, which is 1-2 orders higher than the limiting value of fluid permeation. Both GBG5 and GBG10 samples achieved a fluid permeation rate lower than the limiting value in 90-100 minutes from the start of the experiments. The fluid permeation of both GBG5 and GBG10 samples achieved a minimum value in order of $10^{-10} – 10^{-11}$ m/s. Further, the permeation rates of these samples started increasing and achieved an equilibrium fluid permeation in order of 10^{-10} m/s, which is lower than the limiting value of the fluid permeation. The reduction in fluid permeation rates with high concentration $CaCl_2$ salt solution is attributed to the formation of gel of guar gum upon hydration. Guar gum gel clogs the macro-voids present among the granules of the GB. Moreover, hydrated guar gum gel makes a coating over the bentonite particles, this coating of hydrated gaur gum gel entraps the cations upon hydration with the pore fluid. The reason behind the better performance of guar gum amendment in terms of fluid permeation with $CaCl_2$ environment is due to the formation of more cross-linked biopolymer structure. Due to the formation of more hydrogen bonding of calcium ion with hydroxyl groups present in guar gum as compared to the other monovalent cation like sodium and potassium.

The temporal variations of volume change in terms of normalized thickness GB and GBG samples with high concentration $CaCl_2$ pore-fluid were plotted in Figure 4(b). Both GB and GBG samples showed an initial collapse due to application of the mechanical loading. The samples started swelling upon hydration and achieved original volume within 100 minutes. GB sample achieved an equilibrium normalized thickness of 1.03. Moreover, GBG samples achieved an equilibrium thickness of 1.2 – 1.25, which is

quite higher as compared to alone GB. Higher normalized thickness in GBG samples is attributed to the formation of guar gum gel upon the hydration. Due to presence of calcium as divalent cation from the pore fluid, the repulsive forces among the gel particles were higher, which further resulted in high volume. A slight decrease in volume of GBG10 sample was observed before the equilibrium, such behaviour is observed due to biopolymer elution in GBG10 sample upon permeation with the high concentration of calcium salt solutions.

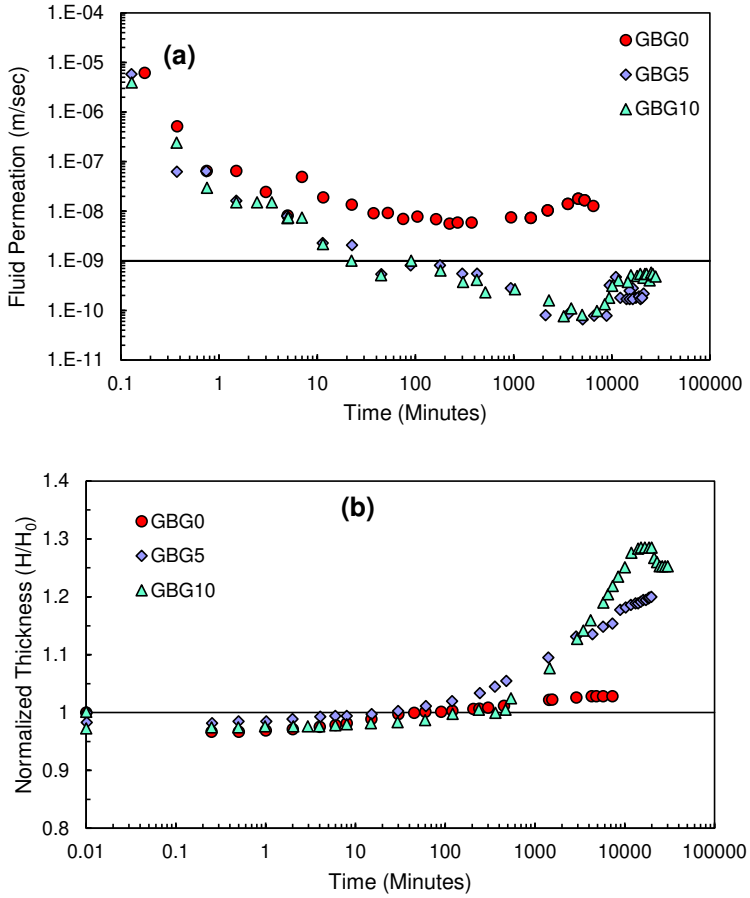

Figure 4. *Temporal variations of (a) fluid permeation; and (b) normalized thickness with guar gum amended GB using 0.5M $CaCl_2$ as pore-fluid under 50 kPa mechanical loading.*

3.2 Comparison of guar gum amended GB with synthetic polymer amended GB

The fluid permeation rates of 0.5M NaCl pore-fluid under 50 kPa mechanical loading of GBG5 samples were compared with 5 percent synthetic polymer amended GB (GBP) in Figure 5(a). Both GBG and GBP samples showed the higher fluid permeation initially, which deceases with time upon hydration with pore-fluid. The fluid permeation of GBG sample decreased rapidly and achieved the limiting value of fluid permeation in 20 minutes from start of the experiment. Further, the fluid permeation of GBG sample started increasing after 8000 minutes and achieved equilibrium fluid permeation slightly higher than the limiting value. Moreover, GBP sample showed decrease in fluid permeation with slow rate and achieved the sealing of the macro-voids in 2500 minutes from start of the experiment. The fluid permeation of GBP sample at equilibrium was found to be in order of 10^{-9} – 10^{-10} m/s. The lower fluid permeation rate of GBP sample is attributed to the formation of the hydrogel in GBP sample.

The volume change in terms of normalized thickness for GBG and GBP samples for 0.5M NaCl salt solution under 50 kPa were plotted in Figure 5(b). Both GBG and GBP samples showed an initial collapse of 1-2 % due to application of mechanical loading. Further, GBG and GBP samples started swelling upon hydration, and achieved an equilibrium normalized thickness of 1.1 and 1.2, respectively. Higher increase in the volume of GBP sample as compared to GBG sample is due to higher moisture holding capacity and formation of hydrogel in case of synthetic polymer. The rate of increase of volume

was found to be higher in case of GBP sample as compared to GBG sample is due to rapid hydration in GBP and formation of more gel in GBP.

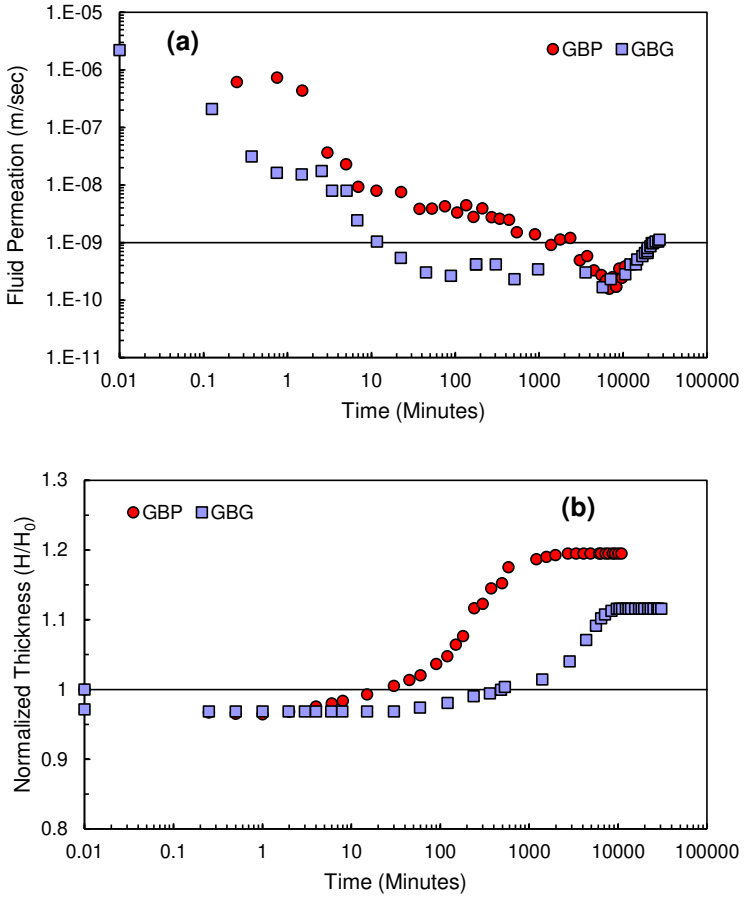

Figure 5. *Temporal variations of (a) fluid permeation; and (b) normalized thickness with GBG and GBP samples using 0.5M NaCl as pore-fluid under 50 kPa mechanical loading.*

The fluid permeation rates of 0.5M KCl salt solution as pore-fluid under 50 kPa mechanical loading for GBG and GBP samples were plotted in Figure 6(a). Both GBG and GBP samples showed higher hydraulic infiltration rates initially, which reduces further upon hydration with pore-fluid. GBG sample achieved equilibrium hydraulic permeation slightly higher than the limiting value. Moreover, GBP sample achieved an equilibrium hydraulic permeation rate in order of 10^{-7} – 10^{-8} m/s. Lower value of fluid permeation for GBG sample is due to formation of guar gum gel. Moreover, the formation of hydrogel gets limited in presence of high ionic salt environment, which results in higher fluid permeation in case of GBP sample.

The volume change in terms of normalized thickness for GBG and GBP samples for 0.5M KCl salt solution under 50 kPa were plotted in Figure 6(b). Both GBG and GBP samples showed an initial collapse of 1-2 % due to application of mechanical loading. Further, GBG and GBP samples started swelling upon hydration, and achieved an equilibrium normalized thickness of 1.1 and 1.22, respectively. GBG sample showed a peak in normalized thickness, which is attributed to the biopolymer elution in presence of high concentration salt solution. Higher increase in the volume of GBP sample as compared to GBG sample is due to higher moisture holding capacity and formation of hydrogel in case of synthetic polymer. The rate of increase of volume was found to be higher in case of GBP sample as compared to GBG sample is due to rapid hydration in GBP and formation of more gel in GBP. High fluid permeation and high swelling of GBP as compared to GBG is observed because polymer and GB are phase separated in case of GBP, which means both are swelling separately due to already existing cross-linking in the polymer. Moreover, the GBG sample has intercalated swelling mechanism, in which the linear biopolymer guar-gum makes cross-linking with the particles of the GB. Such governing mechanism leads to the higher fluid permeation as well as high volume change in case of GBP with KCl.

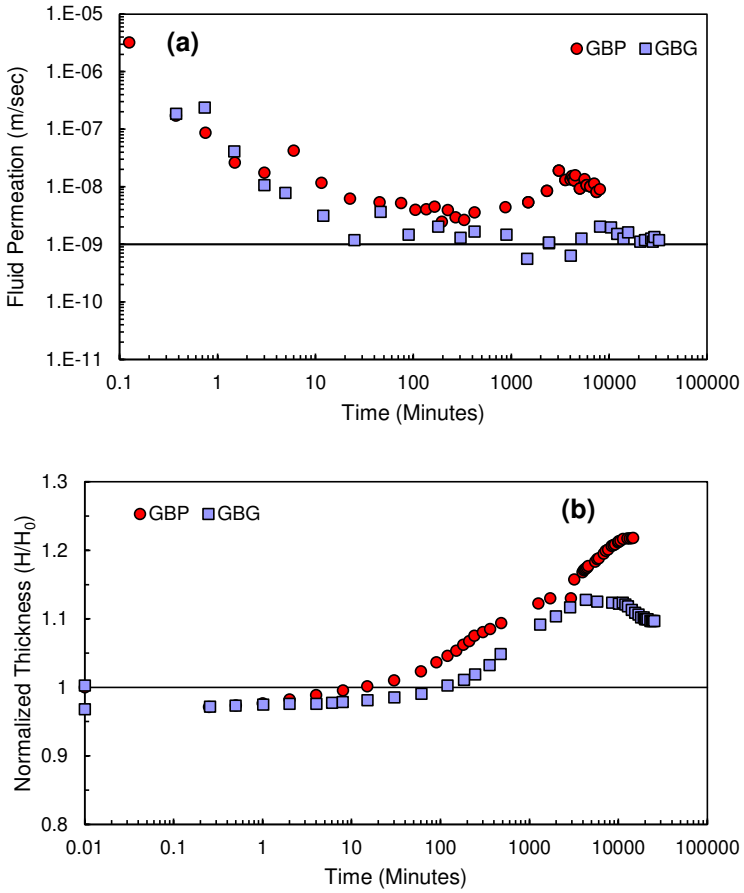

Figure 6. *Temporal variations of (a) fluid permeation; and (b) normalized thickness with GBG and GBP samples using 0.5M KCl as pore-fluid under 50 kPa mechanical loading.*

The fluid permeation rates of 0.5M $CaCl_2$ salt solution as pore-fluid under 50 kPa mechanical loading for GBG and GBP samples were plotted in Figure 7(a). Both GBG and GBP samples showed higher hydraulic infiltration rates initially, which reduces further upon hydration with pore-fluid. GBG sample achieved equilibrium hydraulic permeation lower than the limiting value and sealing time required for sealing of GBG sample was 30-40 minutes. Moreover, GBP sample could not achieve the limiting hydraulic permeation at equilibrium. It achieved an equilibrium hydraulic permeation rate in order of $10^{-7} – 10^{-8}$ m/s. Lower value of fluid permeation for GBG sample is due to formation of guar gum gel and formation of cross-linked polymer chains in GBG sample with calcium. Calcium act as a cross-linker due to its divalent nature and formation of more hydrogen bonds also resulted in lower hydraulic permeation rates of the GBG. Moreover, the formation of hydrogel gets limited in presence of high ionic calcium salt environment due to formation of complexes, which results in higher fluid permeation in case of GBP sample.

The volume change in terms of normalized thickness for GBG and GBP samples for 0.5M $CaCl_2$ salt solution under 50 kPa were plotted in Figure 7(b). Both GBG and GBP samples showed an initial collapse of 1-5 % due to application of chemo-mechanical loading. Further, GBG and GBP samples started swelling upon hydration, and achieved an equilibrium normalized thickness of 1.2 and 1.06, respectively. Higher increase in the volume of GBG sample as compared to GBP sample is due to formation of more guar gum gel. The formation of hydrogel in GBP sample was limited due to the presence of calcium salt environment. The chains of cross-linked polymer collapsed completely with divalent cation. Moreover, presence of Calcium cation enhanced the cross-linking capacity of the linear biopolymer because Ca ion can act as a cross-linker in such case. Due to these reasons, the fluid permeation of GBP is higher and volume change is lower as compared to GBG in case of calcium solution.

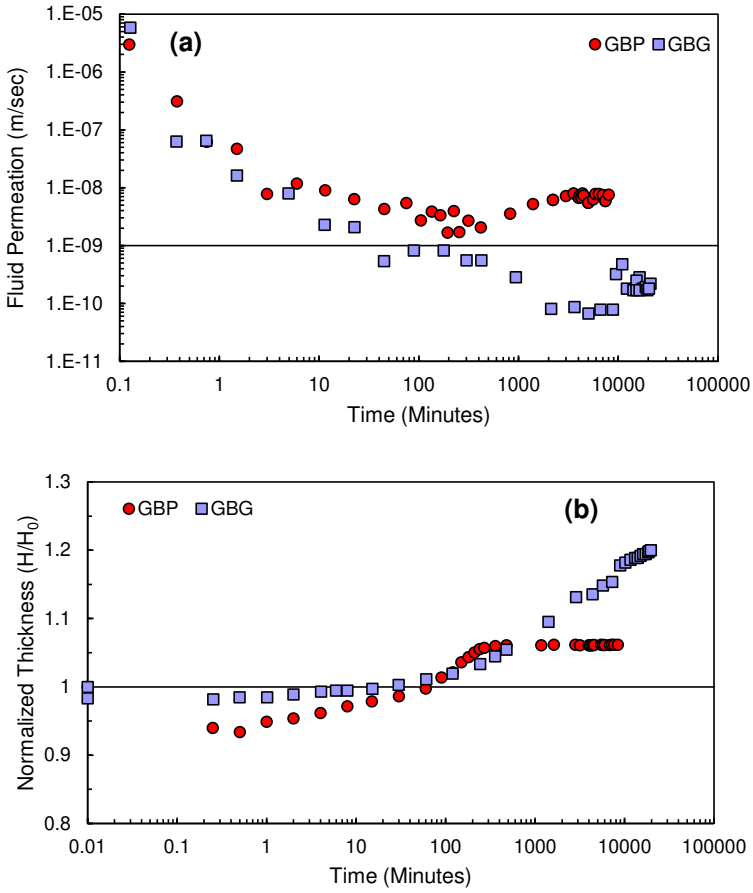

Figure 7. *Temporal variations of (a) fluid permeation; and (b) normalized thickness with GBG and GBP using 0.5M $CaCl_2$ as pore-fluid under 50 kPa mechanical loading.*

4 CONCLUSIONS

The fluid permeation and volume change GBG and GBP were conducted under hydro-chemo-mechanical loading conditions and following conclusions were drawn from the study:

- Guar gum amended GB fails to attain the sealing with high concentration of NaCl and KCl salt environment. Moreover, the fluid permeation rates were found to be lower than the GB alone, which is due to the formation of viscous guar gum gel which clogs the pores between granules of GB, However, GBG samples achieved the sealing with high concentration Ca2+ as pore fluid, which is due to cross-linking of guar gum chains with the help of divalent calcium as cross-linker.
- Guar gum is a linear biopolymer and it forms a gel structure around the GB granules, whereas, sodium polyacrylate is a cross-linked polymer and forms a phase separated matrix with GB.
- GBP samples achieved sealing in case of sodium salt solution but fails to achieve sealing with potassium and calcium salt solution.
- GBP showed higher swelling as well as higher hydraulic conductivity with Ca^{2+} and K^{+} pore-fluids, which contradicts the conclusion from earlier researcher that increase in swelling capacity governs the hydraulic performance of polymer amended GBs.
- GBG could seal under the influence of Ca and GBP sealed under the influence of K. A combination of P&G might work for sealing all the cations, which will be explored by the authors.

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