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Impact of anionic and cationic heavy metals on the geotechnical characteristics of the bentonite-based liner

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ABSTRACT

Leachates from landfills have higher quantities of anionic and cationic heavy metals, which pose a serious threat to ecosystems. These toxic heavy metal ions can negatively impact the long-term performance of landfill liners and enable the hazardous leachate contents to seep into the groundwater and soil. In this study, the effect of a cationic heavy metal, Zn^{2+} and anionic heavy metal, Chromium (VI), on the geotechnical characteristics of landfill liners are examined. Due to their peculiar properties, such as low hydraulic conductivity, high chemical and mechanical stability and better pollutant retention capacity, bentonite-based liners are frequently employed for landfill barrier applications. Therefore, the effect of heavy metals on consistency limits, swelling, strength and hydraulic conductivity of bentonite-based liners is covered in this work. Atterberg limits, free swelling, unconfined compressive strength and consolidation tests were performed on concentrations of hexavalent chromium ions and Zinc ions ranging from 0 to 1000 ppm. The findings showed that the performance of bentonite liners is adversely affected by the presence of both cationic and anionic heavy metals. The swelling capacity, liquid limit and UCS of bentonite considerably decreased, whereas hydraulic conductivity increased as the Zn^{2+} and Cr(VI) concentrations rose from 0 to 1000 ppm. However, the effect of cationic heavy metal on these geotechnical properties was observed to be higher than that of anionic heavy metal ions. These alterations in geotechnical parameters might be due to the change in soil structure and diffused double-layer thickness of bentonite.

Keywords: Hexavalent Chromium; heavy metal; barrier; landfills; swelling

1 INTRODUCTION

Bentonite-based liners have been utilized as landfill covers and liners for waste containment facilities (Mishra et al., 2011). The primary reason for the use of bentonite as a hydraulic barrier is its capacity to swell when exposed to water and to display strong sealing abilities. However, the hydraulic barrier effectiveness is adversely affected when these liners come into contact with dangerous leachate components, which are produced when rainfall percolates through waste materials. Moreover, the concentration of toxic chemicals, including heavy metals and organic pollutants in leachates, has been rising dramatically in recent times as a result of various anthropogenic activities. Ecosystems may be seriously threatened if the toxic substances from leachates permeate through the liner components and contaminate nearby soil and groundwater.

A major environmental and human health concern today is heavy metal contamination, which results from hazardous and municipal waste landfills. Abandoned mines have also become a major source of high-level toxic heavy metal emissions. Recent research revealed that acid mine drainages contain significant quantities of lead, copper, Zinc, cadmium, aluminium, iron, manganese, chromium, and selenium (Liao et al., 2017; Zhang et al., 2021). Zinc is a frequent heavy metal pollutant found in leachates and mine wastes, but it is an essential dietary mineral required for significant functions throughout the body. However, the presence of a higher level of Zinc beyond the permissible limit may cause cellular and tissue damage, respiratory problems and gastrointestinal issues. In addition to human health, it can also impact aquatic life and bacterial diversity in soil (Ray et al., 2020). Chromium is one of the most hazardous heavy metals found in industrial leachates, even in trace amounts. It can exist in the environment in either of the two stable oxidation states - Cr(III) or Cr(VI). Industries, including tanneries, metallurgical, electroplating, and chromium slag landfills, can be a significant source of hexavalent chromium (Cr(VI)) contamination (Foldi et al., 2013). In contrast to other heavy metals that can only exist in cationic states in aqueous solutions, the extremely toxic state of chromium, Cr(VI), may also exist in anionic complexes such as CrO_2^{2-}, $HCrO_4$- and $Cr_2O_7^{2-}$. Therefore, to control the serious threat that these contaminants may generate to nearby soil and groundwater, a thorough study is needed to understand the behaviour of liners when in contact with these toxic and dangerous cationic and anionic heavy metals. In addition to being harmful to ecology and human health, heavy metals can also modify the geotechnical characteristics of soils, making it challenging to reconstruct geotechnical structures in specific locations because the parameters of the polluted soil are unknown.

Numerous research teams have looked into how various cationic heavy metals, including lead, copper, Zinc, and cadmium, affect the geotechnical characteristics of different types of soils. Du et al. (2015) explored the impact of lead contamination on a mixture of calcium bentonite and clay for cut-off wall applications. The chemical compatibility of the mixture was thoroughly studied in view of liquid limit, hydraulic conductivity and compressibility at various concentrations of lead ions. The results displayed that the addition of lead ions alters the properties of the parent soil. There was a decrease in the consistency limits, specific surface area and a rise in specific gravity with an increase in lead concentrations. Du et al. (2014) investigated the engineering properties and microstructural variation on a cement-treated kaolin soil contaminated with Zinc. They found that the contamination by Zinc affected the hydration and pozzolanic reaction induced by the cement and varied the properties of cement-treated kaolin. The undrained shear strength and elastic modulus were observed to be declined with an increase in heavy metal contamination. A detailed experimental analysis was conducted by Souli et al. (2008) on the variation of permeability properties and microstructural changes of clayey soil contaminated with various concentrations of zinc solution. At higher concentrations of Zinc (1M), the soil particles get flocculated and increase the hydraulic conductivity of the soil. Li et al. (2015) examined the consequence of lead contamination on the mechanical and microstructural variation of clay. They concluded that the nature of pore fluid changed the microstructural properties and varied the mechanical properties of clay. The results displayed a rise in hydraulic conductivity and a decline in the shear strength, liquid limit and swelling of clay with a surge in lead contamination. The adsorption behaviour of Cr(VI) contaminated soil was studied by Wang and Zhang (2021). They observed an increase in heavy metal adsorption rate at higher acidic conditions due to the protonation of the clay surface. Yang et al. (2021) did a detailed study on the permeability of various combinations of sand and SHMP-treated calcium bentonite. They found an increase in permeability in the presence of hexavalent chromium contamination.

From the literature, it was noted that the majority of the investigation was conducted with cationic heavy metals. Only a few studies were conducted on the characteristics of bentonites contaminated with anionic heavy metals. The comparison between the engineering behaviour of bentonites under the influence of various types of heavy metals was also not well explored. This study envisages the geotechnical behaviour of bentonite spiked with different concentrations of a cationic heavy metal, Zinc and an anionic heavy metal, Cr(VI). It also compares the geotechnical performance of bentonite under the effect of anionic and cationic heavy metals.

2 MATERIALS AND METHODOLOGY

2.1 Bentonite

Indian bentonite acquired from Rajasthan, a state in India with the properties displayed in Table 1, was chosen for the experimental investigation. The bentonite was in powdered form, and the experiments were conducted on the soil without any prior treatment. The basic geotechnical parameters of the bentonite were determined using American Standard code procedures (ASTM) and are listed in Table 1. The compaction properties of bentonite, such as optimum water content and maximum dry density, were obtained by following the Standard Proctor procedure based on ASTM D698 (2012). The specific surface area and cation exchange capacity of bentonite were determined using the method explained by Cerato and Luteneggar (2002) and the ammonium acetate method, respectively. The specific gravity of bentonite was calculated by using a density bottle method based on ASTM D 854-92 (1994). The pH of the bentonite at a liquid-to-solid ratio (L/S) of 20 was measured using a digital pH meter (Systronics, India).

***Table 1**: Properties of bentonite*

Properties of bentonite	Values	Unit
Specific surface area	340.4	m^2/g
Cation exchange capacity	36.2	meq/100g
Plasticity index	272	-
Specific gravity	2.68	-
pH (L/S = 20)	9.04	-
Maximum dry density	13.34	kN/m^3
Optimum water content	35	%

2.2 Heavy metals

A cationic heavy metal, Zinc and an anionic heavy metal, Cr(VI), were chosen for the present study. These pollutants were selected because they are frequently present in municipal solid waste, hazardous landfill leachates, and mine tailings (Banchhor et al., 2020). The stock solution for Zinc and chromium was prepared by using Zinc nitrate and potassium dichromate powder. Stock solutions of 1000 mg/L Zn^{2+} and Cr(VI) were prepared by dissolving the calculated amount of zinc nitrate and potassium dichromate in 1000 mL of deionized water (DI water). Later, a sufficient amount of stock solutions were used to prepare 100mg/L Zinc and Cr(VI) concentrations. The nitrate of Zinc was selected because nitrate's effect on soil geotechnical characteristics was found to be negligible (Fan et al., 2013; Du et al., 2014; Fu et al., 2021). The pH of 100mg/L and 1000mg/L of Zn^{2+} was noted as 6.64 and 6.04, respectively. And for the same concentrations of Cr(VI) ions, the pH value was obtained as 4.71 and 4.01, respectively. Multiple experiments were performed on bentonite samples mixed with DI water (0 mg/L), 100 mg/L and 1000 mg/L of Zinc and Cr(VI). A maximum heavy metal concentration of 1000 mg/L was chosen in reference to the previous works conducted by Ray et al. (2019) and Banchhor et al. (2020).

2.3 Methodology

For the determination of geotechnical parameters, the powdered bentonite samples were premixed with predetermined quantities of corresponding solutions and kept in a desiccator for at least 24hrs to attain moisture equilibrium. The Atterberg limits and free swelling of bentonite were determined using ASTM D5890 (2006) and ASTM D4318 (2010), respectively. The liquid limit of bentonite samples was found using Casagrande's apparatus since it was the suggested method for clayey soils. The undrained shear strength was obtained by performing the unconfined compressive strength (UCS) test based on ASTM D2166 (2016). To simulate an undrained situation, all UCS tests were run at a 1.25 mm/min strain rate. The measurements of stress and strain were continuously recorded at regular intervals until the sample breaks. The hydraulic conductivities of samples were calculated by an indirect method by performing a series of oedometer tests (ASTM D2435 (1996)). Previous studies displayed comparable hydraulic conductivity values using oedometer experiments in comparison with other direct permeability measurement techniques (Sivapullaiah et al., 2000; Ray et al., 2021; Zhang, 2022). The samples for oedometer experiments were prepared at OMC and MDD corresponding to DI water and stored in a desiccator for one day. The samples were mounted on a consolidation setup and then permeated with respective solutions. Hydraulic conductivity is an essential parameter in selecting appropriate materials for landfill liners and covers. According to US environmental protection agency, a material should require a minimum hydraulic conductivity of 1×10^{-9} m/sec (USEPA, 1988). The detailed procedure for the experimental programme was discussed by Ray et al. (2021). The hydraulic conductivity (k) was calculated on the basis of the following formula proposed by Terzaghi (1943):

$$k = c_v m_v \gamma_w$$

Where, c_v is the consolidation coefficient, m_v is the coefficient of volume compressibility, and γ_w is the unit weight of pore fluid (water).

3 RESULTS AND DISCUSSION

3.1 Consistency limits

The Atterberg limits or consistency limits offer a fundamental understanding of the engineering properties of fine-grained soil. Liquid limits and plastic limits of bentonite under various concentrations of heavy metals were determined using American standard code procedure and are shown in Figure 1. Figure 1 demonstrates how the liquid limit of bentonite rapidly decreases when the concentrations of both cationic and anionic heavy metals increase. The reduction in liquid limit was more prominent for soil contaminated with cationic heavy metal, Zinc. The combined effect of potassium ions and chromate ions may be responsible for the fluctuation in liquid limit in potassium dichromate solutions. Previous researchers demonstrated that there is a significant variation in the geotechnical properties of bentonite when permeated with potassium ions (Quirk and Schofield,1955; Sridharan et al., 1986). This could be due to the drop in diffused double-layer thickness corresponding to the smaller hydrated size of potassium ions. Thus, the contribution of dichromate ions on the variation of liquid limit could be minimal when compared to cationic heavy metal ions.

When the heavy metal concentration increased from 0 to 100 mg/L, a percentage reduction of 14.51% and 12.58% were observed for Zinc and Cr(VI), respectively. And at 1000 mg/L concentrations, the percentage reduction for Zinc and Cr(VI) was found to be 25.81% and 25.48%, respectively. The tendency of bentonite to adsorb divalent Zn^{2+} ions from zinc nitrate solutions and monovalent K^{+} ions from potassium dichromate solutions may be the cause of the decrease in liquid limit. Thus, the consequent weakening in the diffused double layer thickness may be the cause for the reduction in liquid limit with an increase in heavy metal solutions. The decline in interparticle repulsion resulted in faster particle movement even at lower water contents or interparticle distances. Similar results were found for bentonite-based liners in previous literature as well (Du et al., 2015; Ray et al., 2020; Nasab and Keykha, 2021).

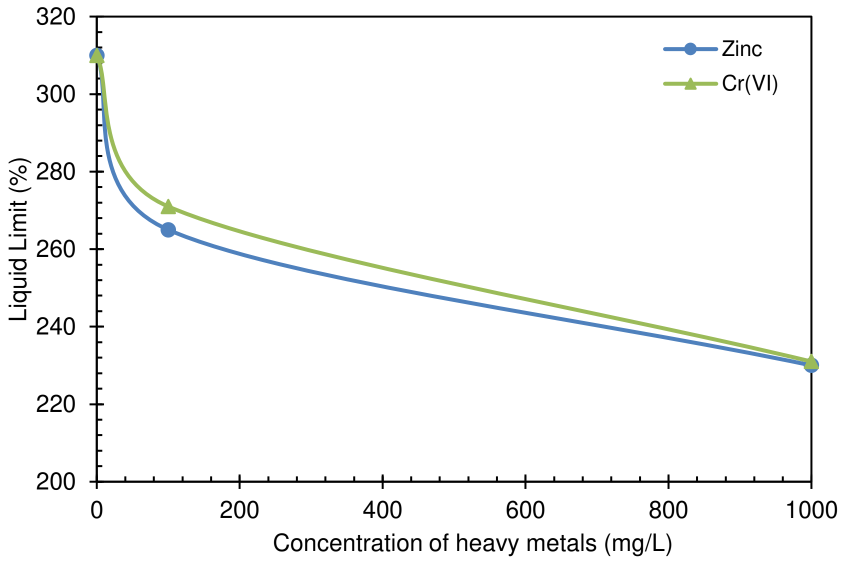

Figure 1. *The liquid limit variation of bentonite with a rise in heavy metal concentrations*

3.2 Free swelling properties

The free swelling shows the increase in bentonite volume brought on by water absorption when there is no external overburden. The swelling behaviour of bentonite depends on several factors, such as the type of bentonite, the nature of permeant solution and the concentration of soluble ions in permeant solution.

The impact of different concentrations of Zinc and hexavalent chromium ions on the free swelling characteristics of bentonite is displayed in Figure 2. The observed trend was similar to that of the liquid limit variation shown in Figure 1. This is because the index properties are one of the controlling factors that affect the swelling behaviour of soil. As the concentration increased from 0 to 1000 mg/L, the free swelling of bentonite reduced from 36 mL/2g to 9 mL/2g and 13 mL/2g for Zinc and Cr(VI), respectively. When the cation concentration in the permeant solution increases, there exists a free energy gradient which could expel the water from the interlayer region (Mishra et al., 2005). Thus, with an increase in cationic and anionic heavy metal solutions, a significant reduction in free swelling was observed. And the reduction was more significant at higher concentrations (1000 mg/L) than that at lower concentrations (100 mg/L). In the case of zinc nitrate solution, the effect of divalent Zn^{2+} ions were much more significant in reducing the diffused double layer thickness of bentonite and hence displayed a substantial reduction in the free swelling index of bentonite. In contrast, in anionic heavy metal solutions, the monovalent K^{+} ions in potassium dichromate solution also affected the alteration in diffused double layer thickness, which could lead to the reduction in free swelling value. Since the monovalent ions significantly reduce the diffused double-layer thickness, the reduction of free swelling in Cr(VI) solutions could be attributed to the cationic K^{+} ions rather than their anionic counterparts. Hence it can be concluded that the effect of cationic heavy metals on the free swelling of bentonite was much higher when compared to anionic heavy metals.

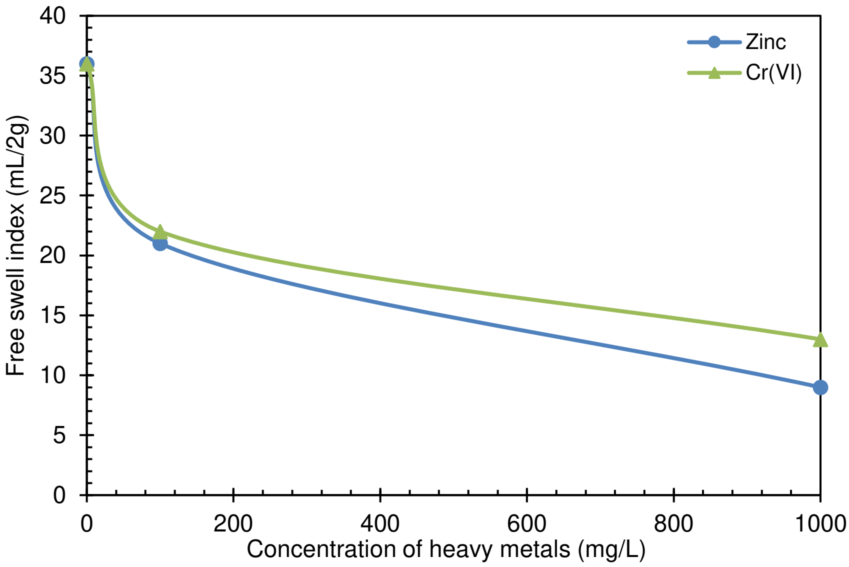

Figure 2. *Free swell index of bentonite in the presence of DI water, cationic and anionic heavy metals*

3.3 Strength properties

A minimum shear strength requirement of 200 kPa must be met by a material before it can be utilized as a hydraulic barrier in order to prevent failure brought on by slope sliding and overburden pressure (Daniel and Wu, 1993; Gahlot et al., 2022). In this context, a sequence of unconfined compressive strength experiments was conducted on bentonite samples mixed with different permeant solutions (Zinc and Cr(VI)) at different concentrations (0, 100 and 1000 mg/L) to obtain the undrained shear strength of samples. The undrained shear strength of bentonite decreased with an increase in anionic and cationic heavy metal ions. The reduction in the strength could be attributed to the alteration in diffused double-layer thickness and particle rearrangement due to the presence of heavy metal ions (Zha et al., 2021). The results from Figure 3 showed that comparatively higher shear strength was observed for cationic heavy metals when compared to anionic heavy metals. A percentage reduction of 9.38% and 9.97% was noted for 1000 mg/L zinc and Cr(VI) contaminated bentonite, respectively. At lower concentrations (0 to 100 mg/L), the reduction in UCS value was comparatively lesser when compared to that with DI water. When the heavy metal concentrations increase, the stress-strain characteristics of the bentonite get shifted from brittle behaviour to ductile behaviour. This will change the particle arrangement, which could lead to strength reduction at greater concentrations. Similar strength behaviour was observed for a Zinc contaminated kaolin soil conducted by Du et al. (2014).

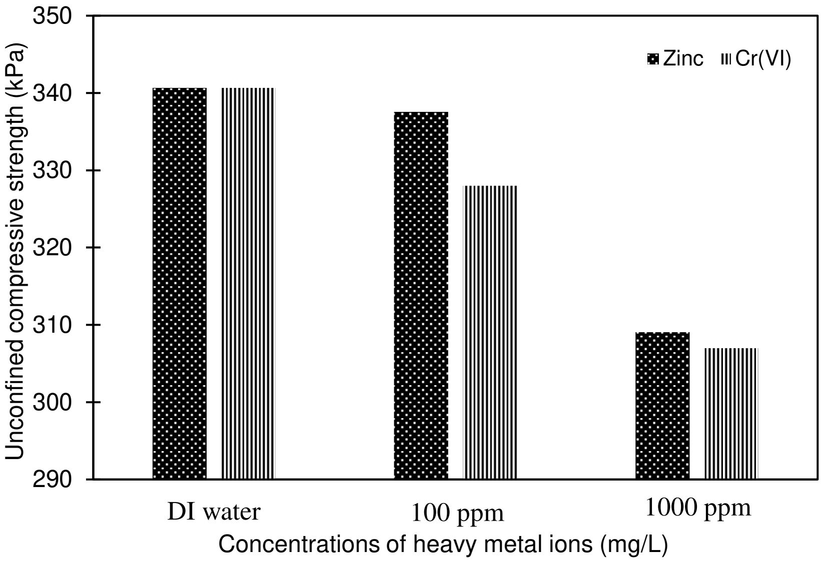

Figure 3. *The unconfined compressive strength of bentonite contaminated with cationic and anionic heavy metal solutions*

3.4 Hydraulic properties

The characteristics of permeant solutions and bentonite soil have a major impact on hydraulic conductivity, which is a crucial metric in determining a material's hydraulic barrier potential (Lambe, 1955). The measurement of the hydraulic conductivity of bentonite using direct methods may take several months to complete the experiment. Hence, in the present study, an indirect method using conventional oedometer apparatus was adopted. This approach has been shown to produce results that are comparable to those obtained using other direct measurements (Sivapullaiah et al., 2000; Zhang, 2022). The experiments were conducted on bentonite samples inundated with various solutions, and the results were used to monitor the minimum hydraulic conductivity requirement of 10^{-7} cm/sec for a hydraulic barrier.

Figures 4 and 5 depict the variation of the coefficient of hydraulic conductivity with the void ratio under the effect of different inundating heavy metal fluids. It was understood that the impact of Zinc on hydraulic conductivity was much higher when compared to Cr(VI) solutions. When the soil gets consolidated, the particles come closer and attain a lesser void ratio. As the void ratio reduces, the coefficient of permeability also reduces. The addition of high concentration of heavy metal solutions increased the hydraulic conductivity of bentonite. However, no samples exceeded the limiting hydraulic conductivity value of 10^{-7} cm/sec. The presence of divalent Zn^{2+} and monovalent K^{+} ions affected the thickness of the diffused double layer of bentonite. At 1000 mg/L, a considerable reduction in diffused double-layer thickness was observed, and particles tried to flocculate, which could lead to the formation of flow paths and, thus, an increase in hydraulic conductivity. In the case of potassium dichromate solution, there will be a competitive effect between cationic K^{+} ions and anionic dichromate ions. The combined effect of these might lead to the lesser influence of potassium dichromate solution on the hydraulic conductivity of bentonite. This study suggests that if the liner is exposed to heavy metals from leachates for an extended period of time, it may seriously impair the swelling and hydraulic capabilities of bentonite.

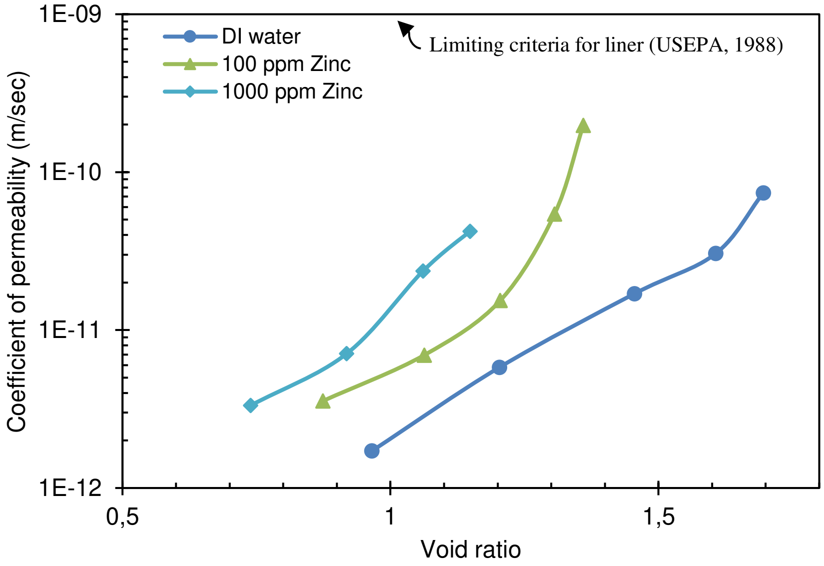

Figure 4. *The coefficient of permeability versus void ratio for bentonite inundated with different concentrations of Zn^{2+} ions*

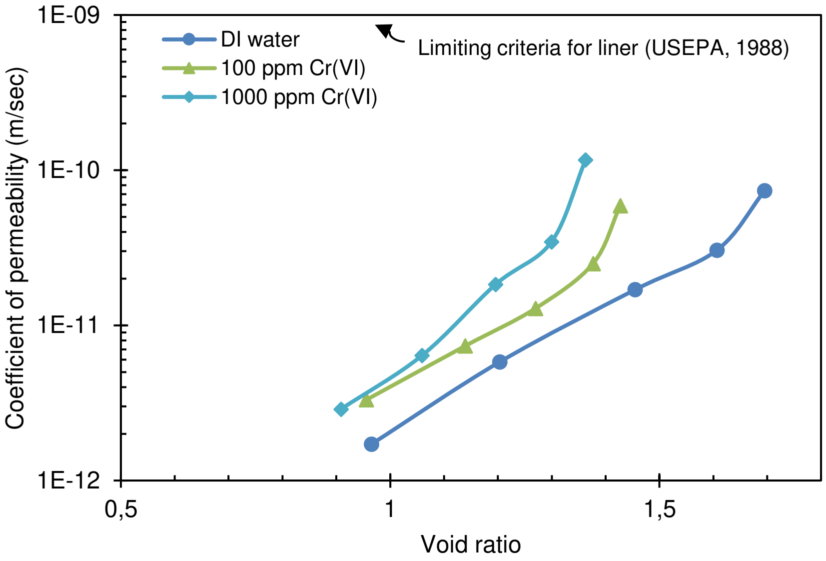

Figure 5. *The coefficient of permeability versus void ratio for bentonite inundated with different concentrations of Cr(VI) ions*

4 CONCLUSIONS

A set of laboratory experiments were performed to understand the geotechnical behaviour of bentonite under the influence of cationic (Zn^{2+}) and anionic (Cr(VI)) heavy metals. The outcomes demonstrated that the contact of bentonite with heavy metal ions significantly changes the geotechnical properties of bentonite, especially at higher concentrations. Reduction in the free swelling, liquid limit and undrained shear strength of bentonite was observed with the addition of heavy metal solutions. The permeability of the bentonite liner was also increased with a rise in heavy metal ion concentrations. The alteration in diffused double-layer thickness and the variation in microstructural properties of bentonite could be the reason for the changes in geotechnical properties. Hence, the outcomes of the study concluded that the hydraulic barrier performance of bentonite liners get adversely affected by the presence of both cationic and anionic heavy metals.

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