The Effect of Saline Fluid on Hydraulic Properties of Clays

Koteswaraarao Jadda and Ramakrishna Bag

Abstract The effect of pore fluid concentration on the hydraulic conductivity of barrier material is one of the key factors which are considered in the long-term performance of a geological repository. The current study presents the effect of various NaCl concentrations on Atterberg limits and hydraulic properties of two bentonites and one kaolin clay. The results indicated that the liquid limit, shrinkage limit, and consolidation characteristics of bentonites such as compression index (C_C) and the time required for 90% of consolidation (t_{90}) were decreased significantly with an increase in salt concentrations. Similarly, the hydraulic conductivity and coefficient of consolidation (C_v) increased drastically with increase in salt concentrations; however, the significant impact was found for the high smectite bentonite. The experimental results showed that the hydraulic conductivity of the clays mainly depends on the particle arrangement rather than percentage of clay fraction and consolidation stress. For kaolin soil, both the liquid limit and shrinkage limit were found to be slightly increased up to 0.5 M NaCl, however, the effect was found to be decreased at further increase in concentrations, whereas hydraulic conductivity was found to be increased with increase in NaCl concentration. Further, the effect of molding water content on the consolidation characteristics of the clays was also investigated. The parameter compression ratio was used to evaluate the impact of initial moisture content on the compression index of the soil specimens. The hydraulic conductivity of the clays was noted to be increased at higher pore fluid concentration, due to the diminishing of the diffused double-layer thickness of clay minerals.

Keywords Bentonite · Kaolin · Compression index · Hydraulic conductivity · Saline fluid

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1 Introduction

Compacted clays have been used as potential buffer/backfill materials in most of the toxic waste disposal repositories due to their inherent properties of low permeability, high swelling potential, self-healing capacity, and good durability under disposal environments. During the long-term operation of a deep geological repository, the contaminants present in the buffer materials or infiltrated pore fluids from the surrounding geological strata may influence the hydromechanical properties of the compacted clays resulting in decreasing the efficiency of the buffer materials. Among all the hydromechanical properties of buffer materials, the hydraulic conductivity is the most important parameter involved in the assessment of contaminant migration in the subsurface and the design of barriers for hazardous waste control. For effective insulation, the barrier material should fulfill some desired specifications such as saturated hydraulic conductivity which should be in the range of 10^{-8}–10^{-10} m/s and the swelling pressure should be more than 1 MPa [15, 16].

The effect of various salt concentrations on the liquid limit and hydraulic conductivity of different soil–bentonite mixtures was studied by Mishra et al. [9]. The hydraulic conductivity of GMZ bentonites by using different electrolyte concentrations of NaCl and $CaCl_2$ was studied by Zhu et al. [23]. Similarly, Singh and Prasad [14] reported the effect of ferric chloride and humic acid on the various engineering properties of barrier materials such as differential free swell index, swelling pressure, and hydraulic conductivity of the clays. Ismeik et al. [6] investigated the effect of seawater concentration on the various geotechnical properties of fine-grained soils. Dutta and Mishra [4] reported the effect of heavy metals concentration on the consolidation characteristic of compacted bentonites. The study suggested that C_C, m_v, and t_{90} of bentonites decreased with increases in salt concertation. Tiwari and Ajmera [21] evaluated the effect of saline fluid on compressibility characteristic of various clay minerals. The study suggested that the effect of NaCl solution was significant in montmorillonite clays as compared to illite and kaolinite clays. Sridharan et al. [19] reported that the increases in the pore fluid concentration cause a decrease in free swell index capacity for the smectite-type clay whereas it was found to be increased for kaolinite-type clays. The temperature effect on the swelling pressure and consolidation characteristics of the soils were studied by Bag and Rabbani [1]. Ören and Akar [11] investigated the effect of landfill leachates on the swelling and hydraulic conductivity of bentonites. The study reported a decrease in the free swell capacity of bentonites with increasing leachates concentration whereas the hydraulic conductivities of bentonites was not affected by the leachates concentration. Thyagaraj and Rao [20] and Chen et al. [3] examined the effect of salinization and desalinization cycles on the osmotic swelling and consolidation characteristic of compacted clays. Siddiqua et al. [13] investigated the effect of pore fluid chemistry on the various geotechnical properties of the barrier materials such as hydraulic conductivity and swelling pressure and stiffness. The study found that hydraulic conductivity increases with electrolyte concentration whereas it decreased with effective montmorillonite dry density. Fan et al. [5] investigated the consolidation characteristics of the sand/clay–bentonite used as backfill materials. The study presented that the effect of sand grain size does not show any significant effect on the compression index and hydraulic conductivity. Komine [8] proposed theoretical equations to evaluate the hydraulic conductivities of buffer and backfilling materials.

Several studies focused on the effect of pore water chemistry on the stiffness and hydraulic conductivity of the clays, however, the impact on the various clay minerals is not very clear. In the current study, the effect of saline fluid concentration on the hydraulic properties of three clays, having different mineralogical characteristics such as Na–bentonite (high smectite content), Ca–bentonite (medium smectite content), and kaolin clay were investigated.

2 Materials and Methodology

2.1 Materials

Commercially available two types of bentonites and one kaolin clay were used in the present study. All the soils were procured from Rajasthan, India. For further discussion, these two bentonites are referred to as bentonite-A and bentonite-B. The basic engineering properties of the clays such as specific gravity, Atterberg limits, and particle size distribution were determined by following Indian Standard (IS) Soil Classification System IS 2720 (Part 3, 1980), IS 2720 (Part 4, 1985), and IS 2720 (Part 5, 1985), respectively. For all the soils used for this study, the percentage of clay fraction was determined from hydrometer analysis. The grain size distribution of the soils is presented in Fig. 1. Here, bentonite-A, bentonite-B, and kaolin have 76%, 82%, and 33% of clay fractions, respectively.

The total specific surface area (SSA) of the soil was determined by using the EGME method [2]. The total cation exchange capacity of the bentonites was determined by using the ammonium acetate method [12]. Cation exchange results suggested that bentonite-A was dominated by sodium cations whereas bentonite-B contains more divalent cations. Therefore, bentonite-A is referred to as Na-type bentonite whereas bentonite-B is classified as Ca-bentonite. The smectite content of the clay was determined as per ASTM C837-09 (2009). The physical properties of the materials used in the current study are listed in Table 1.

In order to find out the specific volume of the clays, the self-compacting dry density of specimens was determined by using the Archimedes principle. After completion of the shrinkage limit test, the dry soil pads were used to determine the dry density of the specimen. The dry density (ρ_d) of the specimen was calculated using Eq. 1.

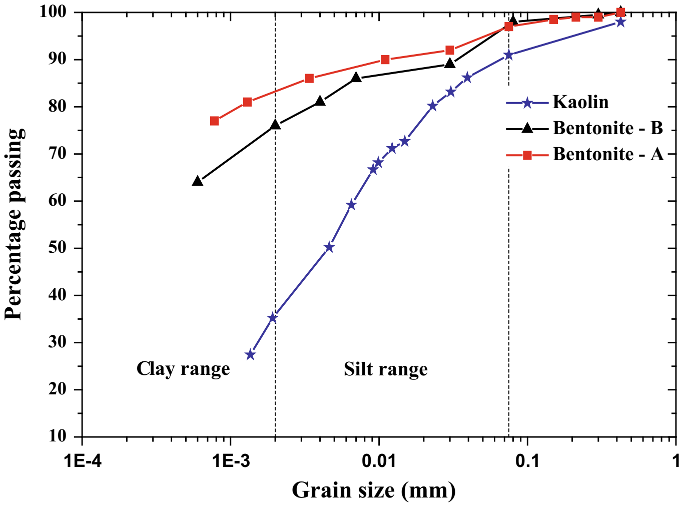

Fig. 1 The grain size distribution of the clays used in the current study

Table 1 Physicochemical properties of the clays used in the current study

Property	Bentonite-A	Bentonite-B	Kaolin
Specific gravity	2.76	2.64	2.62
Liquid limit (%)	220	172	44
Plastic limit (%)	43	52	23
Shrinkage limit (%)	12	26	19
Clay fraction (%)	76	82	33
Cation exchange capacity (meq/100)	65	48	9
Smectite content (%)	76	42	–
Specific surface area (m^2/g)	386	520	58
Soil classification	CH	CH	CI

$$\rho_d = \frac{m_a}{m_a - m_f} \times \rho_f \tag{1}$$

where ρ_f is the density of the immersion liquid (mercury); m_a is the mass of soil specimen in the air; m_f is the mass of soil specimen in mercury (nonpolar liquid).

The measured dry densities of the clays were 1.92, 1.41, and 1.71 Mg/m^3 for bentonite-A, bentonite-B, and kaolin, respectively. Due to the higher specific gravity self-compacting capacity of bentonite-A was found to be higher. Similarly, due to the high specific surface area bentonite-B possesses higher specific volume and less self-compaction density.

2.2 Methodology

Free swell void ratio (FSVR) of the clays is defined as the ratio of the swell volume of the void space that is occupied by the expansive clays under no-load condition to the volume of dry clay solids [7]. The free swell void ratio of the clays depend on the type of exchangeable cations, mineralogical compositions, and the type of pore fluid used. In order to measure the free swell voids ratio of clays, about 2 gm of the soil was taken in a graduated 100 ml glass jar and approximately 80 ml of DI water to the soil. Soil particles stuck to the walls of the cylinder were washed with salt solutions and filled up to 100 ml mark. After 24 h of duration, the free swelling volume was noted as V_f. To evaluate the effect of salt concentration on the free swell void ratio, the similar tests were repeated with (0.1, 0.5 and 1 M) concentrations of the NaCl.

The free swell void ratio (e_f) of the bentonites was calculated from Eq. 2.

$$e_f = \left[\frac{\gamma_w G_c V_f - W_c}{W_c}\right] \tag{2}$$

where W_c is the weight of clay, G_c is the specific gravity of clay, and γ_w is the specific gravity of water.

In order to conduct liquid limit tests for fine-grained soils such as clay soils, the dry soil samples were mixed with different moisture contents of water or electrolyte fluids. The clay–water mixtures were placed in airtight plastic bags and these bags were kept in a desiccator to control the moisture exchange from the external environment. After equilibration of 7 days, the soil specimens were taken out from the desiccator and used for performing liquid limit tests. The liquid limit of the clays was determined by using cone penetration method. For shrinkage limit tests, the bulk volume of the soils was measured using a standard mercury displacement method (IS: 2720 Part 6, 1972). To find the significance of NaCl concentration on the liquid limit and shrinkage limit, similar tests were repeated with various pore fluid concentrations such as 0.1, 0.25, 0.5, and 1 M of NaCl.

The effect of saline solution concentration on the stiffness and hydraulic conductivity of the clays were evaluated by using oedometer tests. The oedometer tests were carried out at different concentrations such as 0.1, 0.5 and 1 M of NaCl. The oedometer tests were performed according to the Indian Standard Code IS 2720-15 (1965). After performing the liquid limit tests, the same soil paste was used for oedometer soil specimen preparation, by maintaining the targeted water content of 1.1 time's liquid limit (LL). The required quantity of the clay paste was carefully transferred to oedometer rings. In order to remove air bubbles in the soil paste, the oedometer ring along with soil samples was pressed and tampered. The soil specimen size of 60 mm diameter and 20 mm height was compacted in the oedometer ring. In ordered to provide double drainage condition, two porous stones along with filter papers were placed on both sides of the soil specimen. The vertical consolidation stress starts with initial stress of 5 kPa and applied up to 800 kPa with increment ratio of 1.

The change in the void ratio due to overburden pressure was calculated from Eq. 3:

$$\Delta e = \frac{\Delta H}{H}(1 + e_0) \tag{3}$$

where Δe and ΔH are the change in void ratio and change in height of the specimen to each of the corresponding load increments; H and e_0 are the initial height and initial void ratio of the soil specimens.

The coefficient of volume compressibility, the coefficient of consolidation, and saturated hydraulic conductivity (k) of the soils were calculated from Eqs. 4, 5, and 6, respectively,

$$m_v = \frac{a_v}{1 + e_0} \tag{4}$$

$$c_v = \frac{T_v H^2}{t_{90}} \tag{5}$$

$$k = c_v m_v \gamma_w \tag{6}$$

where a_v is the coefficient of compressibility, C_v is the coefficient of consolidation, and γ_w is the unit weight of pore water pressure. The settlement of the soil samples corresponding to various time intervals is recorded by using LVDT. The change in displacement of the samples was measured with a precision of 0.001 mm. The coefficient of consolidation (C_v) was evaluated by using Taylor's square root time ($\surd T$) method.

3 Results and Discussions

The effect of NaCl concentrations on the free swell void ratio and liquid limit of the clays were presented in Fig. 2a. The effect of salt concentration on the shrinkage limit of the clays was presented in Fig. 2b. The free swell void ratio of bentonite-A, bentonite-B, and kaolin was found to be 27, 14, and 7.7, respectively. The results showed that the impact of the saline fluid was significant in case of the bentonite having high smectite content; for bentonite-A, both the liquid limit and free swell void ratio were drastically decreased with increase in salt concentration. For both the bentonite-B and kaolin, irrespective of the salt concentration free-swell void ratio was found to be constant. Whereas for the bentonite-B, the liquid limit was decreased linearly with increase in salt concentration, in case of kaolin liquid limit was slightly increased up to 0.5 M of NaCl concentration, subsequently, for higher

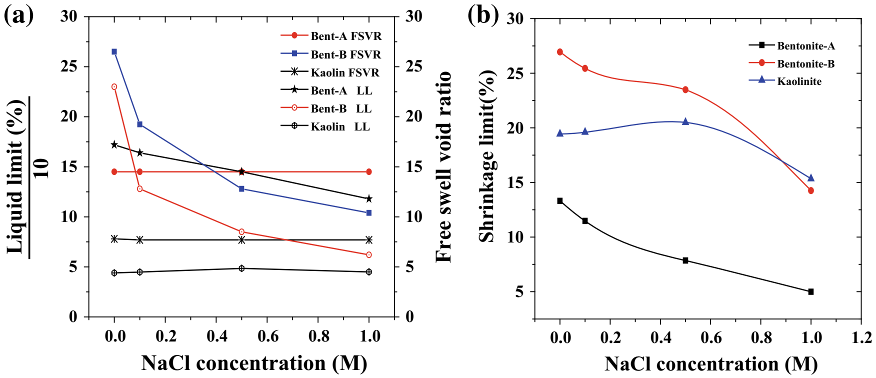

Fig. 2 **a** The effect of salt concentration on the free swell void ratio and liquid limit of clays; **b** the effect of salt concentration on the shrinkage limit of the clays

concentrations, the value was found to be decreased. Therefore, the self-healing capacity of the barrier materials decreased when exposed to higher salt concentration.

According to Gouy–Chapman theory, the diffuse double layer repulsion is inversely proportional to the square root of the electrolyte concentration [18].

$$\theta = \sqrt{\varepsilon_0 kRT/2F^2Cv^2} \qquad \theta \propto \frac{1}{\sqrt{C}} \tag{7}$$

where θ is the Debye length (m), C is the pore fluid concentration, T is the absolute temperature, k is the relative permittivity of the pore fluid, R is the universal gas constant (8.134 J/mol./K), and F is Faraday's constant. Equation 7 suggested that Debye length is inversely proposal to the square to the pore fluid concentration and valency of exchangeable cations present in the clays. In the current study, bentonite-A contained sodium as a predominant cation; thus it possesses higher free swell capacity. The free swell void ratio of bentonite-A was significantly decreased with increase in NaCl concentration whereas for bentonite-B and kaolin, it was found to be constant irrespective of the salt concentration. Therefore, it is difficult to quantify the impact of the pore fluid concentration on the diffused double layer thickness of the clays. The increase in electrolyte concentration causes more flocculation occurred in the crystal lattice of high smectite clays leading to decrease in the total surface area and the absorbed water resulting in decrease in the swell void capacity and the liquid limit.

The shrinkage properties of the clays play a significant role in the performance of the buffer materials. The clays with low shrinkage limit are considered as a good barrier material. Basically, the soil having the higher plasticity index possesses low shrinkage limit. In the current study, both the plasticity index as well as the shrinkage limit of bentonite-B were found to be much higher as compared to kaolin.

It indicates that the shrinkage limit of the soils does not depend upon the plasticity characteristics, and that it primarily depends on the relative grain size distribution of the soil; similar observations made by Sridharan and Prakash [17]. Bentonite-B has a more specific surface area which leads to high micro/mesoporous voids; hence, it possesses the highest shrinkage limit compared to other soils. For all the soils, the shrinkage limit was decreased with increased NaCl concentration, this phenomenon is due to the addition of salt solutions to the clays attributed to bringing the soil particles closer and obtaining a flocculating structure which led to denser clay matrices.

The time versus settlement curves for the various clay soils is presented in Fig. 3. The results showed that both the bentonite-B and kaolin clays possess rapid initial settlement and reach 90% of consolidation (t_{90}) rapidly. For bentonite-A, progressive settlement occurred and the t_{90} values were measured to be much higher than that of the other clay soils. The time required for 90% of consolidation of the clay soils is subjected to effective stress from 10 to 800 kPa which are listed in Table 2. The results showed that irrespective of the consolidation stress, bentonite-A possesses higher t_{90} values. Although bentonite-B contains higher clay fraction, it possesses higher repulsive pressure to any corresponding dry density (observed from *e*-log(*p*) curve); the t_{90} values were found to be much lower as compared to bentonite-A. It can be said that the t_{90} values depend on the particle arrangement and pore size distribution of the clays. The bentonite-A has a higher amount of smectite subjected to a high thickness of the diffused double layer. Although bentonite-B has more specific surface area associated large amount of micro-voids, contains less amount of smectite mineral thus the thickness of the double layer is lower. When the external stress is applied to bentonite-B, the large

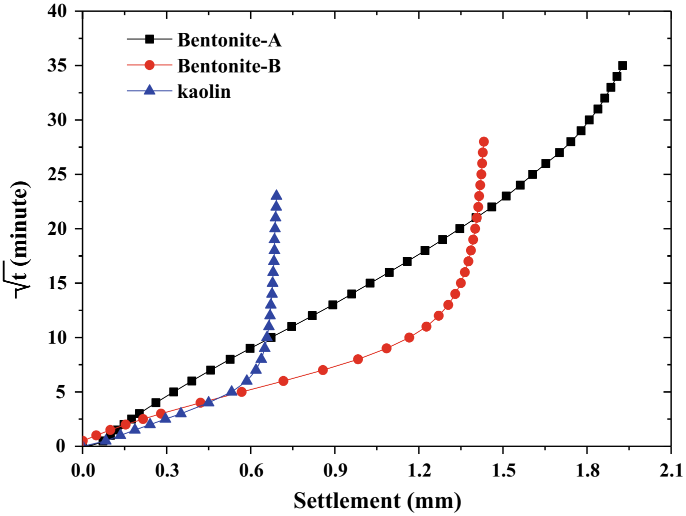

Fig. 3 Typical time settlement curves for different clays for 10 kPa stress by using DI water

Table 2 The time required for 90% of consolidation of the clay soils subjected to effective stress from 10 to 800 kPa

Type of soil	Bentonite-A	Bentonite-B	Kaolin
Consolidation stress (kPa)	Time required for 90% consolidation (min.)		
10	2485	384	58
20	2389	247	29
50	2285	156	19
100	1989	95	12
200	1486	75	9
400	1211	54	8
800	986	45	7

amount of the micro-porous is subjected to rapid settlement subjected to lower t_{90} values. In all the cases, the volume compressibility (m_v) was found to be increased at the initial phase of the loading and decreased to the subsequent loads. As the salt concentration increased, the time required for 90% of consolidation (t_{90}) was decreased whereas the coefficient of consolidation (C_v) significantly increased.

The initial water content plays a significant role in the compression index of the soils. In order to eliminate the effect of initial water content on the compression index, the parameter compression ratio (CR) was used. The compression ratio is the ratio of compression index and initial specific volume [10]. The current experimental results suggested that both the compression index and compression ratio values were slightly decreased with an increase in the pore fluid concentration due to an increase in the NaCl concentration from zero to 1 M; CR values were noticed to decrease from 0.29 to 0.23 for bentonite-A whereas for bentonite-B, it was found to decrease from 0.24 to 0.22. For kaolin, the CR value was noticed to decrease from 0.16 to 0.13. Nagaraj et al. [10] reported that for highly compressible clays, the compression ratio would be relatively close to constant.

The magnitude of the decrease in the void ratio due to the consolidation stress was found to be higher for bentonite-A than that of bentonite-B and kaolin. In the presence of DI water, the compression index values for bentonite-A, bentonite-B, and kaolin were noted as 1.49, 1.14, and 0.26, respectively. The effect of salt concentrations on the consolidation characteristics of the clays was presented in Fig. 4a. The compression index for bentonite-A decreased from 1.49 to 0.62 to the corresponding salt concentration from zero (DI water) to 1 M of NaCl. Similarly, for bentonite-B and kaolin, the compression index values varied from 1.14 to 0.83 and 0.35 to 0.28, respectively. The results indicated that the percentage of decrease in the compression index was maximum for bentonite-A which is about 57%, followed by bentonite-B and kaolin about 27% and 20%, respectively. The effect of salt concentrations on the hydraulic conductivity of the clays was presented in Fig. 5. The saturated hydraulic conductivity of the soils is a monotonic function of its dry density; in a semilogarithmic plane, the hydraulic conductivity of the soils varies linearly with the dry density. The results suggested that for a given dry

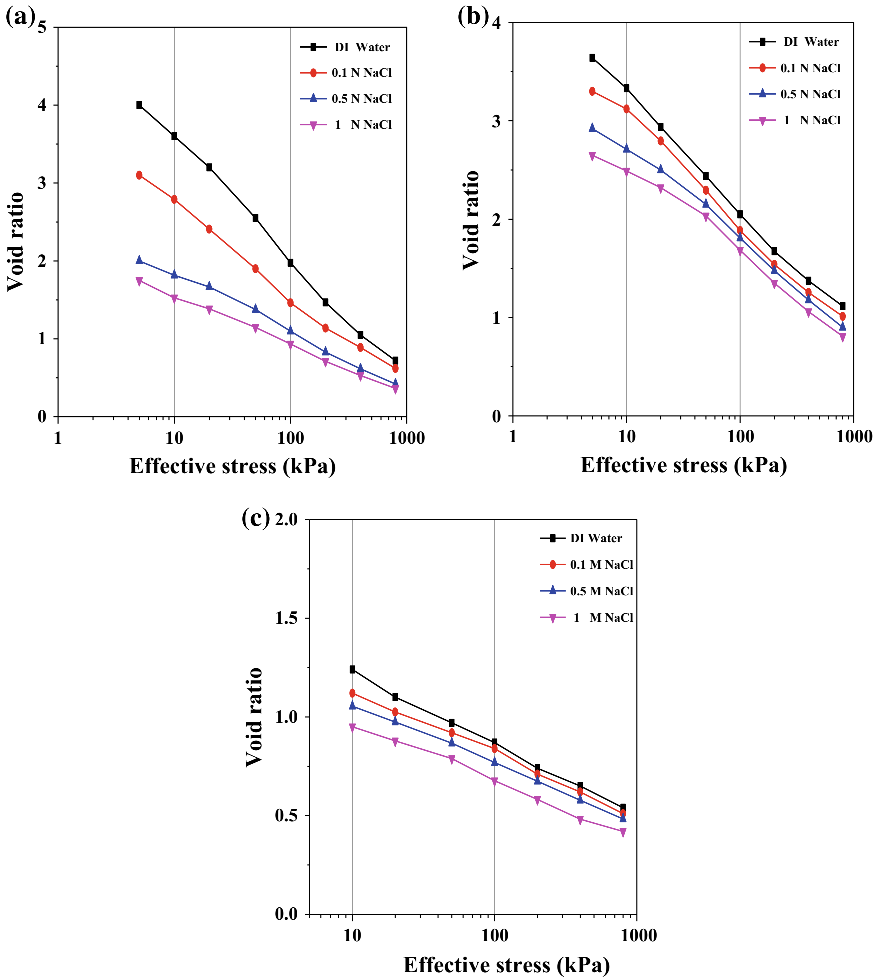

Fig. 4 The *e*-log(*p*) plots for the clay soils **a** bentonite-A; **b** bentonite-B; **c** kaolin

density, the hydraulic conductivity of bentonite-A was found to be minimum in the range of 5.5 × 10^{-9}–3 × 10^{-11} with the corresponding dry density between 0.7 and 1.5 Mg/m^3. Similarly, the kaolin clay possesses higher hydraulic conductivity of about 1 × 10^{-8} to the corresponding dry density of 1.76 Mg/m^3.

The DI water as a pore fluid and for the total consolidation stress of 1.6 MPa the dry density attained by bentonite-A, bentonite-B and kaolin were measured to be 1.6, 1.45 and 1.78 Mg/m^3, respectively. The results suggested that for obtaining the same dry density, bentonite-B required more consolidation stress. The contrasting results were found in the study for a given dry density where bentonite-B possesses more swelling pressure as well as higher hydraulic conductivity as compared to

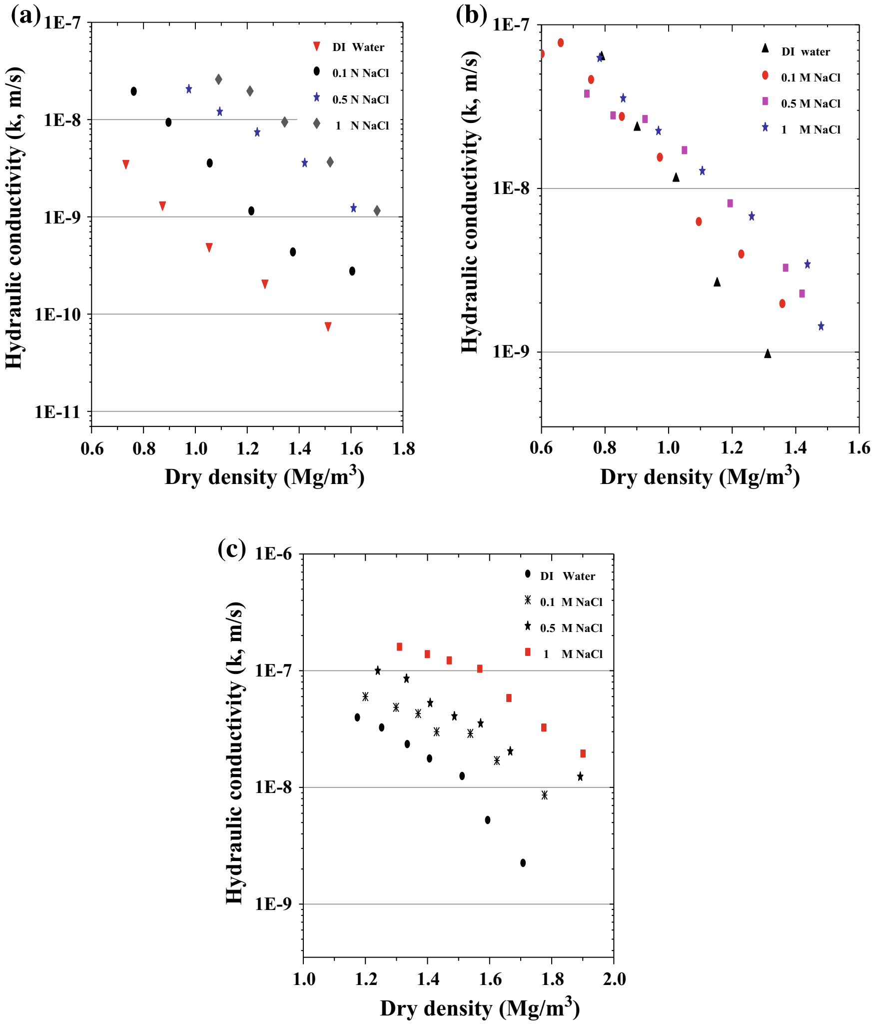

Fig. 5 The hydraulic conductivity versus dry density plots for the clay soils **a** bentonite-A; **b** bentonite-B; **c** kaolin

bentonite-A. Therefore, it can be concluded that the soil structure plays more important role in the hydraulic conductivity of the soils. In order to study the microstructure of clays, scanning electron microscope study was conducted for both the bentonite samples after performing the oedometer tests for DI water conditions; the results are presented in Fig. 7. The microstructure of bentonite-A showed a flocculated structure with equidimensional flakes that have a film-like appearance, whereas, bentonite-B indicates dispersive structure with needle-like poorly distinct, broken structure. Therefore, bentonite-A possesses lower hydraulic conductivity.

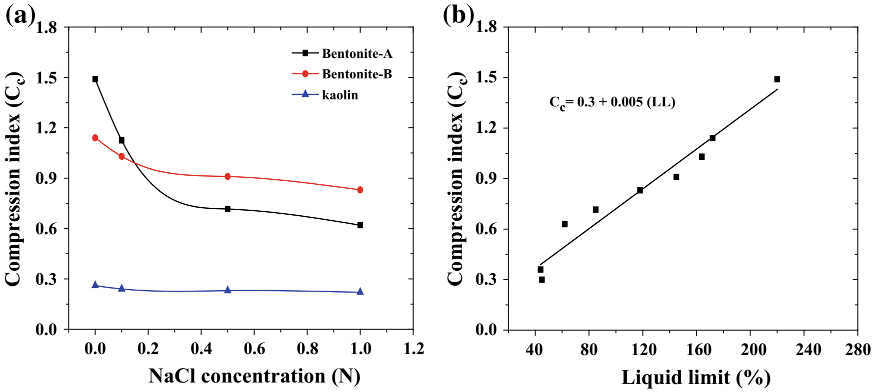

Fig. 6 **a** The C_C versus NaCl salt concentration plots for clays; **b** the liquid limit versus compression index of clays

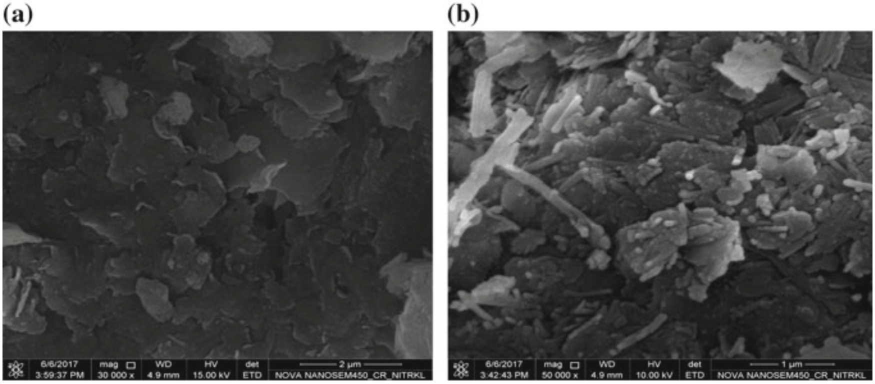

Fig. 7 The FESEM images of the bentonites **a** bentonite-A; **b** bentonite-B

The effect of the salt concentration on the hydraulic conductivity was found to be maximum for bentonite-A, whereas, for bentonite-B and kaolin, the effect was found to be marginal. The hydraulic conductivity of the clays was increased with the pore fluid concentration due to the diminishing of the diffused double-layer thickness of the clay minerals [22]. For a given dry density as the salt concentration increased from zero (DI) to 1 molarity, the k value was found to decrease about 100 times for bentonite-A. Whereas for bentonite-B and kaolinite, the hydraulic conductivity values were marginally decreased. The effect of salt concentration was marginal for kaolinite type of clays due to their small diffused double-layer thickness and the presence of stronger hydrogen bond in the crystal lattice. The results indicate that both Ca–bentonite and kaolin have the maximum endurance against the salt concentration. Based on the test results, the relationship between compression index and liquid limit of the clays is shown in Fig. 6b. The results indicate that both the liquid limit and compression index of the clays are linearly correlated.

SKB report (2004) suggested that the basic criteria for buffer materials are that they should possess swelling pressure of more than 1 MPa and the hydraulic conductivity should be less than 10^{-9} m/s. From the current experimental study, it was observed that both the bentonites meet the basic standards for engineered barriers in terms of hydraulic conductivity. However, in the presence of the salt concentration, the hydraulic conductivity of bentonite-A was drastically increased; hence for the barrier material to be used in the coastal region, a higher density of bentonites is recommended. Although bentonite-B had higher endurance capacity against the salt concentration and also possessed higher k values, for higher dry densities, both the bentonites are recommended as a good barrier material.

4 Conclusions

The current study evaluated the effect of NaCl concentration on the Atterberg limits, free swell capacity, and hydraulic conductivity of two Indian bentonites and kaolin clay. The results suggest that the shrinkage limit of the clays does not depend on the plasticity characteristics; it primarily depends on the pore size distribution of the clays. Due to increase in salt concentration, both the liquid limit and shrinkage limit of the bentonites were found to be decreased whereas for kaolin, it was found to be slightly increased up to 0.5 M NaCl upon that decreases at higher salt concentrations. The experimental results revealed that the salt concentration has a significant effect on the hydraulic conductivity of Na–bentonite containing high smectite content whereas for bentonite with low smectite and kaolin, it does not show any significant effect. For the same consolidation stress, bentonite-A attained higher dry density and possesses low permeability values whereas for bentonite-B, it was found to be the opposite. According to the DDL theories, the thickness of the DDL decreases as the saline concentration increases resulting in increases in the effective pores' size. Therefore, the hydraulic conductivity increases with increase in the pore fluid concentration. The study found a good correlation between the compression index and the liquid limit of the clays.

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