

Effect of Alkaline Environment on the Swell Pressures of Compacted Bentonite under Thermal History

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Abstract. Bentonite is used as a buffer material in the deep geological repository for the safe disposal of high-level nuclear waste (HLW). The repository consists of a natural barrier system that consists of a rock, and its surroundings and an engineered barrier system consists of a buffer material as well as a waste canister. Bentonite is compacted around the waste canister to isolate it from the atmosphere as well as to provide long-term stability to the barrier. The high temperature (150°C to 250°C) of the waste canister reduces over thousands of years till the radioactivity of the waste degrades as well as while the long-term operation of the repository, the concrete components will deteriorate and produce alkaline fluids having $\text{pH} > 12$. This long-term influence of high temperature may create a thermal history on the compacted bentonite, and the highly alkaline solution may alter the bentonite near the concrete affecting the physical and chemical properties of compacted bentonite. Hence it is necessary to investigate the influence of alkaline environment along with the induced thermal history on the swelling pressure of compacted bentonite. The paper discusses an assessment of swell pressures of compacted bentonite (Barmer 1 (B_1)) from Barmer district of Rajasthan, India with an initial dry density of 1.5 Mg/m^3 and subjected to 110°C , hydrated with cement solution and distilled water. The swelling load is recorded with the help of digital load frame and compared with non-heated samples of the same density. The swelling time data of compacted bentonite is presented and compared. Experimental evidence indicated that the swelling pressure of compacted bentonite was less when hydrated with cement water as compared to distilled water. But the time taken for saturation was more when hydrated with cement water as compared to distilled water. However, further investigation is required to understand the swelling mechanism under highly alkaline solutions.

Keywords: deep geological repository, high-level nuclear waste, buffer, swelling pressure, high-temperature canister.

1.0 Introduction

Compacted bentonite is used as a buffer material for constructing a deep geological repository for the disposal of high-level nuclear waste [15] [12] [6]. The bentonite used as a buffer material in repositories is under unsaturated conditions, and these conditions influence the behavior of compacted bentonite, including its hydraulic conductivity, shrinkage, and swelling. The swelling behavior and stability of bentonite depend on its contents and the percentage of swelling clays, i.e., Smectite. [7] reported that smectite remained dominant in Wyoming MX80 bentonite after alteration but significant changes in the morphology of smectite were observed in rich brine solutions of NaCl and MgCl_2 at high temperature ($< 150^\circ\text{C}$). Such interactions of bentonite with different solutions like KOH at 80°C may change the mineralogical composition of bentonite and its swelling characteristics. According to [4] the direction of compaction results in the higher value of swell pressure along with it. [13] observed

a higher value of swell pressure of compacted bentonite in the axial direction than in the radial direction. [18] defined a coefficient of anisotropy as the ratio of radial to axial swelling pressure to describe the bentonite anisotropy. Thus the long-term performance of repository depends on these interactions.

According to [20], when clayey soils are subjected to elevated temperature, soil changes its index and engineering properties. The compacted buffer material is subjected to heat by (i) radiations coming from the waste canister released by waste material and (ii) other end of buffer material which is in contact with the host rock. According to [10], elevated temperature affects the index, physicochemical, and hydro-mechanical properties of bentonite and increases the permeability of the bentonite [1]. [17] observed a decrease in soil suction when the temperature was increased from 22°C to 80°C. [3] observed the decrease in swell pressure of bentonite when the temperature was increased. However, the effect of high temperature on the properties of the bentonite buffer depends on the type of clay minerals present in the sample [19]. These observations have suggested researchers to study the effects of thermal history on the properties of bentonite. Recent studies were reported on the influence of thermal history on the performance of bentonite buffer by [8] [9]. It was observed that with the increase in temperature the properties of powdered bentonite such as swell pressure, liquid limit, plastic limit, specific gravity, specific surface area, cation exchange capacity, etc., were reduced. [5] performed experiments on montmorillonite and concluded that with an increase in time duration of heating and increased temperature results in decreased liquid limit and plastic limit. The swelling potential and swelling pressure tests were conducted on samples with a dry, optimum, and wet side of optimum water content. For dry samples, the swelling potential was higher than the other two cases. Both the swelling potential and swelling pressure depends on the temperature the sample was exposed and the duration of exposure. The swelling pressure decreases with increase in temperature and duration of heating. Increasing the temperature and duration of heating reduces the concentration of Na^+ but no such effect was observed in dry unit weight.

During its operation, pore fluids coming from the host rock, and may get percolated through concrete and becomes alkaline by having cementitious characteristics hydrates the bentonite. Due to hydration, of pore fluids, the technological or natural gaps could be gradually sealed by swelling of bentonite [2]. The properties of compacted bentonite get altered by coming in contact with cement water having a high pH of 12-13.5 due to the release of Sodium (Na^+) and Potassium (K^+). The properties such as cation exchange capacity changes due to the adsorption of these released Sodium (Na^+) and Potassium (K^+) cations. The presence of moisture seeps through the layer of cement from the host rock and creates the alkaline environment ($\text{pH}>12$) inside the repository. The prolonged degradation produces the highly alkaline fluids that diffuse through the compacted bentonite buffer and changes the behavior of bentonite that is useful to act as a buffer in the repository [21] [24] [9]. The alkaline pore fluids may enhance the chances of solubility of radioactive elements [6]. The engineering properties of bentonite are different at room temperature and high temperature, and there is a requirement of suitable swell pressure for the long-term operation of the repository. The buffer must be designed to perform well for safety against both radioactive waste and reactive solutions flowing through the buffer material. Significant studies have been conducted to understand the effects of high temperature on the clayey

and expansive soils in the past, the studies on the effect of thermal history on the bentonite under hyper alkaline environment are still not clear. Hence, a proper quantified study is essential to develop a deep geological repository (DGR). Barmer bentonite is chosen as buffer material for its favorable physical and mineralogical properties. Again, a lot of studies are presented concerning the effects of salinity on the swelling properties of bentonite. However, very few studies are focused on the effect of swelling pressure of bentonite in the presence of alkaline cement solution at elevated temperature. Therefore, in current experiments, swell pressure tests are performed on Barmer bentonite with infiltration of distilled and cement water solutions. The variation in swell pressure of bentonite due to these solutions and elevated temperature is discussed.

2.0 Materials and methodology

2.1 Materials

Bentonite (Barmer 1 (B₁)) from Barmer district of Rajasthan, India was used in this study. The basic characterization of the bentonite was determined by ASTM procedure (ASTM D4318, D7928-17, D720, D698) (Table 1).

Table 1: Properties of bentonite used in the study [8]

Properties	Barmer Bentonite
Specific Gravity	2.79
Clay content (%)	89
Sand Sized Fraction (4.75–0.075 mm)	1.63
Liquid Limit (%)	447.28
Plastic Limit (%)	49.23
Plasticity Index (%)	398.05
Specific Surface Area (m ² /g)	507.74
Maximum dry density (g/cm ³)	1.42
Optimum moisture content (%)	35
Cation Exchange Capacity (meq/100 g)	94.58
Free swell index (%)	833.33
pH	8.29

2.2 Methodology

2.2.1. Determination of swell pressure

The swell pressures of compacted bentonite specimens were determined using constant volume consolidation cell [12] [16]. An air-dried sample of bentonite was compacted in a consolidation ring (diameter = 61 mm; height = 20 mm) and a thickness of 8 mm. The water content of the bentonite during compaction was 14%, and the specimens were prepared at a compaction density of 1.5 Mg/m³. The compacted samples were mounted on the swell pressure assembly consisting of a 10 kN load

cell (capacity= 10 kN; sensitivity=0.01 kN) and LVDT (maximum displacement = 20 mm; sensitivity=0.01 mm). The specimens were saturated using distilled water and cement water. A photograph of the test set up is presented in Fig 1. The load during different stages of saturation was indicated by the load cell were noted down at the regular intervals of time. The experiment was stopped when the load cell showed a constant reading of load [14]. The equilibrium load divided by the cross-sectional area of the specimen gave the swell pressure of the compacted specimen.



Fig 1 Swell pressure apparatus

2.2.2. Preparation of cement solution

The cement pore-water was prepared by mixing the ordinary portland cement with distilled water maintaining the w/c ratio equal to 1.0 and separating the supernatant water. The pH value of the cement solution used in this study is 12.5, i.e., hyper alkaline solution.

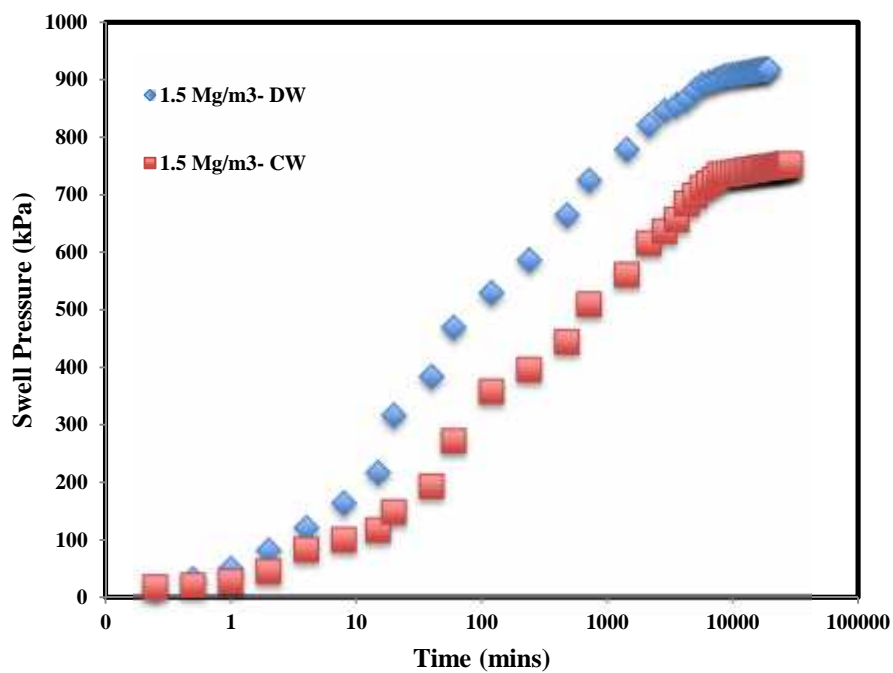
2.2.3. Influence of thermal history

To study the influence of thermal history on the swell pressure of bentonite, the compacted bentonite specimens of density 1.5 Mg/m^3 were exposed to 110°C for about three h, and results were compared with the non-heated specimen.

3.0 Results and discussions

3.1 Time-swelling of compacted bentonites without heating

The plot of swell pressure with time for the specimens compacted at densities 1.5 Mg/m^3 and without heating (WH) for distilled water and cement water is presented in fig 2(a). The specimens compacted to 1.5 Mg/m^3 and without heating started attaining the equilibrium swell pressure after 18,720 min. of starting the test when infiltrated with distilled water. Comparatively, the specimens without heating started attaining the equilibrium swell pressure after 26,640 min of starting the test when infiltrated with cement water. The final swell pressure values of compacted bentonite specimen are 913.78 kPa and 749.45 kPa for distilled water and cement water, respectively.



(a)

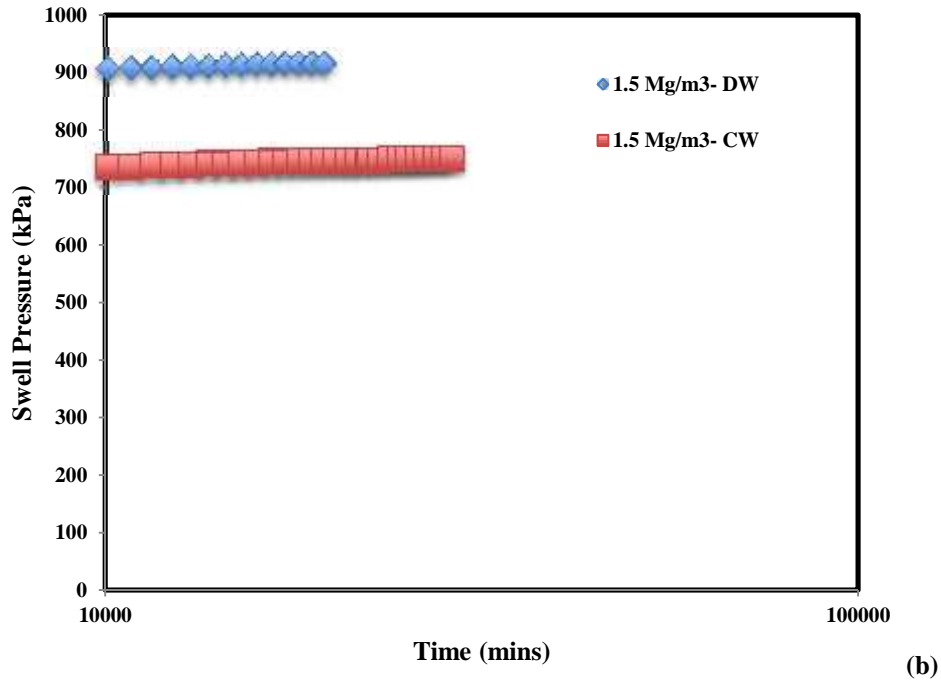
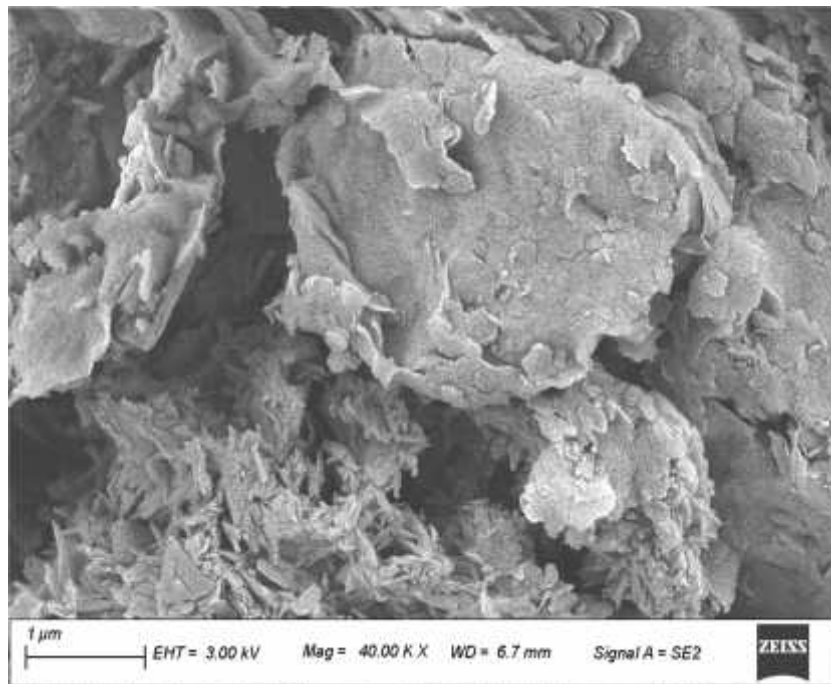
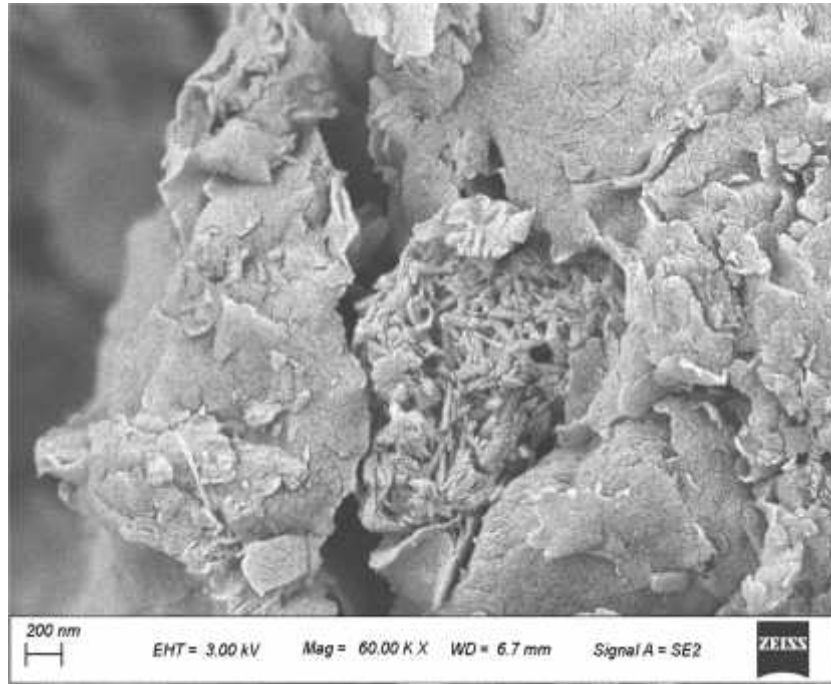


Fig 2(a) Swell pressure Vs time of compacted bentonite (without heating) with distilled water (DW) and cement water (CW) (b) Magnified image of swell pressure Vs. time while saturation.

The important observation found was, though the swell pressure in case of distilled water was greater than the cement water the time of saturation for the sample saturated with cement water was more than in case of distilled water (fig 2(b)).



(a)



(b)

Fig 3(a) FESEM images of compacted bentonite specimen saturated with cement water (WH).

(b) Magnified image (cement water saturation)

In case of cement water, the exact reason behind the long duration taken for the saturation may be due to clogging of pores by the precipitation of cementitious products, which was further investigated by microstructural behavior by FESEM (fig 3 (a) and (b)). The observed swell pressure is less than that of sample hydrated with distilled water. The reason behind the reduced swell pressure may be due to the rapid exchange of Na^+ and Ca^+ ions with the hyper alkaline cement solution. From fig 3(a) and (b), it can be observed that there is a deposition of cementitious material in between the pores of the bentonite particles. The presence of calcium in highly alkaline cement water may have exchanged with the active ions present with the bentonite, and therefore, the deposition is observed.

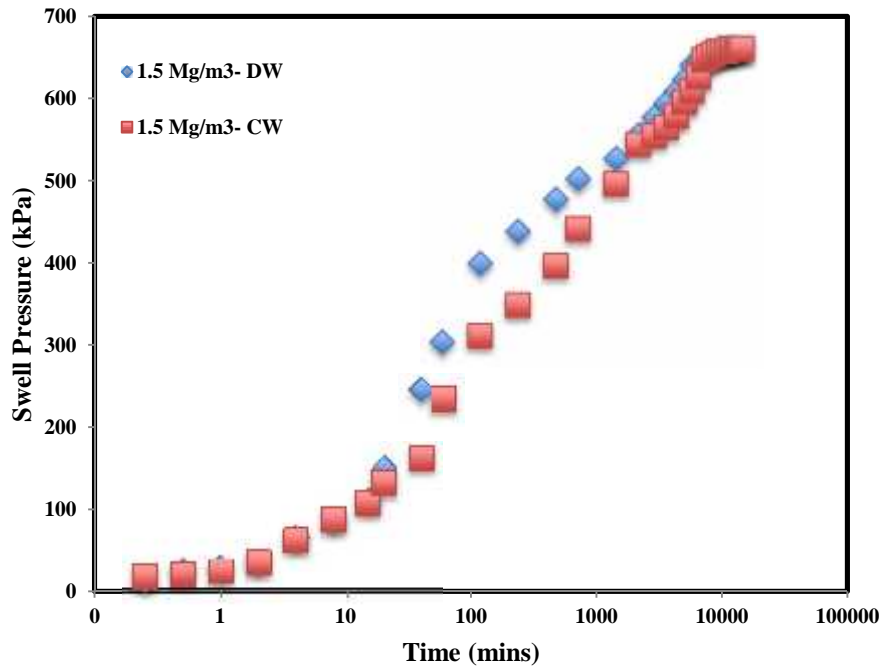
3.2. Time-swelling of compacted bentonites subjected to high temperature

The plot of swell pressure of compacted bentonite against time having compaction density 1.5 Mg/m^3 heated at 110°C for about three hrs for both distilled water and cement water is presented in fig 4(a). The specimens compacted to 1.5 Mg/m^3 and heated at 110°C started attaining the equilibrium swell pressure after 16,640 min after starting the test when infiltrated with distilled water. Comparatively, the specimens without heating started attaining the equilibrium swell pressure after 18,720 min after starting the test when infiltrated with cement water. The final swell pressure values of compacted bentonite specimen having 1.5 Mg/m^3 densities are almost same, i.e., 661 kPa and 660 kPa for distilled water and cement water respectively. However, the time of saturation for the samples in case of cement water was more than that of distilled water (fig 4(b)) (table 2).

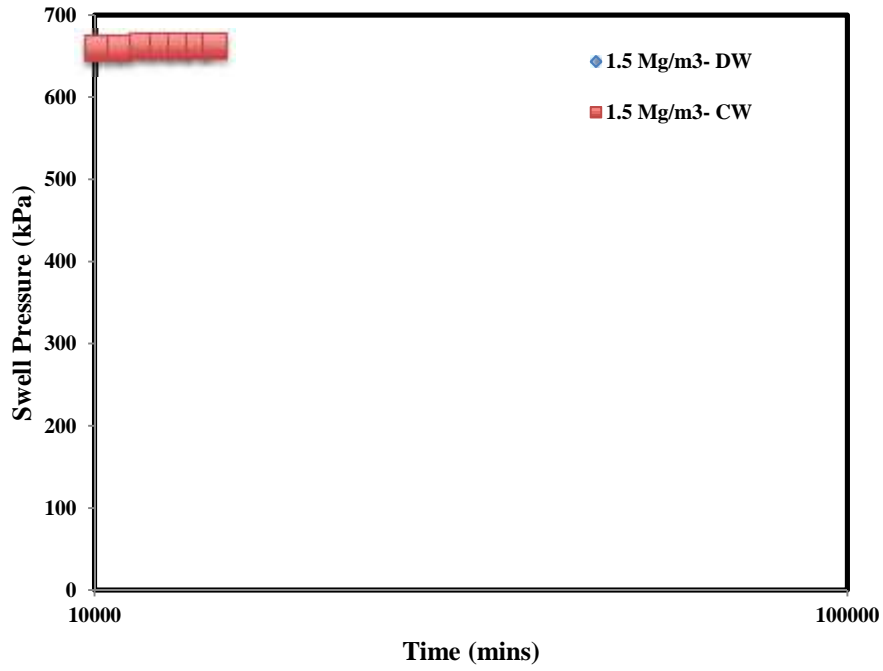
Table 2. Variation in a final time of saturation of compacted bentonite samples.

Temp (°C)	1.5 Mg/m ³ (min)	
	DW	CW
WH	19940	28800
200	12960	14400

In fig 5(a) and (b), the deposition of cementitious material in between the pores of the bentonite particles is observed when the sample was heated at 110 °C. The presence of calcium in highly alkaline cement water may have exchanged with the active ions present with bentonite, and therefore, the deposition is observed. The flaky structure of bentonite is shown in fig 5 (a).

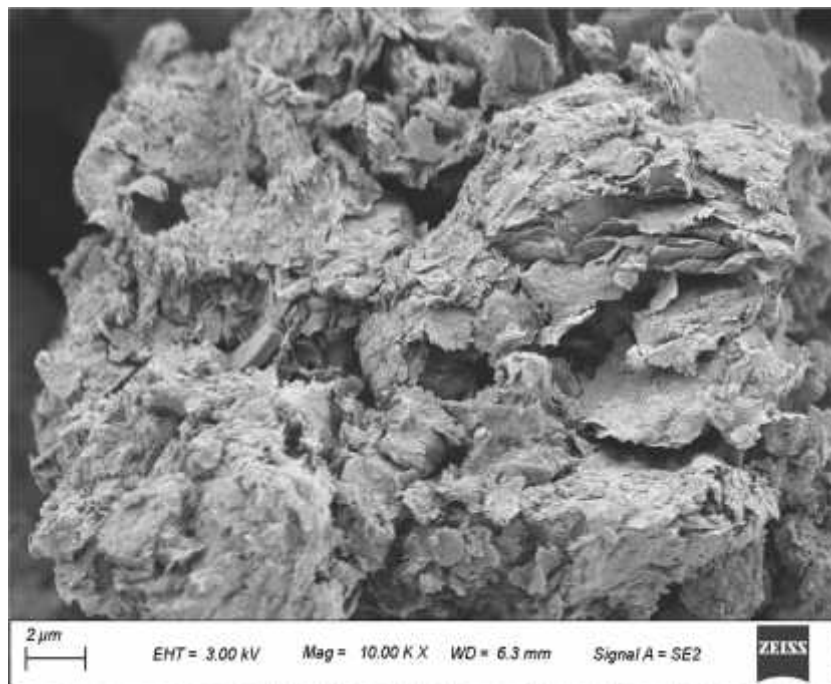


(a)

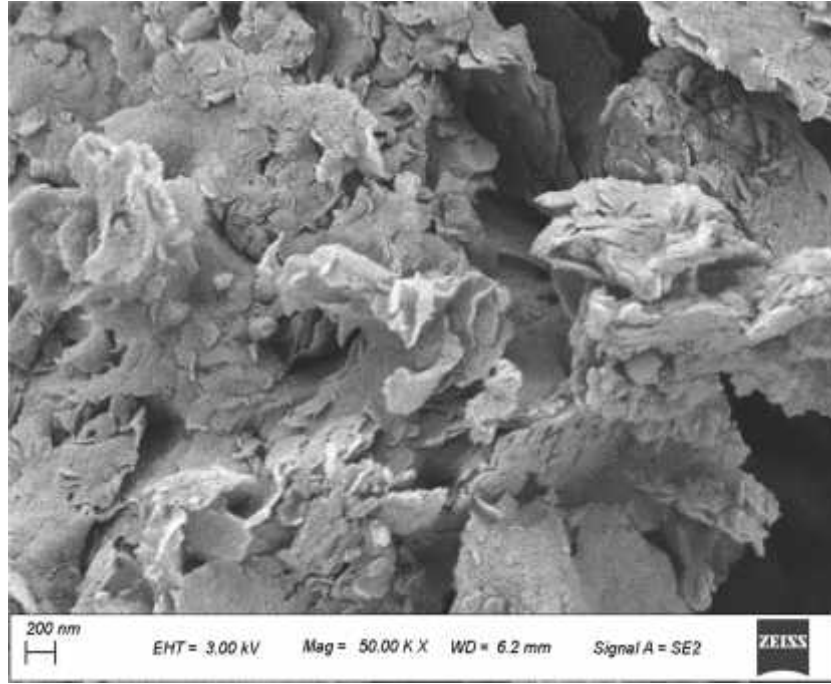


(b)

Fig 4(a) Swell pressure Vs time of compacted bentonite (heated at 110°C) with distilled water (DW) and cement water (CW) (b) Magnified image of swell pressure Vs. time while saturation.



(a)



(b)

Fig 5(a) FESEM images of compacted bentonite specimen saturated with cement water (110°C).

(b) Magnified image with cement water saturation

When the compacted samples are heated at high temperature such as 110 °C, the available moisture will be lost, and the sample is completely dry as can be observed from the fig 5(a) and (b).

Table 3. Percentage variations in swell pressure at different temperatures

Temp (°C)	1.5 Mg/m ³ (kPa)		
	DW	CW	% Change
WH	913.78	749.75	17.95
110	661.71	660.74	0.15

Table 3 shows the percentage of variations in swell pressure at different temperatures when saturated with distilled water and cement water. It is observed that the percentage change is more in case samples without heating than heated at 110°C.

4.0 Conclusion

In this study, swelling pressure tests were conducted on densely compacted Barmer bentonite specimens, which had an initial dry density of 1.5 Mg/m³. Influences of highly alkaline cement water on swelling behavior as well as the combined effect of thermal history and alkaline solution on the microstructural changes of the compacted Barmer bentonite were analyzed.

1. The highly alkaline (pH=12.5) cement solution significantly influenced the swelling properties of the compacted Barmer bentonite.
2. The swell pressures of the compacted specimen of bentonite infiltrated with distilled water were more than the specimens infiltrated with cement water.
3. The swell pressure decreased with the increase in temperature in both the cases of distilled water and cement water.
4. The time taken for saturation in case of cement water was more than that of distilled water.

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