

Technical Note

The effect of thermal history on thermo-mechanical behavior of bentonite-sand mixture

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Abstract

Bentonite-sand mixture is one of the most important candidates for engineering buffer element in nuclear waste repositories; so the analysis of its thermo-hydro-mechanical behavior is important for design purposes. An innovative setup of classic oedometer was used for swelling and compression study at high temperatures in this research. A fully calibration program was utilized to include high temperature effects on measurements. This research shows that the elevation of temperature from 25 to 90°C in 1:1 bentonite-sand mixture in distilled water reduces free swelling potential and strain about 20 percent. The required time for equalization of swelling is less in high temperature due to increasing in permeability. Also, the high temperature causes increasing in compressibility rate and quantity for this buffer. For detection of this effect, XRD analysis showed that an increase in temperature causes a decrease in basal spacing. So, the particles can come near to each other more than lower temperatures and the amount of absorbed water in the microstructure of the clay is smaller. The effect of thermal history on behavior of bentonite-sand mixture has been showed and tried to clarify it. At similar stress-temperature states, thermal history causes different deformation in samples. The highest temperature that bentonite has been experienced, controls its behavior in the next thermal cycles.

Keywords: Thermo-mechanical behavior, Bentonite-sand barrier, Nuclear waste repository, Thermal history, Environmental geomechanics

1. Introduction

At present, bentonite-sand mixture is expected to be the most appropriate as a buffer material of high-level radioactive waste products when they are disposed in the deep ground repositories [1]. The buffer material primarily is required to possess a low mass diffusion coefficient. It should be capable of maintaining its integrity at elevated temperatures (up to 150°C) and in the saline groundwater that may exist in a disposal vault [2]. It should have a good resistance against chemical attacks and a high buffering capacity to adsorb radioactive escaped ions from canisters. To dissipate heat, it should possess a thermal conductivity, similar to that of the host rock [3], and on drying, should not shrink away from either the container or the rock. In addition, its bearing capacity must be sufficient to support the container and

minimize settlement or long-term "creep" deformation [4]. The dry density of pure clays can be as low as 1.3gr/cm³ when compacted in situ; however, by mixing sand with the clay, the attainable dry density can be increased. Additions of sand can be expected to decrease the creep potential of the material and also the cracking potential, increase the thermal conductivity [5], and improve soil strength properties [6]. Moreover, compact bentonite-sand mixtures may possess lower hydraulic conductivities than compacted clay [7]. For these reasons, clay-sand mixtures are being considered as potential buffer materials with the most of these benefits. Therefore, it is important to estimate the mechanical behavior of this material. For this purpose, it is necessary to clarify the stress-deformation behavior of mixtures under various conditions, such as the elevated temperature that occurs around the waste canisters.

One of the missed aspects in this field of geomechanics is the effect of thermal history on the behavior of soils. For instance, when a barrier faces a high temperature and cools after that, we have to know in the next cycle of heating what its behavior is. What mechanical characteristics are affected by the highest temperature that soil has experienced? This research tried to

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show some changes in mechanical parameters of bentonite with special thermo-mechanical paths. The effect of thermal history on microstructure of bentonite was detected by Atterberg's limits as an important indicator in clay behavior. Also, the changes in bentonite microstructure have been investigated via X-ray diffraction tests.

2. Materials and methods

The material is a 1:1 mixture by weight of silica sand and sodium bentonite. With these proportions, the behavior of the mixture is clay-dominated, with the sand essentially acting as filler. The clay is a sodium-rich bentonite from "Iran Barite Group" with liquid limit $W_L = 321\%$ and a plasticity index $I_p = 285\%$. Physical properties such as specific gravity, particle size distribution, Atterberg limits, and moisture content were determined according to ASTM methods [8]. The specific surface area (SSA) was determined using the ethylene glycol mono-ethyl ether (EGME) method described by Eltantawy and Arnold [9]. The soil was classified as CH (clays of high plasticity). The sand is a commercial, natural, medium, sub-angular, poor graded, silica sand, namely Firuzkuh 161. Its specific gravity is 2.66 and D_{50} is 0.3mm. Table 1 presents some characteristics of Iran Barit bentonite. Table 2 shows the 1:1 mixture properties.

Table 1. The Characteristics of Iran Barit Bentonite

Soil Characteristics	Definition
XRD Analysis	Montmorillonite, Quartz, Calcite
Soil Classification	CH
Liquid Limit (%)	321
Plasticity Index (%)	285
$\gamma_{d \max}$ (gr/cm ³)	1.51
Optimum Water Content (%)	24
CEC (cmol/kg soil)	68.5
G_s	2.78
pH (1:10 soil:water)	9.5
SSA (m ² /gr)	420

A Siemens-Diffractometer D8 Advance with Cu-K α radiation was used to obtain the X-ray diffraction patterns of samples. To prepare the samples for XRD tests, 0.2 g of air-dried sample was mixed with 10 ml of distilled water in a small volumetric flask and shaken for 2 minutes. Finally, using a micro pipette, four drops of the prepared solution were positioned on a glass slide. The coatings on glass slides for all samples had similar thicknesses. Then, glass slides were air-dried. Several similar samples on glass slides were prepared and heated up to defined temperatures and were scanned in the 2θ range of 4 to 60° for their XRD spectra.

In preparing specimens for consolidation testing, the quantity of de-aired water needed to produce a specimen with the target dry density first was added to the sand. The bentonite then was combined thoroughly with the sand and water, and the resulting mixture was stored in a sealed bag for 10 days at 25°C to equalize water contents in the clay. After curing in this way, the mixture was compacted statically into a rigid mold in two lifts to form specimens with 60 mm diameter and 15 mm height.

The oedometer with temperature control that was used for this research is an innovative setup of classic oedometer. In this setup (Figure 1), a thermal bath with a pump provided hot water around the sample. The temperature in thermal bath was set and controlled by a thermostat and sent by a thermocouple to data logger. Additionally, two PT100 thermocouples with $\pm 0.1^\circ\text{C}$ accuracy were used close to the sample to send the temperature of water around the sample to the data logger. Insulated tubing has the task of circulate hot water from the bath to the cell and vice versa. As the result of heating waste

Table 2. vThe Characteristics of Bentonite-Sand mixture

Soil Characteristics	Definition
Liquid Limit (%)	290
Plasticity Index (%)	255
$\gamma_{d \max}$ (gr/cm ³)	1.78
Optimum Water Content (%)	21.5

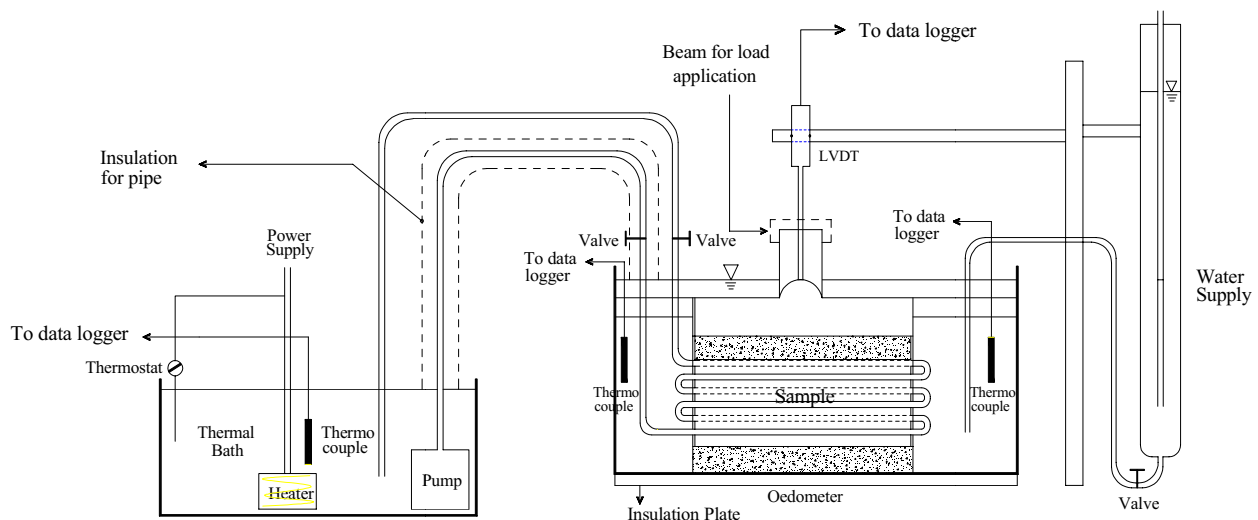


Fig. 1. The schematic setup of oedometer with temperature control

in the system, it different temperatures in thermal bath and around the sample were expected. Therefore, at the first step, the relationship between the temperature in the bath and the water inside of sample location was measured. The measurement of temperature was done by a portable thermometer with an accuracy $\pm 0.1^\circ\text{C}$. After this step, one relation equation was extracted to use for setting temperature at thermal bath as:

$$T_{\text{sample}} = 0.92 T_{\text{bath}} - 0.15 \quad (1)$$

The evaporation of water from cell and bath is the basic problem in this system. So, it is required to compensate the water periodically. An attached burette has been used to keep constant level of water in the cell. To start working by this set, It is essential a fully calibration in different temperatures; because we have to include deformation of cell parts as the effect of thermal and mechanical loading. For this purpose, it was used a steel sample with 60mm in diameter and 15mm in height as dummy sample. In each step, first, the temperature of system was adjusted and after equilibration, the loading started. For each step of loading and unloading, enough time to equilibrate deformation of system has been utilized.

Figure 2 shows the vertical displacement in micrometer versus applied stress in kPa for the mentioned sample at different temperatures. Usually, the target temperature in sample was selected and the temperature in bath adjusted. To calculate deformation of steel sample, it was used 200GPa for elastic modulus and $1.6 \times 10^{-5}/^\circ\text{C}$ for β as thermal expansion coefficient and this relation from elasticity:

$$\Delta_d = h \left(\frac{\sigma}{E} - \beta \Delta T \right) \quad (2)$$

where h is the height of sample, σ vertical stress, E Yong's modulus, and ΔT is the change of temperature. To calculate Δ_{cell} :

$$\Delta_{\text{cell}} = \Delta_{\text{measured}} - \Delta_d \quad (3)$$

And finally for usage in a real test with soil sample, it was used this relation:

$$\Delta_{\text{sample}} = \Delta_{\text{measured}} - \Delta_{\text{cell}} \quad (4)$$

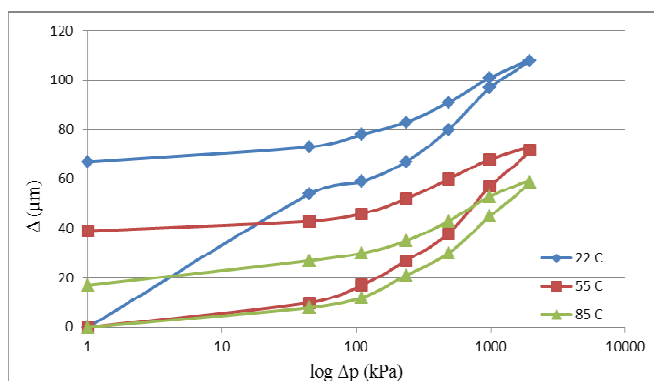


Fig. 2. The calibration of cell with temperature

3. Results

3.1. Free swelling tests

The first series of tests was free swelling of sand-bentonite in three temperatures: 25, 55 and 90°C . Figure 3 shows the evolution of swelling during saturation under a vertical seating load of 15kPa at different temperatures for samples of initial dry density $1.5\text{gr}/\text{cm}^3$. As it is seen, by increasing the temperature, the velocity to reach saturation state and stability in data is increased. The decrease of swelling capacity of the bentonite-sand mixture with the increase of temperature can be observed. From 25 to 90°C , decreasing about 20 percent in swelling strains was measured.

3.2. Oedometer tests

After saturation, the samples were loaded under oedometric conditions. Figure 4 shows the settlement of sample at 90°C . In low stresses, a little swelling during a long time is observed despite the fact of saturation in previous period. As it is expected, the behavior of this material is so time dependent and by a very low permeability, the required time for consolidation in each step is long but shorter than the corresponding time at 25°C .

The variation of void ratio due to vertical stress has been presented in Figure 5. Whereas the start point for consolidation after swelling was different, the vertical axis has been

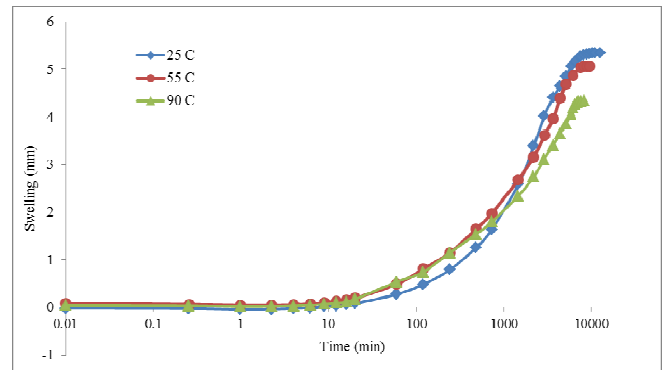


Fig. 3. The swelling of sample in oedometer at 15kPa and different temperatures

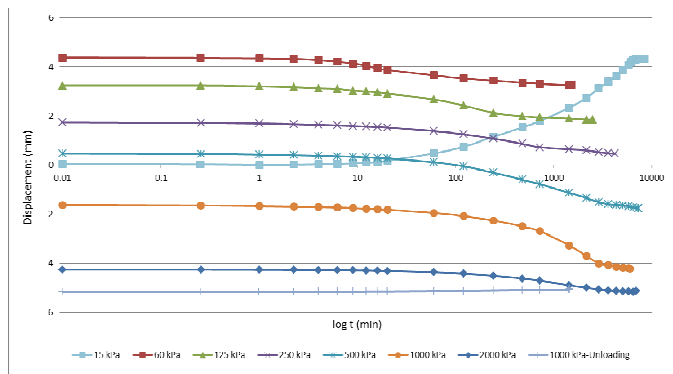


Fig. 4. The displacement of sample in oedometer at 90°C

normalized to facilitate comparison. From this figure, it is obvious that compressive strains increase at higher temperatures. As it is observed, when temperature increases the point of inflection has been changed to lower stresses. As a very important point, after heating the slope of line in loading has increased a little; however in unloading path for all temperatures, difference is negligible.

To analyze the effect of recent thermal history, in 1000kPa loading stage and 55°C temperature, after equalization, temperature was increased up to 90°C and then to 25°C and again returned to 55°C. In each step, equalization was achieved. Figure 6 zooms in void ratio variations. As it is seen in this stress-temperature path, point C is corresponding to 90°C and A to 25°C. By increasing of temperature in constant stress, sample contracts and by decreasing of heating, sample dilates. On the other hand, in figure 7 another path has been showed. This time, void ratio condition in 25, 55 and 90°C after equalization of temperature and then increasing of stress to 1000kPa have been compared. If we concentrate on void ratio variations, in the first state, from 90 to 25°C, only about 0.02 and in the second path about 0.2 means ten times were seen. For more explanation, for instance, at point C in both paths has similar stress-temperature state (1000kPa and 90°C) but how to reach to these points are different. Of course, it is obvious that mechanical loading paths are similar so what is responsible for difference is thermal history.

3.3. Atterberg's limits tests

Atterberg's limits are one of the most important indications to define plasticity characteristics of clays and their changing defines alteration in the microstructure of clays. Figure 8 shows the changes in Atterberg's limits of bentonite between room temperature up to 150°C. As it is seen from diagram, liquid limit (that is dependent to free water in clay structure) has been decreased about 10% in this range of temperature changing. As the result of low changing in plastic limit by heating, plasticity index has decreased in the same quantity. This decreasing can be attributed to reduction of space for water as the result of coming closer in clay pellets.

To investigate the effect of thermal history on plasticity index, the preheated samples up to 150°C, were cooled and then were heated again up to 85°C for 48 hours (one cycle) and again Atterberg's test was examined. This one-cycle process actually showed (fig. 9) that the sample has been preheated and these results are different from previous results in 85°C.

On the other words, when a bentonite sample is faced to a higher temperature, in lower temperatures is resistant and its behavior is similar to the preheating temperature (that means the highest temperature in the sample thermal history).

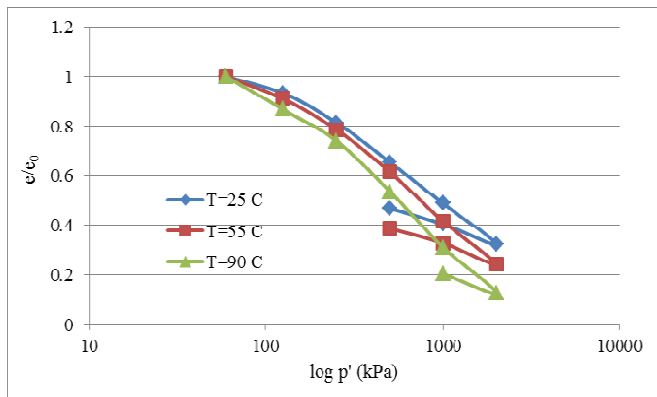


Fig. 5. The variation of normalized void ratio versus compressive stress at different temperatures

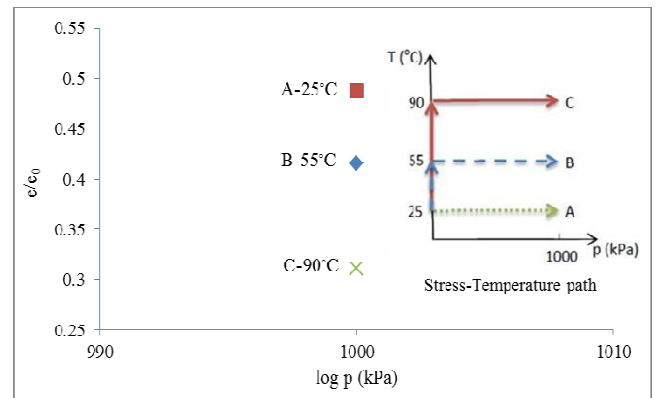


Fig. 7. The variation of normalized void ratio at constant temperature and 1000kPa

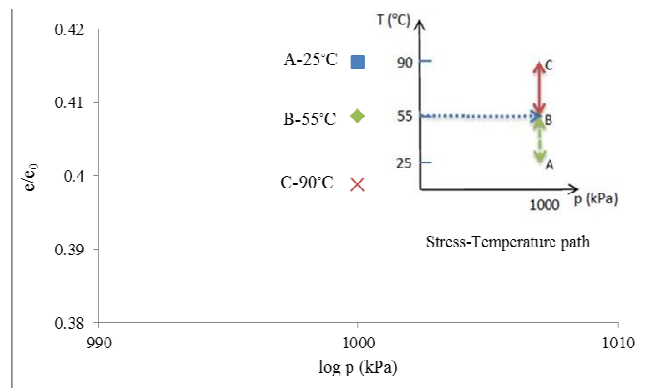


Fig. 6. The variation of normalized void ratio versus temperature at constant stress 1000kPa

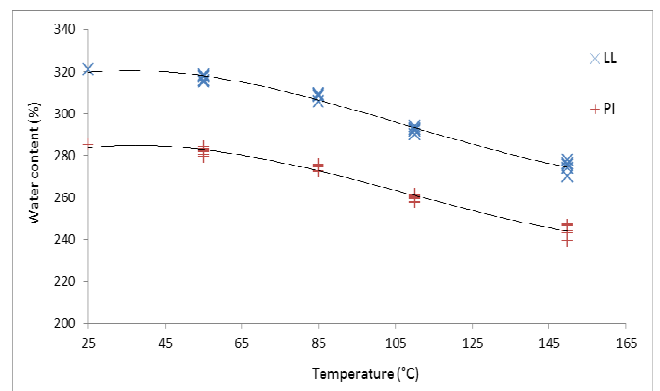


Fig. 8. The variation of liquid limit and plasticity index of Barit bentonite with temperature

4. Discussion

The induced heating by radioactive waste or other heater resources may change the mechanical behavior of soil that act as barrier or surrounding host formation. On the basis of foregoing results, high plastic soils such as bentonite-sand have big deformations in wetting and drying. This is due to the high surface activity of the bentonite-sand mixture reflected by its high plasticity ($W_L = 321\%$, $I_p = 285\%$). As a result of the high surface activity, time dependent behavior is dominant in this mixture and it does not behave like more usual soils. The swelling and consolidation behavior is due mainly to time-dependent creep mechanism rather than the more usual process of dissipation of excess pore water pressure.

Earlier investigations show that the increase in temperature induced insignificant increase in the swelling potential of bentonite (e.g. for Na-bentonite [10], for Ca-bentonite [11]). On the other hand, the reverse is also true (e.g. for Ca-bentonite [10], for Na-bentonite [12], and for FEBEX bentonite which is mainly Ca-Mg-bentonite [13]). From our results, elevated temperature decreases swelling strains in bentonite-sand mixture. It can be attributed to the effect of temperature on double layer state. An increase in temperature causes a decrease in surface potential for a constant surface charge [5].

So, the particles can come near to each other more than lower temperatures and the amount of absorbed water in the microstructure of the clay is smaller [14]. To prove, four X-ray tests were done at 25, 55, 110 and 150°C. Figure 10 compares their analysis diagrams. As can be seen in Fig. 10, by heating the sample, apparently the Montmorillonite peak at usual temperature ($d=12.4 \text{ \AA}$) disappeared, while a peak with different basal spacing is formed at different points. This changing means that basal spacing in Barit bentonite microstructure has been changed for instance to 10.6 \AA at 110°C . This reduction produces a smaller space to keep water.

A reduction in bentonite's plasticity index can demonstrate this effect, too. On the other hand, when a bentonite pellet is subjected to a high temperature and then cooled, some, but not all, of the deformation in structure is reversible. So, when the bentonite is reheated to lower than maximum temperature,

the effect of the sample's previous highest temperature is seen. The plasticity behavior of bentonite is shown in Figure 9.

In addition, high temperature increases soil permeability. Permeability depends inversely on the volume of adsorbed water held to the soil particles. Also, the viscosity of water decreases at high temperatures; thus, permeability increases. Morin and Silva [15], Habibagahi [16], Houston and Lin [17], Towhata et al [18] have all reported an increase in permeability at elevated temperatures due to reduction in water viscosity. Because of this fact, the speed of equalization in swelling and consolidation tests was slightly faster in higher temperatures.

The point of inflection in consolidation curves (called preconsolidation pressure) is considered the pseudoelastic limit, which separates "elastic" pre-yield from "plastic" post-yield behavior in oedometric conditions. Of course, in high plastic clays, this phenomenon is not as clear as with other clays [19]. Nevertheless, we can still evaluate it as an apparent preconsolidation pressure. By increasing temperature, this point has displaced to the lower stresses. Plastic deformation of clays corresponds to an increase in the number of mineral-to-mineral contacts, instead of mineral-to-water-to-mineral contacts. This is because water acts as an elastic material between two clay particles. The aforementioned reduction in adsorbed water of clay from higher temperatures facilitates formation of mineral-to-mineral contacts, thus globally producing plastic strains while reducing the curve's elastic domain.

The increased slope in loading (coefficient of compressibility) after heating (observed in the consolidation curves) means the compressibility rate of sand-bentonite increases. Meanwhile, in a defined stress, the corresponding void ratio in high temperatures is reduced as compressibility increases. The microstructural phenomenon stated above can clarify this observation too. In a bentonite-sand mixture, this behavior is different compared with other clays, such as Illite or Kaolinite, in that their compressibility rate (C_c) is independent from temperature. This is related to the point we described as a different mechanism (time-dependent) for consolidation in high plastic clays.

The behavior of sand-bentonite in a constant stress and making temperature up and down (figures 6 and 7) defines

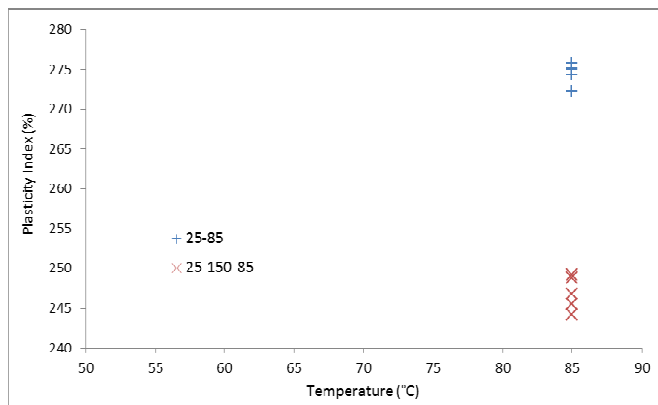


Fig. 9. The variation of PI for Barit bentonite in two thermal paths

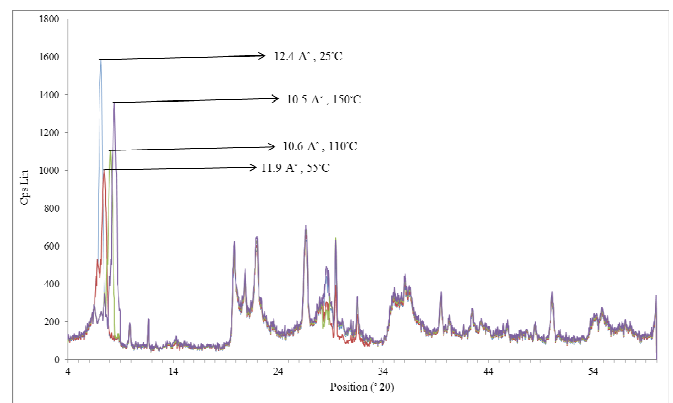


Fig. 10. The variation of basal spacing in Barit bentonite at different temperatures

that thermal loading does not have the same effect of mechanical loading in compression. The amount of volume change in thermal loading is so little; for this kind of loading, only expansion and contraction of water and clay skeleton acts; But at constant temperature, loading causes going out pore water and deformation of particles, pores and solid skeleton that produces bigger compressive volume change. This comparison confirms that thermal loading has quite different effect from mechanical loading on bentonite-sand behavior.

Overall, it can be remarked that elevation of temperature causes increasing compressibility both rate and quantity; however this changing is not significant. This effect has to be included for design of bentonite-sand blocks around nuclear waste canisters.

5. Conclusions

The swelling and compression behavior of sand-bentonite mixture has been detected by a modified thermal oedometer apparatus. Some of main results are:

- The elevation of temperature in bentonite-sand mixtures reduces free swelling potential and strains.
- The required time for equalization of swelling at high temperatures is less rather than low temperatures.
- An increase in temperature causes a decrease in basal spacing. So, the particles can come near to each other more than lower temperatures and the amount of absorbed water in the microstructure of the clay is smaller.
- The high temperature causes increasing in compressibility rate and quantities in bentonite-sand mixture.
- Elevation of temperature causes reduction in plasticity index of bentonite. After many cycles of heating and cooling, a little of this reduction comes back.
- Thermal history of soils is different from mechanical history at different temperatures. However the stress-temperature final state of two samples may be similar, but thermo-mechanical path will produce different results.
- The highest temperature that bentonite has been experienced will effect on its plasticity and mechanical behavior. So when we want to estimate the deformation of clay layers such as clay buffers, we have to consider its thermal history.

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