Teknik Dergi, 2022 12457-12472, Paper 690

Improvement of Shear Strength of Zeolite-Bentonite Liner Material under High Temperatures with Tincal and Pumice*

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ABSTRACT

Thermal changes (high temperature and thermal cycles) occur around energy structures, such as energy piles, nuclear waste repositories, etc. Sometimes these temperature changes affect the engineering properties of surrounding soils undesirably. Hence, there is a need for durable soils that can keep their engineering properties unchanged under high temperature and thermal cycles for a long time. Tincal and pumice are used in the production of temperature-resistant and heat-insulated materials. Therefore, in the present study, 10% and 20% tincal and pumice additives were added to zeolite-bentonite mixtures and the shear strength behavior of the mixtures was investigated under room and high temperature (80°C). According to the results, the maximum shear stress values of zeolite- bentonite mixtures generally increased in the presence of tincal and pumice additives under high temperature. Both additives are effective for improvement the effect of pumice can be more pronounced.

Keywords: Energy geo-structures, pumice, tincal, zeolite, bentonite, shear strength.

1. INTRODUCTION

There has been an enormous increase in the number of energy structures in last decades. Since energy structures are in a direct contact with soil, there has been an increase in the number of studies on the behavior of soils under high temperatures in recent years. The

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Note:

- This paper was received on August 31, 2020 and accepted for publication by the Editorial Board on April 13, 2021.
- Discussions on this paper will be accepted by November 30, 2022.

- https://doi.org/10.18400/tekderg.788422

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* A part of this study was presented at the 8th International Geotechnical Symposium. This article is an expanded version.

former studies have shown that the engineering properties of soils change depending on temperature. This change in engineering properties under high temperatures can often lead to cases that can damage energy structures, decrease their efficiency and affects human health and environment. Therefore, there is a need for the soils and/or soil mixtures that can maintain their properties unchanged at high temperatures or thermal cycles. A soil material that is durable to the thermal changes can be developed by help of thermally resistant materials such as boron, pumice, perlite, or fiber glass.

High temperatures affect the engineering properties of soils. The magnitude of this effect depends on density, water content, soil type, mineralogical and chemical composition [1]. The thermo-mechanical behavior of disturbed and natural clay soils was investigated using triaxial cell tests with controlled temperature (20°C to 60°C) [2]. In this study, it was found that the temperature increment caused change in the sample volume and the particles were rearranged. However, this volumetric deformation may be contraction or expansion. The change in deviator stresses due to high temperature and temperature cycling was investigated by Abuel-Naga et al. (2006) [3]. The water temperature in the triaxial cell was increased to 90°C, then tests were performed under this temperature. In terms of shear strength, higher peak shear strength values were obtained under high temperatures. Similar to this, previous studies show that the shear strength of clays increases with an increase in temperature. The change in the shear strength of clays with an increase in temperature is largely dependent on the volume change caused by high temperature [4]. The volume change that occurs with increase in temperature can be explained by the effects of high temperature on the interparticle forces and the viscous shear resistance of the adsorbed water, which affects the resistance of the clay particles to the fabric change [5]. Wang et al. (2017) reported that as the temperature increases, the viscosity of the adsorbed water decreases under the same consolidation stress and consequently decreases the porosity [6]. Accordingly, it can be concluded that the clays shrink in volume at high temperature. Hong et al. (2013) observed that thermal shrinkage increased the shear strength as a result of their study [4]. Although it is concluded that the shear strength of clays increases with high temperature, there is not much study on the behavior of soil mixtures at high temperature.

Bentonite, a montmorillonitic clay, often used as a liner, has low permeability and high plasticity. However, in cases where the water content decreases, bentonite shrinks and cracks occur. These cracks cause an increase in hydraulic conductivity and this situation will negatively affect the long-term performance of the liner. Therefore, the use of compacted clay is suitable in areas where temperature and humidity changes are insignificant [7]. Due to these adverse conditions when clay is used alone, sand-bentonite mixtures are widely used as barrier materials. The sand component increases the strength and the bentonite component fills the pore gap between the sand grains to reduce hydraulic conductivity [8]. Natural zeolites consist of acidic and volcanic tuffs that form rock-forming minerals [9]. Zeolites have ability to absorb smaller molecules and are therefore called "molecular sieves". It was observed that zeolite-bentonite mixtures were not affected by changes in water content, and also had very low hydraulic conductivity and low volumetric shrinkage potential, and these mixtures are proposed as an alternative to sand-bentonite mixtures [10]. Yukselen-Aksoy (2010) investigated the swelling potential, compressibility, hydraulic conductivity and shear strength behaviors of two different natural zeolites. In this study, it was found that zeolites did not have high compressibility and swelling potential and hydraulic conductivity was suitable for limitation of landfill liners [11]. Galvão et al. (2008) compared sand-bentonite and zeolite-bentonite compression characteristics, volumetric shrinkage strain and hydraulic conductivity. In this study, it was reported that zeolite-bentonite was a good material for regular liner material [12]. The use of zeolite bentonite mixtures in waste storage is more advantageous than sand-bentonite mixtures thanks to the high adsorption property of zeolite [13].

Boron minerals and pumice are two of the materials commonly used in industry for various areas like temperature resistance and heat insulation. Pumice which is used as an additive, is a volcanic rock formed during volcanic eruptions [14]. It is found in two different forms as acidic and basic in nature. Pumice that consists of Si, Al, K, Na and Fe oxides, is also contains a small amount of Ca, Mg, Mn and Ti oxides [15-17] (Table 1). Pumice is used in different fields such as construction, textile, agriculture, chemistry, filtration and brick-ceramic production. Boron is found very commonly in the earth's crust. The boron is present in the minerals form in nature by bonding with oxygen. One of the most widely used boron minerals in the industry is tincal. Tincal is very rich in terms of sodium minerals ($Na_2B_4O_7.10H_2O$). Boron minerals are used in the various areas such as; energy, metallurgy, cement, glass and ceramics in order to reduce the thermal expansion of glasses and protect the glass from acid and scratches. In addition, Alpaydın (2019) reported an increase in the shear strength of tincal added sand-bentonite mixtures under high temperature (80°C) [16].

Due to such superior thermal properties of tincal and pumice, the motivation behind of the present study is improvement of engineering properties of zeolite-bentonite mixtures with these additives under high temperatures. The aim of this study is to investigate the change of the shear strength parameters of 10B-90Z and 20B-80Z mixtures under high temperature (80°C) with 10% and 20% pumice and tincal additives. In this way, durable soil material may be developed with the addition of tincal and pumice against high temperature.

Table 1 - Chemical composition of pumice [17]

	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	Na_2O
%	48.37	12.49	8.07	1.78	8.43	9.58	4.63

2. MATERIAL CHARACTERIZATION AND TEST METHODS

2.1. Material Characterization

In this study, zeolite-bentonite mixtures, pumice and as a boron mineral tincal were used. Bentonite was gathered from Esan Eczacıbaşı Industrial Raw Materials Industry and Trade Inc. and zeolite was obtained from Rota Mining Inc.. The X-Ray analyses of bentonite and zeolite were performed with Rigaku D/Max-2200/PC X-Ray diffractometer (XRD) and Thermo Scientific ARL X'TRA devices, respectively. XRD data of powders were recorded with a 2°/min scanning speed in the range of $20<2\theta<80^o$. The analyses were performed with Win XRD software. After the analyses data were received, the relevant quantitative crystallographic information via Material Analysis Using Diffraction (MAUD) software, based on Rietveld refinement was used. The analyses results are shown in Figure 1. According to the results of the analyses, the bentonite consists of mostly quartz (SiO_2), montmorillonite $Na_{0.3}(Al,Mg)_2Si_4O_4(OH)_24H_2O$ and illite (Figure 1a). Clinoptilotile, silica

(SiO_2), silicon oxide hydrate, sodium tecto-alumosilicate hydroxide, and silicon aluminum oxide were obtained in the zeolite sample (Figure 1b).

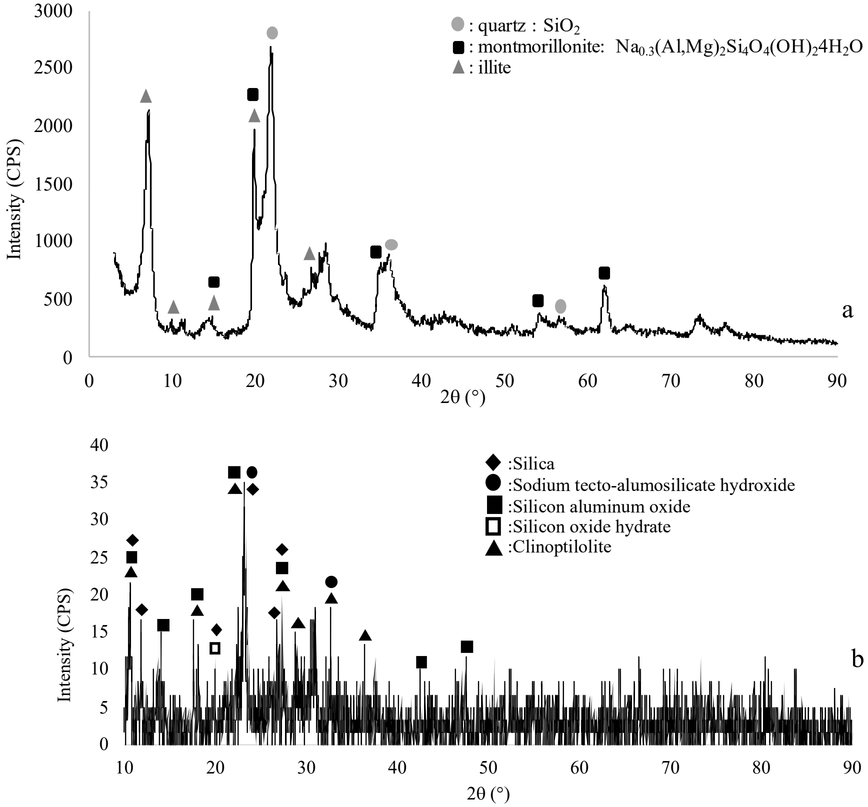


Figure 1 - XRD analysis results a) bentonite, b) zeolite

Pumice sample used as additive was gathered Pumice Export Mining Industry Inc. and tincal was obtained from Eti Mining Operations General Directorate. The grain size distribution of pumice is given in Figure 2. Because of the crystal structure of tincal, this experiment could not be performed for tincal.

According to XRD analyses, silicate and sodium-aluminum silicate compounds were determined in the pumice. Tincalconite, borax, jadeite and silica contents were observed in the tincal mineral.

The structure of the additives used in this study was observed using scanning electron microscopy (SEM). SEM analyses were performed using the COXEM EM-30 Plus device. Physico-chemical properties of all materials used in this study are given in Table 2. For the tests, zeolite and bentonite samples were prepared by drying in an oven (105°C) for 24 h. Pumice and tincal samples were used by sieving through No.40 sieve. Natural water contents of pumice and tincal were determined before the tests and calculations were made by taking these data into consideration. While giving the name to the mixtures, bentonite was abbreviated as "B", zeolite "Z", pumice "P" and tincal "T". For example, the mixture of 18B-72Z-10P contains 10% pumice, 18% bentonite, 72% zeolite. While preparing the mixtures, 10% or 20% of the total weight of the dry mixture was taken as tincal or pumice, 10% or 20% of the remaining amount was bentonite and the rest of was zeolite. The material ratios in the sample were chosen such that bentonite / bentonite + zeolite ratios (B / B + Z) were 10% and 20%. For example, the B / B + Z ratio for an 9B-81Z-10T mixture was 0.10, while it was 0.20 for a 18B-72Z-10T mixture.

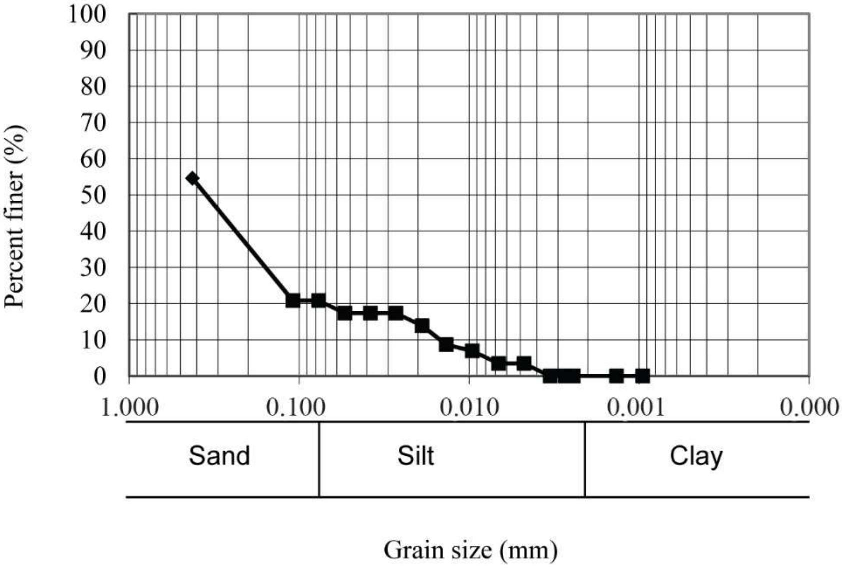


Figure 2 - Grain size distribution of pumice

Table 2 - Physico-chemical characteristics of all materials

	Bentonite	Zeolite	Pumice	Tincal
Specific gravity	2.70	2.30	2.50	1.67
Liquid limit (%)	476.0	42.0	37.1	60.5
Plastic limit (%)	70.1	NP	NP	42.9
pH	9.50	8.50	8.86	9.12

2.2. Test Methods

2.2.1. Compaction Tests

In this study, compaction tests were performed to obtain compaction parameters to be used in sample preparation for the direct shear tests of 10B-90Z and 20B-80Z mixtures in the presence of 10% and 20% pumice and tincal additives. In addition, the changes in the compaction parameters of zeolite-bentonite mixtures with additives were determined. The compaction tests were performed according to ASTM D-698 (2012) standard [18]. While preparing the samples, the natural water content of pumice and tincal was taken into consideration for the calculations. The dry materials were mixed in a bowl with scoop. After mixing the dry materials homogeneously, these samples were prepared at four different water contents. The water was added by spraying method. The prepared samples were kept closed for 24 h in a plastic bag. After the tests were carried out, the samples were kept in an oven to determine the final water content. The Standard Proctor tests were and maximum dry unit weight (γ_{dmax}) and optimum water content (w_{opt}) values were determined.

2.2.2. Direct Shear Tests

In this study, the direct shear tests were performed according to ASTM D3080 (2018) standard [19]. These tests were carried out for 10B-90Z and 20B-80Z mixtures in the presence of 10% and 20% pumice and tincal at room temperature and 80°C. The samples were prepared at the $w_{opt}+2\%$ and γ_d values corresponding obtained from Standard Proctor tests. The samples were placed in 6x6 cm^2 mold by compacting in three layers. The samples were placed as submerged condition in a tray filled with water for 24 h and 25 kPa vertical pressure was applied on the mold to prevent swelling. At the end of this period, the tests were performed under three different normal stresses (49, 98, 196 kPa). In the tests at 80°C, the temperature increment was provided with a specially designed heat rod for the shear box. The temperature of the water in the cell was kept around 80°C by using a thermostat. When the water temperature increased, the thermostat deactivated the heat rod, and when it decreased, it activated the heat rod to ensure that the water temperature

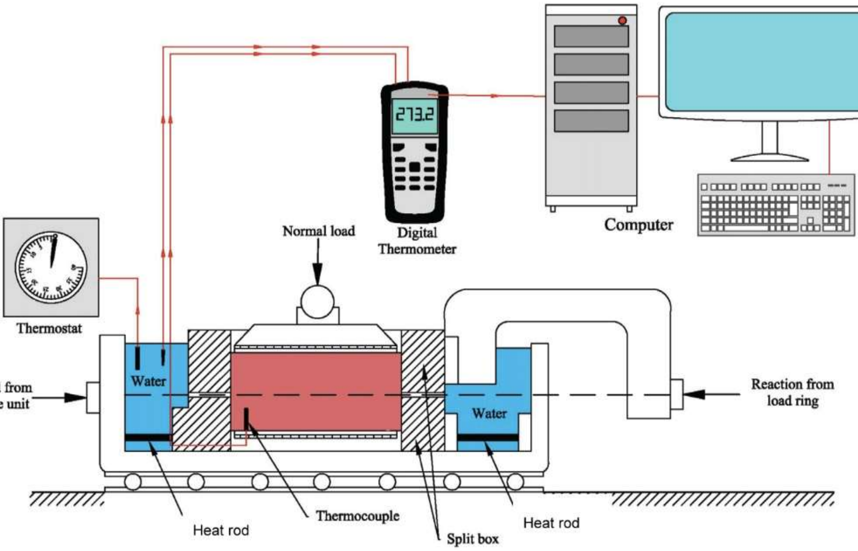


Figure 3 - Schematic presentation of the test set-up

remained at adjusted level. During the tests, the temperature of the sample and water were measured with K-type thermocouples. Measured values were recorded by digital thermometer. The schematic presentation of the test set-up is given in Figure 3.

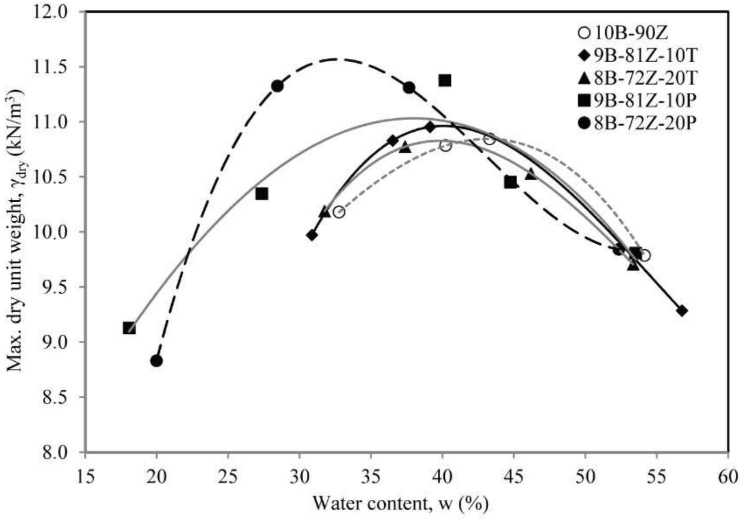
At the end of the tests, the sample was left in the oven (105°C) to determine the water content. The shear strength parameters (internal friction angle, ϕ'; cohesion, c'), maximum shear stress (τ_{max}) of the sample were determined with the data obtained during the tests.

3. RESULTS AND DISCUSSION

3.1. Compaction Test Results

The changes in the compaction parameters of 10B-90Z and 20B-80Z mixtures in the presence of 10% and 20% pumice and tincal were determined and the results are given in Figure 4.

As shown in Figure 4, when 10% pumice was added to 10B-90Z mixture, max. dry unit weight did not change significantly, while 20% pumice additive increased the max. dry unit weight. It should be noted that the specific gravity value of pumice is 2.5 which is higher than zeolite (2.3) and lower than bentonite (2.7). Whereas the optimum water content of this mixture decreased with 20% pumice, it increased slightly with 10% pumice additive. When additive was added to the mixture the bentonite content decreased. The liquid limit values of the bentonite, pumice and tincal are 476%, 37.1% and 60.5%, respectively. For that reason, when bentonite content decreased, the optimum water content values decreased. For 20B-80Z mixture, 10% and 20% pumice additive increased the max. dry unit weight values, while the optimum water content showed a tendency as in the 10B-90Z mixture.



(a)

Figure 4 - Compaction curves of pumice and tincal added zeolite-bentonite mixtures a)10B-90Z mixtures, b)20B-80Z mixtures

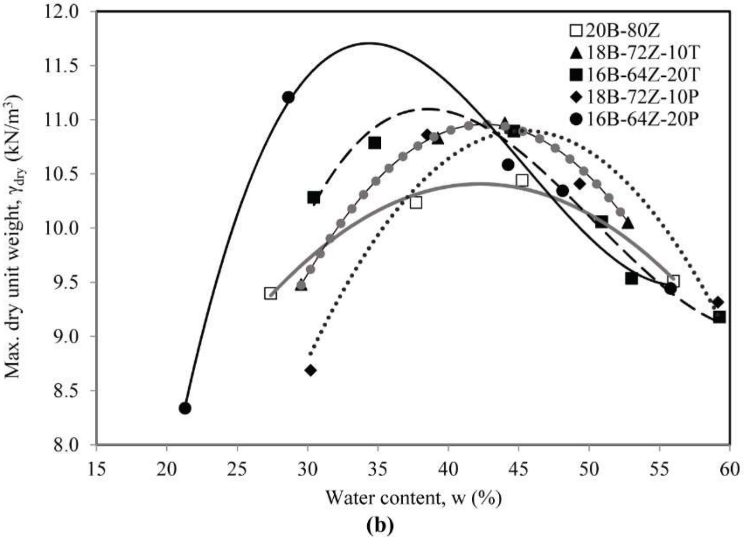


Figure 4 - Compaction curves of pumice and tincal added zeolite-bentonite mixtures a)10B-90Z mixtures, b)20B-80Z mixtures (continue)

As shown in Figure 4a, when tincal added to the 10B-90Z mixture, there was no significant change in the $\gamma_{d,max}$ value, while the w_{opt} value decreased slightly. This decrease was around 3.5% and did not change with increasing tincal additive from 10% to 20%. According to the results given in Figure 4b, when 10% tincal was added to the 20B-80Z mixture, while the $\gamma_{d,max}$ value increased by 0.4 kN/m^3, there was no significant change in the w_{opt} value. When the tincal percentage was increased to 20%, the $\gamma_{d,max}$ value increased slightly (0.3 kN/m^3) while the w_{opt} value decreased.

3.2. Direct Shear Test Results

The change in the shear strength parameters of 10B-90Z and 20B-80Z mixtures in the presence of 10% and 20% pumice and tincal additives was determined at room temperature and 80°C. The shear strength parameters are given in Table 3 and failure envelopes are shown in Figure 5.

The angle of internal friction value of 10B-90Z was determined as 30.9°. The angle of internal friction value of pure zeolite is around 34-44° [11, 20]. According to the direct shear test results, when the temperature increased, the internal friction angle of the 10B-90Z mixture increased from 30.9° to 32.0°, and the cohesion value increased from 5.2 to 9.8 kPa. With increasing temperature, the internal friction angle of the 20B-80Z mixture increased by 5.2° and reached to 27.9°.

When additives (tincal and pumice) were added to the 10B-90Z mixture at room temperature, the internal friction angle and cohesion values generally increased. So much as 20% pumice additive, the internal friction angle value of 10B-90Z mixture increased from 31° to 36°. For

20B-80Z mixtures, while the internal friction angle generally decreased at room temperature, cohesion values increased. As an exception, an inverse trend was observed for the 18B-72Z-10P mixture (i.e. the internal friction angle increased while the cohesion value decreased). The internal friction angle value (32°) of the 10B-90Z mixture at 80°C temperature increased in the presence of 10% additives (tincal-pumice), while it decreased in the presence of 20% additives. The cohesion value of the 10B-90Z mixture at 80°C increased in the presence of pumice and tincal just like at room temperature. The internal friction angle of the 20B-80Z mixture increased in the presence of pumice, but decreased in the presence of tincal at 80°C. The cohesion values of 20B-80Z mixture also increased with additives at 80°C as in general. In addition, the effect of temperature increment on the internal friction angle of 10B-90Z mixtures was not as much as that of 20B-80Z mixtures.

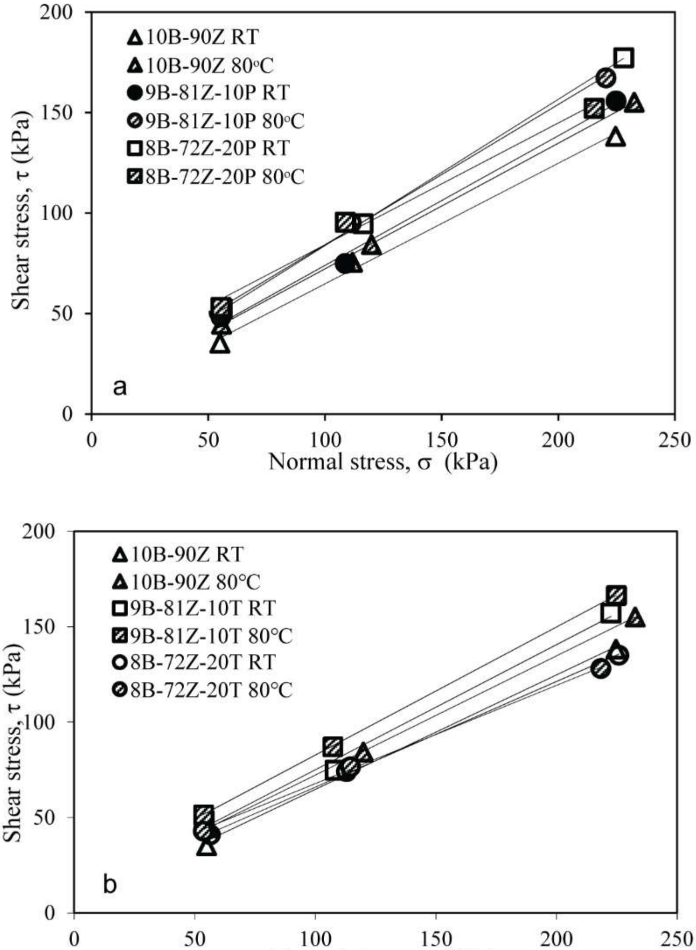


Figure 5 - Shear failure envelopes of 10B-90Z mixtures at room temperature and 80°C in the presence of a) pumice, b) tincal

Table 3 - Shear strength parameters of samples

Samples	Room Temperature		80°C	
	ϕ' (°)	c' (kPa)	ϕ' (°)	c' (kPa)
100 P	36.8	29.6	37.3	13.1
100 T	15.6	40.6	*	*
10B-90Z	30.9	5.2	32.0	9.8
9B-81Z-10P	32.9	9.5	35.0	14.4
8B-72Z-20P	36.1	11.0	31.5	23.6
9B-81Z-10T	33.3	9.5	33.9	15.6
8B-72Z-20T	29.0	10.6	27.2	16.3
20B-80Z	22.7	18.3	27.9	15.4
18B-72Z-10P	25.5	15.1	28.7	12.4
16B-64Z-20P	21.4	36.0	31.5	16.8
18B-72Z-10T	20.5	25.3	22.7	23.4
16B-64Z-20T	18.7	30.8	25.0	20.3

*: Tincal collapsed when it was heated to 80℃

The maximum shear stress values of bentonite-zeolite mixtures in the presence of pumice and tincal at room temperature and 80°C are given in Table 4. According to Table 4, when the temperature of additive-free zeolite-bentonite mixtures was increased, an increase up to about 10 kPa in maximum shear strength values was observed.

The results show that the shear strength of 10B-90Z and 20B-80Z mixtures were not affected negatively much by the additives at both temperatures. However, there was an increase up to 20 kPa in the maximum shear strength in the presence of 20% pumice. However, when the temperature of 20% pumice mixtures (8B-72Z-20P and 16B-64Z-20P) was increased to 80°C, the change in maximum shear strength was almost negligible. The effect of additives other than this on the shear strength was either insignificant or negative at room temperature. In addition to this, another remarkable point was that the maximum shear strength of all mixtures increased only with the effect of temperature increment, while only 20% pumice added mixtures decreased slightly. It can be concluded that the improvement in the shear strength of the mixture obtained by pumice additive occurred independently of the temperature increment.

The maximum shear stress values of bentonite-zeolite mixtures in the presence of additives at both temperatures are presented in Figure 6. As can be seen from this figure, there was generally an improvement in maximum shear stress for 20B-80Z mixtures at 80°C, and this improvement also valid for 10B-90Z mixtures.

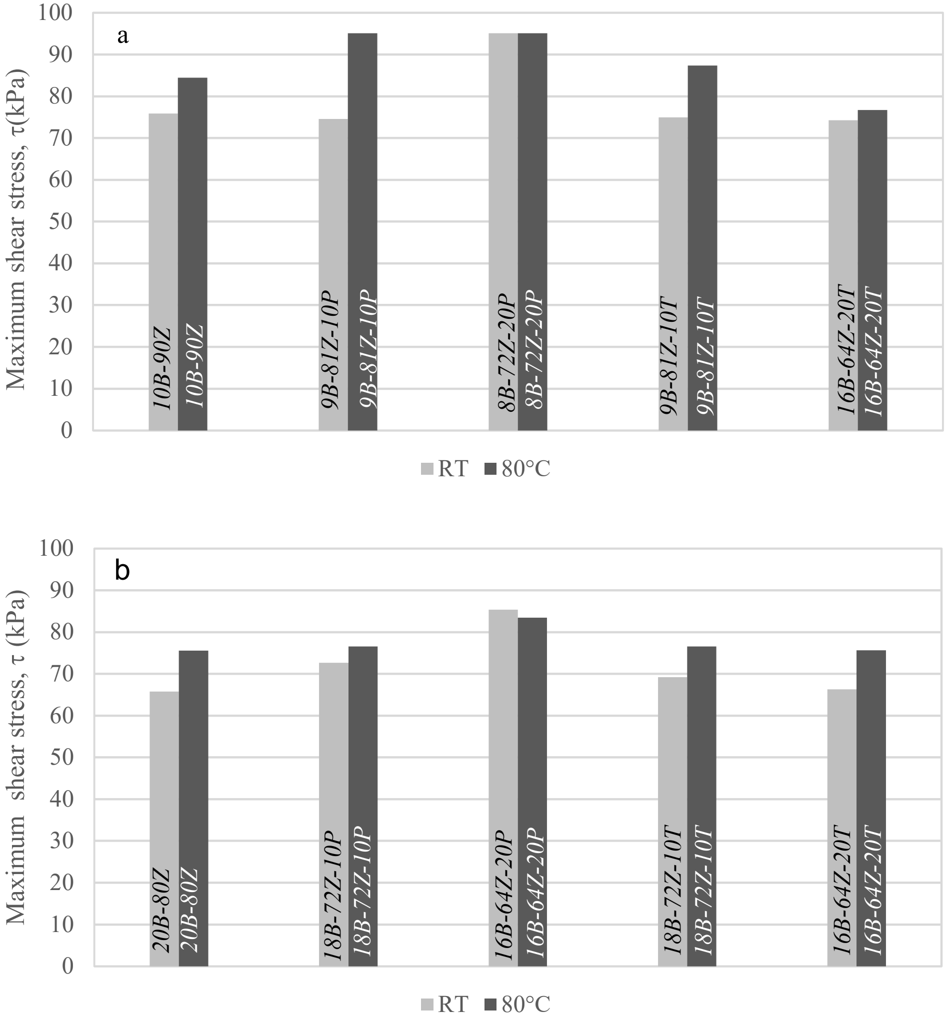


Figure 6 - Maximum shear stress a) 10B-90Z mixtures at room temperature and 80°C, b) 20B-80Z mixtures at room temperature and 80°C

The results have shown that, when temperature increased the maximum shear stress of additive-free zeolite-bentonite mixtures increased slightly. Moreover, generally the addition of tincal and pumice increased the τ_{max} more. In the presence of 20% pumice the maximum shear strength value of 20B-80Z mixture increased from 75.5 to 83.4 kPa under 80℃. However, the effect of tincal on the τ_{max} was is insignificant. Previous studies have shown that the increase in temperature transforms the smectite mineral into more constant silicate phases. Smectite transforms to illite depending on temperature, time and K^+ pore-water doping [21]. As a result of this water retention capability of bentonite decreases. If the ability of water adsorption of clay decreases, the strength of the clay should increase. In terms of zeolite, most zeolites undergo dehydration-induced reductions in volume. Clinoptilolite can undergo a large, very anisotropic decrease in unit-cell dimensions [22]. The water retention capability decreased in bentonite and volume reduction in zeolite caused an increase in shear strength of the zeolite-bentonite mixture by temperature increase. The results of this study confirm this behavior as well. The maximum shear strength value of 10B-90Z and 20B-80Z increased from 75.8 to 84.4 kPa, from 65.7 to 75.5 kPa, respectively. Previous studies showed that at a given water content the suction decreased with increasing temperature [23]. Moreover, as suction decreases, the shear strength increases. Hence, temperature increment causes an increase in the shear strength.

Table 4 - Maximum shear stress of all mixtures at both temperatures

Samples	Maximum Shear Stress (kPa)	
	Room Temperature (~25°C)	80°C
100P	127.5	87.3
100T	70.2	*
10B-90Z	75.8	84.4
9B-81Z-10P	74.5	95.1
8B-72Z-20P	95.1	95.1
9B-81Z-10T	74.9	87.3
8B-72Z-20T	74.2	76.7
20B-80Z	65.7	75.5
18B-72Z-10P	72.6	76.5
16B-64Z-20P	85.3	83.4
18B-72Z-10T	69.2	76.5
16B-64Z-20T	66.3	75.6

*: Tincal collapsed when it was heated to 80°C

The results of this study have shown that the shear strength of both zeolite-bentonite mixtures increased in the presence of tincal. The tincal is held strongly by the aluminum or silicon tetrahedron portion in the clay structure [24]. For that reason, the tincal was held by bentonite particles. Eventually the shear strength of zeolite-bentonite increased as a result of both tincal adsorption and material replacement. Because when the tincal added to the sample both bentonite and zeolite content was decreased. The bentonite material was replaced with tincal mineral which has very high thermal resistivity and very low thermal expansion. For that reason, the material replacement may be another reason for the increase in the shear strength of zeolite-bentonite mixtures under high temperature.

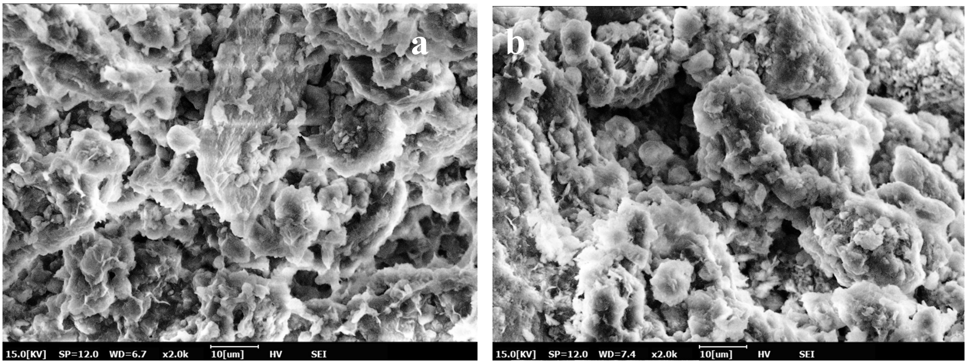


Figure 7 - Scanning electron microscope photographs of 20B-80Z mixture (x2000) at a) room temperature, b) 80°C

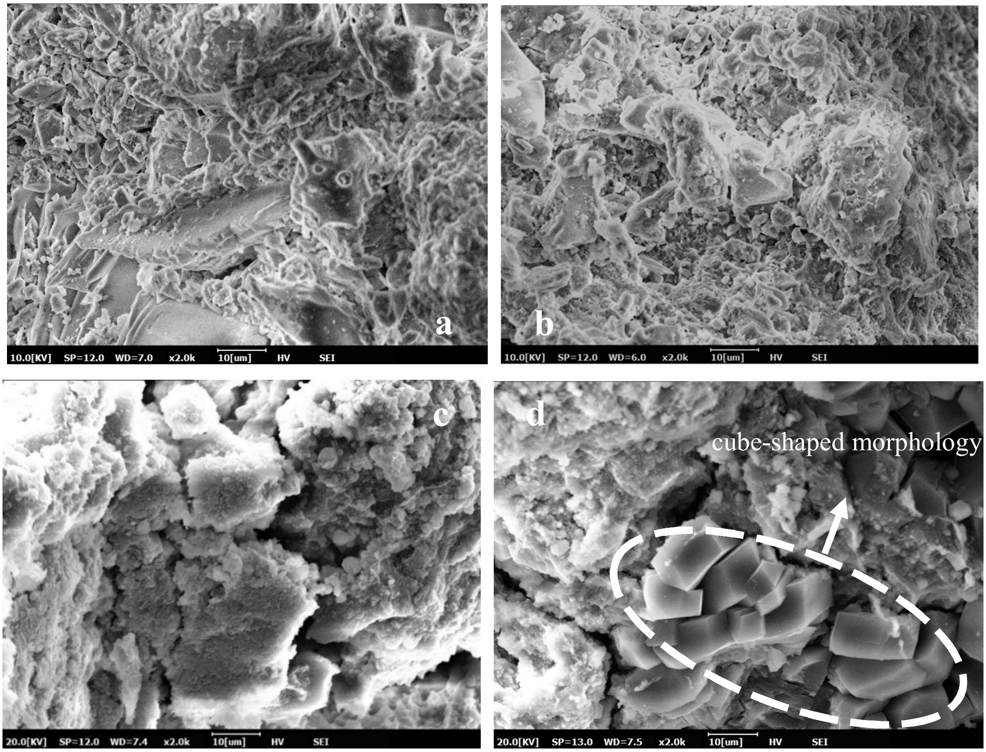


Figure 8 - Scanning electron microscope photographs of additives (x2000) a) pumice (RT), b) pumice (80°C), c) tincal (RT), d) tincal (80°C)

The SEM photos of the additive-free 20B-80Z mixtures at room temperature and 80℃ are given in Figure 7. It was observed that the structure of the mixture almost same between the two SEM photos. This observation supports evaluated slight changes in the shear strength parameters. The SEM photos of the additive materials (i.e. pumice and tincal) at room temperature and 80℃ are given in Figure 8. The SEM photos of the pumice were taken at low voltage for best view. When structural view was compared between room temperature and 80℃, there was no change between these photos. However; the maximum shear stress value decreased when temperature increased. The direct shear test on the tincal collapsed under 80℃. However; at the SEM photo different structure like cubical particles clearly was observed under high temperature. According to the X-Ray analysis jadeite was found in the tincal sample and this cubical form belongs to jadeite mineral.

4. CONCLUSIONS

In this study, boron mineral (tincal) and pumice additives were used in order to improve engineering properties of zeolite-bentonite mixtures under high temperatures. The compaction and shear strength behavior of 10B-90Z and 20B-80Z mixtures were determined in the presence of 10% and 20% pumice and tincal. In addition, the direct shear tests were performed at 80°C and compared with the results at room temperature. According to the compaction test results, when pumice and tincal were added to the 10B-90Z mixture, the optimum water content generally decreased, while the maximum dry unit weight did not change much in the presence of tincal but increased in the presence of pumice. The maximum dry unit weight values of 20B-80Z mixtures generally increased with both additives. According to the direct shear test results, when pumice was added to zeolite-bentonite mixtures, the internal friction angles generally increased at 80°C. On the other hand, tincal additive generally decreased the angle of internal friction and increased cohesion. The highest internal friction angle values of zeolite-bentonite mixtures were obtained in the presence of pumice additive at both temperatures. Also, the shear strength of additive-free 10B-90Z and 20B-80Z mixtures increased under high temperature, when compared with the room temperature. However, this increment was more in the presence of pumice (10% and 20%) and tincal (10%). According to the results obtained from the present study, pumice and tincal additives may be used to increase the thermal durability of zeolite-bentonite mixtures at high temperatures. Considering the results of the present study, pumice and tincal additives increased the shear strength of zeolite-bentonite mixtures under high temperatures. The effect of pumice was slightly higher than that of tincal, however; both additives have positive effects. These mixtures have a good potential to be used as thermally durable materials, with additional research required in this regard. In future studies the hydraulic conductivity and volume deformation behavior in the presence of pumice and tincal should be investigated. In terms of shear strength with tincal and pumice additives promising results were observed. Moreover; in landfills and nuclear waste repository liners these additives are candidate additives in order to improve engineering properties of natural soil mixtures under high temperatures.

Symbols

$\gamma_{d,max}$	is the maximum dry unit weight
$w_{opt.}$	is the optimum water content
ϕ'	is the internal friction angle

c' is the cohesion

τ_{max} is the maximum shear stress

Acknowledgements

The authors greatly acknowledged the support of TÜBİTAK (Grant no:217M553).

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