

Investigation of the influence of temperature on water retention properties of Czech Bentonite B75

Étude de l'influence de la température sur les propriétés de rétention d'eau de la bentonite tchèque B75

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ABSTRACT: Bentonite materials are considered as backfill material and engineered barrier material in high level nuclear waste disposal. In the basic framework of high level nuclear waste management all around the world, the bentonite material near the canister will be heated by the spent nuclear fuel, while the bentonite material on the opposite side will absorb water from the hosting rock. Bentonite will be therefore exposed to both drying and wetting and the influence of temperature on the retention properties is an important aspect of evaluation of the material. In this paper, an experiment was carried out to evaluate the retention properties of Czech compacted bentonite B75 with two different initial dry densities in the range 20-80 °C. Vapor equilibrium method was used to control suction. Results show that with increasing temperature water retention capacity decreased. This effect is more obvious in lower suction range. The temperature has more significant effect on drying path than wetting path for both dry densities.

RÉSUMÉ: Les matériaux à base de bentonite sont considérés comme des matériaux de remblayage et des matériaux de protection techniques lors de l'élimination de déchets nucléaires de haute activité. Dans le cadre fondamental de la gestion des déchets nucléaires de haute activité dans le monde entier, le combustible nucléaire irradié chauffera la bentonite située à proximité du conteneur, tandis que la bentonite située du côté opposé absorbera l'eau des roches hôtes. La bentonite sera donc exposée à la fois au séchage et au mouillage et l'influence de la température sur les propriétés de rétention est un aspect important de l'évaluation du matériau. Dans cet article, une expérience a été réalisée pour évaluer les propriétés de rétention de la bentonite B75 compactée tchèque avec deux densités sèches initiales différentes dans la plage de 20 à 80 ° C. La méthode

d'équilibre de vapeur a été utilisée pour contrôler l'aspiration. Les résultats montrent que, avec l'augmentation de la température, la capacité de rétention d'eau a diminué. Cet effet est plus évident dans la plage d'aspiration inférieure. La température a un effet plus significatif sur le trajet de séchage que sur le trajet de mouillage pour les deux densités sèches.

Keywords: temperature; bentonite; water retention properties; vapor equilibrium method

1 INTRODUCTION

Bentonite is clay with high montmorillonite content, which has a high swelling potential, high cation exchange capacity and low permeability. The main function of the bentonite barrier in high level nuclear waste disposal is to isolate the canister with spent fuel from the geological environment. The assessment of thermo-hydro-mechanical properties of bentonite is important to ensure long term safety operation of the repository. Water retention capacity of compacted bentonite is one of the key characteristics of its hydro-mechanical behavior. However, the retention capacity of the barrier is influenced by radiogenic heat released from the waste container, which propagates to bentonite. The influence of thermal properties of many bentonites have been studied by various researchers through the world in past years, such as MX80 bentonite (Johansson et al., 2007) which is considered to use in Swedish repository, FEBEX bentonite (Villar and Lloret, 2004) considered in Spanish project or GMZ bentonite considered in China (Xu et al., 2016).

In the present paper, the authors focus on the Czech bentonite B75. Several tests have been performed to investigate the influence of temperature on water retention curves of compacted

bentonite under unconfined conditions. The retention curves were studied at constant temperatures of 20 °C, 40 °C, 60 °C and 80 °C respectively. The initial dry densities of compacted bentonite was 1.27 g/cm³ and 1.9 g/cm³ respectively.

2 MATERIALS AND METHODS

The commercial Czech Ca-Mg bentonite extracted from the Cerny vrch deposit (north-western region of the Czech Republic), known as B75 was used in this study. The montmorillonite content of the bentonite is about 60%. The plastic limit, liquid limit and specific density of solid particles were 65%, 229%, and 2.87, respectively. More information such as cation exchange capacity, chemical composition can be found in Sun et al., (2018a). The compacted bentonite was prepared from the bentonite powder with the initial water content around 10%. The required weight of bentonite powder was calculated based on the desired initial dry density, dimensions of compacted samples and the initial water content.

Vapour equilibrium method described in Delage et al. (1998) was applied to control the suction. The relative humidity in the sealed desiccator was controlled by eight different saturated

chemical solutions. The total suction and relative humidity follows Kelvin's law:

$$S_t = \left(\frac{RT\rho_w}{\omega} \right) * \ln\left(\frac{1}{RH}\right) \quad (1)$$

Where S_t is the total suction (kPa); R is the molar gas constant, which equals to 8.314462 J/(mol K); T is the absolute temperature (K); ρ_w is the density of water; ω is the molecular mass of water vapour, which equals to 18.016 g/mol; RH is relative humidity of the system, which is defined as the ratio of partial pressure of vapour over saturation vapour pressure. The relative humidity of each saturated solution at different temperature is listed in Table 1.

The samples of compacted bentonite were splitted into two groups. One group was put in K_2SO_4 desiccator and equilibrated at 20 °C. Then the samples were moved to other seven desiccators to evaluate drying path of the retention curve. Another group was placed in the oven and the samples were dried at 105 °C for 24 hours, which corresponds to a total suction of approximately 1000 MPa (Sun et al., 2018b). Then the samples were placed in desiccators for determination of wetting path as shown in Figure 1. The tests at 20 °C were performed in the air conditioned laboratory with constant temperature. Tests at increased temperatures were performed in temperature controlled box. The typical time of equilibration was 2-3 months. After equilibration, the water content was determined by oven drying.

Table 1. Relative humidities imposed by saturated saline solutions for vapor equilibrium method (adopted from Greenspan (1977), OIML(1996), Tang and Cui (2005)).

Chemical Solution	Relative humidity			
	20 °C	40 °C	60 °C	80 °C
Temperature	20 °C	40 °C	60 °C	80 °C
LiCl·H ₂ O	11.31	11.21	10.95	10.51
CH ₃ COOK	23.11	-	-	-
MgCl ₂ ·6H ₂ O	33.07	31.6	29.26	26.05
K ₂ CO ₃	43.16	-	-	-
NaBr	59.1	53.2	49.7	51.4
NaCl	75.47	74.68	74.5	76.29
KCl	85.1	82.3	80.3	78.9
K ₂ SO ₄	97.2	96.5	96.2	95.5

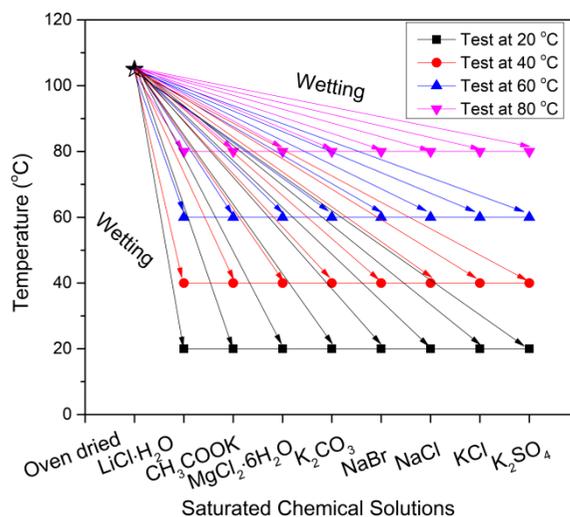


Figure 1. Wetting paths under different temperatures.

3 RESULTS AND DISCUSSION

3.1 Water retention curves at 20 °C

In order to check the state of equilibrium of bentonite with vapor water in each desiccator, the weight of the bentonite was regularly measured

until the sample mass stabilized. Figure 2 shows the change of water content with time for wetting path of 1.27 g/cm^3 samples. The water content increased sharply during the first week, then increased smoothly until the equilibrium state was reached. Except of the suction of 3.29 MPa, where equilibration took significantly longer, all other samples were stabilized in 1-2 months.

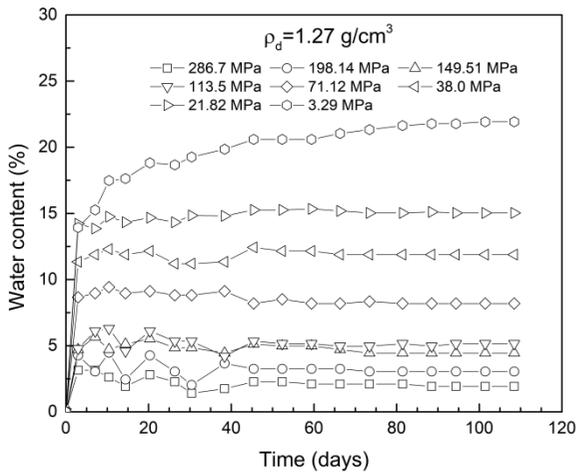


Figure 2. Example of water content evolution in WRC measurements by vapour equilibrium method for initial dry density of 1.27 g/cm^3 (Sun et al., 2018a).

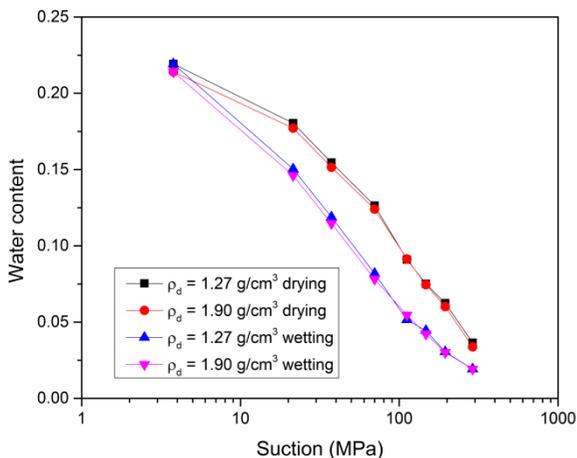
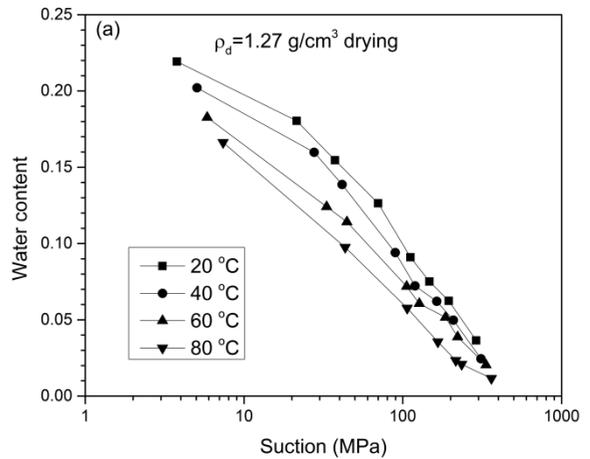


Figure 3. Water content versus suction for water retention curve measurements under wetting and drying path at both dry densities at $20 \text{ }^\circ\text{C}$

Figure 3 shows the water retention curves of samples at two initial dry densities (wetting and drying paths). It can be seen that the initial dry density had little influence on the water content, which increases almost linearly with logarithm of suction. The effect of initial dry density was observed when suction was plotted against void ratio (see Sun et al., 2018a).

3.2 Influence of temperature on drying and wetting path

The water retention curves of two dry densities of 1.27 g/cm^3 and 1.90 g/cm^3 at $20 \text{ }^\circ\text{C}$, $40 \text{ }^\circ\text{C}$, $60 \text{ }^\circ\text{C}$, $80 \text{ }^\circ\text{C}$ are shown in Figure 4 (a-d) respectively.



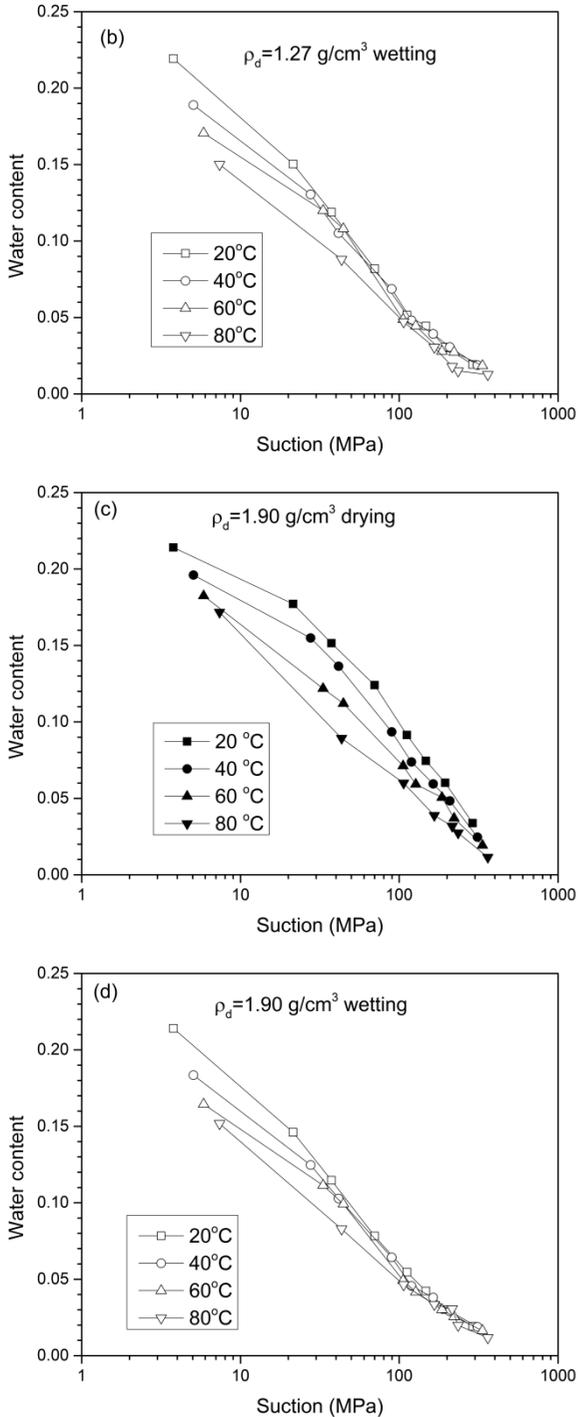
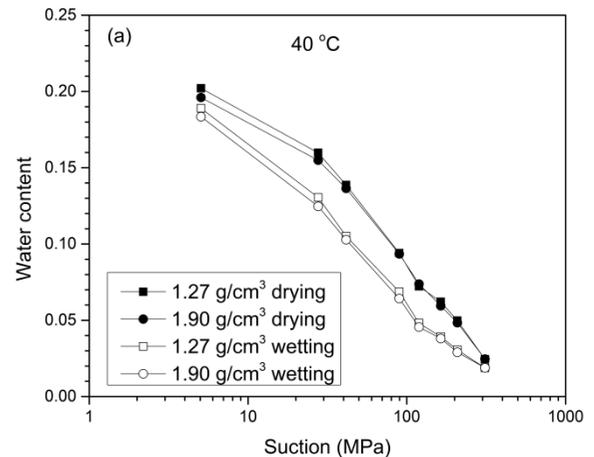


Figure 4. Water retention curves at various temperatures for dry density of 1.27 g/cm^3 ((a) - drying path; (b) - wetting path) and 1.9 g/cm^3 ((c) - drying path, (d) - wetting path).

It can be seen that the water content decreases with increasing temperature for the same initial dry density. This trend is more obvious at low suctions. Comparison of wetting and drying paths shows that wetting curves at high suctions (above 100 MPa) exhibit almost no temperature dependence for both dry densities. The effect of temperature is generally more clear in drying paths.

In order to see the temperature effects on the hysteretic behavior, all the data determined at particular temperature were redrawn to one plot in Figure 5 (a-c). It is clear that the biggest effect of hysteresis can be observed at $40 \text{ }^\circ\text{C}$ and with increasing temperature the hysteretic loop gets narrower. Hysteresis determined at room temperature (Fig. 3) follows this trend. Similarly to room temperature, the initial dry density have little influence on the water content at higher temperatures.



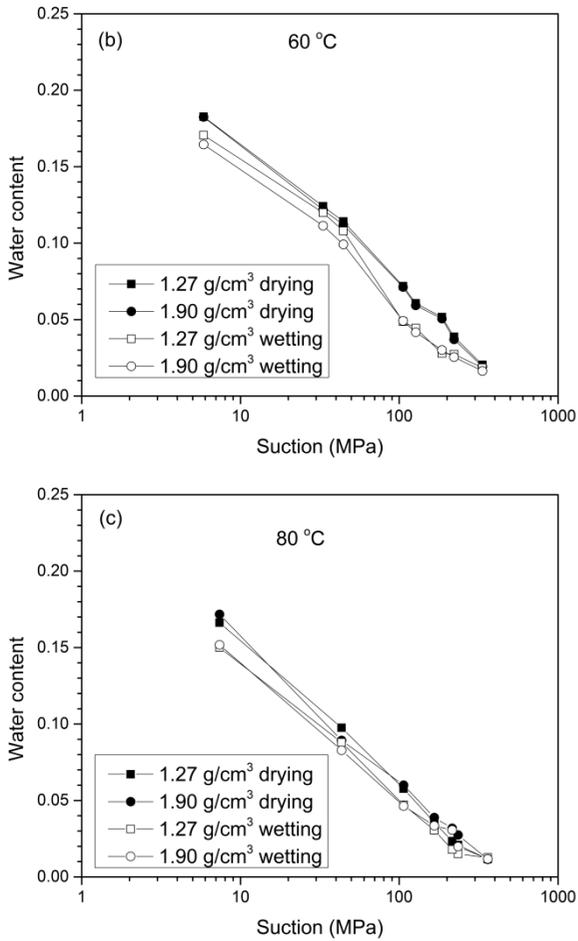


Figure 5. Water retention curves of both dry densities at different temperature of 40°C (a), 60°C (b), and 80°C (c)

The decrease of water retention capacity with increasing temperature is more clearly demonstrated if the water content is expressed in terms of temperature for each salt solution. Figure 6 shows the example of 1.27 g/cm³ drying and wetting test. It shows approximately linear decrease of water content for both wetting and drying tests

and all salt solutions. Slopes of the curves represent sensitivity of water content on thermal change.

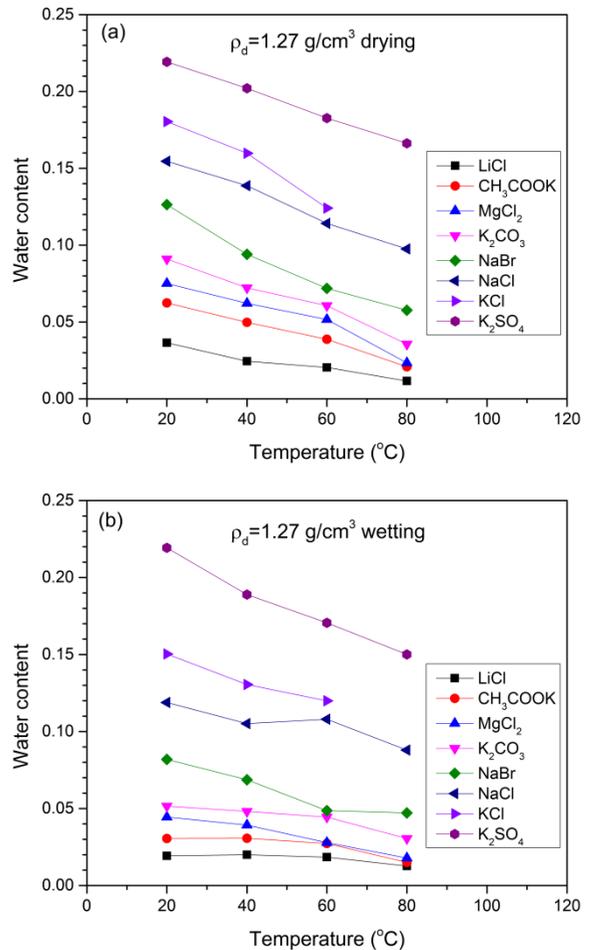


Figure 6. Water content change with temperature for each chemical solution. Drying (a) and wetting (b) paths for the 1.27 g/cm³ samples is presented.

The decrease of water retention capacity with increasing temperature is influenced by the water surface tension, the liquid-gas interfacial tension

or contact angle between water and soil. The increased temperature can also drive volumetric changes of the aggregates. These volumetric changes can include both expansion and contraction, depending on initial dry density and applied suction (Villar and Lloret, 2004; Tang et al., 2008). These structural changes are associated with transfer of the intraaggregate water into inter-aggregate pores, which would explain the lower water retention capacity at higher temperature at certain suction (Villar and Lloret, 2004). The presented results of Czech bentonite B75 are consistent with this theory and confirm that the temperature has an important influence on the water retention capacity.

4 CONCLUSIONS

The retention properties of Czech bentonite B75 were studied in the temperature range 20 – 80 °C. Samples compacted to initial densities of 1.27 g/cm³ and 1.9 g/cm³ were analysed. No variations of retention curves with initial dry density was observed at 20°C and the same result was confirmed at higher temperatures. The water retention capacity of compacted bentonite decrease with increasing temperature for both dry densities. This trend is more obvious for drying paths. For wetting path and suctions over 100 MPa, no obvious change of the retention capacity with temperature was observed. The comparison of drying and wetting paths shows hysteretic behaviour of the retention curves, which is more obvious at higher temperatures.

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6 REFERENCES

- Delage, P., Howat, M.D. and Cui, Y.J., 1998. The relationship between suction and swelling properties in a heavily compacted unsaturated clay. *Engineering geology*, **50**(1-2), pp.31-48.
- Greenspan, L., 1977. Humidity fixed points of binary saturated aqueous solutions. *Journal of research of the national bureau of standards*, **81**(1), pp.89-96.
- Johannesson, L.E., Börgesson, L., Goudarzi, R., Sandén, T., Gunnarsson, D. and Svemar, C., 2007. Prototype repository: A full scale experiment at Äspö HRL. *Physics and Chemistry of the Earth, Parts A/B/C*, **32**(1-7), pp.58-76.
- Sun, H., Mašín, D., Najser, J., Neděla, V. and Navrátilová, E., 2018a. Bentonite microstructure and saturation evolution in wetting–drying cycles evaluated using ESEM, MIP and WRC measurements. *Géotechnique*, pp.1-14.
- Sun H., Mašín D. and Najser J., 2018b. Thermal Water Retention Characteristics of Compacted Bentonite. In: Hu L., Gu X., Tao J., Zhou A. (eds) *Proceedings of GeoShanghai 2018 International Conference: Multi-physics Processes in Soil Mechanics and Advances in Geotechnical Testing*. GSIC 2018. Springer, Singapore.

Tang, A. M, Cui, Y. J. and Barnel, N., 2008. Thermo-mechanical behaviour of a compacted swelling clay. *Géotechnique* **58**(1), 45–54.

Tang, A. M., and Cui, Y. J., 2005. Controlling suction by the vapour equilibrium technique at different temperatures and its application in determining the water retention properties of MX80 clay. *Canadian Geotechnical Journal*, **42**(1), 287-296.

The International Organization of Legal Metrology (OIML). 1996. *The scale of relative humidity (RH) of air certified against saturated salt solutions*. OIMLR 121, France.

Villar, M. V., and A. Lloret. 2004. Influence of temperature on the hydro-mechanical behaviour of a compacted bentonite. *Applied Clay Science* **26**, no. 1: 337-350.

Xu, L. Ye, W. M., Chen, B., Chen, Y. G. and Cui, Y. J., 2016. Experimental investigations on thermo-hydro-mechanical properties of compacted GMZ01 bentonite-sand mixture using as buffer materials. *Engineering Geology* **213**, 46–54.