

# Utilization of Slovak bentonites in deposition of high-level radioactive waste and spent nuclear fuel

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**Abstract** The basic strategic aims in the field of managing high-level radioactive waste and liquidation of nuclear power plants are all contained in the Energy policy of the Slovak Republic. Its aim is to resolve the concept of the backside of the nuclear energetics fuel cycle—long-term deposition of high-level radioactive waste and spent nuclear fuel (SNF). The most important form of high-level radioactive waste and SNF long-term deposition is their deposition in deep geological formations created by natural as well as engineering barriers used to isolate the long-lived radionuclides from the biosphere. The basic components of these barriers are clays, of which bentonite is generally referred to as the most suitable clay material. There are a few significant bentonite deposits in the Slovak Republic: *Jelšový potok*, *Kopernica*, *Lastovce*, *Lieskovec*, *Dolná Ves*. The review article summarizes the information on geotechnical properties of Slovak bentonites published up-to-date, which is inevitable to know for the intention of their use. It highlights the advantages and shows drawbacks of five Slovak deposits. It suggests further research direction, to draw a thorough hydraulic, microbial and radiation profile of Slovak bentonites.

**Keywords** Slovak bentonites · Jelšový potok · Kopernica, Lastovce · Lieskovec · Dolná Ves · Radioactive waste · Spent nuclear fuel · Deep geological repository

## Introduction

The basic strategic aims in the field of managing high-level radioactive (HLRW) waste and liquidation of nuclear power plants are all contained in the Energy policy of the Slovak Republic. Its aim is to resolve the concept of the backside of the nuclear energetics fuel cycle—long-term deposition of HLRW and spent nuclear fuel (SNF).

The review article summarizes the information on geotechnical properties of Slovak bentonites published up-to-date, which is inevitable to know for the intention of their use in deposition of HLRW and SNF. It highlights the advantages and shows drawbacks of five Slovak deposits. It suggests further research direction, to draw a thorough hydraulic, microbial and radiation profile of Slovak bentonites.

The most important form of HLRW and SNF long-term deposition is their deposition in deep geological formations created by natural as well as engineering barriers used to isolate the long-lived radionuclides from the biosphere. The basic components of these barriers are clays, of which bentonite is generally referred to as the most suitable clay material [1–5].

Significant producers of HLRW and SNF in Slovakia are nuclear power plants located in Bohunice and Mochovce [6]. In the Slovak energetics as fuel mainly uranium dioxide is used, enriched by uranium-235 radioisotope, that the average enrichment in the fuel cartridge is 3.82%. One reactor of 1,000 MW power annually produces about 30 t of SNF. Because the fuel has a high density, it represents the volume of only about 1.5 m<sup>3</sup> [7]. One WWPR 440 block annually produces about 220 m<sup>3</sup> of low-level, 90 m<sup>3</sup> of medium-level radioactive waste and 10 t of SNF. It is assumed that blocks of individual nuclear power plants in Slovakia will produce during their project operation period

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2,500 t of SNF and 3,700 t of RW, which under current legislation will not be able to be deposited in the Mochovce RR RW surface type (this estimate includes also the production of RW from NPP A-1 Bohunice) but will have to be deposited in a deep geological repository (DGR).

### Deep repository development in Slovakia

The preparatory and development work on building DGR in Slovakia began in 1996 [8]. DGR should be placed below the ground surface (about 300–700 m) in a stable geological formation, which will form one of the most important barriers of a multi-barrier system (a combination of technical and natural barriers, i.e. geological barrier) [9].

The role of geological barrier as a longest-acting one in the multi-barrier system of repository protection is to isolate the surrounding environment from harmful effects of radiation. Its insulation and protective function must be maintained and sufficient for a period of 100,000 years, till the radiation level has dropped to an acceptable level.

Slovak DGR should be put into operation no later than 2037. Based on preliminary evaluation of existing geological data have been identified 12 territories, potentially suitable for DGR. Further evaluation led to a reduction of this number to five territories in two potential host environments (granitoids and sediments) that have been proposed for more detailed research: Tribec Mts., Veporske vrchy Mts., Stolické vrchy Mts., Rimavska kotlina Basin, Cerova vrchovina Upland [8].

Preparation of models of potentially suitable localities for DGR placement includes extensive multidisciplinary research of interactions between host environment and engineering barriers [10–12]. To construct DGR is needed homogeneous block of geological rock (measuring approx. 1 km × 1 km × 1 km), which behaviour and properties is important to know especially in the considered depth [13].

Currently an option of building an international DGR, where various countries would store would impose their HLRW and SNF seems to be a realistic solution.

### Bentonite

Bentonite and other clay rocks are of paramount importance in the field of environmental and waste management [14–32]. Bentonites are a group of natural nanomaterials composed predominantly of crystalline mineral particles from the group of dioctahedral smectites—montmorillonite [33, 34].

Bentonites from Slovak deposits should be used as part of multi-barrier system in DGR for SNF and HLRW [35–41]. This requires extensive multidisciplinary research

a comprehensive and detailed characterization of bentonites [42–57]. The use of bentonite rocks, or bentonite backfills in the repository is destined to their mineralogical, erosion and rheological properties [33, 34, 58–65, 144], and their favourable adsorption and retardation behaviour to the activation and corrosion products and the fission products of U [66–83]. Study of those products is the subject of still ongoing research [84–103]. Adsorption properties of bentonites are determined by their chemical and mineralogical composition, value of cation exchange capacity (CEC) and specific surface area (SSA) [104–135]. Slovak bentonites are subjected to basic [136–143] and applied [144–157] research for several decades. The first results our department has been published already in 1966 [158]. The purpose of the multidisciplinary study of bentonites is to assess comprehensively in the long term the geotechnical properties and to simulate conditions expected in DGR for SNF and HLRW [159–174].

### Bentonite deposits in Slovakia

In the Slovak Republic there are several important bentonite deposits [8, 9, 17, 25, 33–39, 41]. There is the most popular and long mined bentonite deposit formed by Al–Mg-montmorillonite in the Slovak upland in the locality Stará Kremnička: *Jelšový potok*. In this area, there is one more a partially mined deposit of andesitic bentonite formed by Fe-montmorillonite: *Lieskovec*. In the eastern neovolcanic area, there are two currently mined deposits of rhyolitic bentonites: *Kuzmice* and *Lastovce* (Al–Mg-montmorillonite). Moreover, in this area there are currently not mined deposits Nižný Hrabovec, Fintice, Nižný Žipov, Veľaty and Hliník. Deposit *Dolná Ves* (illite–smectite) is a part of Jastrabská formation.

Genetically are these deposits partly different, even though their common essential characteristic is that they arose as a product of volcanic activity and by subsequent action of alternating processes.

From the perspective of the qualitative composition of the Slovak bentonites can be earmarked two basic groups of Slovak deposits [20, 22, 33, 34, 36, 65]:

- I. *Smectite deposits*—an essential component forming a clay proportion of rock are smectites:
  - (a) *High smectite*—smectite content above 60% (*Jelšový potok* labelled as *J*, *Kopernica* labelled as *K*, *Lieskovec* labelled as *L*),
  - (b) *Low smectite*—smectite content about 40% (*Lastovce* labelled as *LA*).
- II. *Illite–smectite deposit*—an essential component forming a clay proportion of rock is mixed-layer mineral illite–smectite (*Dolná Ves* labelled as *DV*).

The properties of natural chemically modified and irradiated forms from the above-mentioned five Slovak deposits (*J, K, L, LA, DV*) were studied and compared. The fractions of bentonites grounded under 15, 45 and 250  $\mu\text{m}$  were monitored.

From the perspective of the crystal-chemical specifications of the main clay component structure itself can be deposits further classify into [33, 34, 36–39, 41, 43–45]:

- *Al–Mg–montmorillonite*—smectite is identified as montmorillonite and its octahedral structure is composed mainly of Al and Mg (*J, K, LA*).
- *Fe–montmorillonite*—smectite is identified as montmorillonite and its octahedral structure is composed mainly of Fe (*L*).
- *Illite–smectite*—the main clay component is mixed-layer illite–smectite (*DV*).

*J, K* and *L* have similar qualitative mineral composition. *J* and *K* are also close quantitatively. *L* has a significantly lower content of smectite (by about 20%), whose presence is mainly replaced by kaolinite and cristobalite. *LA* differ significantly from the other smectite deposits, especially by very low content of smectite, high content of cristobalite and unavoidable presence of calcite.

### Performance requirements for bentonite barriers

The utilization of bentonites in multi-barrier system inevitably requires assessing a long-term stability of bentonite barrier behaviour, to characterize it from various geotechnical and physico-chemical aspects.

The focus is on:

- *Mineralogical properties* [52, 55, 64, 140, 159, 161, 166, 167, 169, 171, 175–181]: mineralogical composition, the presence of hazardous materials (K-feldspar and biotite, calcite, anthropogenic carbonates), hypergene processes, illitization, kaolinization, interaction of bentonite barrier and the environment, mineral, thermal and microbial stability, technological processing.
- *Physico-chemical properties* [20, 40, 53, 154–156, 165, 168, 170, 173, 182–216]: chemical composition, CEC, adsorption and retardation properties, radiation stability, interaction of metal Fe, buffering capacity.
- *Physico-mechanical properties* [16, 33, 36, 40, 42, 44, 52, 56, 60, 136, 139, 162, 163, 175, 191, 192, 217, 218]: expansion, SSA, plasticity, grain size, hydraulic properties, permeability, compactibility, high strength at the pressure and shear, low compression, high deformation modulus, thermal conductivity, density, consistency limits, strength.

### Mineralogical composition and stability of bentonite

#### Mineralogical composition

The use of bentonites is largely conditioned by the content of smectites, their crystal-chemical composition and by the admixtures of another clay and other minerals [2, 10, 15, 37, 38, 45, 54, 55, 58, 120]. From other clay minerals, kaolinite occurs most frequently, especially in the deposits of hydrothermal origin [33]. Halloysite also in the case of bentonite belongs to accessory present clay minerals and is not crucial to its properties and use [34]. Illite may appear on sedimentary deposits and adversely affect them [46, 47, 50]. From other minerals forming an admixture in bentonite has dominance cristobalite, belonging most often to opal. It appears in all genetic types of bentonite, but mainly in the volcanic-sedimentary deposits, where is formed by the alteration of volcanic glass [36, 41, 50]. Its admixture sometimes reaching up to 40%. It cannot be effectively removed by any treatment and, therefore, significantly limits the use of bentonite in particular for its demanding applications [60, 65].

#### Hypergene processes

The most important process of hypergen sphere is weathering [12, 33, 166, 167, 171]. It is a spontaneous process generated as a result of lack of balance between exogenous conditions influenced by the interaction between atmosphere, biosphere, hydrosphere, and rocks forming the lithosphere [36, 38]. Clays and clay minerals belong among the most important products of weathering [22, 34, 40, 139]. Weathering processes are conditioned especially by original weathering material, climate and landscape morphology [140, 161, 166]. Based on the composition of the attacking solution can be these processes divided into acidolysis, salinolysis, alkanolysis and hydrolysis [60].

#### Illitization

One of the processes leading to the loss of the required properties of bentonite barriers is a mineral conversion of smectite into illite—illitization [12, 33, 36, 52, 53, 166, 169]. This is an irreversible process that causes an irreversible fixation of K atoms in smectite inter-layer area, a loss of hydrated exchangeable cations from inter-layer space and Al for Si substitution in the structure of tetrahedrons [34, 40, 46, 47]. The conversion of smectite into illite via the mixed-layer mineral illite–smectite at elevated temperature is processing not by impacts or saltation, but gradually, depending on the temperature. As a result of dehydration and K fixation in the inter-layer the collapse of the layers occurs, which is accompanied by the loss

of smectite expanding ability, the significant reduction of the SSA, adsorption and CEC, i.e. the total loss of the bentonite retention properties [60, 65, 106, 160, 171–173, 176, 186–192].

#### Kaolinization

Kaolinization process is processing in acidic conditions of acidolysis [30, 33, 36, 60, 196]. Advanced acidolysis is characterized by the higher activity of Al, which is released by octahedron decomposition and migrates into the inter-layer of expanding clay minerals [169]. In slightly acidic conditions (pH 4.3–5.1) of temperate climate zone, there occurred to smectite dissolution, which was accompanied by the disintegration of smectite crystals during cambisol pedogenesis on the contact of Al–Mg–montmorillonite Stará Kremnička deposit (deposit *J*) [34]. At about 2 m gross profile involving soil and bentonite affected by weathering the degradation up to about 60% of the original smectite occurred. A part of amorphous material probably became the source for neoformation of kaolinite, which representation increases towards the surface.

#### Thermal stability

A weakness of bentonites, compared e.g. with natural zeolites (containing mainly mordenite), is greater possibility of distortion of their structure at higher temperatures, which may occur a collapse of smectite structures and a loss of initial adsorption, expanding and other, from a geotechnical perspective significant, properties [25, 29, 36–40, 49, 114, 123, 139, 150, 166, 169, 176]. Bentonite loses the expansion already at temperatures above 150 °C [33, 34, 196, 198, 206, 210, 215]. During the water saturation of bentonite backfill may occur to its evaporation by the decay heat from HLRW and SNF, and thereby to an increase of salt content, which reduces expansion [122, 123, 171, 172]. Similarly smectite illitization can reduce the expansion and adsorption properties of this material [46, 47, 50, 186–192].

#### Microbial stability

Microbial destruction of aluminosilicate minerals, could negatively affect some of their basic properties [176–180]. Pore size of microbes on average generally ranges from 0.1 µm up to several micrometers. The mechanism of microbial destruction by bacteria of the genus *Bacillus* is not stoichiometrically described and fully elucidated yet, but it is known that its effect is a reduction of Si content, the extraction of Al, Ti, U, Au and other elements of silicates and aluminosilicates [179].

Research on microbial activity in the Canadian type depositary, which included the radionuclide microbial

transport, the production of gas and microorganism influence on the corrosion of the containers was confirmed that the compact clay barriers will limit or fully prevent the transport of radionuclides by microbes in view of their small pores [178, 180]. On the container itself, there will not be appropriate conditions for their existence (temperature—drying, radiation). Surviving bacteria will be inactive as long as the environment keeps being dry. After water percolation into the system their re-activation could happen, but retarded by physical conditions of compressed clay barrier.

### Physico-chemical properties

#### Chemical composition

Bentonites *J* and *K* have a very similar chemical composition [43–45, 58, 106, 137, 161]. The most significant difference is the higher Mg content in the *J* bentonite, which is except of removable positions strongly represented also in montmorillonite octahedrons. Bentonites *L* have a significantly higher Fe content compared with the previous two bentonites. This is due to a different parent rock—andesite [52, 64, 78, 79, 134, 167]. When comparing the chemical composition of bentonite fractions with different grain size (15, 45 and 250 µm) from one deposit no significant differences have been shown [186–190]. When comparing *J* and *K* bentonites the differences are minimal. At *L* bentonite samples can be observed minor differences.

#### Cationic exchange capacity

Cation exchange capacity is determined by the ability to adsorb cations and to keep them in the exchangeable positions [12, 33, 34, 60]. In smectites it is called inter-layer space. Quantitative analysis of individual exchangeable cations confirmed the predominance of Ca and Mg over Na and K elements [55, 78, 161, 165, 167, 168]. Distribution of individual initial cations indicates the correlation between the *J*, *K* and *L*, with the difference that *Lieskovec* has a slightly higher content of Na. *DV* bentonites form a separate group, which dominates in the content of exchangeable Na and K. CEC values for individual bentonites deposits decrease in the following order:  $J > K > L > La > DV$  [186–190]. The lowest CEC values, which were measured for the *DV* were up to 4-times lower than in the case of *J* bentonites.

#### Radiation stability

One of the basic prerequisites for the bentonite utilization as engineering barriers is their stability against exposure of

ionizing radiation from radionuclides present in the irradiated fuel cells and HLRW [6, 12, 36, 65, 165, 193–216].

Most activity during the first 100 years falls on the activity of  $^{137}\text{Cs}$ . It is expected that in a box during the first 100 years since fuel cell removal from the reactor is  $3 \times 10^{13} \text{ Bq } ^{137}\text{Cs}$ . Dose rate for  $^{137}\text{Cs}$  with this activity at a distance of 0.5 m is  $26.6 \text{ Gy hod}^{-1}$ . To calculate the absorption of radiation in the 30 cm bentonite layer is used half-width value, which is the same as for concrete, i.e. 3.7 cm. For the 30 cm bentonite layer is then ray photon absorption 99.6%. The annual dose absorbed by the bentonite should be considered with a value of 74 MGy for a year [191, 192].

Interaction of  $\gamma$ -radiation at dose interval of 105, 106, 107 and  $3 \times 10^7 \text{ Gy}$  with various clay minerals such as kaolinite, montmorillonite—dioctahedral Mg-smectite, palygorskite and muscovite was observed in order to detect a change in their physico-chemical properties [209]. There was observed the impact on the type and concentration of the radiation-induced structural defects (in tetrahedrons  $\text{Si-O}^-$  and octahedrons  $\text{Al}^{\text{VI}}\text{-O-Al}^{\text{VI}}$  bond), the degree of the deuterization in consequence of  $\text{D}_2\text{O}$  interaction, the change in the SSA, adsorption and CEC and the effect on the solubility of clay minerals irradiated in heavy water. An important finding was that the degree of accumulation of defects in the structure is a function of the content tetrahedral structural cells ( $\text{Si}_4\text{O}_{10}$ ). In addition to chemical composition, formation of structural defects is significantly affected by the content of  $\text{H}_2\text{O}$  adsorbed on the mineral surface and in the inter-layer space. In general, high water content was slowing down the formation of defects. With increasing radiation dose, there was in the montmorillonite increased the loss waiter of  $\text{Si}^{4+}$  and decreased the loss waiter of  $\text{Al}^{3+}$  from the structure. In kaolinite the situation was reversed. The radiation dose increase from 0 to  $3 \times 10^7 \text{ Gy}$  resulted in the increase of smectite SSA about 8% (for montmorillonite), furthermore in the adsorption capacity increase (15%) and the solubility change.

#### Adsorption and retardation properties

The basic cause of the bentonite adsorption behaviour is determined by the presence of negative charge on the smectite particle surface, which is directly related to the ability to receive and exchange in the inter-layer space so-called exchangeable cations most often Na, Ca, Mg and K [12, 33, 34, 60, 66]. Bentonite adsorption properties are determined by the bentonite chemical and mineralogical composition, CEC and the SSA [54, 55, 58, 61, 62, 82, 107, 108, 110].

Research of bentonite adsorption properties is an essential step for developing a radionuclide migration model [15, 20, 31, 122, 123, 126]. The radionuclide

adsorption is the most frequently studied by the radio-indicator methods, using batch and diffusion techniques [3, 5, 16, 29, 73, 84, 174]. The experiments are carried out under aerobic and anaerobic conditions, in static or dynamic experiment configuration so that the experimental environment approaches as close as possible to the conditions in the deep repository geoenvironment [120, 132, 133, 176, 216–218]. The traditional way of characterization of radionuclide sorption behaviour on the bentonite are sorption isotherms (most often Freundlich, Langmuir, Dubbin-Radushkevitch) [4, 75, 104, 109, 111]. If the experimental isotherms cover a wide interval of concentrations, Langmuir model is the prime candidate to study the sorption behaviour of radionuclides on bentonite.

The focus is on the kinetics of adsorption and the influence of various factors affecting the bentonite barrier adsorption processes, which are applied in the vicinity of stored HLRW in deep geological conditions [2, 65, 69, 70, 129, 135, 159, 173].

There were experimentally studied the adsorption rate, the impact of pH environment change, ionizing radiation, the presence of competing ions, complexing organic agents, synthetic and natural organic ligands on Slovak bentonite adsorption processes in relation to the radionuclides [78–80, 106, 112, 114, 116, 134, 158, 182–192].

#### Adsorption kinetics

Adsorption of  $^{137}\text{Cs}$  and  $^{85}\text{Sr}$  radionuclides in bentonites is relatively quick [68, 182, 184]. The equilibrium of the bentonite sample *J* was reached almost immediately, within 1 min from the beginning of the contact between solid and liquid phase [186, 188]. Comparable values of distribution ratios and percentage adsorption were obtained at interval of 1–480 min. Instantaneous adsorption can be explained by the rapid adsorption process and the cation exchange on the adsorbent surface. At higher concentrations of Sr the capture may also be due to precipitation of Sr in the carbonate form on the bentonite surface [69, 187].

#### pH environments

At the deep environment the changes of pH environment value may occur [2, 4, 6, 69, 127, 135, 161, 171]. Bentonites are characterized by two types of “functional” groups that are in the literature known as “edge sites” and “layer sites” [12, 187]. For the “edge sites”—surface groups, which are usually labelled by symbol  $\text{XOH}$  ( $\text{X} = \text{Al, Si, Fe...}$ ), is characteristic that on them, depending on the pH value, is in the acidic area processing a protonation with the formation of  $\text{XOH}^{2+}$ . In alkaline area there occur to deprotonation with the formation of  $\text{XO}^-$ . To the sorption,

respectively desorption occurs on the protonized, as well as on the deprotonized form with the fact that the character of the reaction mechanism may be complexing (especially  $\text{XO}^-$ ) as well exchangeable (substitution), depending on the type of component that interacts with that group. The effect of the pH value change on the Sr-adsorption in the values range of pH 2–8, in various rates of solid (bentonites *J*, *K* and *L*) and liquid phase (eight different concentrations of Sr) confirmed, that the adsorption of Sr decreases in the order:  $\text{pH} = 8 > \text{pH} = 6 > \text{pH} = 4 > \text{pH} = 2$  [187, 189, 191]. The values of adsorption percentage and distribution ratios increase with increasing pH value (towards the alkaline area also with decreasing initial concentration of Sr in the solution. The value of adsorption percentage close to 99% was reached on the adsorption of Sr-cations on bentonites *J*, *K* and *L* from  $\text{pH} = 8$ . From above it can be concluded that in addition to the basic adsorption mechanism, which is cationic exchange, there are processing at higher pH values the complexing reactions with surface groups of bentonite. The increase of the adsorption percentage value can be attributed to “hydrolytic” adsorption because of the reaction between  $\text{Sr}(\text{OH})^+$  and  $\text{OH}^-$  groups and competition of  $\text{H}^+$  ion is suppressed.

At  $\text{pH} = 2$  there were at the whole studied interval of concentrations observed low values of adsorption percentage, distribution ratio and adsorbed amount of strontium, which is attributable to significant competitive effect of hydrogen ions and disturbed bentonite structure.

#### *Influence of $\gamma$ -irradiation*

On the radiation stability of bentonite barrier are placed high demands because the bentonites will be exposed to the long-term, continuous and immediate effects of ionizing radiation [40, 165, 193, 195]. The initial surface dose rates from  $\gamma$ -ray and neutron radiation are estimated at around  $2 \text{ Gy h}^{-1}$ . The interaction effect of ionizing radiation and bentonites from *J* and *L* deposit have been studied after sample irradiation by cobalt source ( $^{60}\text{Co}$ ) with energy of 1.17 and 1.33 MeV for a period of about 50 days, with a moderate dose rate of  $0.092 \text{ Gy s}^{-1}$  [186, 187]. Total absorbed dose was 390 kGy. At the irradiated samples of bentonites *J250* and *L250* there were detect higher sorbed amount of Cs and Sr than at their non-irradiated forms. Currently there is being conducted a research of Cs and Sr adsorption on bentonites from five Slovak deposits (*J*, *K*, *L*, *LA*, *DV*, native and natrified forms, grain size 15, 45, 250  $\mu\text{m}$ ), that were irradiated in a wider range of doses by  $\gamma$ -source  $^{60}\text{Co}$  with the highest reached dose of 1 MGy. The radiation dose increase resulted in the increase of the SSA, the change of the clay minerals solubility and the possible loss of the initial adsorption properties due to the radiolysis of sorbed water [195, 198–215].

#### *Competition of cations*

Metal cations, which may be present in groundwater, significantly affect the adsorption of radionuclides [2, 5, 12, 15, 18, 33, 66, 73, 78]. The effect can be explained by ion-exchange competing reactions and occupying the active adsorption centres of bentonites. The adsorption is suppressed more by the presence of bivalent cations than by univalent cations. The cause of the different observed effect is in bentonite ability to prefer cations with low hydration energy and small ionic radius. They preferably come into their inter-layer. According to the received ion size the distance between the layers is variably. Smectite inter-layer increases its size by the ion, which is into its structure received. After losing the received ion the structure is changed again. This unique property—expandability is characteristic only for smectites and partially for vermiculites.

The effect of competing cations ( $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{NH}_4^+$ ,  $\text{Ca}^{2+}$ ,  $\text{Ba}^{2+}$ ,  $\text{Mg}^{2+}$ ) on the Cs and Sr adsorption confirmed that the higher distribution ratio values are achieved in the presence of univalent as well in the presence of bivalent cations [182, 186, 187, 189, 191]. Values of the distribution ratios increase with decreasing initial concentration of competing cations in the solution. The Sr-adsorption is the most suppressed by the presence of  $\text{Ba}^{2+}$  cation.

#### *The presence of organic complexing agents, synthetic and natural organic ligands*

Organic compounds such as EDTA, oxalic and citric acid, humic acids and fulvic acids that are the part of the decontamination solutions or are ranged among the organic components of soil and water suppress the adsorption of radionuclides [12]. The effect of EDTA, oxalic and citric acids on adsorption of Cs has been studied in the concentration interval of  $1 \times 10^{-5}$ – $5 \times 10^{-2} \text{ mol dm}^{-3}$  [190]. The results confirmed that the presence of the studied organic and natural ligands significantly affects the adsorption of Cs. By increasing their initial concentration the values of percentage adsorption and distribution rates drop. The adsorption of Cs on samples *J15*, *K15* and *L15* is significantly suppressed at the ligand concentration of  $5 \times 10^{-2} \text{ mol dm}^{-3}$ : EDTA > citric acid > oxalic acid. The adsorption reducing effect can be explained by formation of soluble complex ions, which are too large to enter the available structural positions of bentonite [69, 190, 191].

#### *Interaction with metallic iron*

The prognosis of the long-term stability of bentonite barriers in DGR includes studying the interaction of bentonite-metallic Fe and bentonite-surrounding rock system [40, 55, 64, 126, 133, 134, 154–156, 193, 196]. The processes

processing in the repository were simulated to determine the stability of the Slovak bentonites in the presence of metallic Fe, pyrite and Fe(III)-oxide-hydroxides [168, 170]. The results show a partial destabilization of the smectite structure. The smectites with the highest Fe content in its structure in the presence of metallic Fe have shown as the least stable. It follows that the bentonite barriers that will be contacted with iron structural elements of depository should be without structural Fe.

### Physico-mechanical properties

#### Expansion

It allows in contact with water and polar liquids to expand in volume up to twelve-times [33]. It is important at sealing joints and cracks when a contact of bentonite barrier and water (self-healing) [34, 40, 159]. Expanding pressure of the bentonite barrier may not adversely affect the function of the container and building elements of engineering barrier, respectively the function of natural barrier [10, 36, 60]. Expanding pressure is low order in the case of K-bentonites from DV as well in the smectite type bentonites [34, 65, 192].

#### Specific surface area

The SSA, which characterizes the adsorbent active surface area allows high reactivity with the environment in which the adsorbent—bentonite is [33, 34, 36, 60, 139]. The comparison of physico-chemical properties of the Slovak bentonites showed that natural *J* and *K* bentonites have a significantly higher SSA than bentonites from *L* and *LA* deposits [44, 65]. The same applies to the bentonite hydration ability. Those properties achieved the lowest values at DV bentonites, which fully correlated with their mineral composition [187, 188].

When comparing the chemically activated samples (natrification), the largest SSA have the natrified bentonites from the deposit of *Jelšový potok*, when are the changeable positions in the inter-layer saturated by Na<sup>+</sup> cations [189, 191]. Partially lower values were achieved by *K* samples and natural *J* samples. *L* bentonites showed the lowest values. Natrification allows the best separation of the basic smectite layers, which leads to the obtaining of a new area for adsorption. The decrease of the SSA from the finest to the coarsest fraction (15 → 45 → 250 μm) was moderate and consistent for all samples.

#### Plasticity

The plasticity is in geotechnical sciences defined as the moisture range between the value of the plasticity limit and

fluidity limit [33, 34, 36, 51]. It is expressed by the plasticity index. For the light self-healing of areas of discontinuity it is appropriate to the plasticity index value was the highest. High plasticity index value means that even at large moisture increase over the plasticity limit soil remains plastic and does not pass into the liquid state. In the plastic state bentonite keeps sealing effects. The large increase of plasticity was observed due to the bentonite natrification [65, 188, 191, 192]. Smectitic bentonites of Central Slovakia (*J*, *K*, *L*) achieved the results of plasticity comparable with internationally tested bentonites (*Montigel* a *MX80*). *LA* and *DV* bentonites show significantly worse properties than the above three deposits. K-bentonite from *DV* has the worst physical and mechanical parameters at all indicators. Short-term treatment of temperature on the non-natrified bentonite (7 days at 90 °C) had no noticeable effect on the bentonite plasticity.

#### Permeability

Imperfect preparation of bentonite, thermal and mechanical stress of barrier material may cause the area discontinuity, then the increase of the material permeability [36, 56, 65]. Among the studied Slovak deposits (*J*, *K*, *LA*, *L*, *DV*) the samples of K-bentonites *DV* had the worst hydraulic permeability parameters [162, 163, 192].

### Modification of bentonites

Bentonite can be used in a natural, physically or chemically modified form [25, 37–39, 65, 80, 82, 105, 114, 144, 146, 154, 156]. Modification is made to improve their physical and chemical properties.

Physical modification is a basic mechanical treatment of bentonites, which consists of drying and milling [33, 44]. By milling are bentonites fined down to the desired grain size. When comparing the Slovak bentonite fractions milled under 250, 45 and 15 μm has been shown that it does not make sense to work with the finer grain sizes than 250 μm [187–192]. At the finer fractions (45 and 15 μm) there is no significant improvement of the physico-chemical properties and technological process of bentonite barrier treatment would be pointlessly financially encumbered.

Drying reduces the original moisture content of bentonite (when surface mining of Na-bentonite around 30%, Ca-bentonite around 25%) to the required 7–8% [12, 33, 36, 38, 49, 52, 60, 136]. Drying temperature ranges from 100 to 200 °C at the input and up to 800 °C at the output, depending on the desired application. Smectite structural properties get significantly worse if when is the temperature exceeded.

More exacting bentonite treatment method, nowadays already quite common, is based on the preparation of the chemically modified forms [63].

At the chemical treatment the natural bentonite is cultivated by chemicals for various types of finished products, usually directly in the mining plant [33, 34]. Chemical modification is allowed by a large SSA with active sorption centres, the presence of molecular water and exchangeable cations in the smectite inter-layer space area. There are prepared mono-ion forms, of that is the most often used the protonation (the changeable positions in the inter-layer are saturated by  $H^+$  cations) or natrification (the changeable positions in the inter-layer are saturated by  $Na^+$  cations) [57, 60, 73, 103, 189]. As a nitrification salt is almost exclusively used bicarbonate  $Na_2CO_3$  because of its financial ease and the significant exchange rate. The amount of soda ingredient varies with the intended use and the type of bentonite. Preferable is the wet method of activation, by which is all bentonite transformed into Na-bentonite. Natrification allows the best separation of the basic smectite layers, what leads to the obtaining of a new area for the cations exchange and to an increasing of the full adsorption capacity. The utilization of artificial natrified Slovak bentonites as bentonite barriers in DGR was not excluded yet [189, 191]. The main problem is the technological process, which significantly shifts the pH to alkaline area. On the adsorption of  $Cs^+$  and  $Sr^{2+}$  the formation of colloids occurred that could not be removed even by repeated centrifugation at high speed. Natrification may also cause undesirable excessive pressure from the expansion of the clay barriers. Specific and more complicated method of chemical modification is a intercalation and pillaring of the smectite structure [42, 49, 142, 150, 205, 219, 220]. In the first case, is concerned the process of intercalation by organometallic and organic cations, in which are prepared so-called organobentonites. Pillaring respectively reinforcing of the smectite structure is made due to their very low thermostability. Into the smectite inter-layer area is inserted e.g.  $Al_2O_3$  molecule that performs there a function of the pillar, defending to the smectite structure collapse.

## Conclusion

Bentonites from the Slovak deposits (*Jelšový potok*, *Kopernica*, *Lastovce*, *Lieskovec*) meet most of the geotechnical requirements for that type of barriers. They can be used as filler, damping, respectively sealants materials in the vicinity of a container of radioactive waste and SNF.

The deposit *Dolná Ves* showed the lack of geotechnical, especially adsorption and retarder properties of bentonite. From the long-term barrier stability point of view the

*Lieskovec* bentonites with high content of structural bound Fe appear to be unusable.

Smectite bentonites jastrabská formation—*Jelšový potok* and *Kopernica* can be identified as the most suitable material for practical use when depositing high-level radioactive waste and SNF. Bentonites of these deposits are stable material guarantees a long-term stability.

Hydraulic properties require attention in the further study of the matter as well as radiation and microbial stability of Slovak bentonites. The authors of the article would be extremely grateful to the workplace which could on the studied Slovak bentonites reach absorbed doses of ionising radiation of 10–100 MGy.

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