# Engineering geological clay research for a radioactive waste repository in Slovakia

Renáta Adamcová[1], Jana Frankovská[2] & Tatiana Durmeková[1]

[1]Department of Engineering Geology, Faculty of Natural Sciences, Comenius University in Bratislava, Mlynská dolina G, 842 15 Bratislava, Slovakia; adamcova@fns.uniba.sk; durmekova@fns.uniba.sk
[2]Department of Geotechnics, Faculty of Civil Engineering, Slovak University of Technology in Bratislava, Radlinského 11, 813 68 Bratislava; jana.frankovska@stuba.sk

AGEOS

## Inžinierskogeologický výskum ílov pre hlbinné úložisko rádioaktívnych odpadov na Slovensku

**Abstract:** Research for the deep geological repository (DGR) started in Slovakia in 1996. Results of the engineering geological research of the clay in both, the natural and engineered barriers are summarized in the paper. The Szécsény Schlier from the Lučenec Formation is a potential sedimentary host rock for the DGR in Slovakia of favourable properties, similar to a natural clay barrier. It is a friable calcareous siltstone with intercalations of silty clay and fine-grained sandstone, of low hydraulic conductivity (k≅10-10 m.s$^{-1}$) and acceptable uniaxial compessive strength (from 18.5 to 34.8 MPa). Engineering geological characterisation of this formation is based upon rock samples collected from boreholes and tested in the laboratory. Several deposits in Slovakia might offer suitable buffer/backfill material for engineered barriers of the DGR. Since 2003 five bentonite types have been studied, selected results of the engineering geological evaluation are discussed in this paper. Liquid limit was the first suitability indicator. Grain fraction <0.250 mm was preferably used in the swelling tests, needing lower compaction pressures than finer fractions. Bentonite from Jelšový Potok deposit showed a very good quality of pressed high-density samples (with uniaxial compressive strengths similar to weak rocks) and the best swelling potential. Comparing physical and mechanical properties, succession Dolná Ves<Lastovce≅Lieskovec<Kopernica≅Jelšový Potok can be written. However, bentonite from Dolná Ves was considered as an alternative because of the very good compactibility and, particularly, large amounts available in the deposit.

**Key words:** radioactive waste, deep geological repository, Szécsény Schlier, host rock, engineered bentonite barrier

## 1. INTRODUCTION

Modern life is inherently connected with production of radioactive waste in most countries of the world, even also in those without nuclear power plants. Safe disposal of this waste is a big challenge for the scientists and engineers. After more than 20 years of research (e. g. Bucher & Müller-Vonmoos, 1989), a deep geological repository (DGR) seems to be the best solution for the most dangerous radioactive waste. This is the result of hundreds of technical reports (e.g. Pusch, 2001; OECD NEA, 2003; Witherspoon & Bodvarsson, 2006; etc.) published by organisations responsible for the radioactive waste disposal abroad, e.g. NAGRA in Switzerland, ANDRA in France, SKB in Sweden, ONDRAF in Belgium, etc., describing in detail different aspects of the safe construction and sealing of a DGR. Clays have a special place in DGR projects: as one of the considered host rock environments, as well as the most important material for engineered barriers and pollution confinement. A wide range of scientific subjects, dealing with most of the disciplines concerned by the "Clay Science" applied to waste isolation was covered within international meetings "Clays in Natural and Engineered Barriers for Radioactive Waste Confinement" in France: in 2002 in Reims, in 2005 in Tours, in 2007 in Lille. Proceedings from these meetings (e.g. Aranyossy, 2007, 2008) are full of up-to-date scientific ideas, discoveries and conclusions. Many research papers dealing with special problems of clays in a DGR, and of DGRs in clays can be found also in the Journal Applied Clay Science (Alba et al. 2009, Alonso et al. 2009, Ghorban et al. 2009, Kaufhold & Dohrmann 2009, Sun et al. 2009, etc.).

Learning from foreign experience helped Slovakia partly to over-bridge the gap caused by a late start of own research and investigation, as well as insufficient financial means. The Deep Geological Disposal of Spent Fuel and High Level Waste programme of the Slovak Republic has been funded from the State Fund for Nuclear Facilities Decommissioning and Spent Fuel and Radioactive Waste Management since 1996, aiming at the selection of suitable locality for the DGR. The radioactive waste in Slovakia is produced mainly during operation of two nuclear power plants (NPP), Mochovce and Jaslovské Bohunice, and decommissioning of the latter one (Fig. 1), as well as handling radioactive materials in medicine, industry and research. During the operation of the Slovak NPPs, 2300 tons of spent fuel will approximately be produced. In addition, about 5000 tons of radioactive waste unsuitable for the national near-surface repository at Mochovce should end in the DGR (Matejovič et al., 2006).

A large part of the Slovak territory is located in the Western Carpathians. The Carpathian arc is a tectonically complicated Alpine-type geological structure within the Alpine mountain chain of Europe, including different tectonic units. Tectonic units studied in Slovakia as potential DGR host rock environ-

Manuskript doručený: 4. novembra 2009
Manuskript revidovaný: 10. decembra 2009

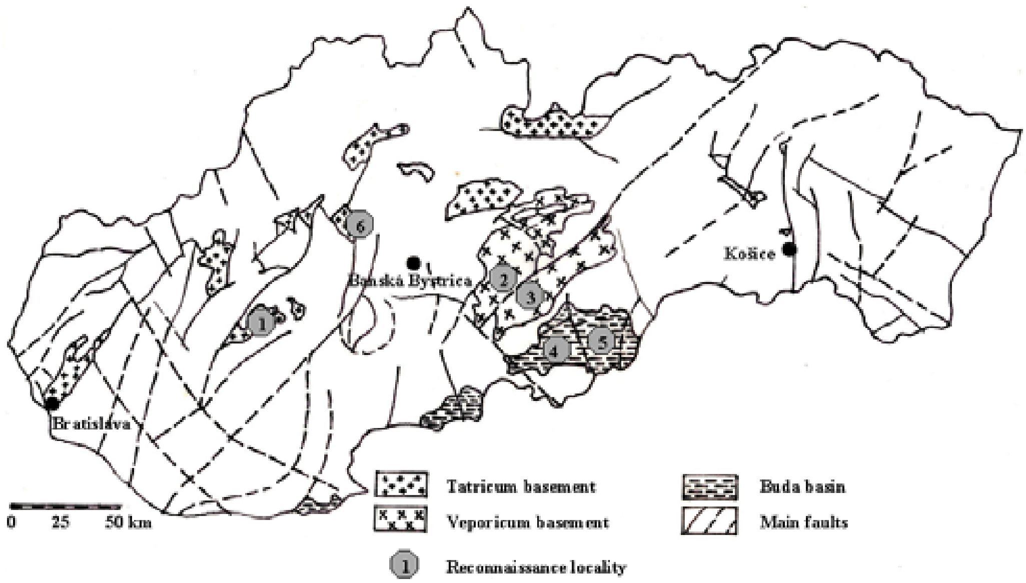


**Fig. 1. Potential DGR sites in studied tectonic units in Slovakia (according to Kováčik et al., 2001): 1 – Tribeč Mts.; 2 – Veporské vrchy Mts.; 3 – Stolické vrchy Mts.; 4 – Rimavská kotlina Basin; 5 – Cerová vrchovina Upland; 6 – Žiar Mts.**

**Obr. 1. Potenciálne lokality pre vybudovanie hlbinného úložiska rádioaktívneho odpadu v skúmaných tektonických jednotkách na Slovensku (upravené podľa Kováčika et al., 2001): 1 – Tribeč.; 2 – Veporské vrchy; 3 – Stolické vrchy.; 4 – Rimavská kotlina; 5 – Cerová vrchovina; 6 – Žiar.**

ments are presented in Fig. 1. The Tatric and Veporic basement units are built up by the Hercynian (Variscan) crystalline rocks (Maheľ, 1986). The whole Tatric Unit contains isolated cores of the crystalline basement (granitoids and metamorphic rocks) partly covered by autochthonous and allochthonous Late Palaeozoic to Mesozoic sediments. The Veporic Unit is the largest granitoid pluton in the Western Carpathians, with a length of about 60 km. The rocks of this unit (mainly granitoids) may potentially provide suitable sites for DGR construction. Four sites in granitoid rocks (granites, granodiorites, tonalites) were selected for detailed geological investigations, namely: the central part of the Tribeč Mountains, the southern part of the Veporské vrchy Mountains, the southwestern part of the Stolické vrchy Mountains, and the central part of the Žiar Mountains (Fig. 1). A characteristic feature of the Western Carpathians is the presence of Neogene post-tectonic basins filled with Miocene sediments (clays, claystones, sands and sandstones predominantly) (Vass, 2002). The Neogene sediments are several thousand metres thick and their pelitic sequences are also potentially suitable as host rocks for DGR facility building. Up to now, two sites were selected: the eastern part of the Cerová vrchovina Upland and the western part of the Rimavská kotlina Basin (parts of the Buda Basin, Fig. 1). Engineering geological research of the Szécsény Schlier as an important part of these two Neogene sites will be described below. Further reduction of the number of candidate sites is foreseen and the final selection of the DGR site is expected around 2010.

Beside the suitable locality, also a bentonite type suitable for engineered clay barriers has to be selected. Slovak bentonites have been studied within 3 research projects, to identify the most suitable of them. Mineralogical and chemical analyses and experiments represent an important step of this research; however, only selected results of engineering geological tests on bentonites from five Slovak deposits are discussed in this paper. The results were compared with data on the bentonites MX-80, Montigel or FEBEX known from international research projects if it was possible.

## 2. SEDIMENTARY HOST ROCK – THE NATURAL "CLAY" BARRIER

### 2.1. Geological description

The Lučenec Formation is a potential sedimentary host-rock for DGR facility building. It occurs at both potential sites, the Cerová vrchovina Upland and the Rimavská kotlina Basin. This geological formation consists of 6 members of marine origin. The formation is of Egerian age (Late Oligocene – Early Miocene). The Szécsény Schlier is the dominant member of this formation, of a monotonous lithological composition: it is a friable jointed calcareous grey or blue-grey coloured siltstone in fresh state, with occasional 10 to 30 cm thick intercalations of hard siltstone, silty clay or fine-grained sandstone. In the lower part of this member, also thicker layers of sandstone can be found (up to 1 m). The Szécsény Schlier is rich in marine fauna (molluscs, foraminifers), calcareous nanoflora and sporomorphs. It was deposited on shelf or in an open sea with depth of 150 to 200 m under saline or slightly hyper-saline conditions (Vass, 2002). On the Slovak territory, maximum thickness of the Szécsény Schlier verified by the boreholes is about 700 m, but it might be thicker near the Slovak-Hungarian border (up to 1300 m). Geophysical measurements indicate a favourably increasing thickness of the formation from N to S, as well (Kováčik et al., 2001).

The tectonic structure of the whole region is relatively complex, with various fault directions, both in the pre-Neogene basement and in the Neogene sedimentary filling, too, that contributes to the horst-and-graben style of the basement configuration. In the potential DGR area, the faults form two systems, the main one is oriented SW-NE and is related to the Carpathian arc uplift, while the secondary system is oriented NW-SE. Most of the faults do not show considerable vertical movements and the dominant tectonic processes ended together with the volcanic activity during Miocene. There are no indications that the volcanic activity could be re-activated (Kováčik et al., 2001).

According to the European Macroseismic Scale (EMS-98), the generally accepted limit of seismicity for a DGR establishment in Slovakia is intensity number VI. The majority of the Slovak territory roughly averages this value. Potential DGR sites are considered to be located at a sufficient distance from recorded earthquake epicentres (Kováčik et al., 2001).

Field investigations included shallow drilling to 250 m below the surface. Rocks and water samples collected during drilling were used for laboratory investigation. Selected results from the borehole RAO-5 are presented in this paper. The borehole is situated in the Rimavská kotlina Basin, SE from Fiľakovo town, in the vicinity of the village Gemerček.

## 2.2. Investigation methods

The laboratory investigation included mineralogical analyses (3 samples, from depths of 161 m, 209 m and 250 m) and determination of basic physical and mechanical properties (13 samples from the interval of 21 to 248 m).

For a preliminary qualitative description of the mineral composition, X-ray powder analyses were carried out using diffractometer DRON-3 with a Co anticathode, $K_\alpha$ radiation (1.78897 Å), and 0.1 °2θ/s goniometer speed. Oriented specimens were analysed after air-drying, as well as after saturation with ethylene glycol (EG).

The following standard laboratory methods were applied for identification and determination of geotechnical properties of the rock samples: determination of dry density $\rho_d$ [g.cm$^{-3}$] and particle density $\rho_s$ [g.cm$^{-3}$] (also called "apparent density" and "real density" according to the standard EN 1936, 2006), porosity $n$ [%] and water absorption (EN 13755, 2008), evaluation of the rock behaviour in contact with water (EN ISO 14689-1, 2003), determination of Atterberg consistency limits (BS 1377, 1990), uniaxial compression test and deformability test (EN 1926, 2006; EN 14580, 2005) and Brazil test for determination of the tensile strength (EN 1997-1, 2004; EN 1997-2, 2007). Hydraulic conductivity was tested in a triaxial cell with pore water pressure control.

## 2.3. Geotechnical properties of the Szécsény Schlier

The analysed samples of the Szécsény Schlier consist of quartz, carbonates (calcite and dolomite), feldspars and sheet silicates. Kaolinite, illite and swelling minerals (smectite or mixed layers, e.g illite/smectite, as applied method did not allow an exact identification) are the main clay minerals in all samples, partly also chlorite at a 161 m depth (Fig. 2). In two samples at 209 and 250 m depth, traces of pyrite and siderite were also identi-

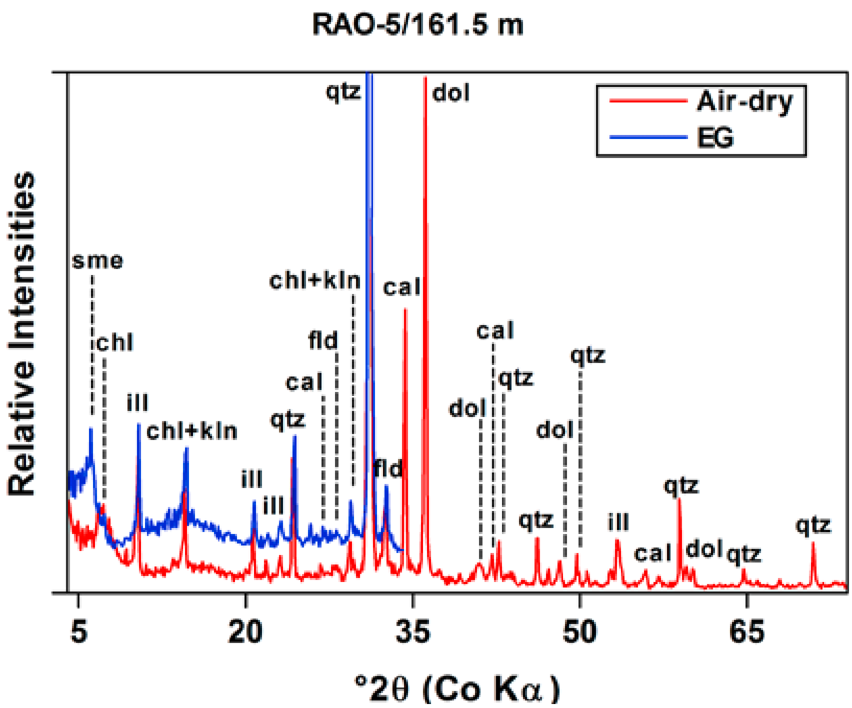


**Fig. 2. Powder XRD patterns of a sample from the borehole RAO-5 (analysed by M. Gregor, not published): qtz – quartz, dol – dolomite, cal – calcite, fld – feldspar, chl – chlorite, kln – kaolinite, ill – illite, sme – smectite and swelling mixed layers.**

**Obr. 2. Záznam práškovej rtg difrakčnej analýzy vzorky z prieskumného diela RAO-5 (analyzoval M. Gregor, nepublikované): qtz – kremeň, dol – dolomit, cal – kalcit, fld – živce, chl – chlorit, kln – kaolinit, ill – illit, sme – smektit a napúčavé zmiešanovrstevnaté silikáty.**

Tab. 1. Geotechnical properties of the Szécsény Schlier Formation from the borehole RAO-5.

Tab. 1. Fyzikálno-mechanické vlastnosti séčenských šlírov z vrtu RAO-5.

| Depth [m] | Dry density [g.cm$^{-3}$] | Porosity [%] | Uniaxial compressive strength [MPa] | Static modulus of elasticity [MPa] | Tensile strength (Brazil test) [MPa] |
|---|---|---|---|---|---|
| **21** | 2.193 | 18.51 | 22.5 | x | x |
| **31** | 2.133 | 20.74 | 23.1 | x | 1.9 |
| **55** | 2.226 | 17.28 | 24.0 | x | 2.6 |
| **91** | 2.189 | 18.65 | 22.2 | x | x |
| **111** | 2.162 | 19.66 | 19.7 | x | 1.6 |
| **146** | 2.199 | 18.28 | x | x | x |
| **176** | 2.195 | 18.43 | 21.7 | 2 939 | x |
| **191** | 2.228 | 16.18 | 18.5 | x | 1.6 |
| **209** | 2.232 | 16.03 | 30.4 | x | x |
| **220** | 2.199 | 17.27 | 26.5 | x | x |
| **227** | 2.228 | 16.18 | 23.9 | x | 1.9 |
| **231** | 2.254 | 15.20 | 34.8 | 4 600 | x |
| **248** | 2.214 | 16.70 | 26.9 | x | x |

Tab. 2. Results of hydraulic conductivity k – the borehole RAO-5.

Tab. 2. Hodnoty koeficientu filtrácie k – vrt RAO-5.

| Depth [m] | 161.5 | 209.5 | |
|---|---|---|---|
| **Hydraulic gradient** | 169.5 | 69.4 | 138.9 |
| **k [m.s$^{-1}$]** | 3.59E-10 | 1.5E-10 | 1.5E-10 |
| **T [°C]** | 18.2 | 17.4 | 17.5 |
| **k10° C [m.s$^{-1}$]** | 2.89E-10 | 1.23E-10 | 1.22E-10 |

fied. Quartz is the dominant phase in all evaluated samples. The presence of the swelling clay minerals from smectite group is important for the host rock properties as a natural barrier.

Basic geotechnical parameters of the samples from the borehole RAO-5 are summarised in Tab. 1. The dry density $\rho_d$ is high (about 2.20 g.cm$^{-3}$) and increases with depth. Due to the high dry density $\rho_d$ and relatively low total porosity $n$ (from 15% to 20%), low hydraulic conductivity and low diffusion coefficients can be expected. The uniaxial compressive strength (UCS) was determined as the maximum vertical stress obtained during the compression test. Results of the compression tests confirm the visual homogeneity of rock samples from various depths. The obtained values of UCS range from 18.5 to 34.8 MPa, with a moderate increase with depth. Only several informative tests of the Young's elasticity modulus and Poisson's ratio were carried out on cylindrical rock specimens from 176 and 232 m depth. The Brazil test as a supplementary method provides an indirect determination of the tensile strength in a forced rupture

**Tab. 3. Host rock mass rating according to the classification of Bieniawski (1989).**
**Tab. 3. Hodnotenie hostiteľského horninového masívu podľa klasifikácie Bieniawskeho (1989).**

| Classification parameter | Maximum score | Szécsény Schlier score |
|---|---|---|
| Uniaxial compressive strength | 15 | 4 |
| Rock Quality Designation | 20 | 20 |
| Discontinuity spacing | 20 | 20 |
| Discontinuity condition | 30 | 25 |
| Groundwater | 15 | 15 |
| Total = Basic Rock Mass Rating | 100 | 84 |

plane. According to the laboratory tests, no significant changes in the strength were detected along borehole RAO-5 that indicates a homogeneous soft rock environment from the surface to the depth of 250 m.

Laboratory investigation showed that siltstones of the Szécsény Schlier are unstable in water and disintegrate into cohesive soil. Their stability grade is 5 that means the whole rock specimens become muddy after 24 hours in water (EN ISO 14689-1, 2003) and can be tested as a soil. Consistency limits of soils depend on the mineral composition of the sediment, especially on the amount and the type of clay minerals. Low value of the liquid limit $w_L$ (from 37.52 to 39.17 %) indicates a very low content of swelling clay minerals. The plastic limit $w_P$ is approximately 24% and the index of plasticity $I_P$ is then from 13.08 to 14.85%. The hydraulic conductivity of the tested samples was of the order of $10^{-10}$ m.s$^{-1}$ (Tab. 2).

According to the rock mass rating (RMR) of Bieniawski (1989), the Szécsény Schlier is qualified as a good rock (Tab. 3) at least along the borehole RAO-5. From all evaluated parameters, only the UCS gave low numbers, for UCS falling within the interval of 25 – 50 MPa receives rating 4. Of course, since the DGR will be located at a depth between 500 and 600 m, the quality of the rock has to be confirmed by further investigation in deeper parts of the rock mass.

## 3. ENGINEERED CLAY BARRIER

### 3.1. Material

Since 2003 five bentonite types considered for DGR engineered clay barriers have been tested in the laboratories of the Comenius University in Bratislava, sampled at candidate deposits in Slovakia: Jelšový Potok (J), Kopernica (K), Dolná Ves (DV), Lieskovec (L), Lastovce (LA). They are all products of the Neogene volcanic activity (rhyolite tuffs and tuffites, andesites), and of different subsequent alteration processes and/or re-deposition. Šucha et al. (2005) summarized papers dealing with previous research, as well as data regarding origin, mineral composition, chemical, and physical properties of all mentioned bentonites. Basic information about the different bentonite types is given in Tab. 4. $Ca^{2+}$ is the main exchangeable smectite cation in all studied bentonites. Natural bentonites were used, but bentonite J was also natrified by adding ca. 4% of soda (symbol $J_{Na}$). All bentonites were industrially dried, powdered in a mill and three different fractions were separated in centrifuges: <0.250 mm, <0.045 mm, <0.015 mm. Samples of different maximum grain size can be recognised by symbols 250, 45 and 15.

### 3.2. Laboratory methods

#### 3.2.1. First phase

The laboratory engineering geological research was organized in two phases. In the first phase, all bentonite types and fractions were tested, and mostly standard methods were applied for the engineering geological classification. Bentonite samples were compared first on the basis of simple methods, such as the determination of the Atterberg liquid limit $w_L$ [%] by the Casagrande method and by the fall cone method (BS 1377, 1990). This criterion was thought to be a useful indicator for a pre-selection, as it reflects well the smectite content. High liquid limit means a high water sorption capacity, indicating also a high swelling capacity, the most important physical property.

Other engineering geological tests were applied to complete the classification and description of the bentonite powder: determination of the Atterberg plasticity limit $w_P$ [%] according to BS 1377 (1990), the granulometry by a SediGraph© and hydrometer (BS 1377, 1990), the particle density $\rho_s$ [g.cm$^{-3}$] by a Micromeritics© helium pycnometer, the moisture content $w$ [%] by oven-drying (BS 1377, 1990), the latter varying even in the prepared powder due to changes in the atmosphere (air pressure, moisture, temperature). The moisture tests were repeated as often as necessary. The plasticity index $I_P$ [%] was calculated as $I_P = w_L - w_P$.

The index of the colloidal activity $A$ (dimensionless) resulted from the mathematical expression $A = I_P / x$, where $x$ [mass %] is the content of clay particles finer than 0,002 mm.

**Fig. 3. First compaction mould prepared to press ∅ 50 mm bentonite cylinders.**

**Fig. 3. Prvá zhutňovacia forma pripravená na lisovanie bentonitových valčekov ∅ 50 mm.**

**Fig. 4. Second compaction mould – for ∅ 100 mm bentonite cylinders.**

**Obr. 4. Druhá zhutňovacia forma – na bentonitové valce priemeru 100 mm.**

Because high-density bentonite blocks will probably be used for the barrier construction, knowledge of the compactibility was another requirement in this study phase. The bentonite powder was compacted to regular cylinders ($r$=2.5 cm, $h$=5.0 cm) in a special mould (Fig. 3) using a hydraulic jack. The „envelope volume" of the cylinder $V=\pi.r^2.h$ and bulk density $\rho_n=m/V$ could be easily calculated ($m$ [g] is mass of the cylinder). The dry density $\rho_d$ [g.cm$^{-3}$] of every pressed sample was calculated with the immediate moisture content $w$ in the powder:

$$\rho_d = \frac{\rho_n}{1+w}$$

The compactability was expressed in plots as the obtained dry densities $\rho_d$ relative to necessary compaction pressures.

At the end of this experimental phase, the results of the liquid limits and of the compactability were critically reviewed and a limited number of bentonite types and fractions was selected for the second phase.

### 3.2.2 Second phase

The tests still had rather a comparative character, whereby bentonites J and DV were most carefully compared. The swelling pressure $p$ [MPa] and the relative expandability $B_0$ [%], as well as the hydraulic conductivity $k$ [m.s$^{-1}$] were the prime important parameters to be addressed in this phase. To answer them, methods based on oedometer measurements were applied. Two types of samples were used according to the oedometer type. At low dry densities and expected low swelling pressures, old oedometers (capacity only 0.8 MPa) were used, where the sample diameter was 100 mm. They required a new compaction mould (Fig. 4). When the high-density samples had to be tested, some tests were carried out at the Technical University Prague in oedometers allowing pressures up to 10 MPa. Now those tests continue at the Comenius University inBratislava in new oedometers (Tecnotest©). High pressures can be reached due to the small sample diameter, i.e. much smaller area is under load. 20 mm high cylinders with a diameter of 50 mm are required, so that samples can be compacted in the first mould adding a small adapter to the piston.

High swelling pressure is required from buffer material to be able to seal any cracks and failures in the barrier (so-called "self-healing" effect). The swelling pressure $p$ due to saturation with distilled water was determined from the counterforce necessary to prevent swelling, i.e. as soon as small deformations occurred, the load was increased to bring the sample back to the starting "0" position.

The relative expandability $B_0$ [%] was determined in a free-swelling test in oedometers, i.e. no vertical load was applied to the sample surface:

$$B_0 = \frac{h_2-h_1}{h_1}$$

**Tab. 4. Basic mineralogical characteristics of the candidate bentonite deposits (Šucha et al., 2005).**

**Tab. 4. Základná mineralogická charakteristika perspektívnych ložísk bentonitov (Šucha et al., 2005).**

| Bentonite | smectite type | | illite-smectite type |
|---|---|---|---|
| | smectite content [%] | main clay mineral | |
| J | >70 | Al-Mg montmorillonite | x |
| K | >70 | Al-Mg montmorillonite | x |
| L | 60 – 70 | Fe montmorillonite | x |
| La | 40 | Al-Mg montmorillonite | x |
| DV | x | x | illite/smectite with 30% of smectitic interlayers |

**Tab. 5 Selected physical properties of studied bentonites.**

**Tab. 5 Vybrané fyzikálne vlastnosti skúmaných bentonitov.**

| Sample | $w_L$ [%] | | $w_P$ [%] | $I_P$ [%] | A | $\rho_s$ [$g.cm^{-3}$] |
|---|---|---|---|---|---|---|
| | Casagrande | Fall cone | | Casagrande | | |
| JNa15 | 643.04 | 454.03 | 53.14 | 589.9 | x | 2.66 |
| JNa45 | 591.2 | 436.57 | 45.94 | 545.26 | x | x |
| J15 | 158.27 | 142.08 | 45.04 | 113.23 | 1.64 | 2.54 |
| J45 | 138.24 | 123.14 | 48.56 | 89.68 | 1.81 | 2.53 |
| J250 | 99.29 | 90.79 | 47.97 | 51.32 | 1.14 | 2.53 |
| K15 | 191.25 | 168.18 | 46.04 | 145.21 | 2.11 | 2.54 |
| K45 | 162.4 | 145.68 | 45.08 | 117.32 | 2.07 | 2.53 |
| L15 | 137.28 | 127.15 | 67.54 | 69.74 | 1.05 | 2.95 |
| L45 | 105.95 | 101.59 | 47.13 | 58.82 | 1.04 | 2.606 |
| L250 | 94.32 | 85.34 | 37.27 | 57.05 | 0.98 | 2.616 |
| LA45 | 130.76 | 114.64 | x | x | x | 2.5 |
| DV15 | 209.64 | 168.04 | 58.95 | 150.69 | 1.84 | 2.66 |
| DV45 | 83.09 | 66.15 | 34.65 | 43.64 | 1.02 | 2.641 |

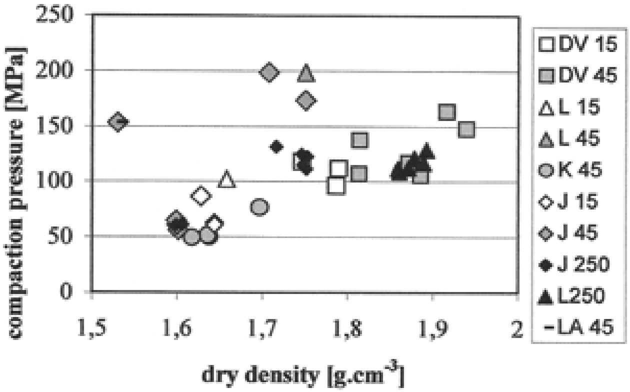


Fig. 5. Compactibility of studied bentonites.
**Obr. 5. Hutniteľnosť študovaných bentonitov.**

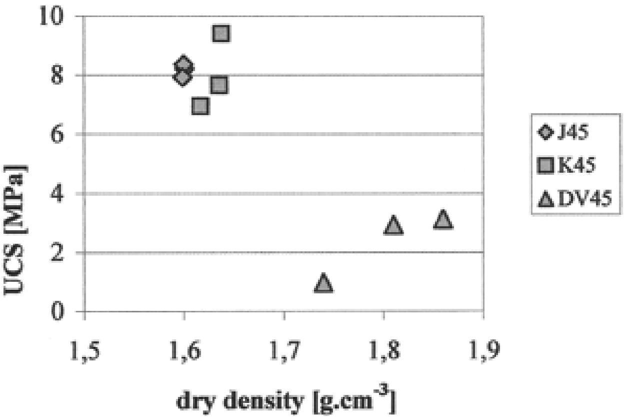


Fig. 6. Selected results of the uniaxial compression strength.
**Obr. 6. Vybrané výsledky pevnosti v jednoosom tlaku.**

where $h_1$ [mm] is the sample height before the test, $h_2$ after the test.

The hydraulic conductivity was derived from the consolidation test of a fully saturated sample according to Dananaj & Frankovská (2004) and STN 72 1027 (1983) In this method, also recommend by Marcial et al. (2001), Taylor's square-root calculation gave more reliable results then Casagrande's logarithmic calculation.

The uniaxial compressive strength was tested on standard 50 mm cubes or cylinders in a hydraulic jack (EN 1926, 2006). This strength is important for the durability of the pressed segments during manipulation and construction of the engineered barrier. Other mechanical tests were either not carried out (shear tests), or the test number is still too low until now (deformation tests).

Preliminary attempts were also made to study the effect of chemical agents and/or high temperature on the bentonite properties. Bentonite powder was treated with 1M $H_2SO_4$ or 1M KOH, well washed in deionised water, separated by centrifugation, dried at 50°C and milled again. One part of all bentonite samples was then heated at 120°C during 1, 2 or 3 months. Chemically and/or thermally pre-treated bentonite powder was used for determination of the Atterberg consistency limits, as well as for compaction and swelling tests.

### 3.3. Results

The liquid limit $w_L$ of the natrified bentonite $J_{Na}$ was too high (>600%), higher than reported for the Na-bentonite MX-80 (Studer et al., 1984). Samples J and K are very similar smectite type bentonites (Tab. 4); the highest clay activity $A$ and slightly higher plasticity of bentonite K (represented by $w_L$) can be due to a little higher total specific surface (Šucha et al., 2005). Depending on the grain size, the bentonite K with the highest content of exchangeable sodium yields $w_L$ from 160% to 190%, and bentonite J showed $w_L$ from 100% to 158%, which is similar to the Ca-bentonite Montigel tested in Switzerland (Studer et al., 1984). These values result from the 4-point Casagrande method that gave always lower figures than the fall cone method. Finer samples always show higher liquid limits (Tab. 5). The difference in $w_L$ between J and L is more evident in finer fractions. But J250 and L250 have similar plasticity, only the clay activity indicated a better suitability of J250. Plasticity of LA45 resembles J45, but it showed bad compactibility (Fig. 5) and coarser fraction (250) was not available, so its engineering-geological research was stopped.

Plots of the uniaxial compaction pressure relative to the dry density allowed a quantitative comparison of the compactibility (Fig. 6), but the quality of the pressed cylinders must be considered, as well. Friction on the bentonite-to-mould interface is a problem in all samples axially loaded in moulds (Akgün et al., 2006). The quality decreased and the pressure increased with the increasing content of both, the fine fraction and the moisture content in the powder. Highest dry densities were always reached with the coarse fraction 250, where the effective porosity is higher, allowing the air to escape from pores during compaction. In the finest fraction, air remaining in very small pores behaved as an elastic material, high compaction pressures were needed (up to 200 MPa). Consequently, the water (the sorbed atmospheric wa-

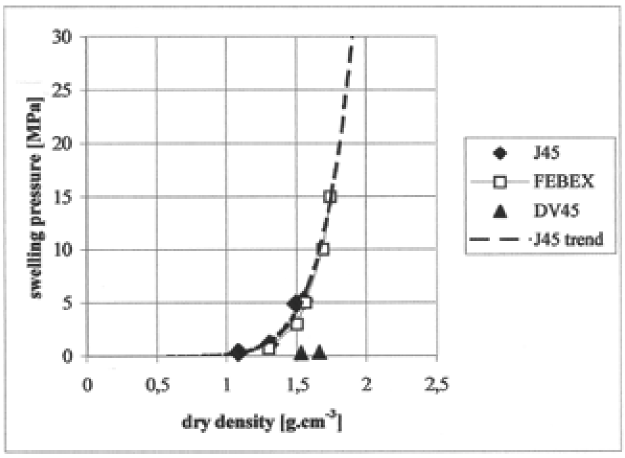


Fig. 7. Swelling pressure of J45 and DV45 compared with FEBEX bentonite.
**Obr. 7. Tlak napúčania vzoriek J45 a DV45 v porovnaní s bentonitom FEBEX.**

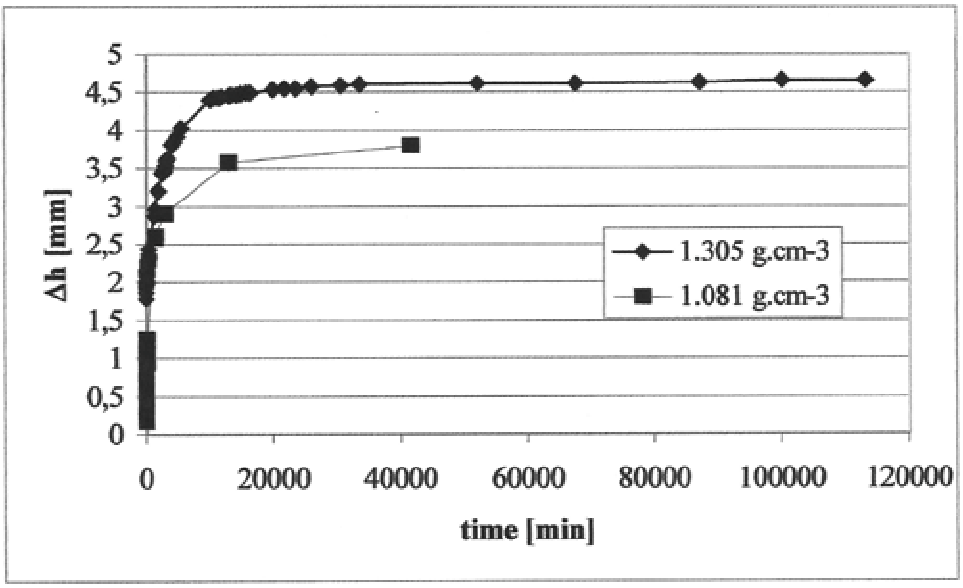


**Fig. 8. Uniaxial free swelling of the bentonite J45 at low initial dry density.**
**Obr. 8. Jednoosé voľné napúčanie bentonitu J45 s nízkou $\rho_d$.**

ter vapour) was pressed out from sample, which caused sticking of the wet bentonite surface to the mould, increased the friction, and induced shear cracks. Therefore, the finest fraction 15 was discarded and coarser fractions with limited risk of cracking will be preferred. At dry densities around 1.8 g.cm$^{-3}$, bentonite DV showed better compactibility than J and other bentonites due to lower smectite content, i.e. less moisture.

Tests of the uniaxial compressive strength (UCS) yielded values similar to soft rocks or weak rocks (from 1 to 10 MPa). If powders with more smectite and higher moisture content were used (J and K), compacted bentonite samples were stronger even at lower dry densities. Though natural bentonite DV45 is easier to compact, the quality of compacted natural bentonite J 45 and K 45 is better (Fig. 6). However, dried at 90°C, the same bentonites behaved as cohesionless soils and pressed samples lost readily their form, suggesting that a certain moisture content is necessary to reach a sufficient quality.

**Tab. 6 Compressibility of selected saturated bentonite samples (with low dry densities $\rho_d$) and their hydraulic conductivity.**
**Tab. 6 Stlačiteľnosť vybraných nasýtených bentonitových vzoriek (s nízkou objemovou hmotnosťou $\rho_d$) a ich priepustnosť.**

| Sample | Initial $\rho_d$ [g.cm$^{-3}$] | Normal load $\sigma_z$ [kPa] | Oedometer modulus $E_{oed}$ [kPa] | Consolidation coefficient $c_v$ [m$^2$.s$^{-1}$] | Hydraulic conductivity k [m.s$^{-1}$] |
|---|---|---|---|---|---|
| **J45** | 1.081 | 100 | 7.08 | 6.84E-08 | 9.66E-11 |
| | | 200 | 5.2 | 7.97E-09 | 1.53E-11 |
| | | 400 | 3.79 | 3.01E-09 | 7.95E-12 |
| **J45** | 1.305 | 100 | 9.14 | 5.15E-09 | 5.64E-12 |
| | | 200 | 6.92 | 4.29E-09 | 6.19E-12 |
| | | 400 | 8.1 | 4.77E-09 | 5.89E-12 |
| **DV45** | 1.659 | 100 | 8.08 | 1.38E-07 | 1.71E-10 |
| | | 200 | 16.15 | 2.08E-07 | 1.29E-10 |
| | | 400 | 24.52 | 3.89E-08 | 1.59E-11 |

As it is presented in Fig. 7, the test results of the bentonite J are very similar to those of the Spanish FEBEX bentonite (www.grimsel.com/febex), when a trendline is drawn into the chart, which means very good swelling. But the swelling capacity of bentonite DV is much lower. The relation between the dry density $\rho_d$ and the swelling pressure $p$ proved to be exponential, therefore good compaction is necessary. During free swelling, the very rapid start turned into a very slow process after short time (an example in Fig. 8).

Already at low dry densities of about 1.3 $g.cm^{-3}$, the hydraulic conductivity $k$ of the bentonite J did not exceed $6.2\times10^{-12}$ $m.s^{-1}$ (Tab. 6). This bentonite showed overall similar or better properties compared to Montigel (resp. Calcigel) (Plötze et al., 2002).

## 4. CONCLUSIONS

From the geological point of view, the Szécsény Schlier is a geological formation with favourable properties as a potential host rock for the deep geological disposal of the radioactive waste. These characteristics include lithological homogeneity, high dry density, low porosity, suitable geomechanical properties, low hydraulic conductivity etc. However, all these parameters are currently based on a relatively low number of observations carried out on material only buried to a depth of 250 m, or obtained by re-interpreting previous measurements and tests. Therefore, they need to be verified by a detailed site investigation and by laboratory tests of deeper rock samples.

The engineering geological laboratory research of five bentonite types showed that Slovakia has own bentonite deposits with suitable material for the buffer/backfill of the DGR. Both research phases had a comparative character, but some tests yielded already quantitative parameters comparable with FEBEX and Montigel bentonites tested abroad. It is clear at this point that bentonite from the Dolná Ves deposit has certain advantages: very good compactibility and, particularly, large amounts available in the deposit. The last is the main reason why a special attention is given to this bentonite type. However, it cannot compete with the bentonite from Jelšový Potok in the sealing effect, because its swelling potential is lower (Tab. 6). But no legislative dispositions were taken to protect the promising Jelšový Potok deposit, and if excavation continues at the present intensity, no more bentonite will be available there when it will be needed for the DGR (Šucha et al., 2005). Therefore, it is recommended to consider another type of bentonite. The good compactibility of the bentonite from Dolná Ves gives still certain hopes due to swelling pressure increasing exponentially with better compaction, i.e. with increasing dry density. However, swelling pressure at very high densities ($\geq 2$ $g.cm^{-3}$) was not tested yet, and its sorption capacity is low (Galamboš et al., 2009a, 2009b). Comparing only physical and mechanical properties of all studied bentonites, succession $DV<La\cong L<K\cong J$ can be written. Kopernica bentonite is also similar to Jelšový Potok bentonite from the mineral composition and stability point of view. Bentonites from Lieskovec and Lastovce deposits have also other disadvantages, mostly of mineralogical/chemical instability (Šucha et al., 2005). As soon as the bentonite type is selected, more tests are necessary for an exact definition of the mechanical properties. Beside its basic behaviour under mechanical load, the study of the thermo-physical properties and thermal load consequences are programmed.

**Acknowledgment:** We acknowledge the Ministry of Education of the Slovak Republic and ENVIGEO, J.S.C. for funding the project of the R&D State programme No. 2003 SP 26/028 0C 00/028 0C 02 and the project VEGA Agency No 1/4045/07 and No 1/0413/09, Austrian Science and Research Liaison Office contributed with the project SK-04-BA-005. We also acknowledge Mgr. M. Gregor from FNS CU Bratislava for PXRD analyses of the host rock, as well as Ing. J. Svoboda, PhD. and Ing. R. Vašíček, PhD. from CEG TU Prague for their swelling tests.

## References

Alba M. D., Chain P. & Orta M. M., 2009: Chemical reactivity of argillaceous material in engineered barrier: Rare earth disilicate formation under sub-critical conditions. *Applied Clay Science,* 43, 3-4, 369-375.

Alonso U., Missana T., García-Gutiérrez M., Patelli A., Siitari-Kauppi M. & Rigato, 2009: Diffusion coefficient measurements in consolidated clay by RBS micro-scale profiling. *Applied Clay Science,* 43, 3-4, 477-484.

Akgün H., Koçkar M.K. & Aktürk Ö., 2006: Evaluation of a compacted bentonite/sand seal for underground waste repository isolation. *Environmental Geology,* 50, 331–337.

Aranyossy J. F. (Ed.), 2007: Clay in Natural and Engineered Barriers for Radioactive Waste Confinement. Selected papers from Andra meeting in Tours, France, 2005. *Physics and Chemistry of the Earth, Spezial Issue,* 32, Elsevier, 965 p.

Aranyossy J. F. (Ed.), 2008: Clays in Natural and Engineered Barriers for Radioactive Waste Confinement (2007). Selected papers from 3rd Int. Meeting on Clay Confinement Properties, Lille, France (Sept. 2007). *Physics and Chemistry of the Earth, Spezial Issue,* 33, Elsevier, 543 p.

Bieniawski Z. T., 1989: Engineering Rock Mass Classification. John Wiley & Sons, New York, 251 p.

BS 1377: 1990: British Standard Methods of tests. Part 2: Soils for civil engineering.

Bucher F. & Müller-Vonmoos M., 1989: Bentonite as a containment barrier for the disposal of highly radioactive waste. *Applied Clay Science,* 4, 2, 157-178.

Dananaj I. & Frankovská J., 2004: Permeability of fine-grained soils. *Slovak Geological Magazine,* 10, 4, 305-312.

EN ISO 14689-1: 2003: Geotechnical investigation and testing. Identification and classification of rock. Part 1: Identification and description.

EN 1926: 2006: Natural stone test methods. Determination of uniaxial compressive strength.

EN 1936: 2006: Natural stone test methods. Determination of real density and apparent density, and of total and open porosity.

EN 13755: 2008: Natural stone test methods. Determination of water absorption at atmospheric pressure.

EN 14580: 2005: Natural stone test methods. Determination of static elastic modulus.

EN 1997-1: 2004: Eurocode 7: Geotechnical design. Part 1: General rules.

EN 1997-2: 2007: Eurocode 7: Geotechnical design. Part 2: Ground investigation and testing.

Galamboš M., Kufčáková J. & Rajec P., 2009a: Sorption of strontium on

Slovak bentonites. *Journal of Radioanalytical and Nuclear Chemistry,* 281, 347-357.

Galamboš M., Kufčáková J. & Rajec P., 2009b: Adsorption of cesium on domestic bentonites *Journal of Radioanalytical and Nuclear Chemistry,* 281, 485-357.

Ghorban A., Cosenza Ph., Revi A., Zamora M., Schmutz M., Florsch N. & Jougnot D., 2009: Non-invasive monitoring of water content and textural changes in clay-rocks using spectral induced polarization: A laboratory investigation. *Applied Clay Science,* 43, 3-4, 493-502.

Kaufhold S. & Dohrmann R., 2009: Stability of bentonites in salt solutions: sodium chloride. *Applied Clay Science,* 45, 3, 171-177.

Kováčik M., Hraško L., Ivanička J., Kohút M., Madarás J., Nagy A., Siman P., Töröková I., Hók J., Gedeon M., Helma J., Marcin, D., Švasta J., Durmeková T., Frankovská J., Král M., Liščák P., Petro L. & Wagner P., 2001: Hodnotenie študijných lokalít – 1. časť. Projekt: Vývoj hlbinného úložiska. Číslo etapy: VYL-01-00. Štátny geologický ústav Dionýza Štúra, Bratislava, 310 p.

Maheľ M., 1986: Geologická stavba československých Karpát. Paleoalpínske jednotky. Veda – SAV, ŠGÚDŠ, Bratislava, 503 p.

Marcial D., Delange P. & Cui Y. J., 2001: Compressibility and permeability of two swelling clays under high pressure. In: Adachi & Fukue (eds.): Clay Science for Engineering. Balkema, Rotterdam, 571-578.

Matejovič I., Hók J., Madarás J., Slaninka I. & Prítrský J., 2006: Status of Deep Geological Disposal Programme in the Slovak Republic, In: Witherspoon, P.A., Bodvarsson, G.S. (Eds.): Geological Challenges in Radioactive Waste Isolation. Fourth Worldwide Review. California, USA, LBNL-59808, 173 – 189.

OECD NEA, 2003: SAFIR 2: Belgian R&D Programme on the Deep Disposal of High-level and Long-lived Radioactive Waste. OECD Publications. 77 p.

Plötze M., Kahr G. & Hermanns-Stengele R., 2002: Alteration of clay minerals in long-term nuclear waste repositories – influence on physicochemical properties. In: Di Maio C. , Hueckel T. & Loret B. (Eds.): Chemo-mechanical coupling in clays. Balkema, 325-337.

Pusch R., 2001. SKB Technical Report TR-02-12. The Buffer and Backfill Handbook, Part 2: Materials and techniques. Geodevelopment AB, 198 p.

STN 72 1027: 1983: Laboratory determination of soil compressibility in the oedometer apparatus.

Studer J., Ammann W., Meier P., Müller Ch. & Glauser E., 1984: Verfüllen und Versiegeln von Stollen, Schächten und Bohrlöchern. Technischer Bericht 84-33. NAGRA, Baden, 220 p.

Sun D., Cui H. & Sun W., 2009: Swelling of compacted sand-bentonite mixture. *Applied Clay Science,* 43, 3-4, 485-492.

Šucha V., Adamcová R., Bujdák R., Haasová Z., Honty M., Komadel P., Kufčáková J., Madejová J., Rajec P., Stríček I., Uhlík P. & Valúchová J., 2005: Fyzikálne a mechanické vlastnosti tesniacich materiálov pre úložisko RAO. Čiastková úloha projektu SP 26/028 0C 00/028 0C 02 Štátneho programu výskumu a vývoja Záverečná správa. Univerzita Komenského, Prírodovedecká fakulta, Bratislava, 281 p.

Vass D., 2002: Litostratigrafia Západných Karpát: neogén a budínsky paleogén. ŠGÚDŠ, Bratislava, 202 p.

Witherspoon, P.A. & Bodvarsson, G.S. (Eds.), 2006: Geol. Challenges in Radioactive Waste Isolation. Fourth Worldwide Review. California, USA, LBNL-59808, 283 p.

**Resumé:** Nájdenie vhodnej geologickej štruktúry a geologických materiálov pre vybudovanie bezpečného trvalého úložiska vysokorádioaktívnych odpadov a vyhoreného jadrového paliva (RAO) je jednou z najnáročnejších geologických úloh súčasnosti. Na Slovensku regionálny výskum pre umiestnenie hlbinného úložiska (HÚ) RAO začal až v 90-tych rokoch minulého storočia. Po predbežnom výbere dvanástich lokalít sa prieskum sústredil na štyri lokality v území budovanom granitoidnými horninami a dve v neogénnych sedimentárnych formáciách. Článok v prvej časti hodnotí vytypované neogénne ílovce a siltovce ako hostiteľské prostredie a potenciálnu prirodzenú geologickú bariéru pre šírenie znečistenia z HÚ RAO. Druhá časť článku sa zaoberá výskumom vybraných slovenských bentonitov ako materiálu pre inžinierske bariéry v HÚ RAO.

Na obidvoch vytypovaných lokalitách v sedimentárnych horninách juhoslovenskej panvy sa nachádza séčenský šlír Lučeneckej formácie (Obr. 1). Ide o pomerne litologicky monotónne vápnité ílovité a prachovité sedimenty čiastočne spevnené, s nepravidelne sa vyskytujúcimi vložkami piesčitej frakcie. Neogénne súvrstvia sa vyznačujú dostatočnou hrúbkou (vrtmi overenou do 700 m, predpokladanou až do 1 300 m), nepatrným tektonickým porušením, nízkou priepustnosťou. Hodnoty priemerných vlastností všetkých testovaných vzoriek zodpovedajú kritériám, charakterizujúcim poloskalné horniny. Objemová hmotnosť v suchom stave je 2,13 až 2,25 g.cm$^{-3}$, pórovitosť je 15 až 21%, hodnoty pevnosti v prostom tlaku sa pohybujú v rozmedzí 18,5 až 30 MPa. Treba upozorniť na skutočnosť, že ide o materiál veľmi citlivý na kontakt s vodou. Koeficient filtrácie bol zistený v rozsahu $10^{-10}$ m.s$^{-1}$ až $10^{-11}$ m.s$^{-1}$. Uvedené údaje poskytujú základnú informáciu o fyzikálno–mechanických vlastnostiach v rôznych hĺbkových horizontoch v rámci doterajšieho prieskumu. Nevýhodou je ich nízka pevnosť a vysoká pretvárnosť. Všetky vlastnosti horninového prostredia boli doteraz zisťované len v laboratóriu, len na obmedzenom počte vzoriek z vrtných jadier, z hĺbky maximálne do 250 m, kam siahali vrty. Preto je v tomto štádiu prieskumu ešte predčasné robiť konečné závery ohľadom vhodnosti hodnotených neogénnych sedimentov ako hostiteľského prostredia HÚ RAO.

Za účelom budovania inžinierskych bariér HÚ RAO z domácich materiálov boli laboratórne testované práškové bentonity (zrnitosť <0,250 mm, <0,045mm a 0,015mm) z ložísk Jelšový potok, Kopernica, Lieskovec, Lastovce a Dolná Ves, avšak nie v rovnakom rozsahu. Najdôkladnejšie sú po stránke fyzikálnych a mechanických vlastností preskúmané bentonity s najväčšími rozdielmi, z Jelšového potoka a Dolnej Vsi. U všetkých boli určené Atterbergove konzistenčné medze (Tab. 5), z ktorých najmä vysoká medza tekutosti indikuje vysoký obsah smektitu, vysokú napúčavosť a schopnosť samohojenia prípadných trhlín. Avšak extrémne vysoká medza tekutosti diskvalifikovala natrifikovaný íl z Jelšového potoka $J_{Na}$. Napúčavosť prudko stúpa s rastúcou hustotou vzorky, preto zlá hutniteľnosť a kvalita výliskov spolurozhodovali pri vylúčení jemne mletých bentonitov z druhej etapy výskumu. Ukázalo sa, že na ich zhutnenie na požadovanú hustotu $\rho_d$ okolo 1,8 g.cm$^{-3}$ je v dôsledku nízkej efektívnej pórovitosti potrebný tlak až do 200 MPa. Ten spôsobil presun adsorbovanej vzdušnej vlhkosti k povrchu výliskov, ktorý sa rozmočil a priľnul k lisovacej forme. Nárast trenia na kontakte bentonit/forma následne porušil zhotovované výlisky strihom. Naopak dobrú hutniteľnosť, teda na vysokú hustotu už pri nízkom tlaku, vykazovali najhrubšie vzorky, ako aj vzorky s nízkym obsahom smektitu (Obr. 5, Tab. 4). Ich výlisky však nie sú tak pevné ako z kvalitnejšieho bentonitu s vysokým obsahom smektitu, kde vyššia vlhkosť zvyšuje súdržnosť materiálu (Obr. 6). Vcelku sa však výlisky pevnosťou v jednoosom tlaku 1 až 10 MPa vyrovnali poloskalným horninám. Kvalitný bentonit z Jelšového potoka potom vykazoval pri rovnakej hustote ako bentonit z Dolnej Vsi aj vyšší tlak napúčania (Obr. 7) a už pri hustotách okolo 1,3 g.cm$^{-3}$ aj dostatočnú nepriepustnosť (koeficient filtrácie 6,2×10$^{-12}$ m.s$^{-1}$). Vyrovná

sa zahraničným bentonitom FEBEX a Montigel. Ak hodnotíme iba fyzikálne a technologické vlastnosti, vhodnosť študovaných bentonitov do HÚ RAO stúpa v rade Dolná Ves < Lastovce ≅ Lieskovec < Kopernica ≅ Jelšový potok. Ložisko Jelšový potok je však ťažené tak intenzívne, že hrozí predčasné vyťaženie zásob. Práve veľké zásoby na ložisku Dolná Ves boli dôvodom, prečo bola tomuto bentonitu venovaná toľká pozornosť. Jeho dobrá hutniteľnosť dáva ešte nádej, že pri extrémne vysokej hustote výliskov ($\rho_d$>2 g.cm$^{-3}$) by exponenciálne závislý tlak napúčania mohol dosiahnuť potrebnú úroveň, treba to však experimentálne overiť. Zhodné alebo veľmi podobné fyzikálne vlastnosti má v porovnaní s bentonitom z Jelšového potoka aj bentonit z Kopernice. Ostatné dva majú horšiu kvalitu. Skúšky mali doteraz prevažne orientačný komparatívny charakter, ale poskytli aj viacero kvantitatívnych údajov. Po výbere ložiska ich bude nutné upresniť, najmä ukazovatele mechanických vlastností k dimenzovaniu inžinierskych bariér.