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**THE EVOLUTION OF THE GYÓD SERPENTINITE FORMATION
AND ITS ENVIRONMENTAL GEOLOGICAL ASPECTS**

PhD thesis

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REASON FOR RESEARCH

The political and economic changes in the late '90s caused obscurity in the deposition of the high level nuclear waste (HLW) of Paks Nuclear Power Plant (NPP). Hungary had to turn over the strategy for the definite closure of the nuclear fuel cycle. The capacity of the operating repository in Püspökszilágyi is not enough to dispose the nuclear waste of NPP. The country has to reposit the high, intermediate and low level nuclear waste within the state border into a deep level repository in accordance with the national directives of the IAEA (International Atomic Energy Association). The currently spent fuel is stored in the Interim Storage Facility (ISF) within the area of the NPP. The official permission of storage allowance is for 50 years. After this relaxation phase of the HLW is placed in a well engineered and well chosen underground repository that provides additional protection from surface hazards.

According to the national practice, the Geographical Research Institute of the Hungarian Academy of Sciences highlighted several possible formations near the well-studied Boda Aleurolite Formation, which was also revealed by mining. This report gave forth for the Gyód Serpentinite Formation as a possible mass for repository of nuclear waste on the basis of the literature.

The goal of the present thesis work was to evaluate the rock bodies of the Gyód Serpentinite Formation with regards to its suitability for long-term nuclear waste reposition.

STUDY AIMS

One basic expectation for the underground repository is that it has to be able to isolate perfectly the nuclear waste for hundreds or thousands of years depending on the level of the nuclear waste.

Obligatory tasks are listed in the screening plan for determining a capable mass that far exceeds the volume of a PhD thesis. That is why only a restricted subject of the tasks was chosen for further investigations. The goal of the PhD was to survey the suitability of the Gyód mass for nuclear waste storage in a petrological sense. To realize this aim I gave all my attention to examine the petrography, petrology, deformation and structural features of the serpentinite body, but I was not concerned with the hydrogeology, nuclear migration features, tectonics and seismicity.

The goal of the thesis was reached with the following steps:

1. The heterogeneity of the serpentinite body was mapped by the investigation of the mineral composition and the geometric relations (texture) of the rock-forming minerals. The generations of the mineral paragenesis were distinguished by the textural and microtextural examinations. I classified the different rock types making up the serpentinite body.
2. The rock-forming minerals were analysed and their chemical composition are useful to calculate the physical parameters of all mineral paragenesis, which were determined by thermometric calculations.
3. Ductile and brittle deformations were described and classified at the meso and micro scale on the core samples. These results were synthesized with the results of the thermometric calculations with the aim to reconstruct the deformation history of the serpentinite body.
4. The traces of minerals during rock–fluid interaction process and the element mobilization, as well chemical alterations were examined by chemical analyses.
5. I determined the tectonic zones and fluid conduits of the classified samples by rock physics methods.
6. I developed a structural model of the serpentinite body which represents anisotropy of the rock in the light of its evolution. This model is also suitable for a hydrogeological flow simulation.

METHODS

At first the samples were described macroscopically, and a 100 slides of oriented thin sections were prepared for microscopic examinations. The thin section collection of the Department of Mineralogy, Geochemistry and Petrology, University of Szeged and applied collection of the thin sections (80 pieces) and thin section (20 pieces) of Gábor Papp were re-examined.

The composition of minerals was determined by X-Ray Diffractometry (XRD), electronmicroprobe, electronmicroscope and Raman spectroscopy. Carbonate minerals were determined in thin sections by the staining method of Evamy (1963). XRD examinations were carried out at the Department of Mineralogy, Geochemistry and Petrology, University of Szeged and at the Department of Environmental Science, University of Pannonia. Hand picked and whole rock samples, including on fraction less than 2 μm were analysed.

Microprobe analyses were performed at the Institute for Geochemical Research of Hungarian Academy of Sciences and at the University of Toronto. Samples were analysed by Raman spectrometry at the University of Leoben. The electronmicroprobe investigations were run by scanning electronmicroscope at the University of Szeged.

The chemical compositions (major and trace elements) were determined in the following laboratories:

Nine samples were analysed in the Geological Institute of Hungary by ICP-OES technique and five samples at the University of Pannonia by XRF. Furthermore eight samples at the Research Institute for Soil Science and Agricultural Chemistry of the Hungarian Academy of Sciences by ICP-AES technique. Trace and REE in five samples were analysed by XRF and mass spectrometry at the XRAL Laboratory in Toronto.

The physical parameters of the rocks were determined at the Pollack Mihály Faculty of Engineering, University of Pécs and the Laboratory of Non-Profit Company for Quality Control and Innovation in Buildings. The computer tomography investigations were performed at the Diagnostic and Radiation Oncology Institute, University of Kaposvár

RESULTS

1. I detected the spatial change of serpentinite types within the Gyód Serpentinite body in relation to the position of the shear zones.

I typified the serpentinite based on representative samples during examining the core samples of the entire borehole. I separated the schistose and the bastite types of typical serpentinite, and the ultramafic-bearing serpentinite. Based on the location of the different types, I determined that they are similar to the kernel structure described by O'Hanley and Offler (1992). In relation to the position of the shear zones and at the same time of the fluid migration trajectories, the decrease of the effect of the infiltration-driven metamorphism can be detected.

An amphibole-talc containing shear zone can be found in the borehole at 105 m. In the immediate vicinity of that zone schistose serpentinite, a bit farther bastite serpentinite and ultramafic-bearing serpentinite can be found, respectively at increasing distances.

2. Based on the mineralogical, textural and geochemical examinations, I identified the protolith of Gyód Serpentinite as an exhausted, of upper mantle origin, harzburgitic peridotite.

The protolith determination of serpentinite varies by authors. Szederkényi (1974a, 1976b, 1977a, b) determined it as pyroxenite, and dunite. According to Ghoneim & Ravasz Baranyai (1969), Ghoneim (1978, 1979), the starting rock could have been lherzolite, pyroxenite. Finally Balla (1983, 1985) determined the precursor rock as olivine–enstatite containing ultramafic rock, harzburgite. Based on the mineralogical and geochemical examinations, the Gyód Serpentinite is homogeneous; I determined its protolith as a mantle derived harzburgite. The primary igneous mineral paragenesis of harzburgite is made up of clinoenstatite–enstatite–olivine–chromite. The sample previously determined as aplite is in fact an ultramafic cumulate (SiO_2 content <42 w%). According to geochemical features, the protolith is a residuum of partial melting of rock containing small amount of basalt components. The high 89–91 Mg#, and the olivine 0.06–0,3 w% NiO content of the rock unambiguously proves its mantle origin, and difference to cumulates.

3. I justified the formation and multiphase retrograde metamorphism of the protolith under upper mantle conditions with thermometric calculations.

According to thermometric calculations and modelling the protolith was formed between 1030–1080 °C and under about 8.5 kbar PT. In the upper mantle harzburgite was exposed to several deformation events and metamorphism. The first detectable change in the mineral components was that clinoenstatite altered to enstatite. Deformed olivine porphyroclasts and chromite grains refer to plastic deformation under upper mantle PT conditions. Small enstatite grains (En₂) with polygonal texture formed by enstatite recrystallisation preserve the traces of high temperature metamorphism.

Anthophyllite deriving from enstatite proves the gradual rising of protolith from upper mantle conditions. Based on thermometric calculations and the composition of the edge zone of enstatite porphyroclasts, an event between 630–680 °C can be detected, which coincides with the formation temperature of amphiboles. Based on the in-out reaction, pressure may have been under 4 and above 2 kbar. During their formation process, amphiboles took up an oriented array along the S₀ foliation.

Talc appearing after the metamorphism of pyroxene–amphiboles at about 400–500 °C proves the increasing hydration during further rising.

4. The uplift of the protolith into the oceanic crust level was confirmed where it suffered a static hydration by subseafloor metamorphism.

The next stage of the evolution of the protolith is the subseafloor metamorphism which took place in the high level of the oceanic crust. The protolith emplaced by tectonically to this zone was altered by the seawater. This process results in a pseudomorphic texture proving that serpentinization took place under static circumstances. Hydration of the ultramafic rocks shows textural differences between domains. The olivine-bearing domain alters to mesh textured serpentinite while bastites form from pyroxene–amphibole-bearing domain. During serpentinization lizardite–chrysotile–clinochlore–magnetite develop from the primary rock-forming minerals. The serpentinization took place at 230-250 °C on the basis of chlorite geothermometry.

The serpentinization was an isochemical process with respect to the SiO_2 , MgO , CaO , TiO_2 , NiO oxides. I pointed out a decrease in the amount of FeO , MnO , Na_2O and an enrichment of Fe_2O_3 , Al_2O_3 .

5. The intimate relationship between structural evolution and hydration was corroborated by findings of the history of deformations.

First of all I examined the deformation features and veins of the Gyód Serpentinite body. Four generations were distinguished of the serpentine veins and a posttectonic carbonate veins. Each generation of veins belongs to a period of serpentinization. The first generation of serpentine veins (E1) formed in the early period of serpentinization when olivine started to decay, which marked the magnetite grains in the vein. The deformation space was different during its formation from the other generations'. The second generation of the veins formed during the main phase of serpentinization, when the rate of volume increasing should have been the highest. These parallel chrysotile veins crosscut the whole body except for the partly serpentinized ultramafic rocks, which proves the role of volume increasing on their formation. The E2 veins show sigmoidal set of en échelon texture which demonstrates shearing. The third generation of veins are banded crack-seal veins formed during the unroofing of the peridotites. The periodicity of tectonism caused banded structure of the veins. During their formation the concomitant shearing resulted in sigmoidal, antitaxial microtextures. The formation of S1 foliation took place in the main phase of the serpentinization. Milonitization was pointed out in the serpentinites belonging to these shear events. The fourth generation of serpentine veins (E4) are characterised by composite, banded and fan-like structures. In the host rock needle shape serpentines (antigorite) occur which represent a prograde metamorphism in contrast to a retrograde evolution of the serpentinite body until this point. The formation of antigorite is the outcome of dehydration.

6. I confirmed the subduction related progressive metamorphism in both of the serpentinite and the S2 foliation zones.

The S2 foliation crosscuts steeply the main foliation (S1). Antigorite and talc build up the S2 zones referring to the dehydration of the serpentinite. From the thermodynamic models it's quite clear that this dehydration process occurred at higher PT conditions than those characterizing the regular serpentinization. The S2 foliation trajectories must thus correspond to the dehydration channels enabling the escape of excessive water. The precipitation of antigorite and talc can be linked to the zones. The increasing temperature also results changes in the texture of serpentinite because the pseudomorphic texture was recrystallised into interlocking and interpenetrating textures, containing antigorite. The increasing temperature was identified as to be related to subduction on the basis of the possible geotectonic environments. During the subduction process the Gyód Serpentinite body did not cross the isotherm of 500 °C.

7. I detected that the evolution of the Gyód Serpentinite body is different from the metamorphic evolution of the crystalline basement of South Transdanubia.

Király (1996) identified the first metamorphism of the polymetamorphic crystalline basement as that of described by Szederkényi (1974a): on Görcsöny Mount 5-7 kbar, 552-648 °C, supposedly in West Mecsek 6-8 kbar, too, 519-603 °C PT features. The second phase took place at the same time of granite formation, which is of intermediate grade, small pressure metamorphism of the restites (<2 kbar, 416-560°C). The third event is potassic–metasomatism resulting in biotite and microcline formation, which is in connection with the Mórággy granite occurrences. The forth and last event is retrograde, low temperature phase, which touched upon the whole area. The evolution of Gyód Serpentinite body cannot be connected to the evolution of the crystalline basement of South Transdanubia. The serpentinite is tectonically interposed in its environment, just like most of the Alpine-type ultramafics.

8. I detected with rock physics examinations that the structures forming during the evolution of Gyód Serpentinite can function as fluid migration paths despite the impermeable features of the rocks. According to this present examination, the Gyód Serpentinite is not suitable for isolating nuclear waste.

During the rock physics examinations I detected that the rheological features of the typified serpentinites are different. The serpentinite containing ultramafics has the highest mechanical resistance despite its foliation. The samples showing S1, S2 foliation are harder to break than

the serpentinites densely foliated along the S1. The structures may be renewed along the foliation when exposed to deformation. I detected that relict ultramafics has the lowest porosity in the serpentinite body, so it can be referred to as impermeable rock just as the serpentinite matrix as well.

During the saturation CT measurements flows were detected along both S1 and S2 foliation planes. The two directions differ in their genetics and composition. The stream is stronger along the S2 foliation since talc has a significant role in forming the zone. The permeability and reaction to deformity of talc containing zones in the serpentinite is different in relation to its surroundings (Boschi et al., 2006, Escartín et al, 2001, 2004, Searle & Escartín 2004). The reason is that talc – considering its rheological features – is a particularly weak mineral.

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