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PRINCIPLE ROCK TYPES FOR RADIOACTIVE WASTE REPOSITORIES

GLAVNI TIPOVI STIJENA ZA ODLAGALIŠTA RADIOAKTIVNOG OTPADA

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Ključne riječi: odlagališta radioaktivnog otpada, evaporiti, šejl, kristalinska podloga, Dinaridi

Abstract

Underground geological storage of high- and intermediate/low radioactive waste is aimed to represent a barrier between the surface environment and potentially hazardous radioactive elements. Permeability, behavior against external stresses, chemical reacatibility and absorption are the key geological parameters for the geological storage of radioactive waste. Three principal rock types were discussed and applied to the Dinarides: (1) evaporites in general, (2) shale, and (3) crystalline basement rocks. (1) Within the Dinarides, evaporite formations are located within the central part of a Carbonate platform and are inappropriate for storage. Offshore evaporites are located within diapiric structures of the central and southern part of the Adriatic Sea and are covered by thick Mesozoic to Cenozoic clastic sediment. Under very specific circumstances they can be considered as potential site locations for further investigation for the storage of low/intermediate level radioactive waste. (2) Thick flysch type formation of shale to phyllite rocks are exposed at the basement units of the Petrova and Trgovska gora regions, whereas (3) crystalline magmatic to metamorphic basement is exposed at the Moslavačka Gora and Slavonian Mts. regions. For high-level radioactive waste, basement phyllites and granites may represent the only realistic potential option in the NW Dinarides.

Sažetak

Podzemno odlaganje visoko i srednje do nisko radioaktivnog otpada predstavlja barijeru između površinskog okoliša i potencijalno opasnih radioaktivnih elemenata. Neki od ključnih geoloških parametara za odlaganje radioaktivnog otpada su permeabilnost, ponašanje prilikom opterećenja, intenzitet kemijskih reakcija i apsorpcija. U članku diskutiramo tri pogodna tipa stijena za odlagališta: (1) evaporiti, (2) šejl, i (3) stijene kristalinske podloge, čije predstavnike pronalazimo i u Dinaridima. (1) Evaporitne formacije smještene su na kopnu duž centralnog dijela karbonatne platforme i u podmorju unutar dijapirskih struktura centralnog i južnog dijela Jadranskog mora. Evaporiti na kopnu sastoje se od gipsa i anhidrita i nepogodni su za odlagališta. Evaporiti u podmorju sastoje se od anhidrita i soli i mogli bi biti pogodni su za odlaganje nisko do srednje radioaktivnog otpada. (2) Debele formacije šejla i filita nalaze su u području Petrove i Trgovske gore, dok su (3) magmatsko-metamorfne stijene kristalinske podloge izložene u području Moslavačke gore i Slavonskih planina. Za odlagališta visoko radioaktivnog otpada, kristalinska podloga i filiti predstavljaju jedine moguće lokacije unutar Dinarida.

1. Introduction

Radioactive waste is classified into low-/ intermediate and high-radioactive wastes, which differ in terms of their hazard for the environment and consequently different criteria for the geological storage must be met (IAEA, 2009). E.g., for high radioactive waste, an increase of temperature is predicted during storage, and the temperature could increase to up to 125 °C. Depending on their class, radioactive waste is usually packed as solids in canisters or barrels, which are stored in an underground environment. For a safe long-term storage, considerations of the development of future societies are also necessary (e.g., Hora and Winterfeldt, 1997).

The disposal of radioactive waste in the deep geological environment is required to ensure a safe long-term storage. The rock type and formation should represent a safe geological barrier between the radioactive waste and the surface environment. The Blue Ribbon Commission on America's Radioactive Future (2012) writes … *"deep geological disposal is the most promising and accepted method currently available for safely isolating spent fuel and high-level radioactive wastes from the environment for very long periods of time"* (Hamilton and Scowcroft, 2012). The isolation from the environment is in the range of a hundred thousand to a million years, and this cannot be guaranteed on the Earth´s surface. The geological formation with the contained radioactive waste, especially high-level, should represent a sufficiently thick geological barrier (minimum 50 to 100 m thick strata) in a geologically stable region of more than 1 million years (e.g., Templeton et al., 2010), which is also relatively inert against chemical attack. Long-term stability also requires protection against erosion by glaciers, flooding etc., which may arise as the result of climate (Stuewe et al., 2009). This implies storage in the geologically safest environment of a particular country. The rock unit, in which radioactive wastes is stored, should not contain valuable resources, to avoid accidental exploration by a future society.

The deep geological repository should fulfill a number of criteria, which includes low permeability of the host rock, protection against water, stable storage, and long-term stability. For long-term storage, a number of geological formations and rock types were proposed, which include (1) salt (or evaporites in general), (2) shale, and (3) crystalline basement rocks (e.g., gneiss or granite).

In this contribution, we first discuss the above principal three concepts of host rocks and show some key parameters for the geological storage separately for (i) low-/intermediate and (ii) high-level radioactive waste. Finally, we apply each of the proposed rock types to the Dinarides.

2. Principle rock types for geological storage of radioactive waste

As already stated, there are three principle rock types for the storage of radioactive waste. Each of the storage rock types has its specific advantages and disadvantages, and the proposals vary from country to country. Some key parameters for storage of radioactive waste in these principal rock types are shown in Figure 1 and some international examples are mentioned.

2.1. Evaporites

Evaporite rocks are usually deposited in hot arid environments, mostly in lagoons. A specific sequence of precipitation is observed, which usually includes carbonates at the base (limestone, dolomite), gypsum, halite as the main rock type, and a thin complex of Mg-K sulphates and K-chloride at the top. In most cases, the precipitation stops at the level of gypsum or halite, and only minor evaporate successions reach the level of K-chloride and K-sulphate precipitation (Warren, 2006). For example, salt bodies of the Eastern Alps, the so-called Upper Permian to Lower Triassic Haselgebirge Fm., include lenses of various sulphates, in which the different structural behavior of halite rocks and embedded sulphate lenses can be studied (e.g., Leitner and Neubauer, 2011; Schorn and Neubauer, 2011). Each of the evaporite minerals has their specific behavior under natural conditions and will be discussed here.

Gypsum [$CaSO_4 \times 2H_2O$] is the most widespread evaporite mineral. At elevated temperatures, gypsum converts to bassanite [$2CaSO_4 \times H_2O$], which is metastable and decomposes to anhydrite [$CaSO_4$]. This reaction is frequent as the depth (i.e. pressure and temperature) of an evaporite formation exceeds 64 °C (Murray, 1964) or max. 85 °C due to overburden (Yamamoto and Kennedy, 1969). The decomposition of gypsum to bassanite and finally to anhydrite reduces the volume by 40 % and releases much water, which results in rheological weakening and mechanical destabilization of evaporite bodies (e.g., Urai et al., 1986) as well as the formation of chemically aggressive brines. This makes gypsum inappropriate for any kind of radioactive waste storage.

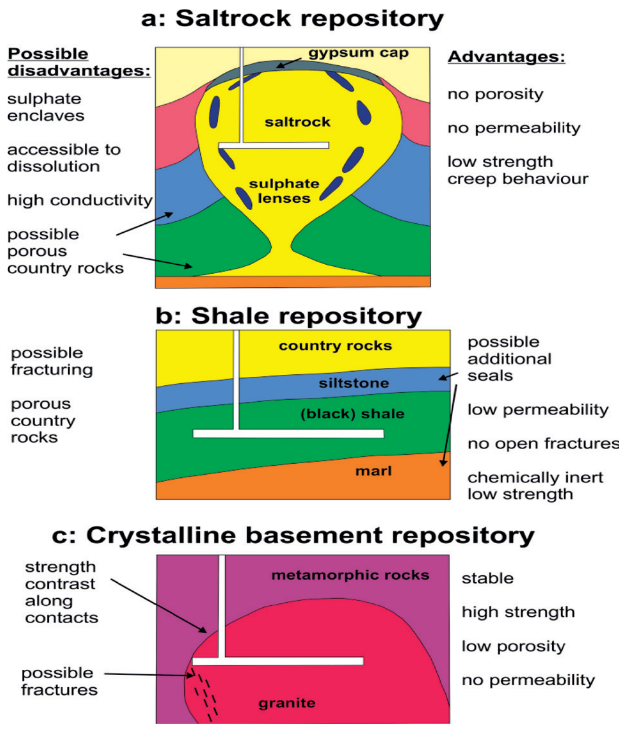

Figure 1. Overview on underground geological storage of radioactive waste. a – Salt diapirs. b – Silty to clayey shale/slate (phyllite). c – Crystalline rocks.

Slika 1. *Pregled podzemnih odlagališta radioaktivnog otpada. a – Solni dijapir. b – Siltozni do glinoviti slejt/šejl (filit). c – Kristalinske stijene.*

Anhydrite [$CaSO_4$] is thermally stable and is potentially good storage stratum for low-/intermediate level radioactive waste (Fig. 2). However, anhydrite behaves brittle and can bear joints and even open fractures. Such fractured anhydrite is a potential pathway for fluids and gas, particularly when fracturing is severe because of tectonic stresses. Anhydrite in reaction with water is transformed to gypsum with the increase of volume of some 40%.

Thick anhydrite layer can be considered as a repository for low-/intermediate level radioactive waste if fulfilling the following two requirements: (1) location above valleys, e.g. in a mountain, where the groundwater level is deep, and (2) a seal both at the top and base to protect the anhydrite layer from water inflow (Fig. 2).

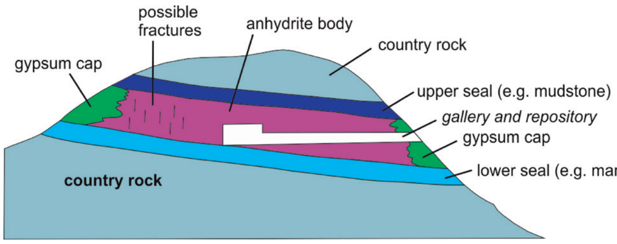

Figure 2. A sulphate body as a repository for low-/intermediate level radioactive waste.

Slika 2. *Odlagalište nisko i srednjeradioaktivnog otpada unutar sulfatnog tijela.*

Halite (NaCl) itself remains thermally stable over the range of temperatures expected in radioactive waste repositories (Figure 1a). The main advantages of halite include: low porosity resulting in a geologically perfect seal, creep behavior of halite even at room temperatures, which enables closure of open fractures. Disadvantages are the high heat conductivity and the high solubility of halite in water. Locations in arid climates are preferred, and the classical example is the Carlsbad site in New Mexico in the United States (Rechard, 2000; Goldstein, 2011). The additional problem and potential hazard is the gypsum/anhydrite which is often embedded within halite in evaporite formations.

2.1.1. Evaporite tectonics

Evaporite bodies, specifically salt bodies, are often used for the geological storage of high-level radioactive wastes. Composition, internal and external structure are important parameters for the understanding of the evaporite bodies as a geological barrier. The long experience in salt mining and oil exploitation related to salt diapirs argue for good knowledge of mechanical and geotechnical properties (Fossen, 2011; Hudec and Jackson, 2007).

Evaporite bodies can become mobile when they reach a high thickness (> 200 m) and under a thick overburden (usually > 1 km). The essential properties for mobility are the extremely low yield strength of halite even at room temperature (e.g., Urai et al., 1986) and density inversion. Usually, a tectonic trigger, extensional or compressional, is responsible for salt mobility. Preferentially halite flows from the concordant stratum to a salt wall, and later to salt diapirs. The resulting bodies are salt complexes or salt diapirs. The process of salt flow takes place until welding of the hangingwall to the footwall in the feeding subhorizontal stratum occurs, and no more salt feeds the diapir. The growth of the diapir stops and the diapir remains in a relatively stable position (Hudec and Jackson, 2007; Fossen, 2006). The structural history also influences the possible culmination of diapir growth, and this is extensional versus compressional history. A salt diapir under compression can also loose the connection to the feeding stratum, and diapir growth, therefore, stops. Figure 3a, b shows a typical salt diapir, Gorleben in Germany, which is under consideration for the storage of high-level radioactive waste (Jobmann et al., 2011; Schneider, 2011).

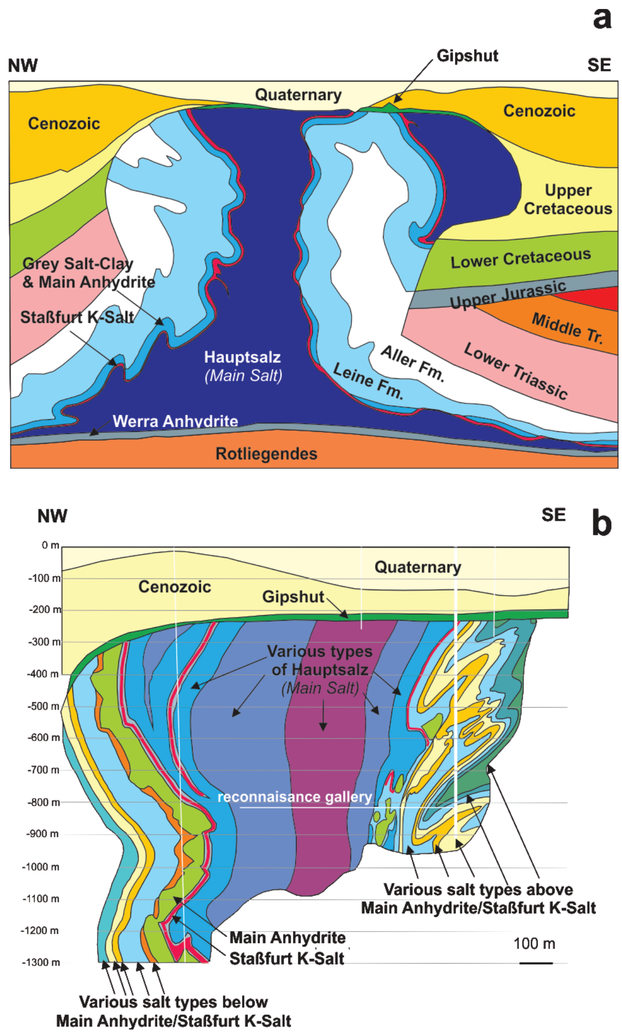

Figure 3. The Gorleben example (Germany) as a radioactive waste repository. a – At depth, a partly hypothetical section showing welding of feeding Zechstein stratum. b – Cross-section at the repository level (from www.endlagerung.de finally accessed February 25, 2012).

Slika 3. *Primjer odlagališta nuklearnog otpada u Gorlebenu (Njemačka). a – Djelomično hipotetski prikaz prestanka prihranjivanja Zachstein naslaga u dubini. b – Poprečni presjek odlagališta (podaci preuzeti iz www.endlagerung.de 25.02. 2012)*

The internal structure of evaporite bodies is usually complex (Hudec and Jackson, 2007; Fossen, 2011). A salt diapir often contain one to ten meters thick lenses of gypsum, anhydrite, limestone and other country rocks, embedded during salt flow. Such lenses, may be traced over hundreds of meters. They remain stiff and brittle within rheologically weak halite representing the main problem in the geotechnical treatment during salt mining, salt exploitation and waste disposal (Warren, 2006). Such lenses can acquire open fractures and are accessible for fluid flow through the rock body if connectivity between such bodies exists. Also, they have the tendency to slowly move downwards through the salt body (density: ca. 2.17 g/cm^3) because of the higher density (e.g. $\rho_{anhydrite} = 2.97$ g/cm^3; $\rho_{gypsum} = 2.36$ g/cm^3; $\rho_{dolomite} = 2.83$ g/cm^3).

2.2. Shale

Silty to clayey shale is considered to represent a good and, therefore, potential host rock for high-level and low-/intermediate level radioactive waste and a recently selected example is the Opalinus Shale in Switzerland (www.nagra.ch; Thury, 2002). There are several reasons for selecting thick shales: low permeability, a quasi-plastic behavior against external stresses, no formation of long cracks or fractures, which are closed because of the low yield strength (Figure 1b). A further advantage is the inert chemical environment and the ability of clay minerals to absorb cations. Shales in stable platform areas are preferred as shown by the Swiss example shows. The stability and therefore the mineability of shale in the geological underground of ca. 1 km depth is limited but manageable when no large caverns are planned.

2.3. Crystaline rocks

Crystalline rocks, meaning *metamorphic rocks (schists)* as well as *magmatic rocks* like granites (Duro et al., 2011) have a low porosity, high strength and therefore stable conditions during the building phase of an underground waste repository (Figure 1c). In tectonically stressed areas, even in stable platforms, crystalline rocks may be fractured at depth, and details are difficult to predict from the surface. Granite host rocks and their interaction with fluids and barrier materials were investigated in detail at the Grimsel Test Site in Switzerland (e.g., Buil et al., 2010 and references). Fractures may represent a pathway of fracture-controlled groundwater and fluids. Oxidizing groundwater moving along such fractures represents a potential hazard for dispersion of radionuclides in such granites (e.g., Dideriksen et al., 2010).

A peculiar host rock for a repository of high-level radioactive waste includes altered *volcanic tuffs* and such a place is Yucca Mts. in the United States (U.S. Department of Energy, 1988; Potter et al., 2004; Bredhoeft and King, 2009). Many detailed investigations were done to assess the long-term stability of that particular site, e.g. the state of stress (Potter et al., 2004) and modeling of the long-term erosion behavior (Stuewe et al., 2009).

Here, we also consider *phyllites* as a further particular rock type for the storage of both high- and low-/intermediate radioactive waste. In their geotechnical properties, quartz-poor phyllites are intermediate between shale and high-grade metamorphic crystalline rocks and granites. As many galleries reveal, quartz-poor phyllites have a relatively low strength, low permeability and the tendency to close fractures under overburden. No long fractures form except in areas of tectonic stress. Particularly dark phyllites should have similar geochemical properties as shale although detailed studies are lacking.

Further potential sites for the repository of radioactive waste were considered, e.g. continental shelves (e.g., Stewart, 2002). However, the risk seems to be higher than in other geological situations, so that no furter detailed work has been done.

3. Dinarides

The Dinarides are mostly composed of carbonates, and such rocks are entirely inappropriate to store even low-level radioactive wastes because of its accessibility to groundwater. However, Mesozoic carbonate sucessions of the central and southern part of the Adriatic Sea contain a number of diapire structures, composed of salt and anhydrite (e.g. Burano Formation sensu Mattavelli et al., 1990). Furthermore, thick, flysch-type pre-Alpine formations, exposed in the area of Petrova and Trgovska gora regions and crystaline basement complexes of the southern Tisia will also be discussed.

3.1. Dinaride evaporite rocks

Dinaride evaporite rocks are located onshore in Croatia and Bosnia and Herzegovina (BiH) along the Paleogene to Neogene thrust fronts (Figure 4) and offshore as diapiric structures in the central and southern part of the Adriatic Sea (sensu Grandić et al., 2004; Geletti et al., 2008).

Similar to the Alps, Dinaridic evaporite rocks act as detachment horizons during thrusting and shortening of the Dinaride orogen. As a result of Paleogene to Neogene shortening various types of sedimentary and magmatic rocks were incorporated within the evaporite formation. These are limestone, early-diagenetic dolostone, rauchwacke, siltstone, sandstone, and rare conglomerate and spilitized basaltic rocks, and are Triassic to Lower Cretaceous in age.

3.1.1. Onshore evaporites

Dinaridic evaporites outcropping onshore in Croatia and BiH consist predominantly of gypsum and anhydrite, basically without any rock salt. They are Permian to Triassic in age, based on sulfur isotopes, palynology and structural data (sensu Herak, 1973; Šušnjara et al., 1992; Šiftar, 1986). The majority of the

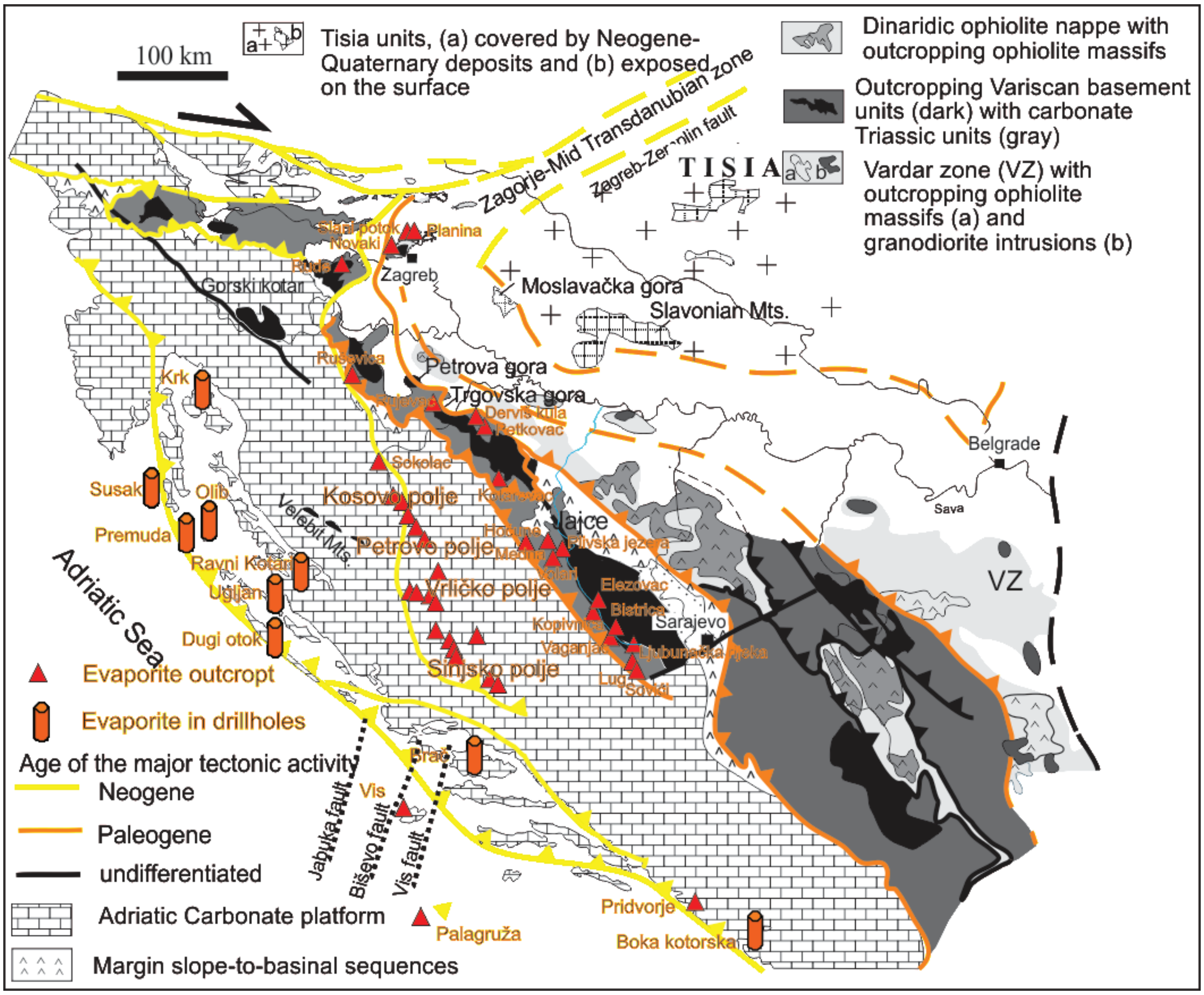

Figure 4. Geological map of NW Dinarides with distribution of evaporites and the location of basement units discussed in text: Petrova, Trgovska, Moslavačka gora and the Slavonian Mts.. Modified after Tomljenović, 2002; Schmidt et al., 2008; and Korbar, 2009.

Slika 4. *Geološka karta SZ Dinarida s distribucijom evaporita i lokacijom podloge koje se diskutiraju u tekstu: Petrova, Trgovska i Moslavačka gora te Slavonske planine. Izmijenjeno prema Tomljenović, 2002; Schmidt i dr., 2008; i Korbar, 2009.*

Croatian evaporite rocks occur within the Adriatic Carbonate platform units along the Neogene thrust fronts (Fig. 4). Most of the outcrops are located within karst poljes and valleys such as: Vrličko, Kninsko, Petrovo and Drniš karst polje within the Lika and Dalmatia areas. These outcrops are large, economically significant and under exploitation (> 1 Mt; Gabrić et al., 2002). They are composed of gypsum in the upper part, and anhydrite in the lower part, and are covered by Neogene sediments (mostly clay). The amount of anhydrite increases with depth, and generally, within investigated deposits gypsum is found within the uppermost 20–40 m (Gabrić et al., 2002). This is due to partial rehydration of anhydrite to gypsum under the influence of meteoric water caused by regional uplift and erosion. Most of the gypsum and gypsum-anhydrite transition zone is cavernous as a consequence of the meteoric water circulation.

3.1.2. Offshore evaporites

The central and southern part of the Adriatic Sea (Adria block) is composed of Permian to Anisian siliciclastic and carbonate sediments interbeded with Burano evaporites, composed of salt and anhydrite (sensu Mattavelli et al., 1990). A compressive regime throughout the Paleogene caused southwestward migration of the Dinaride belt toward the central axis of the Adriatic Sea (Chanell et al., 1979). The compressive tectonics within the Adriatic Sea (as well as within Dalmatia) caused reactivation of some pre-existing faults and the onset of salt diapirism, deforming Mesozoic to Cenozoic sedimentary structures. Examples of such structures are the Vis-Komiža diapir and Palagruža-Jabuka high (sensu Grandić et al., 2004; Geletti et al., 2008).

3.2. Shale within flysch-type sedimentary sequence

Largest flysch-type sedimentary units are located at the (i) Banovina-Kordun area (Petrova gora Mts. - PGM) and (ii) Trgovska gora Mts. (TGM). They are thrust onto the Dinaridic Carbonate platform and floored by units of the Dinaride Ophiolite Zone. PGM is interpreted as a non-metamorphosed sedimentary sequence (Majer, 1964), while TGM was metamorphosed under very low-grade P-T metamorphic conditions. The maximum thickness of these sedimentary sequences is about 600 m (Korolija et al., 1981).

3.2.1. Petrova gora Mts.(PGM)

The PGM are situated within the central part of Croatia, 50 km south of Zagreb, covering an area of about 300 km^2. It is characterized by a number of hills with an elevation below 500 m. The central part of the PGM comprises a pre-Alpine synorogenic flysch sequence, composed of (i) silty shales and siltites in the lower part, and (ii) shales, interlayered by fine- to coarse-grained greywacke, sandstone and subordinate fine-grained conglomerates in the upper part (Korolija et al. 1981). Silty shales consist of fragments of detrital quartz, 10–50 µm in size, sericite, illite, chlorite, feldspar, and opaque minerals within the sericite–illite matrix. The matrix is composed of authigenic sericite and clay minerals, together with rutile, quartz and carbonates. Widespread subgraywacke and sandstones are composed of various detrital particles of quartz, muscovite, feldspars, biotite, chlorite, calcite and rock fragments of various size (60 to 200 µm) within a laminated, microcrystaline matrix which mainly consists of sericite, quartz, organic matter and clays (Jurković, 1957).

3.2.2. Trgovska gora Mts. (TGM)

The TGM is a mountain of low elevation (+557 m) situated 80 km SSE of Zagreb, which occupies an area of about 350 km^2. The pronounced faults from the NW, W and NE divide the TGM from the Mesozoic ophiolites and carbonate rocks and Cenozoic and Quaternary clastic rocks. The core of TGM consists of pre-Alpine synorogenic turbidites. Turbidites are represented by (i) dark-gray clayey schists periodically interbedded with thin layers of silty subgraywacke sandstones (Đurđanović 1968, 1973). Clayey siltstones consist of 10–20 % detrital quartz particles, muscovite, sericite, chlorite and coalified matter, 5–40 µm in size, within microcrystalline illitic-clayey matrix, and (ii) fine- to coarse-grained subgreywackes interbedded with clayey siltstones in the upper part.

3.3. Crystalline basement rocks

The largest crystalline basement rocks crop out in the southwestern part of the Pannonian basin, in the area of (i) the Moslavačka gora Mts. (MGM) and (ii) Slavonian Mts. (SM). They are composed of magmatic to metamorphic cores of pre-Alpine age, surrounded by Alpine sedimentary rocks and sediments. Core of both regions is composed of granites and surrounded by contemporaneous migmatitic zones and regionally metamorphosed sequences at the peripheries. In the area of MGM crystaline rocks are in tectonic contact with Neogene sediments, whereas in the area of SM they are disconformably overlain by mainly Triassic and Cenozoic sediments of the South Pannonian basin.

3.3.1. Moslavačka gora Mts.(MGM)

A crystalline complex of the MGM is located 50 km east of Zagreb, between the Sava and Drava rivers. The MGM covers an area of about 180 km^2. The core of the MGM comprises an S-type granitic pluton surrounded by migmatite rocks and an Abukuma-type metamorphic sequence (Pamić 1990). The granitic pluton comprises monzogranite, andalusite- and sillimanite-bearing granite, granodiorite, and tourmaline-bearing pegmatite rocks (Korolija et al. 1985; Pamić 1990). The Abukuma-type metamorphic sequence consists of amphibolite and amphibole-bearing schists as various sized lenses and bodies interlayered with predominant gneisses, micaschists, cordierite schists, hornfelses and marbles. Amphibolite rocks forms enclaves, more than a hundred meters in size, hosted within granite rocks.

3.3.2. Slavonian Mts.(SM)

The SM (Psunj, Papuk and Krndija) are situated 100 km east of Zagreb and 50 km east of Moslavačka gora. The center part is occupied by S-granite rocks covering an area of about 110 km^2. Granite rocks are surrounded by the migmatite rocks and pre-Alpine Barrovian type metamorphic sequence, composed of medium-grade metamorphic gneisses and mica-schists with subordinate amphibolites and marbles, and very low-grade metamorphic greenschist varieties, chloritoid schists, metasandstones and phyllites (e.g. Radlovac Formation). SM granite consists of granodiorite and monzogranite with associated quartz-diorite and monzodiorite. SM granite rocks are surrounded by contemporaneous migmatite rocks of similar petrography (Pamić and Lanphere 1991). Numerous isolated granite bodies of I-type occur in the southernmost part of the SM. They are up to 30 km^2 in size, consisting of monzogranites and subordinated intermediate and mafic differentiated monzodiorite, gabbro and alpine-type ultramafites (Marci, 1973). I-type granites are conformably interlayered within a Barrovian-type metamorphic sequence. The very low-grade metamorphic unit (Radlovac Formation) is composed of slates, phyllites and sandstones, intruded by 100 m thick metagabbro sills (Pamić and Jamičić 1986). The Radlovac Formation is disconformably overlain by Mesozoic cover sediments and is thrusted onto the pre-Alpine crystalline complex of the South Tisia.

4. Conclusions

Three basic types of host rocks for radioactive waste disposal were discussed: 1) evaporites in general, (2) shale, and (3) crystalline basement rocks, and applied to the Dinarides.

1) Most frequent evaporite minerals: gypsum, anhydrite and rock salt behave differently under geological circumstances. Rock salt deposits, especially in arid climate, are suitable for all types of radioactive waste repositories due to their thermally stability, low porosity and plastic behavior which enable closure of open fractures. Anhydrite with a good seal cap rocks is sometimes considered, whereas gypsum is completely inappropriate. Dinaridic evaporites located onshore are composed predominantly of gypsum and anhydrite, which makes them inappropriate for storage. Offshore salt diapirs in the central and southern Adriatic Sea are composed of salt and anhydrite and hosted within thick Mesozoic-Cenozoic sediments, which under specific circumstances they can be considered as potential site locations for further investigation. Their location (in the middle of the Adriatic Sea) creates additional problems.
2) Shale, especially in a stable platform is a potential host rock for high-level and low-/intermediate level radioactive waste due to its low permeability, a quasi-plastic behavior against external stresses, no formation of long cracks or fractures, inert chemical environment and the ability of clay minerals to absorb cations. Thick shale-phyllite formations are located at Petrova and Trgovska gora Mts.. Intercalation with permeable clastic sediments (mainly various sandstones) is less extent in the area of the Trgovska gora Mts.. Both locations are suitable for further investigation.
3) Crystalline basement is composed of magmatic to metamorphic rocks which have a low porosity and high strength, but are usually fractured at depth which allows fluids (i.e. groundwater) to circulate. Both discussed crystaline massifs (Moslavačka gora and Slavonian Mts.) under specific circumstances can be considered as potential site locations.

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