

World Multidisciplinary Earth Sciences Symposium, WMESS 2015

The Role of Smectite Clay Barriers for Isolating High-Level Radioactive Waste (HLW) In Shallow and Deep Repositories

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Abstract

The major engineered barriers to migration of radionuclides from HLW in repositories are the canister and surrounding smectite clay. They interact physically and chemically by which the properties of both are changed, especially of the smectite “buffer” clay that is examined in the paper. The canisters are made of copper-lined iron according to the Swedish and Finnish concepts, steel being an alternative. The function of the host rock is of importance and the paper examines the role of two repository concepts with long subhorizontal or deep vertical holes for placing the waste.

The hydraulic conductivity of the canister-embedding smectite clay can be significantly raised by high temperature and temperature gradients, which generate precipitation of salt and silica in different parts of the buffer clay. The impact of the degrading processes on the waste-isolating capacity is different for shallow repositories in permeable rock and for very deep disposal with higher temperature. The latter has stagnant groundwater as major barrier to the migration of radionuclides.

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Peer-review under responsibility of the Organizing Committee of WMESS 2015.

Keywords: clayey barrier; high-level radioactive waste; deep repositories; smectite.

1. Scope

The host rock, which has earlier been appointed primary barrier to migration of possibly released radionuclides, merely provides mechanical protection of the engineered barriers, i.e. the HLW canisters and their surrounding clay buffer (Pusch et al, 2014, Pusch 2015a). Their roles are different in different repository types, two of them being considered and compared in the present paper. They both have the form of bored large-diameter holes oriented

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subhorizontally (KBS-3H) or subvertically (VDH), Fig.1 (Svemar, 2005). The different interaction between clay seals, concrete and waste containers of copper and steel is a key issue.

2. Description of considered repository concepts

Fig. 1 shows schematically the two concepts that we will examine with respect to the performance of seals made of smectite clay, separated by concrete cast where the holes intersect active fracture zones.

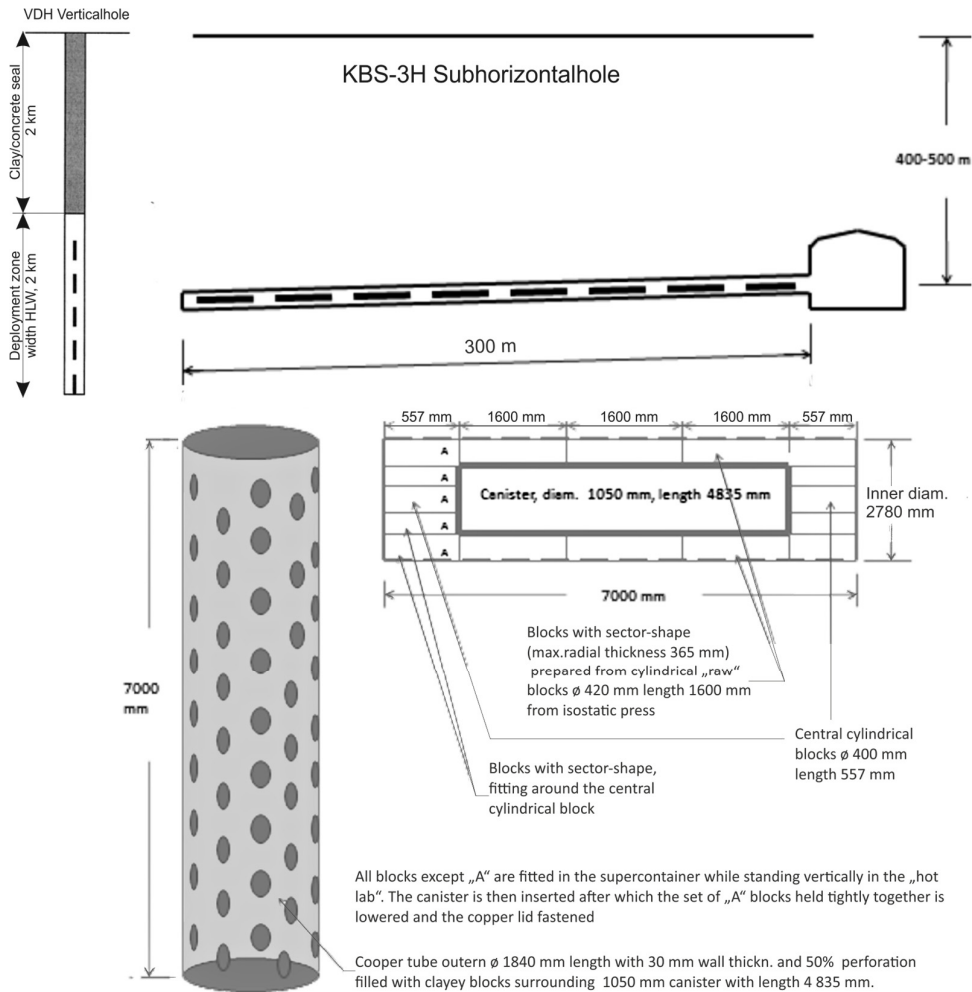


Fig.1. Principles of storing HLW in long, bored holes with waste separated by clayey seals.

Both concepts are based on the assumption that the canisters with gamma-radiating, very hot HLW in the form of spent fuel are placed in “supercontainers” of perforated metal casings, in which the canisters with waste are surrounded by and in tight contact with well-fitting blocks of very dense smectite clay (Fig.2). The copper or steel canisters for KBS-3H have a diameter of 1050 mm and those for VDH have 500 mm diameter. The clay embedding the canisters has a nominal radial thickness of 365 mm for KBS-3H and 100 mm for VDH. The weight of thick-walled steel supercontainers with steel canisters to be placed in KBS-3H holes (tunnels) is estimated at 15-20 tons, while those in VDH can have a weight of 5-10 tons. The supercontainers and canisters will be prepared and charged underground, remote from the deposition tunnels (Pusch, 2008, 2014). For the waste considered by the Finnish and

Swedish organizations POSIVA OY and SKB, which are responsible for the management and disposal of spent reactor fuel, the temperature of the buffer for KBS-3H will be 100°C at maximum and drop to 25°C in about 1000 years (Svemar, 2005). The radial temperature gradient will be up to about 1 centigrade per cm distance from the canister surface. For VDH the temperature of the clay seals, which are enclosed in smaller supercontainers of KBS-3H type, will be 60-70°C down to 2 km depth for any period of time, while for the 2-4 km part, holding waste, it will be 150°C at maximum and drop to about 100°C after 1000 years. At 4 km depth, the temperature will be down to 70°C after 10,000 years.

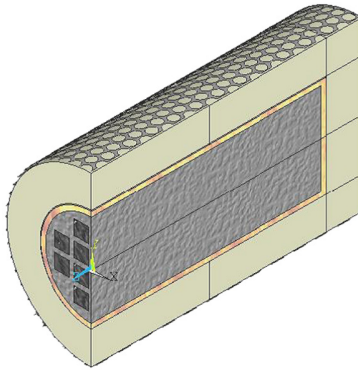


Fig. 2. Example of proposed design of a supercontainer for KBS-3H type repositories.
Upper: Schematic; Central: Detailed design; Lower: Appearance of KBS-3H supercontainer with buffer (light greyish) and buffer-embedded canister (dark greyish in sketch by SKB).

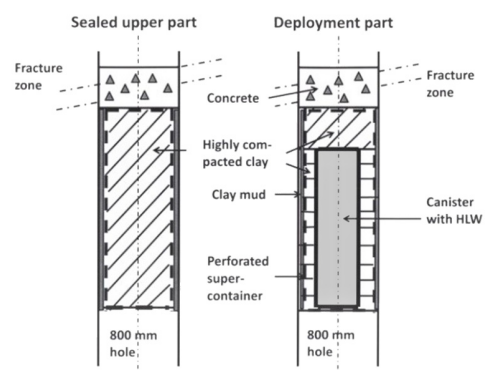


Fig. 3. Supercontainers in VDH to be submerged in smectite clay mud.
Left: Perforated container with dense smectite clay in the upper 2 km part. *Right:* The same type of supercontainer with clay-embedded HLW canisters in the lower 2 km part. The canisters with about 500 mm diameter have a 100 mm envelope of highly compacted smectite clay, which together with the block and mud, makes up the "buffer" clay of the VDH. Extra clay blocks can be placed for separating supercontainers.

Fig 3 shows a vertical section of a VDH-type hole. The left picture illustrates a supercontainer with dense clay in the 2 km upper part, the right one an equally sized supercontainer with a clay-embedded HLW canister in the 2 km lower part (Pusch et al, 2014).

2.1. Construction

The diameter of the holes for deposition of waste containers are 1950 mm for the KBS-3H version proposed by POSIVA OY and SKB, and 800 mm for the VDH concept discussed here (Pusch, 2014). Boring of the subhorizontal holes at 400-500 m depth is made by use of TBM technique under drained conditions, while the 4 km deep VDH holes are bored in clay mud according to techniques used in the oil industry. Both deviate from the intended straight orientation, which has an impact on the allowable length of coupled supercontainers, and on the space between them and the borehole walls. For KBS-3H smectite clay mud is injected in this space (Pusch, 2015b).

2.2. Geohydromechanical site conditions

A number of determinations of the hydraulic and stress conditions in crystalline rock have been made through the years and with some generalization, that the average hydraulic conductivity of crystalline rock likes granite is in the range of E-11 to E-9 m/s excluding major discrete water-bearing fracture zones[†] from the ground surface to depth 1000 m. From 1000 to 2000 m depth, where the average horizontal rock pressure ranges from 30 to 40 MPa, it is estimated to be E-11 to E-12 m/s. From 2000 to 4000 m, where the average horizontal rock pressure is 40-60 MPa, the corresponding conductivity range is E-12 to E-13 m/s excluding major fracture zones (Pusch 2015a). This underlines the necessity of locating the waste-bearing parts in any of the repository types between such zones.

[†] Discontinuities of 2nd and 3rd orders, cf. Pusch, 2008, 2015

2.3. The engineered barriers

2.3.1. Supercontainers and canisters

Copper

Physically intact copper-lined canisters undergo very slow dissolution and the Cu ions released will not migrate more than by a few centimeters in 10,000 years into the clay, causing cation exchange of the Ca ions sorbed in the clay (Atzkarate, 1999, King et al, 2001; Werme et al, 2003, Kursten et al, 2003,). The exchange to copper does not alter the physical properties of the clay significantly (Svemar, 2005). Fig. 4 shows a generalized picture of SKB's and POSIVA's copper-lined iron canister.



Fig.4. Copper-lined iron canister with channels for inserting bundles of spent fuel rods (After SKB).

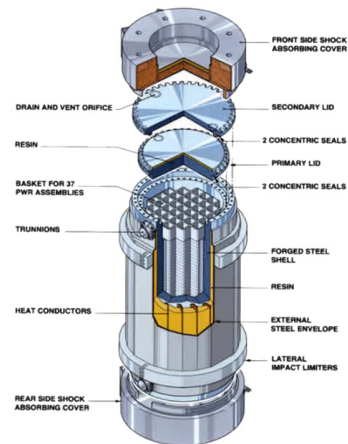


Fig.5. Proposed steel canister for spent fuel (After IAEA).

Iron and steel

A proposed steel canister for HLW in the form of spent fuel is depicted in Fig. 5. Anaerobic conditions will prevail soon after placement, which delays oxidation of canister metals very much. For steel, some magnetite will be formed early by oxygen provided by the air enclosed in the voids of the initially unsaturated clay that embeds the steel canisters and this mineral is believed to be a dominant corrosion product (Grauer, 1986). More magnetite will be formed anaerobically following Eq. 1:



The Fe^{2+} concentration in the magnetite is controlled by the non-congruent dissolution of the mineral, which reacts with the hydrogen and yields hematite (Fe_2O_3) and (FeOOH), (Grauer, 1986; Smailos et al, 1997, 2000). There are also several other expected corrosion products formed in the absence of oxygen, like ferrous hydroxide, $\text{Fe}(\text{OH})_2$ and ferric hydroxide, $\text{Fe}(\text{OH})_3$, which have a strong tendency to form colloids of particles that normally carry a positive charge. Iron ions will under all circumstances be free to react with other materials and become absorbed by the canister-embedding clay through cation exchange, which will make the buffer stiffer and reduce its self-sealing ability in case of large imposed strain. Ion exchange from Na to Fe in combination with heating collapses the clay/water system and widens voids, which promotes channel-type transport of solutions (Gueven and Huang, 1990; Nguyen-Than, 2012; Pusch et al, 2012). As to the use of iron-based metals, the international research on the corrosion and physical behavior of canisters has been extensive and led to various recommendations. Separate national management program, involving the European Commission (EC), has provided lots of scientific data and know-how. The results obtained from work performed i.a at FZK (Karlsruhe, Germany), GNF.IUT (Berlin, Germany) and INASMET (San Sebastian, Spain) for salt environment indicate that carbon steels like the quality TStE 355 are a valid option for thick-walled container concepts in granite. One has to consider that the steel suffers a loss of ductility under these conditions, a behavior that is attributed to hydrogen embrittlement resulting from migration of atomic hydrogen into the metal structure. Despite the low corrosion rates reported, we will assume here that complete corrosion will take place already in 1000 years. The total period time considered here is 10,000 years, after which tectonic and exogenic processes make detailed predictions very hypothetical.

A physical phenomenon of great practical importance is that magnetite has a specific density that is only about 50 % of that of iron, which means that it would occupy twice the initial space. The corrosion depth can be estimated to be a few millimeters in 100 years at free expansion of the iron components and a few centimeters in 1000 years but the expansion is hindered by the compression of the contacting clay and raises the pressure on the walls of the deposition holes to a level that can be critically high. Smectite with octahedral Fe(III) can promote catalytic corrosion of steel (Lantenois et al., 2005). The production of hydrogen gas by corrosion acts similarly (Pusch, 2008, 2015b): if the gas production rate is low the gas can form a successively expanding bubble that compresses the buffer and makes it so tight that the pressure can cause breakage of the surrounding rock. Additionally, this gas is reducing the Eh- and increasing the pH-conditions that offer an additional impact on clay alteration (Nguyen-Thanh, 2012). If gas can penetrate the buffer clay and reach the pervious boring-disturbed zone that extends to a few centimeters distance from the rock surface it will escape via permeable fracture zones that intersect the deposition holes. Geometrical constraints for VDH can make stainless steel a candidate material for thin-walled supercontainers and canisters, while carbon steel is considered the primary choice for the thick-walled canisters of KBS-3H (Smailos, 2000). Here, we will only consider the case of supercontainers and canisters made of carbon steel.

2.3.2 Buffer clay

Degrading mechanisms

The clay blocks fitted around the canisters in the supercontainers and the smectite mud, in which they are submerged, make up the buffer clay. Three issues require attendance here, firstly the criteria for selecting suitable materials, secondly preparation and installation of them, and thirdly their performance at three specific times, 100, 1000 and 10,000 years.

The criteria for selecting suitable clay components are:

- ability to be and stay less permeable than the confining rock for at least 10,000 years,
- ability to exert a swelling pressure of at least 100 kPa on the confining rock and canisters for maintaining tight contact,
- chemical compatibility with rock, canisters, supercontainers, and concrete seals.

The hydrothermal conditions prevailing for many hundred years in the “buffer” regions of a repository will affect the clay in several ways. The initially only partly water saturated dense clay will undergo desiccation and fissuring and the subsequent saturation, caused by uptake of groundwater from the rock taking many decades or centuries, is associated with accumulation of precipitated salt. This process reduces the hydraulic conductivity by blocking voids and channels and proceeds as long as there is a temperature difference between the hot canister (100°C for KBS-3H, and 150°C for VDH) and the rock. The temperature gradient will finally be evened out causing dissolution of some precipitations, thereby raising the hydraulic conductivity. There is consensus on the validity of the empirical reaction formula in Eq.2, (Pusch and Yong, 2006; Kasbohm et al, 2013).



where S: denotes smectite; Fk: K-feldspars; Mi: micas; I: Illite; Q: quartz; and Chl: chlorite. Illite is formed from smectite by uptake of potassium emanating from accessory minerals or from the groundwater. The evolution is via mixed-layer stages or by neoformation through combination of silica, aluminium and potassium released from dissolving minerals or provided by the groundwater (Gueven and Huang, 1990; Pusch and Madsen, 1995; Kasbohm et al, 2013). Smectite buffer clay can change by two reaction mechanisms: “illitization” in open reaction systems, and “smectitization” in closed systems (Kasbohm et al, 2013). Interaction of smectites and metal canisters has been investigated in detail. For copper canisters, the impact on either of them is insignificant at larger distance than a few millimetres from the hot canister surface in 1-2 months long hydrothermal experiments years (Pusch, 2008; Pusch et al, 2012) but presumably reaching some centimeters in 1000-10,000 years (“Kupferschiefer”). For steel canisters the degree of alteration, involving dissolution and subsequent precipitation of crystalline and amorphous silica/iron complexes, depends on temperature and time. Illite will be formed close to the steel canister and kaolinite or pyrophyllite further off via smectitization (Herbert et al, 2008; Kasbohm et al, 2013). The “illitization” process results in higher particle charge and lower swelling pressure. In contrast, the formation of smectite, implying self-healing, reduces the charge and increases the swelling pressure. Uptake of Fe replacing Al in the octahedral sheet of montmorillonite can accelerate the alteration because these large ions cause high crystal stresses and instability

(Ngyuen-Thanh, 2012). Raised temperature increases the Fe-activity and the amount of dissolved Si at the clay/canister interface causing migration towards the colder rock and silicification of the entire buffer in the cooling phase. In a very long time perspective, cementation by precipitation of Si can convert smectite clay to shale (Pusch and Madsen, 1995; Svemar, 2005).

Gas production

The pressure of hydrogen gas formed according to Eq.2 will be under high pressure and cause fingerlike channels in the surrounding buffer if its tightness with respect to gas penetration is low, or a high-pressure gas bubble at the surface of the canisters if the buffer is gas-tight (Pusch and Yong, 2006; Pusch, 2008). The rate of gas production will be a primary determinant of the evolution: if it is very low the gas will dissolve and migrate upwards in the system by diffusion; if it is high migration will take place in gaseous form.

Interaction with concrete

As indicated in Fig.3 there are contacts between clay seals and concrete where fractures zones are intersected and comprehensive work has been made to find what the chemical interaction is when using cement of Portland type and of low-pH type (Mohammed et al, 2014). Degradation of the concrete by dissolution of components like portlandite will change the volume and bearing capacity of the concrete seals and cause breakdown of adjacent clay by OH attack. Organic fluidizers can form organic colloids that can transport radionuclides and inorganic substitutes like talc have been successfully tested. The bearing principle of defining recipes for well performing concrete is 1) to apply packing theory for reaching highest possible density, 2) to use quartzite and finely ground quartz or silica powder for aggregate, 3) to use finely ground low-pH cement, and 4) to use finely ground talc powder as fluidizer (Mohammed, et al, 2014). The shear strength of such concrete is low in the first week but increases to higher strength than concrete with Portland cement has after a few weeks due to chemical reactions between the talc and cement components. The solubility of the reaction products is deemed very low but complete loss of them has still to be assumed, which makes the physical properties of the remainder, the aggregate, very important (Mohammed et al, 2014).

3. Comparison of cases

The function of the smectite seals in the two repository types are compared in Table 1 with respect to the constitution and main physical properties 100, 1000 and 10,000 years after disposal. The estimated content of expandable smectite, the stiffness, and the permeability are the parameters of interest. The same smectite clay, termed “Holmehus” is selected for the assessment (Kasbohm et al, 2013; Pusch et al, 2015c). It is of I/S type with at least 60 % expandable and is assumed here to have an average dry density of 1600 kg/m³ soon after placement.

Table 1. Comparison of estimated physical properties of Holmehus buffer clay, combined with Holmehus clay mud, at different stages after disposal.

| Concept/time in years | Canister/ super- container | Ductility ¹⁾ | Tightness (permeability) ²⁾ | Expanda- bility ³⁾ | Smectite content ⁴⁾ | Risk ⁵⁾ |
|-----------------------|----------------------------------|-------------------------|---|----------------------------------|-----------------------------------|--------------------|
| KBS-3H/100 | Copper | +++ | +++ | ++ | +++ | +++ |
| KBS-3H/1,000 | Copper | ++ | ++ | + | +++ | +++ |
| KBS-3H/10,000 | Copper | + | + | None | ++ | ++ |
| | | | | | | |
| KBS-3H/100 | Steel | ++ | ++ | + | +++ | +++ |
| KBS-3H/1,000 | Steel | + | + | None | ++ | ++ |
| KBS-3H/10,000 | Steel | None | + | None | + | + |
| | | | | | | |
| VDH/100 | Copper | +++ | +++ | + | +++ | +++ top seal OK! |
| VDH/1,000 | Copper | ++ | ++ | None | ++ | +++ top seal OK! |
| VDH/10,000 | Copper | + | + | None | + | +++ top seal OK! |
| | | | | | | |
| VDH/100 | Steel | ++ | +++ | + | ++ | +++ top seal OK! |
| VDH/1,000 | Steel | + | ++ | None | + | +++ top seal OK! |
| VDH/10,000 | Steel | None | ++ | None | + | +++ top seal OK! |

1-4) +++ good, ++ acceptable, + poor. 5) Risk for contamination of local groundwater +++ Insignificant, ++ obvious, + high

The assessment of the various cases manifests the conclusions from the descriptions of the function and properties of smectite buffer clay embedding canisters and supercontainers of copper and steel. The hydrothermal conditions in a KBS-3H repository with clay-embedded copper canisters mean that most of its smectite content in the first 1000 years is retained, but some is lost in the time period 1000 to 10,000 years. In a repository of this type but with steel canisters and supercontainers, degradation of the buffer clay will be more obvious, especially beyond 1000 years, because of the precipitation of cementing Fe compounds. Even higher tightness than for copper canisters may be reached by the consolidating effect of expanding steel undergoing corrosion, which will make the clay impermeable and claystone-like. Its ductility will be definitely lost.

For VDH with copper canisters and supercontainers most of the smectite in the buffer clay will be retained even in 10,000 years also down at 4000 m depth but cementation by precipitated silica and illite will cause very significant stiffening and loss of self-sealing ability in case of shearing caused by seismic and tectonic events. Clay seals in supercontainers of steel down to 2000 m depth will become very stiff and low permeable by cation exchange from Ca to Fe and consolidation under the pressure built up by contacting steel undergoing corrosion. Buffer clay in steel canisters and supercontainers confining steel canisters and supercontainers from 2000 to 4000 m depth stiffens very much by consolidation under the very high pressure caused by the expansion of the steel undergoing oxidation, making the clay very dense, impermeable and claystones-like.

4. Conclusions

The present compilation of information gathered in the comprehensive international research and development of principles for safe isolation of HLW gives indications of ways of proceeding. Assessment of the performance of artificial clay barriers tells us that the least risk of contamination of groundwater in the host rock area after 100 to 1000 years is offered by KBS-3H with copper canisters and supercontainers. After 10,000 years, the best isolation of the HLW can be provided by VDH because of the almost completely preserved tightness of the clay seals in the upper 2 km long parts. Copper canisters and supercontainers isolated by dense smectite clay will be superior to those of steel in the lower 2 km parts, which would make the stiff buffer clay very dense by consolidation under the pressure caused by chemical degradation of the steel components. The real essence of the VDH concept, that makes it competitive, is that the very high salt and heavy groundwater surrounding the waste-bearing lower part of VDH holes makes it stagnant, meaning that possibly contaminated groundwater will not reach to shallow rock (Åhäll, 2006). The major conclusions from the examination of the KBS-3H concept are:

- placement of supercontainers causes difficulties and involves risk of being stuck because the holes (tunnels) will not be straight. Pregrouting of the rock for reducing inflow of water in the construction phase will cause delay in wetting of the buffer clay and temperature rise,
- heat effects will cause insignificant change of the smectite content of the buffer confined in KBS-3H supercontainers and canisters of copper[‡] with mud injected around them. Stiffening of the buffer will increase with time and reduce the self-sealing potential that will be caused by precipitated silicious complexes and neoformed minerals, giving high stresses in supercontainers and canisters in case of shearing by seismic or tectonic movements in the rock. This effect will be particularly strong for canisters and supercontainers of steel. For such components, the expansion caused by the oxidation will compact adjacent clay to become very tight claystone in and beyond a 10 000 year perspective.

The major conclusions from the examination of the VDH concept are:

- placement of supercontainers with clay blocks in the upper mud-filled 2 km part of the holes is simple but must be made within a day or quicker because of the increase in shear strength of the thixotropic mud in the holes and of the consolidation of the mud under the pressure of the expanding clay blocks in the supercontainers,
- heat effects will cause very significant stiffening of the buffer by cementation of precipitated silicious complexes especially if the canisters and supercontainers are made of steel.

[‡] Including Navy Bronze with low content of Zn

The two repository concepts have several problems in common, the most obvious being the stiffening of the buffer that reduces the self-sealing ability in case of shearing by seismic and tectonic events. This is most alarming for KBS-3H since such repositories will have to be located at shallow depth where there is greater risk of shearing of the holes (tunnels) because of the more frequent fractures that can undergo shearing. At depth, the rock pressure is higher and only major fracture zones, which are easily identified in site investigations and avoided in the design and construction phases, may be sheared.

The major differences between the KBS-3H and VDH concepts with respect to clay seals is the simpler installation of them in VDH and the elimination of critical hydraulic gradients at the installation and construction, which takes place under original, stable piezometric conditions, while this can cause considerable difficulties for KBS-3H. A strong advantage of VDH is that the very salty and heavy groundwater surrounding the waste-bearing part of the holes makes it stagnant. This means that possibly contaminated groundwater will not reach the biosphere.

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