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Isolation of hazardous waste in crystalline rock

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Abstract

Radioactive waste from the nuclear industry and hazardous products from chemical industries need to be effectively isolated from the biosphere for a very long time. Highly radioactive waste gives off heat and requires disposal at depth in special repositories while low-level radioactive waste, pesticides and mercury and arsenic, can be stored in deep mines. The multiple barrier principle implies that the rock and engineered barriers combine to provide isolation but assessment of the constitution and performance of crystalline rock reduces its role to provide "mechanical support" to waste containers rather than true isolation of them. Smectitic clay is required for achieving this but its isolating capacity is limited over time, and long-lasting waste containers are needed as well. The waste isolation effect of clay and containers can allow for constructing repositories in rock of rather poor quality, represented by abandoned mines, and waste containers of 100 % copper further reduce the need for very well planned and constructed repositories.

Keywords: Clay, Crystalline rock, Hazardous waste, Repositories, EDZ

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1 Introduction

Deep long-term "geological" disposal is considered for highly radioactive waste encapsulated in robust containers for limiting the risk of release of radionuclides that can migrate via groundwater to the biosphere. A less demanding but still challenging issue is that of permanent disposal of hazardous chemical waste, like pesticides and mercury and arsenic compounds contained in drums or left unconfined. An important matter is if a repository has to be constructed in virgin rock or if deep mines can be adapted to become repositories.

2 Disposal concept

There is general consensus that repositories for highly radioactive waste (HLW) should be located at a depth of a few hundred meters in rock for effective isolation [1]. Engineered barriers in the form of metal containers (canisters, casks) and clay are proposed for creating multi-barrier systems (Figure 1).

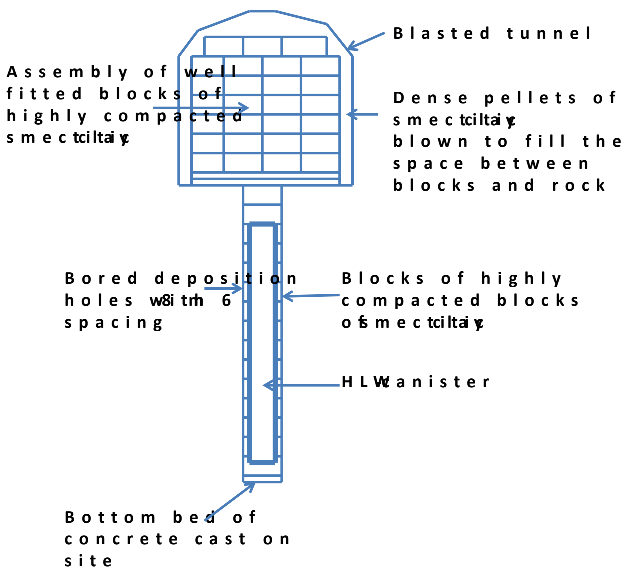

Figure 1: Isolation of HLW. Height and width of tunnels 5 m. Depth of 1.9 m deposition holes for 5 m canisters with 1 m diameter is 8 m [2].

The present paper discusses the performance of rock as barrier and the possibility to accept rock of different quality by selecting engineered barriers that can compensate for its shortcomings. Focus is here on highly radioactive waste (HLW) that represents the most hazardous waste because of the double effect of gamma radiation and release of radionuclides.

3 Rock

3.1 Structural constitution

The structural constitution of the host rock, which is responsible for its mechanical and hydraulic performance, will be only partly known when location and orientation of the tunnels, deposition holes and bulkheads have to be decided. This makes it difficult to work out structural models for which categorization of weaknesses is needed [1, 3]. Systems for this distinguish between 1st order discontinuities (faults) and 2nd and 3rd order discontinuities (major and minor fracture zones). Any drift, tunnel, or shaft will be intersected by several water-bearing and unstable zones of the latter type. Tectonic and seismic events will change their potential to transport water and dissolved hazardous chemical elements. Here, no waste can be placed. There are also discrete, persisting water-bearing fractures of 4th order that will intersect a repository and they will also react to stress changes. These “fractures” can affect the integrity of waste containers. Figure 2 illustrates a structural model with major discontinuities between which the repository is fitted.

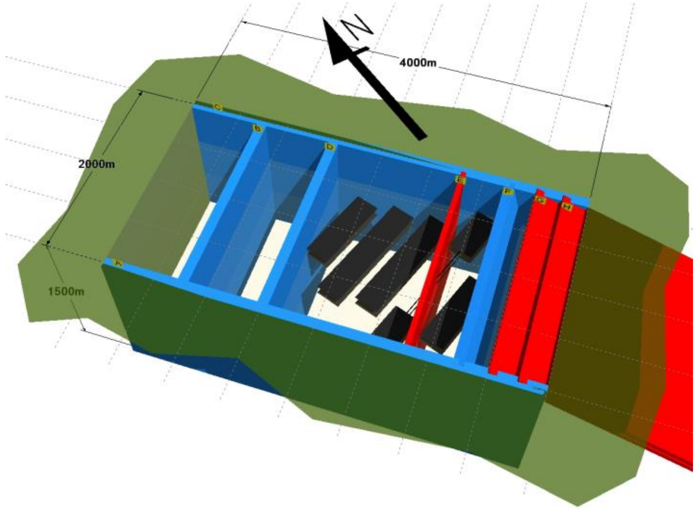

Figure 2: Generalized model of a repository site. Thick plates are large (2nd order) discontinuities with 100 m width, and thin ones less important (3rd order) 10-30 m wide zones. Tunnels with waste are black [4].

Crystalline, argillaceous, and salt rock are host rock candidates depending on the geological conditions in the respective countries. Argillaceous rock like clay shale is very tight but the physical stability of underground rooms at depth is commonly rather poor. Salt rock contains fluid water in local brine lenses and undergoes strong time-dependent strain that can make it difficult to localize and retrieve waste containers after a few hundred years if this is required [1].

3.2 Performance of rock hosting a HLW repository

3.2.1 Permeation

The transport capacity of the rock is determined by its hydraulic transmissivity, which is controlled by the interconnectivity and transmissivity of the system of discontinuities. These systems are networks of openings with varying aperture representing flow paths similar to the water pipes in a building [4]. They are largely hypothetic since accurate structure models cannot be derived until the repository has been constructed. The possibility of making trustworthy calculation of groundwater flow in crystalline rock with its spectrum of permeable features is hence very small. As realized in an EC project on disposal of hazardous waste in abandoned mines, only the major discontinuities, i.e. the fracture zones, can be ascribed hydraulic properties that make gross flow calculations relevant [5]. This project led to the conclusion that groundwater flow primarily takes place in crossings of such zones.

Construction of a repository has two impacts: damage by the excavation process, which is commonly blasting, and expansion or shearing of existing discontinuities, primarily of those of 3rd and 4th orders. Also, there will be an impact by the heat production from the radioactive decay as commented here.

3.2.2 Impact of heat on the host rock

Very tight rock, which would appear to be ideal for locating a HLW repository, is commonly under high pressure which will generate very high hoop stresses. The risk of failure is particularly high in HLW repositories because of superimposed thermally induced stresses. The 6000 waste containers with 0.6 kW power in the Swedish repository will lead to a heave of the ground surface by about 200 mm, causing shearing of 3rd and 4th order discontinuities. This can cause a net increase in bulk hydraulic conductivity [6].

3.2.3 Excavation disturbance - The EDZ

The excavation of underground rooms can be made by blasting, which is proposed for the Swedish HLW repository, or by TBM boring. The latter technique causes higher hoop stresses that can generate failure, while blasting has the effect of relaxation but also formation of a disturbed zone (EDZ) with significantly increased hydraulic conductivity [6]. It will rise by at least 100 times down to about 1 m below a tunnel floor.

An additional EDZ effect comes from the change in stress conditions caused by the creation of a tunnel. They generate shear strain and expansion or compression of 4th order discontinuities as illustrated by Figure 3. The initial average aperture of 10 μm was found to increase to at least 100 μm for certain of them, increasing the average hydraulic conductivity in the direction normal to the section by about 1000 times [6, 7, 8].

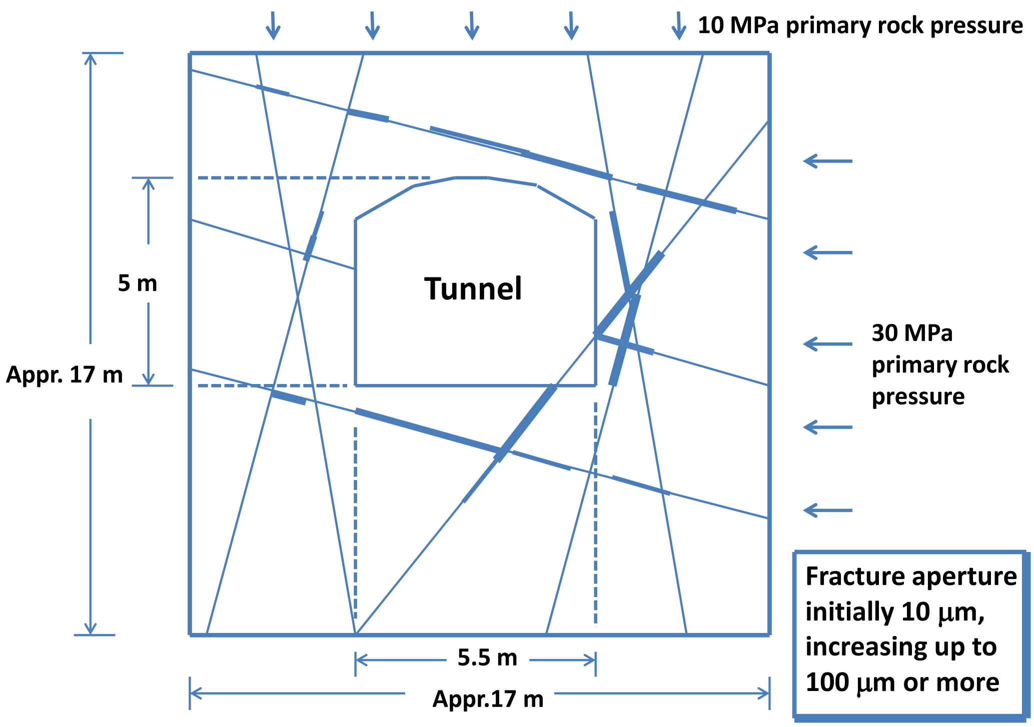

Figure 3: Example of increase in aperture of fractures of 4th order in granite with four major fracture sets. Stripa rock at 360 m depth, (After Hökmark).

3.2.4 Mechanical stability

The rock pressure is locally more than 40 MPa in the region where the Swedish HLW repository is now under construction. Application of statistical methods using the laboratory-determined unconfined compressive strength between150 and 350 MPa as parameter for assessing hole positions, has shown that a few percent of them have to be abandoned. However, taking stress concentrations that can arise where steep 4th order fractures are close to the holes into consideration, and also the impact of heating, the hoop stress is found to lead to failure of most of the deposition holes by fracturing and fissuring of the surrounding rock. The porosity and hydraulic conductivity will thereby increase very significantly.

A further problem is the risk of destruction of waste containers by tectonically induced shearing. Theoretically, it can take place along any discrete discontinuity that intersects the repository rooms and lead to breakage of the containers (cf. Figure 4). If they contain HLW, water will enter and radionuclides start to migrate to the biosphere. The critical strain is 50-100 mm, which can be caused by tectonics, earthquakes and thermal impact, as well as by repeated glaciation and subsequent unloading, in the next 100000 years [9]. The ductility of the clay “buffer” surrounding the containers reduces the stresses generated in them but it will be stiffer with time and reduce the maximum allowed shear strain.

The risk of tectonically generated rock strain that can affect the integrity of waste containers depends on the size of the repository rooms, which is of importance in considering use of abandoned mines for disposal. Large drifts have deeper and more pervious EDZ and more frequently intersecting major discontinuities than small ones. The major drifts will undergo earlier and larger shear strain which will affect a larger number of waste containers.

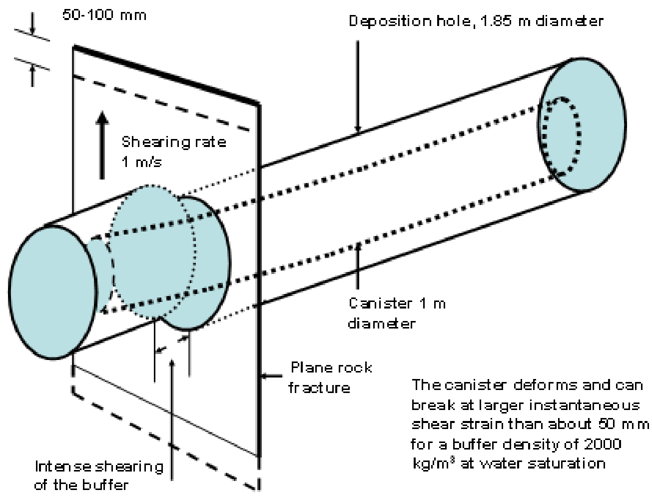

Figure 4: Calculated deformation of clay-embedded container by 0.1 m instant shearing along a fracture that intersects the deposition hole.

The rock in mines of room-and-pillar type has normally undergone significant earlier strain leading to more fracturing and an appreciable EDZ with high conductivity between the pillars. Such mines should be more apt to undergo large strain by tectonic events but the resulting increase in hydraulic conductivity can be compensated by using more effectively isolating clay buffer. A mine with small drifts and moderate EDZ serves even better for disposal of hazardous waste, possibly even HLW (Figure 5).

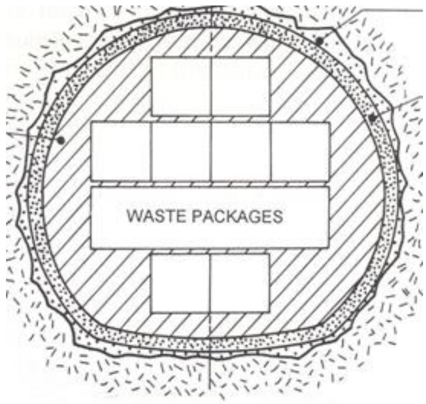

Figure 5: Drift with 5-10 m height and width for disposal of hazardous waste. The rock surfaces are shotcreted (upper pointer) to give a smooth contour for placing tightly fitting blocks of dense clay (right pointer) parallel to placement of containers in clay-based fill (left pointer).

3.3 Can rock serve as barrier?

A first and most important issue is to identify the presence of major discontinuities that determine the positioning of rooms for waste containers in a repository to be built, or in an abandoned mine. The problem with this is particularly obvious for new repositories since even very comprehensive geophysical investigations and borings cannot lead to a sufficiently detailed structural model of the more than ten million cubic meter host rock volume. Much energy and very large sums of money have been spent on making structural models look scientific and reliable by applying statistical methods. There are examples of theoretical models with billions of plane-parallel slots simulating fracture systems in 3D, for predicting groundwater flow in large rock volumes [2, 10]. The reliability is poor, however, since the fracture data come from boreholes with a total volume that is less than 1/10000000 of the host rock volume.

The conditions are different for mines. The number of boreholes made for identifying the boundaries and quality of the ore bodies is large, there are often several hundreds or even thousands of exploration holes, and structural models are much more realistic. From this viewpoint abandoned mines are attractive for disposal of hazardous waste.

Summarizing the arguments for and against constructing a new deep underground repository in crystalline rock there is in fact just one positive: the possibility to create a stable repository because of the high rock strength. The negative arguments are multiple: the difficulty in developing and applying geological and hydrological models for safety analysis, the low degree of utilization because of unpredicted presence of discriminating discontinuities, the risk of unforeseen strain because of seismic, tectonic and heat impact, and above all, the high construction cost.

The low cost for exploitation of a mine to become a repository is positive as well. But there are negative arguments: the EDZ will be more extensive and conductive than for constructed repositories because of the effective demolishing that has been required for extracting ore. A further difficulty is that the usually irregular shape of drifts and rooms makes rational placement and isolation of waste difficult.

One concludes, in summary, that the barrier effect of crystalline rock is merely to offer stability of the host rock and that the comprehensive permeation of groundwater and transport of possibly released radionuclides requires very effective artificial isolation of the waste containers. This can be provided by clay embedding tight waste containers.

3.4 Design principles

Figure 1 is an example of how clay is used for isolating HLW: very dense clay around the waste containers and in the tunnels, both being surrounded by clay pellets or pumped-in clay mud.

3.5 Clay material

3.5.1 Criteria

The hydraulic conductivity of the clay surrounding waste containers shall, by definition be lower than of the surrounding rock and its expandability sufficient to support the surrounding rock, and to self-heal if it is deformed by external forces or internal processes, like temporary desiccation. The clay in the deposition holes, the "buffer", must be able to carry the heavy containers without yielding but also be sufficiently soft to even out stresses generated in the canisters by tectonically induced movements.

3.5.2 A reference clay

The commercially available smectite-rich, strongly expansive Wyoming bentonite (MX-80) was early taken as reference clay for the Swedish and Finnish concepts but other smectite-rich clays perform similarly. Buffer clay in the form of dense blocks manufactured by compacting air-dry smectite granules has been systematically investigated with respect to the essential physical properties, i.e. the hydraulic conductivity, expandability, and ion diffusivity as functions of the density [2].

For the planned density of about 2000 kg/m^3 at water saturation the hydraulic conductivity of smectite-rich clay buffer is as low as E-13 to E-12 m/s and the swelling pressure in the range of 2 to 10 MPa.

3.5.3 Longevity

The matter of long-term chemical stability is of course of fundamental importance for relying on clay as effective barrier. It has been in focus of the R&D of the national organizations that are responsible for safe disposal of HLW and hazardous chemical waste for decades and new findings are currently being published. A fundamental empirical reaction derived from comprehensive mineral analyses of samples from deep boreholes and being relevant to the buffer clay case, is the reaction formula in Eqn. (1), [11].

$$S + (Fk + Mi) = I + Q + Chl \qquad (1)$$

where: S denotes smectite, Fk K-feldspars, Mi micas, I illite, Q quartz, and Chl chlorite.

The formula implies that illite is formed from smectite by uptake of potassium that originates from the rock-forming minerals, formed by cannibalism of the smectite under the hydrothermal conditions prevailing in the buffer, or provided by the groundwater [12]. Interaction of smectites and waste containers has been investigated in detail. For copper containers the impact on either of them is reported to be insignificant [2] but the matter is under dispute.

For containers of steel for disposal of HLW and chemical waste the use of clay barriers is presently in focus in many countries, and the latest and most comprehensive study has led to deepening of the understanding of the longevity of smectite clay in contact with steel [13]. A thermodynamic approach was taken in this study, showing:

- For montmorillonitic smectite, there are two reaction directions: “illitization” in open reaction systems and “smectitization” in closed systems. The degree of alteration is controlled by the concentration of free Fe- and Si. “Illitization” results in higher hydraulic conductivity and lower swelling pressure. In contrast, the formation of smectite reduces the conductivity and increases the swelling pressure.
- The contact with metallic iron causes strong increase in dissolution potential leading to release of Si from the clay particles.

- In the early interaction between montmorillonitic smectite and steel, Si will be dissolved from clay minerals and used up by neoformation of montmorillonite layers giving mixed-layer I/S phases. Si-precipitation occurs if not all the dissolved Si is used up, causing reduction in porosity and brittleness by cementation.

Enhanced temperature raises the Fe-activity and the amount of dissolved Si at the clay/canister interface, causing strong precipitation of Si in the cooling phase. Ion exchange from Na to Fe collapses the clay/water system and widens extra-lamellar voids. This promotes channel-like transport of solutions (Figure 6). In a very long time perspective cementation by precipitation of Si can convert smectite clay to shale.

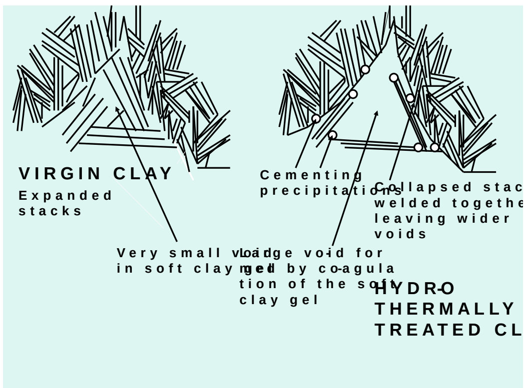

Figure 6: Hypothetic microstructural changes in heated smectite [3].

3.5.4 Conclusive remarks to the need for and use of smectitic buffer clay

The poor barrier function of crystalline host rock and the not yet certified performance of copper and steel containers make buffering clay necessary for retarding migration of possibly released contaminants. The type of waste is of greatest importance: HLW is heat-producing and the temperature of the waste, container and surrounding clay can be critically high especially if the thickness of the clay is large. Its chemical stability drops exponentially with increasing temperature and time but the present understanding is that heating to 90°C for a few thousand years will not cause significant degradation [2, 14]. The thermal conductivity can be increased by mixing the clay with quartz powder for significant reduction of the temperature but this increases the hydraulic conductivity. An optimal composition has to be identified for which material data are available [2, 11]. For hazardous waste that does not produce heat it is of course recommended to make the waste-embedding clay as thick as possible. Today there are methods for very effective compaction of buffers and backfills so that the required high densities can be obtained for fulfilling common criteria concerning hydraulic conductivity and expandability.

The minimum time for effective isolation of HLW is commonly taken as 100000 years because of the risk of genetic damage, while there is no corresponding internationally accepted time for chemical waste despite the fact that wastes like pesticides maintain their risk potential forever. Taking the structural constitution of crystalline rock into consideration one finds that contamination can occur quickly at large distance from a repository [5]. The need for effective artificial barriers is therefore obvious.

One concludes that the barrier effect of smectitic clay is excellent as long as it has not undergone hydrothermally generated alteration to become stiff and significantly more permeable than originally. This makes permanent separation of non-heating hazardous waste from the groundwater achievable, while effective isolation of HLW by clay still has to be proven. Long-term integrity of the waste containers is required and can be provided by containers that remain perfectly tight for at least 100000 years. This is offered by the all-copper HIPOW canister [15], (Figure 7).

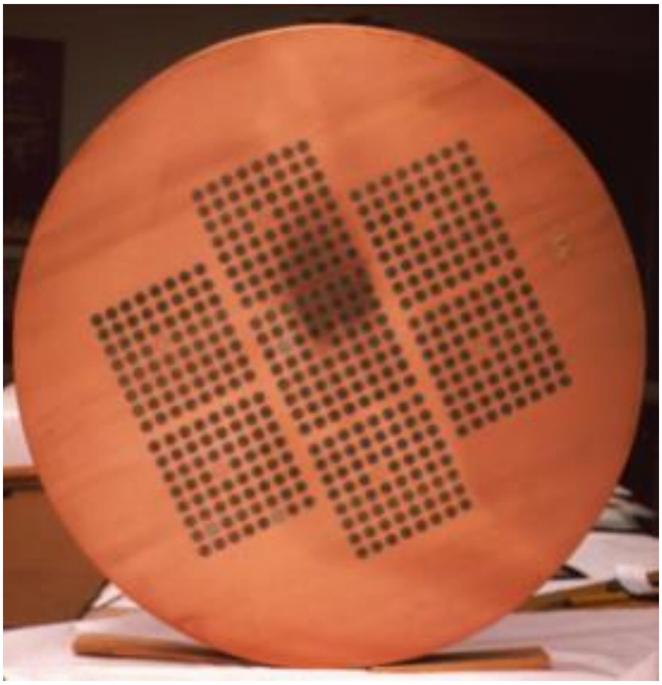

Figure 7: Disc sawn from a 1.6 m long column of HIPOW-compressed copper powder in a copper tube. The spent fuel, simulated by steel pellets in the experiment, is the black spots in the copper mass. Diameter about 600 mm [3, 15].

4 Discussion and conclusions

4.1 Rock issues

Granitic host rock is excellent for constructing stable repositories for hazardous waste since the strength properties are good but the structural constitution needs to be taken into consideration for locating deposition tunnels and holes so that the hoop stress around them will not be critically high. The rock structure cannot be defined with any certainty until the repository is already constructed and the degree of utilization can therefore become low and the cost high. In contrast, conversion of abandoned mines to repositories means that all essential rock structural features are known beforehand and that groundwater permeation can be predicted with much greater accuracy than for new repositories.

For disposal of HLW, construction of a new repository means that the technique employed will create a pervious zone of disturbance and that the stress conditions create a disturbed zone with enhanced hydraulic conductivity around tunnels and shafts. The resulting transient structural changes make it impossible to reliably calculate and predict groundwater flow in the host rock for safety assessment. Engineered barriers are hence needed for certifying isolation of the waste.

Conversion of abandoned mines to repositories means that the rock structure is known beforehand and that the placement of waste can be adapted to the geometry of the drifts and rooms and to the major rock structural features. Excavation-disturbed zones extend deeper in the rock than for new repositories and they have a higher hydraulic conductivity and transmissivity but this can be compensated by constructing long-lasting bulkheads keyed into the rock. For disposal of HLW the effect of heating is expected to convey most of the strain to the disturbed zones, which have lower E-moduli. Their continuity direct groundwater flow through them leaving well confined waste in the drifts hydraulically isolated. The typical trend of convergence of drifts and rooms in mines will tend to compress the clay-embedded waste but the practical positive impact of such strain is marginal.

4.2 Engineered barriers

Use of smectitic clay as barrier is required for effective isolation of radioactive as well as chemical waste placed in repositories in crystalline rock. For low and intermediate level radioactive waste, which does not give off heat, and for chemical waste, smectite clay buffer consisting of montmorillonite can be used since the longevity is sufficient for providing required isolation for at least 100000 years and probably much longer than that. For HLW the generated hydrothermal conditions, prevailing for several thousand years, will cause degradation of the buffer clay. The major part of the smectite content may be preserved for a considerable period of time but the trend of subsequent conversion to less tight illite is obvious. Thermal impact can increase the hydraulic conductivity and there is also risk of stiffening of the buffer clay by precipitation of quartz and other silicious matter. This will cause loss in self-healing potential if thermally and tectonically generated strain takes place. The uncertainty respecting sufficient barrier function of the buffer clay makes it necessary to rely on the integrity and tightness of the waste containers. Canisters of HIPOW-type, made of 100 % copper, have the greatest chance to perform acceptably. Other corrosion-resistant metals may also turn out to be candidates.

4.3 Overall conclusion and recommendations

Crystalline rock provides sufficient strength and stability of repositories but groundwater permeation cannot be predicted with certainty and thermally and tectonically generated container strain may be unacceptably large. For non-heating chemical waste smectite clay provides acceptable isolation for very long period of time but for HLW this has not been proven. Using HIPOW-type containers complete tightness is guaranteed and the risk of release of radionuclides eliminated. Use of such containers strongly reduces the need for effectively isolating, ductile buffer clay, and minimizes the requirement to adapt the location of them to the rock structure. Simple backfilling of repository-converted drifts and rooms in abandoned mines would in fact be sufficient even for highly radioactive waste if it is contained in HIPOW or similar tight containers.

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