

Review

Application of Cement-Based Materials as a Component of an Engineered Barrier System at Geological Disposal Facilities for Radioactive Waste—A Review

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Abstract: Over the past several decades, the international community has been actively engaged in developing a safe method for isolating spent nuclear fuel, high and intermediate level radioactive waste of different degrees of heat generation in deep geological formations on the basis of regulatory requirements existing in each individual country (for example, in the Russian Federation-NP-055-14). Such a storage facility should be equipped with an engineered safety barrier system that combines a range of materials capable of ensuring the safe localization of environmentally and health-threatening nuclear power generation industry and the nuclear industry waste products, in particular. On the basis of the international experience discussed in this article on the design and operation of such facilities, the most universal material in terms of the functions performed as a component of the engineered barrier system is cement and the cement-based product mixed with various components—concrete. Furthermore, due to the possible mutual influence of buffer materials and their transformation over time at interfaces, this work considers the impact of cement-based barriers on other components of engineered barrier systems, the information on which has been accumulated as a result of both analytical laboratory tests and in situ radioactive waste disposal facilities under construction.

Keywords: nuclear power generation industry; radioactive waste; geological disposal facility; cement; concrete



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1. Introduction

The isolation of radioactive waste (RW) at disposal facilities is ensured mainly by the implementation of the multiple-barrier principle [1,2]. Its essence is to create a number of obstacles to the potential migration of radionuclides into the environment, allowing the process to be significantly delayed or completely stopped [1,3,4]. A multiple-barrier protection system typically consists of a host rock of the disposal facility and its surroundings (geological barrier) called the natural barrier system (NBS) and an engineered barrier system (EBS) (Figure 1) [1,2]. The composition of the engineered barrier system is formed mainly of the following parts: waste form, containers with waste, borehole or tunnel lining, sealing, plugging, backfilling and construction of RW disposal facility walls [1,2]. Near-surface disposal facilities (NSDF) and near-surface repositories of radioactive waste (NSR) mainly rely on EBS, while geological disposal facilities (GDF) and deep geological repositories of RW (DGR) use NBS [1] as the main barrier.

The worldwide practice of deep geological and near-surface RW deposition, as well as their designs, widely used cement and materials based on cement as engineered safety barriers (ESB) such as: matrices for immobilization of RW of low and intermediate levels of activity (LLW and ILW), concrete block or cement filling buffer, underground tunnel

concrete plugs involving RW containers, as concrete lining of tunnels or boreholes in the host rock, and as a component of multi-purpose container (MPC) for the disposal of high-level waste (HLW) [5–10].

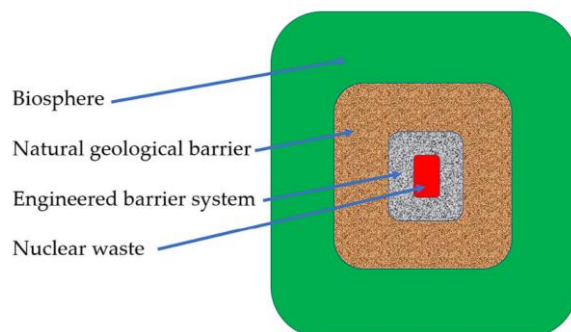


Figure 1. Illustration of the multiple-barrier safety principle for GDF [1].

The use of cement in the disposal of RW and spent nuclear fuel (SNF) requires barriers based on cement to maintain isolation properties for several tens of thousands of years [6]. Therefore, to understand the possible mechanisms of adverse changes in cement-based materials under geological disposal facility conditions, there are a number of experiments carried out in situ at the GDF/DGR under construction (Grimsel test site, Switzerland; Äspö hard rock laboratory, Sweden; ONKALO underground research laboratory, Finland) and modelled under laboratory conditions [6,11–15]. Thus, the objective of this work is to analyze the international experience in the application of cement-based materials and their laboratory testing in projects and at existing (or closed) RW disposal facilities in deep geological formations. Consideration of this is relevant in connection with the development of the concept of a GDF on the territory of the Russian Federation in the Nizhne-Kansky Massif (Krasnoyarsky region).

2. International Cement-Based Materials Application in Design Concepts and Operating Facilities of RW Deposition

2.1. Cement as Matrix for LLW and ILW

Cement has a variety of physicomechanical and chemical properties, making it an appropriate barrier and matrix for immobilizing radioactive and toxic waste [16]. From the chemical perspective, hardened cement has a high pH (its pore solution) and forms hydration products with sorption and ion exchange properties. From a physical point of view, cement is a solid strong material with low hydraulic permeability in its hardened form, ensuring the safe transport and storage of radioactive waste. Cement is also an inexpensive and easily accessible material, durable in its cured form, liquid at the stage of its initial forming and resistant to various forms of radioactive waste, which can be incorporated into it in both its liquid and solid states [16]. Hydrated cement has also been found to be resistant to radiation and capable of acting as a radiation shield.

Three methods of cementing are used for different types of radioactive waste in international practices [16].

1. Cementing directly in the barrel/container. During this process, the components, including radioactive waste, are mixed in a standard 200 L steel barrel using a single or interchangeable mixing element. After the cement is grasped and solidified, the barrel with the resultant compound is closed with a lid and sent to a near-surface disposal facility.
2. Cementing by a mixing plant. In this method, cement and waste are mixed to a homogeneous state and poured into standard 200-L steel drums or containers. The barrel/container shall be locked and, once the strength has been established, disposed of in the near-surface facility.

3. Cementing in situ. Radioactive waste cementing is often carried out on a large scale. For example, in India, 4 m³ tanks are used for radioactive waste conditioning, which are placed in a concrete ditch on a vermiculite layer. The air-conditioning tanks are interconnected at a certain level to avoid overflow. They are also equipped with one-time mixing mechanisms and a cement injection channel. All the tanks in the reinforced concrete trench are connected to an exhaust system equipped with a cyclone separator, filters and a blower. Since immobilization in cement is made inside the conditioning vats, this process is defined as «cementing in situ». This process is also used in the United States, where vats and bunkers of up to several million liters are filled with cemented radioactive waste.

Many countries have developed their approaches to the choice of curing materials, additives and process design. The experience is extensive [17,18]. Table 1 lists countries and enterprises where typical cementing processes and installations are used.

Table 1. International experience in RW cementing [17,19].

Location	Type of RW	Process	Productivity by Initial Waste
Belgium, NPP (three)	Concentrates, slurries	Stirring in drums	2 m ³ /day
UK, Hinckley Point, NPP *	Sludge of spent fuel holding basins	Stirring in drums	12 drums 80-L volume per day
India, Tarapur NPP	Sludge	Cementing at disposal site	500 m ³ /day
India, The Bhabha Atomic Research Centre (BARC)	Sludge	Cementing at disposal site	225 m ³ /day
Netherlands, Center of Nuclear Research in Petten	Sludge, liquid waste	Pre-stirring, final in drums	5 m ³ /day 0.5 m ³ /day
USA, Los Alamos National Laboratory	Concentrates of ILW	Stirring in drums	4 m ³ /day
USA, Brookhaven National Laboratory	Evaporation concentrates	Addition of concentrate to cement mixture with vermiculite (1:3) in concrete containers with a capacity of 4.2 m ³	15 m ³ per 6 months
France, Marcoule, NPP	Solid and liquid RW	Stirring in concrete containers	6 m ³ /day 3–5 m ³ /day
France, Nuclear Research Centre at Fontaine-au-Rose	Evaporation concentrates	Stirring in the drum (cement + vermiculite)	0.3 m ³ /day
France, Centre for Nuclear Research in Saclay	Sludge	Pre-mixing and discharge into concrete containers	100–300 kg/day
France, Nuclear Research Centre in Cadarache	Evaporation concentrates	Stirring in concrete containers	1.7 m ³ /day
France, Waste Management Centre in La Manche	Compacted solid waste	Cementing in drums	20 m ³ /day
Germany, NPP	Evaporation concentrates, sludge	Stirring in drums	2–7 m ³ /shift
Germany, Center for Nuclear Research in Jülich	Concentrates of LLW	Stirring in drums	50 L/day
Germany, Nuclear Research Centre in Karlsruhe	Concentrates of ILW	Stirring in drums	3–4 m ³ /shift
Switzerland, NPP (two)	Evaporation concentrates, sludge, ion-exchange resins	Stirring in drums	10–25 drums/day
Sweden, Ringhals NPP, Oskarshamn NPP	Evaporation concentrated, ion-exchange resins	Stirring in 1 m ³ concrete containers	2–5 containers/day

* There are three nuclear power plants «Hinckley Point»: two operating «Hinckley Point A» and «Hinckley Point B», and «Hinckley Point C», under construction.

Cement matrices serve to immobilize different kinds of waste: spent ion-exchange resins are typically immobilized using slag Portland cement mixtures; sludge and concentrates from LLW treatment are handled with ordinary Portland cement (OPC) with or without additives such as slag or fly ash; liquid ILW (LILW) are usually localized with slag Portland cement mixtures or OPC with vermiculite; mixture of sludge and ion exchange resins are isolated using OPC or slug Portland cement mixtures; secondary waste from the treatment of spent solvents at the processing plant, RW generated during processing SNF based on thorium and evaporator concentrate containing boric acid are immobilized using OPC [16].

For immobilization of high-level waste, the cementing method is not widely used due to the possibility of degradation of cement compounds under high radiation loads, causing radiolysis of water in cement pores to form free radicals with high reactivity [5]. However, in recent years, the Russian Federation's work is underway to investigate the possibility of immobilizing high-level waste by cementing [20–22]. Along with HLW, in the planned variant of geological disposal on the territory of the Russian Federation, the isolation of cemented liquid ILW [23] is considered, which, despite the accumulated extensive experience in using this method in this particular country, requires consideration of international practices for cementing radioactive waste.

2.2. Cement as a Buffer Material for Radioactive Waste Disposal

2.2.1. Cement as a Backfill Material in the Deep Geological Repository Project for Long-Lived (LL) High-, Intermediate-, Low- and Short-Lived (SL) High-Level Waste (Belgium)

In the Belgian repository project [24], it is planned to dispose long-lived high-, intermediate- and low-level radioactive waste at a depth of 230–240 m in boom clays.

Radioactive waste containers will be located in 1 km of horizontal tunnels lined with concrete slabs in order to avoid swelling of the tunnel walls due to the plasticity of the clays. Waste packages will be arranged in relation to each other without gaps by pushing them along the tunnel. The tunnels will be covered with cement. The choice of cement as a backfilling and lining material is due to its ability to hold radionuclides properly for the required isolation time, display shielding properties in relation to ionizing radiation, its low cost and extensive application experience. The tunnels will be connected by a common gallery connected to the surface of the two shafts [25].

It is planned to dispose long-live low-level and intermediate level waste packages by placing them in a concrete reinforced container and then filling the voids between them with cement mortar to form monoliths, which, when placed in tunnels, are proposed to be backfilled with cement with a layer thickness of 10 cm (Figure 2c).

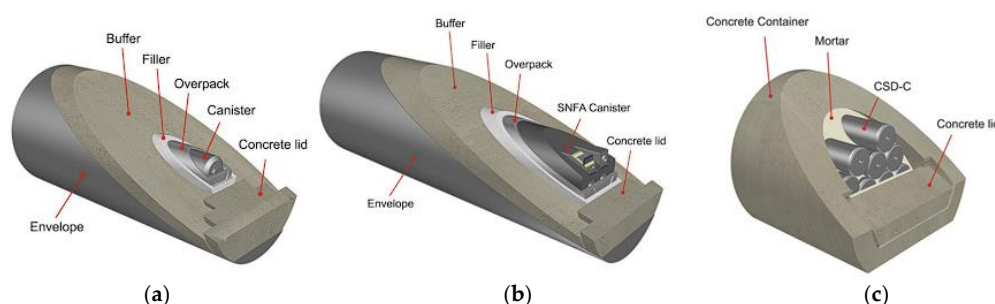


Figure 2. Construction of «supercontainers» for high- (a,b) and long-lived low- and intermediate-level radioactive waste, and monolith (c) for long-lived low- and intermediate-level waste disposal [24].

The Geological Repository Project proposes the use of “supercontainers” for disposal of long-lived and short-lived high-level radioactive waste, which are canisters including RW placed in a carbon steel container filled with cement mortar. The high-level radioactive waste containers shall be filled with a 40–50 cm thick cement buffer [25] in the tunnel. Because the vitrified HLW and spent nuclear fuel assembly (SNFA) canisters emit different amounts of heat, the distance between the individual tunnels should be different: between vitrified HLW—50 m; with SNFA—120 m [25].

2.2.2. Cement Buffer NRVB (Nirex Reference Vault Backfill) in the Concept of Intermediate-Level Radioactive Waste Isolation in Geological Disposal Facility (United Kingdom)

A proposed radioactive waste geological disposal concept in the United Kingdom provided two separate isolation options at depths of more than 500 m below ground: cemented intermediate-level waste in horizontal tunnels, vitrified high-level waste and spent nuclear fuel in vertical boreholes. Cement and clay backfill, respectively, serve as buffers for these options [26].

The so-called high-alkaline cement buffer NRVB [27] was considered for the intermediate-level waste tunnel isolation option, which is a mechanical mixture of OPC with hydrated lime (calcium hydroxide, $\text{Ca}(\text{OH})_2$) and calcium carbonate (CaCO_3) using the ratios presented in Table 2.

Table 2. Composition of cement buffer mixture NRVB [26].

Component	Mass Content, %
OPC (CEM I 52,5N)	26
Calcium carbonate (CaCO_3)	29
Hydrated lime ($\text{Ca}(\text{OH})_2$)	10
Water	35

This buffer material based on Portland cement was developed in the 1990s to meet a number of requirements for geological disposal facilities in the UK [28], such as [28,29]:

- The high hydrogen index of the medium (pH) provided by the dissolution of the various mineral components of the cement buffer by underground water, which makes it possible to significantly reduce the mobility of many radionuclides;
- High porosity and, consequently, permeability to ensure homogeneous chemical conditions, allowing for the removal of gaseous products resulting from corrosion of metal components of radioactive waste (H_2), activity of micro-organisms in aerobic and anaerobic conditions when they decompose organic radioactive waste (CH_4 , CO_2), as well as a number of radioactive gases (containing isotopes ^3H , $^{14}\text{CO}_2$, $^{14}\text{CH}_4$, ^{222}Rn) [27,28] and the creation of a high specific surface area for the sorption of radionuclides;
- Low compressive strength of the material (4.9, 5.9 and 6.3 MPa on 7, 28 and 90 days of curing, unlike OPCs under almost identical conditions—31, 45 and 46 MPa on 7, 28 and 90 days, respectively [30,31]) to create a potential recovery of waste packages [28];

Although Nirex merged into Nuclear Decommission Authority and ceased to exist in 2007 [32], the NRVB cement buffer is being considered as an alternative option for geological disposal in the Nizhne-Kansky massif [33].

2.2.3. Cement Mortar as a Filler for Concrete Containers with Short-Lived Low- and Intermediate-Level Waste in Geological Repository in Bataapáti (Hungary)

The nuclear power plant waste repository in Hungary is carried out in granite host rock at a depth of 250 m. Radioactive waste are converted to a solid state and then placed in steel barrels with a volume of 200 L. Further barrels, in turn, are placed in a concrete container, then filled with a mortar of cement compounds with liquid radioactive waste (Figure 3) [34].

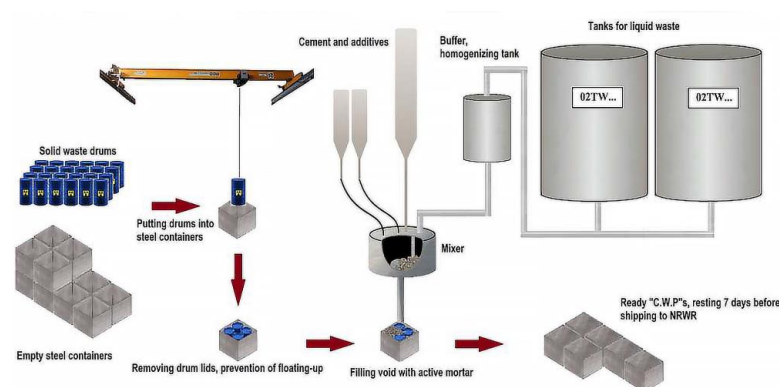


Figure 3. Preparation of solid radioactive waste for disposal at the Hungarian nuclear power plant [35].

In this form, the obtained concrete containers with radioactive waste are transported to the geological repository in Břatapařt, inside which they are placed in a chamber 90 m long [34].

2.2.4. Cement as a Component of the Backfilling Material in the Geological Repository of Low- and Intermediate-Level Waste at Morsleben (Germany)

The radioactive waste repository at Morsleben is located in salt rocks in unused mines. Due to the lack of up-to-date safety requirements for disposal facilities in Germany, the repository is currently being closed down until approximately 2028 [36]. This repository received solid and compacted waste in steel barrels of 200 L.

At the closing stage, mines with radioactive waste to reduce the flow of groundwater will be covered with a special composition: 16% cement, 39% halite, 16% lime, 14% water, 15% sand [25].

2.2.5. Cement Filling of Containers with Low-Level Waste of Geological Disposal Facility Cigéo (France)

According to the Cigéo [37] disposal facility concept, it is planned to be located at a depth of about 500 m in the Callovay–Oxford clay formations. The waste will be disposed of in solid form, arriving at the GDF already packed and further isolated on site in steel canisters, which will be placed in horizontal tunnels with metal lining. Long-lived intermediate- and high-level waste will be disposed in separate parts of the GDF and the size of the chambers for them will be: ILW—a few tens of meters in diameter and several hundred meters long; HLW—a few tens of meters in length and only 70 cm in diameter. When disposing of HLW, waste containers will alternate with buffer material to reduce the temperature in the cells shown in Figure 4.

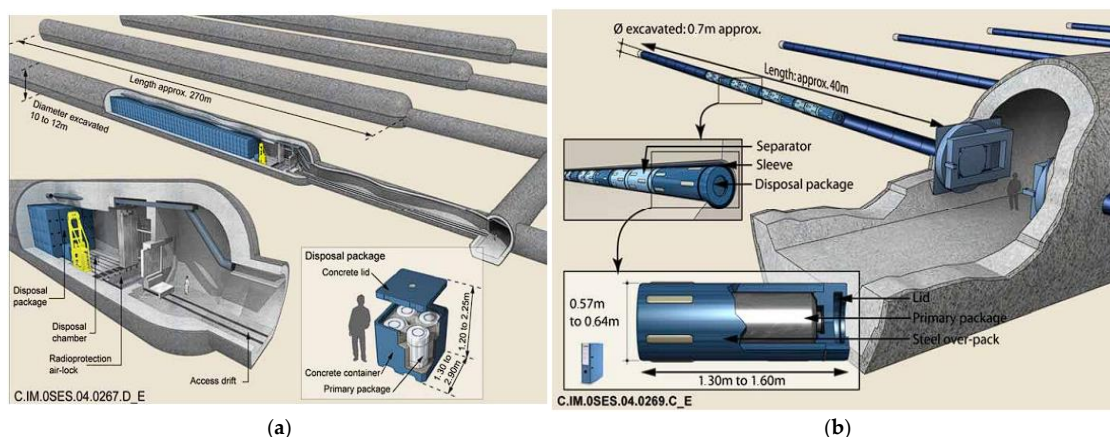


Figure 4. Illustration of containers with intermediate- (a) and high-level waste (b) and the order of their placement in the tunnels of the geological disposal facility Cigéo [38].

Containers with intermediate-level waste canisters, after filling, shall be backfilled with cement mortar [38] to provide additional mechanical protection during transport and to create an additional engineered barrier, reducing the corrosion rate of a steel container when groundwater penetrates the disposal facility.

2.2.6. Special Cement Composition as Radioactive Waste Chamber filling Material for Low- and Intermediate-Level Waste in the Geological Disposal Facility Project (Switzerland)

Disposal of low- and intermediate-level waste in Switzerland will be carried out in geological formations of host rock formed from opalinus clay and/or clayey limestone. The intermediate- and low-level waste are isolated in solid form by means of cementing, bitumizing or alloying in a plasma furnace into a slag-like mass [25,39]. The waste is isolated in metal barrels, which are placed in a concrete container filled with cement mortar (Figure 5).

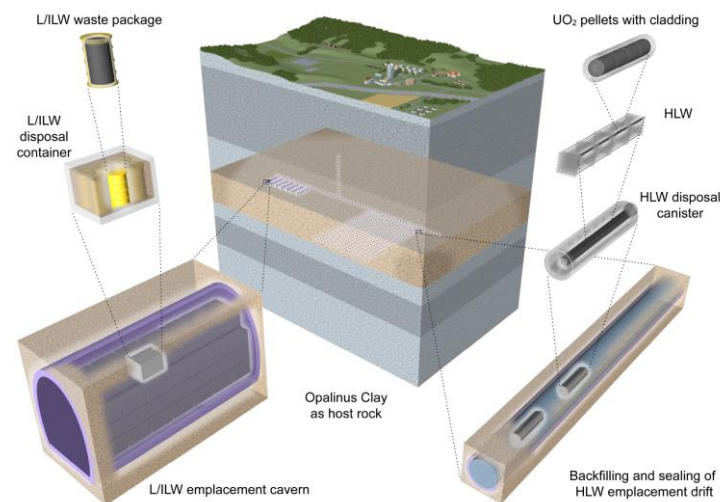


Figure 5. Layout of ILW/LLW and HLW and organization of safety barriers around them in geological disposal facility project [40].

Radioactive waste containers will be located in several tiers in the tunnels of the disposal facility (Figure 5). At closing, the tunnel will be filled with cement mortar of increased viscosity, which will become an additional engineered barrier. The final safety barrier is clayey host rock [25].

2.2.7. Cement Backfilling as One of the Options to Fill the Gaps between Radioactive Waste Containers in the SFL Geological Repository Project for Long-Lived Low- and Intermediate-Level Waste (Sweden)

The SFL repository will be located in crystalline rocks at a depth of 300 m and is designed for long-lived low-level waste. The disposal facility will consist of two drifts: SFL-3 for legacy and operational waste; SFL-5 for decommissioning waste. There are four disposal options [25].

The first option involves filling radioactive waste with gravel, the second—placing the waste inside the concrete structure and filling the free space with cement mortar, the third—placing the waste inside the concrete structure and filling the free space with bentonite briquettes. The fourth version of the concept involves the disposal of waste in a bunker with double concrete walls. The bunker will be installed on a gravel base, the space between the outer wall of the bunker and the surrounding rocks will be filled with gravel. The space between the inner and outer walls will be filled with bentonite (top and sides) and a mixture of bentonite and sand (bottom). After filling, the bunker will be covered with gravel up to the ceiling of the tunnel. The bunker will be 40 m deep and 35 m in diameter [25].

2.3. Concrete as a Material for Lining and Isolation of Tunnels for Radioactive Waste in Geological Repository/Disposal Facility

The use of concrete as a material for lining and isolating repository tunnels is explained by its low cost, extensive experience in construction, the ability to firmly retain radionuclides during the period of radioactive waste isolation, shield ionizing radiation, provide acceptable hydraulic permeability of tunnels from the penetration of groundwater of repository/disposal facility and reduce corrosion rates on surfaces of steel containers [25].

2.3.1. Concrete as Material for Tunnel Lining in the Host Rock of Bataapáti National Radioactive Waste Repository (Hungary)

As a result of the change of the concrete container for repository of low- and intermediate-level waste in the National Geological Repository in Bataapáti (Hungary), the construction of the chambers with radioactive waste was changed: tunnels in the host rock began to reinforce

concrete facing. The chambers shall be divided into blocks for additional reinforcement of the structure and the addition of a step-by-step backfilling of the chambers [41].

2.3.2. Concrete as a Drift Lining Material in High-Level Waste and Spent Nuclear Fuel Deep Geological Repository Project (Spain)

At present, Spain is continuing to develop the concept of high-level waste and spent nuclear fuel repository, which is based on the most widely used in the world: HLW and SNF containers will be placed in drifts in granite or clay rock, surrounded by blocks of compacted bentonite as buffer [42].

It is also planned to ensure lining and plugging of geological repository drifts using concrete [6,13] as an additional engineered safety barrier.

2.3.3. Concrete as a Material of Tunnel Plugging, Isolation of Waste Chambers in the Low- and Intermediate-Level Waste Repository Project in the Municipality of Kincardine (Canada)

The placement of radioactive waste in the geological isolation facility project is planned at a depth of 680 m in clayey limestone under Bruce site, not far from the municipality of Kincardine, Ontario [43]. The proposed repository concept is depicted in Figure 6 and consists of parallel rows of waste cells located on either side of the central access tunnel: on one side there will be cells for LLW and on the other there will be cells for ILW [43].

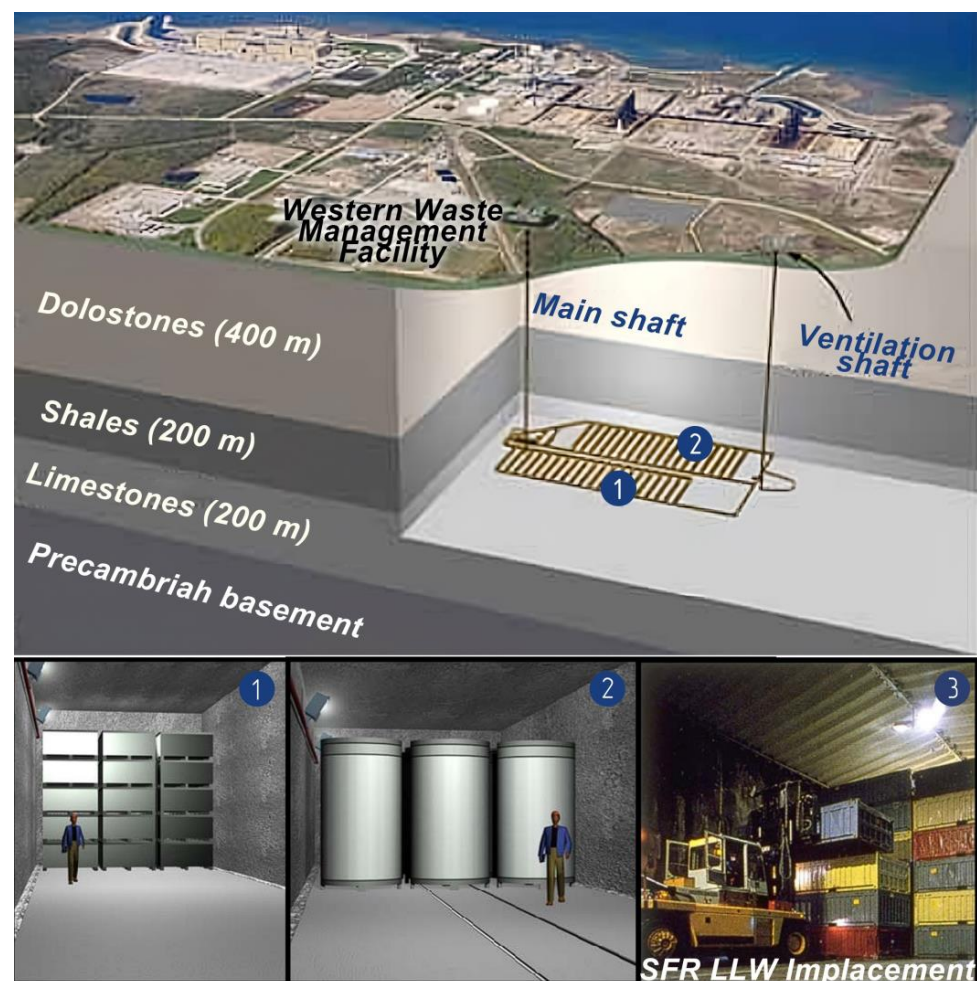


Figure 6. Proposed concept of placing LLW and ILW in the Bruce site repository and illustration of disposal cells with (1) low-, (2) intermediate-level waste and (3) operations to place radioactive waste in SFR repository (Sweden), much like those in the planned Canadian one [43,44].

It is planned to strengthen the floor of the radioactive waste cells with concrete, and after filling it with concrete wall without backfilling and sealing [43]. Access shafts to the cells will be isolated with concrete caps and low-permeable materials, in particular a mixture of bentonite and sand [43].

2.3.4. Concrete as a Lining and Plugging of Tunnels Material with Intermediate- and High-Level Waste in the Project of Geological Disposal Facility Cigéo (France)

Tunnels for intermediate- and high-level waste placement in the Cigéo geological disposal facility [38] are planned to be supported from the inside by the application of shotcrete on the walls with additional strengthening in the form of various reinforcements, after which the tunnel is cladded with non-reinforced concrete (Figure 7).

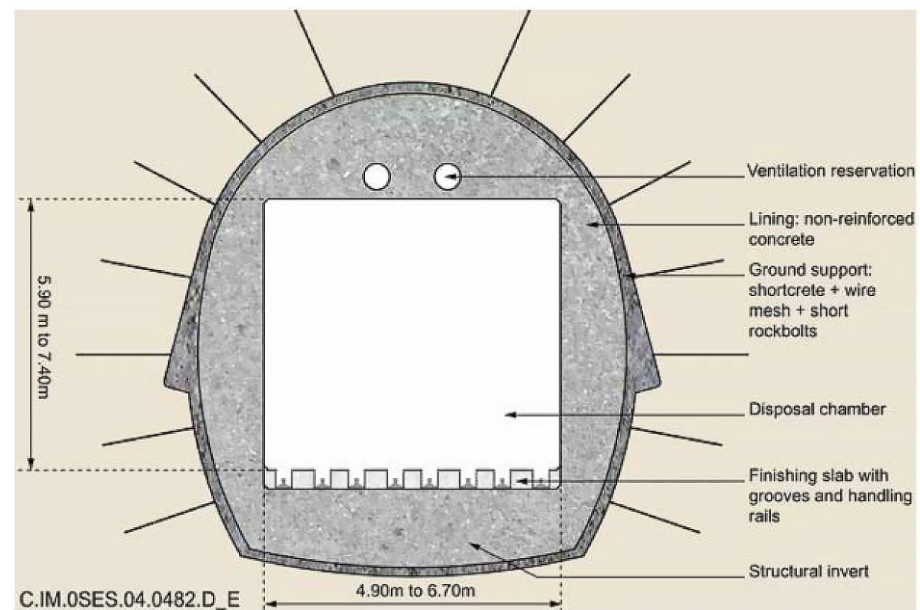
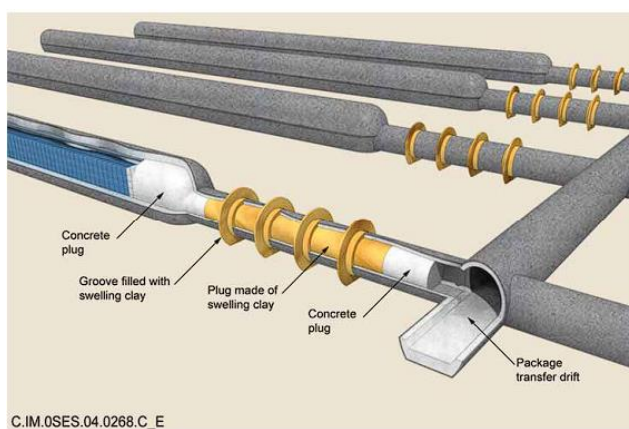
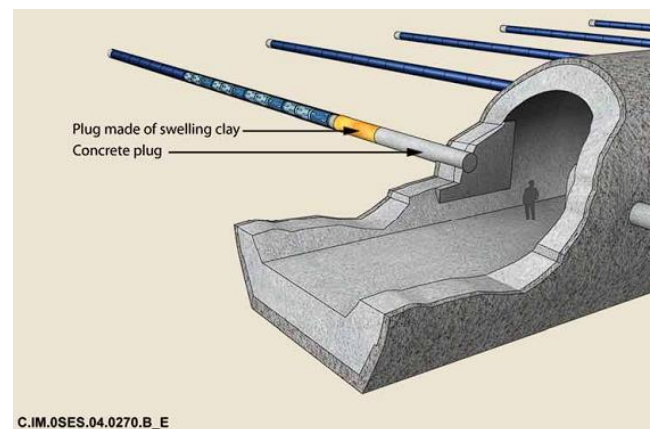


Figure 7. Concrete reinforcement and lining on the example of the cross-section view of tunnel for isolation of the intermediate-level waste in the tunnel of the geological disposal facility Cigéo [38].

After the end of the operation of the disposal site, the waste tunnels will be sealed with bentonite and concrete blocks (Figure 8) [25].



(a)



(b)

Figure 8. Layout of intermediate- (a) and high-level waste (b) in the tunnels of the Cigéo geological disposal facility and their isolation with concrete plugs [38].

2.3.5. Concrete as Bunker Lining Material of the Short-Lived Low- and Intermediate-Level Waste Geological Repository SFR (Sweden)

The SFR repository for the low- and intermediate-level waste (Sweden) is located at the Forsmark Nuclear Power Plant under the Baltic Sea at a depth of 60 m in crystalline host rock [45].

The most active low-level waste is disposed in a cylindrical bunker SILO (Figure 9), located in a shaft with a diameter of 30 and a depth of 60 m, lined with 0.8 m thick concrete, based on sand and bentonite [25]. The low-level waste is emplaced in the bunker in containers made of steel or concrete, the voids between which are filled with cement mortar [45].

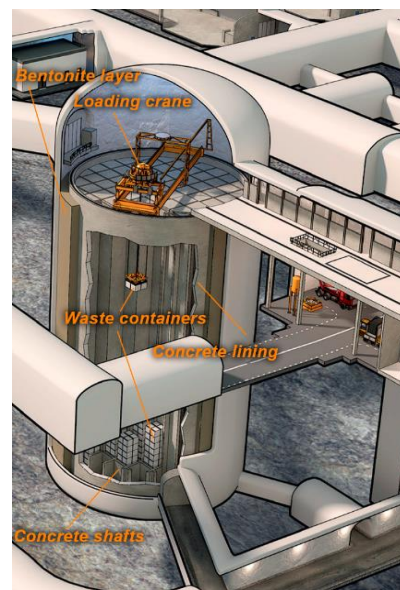


Figure 9. SFR repository SILO bunker cross-sectional view [45].

2.3.6. Low-pH Concrete as Tunnel Plug Material in Design Concepts for High-Level Waste and Spent Nuclear Fuel Deep Geological Repository KBS-3 (Sweden, Finland)

The KBS-3 concept provides for isolation of high-level waste [46] and spent nuclear fuel [47] in crystalline host rock [48]—granite [49]. Two variants of radioactive waste containers emplacement are considered, the schematic of which is shown in Figure 10—vertical in boreholes (KBS-3V) and horizontal in tunnels (KBS-3H) [33,49,50].

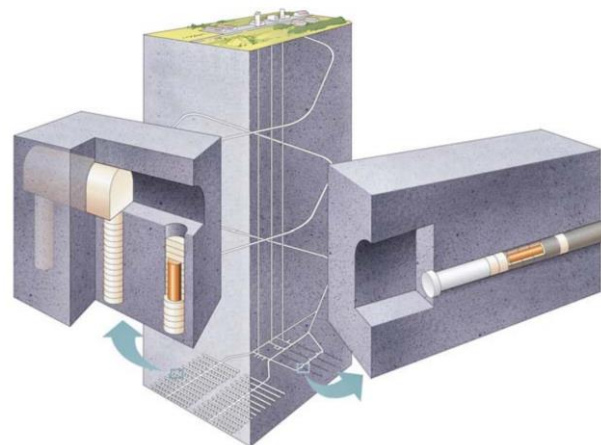


Figure 10. Cross-sectional view of vertical (KBS-3V, left) and horizontal (KBS-3H, right) method of radioactive waste container placement in KBS-3 concept [49].

In either variant, the installation of radioactive waste containers inside the buffer made of compacted bentonite is considered and should be plugged after the final filling (for KBS-3H, additional plugs are also provided inside the tunnel to isolate individual compartments with radioactive waste) [49,50]. In the case of KBS-3V, low-pH concrete, the composition of which is indicated in Table 3 (kg component per m³ concrete) [49,51], while for KBS-3H decided to abandon concrete, since the Äspö hard rock laboratory experiments revealed the following conditions that the reinforced concrete plug could not withstand: hydrostatic pressure (5 MPa) and swelling pressure of the compacted bentonite (<10 MPa) [50].

Table 3. Composition of low-pH concrete B200 for plug construction in KBS-3 repositories [51].

Component	Amount, kg/m ³	Mass Content *, %
Sulphate resistant Portland cement CEM I 42,5 N SR3 MH/LA, produced by Anläggningcement Degerhamn, Sweden	120	5.14
Silica fume or silica fume slurry ** (SiO ₂)	80	3.43
Limestone Limus 25 (CaCO ₃)	369	15.80
Water	165	7.06
Sand 0-8 mm (natural, from Äspö site)	1037	44.41
Gravel (natural or crushed)	558	23.90
Superplasticizer Glenium 51	6	0.26
Total	2335	100.00

* Recalculated in perspective of the mass amount of each component per m³ of the mixture shown in the sources.

** Dense silica fume was used for laboratory testing, and the slurry was used to cast large-scale structures because of the ease of dispersion and safety handling.

The report [52] analyzed the effect of adding superplasticizers on the mobility of radionuclides of the lanthanide and actinide groups under conditions of high pH values typical for cement materials and under conditions of model groundwater solutions. The results of the work in the report showed that under strongly alkaline conditions, there was no significant effect on the sorption of radionuclides Eu, Th and Ni on cement materials. However, in model solutions of groundwater, additions of superplasticizers containing carbohydrates/carboxylates led to effective complexation of trivalent lanthanides and actinides, and at lower pH, to binding of tetravalent actinides. This phenomenon should be taken into account when designing a geological facility for the isolation of radioactive waste, because under these conditions, the mobility of radionuclides increases. The effect of radiolysis products of various types of superplasticizers on the mobility of Eu, Ni and U was analyzed in [53]. Under alkaline conditions, at a concentration of radiolysis products of the order of 10⁻² mol/L, they do not have a significant effect on the mobility of the studied radionuclides. However, in a weakly alkaline medium (pH = 8–10), such radiolysis products as phthalic and acetic acids (at concentrations of about 10⁻² mol/L) increase the mobility of Eu and Ni, while none of the studied radiolysis products had no effect.

As of 2008, two design options for concrete plugs have been proposed: long conical (Figure 11) and short reinforced (Figure 12). Each of the proposed options provided both with and without bentonite intermediate layer [49].

Based on the experimental reports of the Äspö [54–56] hard rock laboratory, SKB has opted for a short reinforced plug with an intermediate layer of bentonite. The use of low-alkaline concrete is explained by the need to preserve the bentonite buffer's previous swelling ability, which is severely degraded in the strong alkaline solutions formed by filtering underground water through concrete structures [49].

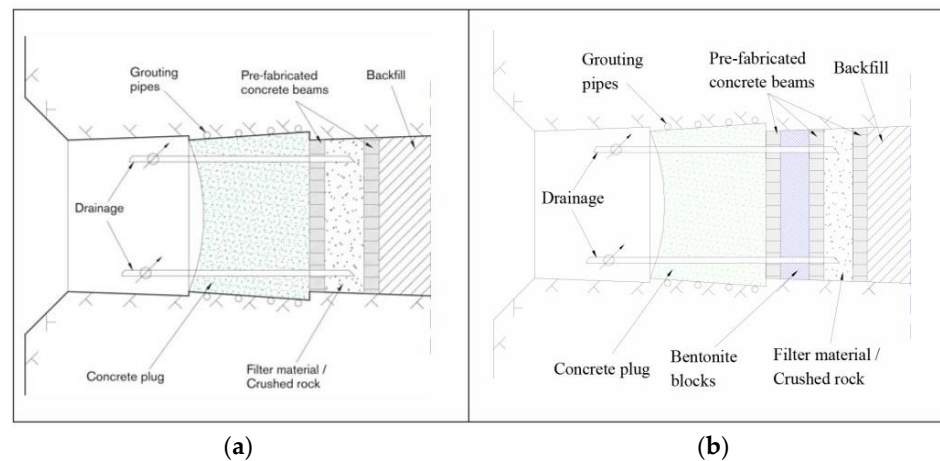


Figure 11. Design of conical concrete plug without (a) and with an intermediate bentonite layer (b) between the filling of the tunnel and the plug itself [49].

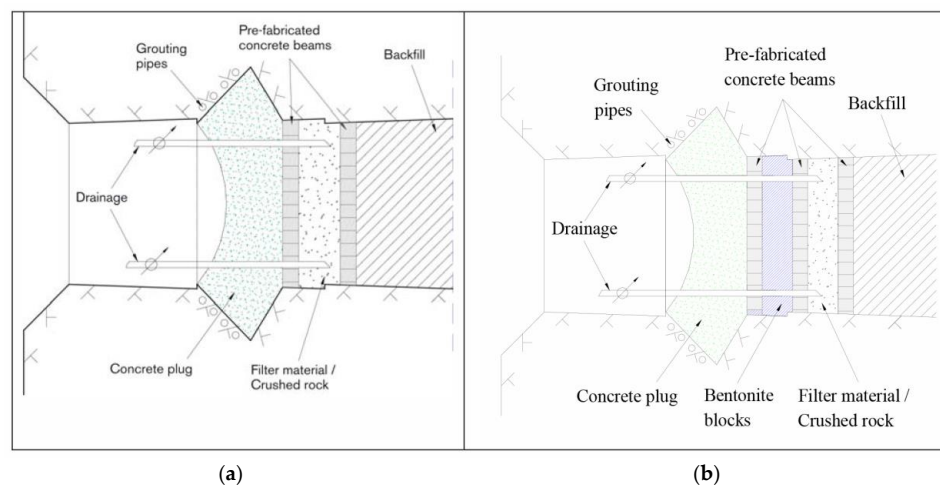


Figure 12. Design of short reinforced concrete plug without (a) and with an intermediate bentonite layer (b) between the filling of the tunnel and the plug itself [49].

2.3.7. Concrete as Lining and Plugging Material in the Geological Disposal Facility Design Concept of Disposal High-Level Waste and Transuranic Spent Nuclear Fuel (Japan)

The Japanese geological disposal facility design concept plans to place transuranic radioactive waste packages in horizontal tunnels (Figure 13) at a depth of ~500 m [57].

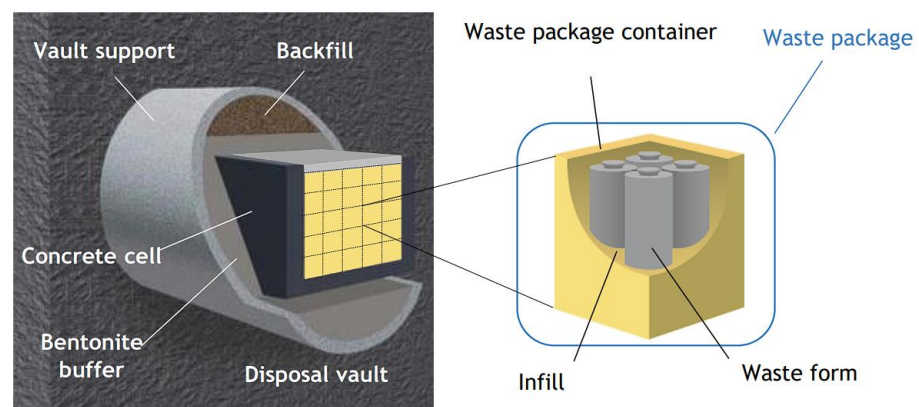


Figure 13. Illustration of transuranic radioactive waste emplacement in Japanese project of geological disposal facility [58].

It is planned to place transuranic radioactive waste canisters inside the container, after which the cement mortar is poured. The containers will be placed in a concrete lined tunnel filled with a bentonite buffer [57,58] and mechanically isolated by a concrete plug (Figure 14) [57,58].

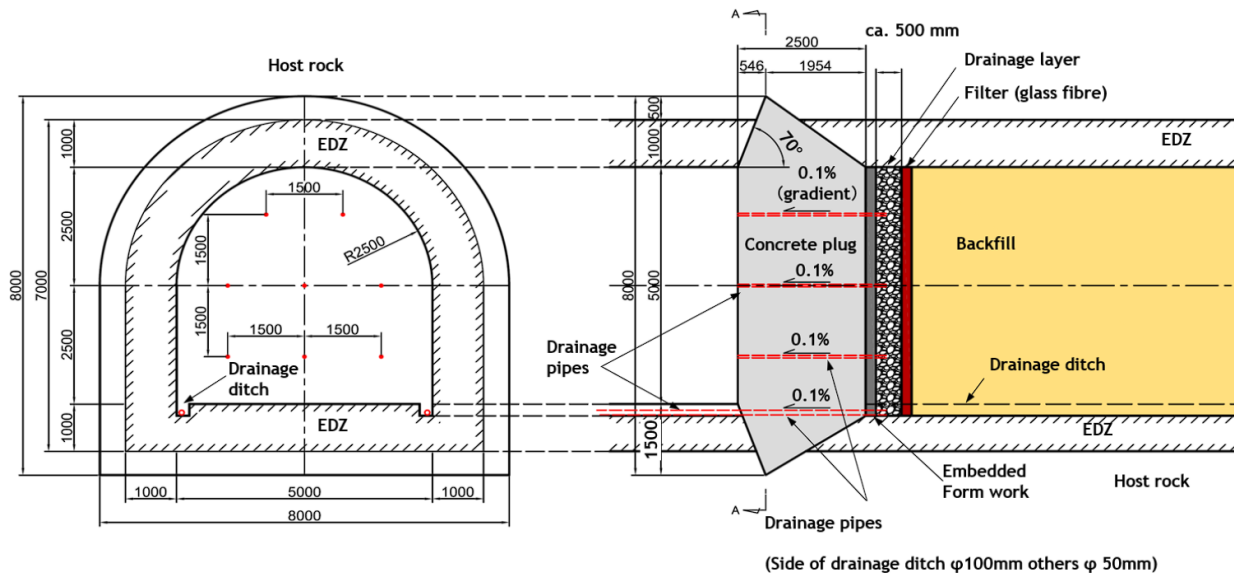


Figure 14. Concrete plug for horizontal tunnel isolation in Japanese geological disposal facility project [58].

In vertical placement, radioactive waste is considered as an option, lining the borehole with shotcrete (Figure 15) to ensure mechanical stability until it is filled with buffer material [58].

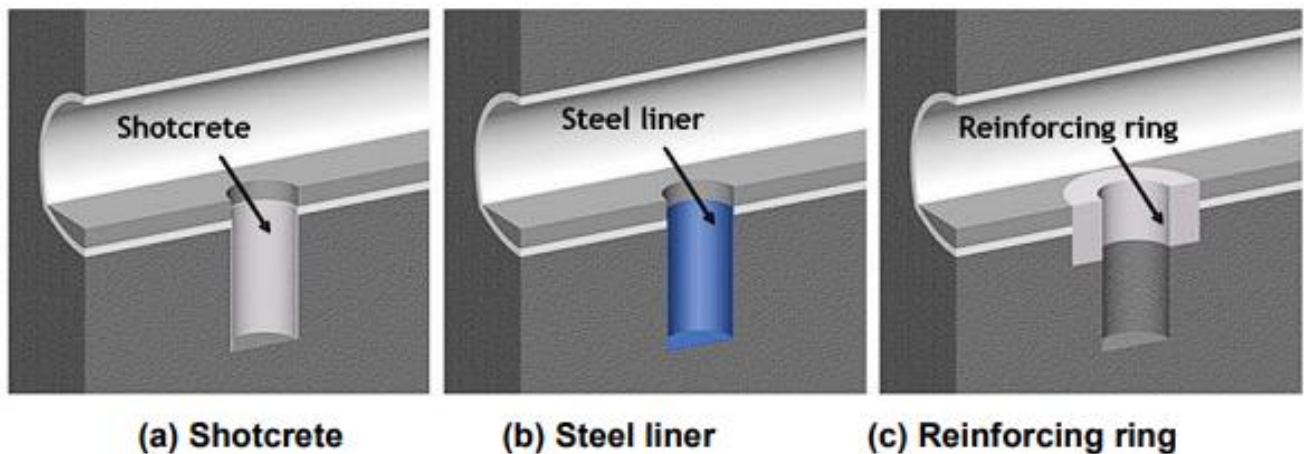


Figure 15. Borehole reinforcing options in Japanese disposal facility development reports [58].

2.4. Conclusions

Table 4 below provides basic information on foreign practices of radioactive waste disposal using cement-based materials, including concrete.

Table 4. Overview of the use of cement-based barriers.

Country	Repository/Disposal Facility	Type of Engineered Barrier
Belgium	DGR project	Drift backfill/One of supercontainer barriers
Canada	DGR project at Bruce cite	Concrete walls isolating RW containing chambers/Concrete caps isolating access shafts
Finland	GDR project (KBS-3V)	Concrete plugs and seals
France	Cigéo GDF project	Concrete plugs/Tunnel buffer mixed with bentonite/Concrete lining
Germany	DGR Morsleben	Mine backfill
Hungary	DGR Bataapáti	RW matrix/Container filling/Drift lining
Spain	DGR project	Concrete lining
Sweden	SFL DGR project for ILW/LLW disposal	Buffer
	SFR DGR for ILW/LLW disposal	Shaft lining
Switzerland	DGR project (KBS-3V)	Concrete plugs and seals
	GDF project for ILW/LLW	Cement matrix/Container filling/Buffer
United Kingdom	GDF project (ceased)	Cement matrix/Buffer/Container filling
Japan	GDF project for HLW and SNF	Container filling/Concrete lining/Concrete plugs

Analyzing the above information, it can be said that in almost all concepts of radioactive waste deposition in deep geologic formations, concrete (cement) is used as an engineered barrier material. The use of cement by its type of application can be classified as follows:

- Radioactive waste contained in cement matrices inside steel drums;
- The material of radioactive waste conditioned container and/or its buffer;
- Buffer material between radioactive waste container and storage walls;
- The material of the lining of the walls of the storage;
- Material isolating tunnels with placed radioactive waste packaging structures (tunnel plugs, walls and seals).

According to the radioactive waste-type arrangement, cement-based geological reservoirs can be classified as tunnels (horizontal) and boreholes (vertical).

In the Russian Federation, a number of radioactive waste GDF concepts in both horizontal and vertical landfills are being considered, each of which assumes the use of cement materials in accordance with the waste composition under consideration. At this disposal facility, the final isolation of RW of classes 1 and 2 is planned [33]. RW class 1 is formed as a result of the processing of SNF containing a number of radionuclides [59] (Table 5), including radioactive isotopes of iodine (for example, ^{129}I), which are especially dangerous for the biosphere. For reliable isolation of radioactive waste containing radioactive iodine, a number of effective sorbents containing silver compounds based on materials with a developed surface such as, for example, bentonite and silica gel [60] are currently being developed. The inclusion of a certain amount of modified bentonite capable of sorbing iodine into the composition of the compacted buffer will preserve the performance properties of bentonites and ensure environmental protection from radioiodine.

Options for creating a buffer zone include pouring cement mortar between the vertical boreholes with the RW class 1 supercontainers (high-level waste with high heat generation) (Figure 16) [61].

As the advantages of using cement backfilling, the authors of the work [61] indicate a decrease in the corrosion rate of the supercontainer in an alkaline medium caused by the dissolution of the cement buffer components by groundwater, as well as the possibility of sealing cracks in boreholes, while the probable scenario of erosion of the bentonite buffer by its «washing up».

Currently, the entire level of uncertainty is being assessed for the final decision on the choice of the concept of the organization of deep deposition of radioactive waste in the Russian Federation. In addition to the analysis of the existing concepts of GDF, it is necessary to analyze and change the key properties of EBS elements, their mutual influence

and other related processes in disposal facility, which will be discussed in the next section of this article.

Table 5. Specific activity of significant radionuclides from VVER-440 and BN-600 nuclear reactors SNF [59].

Radionuclide	Specific Activity, Bq/t U	
	VVER-440 (T = 7 Years)	BN-600 (T = 14 Years)
^{14}C	5.6×10^9	5.24×10^{10}
^{79}Se	8.58×10^8	2.38×10^9
^{99}Tc	7.50×10^{11}	1.44×10^{12}
^{129}I	1.03×10^9	2.29×10^9
^{135}Cs	1.85×10^{10}	1.39×10^{11}
^{234}U	3.23×10^9	1.94×10^{10}
^{235}U	3.62×10^8	1.34×10^{10}
^{237}Np	5.56×10^9	2.87×10^{10}
^{238}Pu	1.20×10^{13}	5.75×10^{13}
^{239}Pu	2.14×10^{13}	7.19×10^{13}
^{240}Pu	2.65×10^{13}	1.15×10^{13}
^{241}Pu	1.11×10^{14}	7.60×10^{13}
^{242}Pu	6.08×10^{10}	1.52×10^8
^{241}Am	9.83×10^{13}	2.48×10^{12}
^{243}Am	1.14×10^{11}	1.24×10^8
^{245}Cm	5.53×10^7	7.15×10^4

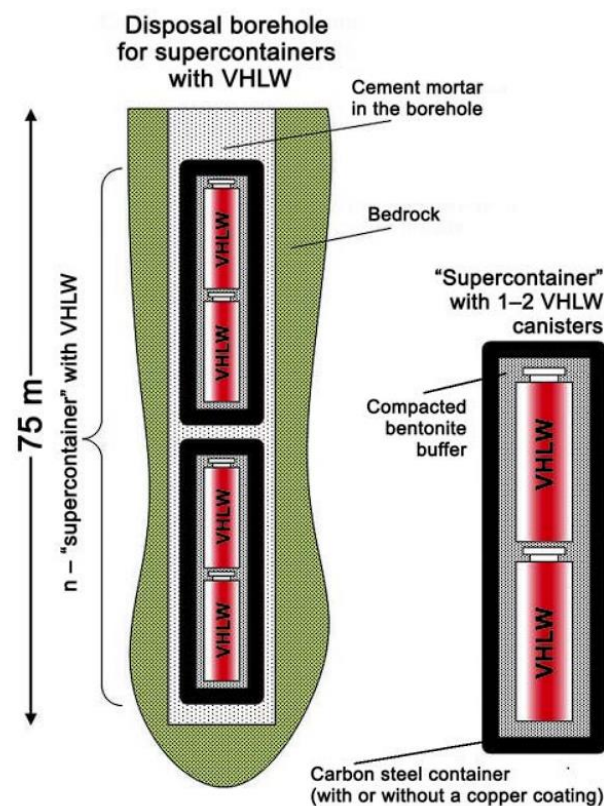


Figure 16. RW class 1 isolation option in cement mortar boreholes [61].

3. Changes in Concrete Engineered Barrier by Means of External Factors of Geological Disposal Facility

Since radioactive waste deposition in deep geological formation should be designed to isolate the waste over a long period of time, it is necessary to predict possible changes in the

main properties of concrete engineered safety barriers based on the analysis of the influence of many factors, such as: the effect of high temperatures and radiation doses, typical for high heat generating high-level radioactive waste; infiltration of groundwater characteristic of the containment environment; exposure of microbial communities and others.

3.1. Concrete Changes Due to Elevated Temperatures (above 100 °C) Typical for RW Class 1 Packages

Cement is being actively investigated as a backfilling material for low- and intermediate-level waste repositories and is also being considered for use in the radioactive waste large heat release geological disposal concept, for example in the Belgian supercontainer concept for high-level waste and spent nuclear fuel. The use of cement as a backfill in geological disposal facilities is conditioned by its ability to provide structural support, its ability to be used as a backfill directly inside the geological disposal facility, its ability to provide alkaline conditions, which reduce the corrosion of steel and the solubility of some important radionuclides and increase their sorption [62].

The effect of continuous exposure to elevated temperatures has been found to cause the following changes in the properties of the concrete [62]:

- Compressive strength decreases by about 25% at the specified maximum temperature of the operating geological disposal facility of 300 °C and continues to decrease with greater force when this limit is exceeded. Strength degradation tends to increase when cement dries.
- Modulus of elasticity decreases in volume than strength and is a more sensitive parameter to external influences. The relative modulus tends to decrease linearly from 100 °C to almost zero at 800 °C but fluctuates strongly at 300 °C—the relative value is in the range of 40 to 90%. However, this significant reduction of the elastic module does not endanger the integrity of the geological disposal facility barriers. Maintaining a high modulus of elasticity is critical if the strain under the load is limited. In the geological disposal facility, the primary load at elevated temperatures will be the result of limiting temperature and shrinkage strains. Under these conditions, achieving a low elastic modulus value is useful because it reduces the stresses caused by limiting deformation and therefore reduces the risk of cracking.
- Temperature expansion coefficient of concrete is approximately constant at the maximum operating temperature of 300 °C, with a value of $10 \pm 2 \cdot 10^{-6}$ mm/(mm·°C). For cement mortar, deformation at elevated temperatures can be reinforced by shrinkage during drying, so the final deformation is caused by shrinkage.
- Thermal conductivity of concrete decreases approximately linearly over the entire range of 20 to 300 °C, with a relative coefficient of thermal conductivity around 0.4.
- Hydraulic permeability of concrete increases with temperature. This conclusion is based on a limited amount of data on the change of porosity of concrete at elevated temperatures and using information on the permeability to porosity ratio, its estimated value, assumed for the maximum operating temperature of 300 °C may be 10 times higher than for the standard temperature. This could result in a high flow rate of groundwater through the geological disposal facility barriers, even though the effect of changes in cement materials would be relatively small and comparable to cracking.

The work [63,64] describes the study of the mechanical and thermal properties of concrete, which were grouped into a visual diagram (Figure 17) that describes the physical and chemical processes that occur in concrete when the temperature increases.

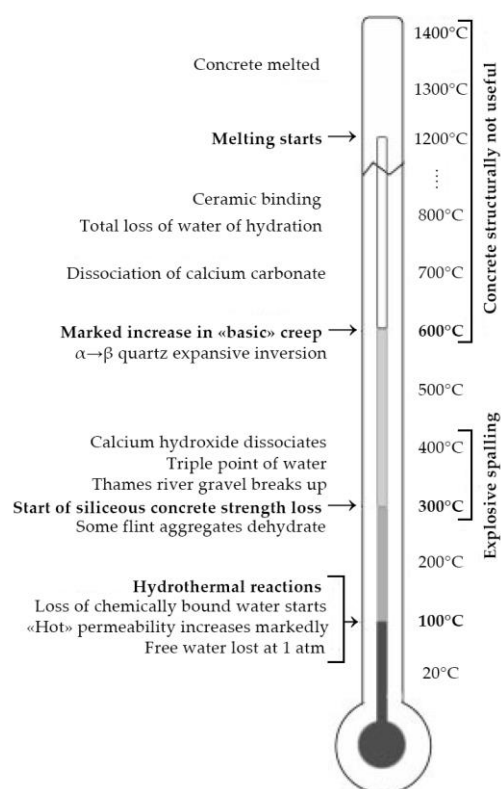


Figure 17. Physicochemical changes in concrete from ordinary Portland cement in heating process [63].

3.2. Changes in Concrete Due to Groundwater in Geological Disposal Facilities

When groundwater enters the geological disposal facility, its active components, such as calcium hydroxide and amorphous cement CSH-gel formed by means of hydration of the ordinary Portland cement, dissolve with cement insertion. Assessments based on accelerated leaching of NRVB cement material (considered to be an analogue of the cement barrier for RW class 2 in geological disposal facility at Nizhne-Kansky massif) showed that it contains a sufficient amount of $\text{Ca}(\text{OH})_2$ to maintain $\text{pH} > 12$ in the volume of underground water passing through it, more than 200 times the volume of cement backfill. Once $\text{Ca}(\text{OH})_2$ is dissolved, high pH will be achieved by dissolving the CSH-gel. The volume of water with $\text{pH} > 10$ is about 800 times higher than the volume of the gel. Consequently, the required volume of groundwater to maintain a high pH is about 1000 times that of NRVB [61]. The dissolution of $\text{Ca}(\text{OH})_2$ groundwater and CSH-gel from the concrete structure should lead to an increase in porosity and, consequently, a decrease in strength.

The evaluation determines that the volume of cement around containers with a diameter of 0.7 and a height of 3.5 m in the geological disposal facility, where the average hydraulic permeability (filtration coefficient) (0.04 m/year) and the hydraulic pressure gradient (0.05) determines the average flow of groundwater at the vertical location of the wells to be approximately 19 m³, which corresponds to a borehole diameter of just over 2.5 m, the flow of which is not subject to technological difficulties [61].

In the absence of changes to the cement geological disposal facility in evolution, except for interaction with groundwater, groundwater conditioning for very high level waste (VHLW) at Nizhne-Kansky massif will be possible for a million years after closure [61]. This conclusion made by the authors in [61] is based, as mentioned earlier, on the approximation that the mineral composition of the cement buffer does not change as a result of groundwater filtration and their following interaction. To obtain more complete data of the duration of chemical containment, it is necessary to take into account the passivation of the mineral surfaces of the cement buffer, which occurs as a result of the interaction of

carbonate- and sulfate-anions contained in the underground water of the Yeniseisky site with $\text{Ca}(\text{OH})_2$, followed by gypsum and calcite precipitation.

In the experiment [65] tests were carried out to determine the hydraulic permeability of concrete (filtration coefficients) on a specially prepared setup (Figure 18). For this experiment, cylindrical concrete samples were prepared with the addition of a silica fume in the ratio of 60% of the ordinary Portland cement, 40% of the silica fume and the amount of water relative to the binder is 0.5 (i.e., water-cement ratio, W/C), measuring 5 cm in diameter and height, and also cores from the plug of the test site Äspö (Sweden) of the same size, made of shotcrete. Samples of concrete with the addition of silica were hardened for 2 years. Through these samples, the granite underground water of the Äspö site was passed under pressure of 0.5 bar to simulate the actual conditions of the further repository [65].

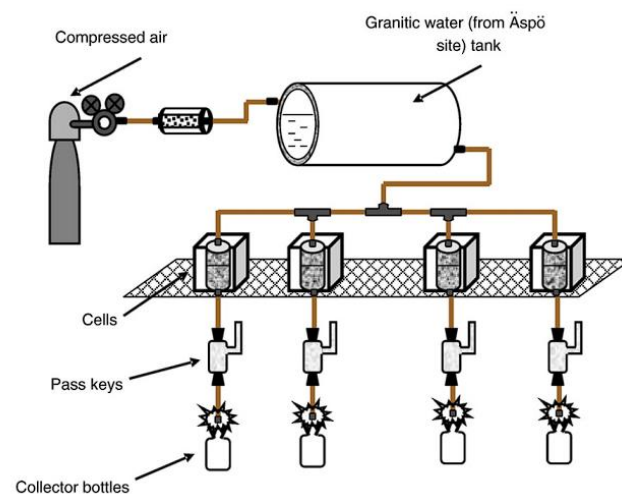


Figure 18. Experimental setup [65].

Cylindrical samples of concrete were placed between two cylinders made of methacrylate with holes for inlet and outlet of incoming water. To pass the water in the right direction, the samples were hydroisolated with epoxy resin shell. For collecting the water passed through the samples, reservoirs are provided (Figure 19). To determine the hydraulic permeability of the concrete, the flow from the granite water concrete cells was measured. These tests were carried out over a period of 14 months for silica-fume concrete and 2 years for shotcrete [65].

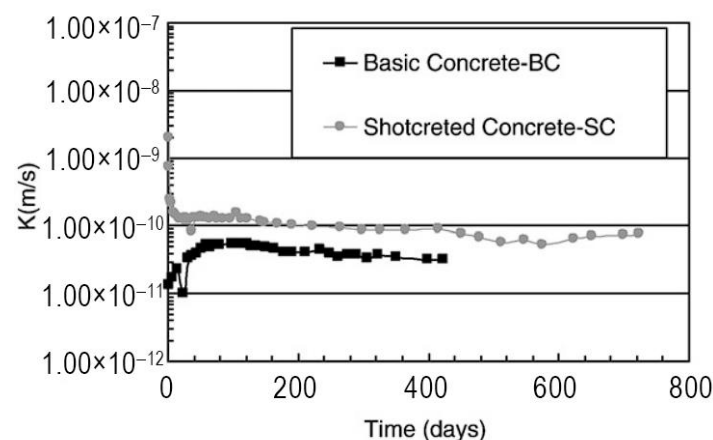


Figure 19. Variation of hydraulic permeability of concrete over time of the experiment [65].

The hydraulic permeability of the concrete must be of rock-like order (which is 10^{-10} m/s) for the granite high-level waste disposal facility. This parameter satisfies this

requirement and is stable throughout the test period (Figure 19), indicating a high stability of the material [65].

It should be noted that the hydraulic permeability of the shotcrete is slightly higher than that of the basic concrete due to the increase in porosity caused in the material by the process of shotcreting [65].

In the paper [66], the experiment on interaction of samples (cylinders 12 mm in diameter and 10 mm in height) of concretes on the basis of cement grade CEM III A/S 42.5 N ($W/C = 0.6$), with model solution of underground water of the Yeniseysky site potential site for the deep geological isolation of radioactive waste in crystalline rock (i.e., Yeniseysky site at Nizhne-Kansky massif). The composition of the model solution was selected on the basis of information in [67,68] by dissolving in distilled water the following mineral salts: $\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$, NaHCO_3 , $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$, KCl . pH of the medium was corrected to 7 by adding small amounts of caustic soda solution. The composition of the model solution produced is shown in Table 6.

Table 6. Composition of the model solution of the underground water of the «Yeniseysky» site before and after the experiment (10^{-4} , mol/L) [66].

Aqueous Phase	Na^+	Al^{3+}	Si^{4+}	K^+	Mg^{2+}	Ca^{2+}	Cl^-	SO_4^{2-}	HCO_3^-	pH
Model solution before experiment	8.87	-	-	1.15	4.95	12.2	25.6	4.95	8.87	6.1
After experiment	67.8	0.01938	0.3877	119.82	0.000411	0.8034	-	-	-	12.1

The concrete and groundwater interaction experiment was conducted in polytetrafluoroethylene (PTFE) cups with a lid in the dryer at a temperature of 90°C for 40 days at a ratio of solid to water of 1:6 [66]. The composition of the modified water phase as a result of the experiment is also given in Table 6.

The study found that the interaction of concrete with the model solution led to the slight dissolution of Al, Si, the transition of a significant amount of Na and K to the aqueous phase, while in the solid phase Cl^- , SO_4^{2-} , HCO_3^- and, to a large extent, Mg and Ca. pH increased almost twofold, from 6.1 to 12.1 [66], which, according to Table 6 and a series of studies [69–71], is due to leaching of Na^+ and K^+ , resulting in increased hydroxyl anion concentrations displacing the equilibrium of the dissolution reaction of Portlandite ($\text{Ca}(\text{OH})_2$) in the opposite direction. The increase in Mg content in concrete at sufficient concentrations in the aqueous phase may be due to a substitution reaction of Ca, or in silicate minerals or the formation of magnesium sulfate, as described in [72].

Phase change of the concrete was studied using the X-Ray Diffraction method (XRD). After the interaction of groundwater and concrete, there was a decrease in the content of the main phases, namely water, portlandite and gypsum, while the amount of $\alpha\text{-C}_2\text{SH}$, alite and belite increased. It was also noted that the mineral phases awfilite and nordstandite [66] were not previously presented in the original samples.

Porosity change has been studied by several methods—scanning electron microscopy (SEM), polymolecular nitrogen adsorption, computer microtomography ($\mu\text{-CT}$). The results of $\mu\text{-CT}$ showed an increase in the relative volume of the macropore from 29.23% to 43.80% (Figure 20c—pores are shown yellow on the concrete model) [66]. The increase in the pores on the exterior surfaces of the specimen is due to the fact that they were phase boundaries when the concrete came into contact with the model solution, which naturally led to an increase in the number of macropores in that region.

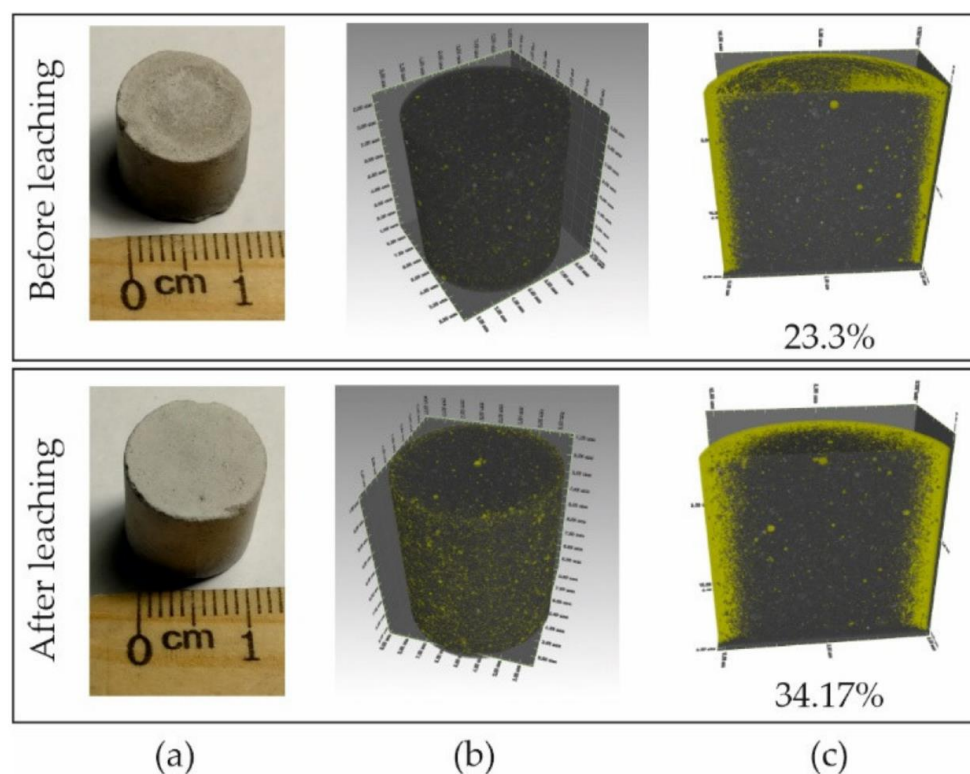


Figure 20. Illustration of the change in porosity of concrete sample as a result of interaction with the model solution. (a)—photo of samples before and after the experiment; (b)—3D model of samples, obtained with the help of μ -CT; (c)—relative porosity, detected using μ -CT (% of volume) [66].

The porosity analysis by SEM and the use of the STIMAN software found that the pore shape on the outer surfaces is more spherical than inside the specimen based on the dependence of the obtained probability density values ($N_i/(N \cdot 1)$) from the coefficient of pore distribution (K_f) (for spherical pores $K_f = 1$). The exposure to groundwater caused a change in the pore morphology outside the specimen to an even more spherical. It was also found that the maximum equivalent pore diameter on the outer sides increased from 162 μm to 259 μm during the experiment. However, the change in porosity is more complex: there was an increase in the contribution of pores $>10 \mu\text{m}$ when the contribution of diameters of 1–10 μm to the total porosity decreased [66].

Thus, the main changes in the porosity of the samples are due to the dissolution of a part of the constituents of the initial samples of concrete phases during the experiment [66].

3.3. Effects of HLW Irradiation on Concrete Properties

In the work [20–22,73], concrete samples were tested at absorbed doses of γ -radiation from 10^6 Gy, used in various sources to determine changes in individual properties of cement compounds under the action of radiation loads, up to the value of 10^8 Gy, regulated by GOST R 50926-96 [74] for cured high-level waste. The results showed that the cement compounds at the dose range had no visual marks of mechanical damage in the form of chipping and cracking, maintained a high compressive strength, which was 4–8 times higher than the regulated value for solidified high-level waste (5 MPa). When comparing the phase composition of samples before and after irradiation in the studied ranges of the values of absorbed doses by the method of XRD, TEM and SEM, it was established that the change does not occur the formation of defects in the structure and changes in the phase composition. Furthermore, by measuring the plasma spectrum of gases emitted by irradiation of concrete samples in a gamma chamber, radiolytic gas formation under high dose loads was estimated. The spectrum was formed as a result of optical breakdown when focusing laser radiation in the volume of the sealed vial. Experimental data show

that the maximum hydrogen release does not exceed 10^{-3} mol/(g of the sample) at 10^8 Gy. Thus, it is shown that engineered barriers based on ordinary Portland cement are able to maintain their physical-mechanical properties, phase composition and structure under the exposure of radiation loads equivalent in magnitude to radiation from high-level waste during disposal.

3.4. Influence of Biogenic Processes on Cement Material Properties

According to the research conducted on the territory of the Russian Federation in near-surface repository of FSUE «Radon» on biodegradation of cemented nitrate-containing radioactive waste, bacteria have been identified from the genus *Pseudomas*, resulting in the formation of carbon dioxide, acetate, propionate- and butyrate-ions, which led to carbonization and transfer of hydrate minerals of the matrix into soluble forms, which entails degradation of physical-mechanical properties of the compound by leaching [75–78]. These phenomena caused microfractures and pores in cemented radioactive waste. In [79], the structure of cement compounds made with and without biocide additives was analyzed and compared after 8.5 years of storage.

Cement matrices without the use of biocidal additives are distinguished by the presence of isolated phases of crystalline nitrate and calcium hydrocarbonate having a high solubility in water. This leads to reduced reliability of immobilization of radionuclides and mechanical strength of cement matrix. This cement compound largely contains biogenic acid breakdown products of calcium hydrosilicates (<10–12%), which causes microstructure breakdown. At the same time, cement matrices with biocidal additives have a monolithic and denser structure [79,80].

The intended materials for use as engineered barrier in the at Yeniseysky site will be in contact with the host rock in the presence of an indigenous and externally impacted microbial communities. There is also a high probability of intensification of microbiological processes due to heat dissipation from packages with radioactive waste and the release of gases that are products of radiolysis and involved in microbial respiration processes (H_2 , CH_4 , CO , etc.), the output of biogenic elements from matrices: phosphate from glass matrices, iron from steel containers, potassium, sodium, calcium, as well as organic matter from clay materials containing waste [75].

Thus, when designing a geological disposal of radioactive waste, it is necessary to take into account the impact on the concrete elements of engineered barrier systems of a number of factors listed in Table 7.

Table 7. Transformation of barriers based on cement materials under external factors of geological disposal facility.

Cement/from Cement Exposure	Changes in Concrete/from Concrete Side Barrier
Temperatures characteristic of RW Class 1 and 2 packages	Reduced concrete compressive strength by 25%
Groundwater flow through pores of concrete	Dissolving Portlandite, cement C-S-H gel, increasing porosity and reducing strength. The hydraulic permeability of concrete with silica fume admixture is at the level of permeability of granite rocks, which meets the requirement for material for deep repositories.
Influence of HLF irradiation on concrete properties	No significant changes of compressive strength, no visible damage of concrete samples and sufficient changes in structure under absorbed dose of high-level waste up to 10^8 Gy. The maximum radiolytic hydrogen release does not exceed 10^{-3} mol/(g of the sample) at 10^8 Gy.
Biogenic processes	Carbonization and neutralization of basic cement stone minerals, resulting in their leaching and reduced strength (for nitrate-containing cement matrices with radioactive waste). It is assumed that the cement materials in the geological disposal facility at Yeniseysky site will contact the indigenous and extraterrestrial microbiota, as well as intensification of microbial processes under the influence of heat emission and radiolytic gases.

4. Influence of Concrete on Individual Component of Engineered Barrier System under Exposure of Conditions of Geological Disposal Facility

During the operation of the geological disposal facility, concrete (due to the creation of a highly alkaline environment) can have a negative effect on other engineered barriers: bentonite, steel containers, glass matrices of class 1 radioactive waste. Therefore, it considers necessary to this impact in order to predict their behavior in the long term in order to possibly prevent a negative scenario.

4.1. Changes of Bentonite Clay Buffer Properties under the Exposure of Model High-Alkali Conditions of Cement/Concrete

Many concepts of deep repositories of radioactive waste in suggest the use of a buffer based on compact bentonite clays [81–87] due to its unique characteristics such as: temperature resistance, essential physico-mechanical and preferred physico-chemical properties [88–96], exceptional swelling ability [97–99], resistance to aggressive media, (simulating bentonite structure destruction) [100,101], high specific surface area, cation exchange properties, their sorption characteristics in relation to radionuclides [81,102–106].

Since the many concepts described in Section 2 intend to place bentonite in the DGR/GDF in contact with concrete structures, it will be mainly influenced by the transfer of dissolved phases of concrete by groundwater to the compacted clayey buffer material [6,13].

Short-term interaction (90 days) of concrete leaching products with model solution of groundwater of granite massif (Table 6) and bentonite of 10th Khutor deposit (Republic of Khakassia, Russian Federation) are in operation [66]. At 90 °C, specific surface area increased by a factor of 1.5, which should have a positive impact on its sorption properties and reduce the number of macropores. In a strong alkaline medium, the dissolution of quartz and feldspars occurred, which influenced the change of phase composition towards an increase in the content of montmorillonite—the main component of bentonite clays [66]. The X-ray fluorescent analysis (XRF) showed an increase in calcium, sodium and potassium content, which may be due to substitution of divalent magnesium with sodium and potassium in the interlayer space [66] that at low compaction densities may lead to a slight improvement in bentonite swelling capacity [81,87,105]. Geochemical modelling in a 450-day period of [66] based on the experimental data obtained showed high bentonite stability: solubility constants in the kinetic equation were significantly lower than those described in the literature [106] including work on mathematical models of bentonite dissolution [107,108].

In [109–111] the influence of alkaline activation (short-term effect of alkali at a temperature of order 100 °C) of bentonite clays has been studied and no negative changes in properties such as cation exchange capacity (CEC), specific surface area (SSA). The results in [109–111] illustrate the increase in the specific surface area, in [111] cation exchange capacity there was a slight change, and in [109,110] there was an increase in the interphase distribution coefficient relative to the radionuclide of ^{137}Cs and the same values for ^{90}Sr .

In [112–115] the influence of strong alkaline solutions on swelling pressure, hydraulic permeability (filtration coefficient) and cation exchange capacity of bentonite clays was investigated. As a result of exposure to bentonite of MX-80 of NaOH solutions with concentrations of 0.1, 0.3 and 1.0 mol/L within 45 days [112] significant reduction of swelling pressure at the concentration of alkali > 0.3 M, corresponding to pH 13.3, which is typical for fresh cement mortar [112]. At a pH close to that of $\text{Ca}(\text{OH})_2$ (12.4, when 0.1M NaOH had a pH of 12.9), characteristic of the hardened concrete, no significant change in swelling pressure was found [112]. In [113–115], the effect of strong alkaline solutions on Chinese bentonite Gaomiaozi (GMZ) has resulted in a significant reduction of cation exchange capacity, swelling pressure and increased filtration coefficients, which are exacerbated by higher temperatures.

4.2. Interaction of Engineered Barrier Materials in the System «Concrete-Steel» under the Exposure of Groundwater

4.2.1. Influence of Low Oxygen Content and High Pore Cement Solution pH on the Integrity of Steel Packages after Closure of Geological Disposal Facility

One of the corrosion effects of steel containers for radioactive waste on the chemical composition of underground water is the absorption of free oxygen. Oxygen absorption will lead to a reducing medium with low oxidative potential. Corrosion products shall henceforth maintain reducing conditions [116]. An assessment of the oxidation-reduction potential of the Nirex-type disposal facility calculated that the oxidative potential would be held in the range of -400 to -600 mV at pH 12.5 and 25 °C. It was found that the oxidizing-reducing potential is maintained by the oxidizing-reducing chemistry of iron and uranium. The study also provided a simple estimate of the duration of the recovery. Based on the quantities of iron and uranium expected in the Nirex disposal facility and typical groundwater flow, the work suggested that the remediation conditions could be maintained in the storage environment for at least a million years [116].

In the long term, according to the study [116], the corrosion rate of a steel container in the Belgian deep geological repository high-level waste and spent-nuclear fuel in an alkaline environment provided by a cement buffer (pH~13.6) should be <0.1 $\mu\text{m}/\text{year}$ with subsequent decrease. When using pressure sensors, the corrosion rate under anaerobic conditions was measured at 0.03 $\mu\text{m}/\text{year}$, which should mean the loss of 3 mm layer of container material over 100,000 years, confirming the high corrosion resistance of steel in a high alkaline environment under anaerobic conditions [116].

4.2.2. Pitting Corrosion and Stress Corrosion of Steel Super Container Material under the Action of Cement Pore Solution with pH~13.6

In [117], it was found that at room temperature (and low concentrations of chloride-anion), the critical potential of pitting corrosion lies in the O_2 release area, but is rapidly shifted to negative values when temperature increases. The presence of sulfide anions (up to 500 mg/L) does not appear to increase the risk of stress steel cracking in a solution simulating the composition of pore concrete solution under oxygen-free conditions.

4.2.3. Formation of Iron Sulphides on the Surface of a Steel Container in Contact with Groundwater with Low-Alkali Cement and Related Corrosion Mechanisms

In work [118], steel samples were immersed in a special cement mortar with low pH, the composition of which was developed for geological disposal facility Cigéo (France), and left for 6 months in oxygen-free conditions at 80 °C. The layers of corrosion products were analyzed using μ -combination spectroscopy as well as XRD and SEM. It turned out that they were mostly composed of iron sulfides, with the addition of magnetite as a secondary phase mixed with cement mortar components. The average corrosion rate was estimated by mass loss measurement and ranged from 3 to 6 $\mu\text{m}/\text{year}$. Corrosion profiles showed local fractures up to 10 μm deep. The heterogeneity of the corrosion layer consisting mainly of conductive compounds (FeS , Fe_3S_4 and Fe_3O_4) is assumed to contribute to the retention of corrosion cells, which may lead to locally aggravated degradation of the metal.

4.3. Influence of Cement Materials on the Solubility Rate of Vitrified RW Class 1

The impact of cement materials on the dissolution rate of the glass matrix for radioactive waste is the least studied, and the available data mainly refer to borosilicate glass, significantly different from sodium-aluminophosphate glass, planned for in first order for disposal in Russia. Laboratory experiments with model vitrifying and solutions show a high dissolution rate, but taking into account the real composition of GW (groundwater) and contacting engineered barriers can reduce conservatism of preliminary estimates. In conditions close to neutral (pH 7), the initial stage of glass-water contact dissolution is characterized by diffusion of water molecules into the glass structure and ion exchange (stage I), and then by hydrolysis of the glass structure, leading to an output into the element solution glass-formers (stage II, «mode of initial dissolution»). These processes lead to the

formation of a structure on the surface of glass (gel) depleted with soluble components. With further dissolution, there is a significant decrease in the speed of the output of glass components into the solution (stage III, «dilution mode»), after which the rate of dissolution becomes relatively constant, by 3–4 orders of magnitude below the characteristic value for stage II. This phase is called stage IV or «residual dissolution mode». In some cases, after stage IV, the dissolution rate again increases (stage V). These mechanisms are well known, but in some cases, they emit less «stages» of dissolution. The dissolution rate of glass in phase II depends on the pH of the solution: it increases significantly in both acidic and alkaline environments [61].

4.3.1. Dissolution of Borosilicate Glass, a Material of Vitrified HLW in Air Conditioning Practice, under the Action of Groundwater Contacting Cement, as a Material of Engineered Safety Barrier

The dissolution rate of glass in phase II depends on the pH of the solution: it increases significantly in both acidic and alkaline environments [61].

Figure 21 shows the dependence of the dissolution rate of borosilicate glass on the temperature and pH of its constituents (aluminium, boron, calcium, silicon and sodium). The coloured areas correspond to the solubility velocity ranges determined by one of the fibers [61,119].

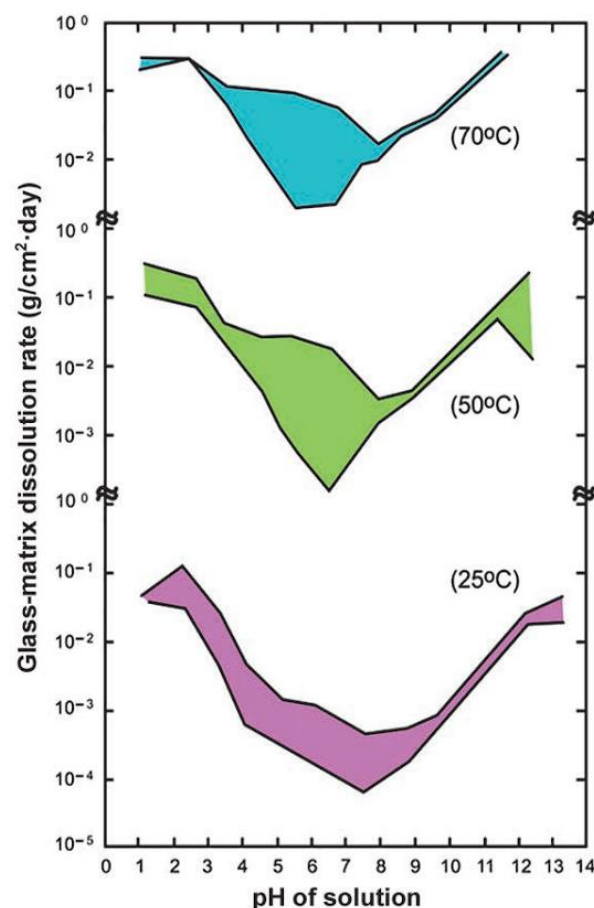


Figure 21. Graphic illustration of the relationship between the initial dissolution rate of borosilicate glass and the temperature of each component of matrix [61,119].

Model experiments [120–124] show that the initial solubility rate of borosilicate panes at temperatures around 100 °C in neutral solutions is in the order of 10^{-3} – 10^{-2} g/cm² days and increases as pH increases by a power law (roughly proportional to $10^{0.4\text{pH}}$) [61].

4.3.2. Dissolution of Sodium-Aluminophosphate Glass, Which Is the Material of Vitrified HLW under the Exposure of Groundwater Contacting Cement, as a Material of Engineered Safety Barrier

The rate of dissolution of Russian sodium phosphate glass at room temperatures by the most soluble components (type Na) does not exceed 10^{-4} – 10^{-5} g/cm²·days, however, when water vapour is heated in the atmosphere, it is quickly opened with a leaching rate of up to three orders of magnitude and the transition of many impurity elements such as radionuclides to colloid form. As a result, sodium-aluminophosphate glass can be inferior to borosilicate glass under typical conditions in geological disposal facility [61].

Experiments with solutions whose salt composition is close to real groundwater have shown a reduction in the solubility rate of glass by about two orders of magnitude compared to solutions with low salt content [61].

For glass that has been in the environment for thousands of years, examination of the profiles of the altered surface has shown that the real dissolve rate of the glass is always lower than that of the existing models [61].

Preliminary estimates show that the increase in the dilution rate of glass is not a determining factor, and its increase even by three orders of magnitude is not critical for the safety of geological disposal facility under the normal operation of other barriers. However, this issue needs to be further elaborated [61].

It can be concluded that the main changes in the properties of the elements of engineered barrier system that are in direct contact with concrete are listed in Table 8 and mainly determined due to the impact of a highly alkaline concrete pore solution.

Table 8. Transformation of engineered barrier system components under alkaline concrete pore solution exposure.

Cement/from Cement Exposure	Changes in Concrete/from Concrete Side Barrier
Effect of alkaline concrete pore solution on compacted bentonite buffer	No significant changes in cation exchange capacity, swelling pressure and hydraulic permeability of compacted bentonite at pH 12.4 which is common for hardened concrete. At values of pH typical for fresh cement mortar (13.3), compacted bentonite buffer shows a decrease in cation exchange capacity, swelling pressure, and an increase in hydraulic permeability.
Influence on steel container from alkaline pore solution of concrete	Under anaerobic conditions, geological disposal facility ensures the integrity of the steel packaging structure (corrosion rate 0.03 µm/year)
Influence on steel container from alkaline pore solution of concrete at temperature 80 °C	Increased corrosion rate of 3–6 µm/year in anaerobic geological disposal facility conditions with occurrence of local corrosion hotspots with thickness of 10 µm.
Effect of alkaline pore solution of concrete on borosilicate and aluminophosphate glass matrices	With increased pH, the dissolution rate of glass matrices increases.

5. Mutual Transformations of Concrete-Bentonite under Exposure of Granitic Host Rock Groundwater

The study of geochemical processes occurring in the engineered barrier system is an important part of the geological disposal facility design and following modelling of long-term evolution of engineered barriers. The results of this study mainly contain mineralogical and chemical changes of barrier materials and determined by transport of dissolved mineral components of the host rock and engineered barriers in groundwater. Full-scale and short-term experimental results and their comparison are necessary to analyze how selected engineered barriers act on each other at their interface in geological disposal facility conditions. Since concrete and bentonite are common in most concept designs of geological disposal facilities and their interactions are key factors determining secure isolation of radioactive waste.

5.1. Mutual Influence of Engineered Barrier Materials in the «Concrete-Bentonite» System under the Exposure of Groundwater in the Long Term Experiments

A unique and oldest long-term in situ experiment was performed in Grimsel Test Site (GTS) underground research laboratory. Since this experiment involved the detailed study of compacted bentonite buffer and concrete plug interaction, it will be particularly described below.

The Grimsel Test Site, located in the Swiss Alps, was founded in 1984 as a facility for underground research projects related to the geological disposal facility of radioactive waste in granite host rock. The FEBEX (Full-scale Engineered Barrier Experiment) experiment is a full-scale study of engineered barriers under natural conditions [6].

The horizontal tunnel, 70 m long and 2.28 m in diameter, was drilled in granite rocks crossed by a lamprophyre layer, in which 2 electric heaters simulating heat release from high-level waste canisters (4.5 m long and 0.9 m diameter) were placed 1 m apart and surrounded by the FEBEX bentonite barrier, 0.7 m thick (Figure 22). However, after five years of uninterrupted use of the heaters, one of them, located closest to the concrete plug of the gallery, was disconnected, removed along with the bentonite and plug surrounding it, replaced with a steel dummy 1 m long. It was re-surrounded by bentonite blocks and a new concrete plug was made. It consists of two layers and was manufactured using the technology of spraying cement mortar directly into the gallery (shotcreting). The first layer of the 1 m thick plug is in direct contact with bentonite and steel chump, and the second layer is 2 m long. Between these layers, a 3–4 cm thick moisture-resistant polymer layer is constructed. After replacing one of the heaters with a steel dummy, the experiment was resumed and continued for 13 years [6].

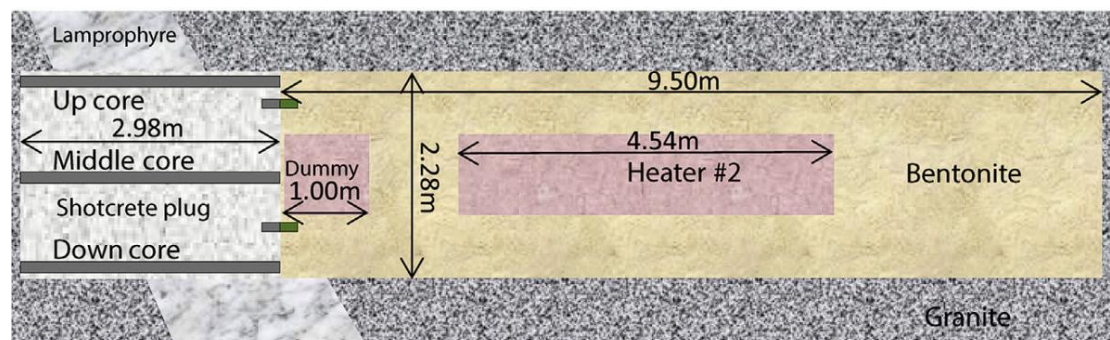


Figure 22. Longitudinal section of the FEBEX setup and location of drilled concrete cores [6].

5.1.1. Concrete-Bentonite Mineral Surfaces Passivation and Formation of Crystalline Phases in the Concrete Pores by Means of Groundwater Exposure

Ca^{2+} content in pore solution of concrete is controlled mainly by dissolution of Portlandite, but closer to the concrete/bentonite interface it may be due to deposition of calcite and gypsum. The presence of gypsum was found in a bentonite layer <1 mm thick, located on the contact surface with concrete, in which the carbonate content was lower than gypsum [13]. The redistribution of $\delta^{13}\text{C}$ isotopes in bentonite carbonates has also been determined, showing the redistribution of the calcite from the bentonite volume to the concrete surface at the point of contact [6].

Mg is precipitated on the concrete surface as stevensite-like phases, as their formation requires high pH (~10) and high Na content, which diffuses from concrete [13].

The mapping of secondary electrons (by means of SEM-EDX) on the scanning electron microscope of the concrete plug sample from the upper core yielded clear effects of ettringite formation from the detected area of its concentration on the concrete surface in contact with bentonite. The presence of a set of ettringite crystals was also detected by SEM [6,13]. The formation of ettringite is mainly due to sulphate-anion diffusion. However, in the manufacturing of a concrete plug, shotcrete additives were used to accelerate its grasping at a maximum of 6% mass which subsequently increased the aluminum content [6].

5.1.2. Transport of Ions in the Concrete-Bentonite Interface (Transfer of Mg^{2+} Ions, SO_4^{2-} from Bentonite to Concrete and Ca^{2+} , Na^+ , K^+ from Concrete to Bentonite) under the Action of Groundwater (Transfer of Cl^- and HCO_3^- Ions Dissolved in Groundwater)

Granite groundwater Grimsel Test Site has a very low ionic force, its pH is approximately 9, and it is the main factor for diffusion of soluble components of concrete and subsequent reduction of pH solution in its pores. pH in the central core (Figure 22) is measured at different distances from the steel dummy and its value in any part is always above 12.65, despite its slightly reduced value at a distance of 1 cm from the dummy. At a distance of 2 cm pH was found to be 12.8. However, the upper and lower cores (Figure 22) have a lower pH, up to a distance of 5 cm from the bentonite barrier and its value measured in the lower region are slightly lower than the upper one. After measurements, pH was determined to be less than 12.2 (even 12 in the lower case) at a distance of 1 cm from the bentonite barrier. The pH decrease is a consequence of several processes, the main of which is the leaching of alkali metal ions (Na and K) with the accompanying dissolution of portlandite ($\text{Ca}(\text{OH})_2$). It is widely known that at pH below 12.5, the presence of portlandite is limited or absent from the concrete structure [6].

Moreover, the main water-soluble ions in the concrete near the interface (Na, K, Ca, SO_4^{2-} , Cl) have different concentrations in the central core relative to the upper and lower. The measurements of their average content in the central core were taken at a distance of 1 cm and 5 cm from the steel dummy and have almost identical values. Results from the upper and lower cores suggest that water sources interacting with concrete have a joint influence on its composition. First of all, a decrease in the content of alkali metals (and Na and K) in the upper and lower cores of the concrete plug, compared to its central part, was determined. This was determined to have been caused by granite groundwater. However, the calcium content is higher in the upper and lower cores compared to the central, which is mainly due to the transfer of Ca^{2+} ions from bentonite at the time it is saturated with granite groundwater, when there is a redistribution of ions between the cation exchange links in the bentonite structure and groundwater. Therefore, the FEBEX bentonite water is enriched with Ca^{2+} ions. Thus, Ca diffuses from the bentonite pore solution to the concrete pore solution and this phenomenon plays an important role in explaining the results.

The interaction of bentonite pore water with concrete is further confirmed by analyses of dissolved sulfates and chlorides in the upper and lower cores of the concrete plug, at the interface with bentonite. Both anions are represented in lower and upper samples at a distance close to the interface of bentonite with concrete, but their content is very low in the central core at the same distances (due to lack of direct contact with bentonite pore or granite groundwater). While Cl penetrates faster, even up to 5 cm from bentonite, high sulphate content is only found on the first centimeter of concrete from the bentonite surface [6].

Granite groundwater in Grimsel Test Site has a fairly low salt content (total dissolved < 10 mg/L), caused mainly by Na-Ca- HCO_3 (pH~9). The deposition of calcite on the contact surface of bentonite concrete confirms the transfer of HCO_3^- ions from groundwater through the concrete cap [6].

5.1.3. Alteration of the Mechanical Properties of Concrete at Concrete/Bentonite Interface Due to Groundwater Characteristic of the Host Rock

In the work [6], the initial compressive strength value of concrete in the plug was obtained was 32 MPa. After 13 years of operation, the compressive strength of the upper concrete core became 30.5 ± 3.9 MPa, 30.2 ± 3.8 MPa of the central and 29.9 ± 2.2 MPa of the lower. Thus, a slight decrease in the strength of concrete can be a consequence of its aging. Consequently, changes in the microstructure of concrete are not yet limited by its mechanical strength, although the mechanical strength of concrete at the point of contact with bentonite cannot be measured [6].

5.2. Mutual Influence of Engineered Barrier Materials in the «Concrete-Bentonite» System under the Exposure of Synthetic Groundwater in the S Term Experiments

On the basis of the obtained data sets at Grimsel Test Site, in articles [11,12] the method of laboratory experiment simulating conditions in Grimsel Test Site was developed. To achieve this, a laboratory unit was developed, the device of which is shown in Figure 23.

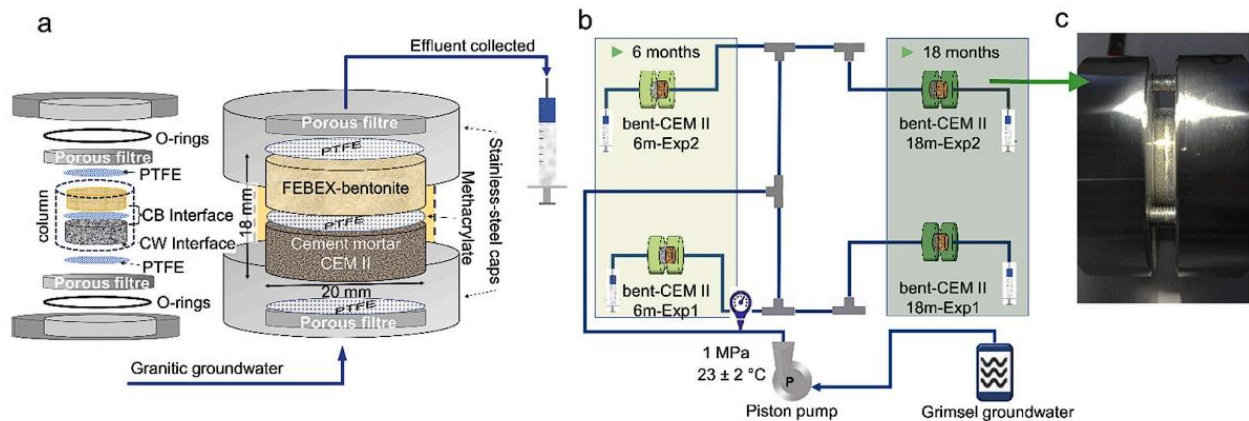


Figure 23. Arrangement of transport cells: (a) cell structure; (b) experimental setup; (c) photo of concrete/bentonite interface in of one cell [13].

Concrete and bentonite FEBEX (compacted to dry density 1.5 g/cm^3) are placed in the housing, which is a methacrylate hollow cylinder closed by two steel flanges, which give the cell a stiffness of the structure, and separated by a polytetrafluoroethylene (PTFE) membrane with a pore thickness of $0.45 \mu\text{m}$. The cell has an internal diameter of 20 mm and a height of 18 mm (9 mm per cement layer and bentonite layer). The cell was designed in such a way that it could be easily disassembled for further analysis of the solid phase changes and the concrete and bentonite could be easily removed from the cell. The design of the cell prevents deformation of concrete and bentonite inside the housing, drying of both materials and isolates from air ingress. The compact outer casing made of stainless steel (together with joints and filters) is able to withstand the high pressure caused by the swelling of bentonite. According to the conditions in the Grimsel Test Site, granite underground water is injected at a constant pressure of 1 MPa through tubes of polyetheretherketone (PEEK) using an automatic piston pump. The elute is collected at the outlet of bentonite and then its volume is measured [11].

Two transport cells are launched for 6 months, and the other two are 18-inch, in virtually isolated laboratory conditions ($23 \pm 2 \text{ }^\circ\text{C}$). The conditions are similar to the expected temperature of the concrete-bentonite system according to the data obtained by the Grimsel Test Site during the FEBEX [12] experiment.

Underground granite water of Grimsel Test Site has the electric conductivity value of $104 \mu\text{Sm/cm}$, $\text{pH} = 8.1$ and the following composition (mg/L): Cl^- (3.5), SO_4^{2-} (9.6), HCO_3^- (42.7), Na^+ (11.5), K^+ (<0.1), Ca^{2+} (12.0) and Mg^{2+} (<0.1) [11].

Papers [11,12] present an assessment of the geochemical reactions occurring at the concrete-bentonite interface at the early stages of their interaction in order to compile the most complete picture of the processes occurring in the course of large-scale and long-term experiments. The results obtained in the course of a full-scale experiment at the Grimsel test site and a short-term experiment in the works [11,12] are consistent and can be summarized as follows:

- Decalcification of C-S-H in a medium in which portlandite dissolves and progressive hydration of anhydrous phases occurs.
- Development of a film of calcite on the interface of concrete with bentonite and dispersed precipitation of calcite in localized zones of bentonites.

- The beginning of the development of Mg-containing components on the interface of bentonite with concrete, associated with the formation of Mg-clay 2:1 sheet silicates as the main neogenic phases expected in the long term.
- Formation of secondary ettringite in concrete at the interface with bentonite.

Thus, the short-term experiment in [11,12] is promising when used to study the interaction of barrier materials, coupled with the use of mathematical modeling of the ongoing processes for their extrapolation to long-term time intervals.

6. Conclusions

This article provides a general review of the concepts of radioactive waste geological disposal facilities/deep geological repositories in terms of the function performed by engineered barriers based on cement, laboratory experiments involving cement materials individually, and when they come into contact with other elements of the engineered barrier systems directly and as products of interaction with model solutions of groundwater. In view of the emerging uncertainty in the concept of the organization of GDF in the Russian Federation, it is necessary to focus on the results of existing concepts, on the actual conditions in prospective geological formations and on the existing requirements for the disposal of radioactive waste. Furthermore, for a number of selected promising concretes, it is necessary to conduct a full cycle of both laboratory and long-term research under engineered barrier conditions with individual analysis of both physical–chemical and operational properties of cement materials, and the mutual influence of all engineered barrier materials. Thus, at the stage of laboratory experiments, it is necessary to determine the filtration coefficients (hydraulic permeability) of concrete and cores of containing rock from the Yeniseysky site in comparison; the selected concrete should have a value lower than for the rock. When selecting a potential concrete as the engineered barrier, the influence of its dissolved components in aqueous environments on the bentonite buffer properties (if the current concept provides for direct contact or indirect contact of the concrete–bentonite phases) and corrosion of stainless steel, often used as material for radioactive waste containers (in the case of cement buffers).

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Abbreviations

μ-CT	Computer microtomography
C/B	Concrete-Bentonite interface
C/W	Concrete-Water interface
CEC	Cation Exchange Capacity
DGR	Deep Geologic Repository
EBS	Engineered Barrier System
EDX	Energy-Dispersive X-Ray Spectroscopy
ESB	Engineered Safety Barrier
FEBEX	Full-Scale Engineered Barrier Experiment
GTS	Grimsel Test Site
GW	Groundwater
HLW	High-Level Waste

IUPAC	International Union of Pure and Applied Chemistry
LL	Long-Lived
LLW	Low-Level Waste
LRW	Liquid Radioactive Waste
NPP	Nuclear Power Plant
NRVB	Nirex Reference Vault Backfill
NSDF	Near-Surface Disposal Facility
NSR	Near-Surface Repository
OPC	Ordinary Portland Cement
PEEK	Polyetheretherketone
pH	Hydrogen index
RW	Radioactive Waste
SEM	Scanning Electron Microscopy
SL	Short-Lived
SNF	Spent Nuclear Fuel
SNFA	Spent Nuclear Fuel Assembly
SSA	Specific Surface Area
STIMAN	Structural Image Analysis
TEM	Transmission Electron Microscopy
TRU	Transuranic
W/C	Water-Cement ratio

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