

HLW DISPOSAL OF IN VERTICAL DEPOSITION HOLES WITH CEMENT BACKFILL – PRO AND CONTRA WITH REGARD TO LONG-TERM SAFETY OF A GEOLOGICAL DISPOSAL FACILITY

Bogatov S. A.

Nuclear Safety Institute of RAS, Moscow, Russian Federation

Article received 18 January 2018

The paper overviews the option of using cement-based materials as an engineered barrier for HLW disposal. This option has a number of advantages. For instance, it provides an effective chemical barrier; decreased corrosion rate of iron containers; is characterized with high gas permeability; ensures the use of the same type of material both for HLW and ILW disposal and easy handling. The main drawbacks associated with cement material use is the increased corrosion rate for the glass matrix containing vitrified HLW, as well as relatively high permeability for weakly absorbed radionuclides, such as ^{129}I and ^{36}Cl . However, advantages of this barrier material are supposed to override all the negative aspects as regards the RW disposal project to be implemented in Nizhnekansk rock massive envisaging compact HLW disposal in 75 m deep disposition holes with bentonite barrier and insignificant ^{129}I and ^{36}Cl content.

Keywords: radioactive waste, geological disposal, engineered barrier, cement-based materials.

Introduction

Nizhnekansk (NKM) disposal concept suggests combined emplacement of HLW and ILW at two levels of a repository located at a depth of 500 m and having a footprint of 300×700 m. HLW are placed into vertical 75 m deep disposition holes, whereas ILW are disposed of in horizontal excavations. Bentonite blocks were proposed as the main barrier material for HLW disposal, whereas a cement-based mix was proposed as a backfill material for ILW disposal [1].

This paper evaluates the possibility of using cement-based materials for engineered barriers construction in HLW disposal facility. This material is being considered as an alternative engineered material for the NKM disposal project. Currently, cement material use is actively discussed under various disposal concepts developed abroad, including the “supercontainer” concept under the Belgian case study [2] and multi-purpose containers for HLW and SNF disposal [3].

1 Advantages of using cement in the engineered barrier system of deep geological disposal facilities

1.1 Physical containment¹

At the post-closure stage, cement backfilling will ensure alkaline conditions for the ground water flowing into the repository (pH > 10) and saturate it with calcium ions. This process will decrease the dissolution rate for materials contained in ILW [5]. At the same time, corrosion of RW packages and containers will shortly result in the uptake of oxygen penetrated to the disposal facility during its construction and waste package emplacement. Low oxygen content and elevated pH will contribute to sustaining the integrity of waste packages at the

¹ This definition is widely used worldwide. The definition presented in [4] means preventing or controlling the release and the dispersion of radioactive substances in the environment (which is similar to Russian wording “localization”)

Disposal of RW

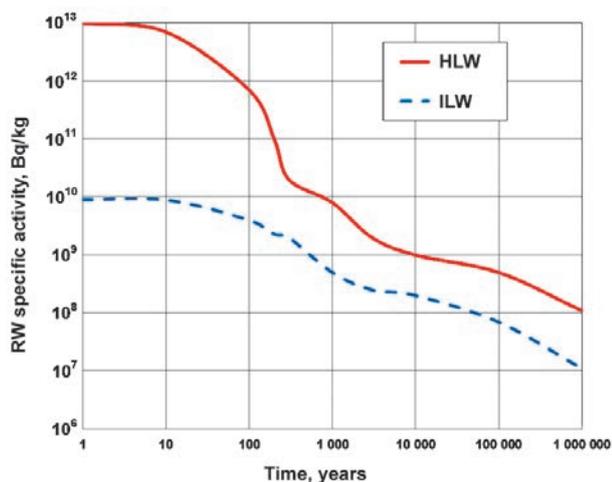


Figure 1. Decrease in the total specific activity level with time for HLW and ILW starting from the waste emplacement time (conservative estimates)

post-closure stage due to the drastically decreased corrosion of iron containers.

The degree to which RW packages preserve their isolation capacities will change depending on chemical properties of relevant radionuclides, RW forms and container backfilling. Estimates show that packages may remain intact from several hundred years for most mobile nuclides escaping fragmented HLW forms via ventilation holes to up to tens of thousand of years for absorbed radionuclides in cemented RW [6].

It seems reasonable to suggest that a period of 1,000 years² can be seen as an average timeframe for which package sealing capacity can be ensured. During this time period HLW activity level will decrease to some 0.1% from the initial value and up to 6% for ILW (figure 1).

1.2 Chemical containment

Physical containment provided by the cement backfill ensures a sufficient time period during which the major part of radionuclides shall remain confined within the package. However, this is not sufficient to confine a small portion of radionuclides having a half-life of over several thousand years [7]. Cement barrier shall primarily act ensuring the containment for a long time period – some 1,000,000 years given all possible changes in ground water flows and geochemical conditions inside the disposal facility. This requirement can be fulfilled through the use of a cement backfill ensuring the

required chemical environment, decreasing solubility and increasing radionuclide sorption.

Ground water high pH level is consistent with the prevailing content of hydroxyl ions (OH)⁻ which is higher than of other ions, for example, nitrates and di-carbonates resulting from RW leaching or introduced with ground water flows. Radionuclides present in the form of soluble chemical compounds react with highly alkaline water producing oxides or hydroxides with low solubility levels in relation to many radionuclides (figure 2, [8]). Precipitation of radionuclides from the solution decreases their transfer with ground water flow.

Cement materials have a large active surface area by means of which radionuclide species can be produced. For example, if granular type material NRVB³ enters aqueous plutonium solution with pH = 10 (plutonium has very low solubility under such conditions), plutonium sorption provides a 10,000-fold reduction in its concentration in the solution [9]. This effect also ensures radionuclide retardation due to which most part of them will decay in-situ.

The goal of maintaining high pH levels in the near-field for a long time period is seen as the primary one for cement buffer use in the disposal facility. Relatively high permeability and porosity provides for a homogeneous chemical environment preventing the formation of particular areas where requirements for sorption conditions and decreased solubility may not be met.

Chemical containment is mostly ensured for actinoids (thorium, uranium, plutonium and etc.) and other elements present in RW in the form positive ions or neutral compounds. These elements provide for the major part of HLW and ILW radioactivity. There are two main types of radionuclides for which chemical containment is considered to be of low efficiency. Although their activity levels in the disposal facility are relatively low, based on these radionuclides actually that the long-term radiological hazard of the system is determined. These radionuclides can form gaseous compounds not interacting with cement (for example, ¹⁴C in methane) and long-lived radionuclides with low solubility, such as ¹²⁹I and ³⁶Cl present in the form of anions.

1.3 Other advantages of cement-based material use

In a disposal facility, corrosion of iron and other metals taking place in anaerobic conditions (no oxygen) results in gas generation. Gas may be generated due to organics decomposition resulting from microbiological activity, radiolysis and radioactive decay. Excess pressure may thus be developed

² This sealing capacity time period is set forth by regulation ND-093-14 Acceptance Criteria for Radioactive Waste Disposal. However, vitrified HLW resulted from SNF reprocessing when U and Pu components are retrieved and long-lived radionuclides (such as ³⁶Cl and ¹²⁹I) are carried over, most of the activity decreases within approximately 300 years (see figure 1) and the time period the package can remain intact may be much shorter with no major damage caused to the environment.

³ Mixture with mass fractions of Portland cement (26 %), ground limestone (29 %), hydrated lime (10 %), water (35 %). In the UK disposal concept, this mixture is proposed as a backfill for HLW and ILW disposal. Whereas, in the NKM project it is proposed as an analogue of cement barrier for ILW

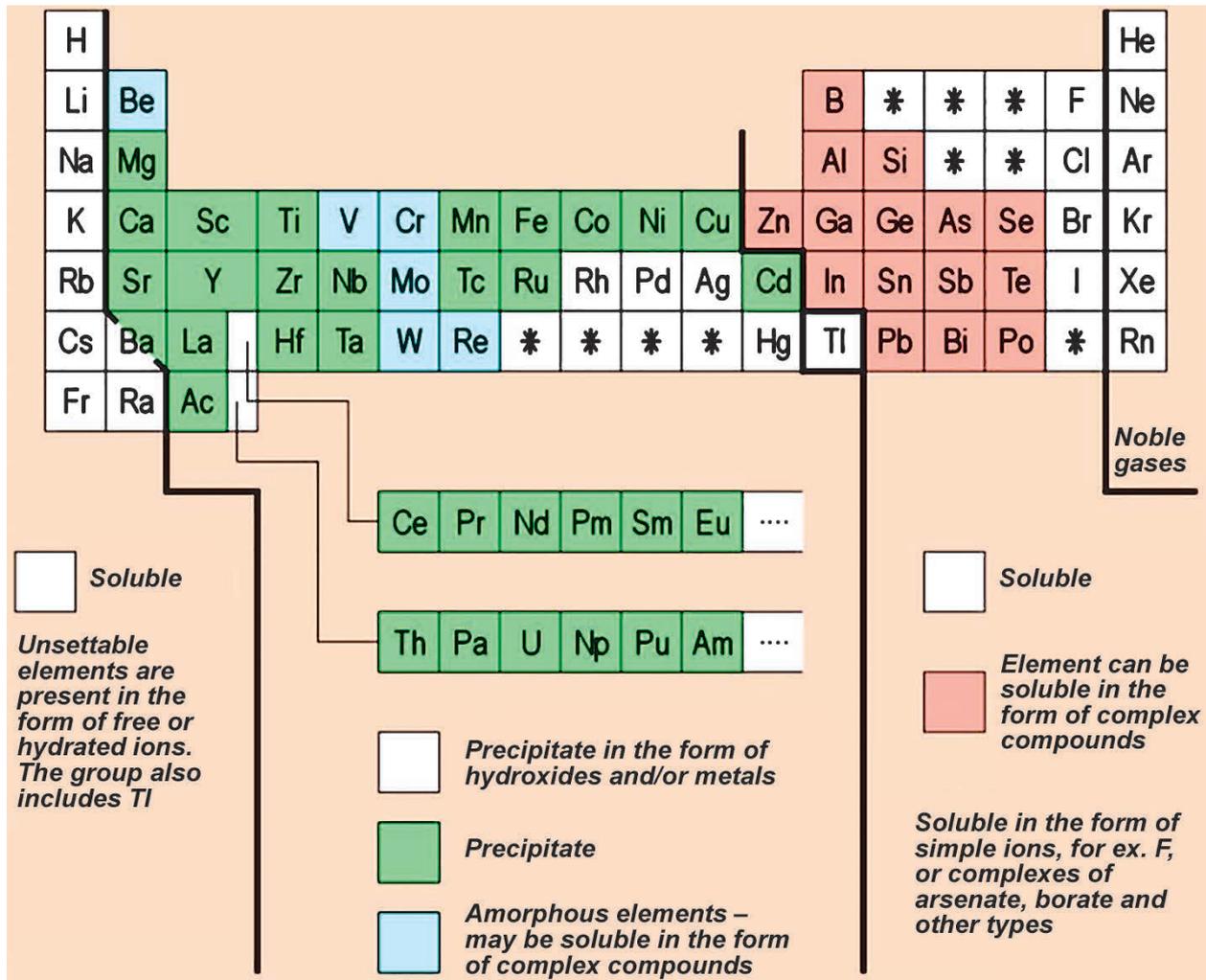


Figure 2. Oxide solubility levels for pH = 12–13, absence of strong complexing agents and oxidants. Data not available is marked with "*" [8]

inside the containers and engineered barriers. To prevent this, the gas has to escape the packages and the near field of the disposal facility. The required permeability of cement-based materials is readily attainable owing to their high porosity (see, for example, [10]). If the requirements set for a disposal facility include the possibility of waste retrieval after the backfilling is installed, cement-based material strength may be considerably low (for example, for hydraulic washing out [11]) with all other necessary properties maintained.

2 Challenges for cement-based materials use

From the long-term safety perspective, the main challenges associated with the use of cement-based materials for barrier construction in deep geological disposal facilities may be exemplified as follows:

- Chemical containment must be ensured during the required period of time – hundreds of thousands of years;
- Chemical containment must be effective all across the disposal area given the heterogeneity of RW emplacement and ground water flows;

- Generation and transport of colloids providing for insoluble form of radionuclides;
- Tolerability of cement-based materials under repository thermal conditions (over 100 °C);
- High rate of HLW glass-matrix dissolution in alkaline media.

These issues have been studied for a long time with some of these findings presented below.

2.1 Extended time scales

Most part of radionuclides will decay inside the near-field of a repository given that the chemical containment conditions retarding radionuclide release are maintained for hundreds of thousands of years. To check if these conditions can be met, a combination of accelerated laboratory tests, modelling and surveys have been performed at a unique site containing “natural cement” – Makarin deposit (Jordan). Some tens of thousands of years ago, bitumen containing clayey limestone caught fire and its burning resulted in a material very similar in its properties to common Portland cement (CPC) [12].

2.1.1 Long-term conditioning of ground water

Having reacted with cement backfill, the ground water flowing into disposal facility becomes alkaline due to the dissolution of backfill active components containing calcium hydroxide and cement gel generated due to CPC hydration. The time needed for such ground water conditioning may be evaluated based on the amount of alkaline materials in the backfill and their solubility rate. Such evaluations based on the accelerated leaching of cement material [13, 14] have shown that it contains enough calcium hydroxide to condition water to the level of over 12 pH and the water volume could be more than 200 times higher than the backfill one. After the calcium hydroxide is dissolved, ground water conditioning will proceed due cement gel dissolution. The volume of water with $\text{pH} > 10$ conditioned via this mechanism will be approximately 800 times bigger than the gel volume. Thus, the total water volume that can be conditioned due to its reaction with cement material of NRVB type to the initial level of $\text{pH} = 7\text{--}10$ is approximately 1,000 times higher than its volume.

Simple estimates show that the amount of cement around 3.5 m tall HLW package with a diameter of 0.7 m in a deep geological disposal facility with average hydraulic permeability values amounting to 0.04 m/year, hydraulic pressure gradient — to 0.05 [15] and vitrified HLW placed in vertical disposition holes, shall approximately account for 19 m³. This amount of cement corresponds to a disposition hole with a diameter somewhat bigger than 2.5 m and its drilling is not considered to be complicated from the technical point of view.

These estimates account for “focusing” effect of the water flow passing through the backfill having higher hydraulic permeability than the bed rock, as well as conditioning of groundwater by other materials in the disposal facility. In particular, formation of acids reducing the pH level may result from the decomposition of organics, for example, polyvinylchloride contained in ILW. Estimates [14] show that the presence of one weight percent of organics decreases the conditioning capacity of cement-based material approximately by 20%. On the other hand, availability of repository structure material containing cement, such as cement grouting in containers and cement backfilling of ILW disposal caverns make up by wide margins for the possible decrease in pH due to organics present [13]. Thus, if during repository system evolution the backfilling material is altered only due to its interaction with ground waters, their conditioning in case of vitrified HLW disposal under NKM project can be ensured within a wide safety margin for a million of years after the repository is closed.

As it was mentioned before, its calcium hydroxide and cement gel contained in the backfill material and generated due to CPC hydration that ensure ground water conditioning. Calcium hydroxide is present in crystalline form which is not subject to

any alterations with time (if no dissolution reaction takes place) unlike amorphous cement gel that can undergo crystallization becoming similar in its form to natural cement materials. It looks quite possible that higher stability of crystalline material will result in lower pH values attained during ground water conditioning compared to those of the cement gel. Under natural temperature conditions crystallization process occurs very slowly. Tens of thousands year old cement gel found in the natural deposit Makarin (see for example [12]) clearly demonstrates this fact. Gel crystallization process was simulated under laboratory conditions: artificial ageing of gel at 80–120 °C [13] resulting in formation of naturally occurring materials. Under these conditions pH values of water dropped approximately by one unit.

2.1.2 Long-term alterations of sorption capacity

Minerals contained in cement are characterized by high radionuclide sorption capacity which was evidenced through a big number of experiments. Thus, this is seen as the second mechanism ensuring chemical containment. Such experiments are carried out using “fresh” cement which is considered to be a potential disadvantage, as a question arises: whether the performance of this material will be maintained during the timescales considered in relevant safety assessments.

This effect was investigated in more detail in [11] focused on uranium sorption by (a) samples of NRVB material leached until complete elimination of calcium hydroxide, and (b) other samples of this material held at 120 °C for 6 months resulting in cement gel crystallization. The experiments showed that such treatment had no effect on material sorption properties. [13] provides the findings of investigations involving samples containing thorium, uranium and plutonium “aged” using the same method. For thorium, 10% of initially absorbed substance passed into solution under the “aging” process. Other actinoids remained in new crystalline phases generated from gel. Uranium was fully contained in newly generated minerals, whereas plutonium passed to newly formed calcium crystals. NRVB ageing resulted in 10-fold decrease of uranium solubility rate. However, plutonium solubility remained the same. These experiments have shown that cement materials “ageing” has only insignificant impact on their sorption properties.

2.2 Heterogeneity

The assessments assumed that homogeneous mixture of backfill, pore liquid and radionuclides (excluding highly soluble and non-absorbed) in the form of oxides or hydroxides generated under high pH values is present in ILW disposal. Permeability of ILW disposal system was supposed to be equal to the one of the backfill, thus, enabling to evaluate cement material conditioning capacity as a whole.

These estimates provide no account for non-uniformity of ground water flow and the ILW disposition inside waste packages.

2.2.1 Non-uniformity of ground water flow

The biggest non-uniformity of ground water flow is supposed to take place if large highly permeable fractures are developed. Such fracturing can result in the formation of channels intensifying ground water flow and, thus, causing local losses of conditioning capacities provided by the cement backfill.

In the extreme case, when the fractures become the only medium for ground water flow, diffusion through the backfill material and waste packages is supposed to be the only mechanism for radionuclide transfer. These considerations demonstrate that the model of a uniform flow moving through hypothetically homogeneous mixture of RW and backfill will be more conservative than the model of fractured backfill and packages.

It appears that some degree of backfill shrinking and its fracturing is inevitable. These processes are expected to occur at the early post-closure stage until full saturation of the repository. [16] provides some attempts to simulate backfill material fracturing processes. Figure 3 shows the key mechanisms that can possibly result in cement matrix and backfill fracturing [16].

The most significant ones are as follows:

- Plastic deformation due to cement grout shrinking caused by fluid losses (horizontal fractures with an opening width of up to 2 mm);
- Early thermal shrinking at the solidification stage (deformation is caused by cooling of the solution earlier expanded due to chemical reactions; formation of vertical fractures with an opening width of up to 0.2 mm);
- Thermal expansion of metal packages and reinforcement metal of concrete containers (the amount of corrosion products is significantly

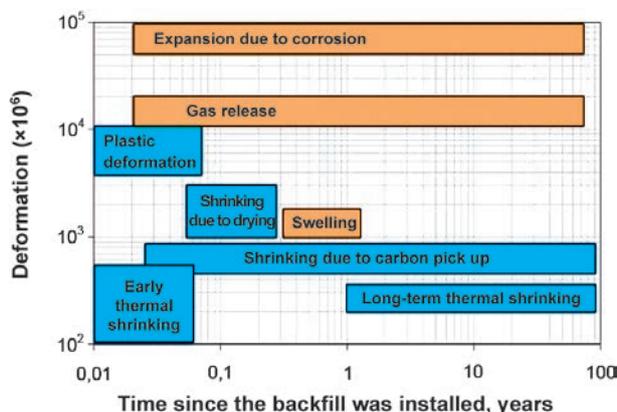


Figure 3. Possible mechanisms inducing changes in sizes at different stages of matrix and backfill material evolution in a deep geological disposal facility. Lighter blue color indicates the increase of liner size. Deformation suggests relative changes in liner size.

higher than the initial amount of relevant material) and the pressure produced by gases generated due to corrosion process.

The latter two mechanisms may result in fracturing with an opening width of up to 5 mm. Quantitative estimates for water transfer, presented in [16], have shown that backfill fracturing may result in some 3-fold increase in ground water flow through a particular disposal chamber.

Products generated due to the interaction of ground water and cement backfill may precipitate at the surface of fractures penetrating the backfill which is another possible impact. Depending on ground water chemical content, for some of the products resulting from such interaction the permeability is low and they may isolate the ground water flow from the main backfill material enabling to achieve high pH as the result of ground water conditioning (see, for example, [7]).

If the backfill is fully intersected with a fracture, this effect produces no negative impact as the limited ground water conditioning also suggests that the sediments present at fracture surface will impede radionuclide migration into fractures. However, if one side of the fracture passes the backfill and the other – waste package surface, and reaction products isolate the backfill surface, then unconditioned ground water may interact with RW. Similar scenarios have been studied for ILW packages located immediately adjacent to each other. Only these cases can result in a decrease of backfill chemical containment capacities due to ground water flow non-uniformity. However, full-scale experiments involving backfill and its cracking show that formation of fractures at the interface between the backfill and the package or between closely adjacent packages can not be considered as the predominant fracturing mechanism. Isolating capacity of products generated due to ground water and backfill interactions is typical only for particular combinations of ground water components.

Sediments are also expected to be produced at fracture outlets in the backfill due to their interaction with ground water resulting in calcite formation (CaCO_3). According to [17] approximately 500 years later the amount of calcite resulting from such interactions will prove to be sufficient to seal smaller fractures and decrease the water flow through the backfill. Experiments focused on carbon pick up have shown that fractures with an opening width of up to 0.1 mm can be sealed. At the same time, Ca transfer from the backfill was retarded by calcite deposits at the walls of fractures having an opening width of over 0.2 mm and fracture sealing process is slowed down.

Dissolution of the backfill at its interface with the bed rock will take place in parallel with sedimentation of newly generated minerals produced due to the interaction of ground water solutions with cement hydration products. Sedimentation of secondary minerals may result in the sealing



Figure 4. Photos of fractures sealed with calcite during alkali plume migration in the Makarin mineral deposit (Jordan) [24]

of fractures penetrating the bed rock and located at the periphery of disposal caverns or boreholes [18]. This process can also slow down the rate of cement barrier degradation due to a lower intensity of ground water flow [19]. Self-sealing of fractures is a quite known process observed in LLW and ILW disposal facilities [20]. Figure 4 shows the calcite mineralization of naturally occurring fractures during alkali plume migration from the Makarin mineral deposit in Jordan [21].

To date, calcination process and fracture sealing can be quite adequately described, that is why it is no longer considered in a conservative manner under quantitative safety assessments.

2.3 Formation of colloid particles

Chemical containment is mainly aimed at decreasing the concentration of radionuclides in the aqueous phase due to their sorption and limited dissolution. So due to colloids present in the flow, the process looks quite more complicated, thus, under certain extreme assumptions regarding colloid concentrations and radionuclide content therein, the rate of contaminant migration in the disposal system can increase significantly.

Colloids may originate from repository structure materials. They can also be introduced to the disposal system with ground water flow. However, colloid-driven radionuclide migration may become significant only if they are stable, mobile, their concentration is high and they have high sorption capacity. The first condition required — colloid stability is hardly attainable when cement backfilling is used: due to the highly alkaline media, colloids will have an opportunity to interact with a big number of other charged particles. During the experiments carried out with cement materials, radionuclide concentrations in the colloid fraction always proved to be insignificant [22–24]. Low colloid concentrations revealed during the experiments are believed to be generated due to high ionic strength of solutions. During the experiments, presence of colloid

fraction resulted in a 30% increase of uranium concentration [13]. This value is somewhat lower than the experimental uncertainty of the solubility limit, and much lower compared to the possible uranium concentration increase due to formation of soluble complexes with cellulose degradation products.

Concentrations of naturally occurring colloids that may be introduced to the disposal facility with ground water flow are believed to be very low. It is not surprising as a significant part of naturally occurring colloids originates from natural organics (humus and microorganisms) and their concentrations at repository depths are quite small. Even if such colloids are introduced to the disposal system they prove to be unstable given the high ionic strength of ground water.

2.4 Alteration of cement properties under thermal impact

Disposal of vitrified HLW suggests that stability of material properties is ensured for a long-term period (some 1,000 years) under high-temperature conditions (over 100 °C⁴). As it was stated in previous sections of the paper, cement materials preserve their properties providing a chemical barrier under “ageing” conditions that were simulated by high-temperature heating. This section summarizes the data on the evolution of mechanical and thermal properties of cement presented in [24].

Compression strength reduces approximately by 25% when the material is heated up to 300 °C, under further heating it drops even more drastically. Cement dehydration is believed to be the cause of strength reduction.

Elastic modulus (EM) decreases faster than the strength and its values are much more sensitive to the ambient environment impacts. Relative EM value decreases as a linear function till 0 when the material is heated from 100 to 800 °C. When heated to up to 300 °C, EM values account for 40–90% from the initial ones. However, this fact does not undermine the strength of the disposal system if the stress loads on the cement material at the post-closure stage is associated with compression. The expected stress impacts on the material will be decreased due to the decrease in EM values, thus, reducing the risk of its fracturing.

Thermal expansion factor for concrete (a mixture of cement, sand and fillings) heated up to a temperature of up to 300 °C is somewhat constant amounting to some $(10 \pm 2) \cdot 10^{-6}$ mm/(mm·°C). Under elevated temperatures, cement grout deformation occurs due to dehydration driven compression, so that negative strain appears.

⁴ Cement is proposed as a buffer material for HLW disposal. This high-level waste results from SNF reprocessing performed according to modern technologies enabling to reduce HLW amount but providing for an increase in their specific activity. Thus, buffer potentially can be heated to up to 300 °C [24].

Heat transfer factor decreases as a liner function to up to 0.4 from the initial value when the temperature rises from 20 to 300 °C.

Concrete permeability increases when the material is heated. Based on the limited data available on the increase of porosity values due to heating and the observed correlations between porosity and permeability, it was identified that when heated to up to 300 °C the possible increase in the permeability may 10 times exceed the initial value. This can result in the increase of ground water flow through the disposal system. However, a much higher increase in porosity is believed to be due to cement fracturing.

Based on [25], **chemical composition** of Portland cement remains constant (not accounting for free and bound water evaporation) till its heated to up to 350 °C. Overheating results in chemical decoupling of silicates present in the gel.

In general, all the research performed involving both common and aluminous cement provide no grounds to suggest that they can be considered unsuitable for their use as buffer and structural material inside disposal facilities under relevant thermal conditions (over 100 °C).

2.5 Impact of cement materials on the dissolution rate of HLW glass matrix

A little research has been done to date on this issue and the currently available data deals mostly with borosilicate glasses proposed abroad that are very different from sodium aluminum-phosphate glasses proposed in Russian designs. High dissolution rate was observed in laboratory tests involving mock-up glasses and solutions. However, the actual content of ground water and interacting engineered barriers may reduce the conservatism of these preliminary estimates.

Under conditions similar to neutral ones (pH = 7), the initial stage of dissolution resulting from glass interaction with water is characterized by water molecular diffusion into glass structure and ion exchange (stage I), followed by glass structure hydrolysis resulting in the glass component release into the solution (stage II, “initial dissolution phase”). These processes result in the formation of a gel structure at the glass surface joined with soluble components. Further dissolution is evidenced through a drastic decrease in the rate of glass component release to the solution (stage III, “slowed down initial dissolution”). Once this process is terminated, dissolution rate becomes rather constant – by 3–4 orders of magnitude lower than the value typical for stage III. The following stage IV is called “residual dissolution stage”. In some cases, once the stage IV is terminated the dissolution rate increases again (stage V) [26]. These mechanisms are well known. However, in some cases a fewer number of dissolution stages can be identified (see, for example, [27]).

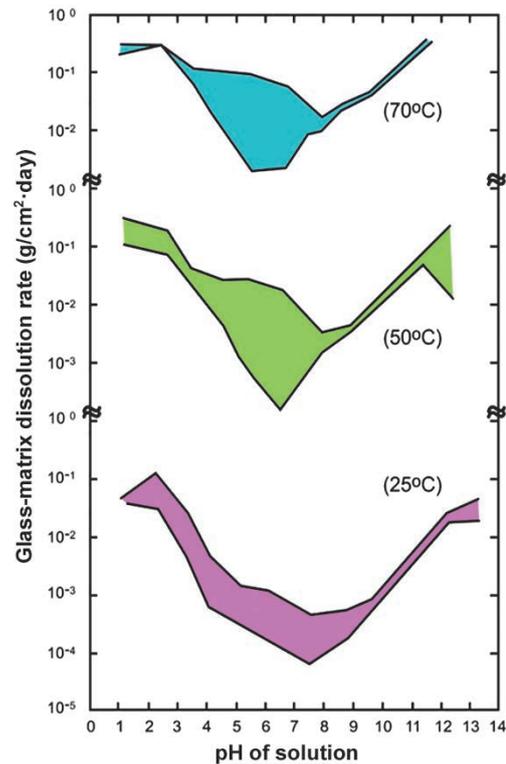


Figure 5. Correlations between the initial dissolution rate of borosilicate glass involving 5 components and the temperature. Filled areas indicated the range of solubility rates identified for one of the glass components

Glass dissolution rate under phase II depends on relevant pH value of the solution: it increases drastically both in alkali and acid media. Figure 5 shows the correlation between borosilicate glass dissolution rate and the temperature, as well as pH values and its component release (aluminum, boron, silicon, sodium) [28]. The filled areas indicate the range of possible dissolution rates identified for one of glass forming components [28].

Modelling experiments [29–30] have shown that the initial dissolution rate for borosilicate glasses under 100 °C and neutral solution accounts for approximately 10^{-3} – 10^{-2} g/cm²·day and increases gradually with the increase in pH level (proportionally to approximately $10^{0.4\text{pH}}$).

The dissolution rate for Russian sodium aluminum-phosphorus glasses under ambient temperature conditions for most highly soluble components (like Na) was lower than 10^{-4} – 10^{-5} g/cm²·day. However, when aqueous vapor was heated enough they started to recrystallize swiftly and the leakage rate increased to up to 3 orders of magnitude with many of residual elements (such as radionuclides) transferred into colloid form [35–38]. Thus, under HLW repository conditions, resistance of sodium aluminum-phosphorus glasses may be lower than the one of borosilicate glasses.

Experiments involving solutions having salt content similar to actual ground waters showed a decrease in glass dissolution rate – approximately by

two orders of magnitude compared to a solution with low salt content [39–42].

The computational model proposed in [43] accounted for saturation of products generated due to corrosion of iron container interacting with the glass. It was shown that “quickly” soluble glass fraction accounts only for some 1% of its initial activity for several decades. The rate of further glass dissolution was shown to decrease by 3 orders of magnitude.

As for glasses that have been staying in natural environment for thousands of years, relevant studies have shown that the actual rate of glass dissolution always remained lower than the one derived from parameter values of existing models [44, 45].

Preliminary estimates have demonstrated that glass dissolution rate should not be seen as a fundamental factor. Even if it increases by 3 orders of magnitude, this fact is not considered to be critical for repository safety given that other barriers maintain their normal performance. Although more detailed study is required on this issue.

Conclusions

Preliminary study of the data available has shown that there are no reasons suggesting that cement materials are not suitable for engineered barrier construction in the Nizhnekansk HLW deep geological disposal facility. Relevant advantages can be exemplified as follows:

- Formation of an effective chemical barrier decreasing the solubility limit for the majority of radionuclides and ensuring an increased sorption capacity;
- Decrease of iron container corrosion rate by several orders of magnitude;
- High gas permeability preventing the excess pressure formation inside engineered barrier system under anaerobic corrosion of steel accompanied by hydrogen release;
- Fractures around the excavation can be “sealed” using special calcite-type materials;
- A similar barrier is used both for ILW (in this case cement is a common solution) and HLW disposal. Negative impacts of cement backfilling present in ILW disposal caverns on the bentonite (when it is used as a barrier material for HLW disposal) may be mitigated using a wider spacing between HLW and ILW disposal areas;
- Cement material is easy to handle so that the backfill can be installed even in deep disposition holes (tens of meters). Installation of bentonite buffer of proper quality may be quite complicated from a technical point of view which outweighs all the positive aspects associated with its use.

The required time period for ground water conditioning (over one million of years) can be achieved through the use of an adequate amount of cement for barrier construction. For Nizhnekansk project, the required diameter of the disposition hole accounts for approximately 2.5 m.

Cement cracking is not considered critical given the low permeability of crystalline rocks. Potential release of weakly absorbed radionuclides — ^{36}Cl and ^{129}I through fractures present in the backfill is offset with a minor content of these radionuclides⁵ in the vitrified waste (these radionuclides are almost completely eliminated during SNF reprocessing).

Fracture surface “reinforcing” and presence of ground water flows at the interface “backfill — container” will hardly play a major role. Although further studies on this issue are required.

Colloid concentration in a solution with a high ion content ensured by cement material is expected to be negligibly low.

Cement heating to the highest possible temperature under repository conditions produces no impact on the chemical containment ensured by cement material. Some deterioration of cement physical properties (such as higher water permeability values) is not considered to be critical.

The most challenging issue is associated with the high rate of HLW glass matrix dissolution in alkali environment. However, it should be noted that this high rate is typical only for early stages of glass dissolution and saturation of steel HLW container corrosion products with glass degradation products. We hope that the total radionuclide release under this mechanism will prove to be insignificant (a few percent).

References

1. Materialy obosnovaniya litsenzii na razmeshcheniye i sooruzheniye ne otnosyashchegosya k yadernym ustanovkam punkta khraneniya RAO, sozdavayemogo v sootvetstviy s proyektnoy dokumentatsiyey na stroitelstvo obyektov okonchatelnoy izolyatsii RAO (Krasnoyarskiy kray. Nizhnekanskiy massiv) v sostave podzemnoy issledovatel'skoy laboratorii (vkluyuchaya materialy otsenki vozdeystviya na okruzhayushchuyu sredyu). FGUP «Natsionalnyy operator po obrashcheniyu s radioaktivnymi otkhodami», Moskva, 2015 g. URL: <http://bezrao.ru/hearings/pzro-zheleznogorsk> (data obrashcheniya 31.08.2017). (In Russian).
2. Poyet S. The Belgian supercontainer concept: Study of the concrete buffer behaviour in service life. *Journal de Physique IV France*, 2006, vol. 136, pp 167–175.
3. Hicks T. W., Baldwin T.D., Hill I. et al. The Feasibility of using Multipurpose Containers for the Geological Disposal of Spent Fuel and High Level Radioactive Waste. Report N°1107-1 of Galson Sciences Ltd. 2011, 59 p. URL: <https://rwm.nda.gov.uk/publication/the-feasibility-of-using-multi-purpose-con>

⁵ It is based on these radionuclides, whose presence is mostly typical for ILW repositories, that the radiation safety of a disposal facility is determined for the long-term perspective. Today, the possibility of segregating RW having high content of these radionuclides from the RW stream and their separate disposal is actively discussed. However, this matter falls beyond the scope of this research.

- tainers-for-the-geological-disposal-of-spent-fuel-and-high-level-radioactive-waste-november-2011/ (data obrashcheniya 15.07.2017).
4. Glossariy MAGATE povoprosam bezopasnosti. Terminologiya. ispolzuyemaya v oblasti yadernoy bezopasnosti i radiatsionnoy zashchity. Vena: Mezhdunarodnoye agentstvo po atomnoy energii, 2007, 295 c.
 5. *Revertegat E., Richet C. and Gegout P.* Effect of pH on the Durability of Cement. Pastes, Cem. Concr. Res, 1992, vol. 22, pp 259–273.
 6. The Scientific Foundations of Deep Geological Disposal. United Kingdom Nirex Limited. Report No: N/016. 2001, 118 p. URL: <https://rwm.nda.gov.uk/publication/the-scientific-foundations-of-deep-geological-disposal-nirex-report-n016-2001/> (data obrashcheniya 15.07.2017).
 7. *Hooper A. J. and Francis A. J.* Made to Measure Safety. International Cement Review, 1994, pp 59–62.
 8. *MacPhee D.E. and Glasser F.P.* Immobilisation Science of Cement Systems, MRS Bulletin, March 1993, p. 66.
 9. *Vines S. and Beard R.* An overview of radionuclide behaviour research for the UK geological disposal programme. Mineralogical Magazine, 2012, vol. 76, no 8, pp 3373–3380.
 10. Geological Disposal. Behaviour of Radionuclides and Non-radiological Species in Groundwater Status Report. NDA Report no. DSSC/456/01. 2016. 131 pp. URL: <https://rwm.nda.gov.uk/publication/geological-disposal-behaviour-of-radionuclides-and-non-radiological-species-in-groundwater/> (data obrashcheniya 15.07.2017).
 11. *McCall A., Holmes J., Harrison M. et al.* Backfill - Development and Placement of an Engineered Barrier. Proceedings of the Sixth International Conference on Radioactive Waste Management and Environmental Remediation: ICEM '97. Singapore, October 12–16, 1997. New York, N.Y.: American Society of Mechanical Engineers, 1997, pp 347–351.
 12. MAQARIN natural analogue study: Phase III. Edited by J A T Smellie, Swedish Nuclear Fuel and Waste Management Co Technical Report TR-98-04, 1998, 401 pp.
 13. *Crossland I.G. and Vines S.P.* Why a cementitious repository? United Kingdom Nirex Limited 2001, Report N/034, 2001, 19 pp. URL: <https://rwm.nda.gov.uk/publication/why-a-cementitious-repository-nirex-report-n034-2001/> (data obrashcheniya 15.07.2017).
 14. *Rochelle C.A., Purser G., Milodowski A.E. et al.* Results of laboratory carbonation experiments on Nirex Reference Vault Backfill cement. FORGE Report D3.19. 2014. 65 pp. URL: <https://www.bgs.ac.uk/forge/docs/reports/d3.19.pdf> (data obrashcheniya 15.07.2017).
 15. *Ozerskiy A. Yu.* Osobennosti filtratsii podzemnykh vod v massive kristallicheskih porod v yuzhnoy chasti Eniseyskogo kryazha / Sb. «Kompleksnyye problemy gidrogeologii»: Tez. dokl. nauch. konf., Sankt-Peterburg, Rossiya, 23–25 oktyabrya 2013 g. — S.-Pb. : Sankt-Peterburgskiy gos. un-t, 2013, S. 110–113. (In Russian).
 16. *Swift B. T., Bamforth P.B., Baston G.M.N.* Cracking in a cementitious backfill and its implications for flow and chemistry. Mineralogical Magazine, 2012, vol. 76, no 8, pp 3071–3082.
 17. *Brodersen K. and Nilsson K.* Pores and cracks in cemented waste and concrete. Cement and Concrete Research. Cement and Concrete Research, 1992, vol. 22, pp 405–417.
 18. *Wang L., Jacques D. and De Canniere P.* Effects of an alkaline plume on the Boom Clay as a potential host formation for geological disposal of radioactive waste, External Report, SCK-CEN-ER-28, Mol, Belgium. October 2007, 193 pp. URL: http://publications.sckcen.be/dspace/bitstream/10038/1397/1/er_28final.pdf (data obrashcheniya 11.07.2017).
 19. Demonstration of disposal feasibility for spent fuel, vitrified high-level waste and long-lived intermediate-level waste. Nationale Genossenschaft fur die Lagerung radioaktiver Abfalle (Nagra) Technical Report 02-05. 2005, 472 pp. URL: [https://www.nagra.ch/data/documents/database/dokumente/\\$default/Default%20Folder/Publikationen/NTBs%202001-2010/e_ntb02-06.pdf](https://www.nagra.ch/data/documents/database/dokumente/$default/Default%20Folder/Publikationen/NTBs%202001-2010/e_ntb02-06.pdf) (data obrashcheniya 11.07.2017).
 20. *Pfingsten W.* Indications for self-sealing of a cementitious repository for low and intermediate-level waste. Nagra Technical Report 01-05. 2001, 112 pp. URL: http://www.iaea.org/inis/collection/NCLCollectionStore/_Public/33/012/33012826.pdf (data obrashcheniya 21.07.2017).
 21. Cementitious materials in safety cases for geological repositories for radioactive waste: Role, evolution and interactions. Nuclear Energy Agency Organization for Economic Co-operation and Development (NEA OECD), Brussels, Belgium. November 2009, 258 pp. URL: <https://www.oecd-nea.org/rwm/docs/2012/rwm-r2012-3.pdf> (data obrashcheniya 21.07.2017).
 22. *Alexander W.R. and Moeri A.* Cementitious colloids: integration of laboratory, natural analogue and in situ field data. Geochimica et Cosmochimica Acta. Conference: 13th Annual VM Goldschmidt Conference, 2003, vol. 67, pp 159–160.
 23. *Swanton S.W., Alexander W.R. and Berry J.A.* Review of the behaviour of colloids in the near field of a cementitious repository Report to NDA RWMD. Technical & Assurance Services Serco Report SERCO/TAS/000475/01. 2009, 193 pp. URL: <http://www.natural-analogues.com/nawg-library/colloids-microbes> (data obrashcheniya 25.07.2017).
 24. *Dickinson M., Holton D., Bamforth Ph. and Cairns M.* Development of Understanding of Disposability of High-Heat-Generating Wastes in the UK. WMS Journal, 2015, vol. 1, no. 2, 13 pp. URL: <http://www.wmsjournal.org/Volume1Issue2/15359.pdf> (data obrashcheniya 25.07.2017).
 25. *Naus D.J.* A Compilation of Elevated Temperature Concrete Material Property Data and Information for Use in Assessments of Nuclear Power Plant Reinforced Concrete Structures, Oak Ridge National Laboratory, Report ORNL/TM-20009/175. 2010,

- 328 pp. URL: <https://www.nrc.gov/reading-rm/doc-collections/nuregs/contract/cr7031/> (data obrashcheniya 27.07.2017).
26. Jantzen C.M., Brown K.G., Pickett J.B. Durable Glass for Thousands of Years. *International Journal of Applied Glass Science*, 2010, vol. 1, pp 38–62.
27. Vienna J., Ryan J., Gin S. Current understanding and remaining challenges in modelling long-term degradation of borosilicate nuclear waste glasses. *International Journal of Applied Glass Science*, 2013, no 4, pp 283–294.
28. Knaus K.G., Bourcier W.L., McKeegan K.D., Merzbacher C.I., Nguyen S.N., Ryerson F.J., Smith D.K., Weed H.C. and Newton L. Dissolution Kinetics of a Simple Analogue Nuclear Waste Glass as a Function of Time, pH and Temperature. *Materials Research Society Symposium Proceedings*, 1989, vol. 176, pp 371–381. MRS Online Proceeding Library Archive 176, doi: 10.1557/PROC-176-176.
29. Gin S., Jollivet P., Fournier M., Berthon C., Wang Z., Mitroshkov A., Zua Z., Ryan J. The fate of silicon during glass corrosion under alkaline conditions: A mechanistic and kinetic study with the International Simple Glass. *Geochimica et Cosmochimica Acta*, 2015, vol. 151, pp 68–85.
30. Cassingham N., Corkhill C.L., Backhouse D.J., Hand R.J., Ryan J.V., Vienna J.D., Hyatt N.C. The initial dissolution rates of simulated UK Magnox-ThORP blend nuclear waste glass as a function of pH, temperature and waste loading. *Mineralogical magazine*, 2015, vol. 79, no 6, pp 1529–1542.
31. Gin S., Frugier P. SON68 glass dissolution kinetics at high reaction progress: experimental evidence of the residual rate. *Materials Research Society Symposium Proceedings*, 2003, vol. 807, pp 175–182.
32. Abratis P. K., Livens F.R., Monteith J. E. et al. *Applied Geochemistry*, 2000, vol. 15, pp 1399–1416.
33. Fournier M., Frugier P., Gin S. Resumption of alteration at high temperature and pH: rates measurements and comparison with initial rates. *Procedia Material Sciences*, 2014, vol. 7C, pp 202–208. URL: <https://www.sciencedirect.com/science/article/pii/S2211812814010694> (data obrashcheniya 30.07.2017).
34. Fournier M., Frugier P., Gin S. Resumption of alteration at high temperature and pH: rates measurements and comparison with initial rates. *Journal of Nuclear Materials*, 2014, vol. 448, pp 348–363.
35. Fosfatnyye stekla s radioaktivnymi otkhodami. Pod red. A. A. Vashmana, A. S. Polyakova. M.: TsNIiatominform, 1997, 171 s. (In Russian).
36. Yuditsev S. V., Malkovskiy V. I., Mokhov A. V. Hidrotermalnyye izmeneniya alyumofosfatnoy steklomatritsy s imitatorami vysokoaktivnykh otkhodov. *Doklady AN*, 2016, t. 468, no 2, s. 196–200. (In Russian).
37. Yuditsev S. V., Pervukhina A. A., Mokhov A. V. Vliyaniye raskristallizatsii fosfatnogo stekla na us-toychivost matritsy otkhodov k vyshchelachivaniyu. *Doklady AN*, 2017, t. 473, no 4, s. 477–483. (In Russian).
38. Laverov N. P., Velichkin V. I., Omelianenko B. I. *in dr.* Izolyatsiya otrabotavshikh yadernykh material-ov: geologo-geokhimicheskiye osnovy. M.: IFZFRAN, 2008, 280 s. (In Russian).
39. Ferrand K., Liu S., Lemmens K. The interaction between nuclear waste glass and ordinary portland cement. *International Journal of Applied Glass Science*, 2013, vol. 4, no 4, pp 328–340.
40. Mercado-Depierre S., Angeli F., Frizon F., Gin S. Antagonist effects of calcium on borosilicate glass alteration. *Journal Nuclear Materials*, 2013, vol. 441, pp 402–410.
41. Harrison M.T. Review of Glass Dissolution Rates for use in the Disposal System Safety Case Performance Assessment Models. *National Nuclear Laboratory Report NNL (10) 10734 issue-3*. 2010, 36 pp. URL: https://tools.nda.gov.uk/publication/nnl_10_10734-review-of-glass-dissolution-rates-final-for-bibliography/ (data obrashcheniya 30.07.2017).
42. Valcke E., Smets S., Labat S., Lemmens K., Van Is-eghem P., Thomas P., Godon N., Jollivet P., Parisot G., Mestre J.-P., Jockwer N., Wieczorek K., Pozo C. CORALUS: An integrated in situ corrosion test on α -active HLW glass. URL: http://cordis.europa.eu/pub/fp6- Euratom/docs/euradwaste04pro_posa-valcke_en.pdf (data obrashcheniya 20.12.2017).
43. Argile – Phenomenological evolution of a geological repository. Andra, Dossier 2005. 2005, 527 pp. URL: <https://www.andra.fr/download/andra-international-en/document/editions/269va.pdf> (data obrashcheniya 20.12.2017).
44. McGrail B.P., Bacon D.H., Meyer P.D., Ojovan M.I., Strachan D.M., Ojovan N.V., Startceva I.V. New developments in field studies of low activity waste glass corrosion and containment transport. In R.J. Finch and D.B. Bullen (eds.), *Scientific Basis of Nuclear Waste Management XXVI*, *Materials Research Society Symposium Proceedings*, 2002, vol. 757, pp 83–95.
45. Verney-Carron A., Gin S., Frugier P., Libourel G. Long-term modelling of alteration-transport coupling: application to a fractured Roman glass. *Geochimica et Cosmochimica Acta*, 2010, vol. 74, no 8, pp. 2291–2315.

Information about the authors

Bogatov Sergey Alexandrovich, PhD, Nuclear Safety Institute of RAS, (52, B. Tulsкая st., Moscow, Russia, 115191), e-mail: sbg@ibrae.ac.ru.

Bibliographic description

Bogatov S. A. HLW Disposal of in Verticale Deposition Holes with Cement Backfill – Pro and Contra with Regard to Long Term Safety of a Geological Disposal Facility. *Radioactive Waste*, 2018, no 1 (2), pp. 17–26.