MATERIALS AND TECHNOLOGIES PROVIDING RADICAL IMPROVEMENT OF RW STORAGE FACILITY WATERPROOFING CAPACITIES

Ilyina O. A.[1], Lundin D. S.[2], Proskurin D. V.[3], Vedernikova M. V.[4], Biryukov D. V.[4]

[1]Bentonite Company Ltd, Moscow, Russia
[2]BentIzol Ltd, Kurgan region, Russia
[3]Bentonite of Khakassia Ltd, Republic of Khakassia, Chernogorsk, Russia
[4]Nuclear Safety Institute of the Russian Academy of Sciences, Moscow, Russia

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The article explores the application of modern clay mineral-based materials and technologies for the development and restoration of engineered safety barriers at facilities holding non-retrievable radioactive waste during their upgrading to provide long-term safe storage configuration (term referred to as conservation in Russian literature) and to establish disposal facilities. It presents the fields of application, available experience of such applications and the main characteristics allowing to expand the practice of their application at nuclear facilities.

Keywords: *radioactive waste, bentonite, BentInject, Geosynthetic Clay Liner (GCL), compacted clay liner (CCL), anti-seepage and anti-migration screens, jet-grouting, "buried wall", injection, engineered safety barriers, disposal facilities for radioactive waste, radioactive waste disposal safety, conservation of disposal facilities for non-retrievable radioactive waste, engineered safety barriers.*

Introduction

Deployment of efforts on the development of new disposal facilities for radioactive waste (DFRW) and conservation (upgrading to safe long-term storage configuration) of facilities holding non-retrievable radioactive waste (NRRWF) requires application of most effective technologies with an unconditional regard for the economic component. The long-term safety of such facilities basically depends on the insulating properties of both the geological environment enclosing these facilities and the engineered materials applied during their construction. Almost all specialists involved in DFRW design development and their safety demonstration have recognized that natural clay-based materials play a key role among barrier materials.

Waterproofing properties of clays are well known: more and more publications exploring their actual or prospective application in the field of atomic energy use have started to appear in scientific literature [1—3]. Certain experience has been gained during the operations performed under FTP NRS-1 and FTP NRS-2, namely, during the conservation of surface storage reservoirs for liquid radioactive waste (pool 354 at MCC site, pools B-1, B-2, B-25 at SCC site, reservoir V-9 at PA Mayak site), PUGR decommissioning (EI-2 at PDC UGR site) and installation of additional engineered safety barriers with clay-based materials used as barrier system components. Until 2030, the plans specified in FTP NRS-2 call for the conservation of PKh-1, PKh-2 tailings at SCC site, reservoir V-17 at PA Mayak site, tailing No. 1 at NZKhK site and seven PUGR. These activities, obviously, require technologies enabling the construction of anti-seepage and anti-migration safety barriers.

In the long term, the number of facilities requiring certain application of clay materials can increase significantly: these are the facilities for which the decision on the final stage of their operational life has been put off for later, as well as fundamentally new facilities and areas of application. According to preliminary data, the number of such facilities may amount to 100.

The key activities to be performed at almost all near-surface DFRW and NRRWF involve the construction of cover systems, drainage structures or drainage systems. When it comes to the facilities holding non-retrievable solid radioactive waste (SRW) (purpose-designed structures, PUGR, etc.), these operations provide for the backfilling of voids with a buffer material, elimination of water ingress into the facility, restoration of waterproofing capacity and etc. Considering facilities located in the areas with high groundwater level, tailings, LRW storage reservoirs and a number of facilities holding non-retrievable SRW (for example, earthen trenches), these activities would involve installation of vertical, inclined or horizontal waterproofing screens and curtains in the ground in case of their absence or failure. In practice all of the above tasks can be addressed through the application of certain technologies suggesting the use of natural clay-based materials.

This article overviews modern technologies and materials, in particular, the BentInject composition, widely used in other industries to adjust or manage local hydrological conditions, seeking for their application at nuclear facilities.

Anti-seepage and anti-migration screens

Compacted clay soils

Simple barrier systems are usually made of compacted clay, so-called CCL — Compacted Clay Liner. Given adequate compaction, CCL may provide fairly low hydraulic permeability factor, for instance, 10^{-9} m/s [4]. However, due to uneven settlement and non-uniform compaction, the density of a clay or loam screen may vary from site to site and its waterproofing capacity will be lower than the expected one. Application of screens made of compacted clayey soils may prove to be effective in case if relevant clay deposits are located close to the DFRW site. Some successful case studies exploring the application of CCL made of low-permeability clays as EBS elements in disposal facilities for very low-level and low-level radioactive waste can be found in [5].

In case of clay material shortage, bentonite (5—15 % by volume) can be used as an additive to improve the waterproofing properties of soils. This engineering solution is not considered as a universal one: optimal dosage of the bentonite additive should be calculated for each specific case. A case study exploring the hydraulic conductivity of mixtures made of tropical Brazilian soils and bentonite assuming their application in CCL construction was considered in [6].

The flowchart providing for the fabrication of a composition featuring local soil and bentonite includes mixing, extrusion and homogenization stages. This method seems to be economically feasible only if the production line is located in a close proximity to the disposal facility itself.

Cold recycling technology is considered as another method providing bentonite introduction into soil: it provides soil strengthening (stabilization) through the application of various binding materials by means of their preliminary milling and mixing [7]. At the milling stage, bentonite suspension, which is mixed beforehand at a mobile mixing unit, is injected into the recycler chamber under pressure (bentonite solution or BentInject composition, which will be described later in the article). In addition to bentonite, depending on the purpose of the resulting coating, polymer additives and cement can be used. Recycling is done to a depth of 250 mm.

Geosynthetic clay liners

Geosynthetic Clay Liners (GCL) enabled substantial replacement or supplementation of the compacted clay liners (CCLs) and bentonite-soil mixtures previously used in the barrier systems. GCL is a rolled geosynthetic material with a thin layer (usually 5—10 mm) of Na-bentonite located inside, anchored by a needle-punched method or by sewing. Table 1 compares CCL characteristics with those of Na-bentonite-based GCL.

Based on the assumptions regarding the long-term shear strength and the oxidation resistance, GCL service life is supposed to be at least 100 years [8]. GCL were listed among the best available technologies for the disposal of production and consumption waste [9]. For example, they were used in the construction of the second section of a near-surface DFRW in the Novouralsk city, namely, as part of an underlying screen in combination with a compacted bentonite clay layer.

A method providing for GCL application as part of a covering insulating barrier of a facility accommodating toxic chemical and radioactive substances at the Kolomenskoye site (Moscow) was presented

Table 1. Equivalent hydraulic permeability factor (k_{sat}), thickness and design life of Na-bentonite-based GCL and CCL exposed to various fluids [4]

Penetrant/parameter	GCL	CCL
Clay content, %	> 90	30–50
Swelling clay mineral content, %	> 80	10
Thickness, L, cm	1	$1\cdot10^2$
Porosity, n	0.65	0.3
Mass of swelling clay[1], kg/m^2	4	30–50
Mass of swelling clay per cm of depth, kg/m^2	4	0.6–1.0
Deionized water		
Hydraulic permeability factor k_{sat}[1], m/s	$1\cdot10^{-11}$	$1\cdot10^{-9}$
Equivalent thickness[3], cm	1	$1\cdot10^2$
Equivalent service time[4], years	41	9.5
0.1 M NaCl		
Hydraulic permeability factor k_{sat}, m/s	$4\cdot10^{-11}$	$5\cdot10^{-9}$
Equivalent thickness, cm	4	$5\cdot10^2$
Equivalent service time, years	10	2
0.1 M $CaCl_2$		
Hydraulic permeability factor k_{sat}, m/s	$1\cdot10^{-10}$	$1\cdot10^{-8}$
Equivalent thickness, cm	10	$10\cdot10^2$
Equivalent service time, years	4	1
1 M brine		
Hydraulic permeability factor k_{sat}, m/s	$8\cdot10^{-10}$	$5\cdot10^{-8}$
Equivalent thickness, cm	80	$50\cdot10^2$
Equivalent service time, days	190	70

[1] According to GCL standard manufacturing specification, the bulk density taken into account for CCL is 2 g/cm^3.

[2] The hydraulic permeability factor k_{sat} is derived from experimental values found in literature sources (e. g., Rowe 2001; Shackelford and Lee 2003; Bouazza et al. 2006).

[3] Equivalent thickness is calculated as barrier thickness required to trap the contaminated liquid against the deionized water.

[4] $\eta L/k_{sat}$ expression is used to estimate the equivalent lifetime; it is used to calculate the time required for 10 cm of liquid to pass through a barrier having a thickness of 1 cm (GCL) or 1 m (CCL).

in [10]. The structure of the covering screen consists of a compacted clay sublayer (CCL), geosynthetic clay liner (GCL), drainage, ballasting (sandy loam, sand) and soil layers. The drainage layer provides the diversion of the surface water flow and prevents topsoil erosion, the GCL layer prevents water flows from entering the storage facility. To arrange for the drainage layer, light geosynthetic drainage materials designed to reduce the mechanical load on the storage structure can be used instead of inert materials (crushed stone, sand), in particular, composites with internal drainage tubes are viewed as quite promising elements providing effective interception and centralized streaming of the wastewater to the collector for further treatment and/or analysis and control [11]. The tubes provide effective performance of these materials even in the presence of local negative surface slopes.

GCL application scope can be much wider:

- elimination of accidental spills and spillages of radioactive materials during their handling, in particular, in case of such incidents during their transportation;
- protection of soil from the ingress of pollutants and radioactive substances on the slopes of roads used for RW transportation purposes;
- construction of anti-seepage screens preventing the ingress of contaminants and radioactive substances into the ground underneath the sites providing temporary accommodation and storage of radioactive waste;
- soil isolation in the event of leaks and accidents that can potentially occur under pipelines with chemical liquids, radioactive waste and other hazardous substances;
- as a part of cover screens of NRRWF and DFRW to prevent water seepage into the storage system.

Injectable bentonite composition BentInject

Curtain-type structures based on poor-filtering clays installed in the ground to prevent the spread of pollutants and radionuclides were first developed a long time ago. The “buried wall” method has been widely used in the construction of dams for slurry storage facilities and tailings, industrial LRW storage reservoirs, as well as during the operations on their safety upgrading and conservation. For example, to increase the stability of the TCR (the Techa cascade of reservoirs) dam, a hydraulic lock was installed in its body along the entire length (1.8 km) to a depth of 7—13 m [12].

Modern materials and methods provide the construction of impervious screens reaching a depth of 20 m or more using special insulating compounds with a dry matter content of only 30%.

Insulating bentonite composition BentInject has been purposely developed to eliminate leaks in underground structures and create anti-seepage (impervious) curtains at the facilities holding non-retrievable RW [13]. BentInject is a dry polymineral mixture based on Na-bentonite, plasticizing and stabilizing additives. Upon being mixed with water, a fluid solution with a high content of clay particles is formed. Material thickening and the buildup of required high waterproofing properties happens within 8—12 hours after its injection. Table 2 summarizes the main characteristics of BentInject.

BentInject can be used to produce vertical, inclined and horizontal waterproofing screens and

Table 2. Main characteristics of the insulating bentonite composition BentInject [14]

Indicator	Unit	Indicator value
Free swelling index, not less than	ml/g	13
Water loss, no more than	ml	16
Immersion of a 76 g cone after 28 days since mixing (mobility (viscosity) of the composition), no more than	mm	16
Density of the composition after 28 days since mixing	g/cm^3	1.2 (±10 %)
Hydraulic permeability of the composition after 28 days since mixing, no more than	m/s	$1{\cdot}10^{-10}$
Porosity of the composition after 28 days since mixing	%	88-90

curtains in the ground, for soil compaction purposes (to reduce the porosity and capillarity), to prevent water seepage into underground structures and premises, to restore deteriorated waterproofing structures.

Installation of impervious vertical trench curtains based on the "buried wall" method

Buried wall method is considered as a common construction method applied to erect foundations, pit fences, retaining walls. Trench is excavated provided the protection by a drilling mud based on a mixture of bentonite powder and industrial water. At the same time, the solution stabilizes the walls of the trench and prevents their sliding in water-saturated soils. After reaching the bottom mark established for the standard "buried wall" method, reinforcement frames are usually lowered into the trench. Then the trench is backfilled with a concrete mixture.

The impervious curtain method providing for BentInject application [15] suggests that upon reaching the bottom mark, the trench is backfilled under a low pressure of 2.0—4, 0 atm with a highly plastic BentInject solution having a density of 1.20—1.25 g/cm^3 while the drilling fluid is displaced and simultaneously pumped out by branch pipes. To ensure the maximum continuity of the curtain body, the trench is required to be filled from the bottom up, while the original drilling fluid is displaced and removed from the trench through the outlet pipes (Figure 1).

This method allows the anti-seepage trench curtain being 0.4 to 1 m wide to reach a depth of up to 60 meters. Its length and configuration are not confined in plan. Trenches are arranged in individual sections having a length of 3—6 meters with the next but one section being excavated. The drilling fluid collected from the outlet is stored and then recycled in the next segments of the trench curtain.

Vertical impervious subsurface engineered barriers can be built:

1) upstream of the groundwater flow moving to the disposal site to prevent the inflow of water to RW disposal structures;

2) downstream along the groundwater flow to exclude contaminant spread beyond the site boundaries;

3) around the disposal site to prevent the contaminant spread.

Installation of vertical impervious curtains using jet-grouting technology

Jet-grouting technology is used for soil stabilization and in the construction and reconstruction of any facilities located in incoherent, unstable and water-saturated soils.

This method involves the destruction of the natural soil structure by high-speed jet energy of an injection solution and its mixing with a solution injected under a high pressure. In this case, a soil-bentonite pile (pillar) is formed in the soil mass (Figure 2) [16]. Piles are spaced depending on the geotechnical conditions of the site: sequentially in one row with overlapping contours or staggered in

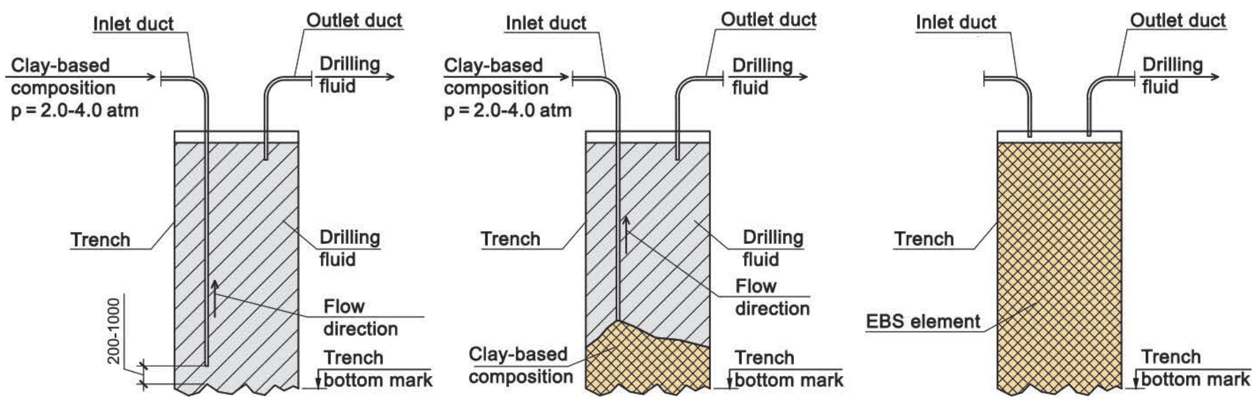

Figure 1. Impervious curtain of a "buried wall" type based on BentInject composition

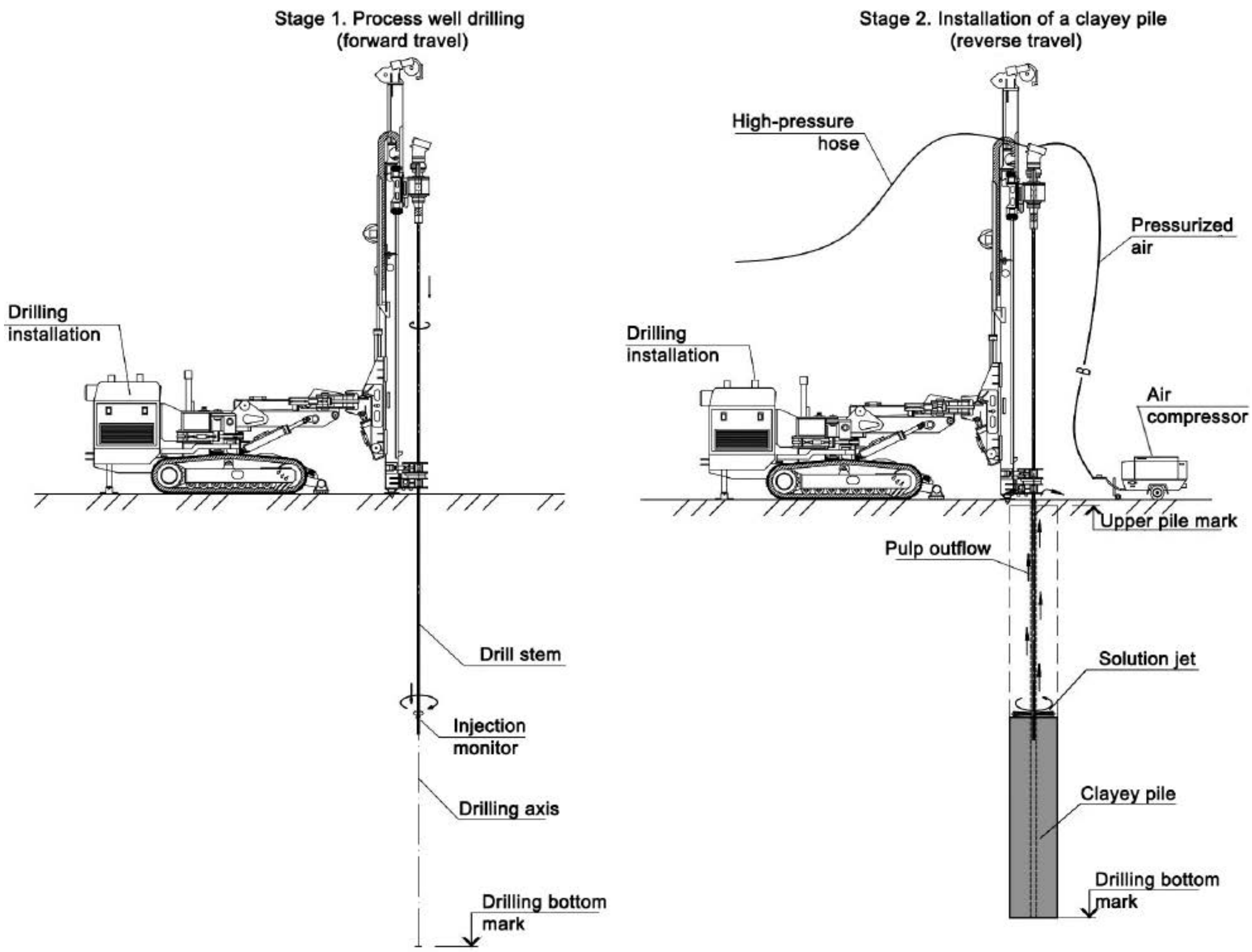

Figure 2. Layout of a soil-clay pile based on BentInject composition

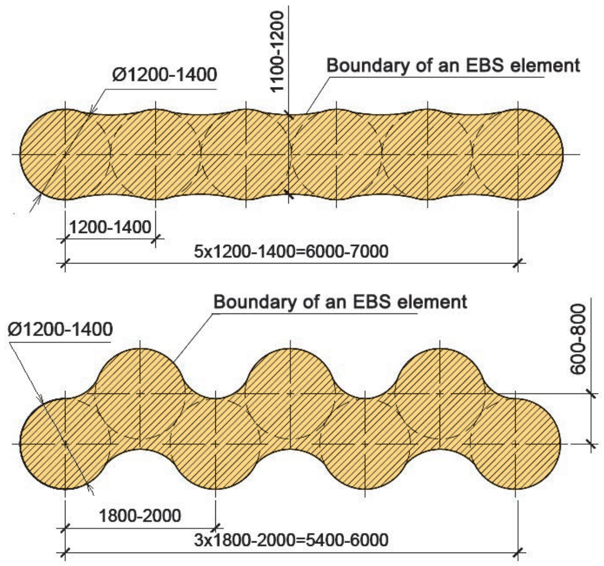

Figure 3. Different screen layouts involving clay piles (barrier elements)

two rows (Figure 3). To create an impervious curtain of a greater thickness in case of staggered orientation, spacing between the pile axes can be reduced. The depth of soil-clay piles can reach 60 meters with the diameter commonly ranging from 0.9 to 1.2 m depending on the soil characteristics and the penetration depth of the injected mixture into it.

Combined application of BentInject material and jet-grouting method provides significant increase in the anti-seepage properties of the soil thanks to a reliable waterproofing screen. After the intended properties are attained, the ultimate screen remains plastic providing a layer of elastic active insulation with self-healing properties that manifest themselves during structure shearing or settlement.

However, BentInject is prone to cracking during drying, for example, close to the surface, if the upper face of the screen is not covered with soil, which is seen as a disadvantage of this method. Nevertheless, upon its saturation, the barrier will fully restore its continuity and waterproofing properties due to the swelling of bentonite clay contained in it.

Ultimate properties of barriers designed based on the "buried wall " and jet-grouting methods may differ: thus, the method for screen construction should be selected depending on the hydrogeological conditions and technical features of the facility in question.

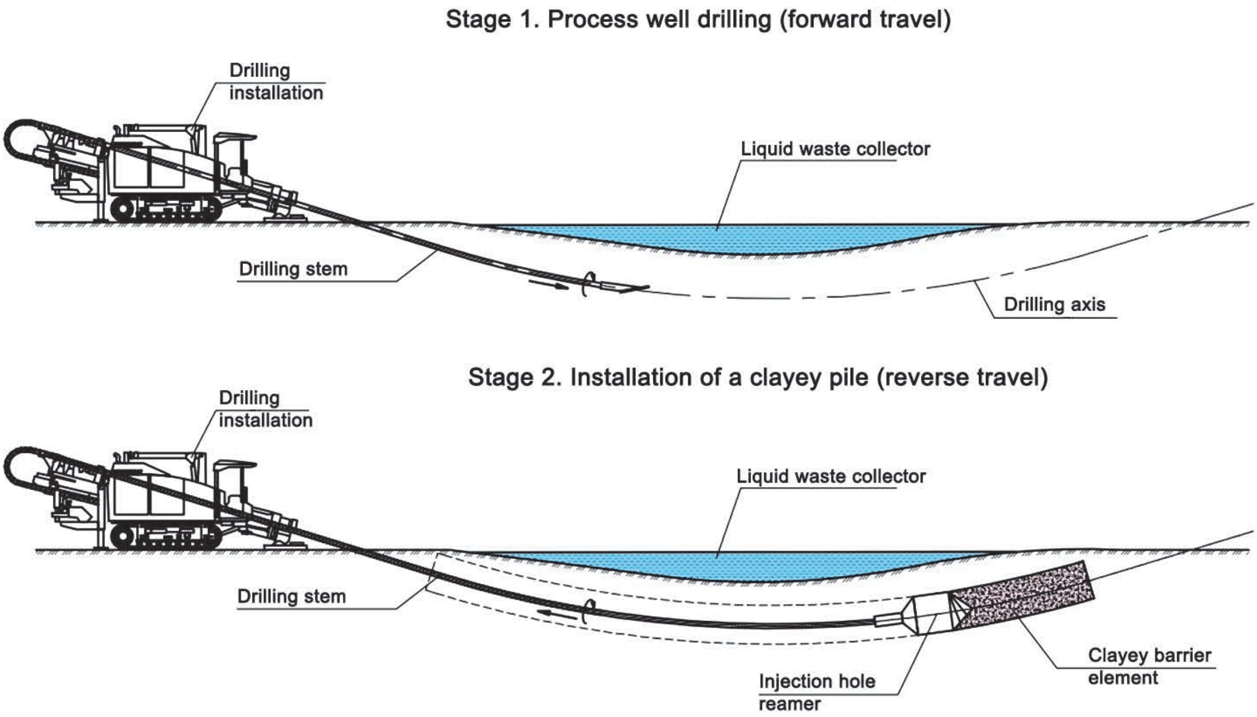

Figure 4. Stages of horizontal EBS clay-based element installation using the BentInject composition

Pilot industrial application of vertical anti-seepage BentInject-based curtains

In 2019—2020, a pilot project on the installation of anti-seepage and anti-migration barriers using jet-grouting and "buried wall" methods was implemented. A total of 3 experimental screen sections was arranged:

- soil-clay pile (jet-grouting) with productive thickness of 1.2—1.4 m reaching a depth of 10 m;
- a section of a "buried wall" being 0.8—1.0 m wide and 6.0 m deep;
- a screen of 6 soil-clay piles being 1.4 m in their diameter and reaching a depth of 10 m was arranged in the immediate vicinity of a NRRWF.

Upon work completion, core samples of screen materials were taken from different depths. In the selected samples, filtration, sorption and diffusion characteristics were identified: hydraulic permeability factor was found to be ranging from 10^{-10}—10^{-11} m/s, the diffusion coefficient of tritium amounted to some 10^{-10} m^2/s and uranium sorption distribution factor amounted to 10—10^3 ml/g [17], which corresponds to the typical requirements set for barrier materials.

Arrangement of horizontal screens without excavation

Vertical barriers along structure perimeter are not always sufficient to provide adequate safety in case of radionuclide migration with groundwater flow. Backfilling of the free space inside a NRRWF with a buffer material and installation of a cover screen preventing the surface water from entering the storage system increases the load on the underlying layer potentially increasing its permeability and contaminant ingress into the soil and groundwater. Methods enabling excavation-free construction of horizontal engineered barriers at nuclear facilities are seen as quite promising in this regard.

To arrange for and restore the waterproofing capacities in the horizontal base plane of facilities and structures (underlying screen) occupying large space areas (for example, reservoirs, sludge collectors, etc.), horizontal directional drilling (HDD) method can be applied [18] along with clay mixture injection during reverse holing with a reamer.

This method combines the construction of pilot drilling channels arranged in a horizontal row using HDD unit with a high-precision navigation system followed by reverse injection of a waterproofing BentInject mixture into the pilot channels (Figure 4). Pilot channel trajectories are arranged in a way allowing to ensure 100% overlapping of adjacent horizontal wells during the injection and the continuity of the underlying screen. Such a screen has a similar waterproofing performance to the one of a vertical anti-seepage curtain made of overlapping soil-clay piles.

Internal barriers

Internal engineered safety barriers at NRRWF and DFRW are designed to avoid the seepage-driven migration of radionuclides or to reduce the contact time of RW with water, retain (absorb) leached radionuclides by eliminating available voids and free spaces [19].

Powders, dry mixes

Crushed clay with a particle size of up to 30 cm and a moisture content of 22—28 % is commonly applied in case of CCL mentioned above. A wider range of products: powders, grains, granules, etc. can be produced thanks to the application of bentonite processing methods.

Bentonite powder can be added to the drilling fluid, in particular, for wells pre-commissioning purposes since a thin protective mud cake is formed providing the stability of the walls. Bentonite can be also used to backfill the wellbore and underground cavities after well abandonment. Bentonite granules can quickly settle to the bottom in standing water: they swell putting pressure on the walls of the cavity, thus, providing a reliable sealing plug. This method was found to be effective in sealing water-filled boreholes at a depth of up to 500 meters [4]. Laboratory studies exploring the potential application of pressed bentonite for deep-well sealing purposes are currently underway in the UK[1].

Extensive experience in the construction of additional internal engineered barrier was obtained during PUGR EI-2 decommissioning at PDC UGR site: a barrier mixture (82.0—89.5 % kaolin, 10.0—15.0 % bentonite and 0.5—3.0 % vermiculite) was poured into the reactor space [20]. Kaolin was chosen as a barrier material base due to its property noted under laboratory conditions: it becomes firmer under its own weight after being saturated with water.

For any clay powder, the initial bulk density of its skeleton would range from 0.9 to 1.1 g/cm^3. Compaction, according to [21], reaches a level of up to 1.6—1.7 g/cm^3 for a kaolin-based barrier mixture versus 1.3—1.4 g/cm^3 in case of 100 % bentonite. Given the resulting densities, the hydraulic permeability factor for a kaolin-based mixture is only an order of magnitude higher (10—11 m/s) than the one for bentonite (10—12 m/s). Such high self-compaction capacity of kaolin prompts barrier material shrinkage and requires staged backfilling to eliminate the available voids before the NRRWF conservation is completed. In this regard, not only is it a more time-consuming option, but it also requires greater amount of barrier material and, thus, increases the cost of operations.

Such degree of compaction is not typical for such swelling clays as bentonite, i. e., upon its contact with water, it swells filling all the free space, cracks, voids. Even in case of a lower density, its hydraulic permeability remains rather low (10—12 m/s). The case of bentonite swelling with its pressure acting on the NRRWD structures was considered in [22]: at a skeleton density of up to 1.45 g/cm^3, the swelling pressure was found to be lower than 1 MPa and the strength of the structure could not be essentially affected by it.

Injection

Injection technologies can be used to arrange and restore the insulation properties of buried structures preventing their degradation due to groundwater flow, as well as possible ingress of radionuclides into them. This issue can arise during the operation of buried structures used for various purposes, including the facilities holding non-retrievable SRW, storage tanks for LRW upon liquid phase removal.

BentInject can be pumped both from the inside of the structure through injection holes (Figure 5) and from the outside through submersible tubular injectors. No excavation is required to restore the insulation properties using the injection method. These operations can be performed under rough conditions and in a constraint environment of a construction site.

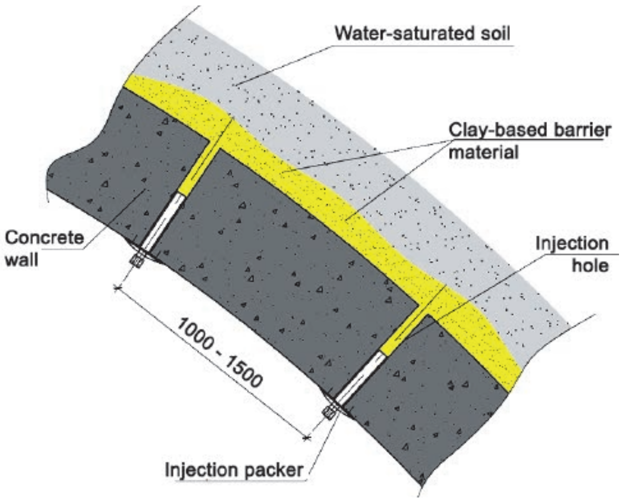

Figure 5. Layout of an insulation method involving contour injection based on BentInject composition

At the initial hydration stage, the material has a fluid-plastic consistency, thus, enabling the application of the injection method. Then it thickens to a stable state of a rigid-plastic consistency. In this state, its hydraulic permeability factor would not exceed $1\cdot10^{-10}$ m/s.

Advantages of contour injection isolation method assuming the application of clay solutions can be summarized as follows:

[1] https://www.iom3.org/resource/testing-starts-on-bentonite-system-to-seal-deep-boreholes.html

- further soaking of concrete structures is avoided;
- minimal corrosion of reinforcement and concrete structures;
- vibration resistance of the resulting waterproofing layer;
- repair and operation of the waterproofing layer is possible (re-injection into pre-arranged injection nests);
- injection mixture material is neutral to the structure concrete;
- mixture material is considered environmentally friendly (no soil and groundwater contamination).

Conclusion

The article overviews both promising technologies and clay-based materials and those that have already gained wide application in enhancing the safety of NRRWF and DFRW providing radical improvement of RWDF waterproofing capacities. In particular, the paper considers the characteristics and options suggesting the application of BentInject bentonite composition that has been developed to eliminate possible leaks in underground structures and to arrange for impervious curtains at NRRWF sites.

The discussed technologies providing the establishment and restoration of NRRWF engineered safety barriers during their conservation and upgrading to RWDF, RWDF construction and closure are considered as quite mature (TRL 7-9). Introduction of these technologies to the nuclear industry assuming their application at nuclear legacy facilities will contribute to better hydrogeological conditions and higher radiation safety level at the sites of operating organizations, such as SCC, NZKhK, CMP, MCC, PA Mayak and others.

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Information about the authors

Ilyina Olga Alexandrovna, Director of Science, Bentonite Company Ltd (office 24, 12/1 Tverskaya st., Moscow, 125009, Russia), e-mail: ilina@bentonit.ru.

Lundin Dmitry Sergeevich, Deputy commercial director for service, BentIzol Ltd (14, Promyshlennaya st., Vvedenskoye set., Ketovsky district, Kurgan region, 641321, Russia), e-mail: lundin@bentizol.ru.

Proskurin Denis Vladimirovich, Head of the service of drilling fluids, Bentonite of Khakassia Ltd (11A, Maxim Gorky st., Chernogorsk, Republic of Khakassia, 655162, Russia), e-mail: proskurin@b-kh.ru.

Vedernikova Marina Vladimirovna, PhD, researcher, Nuclear Safety Institute of the Russian Academy of Sciences (52, Bolshaya Tulskaya st., Moscow, 115191, Russia), e-mail: vmv@ibrae.ac.ru.

Biryukov Dmitry Viktorovich, Junior researcher, Nuclear Safety Institute of the Russian Academy of Sciences (52, Bolshaya Tulskaya st., Moscow, 115191, Russia), e-mail: biryukov@ibrae.ac.ru.

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