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USE OF NATURAL CLAY MATERIALS TO INCREASE NUCLEAR AND RADIATION SAFETY OF NUCLEAR LEGACY FACILITIES

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The paper studies the composition, structure and properties of clay minerals being considered as engineered safety barriers components providing the isolation of nuclear legacy facilities. These considerations may provide a basis for the Program aimed at development and fabrication of clay mineral compositions to ensure the safety of different nuclear facilities.

Key words: *clay materials, nuclear legacy, isolation of radioactive waste, engineered barriers.*

Introduction

The wide scope of clay and clayey material useful properties has been known for a long time providing for their massive production and a variety of different applications. It is also well known that omitting multiple specific features of clayey materials associated with raw material properties and the treatment methods inevitably results in the underuse of their high potential or can cause even more serious problems.

Thus, incomplete knowledge on clayey material properties along with disregarding proper treatment and production methods and technologies used in the fabrication of barrier systems has manifested itself to a fullest extent possible in the nuclear legacy management and in general while performing certain environmental activities on the elimination of nuclear accidents. It was following the Chernobyl accident that clayey materials were most intensively applied to prevent radioactive contamination of water supply sources and surface waters, in particular, the Dnepr River. Water protection measures implemented then, involved the construction of so-called filtering curtains acting as barriers tying up radionuclides and preventing their release into open hydrological network. Unfortunately, the use of clayey materials failed to produce the expected result in some cases even contributing to unfavorable environmental impacts when the nearby forested areas had been flooded [1].

As it comes to nuclear legacy facilities with relevant designs suggesting the use of clayey materials in safety barrier construction, B-25 LRW storage reservoir at FSUE SCC deserves particular attention. Investigations performed in 2003 concluded that the impermeable screen had lost its sealing capacity.

This case has required the development and implementation of additional measures preventing radionuclide release from this radioactive waste (RW) storage facility.

However, best international practices (USA, France, Sweden) suggest the wide scope of clayey materials applications both under nuclear decommissioning projects [2, 3] and construction of RW storage and disposal facilities.

The translation is done by N. S. Cebakovskaya

Application of clay and clayey materials

Clay being applied as engineered barrier material is essential in terms of nuclear safety and may be used:

- To construct barriers with predefined high sorption and low filtration properties to prevent radionuclide migration during the implementation of programs aiming to ensure the long-term safe configuration of nuclear legacy facilities (term referred to as conservation in Russia);
- To construct barriers during the implementation of the ENTOMB decommissioning option;
- To ensure long-term safe storage configuration of RW disposal facility cells (compartments, vaults, sections), transportation tunnels and other auxiliary areas, construction of impermeable dividing walls;
- For RW conditioning as matrix material;
- To construct safety barriers at planned near-surface and deep disposal facilities for RW. The use of clayey materials as part of engineered barriers during the construction of RW storage/disposal facilities may enable:
- To limit ground water inflow to RW;
- To ensure such conditions suggesting that mass transport between waste and ground waters is possible solely via diffusion;
- To ensure effective radionuclide sorption in case if containers (canisters, drums and etc.) possibly lose their sealing capacity;
- To seal open fractures and large pores inside clay rocks due to their swelling.

The properties indicated above result in the wide scope of clayey materials applications in the construction of barrier systems performed to enhance radiation and nuclear safety of nuclear legacy facilities. Initially, during the development of the federal target program Nuclear and Radiation Safety in 2008—2015 (hereinafter, FTP NRS) the use of clayey materials was planned for some 40 nuclear legacy facilities. However, as the design documentation had not been elaborated to the necessary extent, a big number of activities scheduled under the FTP NRS were withdrawn from the plans. Nevertheless, a big scope of activities was performed also considering more elaborated designs for the use of clayey materials with much more comprehensive scientific evaluation of relevant safety aspects. This is the case of decommissioning activities performed at LRW storage facilities and production reactors started as far back as in 1990's. Most indicative is the case of activities performed at SCC and PDC UGR involving comprehensive R&Ds with the engagement of a large number of specialized organizations.

Table 1 summarizes most large-scale efforts on construction of additional clay-based safety barriers performed under FTP NRS [4].

In 2008 conservation of 354 RW storage reservoir was completed at MCC. Its isolation was achieved with the use of barrier system based on a composition of sandy clay, gravel and loam. As the result, thickness of the capping screen totaled some 8.5 meters.

In 2012, B-2 conservation project was fully completed involving the backfilling of its water surface with coarse materials followed by construction of a capping clay screen enabling the hydrauisolation. Construction of additional protective barriers preventing radionuclide migration by injection methods was developed and tested at B-2 and B-25 LRW storage facilities. This method suggests the use of liquid glass and was not provided for in the initial designs. Such injection barriers have been widely used in construction to limit the migration of contaminants and now saw successful application in nuclear industry [5].

2015 saw the completion of EI-2 production uranium graphite reactor (PUGR) decommissioning at PDC UGR site suggesting its entombment with the establishment of a storage facility for non-retrievable (special) RW. Safety of the facility was provided by existing and construction of additional safety barriers. As the result, a set of R&D activities was implemented aiming to develop and evaluate clayey based barrier system construction including material pretreatment and backfilling techniques. Materials considered for construction of additional barriers were specially designed clayey compositions used both for reactor shaft backfilling and construction of the capping screen [6]. Developed and tested technologies are proposed for the decommissioning of other PUGRs with due account of relevant geological, hydrogeological setting and sitting conditions.

Activities implemented under FTP NRS enabled to gain knowledge and knowhow on the closure of surface LRW storage reservoirs, PUGR decommissioning and construction of additional clayey-based safety barriers. However, despite the success achieved one distinctive drawback was revealed

Table 1. Summary of activities on construction of additional safety barriers suggesting the use of clayey materials

№	Facility	Site	Surface area, m^2	Type of constructed protective barrier	Used clayey materials, t
1	LRW storage facility 354	MCC	25	Sorption capping screen preventing radionuclide migration	160,000
2	LRW storage facility B-2	SCC	57.3	Sorption capping screen preventing radionuclide migration	105,600
3	PUGR EI-2	PDC URG	5.6	Sorption barrier and sorption capping screen preventing radionuclide migration	75,200 (including backfilled reactor shafts)

Table 2. Preliminary assessment of clayey material amounts required in the short-term perspective

Facility type	Number of facilities	Types of efforts involving clayey material use	Preliminary assessment of clayey materials required, thousand tons
Production reactors and at reactor RW storage facilities	24	Construction of sorption capping screen preventing radionuclide migration	800
Reservoir storage facilities	13	Construction of sorption barrier and sorption capping screen preventing radionuclide migration	11,000
SRW storage facilities (special structures and near-surface repositories)	> 30	Construction of sorption capping screen preventing radionuclide migration	500

for all these cases — inconsistency when it came to addressing some general issues with no proper understanding of what number of facilities would require the use of clay materials and what should be their composition, rate of production and marketing outlets.

Nuclear facility and RW inventory taking campaigns performed under FTP NRW enabled to obtain the required knowledge to address nuclear legacy challenges. Thus, for over 700 facilities relevant decisions were made on their decommissioning (demolition, in-situ disposal) or modernization to enhance their safety [7]. Almost 70 storage facilities may require the use of clayey materials to provide RW disposal in situ. There are some reasons to expect that the number of such facilities may increase significantly both as some new facilities are going to be added on the list, as well as due to the facilities for which relevant decision making had been postponed. Table 2 provides preliminary estimates of clayey material amounts that may be required in the short-term to implement planned nuclear decommissioning projects. These amounts can be significantly reduced due to the use of innovative materials and production approaches that are already available or soon become available in Russia. A brief overview of potential materials will be presented below.

The list of facilities that are going to be decommissioned by 2030 under ENTOMB option has already been drafted. These are B-25 water reservoir, tailings PKh-1,2, at SCC, water reservoir B-17 at PA Mayak, tailing № 1 at NZCC and etc. Besides, there are seven PURGs with at reactor RW storage facilities [4]. These activities are scheduled under Federal Target Program Nuclear and Radiation Safety in 2016—2030.

Thus, recent activities aimed at enhancing radiation safety of nuclear facilities have evidenced the perspectives of applying clayey materials for different purposes as elements of engineered barrier systems. However, construction technology and engineered barrier material composition should be chosen with due account of RW type and class, layout and geometry of the facility, as well as the geological and hydrogeological setting.

Large-scale efforts aimed to ensure the long-term safety of nuclear facilities require the development of protective engineered barriers preventing radionuclide release into the environment. Barrier systems should have necessary properties enabling safe and robust RW long-term storage whereupon all barrier system elements should remain stable during their entire lifetime.

Barrier properties of clays

Clay rocks and clay-based materials may have different granulometric and mineral contents. Materials containing no less than 30—50 % of fine-grained fraction (< 2—5 μm) depending on clayey material content are considered as most preferable materials for engineered barriers construction. Fine-grained fraction is mostly present in form of clayey minerals containing some feldspar, quartz and other minerals.

Only clayey minerals are characterized with some favorable properties in terms of engineered isolating barrier construction. Such materials can differ significantly by their properties which depends on their specific contents and structure. The following indicators are viewed as key characteristics of clayey materials seen as potential components for isolating barrier construction retarding radionuclide migration: filtration and diffusion coefficients, permeability, cation exchange capacity, sorption radionuclide distribution coefficients, kinetic parameters, quantity and nature of sorption centers, crystal size, specific surface area, swelling index and pressure and etc.

Clayey minerals are referred to phyllosilicate subclass, silicate class. According to the classification of International Mineralogic Association (IMA) they can be divided into two groups [8—10] based on tetrahedral and octahedral bonds present — 1:1 (TO) and 2:1 (TOT) and differ from each other by the number of isomorphous replacements defining layer charge. Clayey mineral structure defines their physical, chemical and mechanical properties. Figure 1 shows pictures of basic clayey minerals made by electronic microscope.

Structure of type 1:1 minerals is based on a bound between one tetrahedral and one octahedral net. Kaolin-serpentine minerals group is referred to this type, including: kaolinite, halloysite, lizardite, chamosite and others. Low isomorphic replacement rate inside layer structure typical for 1:1 type minerals results in its low capacity and sorption properties (table 3). Due to particular structure features, as well as to the fact that rather large particle aggregate formation is quite typical for this type of minerals (an average of 5—10 μm) kaolin clay rocks

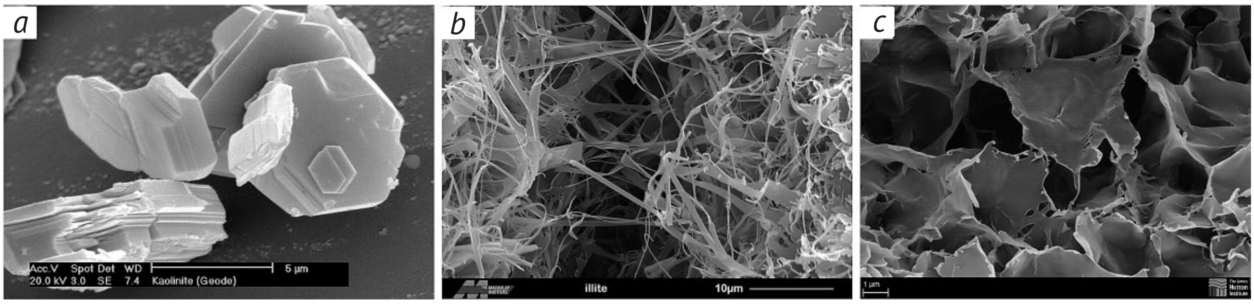

*Figure1. Electronic microphotographs of clayey mineral particles:
a) kaolinite, b) illite, c) smectite (based on http://www.minersoc.org)*

are characterized by low swelling capacity and higher water permeability (high diffusion).

Type 2:1 mineral structure is based on a layer composed of two tetrahedral nets with an octahedral one in between. Water cations and/or molecules are typically present inside this interlayer space (or simply interlayer). Despite similar layer structure of all layered silicates of type 2:1, interlayer composition may differ significantly depending on the nature and number of isomorphic replacements. This type includes groups of true micas (muscovite, biotite, phlogopite, etc.), mica with interlayer deficiency (illite, glauconite), smectites (montmorillonite, saponite, etc.), vermiculite (vermiculite), chlorite group (clinochlore, chamosite and etc.), previously referred to minerals of type 2:1:1.

4 groups of clayey minerals are considered to be most commonly occurring in natural clays, clay loams and other sedimentary rocks (table 3): mica minerals, smectite, kaolinite and chlorite. As it was mentioned above, it's the contents and the structure that defines physical, chemical, mechanical and other properties of materials potentially suitable for engineered barrier construction.

Mica clay physical and chemical properties most drastically differ from smectite ones [11 – 15]. Big number of isomorphous substitution and their predominant localization inside tetrahedral is typical for mica minerals (muscovite, illite and etc.). These result in calcium cation fixation inside interlayer gap with a rather big grain size of some 1 -5 µm, low exchange capacity, low swelling capacity and relatively high water permeability. Mica with interlayer deficiency (illites, glauconites) primary differs from genuine micas by lower number of isomorphous substitutions resulting in higher swelling capacity and dispersion.

Due to their particular layer structure and virtually total absence of isomorphous substitutions, kaolinites are primary characterized with big particle size, low cation exchange capacity, absence of swelling capacity and low water retarding properties. Chlorites are generally contained in insignificant quantities with no commercial accumulation.

Furthermore, kaolinites, illites and chlorites are often present as sedimentary rock components that may be potentially used as additives during clayey engineered barrier fabrication.

Unlike other clayey minerals, particular features particular features of smectite group minerals, primary montmorillonite (relatively low layer charge and isomorphic substitutions mostly confined to octahedral nets) provide its structure liability, inter-crystalline swelling capacity, as well as cation and anion sorption at basal surface and lateral shears. It also results in high swelling capacity, small crystal size, increased specific area and high cation exchange capacity. Thus, montmorillonite and clayey mixtures with high montmorillonite content are considered to be the best components when it comes to construction of sorption, impervious and migration retarding barriers enabling to ensure the safety of RW storage and disposal facilities, as well as other nuclear facilities.

Montmorillonite being the prime component of bentonite clays (70—90 %) is considered as a most perspective material for such nuclear applications. Previously it was deemed that bentonite clay reserves in Russia are low [16]. However, this assumption was proved to be wrong. Currently, bentonite raw material with different properties and montmorillonite content (from 60 to 99 %) is available at Russian market. Below are presented the main features of clay raw material deposits, including bentonite deposits.

It's also worth noting the minerals having stratified structure — palygorskites and sepiolites. Due to the high rate of isomorphic substitution, these minerals have rather high sorption capacity and due to some specific features in their structure no intracrystalline swelling typically occurs.

Quite similar structure typical for all clayey minerals results in formation of numerous mixed layered minerals (MLM) due to synthesis and transformations being part of geological processes occurring. Such MLMs may differ significantly from each other by content and ratio of interstratified packages and layers. This eminently results in a big variety of their properties. Table 3 provides only two examples of mixed layered minerals — kaolinite-smectites and illite-smectite that are commonly present as components of sedimentary rocks and soils. Moreover, MLM properties depend on their contents and primary on the amount of smectite package (layer) and the particular features of its structure.

Table 3. Contents and properties of main clayey mineral groups

Layer type	Group	Examples of minerals	Swelling capacity	Clayey rock with the selected mineral being considered as the main one	Sorption capacity as regards heavy metals and radionuclides	Filtration capacity	Big deposits	Potential of using for barrier construction
1:1	Kaolinites[1,2]	Kaolinite, dickite, nacrite	low	kaolinite	low	high	Yes, available in Russia	intermediate
		halloysite	low		intermediate	high	Yes, available in Russia	intermediate
2:1	Genuine mica[1,2]	muscovite, phlogopite, biotite	low	Polymer clay component	low	high	no	non
	Mica with interlayer deficiency[1,2]	illite, glauconite	low		low	High-intermediate	Yes, not available in Russia	non
	Smectites[3,2]	montmorillonite, beidelite, saponite	high	bentonites	high	low	Yes, available in Russia and CIS	high
	Vermiculites[4,2]	vermiculite	High (for dispersion differences)	vermiculites	High (for dispersion differences)	Intermediate – low (for dispersion differences)	Yes, available in Russia	intermediate
	Chlorites[1,2]	clinochlore, chamosite	low	polymer clay component	low	high	non	non
	Polygorskit*[5,2]	palygorskite, sepiolite	low	palygorskites	intermediate	High-intermediate	Yes, available in Russia	intermediate
1:1 and 2:1	Mixed-layer minerals (MLM)**[2]	kaolinite-smectite	Low - intermediate***	polymer clay component	Low - intermediate***	Low - intermediate***	non	non
		illite-smectite	Low - high***		Low - high***	Low - high***	Yes, not available in Russia	non

* – refers to nonplanar (not layered) phyllosilicates

** – for mixed layered minerals only, the examples of rocks mineral species most common for soils and sedimentary are given

*** – properties of mixed layered minerals differ significantly and depend on the contents and structure of components involved in MLM composition

Numbers indicate clays for which relevant minerals are important in terms of pore formation: 1 – kaolin clays, 2 – mixed polymer clays, 3 – bentonite clays, 4 – vermiculite clays, 5 – palygorskite or sepiolite clays

Palygorskite clays with commercial deposits being available in Russia (Moscow, Kaluga and Arkhangelsk Regions) also seem to be quite perspective in terms of their use for engineered barrier construction. These minerals are characterized with high sorption capacity for heavy metals and radionuclides. However, they do not show high swelling capacity and may be used as additives for sorption barrier construction.

Thus, due to clay material properties, bentonite clay — clayey minerals with high montmorillonite content are considered as most perspective base functional component to be used in engineered barrier construction. It should be additionally noted that clays typically contain organic substance having high sorption capacity comparable with the one of montmorillonite — 150—400 mg-equ/100 g, and high selective sorption capacity regarding a number of radionuclides, for example, uranium. However, presence of such organic matter may undermine the long-term stability of such composition due to microbilogical and radiological degradation. This should be considered during the design development.

For over 60 years, clay rocks have been widely used as natural screens – structures for RW final disposal facility construction. This could be explained by their low permeability and high retardation capacity for mobile radionuclides. For this reason, the use of clayey rocks or clay-based materials allows to consider them as highly effective retardation matter providing long-term isolation of RW. Due to low permeability, diffusion driven mass transfer is considered as the primary migration mechanism for saturated clay rocks and engineered barriers. Migration properties of bentonite screens in particular diffusive were investigated under a number of theoretical studies [17—19]. These studies were basically aimed at modelling diffusion driven transfer accounting for physical and chemical interactions between minerals and mobile agents. All models allowing to forecast radionuclide migration involve diffusion rates for absorbing and non-absorbing species, as well as relevant parameters determining physical and chemical interactions between solution and the rocks that strongly influence barrier performance. For this reason, performance assessment of clay rocks and engineered barriers should be based on experimental data.

Radionuclide chemical properties, clay material type and environmental setting defines the mechanisms by which radionuclides may interact with clayey minerals. Two key mechanisms can be indicated for the interaction of radionuclides producing cations in water-based solutions.

Cation exchange is considered as the first one — radionuclides interact via so called ion-exchange centers, the number and the type of which in turn depends directly on the structure of the clayey mineral in question. The total number of ion-exchange centers for a given sample determines relevant cation exchange rate. However, these centers may include those having high or contrary low selective ones as regards the radionuclide in question. Ion-exchange mechanism is typically common for weakly hydrolysable cations such as alkaline and alkaline-earth elements (^{90}Sr, ^{137}Cs and others). This mechanism is also considered to be common for actinide and lanthanide cations but only in case of relatively low salt content of the solution and its low pH level.

The second mechanism refers to so called edge (peripheral) centers. Such interactions typically result in formation of intraspheric complexes, thus, are characterized with low dependency on ionic strength and pH values. This mechanism is basically common for actinide ions. It should be noted that the number of such peripheral centers do not correlate with cation exchange capacity rate.

Thus, experimental verification of migration parameters seems to be required for the performance assessment of clayey materials being considered suitable for the construction engineered barrier in RW repositories. Such migration parameters involve those responsible for mass transfer through barrier materials (diffusion rate, filtration rate, effective porosity), as well as those associated with the interaction between mobile radionuclides and barrier (cation exchange capacity, crystal size, specific area, concentration of different type sorption centers, sorption distribution rates). Such experimental parameter studies associated with ion exchange parameters should be accompanied by those aiming to investigate diffusive transfer parameters. The latter one should involve samples, as well as existing or newly formed aggregates with inner diffusive kinetics of exchange degrade under normal statistical experiments that increases real (effective) specific surface of exchange.

Barrier material may be manufactured based on one or several natural occurring materials with certain additives introduced to attain the required properties. Clayey materials used for engineered barrier fabrication may be present in form of lumps delivered directly from pits or following some preliminary treatment – plastic deformation, drying, grinding, palletization, sodium activation enabling to achieve better and some additional favorable properties in keeping with relevant requirements to granulometric content, dry density and etc.

Natural bentonite clays are considered as most effective material characterized with a full set of desired properties in terms of radionuclide migration retardation and permeability. At some nuclear sites, bentonite mixtures with sand (common international practice) may be used to enhance the economic attractiveness of this technology with due consideration of relevant safety requirements. "Sand" component introduced to decrease the cost of barrier material may involve some local polymineral clays, sandy loams, loams and soils.

Clay material deposits in Russia and CIS countries

High-quality bentonite reserves currently present at Russian market are quite large: over 10,000,000 tons of Na-Ca bentonites in the Republic of Khakassia involving 3 deposits (hamlet № 10, Karasuksk, Bentoysk), 22,000,000 tons of Ca-Mg bentonite in the Kurgan Region (Zyryansk), over 9,000,000 tons of high-quality bentonites in Republic Kazakhstan (Tagansk deposit), 86,000,000 tons of natural occurring Na-bentonites in Azerbaijan (Dash-Salakhlinsk). The quality of bentonite raw material is ensured by the content and specific features of the main valuable component – montmorillonite [20]. Montmorillonite content in bentonite clay can vary from 65 % (Zyryansk deposit) to some 75—80 % (deposits hamlet 10 and Dash-Salakhlinsk) and even up to 98 % (certain mineral occurrences). Bentonite raw material is produced selectively in pits enabling to obtain the product of a given quality. Figure 2 provides a map of main deposits currently being exploited at the territory of the Russian Federation and CIS countries [21—26]. Thus, it can be firmly stated that bentonite reserves of different quality are more than enough for Russia to address relevant RW management issues.

It's known that to reduce the cost of barrier materials natural or enriched materials can be used as additives with high capacity (vermiculites, ceolites), high density for better backfilling (kaolines and kaoline similar clays), as well as polymineral clays (basically referred to the category of fire-resistant clays). There is only a few number of vermiculite deposits in Russia with relatively low-quality raw material. Most well-known is Potaninsk deposit (Chelyabinsk Region) with balance reserves of category C1 amounting to some 3,200,000 tons and Tatarsk deposit located in the North-Yeniseisk area of Krasnoyarsk Region with category C1 reserves amounting to 1,700,000 tons. Vermiculite content therein averages to some 45 % and this figure can vary much due to specific geological conditions associated with the formation of this deposit. Vermiculite boating technique (baking in special ovens at a temperature of 1,000 °C) widely used in case of other applications cannot be used to produce engineered barrier mixture — as under these conditions, vermiculite loses its capability to retard radionuclide migration and prevent water infiltration due to irreversible alterations in the mineral structure.

Kaolin clays are currently used in a number of Russian projects as main barrier mixture component [6]. However, future projects should account for good bulk density being considered as a favorable property when it comes to barrier mixtures used in the decommissioning of facilities with complex structure (for example, uranium graphite production reactors). On the other hand, it should be noted that they may not ensure sufficient radionuclide retardation and ground water flow reduction.

Figure 2. Overview map of bentonite clay resources available in Russia and some of the CIS countries.

List of deposits: 1 — Biklyansk, Berezovsky and others (Tatarstan); 2 — Zyryansk (Kurgan region); 3 — 10th Hamlet (Khakassia); 4 — Izhberdinsk, Saraybashsk and others (Orenburg region); 5 — Gerpegezhsk, Nalchinsk (Kabardino-Balkaria); 6 — Kalinovo-Dashkovsk (Moscow region); 7 — Nikolsky, Maidan-Bentonite, Podgornoye (Voronezh region); 8 — Lubinsk (Omsk region); 9 — Kamalinsk (Krasnoyarsk Territory); 10 — Tarasovsk, Millerovo (Rostov region); 11 — Tikhmenevsk, Vakhrushevsk. Makarovsk (Sakhalin region); 12 — Zerkalnoe (Primorsky Territory); 13 — Urgalsk (Khabarovsk Territory); 14 — Dash-Salahlinsk (Republic of Azerbaijan); 15 — Tagansk, Dinozavrov (Republic of Kazakhstan)

Bulk density of bentonite clay-based materials may be enhanced by introducing some new products that will become available in Russia in the near future. Primarily this refers to compacted granules (pellets) of clean bentonite or bentonite with different content of sand which is being considered under some European projects [27]. Several large and medium size kaolin clay deposits are being exploited in Russia. These are located in the Chelyabinsk Region (most well-known are Kyshtymsk, Eleninsk, Zhuravlinyi Log and Poletaevsk) holding a total of 35,000,000 tons of clay referred to categories A+B+C1 and some 20,000,000 tons of C2 category clay. Reserve grade (kaolinite) accounts for some 65—85 %. These clays are mainly used in faience industry, paper blanching and etc. and are not considered as a cheap clay raw material.

Finally, in general, fireproof polymineral clays are to larger extent polydisperse clays and may have heterogenous content (illite, kaolinite, chlorite, and usually insignificant quantity of smectite may be present), have relatively low sorption capacity and high filtration rate. Polymineral clay reserves are quite abundant in the European part of Russia and in Siberia. Decisions regarding the exploration of such deposits are made based on accessibility and feasibility evaluations. Such clays are mainly used in construction, for example, in road construction. Thus, for example, 3 deposits of fireproof clays with a total balance reserve of B+C1 clay category accounting for 205,000,000 tons and 224,000,000 tons of C2 clays are being exploited in Chelyabinsk Region.

Thus, Russian clay material reserve estimates show that these are quite available and that barrier mixtures with given properties can be designed and manufactured. Variations introduced to their contents may provide for the most economically feasible options.

Perspectives for development and manufacturing of clayey materials for engineered barrier construction

Nuclear facilities (for example, large facilities such as LRW storage reservoirs, nuclear fuel cycle facilities, production uranium graphite reactors and etc.) have different characteristics. Due to this reason new mixtures should be developed and some changes introduced to already existing sets of barrier mixtures. At present time, development and/or elaboration of standard clayey mixture compositions seems to be a most topical task. Properties of such mixtures will be tested in independent laboratories in Russia and abroad, and verified under joint research projects involving leading international experts.

It should be noted that decommissioning of complex facilities (production uranium graphite reactors, LRW storage reservoirs) carried under FTP NRS has required to address a number of methodical, scientific and engineering issues associated with the selection of most suitable barrier materials. Based on the knowledge accumulated to date on engineered barrier construction in the State Corporation Rosatom and specialized institutes including those of the Russian Academy of Sciences, effective and feasible barrier clayey compositions should be developed to ensure isolation of different nuclear facilities.

Efforts implemented in the nuclear legacy management should directly address existing and future tasks associated with RW storage and disposal facility safety. Based on safety requirements specified for relevant nuclear facilities and system approach, compositions of clayey materials should be developed. These materials should have given properties associated with permeability and sorption.

Since decommissioning efforts were launched in 1990 the list of materials available at Russian market has changed drastically. Besides powder materials, some new ones can be produced with the use of modern technologies — pellets, bricks and disks having various forms and manufactured from compacted bentonite or bentonite-sand mixture.

Bentonite-sand mixtures allow to design feasible compositions of compacted clays with given properties. Advantages of pellet applications are being evaluated under a number of European projects [27] allowing to address some complex engineering issues. Pellets (granules) varying in size and properties are placed between RW packages, as well as between packages and rock surface ensuring multiple favorable properties including high bulk density with high sorption and radionuclide retardation capacity. Pellet (granule), brick, disk properties, as well as clay-sand rate may vary to ensure the desired material properties.

Bentomats (geotextile) (figure 4) are viewed as perspective materials in terms of nuclear facility safety. Variations are possible in material composition and the thickness of bentonite granule layer inside the material to ensure the desired properties [28].

Bentonite mat is a coiled geosynthetic material designed to ensure hydrological isolation, to prevent contaminant release into soils and ground water, as well as to protect the structures from moisture impacts. Operating principle is based on the fact that montmorillonite can swell significantly under saturation conditions. In limited space area such saturation results in a macro molecular structure having solid body characteristics — gel with low water permeability, elasticity and plasticity. Bentonite mat is a flexible and durable needle structure made of polypropylene fibers with natrium bentonite granules homogeneously arranged inside.

To carry out experiments under barrier mixture design and development projects, scientific and research center (laboratory) should be established. Moreover, industrial stakeholders not being associated with the State Corporation Rosatom should be engaged in this work ensuring the development of

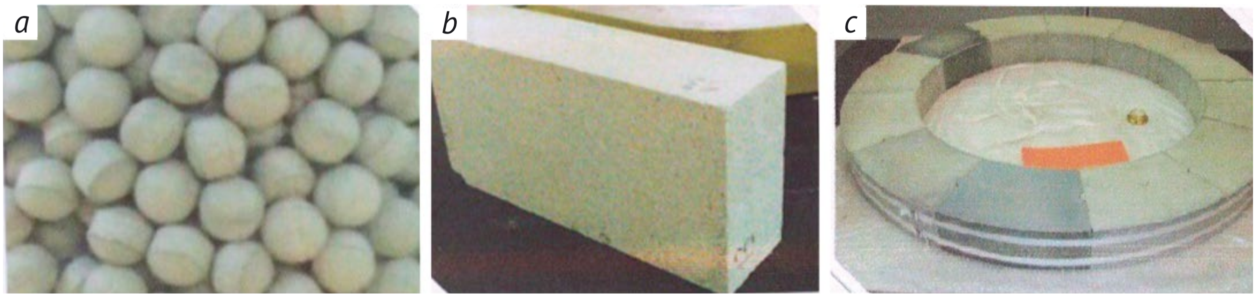

Figure 3. Examples of products manufactured from bentonite clays and bentonite-sand mixtures

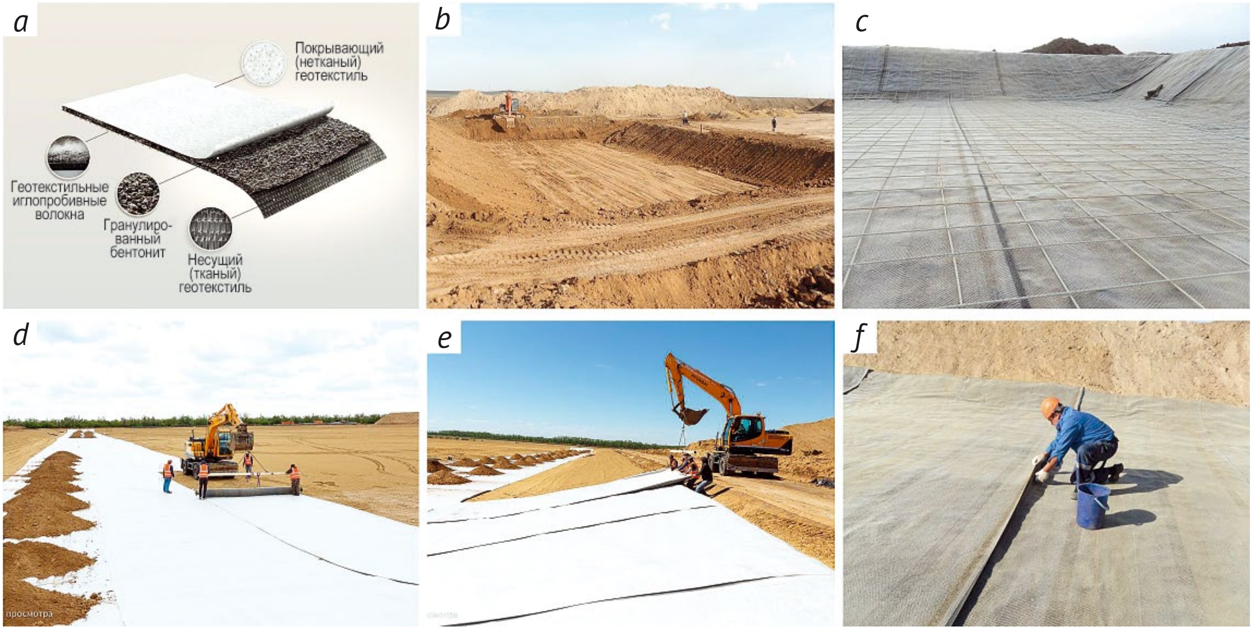

Figure 4. Layout of bentomat (a) examples of its application to ensure isolation of household waste landfills (before geotextile installation (b) after geotextile installation (c)) examples of different layouts (d, e) and manual installation (f) during impervious barrier construction. Materials were presented by JSC Bentonite Company

a process flow enabling grinding, enrichment, mixing of different clayey materials and etc. To enable adequate and independent estimates and to check the compliance of the developed mixtures with relevant safety requirements, expert council involving independent specialists working in different subject areas should be established.

Conclusions

Based on the facts mentioned above it's worth to conclude that the establishment of a program for the development and manufacturing of clayey material compositions ensuring the safety of different nuclear facilities is considered quite a topical challenge for the moment. Just a decade ago the development of an industry wide program on clayey materials seemed quite a premature challenge to be addressed. At present time this problem is considered as a quite urgent one that can be addressed under RW management scope of challenges. The program should be based on the following aspects:

- Identification of existing nuclear facilities requiring clayey material application;
- Taking the inventory of facilities potentially requiring such application;
- Evaluation of previously conducted activities in this field;
- NO RAO plans suggesting the development of very-low level waste disposal systems directly at industrial sites;
- Development of requirements on material property indicators depending on their purpose and future applications;
- Development of new hi-tech clayey materials together with industrial companies;
- Estimating available reserves of clayey materials and existing plans of other industries and national economy sectors, including those associated with the management of highly toxic chemical and household waste;
- Estimating the contents and properties of clayey materials and different clay-based compositions defining their sorption, filtration, diffusion, retardation, colloid and other indicators required

to estimate their potential applications in nuclear industry;

- Estimating critical conditions and given properties of clayey materials enabling to ensure the safety of radiation hazardous facilities;
- Development of a required set of industry-wide standards and, if necessary, safety guides.

According to present date knowledge, the Program on the investigation of clayey materials aimed at enhancing the safety of nuclear facilities should include several stages and involve the following areas of research:

1. Evaluation of all the knowledge on clayey material applications in construction and environmental remediation projects;

2. Evaluating the contents and properties of clayey raw material being available in Russia. These investigations should primarily address the contents and basic properties of clays (filtration, diffusion, sorption, colloid and etc.);

3. Investigating basic properties of clayey materials: behavior during cation and anion sorption, swelling pressure, filtration and diffusion processes, stability under aggressive conditions.

4. Investigating radionuclide behavior in natural clays and their compositions: sorption and desorption, radionuclide migration, the effect that impurity content and amount may produce on sorption mechanisms and parameters as regards certain radionuclides, particular features of colloid formation process for different clayey materials and their impact on radionuclide migration.

5. Development of key parameters covering clayey material structure and properties ensuring their use as certain barrier materials.

6. Experimental and numerical modeling of clayey material interactions with other barrier materials present in nuclear facilities: interaction with concretes, cements, steel and other under similar to storage/disposal facility conditions with due account for pH-Eh values, temperature, pressure and etc.

7. Development of natural clayey material inventory based on which further elaboration of different clayey compositions can be made.

8. Development of clayey materials compositions with given properties. Development of guidelines for design development organizations providing necessary terms of reference on clayey material compositions.

9. Establishment of an expert council ensuring the independent assessment of clayey material composition properties before and after their application in projects associated with RW isolation.

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