# RADIOACTIVE WASTE CEMENTATION VIA THE IN-CONTAINER HOT PRESSING METHOD

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*The paper explores mechanical, physical, chemical and structural properties of cement compounds based on blastfurnace slag produced in metal containers using the hot isostatic pressing (HIP) method. The study allowed to evaluate the radiation resistance of cement compounds under the influence of ionizing radiation up to an absorbed dose of $10^9$ Gy and $10^{19}$ alpha-dispersion/g. The tests confirmed that the matrices complied with the established regulatory requirements. It was demonstrated that cement compound synthesis based on the HIP method provided matrices with enhanced physical and chemical characteristics. It was also shown that under radiation exposure, cement compounds containing high-level waste simulators retained their properties and structure.*

**Keywords:** *radioactive waste, cementation, hot pressing, metal container, compressive strength, structure, radiation resistance.*

## Introduction

The cementation method has been developed to condition intermediate- and low-level waste (RW). Most of scientific and technical papers devoted to this method were published in the period from 1980 till early 2000's. These studies summarize the research on the properties of cement compounds and process parameters relevant for different RW streams, binders, additives. The studies were focused on salt water solutions, pulps of ion-exchange materials, sediments, sludge, liquid organic RW, ash from combustible waste incineration and other liquid and solid RW (LRW, SRW) [1]—[6].

Almost all common construction and purposely-developed cements and their applications were studied: normal- and fast-hardening Portland cements, high-strength, with additions of blast-furnace granulated slag, fly ash, limestone, microsilica, burnt slates, pozzolans, various composite, slag-alkaline, aluminous, magnesian, binder materials based on salts of potassium, magnesium, aluminum, chromium and gypsum.

A few mineral and artificial bulk and liquid additives were studied separately, as well as multicomponent and multifunctional additives providing various effects: strengthening, accelerating or slowing down the setting and hardening processes, increasing density, corrosion resistance, water resistance, reducing radionuclide leaching, etc.

Concrete produced from basic construction cements is widely used as a structural material at nuclear facilities subject to high-level radiation exposure (nuclear reactors, storage facilities for high-level spent ionizing radiation sources, RW containers, etc.) providing high-performance characteristics considered important for the operation of such facilities.

The research findings indicate that the cement compound can withstand high-level radiation exposure and can be potentially applied as a waste form for high-level waste (HLW) immobilization purposes [7]—[9].

Ionizing radiation emitted by the radionuclides contained in the cement waste form can alter its properties. Two possible types of radiation damage are considered: atomic displacement caused by particle interaction and chemical effects due to radiolysis and destruction of molecules. Most probably at low concentrations of alpha emitters in the waste, atomic displacements will not produce any significant degradation effect. Radiation exposure will mainly result in gas formation due to radiolysis. Radiation exposure causes hydrogen release from cemented RW with oxygen being absorbed. The gas will produce only some negligible mechanical effect on the integrity of the cemented RW since the gas permeability of the cement compound is high. Both water in hydrate neoplasms and unbound pore water can undergo radiolysis in it.

RW conditioning by cementation method is used widely in the world since it results in a final product of a required quality and does not involve any high capital and operating costs.

High efficiency is provided by maximum inclusion of waste into the cement compound and a feasible reduction of the composition options, treatment methods and types of equipment involved. There are many waste types the cementation of which is considered challenging and requires the application of some special techniques. Without proper pre-treatment and application of appropriate compositions and equipment, it appears difficult to implement the cementation process and the waste can be included into the final product in small quantities only. It seems feasible to reduce the volume of conditioned RW to a minimal level validated both from an engineering and economic perspective.

It can be argued that by now the RW cementation process has been studied well, therefore, any problems associated with RW conditioning can be addressed. Areas for further optimization of cement compositions, relevant methods and RWs amounts have been identified.

The research and engineering advances geared to the practical application are based on a system of standard requirements for the final form of the cemented RW: their fulfillment provides the preservation of their primary physical and chemical properties and integrity during waste management, transportation and storage for the period required for the contained radionuclides to decay to a safe level. RW conditioning process involves production of waste packages suitable for safe storage and (or) transportation, and (or) disposal. Process safety is also ensured by the quality of the containers applied [10].

Russian national standard GOST R 51883–2002 establishes general technical requirements for the cemented waste form produced by means of incorporating liquid low- and intermediate-level waste into waste form compositions based on inorganic binders. The standard also applies to cement compounds containing ash from RW incineration, and, in fact, establishes minimum levels for the characteristics of cemented waste forms considered sufficient for further safe RW management and storage.

GOST R 51883–2002 primarily imposes requirements on the mechanical compressive strength of cement compounds, which should be not less than 5 MPa and should provide the preservation of their properties after being exposed to an absorbed dose of up to $1{\cdot}10^6$ Gy and subject to frost resistance testing and prolonged exposure to water. The mechanical strength (compression) of cemented waste is seen as a most important characteristic.

The following regulatory requirements have been set forth for high-level waste: the package as a whole should have a mechanical compressive strength of at least 10 MPa, for solidified waste — 9 MPa, with the strength retention provided after a radiation exposure to an absorbed dose of $1{\cdot}10^8$ Gy for beta-/gamma-emitters and up to $10^{19}$ alpha disp./g [11].

Mechanical strength is seen as the main quality criterion for the cemented RW. Usually, the higher it is for the compound, the higher is its resistance to leaching, radiation exposure, prolonged exposure to water and frost resistance. Strength of a cement stone largely depends on its porosity, which is the one that basically affects these characteristics.

Porosity level basically depends on the water amount involved in the cement slurry production, i. e. the water-cement mass ratio (W/C). Its role is not limited to the hydration development of the cement compound structure: it is also responsible for the resulting viscosity of the cement slurry (spreadability), the level of which should be adequate to provide the workability of the mixture and cement mortar mixing, its unloading from the mixer and further transportation. Usually, about 30 % of water by the cement weight (W/C = 0.3) is required to provide complete hydration of Portland cement. For ordinary Portland cement, the water-cement ratio applied during the cement mortar production resulting in a required viscosity level is 0.5—0.7.

For some binder systems, this figure appears to be much higher. Free water, not bound by hydration, remains in the compound and forms a pore structure. Literally, excess water added for water hydration purposes reduces the cement compound quality.

When it comes to HLW cementation, one should note that the higher is the water content in the cement compound, the higher is the hydrogen release due to radiolysis. Some papers [12]—[15] emphasize that it is the unbound (pore) water that is mainly affected by this process occurring in concrete. Some researchers propose to dry the hardened cement compounds to remove it, thus, reducing the hydrogen release. The method seems unlikely to have any practical application, since the drying process would cause the degradation of the hardened hydrate structure. Moreover, it would be a quite challenging task to dry 100—200 liter-cement blocks and even more so in case if the block volume amounts to 1—2 $m^3$.

In case of HLW conditioning, one may improve the properties of the cement waste form containing RW of various morphological compositions, increase their content in the final product by 2—3 times compared to commonly recognized methods and reduce the hydrogen release due to radiolysis by compacting the waste form structure, which may be achieved by reducing the pore volume and the free water content.

To achieve this goal, a promising RW immobilization method was proposed, namely, hot pressing (HP) in a sealed capsule with a reduced W/C ratio. Waste form production by the HP method involves simultaneous mechanical pressing and heat treatment (steaming at elevated water vapor pressure) of the cement mixture (cement, additives, water, RW) in a sealed metal capsule (container) providing compaction of the material during HP treatment and acting as an additional barrier preventing the spread of radioactive contaminants. This process and its parameters (pressure, temperature and processing time) are adjusted in a way to provide the maximum density and strength of the cement compound.

The HP method enables the production of cement waste forms with much lower W/C ratio, thus, providing their high density and impermeability. Nevertheless, the flowability of the cement mortar gets lower. However, since the cement compound is produced directly in a sealed container, high flowability is not required. Water vapor, occupying the entire volume, provides cement hydration and formation of a cement waste form, as well as a uniform distribution of salts and radionuclides in it.

To study the HP method in a sealed capsule, we have considered two pressing methods: in a heated mold and isostatic pressing.

The study was seeking to provide a comprehensive assessment of the cement compound properties produced by the hot-pressing method in a container and to explore the radiation exposure impact on its properties and structure.

## The experimental part

At the first stage, the isostatic pressing method was applied to produce the cement compounds — barothermal treatment of capsules with compressed gas implemented in purpose-designed high-pressure installations and at elevated temperatures. A slag-alkaline binder mixture was chosen as the object of the study (Table 1): ground blast-furnace granulated slag from OJSC Mechel (Chelyabinsk region), bentonite clay (5 wt. % to slag) and various aggregators (NaOH aqueous solution, LRW simulator with a salt content of 545 g/l) [16].

***Table 1. Composition of cement compounds***

| Composition | Binder | Grouting fluid | W/C |
|---|---|---|---|
| 1 | Sludge | NaOH solution | 0.1 |
| 2 | | | 0.3 |
| 3 | | Mock-up RW | 0.1 |
| 4 | | | 0.2 |
| 5 | | | 0.3 |
| Mock-up RW, g/l: NaOH 197; $NaNO_3$ 209; $NaNO_2$ 108; $NaHCO_3$ 18; $K_2Cr_2O_7$ 11; KCl 2 | | | |

Hydration and hardening of such a binder system in a strongly alkaline medium resulted in a three-dimensional geopolymer aluminosilicate structure [17]:

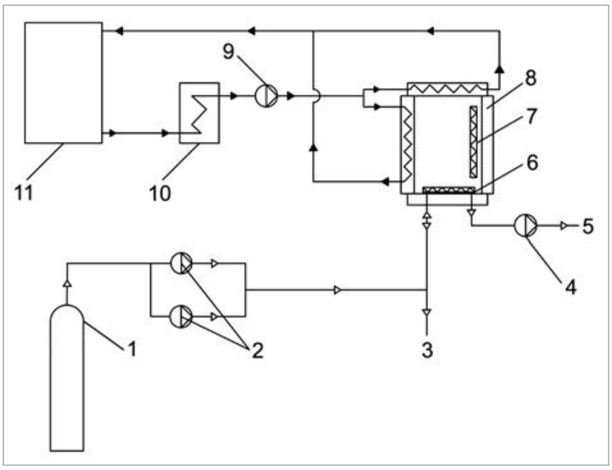
$$M\cdot\{-(Si-O)z-Al-O\}n\cdot w\cdot H_2O, \qquad (1)$$

where M is K, Na or Ca atoms or cations;
$n$ is the polycondensation degree;
$z$ is equal to 1, 2, 3 or more.

The material structure is formed by $[SiO_4]_{4-}$ and $[AlO_4]_{5-}$ tetrahedra interconnected by oxygen bridges. During the synthesis process, silicon and aluminum atoms form strong branched Si—O—Al—O chains with two- and three-dimensional structures, therefore, in terms of physical and mechanical properties, geopolymers do not seem inferior to the rocks. Alkali and alkaline earth metals are parts of the crystal lattice, which is important during RW solidification: they firmly attach such radionuclides as cesium and strontium. The essential advantages of geopolymers are seen in their high strength, density, water resistance, heat, thermal and corrosion resistance.

To produce cement waste forms by hot isostatic pressing (HIP), an ABRA HIRP 1984 HIP unit (Switzerland) (Figure 1) having the following characteristics was used: maximum operating temperature — 1400 °C; maximum working pressure — 200 MPa; heating rate — 15 °C/min.

*Figure 1. HIP unit ABRA HIRP 1984*



1 — argon storage tank; 2 — high pressure compressors; 3 — argon discharge into the atmosphere; 4 — vacuum pump; 5 — air discharge into the atmosphere; 6 — bottom heater; 7 — side heater; 8 — gasostatic extruder; 9 — recirculation pump; 10 — radiator; 11 — cooling recyclable coolant

*Figure 2. HIP unit layout*

Figure 2 presents the layout of the unit.

In the study, the HIP treatment provided for the use of two cylindrical capsule types: the first one being 65 mm in its diameter and up to 200 mm high; the second one with a diameter of 350 mm and a height of up to 300 mm. The wall thickness of the capsules was 4 mm. The capsule was sealed by welding immediately after the mixture was loaded. HIP was implemented at a temperature of 300 °C, a pressure of 100 MPa and an exposure time of 3—4 hours. Upon the process completion, the volume of the first and second type capsules decreased by 7 and 30 % respectively (Figure 3).

*Figure 3. Changes in the metal capsule volume following HIP treatment*

The capsules were opened 7 days after the HIP was completed. The cement compound was removed (Figure 4) with the-shaped (side dimensions equal to 20 mm) and beam-shaped (10 × 10 × 30 mm) samples being cut off (Figure 5).

*Figure 4. Cement compound produced by HIP method*

*Figure 5. Cube-shaped cement samples*

The following mechanical, physical and chemical properties of the obtained compounds were measured: compressive strength after 7, 14, 28 days of hardening; frost resistance; resistance to prolonged water exposure and the radionuclide leaching of radionuclides after 28 days.

To measure the mechanical compressive strength, German testing machine (Testing Cybertronic Cyber-Plus Evolution) was used, frost resistance was evaluated by periodic freezing-thawing of samples using a REOCAM TC-64 heat-cold test chamber (Russia). To evaluate the water resistance, the compounds were placed into bottles filled with water until their complete immersion and kept in water for 90 days. After that, the compressive strength was measured.

To assess the radiation exposure impact on the properties and structure of cement compounds, their mechanical, physical and chemical parameters, phase composition and microstructure content were measured before and after the radiation tests.

To study the phase composition and microstructure of cement compounds, X-ray phase analysis (XPA) was implemented, as well as an analysis involving a scanning electron microscope.

Cement samples were exposed to radiation in various ways: by electron irradiation at the PTM-55 microtron (Russia) up to an absorbed dose of $10^9$ Gy and by alpha-emitters up to $10^{19}$ dis./g at the HVEE cascade accelerator (Netherlands).

Previously [18], it was found that the maximum hydrogen release from cement waste forms based on Portland cement does not exceed $10^{-3}$ mol/(g of sample) at an absorbed dose of $10^8$ Gy.

## Results and discussion

Tables 2 and 3 present the measured levels for regulated properties of the cement compounds listed in Table 1 as regards cement compounds produced using HIP and conventional mixing methods. The given results were averaged over three parallel measurements.

Table 2 shows that on the 28th day of hardening, the strength of cement compounds produced using the HIP method significantly exceeds the strength of conventional compounds, namely, by 3—8 times, depending on W/C ratio: at W/C=0.1 it amounts to 115 MPa, which is comparable to the characteristics of special high-strength concretes. The samples shown in Figures 4 and 5 have dense homogeneous structure without any visible defects and cracks. When tested for frost resistance and resistance to prolonged water exposure, the strength level virtually did not change. Table 2 summarizes the test results compared to the reference samples considering an equivalent hardening time.

The rate of $^{137}$Cs and $^{239}$Pu leaching (Table 3) from cement compounds produced by the HIP method is 5—10 times lower, depending on the W/C ratio as compared to conventional compounds. For $^{137}$Cs this indicator is less than $10^{-6}$ g/(cm$^2$·day), which is comparable to the leaching rate from glass-like and ceramic waste forms.

*Table 2. Mechanical compressive strength of cement compounds during hardening and after the tests seeking to assess the frost resistance and resistance to prolonged exposure to water*

| Composition | Average measured compressive strength, MPa | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | Post-HIP hardening | | | Ordinary mixing, Day 28 | Frost resistance | | Resisence to prolonged water exposure | |
| | Day 7 | Day 14 | Day 28 | | Control | After testing | Control | After testing |
| 1 | 62.9 ± 5.8 | 76.3 ± 6.5 | 115.2 ± 9.2 | 13.5 ± 1.2 | 118.4 ± 10.2 | 120.2 ± 11.1 | 125.1 ± 15.3 | 115.6 ± 10.3 |
| 2 | 43.4 ± 4.0 | 50.8 ± 4.5 | 61.0 ± 5.6 | 15.4 ± 1.3 | 67.9 ± 4.5 | 60.0 ± 5.5 | 73.5 ± 6.2 | 60.4 ± 7.0 |
| 3 | 57.3 ± 5.5 | 70.2 ± 7.0 | 104.0 ± 8.7 | –* | 110.6 ± 9.3 | 101.3 ± 10.5 | 125.5 ± 17.2 | 107.3 ± 10.5 |
| 4 | 26.3 ± 2.1 | 32.6 ± 2.8 | 40.5 ± 3.8 | –* | 47.1 ± 4.2 | 48.3 ± 5.2 | 52.1 ± 4.4 | 47.3 ± 2.9 |
| 5 | 18.6 ± 2.1 | 22.8 ± 3.2 | 31.0 ± 3.4 | 12.3 ± 2.9 | 24.6 ± 2.5 | 22.2 ± 2.0 | 27.1 ± 2.2 | 22.4 ± 1.7 |

*dry compound, cement sample failed to make

*Table 3. Leaching rate of $^{137}$Cs and $^{239}$Pu at the 28th day*

| Composition | Leaching rate, g/(cm$^2$·day) | | | |
|---|---|---|---|---|
| | $^{137}$Cs | | $^{239}$Pu | |
| | After HIP | Ordinary mixing | After HIP | Ordinary mixing |
| 1 | $6.1\cdot10^{-7}$ | $1.0\cdot10^{-5}$ | $1.0\cdot10^{-8}$ | $7.8\cdot10^{-7}$ |
| 2 | $1.0\cdot10^{-6}$ | $3.7\cdot10^{-5}$ | $6.5\cdot10^{-7}$ | $1.0\cdot10^{-6}$ |
| 3 | $9.0\cdot10^{-7}$ | –* | $8.3\cdot10^{-8}$ | –* |
| 4 | $6.6\cdot10^{-7}$ | –* | $1.0\cdot10^{-8}$ | –* |
| 5 | $3.0\cdot10^{-6}$ | $6.1\cdot10^{-5}$ | $4.0\cdot10^{-7}$ | $3.1\cdot10^{-6}$ |

*dry compound, cement sample failed to make

*Table 4. Mechanical compressive strength of cement compounds after testing for radiation resistance, frost resistance and resistance to prolonged water exposure*

| Composition | Average measured compressive strength, MPa | | | | | | |
|---|---|---|---|---|---|---|---|
| | Absorbed dose after electron exposure, Gy | | | Frost resistance | | Resisence to prolonged water exposur | |
| | 0 | $10^8$ | $10^9$ | Control | After testing | Control | After testing |
| 1 | 115.2 ± 9.2 | 116.3 ± 8.7 | 121.4 ± 7.5 | 122.7± 6.1 | 108.3 ± 7.3 | 131.0 ± 15.2 | 112.3 ± 10.1 |
| 2 | 61.0 ± 5.6 | 61.3 ± 6.0 | 53.4 ± 4.8 | 45.5 ± 2.8 | 53.7± 5.0 | 54.5 ± 5.0 | 45.6 ± 5.1 |
| 3 | 104.0 ± 8.7 | 110.1 ± 10.5 | 100.0 ± 8.9 | 100.5 ± 9.5 | 89.8 ± 10.0 | 104.5 ± 9.5 | 93.0 ± 10.0 |
| 4 | 40.5 ± 3.8 | 40.6 ± 4.5 | 44.3 ± 3.2 | 35.0 ± 2.1 | 33.2 ± 3.1 | 40.0 ± 3.5 | 34.5 ± 4.0 |
| 5 | 31.0 ± 3.4 | 31.5 ± 2.5 | 34.4 ± 2.2 | 45.7± 2.9 | 44.1 ± 3.0 | 55.7± 7.2 | 46.3 ± 5.0 |

*Table 5. The rate of $^{137}$Cs and $^{239}$Pu leaching from irradiated samples at the 28th day*

| Composition | Leaching rate, g/ (cm$^2$·day) | | | |
|---|---|---|---|---|
| | $^{137}$Cs | | $^{239}$Pu | |
| | $10^9$ Gy | $10^{19}$ disp./g | $10^9$ Gy | $10^{19}$ disp./g |
| 1 | $8.2 \cdot 10^{-7}$ | $9.8 \cdot 10^{-7}$ | $3.2 \cdot 10^{-8}$ | $4.8 \cdot 10^{-8}$ |
| 2 | $9.8 \cdot 10^{-7}$ | $1.0 \cdot 10^{-6}$ | $7.0 \cdot 10^{-7}$ | $9.3 \cdot 10^{-7}$ |
| 3 | $2.5 \cdot 10^{-6}$ | $3.0 \cdot 10^{-6}$ | $1.0 \cdot 10^{-7}$ | $1.5 \cdot 10^{-7}$ |
| 4 | $6.2 \cdot 10^{-7}$ | $6.8 \cdot 10^{-7}$ | $2.8 \cdot 10^{-8}$ | $4.0 \cdot 10^{-8}$ |
| 5 | $7.5 \cdot 10^{-6}$ | $7.9 \cdot 10^{-6}$ | $6.5 \cdot 10^{-7}$ | $9.8 \cdot 10^{-7}$ |

Tables 4 and 5 summarize the results of tests aimed at evaluating mechanical, physical and chemical properties of irradiated cement compounds. Samples irradiated to the highest dose (by electrons up to $10^9$ Gy) were tested for frost resistance and resistance to prolonged water exposure. Visual inspection of the obtained compounds did not reveal any defects such as cracks and chips. Table 4 presents the test results compared with reference samples considering similar hardening time.

The compressive strength of samples referred to as 1, 4 and 5 increases by (5—10) % under the radiation exposure impact. The effect of radiation hardening was noted in [19]. The decrease in the compressive strength that manifested itself during frost resistance testing and tests on the resistance to prolonged water exposure was found to be less than 20 %, which complied with the regulatory requirements for cemented RW.

Figure 6 presents the results of X-ray phase analysis for cement compound samples before and after irradiation with electrons up to a dose of $10^9$ Gy and alpha-emitters up to $10^{19}$ dis./g. Akermanite $Ca_2Mg[Si_2O_7]$ and merwinite $Ca_3Mg[SiO_4]_2$were revealed as the main identifiable phases in the samples, which are considered characteristic components of blast-furnace slags. Phases of magnesium manganese silicate, calcium magnesium aluminosilicate, hydrated sodium magnesium aluminosilicate were identified as well.

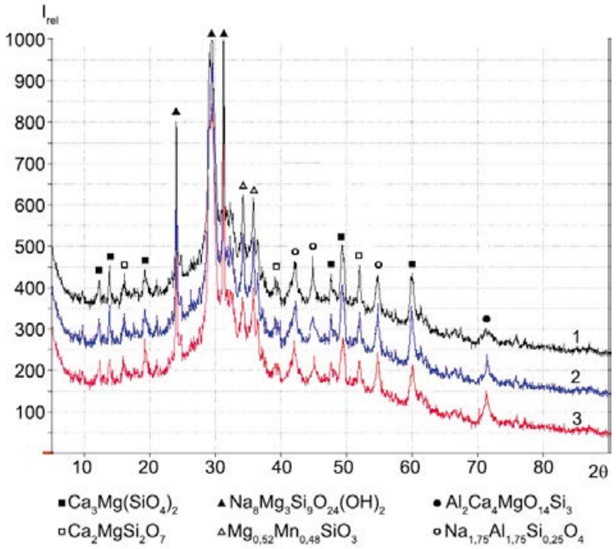
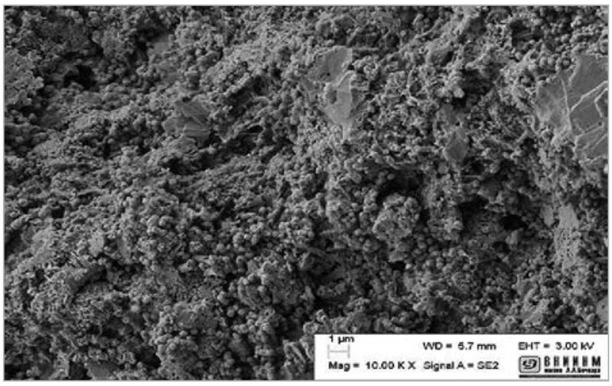


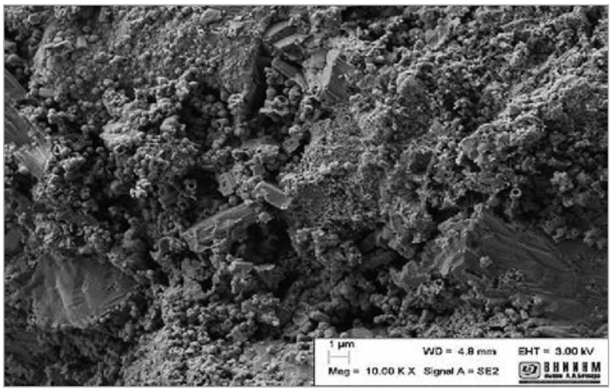
*Figure 6. X-ray pattern of a cement compound sample before irradiation (1), after irradiation with electrons up to a dose of $10^9$ Gy (2) and alpha emitters to up to $10^{19}$ diss./g (3)*

The types of X-ray patterns for non-irradiated and irradiated samples are quite similar, therefore, one may assume the stability of the phases to the ionizing radiation effects.

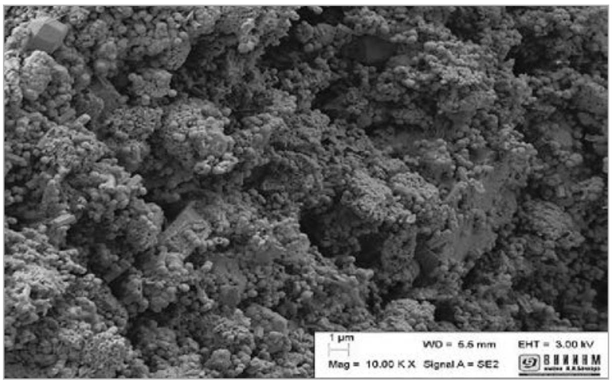
The images obtained by scanning electron microscopy showed that the microstructure and the morphology of cement compounds did not suffer any changes, neither some new structures nor defects have formed under the radiation exposure impact (Figure 7).



a) before radiation exposure



b) after radiation exposure with electrons to a dose of up to $10^9$ Gy



c) after radiation exposure with alpha emitters to $10^{19}$ diss./g

*Figure 7. Microstructure of the cement compound before and after radiation exposure*

To streamline the process, preliminary tests of the pressing method involving a heated mold have been launched: the container was sealed with a lid, installed into a heated mold and pressed from above with a punch of a purpose designed press by (10—20) rev.% at a capsule temperature of (200—300) °C for 2—3 hours. Currently, the optimal process parameters and properties of the cement compound are being evaluated.

The strength characteristics of the cement compound produced by the HP method far exceeded the minimum levels set forth for the characteristics of cemented waste forms intended for low- and intermediate-level waste containment and considered adequate for the safe handling and storage of such RW. This method can be used to maximize the content of the final product containing the radioactive waste. Some research has been launched to study the cementation of borate melt and spent ion-exchange resins seeking to increase the content of these RW in the final product by 2—3 times compared to commonly applied methods. Preliminary results suggest that cement compound with a melt content of up to 45 wt% and the one of ion exchange resin of up to 40 vol.% may be produced.

## Conclusions

The HIP method applied to produce cement compounds with a reduced W/C ratio results in a dense waste form with a structure being uniform over the entire volume of the conditioned RW.

The cement compound strength at the 28th day amounted to (31—115) MPa depending on the type of aggregator and W/C. The level of (104—115) MPa for samples produced using the HIP method significantly exceeds the strength of compounds produced based on the commonly applied method and is comparable with the one of such rocks as marble, the estimated strength of which amounts to (100—250) MPa. The leaching rate of $^{137}$Cs and $^{239}$Pu radionuclides was found to be equal to ($10^{-6}$—$10^{-7}$) g/cm$^2$·day and ($10^{-7}$—$10^{-8}$) g/cm$^2$·day respectively, which is comparable to the characteristics of glass-like and ceramic waste forms. No decrease in the strength following the tests for frost resistance and resistance to prolonged water exposure has been revealed.

High-level radiation exposure to levels equivalent to those emitted by HLW, did not affect the controlled properties and produced no visible changes in the microstructure of the cement compounds. In some cases, the strength characteristics of some samples have increased following the radiation exposure.

The waste form components were mixed directly in the container, while fragments of solid radioactive waste (parts and structural materials of SFAs, filters, pulp sorbents, etc.) could be additionally introduced into the waste form. The use of sealed metal containers for HIP-based waste form synthesis is believed to provide some additional protection.

The method discussed above is thought to have great potential as regards the conditioning of RW with complex chemical composition, such as borate melts from the evaporation of LRW bottom residues at NPP, spent ion-exchange resins, LRW containing organic substances, tritium. HIP method may increase the RW content in the final product by 2—3 times compared to the common cementation techniques.

The volume and shape of the compressed capsules can be optimized to maximize the disposal package filling degree. For example, a NZK-150-1.5P container can be filled with 30 % more compressed capsules compared to the case when it is filled with four 200-liter drums.

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