SCIENTIFIC AND DESIGN ASPECTS OF LIQUID RADIOACTIVE WASTE VITRIFICATION FROM NUCLEAR POWER PLANTS WITH WWER-1200 REACTOR UNITS

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The article presents a comparison of processing technologies for liquid radioactive waste bottom sediment from NPPs with WWER-1200 reactor units. Vitrification and cementing methods were compared based on the state of art in the development of the Unified State System for Radioactive Waste Management, as well as engineering and design study of various processing methods. The research demonstrates that industrial use of the vitrification method can be seen as a promising one when it comes to the processing of liquid radioactive waste from NPPs and radiochemical plants.

Keywords: *radioactive waste (RW), liquid RW (LRW), WWER-1200 NPP, vitrification of RW, induction melter, disposal, disposal facility.*

Operation of a nuclear power plant results in significant amounts of liquid radioactive waste (LRW) with the main sources of its generation accounting for floor drains (effluents from equipment decontamination during routine and repair operations), regeneration solutions and solutions from filter washing. This waste is high-salinity solutions contaminated with fission products, corrosive radionuclides and various substances used to maintain water chemistry and to decontaminate the equipment. Resulting LRW are evaporated with the still bottom residue (BR) subject to solidification and conditioning.

In keeping with the Federal Law On Radioactive Waste Management and on Amendments Introduced to Certain Legislative Acts of the Russian Federation No. 190-FZ, before being disposed of, liquid radioactive waste shall be immobilized and conditioned to meet the acceptance criteria [1]. Pre-conditioning of bottom sediments involves its processing to convert it into a stable solid form. For this purpose, various methods are used:

- BR strong evaporation resulting in solid salts (“salt melt”);
- solidification using binders;
- extraction of radionuclides contained in BR;
- BR vitrification.

Table 1 presents a qualitative comparison of methods applied to treat BRs from nuclear power plants.

Strong evaporation method. BR immobilization by strong evaporation proceeds as follows: BR with

Table 1. Comparison of methods used to manage BR from NPPs with WWER-1200 reactor units

Advantages of the matrix and the technology	Disadvantages of the matrix and the technology
Cementation	
• simplicity of process equipment; • low temperatures	• increased volume of solidified waste; • low hydrolytic stability; • of LRW pre-cementing treatment
Strong evaporation	
• simplicity of process equipment	• high solubility of salt residue; • possibility of final product transition to a fluid state; • high corrosion activity of salt residue
Solvent extraction	
• boric acid can be regenerated	• sophisticated technology; • highly active sorbent requiring its final disposal in a deep repository; • salts with a high content of technogenic radionuclides shall be contained
Vitrification	
• high hydrolytic stability; • significant reduction in the final volume of the solidified waste; • can be applied to immobilize LRW of any activity level; • low leaching rate	• high process temperature; • radionuclide entrainment into gas phase; • relatively high-power consumption for glass melting; • sophisticated process involving high temperatures; • purpose-designed gas purification equipment is required to remove nitrogen oxides generated during vitrification

a salinity of 300—400 g/l is sent to evaporation unit (EU) for additional evaporation to a salinity of 1,500—1,800 g/l. The resulting fluid product is poured at a temperature of about 80 °C into 200-liter cans where its solidification occurs upon its cooling (“salt melt”). This method is applied at Novovoronezh and Balakovo NPPs with WWER reactor units.

To date, a big number of salt-filled cans has been accumulated in SRW storage facilities located at at-reactor sites, which, in accordance with the classification established in the Government Resolution of the Russian Federation of October 19, 2012 No. 1069 On Criteria Used to Categorize Solid, Liquid and Gaseous Waste as Radioactive Waste, Criteria Used to Categorize Radioactive Waste as Non-retrievable Radioactive Waste and Retrievable Radioactive Waste and Classification Criteria for Retrievable Radioactive Waste (Government Resolution No. 1069), refers to retrievable RW Class 3 [2]. However, this waste form provides no full compliance with the established RW acceptance criteria for disposal.

A study [3] showed that to meet the acceptance criteria established for RW Class 3, the cans with salt melt should be subjected to additional containerization, for example, using containers of NZK-150-1.5P type. Although this BR processing method accounting for conditioning, transportation and disposal stages, is associated with the lowest costs compared to other methods applied at Russian NPPs, the high solubility of the salt melt and its transition to a fluid state due to water absorption upon its contact with atmospheric air prompts discussions on the need of additional salt melt processing to bring it into an insoluble form.

BR solidification using binders. Evaporation followed by BR solidification using cement material was adopted as the main LRW processing method in Russian: high-salt aqueous BR solutions are mixed with cement and necessary additives, followed by hardening of the resulting cement mortar.

This method was adopted in the designs of NPPs with WWER-1200 reactor units in Russia (Kursk-2, Leningrad-2, Novovoronezh-2) and abroad (Belarus, El-Dabaa, Paks-2, Hanhikivi-1, etc.).

Despite relative simplicity and low costs involved, this method results in higher volumes of the solidified product compared to the initial BR volume due to relatively low degree of salt inclusion into the compound, requires strict adherence to cementing technology with no sufficient reference experience in cementing waste with LRW chemical compositions adopted under NPP designs providing for a wide range of possible changes in borate content.

The closest LRW solidification process analogue for waste generated at NPPs with WWER-1200 reactor units is the bottoms cementing plant operated for many years at Rostov NPP. Industrial operation of the cementing unit at Rostov NPP was preceded by research, pilot and experimental operations focused on process parameters relevant for reliable operation of the equipment and production of a cement compound with highest possible degree of filling, the quality of which would satisfy the established regulatory requirements [4].

Available experience in LRW cementation at Smolensk NPP cannot be considered as a reference one for the designed WWER-1200 reactor units, since borates are virtually absent in LRW composition.

Extraction of radionuclides. Given normal operating conditions, BR activity mainly depends on the content of cesium, strontium, cobalt and manganese radionuclides which can be absorbed on inorganic sorbents with a very high degree of selectivity. It was this fact that actually prompted the development of this method. The extraction process involves a number of successive stages: ozonation of solutions to destruct organic substances and complexes binding radionuclides and preventing their sorption; filtration allowing to separate precipitates from the previous stage; selective sorption of radionuclides from the filtrate. After such processing, the high-salinity filtrate is subject to evaporation to the maximum salt content, with a solid material (salt mixture) being produced upon its cooling. This method provides concentration of the major part of radioactive substances contained in sediments and sorbents, while the salts are not assigned to RW category given their activity level.

It should be noted that the efficiency and universality of BR ion-selective purification method implemented at Kola NPP has not yet been demonstrated for other NPPs.

BR vitrification. Vitrification method can be considered as an alternative option. Initially, it was developed to provide immobilization of high-level waste (HLW) from SNF reprocessing.

On an industrial scale, this method is used to immobilize waste in France (since 1977), the U.S. (Savannah River, West Valley, since 1980), Germany (Karlsruhe), Belgium (Moth, Pamela melter, since 1990), Russia (PA Mayak, since 1986).

It is generally recognized that among currently available industrial LRW processing methods, it is the vitrification that produces most stable material allowing reliable containment of radioactive substances. Melters of various designs constitute a main unit of RW vitrification facilities: salts contained in the waste and glass-forming materials (feed) are melted inside this unit. To date, two main types of melting devices basically applied for vitrification purposes have been developed and tested:

- Ceramic melters with direct electric heating. In this case, the heat required for melting is emitted when electric current passes through the molten glass in the melter;
- induction melters of two types: a) the energy is released when the electromagnetic field excited by the generator interacts with the conductive wall of the metal crucible (hot crucible) b) the energy is released when electric current is induced inside molten glass melt (cold crucible).

An important aspect that should be taken into account during the selection of a melter type used to immobilize LRW generated from NPP operation is that direct electric heating melters require constant maintenance of the molten glass in the molten state throughout the entire period of their operation. Provided periodic supply of LRW for solidification, this operation mode leads to wasteful energy expenditures.

Hot crucible induction melters that have been developed to date are characterized by an insufficient capacity of about 15 kg/h and a short crucible life of 2,000 hours. Therefore, given the significant amount of waste inventory accumulated at NPPs, the use of this melter type seems to be unfeasible from the engineering perspective.

Various types of melters developed to date and used to vitrify the RW from NPP operation have been evaluated: it was found that the most preferable option suggests the use of units fitted with an induction melter of "cold" crucible type (IMCT).

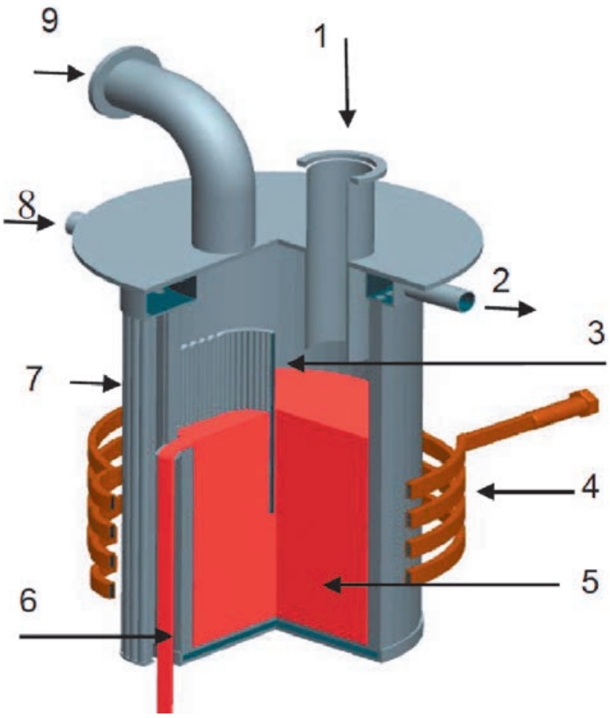

Figure 1. Induction melter with a "cold" type crucible
1 – feeder, 2 – cooling water outlet, 3 – baffle, 4 – inductor, 5 – melt, 6 – crucible lip, 7 – profiled sections, 8 – cooling water inlet, 9 – gas duct

This melter (Figure 1) is a structure with vertical water-cooled metal sections made of stainless steel and isolated from each other (7). Inside the crucible there is a melt of oxides or glass (5) which is electrically conductive and able to absorb the energy of the electromagnetic field generated by the HF generator. This process results in heat generation which covers the losses and allows to maintain given melt temperature.

To supply a high-frequency electromagnetic field, a cylindrical inductor (4) embodying the crucible is used.

During crucible operation, a skull layer is formed on the contact surface of the melt with the cooled wall and the bottom (a layer of melt solidified upon contact with the cooled surfaces of the crucible), which prevents the melt from flowing into the gaps between the crucible sections, protects its material from corrosion and also acts as a lining and bath insulation.

To prevent the ingress of unmelted material into the melt discharge zone, a water-cooled baffle (3) is provided for in the designs. The melt is discharged continuously via a crucible lip (6).

The main advantages of IMCT-type melters are as follows:

- high specific productivity;
- opportunities for maintaining both continuous and periodic operational mode;
- opportunities for remote replacement;
- small dimensions of the melter proving for the development of compact chamber designs accommodating the vitrification system.

Vitrification process implemented for many years at MosNPO Radon to treat ILW can be referred to exemplify the feasibility of IMCT method [3].

Vitrification technology providing for the use of a cold-type crucible induction furnace to treat LLW and ILW was considered by SGN and KEPCO as far back as in 1995. These studies demonstrated the cost-effectiveness of this solution in treating waste from Korean NPP [4].

At present time, industrial RW vitrification technology based on IMCT application and production of borosilicate glasses is being developed under scientific supervision of JSC VNIINM and JSC Radium Institute named after V. G. Khlopin providing for its further implementation at MCC, PA Mayak and SCC Pilot-Demonstration Centers [5—6].

Thus, one can state that the Russian Federation possesses over 30 years of vitrification experience addressing RW of various compositions and activity levels. In the coming years, this experience will be expanded along with the commissioning of new industrial and pilot plants.

Design solutions for BR vitrification from NPPs with WWER-1200 reactor units

Normal operation of WWER-1200 NPPs is expected to generate some 25 m^3 of BR per year with a salinity of about 400 g/dm^3.

Table 2 presents the estimated chemical composition of the BR. Its maximum activity can amount to $5 \cdot 10^2$ MBq/dm^3 with the radionuclide inventory involving ^{137}Cs — ~ 97.5 %, ^{60}Co — ~ 2 % and other radionuclides — less than 1 %.

Table 3 presents the composition of glasses for RW solidification [3].

Based on the composition of borosilicate glasses, the amount of oxides contained in BR and incorporated into the glass amounts to ~ 30 wt %.1

Table 2. The estimated average annual BR inventory

Component	Concentration, g/dm^3	Recalculated oxide concentration, g/dm^3	Chemical oxide-based composition, wt %
BO_3^{3-}	8.083	4.1	1.24
K^+	5.389	6.5	1.97
NH_4^+	$2 \cdot 10^{-3}$	–	–
NO_3^-	65.975	–	–
MnO_2	6.699	6.7	2.03
Cl^-	$0.13 \cdot 10^{-3}$	–	–
Fe^{2+}, Fe^{3+}	8.738	12.4	3.76
CO_3^{2-}	79.374	–	–
Sulfanol	0.874	–	–
Sodium hexametaphosphate	2.039	-	-
Na^+	222.829	300.3	91.0
Total	400.0		

Table 3. Composition of glasses for RW solidification

Site/country/RW	SiO_2	P_2O_5	B_2O_3	Al_2O_3	CaO	MgO	Na_2O	Other	Waste filling
R7/T7, France, HLW	47.2	–	14.9	4.4	4.1	–	10.6	18.8	≤ 28
DWPF, USA, HLW	49.8	–	8.0	4.0	1.0	1.4	8.7	27.1	≤ 33
WVP, UK, HLW	47.2	–	16.9	4.8	-	5.3	8.4	17.4	≤ 25
Pamela, Germany, Belgium, HLW	52.7	–	13.2	2.7	4.6	2.2	5.9	18.7	≤ 30
FSUE PA Mayak, Russia, HLW	–	52.0	–	19.0	–	–	21.2	7.8	≤ 33
FSUE MosNPO Radon, Russia, ILW	43.3	–	6.6	3.0	13.7	–	23.9	9.8	≤ 35

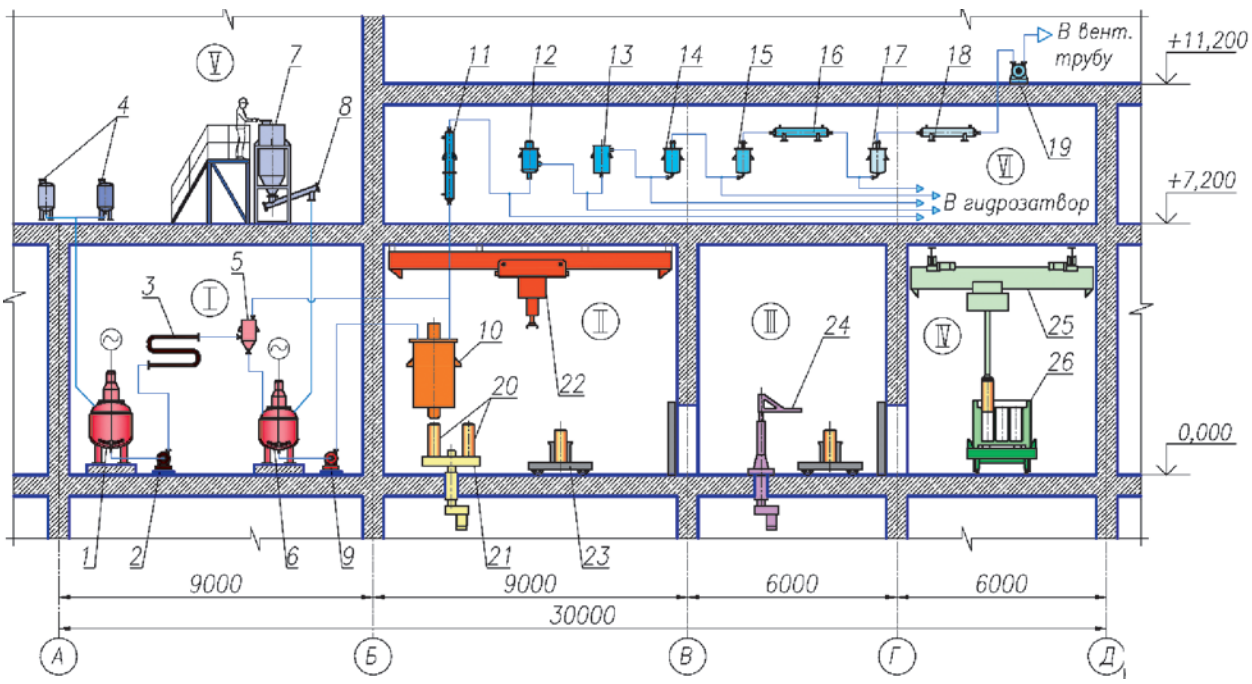

Figure 2. Hardware and engineering layout of a facility for BR vitrification at an NPP with WWER-1200 reactor units
I – BR pre-solidification treatment unit; II – salt melt vitrification system; III – can sealing system; IV – NZK packaging system; V – fluxing additives pretreatment and reagents batching system; VI – gas purification system

Thus, some 1,000 g of borosilicate glass with an activity of $5{\cdot}10^8$ Bq/kg can be produced from 1 dm^3 of BR. Given an average glass density of 2.5 kg/dm^3, the volume of vitrified waste will amount to 0.4 dm^3. The annual vitrification capacity will amount to $2.5{\cdot}10^4$ kg or 10 m^3.

Given IMCT vitrification capacity of about 10 kg/h by glass production, annual BR amount (25 m^3) will be processed in about 100 days assuming continuous operation.

Figure 2 presents the hardware and engineering layout of a facility for BR vitrification at a NPP with WWER-1200 reactor unit.

Below are considered the basic BR vitrification flow chart.

Bottom residue with a salinity of 400 g/l is pumped (2) from a tank (1) at a flow rate of 10 l/h to a direct-flow evaporator (3), where it is evaporated to a concentration of 1,000 g/l. To adjust BR chemical composition, necessary reagents are introduced into it with a correct batching provided by the use of measuring devices (4). From the evaporator, the solution is fed into separator (5): liquid phase at a rate of 4 l/h is discharged into a 0.5 m^3 air-operated concrete pump (6) equipped with a steam jacket and a mixer; the gas-vapor phase is fed to the gas purification system. After the air-operated concrete pump is filled, glass-forming and fluxing additives are fed into it from a hopper-mixer (7) using a screw batcher (8). Homogeneous mixture generated from salt melt mixing with fluxing additives is batched by pump (9) into IMCT (10). The gas-vapor phase is fed into gas purification system involving a heat exchanger (11), self-cleaning coarse filters (12) and fiberglass industrial fine filters (13), bulk reagent capture (14) and SMOG devices (15), heater (16) and a hemi-oxide purification device (17), refrigerator (18) and a vacuum pump (19). Gas purification devices ensure the removal of radioactive aerosols and nitrogen oxides from process gases proving their compliance with sanitary standards.

Molten glass melt containing radionuclide oxides is periodically poured into a metal can (20) with a capacity of 65 dm^3 (Figure 3) mounted on a revolving carrier (21). A filled can is installed by a lifting mechanism (22) on a trolley (23) and moved to the sealing compartment, where a lid is installed and sealed by remote welding (24). The sealed can is transported to the packaging compartment where it is lifted by a crane (25) and installed into a NZK reinforced concrete container with nine slots (26) (see also Figure 3). After nine cans are installed, the container is covered by a lid, sealed and sent for storage and (or) disposal.

Preliminary estimates show that structural volume of about 4 thousand m^3 is required to accommodate the vitrification installation (including electrical equipment and the operator's room).

The hardware design and transport and process equipment of the vitrification facility intended for liquid LLW and ILW from nuclear power plants can be much simpler than in case of HLW vitrification facilities currently being designed for radiochemical production facilities (pilot demonstration power

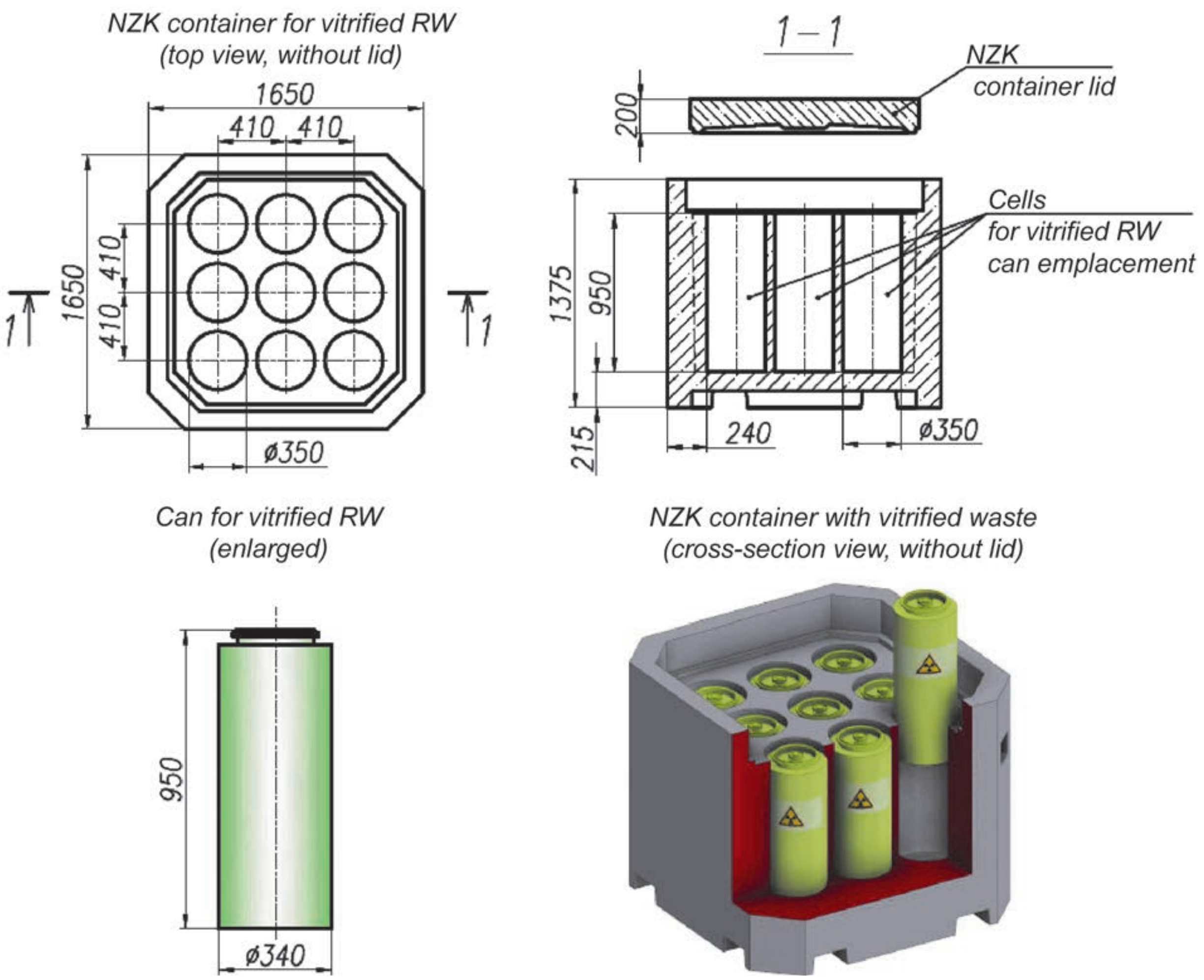

Figure 3. NZK container for vitrified RW cans

complex, pilot demonstration center). In case of ILW vitrification, costly remotely-controlled equipment for HLW handling can be replaced by more affordable manually controlled equipment. Some operations, for example such as IMCT replacement can be performed in the presence of personnel.

Table 4. Estimated annual performance of a vitrification unit

Item	Indicator	Remarks
Annual amount, m^3	25.0	–
Vitrified RW amount, m^3	10	–
Vitrified RW weight, kg	25,000	–
Total activity, Bq	$1.25 \cdot 10^{13}$	–
Specific activity of glass, Bq/g	$5 \cdot 10^5$	RW Class 3
Number of cans, pcs	154	Volume – 65 dm^3
Number of NZK-150-1.5P, pcs	17	Maximum capacity – 9 cans
Total activity per package, Bq	$7.3 \cdot 10^{11}$	A-type package
Dose rate on the surface of a package, mSv	Less than 2	–

It should be also noted that, if a replacement is necessary, spent crucible can be also emplaced into an NZK container.

Table 4 presents the estimated annual performance of a vitrification unit and the characteristics of the final product.

Feasibility assessment

In order to determine the economic advantages of BR vitrification technology and its application at NPPs over the basic immobilization technology (cementation), technical and economic indicators of these methods were compared taking into account relevant costs associated with conditioning, transportation and disposal.

Cementation method involves BR concentration up to a salt content of 800 g/l, solution cooling to a temperature of 60 °C, its 5-minute-long mixing with a binder in a mixer. Given a solution-binding ratio of 0.8—0.9, salt inclusion will amount to ~25 wt.% with a strength of more than 5 MPa, leachability of less than 10^{-3} g/(cm^2·day), compound density — ~2 g/cm^3. The cement compound is poured into metal 200-liter cans or concrete containers with a metal insert. Metal cans are installed into a concrete container NZK-150-1.5P with a capacity of four cans.

Table 5 presents estimated annual performance of a cementing unit and the characteristics of a final product.

Table 5. Estimated annual performance of a cementing unit

Item	Indicator	Remarks
Annual amount, m^3	25.0	–
Cemented RW amount, m^3	38.8	–
Cemented RW weight, kg	77,000	–
Total activity, Bq	$1.25 \cdot 10^{13}$	–
Specific activity of cement, Bq/g	$1.6 \cdot 10^{5}$	RW Class 3
Number of can, pcs	200	Volume – 0.2 dm^3
Number of NZK-150-1.5P, pcs	50	Maximum capacity – 4 cans
Total activity per package, Bq	$2.4 \cdot 10^{11}$	A-type package
Dose rate on the surface of a package, mSv	Less than 2	–

Table 6 shows BR conditioning costs associated with the application of vitrification and cementing methods and conditioned waste disposal.

Table 6. Technical and economic indicators for BR conditioning and disposal, thousand rubles/year

Expenditure heading	Solidification method	
	Vitrification	Cementation
Cans/drums	6,930	6,000
NZK-150-1.5P containers	2,550	7,550
Transportation	504	1,480
Disposal	10,071	29,618
Total	20,055	44,648

BR vitrification costs can be calculated during design studies and task-specific feasibility studies.

Preliminary technical and economic indicators derived for BR cementing and vitrification methods, presented in Table 6, confirm the feasibility of BR vitrification method for NPPs with WWER-1200 reactor units.

Conclusion

This study provided for the development of basic design solutions for the vitrification of bottom residues generated at NPPs with WWER-1200 reactor units: method was compared with a referent solidification method adopted under NPP designs (cementation).

Calculations showed that according to the existing RW classification system for disposal purposes, vitrified bottoms from WWER-1200 NPPs can be attributed to RW Class 3. Thus, the packages with vitrified waste can be disposed of in near-surface disposal facilities.

Proposed engineering solution suggesting that vitrified RW is packaged into NZK-150-1.5P containers ensures compliance with the requirements established for A-type packages according to NP-053-16 and acceptance criteria for disposal considering such indicators as radionuclide content, dose rate on the package surface, preservation of the containment capacity and other requirements stated in federal regulations.

An enlarged technical and economic assessment, as well as a comparison of indicators assumed under the proposed and the reference options taking into account the costs of basic consumables, conditioning, transportation and disposal shows that BR vitrification providing a reduction in waste amount can be more than twice as effective as the cementation method. At the same time, waste handling operations associated with waste storage, transportation and disposal are characterized with a higher level of radiation safety.

The developed solutions can be used to treat liquid ILW generated at radiochemical plants.

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Bibliographic description

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