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DISPOSAL SAFETY JUSTIFICATION FOR SALT MELT GENERATED AT NPP EVAPORATION-TO-THE-MAXIMUM-SALT CONCENTRATION PLANTS AND PACKED IN NZK-150-1.5P CONTAINERS

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The paper focuses on engineering and economic comparison of solidification methods applied to treat still residues from NPP VVER reactors with due account of the final RW management stages. It presents the feasibility and disposal safety study for salt melt packed into non-returnable protective NZK-150-1.5P containers.

Keywords: *radioactive waste, salt melt, conditioning, technical-and-economic indices, disposal safety*

Under the first NPP designs, deferred decision concept was provided for the radioactive waste (RW) management practice. Liquid radioactive waste (LRW) such as still bottoms, ion-exchange resins and perlite pulps and sludge were stored in purpose-designed reservoirs.

From the mid-80s, facilities designed for liquid and solid radioactive waste processing started to be developed. Table 1 provides information on existing and planned facilities for still bottoms (SB) treatment given the following industrial SB processing methods used at Russian NPPs: bituminization, deep evaporation to salts, ion-selective treatment and cementation.

It should be noted that literature sources provide no information on any feasibility studies of the technologies applied.

Table 2 presents a qualitative comparison of solidification methods for SB and resulting secondary waste.

Currently, operation of SB bituminization facilities is limited only to Kalinin NPP site. Deep evaporation method for LRW treatment was adopted as the basic one at Novovoronezh and Balakovo NPPs. Deep evaporation facilities (DEF) produce salt melt with a density of some 1.8—2.1 kg/dm^3 with crystal water content ranging from 5 to 20% (based on different literature sources).

Salt melt is emplaced into metal ZP551 and A2201 containers with a capacity of 0.2 m^3 and stored in purpose-designed facilities. In keeping with relevant provisions of RF Government Resolution № 1069 [2] of October 19, 2012, these RW are categorized as retrievable RW of Class 3 and 4.

In some cases, containers with salt melt considered as RW class 3 do not meet general waste acceptance criteria set for disposal. Namely, they do not comply with the requirements set in certain federal norms and rules [3] in terms of maintaining the insulating ability of RW packages (for at least 100 years) and the gamma radiation dose rate emitted.

Table 1. Existing and planned SB processing facilities at operating NPPs in Russia [1]

NPP	SB processing method			
	Ion-selective treatment	Deep evaporation till salt melt	Cementation	Bituminization
Balakovo	-	since 1992 г.	-	-
Beloyarskaya	since 2022	-	since 2022	-
Bilibino	-	-	-	-
Kalinin	-	-	-	since 1986
Kola	since 2006	-	On stand by since 1995	-
Kursk	since 2019	-	since 2019	-
Leningradskaya	since 2019	-	since 2019	since 1986 Currently on stand by
Novovoronezh	-	since 1991	-	-
Rostov	-	-	since 2005	-
Smolenskaya	since 2015	-	since 2011	-

Operating Planned Shut-down

Table 2. Comparison of methods for the solidification of SB and its secondary waste

Processing method	Advantages	Disadvantages
Bituminization	Relative ease of the process	Fire hazard. Increased waste generation. Stratification of bitumen during storage. Biologically driven corrosion. Gas generation
Cementation	Relative ease of the process, fire safety	The need to adjust SB composition. Increased waste volume
Ion-selective treatment	Optimal radionuclide fractionating. Possibility of valuable component extraction for recycling purposes	Sophisticated technology. Three types of waste generated. Multiple installations are required to be in place
Evaporation to salts	Ease of the process	No matrix

The above stated facts prompted a constant discussion on the need of introducing additional operations for salt melt conditioning enabling to achieve a waste form being suitable for its final disposal, as well as on the opportunities for further introduction of this method at new-built nuclear power plants with VVER-1200 type reactor units.

High efficiency was demonstrated for the ion-selective SB processing method put in place at Kola and Smolensk NPPs in terms of LRW fractionating enabling to generate end products with different length of the potential hazard time-period: sorbent (thermoxide-35) containing $^{137,134}Cs$, sludge mostly containing ^{60}Co and a salt melt not being categorized as RW.

The need of operating several facilities, namely, ozonation, sorption, deep evaporation, sludge cementation or drying facilities can be considered as a disadvantage of the technology. However, its attractiveness can be significantly increased if introduced are the technologies allowing the extraction of boric acid and other reagents from the salt melt.

SB cementation is widely used abroad and at a number of sites in Russia to immobilize low- and intermediate-level waste. Advantages of the method include comparative ease of mixing the LRW concentrate with a binder material, the incombustibility of the cement compound, high strength and sufficiently low leachability of radionuclides, which can be attained in case of strict adherence to cementation process specification.

This SB processing method is assumed as a basic one under the designs of Russian and foreign NPPs with VVER-1200 type reactor units.

The following aspects can be considered as potential disadvantages of the method: low degree of salt inclusion, high content of bound water, the need for strict observance of cementation specifications. LRW cementation method is applied at Smolensk and Rostov NPPs. Before putting this technology into industrial operation at Rostov NPP, relevant process parameters were developed to ensure reliable operation of the equipment, as well as to produce a cement compound with the highest possible degree of filling and the quality meeting relevant regulatory requirements.

Based on R&Ds carried out using simulated and real waste from Rostov NPP, it was found that [4]:

- the quality of cement compound during the cementation of LRW from NPPs with VVER-1000 type reactor units depends on the salt content of the still bottoms, availability, quantity and shape of borates, the alkalinity of the solution, the type and composition of the binder, solution-binder ratio, the temperature of the solutions and others factors;
- optimal conditions for LRW cementation should be identified for each specific composition of liquid waste, binder type and special additives with relevant limits being set up for possible alterations in the salt composition of the waste, salt concentration and solvent-binder ratio.

Vitrification can be suggested as an alternative option to the considered methods enabling to immobilize boron-containing LRW from nuclear power plants allowing to produce borosilicate glass using an induction melter with a cold crucible (IMCC) [5].

Six different options allowing to compare different SB management technologies presented in Table 3 were considered.

Table 3. Still bottom management options

Number of the option	SB solidification method	Primary packaging	Container
B-1	Cementation	Metal drum	NZK-150-1,5P
B-2	Cementation	-	NZK-150-1,5P
B-3	Deep evaporation till salts	Metal drum	NZK-150-1,5P
B-4	Deep evaporation till salts	-	NZK-150-1,5P
B-5	Ion-selective treatment	Filter container. Drum for VLLW salts	NZK-150-1,5P
B-6	Vitrification	Metal drum	NZK-150-1,5P

Table 4 presents the estimates of annual operating costs for the solidification of still bottoms from LRW evaporation calculated per one VVER-1200 unit with due account of relevant costs for reagents, energy, containers and disposal tariffs for the conditioned waste [6]. However, the following costs were not accounted for in the calculations: appreciation of equipment, staff salaries, expenses on lighting, heating, ventilation and other cost items associated with the operation of solidification facilities as these can increase the total costs, but would not produce a significant effect on the changes in relevant ratios.

The table shows that the cementation method providing for cement compound pouring into metal drums followed by their emplacement into protective NZK containers is considered as the most expensive option. When drums are omitted and the cement compound is poured directly into NZK containers, processing and disposal costs are halved. The costs associated with vitrification and ion-exchange treatment options are comparable and fall in between cementation and deep evaporation options. However, in terms of protective properties the vitrified waste form stands head and shoulders above salt melt and cement compound.

Table 4. Annual operating costs associated with the management of SB from LRW evaporation per one VVER-1200 unit, thousand rubles

Cost items	LRW solidification method					
	B-1	B-2	B-3	B-4	B-5	B-6
Processing	300	300	10,720	10,720	17,500	6,460
Drums	6,400	-	900	-	150	2,640
Containers	6,480	3,240	1,440	720	1,440	2,640
Transportation	1,680	840	248	124	264	683
Disposal	33,567	16,790	4,975	2,487	5,282	13,675
Total:	48,426	21,170	18,283	14,051	24,636	26,098
	345%	150%	130%	100%	175%	186%

SB deep evaporation to salt melt commonly used as a treatment method at Novovoronezh and Balakovo NPPs, is considered as a most cost-effective method for boron-containing SB processing.

To date, several tens of thousands of drums with salt melt have been accumulated at Balakovo and Novovoronezh NPPs. Demonstration of storage and disposal safety for the packages containing the salt melt will play a crucial role in the decision making on the further management flow chart for this waste.

Two approaches can be potentially applied to solve the salt melt management challenge: its processing allowing to enclose the radionuclides into a more robust waste matrix and packaging the salt melt into containers without prior processing to obtain a packaging that would comply with relevant waste acceptance criteria for disposal.

Existing and proposed methods can be used to process the salt melt accumulated at NPP sites: dehydration of the salt melt in an induction heating unit, cementation, incorporation into magnesium phosphate matrix, bituminization, incorporation into polyester resins, ion selective sorption, vitrification.

The indicated methods require the development and operation of purpose-designed plants enabling melt extraction from the containers, melt inclusion into particular matrixes, packaging of solidified product into containers allowing to obtain a package complying with relevant requirements of federal norms and environmental regulations. Economic evaluation of these methods was discussed in the first part of this paper with some additional costs that are to be covered to ensure salt melt extraction from the containers and its dissolution allowing its subsequent processing. Apparently, in terms of salt melt conditioning, emplacement of containers into purpose-designed concrete non-returnable protective NZK-150-1.5P containers is considered as the easiest and most cost-effective solution.

In 2003, Rosenergoatom Concern adopted the "Engineering Proposal No. NVAES TR-252 K03 on Changing the Storage Conditions for Metal Containers with Salt Concentrate from Deep Evaporation Units of Novovoronezh NPP" providing for the emplacement of packages into reinforced concrete NZK-150-1.5P containers.

This decision allowed the following:

- to ensure the safe storage of expired containers with salt melt;
- to ensure the storage designs;
- to ensure safety of lifting and handling operations with containers;
- to limit personnel exposure;
- to obtain the packages with the salt melt to be transferred to the National Operator for disposal.

The main condition as regards the package transfer to the National Operator is the adherence to the requirements established by federal norms and rules for RW of this class and RW acceptance criteria for disposal set forth for each particular RW disposal facility (operated or planned for construction).

Table 5 summarizes most important requirements for RW packaging of RW class 3 and 4.

Table 5. Requirements to the packages designed for different RW classes [3]

Requirements	RW class	
	3	4
Dose rate on the surface, mGy/h	No more than 10 mGy/h	No more than 2 mGy/h on RW surface
Mechanical compressive strength	Not lower than similar requirements to A type package (more than 5 MPa)	
Rate of radionuclide release from the package (mass fraction of activity released from RW package per year)	No more than 10^{-2}/year for tritium	
no more than 10^{-3}/year for β,γ-waste;		
no more than 10^{-4}/year for α-waste		
Maintaining RW package insulating capacity, years	No less than 100 yeas	Prior to actual disposal
Thermal Cycle Resistance	preservation of strength and insulating properties after 30 cycles of freezing and thawing (40 ... +40 °C)	
Radiation resistance of the RW package	strength reduction by no more than 20 % of the established limit when irradiated with a dose of 10^6 Gy or a predicted one	

Another issue to discuss is how the characteristics of salt-melt containing packages comply with relevant regulatory requirements. It should be noted that the monograph [7] provides a more detailed summary on the properties of non-returnable protective containers providing radiation safety, strength, insulating ability and durability.

Radiation safety requirements

According to [8], at Novovoronezh NPP, for more than 30 % of A2201 containers the exposure dose on RW container surface exceeded 2.0 mSv/h.

For A2201 containers being placed into NZK containers given certain displacement and backfilling of void space with 2.1 g/cm^3 dense material, the following gamma radiation attenuation coefficients can be achieved:

- about 15 for NZK facets with a minimum layer of buffer backfill;
- about 380 for NZK facets with a maximum layer of buffer backfill.

Following the conditioning performed, for most part of containers, gamma exposure rate measured immediately adjacent to NZK container facet amounted to 0.43 mSv.

Evaluation and demonstration of container's mechanical strength

The task of substantiating the mechanical strength of disposal container was reduced to the evaluation of its structure in terms of its ability to withstand certain static and dynamic loads:
- when containers are stacked in 6—8 rows;
- when the container is dropped on a solid base from a height of 0.5 and 1.2 m.

Factory and certification tests confirmed the mechanical strength of the containers in terms of withstanding relevant static and dynamic loads.

Insulating ability of the packages

Radionuclide release from containers can occur due to the dissolution of salts containing radionuclides and the diffusion of radionuclides in the pore water of container walls, in engineered safety barriers (under near-surface disposal conditions) and in the geological formation (under deep disposal conditions). To predict radionuclide diffusion through container wall resulting in its release into the environment (for example, through the backfill and/or the bedrock), finite thickness equation (container wall) can be applied. Its analytical solution [9] is as follows:

$$C_1(x,t) = C_0\,e^{-\lambda t}\left[\operatorname{erfc}\frac{x}{2\sqrt{D_1^{eff}t}} - h\sum_{n=1}^{\infty}h^{n-1}\left[\operatorname{erfc}\frac{2nL-x}{2\sqrt{D_1^{eff}t}} - \operatorname{erfc}\frac{2nL+x}{2\sqrt{D_1^{eff}t}}\right]\right],$$

$$C_2(x,t) = C_0\,e^{-\lambda t}\,\frac{2K_\varepsilon}{1+K_\varepsilon}\sum_{n=1}^{\infty}h^{n-1}\operatorname{erfc}\left[\frac{x-L+(2n-1)K_aL}{2\sqrt{D_2^{eff}t}}\right], \qquad (1)$$

$$K_\varepsilon = \sqrt{\frac{D_1^iR_1}{D_2^iR_2}},\quad K_a = \sqrt{\frac{D_2^{eff}}{D_1^{eff}}},\quad h = \frac{1-K_\varepsilon}{1+K_\varepsilon},\quad D_i^{eff} = \frac{D_i^i}{R_i},\quad \operatorname{erfc}(x) = \frac{2}{\sqrt{\pi}}\int_x^{\infty}e^{-x^2}dx,$$

where R_i is a delay coefficient in i medium, D_i^i is internal diffusion coefficient in the i medium, C_i is radionuclide concentration in pore moisture of the i medium, L is container wall thickness.

The calculation was performed for ^{137}Cs, which is explained by its significant percentage content in the waste, long half-life and relatively large diffusion mobility in concrete.

Figure 1 shows how ^{137}Cs concentration in the container wall is changing under such conditions.

The calculation was performed for 150 mm thick concrete wall with D^{eff} amounting to $3{\cdot}10^{-13}$ m^2/s, whereas D^{eff} for sand accounts for 10^{-10} m^2/s. Non exceedance of IL^{water} beyond the container was assumed as a safety criterion. The horizontal line in the figure corresponds to the IL^{water} for ^{137}Cs being equal to $1.1{\cdot}10^4$ Bq/m^3. The graph in Figure 1 shows that NZK containers made of concrete with D^{eff} of $3{\cdot}10^{-13}$ m^2/s would ensure the compliance with the selected safety criterion for ^{137}Cs, thus, cesium concentration on the outer wall of the container will never reach the intervention level.

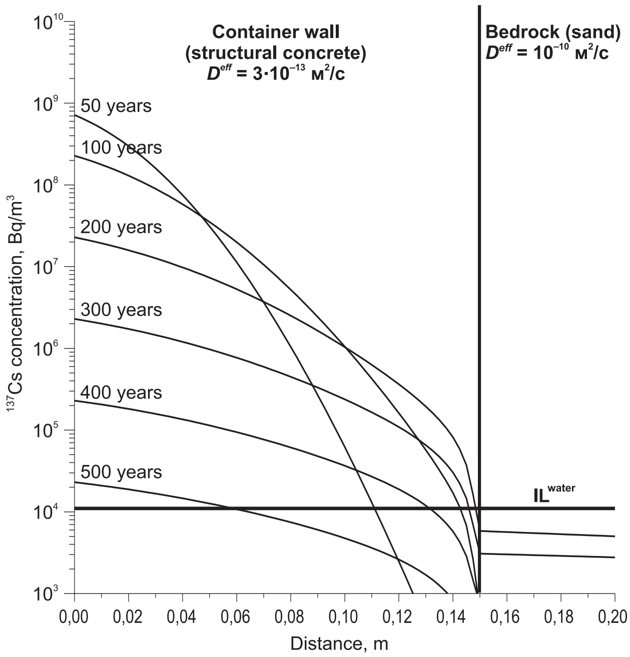

Figure 1. Changing concentration of ^{137}Cs contained in the pore water of a concrete 0.15 m thick container wall

Durability of the packages under storage conditions

Package durability depends both on the properties of the container's structural material and relevant service conditions.

It was proposed to consider the durability of the packages under storage and disposal conditions in a separate manner, since these management stages differ significantly in terms of the natural factors involved.

Table 6. Impacts produced on containers under storage and disposal conditions

NZK management	Storage and disposal conditions	Types of impacts on NZK containers
Storage in unheated surface structures with natural or forced ventilation	Time period: from 1 year to 50 years. Zero temperature crossing — twice per year	Gas corrosion (carbonization). Frost deterioration
Disposal in near-surface RW disposal facilities	Time period: from 250 to 300 years. Temperature: always above 0 °C Medium: weakly permeable and sedimentary rocks (clay, loam, limestone)	Leaching corrosion. Chemical interaction between cement stone and aggressive agents. Accumulation of crystalline neoplasms in concrete pores and capillaries

Table 6 presents the generally accepted NZK container management flow chart, storage and disposal conditions and the impacts produced on the containers.

Gas corrosion (carbonization) and frost deterioration are considered as the main types of impacts produced on concrete containers.

Carbonization

Concrete carbonation involves an interaction of carbon dioxide CO_2 with calcium hydroxide $Ca(OH)_2$ resulting in the formation of calcium carbonate $CaCO_3$ occurring in the surface layer of concrete.

Calcium carbonate solubility, ceteris paribus, is about 100 times lower than the one of calcium oxide hydrate. Therefore, the carbonization of concrete leads to a significant increase in its resistance to the development of Type 1 corrosion. However, during such carbonization, the alkalinity of concrete decreases resulting in an increased corrosion of the reinforcement.

Calculations showed that in 50 years the depth of concrete carbonization will amount to some 0.6—1.3 cm. Container design provides for a 3 cm thick concrete layer to protect the reinforcement from corrosion.

Freeze-thaw resistance

The class of concrete specified according to its freeze-thaw resistance is identified based on the set number of alternate freezing and thawing cycles carried out according to particular methods.

Tests showed that freeze-thaw resistance of the developed concrete composition exceeded F400. After 400 cycles, the samples were removed from the test, because the design limits were exceeded.

Durability of disposal packages

Concrete and reinforcement containers under the groundwater exposure conditions are considered as the main types of impact produced on concrete containers during disposal.

Three main types of corrosion can be distinguished.

The first group (corrosion of type I) involves all the corrosion processes that can occur in the concrete being exposed to water with low hardness, when the components of the cement stone dissolve and are carried away by the water flow.

To confirm this conclusion on the durability of concrete and to obtain quantitative estimates of Type 1 corrosion rate, the following parameters can be calculated: the leaching rate of lime, the allowable water filtration coefficient or the time period during which the concrete can maintain its strength if the water is seeping through it or the concrete surface is washed by the water flow. Below is presented a simplest option allowing to calculate the service life of concrete and reinforced concrete structures being exposed to water flow under pressure, thus, enabling to evaluate the hazard level associated with this type of corrosion.

Table 7 presents the flow chart allowing to calculate the durability of container concrete subject to corrosion of type I.

As an example, let's focus on a case study suggesting that RW container is installed into a bedrock with the following parameters of the ground water flow: hydraulic slope (gradient) — 0.013 m/m, filtration coefficient for the outer environment — 730 m/year (sandy loam), filtration rate — 9.49 m/year. These parameters can be considered as typical for the soils pertaining to the site of the Leningrad branch of FSUE RosRAO's North-Western Territorial Division.

Calculation of concrete durability with a water resistance assumed as W6, filtration coefficient of $2{\cdot}10^{-9}$—$7{\cdot}10^{-9}$ cm/s, showed that the service life of concrete suggesting that no loss of its basic technical properties occurs can range from 450 to 1,600 years. This indicates that at small hydraulic slopes, being considered as quite common under the suggested disposal conditions, filtration flows in then concrete would be virtually absent, and, as the calculations show, the leaching of lime produce no impact on the service life of concrete.

Corrosion types II and III involves processes that can evolve in the concrete under the impact of water containing some aggressive agents interacting

Table 7. Flow chart allowing to calculate the durability of container concrete under the impact of Type 1 corrosion

Calculation steps	Calculation Model or Formula	Limitations or Parameter Values
1 Calculating the amount of water filtered through NZK container wall	$V = K_f \cdot \Delta H/X$ K_f – filtration coefficient; ΔH – hydraulic gradient; X – wall thickness	ΔH – assumed to be equal to 0,013 m/m; X – 0.15 m
2 Calculating the amount of lime that can be removed	$q = k \cdot Ц \cdot ά$ k – leakage percentage for CaO; Ц – cement content in concrete; ά – CaO content in cement	k – assumed to be equal to 10 %; Ц – assumed to be equal to 0.4 g/cm^3; ά – for Portland cement can be taken equal to 0.65
3 Calculating the durability of concrete NZK walls	$T = q/V \cdot C$ C – average concentration of lime in water	C – assumed to be equal to 1.2 g/l

with the constituent parts of the cement stone. These impacts result in the accumulation of soluble salts in pores, capillaries and other voids present in the concrete. The process is enhanced by the cyclical effect of mineral salt solutions, when the saturation of concrete with a solution alternates with its drying.

For NZK containers, these processes can be excluded from consideration since the site proposed for repository construction should be selected based on the characteristics (aggressiveness) of groundwater, thus, the processes resulting in concrete deterioration due to Type II or III corrosion can be ruled out. According to JSC “Construction Research Center” NIIIZhB named after A. A. Gvozdev, being in the solid state, the salt melt poses no threat to the performance of concrete Class B50 with a water resistance of W12. In keeping with relevant provisions of SP 28 13330-2017, the environment is considered as being not aggressive to the concrete.

As it comes to the radiation resistance of the packages, it can be noted that purpose studies were carried out. These have shown that Portland cement-based concrete can be considered as a radiation-resistant material with a total absorbed dose of γ–radiation of at least $1.95 \cdot 10^7$ Gy [10]. In [11] it was shown that the durability of cement compounds can be maintained under the values of up to 10^8 Gy.

Durability of the metal insert

During at least 50 years, corrosive effects produced by the media on the inner and outer parts of the containers under consideration will have a primary influence on the durability of metal containers with salt melt or built-in metal insert when emplaced into NZK-150-1.5P non-returnable protective container.

Exteriorly to the metal container, the environmental conditions of operation involve a sealed reinforced concrete container ensuring no free access of oxygen and water with an alkaline medium available on the concrete surface.

Literature sources indicate that if no oxygen is available in the solution and the pH is greater than 11, carbon steel is practically not suspectable to corrosion. According to the data, even if carbon steel is placed into aerated fresh water with a pH level of 12, the corrosion rate will be about 0.02 mm/year, which corresponds to only 1 mm if calculated for 50 years.

Salt melt placed inside a metal container or metal insert is characterized with high inhibitory properties with respect to structural steel which is due to the presence of sodium hydroxide (NaOH), trisodium phosphate (Na_3PO_4), sodium teraborate ($Na_2B_4O_7$) and alkaline pH.

A survey of actual container state (metal drum) containing salt melt from Novovoronezh NPP, performed at the Central Research Institute of CM "Prometey", showed that the internal corrosion ranged from 0.01 to 0.02 mm/year, which will amount to some 0.5—1 mm over 50 years of storage. It was also found that the presence of brine or softened salt product in the container does not cause an increase in the corrosion rate of steel given that the container does not lose its structure integrity.

Thus, under the corrosion rates presented above and a wall thickness of a metal container of 4 mm, the durability of the container given certain margin is ensured during the entire storage period, namely 50 years.

Conclusions

Comparison of methods used to manage conditioned waste at NPPs with VVER-1000 type reactor units has shown that evaporation of such waste to salts and salt melt emplacement into NZK containers can be considered as the most cost-effective solidification method.

Calculation, analytical and experimental evaluations were performed aiming to demonstrate the disposal safety for a salt melt resulting from LRW treatment at nuclear power plants with VVER-1000 reactor units. Under these evaluations it was assumed that the drums with the salt melt are emplaced into NZK-150-1.5P containers and NZK-150-1.5P(C) containers are used for direct salt melt emplacement. The calculations showed that the NZK packages with the salt melt meet the requirements specified for the packages intended for RW class 3 disposal.

The developed designs of NZK-150-1.5P and NZK-150-1.5P(C) containers and the composition of the concrete used to manufacture the container body provide mechanical strength under given static and dynamic loads, which has been confirmed by laboratory tests of concrete and certification container testing.

The high insulating ability of such packages is assumed to be due to the high quality of the structural material — concrete. Radionuclides release from the package is possible only if driven by diffusion processes. Due to low diffusion coefficient, determined based on relevant experiments, the release of cesium from the package is ruled out provided that the disposal conditions are selected correctly.

Insulating ability for NZK-150-1.5P containers being maintained for 300 years was confirmed by calculations aimed at demonstrating the durability of the packaging under storage and disposal conditions with experimentally determined high level of freeze-thaw resistance (F400) and low water resistance (W18) of the structural material.

Service life of metal containers with salt melt placed into NZK-150-1.5P containers will amount to over 50 years, which is due to favorable storage conditions. The safety of NZK disposal packages with salt melt can be ensured by a correct choice of disposal conditions excluding the corrosion of concrete container walls given all corrosion types:

- absence of groundwater (underground water) flow in the near zone;
- absence of acids and salts being considered aggressive to concrete in the groundwater (underground) waters within the repository site area.

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