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ENVIRONMENTAL EFFECTS OF CAVITATION TECHNOLOGY FOR RADIOACTIVE WASTE MANAGEMENT

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Results from studies of the removal from storage and solidification of low- and intermediate-level liquid radioactive waste in inorganic binders (cementation) are presented. Cavitation technology for recycling radiochemical wastes from nuclear power plant decommissioning increases the extraction throughput and strength of the cement compound.

Keywords: *cement compound, cavitation technology, solid precipitates, dissolution, spent nuclear fuel.*

According to the Joint Convention on the Safety of Radioactive Waste Management, people and the environment must be effectively protected from the harmful impacts of radionuclides and ionizing radiation [1–5]. New technologies for managing radioactive wastes must be improved and developed and special methods for ensuring the safety of humans and the biosphere must be applied because of their specific nature although the amount of radioactive wastes is vanishingly small (~0.5% of all industrial wastes). Long-term storage and burial are the most important stages of managing radioactive wastes with respect to long-term safety. A legal and organizational system for ensuring and regulating nuclear and radiation safety was created in Russia [4, 6].

Incorporation into inorganic binders (cementation) is currently the most widely used process for solidification of low- and intermediate-level liquid radioactive wastes (LRWs). The resulting product typically has high mechanical strength and is non-combustible and stable to radiation and chemicals. External radiation from cemented materials is weak because of their high density.

The increased safety requirements for storing wastes from spent nuclear fuel (SNF) reprocessing are responsible for the need to use fundamentally different science-based technologies, in particular, cavitation technology, which is rather easily implemented, energy efficient, and the only alternative in several instances [7, 8].

The forms and structural features of SNF have been discussed [9]. Practice showed that long-term storage of LRW led to the accumulation of solid precipitates, i.e., sludge. This waste form is the most complicated with respect to reprocessing and recycling and requires the solution of several technical problems, including:

1) evaluation of the effect of temperature on the kinetics of physicochemical processes in the solidifying cement mass and, eventually, on the quality of the obtained concrete;

2) determination of the effect of the physicochemical properties of cavitation-activated water on the quality of the obtained cement compound.

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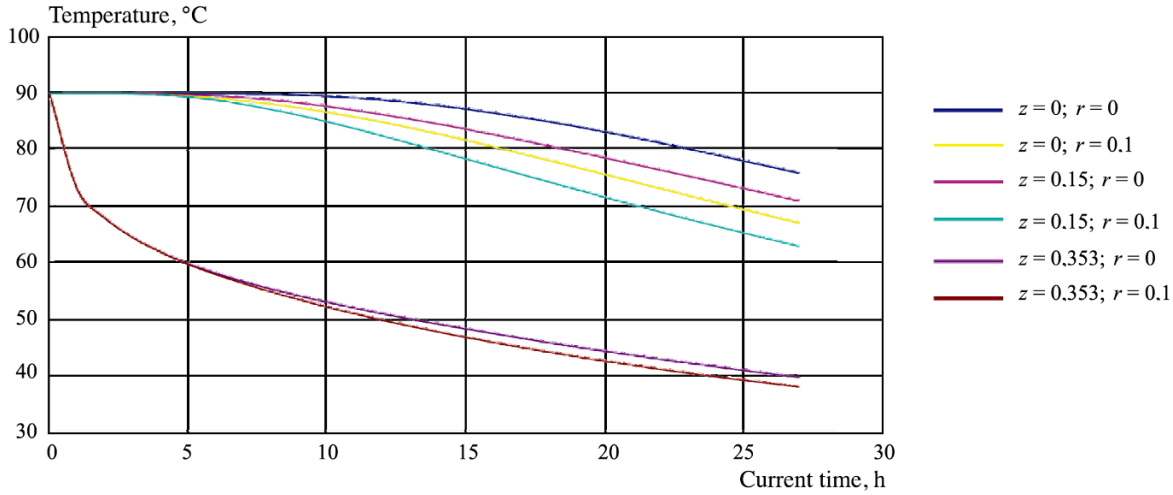


Fig. 1. Time dependences of temperature at various tank points with water cooling and measuring points distributed according to Fig. 2: z, distance from tank bottom along the vertical, m; r, radius of measuring point positioning, m.

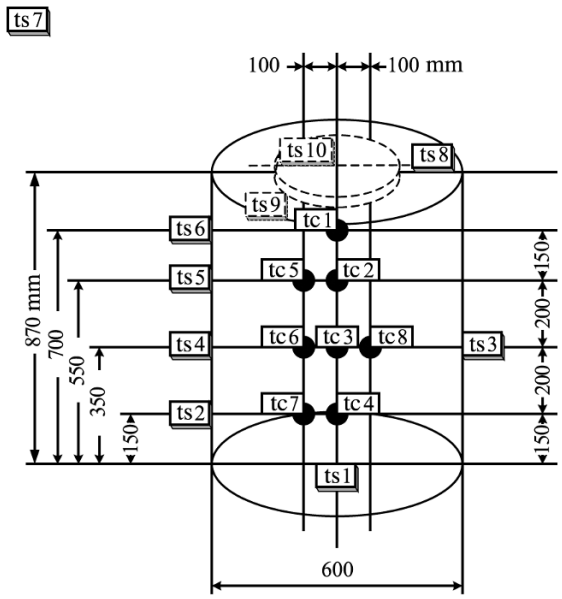


Fig. 2. Diagram of spatial positioning of sensors in tank volume and on its outer surface.

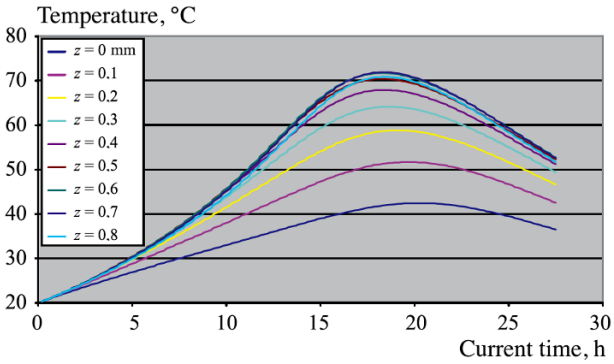


Fig. 3. Time dependences of temperature at various points of the tank during hardening of the cement compound.

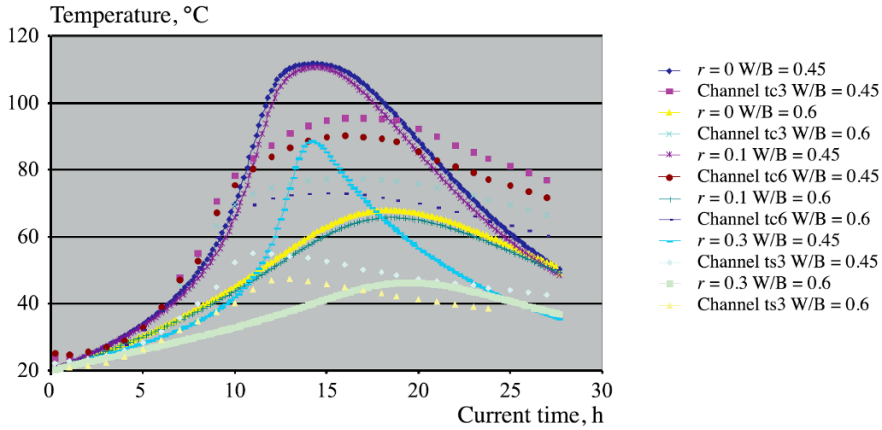


Fig. 4. Calculated and experimentally measured temperatures during hardening of the cement compound at the hottest point inside the tank (tc3), peripheral point tc6, and tank side surface point ts3 for various water-binding ratios.

Numerical study of the temperature field in a standard 200-liter SNF storage tank. The goal of the present heat calculation was to determine the temperature field in a tank with a cement compound during its solidification and subsequent storage. The characteristic features of these processes are:

1) solidification of the cement compound in the tank in the examined time interval is non-equilibrium with respect to heat transfer because the temperature field changes continuously as heat is released during the cement hardening;

2) heat exchange to the environment is non-steady state because of the continuously changing temperature of the heat-exchange surface; and

3) heat exchange of the tank with the environment during long-term storage becomes steady-state because of heat released during radioactive decay of radionuclides contained in the cement compound. A mathematical model and calculation algorithm were published [10].

A comparison of the cooling of water and the cement compound led to the conclusion that both processes obeyed analogous power laws although the cement compound cooled significantly faster than water. According to the temperature profiles in various cross sections of the tank (Fig. 1), cooling was characterized by significant temperature gradients between the central points and points on the heat-transfer surface. Heat transfer was limited by the thermophysical characteristics of the tank contents. Figure 2 presents a diagram of the spatial positioning of sensors within the tank and on its outer surface.

The results (Fig. 3) indicated that the tank inner volume was heated to 70°C and greater (also noted earlier in practice). Central regions were 25–30°C warmer than the peripheral ones. This indicated that the overheating had to be reduced to ensure the cement compound was uniform and of high quality.

The following materials were used according to the experimental program for selecting the composition of the cement compound and the cementing process parameters for preparing the cement compound:

- Portland cement TsEM I 42.5B, GOST 31108-2003 (PTs 500-D0 according to the previous classification) (Mordovtsement);
- sorption additive of ground clinoptilolite from Kholinskoe deposit of fractional composition (wt.%): >1 mm, 2.55; up to 0.5 mm, 13.74; up to 0.25 mm, 12.20; up to 0.125 mm, 34.19; up to 0.1 mm, 15.23; up to 0.05 mm, 20.03; and <0.05 mm, 2.06;
- tap water (technical) and cavitation-activated water (Table 1); and
- insoluble hydroxide sludges.

The cement solution composition was adjusted after the batch was sampled and the flowability of the cement solution was determined (using a Suttard viscometer).

The following procedure was used to mix the components during sample preparation. Clinoptilolite with hydroxide sludge was placed into plastic beakers in given ratios and stirred for 15 min. The calculated amounts of cement and water were

TABLE 1. Physicochemical Parameters of Cavitation-Activated Water with Various Volume Fillings of a Supercavitating Reactor (SC-reactor)

Volume filling of SC-reactor, V, %	Parameters									
	T, °C		CDO, %sat.		S, Ω^{-1}/cm		pH		E, mV	
	α = 10°	α = 20°	α = 10°	α = 20°	α = 10°	α = 20°	α = 10°	α = 20°	α = 10°	α = 20°
100 (before activation)	20.7		101.2		0		5.4		200	
100	40.5	32.6	85.1	96.65	14.5	6	5.65	5.45	133	168.5
65	34.6	27.2	87.65	95.7	5.5	13	6.0	5.7	128.5	147
CDO, concentration of dissolved oxygen; α, top angle of slanted cavitator of SC-reactor; E, redox potential; S, electrical conductivity.										

TABLE 2. Distribution of Solid-Phase Aluminosilicate Components between Solutions Used to Treat It

Composition of solutions used to treat sludge	Degree of dissolution, %			
	test 1		test 2	
	Al	SiO_2	Al	SiO_2
100 g/iter NaOH	2.52	1.81	5.79	4.34
H_2O	2.04	0.70	2.36	0.85
HNO_3 + CPW	1.39	8.57	2.50	15.43
H_2O	0.56	0.57	0.64	0.64
100 g/iter NaOH	8.88	2.74	16.87	6.30
H_2O	5.54	0.94	6.34	1.15
HNO_3 + CPW	1.22	8.65	2.01	14.96
H_2O	0.32	1.01	0.36	1.14
100 g/iter NaOH	4.72	2.57	9.48	5.04
H_2O	6.60	1.28	7.34	1.68
HNO_3 + CPW	1.36	5.54	2.31	9.19
Total	35.6	34.39	56.03	60.72

added. The mixture was thoroughly stirred and poured into fluoroplastic forms for producing samples of diameter 20 mm and height 20 mm. After the mixture set, the forms with the samples were placed into a chamber for normal hardening at 20–30°C with relative humidity 95 ± 5% for 24 h. The samples were removed from the forms and replaced into the chamber for normal hardening for 28 days. Then, the samples were tested for mechanical strength, water resistance, and freeze-thaw resistance. The sample characteristics were determined from three tests to ensure that the results were comparable and accurate.

The effects of hydrodynamic cavitation on the water temperature, concentration of dissolved oxygen (CDO), pH, electrical conductivity, redox potential (ROP), etc., were studied. Results for the changes of these characteristics of cavitation-activated distilled water with the cavitation number constant at $\chi = 0.05$ and regression analysis allowed a reasonable cavitation treatment time to be established.

Cavitation technology to extract sludge from SNF tanks. Tests were conducted using several samples of slightly soluble hydro-aluminosilicate solids [11]. A thermostat (a given working temperature was maintained to less than ±3°C) was used to treat the sludge at elevated temperature. Working solutions were prepared from pure and chemically pure chemical reagents.

The volumes of the solid and liquid phases were measured after the suspension stood for 1 day.

TABLE 3. Tests of Cement Compound Samples

Sample No.	Starting compression strength, MPa	Average, MPa	Strength after freeze-thaw resistance tests, MPa	Average, MPa	Change of strength, %
0-1	36.0	34.2	32.8	29.8	–12.9
0-2	32.5		26.8		
1-1	27.4	27.5	21.3	21.6	–21.5
1-2	27.7		22.0		
2-1	39.2	42.2	50.7	42.4	0.5
2-2	45.2		34.1		
3-1	52.2	47.6	52.9	48.4	1.7
3-2	43.0		44.0		

In instances where the liquid phase did not clarify in this time, the standing time was increased or the solution was filtered through "blue ribbon" filter paper.

Analytical samples of the liquid phase were obtained by decantation.

The experimental results (Table 2) showed that the yield of dissolved sludge components reached 35.6% Al and 34.39% SiO_2 if the traditional technology was used (test 1) whereas use of cavitation-activated water (test 2) gave 56.03% and 60.72%, respectively.

It could be assumed based on the results that use of cavitation technology to leach and dissolve the model sludge solid phase could increase the reprocessing efficiency of highly active wastes and free their storage tanks of accumulated precipitates without increasing the concentrations of chemical reagents and the temperature. This would enable the corrosion potential to be reduced [9, 11].

Cavitation technology to prepare the cement compound. A series of experiments determined how the physicochemical properties of water were affected by cavitation with the maximum effect, i.e., with the cavitation number at a minimum. Figure 3 shows experimental temperatures during hardening of the cement compound at three points, i.e., the hottest point inside the tank tc3 (Fig. 2), peripheral point tc6, and tank side surface point ts3, for various water/binder (W/B) ratios (point designations are the same as the published measuring diagram [12]). The calculated functions are given for these same points.

The calculations and experiments agreed best (Fig. 4) for W/B = 0.6, where the temperature maxima differed by 6°C. The maximum calculated temperature for W/B = 0.45 was 15°C greater than the experimental value. The biggest difference (40°C) was observed for the calculated and experimental temperatures on the tank side surface at point ts3. Obviously, peculiarities of the hardening of the cement compound at temperatures close to the boiling point of water were not considered in the model. However, thermal calculations of the tank using the aforementioned model could be used for W/B ≥ 0.5 with an accuracy sufficient enough for technical work.

Mechanical strength of the samples was determined using the branch instruction "Cement Compounds for Radioactive Wastes." Table 3 presents test results for the mechanical strength of the compound after 28 days and the freeze-thaw resistance.

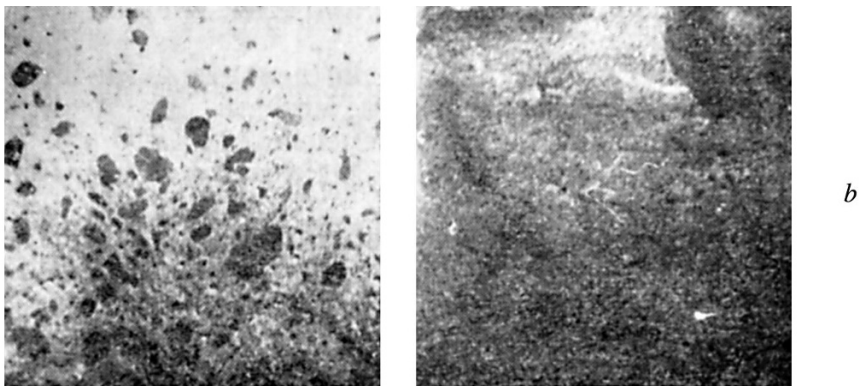
The water/cement (W/C) and W/B parameters were 0.5 and 0.41, respectively (W/B = W/(C + K-factor), K-factor is determined upon adding a thin filler according to the corresponding regulations).

The measured Cs-137 leaching rate (Table 4) was less than the established values given in regulations of the atomic sector.

The increased specific surface area of the cement directly in the aqueous medium after cavitation dispersion enabled its potential properties to be used more fully and its degree of hydration and the surface energy of its particles to be increased. Destruction of the primary large-grained aluminate cement structure produced finely crystalline cement rock, the strength of which was 2–3 times greater (Fig. 5) than that of cement rock obtained from a solution prepared in an ordinary mixer.

TABLE 4. Cs-137 Leaching Rate

Sample No.	A_0, Bq/g	R_n, g/(cm^2·day), for number of days							
		1	3	7	10	14	21	28	56
0-1	5850	$1.03\cdot10^{-2}$	$2.47\cdot10^{-3}$	$2.93\cdot10^{-4}$	$2.00\cdot10^{-4}$	$1.88\cdot10^{-4}$	$1.26\cdot10^{-4}$	$9.46\cdot10^{-5}$	$4.89\cdot10^{-5}$
0-2	980	$9.4\cdot10^{-2}$	$2.78\cdot10^{-2}$	$3.04\cdot10^{-4}$	$2.17\cdot10^{-4}$	$1.55\cdot10^{-4}$	$1.33\cdot10^{-4}$	$9.01\cdot10^{-5}$	$5.67\cdot10^{-5}$
1-1	1840	$6.12\cdot10^{-2}$	$1.51\cdot10^{-2}$	$2.01\cdot10^{-3}$	$1.13\cdot10^{-3}$	$8.08\cdot10^{-4}$	$5.77\cdot10^{-4}$	$4.95\cdot10^{-4}$	$2.52\cdot10^{-4}$
1-2	3840	$6.69\cdot10^{-2}$	$3.29\cdot10^{-2}$	$1.87\cdot10^{-3}$	$1.16\cdot10^{-3}$	$8.47\cdot10^{-4}$	$5.83\cdot10^{-4}$	$5.09\cdot10^{-4}$	$2.34\cdot10^{-4}$
2-1	2050	$5.07\cdot10^{-2}$	$2.82\cdot10^{-2}$	$1.83\cdot10^{-3}$	$1.45\cdot10^{-3}$	$7.03\cdot10^{-4}$	$6.18\cdot10^{-4}$	$5.23\cdot10^{-4}$	$2.55\cdot10^{-4}$
2-2	3150	$7.95\cdot10^{-2}$	$4.36\cdot10^{-2}$	$2.16\cdot10^{-3}$	$1.67\cdot10^{-3}$	$7.31\cdot10^{-4}$	$6.62\cdot10^{-4}$	$5.18\cdot10^{-4}$	$2.71\cdot10^{-4}$
3-1	3680	$7.34\cdot10^{-2}$	$3.78\cdot10^{-2}$	$2.86\cdot10^{-3}$	$1.91\cdot10^{-3}$	$1.81\cdot10^{-3}$	$7.97\cdot10^{-4}$	$6.96\cdot10^{-4}$	$3.20\cdot10^{-4}$
3-2	1400	$1.22\cdot10^{-2}$	$6.07\cdot10^{-2}$	$3.45\cdot10^{-3}$	$2.13\cdot10^{-3}$	$1.74\cdot10^{-3}$	$8.41\cdot10^{-4}$	$7.19\cdot10^{-4}$	$3.57\cdot10^{-4}$



a *b*

Fig. 5. Structure of a block of cement rock (W/C = 0.5, cement M300): *a*) without cavitation activation (bending strength σ_b = 3.6 MPa, compression strength σ_c = 17.3 MPa); *b*) t_{tr} = 60 sec (relative number of cavitations χ/α = 0.34, σ_b = 9.6 MPa, σ_c = 38.1 MPa).

Conclusions

1. It was found experimentally that cavitation-activated water could be used to increase the yield of slightly soluble sludge components to 56.03% for Al and 60.72% for SiO_2 (35.6% and 34.39%, respectively, for the traditional technology). Use of cavitation technology to reprocess nuclear-energy wastes could provide uniform extraction of sludge components, avoiding the accumulation of fissionable materials and reducing the corrosion potential of tank construction elements.

2. The average heat-transfer coefficient for the whole cooled surface of the storage tank for the cement compound was determined experimentally as 9.86 W/(m^2·K). This was almost twice the value calculated using the well-known criterial equation.

3. The results for SNF compounding supplemented previous results [12]. Use of cavitation technology was advisable for recycling radiochemical wastes and helped to increase the strength of the cement compound and to improve the freeze-thaw resistance and leaching rate of radionuclides from the cement matrix (this is one of the main factors determining the reliability of long-term storage and burial of radioactive wastes).

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