Radioactive Wastes Cementation during Decommissioning Of Salaspils Research Reactor - 9188

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ABSTRACT

This paper deals with information on the radioactive wastes cementation technology for decommissioning of Salaspils Research Reactor (SRR). Dismantled radioactive materials were cemented in concrete containers using tritiated water-cement mortar. The laboratory tests system was developed to meet the waste acceptance criteria for disposal of containers with cemented radioactive wastes in near-surface repository “Radons”. The viscosity of water-cement mortar, mechanical tests of solidified mortar’s samples, change of temperature of the samples during solidification time and leakage of Cs-137 and T-3 radionuclides was studied for different water-cement compositions with different additives. The pH and electro conductivity of the solutions during leakage tests were controlled. It was shown, that water/cement ratio significantly influences on water-cement mortar’s viscosity and solidified samples mechanical stability. Increasing of water ratio from 0.45 up to 0.62 decreases water-cement mortar’s viscosity from 1100 mPas up to 90 mPas and decreases mechanical stability of water-cement samples from 23 N/mm^2 to the 12 N/mm^2. The role of additives – fly ash and Penetron admix in reduction of solidification temperature is discussed. It was found, that addition of fly ash to the cement-water mortar can reduce the solidification temperature from 81 ^{o}C up to 62^{o}C. The optimal interval of water ratio in cement mortar is discussed. Tritium and Cs-137 leakage tests show, that radionuclides release curves has a complicate structure. The possible radionuclides release mechanisms are discussed. Experimental results indicated that addition of fly ash result in facilitation of tritium and cesium leakage in water phase. Further directions of investigations are drafted.

INTRODUCTION

The decommissioning of Salaspils research reactor is connected with the treatment of 2200 tons different materials. The largest part of all materials (≈ 60 % of all dismantled materials) is categorized as low level radioactive wastes. Dismantled radioactive materials were cemented in concrete containers using water-cement mortar. According to elaborated technology, the tritiated water (150 tons of liquid wastes from special canalization tanks) was used for preparation of water-cement mortar [1]. Such approach excludes the emissions of tritiated water into environment and increases the efficiency of radioactive wastes management system for decommissioning of Salaspils research reactor. The Environmental Impact Assessment studies for Salaspils research reactor decommissioning (2004) [2] and for upgrade of repository “Radons” for decommissioning purposes (2005) [3] induced the investigations of radionuclides release parameters from cemented radioactive waste packages. These data are necessary for implementation of quality assurance demands during conditioning of radioactive wastes and for safety assessment modeling for institutional control period during 300 years [4].

DESCRIPTION OF THE ACTUAL WORK

The laboratory tests system was developed to meet the waste acceptance criteria for disposal of containers with cemented radioactive wastes in near-surface repository “Radons”. The viscosity of water-cement mortar, mechanical tests of solidified mortar’s samples, change of temperature of the samples during solidification time and leakage of Cs-137 and T-3 radionuclides was studied for different water-Portlandcement compositions with different additives. The commercially available Portlandcement CEM I 42.5, fly ash and Penetron admix and deionized water was used for experiments. The mortar was prepared using programmed mixer, then filled in samples holder for preparation of cubic shape samples with size 40 mm x 40 mm x 40 mm. After fulfillment, the sample holders were inserted in solidification facility for solidification during 28 days (or more) at temperature $20^{o}C \pm 1^{o}C$ and humidity > 90%. Solidified samples were used for mechanical tests. The leakage tests were performed using the methodology described in [5]. Tritium radioactivity in water phase was measured with liquid scintillation counters, but Cs-137 radioactivity in water phase was measured with gamma spectrometer with semiconductor probe [6]. Temperature measurements were performed by using on PC based measuring system with thermo resistance probes. The electroconductivity, pH and concentration of Ca^{2+} in leaching solutions were measured.

RESULTS

Radioactive wastes for final disposal in the vaults of repository "Radons" must be cemented in cubic concrete containers with size 1.2m x 1.2m x1.2m. Disposal technology includes the stabling of 4 levels of containers in the vaults. It defines, that containers with cemented radioactive wastes must be mechanically stable for 300 years period without formations of cracks and damages. The mechanical tests of containers were performed in 2004 (Fig.1) to

Fig. 1. Mechanical tests for containers with cemented wastes.

define the suitability of containers for mechanical stresses up to 25 tons (lower container). Therefore, the cementation's process of solid radioactive wastes in concrete containers must be performed carefully to avoid decreasing of mechanical stability of container with cemented wastes.

Mechanical stability of cemented radioactive wastes

The basic factors, which influence on the mechanical properties of prepared radioactive wastes packages, were identified as following:

- Composition of mortar. It is very important factor since mechanical stability of water-cement rock significantly depends on water/cement ratio and additives. The sample test's results are given in Table I. Experimental data confirmed , that increase the water

Table I. Viscosity of mortar and mechanical stability of water/cement rocks.

No.	Water/ cement ratio	Penetron admix, %	Fly ash/ cement ratio	Dynamic viscosity of mortar, mPas	Mechanical stability of samples *, N/mm^2
1	0.62			74.5	15.0
2	0.55			86.4	18.5
3	0.51			148.2	22.0
4	0.44			1198.1	30.0
5	0.62	2		66.7	21.1
6	0.55	2		93.1	18.9
7	0.51	2		147.4	19.1
8	0.44	2		1247.8	32.0
9	0.61		0,33	210.6	20.9
10	0.55		0,33	168.7	25.6
11	0.51		0,33	945.2	33.4
12	0.44		0,33	inflowing	33.8

*-for samples with size 4cm x 4cm x 4cm after 28 days solidification.

content in sample caused reduction of mechanical stability of water/cement rock. Changes of the water content from 0.44 up to 0.62 decreases mechanical stability 2 times. The additives – flay ash and Penetron admix increased the mechanical stability of samples up to 25 % depending on the water content in sample. Thus, for samples with water content 0.61-0.62 both additives increased mechanical stability from 15 N/mm^2 to 21 N/mm^2 . Observed effect of additives gives a possibility for more effective utilization of tritiated water from Salaspils research reactor's special canalization for solid radioactive wastes cementation processes. It gives a possibility to increase up to 20% of tritiated water load in container without significant decreasing of mechanical stability of cemented packages.

- Viscosity of mortar. This factor has an important role during fulfillment of containers with mortar. According to elaborated technology, the container with solid radioactive wastes (around 4.5 tons weight) is lifted on vibration table and fulfilled with the mortar. Mortar must penetrate in all gaps, tubes and fulfill all empty space to reduce the probability for formation of cavities, bubbles and another macrodefects in the cemented wastes structure. Experimental data show (Table I), that reduction of water content in mortar significantly (by factor 10) increases viscosity of the mortar. It is unpleasant phenomena since increasing of viscosity promotes the formation of macrodefects during cementation of radioactive wastes. Therefore, the optimal water/cement ratio for cementation of radioactive wastes using Portlandcement or it's compositions with additives – fly ash or Penetron admix should be > 0.50.
- Composition of solid radioactive wastes. The sorting of dismantled radioactive wastes must be performed very carefully to avoid the contact of mortar with active metals or another components, which can cause the chemical reactions with formation of gases. The basic problems are connected with cementation of aluminum scraps in concrete containers. In such case proceeds the

formation of hydrogen [7, 8], which creates the porous structure of water/cement rock with inclusions of hydrogen in bubbles. Such structure has low mechanic stability due to increased porosity. To eliminate the formation of increased porosity, the chemically active radioactive wastes are packed in 100 l-200 l drums and cemented in containers together with drum.

- Solidification temperature. Experimental data show (Fig.2.), that during solidification of water/cement matrix proceed heat formation processes. Observed data confirmed, that volume temperature in water/cement cubicle can increase up to 78 ^{o}C. Addition of fly

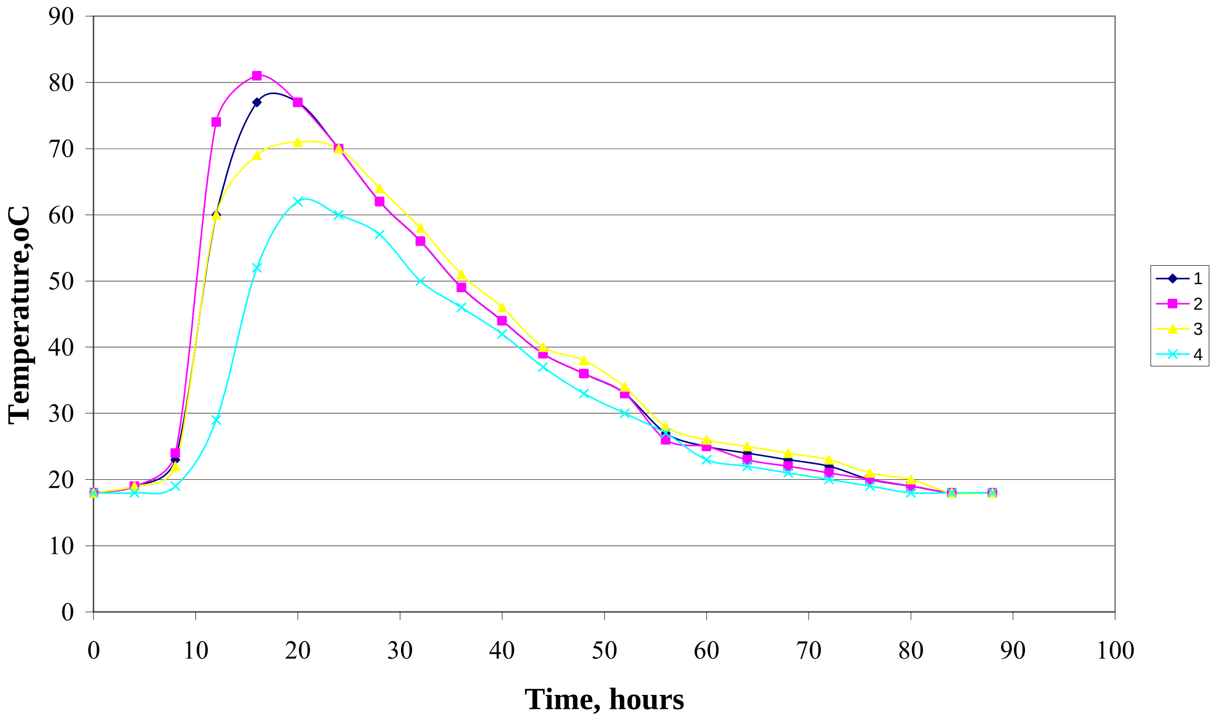

Fig. 2. Temperature measurements during solidification of mortar with water/cement ratio 0.62. 1- water/ Portlandcement ; 2- water/ Portlandcement with 2% Penetron admix; 3- water/ Portlandcement with flay ash, fly ash/ cement ratio 0.33; 4- water/ Portlandcement with 2% Penetron admix and fly ash/ cement ratio 0.33.

ash reduces the temperature up to 70 ^{o}C , but addition of Penetron admix slightly increases the solidification temperature. Increase of the volume temperature during solidification can damage the concrete container and form the cracks in the walls of container. To avoid the damage of container, the container is fulfilled 2 times now. Firstly it is filled up to 50% -60 % of the volume, then after 72 hours it is fulfilled finally. Used technology is not optimal and further investigations are performed for reduction of solidification temperature.

- Radiation stability of composition. Taking into account, that cemented radioactive wastes package must be a barrier preventing migration of radionuclides in environment during 300 years period, additional efforts must be performed to reduce radiation induced effects, which can significantly influence on mechanical stability of packages. It means that materials with low radiation stability can not be acceptable for radioactive wastes cementation. All used additives must have high radiation stability and dismantled radioactive materials with low radiation stability (usually organic substances) can not be packed in containers. It was tested experimentally, that additive Penetron admix has high radiation stability [8] and can be suitable for cementation of radioactive wastes in containers. Therefore, each new component of the mortar must be tested for radiation stability. Materials with low radiation stability generate radiolysis products with high chemical activity –radicals, oxygen, peroxides, which can cause corrosion of container's content and armature and can cause disintegration of cemented package.

Radionuclides leaching tests

Radionuclide leaching tests were performed to fulfill the Waste Acceptance Criteria (WAC) for repository "Radons". According to the WAC, tritium and Cs-137 diffusion parameters must be determined for cemented packages. Leaching tests for both radionuclides were performed according to methodology described in [5]. Experimental results (Fig.3, Fig.4)

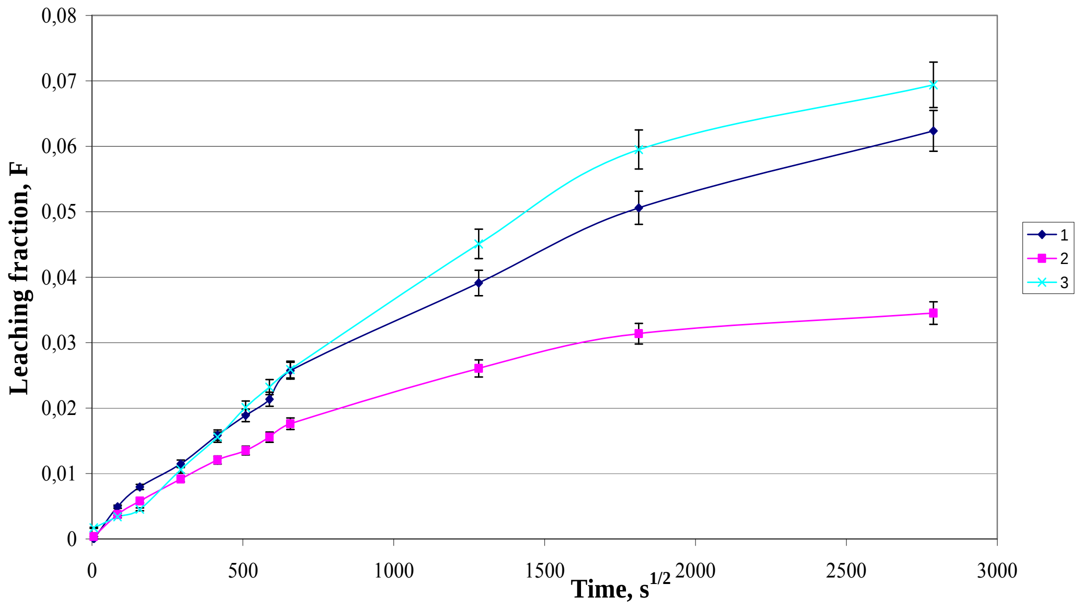

Fig. 3. Cs-137 leaching from cemented samples into deionized water in coordinates $F=f(t^{1/2})$. Water /cement ratio is 0.62. 1- water/ Portlandcement ; 2- water/ Portlandcement with 2% Penetron admix; 3- water/ Portlandcement with flay ash, fly ash/ cement ratio 0.33.

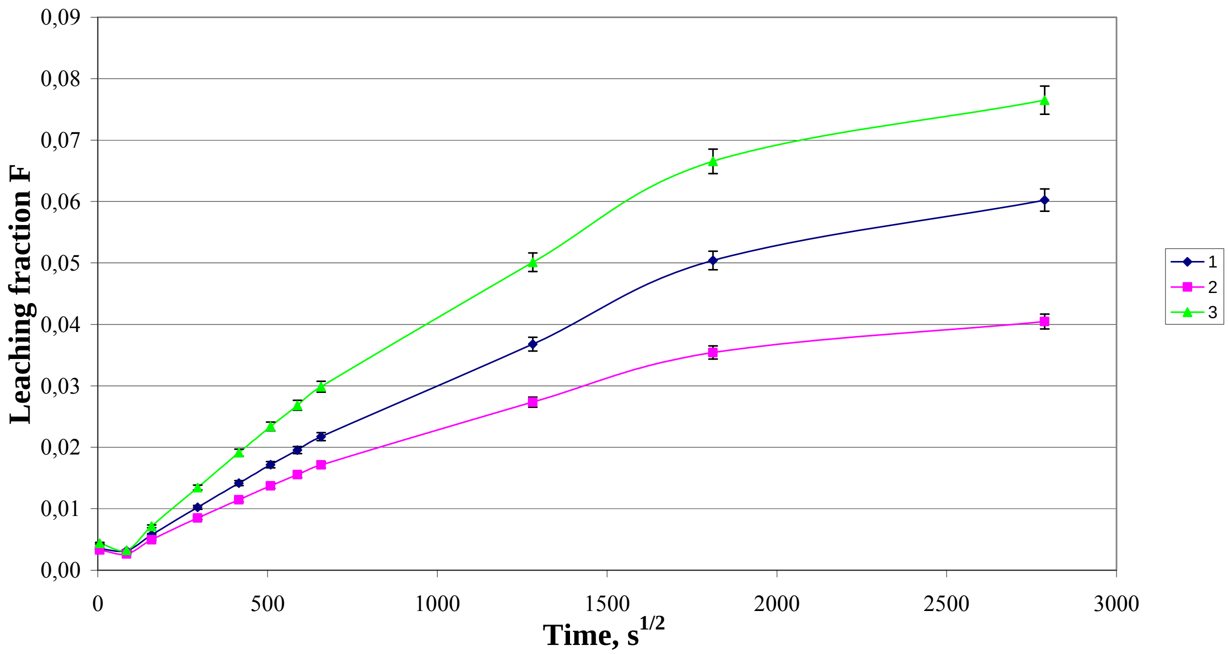

Fig. 4. Tritium leaching from cemented samples into deionized water in coordinates $F=f(t^{1/2})$. Water/cement ratio 0.62. 1- water/ Portlandcement ; 2- water/ Portlandcement with 2% Penetron admix; 3- water/ Portlandcement with flay ash, fly ash/ cement ratio 0.33.

show, radionuclides leaching depends on water/cement rock composition. It was found, that Penetron admix additive significantly decrease tritium and Cs-137 leaching from cemented samples. Addition of fly ash to the water/cement mortar causes the increased radionuclides leaching from cemented samples. It means that fly ash is not suitable for radioactive wastes cementation since additional leaching from cemented wastes packages will increase the irradiation doses for surrounding population. The interpretation of leaching curves in coordinates $F=f(t^{1/2})$ indicates, that leaching curves for both radionuclides are rather complicate and do not fit with linear approximation. It means that leaching process also is complicated including several leaching mechanisms [10-12]. Further investigations are necessary to clarify the leaching mechanisms. It can provide with information on the leaching process during long time period, which is essential for definition of correct leaching parameters and doses calculations. Experimental data show, that increase of pH and electroconductivity proceeds during leaching of radionuclides in deionized water. Obtained leaching curves were used for determination of leaching parameters. Calculated values of diffusion coefficients will be used for safety assessment calculations for repository “Radons”. Additional investigations will be performed for clarification of the leaching of other radionuclides, which dominate in dismantled radioactive materials – Co-60, C-14, Fe-55, etc.

CONCLUSIONS

1. The influence of fly ash and Penetron admix on the mechanical stability of cemented radioactive wastes and radionuclides leaching were studied.

2. It was found, that increasing of water content in water/cement mortar from 0.45 up to 0.65 decreases mechanical stability of cemented samples 2 times. Dynamic viscosity of mortar decreases within factor 10 with increasing of water/cement ratio up to 0.65.
3. It was shown, that solidification temperature depends on addition of fly ash or Penetron admix in the mortar. Fly ash reduces solidification temperature for 16 ^{o}C in comparison with pure water/cement composition.
4. Addition of fly ash to the water/cement mortar increases the leaching of tritium and Cs-137 from cemented samples, but addition of Penetron admix to the mortar significantly reduces the leaching of both radionuclides from cemented samples.
5. It was found, that radionuclides leaching processes includes several radionuclides migration mechanisms. Further investigations are necessary to clarify the leaching mechanisms.

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