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IMMOBILIZATION OF RADIOACTIVE WASTE SLUDGE FROM SPENT FUEL STORAGE POOL

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ABSTRACT

In the last forty years, in FR Yugoslavia, as a result of the research reactors' operation and radionuclides application in medicine, industry and agriculture, radioactive waste materials of the different categories and various levels of specific activities were generated. As a temporary solution, these radioactive waste materials are stored in two hanger type interim storages for solid waste and some type of liquid waste packed in plastic barrels, and one of three stainless steal underground containers for other types of liquid waste.

Spent fuel elements from nuclear reactors in the Vinča Institute have been temporary stored in water filled storage pool. Due to the fact that the water in the spent fuel elements storage pool have not been purified for a long time, all metallic components submerged in the water have been hardly corroded and significant amount of the sludge has been settled on the bottom of the pool.

As a first step in improving spent fuel elements storage conditions and slowing down corrosion in the storage spent fuel elements pool we have decided to remove the sludge from the bottom of the pool /1/. Although not high, but slightly radioactive, this sludge had to be treated as radioactive waste material. Some aspects of immobilisation, conditioning and storage of this sludge are presented in this paper.

Key words: Radwaste, radioactive sludge, immobilisation, cementation, spent fuel storage pool

INTRODUCTION

Since the one of two hangers for temporary radioactive waste storage has been completely filled with radwaste materials that are packed in the metal drums and plastic barrels, and the second one has an effective space for radwaste materials storing only for a few years, attempts are made in the "Vinča" Institute of Nuclear Sciences in developing the immobilisation process for conditioning low and intermediate level radioactive waste materials and their safe disposal into the appropriate disposal system /2,3,4,5/. As an immobilisation process, cementation, for the certain radwaste materials origin and composition are investigated. Developed immobilisation processes have, as a final goal, production of the solidified radwaste-matrix mixture form, that is easy for handling and that satisfies safety and QA requirements for interim storage and final disposal of such materials on the appropriate sites /6,7,8,9,10/. Radwaste materials that were immobilised in the inactive matrices are to be placed into the concrete containers, for the further manipulation and disposal.

CHARACTERISTICS AND QUANTITIES OF RADIOACTIVE SLUDGE IN THE SPENT FUEL ELEMENT STORAGE POOL

In order to estimate storage conditions for the spent fuel elements in the storage pool and characteristics and quantities of the sludge on the bottom of the pool water and sludge samples have been taken on different location in the pool. Analysis of the water from the pool (pH= 8.4 , Conductivity = 446 µS/cm, $[Cl^-]$ = 66 mg/l, [Cu] = 0.05 mg/l, [Zn] < 0.01 mg/l, [Fe] = 0.15 mg/l, $[SO_4]$ = 55 mg/l) shows that the water is highly corrosive to aluminium alloys /11/. Activity concentration of the water from the pool of about 80 - 90 kBq/l of ^{137}Cs, although not of grave concern, is certainly significant, and is incontestable proof that some amount of the fission products are leaking. The activity of ^{137}Cs in the sludge samples is 1.8 ± 0.2 MBq/l /11/.

SLUDGE CONDITIONING AND STORAGE

Total quantity of sludge on the bottom of the RA research reactor spent fuel storage pool is estimated to be about 3 m^3. Estimation were made on the basis of the average sludge height on the bottom of the pool and pool surface. The sludge colour have been a dark red - brown, like a Fe corrosion products. Gamma spectrometry analysis showed that the activity concentration of the sludge is about 1.8 MBq/l ^{137}Cs and about 15 kBq/l ^{60}Co.

Based on the previous experience /6,7,8,9,10/, a technology was developed for sludge immobilisation and conditioning in a cement matrix, inside casks produced using the standard 200-liter metal barrels which have lids supplied with screws. Casks have been produced as containers in standard metal barrels. Thickness of the concrete walls are about 6 – 7 cm. Entire side of the cylindrical concrete wall is a plastic tube with wall thickness of 1 cm, which has been used as a model in forming cylindrical concrete wall. This plastic tube serves as a first barrier in preventing radionuclides leaching from radioactive sludge immobilised in a cement matrix. The bottom cask concrete wall is also 6 – 7 cm thick. In order to prevent or reduce radionuclide leaching, this wall has been covered with epoxy resin. The useful volume of these casks are about 75 l.

The existing pilot cement mixer was reconstructed to enable placing a barrel containing the planned quantity of sludge on its platform without a risk of spilling. A new mechanical manipulator, which provides mixing of the cement matrix with the sludge in the entire volume of the barrel, was constructed. Rooms for conditioning the sludge in a cement matrix, supplied with independent ventilation system, and for storing the casks during the period needed for cement hardening, have been arranged. Arrangement of the modified pilot mixer with concrete container made in metal barrel, placed on the mixer platform with appropriate quantity of sludge are presented in Fig. 1.

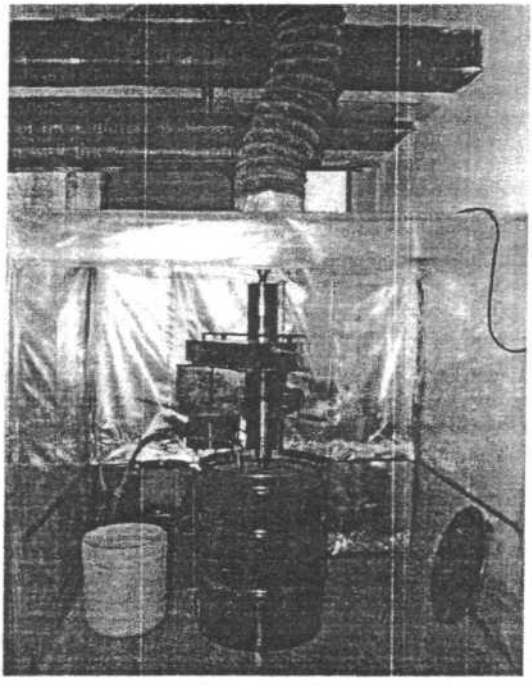

Fig. 1. Arrangement of the modified pilot mixer with concrete container made in metal barrel, placed on the mixer platform with appropriate quantity of sludge.

About 60 – 65 l of sludge are poured at a time from the sedimentation vessel into a previously prepared cask. As soon as a cask is filled up, it is hermetically covered with a lid supplied with screw and transported to the laboratory for sludge conditioning. There, additional settling of sludge is allowed. Separated water is pumped into a plastic can and taken back to the RA reactor spent fuel storage pool. Through the second stage of the sludge settling, volume of the sludge in the cask has been reduced to about 40 l.

When the cask with the settled sludge is placed on the platform of the mixer for further conditioning, the necessary amount of cement (PC-45 MPa), according to the established formula of cement matrix and the cement-sludge ratio, are poured into the cask. Mechanical manipulator will then mix this mixture until a homogeneous substance is obtained. This technology for sludge conditioning eliminates all the risks related to pouring the sludge into the concrete mixer and pouring the cement-sludge mixture into the metal barrel. The barrel with the homogenised mixture is removed from the mixer platform and placed in a separate room for concrete to harden. The time needed for concrete hardening is about 48 h.

The final stage of radioactive sludge conditioning is the covering of radioactive sludge immobilised in a cement matrix with pure concrete cork and, after the concrete cork hardening, cover metal barrel with a lid supplied with screw. The results of three stages of radioactive sludge conditioning: settled sludge in a cask; sludge immobilised in a cement matrix; and immobilised sludge covered with pure concrete cork, are shown in Fig. 2.

Fig 2.The results of three stages of radioactive sludge conditioning: settled sludge; sludge immobilised in cement matrix; and immobilised sludge covered with pure concrete cork.

Taking into account the measured sludge activity concentration, two stage sedimentation process, one in the vessel for sedimentation and the second in the concrete cask - container, and the conditioning technology, it is estimated that each cask containing conditioned sludge contains about 150 - 200 MBq ^{137}Cs and about 7 - 10 MBq ^{60}Co, i.e. activity concentrations of the conditioned radioactive waste in radioactive waste packages are about 0.7 - 1 GBq/m^3 ^{137}Cs and about 35 - 50 MBq/m^3 ^{60}Co. Taking into account composition of radioactive waste packages, the effect of self-absorption in homogeneously dispersed radioisotopes in the cement matrix, and concrete cask walls radiation absorption capability the contact dose rates on the casks surface are in the range from 100 to 150 μSv/h, much less than 2 mSv/h, which is an acceptable value for the radioactive waste packages.

According to the estimated activity, activity concentration and radinuclides composition, estimated heat generation rate is of the order of mW/m^3. Since the activity concentration of long lived α emitters is practically negligible, and thermal power is far beneath 2 kW/m^3, radioactive waste packages – concrete casks in metal barrel with in a cement matrix conditioned radioactive sludge are classified as Low and Intermediate Level Waste - Short Lived (LILW - SL) /12/. Total amount of the sludge taken out from the bottom of the RA reactor spent fuel storage pool have been conditioned in 31 concrete casks in metal barrels and disposed at the existing waste repository at Vinca site.

CONCLUSION

Cleaning the research reactor RA spent fuel storage pool appeared to be a more difficult, more time consuming and certainly more expensive operation than originally estimated. However, the results achieved so far, extraction of the sludge from the bottom of the pool and its immobilisation and conditioning – the first stage of the cleaning, are a sound basis to conclude that the task shall be accomplished successfully.

After the operations explained above have been performed, necessary elements for planning further stages of pool and water cleaning, and treatment of the spent fuel should be obtained. Through many years research and development in radioactive waste immobilisation and conditioning performed experimental experience gave the possibility to choose the best formulation for cement mixture and results gave us certain to claim that described methods and used matrix materials will serve as a barriers to preserve radionuclides migration to the surroundings for at least 300 years. Optimization of the processes and matrix-radwaste mixtures is in further progress and we hope that this work will influence the design of the future Yugoslav storage centre, shallow land burial type for low and intermediate level radioactive wastes.

All performed steps in removing sludge from the bottom of the spent fuel storage pool, pouring the sludge from the sedimentation vessel to the concrete cask, cask with a sludge transportation to the laboratory for immobilisation and conditioning, and immobilisation and conditioning of the radioactive sludge in a cement matrix have been done in accordance with all relevant requirements for radiation safety and radiation protection.

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