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PRACTICAL EXPERIENCE FOR LIQUID RADIOACTIVE WASTE TREATMENT FROM SPENT FUEL STORAGE POOL AT RA REACTOR IN THE VINCA INSTITUTE

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ABSTRACT

The present paper reports the results of the preliminary removal of sludge from the bottom of the spent fuel storage pool in the RA reactor, mechanical filtration of the pool water and sludge conditioning and storage.

Yugoslavia is a country without a nuclear power plant (NPP) on its territory. The law which strictly forbids NPP construction is still valid, but, nevertheless we must handle and dispose radioactive waste. This is not only because of radwaste originating from the use of radioactive materials in medicine and industry, but also because of the waste generated by research in the Nuclear Sciences Institute "Vinca". In the last forty years, in the "Vinca" Institute, as a result of two research reactors being operational, named RA and RB, and as a result of the application of radionuclides in medicine, industry and agriculture, radioactive waste materials of different levels of specific activity were generated. As a temporary solution, radioactive waste materials are stored in two interim storages. Radwaste materials that were immobilized in the inactive matrices are to be placed in concrete containers, for further manipulation and disposal.

1 INTRODUCTION

The RA, research reactor (6,5 MW) was shut down in 1984 in order to reconstruct and improve practically all vital reactor systems. However, for a number of political, administrative, economical and technical reasons, this reconstruction has not been completed.

The spent fuel storage pool in the RA research reactor in the "VINCA" Institute consists essentially of four, six meter deep, inter-connected rectangular basins. The fourth basin contains an annex, which makes the basin L-shaped. Basin 1 is accessible from the reactor via a water-filled transport channel. It is estimated that the basins together contain about 200 tons of stagnant tap-water. The basins were filled in 1960 and had received no attention to water chemistry until 1995. 304 channel type stainless steel fuel containers, receiving up to 18 spent fuel elements each, are placed vertically in the pool. Now these containers contain 1727 of 2% and 894 of 80% enriched spent fuel elements. In order to increase storage capacity, the oldest spent fuel elements were gradually taken out of the original stainless steel containers, and repacked into aluminium barrels. Now 30 such aluminium barrels containing 4929 of 2% enriched spent fuel elements are placed in two layers in the annex of the basin 4 [1].

At present, the reactor RA spent fuel storage pool is in very bad condition. Water in the pool is dirty and its chemical parameters are not maintained to minimize corrosion. Following the recommendations obtained from International Atomic Energy Agency (IAEA), the "Vinca" Institute created a project incorporating the following steps: preliminary removal of sludge and other debris from the bottom of the pool in the RA reactor, washing of deposits from all the surfaces in contact with pool water, venting of the aluminum barrels, mechanical filtration of the pool water, final removal of the sludge, sludge conditioning and storage at the waste repository at the “Vinca” Institute site.

Since one of two hangars 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 material storing only for a few years, attempts were made at the “Vinca” Institute of Nuclear Sciences to develop an immobilization process for conditioning of low and intermediate level radioactive waste materials and their safe disposal into the appropriate disposal system [2, 3, 4, 5]. For radwaste materials of different origin and composition, an immobilization process, cementation was investigated. Developed immobilization 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, according to radionuclide inventory, decay heat, radiation dose rate and contamination, identification, configuration and weight, mechanical integrity and so on [6] for interim storage and final disposal of such materials on the appropriate sites. Radwaste materials that were immobilized in the inactive matrices are to be placed in the concrete containers, for further manipulation and disposal.

2 CHARACTERISTICS AND QUANTITIES OF RADIOACTIVE SLUDGE

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 [1]. Activity concentration of the pool water of about 80 - 90 kBq/l of ^{137}Cs, although not of grave concern, is certainly significant, and is incontestable proof that a certain amount of the fission products are leaking. The activity of ^{137}Cs in the sludge samples is 1.8 ± 0.2 MBq/l [1].

2.1 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. Estimations was made on the basis of the average sludge height on the bottom of the pool and pool surface. The sludge color has been a dark red - brown, like Fe corrosion product. 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 previous experience, a technology was developed for sludge immobilization and conditioning in a cement matrix, inside casks, produced using the standard 200-litre metal barrels which have lids supplied with screws. Casks have been produced as containers in standard metal barrels. Thickness of the concrete walls was about 6 – 7 cm. The inner side of the cylindrical concrete wall was a plastic tube with wall thickness of 1 cm, which serves as a first barrier in preventing radionuclides leaching from radioactive sludge immobilized in a cement matrix. The bottom cask concrete wall was 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 was about 75 l.

The existing pilot cement mixer was reconstructed to enable the placing of 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 an 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 is presented in Fig. 1.

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 a 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, is poured into the cask. The formula of cement matrix and the cement to sludge ratio, is defined in accordance with previous experience and experimental investigations on radwaste cementation and experiments made with this sludge. The best sludge to cement mass ratio for appropriate mechanical strength was approximately 1:1.8

A mechanical manipulator will then mix this mixture until a homogeneous substance is obtained. A homogeneous substance could be obtained by less than one hour of mixing. This technology for sludge conditioning eliminates all contamination and radiation risks related to pouring the sludge into the concrete mixer and pouring the cement-sludge mixture into the metal barrel. The barrel with the homogenized mixture is removed from the mixer platform and placed in a separate room for the concrete to harden. It was experimentally determined that the time needed for concrete hardening is about 48 h. Due to the dynamics of the sludge removal from the spent fuel storage pool, and sludge settling, the final stage of radioactive waste conditioning is 7 – 10 days after the sludge cementation.

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

Taking into account the measured sludge activity concentration, the two stage sedimentation process, one in the vessel for sedimentation and the second in the concrete cask - container, as we performed as 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 0.1 to 0.15 mSv/h, much less than 2 mSv/h, which is an acceptable value for radioactive waste packages.

According to the estimated activity, activity concentration and radionuclides composition, the 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 a metal barrel with a cement matrix conditioned radioactive sludge are classified as Low and Intermediate Level Waste - Short Lived. The total amount of the sludge taken out from the bottom of the RA reactor spent fuel storage pool has been conditioned in 31 concrete casks in metal barrels and disposed at the existing waste repository at the " Vinca " Institute site.

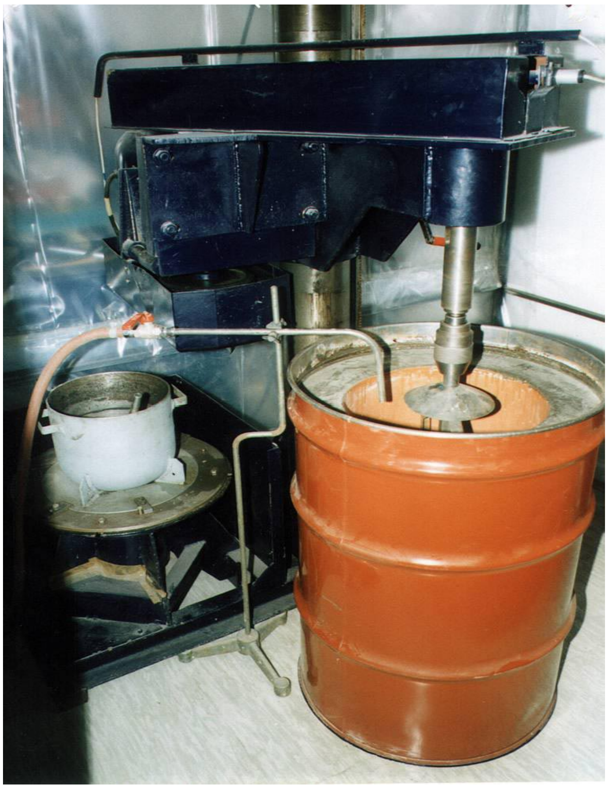

Figure 1: Modified pilot mixer with concrete container made in metal barrel

3 CONCLUSIONS

Cleaning the research reactor RA spent fuel storage pool was 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 immobilization and conditioning – the first stage of the cleaning, are a sound basis for concluding that the task will 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 of research and development of radioactive waste immobilization and conditioning processes, the performed experimental experience enabled us to choose the best formulation for the cement mixture. The results allowed us to claim that the described methods and used matrix materials will serve as barriers to preserve radionuclide 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 a 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 in the concrete cask, cask (with a sludge) transportation to the laboratory for immobilization and conditioning, and immobilization 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 [6].

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