Full-Scale Tests of the Technology for TRU-containing MLW and LLW Decontamination - 10026

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ABSTRACT

The technological layout and engineering embodiment for decontamination of medium- (MLW) and low-level radioactive waste (LLW) containing alpha-emitting radionuclides have been developed. The technology is based upon precipitation, ultrafiltration, and ion exchange methods.

Feed MLW and LLW are conditioned in two separate streams. Alpha-emitting radionuclides present in MLW feed with the concentration up to 10^7 Bq/L are co-precipitated with ferric hydroxide and the resulting suspension is filtered through the ultrafiltration unit. The unit is equipped by tubular ceramic membranes with the filtering layer made of ZrO_2-based ceramics. The permeate is combined with the LLW feed. The concentrate is dried in a microwave unit and stored in drums.

The LLW stream containing up to 10^3 Bq/L α-emitters goes through the analogous ultrafiltration unit. Emerging permeate contains soluble carbonate metal complexes, which are removed from solution on anion-exchange resin. The filtrate from the column contains less than 0.5 Bq/L of α-emitters that meets the sanitary requirements. The concentrate from the ultrafiltration unit is further concentrated by evaporation using rotor-film evaporator units and finally, conditioned in drums by infrared drying.

The gross concentration factor (volume reduction) of the α-bearing waste decontamination facility reaches 15,000.

In our paper creation of the general alpha-bearing LRW processing facility and the development and design of various stages of the technology used are reported and discussed.

INTRODUCTION

Extremely high environmental hazards of liquid radioactive waste (LRW) from processing nuclear materials require that it be treated to reduce the hazard and volume requiring longterm disposal. The volume of liquid radioactive MLW and LLW annually generated at the chemical-metallurgical plant is 1,500-2,000 m^3 and 150,000 m^3, respectively. For the major part the waste in question consists of solutions containing plutonium, uranium, and americium radionuclides in concentrations of 10^3 Bq/dm^3 and 10^7 Bq/dm^3 for LLW and MLW, respectively. LLW solutions have pH values within the range of 7-9 and contain up to 100 mg/dm^3 of solid particles with up to 99.9% of the radionuclide contaminants affixed to them. MLW is nitric acid solutions containing iron, calcium and other elements. The difference in the LLW and MLW composition defines different approaches to the waste processing.

Until recently the waste in question was processed by means of co-precipitation with ferric oxide. The process resulted in large volume of the secondary waste and frequently required re-processing of the waste batches to meet the regulatory values, thus reducing the waste treatment

stage efficiency even further. And the more stringent regulations of the Russian SPORO-2002 standard [1] effected in 2003 made the α-bearing waste treatment task a real challenge. Therefore there was a pressing need to develop a technology that would provide the necessary α-bearing waste decontamination factor and the reasonably high waste volume reduction factor simultaneously. The ultrafiltration technology being a powerful tool for the liquid-solid phase separation seemed to suit the purpose just perfectly. Certain enhancements, namely, the ion exchange, thin film evaporation, and drying techniques were incorporated later. After the tests of separate stages with actual waste showed good results, the reported complex technology was ready for application.

EXPERIMENTAL

A general flow diagram of the technology is shown in Figure 1. It provides LRW and MLW decontamination to the sanitary specifications imposed on the decontaminated effluents to be released into open natural reservoirs. According to the standing SPORO-2002 regulatory document the maximum permissible concentration of α-emitting radionuclides in the effluents to be released into natural water bodies is 5 Bq/dm^3.

Various designs of microfiltration and ultrafiltration units for LLW processing were tested. Tests of various filtering units based on flat metal-ceramic membranes were reported in [1, 2, 3]. The membranes demonstrated high LLW effluent decontamination efficiency; however, certain design and manufacturing issues affected the filtering packet reliability, and due to an insufficient filtering cycle value the desired waste concentration factors could not be reached.

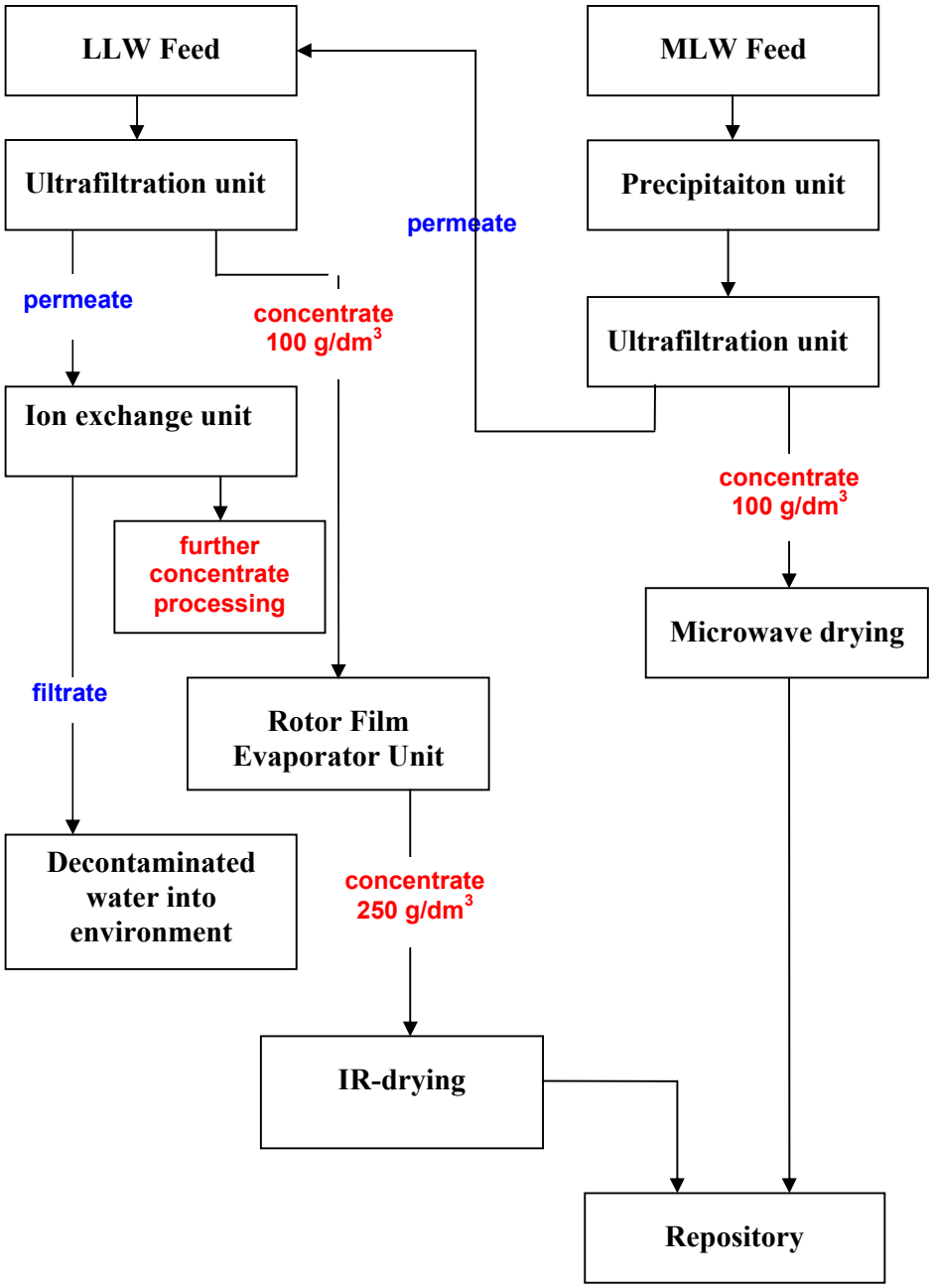
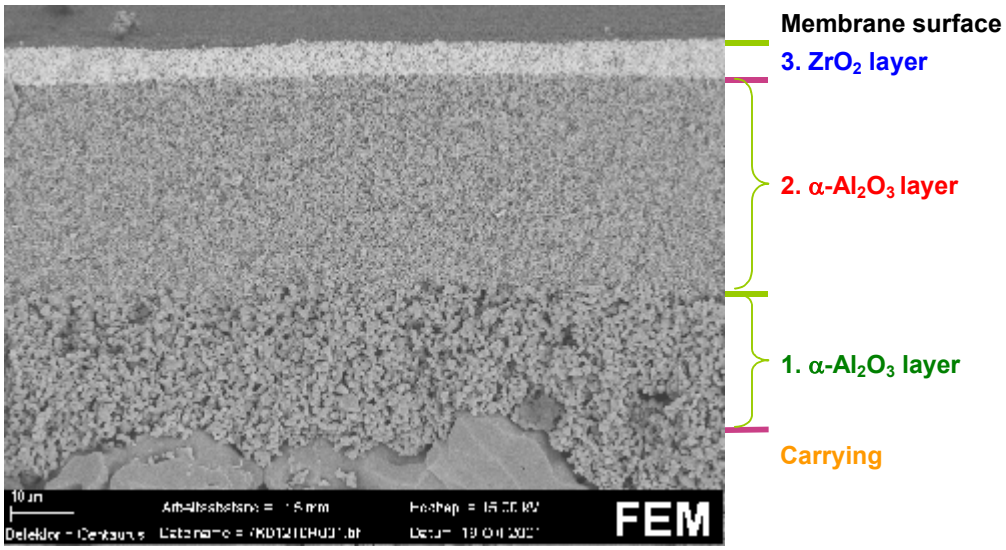


Fig. 1. Flow diagram of the LLW and MLW processing

The principal decision that allowed a significant filtering time increase and a large gain in the concentrating factor was the application of tubular filtering cartridges made of ZrO_2 and α-alumina ceramics. The membrane structure is shown in Figure 2.

Fig.2. Cross-section view of the nano-structured ultrafiltration membrane



In Fig.3 the pilot ultrafiltration facility equipped with the tubular filtering cartridge module design of 0.4 m^2 filtering surface area is shown.

With this design of the filtering unit at 150 g/dm^3 of suspended solids the permeate output was still maintained at 400 dm^3/m^2.

Fig. 3. Pilot ultrafiltration facility equipped with the tubular membrane filter cartridge module design

In Table I, the permeate output of the filtration facility is given as a function of the process temperature and the concentration of suspended solids in the loop.

Table I. Permeate Output Depending on the Feed Temperature and Various Content of Solids in the Loop

Temperature of the feed, ^{o}C	**Permeate output at the mass concentration of solids in the feed, g/dm^3**		
	1.9 thru 11.8	4.6 thru 28.2	9.5 thru 102,0
22±2	72±2	65±5	not determined
35±5	75±5	73±5	75±5
51±1	115±5	110±5	110±5
60±2	140±5	145±5	155±5
75±2	not determined	not determined	160±5

Operating pressure in the loop was 2.0 ±01 bar, the filtering module outlet pressure was 0.3 bar

The results showed a 4-fold increase in the permeate output and a 10-fold increase in the concentration of suspended solids in the loop as compared to the FFFU with Trumem™ membranes. Based on those data the full-scale industrial prototype of the LRW ultrafiltration facility for 3-5 m^3/hr by permeate has been designed and assembled. A general view of the facility during the control unit hardware and software start up phase and hydraulic test runs is shown in Figure 4.

Fig. 4. Full-scale industrial prototype for concentrating suspended solids

The LLW ultrafiltration unit permeate is fed into an end-polishing ion exchange unit. The columns are loaded with high-basic anion exchange resin. The emerging filtrate contains alpha-emitting radionuclides with concentration less than 0.5 Bq/dm^3 and is released into the Techa cascade of natural water bodies. Nitrate resin bed stripping solutions are recycled for the recovery of valuable components.

The LLW ultrafiltration unit concentrate contains 150 g/dm^3 of suspended solids and is further concentrated using rotor-film evaporators with the shaft rotating speed of 150 RPM. The emerging slurry concentration of suspended solids is within the range of 200-250 g/dm^3. It is loaded into certified stainless steel drums of 200 dm^3 volume and conditioned using the IR drying process.

After drying the drums are sealed tight and moved to the repository for temporary long-term radioactive waste storage located on the chemical-metallurgical plant site. Currently a new on-site repository is under construction.

Acidic MLW is neutralized with sodium hydroxide solutions and is fed into the MLW ultrafiltration unit for further concentrating and removal of excess sodium nitrate. Removal of sodium nitrate to the concentration of 1 g/dm^3 in 1 volume of the precipitate requires as many as

14-15 volumes of water in the conventional stirring – settling mode. The washing operation performed by feeding small quantities of water into the filtering loop requires merely 3 volumes of water; hence, the efficiency of washing is almost 5 times as high as with the conventional precipitate decanting process. The MLW ultrafiltration unit permeate is recycled by combining it with the LLW feed. Water from the MLW ultrafiltration unit concentrate is removed in the microwave dryer and the dry powder is packed in drums; the drums are sealed tight and moved to the repository for temporary long-term storage.

The residual water content in the dried waste is in the range of 5-7%. The dried waste loading of the drum volume does not exceed 80%. The amount of α-radionuclides in the dried waste permitted into the repository does not exceed 3×10^8 Bq/dm^3.

The content of alpha-radionuclides in all the effluents released from the facility into open natural reservoir does not exceed the regulatory values mentioned above. The total LRW concentrating factor of the LLW and MLW processing facility reaches the value of 15000.

CONCLUSIONS

The technological layout and engineering embodiment of the facility for decontamination of α-bearing liquid MLW and LLW generated at the chemical-metallurgical plant has been developed. The facility consists of the ultrafiltration and ion exchange units, rotor-film evaporator, IR- and microwave drying units. The facility provides the above waste treatment to the standing sanitary norms established by the Russian SPORO-2002 standard for liquid effluents released into a natural water body. Gross secondary waste volume reduction factor of the facility reaches 15,000. The dried solid waste is stored in the on-site repository for temporary long-term radioactive waste storage.

REFERENCES

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