Technology for Treatment of Liquid Radioactive Waste Generated during Uranium and Plutonium Chemical and Metallurgical Manufacturing in FSUE PO Mayak – 13616

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ABSTRACT

Created technological scheme for treatment of liquid radioactive waste generated while uranium and plutonium chemical and metallurgical manufacturing consists of:

- Liquid radioactive waste (LRW) purification from radionuclides and its transfer into category of manufacturing waste;
- Concentration of suspensions containing alpha-nuclides and their further conversion to safe dry state (calcinate) and moving to long controlled storage.

The following technologies are implemented in LRW treatment complex:

- Settling and filtering technology for treatment of liquid intermediate-level waste (ILW) with volume about 1500m^3/year and alpha-activity from 10^6 to 10^8 Bq/dm^3
- Membrane and sorption technology for processing of low-level waste (LLW) of radioactive drain waters with volume about 150 000 m^3/year and alpha-activity from 10^3 to 10^4 Bq/dm^3.

Settling and filtering technology includes two stages of ILW immobilization accompanied with primary settling of radionuclides on transition metal hydroxides with the following flushing and drying of the pulp generated; secondary deep after settling of radionuclides on transition metal hydroxides with the following solid phase concentration by the method of tangential flow ultrafiltration. Besides, the installation capacity on permeate is not less than 3 m^3/h. Concentrates generated are sent to calcination on microwave drying (MW drying) unit.

Membrane and sorption technology includes processing of averaged sewage flux by the method of tangential flow ultrafiltration with total capacity of installations on permeate not less than 18 m^3/h and sorption extraction of uranium from permeate on anionite. According to radionuclide contamination level purified solution refers to general industrial waste. Concentrates generated during suspension filtering are evaporated in rotary film evaporator (RFE) in order to remove excess water, thereafter they are dried on infrared heating facility. Solid concentrate produced is sent for long controlled storage.

Complex of the procedures carried out makes it possible to solve problems on treatment of LRW generated while uranium and plutonium chemical and metallurgical manufacturing in Federal State Unitary Enterprise (FSUE) Mayak and cease its discharge into open water reservoirs.

INTRODUCTION

As a result of uranium and plutonium chemical and metallurgical manufacturing at FSUE Mayak low and intermediate level LRW is generated. In order to solve the problem of this LRW treatment a technological scheme has been developed, trials of separate units have been carried out, the equipment has been manufactured and assembled, pre-commissioning works for its commissioning have been begun.

DESCRIPTION

Developed technological scheme for treatment of LRW generated while uranium and plutonium chemical and metallurgical manufacturing consists of:

- LRW purification from radionuclides and its transfer into category of industrial waste;
- Concentration of suspensions containing alpha-nuclides and their further conversion to safe dry state (calcinate) and moving to long controlled storage.

The following technologies are realized in an abuilding complex for RW treatment: settling and filtering technology for intermediate level LRW from workshops #1 and #2 (intermediate level waste (ILW1 and ILW2)) and membrane and sorption technology for purifying of LLW from active drain (Table I and Table II).

Table I. Characteristics of initial waste

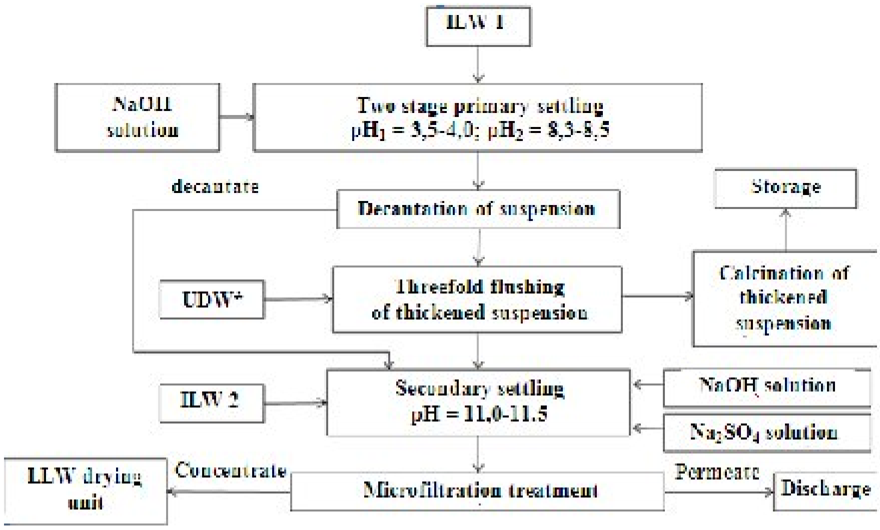
Flow type	Volume, m^3	Specific (volumetric) alpha-activity, Bq/m^3	Mass concentration of solid phase, g/dm^3	Mass concentration of salt, g/dm^3
ILW1	700	10^{10} - 10^{11}	-	330
ILW1	800	10^{8} - 10^{9}	1,5	140
LLW	150000	10^{6} - 10^{7}	0,1	1,5

Table II. Radionuclide content of flows

Flow type	Part of separate radionuclide alpha-activity from total alpha-activity			
	Uranium	Plutonium	Americium	Products of radioactive decay – Radium, Radon, Polonium and so on
ILW1	-	6 ± 2	93 ± 4	-
ILW1	6,5 ± 0,5	43 ± 5	5 ± 2	54 ± 2
LLW	3,3 ± 0,6	57 ± 20	7 ± 3	31 ± 1

Settling and filtering includes primary two-stage settling on transition metal hydroxides with the following flushing and drying of the pulp generated; secondary deep after settling of

radionuclides on transition metal hydroxides with the following solid phase concentration by the method of tangential flow ultrafiltration (Figure 1).
Thickened suspension volume ratio after 1 hour of settling at different conditions of two-stage hydroxide sediments precipitation is (40 ± 10) %, after 2 hours of settling thickened suspension volume ratio decreases up to (25 ± 10) % and after more than 18 hours of settling thickened suspension consolidates up to (15 ± 10) %.
This technology of ILW treatment is based on use of alpha-radionuclides joint settling with iron and other elements hydroxides, forming during increase of solution pH level, sorption of alpha-radionuclides in ion state on the extended surface of forming partially soluble compounds with metals and following formation of pseudocolloid and own solid phase of alpha-radionuclides.
Two-stage settling of metal hydroxides makes it possible to receive sediments of minimal volume and with high consolidation speed as well as decantate without visually determined fine suspension. Main share of activity concentrates in thickened suspension generated in the result of settling and sedimentation. The main contribution to the total thickened suspension alpha-activity (up to 98%) brings Am-241.



* UDW - utility and drinking water supply system

Figure.1 Flow scheme of ILW treatment

Flushing

The thickened suspension generating in the result of settling and sedimentation contains a lot of water soluble dead-weight salts including metal nitrates increasing the calcinate volume and impairing calcination process.
During flushing determinative influence on foliation speed has volumetric ratio between thickened suspension and rinsing water. It was determined that taking into account foliation speed the volumetric ratio “thickened suspension : rinsing solution” should be not less than 1 : 3.

In consequence of threefold flushing according to abovementioned mode the content of nitrate ions decreases at rinsed thickened suspension till mass concentration of less than 50 g/dm^3.

Calcination

Thickened suspension heating is carried out in a working chamber of microwave drying unit due to absorption of microwaves. Steaming of thickened suspension at microwave unit is carried out with continuous feed of suspension into metal container (drum). During thickened suspension steaming dry residue is concentrated and thickened. After that the layer of collected dry residue is calcined at a temperature of (850 ± 50) ^{0}C. During the process of high-temperature calcinations the mass of solid phase decreases up to 30%.

Secondary settling

The secondary settling of alpha-radionuclides is carried out by iron or other metal hydroxides. A significant share of the total activity of ILW has radium. Separation of radium from the solution is based on its suspension coprecipitation with barium sulfate. The amount of barium in initial solution is quite enough for formation of necessary amount of barium sulfate sediment for effective separation of alpha-radionuclides into solid phase. In other cases sodium sulfate can be added to the flow of solution for necessary concentration of sulfate-ions in it.
In particular cases the combined flow of initial decantate and rinsing water is treated with solution of iron sulfate (II) with mass ratio of 1% in order to create mass concentration of $FeSO_4$ of 70-75 mg/dm^3 and after it is neutralized by sodium hydroxide solution with concentration of 550 g/dm^3 up to pH level of 11,0-11,5.
Content of suspended particles (solid phase) in neutralized suspension in dependence of solution completing is up to 3 g/dm^3. Generated suspension is directed to the ultrafiltration unit.

Liquid intermediate level waste ultrafiltration

Ultrafiltration is the process of substances separation on a semi-permeable membrane without formation of new phases. The method of tangential flow ultrafiltration is based on passing through the membrane of water and salts whereas solid phase and colloids with particles sizes exceeding the sizes of membrane's pores are entrapped and concentrated in the facility's loop. Earlier trials on real LRW have been carried out and they had a positive result (1). If membrane filters work at chosen parameters they work in a self-regeneration mode and does not take place loading of membrane's pores.

The temperature of suspension coming to the membrane filter should be in the range of (60±5) ºC. Decrease of thickened suspension temperature leads to viscosity increase what influences on hydraulic resistance of circulation loop and resulted in pressure increase at top manifold.

The solid phase is concentrated up to mass concentration of 100 g/dm^3. The concentrate of solid phase is flushed from salts and directed to concentrate drying module from LLW treatment (rotary film evaporator and drum-dryer). Permeate and rinse water are combined with purified water from filtrates from sorption unit and directed for discharge (Figure 2).

Figure 2 Facility for solid phase concentration

Liquid low-level waste treatment

Alpha-activity of filtered water is caused by Plutonium, Americium and Uranium radionuclides in colloidal and pseudocolloidal form. Uranium radionuclides also can be presented by soluble tricarbonate complex. Because of that ultrafiltration method for purification of active drains water from Plutonium and Americium is enough but for purifying from Uranium additional sorption method of extraction on anion-exchange resin AB-17 is a need (Figure 3).

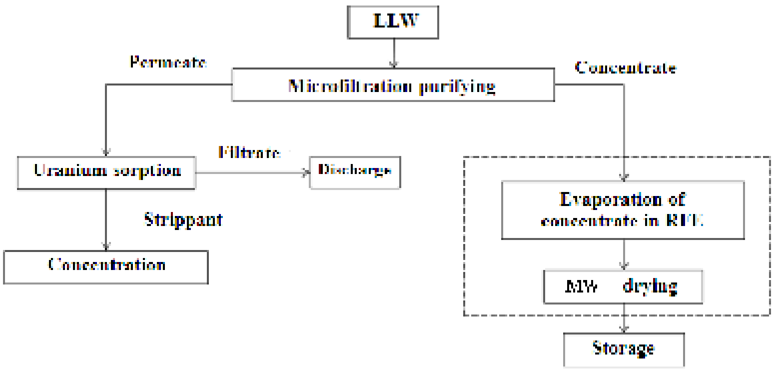


Figure.3 Scheme of LLW flow treatment

Filtered water comes to ultrafiltration unit. Suspended solid particles are concentrated in inner loop of ultrafiltration apparatuses. Concentration of solid particle at concentrate is from 10 to 20 g/dm^3. Obtained concentrate in continuous or periodic mode is directed to rotary film evaporator for evaporation (Figure 4). Presumable content of suspended solid particles after evaporation should be not less than 200 g/dm^3. Evaporated concentrate is directed for drying to the facility with infrared heating (Figure 5). The density of densely packed composition after drying is about 0,9 г/cm^3.

Figure 4. Rotary film evaporator

Figure 5. Facility for radioactive salts drying

Permeate after ultrafiltration treatment is directed to the sorption unit. Total specific activity of alpha-radionuclides in permeate does not exceed 50 Bq/dm^3, at the same time the activity of Pu-239 does not exceed 4 Bq/dm^3 and the activity of Americium 2,6 Bq/dm^3. The duration of working filter cycle takes on 10000 columnar volumes. At the end of filter cycle Uranium is desorbed by nitric acid solution.

DISCUSSION

Final output flows are presented by two liquid and two solid products. Uranium strippanta, contains about 2 kg of Uranium. Combined water flow with the following characteristics: pH level is 7,0-10,0, Uranium mass concentration is less than 0,003 g/m^3. Sum of ratios of specific activities of alpha-radionuclides to the meanings of their minimum significant activity does not exceed 1, which let to refer this flow to the category of industrial waste. About 715 ton of salts will annually go with this flow and one part of them is returning as long as these salts came to the plant with industrial water and another part is invected in the result of technological and treatment processes.

Annually for the long time storage into the storage facility comes about 4,5 m^3 of “Americium concentrate” with volume activity of alpha-emitters about $4{,}4\cdot10^{10}$ Bq/dm^3 (1,2 Ci/dm^3) corresponding to the level of high level solid radioactive waste. Also about 22 m^3 of dry salts concentrate comes to the storage complex with expected volume activity up to $1{,}5 \cdot 10^8$ Bq/dm^3(4,2 vCi/dm^3) by alpha-emitters which does not exceed limits for intermediate level solid radioactive waste (Table III).

Table III. Characteristics of output flows

Flow type	**Volume, m^3/year**	**Specific alpha-activity, Bq/m^3**	**Weight, ton/year**
Uranium Strippanta	150	$3 \cdot 10^7$	0,0023 (Uranium)
Permeate from ILW 1, 2 treatment	4220	до 10^5	-
Filtrate from LLW treatment	~150000	$20 \cdot 10^3$	-
Calcinate after ILW 2 treatment	4,55	$4{,}4 \cdot 10^{13}$	6,4
Dry concentrate after LLW and ILW 1,2 treatment	21,9	$1{,}5 \cdot 10^{11}$	21,66

CONCLUSIONS

1. The technological scheme has been developed for treatment of LRW from chemical and metallurgical production.

2. Capacity for work of all the equipment has been tested for all technological stages. All the resulthickened suspension of testhickened suspension are positive.

3. Assembling of the equipment has been completed for the developed technological scheme for treatment of LRW from chemical and metallurgical production and it is planned ithickened suspension commissioning for industrial exploitation in 2013.

4.The developed technological scheme makes it possible to solve problems on treatment of LRW generated while uranium and plutonium chemical and metallurgical manufacturing and cease ithickened suspension discharge into open water reservoirs.

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Complex of the procedures carried out makes it possible to solve problems on treatment of LRW generated while uranium and plutonium chemical and metallurgical manufacturing in FSUE Mayak and cease thickened suspension discharge into open water reservoirs.