DEVELOPMENT OF TECHNOLOGIES FOR HANDLING LIQUID RADIOACTIVE WASTE OF NUCLEAR POWER PLANTS

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Article received on August 26, 2020

The paper presents the results of a feasibility study focused on two methods designed to treat evaporator bottoms generated from the evaporation of liquid radioactive waste (LRW) at nuclear power plants (NPP), namely, deep evaporation (DE) and ion-selective decontamination (ISD). ISD method proved to be much more efficient and cost effective compared to DE due to a drastic reduction of the conditioned radioactive waste volume intended for disposal. The paper also considers possible opportunities for upgrading particular ISD stages. It provides recommendations on how to upgrade LRW treatment technologies at NPP of a new generation, including separated collection of basic LRW types and development of in situ RW processing and conditioning flowsheets.

Keywords: *radioactive waste, NPP, evaporator bottoms, deep evaporation method, ion-selective decontamination method, operating costs.*

Challenges associated with radioactive waste (RW) processing along with the safety of nuclear technologies themselves have always played a crucial role having major influence on further prospects of nuclear power development. All Russian NPPs have almost identical designs of the process flow diagrams providing the processing of liquid radioactive waste (LRW) from NPP operation that was developed back in 1960s [1] based on evaporation technologies. LRW generated from various sources (regenerated ion-exchange resins, decontamination solutions, waste water from special laundries, etc.) enters a single drainage collection system and then is handed over to evaporation plants of water treatment systems (WES). These units treat water to remove radionuclides and generate salt concentrate as secondary waste which is also called bottom residue (BR) with a salt content of about 300 g/dm^3. Since the 1st unit of Novovoronezh NPP was commissioned in 1964, this flowchart has practically remained the same and the generated BR has to be stored in purpose-designed storage facilities for liquid radioactive waste (LRW SF). To date, Russian NPPs have accumulated about 90 thousand cubic meters of BR categorized as ILW with LRW storage capacities at some NPP sites filled up to 85—90 % [2].

For a number of years, attempts have been made to address the challenge of BR processing by various methods [3, 4] including BR bituminization implemented back in 1986 at Leningrad and Kalinin NPPs. This method did not get widespread acceptance due to a number of serious disadvantages. Due to bituminization, the fire hazard of solidified BR increases, thus, the BR shall be stored only at the at-reactor site. Ultimately, before this waste is removed from the NPP site, it should be subject to some additional final processing and conditioning.

Development of BR cementation method in Russia has been started years ago: since 1995 it has been introduced at a number of NPPs. Its advantages are quite obvious: incombustibility of the compound, simplicity of equipment, strength and durability of the concrete monolith. Nevertheless, it causes significant increase in the waste volume, there are some restrictions imposed on BR salinity level, binder materials and process parameters should be selected on an individual basis provided some strict adherence to the cement composition [4].

In 1987, the first deep BR evaporation unit (UGU) was commissioned at Novovoronezh NPP (NV NPP) providing final conditioning of secondary waste. This process flow ultimately results in a salt melt (SM) with a salinity of 1,500—1,800 g/dm^3 categorized as ILW similarly to the initial LRW and containing some 5—20% of bound water (crystallization water) [3]. Compared to cementing, a 6—8-fold decrease in the amount of waste subject to disposal can be attained.

Long-term operation of UGU-500 unit at NV NPP has shown that its actual capacity was much lower than the design one, which had been assumed as 0.5 m^3/h [5]. This circumstance should be accounted for in any further implementation of the deep evaporation method. To date, with the decommissioning of NV NPP units 1 and 2 started some 5 years ago, a large inventory of such waste still remains in buried LRW SF tanks: some 2,000 m^3 of LRW in a liquid form and some 1,700 m^3 of crystalline hydrates (CH) [5]. During long-term storage, isotopic composition of the waste has changed: it is now categorized as ILW Class 3 with most of it content accounting for ^{137}Cs radionuclides (over 95%) and the rest — for ^{60}Co.

Obviously, such large BR inventory, which should be basically absent at an NPP site at the stage of its decommissioning according to the "immediate dismantling" option, significantly complicates the operation of standard LRW processing systems that have long exhausted their operational capacity. The situation is aggravated by the unresolved problem of BR retrieval from the buried LRW SF tanks. All these factors acting together can significantly complicate all decommissioning operations at units No. 1 and No. 2 of NV NPP, as well as impede the Rosenergoatom Concern from meeting the established deadlines [6].

To treat BR, a selective treatment method was proposed in 1999 [7] with the first Russian ion-selective treatment unit (UISO) developed by ZAO RAOTECH that has been operated at the Kola NPP since 2006 [8].

The main advantage of the UISO method is associated with a dramatic decrease in the volume of ILW subject to disposal (over 100-fold decrease) due to selective extraction of radionuclides from the total mass of ballast salts: ^{137}Cs is absorbed in filter containers (FC) by a selective ferrocyanide sorbent; ^{60}Co in the form of hydroxide microcrystals resulted from the the break down of its complex with EDTA and other complexones in an alkaline medium are absorbed on microfilters (MF) with ceramic porous membranes. The sludge formed at the preliminary treatment stage and the microfilters themselves are then cemented inside NZK 150-1.5P containers. After the constituent sorbent drying to a free moisture content of less than 3%, these containers together with FCs having the required degree of protection are handed over for disposal as ultimately conditioned secondary SRW.

Since 2015, such a LRW processing complex has been operating at Smolensk NPP, similar complexes are being built at three more Russian NPPs.

Let's take a closer look on the performance and economic indicators of the two waste water conditioning methods (suggesting the use of UGU and UISO) when it comes to the processing of LRW accumulated at units No. 1 and No. 2 of NV NPP. Figure 1 presents basic flowcharts of these two methods.

The first stage is common for both methods. It involves the retrieval of both liquid BR fraction and available CH with their subsequent dissolution in BR tanks (BRT). These operations are not considered in this paper since they require a particular design solution [6]. Then the resulting solution is pumped into an intermediate tank (IT). After CH is dissolved and the composition of BR stored in LRW SF is averaged to a conventional salt content of 300 g/dm^3, its total inventory will account for 11,500 m^3 [5]. This is the LRW amount that is to be considered in further cost assessments focused on LRW treatment according to the above two methods.

According to the first method, the generated BR is handed over immediately from the IT to the deep evaporation unit UGU-500. Resulting SM is poured into purpose designed drums of A2201 type with a capacity 0.2 m^3 each (primary packages). After cooling and crystallization, these packages are installed into non-returnable protective containers of NZK 150-1.5P type: each container can accommodate 4 packages. These NZK are exactly considered as final packages to be sent for disposal. Secondary steam is condensed and fed to ion exchange filters (IE) for treatment. The treated water is discharged through a control tank (CT). To reduce water consumption, it can be reused for washing purposes in BRT. The ion exchange regenerates, as well as the wash water are discharged into a special sewer drain, evaporated using standard evaporation units

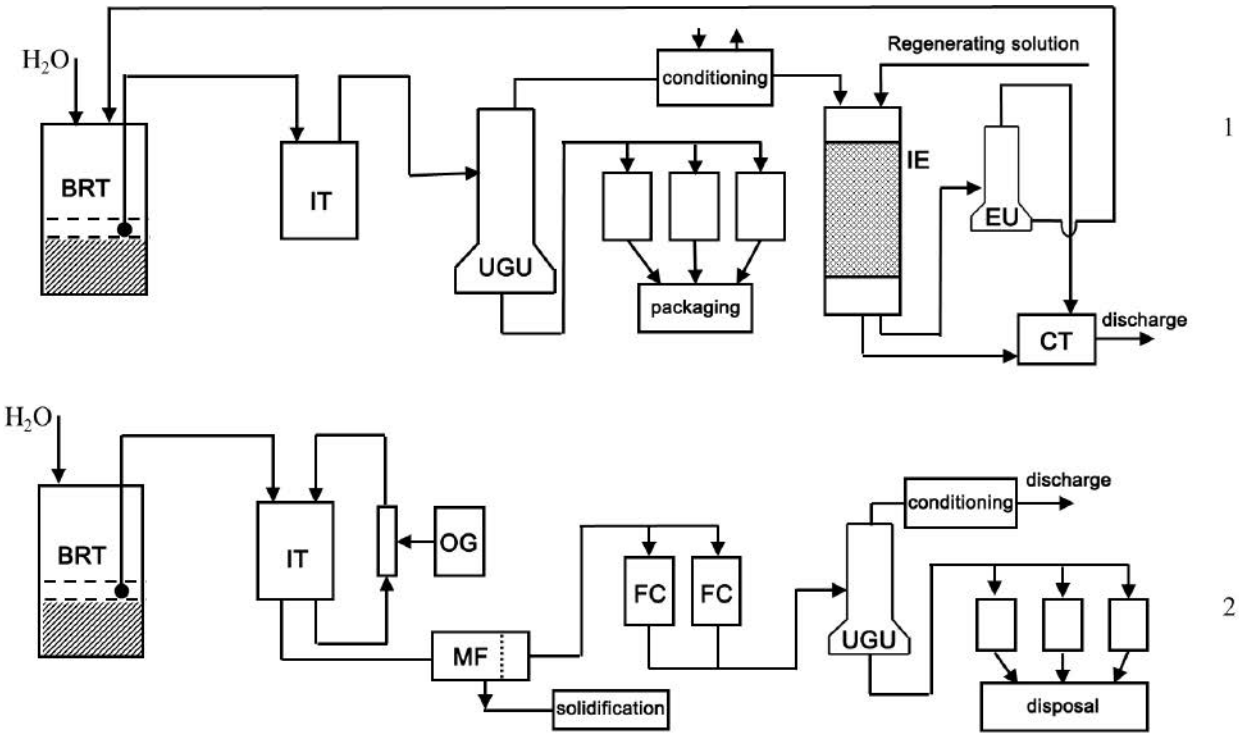

Figure 1. Flow charts for BR deep evaporation method (1) and BR ion-selective treatment method (2)

(EU) of water treatment systems (WTS) and then sent for recycling.

According to the UISO designs, the intermediate tank is used as a chemical reactor with ozone being supplied into it from ozone generator (OG) to provide oxidation of free organic matter and Co-EDTA complexes. The main stages of LRW treatment using this technology were presented above. Ultimately, the UISO method results in two main types of conditioned waste: NZK 150-1.5P with cemented sludge and MF, as well as spent FC with selective sorbents. At the final stage, solution being almost completely free from radionuclides is fed into a deep evaporation unit similar to the USU-500. Resulting SM categorized as VLLW is poured into metal drums and cooled to allow crystallization.

Calculated operating costs accounting for the processing of 11,500 m^3 of waste water using these two methods are presented below.

If USU-500 is used for BR conditioning purposes, the entire volume of conditioned waste is to be categorized as ILW. At first, the unit generates some 2,300 m^3 of SM that can be categorized as ILW with a concentration of about 1,500 g/dm^3.

The number of purpose-designed A2201 drums with a capacity of 0.2 m^3 needed to package relevant SM inventory with a filling degree of 0.9 will account for 12,800 pcs.

The number of NZK 150-1.5P containers will amount to 3,200 pcs. given that each container accommodates 4 drums.

Taking into account the overall NZK 150-1.5P capacity of 3.74 m^3, the total volume of secondary intermediate-level SRW generated during the processing of waste water according to this flow chart will amount to 12,000 m^3 (3,200 × 3.74): in fact it will be comparable and even slightly exceed the volume of initial LRW.

The second method suggesting the use of UISO will result in two waste categories: ILW and VLLW.

Let's calculate the total volume of ILW assuming that when cesium radionuclides are absorbed by a ferrocyanide sorbent, the degree of its saturation should not exceed half of the maximum specific activity limit established for ILW Class 3 waste in a package [9]. Assuming this, its specific activity will amount to $5 \cdot 10^9$ Bq/kg (given the maximum specific activity limit of $1 \cdot 10^{10}$ Bq/kg). Since the useful capacity of the sorbent according to FC designs developed by ZAO RAOTECH accounts for 120 dm^3, 144 m^3 of LRW can be supplied through one FC under given conditions. Thus, a total of 80 FCs will be needed to treat 11,500 m^3 of waste water with the spent FCs categorized as ILW.

To dispose of cemented sludge and MF, a total of 150 NZK150-1.5P containers will be required corresponding to 560 m^3 of ILW.

Given the overall FC volume of 0.6 m^3, the total volume of 80 FC subject to disposal will amount to 50 m^3 and the total volume of all conditioned ILW Class 3 waste from UISO treatment will be equal to some 610 m^3.

The SM generated at the final stage of processing by UISO units will have a specific activity of 0.5—0.6 kBq/kg, thus, corresponding to the VLLW category according to effective standards [10], which provides for its packaging into ordinary metal drums. The total number of these drums would be some 12,000 pieces. Therefore, the total VLLW volume will amount to some 2,400 m^3.

Table 1 presents the calculated operating costs for these two processing methods. The prices for packaging and the cost of services were calculated considering the recommendations and data from [3, 10—12].

Table 1. Total operating costs for BR management at Units 1 and 2 of NV NPP

Cost item	Unit price, mln RUB	Cost, mln RUB	
		UISO	UGU
Processing	–	800	500
Purpose designed drums A2201	0.10	–	1,280
NZK containers	0.15	22	480
Filter containers	1.25	100	–
Metal drums	0.003	36	–
Transportation	-	5	60
ILW disposal	0.158/m^3	97	1,900
VLLW disposal*	0.025/m^3	60	–
Total		About 1,124	About 4,200

* It should be noted that the VLLW management procedure, including the potential cost of waste disposal in near-surface disposal facilities has not yet been approved formally in the Russian Federation [10—12]. [10] indicates only some general principles associated with the management of this waste category, whereas [11] provides no price indicators for its disposal. Therefore, the calculations were made based on the recommendations from [12] referring to global best practices and indicating that VLLW disposal cost should be 5—10 times lower than the one of LLW.

For comparison purposes, costs required to cement some 11,500 m^3 of BR can be given. In this case, the initial waste volume will increase by about 1.7 times assuming the generation of some 20,000 m^3 of cement compound. A total of 13,400 NZK150-1.5P containers will be required with the corresponding purchasing cost amounting to some 2.0 billion rubles and the one needed to dispose of some 50,000 m^3 of ILW would amount to some 7.9 billion rubles. Thus, if cementing method is applied, the total costs accounting for those two items alone will be more than 2 times higher than those suggested under the UGU method and 8 times higher than those associated with the UISO method.

Given all the relativity of the above cost calculations, it seems obvious that the gain in total costs associated with UISO method almost entirely depends on a dramatic decrease in the volume of ILW requiring disposal.

Despite the fact that UISO has some obvious advantages over other LRW conditioning methods, operational experience at Kola NPP [8] did reveal some problems associated with the instability of operation at certain treatment stages, and primarily at the ozone oxidation stage involving the initial solutions. Almost immediately following UISO commissioning at Kola NPP, unstable ozone generator of Russian designs was replaced by a similar device from a German company VEDEKO and the ejector system designed for ozone injection into the reaction mixture was also upgraded [8].

However, despite this upgrading, oxidation stage is still considered as the most challenging one assuming the application of this method due to partial oxidation of organic BR components [13] preventing complete ^{60}Co extraction at the microfiltration stage. It is the increased content of ^{60}Co in the SM obtained at the final stage that ultimately determines its residual activity and does not allow for its categorization as common industrial waste in accordance with the provisions of [10]. Therefore, at all NPPs with operated UISO, the resulting SM categorized as VLLW should be now temporarily stored in drums at at-reactor sites.

Obviously further improvement of this method can contribute to the development of a more advanced and efficient approach that would provide the management of BR inventory accumulated at all NPPs.

These tasks can be addressed in different ways. First of all, it seems worth to improve the oxidation efficiency of organics present in BR. The problem can be solved both by increasing the concentration of ozone in water, which is very low [13], and by increasing the oxidizing capacity of the ozonation process itself.

The first solution potentially assumes the application of fundamentally new supersonic steam-gas-liquid ejectors: relevant experiments show a twofold increase in the concentration of ozone dissolved in water and a corresponding increase in the oxidation rate [14]. The second solution may suggest the use of a combined ozone oxidation method intensified by various physical processes, for example, electromagnetic radiation [15].

Most dramatic advance in this area would be, however, associated with the introduction of some fundamental changes into the method used to manage treated radionuclide-free BR using the UISO. This option suggests that the final processing stage would no longer require their evaporation-based concentration, nevertheless, providing for their

segregation into separate components using the electro-membrane method. A device equipped with bipolar membranes will help to generate nitric acid, mixed potassium-sodium alkali and fairly pure boric acid directly from saline solutions [16]. The first two products can be potentially recycled at NPPs providing the regeneration of ion exchangers in the water treatment systems and in the chemical water treatment plant without any additional purification. Whereas, boric acid should be additionally treated and concentrated. This approach completely eliminates the problem of at-site SM (VLLW) storage due to the absence of this waste category.

According to modern concept adopted JSC Rosenergoatom Concern, no SF LRW for BR storage purposes are provided for at the sites of new-built NPPs. This approach was first implemented during Unit 1 construction at NV NPP-2. Current operation of such units assumes that the resulting "fresh" LRW will contain not only commonly generated radionuclides (^{137}Cs, ^{90}Sr and ^{60}Co), but also some isotopes of Mn, Sb, Cr, etc. [17], and their specific activity may even exceed the one of cesium and cobalt. Therefore, the UISO method, which was originally proposed for the "cooled" BR processing, cannot cope with such LRW treatment. This is quite obvious, since the term "selective treatment" makes sense only if we are talking about two or three isotopes, and not about the whole range of elements.

Given the new tasks, a different approach to the arrangement of LRW management systems at NPPs seems worth to be considered. It should be based on a concept suggesting separate collection of various LRW types provided subsequent development of closed cycles designed for their separate processing. A number of factors prompting the implementation of such solutions can be noticed today, which, assuming their further development and implementation, will significantly increase the reliability and reduce the LRW processing costs.

Such a local LRW processing flow chart can be exemplified by a unit developed by AREVA and already implemented at a number of NPPs abroad [18]. It is designed to provided closed cycle treatment of decontamination solutions. At the same time, softer organic acids are used instead of EDTA for the in-circuit decontamination itself: most likely mixtures of oxalic and formic acids, which are quite easily decomposed under the impact of ultraviolet irradiation. At the next stages, similarly to the UISO method, the hydrolyzed oxides are separated by microfiltration and the filtrate is ultimately treated using ion exchange resins. The treated loop water is recycled and all secondary SRW in the form of cemented concentrate and spent ion-exchange resins are subject to disposal. This approach completely excludes the ingress of "heavy" complexions into the drainage water, and hence into all the water supply systems. Obviously, the introduction of a method similar to the one developed by AREVA to treat certain LRW types will not only simplify further waste management process, but also result in some significant economic benefits.

Russian research and production enterprise Alexandra-Plus is currently working on the development of an advanced submersible decontamination method suggesting the use of hard ultrasound involving an autonomous unit for secondary LRW treatment [19]. Therefore, wider introduction of similar [18, 19] methods accounting for all types of disinfectants generated both during NPP operation and its decommissioning will greatly reduce the complexity of the LRW management flow chart.

Another case in point is the development of an autonomous unit designed to process regeneration solutions from ion-exchange filters. Since regenerates making up no more than 5% of the total LRW amount generated at NPPs and contain the bulk of all radionuclides and a large amount of ballast salts [1], their local treatment would significantly reduce both capital and operating costs. According to our data, such a local "turnkey" installation has never been developed before, but the use of electromembrane jointly with evaporative methods seems quite promising for these purposes [20, 21].

An autonomous system developed to process the water from a special laundry room seems worth to be noted as well. These specific low-level LRW constitutes up to 30% of all waste generated at NPPs, but due to an extensive surfactant inventory its evaporation seems a quite challenging task to accomplish [1]. At a local processing unit used to treat LRW from special laundry both selective sorption with nanosized sorbent particles and membrane ultrafiltration can be applied, which will not only completely eliminate the ingress of surfactants into the drainage waters, but also provide their recycling during the decontamination of workwear [22].

It seems quite obvious that segregation of various LRW types generated during NPP operation performed at waste collection stage with their subsequent separate processing and conditioning will significantly reduce the amount of waste intended for disposal. This approach was originally implemented in the Russian Federation under a complex LRW processing flowchart developed by JSC VNIIKhT for the nuclear fleet LRW: it has been implemented since 2000 at the ship repair center Zvezdochka in Severodvinsk. It has provided quite significant reduction in the amount of secondary waste and operating costs in comparison with standard evaporation method commonly used for its processing [23].

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Bibliographic description

Penzin R. A, Svitsov A. A. Development of Technologies for Handling Liquid Radioactive Waste of Nuclear Power Plants. *Radioactive Waste,* 2020, no. 4 (13), pp. 90—98. (In Russian). DOI: 10.25283/2587-9707-2020-4-90-98.