

LOCAL SYSTEMS ENABLING THE CONCEPT OF SEPARATE COLLECTION AND PROCESSING OF LIQUID RADIOACTIVE MEDIUM AT NPP WITH WWER REACTOR UNITS

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The study shows that the engineering methods applied in radioactive waste (RW) processing are mainly focused on long-term compact storage of accumulated RW inventory at the industrial sites of NPPs. Therefore, the RW should be additionally treated to obtain a final product that would meet the final disposal acceptance criteria. The transition from liquid radioactive effluent mixing practice to its separate collection and processing at purpose-designed local units with relatively low productive capacity will enable selective concentration and separation of biologically hazardous components. The paper presents physical and chemical features of the sorption concentration processes, coprecipitation of radionuclides and the containment of boron-containing and nitrogen-containing chemical waste being considered typical for nuclear power plants.

Keywords: *radioactive waste, liquid radioactive medium, radioactive concentrates, separate processing of radioactive waste, ion-selective inorganic sorbents.*

To date, the environmental safety of nuclear power has been recognized solely based on the absence of emissions containing so-called greenhouse gases, primarily carbon dioxide. However, the environmental impact of all power generating facilities without any exception is believed to be much wider.

Thus, all thermal power units based on steam-power cycle are still considered active hydrosphere pollutants due to discharged chemical by-products. Spent process solutions are seen as common pollutants for all these units: these solutions are used to restore the performance of chemical water treatment (CWT) systems and their elements and unit-type treatment facilities (UTF). When large amounts of heat are removed from the turbine condensers, average annual temperature of water in hydraulic engineering installations grows contributing to excess biomass development. Increased

concentrations of salts and microorganisms adversely affect the performance of heat power equipment (biofouling reduces heat removal to a level that can be lower than the recommended standards) and water treatment systems (i. e., by decreasing the operating time of filters) [1]. The only method that can be currently implemented to maintain the salt background in industrial reservoirs is the water exchange between them and the natural hydrographic network, which ultimately disrupts the established ecological equilibrium.

Under normal operation of a nuclear power plant, another problem is faced as well: radioactive substances are released into the environment with their transfer outside the primary circuit that is yet impossible to prevent.

Early in the nuclear power development, a decision was made, which seemed quite reasonable for

that time: the amount of accumulated waste had to be reduced radically by a universal method — evaporation, which was facilitated by a sufficient amount of available thermal energy. Vented steam recycling reduced the needs in demineralized water produced at CWT plants, as well as salt release into the hydrosphere.

The main reason for the application of nitric acid as a regenerating agent was the requirement on keeping to minimum the volume of radioactive waste subject to long-term storage: all nitric acid salts are highly soluble with practically acceptable corrosion activity level. Bituminous compound [2, 3] and salt melt [4] were considered as final forms for LRW immobilization. Since steadily operating RW conditioning units were not available at most NPP, containers for the temporary storage of radioactive concentrates (bottom residues), CEC got overfilled which literally turned them into storage facilities for solid radioactive salts.

Sharp reduction in the operational free CEC capable of accepting new portions of still bottoms generated under normal operating conditions prompted the development of some new storage facility emptying methods. Under relevant studies, several options were proposed. These can be divided into three main groups:

- joint extraction of biologically hazardous components (chemical and radioactive substances) with the salts being further conditioned to salt water [4], cement [5, 6] or geocement [7] compounds;
- separation of bottom residue components into radioactive and general chemical components with each of the them subsequently emplaced into an appropriate repository according to the hazard class [1, 8–11, 13];
- processing of salts contained in radioactive concentrates providing their recycling at nuclear power plants and conditioning of radioactive substances together with the salts considered of no practical interest [14–16].

Feasibility of the third option seems uncertain: it's believed unlikely that a flowchart resulting in products of a commercial quality and supported by appropriate certificates can be arranged at NPPs.

The first option currently stands for the standard LRW management method with relevant improvements introduced at the final RW conditioning phase.

The above results in at least 3 challenges: waste package transportation logistics, stability of the package itself under long-term storage conditions and the natural safety of biologically hazardous components emplaced into it. It is clear that with a guaranteed service life of NZK-type concrete containers of 50 years [17] and a comparable period of

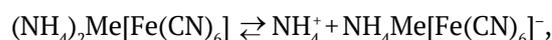
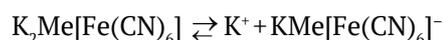
cement compound stability, their behavior cannot be reliably predicted even for 300–500 years ahead. The uncertainty in the long-term stability of engineered safety barriers should promote the development of compounds that are also characterized with natural safety, the level of which primarily depends on the form of substances contained in them. It seems quite obvious to include into the cement composition some compounds that would stay inactive during their interaction with groundwater, for example, due to extremely low solubility, which drastically reduces the concentration of radioactive substances in the pore water and, accordingly, the diffusion flux of these substances.

This reasoning is most consistent with the second approach, which in some Russian scientific papers devoted to this problem was called "ion-selective sorption of radionuclides" [18].

The approach assumes that certain materials can effectively absorb radionuclides regardless of the process solution composition. To validate this hypothesis, ^{137}Cs sorption by mixed hexacyanoferrate precipitates is considered which, from a scientific perspective, is believed to be an extremely flawed example [19–21]. The ion-exchange properties of these sparingly soluble double salts have long been proven [22–24] and the corresponding selectivity series have been identified excluding the specificity of these compounds to cesium radionuclides.

The ion-exchange nature of the sorption process with its inherent reversibility means the presence of a rigid relationship between the efficiency of the material governed by its internal structure [25] and the total chemical composition of the solution subject to treatment. For all ion-exchange materials, the equilibrium exchange capacity depends on two factors: the number of groups capable of exchange and the relative concentration of absorbed ions.

Usually sparingly soluble double salts of potassium or ammonium are used as sorbents with their sorption characteristics depending on an available set of equilibria developed between ionogenic groups and ions contained in the double ionic layer of the solution adjacent to the granule surfaces. In this regard, the initial sorbent form dissociation plays a vital role:



where Me are Ni^{2+} or Cu^{2+} cations.

It should be noted that dissociation in the second step means salt transition into a dissolved form.

Quite likely that formation of most exchangeable groups on the surface of a polycrystalline layer

covering the sorbent carrier granules may be described by the Henderson-Hasselbach equation.

$$p[K^+] = pK_{dis} + \lg \frac{\alpha}{1-\alpha}$$

$$p[NH_4^+] = pK_{dis} + \lg \frac{\alpha}{1-\alpha},$$

where K_{dis} is the apparent dissociation constant of functional groups along the i -step; α is the dissociation degree of functional groups belonging to the considered dissociation stage at a fixed pH value, calculated as:

$$\alpha_1 = \frac{KMe[Fe(CN)_6]^-}{K_2Me[Fe(CN)_6]^-},$$

$$\alpha_1 = \frac{NH_4Me[Fe(CN)_6]^-}{(NH_4)_2Me[Fe(CN)_6]^-}.$$

For nonporous crystalline salts, the concentration of exchange ions from the dissociation of functional groups is assumed to be concentrated in a double ionic layer and, in the first approximation, is equal to the concentration of these ions in the solution. Thus, their increased concentration in the purified solution will decrease the equilibrium and, consequently, the working capacity of the sorbent.

In mixed solutions, the concentration of electrolytes in the double ionic layer is proportional to their concentration in the solution. Due to sorbent interaction with such a solution, all mobile ions get redistributed. Radiocesium ions contained in liquid radioactive media interacting with free ionogenic groups practically do not change their equilibrium concentration due to the analytical insignificance up to reaching the maximum allowable specific activity level established for the sorbent. Fluctuations in the concentrations of structure-forming counterions in the treated solution will change significantly, firstly, the amount of cesium radionuclides capable of absorption and, accordingly, the rate of active group absorption thereby affecting the solution decontamination efficiency.

The use of weakly dissociating ion-exchange resins has proved that the number of groups capable of sorption can be increased by changing the ionic form of the commodity supply to counterions, for which the functional groups of the ion exchanger show less affinity. In the considered case, such ions are the light alkali metal ions of lithium and sodium. This operation can be implemented both at the stage of sorbent preparation and directly during its application. Due to the replacement of initially structure-forming potassium and ammonium cations by sodium ions, which occurs in solutions with

salt background governed by salts of the latter, the dissociation degree increases, as well as the working exchange capacity. This may explain the phenomenon of hexacyanoferrate sorbents capable of removing cesium radionuclides from NPP concentrates with their salt background based on sodium nitrate. Therefore, one can conclude that sodium ion concentration should also fall within a certain range, in which the displacement of commodity form counterions has already been provided, but the suppression of dissociation has not yet occurred.

Thus, all external manifestations of hexacyanoferrate sorbent specificity can be explained by well-known general concepts associated with ion-exchange processes.

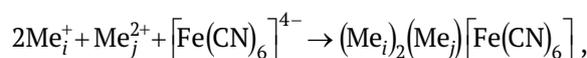
Therefore, in case of solutions with a variable composition, the required degree of radiocesium extraction via their treatment and practically acceptable specific activity of the sorbent can be basically achieved if:

- specific consumption of the sorbent remains constant by adjusting the contact time with the process solution;
- contact time remains constant by adjusting the specific consumption of the sorbent or the composition of the solution.

A most advanced approach enabling simultaneous implementation of these options provides for the application of an ion-exchange filtration method with parallelly connected elements. The system can be set easily according to the features of liquid radioactive medium (hereinafter referred to as LRM) and sorbent if a single condition is met: hydrodynamic, physical and chemical properties of a sorbent batch loaded into parallelly connected filters should be stable.

If sorbents with strictly reproducible characteristics set forth in regulatory and technical documentation cannot be reliably supplied, it hinders the development of a unified LRM management technology, similar to CWT or UTF (condensate cleaning) systems. In this regard, we believe that the performance characteristics of the Termoksid-35 sorbent [26] could be taken as a standard for the development of new sorbents, the acceptable reproducibility of which has provided its popularity.

Basically, co-precipitation of radionuclides with hexacyanoferrates directly into LRM also seems a quite feasible method deserving further development. The formal reaction for the double hexacyanoferrate formation can be represented by the following equation:



where Me_i are alkali metal and ammonium cations; Me_j are 3d-metal cations.

Thus, sparingly soluble double salts form all the cations found in the drain water. It should be noted that the radionuclides are not capable of forming a solid phase on their own, but the presence of their chemical analogues provides solution purification due to the coprecipitation reaction with isomorphic carriers. Given this approach, a reducing environment should be maintained in the process solution providing the cationic form of corrosion products with a charge of 2+.

It is well known that quite low solubility of double hexacyanoferrates promotes the formation of colloidal forms that basically cannot be treated by the accepted sedimentation and filtration methods. Studies show that the generated fine precipitates are most effectively separated from the mother liquor by membrane filtration methods. The use of anisotropic cartridge elements for microfiltration purposes in this mode provides the retention of the largest particles in the pores forming an auxiliary filter layer (Figure 1) characterized with sorption properties [27].

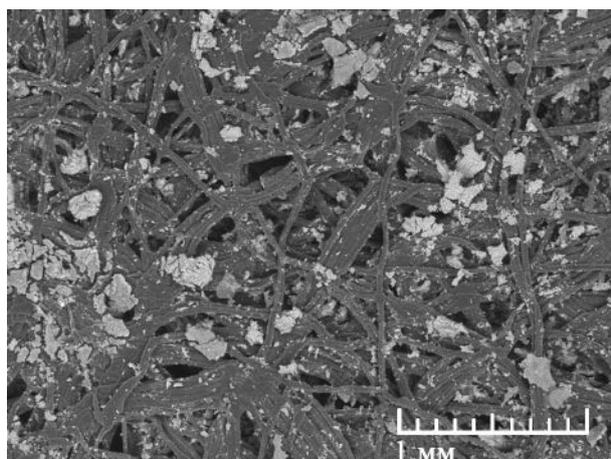


Figure 1. Morphology of a layer formed on the outer surface of a filter element

There is every reason to believe that the accumulated mass of the sorbent, which can occupy 70–80% of the geometrical cartridge volume, will continue to perform the ion exchanger functions.

If the sorbent quality proves to be the main reason for the unattainability of the desired performance in terms of solution treatment from radioactive cesium, then the problems associated with activated corrosion products extraction from LRM, primarily ^{60}Co radionuclides, can be explained by anions found in the solution and capable of binding these metals into complex forms [11, 28–33]. It is believed that these forms are inactive in sorption and precipitation processes and retain a significant part of the radionuclides in the treated solution.

During binary complex formation, the proportion of the complex metal form α is related to the concentration of free ligand $[L]$ and stepwise stability β according to the following equation:

$$\alpha = \frac{\beta \cdot [L]}{1 + \beta \cdot [L]}.$$

Uncharged complex compounds remain inactive in the sorption processes, including those providing co-precipitation of radionuclides with non-isomorphic carriers: in the considered case, these are represented by doubly charged anions: tetraborates, monohydrophosphates, carbonates, sulfates and oxalates.

It should be noted that EDTA ions form negatively contaminated complex compounds that are active in sorption process, but only when it comes to anion-exchange ones, which also occur on metal oxyhydrate compounds during their coagulation in a weakly acidic region.

Inorganic ligands form complex compounds with doubly charged non-ferrous metal cations accounting for most of the corrosion products in structural materials: their formation constant is ~ 100 , while for complex forms with oxalate ions it amounts to $\sim 10^5$ and for those with EDTA ions to $\sim 10^9$. Assuming the inactivity of complex forms in existing treatment systems, the solution purification factor K_{pur} is inversely proportional to the fraction of the complex form:

$$K_{\text{pur}} = \frac{1}{\alpha}.$$

In actual operating systems $K_{\text{pur}} \geq 100$, i.e. $\alpha \leq 0.01$. In this case, the equilibrium concentration of the ligand can be calculated as:

$$[L] = \frac{\alpha}{\beta} = \frac{1000 \cdot \text{MB}}{\beta \cdot K_{\text{pur}}},$$

where MB is the molecular weight of the ligand, or, expressing the concentration in mg/dm^3 accepted in practice:

$$[L] = \frac{1000 \cdot \text{MB}}{\beta \cdot K_{\text{pur}}}.$$

Thus, a purification factor of 100 for activated corrosion products can be achieved at a concentration of oxalate ions of $\sim 100 \text{ mg}/\text{dm}^3$, and of EDTA-ion level of only $0.1 \text{ mg}/\text{dm}^3$. However, such amounts of oxalate ions are quite unlikely to be present in the distillation residues. Nitrate ions are very weak oxidizing agents, although due to high temperature close to the boiling point of a saturated sodium nitrate solution ($\sim 108^\circ\text{C}$), presence of catalysts and the evaporation duration, they somehow intensify the oxidation of oxalate ions [34], but still are not

able to destroy thermally stable compounds, namely, EDTA ions and relevant complex compounds. It seems unlikely that such low equilibrium concentrations of these ligands may be reached using other available oxidizing agents.

Permissible concentration of inorganic ligands is already considered in the gram range, which can be reached in radioactive concentrates resulting from drain water evaporation. It is considered most likely that it is precisely these circumstances prevent high-level purification of solutions when the CEC crystal phase is washed out, even after the organic compounds get oxidized with ozone [35]. A most interesting point should be noted: sufficiently fine purification from ^{60}Co could be achieved due to the precipitation with sulfur-containing ligands [36], in particular, diethyldithiocarbamate. In fact, the purification was provided by the exchange of ligands, which resulted in a complex form with a much lower solubility.

A similar method changing the ligands' form of existence in LRM can be also implemented by replacing the central atoms. This approach is most typically used in the ion exchanger regeneration when large amounts of ions, to which functional groups show less affinity, displace ions with a much higher binding energy. This challenge can be most easily addressed assuming the use of calcium ions forming sparingly soluble compounds with all inorganic ligands and oxalate ions and strong complexes with EDTA ions. In addition, the excess of these central atoms due to rather high solubility of calcium hydroxide even in weakly alkaline media characteristic of radioactive concentrates results in the displacement of corrosion products in

microquantities from their complex forms followed by co-precipitation in the form of oxyhydrates with extremely low solubility. It should be noted that such a process can be implemented under homogeneous precipitation conditions resulting in solid solutions of activated corrosion products in a matrix of calcium carbonate or iron and manganese oxyhydrates. However, this reaction is only possible in case of equally charged deposited cations, in our case it should be equal to $2+$. Thus, to create optimal conditions for co-crystallization, the oxidation mode should be replaced by a reducing environment, for example, by introducing hydrazine hydrate.

Sufficiently complex systems for radionuclide removal seem feasible only if RW are shipped to the final disposal sites in a regular manner with high costs covered by operating organizations. Most likely this is the case of NPP decommissioning, so the main purpose of the above reasoning is to create awareness of those developing relevant engineering technologies and equipment, as well as of the design organizations for the fact that the decontamination of multicomponent solutions, which are radioactive concentrates, is not such a simple task that can be solved by some universal methods. This approach contradicts the principle that evolved in hydrometallurgy [37] — matching the properties of interacting media with operating modes, the neglect of which leads to the results given in [13]. In the light of the foregoing, only local purification systems are supposed to be highly efficient with their structure being adapted to the composition of a particular waste stream (Table 1).

Table 1. List of streams and volume of discharges into the wastewater treatment system of new generation NPPs with WWER-640 reactor units [38]

Stream	Average salt content, g/dm ³	Volume, m ³ /year		Average consumption, dm ³ /h	Specific activity of the streams, GBq/m ³
		Power operation	Under PPM		
Water from hydraulic discharge, loosening and filter cleaning	0–2	432	148	66	10^3 – 10^{-3}
Regeneration solutions of ion exchange filters	40–50	108	–	12	10^{-1} – 10^{-4}
Evaporator chemical washing solutions	40	50	50	11	10^1 – 10^3
Washing water from chemical washing of evaporators	0–2	50	50	11	10^2 – 10^{-5}
Spent decontaminating solutions	5–30	20	75.5	11	1 – 10^{-7}
Wash water from the decontamination of equipment and premises	0–2	50	200	29	10^2 – 10^{-2}
Water from sampling and radiochemical laboratories (including the primary circuit)	0–40	650 (3)	80	83	10^2 – 10^{-2}
Uncontrolled leaks from auxiliary system equipment, drains of auxiliary system equipment and unaccounted leaks (20%)	–	268	116.5	44	–
Total volume of water treated by the system	–	1,630	720	268	–

The above list demonstrates that only three streams may account for significant amounts of complexing substances: spent decontaminating solutions, washing water from chemical washing of evaporators and water for the hydrotransfer of spent sorbents.

As follows from the data shown in Figure 2, the drains of the spent sorbents storage tank will necessarily contain boric acid and, therefore, must be sent to already available system for its recycling.

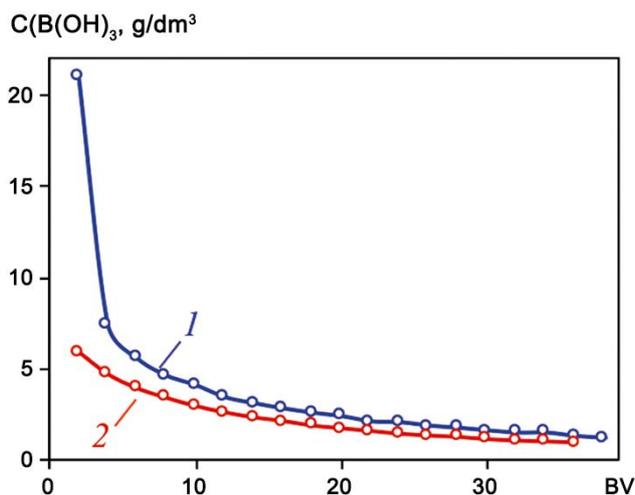


Figure 2. Output curve of boric acid inflow into transport waters during hydraulic unloading and loosening of the borate form of the AB-17-8 anion exchanger saturated with boric acid with a concentration of 40 g/dm³ (1) and 6 g/dm³ (2), BV is the column volume

As already noted above, the complexonates of activated corrosion products are anions that are well sorbed by low-basic anion exchangers, the production of which has already been mastered by Russian industry [39]. Inactivation of inorganic ligands, usually represented by various phosphates, will be provided by protonated forms of cation exchangers. The minimum output of the unit providing continuous processing of these LRM will be 0.05 m³/h. The spent ion exchangers are transferred to the spent sorbent storage tank. If filter containers are used, these are disposed together with the sorbents.

The use of ion-exchange methods providing the complexones and complexonates removal from washing and decontamination solutions is practically excluded due to the strong competitive action of nitrate ions. In these systems, preference should be given to precipitation methods followed by radiocesium removal. The estimated capacity of such a continuous system is 0.025 m³/h.

The remaining solutions do not contain complexing substances, therefore, for their deactivation a system may be used providing separation of

dispersed impurities, isolation of activated corrosion products on a complexing cation exchanger with carboxyl or phosphoric acid groups and removal of radioactive cesium on a hexacyanoferrate sorbent. Similar approach was tested during LRM processing by the NAVY of the Russian Federation [40].

All these units may provide the supply of “pure” solutions to the evaporator and dryer for salts and fully implement the “ideal” environmentally friendly method of NPP LRM processing declared in [1, 10, 13] avoiding any biologically unacceptable substances from being released into hydraulic structures. It is clear that this concept may be implemented only if adequate storage capacities are available enabling collection of the corresponding LRM, installation of which at existing and already designed NPPs is difficult, and therefore application of the considered approach seems unlikely in a reasonable period of time. We hope that the authors of this promising development will be able to provide its commissioning.

In the meantime, under the proposed NPP designs, a traditional drain water processing system is generally accepted. To reduce the waste volume subject to final disposal, it is proposed [41]:

- to prevent the release of boric acid into the drain water;
- to avoid the recycling of filters from special water treatment systems, the service life of which exceeds a year;
- to provide the removal of regeneration solutions from the system regulating the water-chemical mode of steam generators, water from a special laundry room and sanitary inspection rooms to the hydraulic NPP structures.

The latter point seems to be of a greatest interest. It is assumed that an autonomous system based on ion-selective inorganic sorbents should provide the protection of the NPP hydrosphere [41]. Therefore, these can be upgraded up to the establishment of flexible flowcharts adapted to the composition of decontamination solutions used to treat protective clothing. Therefore, permissible discharge standards can be certainly provided.

The salt background of spent regeneration solutions planned to be discharged into the spray pool involves nitrate salts of sodium, ammonium, and monoethanolamine. It is well known that nitrogen compounds contribute to the biomass growth, therefore, in the long term, it will lead to accelerated biofouling of heat transfer surfaces in cooling towers and separating surfaces of baromembrane apparatuses, which are already widely used in cold water systems at nuclear power plants.

According to the SanPiN standards, the allowable concentration of monoethanolamine in household

waters is set to 0.5 mg/dm³. Oxidation is considered a basic method used to remove this compound from process waters. It is well known that it proceeds most easily in case of alcohol group eventually resulting in α -aminoacetic acid, a compound well known and used even in pharmacology under the name of glycine. This primary amino acid constituting to the structure of many proteins is well absorbed by microorganisms, thereby contributing to the increased content of various organic substances in the internal water bodies at nuclear power plants. The ambiguity of this compound is also determined by its complexing properties providing the retention of non-ferrous and heavy metal ions in solution, decreasing the performance of CWT systems providing nuclear power plants with high-purity water.

Thus, a local treatment system for spent regeneration solutions should provide not only the non-exceedance of the standards establishing the maximum radionuclide concentrations in wastewater, but also their quality in terms of the components, the release of which into industrial NPP reservoirs can, in the long-term, result in environmental issues overgrowth into the challenges associated with the safe operation of power units.

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