Low-Waste Process for Chemical Decontamination of Primary Circuits of Water-Cooled Nuclear Power Installations Using Ion-Exchange Methods for Treatment of Decontaminating Solutions

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Received June 18, 2010

Abstract—The possibilities of improving the efficiency of “low-concentration” processes for chemical decontamination of primary circuits of water-cooled nuclear power installations are examined. Laboratory experiments demonstrated the advantages of chemical dissolution of radioactive deposits of the circuits, combined with sorption treatment of the decontaminating solutions. The influence of equilibrium pH of solutions on the volumetric distribution coefficients of Fe(II), Fe(III), Co(II), and Cr(III) ions between decontaminating solutions based on complexing acids and modified AV-17-8 chS strongly basic anion exchanger was studied. The main characteristics of processes involving sorption on the anion exchanger in the EDTA form from solutions containing ethylenediaminetetraacetic, oxalic, and citric acids and on the anion exchanger in the HEDP form from solutions containing 1-hydroxyethane-1,1-diphosphonic acid, hydrazine, and ammonia are compared.

Keywords: decontamination, coolant, primary circuit, nuclear power installations

DOI: 10.1134/S1066362210060044

Among procedures developed for decontamination of primary circuits of nuclear power installations (NPIs), the most widely used in the world are liquid, circulation procedures for chemical treatment of their internal surfaces. Radionuclides affecting the radiation situation in the maintenance area of a nuclear reactor, irrespective of their origin in the primary circuits [activated corrosion products (ACPs), fission products (FPs)], are removed from the equipment surfaces and converted to the soluble state together with corrosion product (CP) deposits as a result of the action of solutions of specially chosen reagents. The solutions should efficiently dissolve CPs, firmly retain chemical elements and radionuclides present in them in the dissolved state, and, at the same time, exert a minimal corrosive effect on structural materials (SMs) of the circuits.

The main result of decontamination of the primary circuits of NPIs in the assembled state is reduction of the γ-radiation dose rates from the surfaces of the circuit equipment. The generally accepted characteristic of the decontamination efficiency is the decontamination factor Kdec, i.e., the ratio of the levels of the radioactive contamination of the equipment before and after treatment.

Irrespective of the decontaminating solution composition, such processes include a number of similar steps: introducing solutions of the reagents into the primary circuit; adjusting the conditions for their circulation along the circuit and the temperature, chemical, and hydraulic parameters of the dissolution of radioactive contaminants; removing spent solutions; bringing the water quality in the circuit to the purity parameters prescribed by the regulations.

Despite apparent simplicity, the final decontamination steps involve significant “hidden” problems. In the existing processes, the decontaminating solutions and wash waters are always drained. In so doing, their circulation in the circuit is, as a rule, stopped. The drain-
The initial values, in the radionuclides, which leads to an increase, relative to secondary contamination of the treated surfaces with ready existing suspensions, on the circuit surfaces. Of the newly formed precipitates, along with the ions of elements present in CPs leads to deposition thermore, the use of reagents forming precipitates with removal of the deposited radioactive suspensions. Furthermore, the use of reagents forming precipitates with the ions of elements present in CPs leads to deposition of the newly formed precipitates, along with the already existing suspensions, on the circuit surfaces.

Deposition of suspensions and precipitates leads to secondary contamination of the treated surfaces with radionuclides, which leads to an increase, relative to the initial values, in the γ-radiation dose rates from blind sections of the circuit after decontamination. The subsequent transfer of the precipitates in the course of starting-up NPI and bringing it to the required power can lead to still more hazardous consequences. For example, transfer of the suspended material to the reactor core can lead to a decrease in the coolant flow rate, to temperature anomalies, and to overheating of fuel assemblies.

In addition, irrespective of the solution composition, the use of technologies involving in the final step drainage of decontaminating solutions and wash waters, leads to the formation of large volumes of liquid radioactive wastes (LRWs). The need for alternative solutions to the problem largely determined the lines of improvement of processes for decontamination of NPI primary circuits.

Initially the researchers’ efforts were aimed to attain the maximal decontamination factors for the equipment. Processes using relatively concentrated reagent solutions (so-called “hard” or “high-concentration” processes) were preferred. In these technologies, solutions with reagent concentrations of 1 wt % and higher were used for CP dissolution. Citric, oxalic, and phosphoric acids and their salts, and also mixtures of acids in combination with complexing agents [mainly ethylenediaminetetraacetic acid (EDTA) and its salts] were used as main reagents [1–4].

“Hard” decontamination processes involve alternating introduction into the primary circuit of alkaline oxidizing and acidic reducing solutions with intermediate drainages of the solutions and washings to remove their residues. The minimal number of steps, each including introduction of solutions into the circuit, their removal from the circuit by drainages, and washing of the circuit with HPW, is three. Even if we assume that, in each step, before introducing the next solution, the discharge volume is equal to two circuit volumes, the LRW amount by the end of decontamination will reach six circuit volumes. In practice it is, as a rule, considerably larger.

Intrinsic drawbacks of “high-concentration” processes, associated primarily with the formation of large LRW volumes, led to the fact that, since the late 1970s, the general trend in the development of circuit decontamination processes was toward a decrease in the concentrations of reagent solutions. “Low-concentration” processes and new principles of organization of NPI decontamination were brought into practice [1, 3–6]. The decrease in the reagent concentrations to the levels that do not critically affect the efficiency of dissolution of radioactive deposits had two major goals. The first goal was to decrease the LRW volume by decreasing the filling factor of the circuit with HPW in the traditional “drainage” scheme of the final step of the decontamination. The second goal was to create conditions for replacing drainage by dissolution of internal circuit contaminations, combined with removal of major CP components and radionuclides from solution on ion-exchange cleanup filters. Removal of ACPs and FPs from solutions on ion exchangers simultaneously allows ion-exchange regeneration of the reagents in the course of decontamination. Removal of the reagents, CP elements, and radionuclides from the circuit on ion-exchange filters decreases both the volume and the activity of LRW and simplifies its processing. At the same time, with respect to a decrease in the dose rate from the equipment ($K_{dec}$), the known processes based on these principles are inferior to the traditional processes.

The above drawback is not very significant, because it is compensated to certain extent by the possibility of more frequent decontaminations with lower time and material expenditures, owing to lower demand for the reagents and high-purity water, lower corrosion loss of SMs, and decreased LRW volume [6]. The general concept of “low-concentration” technologies with ion-exchange solution treatment was developed for the first time for decontamination of NPI...