ASSESSMENT OF CAPABILITIES FOR EFP CARTRIDGE FILTER DECONTAMINATION

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Under this study, tests were carried out to assess the decontamination options for cartridge filters of EFP type and it was established that a mixed method should be used to achieve maximum efficiency of EFP cartridge filter decontamination. The method should combine sequential processing using water and/or acid solutions in hydraulic impact mode and ultrasonic decontamination by immersion. The developed decontamination technique provides quite complete recycling of the target element to enable its further use in the industrial process and prevents the release of nuclear material into the environment during further radioactive waste management under long-term storage conditions.

Keywords: *decontamination, filter cartridge, radioactive contamination, decontamination efficiency, filter holder, ultrasound, hydraulic impact mode, dynamic mode, radioactive waste.*

Radiochemical production basically provides for the use of various filters involving reagents and substances with different properties — from water to aggressive acids and alkalis. Moreover, filtration processes often occur at high temperature and pressure levels. For this reason, in the filter production, materials such as Teflon, polypropylene, fluoroplastic, polyester, polyphenylene sulfide and other substances considered resistant to the above factors are used.

Mayak has accumulated certain inventory of spent polypropylene or polyethylene filters of the EKOPLAST brand manufactured by Express-Eco-Filter LLC (Obninsk) contaminated with various radionuclides, including plutonium-239. Sufficiently complete recycling of the target component in the production process has required the development of a method providing the decontamination of EFP-type cartridge filters (cartridge filter element) that would ensure maximum degree of their decontamination. The study is seeking to prevent the spread of nuclear material into the environment during further management of the spent filters, which are considered solid radioactive waste (SRW) under long-term storage conditions.

Under the study, EFP-type cartridge filters of the EKOPLAST brand were evaluated considering the prospects for their decontamination using ultrasound, as well as via their treatment with counter-current purified water or acid solutions.

Filters of the EFP-101-L type were selected for testing (Figure 1). The EFP-101-L (EFP-PE) depth filter element was designed to remove particles of over 1 μm in the diameter from liquid media, including highly aggressive ones, in a wide temperature range. It is basically applied for the preliminary and final decontamination of liquids from solid mechanical particles [1].

Figure 1. Appearance of the EFP-101 depth filter element

EFP-type cartridge filter elements operate according to the dead-end microfiltration principle, namely, this process can be referred to depth filtration method (due to the heavy thickness of the filter membrane). Solid particles retained by the filter element get accumulated inside the membrane up to a certain hydraulic resistance level. The multilayer porous structure of the filter element provides high dirt-loading capacity of the elements, especially as it comes to solid mechanical impurities.

The EFP-101 type filter element is made of ultra-high-molecule polyethylene (UHMWPE) (TU 2211-001-98386801) with no additives and fillers: it is a two-layer hollow porous cylinder with a height of 90 to 250 mm, in which the total porosity and size of the pores decrease from the outer layers to the inner ones. The layers are formed from UHMWPE powders with different dispersity levels. Thermal bonding of the powders occurs during product sintering in a mold. The liquid is filtered from the outside part to the inside part of the filter [2].

The filter element has a dead-end design with one end piece having no hole, while the other through-hole one has a threaded hole. Its faces are made of polypropylene. Particle retention efficiency amounts to over 98% at the stated filtration rating and the recommended fluid flow rate.

Table 1 presents the key characteristics of the tested EFP with a height of 125 mm.

Table 1. Main technical data for the filter element

Indicator	Value
Brand of the element	EFP-101-L/1(10)-125-R
Filtration rating, μm	1
End and adapter element color	Red
Particle retention efficiency, not less than, %	98
Nominal fluid flow rate (μ = 1 cPs, t = 20 °C), m³/h	0.5
Initial hydraulic resistance in water at 20 °C and nominal flow rate, MPa	25.3
Operating temperature range, °C	from –60 to +100
Maximum pressure drop, MPa	1.6
Threshold reverse differential pressure, MPa	1.2
pH range	1–14

High thermochemical UHMWPE resistance, similar to that of fluoroplast-4, makes it possible to use filter elements to filter aggressive liquids, acids, alkalis, alcohol-containing products, including highly aggressive ones at normal and elevated temperatures ranging from –60 to +100 °C [2]. The high mechanical strength of the EFP brand elements enables the filtration process at high pressure drops and allows applying them in case of viscous media.

For research purposes, a single-cartridge filter holder of the DFP 201D-125AO brand was used (Figure 2). According to its designs, it can accommodate a single filter element with its possible height ranging from 125 to 1,000 mm and can be applied in neutral and aggressive liquid media to decontaminate them from mechanical and colloidal particles with a capacity of not more than 2 m³/h (for water) [3].

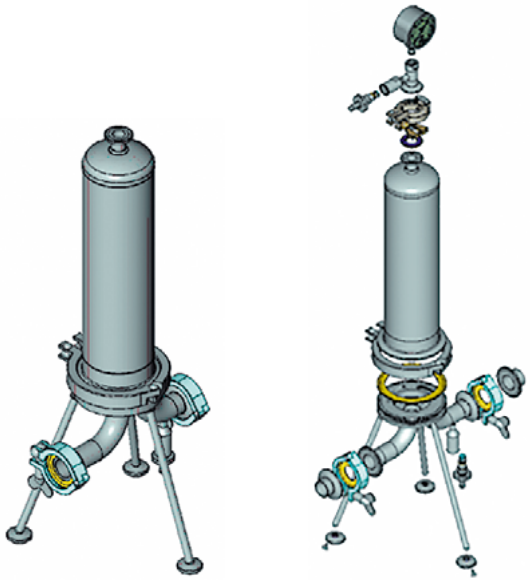

Figure 2. Filter holder brand DFP-201D-125AO

The ultrasonic decontamination method involved the following operations: the filter was immersed into the working volume of an ultrasonic bath filled with decontamination solutions at a temperature of 20 to 25 °C. The following decontaminating solutions were applied:

Solution 1 — 30 g/dm³ HNO_3;
Solution 2 — 60 g/dm³ HNO_3 + 5 g/dm³ $H_2C_2O_4$;
Solution 3 — 250 g/dm³ HNO_3 + 10 g/dm³ $H_2C_2O_4$;
Solution 4 — 150 g/dm³ H_3PO_4 + 5 g/dm³ of urea.

Washing was carried out in cycles of 30 min. For solution No. 3, the duration of each cycle amounted to 60 min. After each decontamination cycle, the decontamination solution was replaced and the filter was washed under running water for 5 min, then the filter was dried in a fume hood for at least 1 day.

Since the operations involved a single filter element, it was re-contaminated after being washed.

Table 2. Decontamination results for a filter contaminated with alpha-emitters

Number of decontamination solution	Initial contamination, part./(cm²·min)		Residual contamination, alpha part./(cm²·min), after					
			first cycle		second cycle		third cycle	
	APFD	smear	APFD	smear	APFD	smear	APFD	smear
1	65,000	118–1,226	49,000	51–670	30,000**	40–610	Not performed	
2	30,000	40–610	8,000	35–450	6,200	24–388	4,500**	15–342
3	4,500	19–342	3,200	10–60	1,511	6–12	300	4–7
4***	7,550	250–1,200	2,807	13–63	445	4–43	365	2–27

* APFD - alpha particle flux density.
** Filter element decontamination was continued in the next decontamination solution.
*** The re-contaminated filter element was subjected to decontamination.

To do this, the contaminant solution containing such radionuclides as ^{238}Pu, ^{239}Pu, ^{241}Am, ^{234}U, ^{237}Np, etc. was passed through the filter installed in a DFP-201D-125AO filter holder.

The initial and residual contamination of the filter was assessed: the particle flux density was measured directly at its surface, as well as using the method of a "dry" smear taken from it by a filter paper.

Radioactive contamination of the filter was measured using a radiometric unit MKS-AT1117M. Residual removable radioactive contamination was checked at 1—3 points. The contamination level was calculated as the arithmetic mean of three to five consecutive measurements made at each point.

To evaluate the filter washing efficiency, the decontamination factor K_D was calculated using the formula below:

$$K_D = A_0/A_R, \quad (1)$$

where A_0 is the initial contamination of the filter, part./(cm²·min), A_R is the residual contamination of the filter after the decontamination stage, part./(cm²·min).

Tables 2—4 summarize the results of the decontamination process. Table 2 shows that decontamination solutions No. 3 and No. 4 can be considered as most effective when it comes to the removal of alpha radionuclides. Residual removable alpha contamination after three cycles of decontamination did not exceed 27 part./(cm²·min), APFD did not exceed 365 part./(cm²·min).

Nitrogen-oxalate solution No. 3 (due to increased reagent concentrations) turned out to be more than twice as effective as solution No. 2. For three cycles of filter processing in the presence of ultrasound, it was possible to reduce the radioactive contamination from 6 to 20 times (Table 3).

During the study, it was noted that the decontamination efficiency increased along with the washing time. Thus, for example, when the filter was treated with solution No. 3 in the third decontamination cycle, a two-fold increase in the exposure time has allowed to increase the decontamination factor to 5.

Table 3. Filter decontamination efficiency for alpha radionuclides

Number of decontamination solution	Initial contamination, part./(cm²·min)	Decontamination factor K_D after			Total K_D
		first cycle	second cycle	third cycle	
1	65,000	1.3	1.6	–	2.2
2	30,000	3.8	1.3	1.4	6.6
3	4,500	1.6	1.9	5.0	15.0
4*	7,650	2.8	5.8	1.3	21.1

* re-contaminated filter was subjected to decontamination.

Table 4. Results of filter decontamination from beta-radionuclides

Number of decont. solution	Initial contamination, part./(cm²·min)		Residual contamination, beta part./(cm²·min), after					
			first cycle		second cycle		third cycle	
	BPFD	smear	BPFD	smear	BPFD	smear	BPFD	smear
1	200	180–216	180	101–115	120	37–76	Not performed**	
2	120	37–76	72	20–40	45	20–40	20	3–20
3	220	41–213	190	background –176	60	background –50	30	background –25
4*	38	15–23	27	10–16	22	9–13	19	6–11

* BPFD is the beta particle flux density.
** Filter decontamination was continued in decontamination solution 2.
*** The re-contaminated filter element was subjected to decontamination.

For three treatment cycles involving solutions No. 3 and No. 4, the decontamination factor turned out to be equal to 15.0 and 21.1, respectively.

Table 4 shows that the levels of initial beta contamination of the filter elements were negligible. All decontamination solutions provided high decontamination efficiency as regards beta emitters. The residual removable contamination after three treatment cycles did not exceed 25 part./(cm^2·min)and the beta particle flux density did not exceed 30 part./(cm^2·min).

Alpha-spectrometric [4] analysis of spent decontamination solution samples showed (Figure 3) that their volumetric activity decreased along with an increase in the number of treatments (decontamination cycles), which could be explained by a decrease in the overall level of radioactive filter contamination after each subsequent cycle. Solution No. 3 has demonstrated the highest decontamination ability. The volumetric activity of the spent decontamination solution after the third cycle decreased by three orders of magnitude compared with the result obtained after the first cycle.

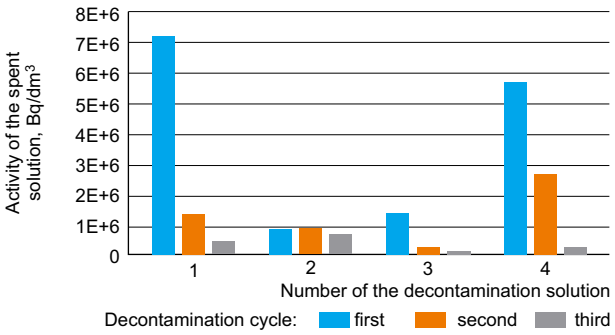

Figure 3. Changes in the volumetric activity of spent solutions depending on the decontamination cycle

Before recontamination (when solution No. 4 was applied), the filter installed in the filter holder was pre-washed with water. Wash solution sample was analyzed: it was discovered that the activity concentration of the solution was two orders of magnitude higher than the value obtained from the analysis of a sample taken from the spent solution No. 3 after the third decontamination cycle. Thus, it can be argued that only the outer surface of a filter can be washed if the immersion method with ultrasound is applied for decontamination purposes. It is impossible to achieve high-quality washing of the filter over the entire volume of the filter element by this method due to the shielding of the inner layers by the outer ones (the filter has a multilayer porous structure).

At the next stage, the prospects of cartridge filter decontamination by flushing the latter in the forward and reverse directions both in the dynamic mode and under hydraulic pressure mode were evaluated.

The filter was washed in a dynamic mode: the washing solution was successively passed through the filter holder under vacuum at a flow rate of 2—5 cm^3/s.

Hydraulic shock mode washing was performed with the washing solution passed through given periodic switching of the vacuum for 30 s and its turning off for 5 s.

The following solutions were applied as the washing ones:

- nitric acid with mass concentration from 30 to 100 g/dm^3;
- nitrogen-fluoride solution containing 380 g/dm^3 HNO_3 + 3 g/dm^3 HF.

The experiments were carried out both at room temperature of the solutions and at 60 °C.

Before each decontamination stage, the filter element was re-contaminated with a solution being a mixture of a clarified part and a precipitate. Alpha spectrophotometric analysis of the clarified part and precipitate showed that ^{239}Pu radionuclide, as well as ^{241}Am, ^{238}Pu, ^{232}Th, ^{235}U, ^{238}U and ^{239}Np and other alpha-active nuclides were those providing the most important contribution to the alpha activity of the solution. The precipitate volume was insignificant and amounted to no more than 5%. The total alpha activity concentration of the initial solutions containing contaminants ranged from $4.5 \cdot 10^3$ to $2.3 \cdot 10^7$ Bq/dm^3. The basis of the precipitate was barium and sulfur compounds. It also contained trace amounts of silicon, chlorine, phosphorus, iron and chromium.

At the post- decontamination stage, samples of spent washing solutions were taken to measure their total alpha activity [4]. Then the filter element was dried for at least 1 day. After being dried, the residual surface radioactive contamination of the filter element material was measured both in the flow and in the smear using the MKS-AT1117M radiometric unit.

Intermediate washings of the filter after the solution with the contaminants was treated and between the application of the basic decontamination methods (dynamic mode and hydraulic shock mode) were done using a nitric acid solution with a molar concentration of not more than 30 g/dm^3.

Tables 5 and 6 and Figures 4—5 summarize the results of the experiments.

Table 5 demonstrates that the filter decontamination with nitric acid solution by reverse flow in a dynamic mode amounted to 81.6%. Additional filter decontamination was done by reverse flow in the hydraulic shock mode. The total yield of the target component amounted to 93.2%.

Table 5. Results of filter element decontamination experiment involving nitric acid solution with a mass concentration of 100 g/dm³ at a room temperature

Operation	Solution volume V, cm³	Activity concentration of solution A, Bq/dm³	Total activity of solution ΣA, Bq	Degree of decontamination from ^{239}Pu, %
Filter contamination	1,400.0	4,546.4	6,365	–
Filtrate	1,325.0	276.0	366	–
Dynamic mode involving acid solution	590.0	8,300.0	4,897	81.6
Intermediate washing in a dynamic mode	570.0	480.0	274	6.9
	595.0	234.0	139	
Hydraulic shock mode involving acid solution	582.0	119.9	70	4.2
	595.0	110.1	66	
	580.0	145.0	84	
	335	94.9	32	
	335.0	94.9	32	
Intermediate washing in a dynamic mode	600.0	41.3	25	0.5
	400.0	8.0	3	
Total				93.2

Table 6. Results of the experiment on the filter cartridge regeneration with a nitrogen fluoride solution

Operation	Solution volume V, cm³	Activity concentration of solution A, Bq/dm3	Total activity of solution ΣA, Bq	Degree of decontamination from ^{239}Pu, %
Filter contamination	740	$2.28 \cdot 10^7$	$1.69 \cdot 10^7$	–
Filtrate	575	$8.8 \cdot 10^3$	$5.06 \cdot 10^3$	–
Dynamic mode involving nitrogen fluoride solution at a temperature of 20 °C	1,010	$1.5 \cdot 10^7$	$1.52 \cdot 10^7$	89.7
Intermediate washing in a dynamic mode	500	$1.44 \cdot 10^7$	$7.2 \cdot 10^5$	7.4
	563	$9.4 \cdot 10^5$	$5.3 \cdot 10^5$	
Hydraulic shock mode involving nitrogen fluoride solution at a temperature of 20 °C	550	$5.09 \cdot 10^5$	$2.8 \cdot 10^5$	1.9
	420	$9.9 \cdot 10^4$	$4.2 \cdot 10^4$	
Intermediate washing in a dynamic mode	560	$2.19 \cdot 10^4$	$1.23 \cdot 10^4$	0.1
	400	$2.21 \cdot 10^4$	$8.86 \cdot 10^3$	
Dynamic mode involving nitrogen fluoride solution at a temperature of 60 °C	550	$1.90 \cdot 10^5$	$1.04 \cdot 10^5$	0.7
	570	$2.99 \cdot 10^4$	$1.71 \cdot 10^4$	
Intermediate washing in a dynamic mode	565	$2.33 \cdot 10^4$	$1.32 \cdot 10^4$	0.1
	570	$1.58 \cdot 10^4$	$9.03 \cdot 10^3$	
Total				99.9

Table 6 presents the results of the experiment on filter cartridge washing with nitrogen-fluoride solution.

The table shows that filter washing with a nitrogen fluoride solution in a dynamic mode at a room temperature allowed reaching 97.1 % in its decontamination (taking into account filter washing with a weakly acidic solution of nitric acid in a dynamic mode). Further washing of the filter in the hydraulic shock mode reduced the number of radionuclides in it by 2.0 %. In general, nitrogen-fluoride solution applied as a decontamination solution allowed increasing the cartridge decontamination degree by 8 %.

The above data evidence that maximum extraction of the target component can be achieved by combining the two decontamination methods (dynamic mode and hydraulic shock mode).

To increase filter element decontamination level and to reduce the ADER, a combined method was tested. It combines sequential treatment involving water and (or) acid solutions in the hydraulic shock mode and decontamination by submersion involving ultrasound.

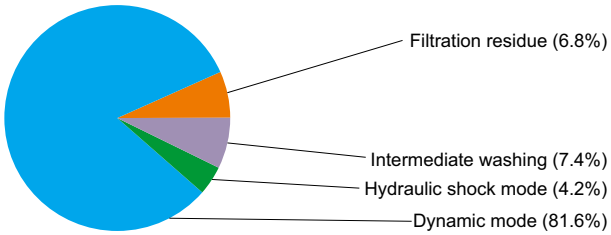

Figure 4. Diagram showing activity distribution in solutions (%) after washing the plutonium-contaminated filter cartridge with a nitric acid

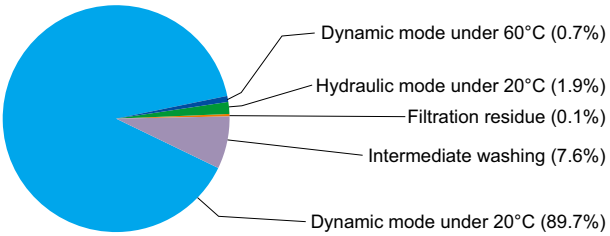

Figure 5. Diagram showing activity distribution in solutions (%) after washing the plutonium-contaminated filter cartridge with a nitrogen-fluoride solution

The results showed that introduction of an additional operation (ultrasonic decontamination) provides high-quality extra washing of the outer filter element layers and decreases by 2 times the ADER emitted by the filter, thereby reducing the personnel exposure during further management of filters.

Conclusion

The study evaluated the potential of EFP-type cartridge filter decontamination. Ultrasonic decontamination method turned out to be effective in dealing with the surface contamination of the cartridge providing high-quality washing of the filter element only as regards its outer layers. Acid washing of the filter cartridge with a reverse flow both in dynamic mode and in the hydraulic shock mode provided almost complete removal of radioactive contamination from its internal filter layers.

To achieve maximum level of EFP-type cartridge filter decontamination, a combined method should be used involving sequential treatment with water and (or) acid solutions in the hydraulic shock mode and decontamination by submersion using ultrasound.

The study showed that in all experiments considered at the post-decontamination stage, the residual content of plutonium-239 in the filter material matrix did not exceed a threshold allowing to treat the decontaminated filters as solid radioactive waste containing no nuclear materials. Moreover, after such decontamination is completed, the waste can be assigned to Class 3 RW (solid radioactive waste) instead of Class 1.

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Bibliographic description

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