

# REFINEMENT OF A DECONTAMINATION TECHNOLOGY FOR RADIOACTIVELY CONTAMINATED EQUIPMENT

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*This article analyzes the data on chemical decontamination methods dealing with radioactively contaminated surfaces. It considers the composition of solutions most commonly used for decontamination purposes. Numerical data are presented to illustrate the effectiveness of various decontamination methods. The paper considers an experiment on the decontamination of stainless-steel samples with water following cavitation treatment. The study reveals a dependence between the decontamination efficiency and the treatment time of the contaminated surface with a decontamination solution based on cavitation-activated water, which appears to be comparable with the results of the one involving an alkaline solution.*

**Keywords:** *radioactive waste, decontamination, surface contamination, decontamination solution, cavitation treatment, decontamination factor.*

## Introduction

Many countries are involved in the refinement of methods providing the decontamination of equipment and PPE during the operation of nuclear facilities, mining and processing of natural resources (oil, gas, non-ferrous metals, etc.) [1–9].

These tasks are closely related to the challenges being addressed in radioactive waste (RW) management within the framework of the general strategy for nuclear power development until 2025 [10–13].

Due to high RW disposal tariffs, the task providing for the direct disposal of all RW without its prior decontamination or reduction of its amount appears to be unsolvable from the economic point of view. At the same time, the topic of high-quality metal recycling after its decontamination has gained considerable interest.

In addition, the levels to which the working surfaces of equipment were contaminated affect the workplace environment for the personnel [7]. Therefore, to exclude additional sources of personnel exposure, formation of radioactive aerosols and the spread of radioactive contamination, timely decontamination of radioactively contaminated surfaces is required [8, 9]. This challenge can be addressed through the use of decontamination solutions based on water treated via the application of cavitation effects, i. e., cavitation method [5, 14–19].

This article summarizes the efforts focused on the improvement of the equipment decontamination method based on hydrodynamic cavitation effects.

### Existing decontamination methods for radioactively contaminated surfaces

Natural radionuclides of  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and their decay products are seen as sources of radioactive contamination during oil/gas production. In nuclear industry, radioactive contamination is mainly caused by uranium and plutonium isotopes,  $^{137}\text{Cs}$ ,  $^{90}\text{Sr}$ ,  $^{60}\text{Co}$ .

A lot of papers [20–25] were devoted to theoretical and experimental studies of decontamination processes with chemical decontamination methods using decontamination solutions seen as most common ones. During such decontamination, the bond between the contaminant and the surface is destroyed, radioactive contamination is removed from the surface and the radioactive substance passes into the decontamination solution [20]. Water, surfactants and complexing agents, acids, alkalis and some salts are considered as main components of decontamination solutions. In most cases, complex solutions are used, the composition of which is chosen depending on the forces holding the contaminant on the surface. Alternating use of different solutions, for example, acidic and alkaline, oxidizing and reducing often proves to be quite effective [26, 27].

Surfactants (surfactants) can be also used as decontamination solutions providing a decrease in surface tension and adsorption at the interface. A big number of colloidal-chemical properties that bind the radionuclide in the solution in the form of ions is responsible for this ability allowing to retain radioactive contaminants in solution and ensure their removal from the treated surface. Chelating compounds such as ethylenediaminetetraacetic acid (EDTA) and its sodium salt are introduced as complexing agents into decontamination solutions [21].

Condensed phosphates or polyphosphates, which form complex compounds with cations of alkaline earth and heavy metals, are also added to decontamination solutions. These complexing agents include: sodium diphosphate (pyrophosphate) ( $\text{Na}_4\text{P}_2\text{O}_7$ ), sodium tripolyphosphate ( $\text{Na}_5\text{P}_3\text{O}_{10}$ ), sodium tetrakisphosphate ( $\text{Na}_6\text{P}_4\text{O}_{13}$ ) and sodium hexametaphosphate (HMPN), which are high molecular weight compounds containing 34–38 ( $\text{NaPO}_3$ )<sub>n</sub> groups. In addition to complexing properties, condensed phosphates additionally impart suspending properties to decontamination solutions, thereby enhancing the colloidal-chemical action of surfactants. Certain organic and inorganic additives are introduced into decontamination solutions acting as washing process activators. Inorganic electrolytes regulate the pH level of the solution, increase the adsorption of surfactants, reduce its critical micelle formation concentration, thereby reducing the flowrate [22].

Application of decontamination solutions can be exemplified by methods of submerged decontamination and decontamination by levigation (rubbing) [12, 13]. Due to its simplicity and effectiveness, submerged decontamination is considered as a most widely used method. The method is implemented by immersing the contaminated product into a bath filled with a decontamination solution. This method can be used to process products of various configurations and sizes; the efficiency of submerged decontamination increases with stirring, circulation of the solution and temperature increase [28, 29]. The method is used to decontaminate removable contouring equipment, rigging, fittings, tools. When large equipment (steam generators, heat exchangers, large containers) are subject to decontamination, they are filled with a decontamination solution with its circulation and heating provided. High flowrate of the decontamination solution and, as a consequence, large volumes of generated liquid radioactive waste (LRW) is seen as a disadvantage of this method [30].

During levigation-based decontamination being considered as a simple, universal and widespread method, combined are the solution decontamination effect and the mechanical action. According to [20], the solution flowrate amounts to some 2–3 l/m<sup>2</sup>. In case of stainless steel, the decontamination factor amounts to 5–20. Employment of manual labor under radiation hazardous conditions is seen as a disadvantage of this method.

To intensify the process of chemical decontamination, ultrasound can be used, which accelerates the decontamination of metallic materials [23] providing sufficiently high processing efficiency achieved in a relatively short time, thereby reducing the volume of secondary LRW. However, firmly fixed contaminants, especially those that have diffused deeply into the base metal, are poorly removed, even if aggressive solutions are applied.

In addition to chemical decontamination methods, physicochemical and physicochemical methods are used as well [31–35]. During mechanical processing, the surface layer of material with radioactive contamination contained in it is removed.

Physicochemical methods include electrochemical decontamination, decontamination with pastes, melts and removable coatings, powdery metallized compositions, etc.

### Development of a theory explaining cavitation effects on water systems

The water modified via hydromechanical treatment (or activated water) can intensify a number of processes [14, 15]. Here, “activated water” is

thought to be a generally accepted concept of an active medium, i. e., a substance in which the distribution of particles (atoms, molecules, ions) over energy states is not equilibrium and population inversion occurs for at least one pair of energy levels.

Activation of solid and liquid systems causing some alterations in their physical and chemical properties, reactivity, defective (impurity) structure, etc. can be achieved through the application of various external influences: weak and strong. Such influences include, in particular, mechanical, magnetic, ultrasonic treatment, radiation exposure (for example, irradiation with gamma quanta and ion beams), as well as heat treatment. Activation methods can be divided into those in which the sample is destroyed entirely (dispersion) and those in which only the defective structure undergoes some changes.

Hydromechanical water treatment (considered as a rather strong effect) has been studied: it was shown that its subsequent activity is manifested both at the macroscale and at the microlevels (i. e., at molecular and sub-molecular levels). The essence of hydrodynamic action can be reduced to two mechanisms: propagation of shock waves near the collapsing cavitation microbubble and shock action of cumulative microstructures due to asymmetric collapse of cavitation microbubbles. Moreover, in this context, it does not matter which method is used to generate the cavitation microbubbles. These basic mechanisms are accompanied by temperature and pressure increase near the bubble making the local area around it a unique reactor allowing to implement various reactions and processes. The preliminary results of these studies were partially summarized in monographs [15–17]. In [14, 15], the boundary-value problem of conjugation for a vapor-gas bubble in a liquid was considered: detailed calculations allowed to identify all the features of bubble development from an embryo until its collapse at a solid wall. Based on this problem, these papers also consider a specific case of a spherical bubble in a stationary liquid. The results obtained by various authors indicate that symmetric collapse of the bubble triggers high-intensity pressure (up to 5–10 thousand atm.) and temperature (up to 15,000 °C) fields.

This phenomenological model allowed the design development of some reactors, agitators, mixers and other equipment based on super-cavitation (SC) effects — SC devices designed to treat water and other liquid media [36]. Designs of these devices have been constantly improved based on numerical and physical experiments [36–44]. The results obtained enabled well-targeted specification of the design parameters for the SC-devices during their design development.

## Experimental research technique

The experimental study was focused on stainless steel samples from pipe fragmentation: the pipe was used during the operation of a radiochemical plant for 40 years. The fragment size was as follows:  $\varnothing 50 \times 3$ ,  $L = 100$  mm [45]. Radionuclide contamination of the samples was caused by  $^{137}\text{Cs}$ .

Radionuclide composition of the presented samples was measured using Inspector-1000 spectrometer with a NaI(Tl) scintillation detector. The spectra were processed using Genie-2000 software. At pre-decontamination stage, gamma radiation from the sample and surface beta radioactive contamination was measured using MKS-AT1117M dosimeter-radiometer with BDPB-01 and BDKG-03 detecting units. Measurement conditions (environmental parameters, external background of gamma radiation) corresponded to technical characteristics of the measuring instrument specified in the operational documentation:  $t = 24.7$  °C,  $\psi = 61$  %,  $P = 749$  mm Hg. The microclimate parameters were measured with IVTM-7M thermohygrometer. The external gamma background amounted to  $H_{\gamma} = 0.27$   $\mu\text{Sv/h}$ . Direct measurement method was used to measure gamma radiation from the samples. Measurement of the dose rate from the samples was done assuming automatic background subtraction.

Indirect method was used to assess surface radioactive beta contamination. Removable surface contamination was determined by a smear method. To implement this method, sheets of filter  $10 \times 10$  cm paper have been prepared with 2–3 % of these tested for purity. The dry-smear sampling procedure involved contaminated area wiping. In this case the sample was wiped with filter paper: the paper was pressed against the surface with a force of 0.5–1 kg and passed over the controlled area. This operation was repeated three times. Then, the flux density of beta particles from the filter paper samples was measured.

To measure the flux density of beta particles, a BDPB-01 detection unit was used: it was installed above the measurement point so that the surface of the sensitive detection unit volume could be located at a distance of 5–15 mm from the studied surface. At each point, the measurements were repeated at least five times. The measurement of beta particle flux density was implemented providing background subtraction.

The following technique was used in the experimental study of sample decontamination (a total of 30 samples have been decontaminated):

- two samples were decontaminated with plain water;

- 4 samples were decontaminated with an alkaline solution containing water — 99.3%, NaOH — 0.5%,  $\text{KMnO}_4$  — 0.2%;
- 4 samples were decontaminated with volgonate solution, which is a detergent consisting of water — 99.3%, sulfonol — 0.5%,  $\text{H}_2\text{C}_2\text{O}_4$  — 0.2%;
- 20 samples were decontaminated with water that has undergone cavitation treatment.

To obtain cavitation-activated water, rotary-type supercavitation reactors (SC-reactors/mixers) were used [14]. The effect of hydrodynamic cavitation on water results in an increased concentration of molecular oxygen, which may be produced via chemical processes triggered by cavitation associated with the decomposition of  $\text{H}_2\text{O}$  molecule into radicals in the cavitation cavity. Mechanolysis of water in this case includes reactions, the products of which are molecular oxygen and hydrogen, hydroxyl groups and hydrogen peroxide [14]. Along with radiation and dissipation of excess energy into heat, an activated water molecule can dissociate. At the same time, the structure of water is changing along with the formation of free hydrogen bonds resulting in its increased activity and reactivity.

### Processing the results

To evaluate decontamination effectiveness, decontamination factor, required and reduced decontamination factors were calculated. To calculate the decontamination factor, the following ratio was used:

$$K_d = \frac{F_n}{F_k}, \quad (1)$$

where  $F_n$  is the pre-decontamination particle flux density;  $F_k$  stands for the post-decontamination particle flux density.

Relation (1) cannot allow a judgment regarding the fact whether the decontamination goal has been achieved or not, since given the same decontamination factor, the residual amount of contamination can be higher than the levels stipulated in the radiation safety standards. In order to take this circumstance into account, introduced was the concept of required decontamination factor  $K_{req}$ . In case of radioactively contaminated surfaces belonging to various items, this factor can be calculated as follows:

$$K_{req} = \frac{F_n}{F_{al}}, \quad (2)$$

where  $F_{al}$  stands for the allowable contamination level.

To ensure that the required decontamination factor reflects the efficiency and the completeness of

processing to the fullest extent possible, the concept of reduced decontamination factor  $RK_d$  was introduced in the following form:

$$RK_d = \frac{\lg K_d}{\lg K_{req}} = \frac{\lg F_n - \lg F_k}{\lg F_n - \lg F_{al}}. \quad (3)$$

Based on formula (3) one can conclude that the  $RK_d$  takes into account the achieved decontamination efficiency ( $K_d$ ) and the required one ( $K_{req}$ ). If during decontamination the goal is achieved and the ultimate contamination of the item appears to be equal to the allowable level, i. e.,  $F_k = F_{al}$ , then  $RK_d = 1$ . If  $F_k = F_n$ , i. e., deactivation did not occur,  $RK_d = 0$ . Therefore, decontamination appears to be effective if  $RK_d \geq 1$ . Thus, the quality of decontamination operations performed in different ways and under different conditions can be compared according to the  $RK_d$  value. To calculate the  $RK_d$ , allowable level of radioactive contamination of 2,000 part/cm<sup>2</sup>·min was set for the surfaces inside the premises, where the personnel was assumed to be permanently present, and the equipment located inside [46].

### Experimental results and their discussion

The implemented experiment can be conditionally divided into three series. The first series consisted of sample No. 1 and No. 2 decontamination with ordinary water. The samples were immersed into containers with water. The contact time amounted to 20 minutes. After that, the samples were removed from the containers and wiped off with a rag. The exposure time amounted to 2 minutes. After that, relevant radiation parameters were re-measured and the required and reduced decontamination factors were calculated.

The second series involved the decontamination of samples No. 3–10 in containers using the chemicals listed above. Samples No. 3–6 were deactivated with an alkaline solution of the following composition: water — 99.3%, NaOH — 0.5%,  $\text{KMnO}_4$  — 0.2%. Samples 7–10 were deactivated with a solution of volgonate, which consisted of water — 99.3%, sulfonol — 0.5%,  $\text{H}_2\text{C}_2\text{O}_4$  — 0.2%. The processing algorithm for the samples was similar to the one described above.

The third series was focused on the decontamination of samples No. 11–30 with water that has undergone cavitation treatment. The primary task of the experiment was to identify the optimal immersion time of the samples in the cavitation-activated water. For these purposes, samples No. 11–20 were immersed into containers with cavitation-activated water. Every 5 minutes, the samples were removed and wiped off with a rag. Then the surface

radioactive contamination was measured and the decontamination factor was calculated. After that, taking into account the optimal processing time identified for the samples, 10 more samples were deactivated, namely, No. 21–30.

Table 1 presents the decontamination factors, the required decontamination factors, the reduced decontamination factors calculated for samples No. 1–10.

**Table 1. Calculated decontamination factors for samples No. 1-10**

| Number of the sample | $F_n$ , part/cm <sup>2</sup> ·min | $F_k$ , part/cm <sup>2</sup> ·min | $K_d$      | $K_{req}$ | $RK_d$ |
|----------------------|-----------------------------------|-----------------------------------|------------|-----------|--------|
| Water                |                                   |                                   |            |           |        |
| 1                    | 10,600 ± 2,141                    | 6,200 ± 1,308                     | 1.7 ± 0.5  | 5.3       | 0.3    |
| 2                    | 22,300 ± 4,950                    | 14,800 ± 3,034                    | 1.5 ± 0.4  | 11.2      | 0.2    |
| Alkaline solution    |                                   |                                   |            |           |        |
| 3                    | 5,600 ± 1,128                     | 1,030 ± 216                       | 5.4 ± 1.6  | 2.8       | 1.6    |
| 4                    | 35,000 ± 7,280                    | 1,400 ± 283                       | 25 ± 7.2   | 17.5      | 1.1    |
| 5                    | 13,000 ± 2,746                    | 2,000 ± 445                       | 6.5 ± 2    | 6.5       | 1      |
| 6                    | 27,000 ± 5,751                    | 906 ± 201                         | 29.8 ± 9.1 | 13.5      | 1.3    |
| Wolgonate            |                                   |                                   |            |           |        |
| 7                    | 20,000 ± 4,463                    | 890 ± 202                         | 22.5 ± 7.2 | 10        | 1.4    |
| 8                    | 38,000 ± 7,853                    | 1,600 ± 405                       | 23.8 ± 7.8 | 19        | 1.1    |
| 9                    | 14,000 ± 3,416                    | 1,200 ± 368                       | 11.7 ± 4.6 | 7         | 1.3    |
| 10                   | 5,200 ± 1,087                     | 370 ± 79                          | 14.1 ± 4.2 | 2.6       | ·      |

It can be seen that after decontamination, the flux density of beta particles for samples 3–10 immersed into decontamination solutions with chemical reagents was found to be lower than the limit established in [46]. Decontamination of samples No. 1 and 2 has proved to be ineffective, since the flux densities identified for the beta particles exceeded the established limits. The decontamination efficiency started to decrease after 20 minutes of sample contact with cavitation-activated water which was due to the transfer of contamination from the decontamination solution back to the contaminated surface. After the optimal treatment time was identified for the samples No. 11–20 treated with cavitation-activated water (Figure 1), 10 more samples, namely, No. 21–30 were decontaminated. To evaluate the effectiveness of the decontamination performed, radiation parameters of the studied samples were re-measured. Table 2 summarizes the calculated decontamination factors for samples No. 11–30.

It can be seen that the decontamination factor for samples decontaminated with water that has undergone cavitation treatment ranges from 2 to

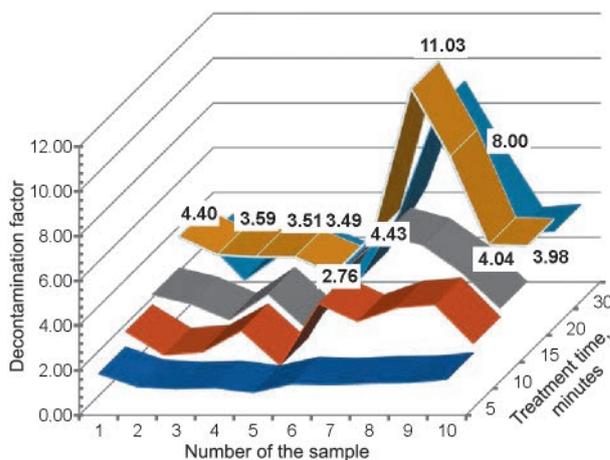


Figure 1. Calculated optimal processing time

**Table 2. Calculated decontamination factors for samples No. 11-30**

| Number of the sample       | $F_n$ , part/cm <sup>2</sup> ·min | $F_k$ , part/cm <sup>2</sup> ·min | $K_d$      | $K_{req}$ | $RK_d$ |
|----------------------------|-----------------------------------|-----------------------------------|------------|-----------|--------|
| Cavitation-activated water |                                   |                                   |            |           |        |
| 11                         | 4,500 ± 1,198                     | 1,020 ± 245                       | 4.4 ± 1.5  | 2.3       | 1.8    |
| 12                         | 2,600 ± 590                       | 722 ± 143                         | 3.6 ± 1.1  | 1.3       | 4.8    |
| 13                         | 6,900 ± 1,938                     | 1,980 ± 470                       | 3.5 ± 1.3  | 3.45      | 1      |
| 14                         | 2,900 ± 661                       | 830 ± 170                         | 3.49 ± 1.1 | 1.5       | 3.0    |
| 15                         | 5,900 ± 1,222                     | 2,100 ± 463                       | 2.8 ± 0.8  | 2.9       | 1      |
| 16                         | 4,900 ± 1,022                     | 1,120 ± 282                       | 4.4 ± 1.5  | 2.5       | 1.6    |
| 17                         | 69,100 ± 14,190                   | 6,300 ± 1,380                     | 11 ± 2.9   | 34.6      | 0.7    |
| 18                         | 17,200 ± 3,486                    | 2,100 ± 510                       | 8.2 ± 2.6  | 8.6       | 1      |
| 19                         | 9,100 ± 1,842                     | 2,200 ± 555                       | 4.1 ± 1.2  | 4.6       | 0.9    |
| 20                         | 10,200 ± 2,056                    | 2,500 ± 1,039                     | 4 ± 0.6    | 5.1       | 0.7    |
| 21                         | 7,600 ± 1,563                     | 2,000 ± 470                       | 3.8 ± 1.2  | 3.8       | 1      |
| 22                         | 39,000 ± 8,307                    | 2,600 ± 556                       | 15 ± 4.5   | 19.5      | 0.9    |
| 23                         | 8,600 ± 1,734                     | 4,200 ± 876                       | 2.1 ± 0.6  | 4.3       | 0.5    |
| 24                         | 5,100 ± 1,076                     | 1,300 ± 364                       | 3.9 ± 1.4  | 2.5       | 1.5    |
| 25                         | 38,000 ± 7,852                    | 2,400 ± 520                       | 15.8 ± 4.7 | 19        | 0.9    |
| 26                         | 30,100 ± 6,026                    | 5,300 ± 1,076                     | 5.6 ± 1.6  | 15.1      | 0.6    |
| 27                         | 7,900 ± 1,647                     | 1,250 ± 373                       | 6.3 ± 2.3  | 3.9       | 1.4    |
| 28                         | 4,100 ± 896                       | 1,400 ± 342                       | 2.9 ± 0.9  | 2.1       | 1.4    |
| 29                         | 5,200 ± 1,080                     | 1,100 ± 280                       | 4.7 ± 1.5  | 2.6       | 1.6    |
| 30                         | 5,000 ± 1,041                     | 1,200 ± 442                       | 4.2 ± 1.8  | 2.5       | 1.6    |

15.8, which is higher than the decontamination factor calculated in case of ordinary water application (according to literature sources, the maximum value amounts to 2.5; whereas values ranging from 1.5 to 1.7 have been obtained experimentally) and is comparable with the decontamination factors

calculated suggesting the application of surface treatment decontamination solutions based on chemical reagents.

Analysis of reduced decontamination factors evidences that:

1. Decontamination with alkaline and wolgonate solutions has appeared to be effective for all samples —  $PK_d > 1$ ;

2. Decontamination of samples with water has proved to be ineffective —  $PK_d < 1$ ;

3. For 13 samples out of 20 deactivated by cavitation-activated water, deactivation has proved to be effective —  $PK_d > 1$  with the  $PK_d$  values ranging from 1 to 4.8. For three samples, the  $PK_d$  values were found to be equal to 0.9.

Figure 2 presents a model of an automated system proposed for the decontamination of metal RW.

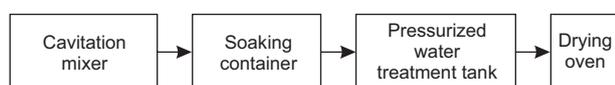


Figure 2. Diagram of a cavitation-based decontamination system

The cavitation-activated water is supplied to the treatment tank from a cavitation mixer. The container with the decontaminated equipment is installed by a crane installation into the first soaking container. After submerged decontamination in the first tank, the container is transferred to a pressurized water treatment tank. When the processing is completed, the container is transferred to a drying oven which is done using a girder crane with an electric hoist.

The contaminated cavitation-activated water can be recycled in other processes allowing to reduce the volume of secondary LRW [2, 5, 18, 19, 47, 48].

### Conclusion

The following conclusions can be drawn from the implemented experiment:

1. Relationship between the decontamination efficiency and the contaminated surface treatment time with a decontamination solution based on cavitation-activated water has been identified. The optimal sample treatment time was found to be equal to 20 minutes.

2. Decontamination of samples with water that has undergone cavitation treatment has appeared to be more effective than the one involving ordinary water and comparable to the results of decontamination with an alkaline solution.

3. Contaminated cavitation-activated water recycling will reduce the amount of generated liquid radioactive waste.

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