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RESEARCH AND DEVELOPMENT OF RESOURCE-SAVING TECHNOLOGIES FOR RECYCLING OF TECHNOGENIC WASTE

ТЕХНОГЕНДІК ҚАЛДЫҚТАРДЫ ҚАЙТА ӨҢДЕУДІҢ РЕСУРСТЫ ҮНЕМДЕЙТІН ТЕХНОЛОГИЯЛАРЫН ЗЕРТТЕУ ЖӘНЕ ӘЗІРЛЕУ

ИССЛЕДОВАНИЯ И РАЗРАБОТКА РЕСУРСОСБЕРЕГАЮЩИХ ТЕХНОЛОГИЙ РЕЦИКЛИНГА ТЕХНОГЕННЫХ ОТХОДОВ

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Decontamination, ultrasound, radioactivity, uranium, waste, metal, filter cloths, recycling.

ABSTRACT
The process of decommissioning uranium production equipment and technological tools leads to the formation and accumulation of metallic radioactive waste. The majority of this waste falls into the category of low-level radioactive waste and requires special conditions for storage. The use of ultrasonic treatment of TRW contributes to an increase in decontamination coefficients compared to traditional methods, while reducing the duration of the process. It has been established that the types of filter fabrics considered are subject to regeneration with the possibility of their subsequent use in industrial conditions. The effect of various solutions on the efficiency of the decontamination process of metal TROs has been studied. The possibility of single-stage cleaning of metal TROs in an ultrasonic field using HNO3 solution to obtain commercial metal with surface contamination of less than 1 alpha particle/cm2·min at an acceptable level of less than 1.2 alpha particles/cm2·min has been demonstrated.

Түйінді сөздер:
Дезактивация, ультрадыбыс, радиоактивтілік, уран, қалдықтар, металл, сүзгі маталары, қайта өңдеу.

ТҮЙІНДЕМЕ
Уран өндіруде қолданылатын жабдықтар мен технологиялық қондырғыларды пайдаланудан шығару процесі металлдық радиоактивті қалдықтардың түзілуіне және жиналуына әкеледі. Бұл қалдықтардың басым бөлігі төмен деңгейлі радиоактивті қалдықтарға жатады және сақтау үшін арнайы жағдайларды талап етеді. ТРҚ-ны ультрадыбыстық өңдеуді қолдану дәстүрлі әдістермен салыстырғанда дезактивация коэффициенттерінің артуына және процесс уақытының қысқаруына ықпал етеді. Зерттеу нәтижелері бойынша қарастырылған сүзгі маталарының түрлері қайта өңдеуге жатады және оларды өнеркәсіптік жағдайда қайта пайдалануға болады. Металл ТРО-ларды дезактивациялау процесінің тиімділігіне әртүрлі ерітінділердің әсері зерттелді. HNO_3 ерітіндісін

қолдана отырып, ультрадыбыстық өрісте металл ТРО-ларды бір сатылы тазалау арқылы бетіндегі ластану деңгейі 1 альфа бөлшек/см^2·мин-нен аз, қабылдалатын 1,2 альфа бөлшек/см^2·мин-ден төмен коммерциялық металл алу мүмкіндігі көрсетілді.

Ключевые слова:
Дезактивация, ультразвук, радиоактивность, уран, отходы, металл, фильтровальные полотна, рециклинг.

АННОТАЦИЯ
Процесс вывода из эксплуатации оборудования и технологической оснастки уранового производства приводит к образованию и накоплению металлических ТРО. Основная доля которых, относится к категории низкоактивных РАО и требует обеспечения особых условий. Применение ультразвуковой обработки ТРО способствует увеличению коэффициентов дезактивации по сравнению с традиционными методами, при этом продолжительность процесса сокращается. Установлено, что рассмотренные типы фильтровальных тканей подлежат регенерации с возможностью последующей их эксплуатации в промышленных условиях. Изучено влияние различных растворов на эффективность процесса дезактивации металлических ТРО. Показана возможность одностадийной очистки металлических ТРО в УЗ поле с использованием раствора HNO_3 для получения товарного металла с поверхностным загрязнением менее 1 альфа-частицы/см^2·мин при допустимом уровне менее 1,2 альфа-частицы/см^2·мин.

INTRODUCTION

An analysis of the energy sector indicates that nuclear power offers significant advantages over conventional fossil fuel–based energy systems in terms of fuel efficiency, pollutant emissions, and waste generation (Fraenkl et al., 2022; Fränkle et al., 2023; Grković & Doder, 2022; Gurau D. et al., 2023). At the same time, the key factor governing the advancement and optimization of all stages of nuclear energy development is the environmental impact associated with the operation of nuclear fuel cycle (NFC) facilities. The environmental effects of NFC enterprises are diverse and include land allocation for infrastructure development, large-scale withdrawal of freshwater for industrial use, discharges of non-radioactive toxic substances, and long-term storage of radioactive liquid and solid wastes. Consequently, NFC enterprises place particular emphasis on improving methods for radioactive waste management and processing. This approach contributes to the achievement of global sustainable development goals and, in turn, enhances the competitiveness of nuclear energy relative to other energy sources.

Currently, Kazakhstan ranks among the world's leading uranium producers; therefore, the issue of radioactive waste (RW) conversion remains highly relevant. An analysis of the environmental impacts of NFC enterprises has shown that the most challenging aspect is the safe decommissioning of radioactive materials, particularly with respect to their processing and compliance with regulatory limits for radioactive contamination. In addition, the relevance of this issue is driven by several factors:

- the substantial volume of accumulated and newly generated radioactive waste, which raises concerns among the public and regulatory authorities due to the potential radiation hazards associated with RW storage facilities;
- the need to improve existing radioactive waste management practices in response to stricter regulatory requirements and increasingly stringent environmental standards;
- the growing influence of society on decision-making related to environmental aspects of activities carried out by nuclear industry organizations.

The annual accumulation of waste generated by NFC enterprises amounts to tens of thousands of kilograms, while the concentration of valuable components in such waste often exceeds that found in primary ore materials. Accordingly, the strategic objective is the maximum recycling of technogenic raw materials, which enables the following outcomes:

- increased efficiency in the use of non-renewable natural resources and conservation of depleting mineral reserves;
- enhanced labor productivity through economically viable processing of materials withdrawn from technological production cycles;
- reduction or elimination of sources of environmental contamination;
- development of low-waste, low-operation, and energy-efficient technological processes.

Radioactive contamination of metallic material surfaces is primarily governed by intermolecular interactions, chemical bonding, and various sorption mechanisms, including physical adsorption, cation exchange, chemisorption, and absorption. In general, the extent of contamination depends on the physicochemical properties of the material undergoing decontamination, the condition of its surface, and the nature of the contaminants (Hagedorn et al., 2022).

At present, decontamination methodologies employ a broad range of techniques for radionuclide removal from surfaces. In addition to simple methods, such as surface wiping or mechanical brushing, more advanced technologies requiring specialized equipment are widely applied. In practice, combined physicochemical and mechanical approaches are most commonly used (Liu et al., 2022; Nakahara et al., 2024). Alternative decontamination methods include electropolishing, ultrasonic cleaning, and metal melting.

Ultrasonic (US) cleaning is based on the conversion of high-frequency acoustic energy into mechanical vibrations. From a physical perspective, the fundamental principles governing sound wave propagation are identical across frequency ranges; however, ultrasonic waves exhibit distinct characteristics due to their high frequencies and correspondingly short wavelengths. The primary nonlinear phenomenon occurring in an ultrasonic field is cavitation, during which pulsating bubbles form in a liquid, containing vapor, gaseous products, or their mixtures (Zhang et al., 2025; Zupanc et al., 2023). Depending on the type and characteristics of contamination, cavitation manifests through micro-impact effects, microstreaming, and localized temperature increases.

In technologies for processing uranium-containing compounds, specific types of filter fabrics are employed at various stages (Dubrovskiy & Zenkov., 2024; Kulagina & Shelenkova., 2017). These materials differ in both composition and operating principles and include cotton-based and synthetic fabrics.

During operation, the hydraulic resistance of filter fabrics progressively increases, resulting in a decrease in filtration rate. To maintain the required permeability within specified limits, filter elements are periodically replaced or regenerated (Kuznetsova., & Sakhnenko, 2022; Savkin et al., 2007). Regeneration allows for partial or complete removal of deposited sediments and restoration of the original filtration properties.

Previous studies have reported the decontamination of glass-fiber filters, which serve as alternatives to Petryanov filters used for the removal of technological gaseous wastes generated during the process cycle of FSUE "Mayak Production Association". The proposed decontamination approach involves ultrasonic treatment of glass-fiber filters in nitric acid solutions at elevated temperatures. This method enables the removal of up to 98% of radionuclides from the treated material and provides near-complete recycling of valuable components (Federal Service for Surveillance on Consumer Rights Protection and Human Wellbeing, 2013).

Optimization of acoustic field parameters, physicochemical properties of the cleaning solution, gas content, and external conditions such as pressure and temperature allows effective control of the contamination removal process across a wide range of materials and contamination types (Ministry of Health of the Republic of Kazakhstan, 2020). An additional intensifying factor in refining processes is the acoustic capillary effect, which facilitates penetration of the decontaminating agent into microcracks and pores. Moreover, the mechanical effects of ultrasound significantly enhance dissolution processes and particle detachment from treated surfaces.

The primary objective of this study was to evaluate the efficiency of ultrasonic cleaning for the decontamination of various radioactive materials used in the processing of uranium-containing compounds at Ulba Metallurgical Plant JSC, to identify optimal treatment media, and to assess the feasibility of material recycling after decontamination.

MATERIALS AND METHODS

When deciding on the necessity of decontamination of various materials, the feasibility of the procedure is assessed by substantiating its benefits in relation to the associated costs, as well as by analyzing the amount of secondary waste generated and available disposal methods. At the same time, the level of residual contamination of the materials after decontamination must ensure the safety of personnel and the environment. In accordance with the requirements of Clause 13 of the Sanitary Rules for Ensuring Radiation Safety (SP SETORB) (Grković & Doder, 2022), the following conditions must be met when releasing a batch of scrap metal:

1. the ambient equivalent dose rate (gamma radiation) from the surface of the scrap does not exceed 0.2 μSv/h above the natural background radiation level;
2. the alpha radiation surface contamination density does not exceed 0.04 Bq/cm^2;
3. the beta radiation surface contamination density does not exceed 0.4 Bq/cm^2.

In conducting the present study, the regulatory framework of the Russian Federation was applied. According to document (Liu et al., 2022), a beta surface contamination level of 0.4 Bq/cm^2 corresponds to 12 particles/(cm^2·min), while an alpha contamination level of 0.04 Bq/cm^2 corresponds to 1.2 particles/(cm^2·min).

The objects selected for decontamination in this study included:

- filter fabrics of various grades used in the processing of natural and enriched uranium;
- scrap metal and structural components manufactured from different steel grades.

Two conceptual approaches were considered during the studies conducted at Ulba Metallurgical Plant JSC:

- "deep" decontamination of filter fabrics aimed at reducing the residual radionuclide content, followed by disposal of the treated materials;
- regeneration of filter fabrics for their subsequent reuse in the technological process.

The objects of investigation comprised two types of filter fabrics used in the filtration stages of uranium-containing solution processing: polypropylene fabric (art. 1002k) and cotton–polyester fabric (art. 2411). Samples of decommissioned filter fabrics collected from several production areas were used to develop decontamination regimes. During the experiments, optimal decontamination conditions for filter fabrics were determined, with particular emphasis on the effects of washing solution composition and ultrasonic treatment duration.

The experimental studies were carried out using the following equipment: an ultrasonic decontamination unit, model MO-646 (manufactured by Alexander Plus, Russia), and a laboratory ultrasonic bath Laborette-17 (FRITSCH).

RESULTS AND DISCUSSION

Figure 1 presents fragments illustrating the fiber structure of the filter fabrics before and after the washing process.

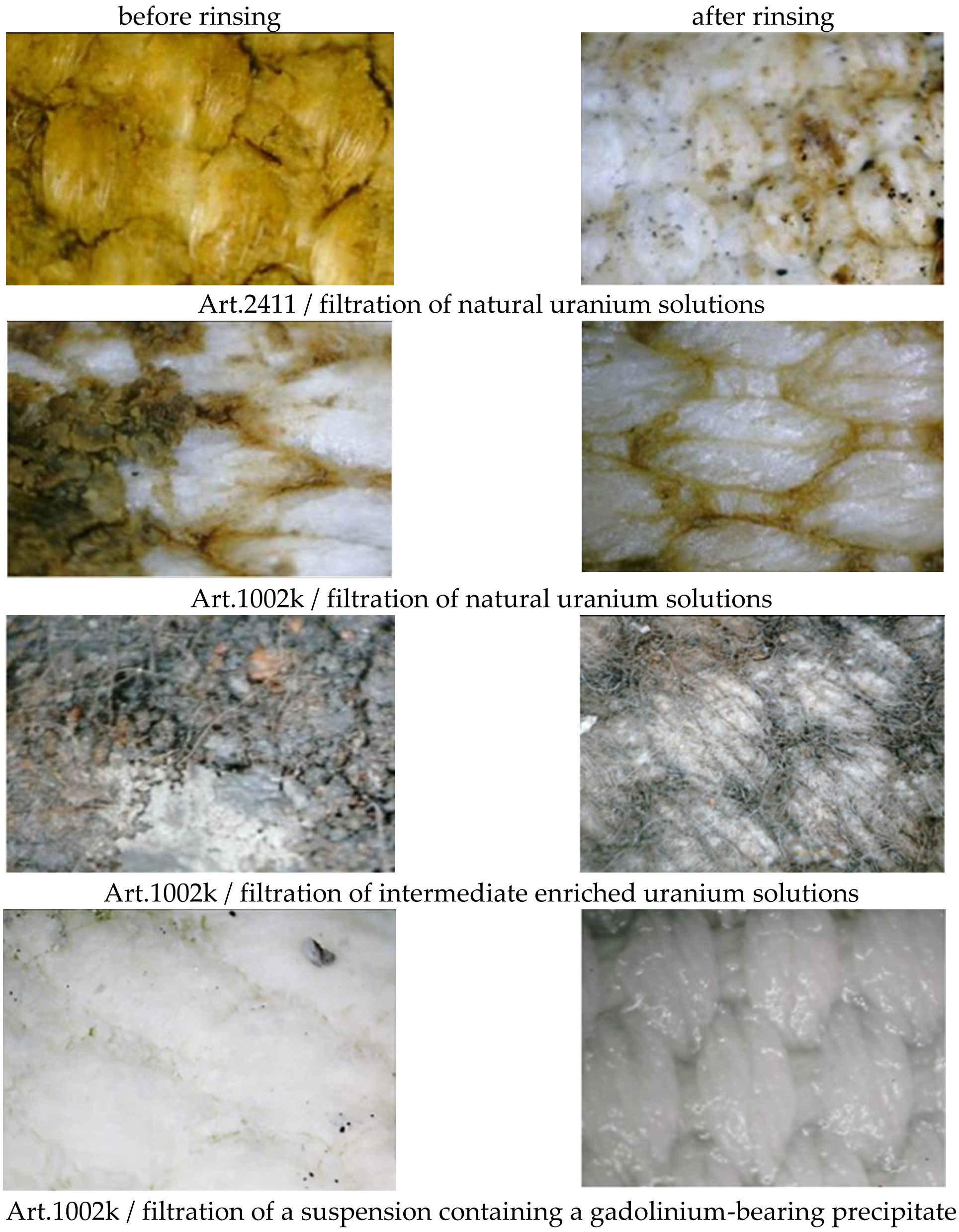


Figure 1. Fragments of filter fabrics from different technological operations before and after decontamination

Note – prepared by the authors

Based on the results of the conducted experiments, optimal decontamination regimes for various filter fabrics were determined. It was established that, for a given duration of ultrasonic treatment and under media of specified composition, the uranium content in filter fabrics can be reduced by a factor of 7 to 70, depending on the initial degree of contamination, achieving uranium concentrations in the material below 0.3 wt.%.

In studies aimed at assessing the feasibility of reusing regenerated filter fabrics, material samples treated by different methods under optimal regimes were employed. The quality criteria for regeneration included water permeability and the content of suspended solids in the filtrates.

Water permeability was evaluated based on the filtration rate of uranium-containing suspensions through the above-mentioned types of filter fabrics. Filtration quality (i.e., the efficiency of dispersed phase separation) using regenerated filter fabrics was assessed by measuring the optical density of the filtrates with a KFK-2MP photocolorimeter.

The obtained results indicate that regeneration efficiency is largely determined by the physicochemical composition of the deposits forming a layer of solid particles on the surface of the filter fabric and/or retained within the pores of the material during filtration of suspensions from different technological units.

Experimental studies demonstrated that all investigated types of filter fabrics, when treated under the most effective processing conditions, can be successfully regenerated and subsequently reused under industrial operating conditions.

Depending on the type of filter fabric and the technological unit in which it is applied, the mass concentration of dissolved uranium in the washing solutions, which represent a suspension, may vary from 0.2 to 1.8 g/L. Processing of solutions generated during filter fabric regeneration should be carried out by sedimentation of the solid phase followed by decantation of the liquid. The resulting sediment is expediently directed to the appropriate processing unit.

When considering radioactive metal waste, it should be noted that the suitability of metals for repeated reuse in the manufacture of new products makes them an important industrial raw material. The absence of effective technologies for the processing and disposal of radioactive metal waste accumulated during the hydrometallurgical processing of uranium-containing raw materials leads to a continuous increase in their volume. An additional challenge arises from the need to perform repair operations on process equipment (e.g., pumps) during industrial operation, which is impossible without prior removal of radioactive contamination from equipment surfaces. Furthermore, certain equipment components and auxiliary fixtures are periodically decommissioned and/or dismantled, resulting in the generation of significant amounts of metal waste, a substantial portion of which contains residual radioactive contamination. To ensure safe disposal of such materials and equipment after dismantling, decontamination is required.

In the experiments, carbon steel samples were treated with various solutions in an ultrasonic field. The compositions of the decontaminating solutions were as follows:

- industrial water;
- nitric acid solution with a concentration of 100 g/L;
- foaming agent solution with a concentration of 5 g/L.

The duration of a single treatment stage was 15 minutes, with a solid-to-liquid mass ratio of T:L = 1:3. After the specified treatment time, a fresh portion of solution was introduced. Following each ultrasonic treatment stage, the mass and thickness of the samples were measured, and the degree of reduction in these parameters was calculated. The obtained results are presented in Table 1.

Table 1. Changes in the parameters of St3-grade steel samples depending on the duration of treatment and the composition of the solution

Processing time, min	Composition of the solution					
	H_2O	HNO_3	A/R	H_2O	HNO_3	A/R
	Mass reduction, %			Thickness reduction, %		
15	0,03	8.14	0.01	0.07	6.69	1.70
30	0.04	17.89	0.02	0.58	8.52	2.01
45	0.04	27.97	0.03	0.65	27.54	2.25
Note – prepared by the authors						

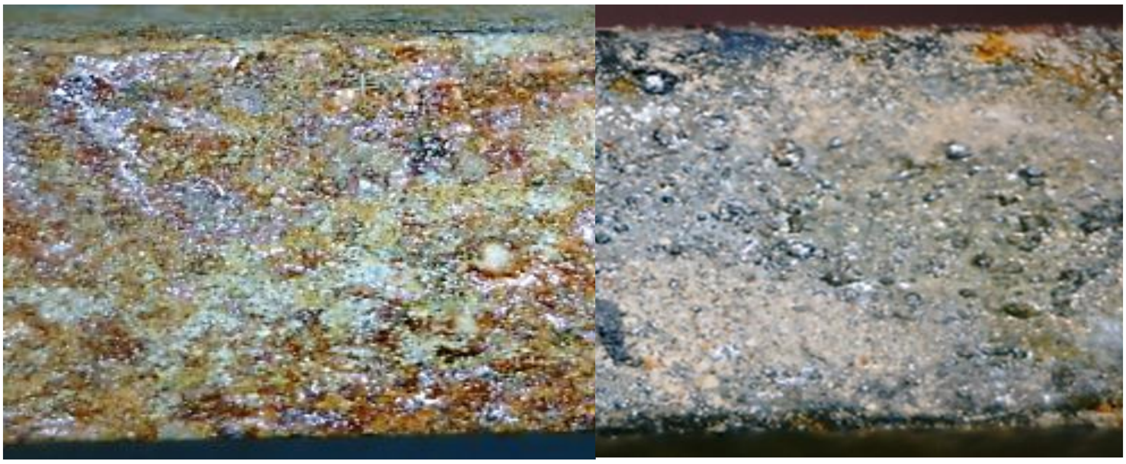
Analysis of the results presented in Table 1 showed that treatment of the samples in nitric acid resulted in the maximum mass loss, reaching up to 28 wt.% of the initial sample mass.

The uranium concentration in the working solutions after decontamination was as follows:

- aqueous solutions: less than 0.003 g/L;
- nitric acid solutions: from 0.128 to 0.410 g/L;
- in foaming agent solutions, uranium concentration was not measured.

The efficiency of the decontamination process after three washing cycles was evaluated based on the residual surface contamination of the samples with alpha-emitting radionuclides. The corresponding values of decontamination efficiency (Aeff) and the decontamination coefficient (Kd) were calculated.

Fragments of the surface of a St3-grade steel sample before and after ultrasonic decontamination are shown in Figure 2.



Initial state of the sample — After ultrasonic decontamination

Figure 2. Macrostructural analysis of St3-grade steel sample surface fragments before and after washing

Note – prepared by the authors

As can be seen from the presented images, the initial surface of the sample is characterized by pronounced heterogeneity, as well as the presence of corrosion deposits and surface contaminants. After decontamination, a noticeable reduction in surface contamination, smoothing of the microrelief, and a decrease in the number of inclusions are observed, indicating the effectiveness of the applied washing method. The results of calculations and measurements are presented in Table 2.

Table 2. Effect of solution composition on the decontamination efficiency of St3-grade steel samples

Medium	Alpha particle flux density, particles/(cm^2·min)	Aeff, %	Kd
No treatment	4.2	-	-
H_2O	1.8	57.1	2.3
HNO_3	1.0	76.2	4.2
Foaming agent	1.2	71.4	3.5
Note – prepared by the authors			

The results presented in Table 2 indicate that a nitric acid solution is the most effective aqueous agent for the decontamination of the examined sample. However, the significant mass losses of the treated material, reaching up to 28%, render decontamination in a nitric acid medium impractical. The use of a foaming agent solution under the specified processing conditions allows the stated objectives to be achieved.

CONCLUSIONS

It was established that the selection of the most effective regeneration regime for filter fabrics largely depends on the physicochemical composition of the solid phase removed from the solutions. The application of ultrasonic treatment during decontamination makes it possible to reduce the uranium content in filter fabrics by up to a factor of 70. The feasibility of reusing regenerated filter fabrics was investigated, and it was demonstrated that all examined types of filter fabrics are suitable for regeneration with the possibility of subsequent operation under industrial conditions.

The reuse of regenerated filter fabrics in technological processes can significantly extend their service life, reduce material consumption in the processing of uranium-containing compounds, and minimize the generation of radioactive waste. Overall, this contributes to the implementation of low-waste technologies at industrial facilities.

In the decontamination of radioactive metal waste, the influence of various solutions on the efficiency of the decontamination process was investigated. The possibility of single-stage ultrasonic cleaning of radioactive metal waste using an HNO_3 solution was demonstrated, enabling the production of marketable metal with surface contamination levels below the permissible limits.

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