

DECONTAMINATION OF SOIL CONTAMINATED BY URANIUM USING HYDROSEPARATION METHOD WITH FURTHER REAGENT TREATMENT OF FRACTIONS

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The article presents a decontamination technology used to remove uranium from radioactively contaminated soils at JSC AECC with the results of its development and testing being discussed as well. These soils, considered as VLLW, are expected to be generated during the decommissioning of sublimate production facility. The paper discusses testing stages of the decontamination technology with a laboratory installation in place applying hydroseparation method with further reagent treatment of the target fraction. Testing results will be used to develop an installation enabling to treat radioactively contaminated soils of VLLW category from JSC "AECC".

Keywords: *radioactively contaminated soil, radioactive waste, hydroseparation, reagent treatment, target fraction, fine fraction, sand fraction, laboratory installation, decontamination factor.*

Following a long-term operation, time has come for the decommissioning being planned for a number of nuclear and radiation hazardous facilities (hereinafter, nuclear facilities) at JSC AECC, as well as at other nuclear power sites. Normally such operations result in some territories and sites that need to be cleaned and remediated. For example, it has been estimated that the dismantlement of buildings and structures at JSC AECC will generate up to 100,000 m³ of secondary radioactive waste (soil) [1], including up to 30,000 m³ of soils being potentially contaminated with uranium and categorized as VLLW. Based on these estimates, JSC AECC considers possible options allowing to reduce the volume of radioactively contaminated soil through its deactivation using a most effective technology.

A similar problem was solved at JSC VNIINM: a soil decontamination installation based on hydroseparation method having a capacity of up to 1,000 kg/h was developed and tested there to reduce the volume of radioactively contaminated soils (RCS) [2–4]. JSC AECC conducted a number of tests to assess the effectiveness of the VNIINM's technology with respect to soil decontamination at sublimation production site (SP): 15 m³ of soil from AECC site contaminated with uranium was used in the testes [2]. These have demonstrated both the efficiency of the technology (RCS volume was reduced by 3.6 times) and its lower efficiency in addressing uranium contamination of the soil.

Compared with the data on the removal of cesium-137 from radioactively contaminated VNIINM

soils, the indicators were found to be lower in terms of decontamination and volume reduction coefficients due to the differences in physical and chemical radionuclide speciation in AECC soils.

This article discusses some solutions and results allowing to upgrade the soil decontamination technology.

Laboratory bench testing of VLLW soil decontamination technology by hydroseparation

Laboratory bench discussed in detail in [2] was used to determine the process parameters of AECC's VLLW soil decontamination by hydroseparation and to estimate material fraction not being cleared from radiation control following decontamination. The tests involved some 40 dm³ of soil being categorized as VLLW and contaminated by uranium-234, -235, -238 isotopes.

Hydroseparation method was used to separate a fraction of finely dispersed (0.1–0.7 mm) and coarse (0.7–3.0 mm) sand, gravel fractions (3–8 mm and more than 8 mm), plant origin fragments of different sizes, cake of finely dispersed fraction (up to 0.1 mm) and treated water (Figure 1).

The volume of the initial soil attributed to VLLW category with a total specific activity of uranium amounting to 15.8 Bq/g decreased as a result of its decontamination. The diagram shows that hydroseparation resulted in the separation of fractions corresponding to VLLW by their activity level (finely dispersed cake (47 Bq/g) and vegetation residues (83 Bq/g)) amounting to 30% of the original soil amount. Thus, VLLW volume reduction ratio amounted to 3.4.

Specific activity measurements for gravel (2.6–8.4 Bq/g) and sand (3.7–5.5 Bq/g) fractions indicate that these materials shall not be considered as radioactive waste, but their treatment should be restricted in keeping with relevant provisions of OSPORB-99/2010. Thus, overall VLLW soil decontamination coefficient appears to be equal to 3.6.

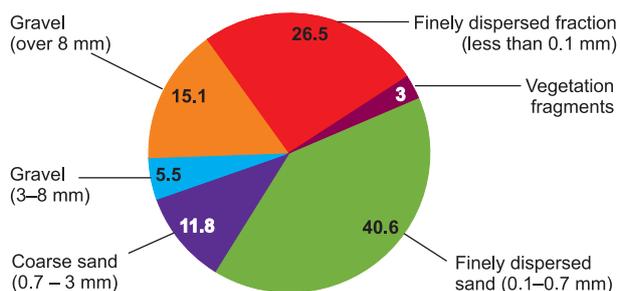


Figure 1. Breakdown of fractions segregated following VLLW soil decontamination

Fine sand fraction appears to be the largest accounting for over 40%. This fraction was assumed as the target one, subject to reagent treatment to achieve a higher coefficient of soil decontamination.

Based on preliminary tests of the decontamination process involving soils contaminated with uranium, a decision was made on introducing a reagent treatment stage to address the contamination of AECC's target soil fraction.

Investigating the physical and chemical speciation of radionuclides in soil fractions. Specifying reagent treatment modes for the target fraction

Radionuclide speciation in the soil accounts for a factor that was taken into account during the development of the reagent treatment process. It's worth noting that the uranium contaminant was distributed extremely unevenly in the studied soil.

Successive leaching studies have shown [5] that uranium in the soil fractions is available both in easily recoverable (exchange and carbonate forms, 50–70%) and in hardly recoverable forms (Fe/Mn oxides, organic and insoluble) in amounts being proportional to their specific activity (Figure 2). Moreover, uranium associated with the form considered as the most mobile out of the latter three (i. e. Fe/Mn oxides) makes up about 30% of the total uranium content in the soil sample.

A relationship is observed between uranium content in the fractions and their mineral and chemical composition. In particular, the target fraction shows a correlation between the iron content and the Fe/Mn oxide-uranium fraction. The low iron content corresponds to a low concentration of uranium being available in Fe/Mn oxide forms. Different calcite content explains the uneven distribution of uranium in carbonate-bound form. Spent reagent solutions show a correlation between uranium and humic compounds extracted from the soil.

According to X-ray absorption spectroscopy (XANES/EXAFS), in the studied soil samples,

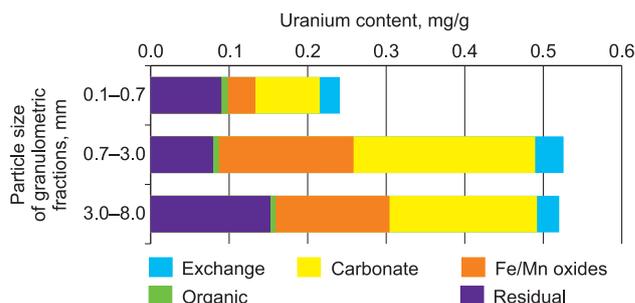


Figure 2. Breakdown of uranium speciation by soil fractions

uranium is available as hexavalent uranium in the form of UO_2^{2+} cation. It's a common knowledge that the hexavalent uranium is mobile, capable of forming complexes, in particular, with carbonate- and sulfate ions, various organic chelators. This suggests that uranium can be extracted from the soil by treating it with simple and cheap reagent solutions.

Comparative tests on reagent treatment of the target fraction with sodium carbonate, sulfuric and nitric acid solutions, EDTA and distilled water, enabled to identify the process modes for the reagent treatment stage: sequential mixing of the target fraction for 0.5–3 hours in sodium carbonate and sulfuric acid solution with a 20–50% (mass) solids content in the solutions.

Study of physical and chemical speciation of radionuclides [5] in the target soil fraction segregated by hydros separation method, testing of reagents applied to treat the soil fraction enabled the development of a reagent treatment stage and a laboratory test reagent treatment unit (Figure 3). The following requirements were considered during the development of the reagent treatment stage:

- plainness of hardware designs;
 - multiple reuse of reagent solutions;
 - minimum amount of secondary waste;
 - reagent safety for personnel and the environment;
 - low cost and minimum consumption of reagents.
- The reagent treatment unit featured the following components and equipment:
- reagent solution dispenser;

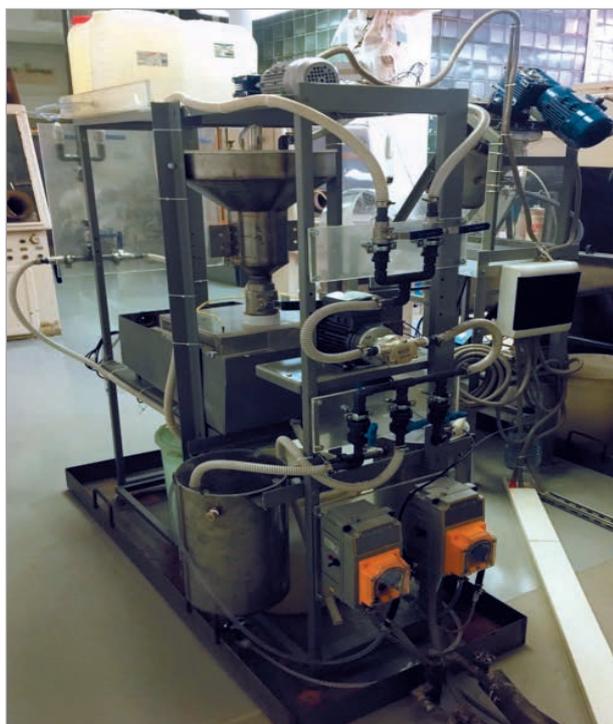


Figure 3. General view of the laboratory bench reagent treatment unit

- reactor with a stirrer for soil treatment with a reagent solution;
- tanks for reagent solution accumulation and their regeneration by coprecipitation;
- unit for target fraction separation and washing after reagent treatment;
- vacuum filtration unit for sludge dewatering after reagent solution regeneration.

Thus, the following operations were performed in the reagent treatment unit:

- mixing up the reagent solution;
- target fraction treatment by stirring;
- separation of the spent reagent solution;
- washing the fraction with water after treatment;
- precipitation of impurities from the spent solution with a calcium hydroxide suspension;
- mixing up new solutions based on wash water and neutralized solutions;
- sludge separation, thickening and dewatering.

Laboratory bench testing of reagent treatment decontamination method

Laboratory bench tests were performed to precise and adjust the process modes for the reagent treatment stage involving target soil fractions. A total of 20 target fraction samples, 2 kg each, were used to fine-tune the process modes via parameter variations:

- concentration of sodium carbonate in the solution ranging from 5.2 to 21 g/l;
- sulfuric acid concentration in solution ranging from 5 to 25 g/l;
- treatment time with sodium carbonate solution ranging from 0.5 to 3 hours;
- treatment time with sulfuric acid solution ranging from 0.5 to 3 hours;
- the number of "solution treatment – regeneration" cycles.

Figure 4 shows some milestones for the target fraction treatment by stirring sodium carbonate and sulfuric acid solutions (Figure 4a, c) and separation of the target fraction from the spent solution (Figure 4b, d).

Reagent treatment of the target fraction contaminated with uranium radionuclides resulted in the following compounds:

- precipitate cakes involving finely dispersed fraction separated following sodium carbonate solution, calcium sulfate precipitate formed due to sand interaction with sulfuric acid solution and a mixture of metal hydroxides and uranium compounds after precipitation under alkaline conditions;
- sand fraction purified from radionuclides and washed with water with a total specific activity of uranium of less than 300 Bq/kg;

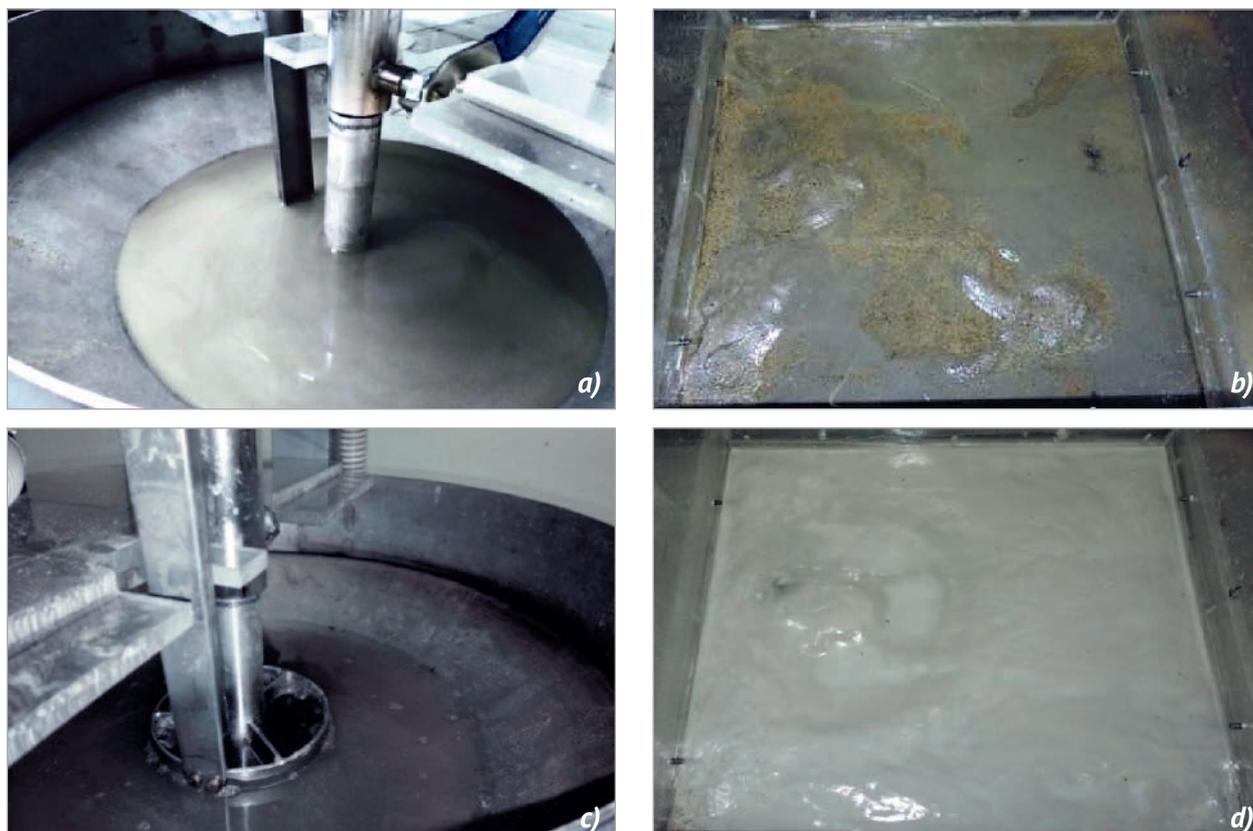


Figure 4. Target fraction treatment by stirring in sodium carbonate (a, b) and sulfuric acid (c, d) solutions

- spent neutralized saline solution (10–20 g/l) with a low specific activity of uranium with its amount being equal to the mass of the decontaminated sand fraction;
- washing water in an amount being equal to the mass of the purified sand fraction.

The following optimal parameters were identified based on a comparative analysis of target soil fraction decontamination coefficients considering different process modes with the resulting decrease in the total specific activity of uranium of less than 300 Bq/kg:

- consecutive 0.5-hour treatment of the sandy soil fraction involving a 50%-mass sodium carbonate solution with a concentration of 5.2 g/l and sulfuric acid with a concentration of 12.3 g/l;
- reagent solution regeneration method — coprecipitation of extracted uranium radionuclides with calcium hydroxide, separation of the precipitate formed and restoration of the reagent concentration in the solution (the number of regenerations is limited to four);
- reuse (up to four times) of reagent solutions after regeneration by calcium hydroxide precipitation.

VLLW soil decontamination by hydroseparation method involving a stage of the segregated target fraction reagent treatment yields the following benefits:

- 40% of clean soil with a decontamination coefficient of at least 90;
- 30% of soil considered as restricted-use materials with a decontamination coefficient of at least 3.6;
- 30% of the soil considered as VLLW with a volume reduction factor of 3.3.

The amount of secondary SRW available in the form of dehydrated sludge resulting from the reagent treatment of the target fraction accounts for some 3% of the initial soil mass.

It has been estimated that this option yields a more than 2-fold reduction in VLLW soil management cost after its decontamination compared to the untreated soil disposal option, taking into account relevant tariffs for RW transportation and disposal, operating and container costs.

Conclusion

The developed RCS decontamination method involving hydroseparation followed by reagent treatment of the separated fraction and the identified process parameters will be used in the development of a pilot-industrial decontamination facility being designed to reduce RCS volume at AECC site, as well as during decommissioning and remediation of similar sites.

The developed method for RCS decontamination from uranium radionuclides by hydroseparation with a reagent treatment stage can be applied to decontaminate VLLW RCS and to remove most part of RCS from the category of ILW.

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