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Article

IAEA-Assisted Treatment of Liquid Radioactive Waste at the Saakadze Site in Georgia

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Abstract: 50 m³ of legacy liquid radioactive waste at the Saakadze site in Georgia was treated using a modular type facility with apparatuses encased in three metallic 200 L drums using as purification method the sorption/ion exchange technology. The main contaminant of water in the underground tank was the long-lived radionuclide ²²⁶Ra. The casing of processing equipment enabled an effective conditioning of all secondary waste at the end of treatment campaign which resulted in the fully purified water stored on site for further reuse or discharge, and three 200 L metallic drums with cemented radioactive waste which are currently safely stored.

Keywords: treatment; liquid radioactive waste; sorption; ion exchange; purification; conditioning



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1. Introduction

Nuclear waste management has received considerable attention due to the important link between the safe management of radioactive waste and public acceptance of nuclear facilities and peaceful applications of nuclear energy. Radioactive waste management is typically divided into predisposal and disposal steps, where predisposal comprises all the steps in the management of radioactive waste from its generation up to disposal. The predisposal includes processing technologies that are primarily intended to produce a wasteform that is compatible with the selected or anticipated disposal option and complies with established waste acceptance criteria [1]. Liquid radioactive waste is highly mobile, and its storage is associated with hazards from potential leakages and contamination, so it cannot be considered passively safe. International disposal standards prescribe only solids as being acceptable forms for disposal facilities [2]. Consequently, liquid radioactive waste is typically processed to decrease the waste volume, solidifying the treated waste (typically a sludge) using durable wasteforms such as cements [3]. Treatment of aqueous waste separates it into two streams: a small volume fraction of concentrate containing the bulk of radionuclides and a large volume of water which has a sufficiently low level of contamination to permit discharge to the environment or recycling. Effective liquid treatment separates as much as of the radioactive contaminants as possible from the primary waste in the concentrated fraction, which typically requires conditioning prior to storage and disposal. A flow chart for managing aqueous radioactive waste is given in Figure 1.

The radioactive waste packages produced after treatment and conditioning contain the solidified waste and are then stored in a passively safe condition pending a disposal route becoming available.

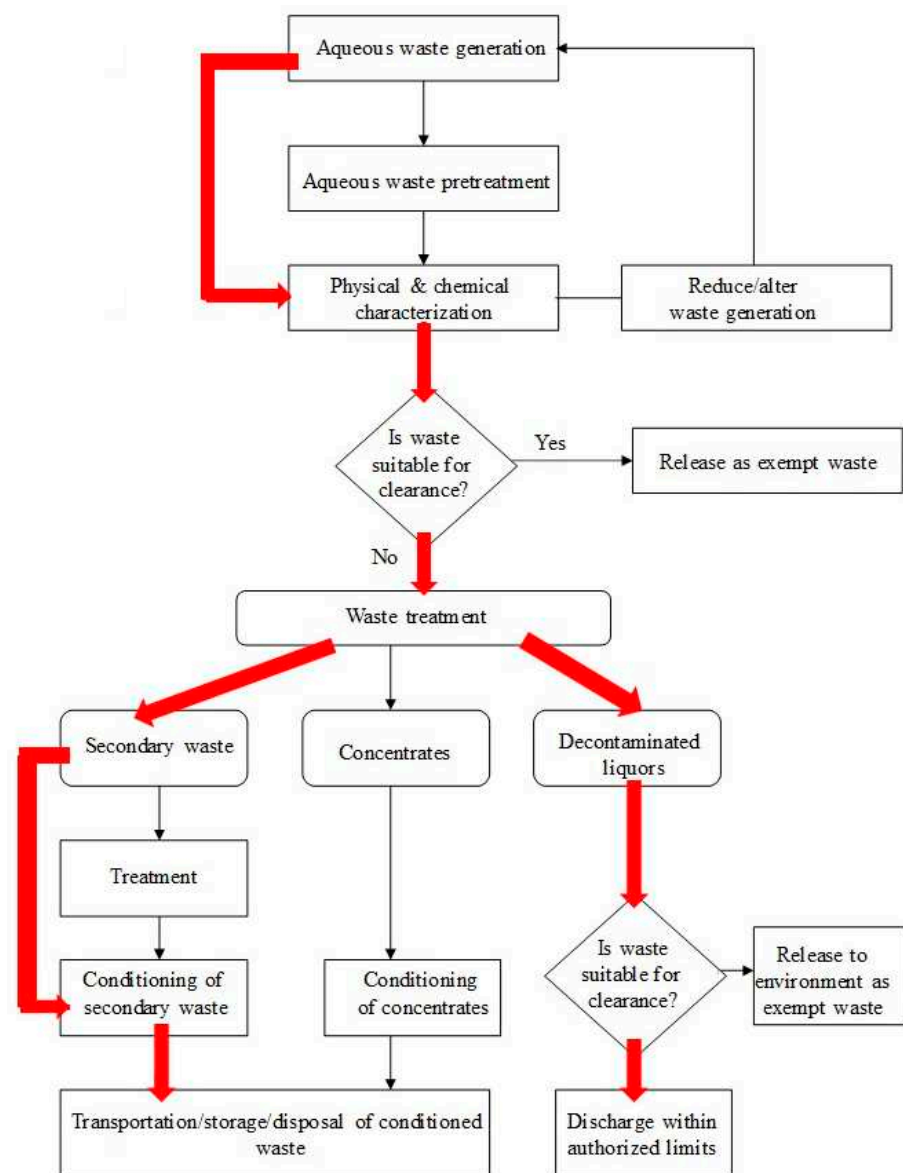


Figure 1. Management roadmap of aqueous radioactive waste following [1] with pathway utilized in the current work shown by red arrows. Reproduced with permission of IAEA.

In recent decades, Georgia has taken active measures for developing a national system for radioactive waste management, including a national strategy for radioactive waste management for the period from 2017 to 2031 that was officially adopted in 2016. The strategy is focused on the predisposal management of radioactive waste and sets six main goals. The first goal of the strategy considers location of all radioactive waste facilities on one site that is properly selected and licensed. Many investigations were conducted to identify such a site within the country. Based on the results and the legacy of former nuclear activities [4], including the “Radon” type facility which operated nearby in the past, the government of Georgia took the decision to define the s.c. Saakadze site as the preferred location. The detailed site investigation and assessment started several years ago with projects supported by Sweden’s Radiation Regulatory Authority (SSM) and the European Union (EU) as well as by the International Atomic Energy Agency (IAEA). Special attention was paid within these projects to the legacy liquid radioactive waste underground tanks on the Saakadze site, which were an historical remnant from the past operations of the national “Radon” central radioactive waste storage facility. The investigation showed that about 50 m³ of the legacy aqueous waste stored in these tanks has been contaminated by long-

lived radionuclide ^{226}Ra . Thus, a significant part of activities dealt with characterization and processing of legacy liquid radioactive wastes on the Saakadze site, particularly with assistance from the IAEA.

The aim of this paper is to describe the works completed under IAEA assistance on the Saakadze site in Georgia to treat and condition the legacy liquid radioactive waste, which has resulted in an increased safety of on-site storage of conditioned waste, and will enable its safe disposal on this site.

2. Liquid Radioactive Waste Characterization

Three liquid radioactive waste underground tanks are situated on the Saakadze site some 30 km from the capital of Georgia, Tbilisi. It is important to emphasize that all documents describing site conditions and its facilities were completely lost during the civil war in the country during the 1990s. The first investigation of the site was conducted jointly by local and SSM specialists who identified the presence of contaminated water in the first of the three underground tanks. The detailed investigation of the tanks was conducted within the EU project G.4.01.08, including identification of tank dimensions and containments [5]. Figure 2 presents a schematic of the orientation of the tanks, their sizes, design and construction materials.

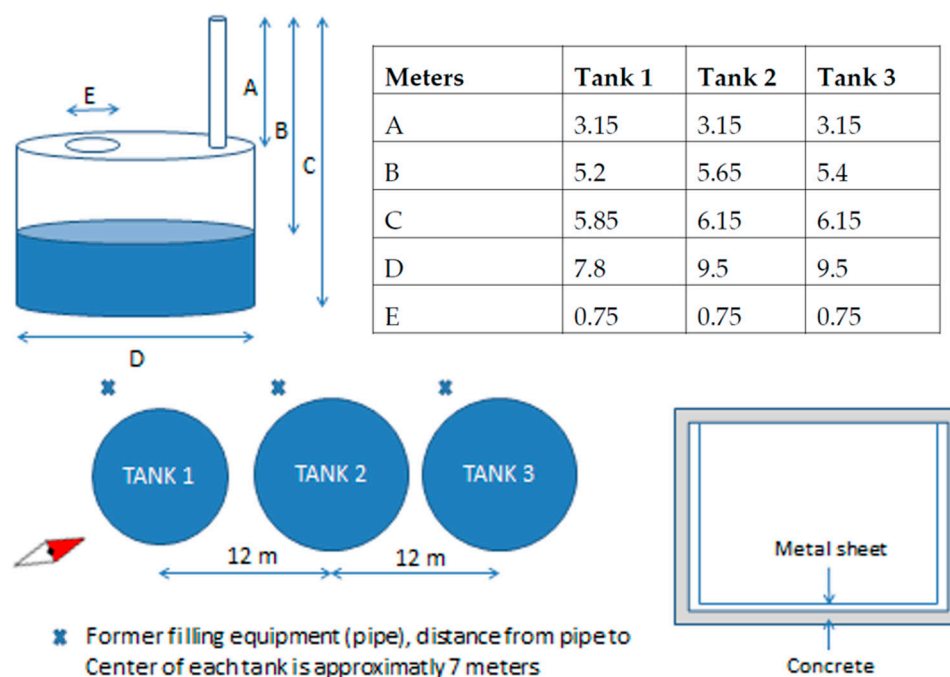


Figure 2. Schematic of liquid radioactive waste underground tanks at the Saakadze site, Georgia.

It was estimated that the volume of contaminated water in the first tank is about 41.5 m^3 . Samples of contaminated water from the tank were analyzed and several radionuclide contaminants were found, among which the long-lived ^{226}Ra had the highest concentration (Table 1). Water sampling was from the bottom of tank. Five samples taken contained 20 mg/L of suspended particles, although their chemical and dispersed composition were not analysed. Unfiltered water was analyzed, and Table 1 gives data after averaging of the results obtained.

In line with Georgian legislation and established clearance level for liquids, which specifies for ^{226}Ra that the discharge can be done at its concentrations in water below 10^3 Bq/kg [6], the given contaminated water is assigned as radioactive waste, with ^{226}Ra as the main contaminant.

Table 1. Concentration of radionuclides in the water stored in the first underground tank.

Radionuclide	Activity, Bq/kg	Measurement Error, %
²² Na	3.1	43
⁴⁶ Sc	16.5	49
¹⁴⁰ La	5.3	66
¹⁵⁵ Eu	16.0	61
²¹⁴ Pb	37.8	1.1
²⁰⁷ Bi	1.7	110
²¹⁴ Bi	8.8	1.4
²²⁶ Ra	1215	5.2

3. Treatment of Waste

Several well-established aqueous waste treatment methods are currently available with Table 2 summarizing their main features and limitations [3,7–19].

Table 2. Main features and limitations of aqueous waste treatment methods.

Method	Features	Limitations
Filtration	Removal of suspended solids. Use as polishing step after chemical treatment. Use upstream of ion exchanger. Backwash is possible.	Not suitable for colloids. Need to replace filter media.
Evaporation	High DF ¹ > 10 ³ . Well established technology—many different designs. High volume reduction factor. Concentrate can be directly immobilized or dried to produce a salt cake. Utilized mostly as batch process. Condensate may require polishing depending on the activity.	Not suitable for small volumes of aqueous waste generation. Process limitations (scaling, foaming, corrosion, volatility of certain radionuclides). High capital and operating cost with high energy consumption.
Sorption/Ion exchange (organics)	DF high on low salt content (10 ²). High DF also possible for high salt content by use of specific resins. Regeneration of resins possible.	Some colloidal particles and resin fines may pass straight through to the treated water. Limited radiation, thermal and chemical stability of the resins. Resins cost. May require some chemical treatment before conditioning.
Sorption/Ion exchange (inorganics)	10 < DF < 10 ⁴ . Chemical, thermal and radiation stability better than organic ion exchangers. Relatively easy immobilization. Mostly used as once through cycle.	Some colloidal particles and sorbent fines may pass straight through to the treated water. Possible high cost for specific sorbents.
Microfiltration	Removal of fine particulates. Pore sizes range from 0.05 and 5 µm. Low pressure operation (100–150 kPa). High recovery (99%). Low fouling when air backwash is employed. Mostly used as the first step in treatment.	Used for suspended fine particles, but not colloidal matter. Backwash frequency depends on solids content of waste stream. Short lifetime of organic membranes. Inorganic membranes exhibit greater mechanical durability than polymeric membranes. High cost of inorganic membranes.

Table 2. Cont.

Method	Features	Limitations
Ultrafiltration	Removal of colloidal materials and large dissolved molecules. Pore sizes range from 0.001–0.01 μm . Pressure < 1 MPa. DFs in the region of 10^3 for α and 10^2 for β and γ emitters. High volume reduction factor can be achieved. Good chemical and radiation stability for inorganic membranes.	Fouling—need for chemical cleaning and backflushing. Organic membranes subject to radiation damage. Short lifetime of organic membranes. Inorganic membranes exhibit greater mechanical durability than polymeric membranes. High cost of inorganic membranes.
Nanofiltration	Separation of salts with charge differences and separation of high molecular weight organics from high concentration monovalent salt solutions. Pore sizes between 0.001 and 0.01 μm . Pressure from 0.3 to 1.4 MPa. Functions between ultrafiltration and reverse osmosis, and is often termed ‘loose reverse osmosis’.	Organic membranes subject to radiation damage. Short lifetime of organic membranes.
Reverse osmosis	Removes dissolved ions and small molecules that contaminate aqueous solutions. $10 < \text{DF} < 10^2$. Well established for large scale operations. Compete with other separation processes (such as evaporation). Suitable for waste streams with complex radiochemical compositions.	High pressure system, limited by osmotic pressure. Non-back-washable, subject to fouling.

¹ DF stands for the decontamination factor, which is defined as the ratio of the initial specific radioactivity of waste to the specific radioactivity of purified water.

The preliminary analysis of treatment methods commercially available has identified the sorption and ion exchange method as the most effective technology to treat the aqueous waste on site (see the pathway selected in Figure 1). The contaminated water treatment (purification) was conducted within the IAEA Technical Cooperation national project GEO9013, involving both local and international experts. Based on the Statement of Work (SoW) jointly prepared by local and IAEA experts and the results of the tender conducted by the IAEA, the Federal State Unitary Enterprise “Radon”, Russia, was identified as the subcontractor to complete the works on site jointly with local experts [20].

Modernized manganese dioxide (MDM) [21] and strong acid cationic resin KU-2-8, which is an analog of DOWEX HCR-S resins, were selected as basic materials and tentatively tested to identify the main technological parameters of the purification process. The tests proved an effective reduction of ^{226}Ra concentration using the sorption-ion exchange method, with expected operational lifetime of materials used as follows:

- MDM—100–1000 bed (column) volumes;
- KU-2-8—50–200 bed (column) volumes.

The flow rate used was up to 10 bed volumes per hour.

Based on the SoW and test data obtained a modular facility was devised by FSUE “Radon” similar to [13], which was manufactured so that the technological equipment is housed inside metallic 200 L drums. The technological scheme of the equipment is shown in Figure 3.

The initial aqueous radioactive waste was pumped out from the first underground tank (IT) and fed to the plant by pump P1, at the flow rate of 0.45 m^3/h . The flow rate was determined by a rotameter at the outlet and controlled by a water meter. The excess of aqueous radioactive waste was dumped back into tank IT via the bypass line, supplied with a valve V1. Adjustment of the flow rate of aqueous radioactive waste through the plant was carried out using valve V1.

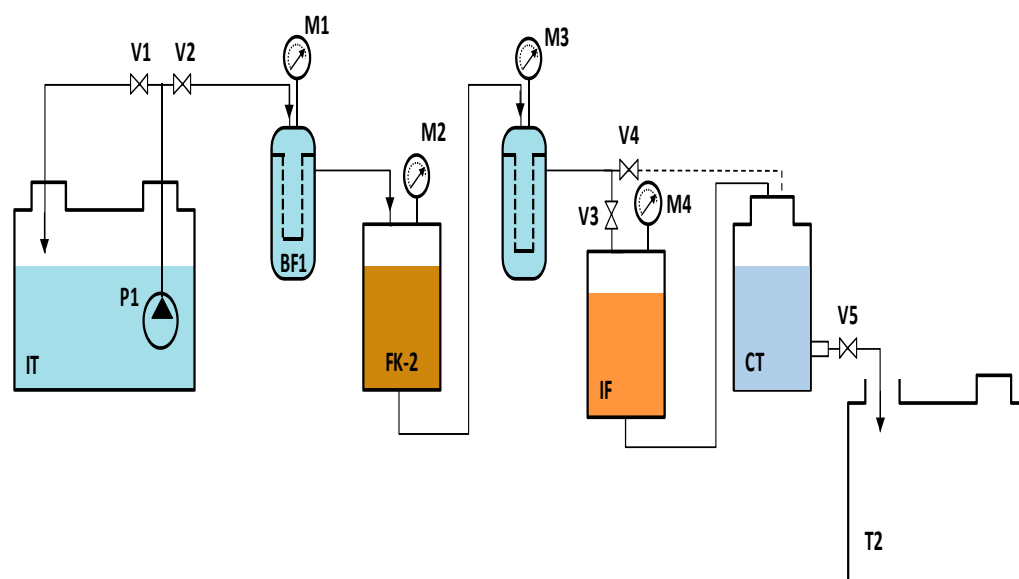


Figure 3. Diagram of water purification installation used on site.

At the first stage of treatment, the aqueous radioactive waste was passed through a bag filter BF1 with pore size of 1 μm .

Then the aqueous radioactive waste was further cleaned of ^{226}Ra in the filter-container FK-2, containing as sorbing agent the selective sorbent MDM.

The bag filter BF2 with pore size of 1 μm was used to further purify the aqueous radioactive waste from suspended particles.

The ion exchange filter IF containing the cation-exchange type resin KU-2 in Na-form was used for purification of aqueous radioactive waste from the ^{226}Ra decay products.

The purified water was collected in the control tank CT. Every 10 hours, representative samples were taken from CT for gamma spectra analysis in the laboratory, using a high purity Ge detector.

The water stream purified was accumulated at the second tank (T2) for its potential use on site, or alternatively for its discharging into the environment following regulatory procedures.

During the treatment work, which lasted 12 days, several replacements of the filtering elements in the filters BF-1 and BF-2 were carried out. During the procedure it was found that the actual volume of the aqueous radioactive waste in the tank was larger than had been initially assessed due to the unexpected shape of the bottom of the tank. The actual volume of the contaminated liquid was 50 m^3 . After processing of 41.5 m^3 of the aqueous radioactive waste, the cleaning efficiency of the filter FK-2 began to decrease. The hydraulic resistance of the filter increased up to 0.1 MPa. Therefore, the FK-2 filter was disabled and the ion exchange resin in the filter was replaced. The purification was continued using the BF-1 filter and the IF filter until the first tank containing the aqueous radioactive waste was completely emptied. The bottom sediment in the tank was left in place and thus might require further processing depending on further use of this tank.

4. Discussion

During the purification process express analyses were conducted by measuring the radiation levels and radionuclide content in the aqueous radioactive waste passing through the apparatuses by gamma spectrometry means. The first express analysis of cleaned water showed an increased level of beta radiation [22]. Spectroscopic measurement conducted by Falcon-500 at measurement times of 15–30 min identified the increase of radium daughter product ^{214}Bi , which has a very short half-life of 19.9 min (Figure 4).

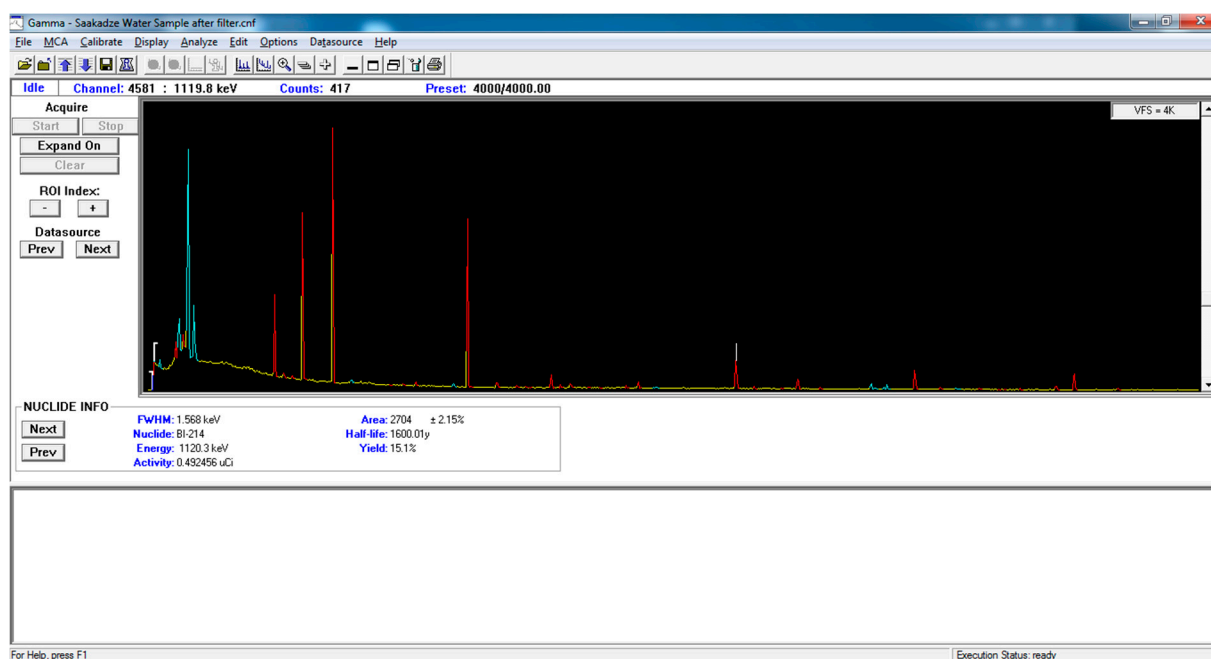


Figure 4. A typical sample of gamma spectra of purified water showing the presence of ^{226}Ra decay product ^{214}Bi .

After keeping the purified water for about 2 to 3 h, the radioactivity levels reduced dramatically, as shown in Table 3, which gives data on radionuclide contents in purified water, where the measurement was conducted for 8 h.

Table 3. Radionuclide concentrations in the purified water.

Radionuclide	Activity, Bq/kg	Measurement Error, %
^{22}Na	1.1	43
^{40}K	15.1	95
^{46}Sc	12.9	19
^{109}Cd	98.8	22
^{134}Cs	1.3	39
^{140}Ba	3.1	60
^{140}La	2.5	25
^{143}Ce	5.7	49
^{155}Eu	13.9	19
^{237}Np	7.0	98
^{226}Ra	<23.1	- 1

¹ When determining the concentration of radium, only the maximum level is given which is very small and therefore the error is undefined.

These data show that specific levels of purified water are acceptable for discharge and that the water purified can be assigned as clean water acceptable for reuse or discharge.

5. Conditioning of Secondary Waste

The following secondary radioactive waste was generated during the processing activities on site:

- spent sorbents;
- spent ion exchange resins;
- spent filter elements;
- pump and piping.

All this waste was immobilized using a purposely prepared cementitious matrix, which is typical for low level radioactive waste immobilization [23]. A significant advantage

of the equipment used was that its technological elements (shown above in Figure 4) were installed inside prefabricated metallic drums. Therefore, the spent sorbents, ion exchangers and filter elements were not removed from their housings and could be directly cemented inside the drums, see Figure 5.



Figure 5. The equipment prepared for immobilization in the cementitious matrix.

The content of ion exchange resins in the cementitious compound was 10% in terms of dry resin. The water-cement ratio was 0.5. The spent filter elements, pipelines and the pump were placed in the same drum with bag filters (Figure 5). As a result of aqueous radioactive waste processing, three metallic 200 L drums containing cemented radioactive waste were produced (Figure 6).



Figure 6. The three drums containing the secondary waste from treatment of 50 m³ liquid radioactive waste immobilized in a cementitious matrix. The fourth drum in the back was used as operational waste collection container during working shifts.

The drums with conditioned radioactive waste were transported to the Centralized Storage Facility (CSF) for safe storage pending final disposal. The overall volume reduction factor (VRF) achieved through treatment followed by conditioning of waste was thus $VRF = (50/0.6) = 83.3$.

6. Conclusions

The IAEA-assisted treatment of 50 m³ of legacy aqueous waste conducted at the Saakadze site in Georgia was successfully completed within a month as a result of the important preparatory works, which comprised characterization of waste and identification of the most suitable treatment technologies. It has, nevertheless, revealed that the actual parameters differed from those tentatively assessed: e.g., the total volume of waste processed was 50 m³ instead of the 41.5 m³ assumed. Another unexpected feature was the increased level of radioactivity immediately after treatment, which was due to concentration of the very short-lived ²²⁶Ra decay product. A positive lesson learned was the organization and successful operation of the international team led by the IAEA.

The treatment of legacy liquid radioactive waste at the Saakadze site has utilized the sorption/ion exchange technology implemented by a prefabricated modular type of facility, which had its parts encased in metallic drums. This form of housing enabled an effective conditioning of all secondary waste at the end of the treatment campaign. The treatment process resulted in the fully purified 50 m³ of water being left on site for further reuse or discharge, and three 200 L metallic drums with cemented radioactive waste currently in safe storage.

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