

IMPROVEMENT OF WATER PURIFICATION PLANT IN SIA "RADON"- 10189

Savkin A.E., Karlin J.V., Flit V.J., Ermakov A.I.
SUE SIA " Radon ", Moscow

Abstract

Experiments on definition of radionuclides forms present in low saline liquid radioactive waste (LRW), generated during processing and storage of radioactive waste (RW) at SIA "Radon" Moscow have been carried out. For LRW treatment from most radionuclides and salts a two-stage reverse-osmosis apparatus was tested. Laboratory experiments on aftertreatment of permeate from stage 1 of the reverse osmosis apparatus were carried out. The new technological scheme for a water purification plant was offered, considerably reducing the amount of secondary radioactive waste. Putting the treatment scheme into operation is planned at the end of 2009.

1. Introduction

Liquid radioactive waste (LRW) treatment, as a rule, is aimed to solve two primary tasks:

- LRW treatment of radionuclides and harmful chemical substances to regulatory requirements;
- Maximum reduction of the amount of secondary radioactive waste generated.

Task 2 considerably reduces the cost of long-term storage/disposal which generally defines the cost of LRW management.

At SIA Radon Moscow, for treatment of internal low saline LRW generated as a result of plant operation (decontamination of the equipment and special vehicles for RW transportation, as well as storage of solidified radioactive waste forms) a water purification plant is used. The plant includes devices for LRW treatment of mineral oil, suspensions, dissolved organic compounds and ion-exchange resin. Last year, because of the change of quality and volume of the influent LRW the ion-exchange resin operation cycle was significantly reduced. This considerably increased the volume of secondary regenerant solution to be directed to long-term storage after conditioning.

To determine the reason for the resin cycle reduction experiments were carried out under laboratory conditions. Microfiltration, ultrafiltration, reverse osmosis, and coagulation and reagent oxidation (permanganate and peroxide oxidation) were tested. Based on results of laboratory experiments, a two-stage reverse osmosis apparatus was proposed as the basic demineralizing equipment for the water purification plant. Trial tests carried out with capacity up to 10 m³ / hour on real LRW have shown the efficiency of the apparatus. For radionuclides aftertreatment of permeate from reverse osmosis stage 1 to regulatory levels, filtration through a ferrocyanide sorbent and cation exchanger with anion exchanger was tested.

Based on the experimental results the recommendations for improvement of the technological scheme of water purification plant for low salt LRW treatment were given.

2. Experimental part.

2.1. Operation of water purification plant.

The water purification plant was put in operation in the late 1970s [1]. Principal process diagram of the plant is shown in Fig. 1.

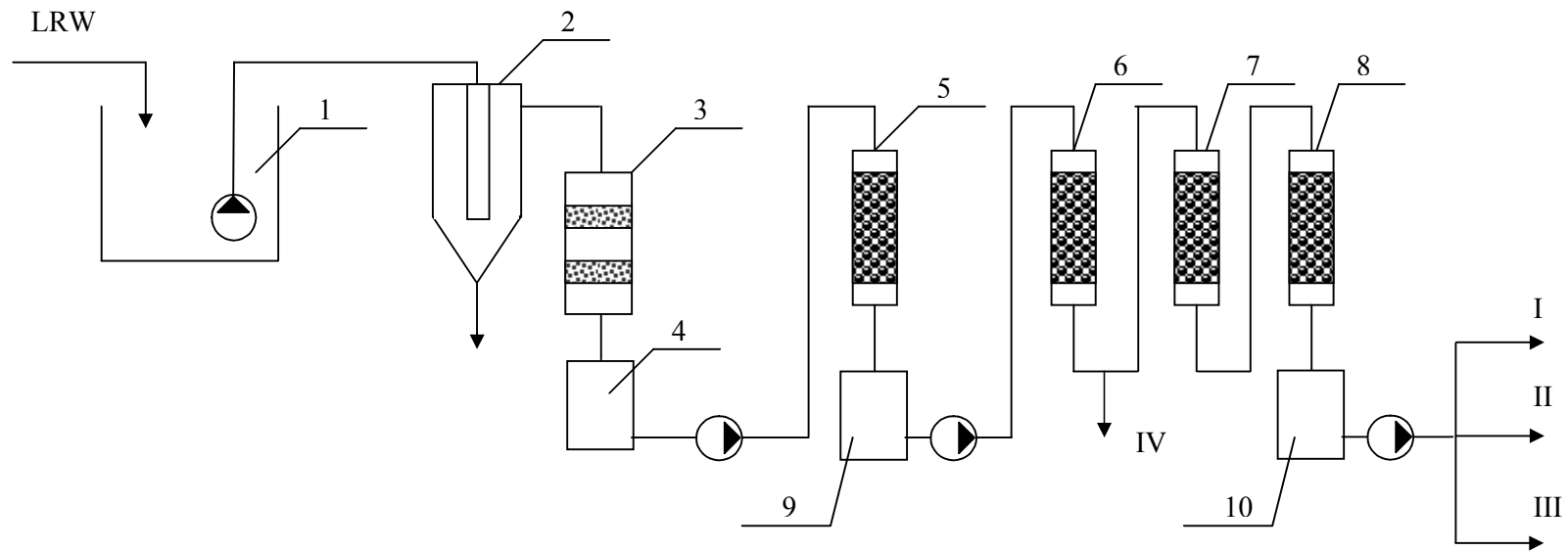


Figure 1. Process diagram of the water purification plant

1 - reception tank, 2 – oil collector, 3 – filter with a waterproof material, 4,9 – intermediate tank, 5 – sand filter , 6 - charcoal filter, 7,8 –ion exchange filter, 10 – control tank,
 I, II – for internal consumption, III – to hydrosphere, IV – to ROA

LRW from various sources comes into reception tank 1. Then LRW from tank 1 is directed by pump for treatment of non dissolved organic substances into an oil collector 2 and then into filter 3 filled with waterproof material. Filtrate from tank 4 is pumped through the sand filter 5, and then LRW from tank 9 is directed for treatment of the dissolved organic substances into the charcoal filter 6, and salts and radionuclides are directed into vessels 7, 8, filled with cation exchange and anion exchange resin, respectively. Regeneration of the filter 5 is carried out by reverse flushing with filtrate, and vessels 7 and 8 are regenerated by solutions of nitric acid and alkali.

The plant is intended for radionuclides treatment of the following LRW:

- from decontamination of the equipment and special vehicles for RW transportation,
- trap and shower waters from the main technological building (MTB),
- drainage waters from RW storage site.

Treatment of LRW, generated in emergency situations and meeting the reception criteria is possible.

Final product resulted from the plant operation is water cleaned up to levels that allow discharge to a special sewage system (stream III) or its further reuse: additional charging of circulating water supply and decontamination special vehicles for RW transportation and equipment (streams I, II). A secondary waste is generated from ion exchange vessels. Last century the annual regenerating solution volume was 50- 60 m³, and in the last few year it reached 200 – 250 m³. Apparently, a principal cause of ion-exchange cycle reduction is the increase in concentration of organic substances in drainage waters from the RW storage site. Last century [2], especial in the 1960 – 70 s, joint disposal of unconditioned solid radwaste (SRW) and cemented salt waste LRW forms at near surface facilities was carried out. During disposal of unconditioned SRW containing significant share of organic materials (paper, rags, wood, carcasses of experimental animals, etc.) there was a natural decomposition of organic substances. It gradually has resulted in increase concentration of organic substances in drainage waters and to formation of various radionuclides forms in liquid waste. To define these radionuclides forms the laboratory experiments were carried out.

2.2. Definition of radionuclide forms present in LRW.

Experiments were carried out on LRW from one tank of collected waters from drainage wells of RW storage site. Radionuclides and the chemical structure of the LRW is given in Tables 1 and 2.

Table 1. Radionuclides structure of drainage waters, Bq/l

$\Sigma \alpha,$	$\Sigma \beta,$	Cs ¹³⁷	Sr ⁹⁰	Y ⁹⁰
40	670	650	60	11,7

Table 2. Chemical structure of drainage waters, mg / dm³

pH	Salt content	NO ₃ ⁻	NO ₂ ⁻	Cl ⁻	SO ₄ ⁻²	Na ⁺	K ⁺	TOC, mgO ₂ /dm ³
9	1351	109	14,2	20	70	350	130	150

As seen from the tables, the main radionuclides in the LRW are Cs¹³⁷ and Sr⁹⁰. The alpha – activity is caused by presence of uranium and plutonium isotopes. Chemical structure of the LRW is defined mainly by sodium nitrate content.

To define radionuclides forms present in the LRW filtration through a microfilter (0.2 microns pore size), an ultrafilter (<0.05 microns pore size) and a reverse osmosis unit were used. Results of the analysis of the filtrates obtained are given in Table 3.

Table 3. Specific activity of the filtrates, Bq/l

Filtration method	$\Sigma\alpha$	$\Sigma\beta$	Cs ¹³⁷	Sr ⁹⁰	Y ⁹⁰
Microfiltration 0.2 microns	17	580	570	48,8	8,32
Ultrafiltration	0,14	580	470	14	-
Reverse osmosis	<0.1	4	3.6	0.24	-

From the tables, cesium – 137 radionuclides are present in LRW mainly in the ion form. Alpha and strontium – 90 nuclides are present both in the ionic form and in the suspension colloid form (large colloids).

Concentration of organic substances in LRW was estimated on TOC (total organic carbon). For elimination of influent organic substances to ion exchange treatment the following processes were tested:

- Coagulation by iron chloride;
- Peroxide and permanganate oxidation.

After the calculated amount of iron chloride has been introduced and blended, the mixture was settled within 5 hours. After that a decantate was sampled and subjected to analysis. Reagent oxidation was carried out by introduction of an oxidizer solution and mixing for 1 hour. The formed deposit was removed upon filtration and filtrates were directed for analysis.

Results of these experiments are given in Table 4.

Table 4. Results of drainage waters processing, Bq/l

Process	$\Sigma\alpha$	$\Sigma\beta$	Cs ¹³⁷	Sr ⁹⁰	TOC, mgO ₂ /dm ³
Coagulation	4,4	720	740	35	38
H ₂ O ₂	0,47	640	570	<3,7	<30
KMnO ₄	<0.1	580	470	≤4,5	<30

As can be seen from the results, each of these processes enables a considerable decrease in TOC and specific activity for alpha -nuclides and strontium-90 . Although, permanganate oxidation is the most effective process. It is comparable to the reverse osmosis efficiency in respect of radionuclides decontamination factors (except caesium-137).

2.3. Improvement of the technological scheme

Let's try to compare permanganate oxidation in combination with other treatment methods and reverse osmosis method by quantity of the secondary waste. We compared two schemes:

1 - a preliminary filtration from organic substances (mineral oil, dissolved substances) and suspensions + permanganate oxidation + ion exchange cleaning;

2 - a preliminary filtration from organic substances (mineral oil, dissolved substances) and suspensions + reverse osmosis.

Under scheme 1 the main secondary LRW are the following:

- manganese dioxide deposit from LRW oxidation ;

- salts present in LRW and adsorbed onto ion exchange resins;
- nitric acid and alkali required for regeneration of the resins.

Under scheme 2 the main secondary LRW are salts present in LRW and removed by the reverse osmosis membrane. It is clear, that the secondary LRW generated under scheme 2 is much less. Therefore, at the existing water purification plant the two-stage reverse osmosis apparatus (ROA) was mounted in a separate location and connected (Fig. 1, stream IV). The ROA process diagram is shown in Fig. 2.

Each stage of the apparatus consists of 5 rolled elements. Permeate from stage 1 is directed to control tank 10 (see Fig. 1), and a concentrate is directed to stage 2. Permeate from stage 2 is returned to an inlet of stage 1, and a concentrate from stage 2 is directed to a tank from which the concentrate is directed to cementation or cleaned of radionuclides by sedimentation methods with the aim of subsequent vitrification.

The ROA was tested on mixture of drainage and trap waters from the MTB. ROA capacity was 10 m³ / hour on permeate from stage 1, and more than 200 l/hour on concentrate from stage 2. Results of the analysis of permeate from the first stage are given in table 5.

Table 5. Results of the analysis of permeate from ROA stage 1, Bq/l

$\Sigma\alpha$	$\Sigma\beta$	Cs137	Sr ⁹⁰	Salt content, mg / dm ³	TOC, mgO ₂ / dm ³
<0.1	3	2-3	<1	20-50	<30

It is necessary to note, that LRW was sent to the ROA at pH = 7 – 8. In the course of LRW treatment in the concentrate from stage 2 a hardness salts deposit was being formed. It, finally, could lead to irreversible membranes clogging. Therefore, it was offered to adjust pH of LRW to value 4 – 5 by nitric acid. Now these works are at a stage of assembling and installation of the equipment.

Values of specific activity on beta – nuclides, given in table 5, negligible exceed the control levels for treated waste discharge into the environment. Therefore, under laboratory conditions 2 circuits of aftertreatment were tested:

- Ion exchange by cation exchanger + anion exchanger;
- Filtration by a ferrocyanide sorbent such as NZS.

Through columns filled with above mentioned sorbents 4000 column volumes of permeate from stage1 were passed. In both cases specific activity on Cs¹³⁷ was below control levels. From the point of view of spent sorbent management the NZS option is preferable.

As was mentioned above, for elimination of sedimentation process on reverse osmosis membranes it is necessary to adjust pH to 4 – 5. This, however, results in a change of radionuclides sorption conditions and also infringement of standard pH values of the cleaned solution (water) which should be 6 – 8. Therefore, permeate from stage 1 was adjusted to pH = 4.5 and passed through columns filled with limestone and NZS. During passing of 2000 permeate column volumes the pH changed in a range of 7.5 – 6.5. The specific activity of the filtrate has not exceeded control values.

Thus, from laboratory results and full-scale experiments a low salt LRW treatment process should consist of the following stages:

- preliminary filtration of organic substances (mineral oil, dissolved substances) and suspensions;
- adjusting pH to 4 – 5 with nitric acid;
- two-stage reverse osmosis;
- pH adjustment through limestone columns;
- aftertreatment by NZS sorbent.

Placing the proposed scheme in operation is planned for the end of 2009.

3. Conclusions

3.1 Experiments to determine the forms of radionuclides present in low salt LRW, generated during processing and storage of RW at SIA "Radon" Moscow have been carried out.

3.2 For LRW treatment of radionuclides and salts the two-stage reverse osmosis apparatus was tested.

3.3 Laboratory experiments on aftertreatment of permeate from stage 1 of the reverse osmosis apparatus have been carried out.

3.4 The new technological scheme for the water purification plant was proposed to reduce the volume of secondary radioactive waste.

References

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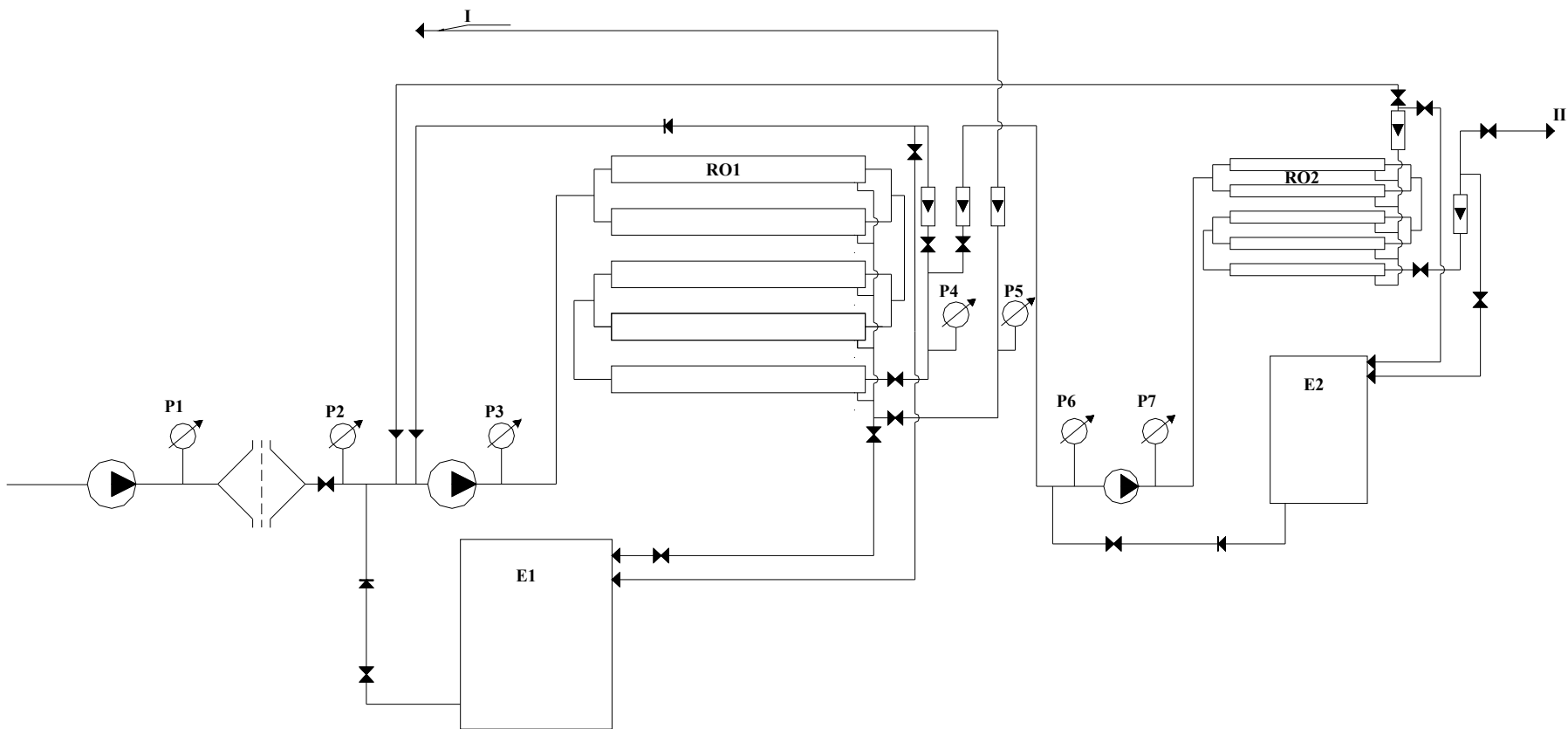


Fig.2. Process diagram of the ROA

RO 1 - reverse osmosis apparatus of stage 1

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