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REPROCESSING OF LOW SALINITY LIQUID RADIOACTIVE WASTE IN MOBILE PLANT «ECO-3» AT RUSSIAN STATE CENTER FOR ATOMIC SHIPBUILDING IN SEVERODVINSK

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ABSTRACT

A number of mobile plants for the liquid low and intermediate level radioactive waste (LLILW) reprocessing was developed and assembled at Moscow SIA "Radon". These plants can be used at places where LLILW is generated.

During the utilisation and repairing of atomic submarines at FGUP «Zvezdochka» of Russian centre for atomic shipbuilding in Severodvinsk, Arkhangelsk region saline liquid radioactive wastes are formed. The mobile plant of SIA "Radon" «ECO-3» was applied for the purification such waste [1].

About 820 m^3 of low salinity LLILW from the tanker «Osetia» was purified in the plant «ECO-3» within the first half of 1999. The plant «ECO-3» operation was based on sorption and electromembrane technology of low level salted LLILW purifying and concentrating with the use of selective sorbents and electrodialysis apparatus. Also the conditioning of formed secondary waste was carried out.

The report contains an apparatus technological flow sheet, results of radiometric, spectrometric measurements and chemical analysis of initial and purified LLILW. Coefficients of purifying and concentrating for different radionuclides are represented as well as overall technical and economical parameters.

INTRODUCTION

Various liquid radioactive wastes are produced at the state enterprise FGUP «Zvezdochka» of Russian center for atomic shipbuilding in Severodvinsk Arkhangelsk region during the repair and maintenance of nuclear submarines. Some of liquid radioactive waste are saline LLILW. The Moscow SIA "Radon" started its activities on purification of these LLILW beginning 1996. A mobile technological plant "ECO-3" was developed by SIA "Radon" specialists for that purpose. This plant is assembled into a standard 20-foot sea container.

TECHNOLOGY

The chemical composition of LLILW to be treated is given in the Table 1. As one can see from this table, the waste is slightly mineralised: total mineralization is 2.6 g per litre with relative high content of chlorides, nitrates, phosphates and sulphates.
The radiometric and spectrometric composition of initial LLILW is shown in the Table 2.
The initial LLILW at FGUP "Zvezdochka" is a salt solution, in which the content of five radionuclides is higher than permitted concentrations, namely:

- Caesium-137, 70 times;
- Caesium-134, 3.5 times;
- Strontium-90, 35 times,
- Cobalt-60, 12 times,
- Tritium, 10 times.

The content of alpha-bearing radionuclides is below of MPC limit for any of these radionuclides. Beta-radionuclides Carbon-14 and Nikel-63 were identified also.

A sorption-membrane technology was recommended for purification of LLILW and concentration of salts, which is followed by cementation of obtained concentrates and disposal of cemented waste into repositories.

The flow sheet diagram of the mobile plant "ECO-3" is shown in Fig.3. This mobile plant consists of two blocks of purification-concentration, namely:

1. Sorption unit;
2. Electrochemical membrane unit.

The sorption unit comprises cartridge filters (or filter-containers) with inorganic sorbent "Phoenix" produced by SIA "Radon", sorption filters with the strong acidic cationic ion-exchange resin KA-11 in sodium form.

The electrochemical membrane unit comprises electrodialysis apparatuses for desalinisation and concentration.

It is necessary to mention that the membrane electrodialysis method is one of the most ecological clean and economical efficient purification methods in application to slightly mineralised LLILW. R&D works at SIA "Radon", which are carried out beginning 1970-s in this area, confirmed this conclusion.

The choice of the sorbent to be used before electromembrane desalinization and concentration was governed by three factors:

1. Radionuclide composition of LLILW;
2. Maximum reduction of dose rate in the plant;
3. Prevention of sedimentation in the electrodialysis and electroosmosis apparatuses.

Taking into account the radionuclide composition of LLILW the following sorbents were chosen:

- For the removal of caesium – ferrocianide sorbent "Phoenix-A";
- For the removal of strontium - strong acidic cationic ion-exchange resin KA-11 in sodium form, which also possess the selectivity to cobalt ions on the sodium ions background. .

In order to reduce the dose rates in the plant the removal of caesium is carried out at the beginning of the purification process through two filters-containers (cartridge filters) with the volume of inorganic sorbent 30 litres each. This permitted to resolve two tasks:

To remove 98% of caesium at the beginning of process and to prevent following accumulation of caesium in other apparatuses of plant;

To eliminate following handling of waste, since cartridge filters are originally inside of metal-drums – containers and are suitable for transportation and disposal.

The employment of sorption technology for radionuclides removal permitted to obtain decontamination factors for caesium 1,000 and for strontium 100 before the electrochemical unit of plant.

Taking into account the enhanced general content of salt in initial LLILW as well as content of such species like chlorides, nitrates, sulphates, phosphates, ammonia with concentrations larger than MPC limits for exempt releases, following purification of LLILW was conducted through two electrochemical membrane apparatuses. Technical parameters of these two electrodialysis apparatuses are given in the Table 3.

The technological parameters of membrane apparatuses for desalination and concentration are given in the Table 4. As one can see from this table the general specific consumption of electric power was below 10 kW h per cubic meter of purified LLILW (including pumping of working solutions).

A particular feature of given technology consists in application of electroosmosis apparatus for concentration with non flowing liquor chambers, which has the dilute circuit as a bypass pipeline for the LLILW circulation pump through liquor loop of the demineralisation electrodialyser. The purpose of this scheme is to enhance the degree of concentration of radionuclides in a small volume and to increase the operation reliability of the demineralisation electrodialyser. Due to this feature it was possible to obtain the final concentrate of LLILW with the salt content between 120 and 200 g per litre, this concentrate being solidified by cementation.

RESULTS

The results of radiometric and spectrometric analyses of purified LLILW are shown in the Table 5. One can see that for all radionuclides their concentration are much below MPC given in the national regulatory document NRB-76/87.

The results of chemical analyses of purified LLILW are given in the Table 6. Herein are given also MPC for releases in fish hatchery basin.

407 cubic meters of LLILW from the tank A-02/1 were purified in 1996-1997 during 800 operation hours of mobile plant ECO-3. Only 2.5 cubic meters of solidified (cemented) radioactive waste were produced as the result of purification campaign. The purified water was discharged into the industrial sewage system with the permission of controlling authorities. This purification campaign was carried out by SIA "Radon" specialists.

500 m^3 of LLILW from the tanker "Osetia" were purified in March – April 1999 during 1000 hour operation of the mobile plant ECO-3. 5 cubic meters of solidified radioactive waste were produced as the result of this purification campaign. The purified water was discharged into the industrial sewage system with the permission of controlling authorities. This purification campaign was carried out jointly by FGUP "Zvezdochka" and SIA "Radon" specialists.

320 m^3 of LLILW from the tanker "Osetia" were purified in July – August 1999 during 650 hour operation of the mobile plant ECO-3. The tanker "Osetia" was completely emptied from LLILW thereafter it was transferred to a dry dock for capital repair works. 3 cubic meters of solidified radioactive waste were produced as the result of this purification campaign. The purified water was discharged into the industrial sewage system with the permission of controlling authorities. This purification campaign was carried out by FGUP "Zvezdochka" specialists under the guidance of SIA "Radon" specialists.

CONCLUSION

1. 1227 m^3 of LLILW were purified in industrial conditions by using sorption-membrane technology and applying domestic materials and equipment. Obtained small volumes of concentrates were solidified resulting in 10.5 m^3 of conditioned waste, which were disposed of. Thus the task was resolved to empty the tanker "Osetia" and accumulating waste tank. This allowed to FGUP "Zvezdochka" to receive a number of orders on repair and utilisation of nuclear submarines.

2. The approximate price was estimated to be 300 $ for purification of 1 m^3 of LLILW.

LITERATURE

1. Sobolev I.A., Demkin V.M., Panteleev V.I. et. al., Decontamination and utilization of salt liquid radioactive waste at Zvezdochka State Engineering enterprise in the city of Severodvinsk, Proceedings of International Symposium Waste Management’98, CD-ROM, 1998.

Table 1

The chemical composition of LLILW in the tanker "Osetia"

Parameter	Amount
Total salt content, mg/l	2600
Suspended particles, mg/l	20
PH	7.25
Chlorides, mg/l	720
Nitrates, mg/l	500
Phosphates, mg/l	670
Sulphates, mg/l	432
Surfactants, mg/l	0.58
Ammonia, mg/l	54
Oils, mg/l	4.38
General hardness, mg-eqv/l	2.9

Table 2.

Radiometric and spectrometric composition of LLILW from tanker "Osetia"

Radionuclide	Specific radioactivity		Maximum permitted concentration MPC (DKb, Table 8.3 from NRB 76/87)		Limit controlling levels (MPC) for radionuclides in the seawater. Guidance documents.	
	Bq/l	Ci/l	Bq/L	Ci/l	Bq/l	Ci/l
Caesium-137	37000	$1.0 \bullet 10^{-6}$	555	$1.5 \bullet 10^{-8}$	0.8	$2.0 \bullet 10^{-11}$
Caesium-134	966	$2.9 \bullet 10^{-8}$	318	$8.6 \bullet 10^{-9}$		
Cobalt-60	28120	$7.6 \bullet 10^{-7}$	3330	$9.0 \bullet 10^{-8}$	1.5	$4.0 \bullet 10^{-11}$
Iodine-129	0.1	$2.5 \bullet 10^{-12}$	7	$1.9 \bullet 10^{-10}$		
Nickel-63	370	$1.0 \bullet 10^{-8}$	1036	$2.8 \bullet 10^{-8}$		
Antimony-125	311	$8.4 \bullet 10^{-9}$	3663	$9.9 \bullet 10^{-8}$		
Carbon-14	318	$8.6 \bullet 10^{-9}$	30340	$8.2 \bullet 10^{-7}$		
Tritium	$1{,}77 \bullet 10^{6}$	$4.8 \bullet 10^{-5}$	148000	$4.0 \bullet 10^{-6}$		
Strontium-90	4810	$1.3 \bullet 10^{-7}$	14.8	$4.0 \bullet 10^{-10}$	0.3	$8.0 \bullet 10^{-12}$
$\Sigma\alpha$	0.5	$1.4 \bullet 10^{-11}$	1.1	$3.0 \bullet 10^{-11}$		

Table 3

Technical parameters of membrane apparatuses

№№	Characteristic of apparatus	Apparatus type	
		EDMS (desalination)	EKDSP (concentration)
1	Ion-exchange membrane type	MK-40, MA-40	MK-40, MA-40
2	Number of ion-exchange membranes	602	91
	Cationic	302	46
	Anionic	300	45
3	Electrode material -anode -cathode	 Impregnate graphite Stainless steel	 Platinized titan Stainless steel
4	Number of electrodes	4	2
5	Frame-separator type	Labyrinth spiral with changed cross-section	Slotted with mortgage separation net in dilute chamber and in-contact membranes in concentration chambers
6	Useful surface of ion-exchange membranes, m^2	35	5
7	Patents	D.V. Adamovich, Patent No.1793949 (1993) "Filter-press type electrodialyzator"	V.I. Demkine et. al. Patent No.102948 (1980) "Electrodialyzator electroosmosis appatus"

Table 4

Technological parameters of membrane apparatuses

Type of membrane apparatus	Working regime	Operation regime		Capacity, m^3/h	Pressure in dilute tract, MPa (kg/cm^2)	Electric energy consumption kW h /m^3	
		Voltage, V	Current, A			Desalination	Over pumping
EDMS (desalination)	Circulating with permanent sampling	250	10	0.6	0.2 (2.0)	6.7 per 1 m^3 of desalinated water	2
EKDSP (concentration)	Circulating	60	22	0.005	0.05 (0.5)	1.25	2

Table 5

Radiometric and spectrometric composition of purified LLILW

Radionuclide	Specific radioactivity of purified LLILW in plant ECO-3		Maximum permitted concentration MPC (DKb, Table 8.1 from NRB 76/87)		Limit controlling levels (MPC) for radionuclides in the seawater. Guidance documents.	
	Bq/l	Ci/l	Bq/l	Ci/l	Bq/l	Ci/l
Caesium-137	27.8	$7.5 \bullet 10^{-10}$	555	$1.5 \bullet 10^{-8}$	0.8	$2.0 \bullet 10^{-11}$
Caesium-134	0.4	$1.2 \bullet 10^{-11}$	318	$8.6 \bullet 10^{-9}$		
Cobalt-60	18	$4.9 \bullet 10^{-10}$	3330	$9.0 \bullet 10^{-8}$	1.5	$4.0 \bullet 10^{-11}$
Iodine-129	0.1	$2.3 \bullet 10^{-12}$	7	$1.9 \bullet 10^{-10}$		
Nickel-63	218	$5.9 \bullet 10^{-9}$	1036	$2.8 \bullet 10^{-8}$		
Antimony-125	7	$1.9 \bullet 10^{-10}$	3663	$9.9 \bullet 10^{-8}$		
Carbon-14	141	$3.8 \bullet 10^{-9}$	30340	$8.2 \bullet 10^{-7}$		
Тритий H-3	$1.7 \bullet 10^{6}$	$4.6 \bullet 10^{-5}$	148000	$4.0 \bullet 10^{-6}$		
Strontium-90	1.5	$4.0 \bullet 10^{-11}$	14.8	$4.0 \bullet 10^{-10}$	0.3	$8.0 \bullet 10^{-12}$
$\Sigma\alpha$	0.23	$6.2 \bullet 10^{-12}$	1.1	$3.0 \bullet 10^{-11}$		

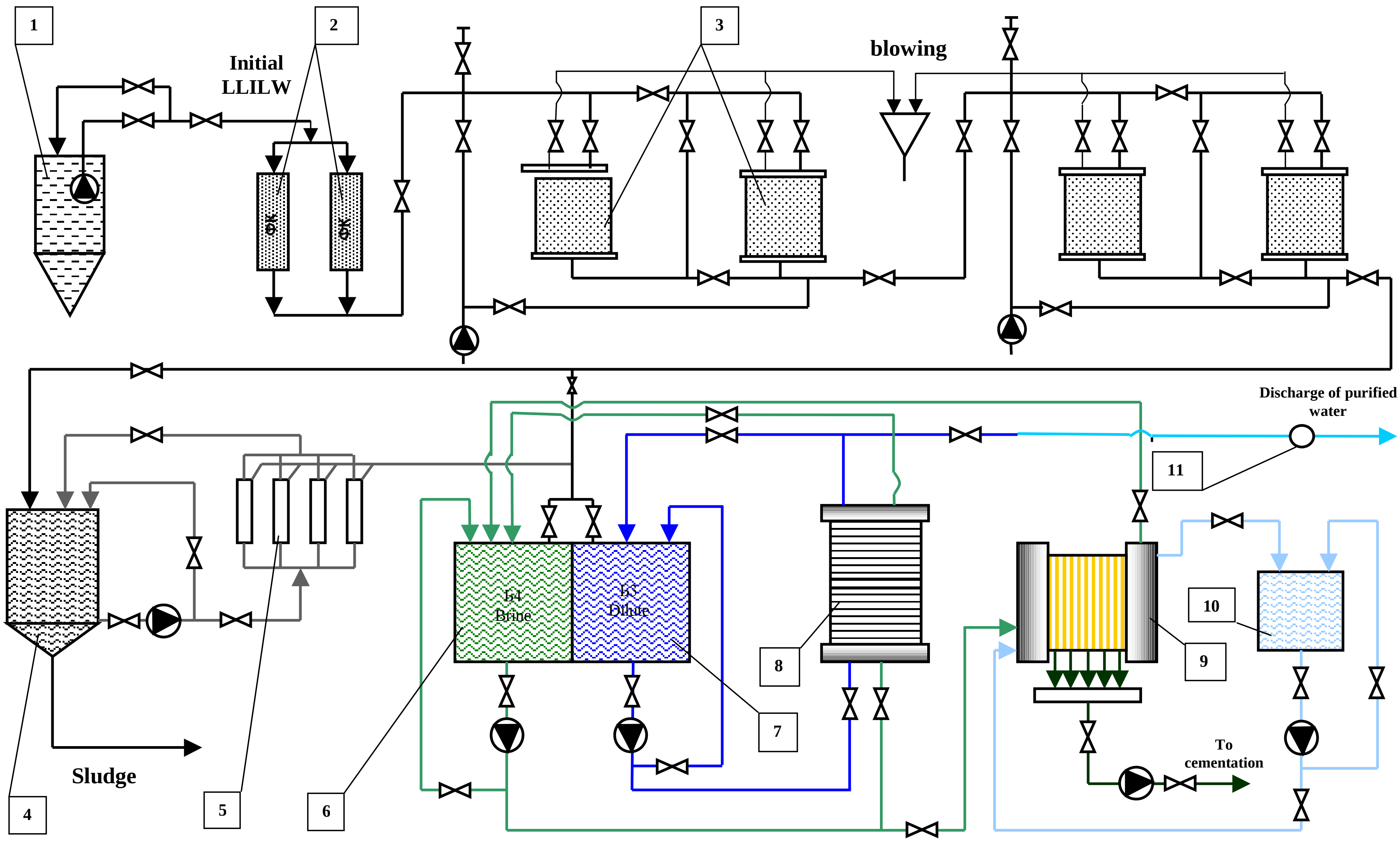
Table 6

Chemical composition of LLILW treated in plant ECO-3

№№	Parameter	Initial LLILW	Purified solutions	Permitted concentrations of chemical substances*
1	Total salt content, mg/l	2600	440	-
2	Suspended particles, mg/l	20	5.85	-
3	pH	7.25	7.1	6.5-8.5
4	Chlorides, mg/l	720	78	150
5	Nitrates, mg/l	500	75	8.1
6	Phosphates, mg/l	670	17	3.3
7	Sulphates, mg/l	432	32	50
8	Surfactants, mg/l	0.58	3	-
9	Ammonia, mg/l	54	12.5	16
10	Oils, mg/l	4.38	0.16	-
11	General hardness, mg-eqv/l	2.9	0.17	-

*The list of Maximum Permitted Concentrations for relative safe levels of exposure of hazardous substances for the water in fish hatchery basins, Approved by the Committee for pisciculture of Russian Federation, 31.12.92. № 54.

Fig.1 Technological scheme of purification of LLILW from tanker "Osetia"



1. Tank with initial LLILW; 2.Filter-container; 3. Filter; 4. Ultrafiltration tank; 5. Ultrafiltration module; 6. Brine tank; 7. Dilute tank; 8. Electrodialyzator EDMS; 9. Electrodialyzator-concentrator EKDSP; 10. Tank for washing solution; 11. Counter for purified wa ter.