Innovative Process for Comprehensive Treatment of Liquid Radioactive Waste -12551

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ABSTRACT

This paper presents the results of research activities aimed at creation of a principally new LRW distilling treatment method. The new process is based on the instantaneous evaporation method (flash process) widely used in distillation units. The main difference (know-how) of the proposed process is that the vapor condensation is conducted without using heat exchangers in practically ideal mode (thermodynamic equilibrium) by way of direct contacting in a vapor-liquid system. This process is conducted in a specially designed ejector unit in supersonic mode. Further recuperation of excess heat of vaporization is carried out in a standard heat exchanger (liquid-liquid system).

Such an arrangement of the process, together with use of the barometric height principle, allows to carry out LRW evaporation under low temperatures (within an interval of 40 to 50 °C), which enables to use excess heat from NPS for heating initial LRW. Thermal calculations and model experiments have revealed that, in this case, the expenditure of energy for LRW treatment by distilling will not exceed 3 kilowatt-hour/m³, which is comparable with the reverse-osmosis desalination method.

Besides, the proposed devices are 4 to 5 times less metal-intensive than standard evaporation units.

These devices are also characterized by versatility. Experiments have revealed that the new method can be used for evaporation of practically any types of LRW, including those containing a considerable amount of oil products. Owing to arrangement of the evaporation process at low temperatures, the new devices are not sensitive to «scale formation». This is why, they can be used for concentrating brines of up to 500-600 g/l.

New types of such evaporating devices can be required both for LRW treatment processes at nuclear-power plants under design and for treating «non-standard» LRW with complex physicochemical and radionuclide composition resulting from the disaster at the Fukushima I Nuclear Power Plant.)

As a result of accidents at nuclear energy objects, as it has recently happened at NPP "Fukushima-1", personnel faces the necessity to take emergency measures and to use marine water for cooling of reactor zone in contravention of the technological regulations. In these cases significant amount of liquid radioactive wastes of complex physicochemical composition is being generated, the purification of which by traditional methods is close to impossible.

According to the practice of elimination of the accident aftereffects at NPP "Fukushima" there are still no technical means for the efficient purification of liquid radioactive wastes of complex composition like marine water from radionuclides.

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Therefore development of state-of-the-art highly efficient facilities capable of fast and safe purification of big amounts of liquid radioactive wastes of complex physicochemical composition from radionuclides turns to be utterly topical problem.

Cesium radionuclides, being extremely dangerous for the environment, present over 90% of total radioactivity contained in liquid radioactive wastes left as a result of accidents at nuclear power objects.

For the purpose of radiation accidents aftereffects liquidation VNIIHT proposes to create a plant for LRW reprocessing, consisting of 4 major technological modules:

- Module of LRW pretreatment to remove mechanical and organic impurities including oil products
- Module of sorption purification of LWR by means of selective inorganic sorbents
- Module of reverse osmotic purification and desalination
- Module of deep evaporation of LRW concentrates

The first free modules are based on completed technological and designing concepts implemented by VNIIHT in the framework of LLRW Project in the period of 2000-2001 in Russia for comprehensive treatment of LWR of atomic fleet. These industrial plants proved to be highly efficient and secure during their long operation life. Module of deep evaporation is a new technological development. It will ensure conduction of evaporation and purification of LRW of different physicochemical composition, including those containing hardness salts (Ca and Mg salts), resulted in generation of LRW concentrate 300-600 g/l. The method is based on utilization of supersonic ejector for intensification of thermal physic processes and performance of evaporation in brine recycling mode.

All proposed technological solutions are totally based on patented Russian developments.

EJECTOR EVAPORATION PLANT OPERATION PRINCIPLE

The main difference between proposed plant and traditional evaporating facilities consists in principle of contact condensation of steam applied by it. This condensation takes place in supersonic steam-gas-liquid flow, created by special construction of the ejector. Thus, there is a possibility to use instead of vertical tubular heat exchanger-condenser much more compact horizontal heat exchanger of much simpler construction, being utilized in liquid-liquid system. And this, in turn, will allow reducing of its size significantly and, first of all, its height. Due to such constructional features there is a possibility to place all units of evaporation plant in 20-foot high cube container. Boiling temperature of the process is set beforehand by barometric height-H. In case of installation of the equipment (item 6, 8, 10) at H height, exceeding the upper level of the brine in vessel-2 by 1-1.3 m, evaporation process will be conducted in temperature interval of 80-90°C. If such plant is constructed for NPP needs, where H value will not be limited, and the equipment is placed on the assumption that H is about 8.5 m, then evaporation process can be conducted at 40-50°C. This will allow using of excessive heat of NPP itself for evaporation.

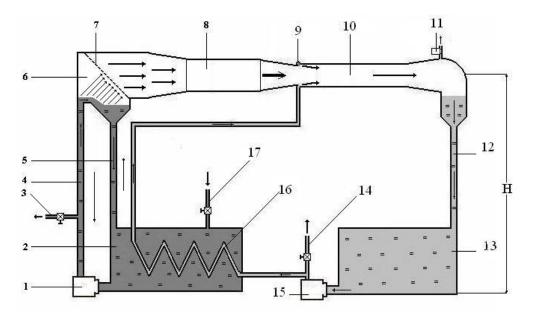
Utilization of compressor for steam compression saves an operator from the necessity to use external cooling source and ensures compact size of the plant. In this case circulating brine will be used as cooling liquid in heat exchanger-16. Thus, for example, during the process of LRW brine evaporation in evaporation chamber-6 at the temperature of 91 °C temperature of the brine, being removed after evaporation of design amount of steam, through main of brine removal-5 in vessel-2 will make up 81 °C. Pressure and temperature of the steam increase after passing the compressor. The temperature of the steam in the outlet of the compressor will make up about 110 °C, and the speed of

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compressed steam in the inlet of supersonic ejector -300 m / s. Design amount of desalinated water with the temperature of 81 °C is being supplied to nozzle unit of the ejector under the pressure of 0.1 mPa. Then water flow with the temperature of 91.2 °C will be formed in the outlet of ejector diffuser. Passing the heat exchanger-16 and transmitting excessive heat to the brine this water will be cooled down to 81 °C and again supplied to ejector for condensation.

For realization of this evaporation process it is necessary to set the ratio of input LRW and fresh water, supplied to fresh water condensation circuit ejector, correctly. In the above given example the proportions will be approximately equivalent.

Fig.1 Flow chart of the evaporation plant with ejector condensation



- 1. Brine pump
- 2. Brine vessel
- 3. Brine outlet manifold
- 4. Brine Circulation Pipe
- 5. Brine Circulation Pipe
- 6. Evaporation chamber
- 7. Dephlegmator
- 8. Compressor
- 9. Ejector Nozzle
- 10. Ejector mixing chamber
- 11. Vacuum pump
- 12. Condensate Circulation Pipe
- 13. Condensate Tank
- 14. Condensate removal manifold
- 15. Condensate pump
- 16. Heat exchanger
- 17. Initial LRW supply manifold
- H barometric height

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BRIEF DESCRIPTION OF THE EJECTOR EVAPORATION PLANT

The plant contains 2 circuits: evaporating circuit and condensation circuit. The fist one is used for continuous circulation of preheated LRW brine from brine vessel-2 to evaporating chamber-6 by brine pump-1.Outlet of circulating mains of brine-4 is equipped with ejector. Due to ejector operation the level of pressure inside evaporation chamber-6 is maintained lower than atmospheric and volume boiling up is taking place at reduced temperature. Steam, generated as a result of boiling up, is supplied to compressor-8 through dephlegmator-7, and brine residue is being returned to brine vessel-2 through circulating main of brine-5.

The steam is being compressed in about 1.3 times in compressor-3, after that it is being supplied to condensation circuit through steam line. This circuit consists of supersonic ejector (9, 10), condensate vessel-13, condensate pump-15 and recuperation heat exchanger-16, installed in brine vessel-2. Nozzle unit of the ejector-9 is set at the point of steam supply, out of (9) prepared in advance desalinated water is being injected to ejector mixing chamber-10 from condensate vessel-13 by condensate pump-15. As a result of mixing of steam and fresh water flow practically complete vapor condensation takes place in ejector mixing chamber-10, and generated gas-liquid mixture (water and solute gases escaped from brine in boiling) is fed to ejector diffuser, where stagnation of supersonic gas-vapor-liquid flow takes place. Condensate coming out of the diffuser is of high temperature due to transmission of excessive heat of vaporization to it. Solute gases, escaped in boiling of the brine, are collected in the upper section of the diffuser and are being withdrawn from the plant by vacuum pump-11. Heat exchange between hot condensate and brine, circulating in evaporating circuit, takes place in heat exchanger-16. Condensate, generated during evaporation of LRW, is being removed through manifold-14. Control over salt content in the brine and its periodic discharge are performed through manifold-3.

We have constructed the pilot plant of 10 dm^3/hr output by condensate. It was used for the model NPP wastewater evaporation experiments using the direct condensation principle. During the tests the brine concentrates with nitrate salt concentration up to 450-600 g/dm³ have been obtained.

In case of implementing ejector evaporation scheme under normal conditions using a compressor and without creation of the barometric altitude the whole process total power consumption will make up 30 kW*h / m³ of generated condensate. If the plant units are located at the barometric height the power consumption will make up about 15 kW*h / m³. At the stationary plants with direct contact condensation of low-pressure steam it is about 3 kW*h / m³.

MAJOR TECHNOLOGICAL AND EQUIPMENT CONCEPTS OF THE PROPOSED PLANT

The plant should consist of 4 technological modules, which can be used either together (in case of sequential connection) or independently (separately). They will be constructed to fit standard 20-foot marine container.

Module for selective purification of all types and categories of LWR from cesium radionuclides will be completed with special protective filter-containers filled with selective inorganic sorbents NFA (nickel-ferrum-cyanide aluminosilicate) type able to absorb up to 120 Cu of Cs-137. Therefore, in case of need, liquid radioactive wastes of medium and high radioactivity also can be purified by these filters.

Cesium radionuclides purification coefficients achieved by using of these filters will make up not less than **20000** in case of two-phase connection. Parallel-series assembling of four

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filters installed in the first module , in case of simultaneous connection, will ensure the possibility of reprocessing of 200 m³ of LRW per day per one module.

Design of the filters allows to disconnect them easily of the system and place for long-term safe storage after their saturation. Both filters themselves and sorbents are certified in the Russian Federation for application in nuclear industry.

The module of reverse osmotic desalination, additionally equipped with sorption filters of afterpurification with inorganic sorbents, will be used for purification of all types of liquid radioactive wastes of medium and low radioactivity to meet all sanitary standards in terms of all chemical and radionuclide impurities. It will ensure achieving of coefficients of purification from all beta- gamma-active radionuclides at the level of **2.000-10.000**.

The module of deep evaporation will be used for brines concentration after reverse osmosis up to salts concentration of 300-600 g/l. The level of concentrate purification from radionuclides will make up about 80.000.

CONCLUSION

Proposed work will allow to construct modular plants, which will be totally prepared for efficient purification of any types of liquid radioactive wastes from radionuclides in case of force majeure. According to proposed scheme concentration level of cesium radionuclides in safe-for-storage form (resistant to radiation inorganic matrix) will make up not less than 5000. With respect to purification from cesium radionuclides of liquid radioactive wastes

stored at NPP "Fukushima" (about 100000 m³) about 10 t of inorganic sorbents, loaded in 160 protective filter-containers, will be required for solving this problem. The amount of secondary wastes (salts concentrates) will be reduced approximately in 5 times in comparison with traditional schemes, applied in purification of secondary LRW of Fukushima-1 by Areva (France) and Kurion (USA) companies.

All units of modular plants will be constructed and manufactured as totally automated, providing their twenty-four-hour safe operation. Modular design will ensure efficiency and let optimize the costs of secondary LRW treatment. In order to ensure off-line operation in emergency conditions the plant should be equipped with auxiliary modules: energy and ventilation ones.

Under normal conditions these modules can be stored in "mothballed" condition at special warehouses under the authority of federal bodies. It will be reasonable to choose required transport facilities (ice class tankers), the most suitable for transportation of modules to target destination beforehand, using vessel classification list.

In Fig. 2-4 the optimum layout is shown of the three major technological modules required for the remediation of emergency situations similar to that happened at "Fukushima-1" NPP.

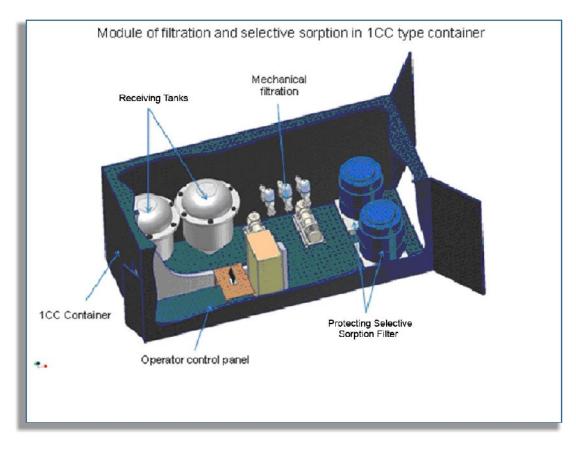


Fig.2 General layout of the radioactive waste selective decontamination module in a marine container

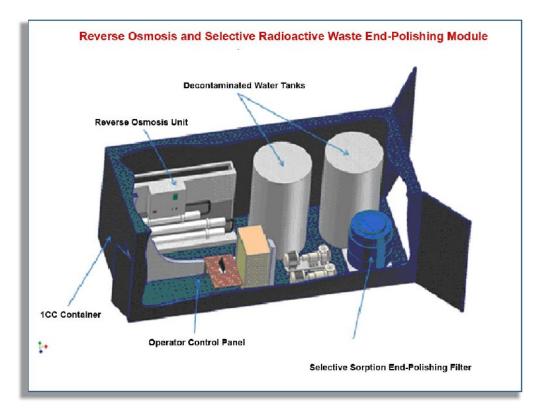


Fig.3 Basic layout of reverse osmosis desalination and sorption end-polishing module in a marine container

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Fig.4. Ejector evaporation plant module

