IV. Conclusions

The mathematical simulation procedure and computer code "Cry_En" for a single cryogenic column used for hydrogen isotope separation have been developed. The code allows to carry out the calculation in a wide range of conditions and has good convergence characteristics though needs further enhance (to make shorter a computation time).

The simulation procedure and computer code "KIO" for a multistage water/hydrogen exchange column has been developed. The static behaviour of a similar column can be studied in detail. The procedure is applicable in cases with any deuterium concentration and has good convergence characteristics.

Both steady-state procedures do not use Newton-Raphson method. This is due to the fact that the convergence characteristics of the plate separation factors are much less sensitive to the type of the iterative method than plate temperatures (or concentrations). That's why large computation efforts are not needed and computer storage requirement is small.

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TWOFOLD PURPOSE DETRITIATION PLANT

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I. Introduction

At present, two plants operate for the protium and tritium extraction from heavy water and it's concentration at the research reactor (at Laue-Langevin Institute, Grenoble, France [1]) and at the nuclear power stations (at Darlington, Canada [2]). The final purpose of this plants is the maintenance of tritium and protium concentrations on the certain level (< 70 GBq/kg of D_2O and 99.8% D_2O). The construction of similar plant was begun for the reactor PIK in PNPI.

In order to use the operating detritiation plants for the reprocessing of tritiated heavy and light water wastes, a new technology should be develop. It allows to create the detritiation plant of twofold purpose. The description of the PNPI full scale experimental base for the development of new technologies is given.

Formation and accumulation of the radioactive tritium together with other radionuclides take place during operation of the light and heavy water reactors. At present it is developed and introduced in practice the technologies of complete purification of heavy and light water coolants and radioactive wastes practically from any radionuclides except tritium. Tritium is formed in the next nuclear plants at present:

- in moderator and coolant of heavy water reactors mainly with rate [3] is equal 90–150 $GBq \cdot kg^{-1} \cdot a^{-1}$ (in moderator) and accumulation is a linearly with time in a modern power and research heavy water reactors if He-3 is not lost. An annual tritium accumulation is on the level $(5.5\text{-}8{,}8) \cdot 10^8$ $GBq \cdot a^{-1}$ or 1.5–2.4 $kg \cdot a^{-1}$;
- in the fuel of light water reactors with the rate of 3 $kg \cdot a^{-1}$. It seems likely that tritium in light water reactors is not a health hazard and for environment, as it is held by fuel cladding. Tritium concentration in the coolant is small (less 10^{-3} GBq/kg water).
- the accumulated tritium in a spent fuel releases completely in the aqueous solutions during reprocessing of the fuel, and the formation of liquid tritium wastes with concentration about 4 $GBq \cdot kg^{-1}$ occurs the rate of 5-7 ton per 1 ton of a fuel. It should be noted that tritium permissible concentration in the water removed in the environment was established equal $1.5 \cdot 10^{-4}$ $GBq \cdot kg^{-1}$ of water;
- at the reprocessing of the disarmed nuclear weapons. The deuterium-protium mixture contaminated by tritium is recovered after initial processing. The mixture can contain tritium from 10^{-3} to 10 GBq per 1 kg liquid wastes. The relationship between H and D in the wastes also can be different;
- the tritium problem will become more urgent in the case of the development of thermonuclear energy, where even in an experimental reactor (e.g. program ITER) the tritium content is evaluated as 1-2 kg. During operation of thermonuclear reactors the wastes can be produced with the different relationship of H:D:T both in a liquid and gaseous state.

As is evident from foregoing, on a global scale the rate of the tritium accumulation at present exceeds the rate of tritium formation by natural way. At present time the tritium does not produce an appreciable effect on plant and animal life in a global scale because of small value of the tritium emission into atmosphere. However in the site of tritium plants it will exist the potential danger of a large tritium emission to the environment and the contamination of the ground over authorized limits. In addition the leakage of heavy water from reactor systems and from fuel reprocessing plants both in a normal and in accident modes of operation can expose of the staff over permissible limits.

Therefore it is necessary to extract the tritium from a large complicated system and transfer it to the small size compounds for the long-term and safe storage.

II. The Lines of Attack on the Problem

The tritium contaminated water and other wastes may have a wide spectrum of isotope concentrations and correlation one with another. Reprocessing of these wastes is expensive due to a small tritium concentration with respect to content of other hydrogen isotopes as well as the small value of separation factor. The method of the catalytic isotopes exchange between the heavy water vapor and deuterium at temperature 200°C is used for the tritium and protium extraction from water and their transfer to a gas phase in the operating plants. The tritium and protium concentration are performed by the low temperature distillation of hydrogen isotopes. The extraction of protium and tritium from heavy water (70%) at these plants is done by isotopic exchange reaction between deuterium and heavy water vapors and

limited by equilibrium constants. However, it does not allow to carry out a complete (a few orders) extraction of tritium from the water. To totally purify the water, the construction of a cascade of a such devices for detritiation of the water would complicate the scheme and make it technologically impracticable and expensive. Therefore the operating plants can not be used for the reprocessing contaminated by tritium of light water and heavy water wastes without using of the new technologies.

The complete tritium extraction from the heavy water reactor coolant and tritiated wastes may be realized on the base of catalyst isotopic exchange on the wetproofed catalysts and electrolysis combined with the cryogenic distillation for the final tritium concentration. The advantage of using of the methods is more high values of separation factors in a convenient temperature interval and accessibility and ecological safety of working substances. The stable and high-effective wetproofed catalyst were developed which allow to realize the isotope exchange process in devices of the column type. The solid polymer membrane electrolyzer used to operate the recirculating hydrogen gas. There are many publications concerning to the study of a wetproofed catalytic isotope exchange among them devoted the development of the technology [4–7]. The published data showed also that many difficulties emerge when passing from bench-scale experiments to the industrial ones. Unfortunately we have not any information about the operation of industrial facilities and about investigations carried out on it.

III. Experimental Assembly of the PNPI

Therefore for the development of the tritiated aqueous wastes reprocessing technology it was decided to create in the Petersburg Nuclear Physics Institute the assembly of semi-industrial type using [7–9]:

- existing cryogenic assembly with the column for the distillation of liquid hydrogen with 300 mm diameter and circulating flow in the column up to 10 kmol/h and its auxiliary systems. The description of this assembly and results of the studies performed on it is given in Reference [8];
- the alkali and solid polymer membrane electrolyzers with capacity up to 10 m^3 H_2/h each;
- water-hydrogen isotopes exchange column of 100 mm diameter and 6,5 m height with wetproofed catalysts. Column has a few (3-5) mass exchange sections for the study of various species of packing and catalyst and various modes of its accommodation in the sections of the column as well as for the study of different design of the sections;
- catalytic burners of 4 sections with capacity of 2,5 m^3/h each;
- 4 tanks with volume of 0,75 m^3 each. The schematic diagram of assembly is presented in Fig. The technological scheme allows to connect and operate the different units of assembly as the next combinations. Each of these combinations forms a closed scheme for recovering tritium from water.
- electrolyzer–exchange column–burners–tanks for the products (CECE);
- electrolyzer–hydrogen purification system –cryogenic column–burners for the bottom and upper (and intermediate too) products–tanks for the products (E+CD);
- exchange column with the packing and wetproofed catalyst–hydrogen purification system–cryogenic column–tanks for the products (LPCE+CD);
- electrolyzer–exchange column with packing and catalyst–hydrogen purification system–cryogenic column– burners–tanks for the product (CECE+CD).

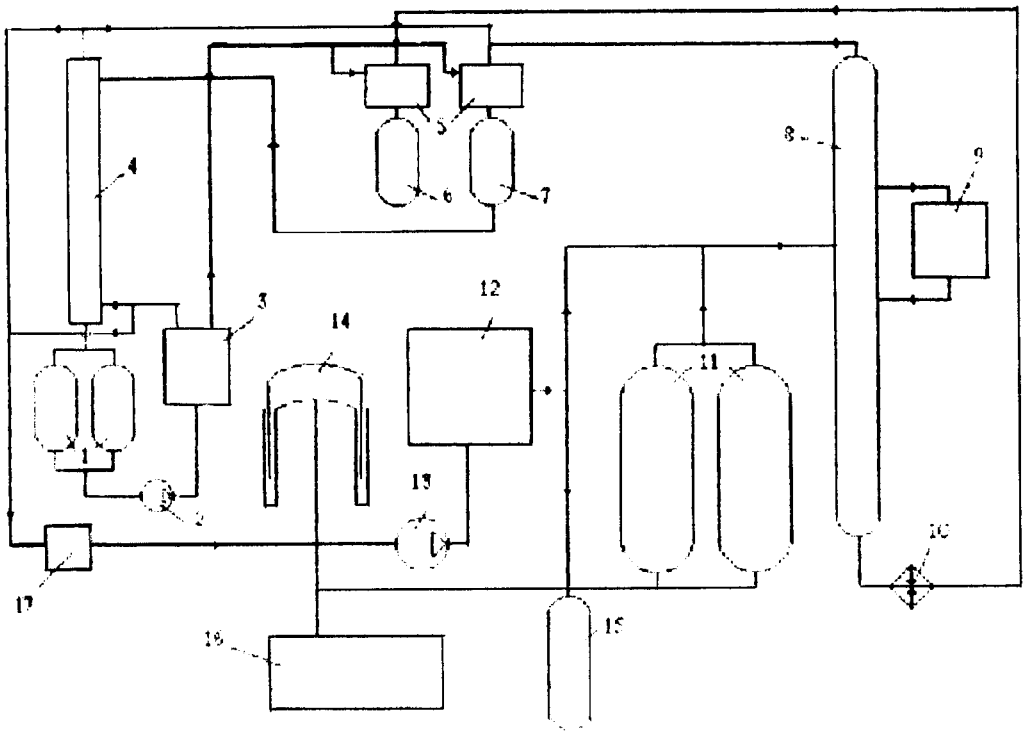

Fig. Schematic diagram of assembly. *1* – tanks for an initial product as well as for a final product; *2* – pump; *3* – electrolyzer; *4* – column for isotopes exchange; *5* – burners; *6* – tank for the bottom product of the cryogenic column; *7* – tank for the top product of the cryogenic column; *8* – unit of cryogenic separation of the hydrogen isotopes; *9* – equilibrator; *10* – liquid hydrogen evaporator; *11* – receivers; *12* – purification unit; *13* – membrane compressor; *14* – gasholder; *15* – recipient; *16* – hydrogen manifold; *17* – unit of preliminary hydrogen isotope purification

At present the equipment of CECE part is mounted and is under commissioning and testing. Testing will concentrate on removing tritium and protium from D_2O. Tritium tests will be limited to 0.01 Ci/kg.

The assembly is a flexible large-scale pilot facility with extended experimental potentialities. We hope that experiments at the assembly will enable us to obtain detail information for development of tritium wastes reprocessing plants.

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