

Technology proposal to prevent the release of Fukushima tritium into the ocean

UZI KOV Vitaly, engineer, Russia (uzikov62@mail.ru)
 UZIKOVA Irina, engineer, France (iuzikova@assystem.com)

The need for isotope separation of water molecules with protium (ordinary hydrogen), deuterium and tritium arises in various industries: obtaining heavy (D_2O) water for the nuclear industry; purification of radioactive water from tritium during the elimination of the consequences of the accident at the Fukushima NPP and heavy water at nuclear reactors; reduction of the natural concentration of heavy hydrogen isotopes in water for biological and medical purposes, etc. But first of all, our proposals are an attempt to propose a technology that could prevent the discharge of water contaminated with tritium into the world ocean after the accident at the Fukushima nuclear power plant.

Isotope separation technologies

Various methods of separating water from hydrogen isotopes exist nowadays:

- isotopic exchange in the presence of palladium and platinum,
- water electrolysis in combination with catalytic isotopic exchange between water and hydrogen,
- column rectification,
- vacuum freezing of cold vapor followed by thawing, and others are used.

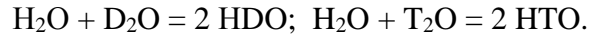
The existing technologies for the hydrogen isotopes separation have a common feature - *they are all extremely energy-intensive* (Table 1) and *require the use of expensive equipment*.

Table 1 – Used technologies for separation of hydrogen isotopes [1]

Method	Operation temperature, K	Split ratio		Energy costs, MW · h / kg D_2O	Application for:
		α_{HT}	α_{HT}		
Water electrolysis	313-333	1.5-10	2-15	120-150	D_2O production
Rectification					
H_2 * rectification	23	1.47	1.90	4-5	D_2O production Intermediate and final concentration of tritium
NH_3 rectification	253	1.036	-	8	D_2O production
H_2O rectification	333	1.047	1.055	40	D_2O production Removal of protium from heavy water coolant and moderator
Chemical isotope exchange					
H_2S - H_2O system (two-temperature)	303 / 403	2.34 / 1.82	3.34 / 2.36	2.8	D_2O production
NH_3 - H_2 * system (two-temperature)	248 / 338	5.16 / 2.87	-	1.0	D_2O production
CH_3NH_2 - H_2 * System (two-temperature)	248 / 333	6.12 / 3.12	-	1.0	D_2O production
H_2O - H_2 * system (single temperature)	328	3.22	5.40	65-70	Waste processing Water detritisation

* – systems requiring the use of hydrogen activation catalysts

When choosing a technology, it is logical to consider, the methods and processes of isotope separation, in which the behavior of molecules of ordinary (protium), deuterium and tritium water has the maximum difference. In addition, the energy for these processes should be minimal. **The phase transition when water freezes** is well suited to these requirements. However, it should be noted that in a mixture of "light" (H_2O) and heavy ($\text{D}_2\text{O} + \text{T}_2\text{O}$) water, isotope exchange occurs:



Therefore, deuterium and tritium are usually found in water in the form of HDO and HTO.

In this case, the freezing point for D_2O is $+3.8^\circ\text{C}$, and for T_2O $+9^\circ\text{C}$, HDO and HTO freeze at $+1.9^\circ\text{C}$ and at $+4.5^\circ\text{C}$, respectively. It has been established that, for example, at temperatures ranging from 0 to $+1.9^\circ\text{C}$, water molecules with deuterium and tritium, in contrast to "light" (protium) water, are in a metastable-solid inactive state [2]. The water temperature and the pressure of saturated water vapor are interrelated and the difference in the isotopic composition of hydrogen atoms in water molecules leads to a noticeable change in the temperature dependence of the saturated vapor pressure (Fig. 1). Taking into account the significant change in the freezing point of water for molecules with different isotopic composition of hydrogen atoms and the corresponding difference in the saturated vapor pressure, it is possible to use the differing change in the properties of water with a decrease in temperature in the range from 0 to 9°C for isotope separation.

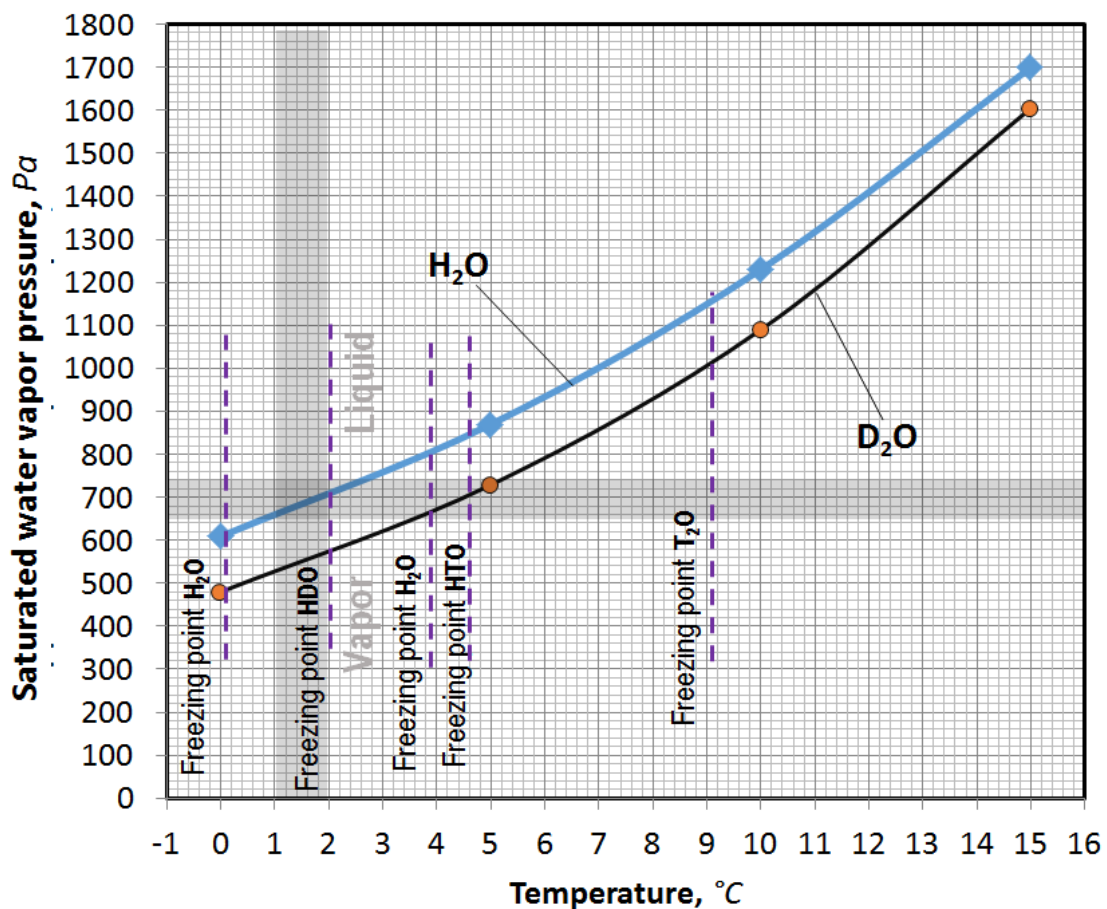


Fig. 1 - Parameters of phase transitions of boiling and freezing for heavy and light water at low temperatures and pressures

This feature underlies the fractional separation of "light" and heavy water by creating a deep vacuum above the water surface at low temperatures. "Light" water evaporates intensively and then condenses. "Heavy" water, being in an inactive state and having a much lower partial pressure, remains mainly in the evaporation tank in the source water.

This principle is used in the industrial installation VIN-4 "Nadia" for the production of "light" water with a 30-35% reduced content of heavy isotopes of deuterium and tritium (Fig. 2) [2] constructed by G.D. Berdyshev and I.N. Varnavsky, together with the R. Kavetsky Institute of Experimental Pathology, Oncology and Radiobiology of the Russian Academy of Sciences of Ukraine.

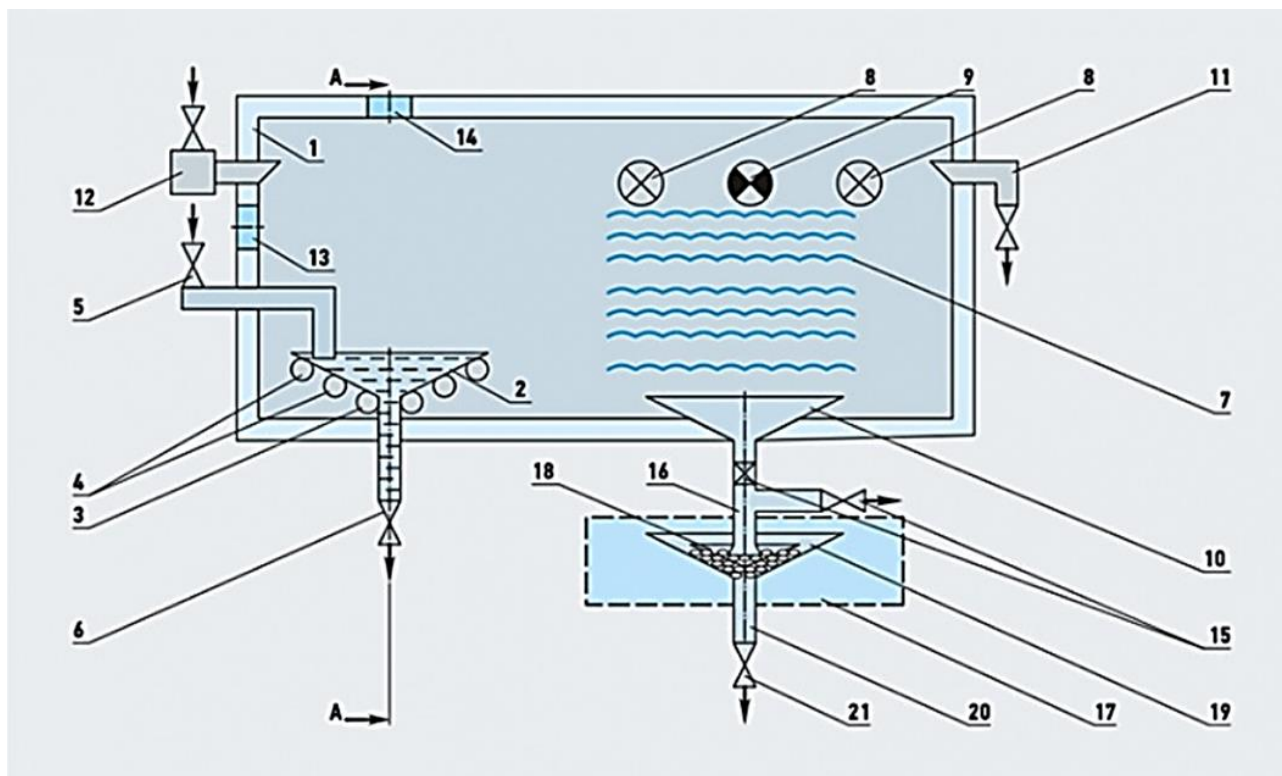


Fig. 2. Schematic representation of the VIN-4 "Nadia" installation: 1 - the installation housing; 2 - evaporation capacity; 3 - device for heating water; 4 - device for cooling water; 5 - valve for water supply to the evaporator; 6 - valve for draining the spent residue; 7 - device for condensation and freezing of cold steam; 8 and 9 - sources of ultraviolet and infrared radiation; 10 - tank for collecting melt water; 11 - vacuum pump; 12 - device for supplying purified air or a mixture of gases; 13 and 14 - portholes for observing the processes of evaporation and freezing of steam and melting of ice; 15 - valve for melting water discharge; 16 - branch pipe; 17 - block for forming the structure and properties of melt water; 18 - inner conical container with minerals; 19 - outer conical container with minerals; 20 - adsorption filter; 21 - drain valve

The installation consists of a housing 1, in which an evaporating tank 2 for source water is installed with heating devices 3 and cooling water 4. There is also a valve 5 for supplying water to the evaporator and a valve 6 for draining the spent residue enriched in heavy hydrogen isotopes. The housing also contains a device 7 for condensing and freezing cold vapor in the form of a set of thin-walled tubular elements that are connected to a pump for pumping refrigerant through them. The device 7 together with the sources of ultraviolet 8 and infrared 9 radiation is placed above the tank

10 for collecting melt water. The inner cavity of the housing 1 is connected to the pipe 11 with a vacuum pump for the air discharge. In addition, the housing 1 is equipped with a device 12 for supplying purified air or a mixture of gases into the internal cavity of the installation. Additionally, the VIN-4 “Nadia” unit is equipped with a thermal control system in the cavity of the evaporation tank 2 to control the set temperature of the evaporation process of the initial treated water. The housing contains windows 13 and 14 for observing the evaporation processes, freezing of cold vapor and melting of ice. The melt water collection tank 10 is equipped with valves 15 for draining the melt water and a branch pipe 16 for connection with the block for forming the structure and properties of melt water 17. Block 17 includes an internal conical container 18 with minerals. An adsorption filter 20 and a drain valve 21 are installed at the outlet of the tank 19.

The installation VIN-4 “Nadia” operates as follows. From the water supply, the evaporation tank 2 is filled with water and the refrigerant is pumped through the device 4. When the temperature threshold is reached (less than +10 °C), the water cooling process is stopped. Then the housing 1 is sealed and air is pumped out through the nozzle 11, creating a vacuum in the internal volume of the installation housing. The creation of a vacuum is accompanied first by an intensive release of gases dissolved in it from the entire volume of the initial water and their removal, and then by intensive vaporization until the water boils, which is observed through windows 13 and 14. The resulting cold vapor condenses and freezes on the surface of the tubular elements of the freezer 7. When the ice reaches a predetermined thickness, the evaporation process is stopped. The vacuum pump is turned off, include sources of ultraviolet 8 and infrared 9 radiation, and through the device 12 is introduced into the cavity of the housing 1 purified air or a mixture of gases; then the pressure in the housing 1 is brought to the atmospheric level or higher. The remainder of the water in the tank 2, enriched in heavy isotopes, is poured through the valve 6 into separate containers or poured into a storage device. As the ice is irradiated and melted, the melt water enters the container 10, then into the block for the formation of the structure and properties of melt water 17. Passing through the minerals of the inner 18 and outer 19 conical containers and then through the filter 20, the melt water completes its journey, acquiring medicinal biologically active properties.

Most likely the announced large decrease in heavy water isotopes (30-35%) is not achieved in this installation after one evaporation-condensation cycle, but a good effect of this method is also confirmed by works on glaciology (the science of natural ice).

Interfacial fractionation of water by hydrogen isotopes at low temperatures

In isotopic geochemistry, it is usual to express the isotopic composition not in terms of the absolute concentration of heavy isotopes in the sample under study, but as relation to the concentration of these isotopes in the "isotope standard" - the sample of the studied substance taken as a standard. When studying the isotope composition of water, the main isotope standard is VSMOW-2 - Vienna Standard Mean Ocean Water, prepared, as the name suggests, from seawater. The VSMOW-2 standard is characterized by the following isotope composition values:

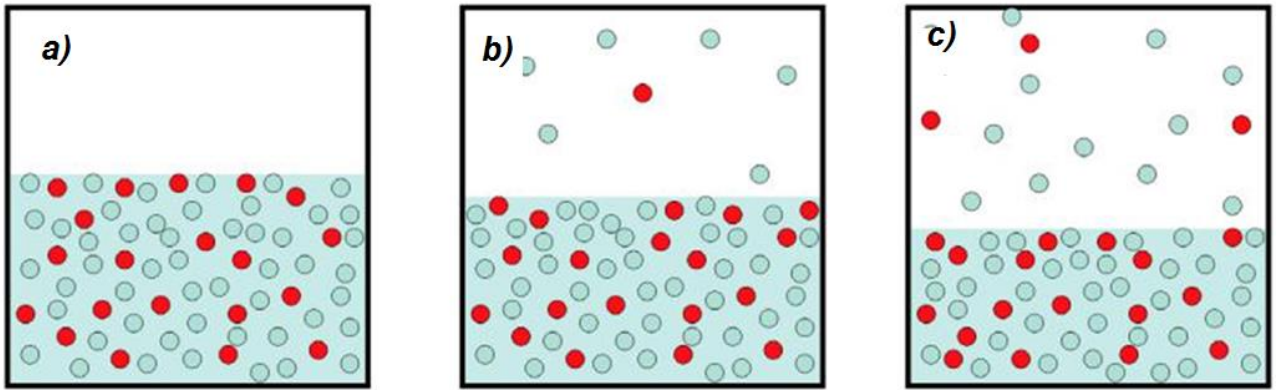
- *the concentration of ^2H is 155.76 ± 0.1 ppm;*
- *the concentration of ^{18}O is 2005.20 ± 0.43 ppm;*
- *the ^{17}O concentration is 379.9 ± 1.6 ppm.*

In order to express the isotope composition of a sample relative to VSMOW-2, the following formula is used [3]:

$$\delta = \frac{R_{\text{sam}} - R_{\text{st}}}{R_{\text{st}}} \cdot 1000, \quad (1)$$

where δ – δD , $\delta^{18}\text{O}$ or $\delta^{17}\text{O}$, respectively, R_{sam} and R_{st} are the absolute concentration of deuterium, oxygen-18 or oxygen-17 in the sample and the VSMOW-2 standard, respectively. Multiplying by 1000 means that the isotopic composition is expressed in ppm.

Due to the small difference in mass, isotopes behavior is slightly different during phase transitions (evaporation - condensation, melting - freezing) and some other physical processes. This leads to the so-called fractionation, i.e. separation of heavy and light isotopes. If such a division did not exist, the isotope composition of water in the entire hydrosphere of the Earth would be the same. In work [3] the fractionation of isotopes is considered on the example of a simple closed system consisting of water and steam (Fig. 3).



Blue and red points denote light and heavy molecules respectively... Ratios between light and heavy molecules don't correspond to the reality and are given only as example

Fig. 3 – Isotopes fractionation in the water evaporation process [3]

a - the system before the evaporation process, b - the initial moment of the evaporation process, c - the system is in equilibrium (water vapor has reached the saturation line).

At the beginning of the experiment, there is a certain volume of water with an isotope composition R_w , where R is the absolute concentration of heavy isotopes, as in formula (1) (Fig. 3 a). At this point, the air above the liquid is completely dry. Immediately after the beginning of the evaporation process, the first portion of water vapor turns out to be depleted in heavy isotopes in comparison with water (Fig 3b). This is due to the difference in the movement speed of heavy and light molecules. Indeed, the kinetic energy of molecules is proportional to their mass and speed. Since the average kinetic energy of the molecules is the same, heavy molecules move slower than light ones. It is easier for light molecules to escape from the liquid and go into the gaseous phase. In addition, it is important that lighter molecules diffuse more easily through the air, moving away from the evaporation surface (i.e. have a higher diffusion coefficient). Since the system is closed and the

available volume is limited, over time, heavy molecules will "catch up" with light ones, and the concentration of heavy molecules in water vapor slightly increases.

At some point, water vapor reaches saturation under these conditions (Fig. 3 c). This means that the number of molecules leaving the liquid becomes equal to the number of molecules returning from the water vapor to the liquid. Heavy molecules reach saturation at a lower concentration in the air than light molecules, since it is more difficult for them to leave the liquid and it is easier for them to return to it. Since the concentration of water molecules determines the pressure of water vapor (along with temperature), it can also be said that the pressure of saturated water vapor, consisting of heavy water molecules, is less than the pressure of saturated water vapor, consisting of light molecules, at a given temperature (Fig.1).

Thus, under conditions of thermodynamic equilibrium between water and water vapor located above its surface, the concentration of heavy isotopes in water vapor will be less than their concentration in water:

$$R_V < R_L$$

The relationship between the two is called the fractionation factor and is usually denoted by the Greek letter α :

$$\alpha = R_L / R_V , \quad (2)$$

The term "fractionation factor" is used to characterize the change in the isotopic composition during any phase transitions, for example, water - water vapor, water - ice, vapor - ice. Therefore, in order to clarify what kind of fractionation factor we are talking about, the corresponding indices are added to the symbol ($w-v$, $w-i$, $v-i$ etc.). If we are talking about fractionation between water and water vapor, which are in equilibrium (ie, water vapor is in a state of saturation at a given temperature), we speak of the "equilibrium fractionation factor".

In nature, evaporation rarely occurs under equilibrium conditions, since the relative humidity of the air is usually significantly less than 100% (i.e. water vapor is far from saturation). If the system was closed, then sooner or later equilibrium would come, but in real conditions this does not happen. In this case, the situation depicted in Fig. 3 b, when water vapor is more depleted in heavy isotopes than it would be under equilibrium conditions, i.e. the effective fractionation factor is greater than the equilibrium one. Such additional fractionation, associated with a higher rate of the process (in this case, evaporation) for light molecules as compared to heavy ones, is called "kinetic fractionation" [3].

In Fig. 4 the values of the fractionation factors are shown in the graphics [3]. First of all, attention is drawn to the fact that the fractionation coefficients at a given temperature are significantly higher for deuterium than for ^{18}O . For example, at a temperature of 0 °C, the fractionation coefficient in the water - steam system is 1.012 for ^{18}O and 1.11 for deuterium. This means that water vapor in equilibrium with water at a temperature of 0 °C will be depleted in ^{18}O by 12 ‰ (ppm), and in deuterium by 100 ‰. Due to this, for the same sample, the δD values are, as a rule, an order of magnitude greater in absolute value than the $\delta^{18}\text{O}$ values.

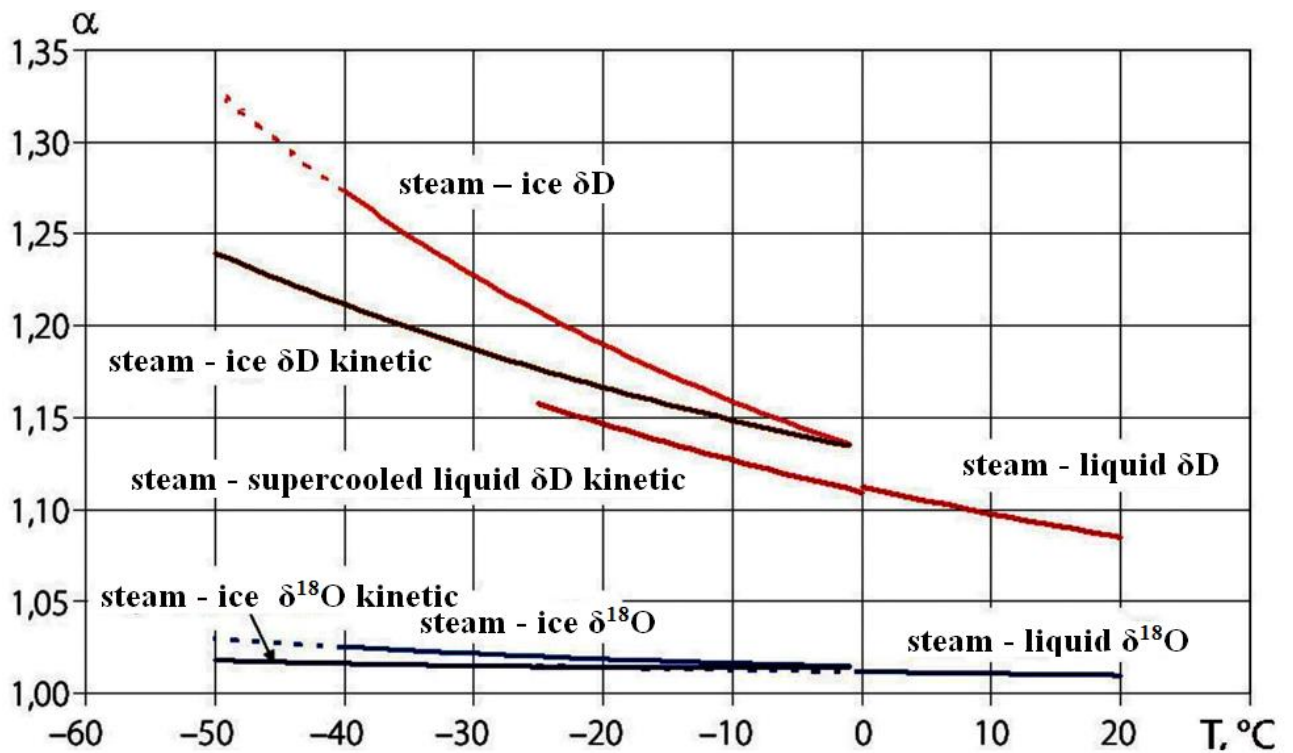


Fig. 4. Dependence of the fractionation coefficients (α) on temperature (T):
curves for *steam - water* and *steam - ice* systems for oxygen-18 and deuterium;
Solid curves - experimentally determined values,
dashed - extrapolated values

Vacuum isotope distillation

The proposed technology of isotope separation is based on the fact that air is completely removed from the system and the pressure in it is reduced to the level of partial pressure of water vapor ($\sim 600..700$ Pa) at a temperature of $1..2^{\circ}\text{C}$, if it is necessary to concentrate deuterium and tritium, obtaining an enriched against water. The difference between the proposed technology consists in organizing a continuous process of evaporation of predominantly water with light hydrogen isotopes and concentrating the heavy water fraction on the wetted surface of the nozzles. As nozzles, a good choice would be, for example, the spiral-prismatic Silivanenko nozzles (SPN) or the regular Panchenkov nozzle (Fig. 5).



Fig. 5 – Spiral-prismatic Silivanenko packing nozzles (top) and regular Panchenkov packing nozzles (bottom)

I.L. Selivanenko patented "A device for obtaining water with a reduced content of heavy molecules" (RU 2612667 C1), which refers to rectification devices for purifying water from impurities in the form of water molecules containing heavy isotopes of hydrogen and oxygen.

The innovative solution for isotope separation, is described below.

The proposed instalation includes a distillation column of 6 m high, operating at a temperature of 60 °C and under a vacuum of 20 kPa, an evaporator, a condenser and a freon heat pump with a 48 kW compressor. The process of enrichment of water vapor with the lightest water molecules occurs in a rectification column on the surface of the packing. The output of condensed light water with a deuterium content of 10 ppm is 6 l/h, which is a very good indicator.

The proposed technology for the water isotopes separation is aimed at changing the method of the nozzles irrigation in the rectification column from the top of the filling for the method of the periodic wetting of the nozzles placed in rotating structures when they are immersed in water located in the installation housing (Fig. 6). In this case, the water molecules with heavy isotopes accumulated in the film nozzles diffuse into the water located in the lower part of the installation housing, which leads to its enrichment with heavy isotopes. In fact, the presented rectification plant with annular packed structures is similar in functional use with rectification columns in horizontal state and with modified systems for wetting and washing the packing with cold steam at complete absence of non-condensable gases.

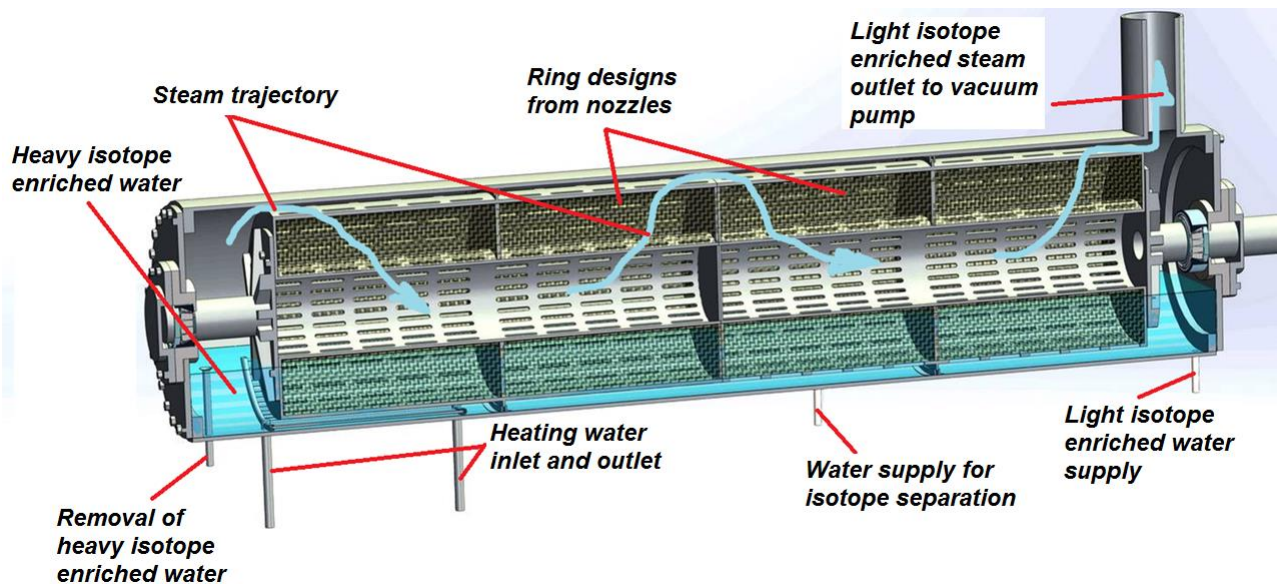


Fig. 6 - Horizontal rectification plant with annular packing structures

This design of the packing device completely removes the problem of limiting the flow surface area of the packaging fillings, and, consequently, of limiting the device's productivity. In addition, all requirements for the height of the rectification columns are removed.

The technical results that can be achieved using the proposed technology are an increase in the efficiency of isotope separation of water using a rectification plant with rotating annular packing structures during vacuum distillation and a decrease in energy costs per unit of the finished product.

The proposed method for the isotope separation of water molecules is based on the technology of vacuum distillation, which provides for the mass exchange of water molecules with light and heavy hydrogen isotopes on the wetted surface of the packing devices when steam passes through them. This method is characterized by the fact that the process is carried out at a temperature not exceeding the freezing point of water with molecules of the target heavy hydrogen isotopes. For example, to enrich the liquid phase of water with deuterium, the process temperature should not exceed 1.9 °C (Fig. 1). In this case, the required temperature of the mass transfer process on the packing devices is maintained by evaporative cooling of water under the conditions of vacuumization of the isotope separation unit by vacuum pumps and the simultaneous supply of thermal energy to the liquid to maintain steady state conditions.

The considered isotope separation technology is based on the fact that the system is evacuated, ensuring the removal of all non-condensable gases and the pressure in it is reduced to the level of the partial pressure of water vapor ($\sim 500..800 \text{ Pa}$). At a given vacuum, the corresponding water temperature drops to a level lower by 0.2 ... 1.5 °C than the freezing temperature of water with the target hydrogen isotope. Evaporation of steam by a vacuum pump is accompanied by evaporative cooling of water, and the resulting steam passes through the layers of nozzles located in the sections of annular packed vacuum structures, providing mass transfer between the vapor and a liquid film on the surface of the nozzles, while enriching the films with water molecules with heavy hydrogen isotopes (Fig.7).

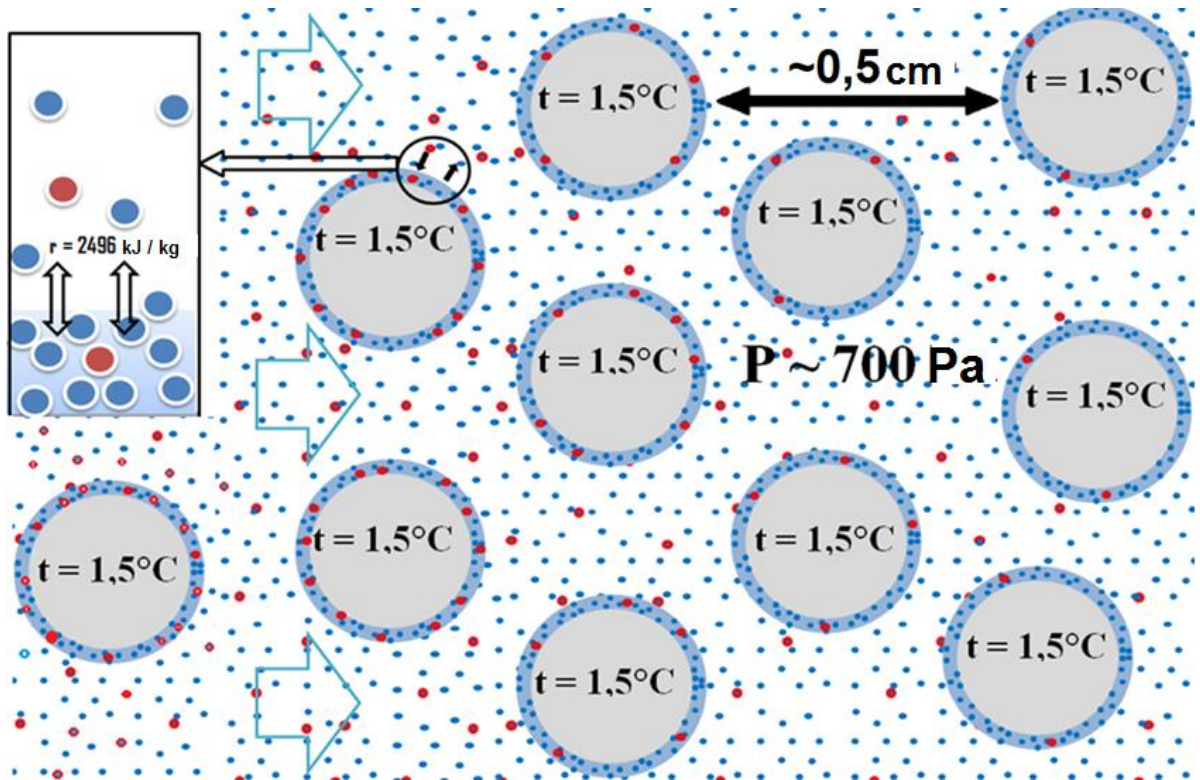


Fig. 7. Scheme of enrichment of water films on the surface of packings during the steam pass by molecules with heavy hydrogen isotopes (Blue and red points represent light and heavy molecules, respectively. The relationship between the number of light and heavy molecules does not correspond to reality and is given for example only)

When the specified parameters of water enrichment with heavy isotopes are reached, the water is replaced in a periodic or continuous mode. The removal of steam by a vacuum pump is accompanied by intensive evaporative cooling, which is compensated by a water heater in the lower part of the apparatus (Fig.6).

The fact that evaporative cooling during evacuation can freeze water is easy to see by looking, for example, this video

https://www.youtube.com/watch?v=3EFrGEXQFz0&t=2s&ab_channel=HarvardNaturalSciencesLectureDemonstrations (Fig.8). To prevent complete freezing of water and to maintain the temperature at a given level, it is necessary to compensate the pumping out of heat with steam by supplying thermal energy and the replenishing the entrained moisture with supplying water.

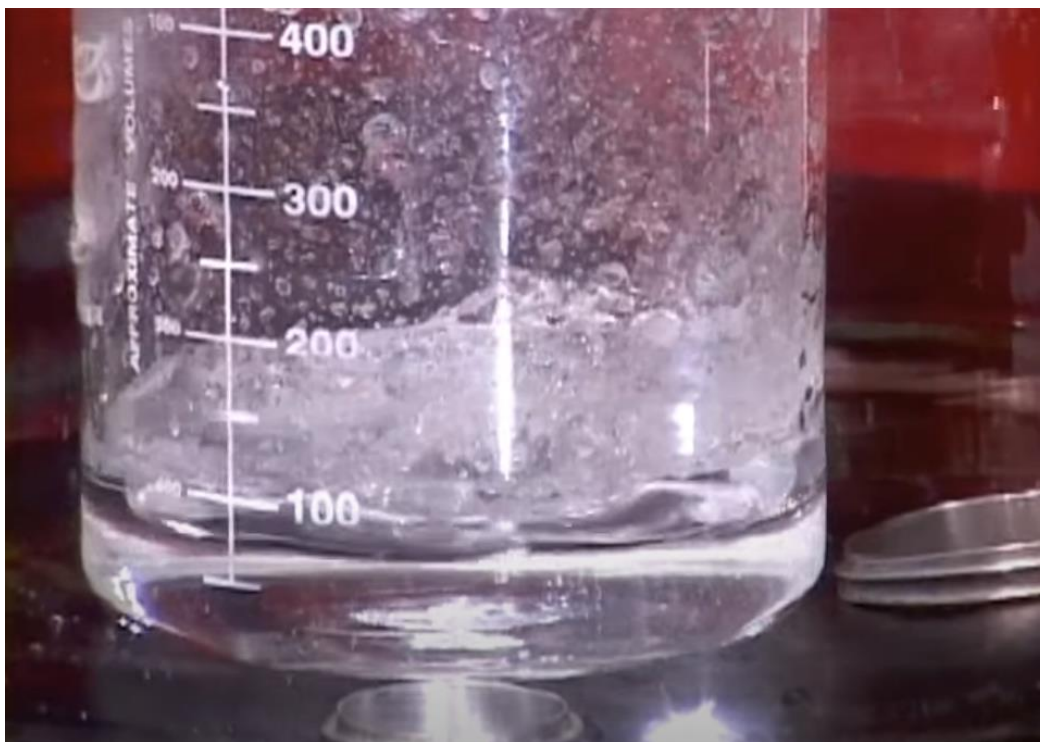


Fig. 8 – Freezing water during vacuum creation

(https://www.youtube.com/watch?v=3EfrGEXQFz0&t=2s&ab_channel=HarvardNaturalSciencesLectureDemonstrations)

In contrast to the VIN-4 «Надія» installation, evaporation is carried out not only from the surface of the water mirror, but also from the developed surface of the nozzles in annular nozzles, which are rigidly connected to the rotating shaft, therefore, the intensity of the isotope separation process shall be significantly higher than in this installation. The required water temperature in the evaporator depends on the concentration of heavy isotopes and increases with increasing concentration so that the water does not turn into ice.

The proposed technology for the separation of water isotopes, as well as the installation according to the patent ***RU 2612667 C1***, is based on the use of a heat pump, however, the role of the refrigerant in the system is played not by freon, but by water itself. All the heat spent on evaporation is generated with excess during the operation of vacuum pumps and heating of the heat carrier when compressed steam condenses on it in the scrubber, and the excess of this heat is removed through the heat exchanger (Fig. 9). As an example of a specific implementation, an installation for the isotope separation of water molecules is shown, with a capacity of 48 kg/h for water enriched in a light isotope (protium). The parameters of a horizontal rectification plant with rotating annular packing structures wetted with water in the lower part of the body are shown in Table 2.

The operating parameters of the facility near the freezing phase transition of water with heavy isotopes allow expecting a sharp decrease in the mobility of these molecules during mass transfer between vapor and a liquid film on the surface of the packings and an increased rate of enrichment of films on the packings with heavy water isotopes. Although, the experiments should be executed to justify this point.

Table 2 - Parameters of a horizontal rectification plant with annular packing structures

Parameter	Value	Units
Body dimensions (Diameter × Length)	2.1 × 6.5	m
Pressure	650	Pa
Type of nozzles SPN 4 × 4 × 0.2 mm		
Inner diameter of packing structure	1.0	m
Outside diameter of packing structure	1.8	m
Total length of packing structure	6	m
Backfill volume	10.5	m ³
Backfill specific surface	2700	m ² / m ³
Backfill density	220	kg / m ³
Surface of nozzles	28500	m ²
Steam discharge rate	13	g / s
Vapor density	0.0055	kg / m ³
Steam volumetric flow	2.42	m ³ / s
Backfill weight	2322	Kg
Vaporization heat	2496	kJ / kg
Heat input	33.3	kw

The simplified process diagram shown in Fig. 9 shows the operation of a heat pump during evaporation, compression and condensation of steam on droplets of circulating water sprayed in a scrubber. At the same time, the water heats up, and then transfers heat for water evaporation during passing through a horizontal rectification unit. The supply of source water, as well as the removal of water enriched in heavy and light hydrogen isotope, is carried out according to a scheme similar to that for rectification plants.

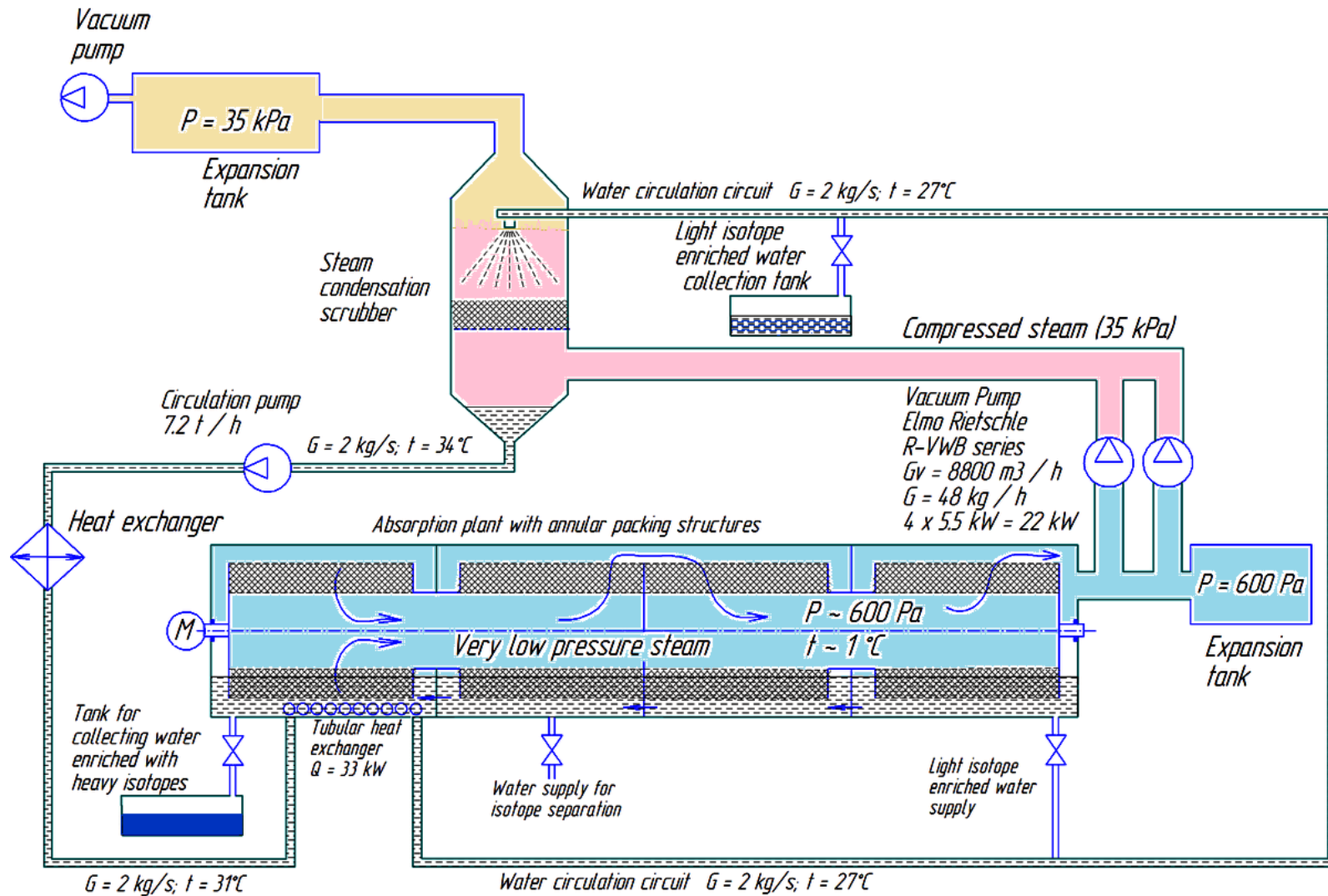
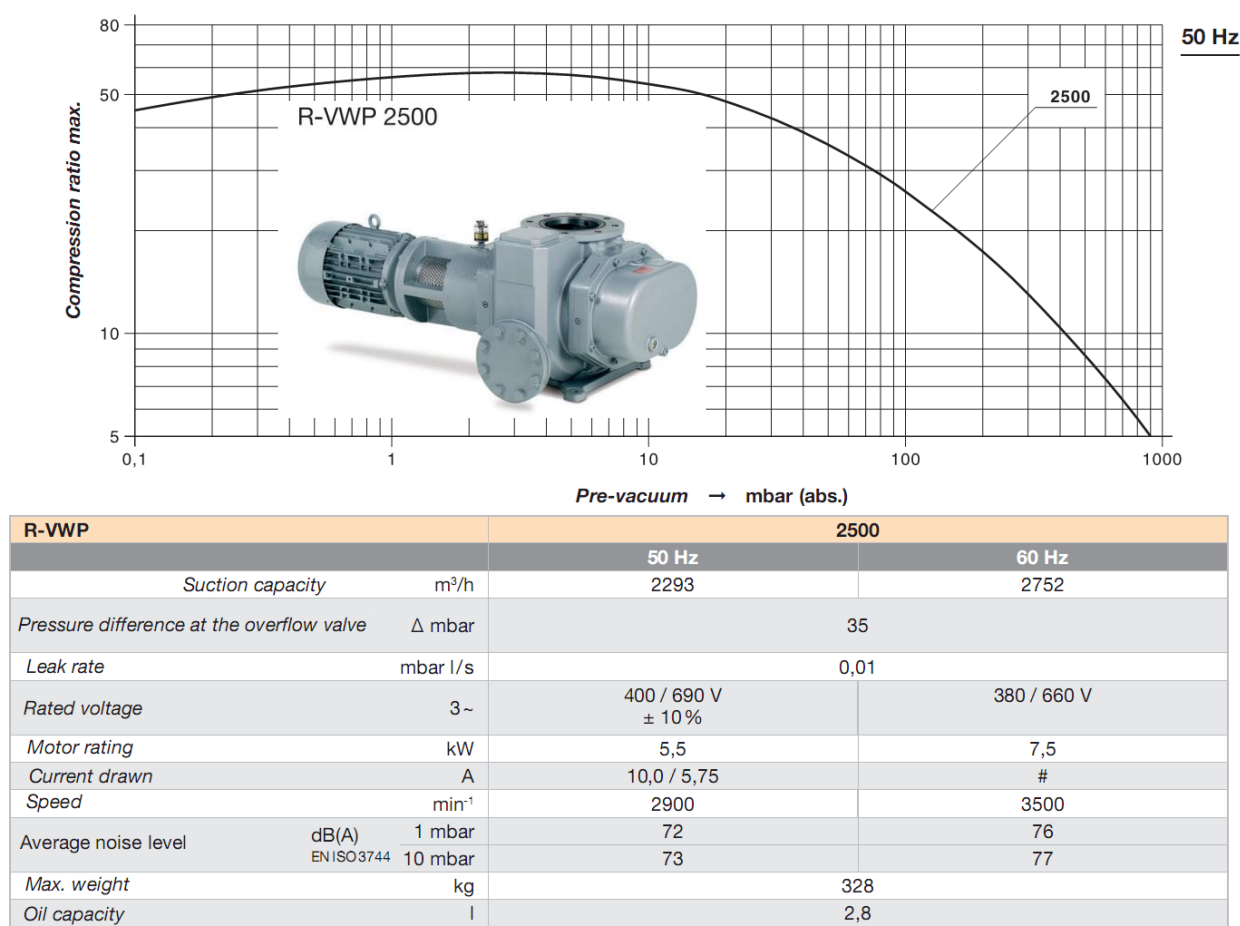


Fig. 9 – Technological scheme for the separation of water molecules by hydrogen isotopes using a horizontal rectification plant with annular packed structures.

The creation of the required conditions for mass transfer by pressure is ensured by the use of 4 Elmo Rietschle R-VWB series cam vacuum pumps, providing a volumetric steam flow $G_v = 8800 \text{ m}^3/\text{h}$ ($G_m = 48 \text{ kg/h}$) with a total power of 22 kW , the parameters of which are given below (Table 3).

Table 3 - Parameters of Elmo Rietschle vacuum pumps of the R-VWB series



To ensure the normal operation of the Elmo Rietschle vacuum pumps, the scrubber is maintained at a reduced pressure of 35 kPa using another small vacuum pump that removes non-condensable gases from the system.

The physical model of the columnar crystallization of ice and the scheme of the installation for ice separation ($\text{HDO-D}_2\text{O}$ in H_2O in the selective mode) are given in [4]. Deuterium is transported in the column by diffusion in the liquid phase and mass transfer from the solid to the liquid phase [5] as a result of successive processes of melting and ice formation. In view of the higher temperature of ice formation of deuterium water in comparison with the temperature of ice formation of protium water, during melting, first of all, protium molecules pass into the liquid; when ice forms, deuterium molecules pass into its structure first of all. In a horizontal vacuum rectification unit, in the mode of ice formation on the surface of the packings, this will lead to the deposition of predominantly deuterium molecules on the packings and to an increased entrainment of protium water molecules with the vapor.

The operating temperature parameters of the unit can vary from ice-free conditions to the beginning of ice formation on the surface of the nozzles. In this case, it is possible to use the periodic change of these modes by changing the power of water heating in the rectification unit, thereby increasing the pressure and saturation temperature.

Possible fields of application of vacuum separation of water molecules by hydrogen isotopes

Obtaining drinking water with a reduced deuterium content

Many recent studies have shown that light water with a low deuterium content is a good biostimulant that increases the functionality of the body at the cellular level and has healing properties. According to some statements, even not deep purification (about 5-10%) of water from deuterium can significantly improve it, giving water immuno stimulating and rejuvenating properties, not to mention deep water purification. It has been reported that the pigs, rats and mice who consumed water, which contained 25% deuterium below the norm, gave birth to offspring much larger than usual, and the chickens' eggs quantity has doubled. Wheat watered with light water ripens earlier and yields higher yields.

However, it should be recognized that many authors of articles on medicinal or beneficial properties are directly related to companies producing light water, and water of their trade marks was often used as a test sample, therefore, such studies are sometimes difficult to classify as independent.

Heavy water production

There are several applications for heavy water.

1. **Nuclear power.** There are nuclear reactors in which heavy water is used as a coolant (for example, the Canadian CANDU reactor, Fig. 10). Its use allows the use of natural uranium of a lower enrichment degree. Heavy water can also be used as the most efficient and technologically advanced reflector of the cores of research reactors such as ORPHEE (France), HANARO (South Korea), OPAL (Australia), FRM-II (Germany).

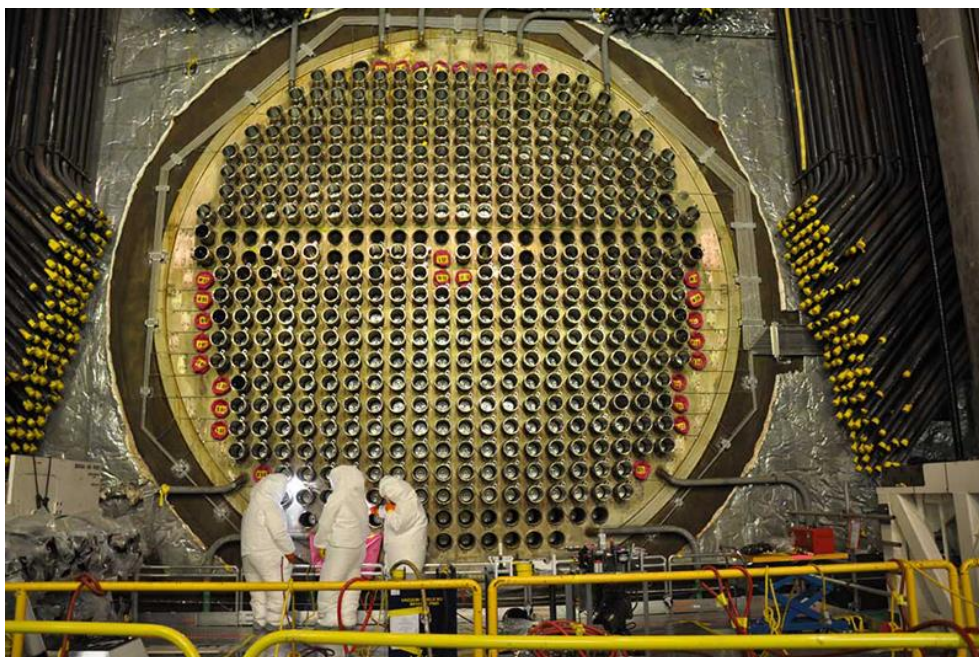


Figure 10 - View of the core of the Canadian CANDU reactor

2. **In laboratories.** As an indicator and solvent in physical and chemical analysis, in studies of biological objects, as well as of living organisms, including humans.
3. **In medicine.** In addition to human laboratory research, heavy water can be used to treat fungi and bacteria, hypertension, etc.

Tritium removal from water

Unlike deuterium, tritium is a particularly dangerous isotope for the human body (β emitter of energy 18.6 keV with a half-life of 12.32 years), since it is easily incorporated into the cellular structure and is a source of internal radiation. In nature, tritium is formed in the upper atmosphere when cosmic ray particles collide with atomic nuclei, such as nitrogen, but nuclear reactors have also become an important source of tritium accumulation in the environment, especially in nuclear disasters such as Fukushima. In addition, during operation of reactors with heavy water, tritium produces and accumulates in it. The heavy water must be purified from it.

For this purpose, water purification in rectification columns is used (the simplest and most energy-intensive method).

Another method of purification is combined with electrolysis isotopic exchange of **water - hydrogen** on a catalyst. However, due to the use of electrolysis, energy consumption is very significant. Also, for these purposes, two-temperature isotope exchange is used in the systems **water - hydrogen** or **water - hydrogen sulfide**.

From the point of view of the economic acceptability of the technology for purification from tritium, due to the relatively short half-life of tritium (12.32 years) with large volumes of processing - more than a million tons (as in Fukushima), it is cheaper to withstand tritium until the main mass decays than to use too expensive technologies.

Therefore, Fukushima requires an efficient and low-cost cleaning technology, and perhaps the proposed technology will be in demand.

There are not so many operating industrial installations for purification of tritium. The largest plant is for the treatment of heavy water at the CANDU reactors in Canada (CIRCE, 300-500 kg/h). Smaller units exist in South Korea (Wolsong tritium removal facility, 100 kg/h) and Romania (ICSI-TRF, 4-8 kg/h).

The economics of the process is determined by two main criteria - *the productivity* of the process and *the degree of concentration*. In addition, in the economic assessment of technology, the most important characteristics to be considered are *capital investments* and *operating costs*. The comparison of the rectification treatment and combined treatment with electrolysis shows that in one case the main problem is capital investment, and in the other case it is operating cost.

The specialists of *RosRAO* and the *Radium Institute* developed their own three-stage process, where they tried to combine the advantages of known technologies from tritium, and which they proposed to solve the problem of tritium waters in Fukushima. This process should be tested in 2017 at a demonstration facility for detritisation of liquid radioactive waste and trial cleaning with a 48 cubic meter of model solution in Sosnovy Bor (Fig. 8). The heating steam consumption in this unit is 6.5 t/h (thermal power $Q = 4434$ kW) at a design capacity of 197 kg/h for treated water.

Follow the link <https://youtu.be/9PrIfnpkxgY> to watch a video with a detailed description of the operation of this installation.



Fig. 10 - Demonstration plant with a rectification column for detritisation of liquid radioactive waste at the Fukushima NPP (Radium Institute, Sosnovy Bor)

Unfortunately, the data on the results of the tests carried out for this installation and its effectiveness are not available.

Conclusion

The proposed innovative technology of tritium separation can allow to avoid the discharge of Fukushima tritium water in ocean.

The advantages of the considered solution are the following:

- The proposed technology for isotope separation during vacuum rectification near the water freezing point of water is characterized by low capital investments and operating costs;
- The operation of the isotope separation unit does not require a heat source, but only a heat exchanger to remove the heat excess;
- The main sources of energy consumption in the proposed technology are conventional industrial pumping equipment for creating vacuum and water circulation;
- The dimensions of the equipment allow it to be placed in almost any production room;
- The capacity of the plant for water enriched with a light hydrogen isotope is not inferior to the best industrial plants.
- The proposed technology can be effectively used for the treatment of tritium water at Fukushima due to low capital investments and low operating costs.

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