Detritiation of the Electrostatic Spectrometer of Troitsk Nu-mass Experiment

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Abstract — This paper describes the methods and presents the results of the “Troitsk Nu-mass” experiment spectrometer cleanup after the inner volume (40 m³) and surfaces (160 m²) were contaminated by 5.2 GBq of tritium. The Troitsk Nu-mass experiment of the Institute for Nuclear Research of the Russian Academy of Sciences (Moscow) is designed to measure the spectrum of electrons from tritium decays in order to search for hypothetical particles—sterile neutrinos. Due to some equipment failures, the spectrometer internal volume was contaminated with tritium. The contamination made measurements impossible, and the research program stopped. Different methods were used for cleanup: vacuum extraction, hydrogen soaks, and water vapor soaks. As a result of detritiation, the background level of the main detector of the Troitsk Nu-mass spectrometer was reduced approximately by more than ten times, which made it possible to resume work. The results are consistent with the literature data obtained earlier for normal conditions in the air and can be used for detritiation of similar installations.

Keywords — Tritium contamination, Troitsk Nu-mass setup, tritiated water, detritiation, tritium removal.

Note — Some figures may be in color only in the electronic version.

I. INTRODUCTION

At present, a large amount of data have been accumulated on the detritiation of various materials and substances used in tritium handling.1–7 However, the detritiation process is still a complicated, resource- and cost-expensive procedure in view of the tritium removal process specifics and the requirements of low levels of residual tritium in materials.8 The investigation of tritium retention in materials and their detritiation has widely been carried out in different laboratories.1–4,9,10 But the approaches that can be applied to the detritiation of large facilities with a relatively low concentration of tritium in materials have not been experimentally verified enough. So, any experimental results on tritium removal at large scientific facilities are of interest for the operation of fusion reactors, mainly for following reasons:

1. Various parts of the fuel cycle of fusion devices use mixtures with a relatively low tritium content.

2. These systems will be subjected to a detritiation procedure as part of periodic maintenance to prevent tritium accumulation; procedures proposed in this paper can be of use.

3. After completion of operation, fusion devices are subject to a decontamination procedure to the minimum achievable level of tritium content; the methods proposed in this paper can be considered for this procedure.

4. Effective procedures proposed in this paper can be tested at a higher tritium concentration.
In this work, detritiation was required to levels of tritium contamination much lower than the normative established ones (from 200 to 8000 Bq/cm² for different types of surfaces and for premises of different classes of radioactive hazards), which is an even more difficult task in the detritiation of facilities and equipment. As opposed to individual contaminated materials and radioactive waste, some additional difficulties arise. These are the operational limits of the facility, such as permissible temperature regimes, pressure ranges, etc. In the case of the Troitsk Nu-mass setup, pressure in the spectrometer was limited to 2 mbar and the heating temperature of the outer jacket could not exceed 110°C. These limitations reduce the range of possible methods and lead to additional costs, in particular, to an increase in the duration of the detritiation.

In this work we describe the procedure and the results of tritium decontamination in the Troitsk Nu-mass spectrometer.

II. TROITSK NU-MASS EXPERIMENT

The Troitsk Nu-mass setup\(^\text{11}\) was designed to measure the tritium decay spectrum to estimate the electron neutrinos mass and to search for hypothetical particles—sterile neutrinos.\(^\text{12}\) The setup consists of two main parts: a gaseous windowless tritium source (WGTS) and an electrostatic spectrometer with magnetic collimation (Fig. 1).

II.A. Windowless Tritium Source

The WGTS (Figs. 1 and 2) is designed to form an electron flux generated by tritium decay inside the source tube and to transport it along the magnetic field lines to the spectrometer. In the source, tritium circulates in a closed loop (Fig. 2). A booster pump B provides pumping. Tritium from the sorption storage in the form of deuterium-tritium (DT) molecules is injected into the center of the decay volume (Tritium pipe in Fig. 2), and then gas is pumped out from both sides by pumps R1 through R4 into a common collector. Pumps R2, R3, and R4 are located on the side of the spectrometer and provide differential pumping of the working gas to prevent tritium penetration into the spectrometer. Additionally, zigzag-shaped vacuum solenoids with magnetic field and total length of about 1 m are installed to transport electrons from the source to the spectrometer. These solenoids contain copper cryopanel traps at a temperature of about 5 K, with frozen argon to adsorb residual tritium. In order to increase the adsorption surface, these copper cryopanels are manufactured as a series of 25 diaphragms with inner and outer diameters of 20 and 50 mm, respectively. The argon frost is prepared by injection of gaseous argon through three perforated capillaries placed along the cryopanels. In total, the suppression coefficient of tritium and other gases before entering the spectrometer is not worse than 10\(^{10}\). At the same time, this does not create obstacles for electrons to enter the spectrometer.

The gas in the WGTS sequentially passes through the circuit warm part at room temperature, and then through the central part at a temperature of about 30 K, located inside the superconducting solenoids. About 3 GBq of tritium is injected into the circuit at a time. During measurements, part of the tritium is adsorbed on the pipe surface and argon frost. As a result, during a long-term operation the source

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**Fig. 1.** The Troitsk Nu-mass setup scheme: (a) the WGTS, a windowless tritium gaseous source and (b) an electrostatic spectrometer (1 is the prespectrometer with its pump, 2 is the prespectrometer valve, 3 is the spectrometer valve, and 4 is the argon cryopanel).
intensity decreases by nearly two times in 5 to 6 days. This leads to the need to add tritium to the circuit. Thus, in 2 weeks of operation of the installation, ~4 to 5 GBq of tritium is adsorbed in the circuit cold sections. In addition to tritium, impurity gases from the atmosphere (mainly nitrogen, oxygen) begin to be adsorbed over time in the cooled parts of the source. In the warm part of the source, where the main portion of the working gas is concentrated, the pressure does not exceed 1 to 2 mbar. A gas mass analyzer controls the gas composition in the circuit warm part in front of the R1 pump. According to records, the partial pressure of water is five to six times less than the pressure of nitrogen. After finishing work with the source, the vacuum valves between the source, the prespectrometer, and the spectrometer are closed. The absence of tritium inside the spectrometer guarantees high-quality measurements of the energy spectrum for electrons generated in the WGTS.

At the start of one physics run, it was found that the electron background count rate by the main Si detector located at the end of the spectrometer increased 100 times relative to the previous values and reached 18 000 counts per second (cps), which indicates the presence of tritium in the spectrometer. Activity originated by tritium decays from the walls and was seen by the Si detector as electrons with energy close to the potential on the electrode. Such a high background level was unacceptable for further measurements, and it should have been reduced by at least an order of magnitude. Later, it was discovered that, after the previous measurement session, the vacuum valve between the source and the prespectrometer did not close tightly, and tritium penetrated to the prespectrometer volume. Unfortunately, shortly after that, another valve between the prespectrometer volume and the main spectrometer was opened for a few minutes for maintenance. This led to tritium penetration into the spectrometer. Thus, we can assume that leakage was short and the spectrometer was under pressure of ~10^{-4} Pa for about 2 months, when it all was found out in preparation to the next physics run. During these 2 months, the outer temperature in the experimental hall was between 15°C and 20°C and no special treatment nor maintenance procedures were undertaken to the spectrometer.

It should be emphasized that most of the tritium was pumped from the cold part of the source, and only after that, were the pumps (R1 through R4 in Fig. 2) stopped. Therefore, only part of the tritium, which was adsorbed on the surfaces of the cooled pipes and the argon trap, could get into the spectrometer. When the source was heated from 30 K to room temperature, defrosting and desorption of impurity gases (nitrogen, argon, oxygen, and water vapor) occurred. These gases and tritium compounds could penetrate into the spectrometer.

Fig. 2. Diagram of a WGTS where R1 through R4, B, and N are steam diffusion mercury pumps, ZT is a zeolite trap, PS is a pressure sensor, C1 is the tritium sorption storage cartridge, C2 is the purifying cartridge, C3 is the transport container, and GUN is an electron gun. The big arrows show the directions of gas circulation.
II.B. Spectrometer

A spectrometer cross section is shown in Fig. 3. The total spectrometer length is approximately 10 m, and the internal volume is approximately 40 m³. Inside the vessel, there is a central electrode mounted on several dielectric supports. The vessel and inner electrode are made of stainless steel type 12X18H10T (United States, analog 321, 321 H). The inner surface of the vessel and the whole surface of the electrode were electrochemically polished. The spectrometer outer diameter is 2.75 m, the cylindrical part length is 6 m, and the inner electrode diameter is 2.4 m. It should be stressed that the electrode increases the total surface inside the spectrometer about three times relative to the outer vessel. As a result, the total metal surface is about 160 m². In addition, two small duralumin electrodes (total surface of about 5 m²) and superconducting magnets are installed at the spectrometer ends. The magnets have cupper cooling screens with a surface of about 8 m². In addition, the cooling screens are covered by cryogenic “super insulation,” a 20-layer stack of superfine glass fiber and aluminized Mylar film. This super insulation doubles the total surface.

II.C. Detritiation Condition Limitations

It was decided to perform the detritiation of the spectrometer volume without its opening since the filling and pumping cycle of such a big volume takes several months. Necessary safety measures were taken to work with tritium and its compounds. Operating parameters and safety considerations limited the maximum pressure (2 mbar) and the temperature of the spectrometer vessel warming up (110°C). The cryostats of the superconducting magnets at the input and at the end of the spectrometer have indium seals. Heating the spectrometer jacket above 110°C can damage the seals and break the cryostat tightness. One cannot provide good heating of the inner electrode surface by limiting pressure in the spectrometer. The temperature on the electrode surface was not controlled, but calculations showed that after 8 h of warming up, the electrode temperature could reach a value of 60°C. In addition, layers of super insulation based on aluminized Mylar are installed around the cryostats. Thin layers of aluminum are very sensitive to ammonia. Viton gaskets, which are used to seal the spectrometer, are sensitive to ammonia and high temperatures.

III. DETRITIATION PROCEDURE FOR CONTAMINATED VOLUMES

For the detritiation, we used the vacuum extraction method and hydrogen and water vapor soaks.

III.A. Vacuum Extraction

The Troitsk Nu-mass vacuum system allows one to reach vacuum in the spectrometer at a level of $10^{-11}$
mbar. This procedure removes tritium from the gas phase and tritium adsorbed on the surface. The spectrometer did not warm up during this process. The method has low efficiency at room temperature since the tritium and its compound desorption rate is $4.4 \times 10^{-2} \pm 1.4 \times 10^{-2}$ dpm/cm$^2 \times$ s (Ref. 2), which is confirmed by the data of the detritiation of contaminated samples$^3$ and the tritium removal from tokamaks JET and TFTR (Refs. 13 and 14).

III.B. Hydrogen Soaks

In this case, the spectrometer was filled with dry hydrogen up to a pressure of $10^{-2}$ mbar and kept for several days without heating the body, and then it was pumped out to a pressure of $\sim 1 \times 10^{-5}$ mbar. This method was applied for six sessions.

III.C. Water Vapor Soaks

The spectrometer or prespectrometer volumes were filled with a small portion of hydrogen (protium or deuterium) with the addition of water (except for some cases). Through a special gasket, 20 mL of distilled water was injected with a syringe. The water instantly evaporated, which was recorded by an increase in pressure. Then, gaseous hydrogen was added until a pressure of about 2 mbar in the spectrometer. The spectrometer vessel was cyclically heated to 110°C, mainly for 8 h during the daytime. The actual change of the temperature was the following (see also Fig. 4). Starting from 9 a.m. until 12 p.m., the temperature was rising, and then stayed flat at 110°C until 5 p.m. After that, heaters were switched off and the vessel exponentially cooled down for about 16 h so that the next morning the temperature was about 35°C. Warm-up time was sometimes increased up to 12 h, the temperature rise was also 3 h. On some days, the spectrometer was not heated due to nonworking days or other reasons. Warm-up time, as mentioned in Table II, is the sum of these cycles. After several days, the volume of the spectrometer was pumped out by a standard vacuum system to a pressure of $\sim 1 \times 10^{-5}$ mbar. A cold trap with liquid nitrogen was used to collect water vapor. The exhaust from the pumping system was directed to a special ventilation system and then to the atmosphere outside the building.

The duration of exposure, the duration of the heating, and the composition of admitted gases of the procedure were varied (in one case, water was replaced with a 10% aqueous solution of ammonia, and in the other, with a 3% solution of H$_2$O$_2$).

The prespectrometer was detritiated in a similar way as described previously. The conditions were slightly changed: maximum pressure was increased to about 80 mbar, formed by 20 mL of water, and the rest by hydrogen (protium or deuterium). The detritiation of the prespectrometer was performed in parallel and independently of the work on the main spectrometer.

This detritiation procedure with water vapor was applied for eight sessions for the spectrometer and six sessions for the prespectrometer.

III.D. Methods for Assessing the Efficiency of the Detritiation

The detritiation efficiency was controlled in two ways: by measuring the activity of the collected tritiated

![Fig. 4. Temperature-time diagram of the water vapor soak procedure (the timeline is indicated conditionally).](image-url)
water and by checking the count rate by the Si detector of the spectrometer.

Water from the spectrometer was collected by a cold trap with liquid nitrogen. We did not use catalytic oxidation of molecular tritium (hydrogen) to evaluate its amount in the spectrometer. Therefore, this was only an estimate of the tritiated water content.

The specific activity of the released water \( A_{\text{HTO}} \) MBq/ml was measured by the scintillation method on a Tri-Carb 2810 counter. The Tri-Carb 2810 detection limit is 0.6 Bq in the case of tritium water and scintillator Ultima Gold. The measurement error was 10%.

Measurement of the spectrometer Si detector background is an indirect method for assessing detritiation efficiency. After each session, the spectrometer was pumped to a vacuum of \( 10^{-5} \) mbar. This vacuum level makes it possible to register the electrons generated inside the spectrometer. The sources of this kind of electrons are cosmic rays, which knock out delta electrons from the electrode surface, the natural radioactivity of the electrode material, and internal tritium contamination. For measurements, a high negative voltage (from 15 to 20 kV) was applied to the electrometer without magnetic fields because even a few Gauss magnetic fields turn the low-energy electrons back onto the electrode material. The electrons produced on the inner surfaces are accelerated to the energy equal to the voltage on the electrode. Before the tritium contamination of the spectrometer, the background count rate of the Si detector was 160 to 180 cps. This count level is a good criterion for spectrometer purity. After contamination by tritium, the Si detector background increased to 18 000 cps. Due to the geometric configuration of the additional electrodes and the aperture of the superconducting magnets, the detector registers electrons only from the inner surface of the electrode. Other contaminated surfaces remain outside the sensitivity range. Measurements with an Si detector are qualitative; however, they help estimate the degree of decontamination. We estimate the Si detector measurement error to be 5%.

IV. SPECTROMETER DETRITIATION RESULTS AND DISCUSSION

The results of the spectrometer detritiation by hydrogen and water vapor soaks are presented in Table I.

The values in the column “Detector Background \( (10^3 \text{ cps}) \)” indicate the detector readings prior to the start of each detritiation procedure. The values in the column “Released Activity (MBq)” indicate the amount of tritium removed from the spectrometer as a result of detritiation. These values were obtained by multiplying specific activity of the released water \( A_{\text{HTO}} \) by the amount of water \( N_{\text{HTO}} \) introduced into the spectrometer, which is indicated in the column “Reagents.”

The detritiation results of the spectrometer volume are presented in Table II. The only way to control the detritiation process of the spectrometer was to measure the recovered water activity.

IV.A. Initial Tritium Contamination

The initial tritium contamination was estimated based on comparison of the released activity in the form of tritiated water and the Si detector background.

Figures 5 and 6 show the correlation between these values. A high residual background of the detector (1860 cps) and low released tritium amount in the last soaks indicate the tritium presence in the spectrometer in such a form (or in such places) that its further extraction by the selected methods won’t be effective. Using a linear dependence coefficient \( (0.0044 \pm 0.00061) \) and taking into account the error of the measuring instruments, we can estimate initial tritium contamination of the spectrometer as \( (4.09 \pm 0.73) \) GBq with an initial detector background of 18 000 cps. Assuming that prespectrometer detritiation leads to similar results, we can use the ratio of the initial contamination of the spectrometer to the released tritium (1.79) to estimate the initial tritium content in the prespectrometer, which was \( (1.09 \pm 0.22) \) GBq. The total contamination was \( (5.18 \pm 0.92) \) GBq \( (1.4 \times 10^{-5} \) g in terms of pure tritium in the molecular form or less than \( 9 \times 10^{-5} \) g in HTO form).

The stabilization of the residual background less than 2000 cps in comparison to the pure spectrometer count rate of 180 cps indicates that the residual tritium content on the inner surfaces is at a level of \( 420 \pm 75 \) MBq. The specific activity of tritium on the surface is \( \sim (250 \pm 47) \) Bq/cm² (in case of even spreading over the area of \( \sim 160 \) m²).

IV.B. Effectiveness of Detritiation Methods

IV.B.1. Vacuum Extraction

Vacuum extraction didn’t give significant results. Pumping out the spectrometer a few times and maintaining the vacuum at a level of \( 1 \times 10^{-5} \) mbar at room
TABLE I
Results of Each Session of the Spectrometer Detritiation

| Number | Detector Background (10^3 cps) | Exposure Time (day) | Warm-up Time (h) | Released Activity (MBq) | Reagents                     |
|--------|-------------------------------|--------------------|------------------|-------------------------|------------------------------|
| 0      | 18                            | 30                 | Room temperature| —                       | H₂, 10⁻² mbar, initial background |
| 1      | 14                            | 30                 | 80               | 1840                    | 20 mL H₂O + H₂              |
| 2      | 6.0                           | 8                  | 28               | 231                     | 60 mL H₂O + H₂              |
| 3      | 3.95                          | 8                  | 72               | 137                     | 20 mL H₂O + H₂              |
| 4      | 2.76                          | 4                  | 24               | 32                      | 20 mL H₂O                   |
| 5      | 2.44                          | 5                  | 24               | 22                      | 20 mL (97%H₂O + 3%H₂O₂)     |
| 6      | 2.11                          | 12                 | 87               | 31                      | 20 mL H₂O + H₂              |
| 7      | 2.05                          | 3                  | 12               | 8                       | 20 mL (90%H₂O + 10% NH₃)   |
| 8      | 1.9                           | 6                  | 48               | 29                      | 20 mL H₂O + D₂              |
| 00     | 1.86                          |                    |                  |                         | Residual background         |

TABLE II
Results of Each Session of the Prespectrometer Detritiation

| Number | Exposure Time (day) | Warm-up Time (h) | Released Activity (MBq) | Reagents |
|--------|--------------------|------------------|-------------------------|----------|
| 1      | 20                 | 40               | 426                     | 10 mL H₂O |
| 2      | 8                  | 32               | 57                      | 10 mL H₂O |
| 3      | 9                  | 40               | 29                      | No hydrogen, 20 mL H₂O     |
| 4      | 12                 | 64               | 40                      | 20 mL H₂O |
| 5      | 18                 | 95               | 40                      | 20 mL H₂O |
| 6      | 10                 | 50               | 30                      | 20 mL H₂O |

temperature did not lead to a decrease in the detector background.

IV.B.2. Hydrogen Soaks

The detritiation process by hydrogen soaks was not monitored between iterations. Only the initial and final (after six cycles) detector backgrounds are available for analysis, 18 000 and 14 000 cps, respectively. These results indicate a low efficiency of the method.

The authors of Ref. 14 call hydrogen soaks completely ineffective under the conditions of the JET tokamak and also refer to similar results for the TFTR (Ref. 13) and TEX-TOR tokamaks. Tritium release behavior from the JT-60 (Ref. 15) shows that the pressure in range 0.5 to 3 mbar and gas type during purges with H₂, He, and Ar do not significantly affect the tritium release from the tokamak walls. This indicates an insignificant effect of isotope exchange under these conditions because otherwise hydrogen purges would show greater efficiency. It should be stressed that in tokamaks, almost all tritium is in absorbed form and is deeply embedded in materials, which is a consequence of high operating temperature and plasma loads during tokamak operation and transmutation reactions. In our case, the molecular tritium sorption is possible in a small amount and directly on the material surface only due to low pressure and room temperature. Therefore, it seems that it should be easier to remove tritium, but our observations show that was not enough to restore the spectrometer.
A completely different detritiation behavior was described in Refs. 16 and 17. In these studies, 2.6 GBq of molecular tritium was injected into a special box, then the tritium was removed by a ventilation system. Under these conditions, tritium was completely (to the level of the background of the detectors) removed from the box, on the average, with an eightfold exchange of the atmosphere. The authors of Refs. 16 and 17 emphasize that when molecular tritium is injected into a volume of 12 m$^3$ filled with air under normal conditions, there is neither significant sorption of molecular tritium by the walls of the box nor formation of the tritiated water as a result of oxidation or isotope exchange. That is, the incoming clean air simply displaces tritium from the researched volume.

In our work, there was no displacement, but there was a dilution of the initial atmosphere inside the spectrometer; the dilution degree was at least 6000 times (six
sessions, the ratio of the initial and final pressure ∼1000). As a result, almost all the tritium in the gas phase was removed. Despite such a significant atmosphere dilution, the detector background decreased only by ∼20%. This allows us to conclude that initially tritium in the spectrometer was distributed between the gas phase and the solid phase (in adsorbed form). After hydrogen soaks, tritium was predominantly in adsorbed form as hydrogen (HT, DT) or water (HTO) molecules.

**IV.B.3. Water Vapor Soaks**

After the first attempt, the spectrometer Si detector background was significantly reduced. The tritium concentration in the collected water was 92 MBq/mL, which corresponds to a total recovered activity of about 1840 MBq. Starting with session 5, the background level actually stabilized and the amount of removed activity decreased significantly. The change of the procedure had no significant effect on the detritiation efficiency. A water amount increase (session 2) only reduced the specific concentration of tritium per milliliter in recovered water and did not noticeably affect decontamination. A hydrogen peroxide solution (3%) was used in session 5 without adding hydrogen gas. No significant effect was found; rather, a drop in the recovered activity was observed. Ammonia (10%) in water was used in session 7 to see its potential benefit. For fear of noticeable corrosion of internal components, the exposure time was limited to 3 days. Due to the low concentration of ammonia and the short exposure time, the recovered activity dropped by a factor of 4. Replacing hydrogen (protium) with deuterium (session 8) did not improve detritiation either. The stable and relatively small amount of tritium in samples 5 through 8 probably indicates that the limits of the method have been reached. For further detritiation, a higher temperature (especially on the electrode surfaces), pressure, and water vapor amount are needed.

The detritiation results of the prespectrometer volume correspond to those described previously for the spectrometer. The amount of released tritium stabilized after session 3. A small value in the recovered activity in session 4 may indicate the importance of the hydrogen presence during decontamination. A similar effect was observed in sessions 4 and 5 for the spectrometer.

The first injection of water in the spectrometer removed 79% of tritium in water form (1.8 of 2.3 GBq) and 45% of tritium in all forms (1.8 of 4.1 GBq), as estimated previously. After all water vapor soaks the Si detector readings dropped by ∼70%, indicating a high efficiency of the method. Such a high efficiency should be related to a high rate of exchange of adsorbed tritium water with water vapor in the gaseous phase.

In Refs. 16, 17, and 18, a mechanism of large-volume detritiation in the presence of tritiated water is proposed. In these works, in addition to experiments with tritium in molecular form, a mixture of molecular (97% to 99%) and oxidized (water, up to 3%) tritium with an activity of 2.6 GBq was injected into the box. In the presence of tritiated water, the character of the box detritiation changed significantly. This was revealed in an increase in the time required to evacuate tritium and residual contamination from the inner surfaces, and in the influence of the purged air humidity on the detritiation rate. The authors of Refs. 16, 17, and 18 associate these effects with the process of adsorption and desorption of tritiated water on the inner surfaces of the box. The authors showed that the results of simulations of these processes using the Freundlich equation are in good agreement with the experimental results. This case resembles in the main details the detritiation process we observed.

Later, these results were used to test the tritium removal from the JT-60 tokamak. The JT-60 tokamak was filled with nitrogen or air with controlled humidity to a pressure of 1 bar for 24 h and then pumped out. It was shown that water vapor enhances tritium release from tokamak surfaces even in ambient temperatures. Nearly the same procedure was used in the Large Helical Device (LHD) stellarator during the maintenance activity. In the initial phase, the tritium release amount was increasing with the increase of the humidity level. Then, the influence of the humidity level on the tritium release decreased, which indicated the prevalence of diffusion-limited tritium release from the material bulk. These results suggest the possible detritiation mechanism of the Troitsk Nu-Mass setup and show the usefulness of water vapor usage in effective mild methods of detritiation, like gas purges and air ventilation, for tritium facilities.

**IV.C. Detritiation Mechanism Discussion**

The detritiation mechanism depends mainly on the form of tritium (molecular, oxidized, or other). The main difficulty in describing the detritiation mechanism is the lack of data on the composition of the gas that entered the spectrometer due to equipment failure. We do not know the ratio of tritium in various forms (molecular or oxidized or other) during detritiation since the sampling technique allowed us to capture only tritiated water vapor. Additionally, the detritiation process wasn’t
investigated systematically; the detritiation conditions such as exposure time, warm-up time, and gas composition differed significantly for each session. The main goal of the detritiation process was the earliest possible return of the Troitsk Nu-mass unit to work. With this in mind, we discuss the possible detritiation mechanisms.

When considering the mechanism of detritiation, we assume two possible options for the initial spectrometer contamination:

1. The main contaminant was tritium water that got into the spectrometer from a tritium source.

2. The main contaminant was tritium in hydrogen form.

In the first case, the tritiated water was easily adsorbed on the inner surfaces of the spectrometer. The high efficiency of the water vapor soaks can be explained by isotopic exchange between water in the gas phase and adsorbed tritiated water and the subsequent removal of the gas phase by pumping. The inefficiency of the hydrogen soaks is associated with low hydrogen adsorption and the low isotope exchange rate between hydrogen and water. This picture can be compared with data obtained in the Caisson Assembly for Tritium Safety (CATS) experiment and Tritium Systems Test Assembly facility and the results of the JT-60 and LHD (Refs. 15 and 20). As was described previously, the initial contamination in that case was less than $9 \times 10^{-5}$ g HTO, which was evenly spread on 160 m$^2$ of the spectrometer inner surface.

In the case of contamination with tritiated water, it is recommended to fill the volume with water vapor and hydrogen (or inert gas since the tritium release is not significantly affected by the gas type). The exposure time depends on the total pressure, water vapor content, and bulk temperature. It should decrease with increasing these values. After exposure, the volume should be pumped down, and the tritium water collected by cold trap or molecular sieve bed. This procedure is suitable for high vacuum facilities that can’t be ventilated with humid air (or other purge gases).

However, as follows from the description of the tritium source operation, tritiated water should not be there. Therefore, the assumption of its presence in the initial contamination composition may seem unreasonable and another mechanism should be proposed.

In the case of only molecular tritium (HT, DT) contamination, a large internal and well-prepared spectrometer surface (almost always under a deep vacuum) can adsorb a fraction of molecular tritium. This fraction shouldn’t be significant considering the low specific content of tritium, low pressure (10$^{-6}$ mbar), and ambient temperature during contamination. However, in some cases adsorption might be significant. In Ref. 22, a gold-coated sample was exposed to tritium at a pressure of 10$^{-5}$ mbar, and the determined surface activity of tritium was $5.4 \times 10^7$ Bq/cm$^2$, which is much higher than our estimation for the spectrometer (250 ± 47 Bq/cm$^2$). Moreover, tritium can interact with adsorbed water, other oxides, and compounds on inner surfaces, as well as organic compounds (like Mylar) and some glasses, to form hydroxyl groups that can easily exchange isotopes with water.

We need to consider, however, that the isotopic exchange of tritium in hydrogen form with hydroxyl groups or other compounds is significant in the case of high tritium concentration because it self-catalyzes due to decay. In our case, the tritium concentration was low, so an additional amount of tritium in the oxide layers and organic materials should be small. The presence of tritium in the HTO form that we collected can be explained by isotope exchange reaction between the adsorbed tritium, tritium in oxide layers, organic materials, and water. However, it is difficult to explain the low efficiency of hydrogen soaks since the adsorbed tritium should be significantly diluted during the exposure and subsequent pumping.

To substantiate the mechanism of detritiation, additional measurements are required, in particular, the analysis of impurities in a WGTS.

IV.D. Additional Measures to Compensate the Influence of the Residual Tritium Content

To ensure the operability of the Troitsk Nu-Mass setup in the presence of residual tritium in the spectrometer, it was decided to increase the magnetic field in the center of the spectrometer by an additional solenoid winding over the vessel. The additional magnetic field functions as follows. Almost all the electrons, which are seen by the Si detector, are generated by tritium decay on the electrode inner surface inside the bulk and have low energy, no more than a few electron volts. Even a small magnetic field is enough to turn them around and direct them back into the electrode body. A technical run was carried out involving all the elements, including the superconducting magnets. A positive result was obtained: After detritiation and the inclusion of an additional magnetic field, the Si detector background with an open gas tritium source (without tritium filling) turned out to be higher than before by a factor of 3 to 4, but at an acceptable level for measurements. We want to stress that during the physics run, the observed low level of the background may play a role for the measurements only at the
very end of the tritium spectrum, where the count rate is low. At the rest of the spectrum, it is negligible.

After detritiation, the spectrometer periodically was pumped out to the pressure less than 10^{-9} mbar. A few months later, successive physics runs did not show a significant change in the tritium background; it stays at the level as just after the detritiation.

V. CONCLUSION

Due to equipment failure, the Troitsk Nu-Mass setup internal volume was contaminated with tritium. Detritiation was performed under pressure (2 mbar) and temperature (110°C) limits in the setup. A minimum amount of reagents was used, resulting in a small amount of tritium-containing waste.

The efficiency of various detritiation methods was evaluated. The most effective method is the water vapor soaks. An addition of gaseous hydrogen seems to be quite effective. It is shown that the addition of ammonia, peroxide, and replacement of protium by deuterium does not lead to an increase in the efficiency of the detritiation. The usage of dry hydrogen soaks is ineffective.

Water vapor soaks as a detritiation method are advisable for high-vacuum facilities that can’t be ventilated with humid air (or other purge gases).

Different possible mechanisms of detritiation are discussed.

As a result of the detritiation, the estimated initial tritium contamination of (5.18 \pm 0.92) GBq in the spectrometer and prespectrometer was reduced to (0.42 \pm 0.07) GBq. This tritium amount is negligible to resume research program on the Troitsk Nu-Mass setup, however, it requires some additional measures. Residual activity can be associated with the surface contamination of unheated parts of the installation, diffusion, or the chemical interaction of tritium with the internal materials of the spectrometer. The main goal of returning the Troitsk Nu-Mass setup to working condition was achieved.

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