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# Helium3 isotope separation and lambda front observation

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ABSTRACT

We present the method of the liquid helium enrichment with He3 isotope based on a special solution of the helium bath cryostat (25 l LHe) equipped with the heat exchanger and entropy filter to stimulate quantum filtration process in low temperature region. The cryostat design makes it possible to increase the initial concentration of He3 more than one order of magnitude. While the temperature of lambda point was achieved close to the capillary-equipped heat exchanger, the movement of lambda front within the liquid helium is observed. A consistent description of the various stages of the cryostat operation with cooling process above the lambda transition, during the long period of lambda front movement and in a superfluid region, with the acquisitioning of He3 isotope in all of these regions, is devoted to the design of a helium isotope flow-separator on an industrial scale.

## 1. Introduction

Strong fluctuations on the He3 isotope market appeared in the period of last decade [1,2]. After the attacks on September 11, 2001 demand for neutron detectors strongly increased and He3 isotope starts to be of the great importance due to its sensitivity to the radioactive materials. He3 availability is also important from the point of view of the future clean energy production - nuclear fusion in the D-He3 system can play the crucial role. Therefore serious plan to utilize He3 from the Moon and planets such as Jupiter or Saturn are developed [3,4]. In contrast to experimental works, where the filtration and/or rectification methods have been described [5–9], theoretical studies are dedicated rather to the specific membranes, which can help to separate He4 and He3 isotopes [10–15]. In practice, due to the He3 isotope market demand, patents are also a goal [16,17]. One of the most important methods to acquire He3 here on Earth is separation of this isotope from the liquid He4/He3 mixture using two processes - filtration based on entropy filter or rectification procedure. Both methods should be the most profitable if performed directly at a helium plant [1,5]. Many cryogenic applications are connected with exploitation of a bath cryostats. These cryostats are usually dedicated to work at temperatures near 4.2 K (liquid helium boiling temperature) and below, down to 1 K. In this case the system needs to be cooled down at least to the lambda point ($T_\lambda$ = 2.18 K). There are essentially two ways to reach this temperature level. Both are well known: reducing vapor pressure above the liquid helium surface, usually with a simple rotary pump ("pumping procedure") or using various kinds of heat exchangers. Some solutions were designed for superconducting magnets. These cryostats are called lambda point refrigerators and are used to cool down the superconducting magnets to approximately 2.2 K using the needle valve to control the liquid helium flow around the lambda plate (refrigerating copper coils) [18]. In these refrigerators lambda plate is located in the top of the superconducting magnet to use the convection of helium according to its density which increases with decreasing temperature until the lambda point is reached [19]. During the helium pumping, more than 30% of helium is lost, together with the He3 isotope. Simultaneously the He3 evaporation from the mixture strongly increases [20]. It is because the difference between partial pressures of He3 and He4 components increases as the temperature is going down - below 1 K this difference reaches three orders of magnitude [21,22]. In our experiment a special solution using the liquid nitrogen shielded bath cryostat, together with the heat exchanger is used. This solution enabled us to observe the moving lambda front within the volume of liquid helium without removing the He3 isotope during the cooling procedure. It means that in this experiment the whole liquid helium bath can be subjected to the He3 filtration procedure. This method gives the possibility to perform the He3 isotope enrichment in a quite effective way. To find the best power range necessary to activate the

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entropy filter for the filtration process, with the use of the helium fountain effect of superfluid helium, the standard double glass Dewar container was used.

## 2. Results and discussion

Schematic presentation of the glassy cryostat is shown in Fig. 1. It operates in the power range of 100 mW–1000 mW giving the smooth, laminar flow of the He4 isotope through the capillary fountain system. Using the instrumentation presented in Fig. 1 we also calibrated the quadrupole mass spectrometer (QMS 700) produced by Pfeiffer Vacuum GmbH which allows for detection of $10^{-3}$ ppm of He3 (QMS 700 parameters are: mass range 1–128 amu, sensitivity - single ion $10^{-19}$ A, resolution - one neutron). This part of the experiment confirms that the pumping procedure at the temperature level of 1.5 K removes He3 efficiently. This effect (as mentioned before) is connected with the increasing difference between the partial pressures of He4 and He3 at temperatures below 2 K. In order to observe the difference in concentration of He3 during the filtration process, two samples had to be collected: the first one before the mixture enters the entropy filter and the second one behind the filter. Decrease of the He3 isotope concentration during overfilling of the burette is shown in Fig. 2 - albeit the He3 level behind the filter still is lower than in front of them, filtration process occurs, but not effectively.

In order to avoid the above mentioned loss of the He3 isotope, the 25 L bath cryostat with the heat exchanger was constructed. Schematic presentation of this apparatus is shown in Fig. 3. Cooling power of the heat exchanger was kept at the level of 700 mW. Cooling process was realized with the capillary of 0.01 mm diameter with a steel wire slightly thinner than the capillary inside. It allowed for the stabilization of the helium transfer during the cooling process at the very low level. Helium evaporated in the coper coil which worked as the Joule-Thomson valve. This system reduced the helium consumption by approximately 10% during the process of cooling to the lambda transition. Heat exchanger was located at the bottom of the cryostat - opposite side to the lambda point refrigerators described by Belshaw [18]. As the He4 density reaches maximum close to the lambda point [19], the convection drives the temperature changes in the whole helium volume. Thus the gradients of density and temperature appear (in the opposite direction than in the lambda point refrigerators). It allows the observation of the front lambda moving up the cryostat.

Extremely high thermal conductivity of the superfluid helium (below lambda point it reaches values more than five orders of magnitude larger than that of helium above $T_\lambda$ [21]) ensured us that the temperatures below $T_\lambda$ could only occur after the whole helium volume was cooled to the lambda point. The lambda front movement was observed via the set of thermometers located at different heights inside the cryostat. Thermometers where mounted at the points crucial for the cryostat operation. Front lambda movement is shown in Fig. 4. This figure also presents the schematic thermometers location, i.e. at the heat exchanger capillary (blue[1]), entropy filter (magenta), the green thermometer is located behind the entropy filter - there is fountain effect (inside a steel pipe) and additional point located in the intermediate position (red). The last T indication comes from the equilibrium pressure inside cryostat (orange).

The first part of the experiment (time interval 10–15 ks) shows the front lambda ($T_\lambda$ = 2.18 K) moving up the cryostat. The effect could be traced as the thermometers reached $T_\lambda$ one by one, starting from the bottom one (blue). Rapid temperature drops to $T_\lambda$ observed for all thermometers were the result of the strongly increasing thermal transport in the vicinity of thermometers - the Joule heat, generated by the wires and thermometers themselves, was efficiently removed at $T_\lambda$.

[1] For interpretation of color in Fig. 4, the reader is referred to the web version of this article.

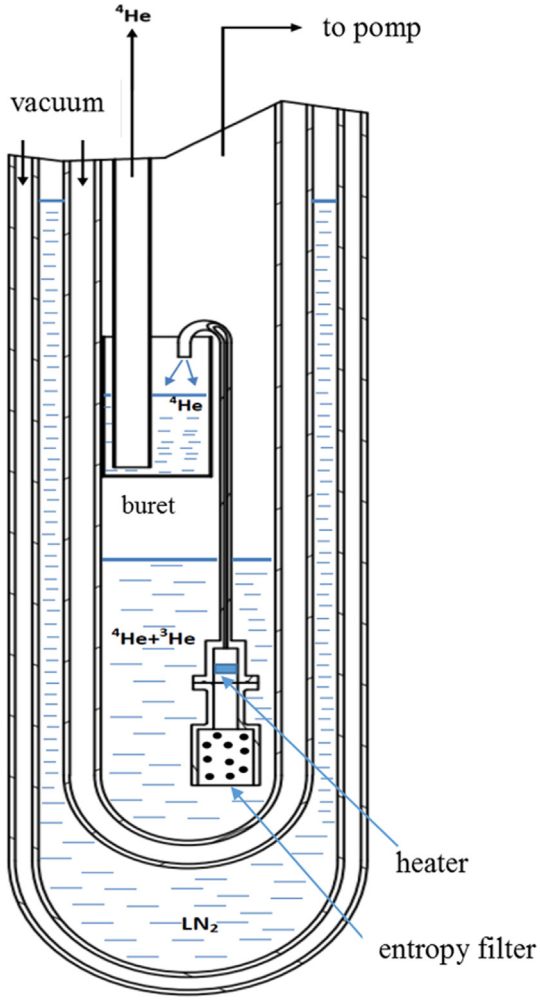


**Fig. 1.** Glassy cryostat system with the entropy filter inside. Helium during the filtration process was removed from the test-tube.

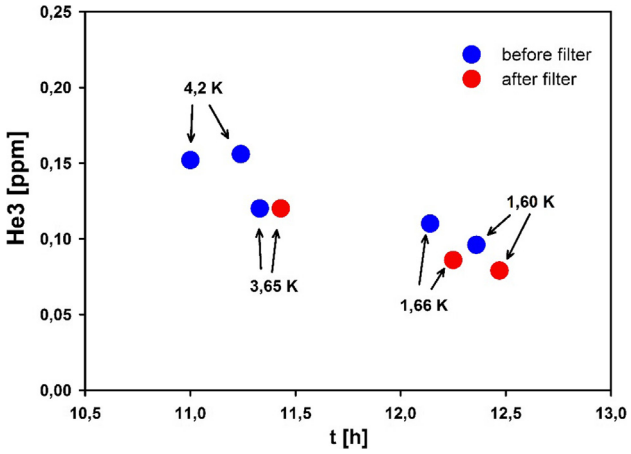


**Fig. 2.** Decrease of He3 concentration during the filtration process carried out in the glassy cryostat in which the whole volume of helium were connected to the pomp system.

Due to the extremely high thermal conductivity of the superfluid helium the system had to wait for the lambda transition to occur in its whole volume (up to the helium surface), before further cooling was possible. This stage was recorded by measuring the pressure above the helium surface (orange line in Fig. 4). The final drop was the result of thermal equilibration of the gas and liquid phases (above $T_\lambda$ disturbed

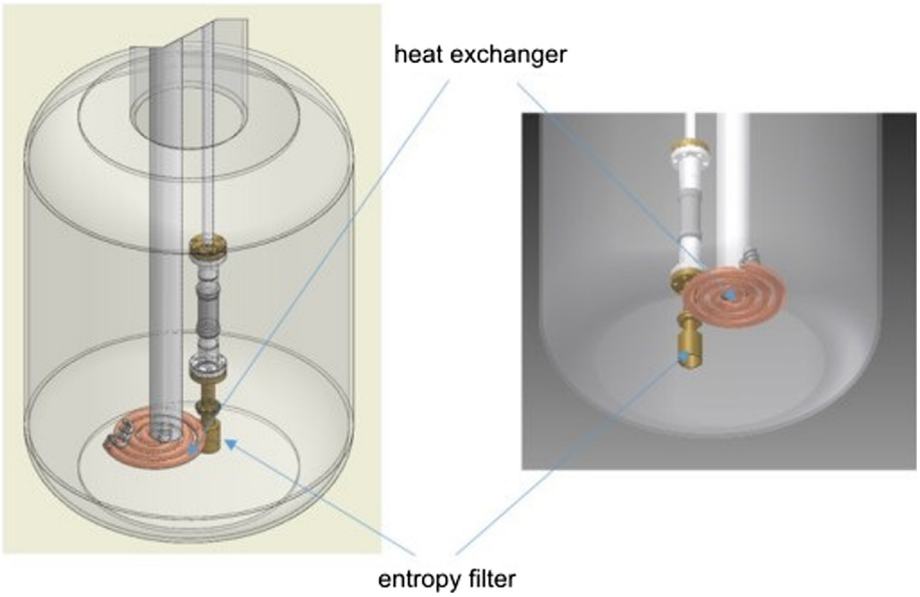


**Fig. 3.** Liquid helium cryostat insert with heat exchanger and entropy filter.

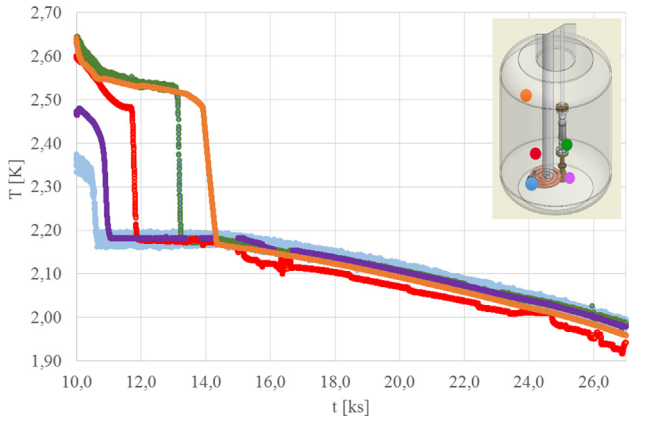


**Fig. 4.** Moving of the lambda front during process of cooling with using the heat exchanger. Thermometers location inside the cryostat is marked by appropriate colors – full explanation in text.

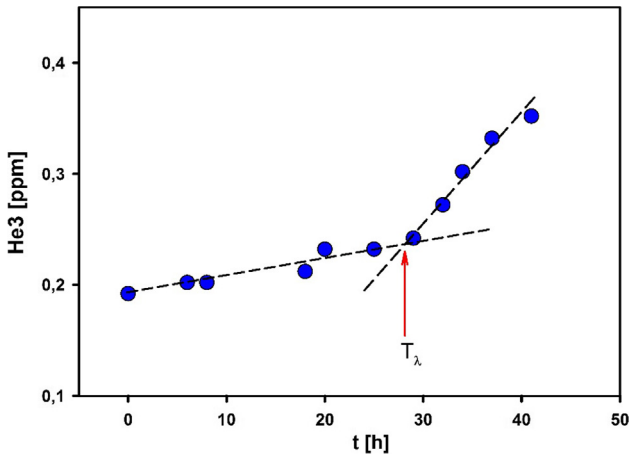


**Fig. 5.** Initial He3 filtration process generated by the capillary. Presented concentration of He3 was measured for the sample taken from the whole bath cryostat volume - before the entropy filter.

by the contact with the cryostat walls). After the whole volume reached the lambda point, the temperature started to decrease to 1.6 K, (t > 15 ks in Fig. 4) where the filtration process could be effectively conducted.

Before the main filtration process is initiated by the heater, it is possible to observe a minor He3 enrichment, as depicted in Fig. 5. In this figure very weak increase of the He3 concentration in the period of 0–30 h is connected with the decrease of temperature to 2.18 K (lambda transition). During this process the difference between the partial pressures of He4 and He3 is increasing (compare description of the Figs. 2 and 3). Next part of this plot, after 30 h, describes the capillary effect. Capillary (as a part of the heat exchanger) with a thin wire inside, can also create percolation paths necessary for quantum filtration. The initial increase in He3 concentration connected with filtration effect is indicated by the arrow where the lambda transition is detected by thermometer located in the vicinity of the capillary - the blue line in Fig. 4. To get this result, very slow experiment were performed - longer observation allowed for the clear indication of the point in which lambda front starts. This way we show also that capillary plays an additional important role in the described construction by slightly supporting the whole separation process, even above lambda transition.

The main separation process was initiated with the heater located just above the entropy filter. After the heater is switched on the temperature reached the level of 1.75 K and was stable during the filtration process. He4 transferred through the entropy filter was removed with the same pumping line and stifle valve used for stabilization of temperatures (and pressures) in front of and behind the filter. Filtration process took about 36 h and 18 l of helium was filtrated. Starting and final levels of He3 are shown in Fig. 6. Results in Fig. 6a) and b) show that our apparatus with only 25 l of He4/He3 mixture and initial He3 concentration at the level of 0.2 ppm (Fig. 6a) efficiently filtrated He3 giving serious increasing concentration of He3 in the He4/He3 mixture. The final concentration of He3 was 2.4 ppm (Fig. 6b).

Additional peaks shown in Fig. 6 at mass 1 and 2 came from different hydrogen ions. These peaks are always present in mass spectra, however, their amplitudes are lower than the background line of the empty chamber. We decided to present these peaks together with the helium peaks (mass 3 and 4) to show that the level of purity in our experiment was very high. Hydrogen ions with mass 1 and 2 can combine, resulting in overestimated intensity of the mass 3 peak [23], thus disturbing the main result. Fortunately, similar intensities of these peaks measured before and after the filtration process ensured us that no additional hydrogen appeared during the experiment and the observed change of the intensity of the He3 and He4 peaks reflected the

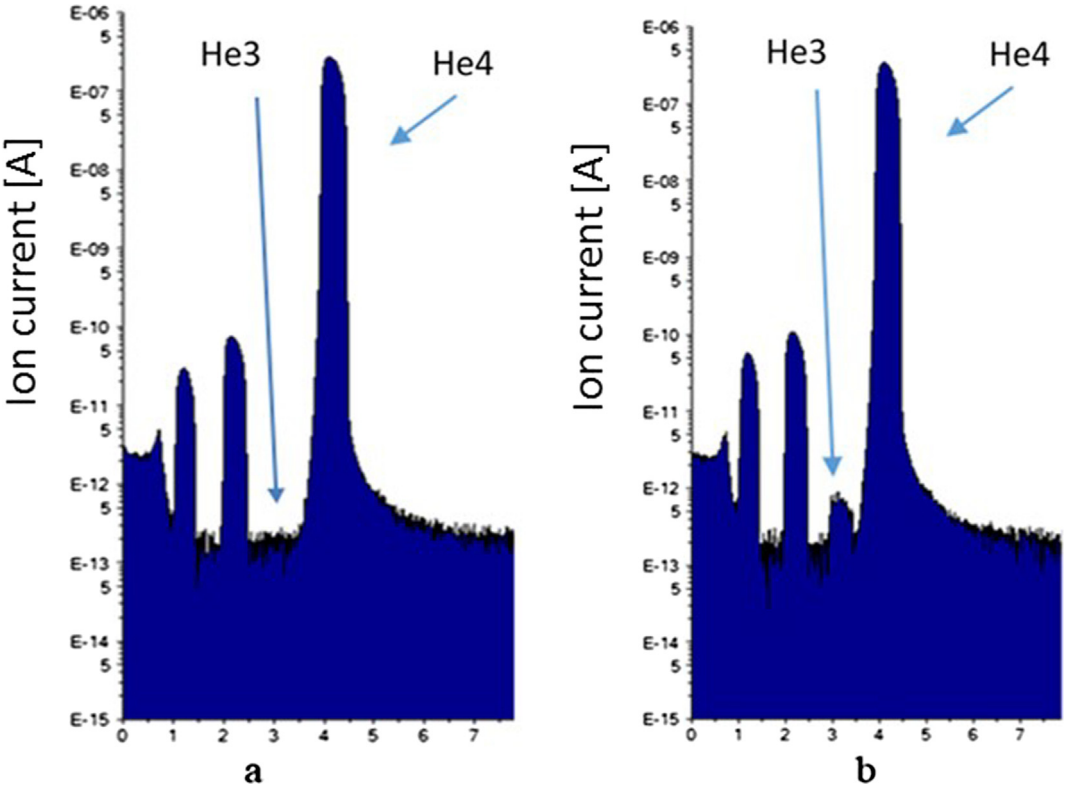


**Fig. 6.** He3 level in the beginning of the experiment (a) and in the end (b).

real change in the concentration of both components.

The most important feature of the presented cryostat is the heat exchanger with a capillary located close to the entropy filter. It enabled the observation of the moving front lambda during the cooling procedure and effective liquid helium enrichment with He3 isotope. Smooth increase of He3 concentration was observed during the whole cooling procedure, also above the lambda transition and long period of lambda front movement. Presented experiment is the first laboratory step to construct the large-scale industrial flow-separator dedicated to collecting the He3 from liquid helium at the low temperature region. Main parameters and technological solutions of this cryostat were published in paper [24]. Described technical solutions can be used to perform tests aiming at comparison of various kinds of entropy filters based on new types of nanomaterials, amongst them the special class of materials called auxetics [25].

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## Declarations of interest

The authors declared that there is no declarations of interest.

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