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Continuous Flow System for Controlling Phases Separation Near λ Transition

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Abstract. As demands on 3He are increasing and conventional 3He production through tritium decay is decreasing, alternative 3He production methods are becoming economically viable. One such possibility is to use entropy filters for extraction of the 3He isotope from natural gas. According to the phase diagram of the 3He, its solidification is impossible by only lowering of the temperature. Hence during the cooling process at stable pressure we can reach λ-point and pass to the special phase – He II. The total density of HeII is a sum of the two phases: *normal* the *superfluid* ones. It is possible to separate these two phases with an entropy filter – the barrier for the classically-behaving normal phase. This barrier can also be used to separate the two main isotopes of He: 4He and 3He, because at temperatures close to the 4He-λ-point the 3He isotope is part of the normal phase. The paper presents continuous flow schemes of different separation methods of 3He from helium commodity coming from natural gas cryogenic processing. An overall thermodynamic efficiency of the 3He/4He separation process is presented. A simplified model of continuous flow HeI –HeII recuperative heat exchanger is given. Ceramic and carbon porous plugs have been tested in entropy filter applications.

Keywords: Helium, entropy filter, separation.
PACS: 67.60.-g, 67.25.D

INTRODUCTION

The world is experiencing a shortage of 3He, a rare isotope of helium which is crucial in many applications such as homeland security, national security, medicine, industry, and science. Up to now, the majority of helium-3 was obtained through tritium decay as a byproduct of tritium production for nuclear weapons programs. Tritium decays into helium-3 and it was the need for tritium of the nuclear weapons programs, not the demand for helium-3 itself, that determined the amount of 3He produced. Due to disarmament policies, a dominant source of 3He – nuclear tritium decay is in a global decrease phase, creating a serious 3He supply problem. Moreover, the situation is aggravated by deployment of neutron detectors, especially at US borders. According to [1], potential additional sources of 3He include increased production of tritium in light-water nuclear reactors (beyond the amount already produced for the weapons program); extraction of tritium produced as a byproduct in commercial heavy-water nuclear reactors; production of either tritium or helium-3 using particle accelerators; and extraction of naturally occurring helium-3 from natural gas or the atmosphere. Until recently these alternative sources were not considered economically attractive. With the current shortage, this consideration may change.

This makes alternative 3He sources, like helium commonly separated from natural gas, potentially competitive for the present 3He price level. In spite of very low 3He concentration in the helium extracted from natural gas, the 3He/4He mixture separation seems to be an attractive alternative that may allow overcoming the 3He supply problem. Assuming 3He concentration in 4He at 0.1 ppm level, the thermodynamic minimum for the light isotope separation is 43 kJ/mol. The thermodynamic minimum has been calculated as a work of isothermal compression of the gas from its partial to ambient pressure. However, the real power demand probably exceeds the thermodynamic minimum by many orders of magnitude and all relevant factors are not yet fully recognized. Potentially feasible technologies that enable 3He separation from helium rectified from natural gas require helium liquefaction and cooling down to temperature of about 1,5 K, i.e bellow λ transition. This allows 3He separation using entropy filters or rectification columns.

Helium-3 separation from commodity helium was investigated in the middle of XX century [2, 3]. A number of experiments have been performed to demonstrate the possibility of 3He separation and to verify the practical

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concentration limits. The experiments used the thermo-mechanical effect, a rectification column or the combination of two. The experiments showed that the 3He content enrichment in the mixture of initial content of the light isotope equal to about 10^{-8} was possible up to 0,01%, i.e. four orders of magnitude. Figure 1 shows the schemes of the 3He separation test stands developed in the former Soviet Union.

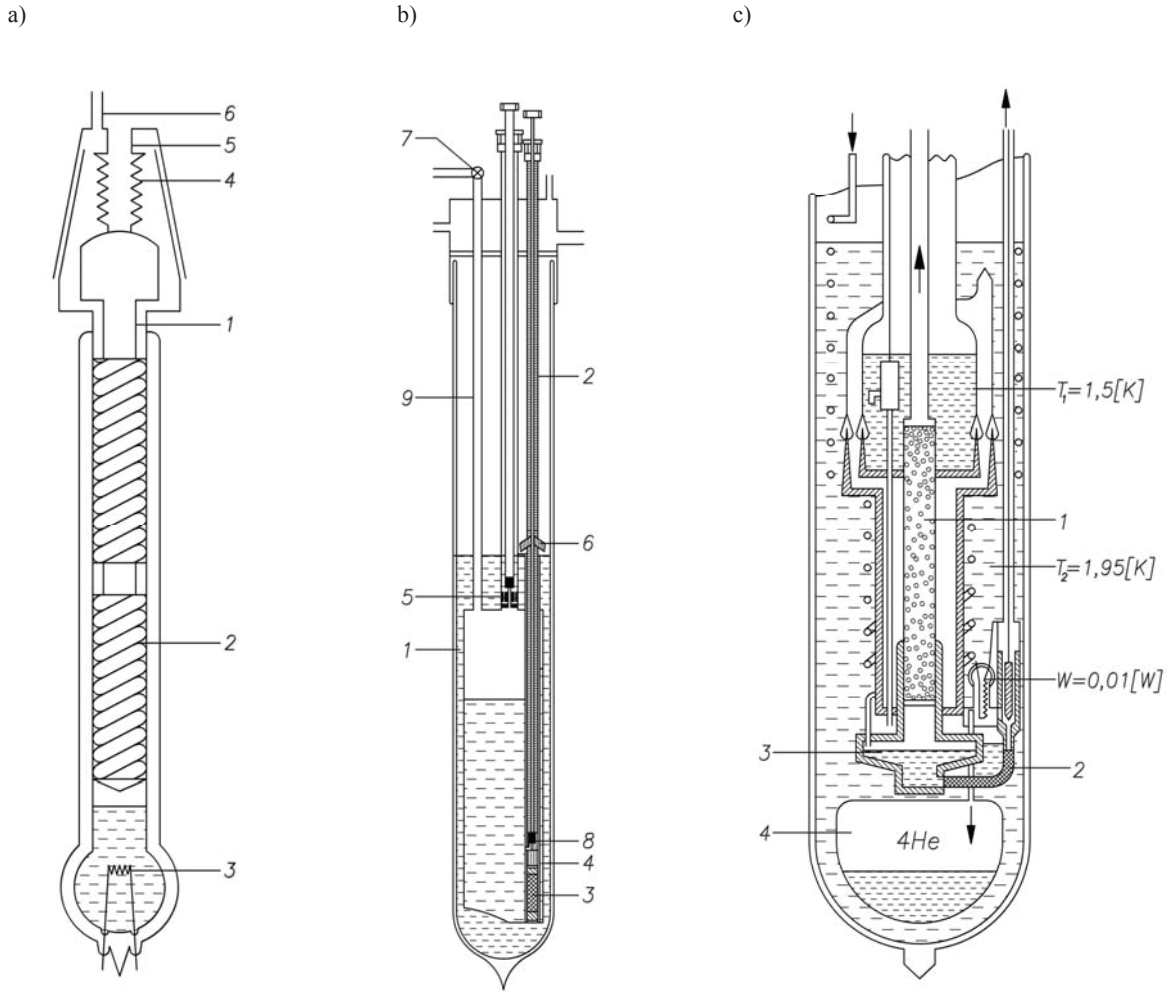

FIGURE 1. Schemes of 3He/4He separation cryostats [3], a) – rectification column, 1 – copper screw with thread, 2 – glass pipe, 3 – heater, 4 – bellow, 5 – support pipe, 6 – helium outflow pipe; b) - separation cryostat with entropy filter, 1 – brass vessel, 2 – pipe, 3 – entropy filter, 4 – heater, 5 – valve, 6 – fountain outflow slot, 7 – valve, 8 – valve, 9 – helium outflow pipe; c) - cryostat with combined thermo-mechanical effect and rectification column, 1 – rectification column, 2 – entropy filter, 3 – evaporator, 4- 4He vessel after the entropy filter.

Rectification column shown in Figure 1.a) was 150 mm long and the power of heater 3 was $5x10^{-3}$ W. Initial 3He content was 0.01%. The mixture leaving the column via the pipe 6 was enriched with 3He to 1,5%. Moreover cascaded operation is possible: the same column supplied with a mixture of 1 – 2 % 3He enriched it up to 53%.

Thermo-mechanical effect cryostat depicted in Figure 1.b) allowed 3He enrichment from about 10^{-8} to 10^{-4} (molar content). Further enrichment was possible with rectification column organized as a separate device. The device depicted in Figure 1.c) combined the entropy filter with the flow forced by thermo-mechanical effect and rectification column.

The presented apparatuses were characterized by practically cyclic operations and low capacity not exceeding 100 liters of processed mixture at normal conditions. Nevertheless they demonstrated the feasibility of the entropy filter for 3He separation starting from initial content characteristic for commodity helium separated from natural gas.

The devices were not optimized from power consumption point of view and were not considered as competitive towards the tritium decay method.

CONTINUOUS FLOW SYSTEM FOR 3HE SEPARATION

Assuming an initial content of 3He at the level of 10^{-7} only high capacity installations may produce a light helium isotope in commercially significant conditions. To illustrate, under the assumption of full 3He recovery, the production of 1 normal liter of 3He requires processing 1.8 ton of commodity helium. It means that the capacity of a prospective 3He separation plant should in practice match the capacity of helium recovery from natural gas installation. The process must be oriented towards low power consumption, making vast use of heat recuperation.

Figure 2 shows a flow scheme of continuous separation process. In the 3He separator an incoming stream *m* of the liquid helium is split into two streams at point 1. The stream *x* supplies the refrigeration Joule-Thomson loop, while the *m-x* stream is first cooled-down to about 1.5 K to be entropy filtered for 3He enrichment in the separator vessel before the entropy filter.

The stream *x* flows through a high pressure channel of a recuperative heat exchanger (Recuperator 1). Helium at about 1 bar cools down and then it is throttled in the Joule-Thomson valve reaching a pressure of about 5 mbar. Downstream the valve, a low pressure is maintained by a warm vacuum pump. The originated cooling power is used to cool down the *m-x* helium stream aimed at further filtration. The *m-x* stream of the helium is first pre-cooled in a recuperative heat exchanger 2.

The separator enables gathering 3He enriched helium and its later removal with the vapors leaving the vessel. The ratio of 3He and 4He partial pressures at 1.5 K is of about 15. The liquid from the separator is filtered at a porous entropy filter allowing a flow of superfluid component only. Light helium isotope 3He remains at 1.5 at normal state and is not transferred through the filter except by diffusion. The driving force of the entropy free superfluid component transfer through the filter is a temperature difference imposed by the heater and/or recuperative heat exchanger 2.

After passing the entropy filter and warm-up at recuperative heat exchanger 2, the helium leaves the system in a liquid phase and is returned to commodity helium container. Not taking into the account the 3He enriched vapor stream, the only helium vaporized and leaving the system in a gaseous phase is the *x* stream. The system optimization requires the ratio *(m-x)/x* as high as possible. This will guarantee a low consumption of energy defined by the pumping and further recondensation of *x* stream. The characteristic feature of the system is that the bulk of helium that forms the *m-x* stream is returned as a liquid and therefore does not entail additional energy consumption.

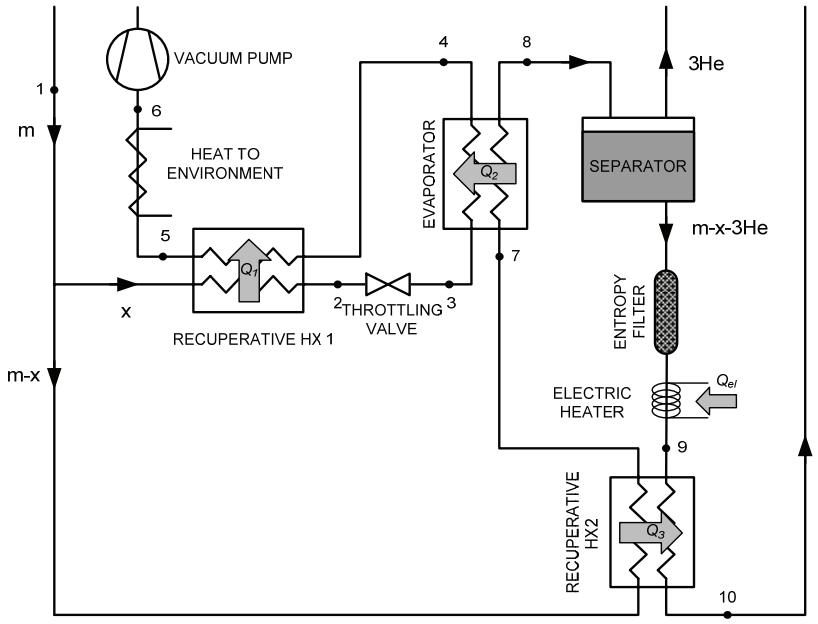

FIGURE 2. Flow scheme of 3He separation process

Selection of the Separation Temperature

In the presented system a final enrichment of the 3He content in the vapor leaving the separator is realized in two processes: first the content of 3He is increased due to HeII superfluid component passage through the entropy filter (and leaving a normal component with 3He isotope behind), second the mixture is rectified due to the large ratio of 3He and 4He partial pressures in the separator. Since the entropy filter blocks flow of HeII normal component and 3He isotope, the liquid enrichment by the filter is inversely proportional to the normal component fraction in HeII. In accordance with Two-Fluid Model the enrichment will increase with a decrease of the separation temperature. Similarly, the content of 3He in the separator vapor will increase with the temperature decrease. The pressure of saturated helium vapor in the evaporator will decrease with the separator temperature decrease. Thus, for a fixed pumping capacity (in m^3/hour) of the vacuum pump, the cooling capacity of the JT loop and in consequence, the *m-x* helium mass stream will decrease. Taking as the optimization objective function the mass stream of the filtered helium, an optimal filtration temperature of 1.5 K has been estimated – see figure 3. The maximum of the curve "Filtration optimization product" indicates the temperature when the highest amount *x* of the helium can be filtered with a given vacuum pump, according to the scheme depicted in figure 2.

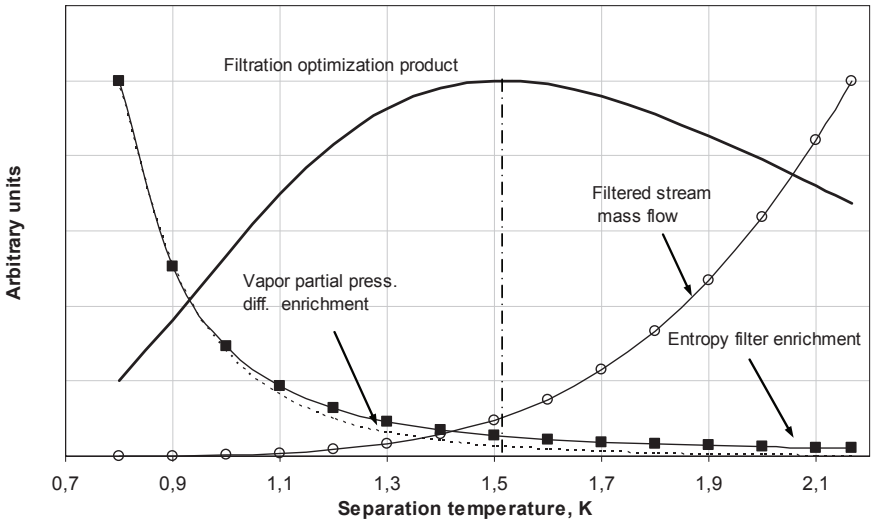

FIGURE 3. Optimization of separation temperature.

Calculation Methodology of Recuperative2 Heat Exchanger

Assuming a perfect system insulation and neglecting the pressure drops in pipes and heat exchangers as well as temperature difference in the evaporator, the mass flow *x* through the Joule-Thomson cooling loop can be calculated from equation 1:

$$x = \rho_6 \cdot S\,, \tag{1}$$

where: ρ_6 is the density of the helium at 300 K and pressure 5 mbar (saturation pressure of helium at 1.5K) and S stands for vacuum pump capacity.

The cooling power of the Joule-Thomson loop Q_2 is given by:

$$Q_2 = x \cdot (h_4 - h_3)\,, \tag{2}$$

where *h* is the enthalpy.

The efficiencies of recuperative heat exchangers 1 and 2 can be defined by equations (3). Since the h_3=h_2 (isenthalpic throttling process on the JT valve) and x, as well as S and ρ_6 are constant, it can be found from equation (2) that the cooling power Q_2 increases when h_2 decreases. The enthalpy and temperature in points 2 and 7 depend on the efficiencies of the recuperators ε_{R1} and ε_{R2} which can be defined as:

$$\varepsilon_{R1} = \frac{h_1 - h_2}{h_1 - h_{2T}}, \quad \varepsilon_{R2} = \frac{h_1 - h_7}{h_1 - h_{7T}}, \tag{3}$$

where h_{2T} , h_{7T} are minimal enthalpies that can be achieved in points 2 and 7, assuming 1.5 K helium temperature at the inlets to the recuperators, i.e. helium enthalpy for temperature 1.5K (temperature of helium after the entropy filter inflowing to the recuperator 2 in point 9) and 1 bar pressure. It can be found that $h_{7T} = h_8$.

The Joule-Thomson loop cooling power Q_2 is causing the temperature drop of the filtered helium stream m-x from T_7 to T_8:

$$Q_2 = (m - x) \cdot (h_7 - h_8) \tag{4}$$

Rearranging the equations (2) – (4) allows finding the relation of $(m - x)/x$ ratio and efficiencies of recuperative heat exchangers – equation 5:

$$\frac{m - x}{x} = \frac{h_4 - h_1 + \varepsilon_{R1}(h_1 - h_{2T})}{h_1 - h_8 - \varepsilon_{R2}(h_1 - h_8)}. \tag{5}$$

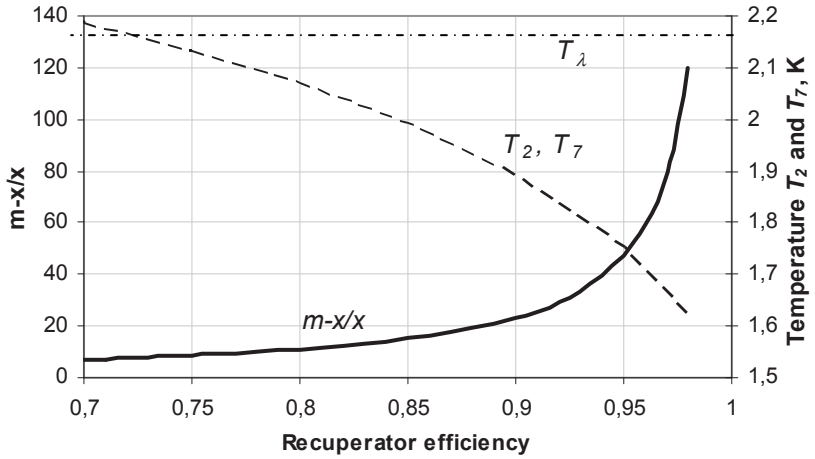

FIGURE 3. Filtration ratio $(m - x)/x$ and temperature of helium streams after the recuperators in function of the recuperators efficiency ($\varepsilon_{R1} = \varepsilon_{R2}$).

Figure 3 presents the $(m - x)/x$ ratio and temperatures of helium streams after the recuperators, calculated from equation (6) in function of the recuperators' efficiency (assuming $\varepsilon_{R1} = \varepsilon_{R2}$). It is visible from Figure 3 that high filtration ratio $(m - x)/x$ requires that the temperatures T2 and T7 are well below the lambda point. It means that the lambda transition has to be observed in the heat exchangers' channels.

Simplified mathematical model of recuperative heat exchanger 2

Recuperator 2 is designed as a tube-in-tube heat exchanger. In a steady state conditions the energy balance for high temperature side of the exchange is given by:

$$m\frac{dh_h}{dx_h} + A_h \frac{dq_{liq\_h}}{dx_h} + \frac{dQ_{HX}}{dx_h} = 0, \tag{6}$$

while for the low temperature side is:

$$-m\frac{dh_l}{dx_l}-A_l\frac{dq_{liq\_l}}{dx_l}-\frac{dQ_{HX}}{dx_l}=0\,, \tag{7}$$

where: m is liquid helium mass flow, h is enthalpy, q_{liq} is liquid heat conduction, A is cross section of the liquid in normal direction to the flow, Q_{HX} is the heat transfer thought the heat exchanger material. Indexes h and l are corresponding to the high and low temperature helium flows.

Assuming relatively low velocities (below speed of sound) in the heat exchanger channels, the heat conduction along the liquid for normal and superfluid helium state ($T \le T_\lambda$) can be calculated as:

$$q_{liq}=\begin{cases}-\lambda\dfrac{dT}{dx} & for \quad T>T_\lambda\,,\\ -\left(\dfrac{1}{f(T,p)}\dfrac{dT}{dx}\right)^m & for\ T\le T_\lambda\,,\end{cases} \tag{8}$$

where λ is the thermal conduction of the liquid at normal state [W/mK], *f(T,p)* is the superfluid helium heat conductivity function [W^3/m^5K], m is exponent value resulting from superfluid helium heat transfer power law.

The heat transfer thought the heat exchanger material is:

$$Q_{HX}=k_L\cdot dx\left(T_h-T_l\right), \tag{9}$$

where dx is elementary length of the heat exchanger [m] and k_L is the heat exchanger linear thermal conductance [w/mK], defined as

$$k=\frac{2\pi}{\dfrac{1}{\alpha_i\dfrac{d_i}{2}}+\dfrac{1}{\lambda_s}\ln\left(\dfrac{d_e}{d_i}\right)+\dfrac{1}{\alpha_e\dfrac{d_e}{2}}}\,, \tag{10}$$

where: λ_s – heat exchanger tube material thermal conductivity, α_i, α_e –heat transfer coefficient for internal/external side of the tube, d_i, d_e – internal/external diameter of the pipe .

The heat transfer coefficient for the helium at normal state can be found from the following equation:

$$\alpha=\frac{Nu\cdot\lambda}{d}\,, \tag{11}$$

where: Nu – dimensionless Nusselt number; λ – fluid thermal conductivity, d – diameter wetted by the fluid.

Nusselt number for internal flow through a smooth pipe can be calculated from [4]:

$$Nu_i=0.026\cdot \mathrm{Re}_w^{0.8}\,\mathrm{Pr}_w^{0.33}\,. \tag{12}$$

The heat transfer coefficient for the helium at superfluid state can be approximated by the Kapitza conductance:

$$\alpha=aT^3\,, \tag{13}$$

where a is the experimental constant and T is helium temperature.

The set of equations (6) – (13) has been solved with the use of finite elements method, where the element length was chosen as 10 mm. At the current system design phase the different geometries of the recuperator 2 are being examined to find the minimum recuperator length, allowing to obtain sufficiently low temperature T_7 for a vast range of helium mass stream. The criterion is $T_7 < 1.6$ K. Two sizes of the market available inner tubes for high temperature stream (6×1 mm and 8×1 mm) and outer tubes for low temperature streams (12×1.0 and 14×1mm) are taken into the account. The influence of the high temperature cross section to low temperature cross section ratio

(A_h/A_l) can be determined. The modeling program considers examination of 3 different heat exchanger lengths: 20, 30 and 40 m.

A smooth cooper tube has been proposed for the heat exchanger. The value of the experimental constant a in equation (14) is a=450 W/m^{2}K^4 [5]. The calculations are performed for inlet high helium temperature (point 1 in Figure 2) equal to 4.2 K and inlet low temperature (point 10 in Figure 2) equal to 1.5K.

Oerlikon Sogevac SV630 B rotary vane vacuum pump has been chosen. The pump is characterized by nominal pumping speed of S = 630 m^3/h at 50Hz. It results from equation (1) that the mass stream x of helium circulated in the refrigeration JT loop will be of about 0.1g/s. According to equation (6) and taking into the account the results presented in Figure 3, it can be expected that the mass stream of helium flowing through the recuperator 2 will be of the order of a few g/s. In the present analysis recuperator 2 mass streams equal to 1, 2 and 5 g/s have been assumed.

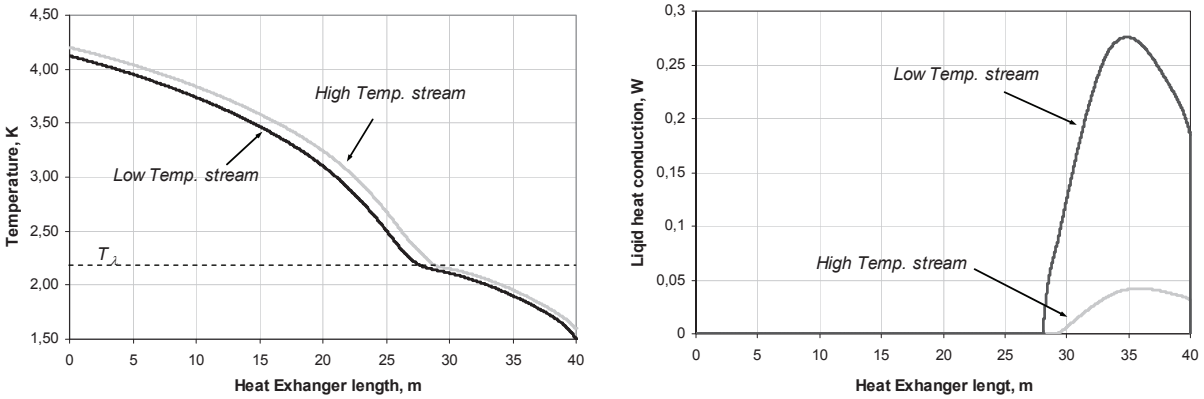

FIGURE 4. Exemplary modeling results – 8x1, 12x1, q_m=5g/s

Figure 4 present the exemplary modeling results of the high and the low temperature streams temperature profile and heat transfer along the streams for inner tube 8x1 mm, outer tube 12x1 mm and m-x=5g/s case.

a) b)

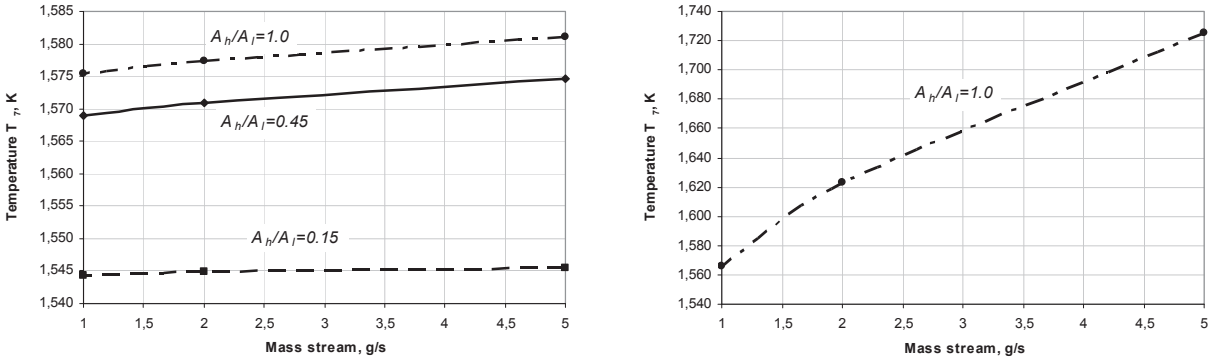

FIGURE 5. High temperature stream T_7 at the outlet from recuperative heat exchanger 2 – modeling results, a) heat exchanger length 40 m, b) heat exchanger length 20 m

Figure 5a shows that for the 40 m long recuperative heat exchanger the temperature at the outlet is below 1.6 K and it is not very sensitive to the mass stream and tubes' cross sections ratio values. It can be also noted that the outlet temperature is decreasing with A_h/A_l ratio. This can be explained by the decrease of the heat conducted through the superfluid helium along the heat exchanger. For a shorter heat exchanger (Figure 5b) the influence of the mass stream on the temperature of the outlet helium T_7 is visible.

Feasibility tests of entropy filters

A number of entropy filters have been tested at Institute for Molecular Research in Poznan, Poland, to evaluate the optimal power of the heater and the filter overall efficiency. The filters have been based on ceramic and carbon materials. Figure 6 presents a glass test rig that allows direct observation of the fountain effect with respect to the heater power (left) and exemplary characteristic of a filter (right). The observed fountain height allows calculations of allowable pressure differences in cryostat channels filled with the filtered helium.

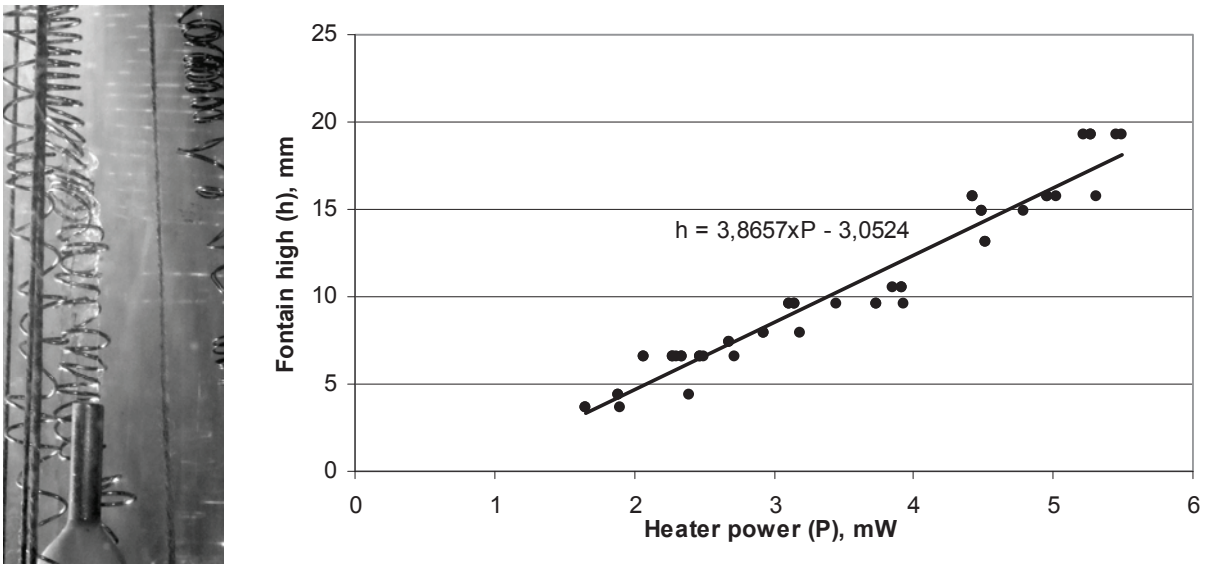

FIGURE 5. Entropy filter, test rig – left, exemplary characteristic for carbon porous plug filter - right

CONCLUSIONS

World is observing a global shortage of 3He. Due to increases in consumption and decreases of production from tritium decay process, the price of light helium isotope has significantly increased. This makes economically justified 3He recovery from commodity helium separated from natural gas. A continuous flow 3He separation system based on superfluid helium entropy filtering has been proposed. The system guarantees exergy recovery from filtered helium thus limiting energy consumption. A prototype test cryostat is under construction and various entropy filters including carbon nano structures are under investigation. The challenging elements of the cryostat are recuperative heat exchangers with lambda transition inside. A simplified model of the heat exchanger has properly reflected the λ transition along the channel.

ACKNOWLEDGMENTS

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