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Separator of 3He isotope from liquid 4He

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Content

- 3He applications and sources, motivation of work
- 3He separator from liquid 4He
  - Separation mechanisms
  - Optimum separation temperature
  - Separator conceptual design
- Challenges in the separator design
  - Heat exchangers with λ transition design
  - Entropy filter geometry
  - Determination of the 3He concentration
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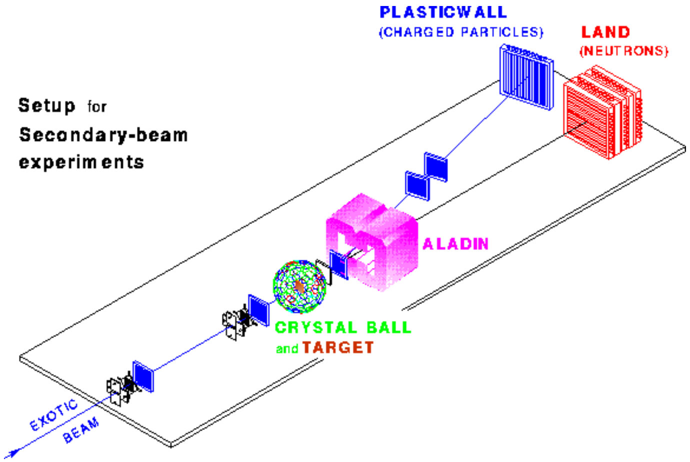
Helium-3 applications

- Neutron-emitting radioactive materials detection at the border and in ships

- Large Area Neutron Detectors for science

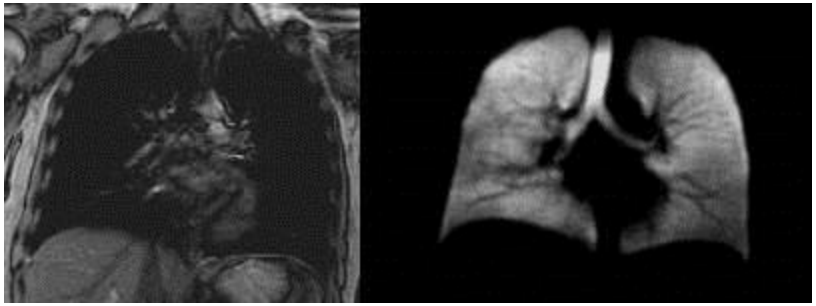


Helium-3 applications

MRI image of the lungs

Gas mixtures containing the hyperpolarized helium-3 gas can be imaged with an MRI scanner to produce anatomical and functional images of lung ventilation

KEOPSYS



discovery.com

Helium-3 applications

Dilution refrigeration

Possibility to produce down to 2 mK

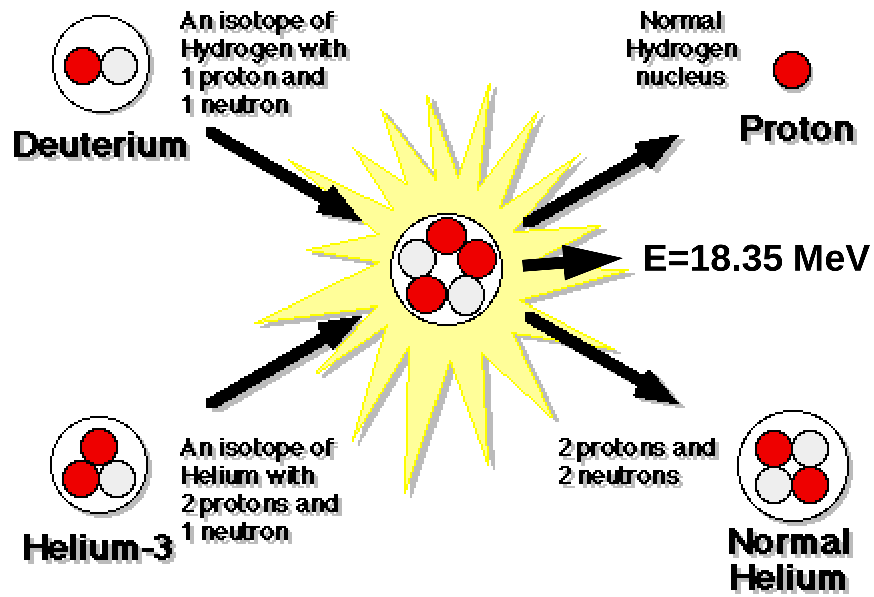
Janis Research

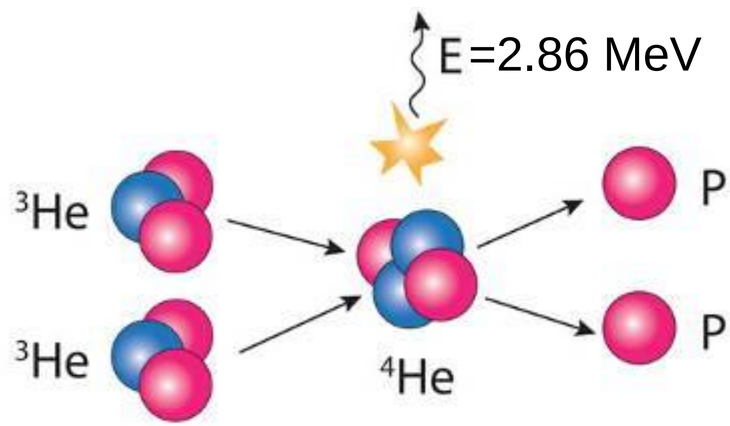
Helium-3 applications

Second and third generation fusion fuel

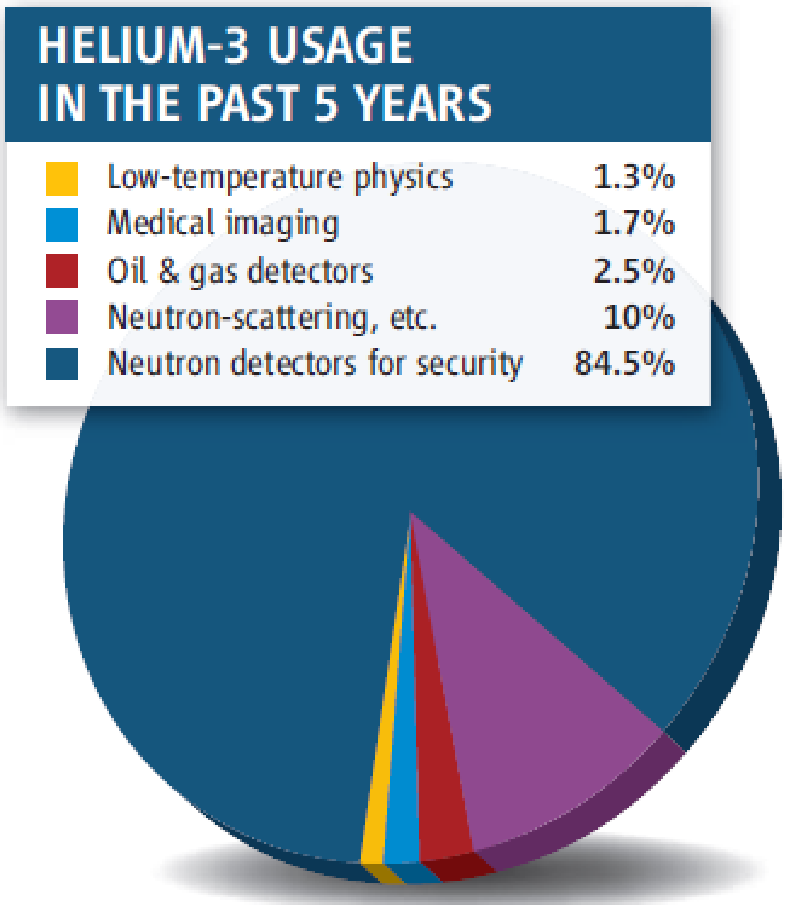
$^{2}D + {}^{3}He$ reaction can provide the higher amount of energy from fusion with much smaller activation of the materials

$^{3}He + {}^{3}He$ produces no neutrons= no material activation/clean energy



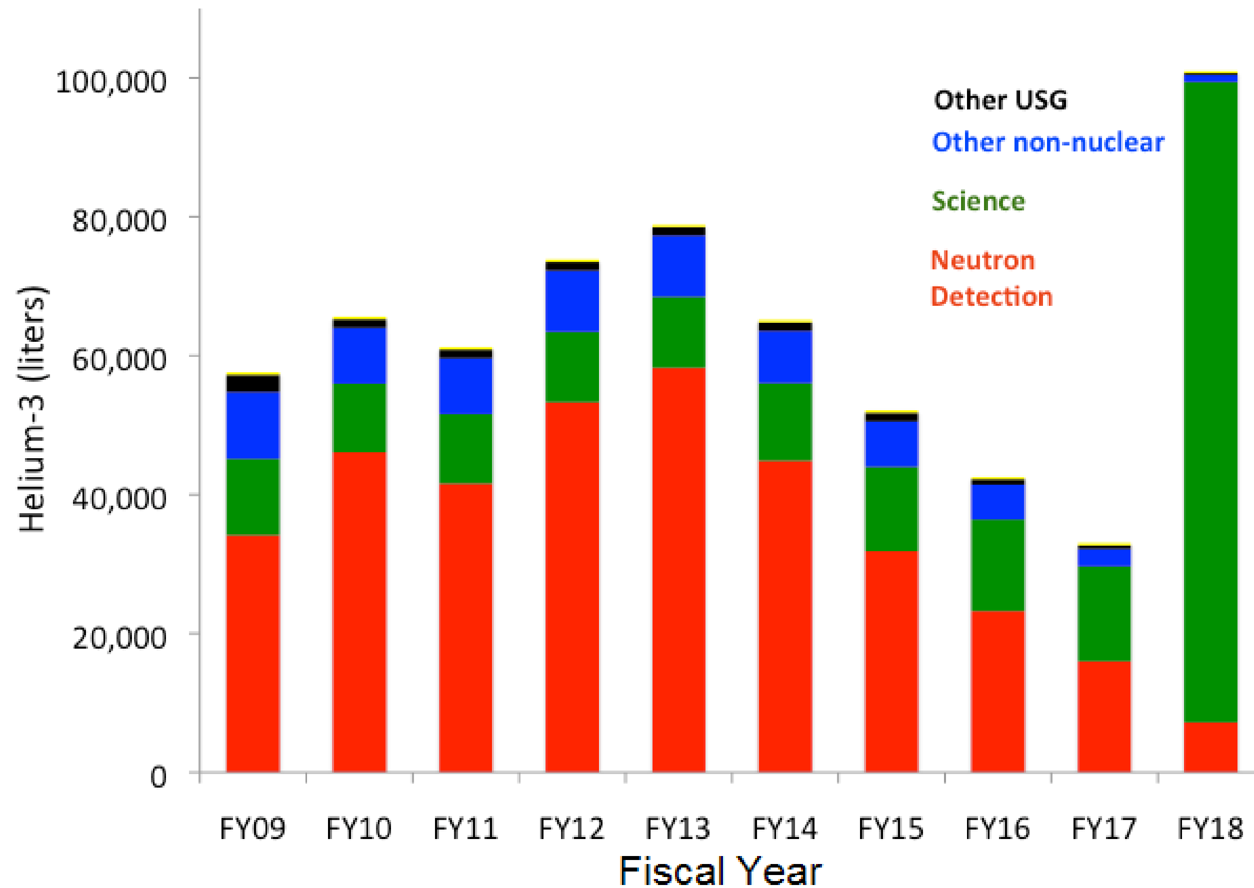


Helium-3 applications



www.sciencemag.org

Helium-3 demand



U.S. Congressional Research Service*: The Helium-3 Shortage: Supply, Demand, and Options for Congress.*

Helium-3 production

Nuclear weapon maintains

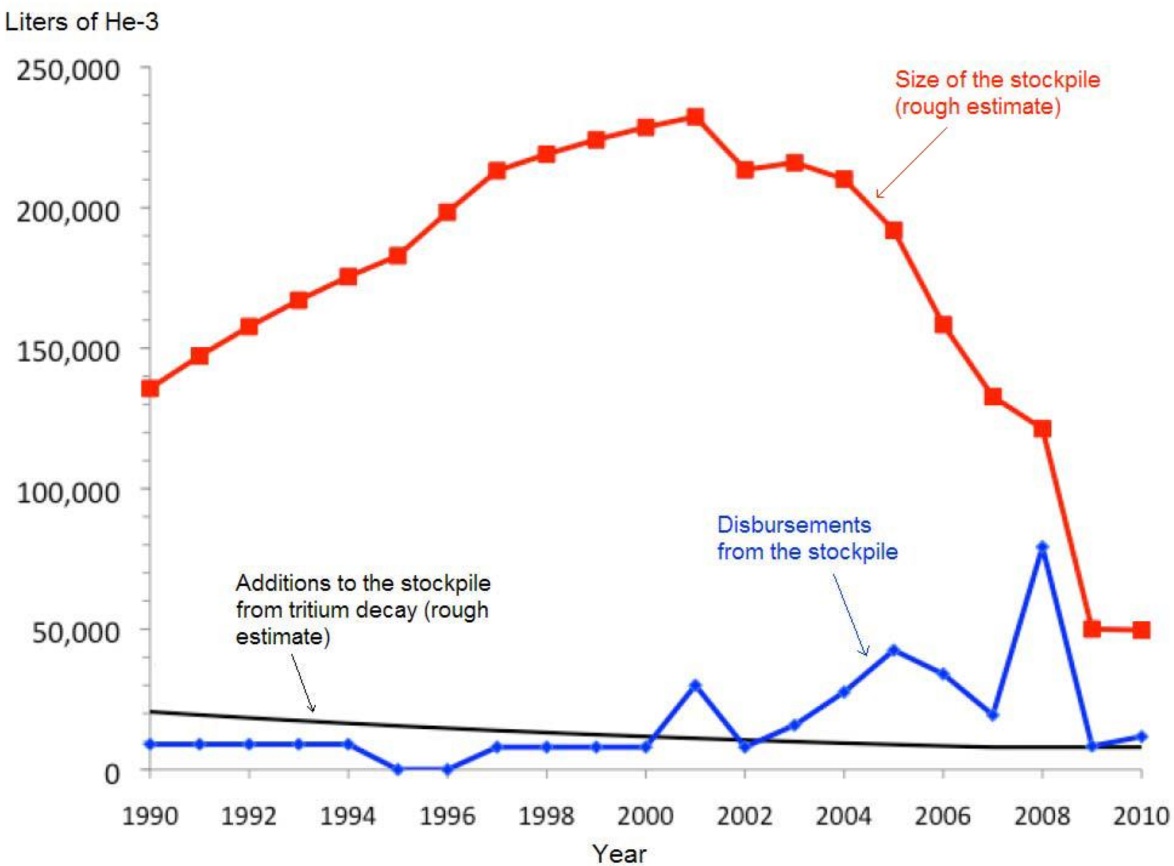
It is the mine source of 3He as it is produced due to tritium decay.

Due to tritium decay (half-life 12.3 years) it need to be exchange each few years

Because of nuclear weapon program over the World is reduced, the main source of 3He is also reduced

Current yearly production of 3He from nuclear weapon in U.S. is estimated for 8 000 liters

Helium-3 U.S. production and stockpile



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Helium-3 production

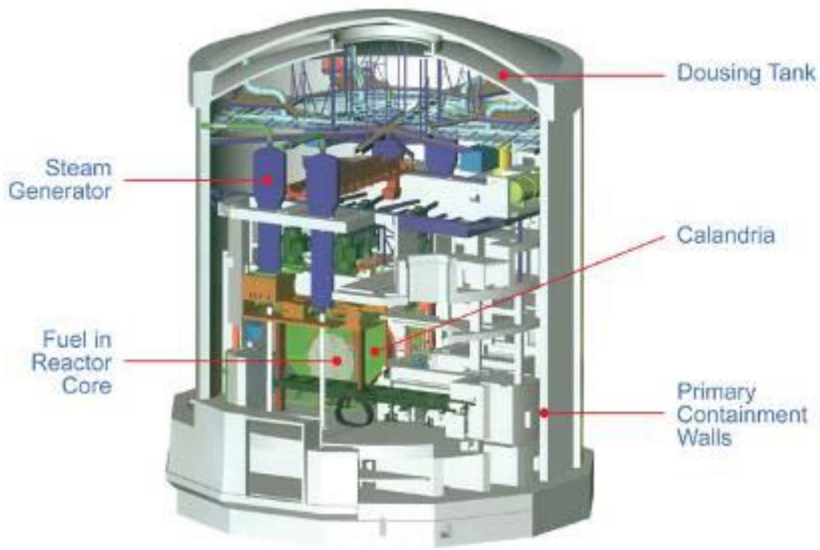
Nuclear power plant

Increase production of tritium in the light-water-reaction, or

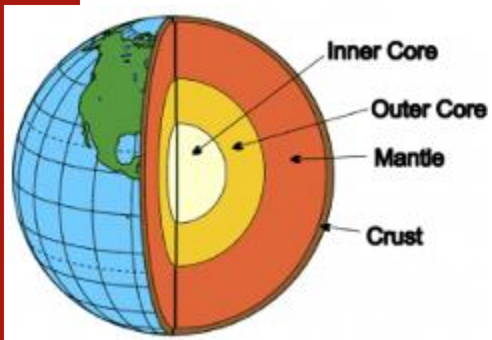
Start tritium extraction in commercial heavy-water nuclear reactors

Due to tritium decay (half-life 12.3 years) the 3He will be available in few years now

Canadian CANDU Reactor

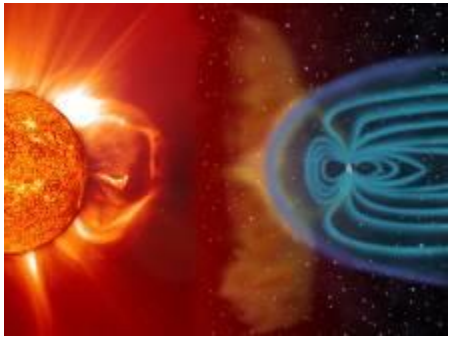


Helium-3 production – alternative sources



Earth's crust and mantle
3He/4He = 200-300 ppm
3He Inventory = 100-1000 kT

Disadvantages: access, environment degradation



Solar wind
3He/4He = 480 ppm
3He/Wind = 20 ppm

Disadvantages: access, lack of technology

Earth's atmosphere
3He/4He = 1.38 ppm
3He/air = 7.2 ppt
3He Inventory = 37 kT

Disadvantages: cost of cryogenic separation of He from air

Moon regolith
3He/regolith = 15 - 50 ppb
3He Inventory = 1000 kT

Disadvantages: access, lack of technology

Helium-3 production - alternative sources

Natural Gas

3He/4He = 0.5 - 5 ppm
3He/NG = 2.5 - 25 ppt

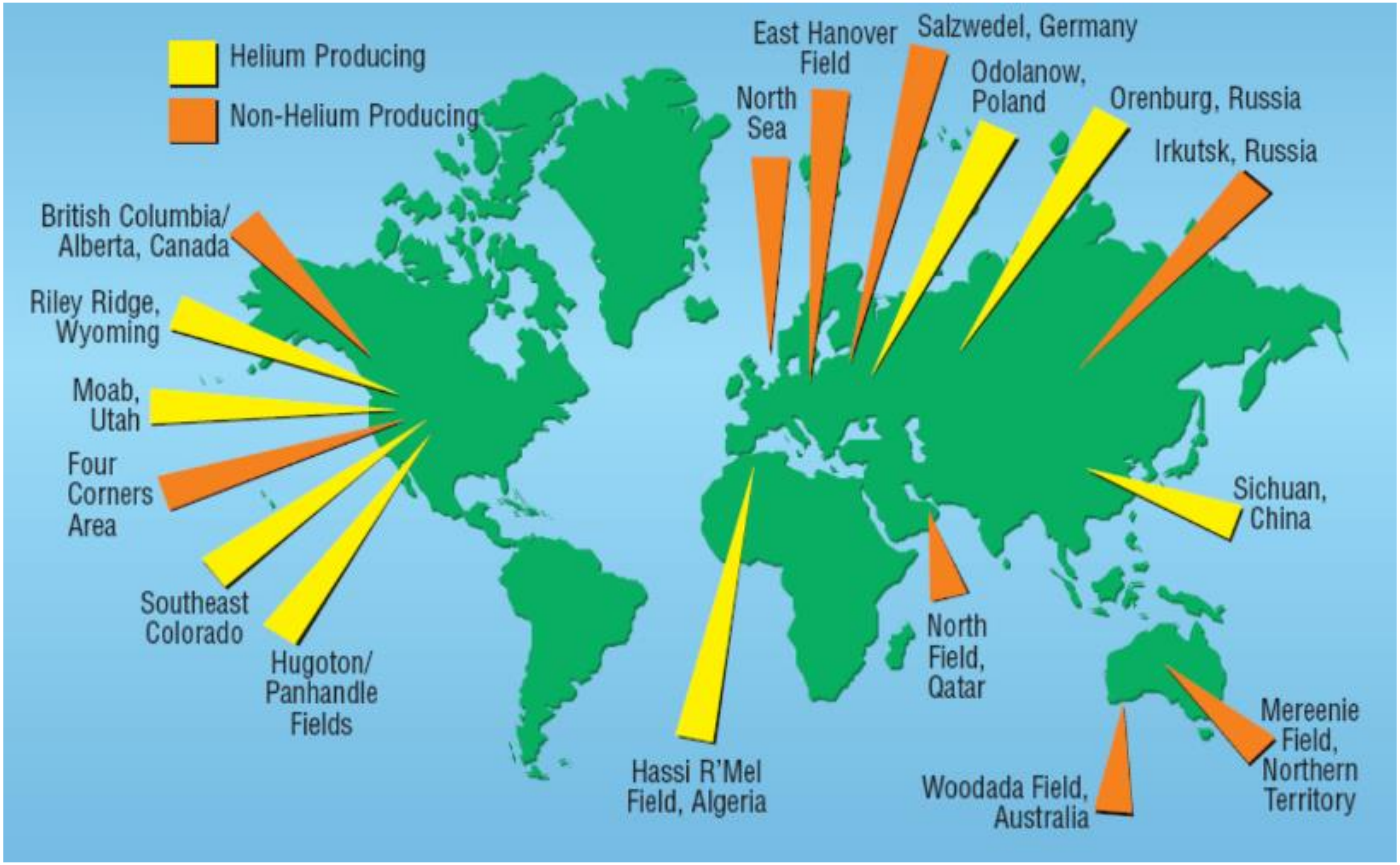
Advantages:

- Unlike air, costs of helium liquefaction are partially covered by the natural gas cryogenic purification costs

- After 3He separation from liquid LHe, the liquid LHe don’t losing the market value

-<u>Separation of 3He from the Natural Gas can be economically justified at the liquid helium production plant sites</u>

World production of commodity helium



Potential of 3He production by separation from commodity helium

Helium global production from NG is:

75 tons/day = 27 375 tons/year

Asuming 0.2 ppm of 3He in 4He

A yearly production of 3He potential is 4500 Ndm^3

Low-temperature physicists say they need between 2500 and 4500 liters of helium-3 per year, primarily to fill new dilution refrigerators.

Polish Oil and Gas Company (PGNiG) branch Odolanow

Raw feed gas stream: 136 000 NCMH

LHe production: 2 000 kg/day

3He potential production:
0.42 g/day = 3.5 Ndm^3/day = 1.27 NCM/year

Motivation of work

- The global helium-3 shortage is visible from a few year now
- One of alternative source of helium-3 production is Natural Gas
- The economical justified production of helium-3 isotope seems to be the NG purification plant coupled with liquid helium production
- Therefore, Wroclaw University of Technology in cooperation with PGNiG branch Odolanow and Institute of Molecular Physics of Polish Academy of Science is developing and will be testing the Separator of 3He from liquid helium

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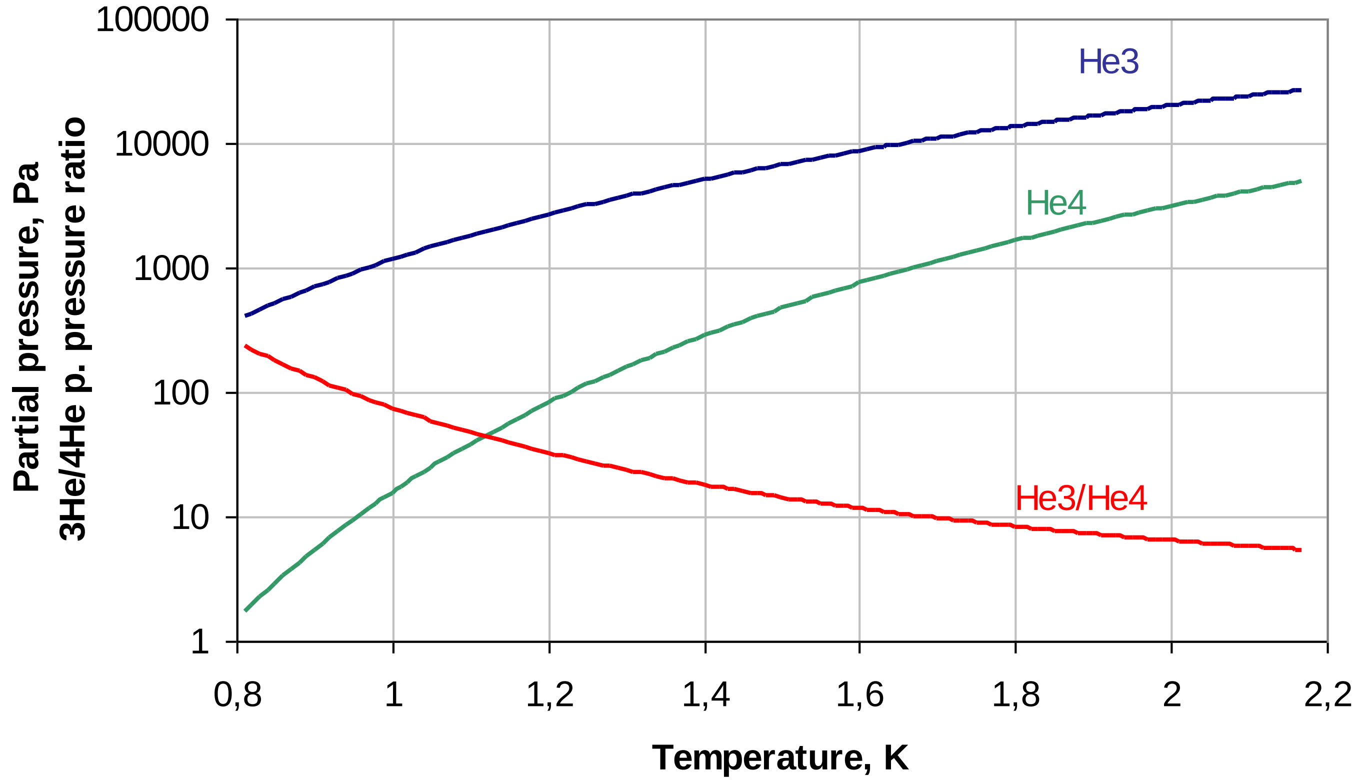
Separation due to partial pressure of 3He and 4He difference



$$\frac{p_{3He}}{p_{4He}} = \exp\left(4.31 \cdot T_{sep}{}^{-1.21}\right)$$

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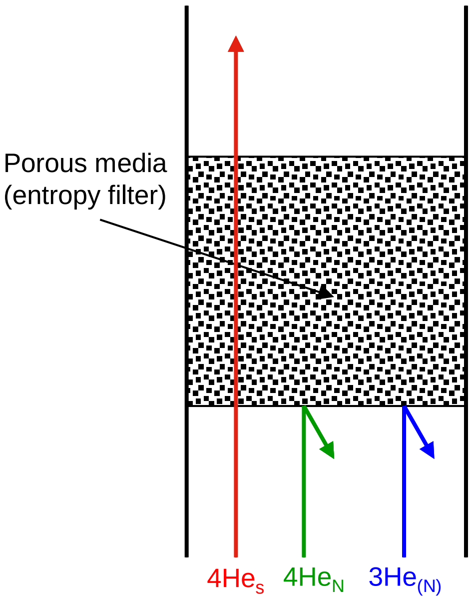
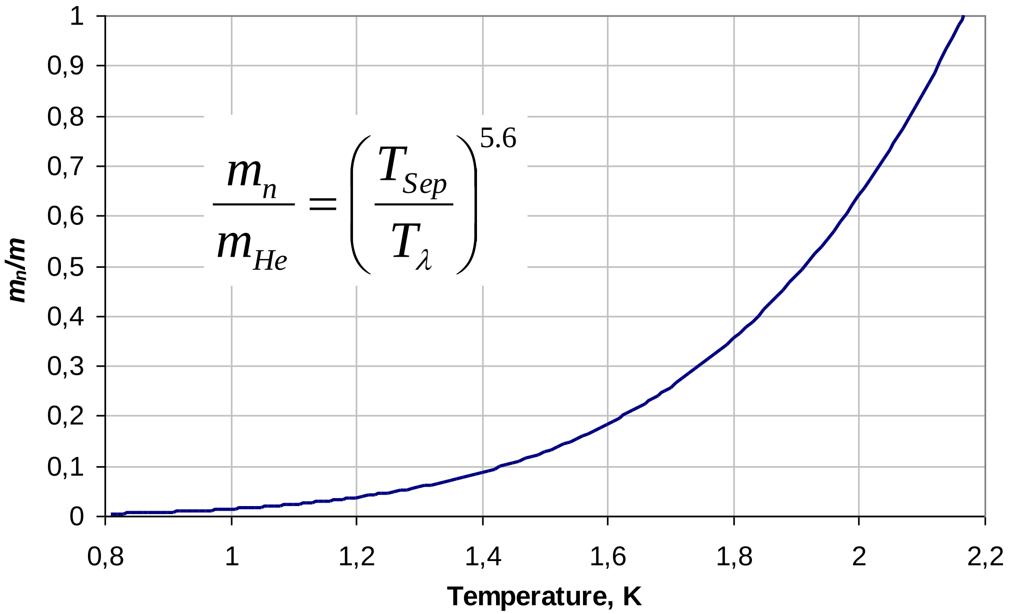
Separation on the entropy filter

Entropy filter idea



Ratio of He normal component in superfluid helium



Optimum separation temperature

Total enrichment of 3He/4He mixture in the separator:

$$\frac{y(T_{Sep})}{y_0} = \left(\frac{T_{Sep}}{T_\lambda}\right)^{-5.6} \exp\left(4.31 \cdot T_{sep}^{\;-1.21}\right)$$

where y_0 refers to original concentration of the 3He in the mixture

After separation the enriched mixture need to be subjected to additional process of separation. The minimal work of this process W_{sep}:

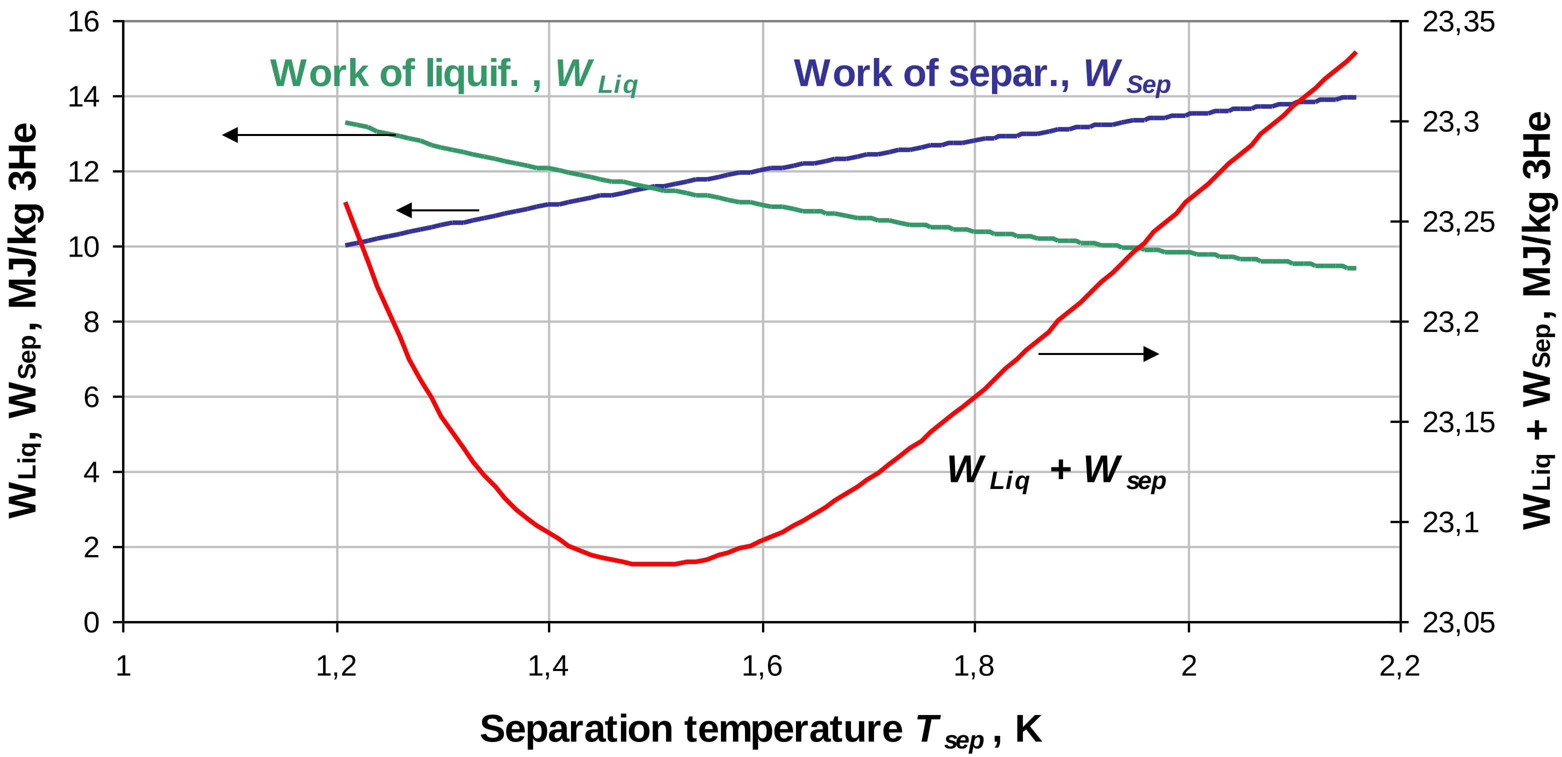
$$W_{Sep} = R_u \cdot T \cdot \left[y(T_{Sep}) \cdot \ln\left(\frac{1}{y(T_{Sep})}\right) + \left(1 - y(T_{Sep})\right) \cdot \ln\left(\frac{1}{1 - y(T_{Sep})}\right)\right] \frac{1}{y(T_{Sep}) \cdot M_{3He}}$$

The minimal work of helium liquefaction at separation temperature W_{Liq}:

$$W_{Liq} = T_0\left(s_1 - s_{Tsep}\right) - \left(h_1 - h_{Tsep}\right)$$

where *1* refers to standard conditions

Optimum separation temperature



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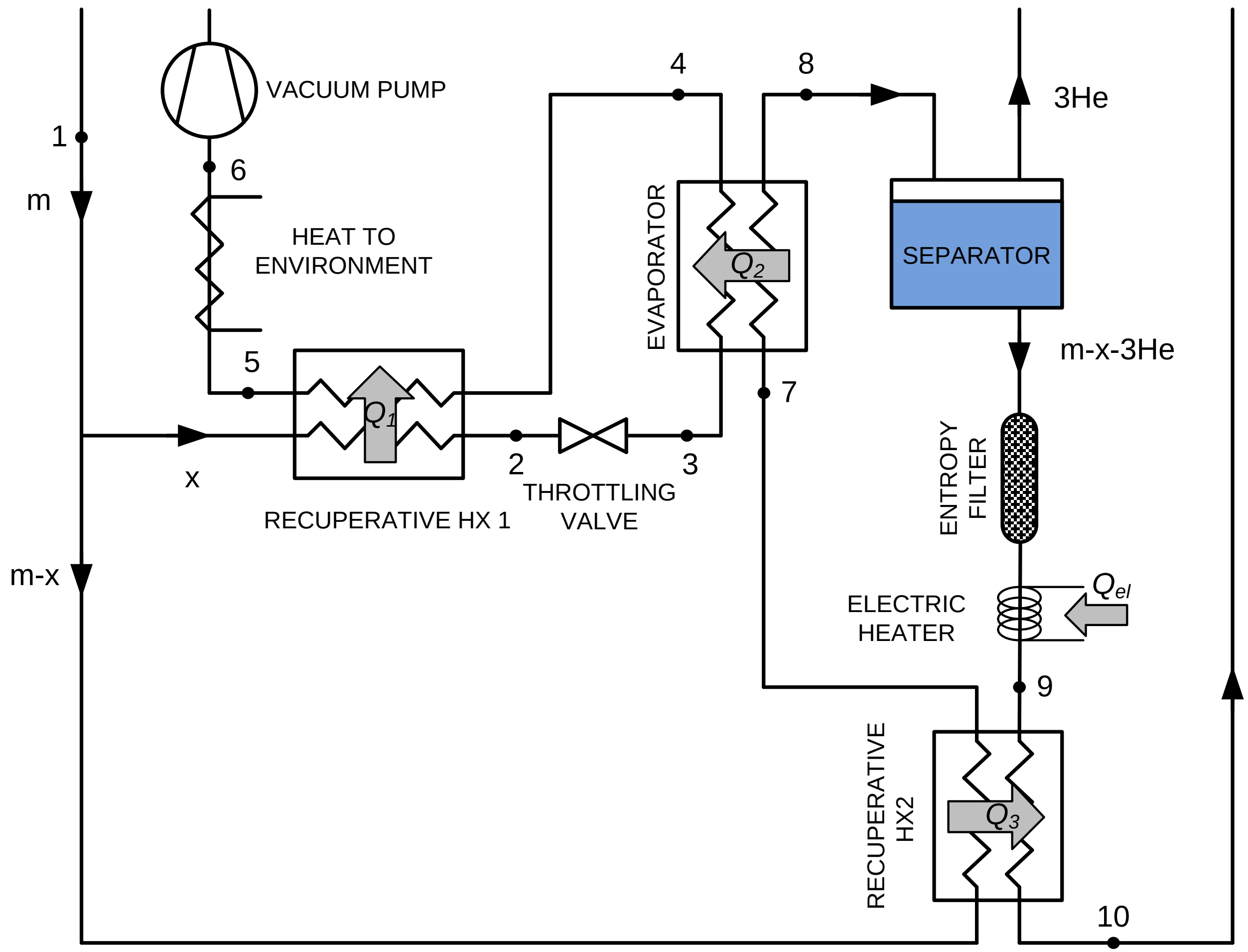
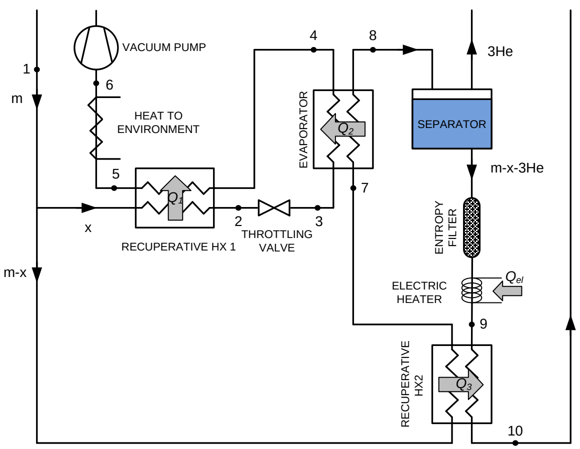
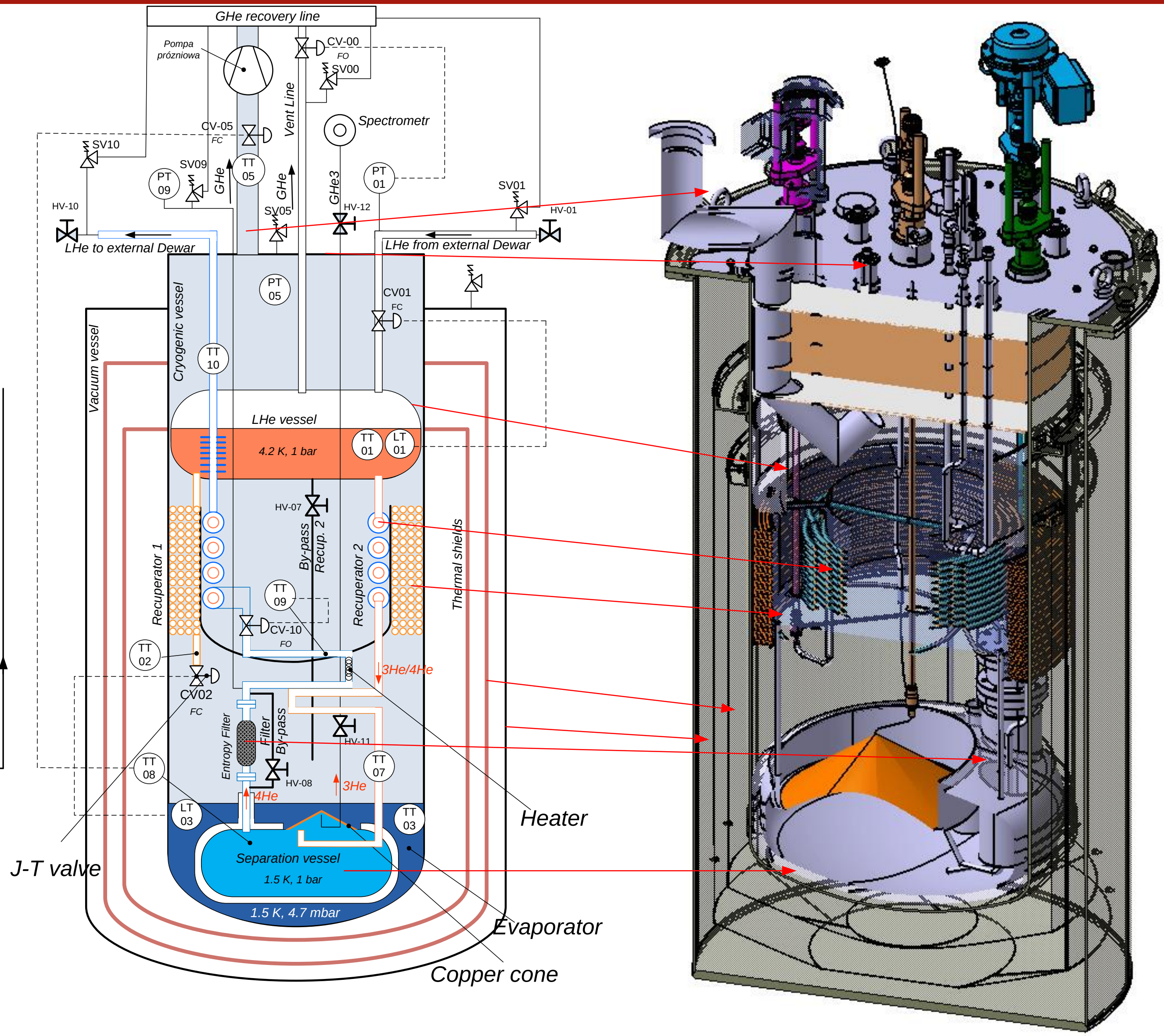
Continuous flow 3He separation system



Separator conceptual design





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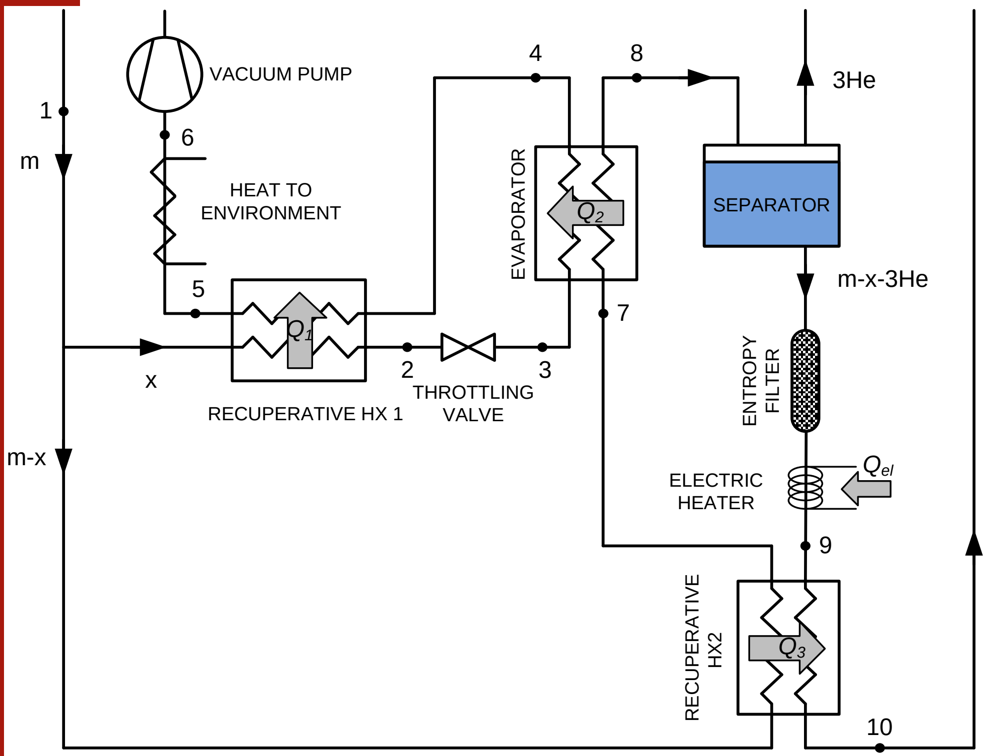
PGNiG

Recuperative heat exchanger model



JT loop cooling power

$$Q_2 = x \cdot (h_4 - h_3)$$

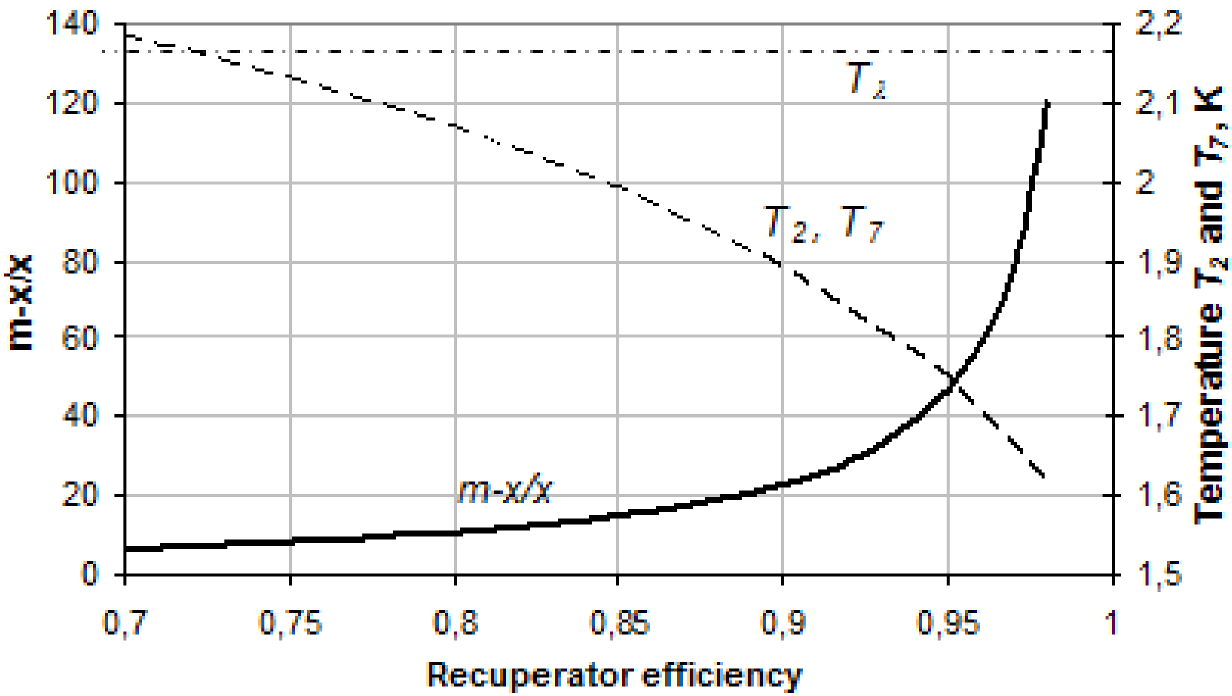
Filtered mixture cool-down

$$Q_2 = (m - x) \cdot (h_7 - h_8)$$

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

Primary helium stream ***m*** is split to JT loop strem ***x*** and filtration stream ***(m-x)***

Recuperator efficiency and separation ratio



$$\varepsilon_{R1} = \frac{h_1 - h_2}{h_1 - h_{2T}}$$

$$\varepsilon_{R2} = \frac{h_1 - h_7}{h_1 - h_{7T}}$$

For recuperators efficiency exceeding 95%, less than 2% of helium must be directed to Joule-Thomson loop

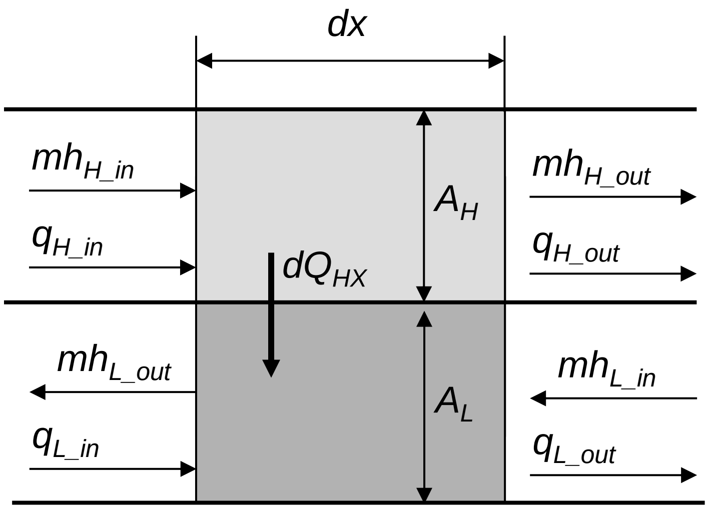
Thermal model of tube-in-tube type Recuperator2 heat exchanger

High temperature side

$$m\frac{dh_H}{dx} + A_H\frac{dq_{liq\_H}}{dx} - \frac{dQ_{HX}}{dx} = 0$$

Low temperature side

$$-m\frac{dh_L}{dx} + A_L\frac{dq_{liq\_L}}{dx} + \frac{dQ_{HX}}{dx} = 0$$



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

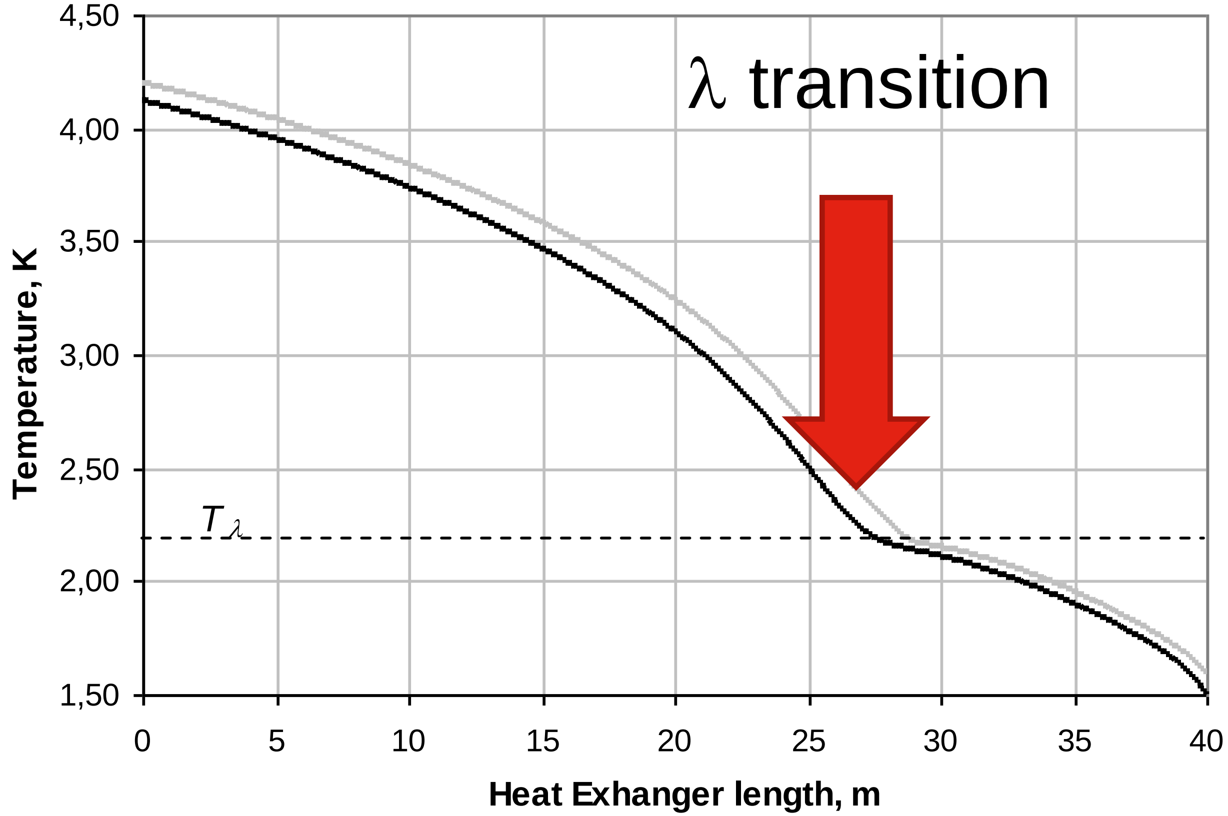
Heat conductivity takes into account lambda transition

Thermal model of tube-in-tube type Recuperator2 heat exchanger

The following configurations of the Recuperator2 have been tested:

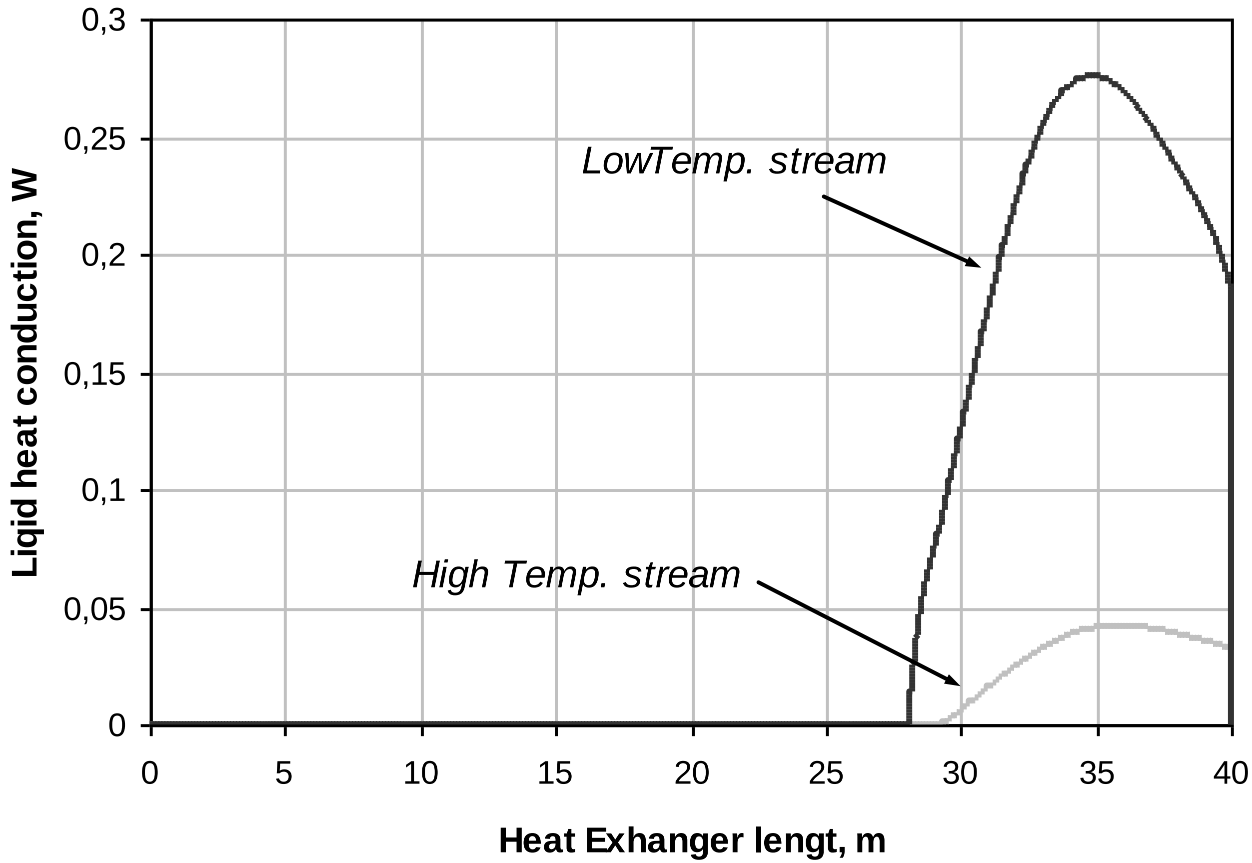
- Inner pipe dimensions: 6x1, 8x1 mm^2
- Outer pipe dimensions: 12x1, 14x1 mm^2
- Heat exchanger length: 20m, 40m
- Helium mass stream: 1.0, 2.0 and 5.0 g/s

Typical temperature profile along „tube-in-tube” Recuperator



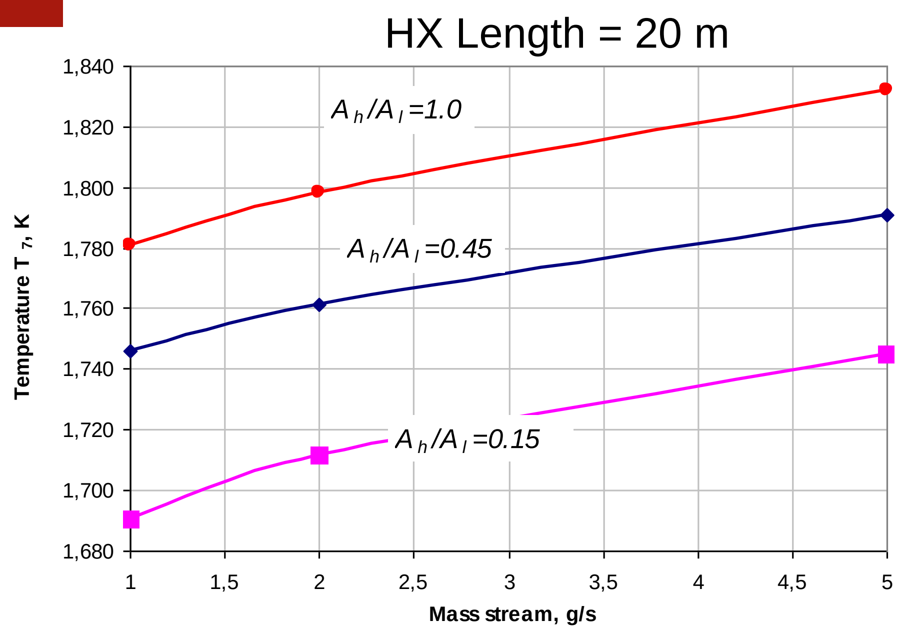
Mass flow 5 g/s

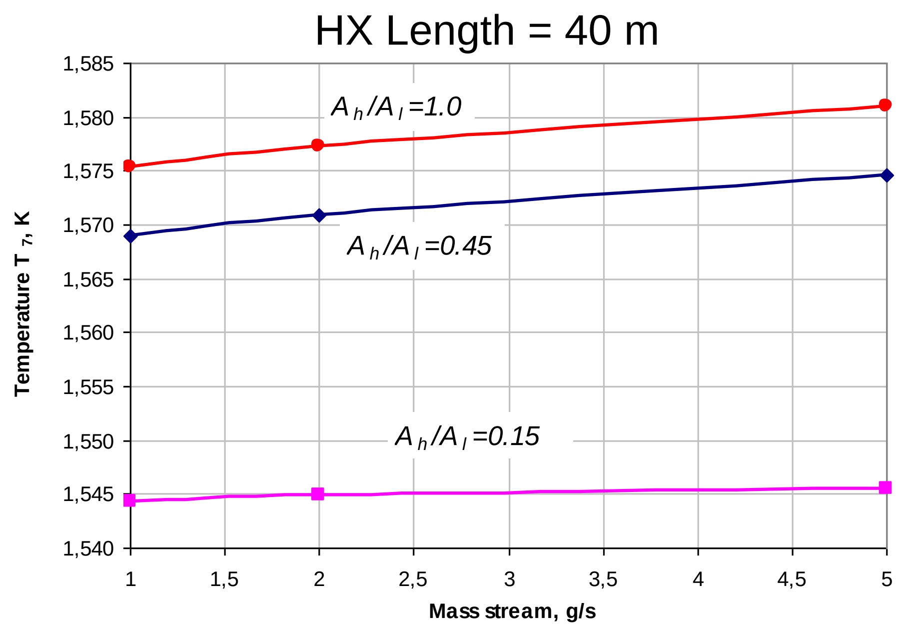
He heat conductivities along heat exchanger



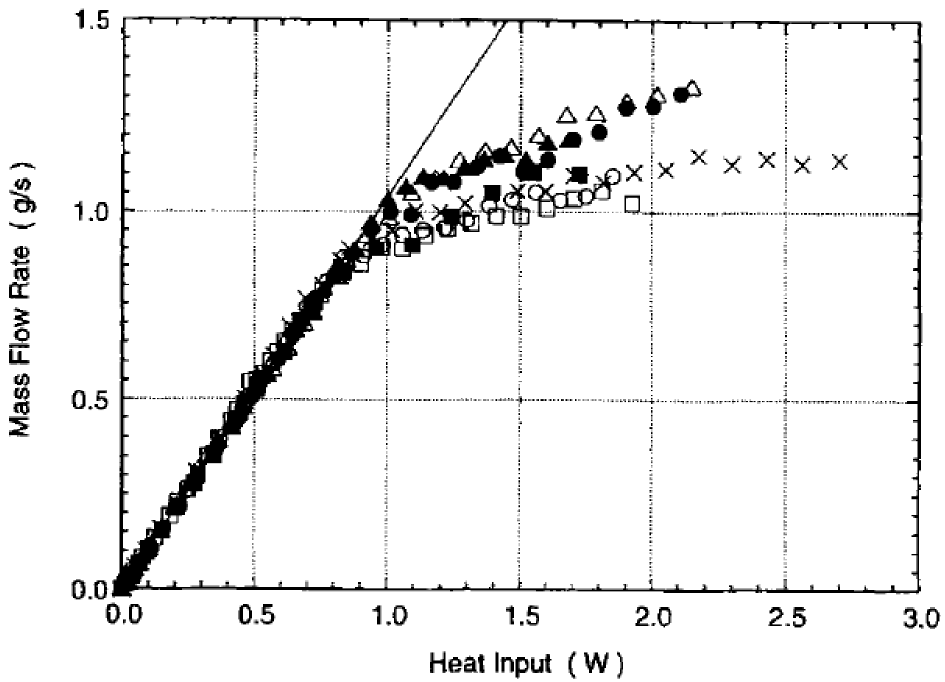
Inner pipe 6x1 m^2
Outer pipe 14x1 m^2
A_H/A_L=0,15

Calculation results of incoming stream outlet temperature





Mass transfer through porous media - determination of the entropy filter diameter



H. Nakai, N. Kimura, M. Murakami *, T. Haruyama and A. Yamamoto, Superfluid helium flow through porous Media, Cryogenics 36 (1996) 667-673

$$Q = \dot{m}ST = \rho_s ST \cdot \varepsilon \cdot A \cdot v_{sc}$$

$$v_{sc} \cdot d^{0.25} = 2 \div 4 \times 10^{-3}\ m^{1.25}/s$$

Q – heat input, W
m - superfluid helium mass flow, kg/s
S - superfluid helium entropy, J/kgK
T - superfluid helium temperature, K
ρ_s – superfluid helium density, kg/m^3
ε – plug porosity = 0.2 – 0.6
A – plug area, m^2
v_{sc} – crit. velocity of superfluid component, m/s
d – mean pore diameter = 0.4 – 10 μm

Entropy filter diameter:

$$d_f = \sqrt{\frac{4 \cdot \dot{m} \cdot d^{0.25}}{\pi \cdot \rho_s \cdot \varepsilon \cdot 2 \div 4 \times 10^{-3}}}$$

For *m*=5 g/s at T_{sep}=1.5K:

$$d_f = 23 \div 83\, mm$$

3He concentration measurements

Pfeiffer Vacuum QMS 700 – Quadrupole Mass Spectrometer

Operation principle: second Mathieu stability region for small values of amu

Mass range: 1÷128 amu,

Sensitivity: single ion (10^{-19} A), concentration: 10^{-3}ppm

Calibrated with samples from Hungarian Academy of Science

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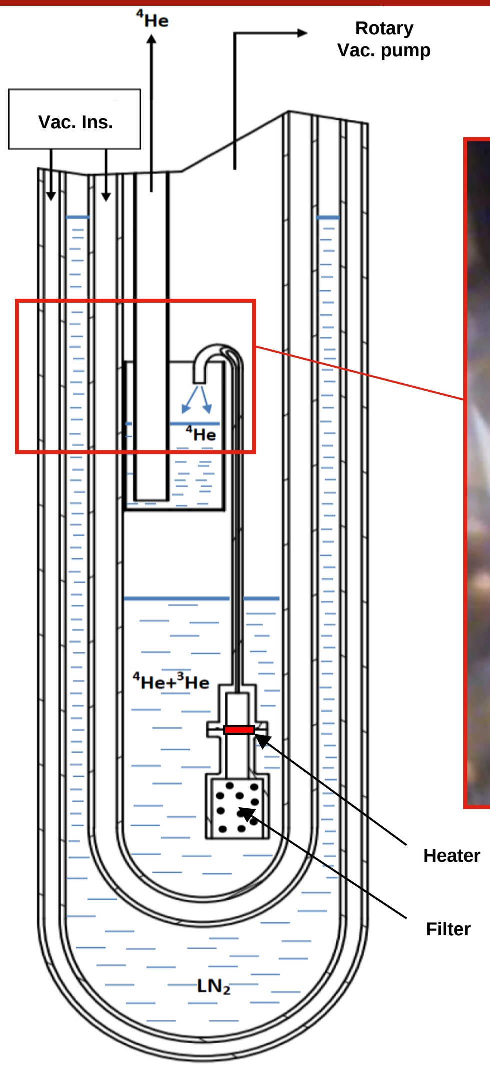
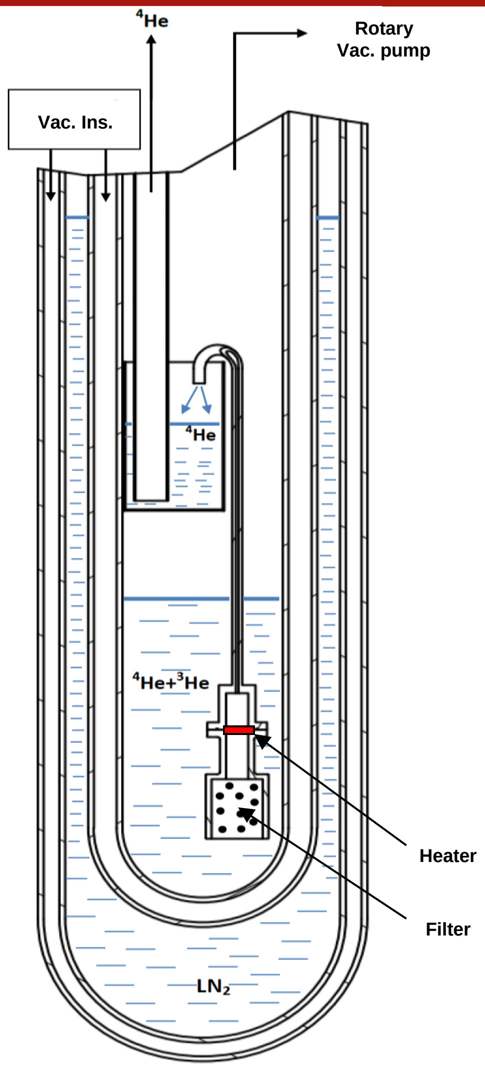
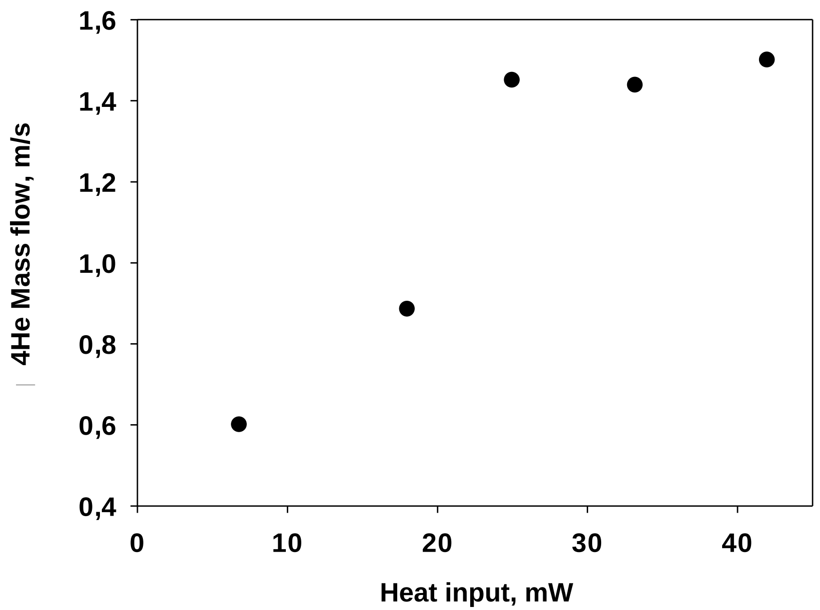
Entropy filters tests



Entropy filters tests



Filter: MultiWalled Carbon NanoTubes (MWCNTs)



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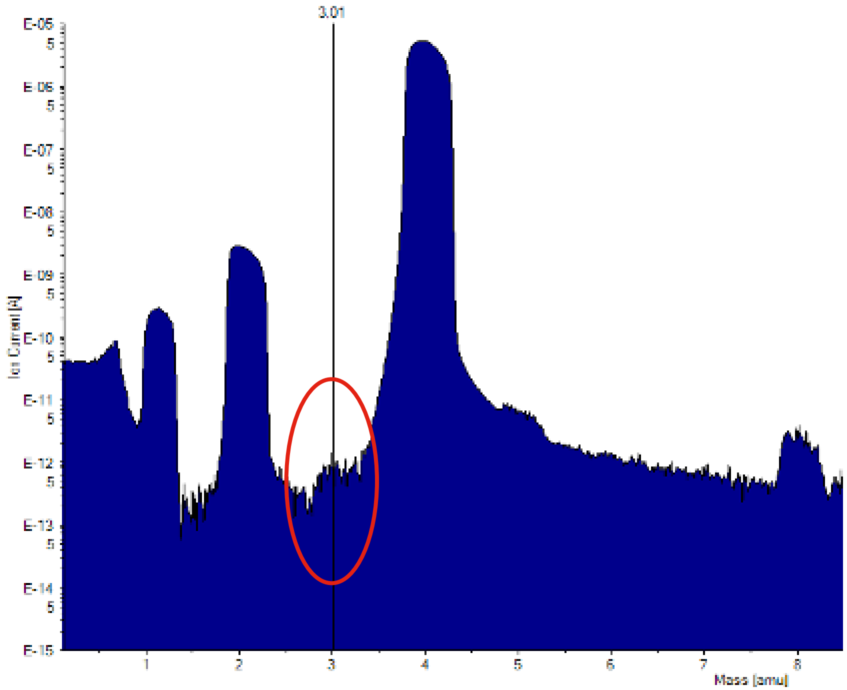
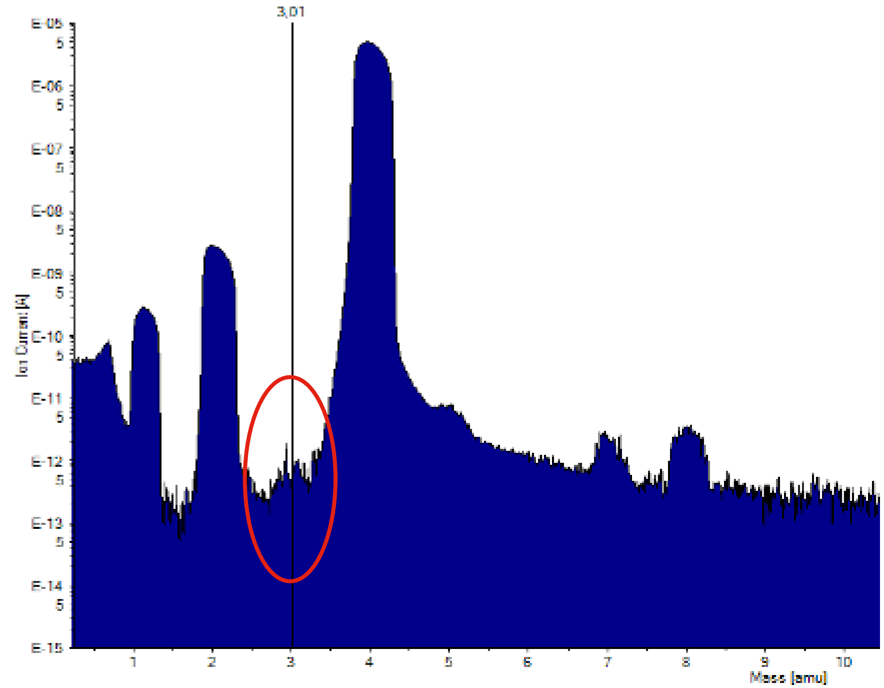
3He concentration measurements



	Current	Minimum	Maximum	Average	Std Dev
H2	4.0550E-03 ppm	3.0646E-03 ppm	4.7670E-03 ppm	4.2366E-03 ppm	4.6880E-04 ppm
3He	1.1337E-05 ppm	0.0000E00 ppm	1.1817E-05 ppm	9.8118E-06 ppm	2.9716E-06 ppm
4He	9.9996E01 %	9.9995E01 %	9.9997E01 %	9.9996E01 %	4.7057E-04 ppm

Downstream the filter



	Current	Minimum	Maximum	Average	Std Dev
H2	2,8788E-03 ppm	2,3670E-03 ppm	4,4349E-03 ppm	3,4268E-03 ppm	5,7751E-04 ppm
3He	7,4115E-06 ppm	0,0000E00 ppm	1,0271E-05 ppm	6,8967E-06 ppm	2,7643E-06 ppm
4He	9,9997E01 %	9,9996E01 %	9,9998E01 %	9,9997E01 %	5,7956E-04 ppm

Upstream the filter

Conclusions

- In view of global 3He shortage the NG purification plants coupled with liquid helium production line have been shown as potential economic 3He source
- WUT in cooperation with PGNiG branch Odolanow and IMP of PA Science is developing and will be testing the Separator of 3He form liquid helium
- The key issues, as separation methods, separation temperature, HX with λ transition, entropy filter design as well as measurement of the 3He concentration have been studied and solved
- The first Separator run is expected till the end of 2014

Thank you for attention

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