Helium Recovery from Guard Vessel Atmosphere of the ALLEGRO Reactor

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Abstract. ALLEGRO is a helium-cooled experimental fast reactor, which is under development by the consortium „V4G4 Centre of Excellence" (institutes and companies from Czech Republic, Hungary, Poland and Slovakia) associated with CEA (France). The current pre-conceptual stage of development of ALLEGRO by the V4G4 CoE is based on the 75 MWt concept presented by CEA in 2009. The main purpose of ALLEGRO is 1) Demonstration of viability of the gas-cooled fast reactor (GFR) technology in pilot scale, 2) Testing of innovative carbide-based refractory GFR fuels in the start-up oxide core driver, 3) Qualification of other GFR-specific technologies such as components of the primary circuit, helium-related systems, fuel handling etc..

One of the GFR-related helium technologies is the recovery system for the helium leaked from pressurized boundaries such as primary circuit into the gas-tight guard vessel (GV - a metallic pressure boundary around the primary circuit filled with 0.1 MPa nitrogen, whose main function is to provide backpressure of at least cca 0.4 MPa to make the emergency cooling system functioning properly in loss-of-coolant accident conditions). The helium recovery system will be able to separate the leaked helium from the nitrogen-helium GV atmosphere and return it back into the helium storage system. This feature will make the future GFRs (including ALLEGRO) much less dependent on the helium market.

The paper describes a helium recovery system with emphasis on both the separation method and the proposed (pre-conceptual) technical solution. The system is based on multi-cycle semi permeable membrane separation and is expected to be operated not continuously, only when the helium concentration in the GV exceeds a certain limit. Up to 90% helium purity can be reached by this process, while the final purification is planned to be performed by other methods, e.g. by using a pressure swing adsorption (PSA) technology, which can achieve high purity helium. For good yield of helium a membrane with high selectivity of nitrogen/helium should be used. A membrane-based recovery system was developed, tested on laboratory scale and a first concept of the recovery system was proposed for the conditions of ALLEGRO. The work has been so far supported mainly by the Technology Agency of Czech Republic.

Key Words: Generation-IV demonstrator, gas-cooled fast reactor, helium separation, gas mixture.

1. Introduction

The gas-cooled fast reactor (GFR) system, namely the high-temperature helium-cooled fast-spectrum reactor with a closed fuel cycle, is one of the six promising Generation-IV systems presented in 2002 [1]. As no GFR has ever been built, the development of the first-of-a-kind experimental (or demonstration) GFR is still a challenge in the last ~50 years from the point of view of both safety and technology.

CEA (France) was working on a concept of a large industrial GFR (2400 MWt) as well as a small experimental/demonstration unit ALLEGRO 75 MWt (ALLEGRO CEA 2009) in the

period 2002-2010 [2]. The development of ALLEGRO (based on ALLEGRO CEA 2009) currently continues in the frame of a Central European consortium „V4G4 Centre of Excellence" established in 2013 by its members ÚJV Řež (Czech Republic), MTA EK (Hungary), NCBJ (Poland) and VUJE (Slovakia) associated with CEA (France). The main purpose of ALLEGRO is 1) Demonstration of viability of the gas-cooled fast reactor (GFR) technology in pilot scale, 2) Testing of innovative carbide-based refractory GFR fuels (UPuC in SiC-SiC_f cladding) in the start-up oxide fuel driver core, 3) Qualification of other GFR-specific technologies such as components of the primary circuit, helium-related systems, fuel handling etc.. The main design characteristics of the reactor are summarized in Table I.

TABLE I: MAIN DESIGN CHARACTERISTICS OF ALLEGRO CEA 2009.

Parameter [Unit]	**Value**	**Comment**
Nominal power thermal [MW]	75	The thermal power is subject of change.
Nominal power electrical [MW]	0	Power conversion system is not required.
Core inlet / outlet temperatures [°C]	260 /530 400 /800-850	Start-up core with oxide fuel. Full refractory core.
Primary pressure [MPa]	7	
Fuel (wire-wrapped rods in hex. tube)	MOX / SS cladding	Start-up core: Feasibility of UOX fuel (<20% U235) is being investigated.
	UPuC in SiC-SiCf	Target full refractory core.
No. of fuel rods per fuel assembly (FA)	169	Oxide fuel start-up core.
No. of FAs / Exp. FAs / C&SD	81 / 6 / 10	C&SD: Control & Shutdown assemblies.
Primary / Secondary / Tertiary circuits	He / Water / Air	Water will be replaced with suitable gas.
No. of primary / secondary / DHR loops	2 / 2 / 3	---
Coolant of DHR intermediate circuit	Water	DHR heat sink: Water pool.
Number of nitrogen safety accumulators	3	Still subject of optimization.
Nominal pressure in guard vessel [MPa]	~0.1	Nitrogen atmosphere.

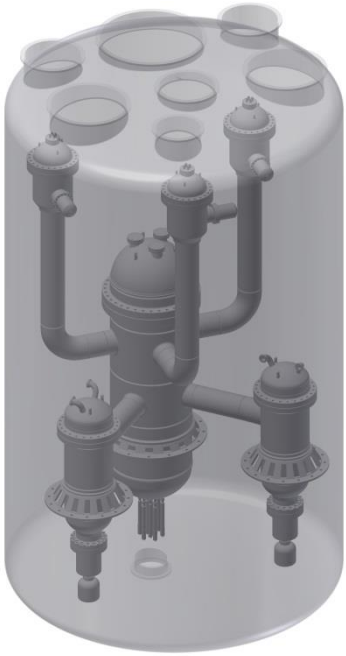
Although the safety of ALLEGRO in general is currently the main issue to be solved, the development of helium-related technologies for ALLEGRO continues in parallel in the Czech Republic in the frame of V4G4 CoE. One of the GFR-related helium technologies is the recovery system for the helium leaked from the pressure boundary such as primary circuit into the gas-tight guard vessel (GV). The GV is a metallic pressure boundary around the primary circuit filled with ~0.1 MPa nitrogen, whose main function is to provide backpressure of at least ~0.4 MPa to make the emergency cooling system (based on active systems) functioning properly in loss-of-coolant accident conditions. The helium recovery system will be able to separate the leaked helium from the nitrogen-helium GV atmosphere and return it back after final purification into the helium storage system. This feature will make the future GFRs (including ALLEGRO) much less dependent on the helium availability & price on the market.

This paper describes the first considerations on the helium recovery system with emphasis on both the potential separation methods and the proposed pre-conceptual technical solution. The idea to recover helium from the GV atmosphere was applied for a patent in the Czech Republic.

2. Background of the helium recovery from the ALLEGRO guard vessel atmosphere

The system for the recovery of leaked helium from pressurized boundaries is applicable with advantage for GFRs only, because the primary circuit (& some helium-related technologies) are located inside the pressure-tight GV, see FIG. 1 including main parameters. This system, thus, cannot be applied for V/HTRs due to absence of GV in the V/HTR design.

Although the typical He leakage from a VHTR does not exceed approximately one primary circuit inventory per year, the estimated maximum helium leakage in ALLEGRO into the GV is conservatively estimated not to exceed ~3 primary circuit inventories per year through solid wall diffusion, sealing imperfections and potential imperfections in the fuel handling/ refuelling technology. This conservatism originates from the lack of data associated with all these technologies (still under development). It is also to note that undesirable pressurization of the GV by the leaking helium would increase its irreversible losses from the GV.



Parameter	Unit	Value
Diameter	m	~17
Height	m	~30
Wall thickness	mm	~46
Nominal temperature	°C	~65
Nominal pressure	MPa	~0.1
Atmosphere in GV	-	nitrogen
Total volume	m^3	6800
Free volume	m^3	~2600

FIG. 1. ALLEGRO primary circuits with the decay heat removal system inside the guard vessel including its main parameters (concrete structures are not shown for simplicity).

This leaked helium can be in ALLEGRO (or in any future GFR) returned from the GV atmosphere to helium storage tanks via regeneration/recovery process. A potential incidental excessive helium leakage, thus, will not be penalizing the economy (& performance) of the ALLEGRO operation. A question arises, whether the helium recovery system in a GFR (or in ALLEGRO, respectively) is economically & technically feasible.

2.1. Economical aspect of the helium recovery system

Helium is a finite, non-renewable resource that can be extracted from only a few natural gas fields around the globe. The entire helium supply of almost ~180 billions of cubic meters per year [3] depends on roughly 20 liquefaction plants, located in the United States, Poland, Russia, Algeria, Qatar, China and Australia. As industry needs helium in e.g. welding applications and electronics manufacturing (semiconductors, flat-panel display, optical fibers), the helium market price is expected to rise in the future at a rate of about 5% per year.

As the helium used especially in the U.S. in large-volume applications is seldom recycled, the cost/benefit ratio that gas recovery and recycling systems can provide, thus, becomes an issue also for the operational economy of the future GFRs. The using of recovery/recycling systems can reduce the helium costs by about one-third. The estimate of the helium price evolution in the future as deduced from [3] is shown in FIG. 2.

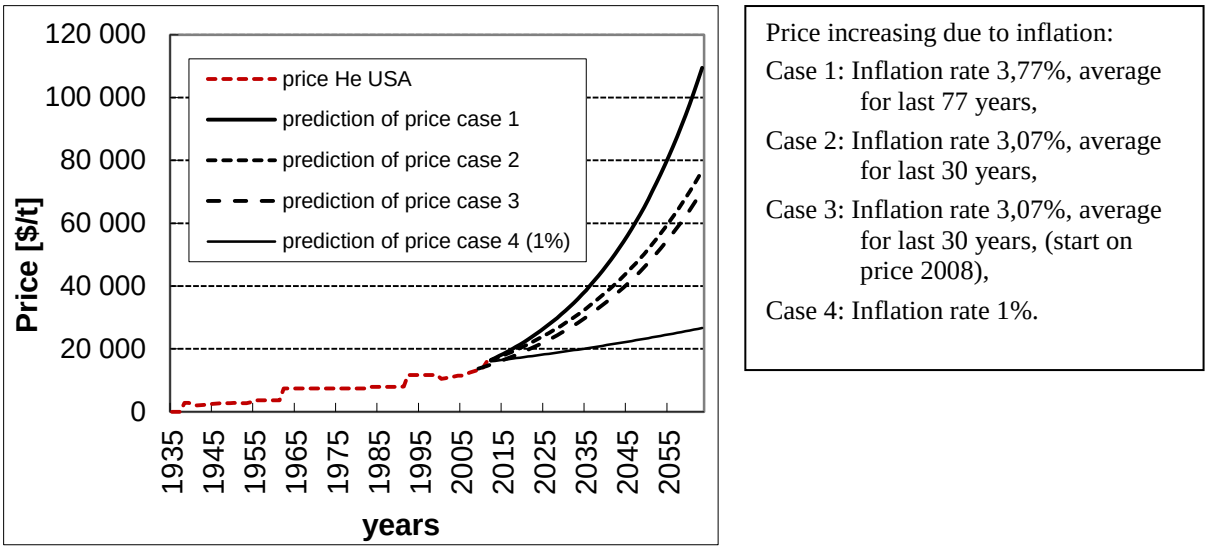


FIG. 2. Estimate of grade-A helium price evolution in USA (99.995 % purity = 4.6) [3].

2.2. Requirements onto the helium recovery system for a GFR

This system intends to recover helium from the GV atmosphere (~0.1 MPa nitrogen, ~60-80 ºC) during the normal reactor operation. As the GV atmosphere must be cooled (using forced convection) & filtered/purified during the normal reactor operation, helium is not expected to accumulate in the upper part of the GV in important amounts. Only a purified GV atmosphere free of dust & other impurities is allowed to access the helium recovery system. The potential impurities in the GV atmosphere will be removed using the GV atmosphere purification system. If excessive activity appears in the GV, the helium recovery system will be stopped or bypassed. Presence of activity in not desired in the helium recovery system. The recovered helium is expected to undergo final purification before its final way to the storage system. The entire helium recovery system must be minimized in space (in order to fit into the containment) and should not require excessive amounts of energy. The recovery system must be located close enough the GV ventilation & filtration systems. A question arises, whether the recovery system can meet all these requirements. Selection of a suitable separation method is, thus, of crucial importance for answering this question. The potentially applicable methods are briefly described in the next chapter.

3. Methods suitable for helium separation from the nitrogen-helium mixture

There are three potential methods available: 1) Low-temperature distillation (cryogenic separation), 2) Pressure swing adsorption (PSA), 3) Diffusion (membrane) separation. Suitability of each method (or a potential combination of these methods) depends largely on the energetic & economic performance and efficiency of each separation method.

3.1 Cryogenic separation

Cryogenic separation requires a very tight integration of heat exchangers and separation columns to obtain a good efficiency. This process is very demanding on energy. All the energy for refrigeration is provided by the compression of gas at the inlet of the unit. This method is particularly used for obtaining pure oxygen, nitrogen and argon from air. As a secondary product, rare gases (Kr, Xe, Ra) are also obtained including helium in minor amounts in a mixture with nitrogen. This method is well mastered by the industry.

Advantages (especially in nitrogen production): 1) Low amount of electricity per gas unit, 2) Very high gas purity achievable, 3) Liquid nitrogen generated for storage on site.

Disadvantages: 1) Large site space & utility requirements, 2) High capital costs, 3) Limited scalability in production, 4) Time-demanding start-up and shutdown of the facility.

3.2 Pressure swing adsorption (PSA)

Pressure swing adsorption is a technology used to separate some gas species from a mixture of gases under pressure according to the species' molecular characteristics and affinity for an adsorbent material. Using two adsorbent vessels allows nearly continuous production of the target gas. It also permits so-called pressure equalization, where the gas leaving the vessel being depressurized is used to partially pressurize the secondary vessel. This results in significant energy savings. Aside from their ability to discriminate between different gases, adsorbents for PSA systems are usually very porous materials chosen because of their large surface areas. Though the gas adsorbed on these surfaces may consist of only few (or even one) molecules thick layer, surface areas of several hundred square meters per gram enable the adsorption of a significant portion of the adsorbent's weight in gas. Typical adsorbents are activated carbon, silica gel, alumina and zeolite.

Advantages: 1) Low to moderate capital costs, 2) Cost-effective nitrogen production of relatively high purities, 3) Quick installation and start-up of the facility.

Disadvantages: 1) High maintenance equipment, 2) Noisy operation, 3) Limited scalability.

3.3 Membrane separation

Membrane separation technologies currently use a hollow-fibber membrane consisting of porous polymer fibbers coated with a separation layer. A porous fibber has a complex asymmetric structure, with the polymer density increasing towards the fibber external surface. The application of porous support layers with asymmetric structure allows separating gases under high pressures (up to 6.5 MPa). The thickness of the fibber gas separation layer does not exceed 0.1 μm, ensuring a high relative permeability of gases across the polymer membrane.

The existing level of the technological development makes possible the production of polymers with a high selectivity for various gases, and consequently, capable of delivering high-purity gaseous products. A modern membrane module used for the membrane gas separation technology comprises a removable membrane cartridge and a body. The density of fibbers packaging in the cartridge is estimated at some 500–700 m^2 per the cartridge cubic meter, which helps to minimize the dimensions of gas separation plants.

Advantages: 1) Low capital costs, 2) Production output is very flexible, 3) Quick installation and start-up, 4) Easy to vary purity and flow rate.

Disadvantages: 1) Not economical for high purity requirements, 2) Not economical for large outputs, 3) Requires relatively large amount of electricity per nitrogen unit produced.

When analysing the above arguments, the most suitable for effective separation of high purity helium from the GV atmosphere seems to be the combination of the membrane separation and the pressure swing adsorption.

4. Laboratory tests of membrane separation of helium from nitrogen-helium mixtures

The membrane separation of helium from helium–nitrogen mixtures was tested using commercial membrane module originally designed for separation of nitrogen from air. The goal of these tests was to verify the application of this type of membrane for helium separation from nitrogen atmosphere and evaluation of efficiency of this method.

A PRISM® membrane, limited to max. 2 MPa and 65 °C, was tested in a laboratory device proposed and constructed for these tests, see FIG. 3. The testing device consisted of pressure vessels with nitrogen and helium, mixing station allowing required gas mixture N_2-He preparation, the tested membrane module, sampling lines and the gas analyser. The used analyser for determination of helium content in gas worked on principle of measurement of speed of sound in the gas. The composition of the inlet gas was adjusted by accurate gas flow controllers and the helium content was measured at both the inlet and the outlet of the permeate and retentate side of the membrane.

FIG. 3. Membrane module PRISM® used for tests in the testing device.

The output composition of the gas mixture (more precisely the helium concentration in the permeate flow) was investigated in the experiments as function of the helium concentration in the input gas, inlet gas pressure and the retentate mass flow rate. During tests the input parameters were varied in the following range:

- inlet gas pressure : 1.5 – 12 bar
- helium inlet concentration : 3 – 50 % by volume
- permeate-to -retentate ratio : 0.2 – 4

The experiments revealed that the efficiency of the one-step helium separation increases with the retentate flow. The increasing retentate flow, however, lowers the whole separation process (the higher is the retentate flow the higher is the energy needed for gas pumping). For illustration (using inlet gas mixture with 10 vol. % helium), the increase of the helium concentration in the permeate at the outlet of the membrane is shown in FIG. 4 from [4] as a function of the inlet gas pressure and the retentate flow (expressed as multiples of the permeate flow). The parameters will, thus, be subject of optimization.

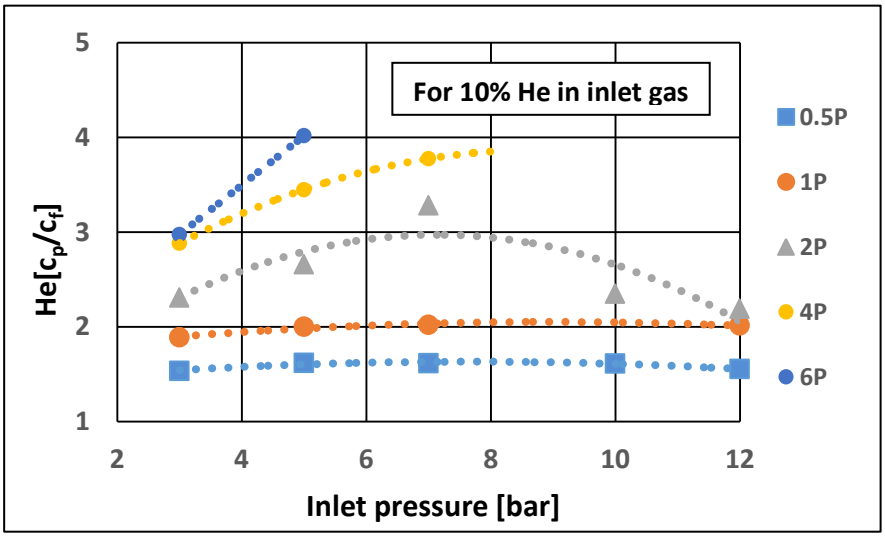


FIG. 4. The dependence of helium concentration in permeate on inlet gas pressure and retentate flow expressed as multiples of permeate flow
(cp – helium concentration in permeate, cf – helium concentration in inlet gas) [4].

The dependence of the helium separation efficiency on the inlet gas pressure is less strong than expected. This dependence illustrated in FIG. 4 was even lower for higher helium concentrations in the inlet gas and lower retentate flows. The optimum inlet gas pressure is somewhere between 4 – 7 bar. To reach the optimal efficiency of the process the gas need not to be compressed to very high pressures. The energy needed for compressing the gas is, thus, lower than originally expected. The results of the experiments indicate that the tested PRISM® membrane for nitrogen separation from air seems to be suitable also for the helium separation from nitrogen. To reach sufficiently high purity helium (90% and higher) 3 to 5 passes of the permeate gas through the membrane are expected to be sufficient.

Future experiments should concentrate at testing of another membrane types available on the market, testing gas mixtures at higher temperature (to simulate the conditions in the GV of ALLEGRO) and also testing of helium separation from more complex gas mixtures (e. g. air). Foreseen is also improvement of the testing device, e.g. implementation of more precise gas-flow controllers & analyser, or constructing larger semi-continual device.

5. Pre-conceptual design solution of the helium recovery system for ALLEGRO

The main designer of large structures of ALLEGRO such as containment & guard vessel needs to know at least approximate information about the systems to be located inside these structures. This is why at least a simplified pre-conceptual design is needed also for the helium recovery system to indicate space & media requirements.

The suitable location of the helium recovery system (HRS) in the containment depends on the layout of the remaining technology. The helium recovery system is designed as a subsystem of the GV ventilation/filtration system. Therefore, it has to be located as close as possible to this system, but also as close as possible to the GV itself to secure the return of the retentate into the GV. The area required for the HRS in the containment will be determined by the required capacity of the membrane device. The current pre-conceptual design of the HRS indicates the required area of ~162 m^2. The capacity & technical parameters of the system are determined by the requirements onto the quality of the GV atmosphere and the economic benefits of the integration of this system.

The HRS is assumed to be used during normal reactor operation only in a not continuous mode (starting only after reaching a threshold level of helium in the GV). Table II shows a preliminary estimate of the HRS parameters:

TABLE II: PRELIMINARY PARAMETERS OF THE HELIUM RECOVERY SYSTEM

Parameter	**Value**	
Volume of helium required for regeneration	~79	m^3
Estimated time to reach the threshold concentration in the GV	~7 to 8	days
Basic number of cycles to regenerate the helium by the HRS	7	
Helium released into the GV during regeneration	~36	m^3
Adjustment number of regeneration cycles	10	
Remaining helium in the GV after the completion of the regeneration	~6	m^3

The nitrogen-based GV atmosphere is expected to contain except the leaked helium also dust particles, some other gases such as oxygen and potentially also fission products (mainly rare gases and isotopes of iodine). The HRS will be connected to the outlet of the GV ventilation/filtration system, where, most of these impurities will already be removed. For this moment, however, it is difficult to quantify the level of these impurities that would enter the HRS.

As indicated in chapter 3.3, the membrane separation was selected for the HRS. The applicability of membranes for helium separation was demonstrated in the previous chapter using the PRISM® membranes limited to relatively low operating temperature. The current pre-conceptual design of HRS assumes the use of borosilicate membranes, which can be operated at higher temperatures (up to ~300 °C).

The operation of HRS starts after the helium concentration in the GV reaches a threshold value. A selected amount of gas mixture is extracted from the ventilation system and after passing through the membrane modules it is led to the dedicated storage device. At present, only one membrane module is assumed in the separation process, but parallel modules are assumed to be used in the future HRS for ALLEGRO.

Concentration takes place by multiple passages of the gas mixture through the membrane device (16 passes) until the desired concentration of 90% He is reached. The necessity of multiple passages to reach reasonably high yield of pure helium is illustrated by computational simulation in FIG. 5, where the 90% He yield is shown in dependency on the number of membrane passages. The resulting permeate is collected in a storage device after each pass and the retentate is returned back to the GV, see FIG. 6. The equipment is designed so that it is able to recycle 40% of the helium from the atmosphere each donation to GV.

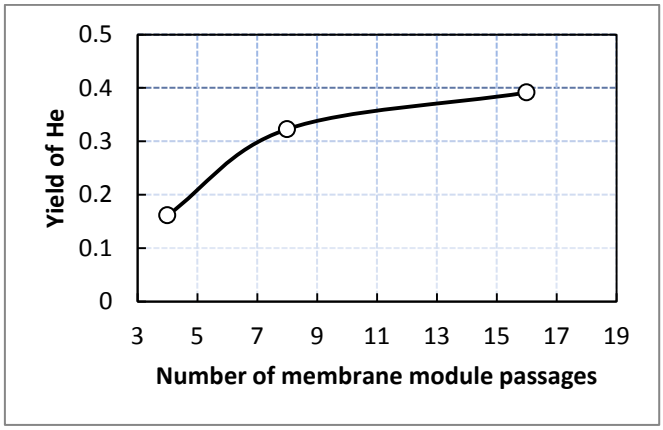


FIG. 5. The dependency of He yield concentrated from 10 to 90%vol. on the number of gas passages through the membrane module [4].

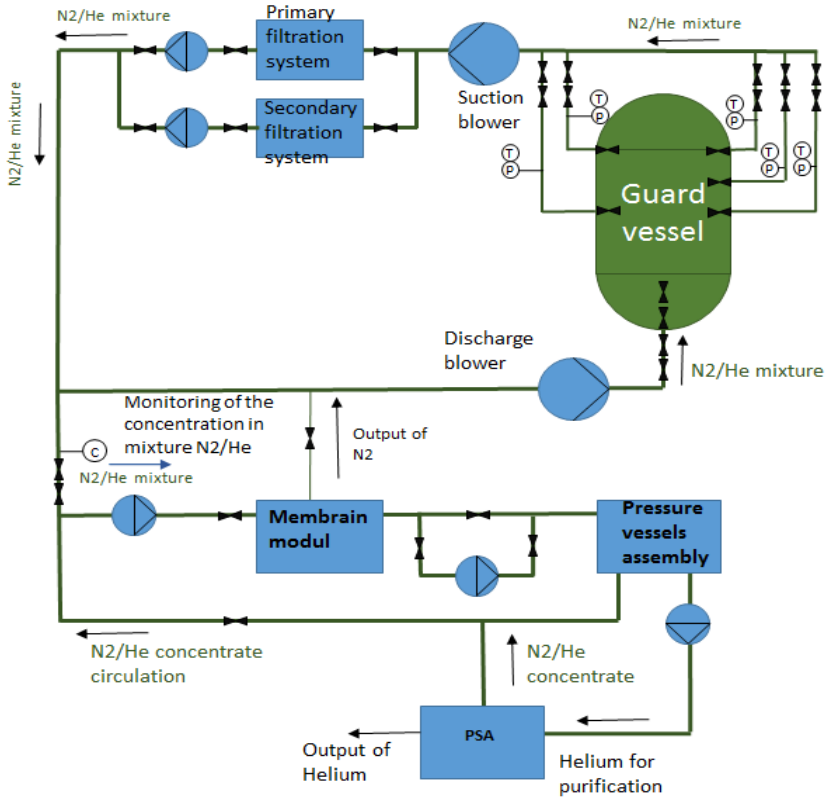


FIG. 6. Pre-conceptual scheme of the He recovery system (HRS).

Borosilicate membrane prototypes of this scale has not been built yet, and in case of fabrication the process reliability and durability should be verified. Also the behaviour of radioactive isotopes of iodine, Kr, Xe and other possible impurities on the borosilicate membrane has to be studied in order to implement these findings to the design of the membrane module´s structure.

In the current design we assume that the system would be operated remotely. The handling/maintenance/replacement of the membrane cartridges in ALLEGRO has not yet been studied. A PSA system is assumed for the final purification of the helium.

The technical solution for a large scale facility assumes two separation circuits. The first separation circuit would consist of a membrane module (& compressor systems) and would concentrate helium to cca 90%. The second circuit would be used for the final purification of helium to cca 99%. Refining will be carried out by two alternatively operating sorption columns. Both circuits are connected to the array of pressure vessels, which will be an exchange of gas and measurement of required data. Both circuits are able to operate independently and also in relation to each other for testing the entire process of regeneration helium. The HRS system layout is shown in FIG. 7.

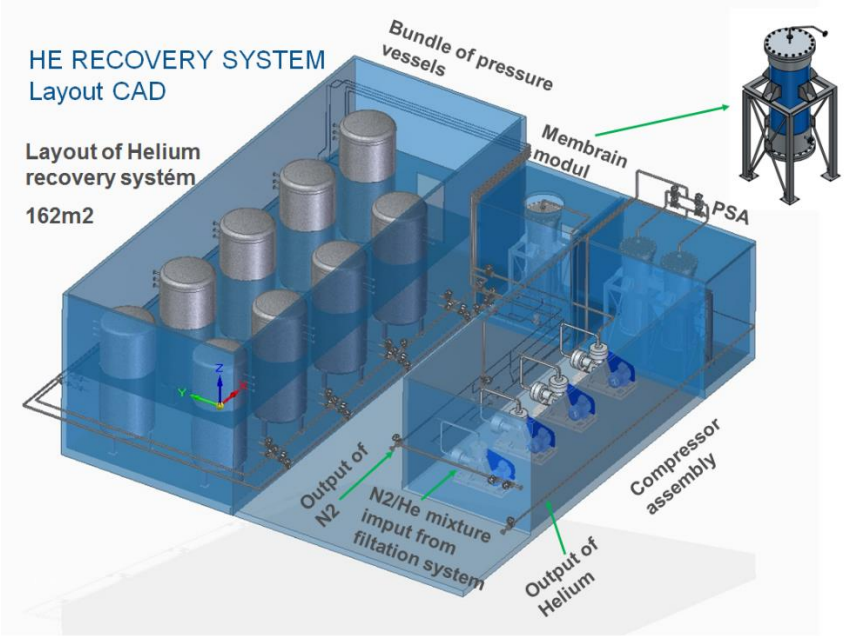


FIG. 7. Pre-conceptual layout of the He recovery system (HRS).

6. Conclusions

This paper presents first considerations on the helium recovery system for ALLEGRO, which would return the helium leaked into the guard vessel back into the helium storage tanks. It was experimentally verified that the existing membrane technology can be used also for the helium separation from nitrogen. A pressure swing adsorption method can be later combined with the membrane technology potentially based on temp. resistant borosilicate membranes.

Although the design of the He recovery system is relatively a routine task, its cooperation with other systems in ALLEGRO such as GV cooling, ventilation, filtration, He storage and He make-up is a subject of rather complex optimization for the ALLEGRO designer. Further steps in the design of the He recovery system will be devoted to small scale experimental demonstration of the complex helium recovery system scalable to ALLEGRO requirements.

7. Acknowledgement

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