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Purification of recovered helium with low level of impurities: evaluation of two different methods

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Abstract

Helium gas coming from low temperature experimental systems is recovered to avoid losses of this scarce gas on Earth. Once this helium gas has been recovered and before its liquefaction, the impurities contained should be removed. It is possible to achieve a low level of impurities by using the proper materials and procedures on the road to helium recovery. A comparison of two different methods applied for the purification of recovered helium with low level of impurities is reported in this paper. One method is the use of liquid nitrogen traps and the other one is the application of a purification system based on getter materials. The cleaning efficiency has been probed experimentally for both methods through the analysis of the purified He gas. The evaluation covers the life time between regenerations, the everyday care as well as the long term, the energy consumption, the initial investment besides the cost of maintenance of both methods.

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Keywords: helium recovery; medium pressure helium recovery plants; helium liquefaction; liquid nitrogen trap; heated getter

1. Introduction

Helium is one of the most abundant elements in the universe, but ironically the low-temperature physics research laboratories, instrument manufacturers, hospitals and other business that depend on liquid helium for their work

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have been suffering during last years frequent worldwide helium shortages [1-2]. The current shortage having several causes [3], there is a well-known solution to cope with this situation in universities, hospitals and industry; that is to recover the helium gas which is evaporated from cooled instruments, and, afterwards, to liquefy again the recovered gas.

Nowadays the helium gas recovery and re-liquefaction can be implemented by using different technologies. The more adequate system selected by any university, hospital or industry would depend on many variables: the rate of helium gas evaporated, the number of cooled instruments, the rate of liquid helium transfers to the instruments, the available room for the recovery setup installation, the current legislation concerning pressurized devices (bottles, compressors), the budget, etc. A major concern affecting the final cost is related with the fact that recovered helium gas contains impurities, mainly nitrogen and oxygen coming from air diffusion trough the materials which are making up the recovering plant. These impurities should be removed before re-liquefying the helium gas.

The University of Zaragoza (UZ) and the Spanish Research Council (CSIC), in collaboration with Quantum Design Inc. (QD) and GWR Instruments, have been active in the research and development of new concepts regarding Helium Recovery Plants (HRPs). In those HRPs, it is possible to achieve a low level of impurities, in the range of tens ppm of global impurities, by using the proper materials and procedures on the road to helium recovery. The helium contamination through some pipe materials used in HRPs has been investigated and the most relevant obtained results are presented in this conference [4]. The elimination of this low level of impurities in helium recovered gas could be achieved by different methods of gas purification. Two of these methods are reported here, which are the liquid nitrogen cooled traps and the gettering process. The physisorption is the mechanism for removing impurities from liquid nitrogen (LN_2) cooled traps, while the heated getter is based in the chemisorption process.

2. Experimental

The two methods tested to eliminate low level impurities in recovered helium gas are implemented in the Medium Pressure Helium Recovery Plant (MP-HRP) [5]. The MP-HRP is one new and simple setup to recover helium gas, which avoids the use of large recovery balloons, high pressure compressors, large high pressure storage capacity and complex purification techniques. This new setup has been recovering 95% of our helium boil-off. Fig. 1 shows the schematic of the setup in which the circle indicates the position of the two different methods tested in this work.

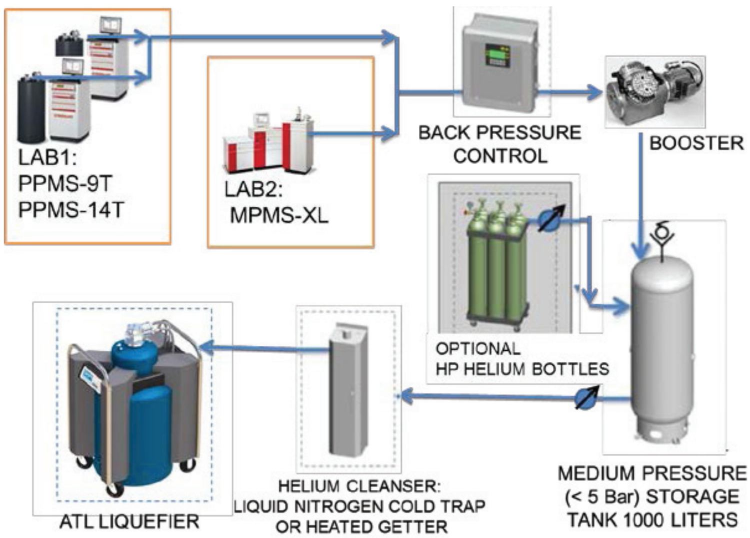

Fig.1. MP-HRP implemented to recover helium boil-off from PPMS and MPMS instruments. The circle indicates the position of the two cleansing methods tested in this work.

By adequate design of the booster pump size and the number of storage tanks and ATL liquefiers, this setup may allow the recovery near 100% of the helium evaporating from low-temperature instruments. The helium gas is conducted exclusively through metallic pipes to eliminate contamination via air diffusion. This is critical to keep oxygen content below 10 ppm. A back-pressure control system maintains the pressure seen by the instruments at a programmable constant value, typically 1 bar absolute with variations below 0.1%. Using a booster pump, the helium is pumped and then compressed and stored into one or several medium pressure (MP) storage tanks to up to 5 bar relative. The ATLs are fed by helium gas coming from the storage tanks, after passing through a helium cleanser, liquid nitrogen trap or heated getter. The cleanser purifies the helium to ultra-high purity grade (impurities < 0.5 ppm of O_2) in order to guarantee the maximum liquefaction rate of the ATL liquefiers. High purity helium bottles can be optionally added to maintain the tank pressure above a minimum (e.g. 1.4 bar) independently of the instrument boil-off, so that enough helium reaches the liquefier to take profit of its maximum liquefaction capacity when needed.

The analysis of the purified Helium gas has been performed by measuring the oxygen content through a gas analyzer SERVOPRO multi-exact from SERVOMEX company. The fundamental of this instrument is based on a zirconia cell, which offers an accurate and stable measurement in a design that gives a fast response and exceptionally long service life. The measurement resolution of this analyzer is 0.01 ppm of O_2. To analyze the quality of the purified helium gas, from the output of the cleanser a small flow of 0.4 L/min is deviated to the gas analyzer.

3. Results and discussion

A preliminary test on the purifying effectiveness of these helium cleansers that can be done very straightforward consists of detecting the possible influence the final gas purity has in the liquefaction rate of the ATL liquefier. The system used in the experimental plant is the ATL160, the full description of this system can be found in the QD web page www.qdusa.com. The average liquefaction rate of ATL160 is 22 liters/day, but this value depends on input helium quality and pressure. In both cases, when the liquid nitrogen cold trap or the gettering processes have been applied to the MP-HRP, the average liquefaction rate has been maintained always within the average of 22 liter/day. This is a first rough indication that both methods can be applied to the experimental MP-HRP setup and that both of them are functioning properly.

The quantitative results of the O_2 trace contents in helium gas using the gas analyzer SERVOPRO multi-exact has been performed for short periods of time periodically. The commercial helium gas of certified quality N50, i.e. 99.999 % of purity with < 2 ppm O_2 guaranteed, provided in a bottle of 50 liter volume at 200 bar, was also measured. The average values of the obtained results are shown in table 1.

Table 1. Oxygen contents in helium gas.

Helium gas analyzed	O_2 (ppm)
He N50	0.75
LN2 cold trap	0.57
Heated Getter	0.02

The liquid nitrogen cold traps have been used in the MP-HRP experimental setup for a period of approximately two months. The experimental MP-HRP is a non-stop system, so the cold traps are duplicated and when one of them is cooled, the other one is in the regeneration status, so that they are alternative changing their status. Each trap is refrigerated by immersion in an open dewar of ten liters volume of liquid nitrogen. The regeneration consists on disconnecting the trap from the circuit and heating the cold trap to room temperature. This process is performed approximately once per week for each trap alternatively. To maintain the cooling in the trap connected to the MP-HRP plant, it is necessary to refill the open dewar every day by six liters of liquid nitrogen.

The heated getter replaced the liquid nitrogen cold traps in the MP-HRP and afterwards it has been in operating status for one year of time approximately. The duplicity of this method is not necessary since the regeneration (replacement of the getter material) is required only about once per year and it can be done in few minutes.

In order to compare the economic cost of each purifying method, the parameters we did take into account were: (1) initial investment, (2) liquid nitrogen/electric power maintenance, (3) manpower maintenance and (4) regeneration cost. Some of these values can be quite different from one laboratory to another; the estimation is made for our laboratory at University of Zaragoza. The numbers in the calculation reported in what follows are just an approximation for the case study at UZ, and they are summarized in Table 2.

Table 2. Economic cost of two methods applied to remove low level impurities in recovered helium gas.

Method for gas purifying	Initial investment (€)	LN2/electric power (€ per year)	Manpower (€ per year)	Regeneration (€ per year)
LN2 cold trap	4000	1314	2737	0
Heated Getter	7000	282	8	2700

The initial investment depends on both cases on the price obtained from the provider company. The case study here has been done for a double liquid nitrogen cold trap, because it is necessary to maintain the MP-HRP experimental setup in a non-stop mode. The price of the heated getter systems depends on the maximum gas flow rate for which they are designed. The two systems considered here can supply similar gas flows, nominal flow rate is $8.3x10^{-5}$ m^3/s and maximum flow rate is $8.3x10^{-4}$ m^3/s.

The calculation for the cost of the electric power supply as well as the cost of liquid nitrogen are different for each country and even it could be different for each university. In the case under discussion these values are 0.124 € per kWh and 0.60 € per liter of liquid nitrogen. The electrical power required by the heated getter it is also depending on the commercial system used. The heated getter applied in the experimental setup requires power supply of 260 W. The liquid nitrogen consumption is 6 liters per day. To maintain the refrigeration of these LN_2 traps the consideration is that a technical operator fills these open dewar manually. Other possibilities for cold traps refilling exist on the market, for example systems which can be refilled automatically, so these numbers should be taken only as an approximation case study. The cost of manpower is considered as 15 € per hour. The estimation of working time needed to refill the cold trap is about 30 minutes per day. The time required for replacing the cartridge of a heated getter is about 30 minutes, but this is done only once per year approximately. The elapsed time for changing the cartridge depends on the incoming impurities contained in the recovered helium gas. The consideration made here is that the impurities level is around a few tens ppm in total, i.e. considering impurities of oxygen and nitrogen mainly. The price for the replacement cartridge depends on the commercial heated getter applied in the MP-HRP. This value depends on the provider and also on trade agreement. Figure 2 shows the accumulative cost of each method from the initial investment, considering a full time operating MP-HRP setup.

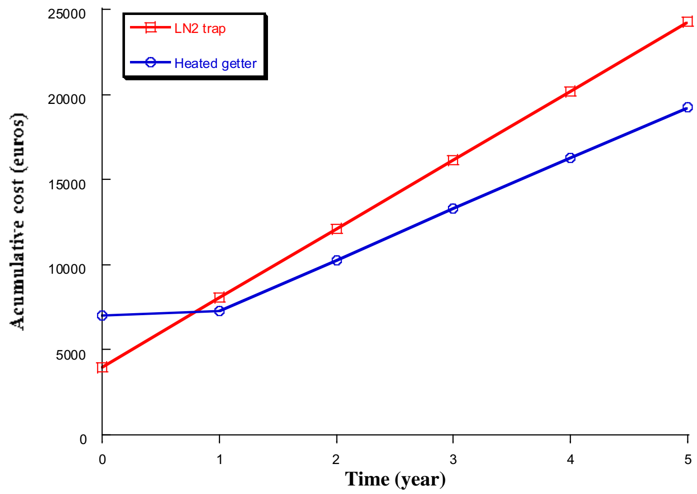

Fig. 2. Economic cost comparison of LN_2 cool trap and heated getter over 5 years of operation.

4. Conclusion

A comparison of two different methods applied for the purification of recovered helium with low level of impurities is reported in this paper. One purification method is the use of liquid nitrogen traps and the other one is the application of a purification system based on getter materials.

Both methods have been tested in the MP-HRP experimental setup and both of them are effective for achieving the proper average liquefaction rate in the ATL160. However the results obtained on oxygen traces content reveals that the gettering purification system reaches lower oxygen content on purified helium.

The initial investment to install one of these purification methods is quite comparable; the budget of both of them falling in the same order of magnitude. The final cost depends on trade agreements. The maintenance evaluation depends on values that could fluctuate between countries: price of liquid nitrogen liters vs. price of power electricity supply and price of technician manpower.

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