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Removing Gaseous Contaminants in ^3He By Cryogenic Stripping

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The Tritium Operations Group at LLNL, Tritium Facility has recently developed a ^3He recovery system to remove argon, xenon, neon, hydrogen, and all other contaminants from the ^3He stream in an Accelerator Production of Tritium (APT) experimental apparatus. In this paper we will describe in detail the background information, technical requirements, our design approach, and the results of our experimental tests.

Background

It is anticipated that during the course of APT experiments undesirable by-products may be introduced into the ^3He gas system. These contaminants may include neutron-activated spallation products from the test chamber, particulate matter, and undesirable gases, such as methane. Additionally, gases intentionally introduced into the APT apparatus as tracers must later be removed from the ^3He stream. These tracer gases include hydrogen, argon, xenon, krypton, and neon.

Many of the anticipated gaseous contaminants could easily be removed from the gas phase by LN_2 cryo-trapping at 77 k. However, a potential by-product of accelerator experiments, is not effectively cryo-condensed at 77 k. Although the quantity of neon produced per experiment is not known, removing neon from the process gas is desirable to prevent a neon "build-up" within the APT test loop. To separate neon from ^3He , it is necessary to operate a cryogenic trap at a temperature below the condensation temperature of neon, (approximately 24 k at atmospheric pressure, see figure 1). To achieve this temperature without the use of expendable liquefied gases a cryogenic refrigerator was required.

Technical requirements

- 1) ^3He purity: all contaminants are to be reduced to concentrations below the detection limits of gas phase mass spectrometry instrumentation (2 to 10 ppm).
- 2) Gas flow rate: $300\text{ cm}^3/\text{minute}$ (0.005 liters/sec).
- 3) Recovery system physical size: system must be portable and able to move through standard width hallways.
 $60'' \times 30'' \times 50''$ (L x W x H).

Design approach

It was decided that the only practical way to capture unwanted gases and by-products effectively and cleanly was to separate them from ^3He by cryo-condensation. We carefully reviewed commercially-available hardware used for cryogenic gas purification. Time and financial constraints compounded with stringent technical requirements made it impossible to purchase and reconfigure a commercially available gas purification system on-time and on-budget. Additionally, the system we investigated most carefully used charcoal adsorbers. We were concerned these charcoal adsorbers would cryo-condense helium. With these factors in mind, we opted to modify a compressed helium cryogenic vacuum pump to meet the stringent gas purification requirements within the operating schedule.

The design concept for our cryo condenser included a closed-loop system in which gas at room temperature and atmospheric pressure could be brought into the body of a cryogenic vacuum pump through a ceramic thermally-insulating feedthrough (see figure 2). On the inside of the vacuum pump body thin wall stainless steel bellows were used to connect to the copper condensation loop attached to the 15 k stage of the cryo refrigerator. In addition to providing a flexible conduit, the stainless steel bellows served as a thermal insulator isolating the copper condensation tube from external heat loads.

A commercially available six-inch CTI cryogenic vacuum pump was selected as it contains a cryogenic refrigerator the second stage of which routinely achieves a temperature of 15 k. The refrigeration capacity of this model is approximately 80 W at the first stage (80 k) and 2 W at the second stage (15 k). The pump body and radiation shield provided the vacuum jacket required to prevent excessive heat

load to the 15 k cold stage. To accommodate our copper condensing coil, stainless steel bellows and required fittings, both the 80K chevron inlet array and the 15 k charcoal array were removed.

We designed the system to achieve a flow rate through this coil of approximately two liters/minute at atmospheric pressure. Cryo-condensing coils constructed from stainless steel and copper were evaluated, as were various methods of bringing the gas through the body of the pump. After several design iterations, we determined that a copper condensing coil, attached to the 15 k second stage, thermally insulated from external heat loads using thin-wall stainless bellows and ceramic thermal insulators in the vacuum pump feedthrough was capable of achieving our gas purification goals.

The condensing coil consisted of one turn of 1/4" OD OFHC copper furnace brazed to a 1.5" diameter copper disk, which was attached to the top of the second stage. Indium foil was used to ensure thermal contact between the 15 k second stage and the condensing coil mounted brazed to the copper disc.

Each design iteration of the cryo-condensing coil was evaluated to ensure that thermal load to the cryo-refrigerator would not overload its cooling capacity. Following assembly of the ^3He purification system, it was operated without gas flowing to verify that the heat load due to thermal conduction in the closed-circuit loop was acceptable. Helium gas was then flowed through the closed-circuit loop to evaluate the heat load due to a flow rate of two liters per minute of helium at atmospheric pressure. The final test consisted of flowing helium gas containing approximately one percent each of the following gases: neon, krypton and argon. During each phase of the evaluation, pressure in the cryogenic vacuum pump was recorded as well as temperature measured at the second stage of the cryo-refrigerator. Gas phase mass spectrometry was performed on gas samples collected at the outlet of the cryo-condensing loop. The results of these analyses indicate that the tracer gases having an initial concentration of approximately one percent by volume were reduced to or below the detection limits of the mass spectrometer by the cryo-condensing loop.

A detailed schematic of the ^3He Recovery System is presented in Figure 3.

Results

The Analytical Sciences Division of the Chemistry and Materials Science Directorate at LLNL provided gas phase mass spectrometry for our sample analysis. Samples of the test gas were collected as the gas exited the cryogenic stripper. Analysis of the gas samples collected from several stripping operations revealed that the concentration of noble gases mixed with the helium were below detectable levels, (10ppm). A copy of the analytical laboratory results is presented in Appendix A. The analytical results also demonstrate that the cryo-condensation system was able to remove nitrogen to below detection limits (2ppm), and oxygen to below 5ppm.

Conclusions

We have developed and tested a gas purification system designed to remove gaseous contaminants from ^3He . This system consists of a modified cryogenic vacuum pump and a closed loop cryo-condensation circuit thermally attached to the 15 k stage of the cryo-refrigerator. Tests were conducted to evaluate the ability of the system to remove the noble gases, argon, xenon, and neon. Analytical results confirm that contaminant gases are reduced to the detection limits of mass spectrometry instruments (~2 to 5 ppm) by the system. We believe this gas purification system may have other applications, as it provides at a reasonable cost an efficient method for purification of gaseous helium.

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Figure 1. Melting points of gaseous elements at 1 atm pressure.

Figure 2. Modified compressed helium cryogenic vacuum pump. Inlet and 15 k arrays have been removed from the pump. Gas to be purified enters through a ceramic-insulated feedthrough, and passes through a thin-wall stainless steel bellows. Gases other than helium are cryo-condensed in the copper coil attached to the 15 k second stage of the cryo-refrigerator. Purified helium flows on through the copper condensing coil, stainless steel bellows and exits the pump via another ceramic feedthrough.

SCHEMATIC HELIUM-3 RECOVERY SYSTEM

Figure 3. Schematic of the ^3He Recovery System.

Appendix A: Analytical Results

LAWRENCE LIVERMORE NATIONAL LABORATORY
CHEMISTRY AND MATERIALS SCIENCE DEPARTMENT
Analytical Sciences Division
Organic Analysis Group

ANALYTICAL GAS MASS SPECTROMETRY SAMPLE ANALYSIS REPORT

REQUESTOR: Coombs
ACCT. NO.: 5284-37

DATE: August 17, 1995

SAMPLE: 7:20
RUN NO'S.: 15108-15110

RESULTS: All results are expressed in mole (volume) percent unless otherwise specified.

Component	Concentration	Std. Dev.
nitrogen N2	8 ppm	2 ppm
helium He	99.998	0.011
oxygen O2	9 ppm	3 ppm

NOTE: A trace analysis was performed on this sample specifically for neon, argon and xenon. None of these compounds were detected. The detection limits are given below:

neon Ne	10 ppm
argon Ar	10 ppm
xenon Xe	10 ppm
nitrogen N2	2 ppm
oxygen O2	5 ppm

If you have any questions feel free to call me at X38530.

Analyst: Jean A. Lindsey
Jean A. Lindsey
Chemist

LAWRENCE LIVERMORE NATIONAL LABORATORY
CHEMISTRY AND MATERIALS SCIENCE DEPARTMENT
Analytical Sciences Division
Organic Analysis Group

ANALYTICAL GAS MASS SPECTROMETRY SAMPLE ANALYSIS REPORT

REQUESTOR: Coombs
ACCT. NO.: 5284-37

DATE: August 17, 1995

SAMPLE: 7:55
RUN NO'S.: 15111-15113

RESULTS: All results are expressed in mole (volume) percent unless otherwise specified.

Component	Concentration	Std. Dev.
nitrogen N2	4 ppm	2 ppm
helium He	99.999	0.012
oxygen O2	5 ppm	3 ppm

NOTE: A trace analysis was performed on this sample specifically for neon, argon and xenon.
None of these compounds were detected. The detection limits are given below:

neon Ne	10 ppm
argon Ar	10 ppm
xenon Xe	10 ppm
nitrogen N2	2 ppm
oxygen O2	5 ppm

If you have any questions feel free to call me at X38530.

Analyst:

Jean A. Lindsey
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Chemist

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CHEMISTRY AND MATERIALS SCIENCE DEPARTMENT
Analytical Sciences Division
Organic Analysis Group

ANALYTICAL GAS MASS SPECTROMETRY SAMPLE ANALYSIS REPORT

REQUESTOR: Coombs
ACCT. NO.: 5284-37

DATE: August 17, 1995

SAMPLE: 8:08
RUN NO'S.: 15105-15107

RESULTS: All results are expressed in mole (volume) percent unless otherwise specified.

Component	Concentration	Std. Dev.
nitrogen N2	14 ppm	3 ppm
helium He	99.997	0.013
oxygen O2	8 ppm	3 ppm

NOTE: A trace analysis was performed on this sample specifically for neon, argon and xenon. None of these compounds were detected. The detection limits are given below:

neon Ne	10 ppm
argon Ar	10 ppm
xenon Xe	10 ppm
nitrogen N2	2 ppm
oxygen O2	5 ppm

If you have any questions feel free to call me at X38530.

Analyst:

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Analytical Sciences Division
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ANALYTICAL GAS MASS SPECTROMETRY SAMPLE ANALYSIS REPORT

REQUESTOR: Coombs
ACCT. NO.: 5284-37

DATE: August 17, 1995

SAMPLE: Gas Mix Before (1% Xe, 1% Ne, 1% Ar, 97% He)
RUN NO'S.: 15084, 15087

RESULTS: All results are expressed in mole (volume) percent unless otherwise specified.

Component	Concentration	Std. Dev.
nitrogen N2	0.030	0.003
argon Ar	1.15	0.01
helium He	96.71	0.27
neon Ne	1.07	0.02
xenon Xe	1.04	0.01

If you have any questions feel free to call me at X38530.

Analyst:

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ANALYTICAL GAS MASS SPECTROMETRY SAMPLE ANALYSIS REPORT

REQUESTOR: Coombs
ACCT. NO.: 5284-37

DATE: August 30, 1995

Test using ProType No. 3

SAMPLE: Sample Test #2 10:43
RUN NO'S.: 15150, 15151, 15152, 15160

RESULTS: All results are expressed in mole (volume) percent unless otherwise specified.

Component	Concentration	Std. Dev.
nitrogen N2	0.0020 - 20 ppm	0.0002
helium He	99.998	0.013

NOTE: A trace analysis was performed on this sample specifically for neon, argon and xenon. None of these compounds were detected. The detection limits are given below. The type of analyses performed were identical in scope and precision to the analyses previously reported on August 17, 1995.

neon Ne	0.0010
argon Ar	0.0010
xenon Xe	0.0010
nitrogen N2	0.0002
oxygen O2	0.0005

If you have any questions feel free to call me at X38530.

Analyst:

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Analytical Sciences Division
Organic Analysis Group

ANALYTICAL GAS MASS SPECTROMETRY SAMPLE ANALYSIS REPORT

REQUESTOR: Coombs
ACCT. NO.: 5284-37

DATE: August 30, 1995

Test using prototype No. 3

SAMPLE: Sample Test #3 11:02
RUN NO'S.: 15153, 15154, 15155, 15161

RESULTS: All results are expressed in mole (volume) percent unless otherwise specified.

Component	Concentration	Std. Dev.
helium He	100.000	0.009

NOTE: A trace analysis was performed on this sample specifically for neon, argon and xenon. None of these compounds were detected. The detection limits are given below. The type of analyses performed were identical in scope and precision to the analyses previously reported on August 17, 1995.

neon Ne	0.0010
argon Ar	0.0010
xenon Xe	0.0010
nitrogen N2	0.0002
oxygen O2	0.0005

If you have any questions feel free to call me at X38530.

Analyst:

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ANALYTICAL GAS MASS SPECTROMETRY SAMPLE ANALYSIS REPORT

REQUESTOR: Coombs
ACCT. NO.: 5284-37

DATE: August 29, 1995

SAMPLE: Gas Mix Before (1% Ne, 1% Ar, 1% Xe, 97% He)
RUN NO'S.: 15131, 15136

RESULTS: All results are expressed in mole (volume) percent unless otherwise specified.

Component	Concentration	Std. Dev.
nitrogen N2	0.026	0.001
argon Ar	1.20	0.01
helium He	96.57	0.59
neon Ne	1.11	0.01
xenon Xe	1.09	0.00

If you have any questions feel free to call me at X38530.

Analyst:

Jean A. Lindsey
Jean A. Lindsey
Chemist

LAWRENCE LIVERMORE NATIONAL LABORATORY
CHEMISTRY AND MATERIALS SCIENCE DEPARTMENT
Analytical Sciences Division
Organic Analysis Group

ANALYTICAL GAS MASS SPECTROMETRY SAMPLE ANALYSIS REPORT

REQUESTOR: Coombs
ACCT. NO.: 5284-37

DATE: August 30, 1995

Test using prototype No. 3

SAMPLE: Sample Test #1 10:05
RUN NO'S.: 15150, 15151, 15152, 15159

RESULTS: All results are expressed in mole (volume) percent unless otherwise specified.

Component	Concentration	Std. Dev.
nitrogen N2	0.362	0.025
helium He	99.633	0.053
oxygen O2	0.0050 - <i>50 ppm</i>	0.0005

NOTE: A trace analysis was performed on this sample specifically for neon, argon and xenon. None of these compounds were detected. The detection limits are given below. The type of analyses performed were identical in scope and precision to the analyses previously reported on August 17, 1995.

neon Ne	0.0010 - <i>10 ppm</i>
argon Ar	0.0010 - <i>10 ppm</i>
xenon Xe	0.0010 - <i>10 ppm</i>
nitrogen N2	0.0002 - <i>2 ppm</i>
oxygen O2	0.0005 - <i>5 ppm</i>

If you have any questions feel free to call me at X38530.

Analyst:

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