Improved helium exchange gas cryostat and sample tube designs for automated gas sampling and cryopumping

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In order to eliminate the use of liquid helium for the extraction of atmospheric gases from polar ice cores, two units of a redesigned top load helium exchange gas cryostat were built and tested. The cryostats feature the shortest and largest diameter sample wells built to date, a base temperature below 7 Kelvin, and a sample well without baffles. The cryostats allowed shortening the length and thus increasing the gas pressure inside our sample tubes by 58% and increasing the amount of sample ending up in the mass spectrometer by 4.4%. The cryostats can either be used as mobile stand-alone units for manual gas processing lines or integrated into a fully automated vacuum extraction and gas analysis line. For the latter application the cryostat was equipped with a custom-designed automated changeover system.

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1. Introduction

Our laboratory uses stable isotope ratio mass spectrometry for analyzing the chemical composition and stable isotope ratios of air extracted from polar ice samples at high temporal resolution. Of particular interest are the causes and consequences of past abrupt climate changes dating back several 10,000 years [e.g., Taylor et al., 2004; Caillon et al., 2003; Severinghaus et al., 1998, 2003; Brook et al., 2000; Severinghaus and Brook, 1999]. We are currently operating five stable isotope ratio mass spectrometers and two manual vacuum processing lines for extract-
ing and processing air samples. We are also in the process of building a third, fully automated vacuum extraction line.

Traditionally, air samples extracted from ice cores are cryopumped into long stainless steel sample tubes partially immersed in a liquid helium tank. (Consequently, the sample tubes are customarily referred to as “dip tubes”.) Because of the high sample throughput, which may reach 200 samples per week, our laboratory consumes two to three 60 L liquid helium tanks over a 2 week time frame. At a cost of ~$350 per tank this substantially adds to the operating costs of the laboratory. Helium is also a nonrenewable resource that is permanently depleted under the current practice. For these reasons, and to eliminate the inherent risks involved in the use of liquid helium (A leak in the vacuum line may cause large amounts of air to be cryopumped into the helium tank followed by a catastrophic release of the frozen air, once the liquid helium is used up.), we decided to eliminate liquid helium in our laboratory altogether, and to build two units of a custom-designed helium exchange gas cryostat.

Helium exchange gas cryostats are the alternative to using liquid helium and have been successfully used for the collection of air and gas samples by several other geochemical laboratories. Depending on the design, existing cryostats allow the simultaneous cooling of up to eight dip tubes. Integrated in an automated extraction line, the cryostats need to be warmed up before the air samples can be transferred into the mass spectrometer, and cooled down again to base temperature. Each warming/cooling cycle requires at least 4 hours, which limits the sample throughput through the extraction line. For this reason, we specified that our two new helium exchange gas cryostats should allow the removal of just a lightweight dip tube containing the sample, while the bulk of the cryostat remains cold. Further, the cryostats should allow cooling a minimum of two 6.35 mm diameter stainless steel dip tubes to a base temperature of 10 K or lower. The length of the sample well was specified to be considerably shorter than the sample well of a cryostat that represented the previous state of the art. This would allow us shortening the dip tube and maximize the amount of gas sample that can be transferred into the mass spectrometer.

2. Cryostat Design

The starting point for the new cryostat design was an optical ARS OMNIPLEX top load exchange gas cryostat. The following modifications were made (Figure 1): (1) A 177.8 mm long protrusion at the bottom of the cryostat, which would normally house a sample holder for spectroscopic experiments was replaced with a removable plate. (2) The sample well (also referred to as exchange gas volume) was shortened and bolted directly to the second stage of an ARS DISPLEX DE-204NI closed cycle expander via a robust second stage thermal link. (3) The exchange gas volume was made from an ultrathin wall stainless steel tube and a one-piece sample well made from oxygen-free, high conductivity (OFHC) copper. (4) The top flange shown in Figure 2 replaced the access port to the sample well and the instrumentation interface, including the connections for the helium exchange gas line and a pressure relief valve. Four bored-through Ultra-Torr adapters with 6.35 mm bores (Swagelok, SS-4-UT-A8-BT) allow inserting up to three dip tubes and a rod-mounted silicon diode temperature probe (Lake Shore, DT-670B-SD) into the sample well. The helium exchange gas line and the pressure relief valve were attached to a radial extension tube ending in a central bore of the top flange. The sample well is 274.1 mm long (measured from the top surface of the top flange to the bottom of the well) and 50.8 mm in diameter. For comparison, the cryostat of the previous state of the art was equipped with a 305 mm long and 25.4 mm diameter sample well (S. Azer, Janis Research Company, personal communication, 1 October 2003).

Closed cycle helium cryostats use high-pressure helium gas to produce cooling. Depending on the expander model, base temperatures between 4.2 K and 35 K can be reached. The cooling process is based on a modified Gifford McMahon cycle. A helium compressor provides high-pressure gas to the expander through a flexible gas line. The expansion of the gas at different stages produces the refrigeration. Low-pressure gas is then returned through another gas line, where it is recycled through the compressor. This closed loop cycle can be continuously repeated and maintained as needed to produce the desired refrigeration.
sample well is filled with helium gas at atmospheric pressure, which acts as a cold bath. The sample well is filled using the following method: At the initial start-up, and whenever the cryostat has not been in use for some time, the sample well first needs to be flushed with helium to expel any atmospheric air that may have entered. A flexible Dekoron tube (Dekoron/Unitherm, Cape Coral, FL) connected to a helium gas cylinder is carefully inserted (with helium flowing) into the bottom of the sample well through one of the Ultratorr adapters on the top flange. The helium gas pressure is adjusted to a slight overpressure and the sample well flushed for about one minute. The presence of a helium gas flow is confirmed by touching the pressure relief valve. (A slight stream of gas should be felt. Depending on the surrounding noise level, a weak hissing sound may also be audible.) After pulling the flexible helium supply line out and plugging the Ultratorr adapter (or reinserting the item that was installed before), the helium gas cylinder is switched over to the permanently installed helium supply line extending to the left in Figure 2. During operation of the cryostat, the permanent helium supply line is used to top off the sample well with helium gas to compensate for changes in barometric pressure, contraction of the helium exchange gas during cooling, or the removal of a dip tube. A pressure increase inside the exchange gas volume is vented through the pressure relief valve. In our current (improvised) setup, the helium supply lines are connected via two ball valves and a Tee connector to the dual-stage pressure regulator of the helium gas cylinder. Since the pressure inside the sample well is currently not monitored, the only way of knowing that the pressure inside the sample well is sufficiently high, is by adjusting the gas regulator to a slight over-pressure, i.e., just enough that a very light gas stream can be felt at the outlet of the pressure relief valve. In a more sophisticated permanent setup, a pressure transducer monitoring the pressure inside the sample well would be connected to a relay and a solenoid valve on the helium supply line. This alternative setup would top off the helium exchange gas volume only when needed, i.e., when the pressure falls below atmospheric pressure. The additional cost of installing a pressure-monitoring unit would be recuperated over time by savings in helium gas. (The current improvised setup consumes about one 50 L helium gas cylinder in two weeks of daily cryostat operation.) Note that we did not observe any change in the base temperature, if we intermittently turned off the helium gas
Thus the slight flow of helium through the top layers of the helium cold bath (which are at room temperature) does not introduce significant turbulence in the cold bath and does not measurably increase the heat load for the expander.

No baffles were installed in the sample well, although the installation of baffles is normally recommended for helium exchange gas cryostats. The baffles disrupt the formation of convective gas loops in the helium exchange gas, which in turn increase the heat load on the expander and increase the base temperature inside the sample well. However, it would be straightforward to retrofit the top flange with baffles that extend into the sample well, if needed for future applications.

3. Dip Tubes

The dimensions of our 60 L liquid helium tanks (Figure 3a) dictated the length of our existing long dip tubes. The new cryostats allowed shortening the dip tubes from 780 mm to 326 mm. The new dip tubes extend 272 mm into the sample well providing 2 mm of free space between the bottom of the dip tube and the floor of the sample well. This gap prevents a collision between the sample well and the dip tube, which may result in damage to the dip tube or negatively impact the integrity of the joint between the copper sample well and the thin wall stainless steel tube. The dip tubes are connected to the valves via VCR face seal fittings and copper gaskets. The dip tubes are equipped with manually operated bellows valves (Swagelok SS-4H-VCR), manually operated quarter turn diaphragm valves (Carten Controls, MDQ250-03-LV), or pneumatically actuated diaphragm valves (Carten Controls, MDA250-03-LV-NC). (Note that the valve body and seats of the quarter turn and pneumatic valves from Carten Control are identical.) In order to increase the internal surface, on which the gas samples adsorb during cryopumping, the dip tubes were equipped with 50.8 mm long and 3.175 mm diameter stainless steel tube inserts. (We decided against using stainless steel balls or filings, as these may damage the valve seats.)

Figure 4 shows a prototype of a short dip tube with integrated silicon diode temperature sensor. It was made from a heavy wall 6.35 mm stainless steel tube (wall thickness 1.651 mm). The temperature sensor was mounted in a groove on the bottom of the dip tube. The electrical leads were guided through a matching groove along the side of the tube and threaded through a bore on a female VCR nut that was spot welded to the dip tube. The grooves were filled with a low-temperature resistant epoxy to protect the sensor and wiring.

Dip tubes with integrated temperature sensors have two main advantages. First, integrating the temperature sensor into the dip tube frees up the sample port currently occupied by the temperature sensor rod. Second, more accurate information about the sample temperature is obtained, as the temperature is measured at the sample tube, and not at a different location inside the sample well. The temperature of the dip tube can be monitored at all times, including during warm up and cool down. It is also possible to cool the dip tube to known intermediate temperatures by partial insertion into the sample well.

4. Automated Changeover System

The first cryostat was integrated in the automated vacuum processing line. It was equipped
with the automated changeover system shown in Figures 1b and 5 (University of California, San Diego, Technology Transfer and Intellectual Property Services, Automated Changeover System for a Cryostat, case SD2005-239/SD2005-837, http://invent.ucsd.edu/technology/cases/2005/SD2005-239.htm). Two dip tubes with pneumatically actuated valves were permanently mounted on two

**Figure 3.** (a) Comparison of the dimensions of the long dip tubes used to freeze out gas samples in liquid helium and the new short dip tubes in front of a standard 60 L liquid helium tank. (b) View of the mobile helium exchange gas cryostat that will replace the liquid helium tanks.

![Figure 3](image1)

**Figure 4.** Prototype of a short dip tube with integrated silicon diode temperature sensor (a) before and (b) after the installation of the silicon diode. The groove protecting the sensor and electrical leads is filled with a low-temperature resistant epoxy.

![Figure 4](image2)
piston-actuated translation stages. Four solenoid valves on a solenoid manifold card (Clippard, EMC-12-24-40) switch the pressurized air operating the upward and downward movement of the two pistons (Clippard, UDR-20-10-MB). The solenoids are actuated by four LabVIEW-controlled relays on an SCXI-1160 relay board (National Instruments). The pistons were equipped with permanent magnets that allow sensing the piston position via Hall effect sensors (Clippard, HS-9901) mounted along the side of the cylinders. The output signal of the Hall effect sensors is read by a PXI-6052E data acquisition board (National Instruments) and used in a feedback loop to operate the solenoid valves controlling the air supply to the pistons. This setup allows lowering the dip tube into the cryostat in several steps, which allows optimizing the cooling of the dip tubes, and the cryopumping efficiency. (Experience has shown that the gas transfers are more complete, if the dip tube is inserted in several steps into the cryostat or liquid helium tank. A possible reason is that each insertion step exposes fresh surface to the gas sample for adsorption.)

The rack holding the cryostat and the changeover system was constructed from 40 × 40 mm aluminum profiles (Maytec, 1.11.040040.43L). The two pneumatic cylinders were mounted on a 40 × 80 mm aluminum profile (Maytec, 1.11.040080.64L), which also acts as the base for the two translation stages (visible in Figures 2 and 5). During operation, the changeover system alternates between the two dip tubes: While one dip tube is inserted into the cryostat to freeze out a gas sample, the other dip tube is pulled out to allow the previous gas sample warming up to room temperature before being expanded into the mass spectrometer for analysis. For the next sample, the two dip tubes switch roles, and so on.

5. Mobile Standalone Unit

The second cryostat was mounted on the cart shown in Figure 3b. The cart was built using the same 40 mm aluminum profiles as the automated extraction line. It has a form factor similar to the standard 60 L liquid helium tanks. The cart features a 500 × 500 mm square base and a height that is adjustable from 820 to 1300 mm. With the frame in its upper position, the cryostat top flange is positioned at the same height as the inlet of the liquid helium tank. In its lower position, it is possible to insert the existing long dip tubes into the cryostat. The cart was equipped with a bottom shelf for stowing instrumentation or lead (Pb) bars to lower the center of gravity. (There exist several other solutions to lower the center of gravity, e.g., enlarging the footprint of the cart, or raising the location of the wheels. However, these alternatives would considerably change the form factor of the cart.) To reduce the vibration from the Displex expander, a 25.4 mm thick natural rubber collar (Mason Industries, BBNR pad) with a central 203.2 mm diameter cutout was placed around the...
vacuum shroud of the cryostat and seated on the aluminum profile frame. An instrument panel equipped with a temperature indicator (Scientific Instruments, Model 1900) and a digital thermocouple gauge (Duniway Stockroom, DTC-06M-115) allows checking the temperature inside the sample well and the residual pressure in the vacuum shroud from across the laboratory.

Our water-cooled compressors are permanently installed in a separate room. Thus the mobility of our cryostat is currently limited to the range of the pressurized helium supply lines. It is possible to convert the cryostat into a completely mobile unit by mounting the compressor, a closed cycle water cooler, and a dedicated vacuum pump on the same cart. Alternatively, an air-cooled compressor may be used. In a laboratory setting, the inclusion of adequate provisions for noise insulation of the cart should also be considered.

6. Results and Discussion

6.1. Performance Tests of the Two New Cryostats

6.1.1. Base Temperature

The base temperature of the cryostats depends on how many dip tubes are inserted into the sample well. Each stainless steel dip tube (6.35 mm diameter, 0.889 mm wall thickness) adds approximately 0.2 W of conductive heat load that needs to be dissipated by the expander. For the performance tests of the first cryostat, the cryostat shroud was evacuated to 12 mTorr using a turbomolecular pump (Pfeiffer Vacuum, TMU071 Turbo Drag Pump) connected to an oil-free forevacuum pump (Varian, Model SH-100 Dry Scroll Pump). For measurements under normal operating conditions, the cryostat was evacuated to 38 mTorr using the dry scroll pump alone (specified final pressure 50 mTorr). For our application, these residual pressures are sufficient. The shroud was not baked out during pumping, and we did not wait for the pressure to reach the final pressure of the turbomolecular pump, which is specified as 7.5 \(10^{-5}\) mTorr. Note that as soon as the cryostat reaches a temperature of 100 K, it is recommended to disconnect the cryostat from the vacuum pump to prevent air from being cryopumped back through the pump into the vacuum shroud.

Table 1 compares the calculated and measured base temperatures of the two cryostats. With base temperatures well below 10 K, the performance of both cryostats is excellent and exceeding our specifications.

Table 1. Comparison of Calculated and Measured Base Temperatures for the Two Cryostats With Up to Three Dip Tubes and One Temperature Sensor Rod Simultaneously Inserted in the Sample Well

<table>
<thead>
<tr>
<th>Base Temperature</th>
<th>Pressure</th>
<th>No Dip Tube</th>
<th>One Dip Tube</th>
<th>Two Dip Tubes</th>
<th>Three Dip Tubes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Calculated</td>
<td>&lt;8 K</td>
<td>~8.5 K</td>
<td>~9 K</td>
<td>~9.5 K</td>
<td></td>
</tr>
<tr>
<td>Measured</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Serial number 04-C291</td>
<td>~12 mTorr</td>
<td>6.3 K</td>
<td>6.6 K</td>
<td>7.1</td>
<td>7.3 K</td>
</tr>
<tr>
<td>Serial number 04-C291</td>
<td>~38 mTorr</td>
<td>6.8 K</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Serial number 04-C292</td>
<td>~38 mTorr</td>
<td>5.8 K</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

The pressure is the residual pressure in the vacuum shroud.

6.1.2. Cool Down Rate and Temperature Stability

Figure 6 shows the temperature vs. time curve during testing of the first cryostat at 12 mTorr residual pressure. All dip tubes were filled with air at room pressure and temperature and the inlet valves closed. The initial cool down rate (without dip tubes) was 4 to 5 K/min. A base temperature of 6.3 K was reached after 2.4 hours, and remained stable for over 22 hours. At this point, the first dip tube was inserted into the sample well in several steps, and fully inserted after about five minutes. After full insertion, the temperature increased to 7.8 K. Within 20 min, the temperature fell to 6.7 K and reached a new base temperature of 6.6 K after
45 min. The second dip tube was inserted all the way into the sample well within a few seconds. The temperature jumped to 23.2 K, then cooled down to 10 K within 3 min and reached a new base temperature of 7.1 K after 17 min. The third dip tube was inserted in two stages. First, it was inserted halfway down and kept at this depth for two minutes. Then it was fully inserted, at which point the sample well temperature rose to 10.2 K. The sample well cooled down to 8.1 K in 6 min, and reached a new base temperature of 7.3 K after 29 min. The experiment was concluded after 29 hours. Under normal operating conditions the cryostat was operated at base temperature for several days at a time.

6.2. Isotope Ratios of Air Samples Collected With the New and Existing Methods

In order to compare the quality of analyses of air samples collected in the short dip tubes and cooled with the new cryostat with the quality of analyses of air samples collected in the long dip tubes and cooled with liquid helium, air samples from the same air standard were transferred into the respective dip tubes by the two methods. The air samples were then compared to the same air standard from a storage tank by stable isotope ratio mass spectrometry. Any alteration of the air samples (i.e., isotopic fractionation) during sample preparation would show up as deviations of the measured from the expected isotope ratios that are larger than the combined uncertainty of the current manual extraction method and the mass spectrometric analysis. The theoretical isotope ratios were 0.000 per mil for both $\delta^{15}N$ and $\delta^{18}O$, calculated as

$$\delta^{15}N = \left( \frac{^{15}N/^{14}N_{\text{sample}}}{^{15}N/^{14}N_{\text{atmosphere}}} - 1 \right) \cdot 10^3 \; \text{%o},$$

$$\delta^{18}O = \left( \frac{^{18}O/^{16}O_{\text{sample}}}{^{18}O/^{16}O_{\text{atmosphere}}} - 1 \right) \cdot 10^3 \; \text{%o}.$$

Table 2 shows that the analysis results compare well with each other and suggest that the three sampling and cooling methods may be used interchangeably in the future. Note also that the shorter dip tubes allowed us increasing the sample pressure in the mass spectrometer by 4.4%.

For experienced users of the traditional method, the higher residual pressures in the vacuum line after completion of the gas transfer are in need of getting used to. In contrast to gas transfers into long dip tubes that are immersed in liquid helium, a helium exchange gas cryostat is not capable of freezing out the helium contained in the air sample. Thus a measurable residual pressure will always remain. For most applications, this is not a concern though. For all practical reasons, instead of watching the end point of the transfer on the vacuum
Table 2. Comparison of the Isotope Ratios of Standard Air Samples Collected by the Traditional Sampling Method and the New Methoda

<table>
<thead>
<tr>
<th>Method</th>
<th>Type of Transfer</th>
<th>Number of Transfers</th>
<th>$\delta^{15}\text{N} \pm 1\sigma$</th>
<th>$\delta^{18}\text{O} \pm 1\sigma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Traditional sampling method</td>
<td>long dip tubes cooled by liquid helium</td>
<td>5</td>
<td>0.007 ± 0.004</td>
<td>0.011 ± 0.011</td>
</tr>
<tr>
<td>New method</td>
<td>short dip tubes cooled by helium exchange gas cryostat</td>
<td>10</td>
<td>0.003 ± 0.003</td>
<td>0.004 ± 0.013</td>
</tr>
<tr>
<td>Combined method</td>
<td>long dip tubes cooled by helium exchange gas cryostat</td>
<td>4</td>
<td>−0.003 ± 0.005</td>
<td>0.005 ± 0.006</td>
</tr>
</tbody>
</table>

aFor comparison, a few transfers were also made into long dip tubes that were cooled by the new cryostat. The short dip tubes were equipped with stainless steel tube inserts, as described in section 3. The air samples were compared to the same air standard from which the air samples were produced.

gauge (the pressure falls to “zero”), the progress of the transfer can be followed by recording the pressure curve in the vacuum line using a high-precision Baratron with inconel diaphragm (e.g., MKS Baratron 627 B, temperature regulated to 45 °C, range 10⁻⁵ to 1 Torr). A horizontal leveling off of the pressure curve indicates the completion of the transfer. In applications where helium is of interest, or to check whether there is a leak in the vacuum line, the helium in the vacuum line can be frozen out into a separate dip tube that is filled with activated charcoal at the end of the transfer.

6.3. Performance of the Automated Changeover System

[22] Figure 1b shows one module of the automated changeover system during testing. For these preliminary tests, the dip tube was connected to the vacuum system via flexible Dekoron tubing. For the tests described in the preceding chapter and all subsequent gas transfers, the Dekoron tubing was replaced with a flexible metal bellows (Swagelok, SS-FL4RM4RF4-27H).

[23] The first test series was conducted with the cryostat at room temperature. A simple LabVIEW program operated the piston and automatically inserted and removed the dip tube from the cryostat at 1 min intervals. (Under normal operating conditions, the piston will be actuated once every 40 min.) After some initial friction problems that were solved by adding graphite dry lubricant to the outside of the dip tube, the dip tube could be smoothly inserted and removed from the cryostat several hundred times, at which point the room temperature tests were concluded.

[24] A second test series was performed with the cryostat at base temperature. A simple LabVIEW program operated the piston and automatically inserted and withdrawn 5 min intervals. As before, the dip tube could easily be inserted and removed every time it was actuated, but the actuation was less smooth than during the room temperature tests. This was caused by humidity that froze out on the dip tube as it was pulled out of the cryostat, then melted as the dip tube warmed up, accumulated on the Ultra-Torr adapter, and flushed off the dry lubricant. After 21 cycles, the tests at cryogenic temperature were concluded.

7. Conclusions

[25] Two units of a top load helium exchange gas cryostat with an extremely short (274.1 mm) and large diameter (50.8 mm) sample well have been built and tested. The performance of the cryostats is excellent and exceeds the initial design specifications. Up to four stainless steel dip tubes of 6.35 mm diameter can be inserted simultaneously and cooled to temperatures below 8 K. The top load helium exchange gas sample well allows changing the dip tubes while the cryostat remains at base temperature. A dip tube can be cooled to temperatures below 10 K in about 10 min, which is comparable to the dip tube cooling times in a liquid helium tank. Note that it may be possible to retrofit existing top load helium exchange gas cryostats with similar automated changeover systems. The only part that would need to be customized is the top flange that closes off the sample well.

[26] A good insulating vacuum in the vacuum shroud is essential for operating the cryostat at peak performance. The user manual specifies a residual pressure of 50 mTorr, but we observed that reducing the residual pressure to 12 mTorr...
allows lowering the base temperature by an additional 0.5 K. For other applications, e.g., the quantitative separation of the noble gases for noble gas mass spectrometry, it is very important to maintain the best possible vacuum in the vacuum shroud in order to consistently reach and maintain the lowest base temperature. The additional cost for a turbomolecular pump and heat tape (for periodically baking out the vacuum shroud of the cryostat) is justified by the added cryostat performance. If funds for a dedicated turbomolecular pump are not available, the cryostat can be connected to an existing vacuum line using a vacuum interface similar to the one shown in Figure 7. After the valve separating the shroud from the pump has been closed (at a temperature of 100 K, see above), the turbomolecular pump is no longer needed, and the cryostat can be disconnected from the vacuum line. For these high precision applications, the optional installation of baffles in the sample well will allow lowering the base temperature even further. For our application, this was unnecessary, though.

[27] A stepwise insertion of the dip tubes into the sample well gives the dip tube time to cool to an intermediate temperature before reaching the coldest region of the sample well. Thus the well temperature can be kept below 10 K at all times.

[28] We designed and tested a set of very short dip tubes that maximize the amount of sample ending up in the mass spectrometer. A comparison of the isotope ratios of standard air samples collected with the long dip tubes and with the short dip tubes showed that the analysis results compare well with each other and that the two types of dip tubes and the two cooling methods can be used interchangeably. Two prototypes of dip tubes with integrated temperature sensors await further testing under normal operating conditions.

[29] A fully automated changeover system was built that is interfaced with an automated vacuum extraction line on one side, and will be interfaced with a dedicated mass spectrometer on the other side. This automated changeover system will allow the continuous unattended operation of the automated extraction line. While one dip tube is warming up before the gas is transferred into the mass spectrometer, the next sample is processed and cryopumped into the other dip tube. After every extraction/analysis cycle, the two dip tubes switch roles. The tests of the changeover system at room and cryogenic temperatures showed that the system can be operated continuously over extended periods of time. Occasional addition of a graphite-based dry lubricant will ensure smooth gliding of the dip tubes inside the bores of the Ultra-Torr adapters. (Automated dry lubricant dispensers are commercially available.) The accumulation of moisture on the dip tubes after removal from the cryostat can be reduced by gentle heating to room temperature with a radiant heater.

8. Outlook

[30] We envision two additional potential applications of the described helium exchange gas cryostat design with automated changeover system. The first application is the cryopumping of vacuum systems processing ice cores and humid air. A cryostat with switchable cold surfaces can handle much higher gas loads than a turbomolecular pump. The highly efficient pumping capacity of a helium exchange gas cryostat equipped with an
automated changeover system can thus be utilized to achieve faster pump down rates, especially in these difficult to pump “wet” vacuum systems. It is straightforward to design a changeover system with four translation stages that could handle two cryopumping modules and two dip tubes, as shown in Figure 8. The top flange can be modified to accommodate dip tubes dedicated for cryopumping with diameters up to 12.7 mm. Since the pumping and gas sampling functions of the cryostat would not be used simultaneously (during the transfer of a gas sample, the vacuum line is isolated from the pump station), the cryopumping and gas sampling modes of the cryostat are not expected to interfere with each other.

The second application is the quantitative separation of air and other gas mixtures, and the stepwise concentration of trace gases extracted from multiple gas samples to increase the analytical sensitivity and precision. For these applications the helium exchange gas cryostat would be equipped with an automated changeover system similar to the one depicted in the conceptual drawing shown in Figure 9a, and used together with the dip tubes with integrated temperature sensors shown in Figure 4. Also, the pistons would be replaced with motorized linear actuators in order to insert the dip tubes into the different temperature zones of the cold bath with higher precision and repeatability. An application example is the quantitative separation of the noble gases for noble gas isotope ratio mass spectrometry described by Lott [2001]. (This application requires the quantitative removal of all reactive atmospheric gases from the air sample by gettering prior to the separation step.) The method currently requires a thermal cycling step before the analysis step, i.e., the gas sample is warmed to 35 K for 30 s followed by refreezing to base temperature (<9 K [Lott, 2001]). The freezing out of an air sample inside a cryogenic trap or a dip tube is a very dynamic process. Because of this, the individual gases in the air sample tend to be adsorbed on the container walls as a mixture. In noble gas analysis, this is particularly a problem for neon gas trapped in the argon ice. (The trapping of other gas species in the krypton and xenon ice is negligible due to the usually very low concentrations of these species in the noble gas mixture.) Thermal cycling rereleases the gas mixture into the sample container volume, which at this point is isolated from the rest of the vacuum system. In the second freezing step, the individual gas species are sequentially readsobered on the container walls in a nondynamic process, as the temperature of the container is gradually falling to base temperature. When the sample container is warmed up again at 10 K/min [Lott, 2001], the individual gas species are released in the opposite sequence and can be transferred into the mass spectrometer for analysis.

Using a helium exchange gas cryostat with automated changeover system and dip tubes with integrated temperature sensors, thermal cycling could be accomplished as follows: After isolation from the vacuum system, the dip tube is partially pulled out into a warmer temperature zone of the cold bath and allowed warming up to an intermediate temperature of 35 K (noble gas mixtures) or 100 K (CO$_2$-free and dry air). This rereleases the gas mixture into the dip tube volume. When the dip tube is gradually reinserted into the cryostat, the individual gas species freeze out one after the other in layers with the least volatile gas (xenon) at the bottom, as depicted in Figure 9b. The insertion depth for quantitatively freezing out each gas species depends on the temperature profile of the individual helium exchange gas cryostat and needs to be determined experimentally.
The refreezing method described here could be utilized for the separation of a gas mixture into its pure components. By gradually pulling the dip tube up into the warmer temperature zones, the individual gas species would be sequentially released into the head space of the dip tube and could either be transferred into one of several “sample” dip tubes, or collected in a designated “waste” dip tube. By repeating this process for multiple air samples, it would be possible to concentrate the rare noble gases (e.g., krypton) in the same dip tube. This in turn would allow substantially increasing the sensitivity of the subsequent mass spectrometric analysis of these rare gas species.

Like the method described by Lott [2001], the method described here does not allow freezing out the helium component of the air sample. However, for applications where helium is of interest, it would be very easy to insert a piece of activated charcoal into a separate dip tube, add it to the system, and freeze out the helium at the end of the thermal cycling step. This in turn would allow substantially increasing the sensitivity of the subsequent mass spectrometric analysis of these rare gas species.

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