

Gas Contamination Effects on Pulse Tube Performance

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ABSTRACT

Experiments were performed to quantify the effect of contamination in the helium working gas of a typical pulse tube cryocooler operating at 60 K with 1 W of applied heating load. A quadrupole mass spectrometer with a closed ion source was used to measure the gas compositions both before and after pulse tube operation. Five tests were performed with different combinations of water, carbon dioxide, nitrogen and argon gas contaminating the helium across a range of 20 to 420 parts per million (ppm). Except for hydrogen, no other contaminants were detected in any experiment either before or after running the pulse tube. A decrease in pulse tube refrigeration performance was observed in all five tests, with asymptotic steady-state temperatures reached on a time scale of 7 to 10 days. Under a 1 W applied load condition with a constant piston stroke, the cold head temperature rise ranged from 0.6 to 2.8 K depending on contamination level. Comparisons between cryopumped and non-cryopumped vacuum chambers appeared to rule out the possibility that the contamination was external to the pulse tube. Water and carbon dioxide were the most important contaminants at the nominal operating temperature of 60 K, although there was evidence for nitrogen condensation under some conditions. Warming up of the pulse tube to room temperature tended to restore the original performance, although this improvement degraded more rapidly the second time around to the asymptotic steady-state temperature.

INTRODUCTION

One of the potential life-limiting factors for cryocoolers is internal contamination of the helium gas. Condensible species such as water and carbon dioxide will freeze on cold surfaces at sufficiently low temperatures, leading to blockage of the flow and possibly increased parasitic heat conduction. For example, blockage problems have been observed in the small orifices of J-T cryocoolers and studies have been conducted to understand and mitigate the problem.^{1,2} Long-term testing of Vuilleumier coolers has also addressed the contamination issue.³ Finally, life test studies of the Oxford ISAMS prototype Stirling cooler, the Hughes/Raytheon ISSC BE Stirling cooler and the TRW SBIRS-low mini pulse tube cooler have all showed some evidence of performance loss due to internal contamination.

In order to address the problem of contamination in pulse tube cryocoolers, it was decided to conduct a set of experiments designed to identify the specific contaminant species and the levels at

which they produce a given degradation of performance. The available equipment consisted of a small pulse tube that had no capability for flow visualization or any other means of measuring the formation and distribution of frozen contaminants inside the cryocooler. Therefore, contamination effects were deduced from measured refrigeration performance (cold head temperature and compressor input power) at constant operating conditions, along with measurement of the gas composition before and after pulse tube operation. This enabled us to quantify the contamination effect without being able to explore the specific internal mechanisms of ice formation and performance loss. A new quadrupole mass spectrometer was acquired to perform the composition measurements to a precision of 1 ppm across an atomic mass number range of 1 to 200. The nature of the test setup did preclude, however, the ability to sample the gas during pulse tube operation.

The tests conducted in this study were of 7 to 10 days duration, usually with a follow up test of the same gas after a brief shutdown period. This is a short time scale compared to the many year lifetime requirement of space-based cryocoolers; however, a first order estimate of the mass diffusion time scales within the pulse tube of this test yielded 1 to 2 weeks, suggesting that the primary impact of initial gas contamination would be observed within the chosen time frame. This was confirmed by the experiments. The issue of longer term degradation due to outgassing within the pulse tube awaits further study.

EXPERIMENTAL APPARATUS AND PROCEDURE

Pulse Tube

These experiments were conducted using a device that consisted of a TRW double-inlet orifice pulse tube driven by a Lockheed two-piston back-to-back linear compressor (Fig. 1). The compressor had a maximum swept volume of 5.0 cm³ (2.5 cm³ per piston). The system was typically operated at 44 Hz with helium gas at a pressure of 1.8 MPa (255 psia). The entire assembly was mounted in a vacuum chamber and operated in the pressure range of less than 1×10⁻⁵ torr. The nominal performance under these conditions was 1 W of cooling at 60 K cold tip temperature and 293 K heat rejection temperature. The power input to the compressors was

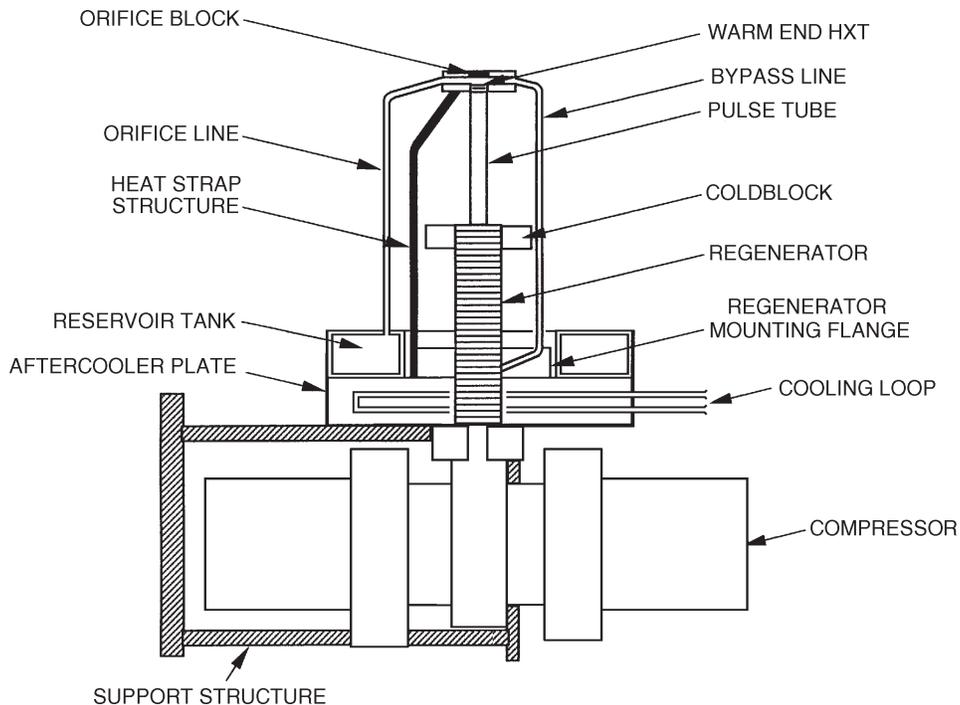


Figure 1. Schematic Layout of Pulse Tube and Compressor

approximately 62 W under these conditions, resulting in a nominal specific power of 62 W/W at 60 K and a Carnot efficiency of 6.3%.

The cold tip temperature was measured with a Lakeshore DT 470 cryodiode connected to a Lakeshore 820 Cryogenic Thermometer readout. For some of the experiments, these data were also recorded on a portable PC computer using National Instruments LabView data acquisition software. Otherwise, the temperature data were recorded by hand. Compressor input power was measured by a Valhalla 2300L Programmable Three-phase Digital Power Analyzer. This measurement was taken between the compressor and the drive electronics and therefore does not account for drive electronic inefficiencies. The heat rejection temperature of 293 K was maintained with a water chiller located outside the vacuum chamber and connected to the fluid loop shown in Fig. 1.

The nominal test procedure was to operate the pulse tube continuously for a minimum of one week. Some of this time would be under no-load conditions, and some under a 1 W of applied load. Satisfactory operation would be verified periodically by inspection of the temperatures, power input, vacuum level, and by compressor piston waveform plots on an oscilloscope. After the first week, the pulse tube was usually turned off and allowed to heat back up to room temperature in order to reverse the effects of condensed impurities in the pulse tube. The pulse tube was then reactivated and performance monitored for a further period of hours or days. One test (G3) employed the use of a Gifford-McMahon cryopump during the second week in order to assess the possibility of condensation on the outside of the pulse tube. Further test details can be found in the Results section.

Clean up of the pulse tube gas after a test was accomplished with a vacuum bakeout technique. The pulse tube was placed in an oven and heated to 50°C. Quarter-inch tubing connected the pulse tube inlet to a vacuum pump, either an Alcatel drag pump (to clean prior to G1, G3 and G4) or a Leybold turbomolecular pump (to clean prior to G5). Typically a vacuum bakeout of 5 to 7 days was performed using up to five purges with 7 atm. (100 psi) high purity helium gas sequenced through the bakeout. Cleaning was acceptable for all cases except for G4, for which a large amount of excess nitrogen was measured after the pulse tube test. Test G2 was conducted with the original gas loaded by TRW.

Mass Spectrometer

Pulse tube gas compositions were measured using a Stanford Research System CIS 200 closed-ion source quadrupole mass spectrometer. This device can measure compositions to a precision of approximately 1 ppm across a mass range of 1 to 200 atomic mass units at an inlet pressure of 1×10^{-3} torr. Accuracies are typically ± 1 ppm up to 20 ppm and $\pm 3\%$ of the reading above that. A gas inlet system was designed and built to interface the high pressure pulse tube gas to the mass spectrometer inlet pressure requirements. It consisted of an intermediate 7 liter vessel into which a small gas charge from the pulse tube could be loaded. This gas was then flowed through a capillary tube into the mass spectrometer inlet. Hence, the inlet system functioned in a batch mode to achieve the 10^{-10} torr pressure reduction. Two turbomolecular vacuum pumps were used to evacuate the system before a test and to provide the necessary differential pumping for the closed ion source operation.

Three different calibration gases were used to verify proper operation of the mass spectrometer and to adjust calibration coefficients for the various gas components. These gases are listed in Tables 1a through 1c along with independent measurements made by the analytical chemistry group at JPL for gases 1 and 3. The third gas was acquired to check the nitrogen sensitivity given the vendor's disagreement with gases 1 and 2. Calibration gas 3 confirmed the JPL measurements and therefore we concluded that the nitrogen concentrations reported by Matheson for gases 1 and 2 were slightly in error.

Hydrogen gas and water vapour posed significant experimental problems for these tests. The detector in the CIS 200 is calibrated with nitrogen gas and therefore does not accurately measure the hydrogen level. Stanford Research Systems indicated in a private communication that the hydrogen signal can be in error by an order of magnitude in this mode. Unfortunately, if the

Table 1a. Calibration Gas #1 (Matheson Gas Products).

	CO ₂	Ar	N ₂	H ₂ O	He
Matheson	110 ppm	108	115	108	balance
JPL chem	99	101	132	50	balance
Authors	102	98	131	90	balance

Table 1b. Calibration Gas #2 (Matheson Gas Products).

	CO ₂	Ar	N ₂	H ₂ O	He
Matheson	8.6 ppm	12	13	10	balance
JPL chem	n/a	n/a	n/a	n/a	n/a
Authors	10	11	22	10	balance

Table 1c. Calibration Gas #3 (Air Products and Chemicals).

	CO ₂	Ar	N ₂	H ₂ O	He
Air Products	n/a	n/a	108 ppm	n/a	balance
JPL chem	n/a	n/a	111	n/a	balance
Authors	2	0	106	20	balance

detector is recalibrated for hydrogen, then the error shifts to the other higher mass components. We decided to leave the calibration as it was and essentially ignore the hydrogen signal in our data analysis. Since the hydrogen will not condense in the pulse tube under our operating conditions, this seemed like a reasonable approach.

Water was a problem because of its ability to attach itself to the walls of the mass spectrometer system. Even though the inlet plumbing of the mass spectrometer was kept at 80°C for all tests, significant amounts of water were absorbed on the walls when a new gas sample was injected into the system, thereby under-measuring the true water content. Even the use of electropolished tubing could not solve this problem. The solution was to pre-load the plumbing with the sample gas in order to accelerate equilibration between the gas and the wall. Typically this was done by flowing the pre-loaded gas for 5 minutes at a pressure of approximately 0.5 to 1 torr in the ionization chamber. The validity of this technique was verified by accurately measuring the calibration gases listed in Table 1. Nevertheless, the measurement accuracy for water was approximately ± 2 ppm up to 20 ppm and $\pm 5\%$ of the reading above that, which is half as good as for the other gas species.

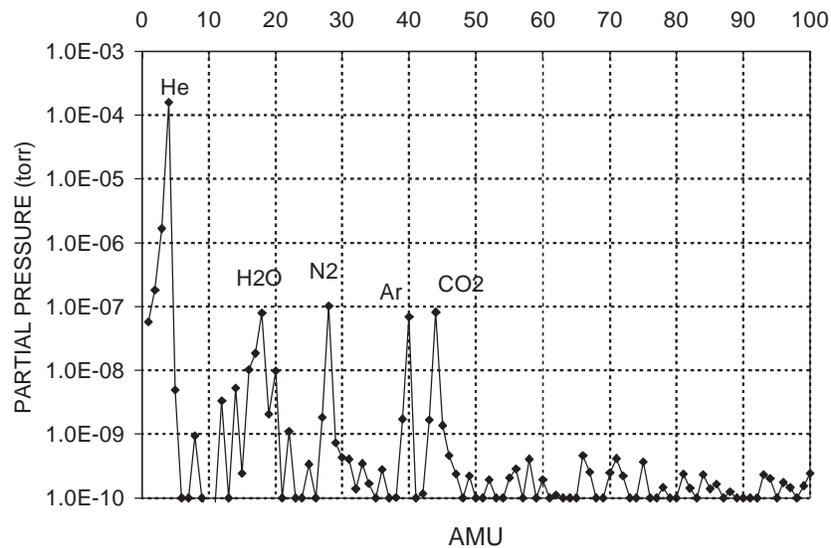
RESULTS AND DISCUSSION

Table 2 lists all five gases that were tested in this study. They have been ordered from highest (G1) to lowest (G5) in contaminant level. Compositions were measured with the mass spectrometer both before and after running the pulse tube. Note that the “at finish” values listed in the table are averages derived from multiple mass spectrometer measurements made as the gas was removed from the pulse tube. Averages are required because the contaminant species in the pulse tube develop spatial concentration gradients during condensation, gradients that persist for some time after the pulse tube is turned off. A typical mass spectrum is shown in Fig. 2 with the main peaks labeled. It corresponds to Calibration Gas 1, which is also test gas G1. The partial pressure values in this plot have not been adjusted for the relative ionization sensitivities of the different gases. When this is done, the true partial pressure of helium rises to roughly 1×10^{-3} torr while the other labelled species do not change appreciably. Therefore, a value of 1×10^{-9} torr corresponds to one part per million.

The time histories of all five pulse tube tests are shown in Fig. 3. The data are steady-state cold tip temperatures for a nominal 1-W applied heater load at constant piston stroke. All tests

Table 2. Summary Data of All Test Gas Compositions.

Gas	At Start				At Finish			
	CO ₂	Ar	N ₂	H ₂ O	CO ₂	Ar	N ₂	H ₂ O
G1	106 ppm	98	134	105	113	97	120	93
G2	3	2	50	70	9	2	54	110
G3	2	1	5	20	3	0	8	12
G4	2	1	17	2	5	1	70	8
G5	1	0	5	6	3	1	12	6

**Figure 2.** Mass Spectrum for Calibration Gas 1 and Test Gas G1.

show a temperature rise with time, which is indicative of a loss of performance due to contamination. The performance degradation is corroborated by a decrease in measured Carnot efficiency based on compressor power measurements. The temperature rise and Carnot efficiency data are presented in Table 3.

Generally speaking, the different tests all start at roughly the same temperature and rise by 0.6 to 2.8 K at the end of the testing period of 7 to 10 days. This test duration was chosen because an asymptotic temperature appears to have been achieved in all cases. The most contaminated gas, G1, starts with a slightly higher temperature than the other cases, presumably due to immediate

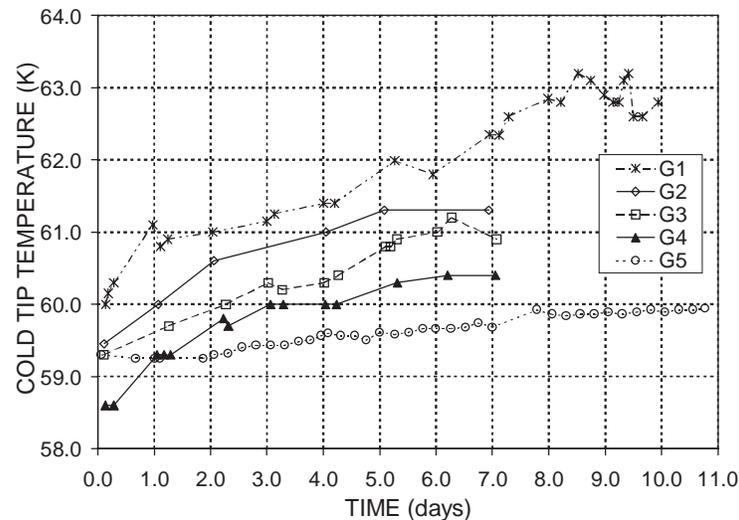
**Figure 3.** Time traces of all pulse tube test.

Table 3. Summary of Test Gas Refrigeration Performance.

Gas	CO ₂ +H ₂ O concentration		Cold tip temp. at 1 W load (K)			Carnot Efficiency	
	At Start	At Finish	At Start	After 1 week	ΔT	At Start	After 1 week
G1	211 ppm	206	60.0	62.3*	2.3*	6.27%	6.07%
G2	73	119	59.5	61.3	1.8	6.40%	6.24%
G3	22	15	59.3	60.9	1.6	6.37%	6.24%
G4	4	13	58.6	60.4	1.8	6.49%	6.32%
G5	7	9	59.3	59.7 ⁺	0.4 ⁺	6.32%	6.24%

*After 10 days, the cold temperature had risen to 62.8 K (+2.8 K).

⁺After 11 days, the cold temperature had risen to 59.9 K (+0.6 K).

condensation of large amounts of CO₂ and H₂O. The reason for the low starting temperature of gas G4 is not clear. It was not the cleanest gas tested; therefore, either the presence of extra nitrogen somehow yielded a slightly more efficient compression process, or the measurement reflected an instrumentation offset error.

The temperature increase data do, however, show a clear trend of greater rises for greater contamination levels. The clean gas test of G5 shows a distinctly different behaviour than the other cases, even though its level of CO₂ + H₂O contamination is only slightly greater than that of test G4. We therefore conclude that the presence of 70 ppm of nitrogen in test G4 was the cause of the difference, indicating that some nitrogen condensation occurred despite the fact that the 60 K is well above the freezing temperature of nitrogen at this concentration. Table 4 lists the freezing temperatures of the four contaminant gases at the 1, 10 and 100 ppm levels in the pulse tube. Based on this data, we would expect only CO₂ and H₂O condensate to be present in the pulse tube during the 1 W at 60 K operation. The reason why the frozen nitrogen does not evaporate when the temperature is raised to 60 K is unclear. Perhaps there is some unusual surface chemistry associated with a combined N₂, CO₂ and H₂O ice that retains N₂ even when the temperature rises above its freezing point. Alternatively, there could be a capillary condensation effect in the regenerator matrix that depresses the nitrogen vapor pressure. Or, perhaps the nitrogen is freezing in the region of the pulse tube downstream of the cold head where the temperature is lower than the nominal 60 K. Although nitrogen contamination is seen to be a factor, the similarity of results for tests G3 and G4, despite them having very different nitrogen concentrations, suggests that N₂ is not as strong a degrading influence as CO₂ and H₂O at a given concentration.

The source of gas for tests G3, G4 and G5 was commercially available “five-nines” pure helium. Concentration differences arose because of different cleanliness levels in the connecting plumbing and in the pulse tube itself. Even the cleanest gas in G5, however, did not avoid performance degradation due to contamination. This suggests that total impurity levels need to be lower than 10 ppm to completely prevent performance loss on a time scale 7 to 10 days. Note, however, that the performance decrease is relatively small. If the change in performance were due to an increase in parasitic heat load at 1 W nominal, rather than due to a decrease in second law efficiency, then the most contaminated gas (G1) equates to an increase of 40 mW after 10 days, while the least contaminated gas (G5) equates to an increase of only 12 mW. However, pulse tube warm-up tests (measuring the temperature rise of the cold head when the pulse tube was turned off) aimed at measuring this additional parasitic load did not detect any difference between the clean and contaminated states.

Table 4. Freezing Temperatures of Pulse Tube Gases at Different Concentrations.⁶

Gas	T _{fr} (at 1 ppm)	T _{fr} (at 10 ppm)	T _{fr} (at 100 ppm)
N ₂	38 K	42	47
Ar	44	49	56
CO ₂	115	125	140
H ₂ O	215	230	260

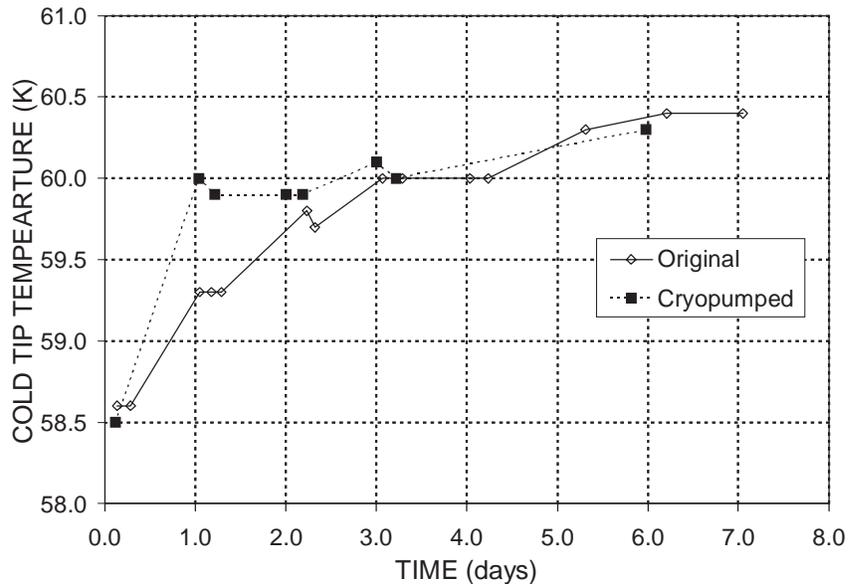


Figure 4. Time traces for cryopumped test of G4.

It is worth emphasizing that a contamination level of 100 ppm of water will form a sphere only 1.5 mm in diameter if all of it in the pulse tube were collected into a single frozen droplet. Note that in this pulse tube approximately 85% of the gas is on the backside of the compressor pistons, and that all of the water in this space must migrate through the piston-wall gap to form a droplet of this size. It seems somewhat surprising that such a small volume of water could produce the observed effects unless it were distributed across a thin (i.e. tens of microns) layer on a regenerator screen, for example. In such a scenario, significant blockage could occur despite the paucity of frozen material. However, our experimental set up did not allow us to measure the distribution of frozen material in the pulse tube, and therefore this issue remains unresolved.

The possibility that the observed performance degradation was due to external, rather than internal, contamination was explored in an additional test on gas G4. The hypothesis is that contaminant species could condense on the cold outside surface of the device and compromise the effectiveness of the multilayer insulation. This issue was addressed by running the pulse tube with gas G4 for another week after the initial test and after first warming it back up to room temperature to drive off any frozen gas that had collected on the outside. In addition, a Gifford-McMahon cryopump was operated in the vacuum chamber for the second test to drastically deplete the partial pressure of the chamber. Measured vacuum levels were down a factor of 50 to 2.6×10^{-8} torr. The result is shown in Fig. 4, where the second cryopumped curve is shown superimposed on the first nominal curve. Clearly the cryopump made no appreciable difference. The second curve rises more quickly to the asymptotic temperature, but this would be expected because the internal contaminants could not completely disperse throughout the pulse tube in the one day that it was inactive at room temperature.

Past studies^{4,5} have indicated that multilayer insulation performance starts to degrade when a thickness of roughly 0.1 microns of H_2O or CO_2 ice forms on it. In a vacuum system, such a thickness represents a vast amount of material, on the order of many hundreds of molecular monolayers. Simple first order analyses indicate that such large amounts of water are inconsistent with the observed vacuum chamber pressures of less than 1×10^{-5} and the capabilities of our pumping system. This, combined with the cryopump test results lead us to the conclusion that the observed performance degradation was due to internal, not external contamination.

For all gases except G2 it was observed that any brief shutdown and warm-up to room temperature of the pulse tube had the effect of restoring the pulse tube to its original performance. However, this original performance tended to be relatively short-lived, as seen in Fig. 4 for gas G4. We hypothesize that the condensed gases in the pulse tube diffuse away from their frozen location during the few hours of shutdown, but do not get very far away. Therefore, upon reacti-

vation of the pulse tube, it takes significantly less time for this gas to migrate to the cold, condensing part of the pulse tube. However, this behaviour was not observed for gas G4. This gas showed no improvement in performance after a two day shutdown, but did show almost complete recovery of the original performance after a six day shutdown. The reason for this difference in performance is not known, although it is interesting to note that this gas was the original gas loaded by TRW. As had been originally requested, they did not bake out the pulse tube prior to loading of this gas. Moreover, the gas sat in the pulse tube for almost two years before this study was conducted.

CONCLUSIONS

Five different gases were tested in a TRW/Lockheed pulse tube to determine the effects of contamination in otherwise pure helium. Total contamination levels ranged from approximately 20 to 420 ppm of water, carbon dioxide, nitrogen and argon. No other contaminants were present above a level of 1ppm except hydrogen, but due to instrumentation restrictions the hydrogen levels were not accurately measured. A decrease in pulse tube refrigeration performance was observed in all tests, with asymptotic steady-state temperatures reached on a time scale of 7 to 10 days. Under a 1-W applied load condition with a constant piston stroke, the total cold head temperature rise due to contamination ranged from 0.6 to 2.8 K depending on contamination level. Comparisons between cryopumped and non-cryopumped vacuum chambers appeared to rule out the possibility that the contamination was external to the pulse tube. Water and carbon dioxide were the most important contaminants at the nominal operating temperature of 60 K, although there was evidence for nitrogen condensation under some conditions. Warming of the pulse tube to room temperature tended to restore the original performance, although this improvement degraded more rapidly the second time around and returned to the asymptotic steady-state condition.

ACKNOWLEDGMENTS

The work described in this paper was carried out by the Jet Propulsion Laboratory, California Institute of Technology, and was sponsored by the NASA EOS IMAS TechDemo Project through an agreement with the National Aeronautics and Space Administration. The authors would like to acknowledge the laboratory assistance of Scott Leland and Gary Plett, and technical discussions with Dr. Simon Collins, all at JPL.

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