Proton Conductive Membrane Compressor-Driven Pulse Tube Cryocooler

James R. Muller[1], Lonnie G. Johnson[1] , Carl S. Kirkconnell[2] and Robert Hon[2]

[1]Johnson Research and Development
 Atlanta, Georgia, USA 30312

[2]Raytheon Space and Airborne Systems
 El Segundo, California, USA, 90245

ABSTRACT

Johnson Research & Development (JRD) and Raytheon are collaboratively developing pulse tube and Joule Thomson (JT) cryocoolers based on a novel solid-state proton conductive membrane compressor (PCMC). By eliminating all moving parts from the system, this approach, if successful, would improve upon the current state-of-the-art in which positive-displacement compressors using linear motors are used as the pressure wave generator. Cryocooler system performance predictions based upon recent proton conductive membrane (PCM) experimental measurements reveal that the solid state compressor is a viable technology for cryocoolers and can potentially be competitive with existing compressor technology. Experimental results show that proton conducting membranes can be used to pump hydrogen using either AC or DC voltages for use in cryocooler systems. If the PCMC can be made competitive efficiency-wise with existing compressor technology, it may offer significant additional advantages including: (1) all solid state system, (2) extremely low vibration, (3) flexible form factor, (4) low cost electronics and control systems, (5) high reliability, (6) scalable designs, and (7) potential elimination of cryocooler redundancy requirement for space-based systems.

INTRODUCTION

Cryocoolers play a vital role in cutting edge space science with applications such as thermal management of infrared (IR) focal plane assemblies (FPA) and sometimes the associated optics. The cryocooler is generally considered one of the highest risk components in a space IR sensor, so reliability enhancements at the cryocooler component level are well-leveraged risk reduction measures at the sensor level. The stressing lifetime and reliability requirements typical of space surveillance missions result in tight tolerances, labor-intensive assembly procedures, and costly materials. The vibration output requirement also contributes to the design complexity of the traditional cryocooler and necessitates expensive control electronics that mitigate the vibration output through closed-loop control of the input current waveform. These characteristics are typical of the baseline linear (Stirling and pulse tube) cryocooler technology presently envisioned. Typically, these linear cooler systems are expensive and may necessitate the inclusion of a redundant cryocooler to achieve the required system-level reliability requirements. The inherent reliability of our solid-state ap-

proach could eliminate the need for a redundant cryocooler, which provides obvious cost savings and reduces the parasitic "off-cooler" load that the operational cryocooler must otherwise carry. Based upon our efforts and analysis to date, we believe we can meet and potentially exceed current space cryocooler performance and, in addition, provide the following:

- Lower manufacturing cost;
- Longer lifetime;
- Higher reliability (no moving parts);
- Scaleable from small MEMs applications to larger applications;
- Permit high heat rejection temperature (up to 373 K) without significant performance degradation.

The key component in enabling the use of the solid state Proton Conductive Membrane Compressor (PCMC) is a membrane electrode assembly (MEA). MEAs consist of a proton conductive membrane (PCM) sandwiched between two electrodes and can be connected together in series or in parallel as in standard fuel cell systems to form stacks. The mass flow rate through a single MEA cell is directly proportional to the current flowing through the cell. Applying a voltage across the MEA "pumps" hydrogen gas (the working fluid for the solid state cryocooler systems) from a low to high pressure in accordance with the Nernst equation. The PCMC is viable with either a DC (JT systems) or AC (PT systems) voltage profile.

PROTON CONDUCTIVE MEMBRANE COMPRESSOR (PCMC)

The cryocooler systems proposed are solid-state devices that utilize a membrane electrode assembly (MEA), similar to those in fuel cells, comprised of a proton conducting membrane (PCM) sandwiched between two electrodes. Figure 1(a) shows a schematic of an MEA separated by two fixed volume chambers, each containing hydrogen gas. If a voltage is applied across the MEA, hydrogen gas is oxidized on the low-pressure side into two (2) protons and two (2) electrons. The PCM will only allow positively charged particles to pass through it, so the protons are transported to the high-pressure side while the electrons are routed through the external circuit. At the high-pressure side electrodes, the protons are reduced with the electrons to reform hydrogen gas, i.e. hydrogen is transported from low pressure to high pressure. This capability has been demonstrated both at JRD using Nafion® based MEAs and in published literature using ceramic proton conductors.[1,2] The maximum theoretical pressure ratio, P_{Hi}/P_{Low}, is the open circuit Nernst voltage as given by

$$V_{OC} = \frac{\bar{R}T}{2F} \ln\left(P_{Hi} \Big/ P_{Low} \right) \tag{1}$$

where V_{OC} is the open circuit voltage, $\bar{R}$ is the universal gas constant, T is the cell temperature, F is Faraday's constant, P_{Hi} is the pressure on the high pressure side, and P_{Low} is the pressure on the low pressure side. Figure 1(b) shows the Nernst voltage plotted as a function of temperature at various pressure ratios. (Note that the plot is of pressure ratios and not absolute pressure.) The expected
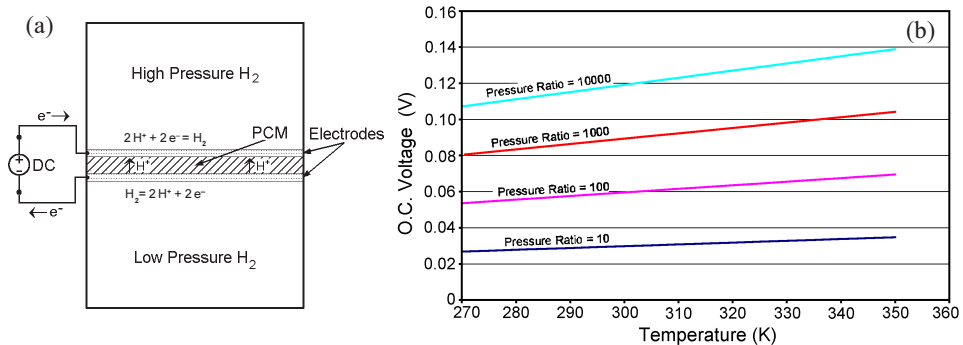

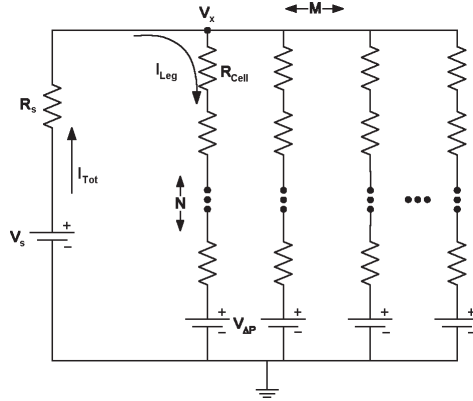**Figure 1.** (a) PCM pump schematic; (b) Nernst voltage plot

Figure 2. Electrical representation of PCMC.

operating temperature of the PCMC is 300 K, so as can be seen in Figure 1(b), in order to achieve a pressure ratio of 10, a voltage of at least 0.03 V must be applied across a single MEA.

For a single MEA the mass flow rate is directly proportional to the current flowing through it according to

$$\dot{m}_{cell} = I/F \qquad (2)$$

where $\dot{m}_{cell}$ is the mass flow rate through a single MEA, I is the current through the MEA, and F is Faraday's constant. As an example, 1A of current is equivalent to a mass flow rate of approximately 1.04e-5g/s of hydrogen gas flowing through a single cell. Note that cells can be stacked in series to increase the mass flow rate, e.g., for 1000 cells in series, the current is still 1A, but the total mass flow rate is now increased to 1.04e-2g/s of hydrogen gas. Additionally, the entire assembly for a single MEA is anticipated to be less than a millimeter in thickness, so the compression process is expected to occur isothermally since such a thin structure cannot support a significant temperature gradient unless adequate heat is transferred from the membrane through its substrate.

PCM COMPRESSOR MODELING

The proton conductive membrane compressor (PCMC) is at the heart of the solid-state cryocooler technology and the focus of this effort. An electrical representation of the PCMC is shown in Figure 2. In this diagram, the applied voltage is represented by the source V_S and the source resistance is shown as R_S. The PCMC is shown as a combination of series and parallel resistances, where a resistor represents each MEA in the stack. For this diagram, there are M legs consisting of N MEAs. Within each leg there is an additional voltage source that represents the sum of the back EMF from each MEA in the leg. This back EMF is produced by the pressure ratio across the cell from the pumping action and is equal to the Nernst voltage (see Figure 1).

Based on this representation, we can derive computational equations that are implemented in the numerical models. The cell resistance, R_{cell}, is calculated by fixing the PCM geometry (i.e., the area and thickness) and operating temperature according to

$$\dot{m}_{cell} = I/F \qquad (3)$$

where t is the cell thickness, A is the active area of the PCM, and σ is the PCM conductivity, which is a function of temperature. The resistance in a single leg is then calculated from

$$R_{leg} = NR_{cell} \qquad (4)$$

In addition to the electrical losses in each cell, the model includes a formulation to predict the gas diffusion losses through the thin membranes. In the PCMC, the gas diffusion losses are manifested as hydrogen gas leaking from the high-pressure side back to the low-pressure side, resulting

in higher power to achieve the required cooling rate. Mass diffusion through a solid medium is defined as

$$\dot{m}_{Diff} = \frac{\Delta P \ D_{AB} \ A_{Cell}}{t_{cell} T_{cell} \overline{R}} \tag{5}$$

where ΔP is the pressure differential across the cell, D_{AB} is the diffusion coefficient, A_{Cell} is the cell area, t_{cell} is the membrane thickness, T_{cell} is the cell temperature, and $\overline{R}$ is the ideal gas constant. The total mass flow rate through a cell to achieve a required mass flow rate from the pump is then defined as

$$\dot{m}_{Tot,cell} = \dot{m}_{req} + \dot{m}_{Diff} \tag{6}$$

The total current for the cell is then calculated as

$$I_{Tot} = \dot{m}_{Tot,cell} \ F \tag{7}$$

The function of the PCMC in a pulse tube cryocooler is distinctly different than the DC pumping function in the Joule Thomson Cycle. The pulse tube utilizes an AC pressure oscillation to produce the refrigeration. The power calculation becomes significantly more complex as a result. To perform an optimization with the existing numerical model, a closed form solution for the input power is necessary and can be derived from the expressions previously presented for the source voltage, V_s, and the total source current, I_{Tot}. The input power is then the product of the source voltage and current. The source voltage is found by writing a voltage loop equation:

$$V_s = I_{Tot} \left(R_s + \frac{R_{Leg}}{M} \right) + V_{\Delta P} \tag{8}$$

where additional relations were used to simplify the expression. For AC drive, such as that used for a pulse tube cryocooler, the pressure ratio will be

$$P_R = \frac{\overline{P} + \widetilde{P} \sin(wt - \varphi)}{\overline{P}} \tag{9}$$

Sinusoidal pressure oscillation from the drive side of the PCMC and DC pressure at the plenum side are approximations. Large pressure fluctuations typically begin to deviate from this approximation due to the asymptotic nature of the compression and expansion process. Additionally, the mass flow rate will be approximately a sine wave at zero reference phase:

$$\dot{m} = \widetilde{m} \cdot \sin(wt) \tag{10}$$

Using these representations, the source current and voltage become

$$I_{Tot} = \frac{\widetilde{m} \cdot \sin(wt) \cdot F}{N} \tag{11}$$

and

$$V_{\Delta P} = \frac{N \cdot \overline{R} \cdot T}{2F} Ln \left(\frac{\overline{P} + \widetilde{P} \sin(wt - \varphi)}{\overline{P}} \right) \tag{12}$$

The instantaneous input power can now be calculated as

$$P_s = I_{Tot}^2 \left(R_s + \frac{R_{Leg}}{M} \right) + I_{Tot} V_{\Delta P} \tag{13}$$

Pulse Tube Parametrics

A study was conducted to generate an optimized system-level design that balances PCMC efficiency with the efficiency of the thermodynamic systems, such that the single-stage system can provide 1.6 W of heat lift at a cold tip of 85 K for a minimum of input power. The goal for the two-stage system has been production of 1.0 W of heat lift at 85 K with a simultaneous heat lift of 0.5 W at 35 K. A safety factor of 2 was used throughout this work, meaning that the single stage thermodynamic model was actually designed to provide 3.2 W of heat lift at 85 K instead of the 1.6 W goal. Similarly, the two-stage model intention was to produce 2.0 W at 85 K and 1.0 W at 35 K.

Single-Stage Modeling

The model used 5000 cells at a thickness of 1 μm and a cell area of 25 cm². This specific model made use of a stepped inertance tube at the warm end of the pulse tube, connecting back to the PCMC housing volume which served as a type of surge volume. This volume had been included in the optimization runs as a variable, and had settled to a value that was similar in size to the compression volume.

The SAGE modeling code was used to directly calculate the PV power in both the compression and housing volumes; these values were summed to represent the total PV power output of the PCMC. The formula for PCMC I²R power dissipation is still accurate, and the total PCMC input power is calculated as the sum of the derived I²R power and the PV power. In the optimizations that followed, the goal of the optimization was generally to minimize this total input power value.

The surge volume and housing volume were separated into different, thermodynamically isolated model components. This largely eliminated the long model run times and allowed optimizations to be run in reasonable amounts of time.

Single-Stage Optimization

The initial optimization runs were aimed at investigating the following system changes:

• Use of a dual-density regenerator matrix
• Elimination of the stepped inertance tube in favor of a single-diameter inertance tube
• Low-frequency (≤10 Hz) operation

The use of a regenerator consisting of two different screen types (differing porosities and wire diameters) was found to increase system performance and was therefore carried through the rest of the optimization runs. The system model consistently optimized to low frequencies, eventually settling out in the 2-3 Hz region. This had the secondary effect of driving the regenerator and pulse tube dimensions to larger values. Elimination of the stepped inertance tube was found to have a negligible effect on system performance, and was therefore carried through the rest of the optimization runs as a way of reducing model complexity.

The second phase of model runs consisted of straightforward optimizations with the goal of providing appropriate heat lift at 85 K while minimizing PCMC input power. The system settled out to a specific power of ~68 W/W, equal to the total PCMC input power divided by the heat lift at 85 K. The compressor efficiency in this case, equal to the PCMC PV power output divided by the total PCMC input power, was ~43%. The PV specific power, equal to the PCMC PV power output divided by the heat lift at 85 K, was ~29 W/W. The compressor efficiency of 43% is an indication that the PCMC is underperforming significantly in comparison to traditional linear motor compressors (65% typical efficiency). Likewise, the PV specific power, a measure of the thermodynamic efficiency of the pulse tube system, was higher than would be expected for an 85 K heat lift temperature.

It was believed that the goal of minimizing total input power was forcing the thermodynamic system to a very inefficient configuration. In an effort to determine whether this was the case, the model was re-optimized with the goal of minimizing PCMC PV power output instead of total PCMC power input; this effectively removes the PCMC inefficiencies from the optimization. The optimization was successful in that the new PV specific power value was ~16 W/W, almost a factor of two better than the previous case. This confirmed that the PCMC inefficiency was hindering the thermodynamic optimization, and indicates that PCMC efficiency will have to improve significantly if this type of system is to be competitive on a power basis with traditional cryocoolers.

The tendency of the model to run at very low frequencies is a departure from past pulse tube design experience. The likely reason for this effect is that the mass flow rate and PCMC drive current are directly related; by moving to lower frequencies, the mass flow rate is reduced, the PCMC current is reduced, and the I²R power loss is reduced. A third set of optimizations was therefore run with a constraint that the operational frequency be greater than or equal to 30 Hz. As before, the optimization was first run with a goal of minimizing total input power; both the normal specific power as well as the PV specific power suffered significantly. The optimization was then run with the goal of minimizing PCMC PV power output, with very interesting results. The PV

Table 1. Parametric Analysis

PARAMETER	DESCRIPTION	RANGE
Number of cells in series in each leg	This parameter determines the number of cells connected in series in each leg of the PCMC.	50, 100, **500**, 1000
PT load temperature	This is the load temperature for the pulse tube stage interface	75, 80, **85**, 90, 95 [K]
Membrane thickness	This is the thickness of the proton conduction layer of the membrane electrode assembly	0.1, **1**, 10 [μm]
Throttle ratio	This is the pressure ratio across the throttle at the low temperature end.	7, **10**, 15, 25
Source HX efficiency	This is the target efficiency for the source heat exchanger	0.80, 0.85, **0.90**, 0.95

specific power was reduced to ~15.5 W/W, the most efficient result so far. However, the amount of I^2R loss in the PCMC increased greatly, leading to an overall specific power of ~630 W/W (PCMC efficiency of ~2%). This is a clear indication that the PCMC's lack of efficiency at higher frequencies is a hindrance to optimization of the overall system.

JT Parametrics

The baseline system specifications for the JT study are given in Table 1. The parametric analysis for the hybrid system was run using the parameters listed in Table 1 (units are shown where necessary). Bold values are the baseline case.

The data shown in Figures 3 through 5 are for a proprietary proton conducting material, PCM1. Figure 3 shows the relationship between the cell thickness and the required input power to the JT compressor. Recall that internal electrical resistance is directly proportional to the thickness while the diffusion loss is indirectly proportional to the thickness. From this chart it can be seen that for very thin membranes, the diffusion is dominant and drives the power higher. Also, the input power is getting larger as the thickness moves to 10 μm, showing that electrical resistance is becoming the dominant loss. We are targeting membranes in the 1-5 μm range for our system, which should be in the optimal range. Figure 4 shows the relationship between the 2nd stage temperature and the compressor power. The data show that the lower the second stage temperature, the lower the input power to the JT compressor. Note that this is deferring some of the cooling load from the JT to the PT stage. Figure 5 shows the relationship between the number of cells in series to the required compressor power. This chart shows that increasing the number of cells decreases the power input. Mass flow is parallel through the cells so that an increase in cells at the same current increases the total mass flow rate; however even though the power input is lower, the specific power (W/cm³) may not improve with more cells. As the manufacturing design progresses, these results will be re-examined to determine the specific power.
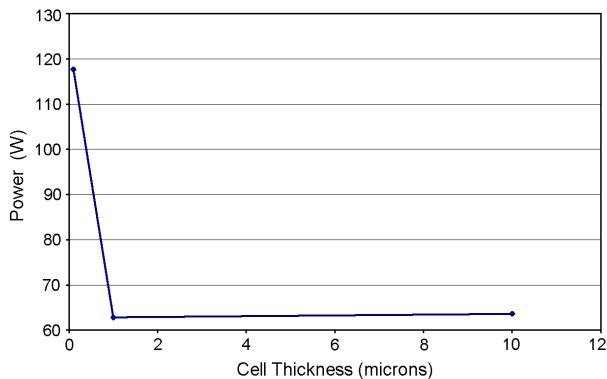
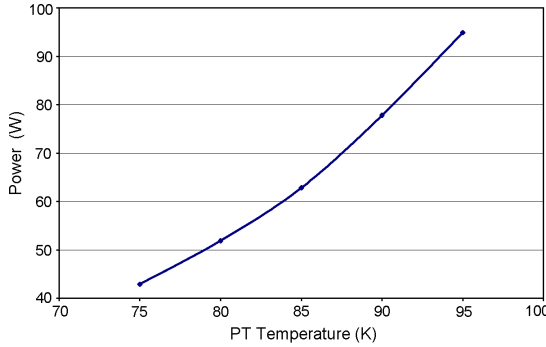**Figure 3.**   JT compressor power versus cell thickness

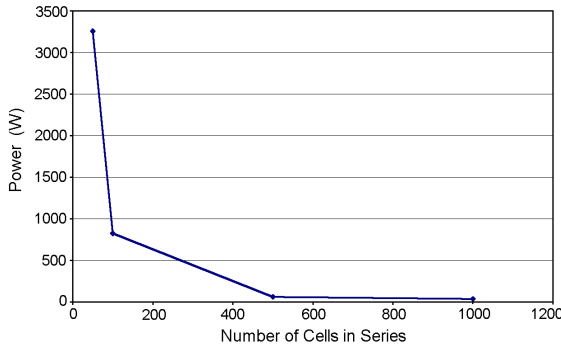**Figure 4.** JT Compressor power versus 2nd stage temperature

Figure 5. JT compressor power versus number of cells in series

PCM COMPRESSOR TESTING

Proton conductive membranes (PCMs) were tested for hydrogen pumping capability in both DC mode (Joule Thomson) and AC mode (pulse tube operation). Summary results are presented in the following sections.

DC Pumping

Several DC pumping tests were conducted on a single MEA to confirm the use of MEAs for solid state compressors. An initial dead-headed pumping test was performed to test the system. In this test, the two sides of the MEA were sealed and 1V signal was applied across the MEA. The measured output current for the run was approximately 900 mA, which is low considering the membranes can operate up to 1-2 A/cm². Figure 6 shows the absolute pressure of each side of the test rig during this initial pumping test.
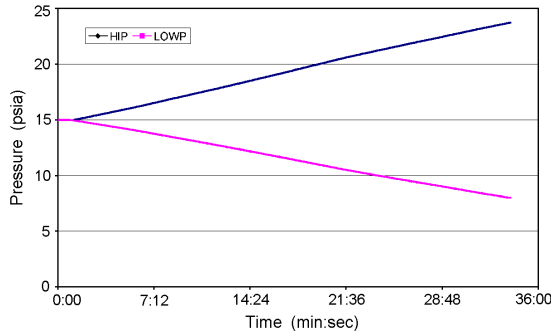
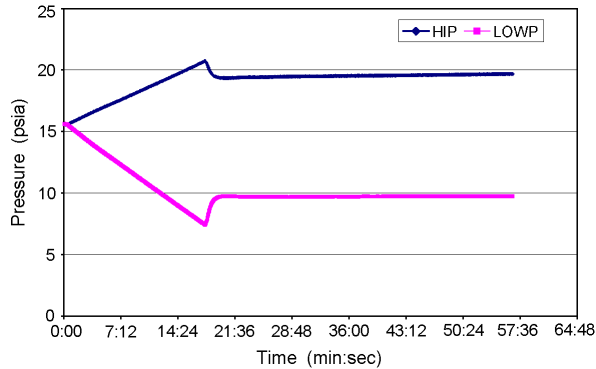**Figure 6.** Absolute pressure versus time for initial pumping test

Figure 7. Transient DC pumping pressure plot for PCM

In this case, the PCM was used to pump around a loop where the initial pressure was set to 15 psia [103 kPa] and a mass flow controller was set to loop on a downstream pressure of 10 psia [69 kPa]. The pressure plot for this run is shown in Figure 7. Notice that the pressures diverge until the low side pressure moves below 10 psia [69 kPa] at which point the mass flow controller begins to regulate mass flow to maintain the set point.

AC Pumping

Pumping capability of the membranes under AC waveforms was studied using the same test rig. The testing was done for both square and sinusoidal waveforms over the frequency range 0.1-5 Hz. For all tests the applied voltage was kept between +/-2 V to ensure that the potential break-down voltage of the material was not exceeded.

Figure 8 shows the measured pressure wave for the PCM using a sinusoidal voltage. Recall that current is proportional to mass flow rate, and for this case, a negative current implies mass flow from the high pressure side to the low pressure side. The pressure rise and drop is within 20% of the value calculated using the ideal gas law for a closed system where a change in mass occurs. This difference can be attributed to inaccuracy in the volume measurement and in sensor calibration.

The same test was run at 0.1 Hz, except a square wave was applied to the MEA. Figure 9 shows the measured pressure wave for the PCM using the square voltage wave. Notice that the pressure rise and fall is larger than with the sine wave, which is due to a larger mass flow over the period. Once again the pressure rise and drop is within 20% of the value calculated using the ideal gas law for a closed system where a change in mass occurs.

In order to show that the membranes are capable of pumping at higher frequency, the conductivity of the PCM was measured from 1 Hz to 1 MHz using a Zahner IM6e impedance spectroscopy
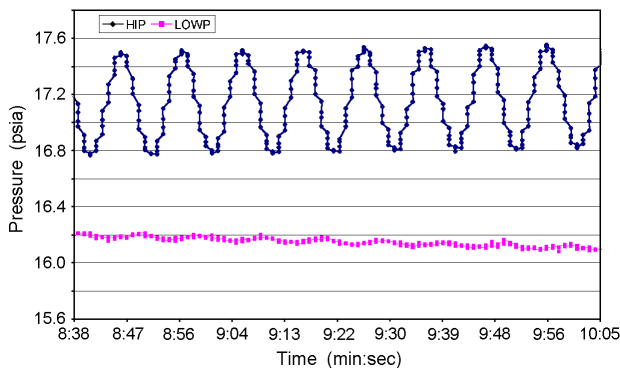

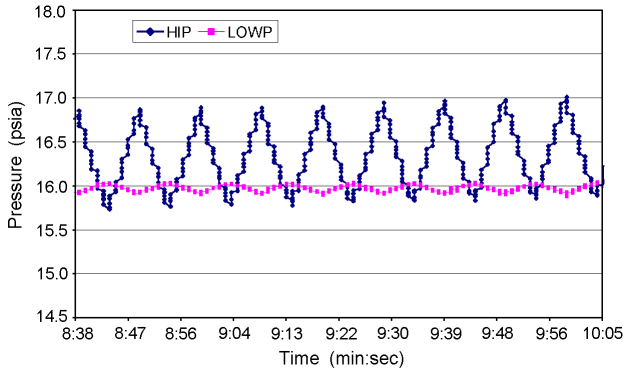**Figure 8.** Pressure wave for PCM using 0.1 Hz AC sin wave

Figure 9. Pressure wave for PCM using 0.1 Hz AC square wave
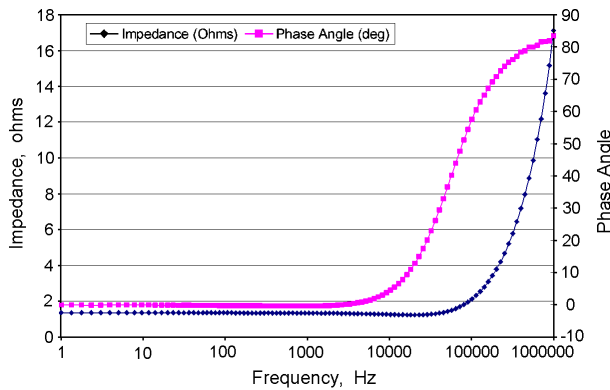

Figure 10. Impedance and phase angle plot versus frequency for PCM

unit. The test was run in pure hydrogen in the test at approximately 25 psia [172 kPa] and the results are shown in Figure 10. As can be seen from the plot, there is no increase in resistance through 1 kHz, which implies that electrochemically there is nothing preventing operation at higher frequencies for pumping hydrogen. Based on this data, it is clear that dead volume in a stack design can potentially degrade performance of the compressor.

CONCLUSIONS

A proton conduction membrane compressor (PCMC) was investigated and the results show that the solid state compressor is a viable technology for cryocoolers and can potentially be competitive with existing compressor technology. Experimental results show that proton conducting membranes can be used to pump hydrogen using either AC or DC voltages for use in cryocooler systems. Numerical simulations for a pulse tube system using the solid state compressor show the system operates at much lower frequency than traditional cryocoolers. Impedance measurements for a proton conductive material through frequencies over 1 kHz confirm that there is no change in resistance for the material, and at this time it is believed that low frequency operation is a result of the coupling of the mass flow rate and the compressor current and therefore the IR losses (lower current results in lower IR losses). Further testing will be necessary to confirm the SAGE computer model prediction of optimal system performance at low frequency. DC operation in a Joule-Thomson/Pulse tube system can result in a competitive system compared to existing technologies.

ACKNOWLEDGEMENTS

REFERENCES

1. Schober, T., Meuffels, P., "High Temperature Proton Conductors for the Injection of Hydrogen into a Vacuum," *J. Am. Ceram. Society*, Vol. 84 (2001), pp. 1996-2000.

2. Schober, T., Meuffels, P., "Electrically operated hydrogen and oxygen vacuum leaks using ceramic high temperature ionic conductors," *J. Vac. Sci. Technology A*, Vol. 19 (2001), pp. 958-962.