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Performance Testing of a Prototype Pd-Ag Diffuser

G. A. Morgan

B. J. Hodge

August 31, 2016

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EXECUTIVE SUMMARY

The fusion fuel cycle has gained significant attention over the last decade as interest in fusion programs has increased. One of the critical components of the fusion process is the tritium fuel cycle. The tritium fuel cycle is designed to supply and recycle process tritium at a specific throughput rate. One of the most important processes within the tritium fuel cycle is the clean-up of the of the process tritium. This step will initially separate the hydrogen isotopes (H_2, D_2, and T_2) from the rest of the process gas using Pd-Ag diffusers or permeators. The Pd-Ag diffuser is an integral component for any tritium purification system; whether part of the United States' defense mission or fusion programs.

Domestic manufacturers of Pd-Ag diffusers are extremely limited and only a few manufacturers exist. Johnson-Matthey (JM) Pd-Ag diffusers (permeators) have previously been evaluated for the separation of hydrogen isotopes from non-hydrogen gas species in the process. JM is no longer manufacturing Pd-Ag diffusers and a replacement vendor needs to be identified to support future needs. A prototype Pd-Ag diffuser has been manufactured by Power and Energy, and is considered a potential replacement for the JM diffuser for tritium service.

New diffuser designs for a tritium facility for any fusion energy applications must be characterized by evaluating their operating envelope prior to installation in a tritium processing facility. The prototype Pd-Ag diffuser was characterized to determine the overall performance as a function of the permeation of hydrogen through the membrane. The tests described in this report consider the effects of feed gas compositions, feed flow rates, pump configuration and internal tube pressure on the permeation of H_2 through the Pd-Ag tubes. Three gas compositions (2% H_2, 50% H_2, and 96% H_2 with the balance N_2) were tested at flow rates ranging from 100 sccm to 3000 sccm. In addition, two different internal tube pressures (700 and 1140 Torr) and two different pump configurations (Normetex and Edwards scroll pumps) on the permeate side of the diffuser were evaluated.

For the 96% H_2/4% N_2 mixtures, nearly all of the H_2 permeated through the Pd-Ag membrane at flow rates up to 3000 sccm. However, results for the 50% H_2/50% N_2 gas composition show that 100% permeation is only achieved up to a flow rate of 1000 sccm and there is hydrogen remaining in the bleed stream at flow rates above 1000 sccm. There was also a significant reduction in the hydrogen permeation for the 2% H_2/98% N_2 cases. This reduction in the hydrogen permeating through the membrane can most likely be attributed to a blanketing effect by the nitrogen The pressure drop across the Pd-Ag diffuser assembly is < 10 Torr for all gas compositions and internal tube pressures up to a flow rate of 1000 sccm. The small pressure drop across the tubes indicates that several diffusers of this design can be operated in series without the need for additional equipment. The results presented here indicate that this prototype Pd-Ag design is not acceptable for low to mid hydrogen gas compositions even if several diffusers are utilized in series. This particular Pd-Ag diffuser design performs exceptionally well for gas streams containing a high initial hydrogen concentration. As the amount of hydrogen in the initial feed stream decreases, the overall performance of the Pd-Ag diffuser decreases as well.

This Pd-Ag diffuser design is not suitable for a tritium purification system within the fusion energy fuel cycle. Typical tritium purification systems can be expected to see a range of

hydrogen isotope concentrations and this particular prototype diffuser is only suitable for process streams containing high concentrations of hydrogen isotopes.

Significant efforts should be undertaken to identify additional commercial vendors for Pd-Ag diffusers. It is of critical importance to identify, procure, and test different Pd-Ag designs that can perform well over a range of hydrogen isotope concentrations for tritium gas processing applications.

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LIST OF ABBREVIATIONS

2TS	20 Standard Liter Per Minute Test System
DAQ	Data Acquisition System
JM	Johnson-Matthey
MFM	Mass Flow Meter
MFC	Mass Flow Controller
MKS	MKS Instruments, INC.
M&TE	Measurement and Test Equipment
PD	Pressure Display
Pd-Ag	Palladium Silver
PT	Pressure Transducer
RGA	Residual Gas Analysis
sccm	Standard Cubic Centimeters per Minute
SLPM	Standard Liters per Minute
SRNL	Savannah River National Laboratory
SRS	Savannah River Site
TC	Thermocouple
VP	Vacuum Pump
μGC	Micro Gas Chromatograph
UPH	Ultra-High Purity Hydrogen

1.0 Introduction

The fusion fuel cycle has gained significant attention over the last decade as interest in fusion programs has increased. One of the critical components of the fusion process is the tritium fuel cycle. The tritium fuel cycle is designed to supply and recycle process tritium at a specific throughput rate. One of the most important processes within the tritium fuel cycle is the clean-up of the of the process tritium. This step will initially separate the hydrogen isotopes (H_2, D_2, and T_2) from the rest of the process gas using Pd-Ag diffusers or permeators. The Pd-Ag diffuser is an integral component for any tritium purification system; whether part of the United States' defense mission or various fusion programs, such as SHINE and ITER. Domestic manufacturers of Pd-Ag diffusers are extremely limited and only a few manufacturers exist.

Johnson-Matthey (JM) Pd-Ag diffusers (permeators) have previously been evaluated for the separation of hydrogen isotopes from non-hydrogen gas species in the process [1]. Pd-Ag diffusers have been used for many years at SRS with great success [2] [3]. JM is no longer manufacturing Pd-Ag diffusers and a replacement vendor needs to be identified to support future needs. This not only an issue for the SRS tritium facilities, but also for those facilities that are using tritium to support the fusion fuel cycle. A prototype Pd-Ag diffuser has been manufactured by Power and Energy, and is considered a potential replacement for the JM diffuser for tritium service.

Many of the companies that are currently in the hydrogen separation business are actually in the hydrogen purification business. A hydrogen purifier is designed to purify an impure hydrogen gas and is not typically designed for tritium service. In the design of a hydrogen purifier, a customer desires a certain flow rate of ultra-high purity hydrogen (UPH, 99.99999% or greater). The purifier is then designed to deliver a certain flow rate of UPH. Hydrogen purifiers are not typically designed to remove all of the hydrogen isotopes from the feed streams and therefore, a percentage of the hydrogen isotopes remain in the bleed stream. When the hydrogen diffusers are used for tritium service, it is desirable to remove as much of the tritium as possible to limit the need for additional reprocessing. The near-complete removal of the tritium ultimately reduces emissions from the facility.

Over the last several years, SRNL has been heavily involved in the identification of potential vendors for the manufacture of tritium compatible Pd-Ag diffusers. One of the largest challenges to overcome is the limited number of domestic vendors. Pd-Ag diffusers for tritium service are typically custom components based on the desired flow rates and pressures. Therefore, the components have a relatively long lead time compared to off-the-shelf components.

A prototype Pd-Ag diffuser was recently purchased from Power and Energy. The prototype Pd-Ag diffuser assembly is shown in Figure 1.1. The feed and bleed ports are located on the same side of the diffuser. The permeate stream exits from the opposite side of the diffuser. The feed stream typically contains a mixture of non-hydrogen gases, hydrogen isotopes, and various process impurities such as methane, carbon monoxide, carbon dioxide, and ammonia. The bleed or raffinate stream ideally does not contain any hydrogen isotopes and only consists of the non-hydrogen gas(es) and impurity species. Ideally, the permeate stream should be composed of 100% hydrogen isotopes.

The Pd-Ag diffuser was characterized to determine the overall performance of the diffuser as a function of the permeation of hydrogen through the membrane. The tests described in this report consider the effects of various feed gas compositions, feed flow rates, internal tube pressures, and pump configurations on the permeation of H_2 through the Pd-Ag tubes. The overall trends reported here may be extended to other configurations for hydrogen permeation through Pd-Ag diffusers although these results are specific to this particular diffuser (flow rates, configuration, etc.).

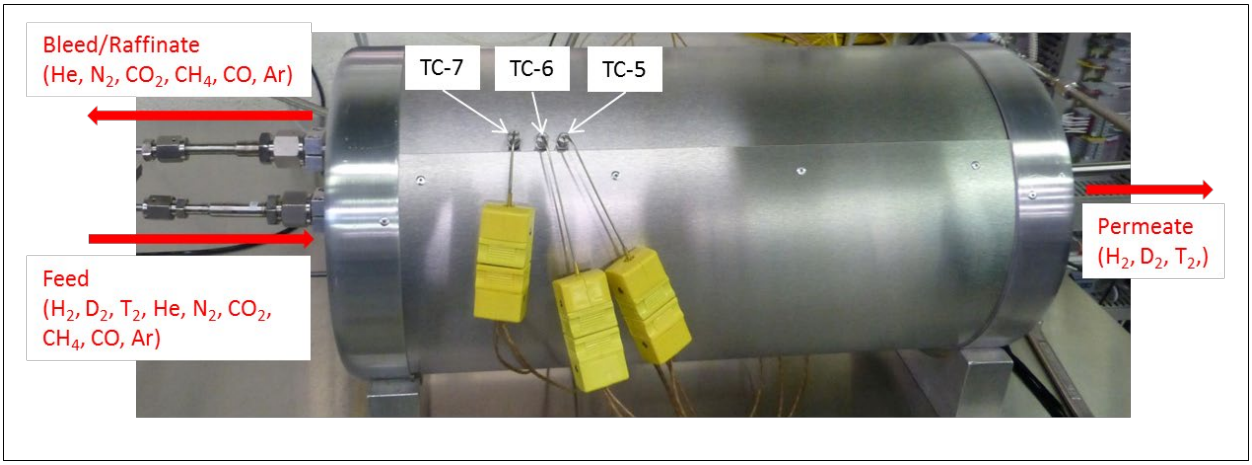

Figure 1.1: Prototype Diffuser Assembly

2.0 Experimental

2.1 Prototype Pd-Ag Diffuser Design Features

The prototype Pd-Ag diffuser manufactured by Power and Energy uses Pd-Ag micro channel membranes. This technology was developed and patented by Power and Energy [3]. This micro-channel membrane design, as shown in Figure 2.1, forces the feed gas to pass within 300 μm of the hydrogen permeable membrane. This results in a more efficient removal of the hydrogen isotopes from the process gas stream and also ensures a uniform processing of the feed gas. The micro-channel membranes have the benefit of large surface areas in a compact footprint, thereby enabling operating at standard temperatures with minimal thermal loss. [3]

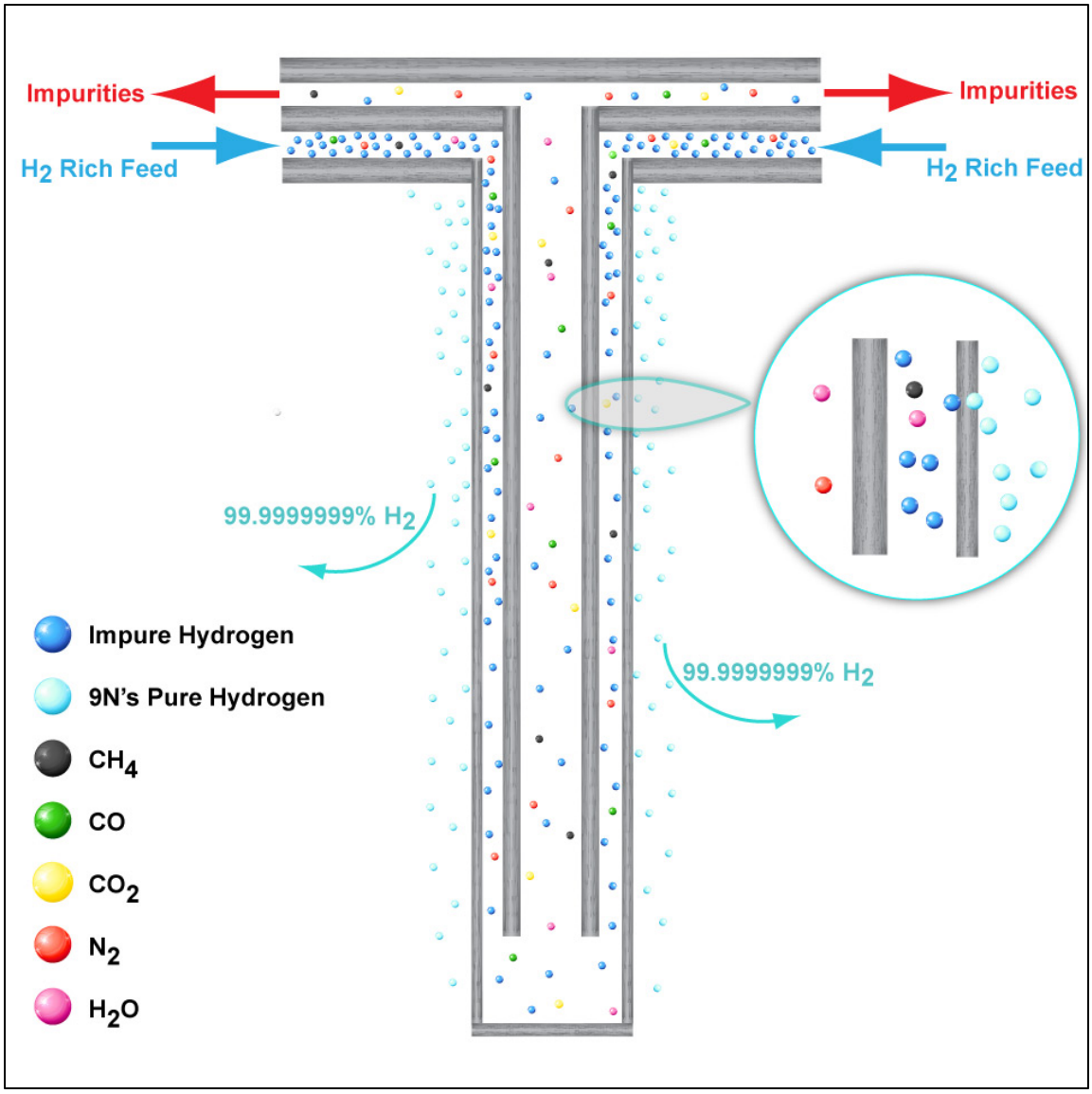

Figure 2.1: Schematic of Pd-Ag Micro-Channel Membranes [3]

The feed gas flows the length of the diffuser in a stainless steel tube to preheat the gas prior to entering the Pd-Ag membranes. The gas enters a plenum which distributes the flow into the Pd-Ag tubes, sometimes called Pd-Ag membranes. At normal operating temperatures, the H_2 in the feed gas permeates through the wall of the Pd-Ag tubing into the lower pressure cavity of the diffuser shell. This H_2 gas is referred to as the "permeate" or pure stream. The non-hydrogen components of the feed stream continue through the tubes and discharge into a common plenum. The gas remaining in the tube after permeation is referred to as the "raffinate", "retentate", or bleed stream.

This type of diffuser configuration is often times referred to as inside out where the feed gases flow through the inside in the Pd-Ag tubing (membrane), and the hydrogen permeates through the membrane wall into the open evacuated shell space. The outside-in configuration flows feed

gases into the shell and the hydrogen permeates through the Pd-Ag membrane into the inside of the tubing, and will not be discussed in this report.

This prototype diffuser was designed to remove hydrogen isotopes from a feed stream at 200 sccm containing 500 ppm non-hydrogen species with the balance being hydrogen isotopes. The design criterion limits the raffinate stream to less than 0.03% hydrogen isotopes. The design temperature of the Pd-Ag diffuser is 525°C and the desig pressure is 60 psig.

2.2 Performance Testing at Vendor

As part of the acceptance of the new Pd-Ag, a series of initial performance tests were completed at the vendor's facility. The test system had mass flow controllers and pressure transducers on the feed and UPH sides of Pd-Ag diffuser. The UPH stream was discharged directly to the atmosphere and there was no pump on the UPH side of the diffuser. The experimental set-up at the vendor's facility is shown in Figure 2.2. The vendor's set-up did not have the means to measure the hydrogen that remained in the bleed stream.

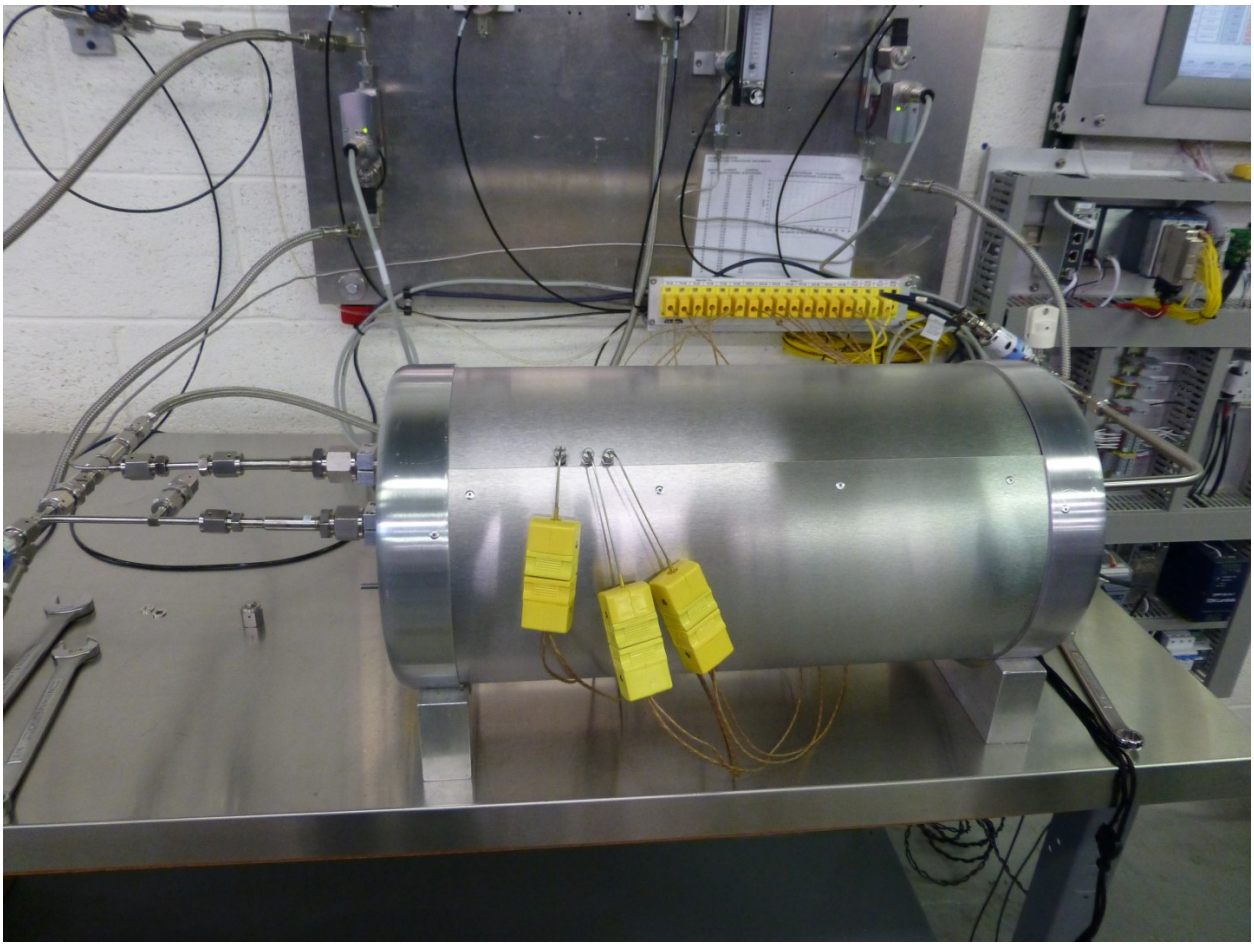

Figure 2.2: Experimental Test Set-up at Vendor's Facility

The Pd-Ag diffuser was brought to the operating temperature of 400°C. The H_2 feed pressure was controlled to 2327 Torr. The feed to the diffuser was pure hydrogen. The pressure differential across the Pd-Ag membrane allowed the hydrogen to permeate through the membrane.

2.3 Experimental Manifold

A flow test manifold was assembled to test Pd-Ag diffusers and is shown in Figure 2.3. A schematic of the experimental test system is shown in Figure 2.4. The 20 standard liters per minute (SLPM) test system (2TS) was initially configured to test the Johnson Matthey (JM) diffuser. The 2TS system was designed to feed a variety of gas mixtures to the diffuser at constant pressure using a series of flow controllers, pressure transducers, and back pressure regulators.

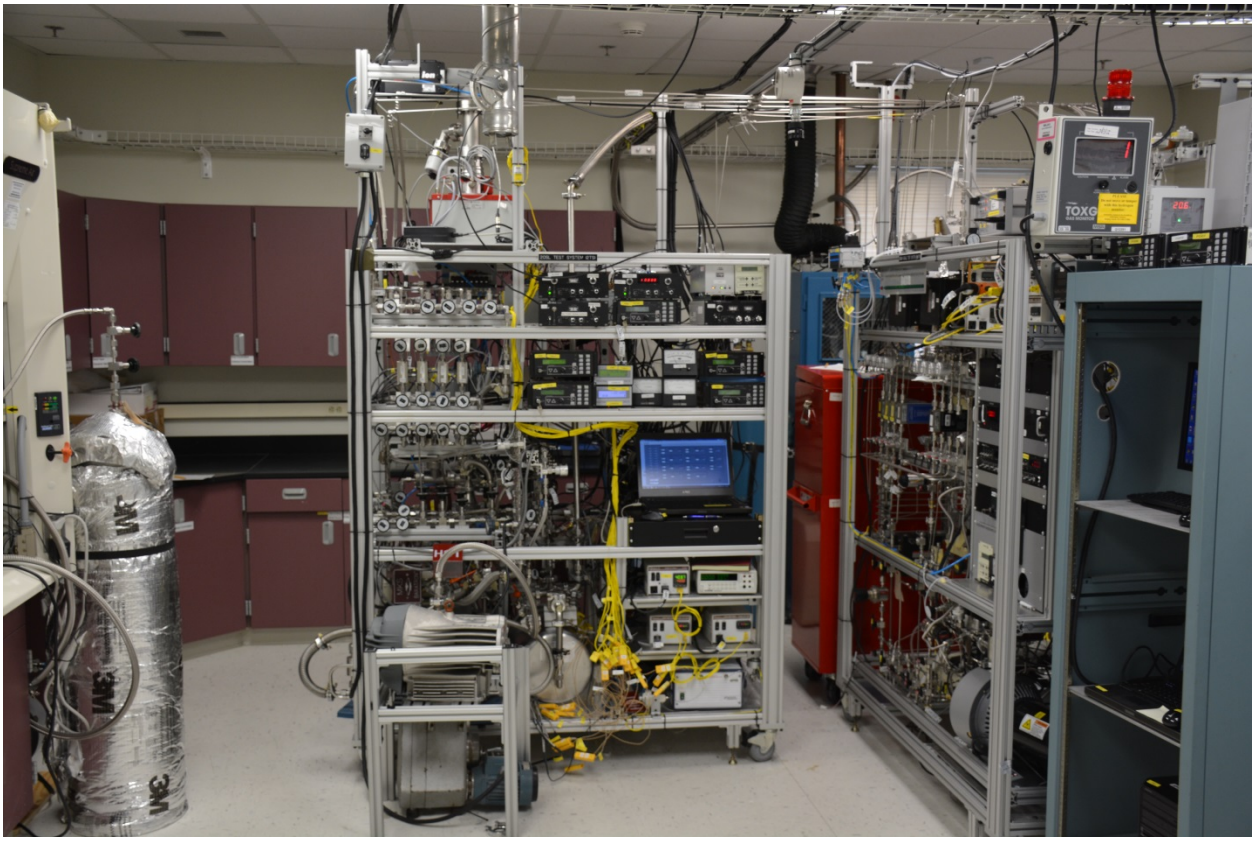

Figure 2.3: 2TS Diffuser Test System

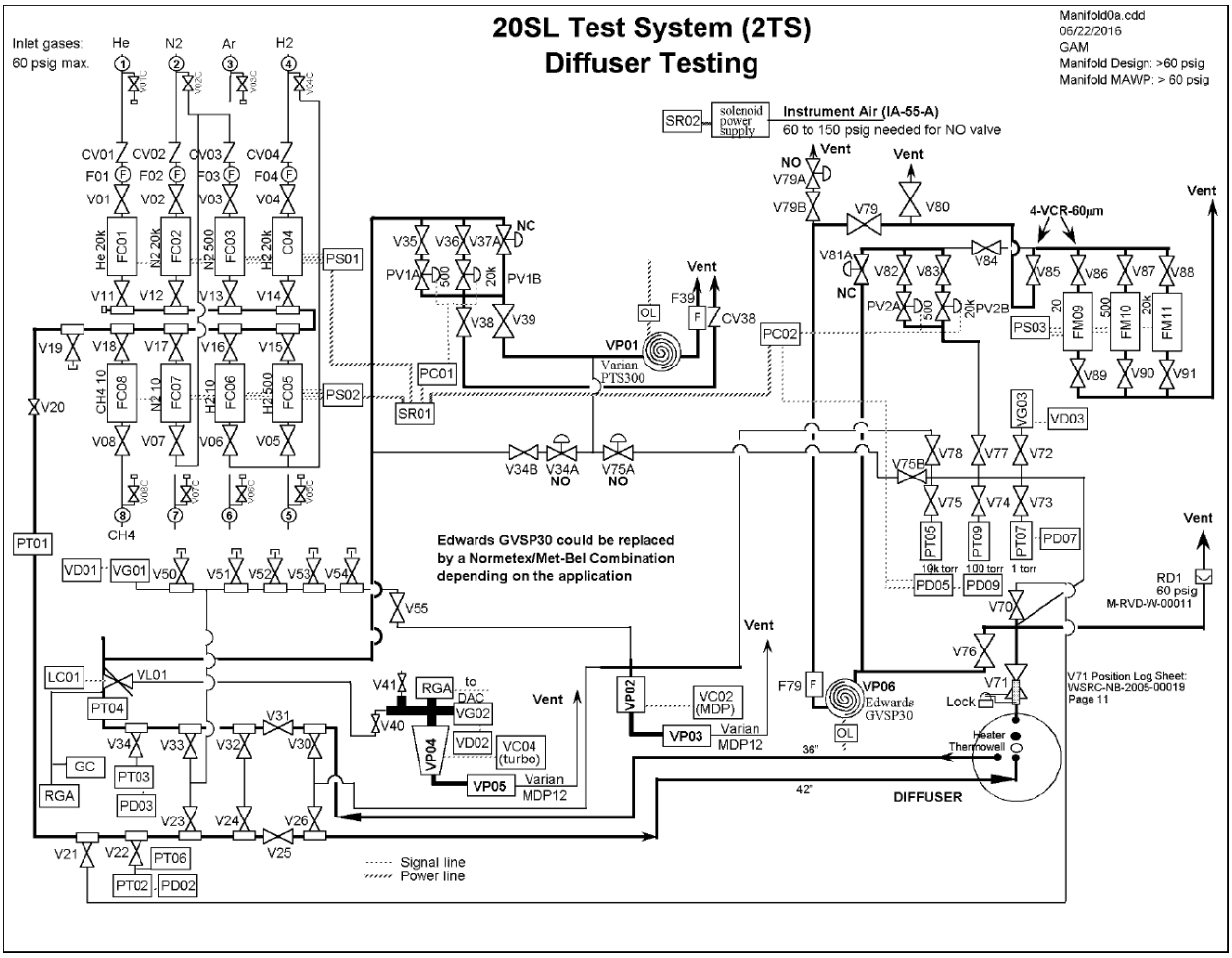

Figure 2.4: Schematic of the 20SL Test System.

Hydrogen and nitrogen gases were supplied to the diffuser feed tube by mass flow controllers (MFCs). The raffinate, or bleed, tube was connected to a back-pressure control valve followed by a vacuum pump (Varian scroll pump). The diffuser shell (permeate gas) was evacuated by a Normetex model 15 scroll pump (or by an Edwards nXDS15iC) backed by a MB-601 Metal-Bellows vacuum pump with the two heads connected in series. VP06 in Figure 2.4 was replaced by the previously mentioned Normetex/Metal-Bellows pump combination or the Edwards nXDS15iC/Metal-Bellows pump combination. The exhaust from the MB-601 was directed to the mass flow meters for direct measurement of the permeate flow.

The bleed composition and flow rate can be calculated by knowing the feed flow rate, feed composition, and permeate flow rate. The system was equipped with a Dycor Dymaxion DM200M Residual gas Analyzer (RGA) for analysis of the gas compositions of the feed and bleed streams. The system is also equipped with an Inficon 3000 micro gas chromatograph (µ-GC) containing dual (A/B) molecular sieve columns (14m x 320µm x 30µm) and a thermal conductivity detector. The (µ-GC) will be used to analyze the feed and bleed gas compositions to determine the amount of hydrogen present in the bleed stream. The sample inlet, sample injector, and the column were controlled to 60°C. The sample injection time was 20 milliseconds and the

sample pump time was 10 seconds. The column pressure was 25 psi for the carrier gas. Helium carrier gas was used in Column A and argon carrier gas was used in Column B for gas analysis.

The diffuser has 4 band heaters with a total power of 2000 watts. The four band heaters are wired in parallel and each heater contains two thermocouples. There are three dual element Type K thermocouples installed on the diffuser for monitoring the temperatures of the feed manifold, ~~the~~ over temperature limits, and the control temperature. The thermocouples are identified in the schematic shown in Figure 1.1. Each of the dual element thermocouples is identified as TCX-1 and TC0X-2, where the X is the number of the thermocouple (1-7). There is one dual channel thermocouple per band heater, numbered TC01, TC02, TC03, and TC04. TC05-1 is connected to the heater controller for monitoring/controlling the diffuser temperature. TC06-1 and TC07-1 are connected to ~~the~~ over temperature controllers. The remaining channels of the thermocouples and the heater thermocouples are connected to the LabVIEW Data Acquisition System. The locations of the thermocouples are summarized in Table 2.1. Some of the thermocouples are not visible in Figure 1.1. The heater wires and the corresponding thermocouples protrude through an opening in the insulation jacket on the permeate side of the diffuser

Table 2.1: Thermocouple Locations and Connections

Source Thermocouple	LabVIEWConnection
TC05-2 (Control)	TC01
TC06-2	TC02
TC07-2 (Feed Gas)	TC03
TC01-1 (Heater 1)	TC04
TC01-2 (Heater 1)	TC05
TC06-1	TC06
TC02-2 (Heater 2)	TC07
Room Temperature Measurement	TC08
TC07-1 (Feed Gas)	TC09
TC03-2 (Heater 3)	TC10
TC04-1 (Heater 4)	TC11
TC04-2 (Heater 4)	TC12
TC05-1 (Control)	Heater Controller #1 (HC01)
TC02-1 (Heater 2)	Over Temperature Controller #1 (OT01)
TC03-1 (Heater 3)	Over Temperature Controller #2 (OT02)

The pressure transducers and associated readouts are all calibrated Measurement and Test Equipment (M&TE) with current calibrations. The system contains eight MKS mass flow controllers labeled FC01 through FC08 and three MKS mass flow meters (MFMs) labeled FM09 through FM11. The flow controllers were calibrated by the manufacturer for He, H_2, N_2, or CH_4. FC01 is for helium service up to 20K sccm of gas flow. FC02, FC03, and FC07 were utilized for N_2 service at flow rates of 20K sccm, 500 sccm, and 10 sccm, respectively. FC04, FC05, and FC06 were designed for H_2 service with flow rates of 20K sccm, 500 sccm, and 10 sccm, respectively. FC08 had a maximum flow rate of 10 sccm for CH_4. The three mass flow meters (FM09, FM10 and FM11) were specifically designed to measure the flow rate of hydrogen at 20 sccm, 500 sccm, and 20K sccm, respectively. The MFCs and MFMs were calibrated with a commercially calibrated laminar flow element procured from DHI instruments. The ranges and primary gases of the mass flow controllers and flow meters are summarized in Table 2.2.

Table 2.2: Ranges and Primary Gases of Flow Controllers

Equipment ID	Range	Primary Gas
FC01	20,000 sccm	He
FC02	20,000 sccm	N_2
FC03	500 sccm	N_2
FC04	20,000 sccm	H_2
FC05	500 sccm	H_2
FC06	10 sccm	H_2
FC07	10 sccm	N_2
FC08	10 sccm	CH_4
FM09	20 sccm	H_2
FM10	500 sccm	H_2
FM11	20,000 sccm	H_2

The 2TS system contains seven MKS pressure transducers (labeled PT01, PT02, PT03, PT04, PT07, PT08, and PT09) and two Paroscientific pressure transducers (PT05 and PT06) at various locations on the system. PT01 (M&TE HTS001669) and PT04 (M&TE HTS 001640) are 10K Torr pressure transducers that only have a digital output to the data acquisition system (DAQ). PT02 (M&TE HTS009143 and associated readout HTS009144) and PT03 (M&TE HTS001667 and associated readout HTS001562) are 10K Torr MKS Baratrons that measure the pressure in the gas line before the diffuser and after the diffuser, respectively. PT05 (200 psia, M&TE HTS009145 and readout HTS009152) is also used to measure the inlet pressure to the diffuser. PT06 (200 psia, M&TE HTS009147 and readout HTS009146), PT07 (100 Torr, M&TE HTS001672 and readout HTS001671) and PT09 (1 Torr, M&TE HTS001418 and readout HTS001333) are used to measure the shell pressure of the diffuser. PT08 (10K Torr, M&TE HTS001507 and associated readout HTS001508) is used to measure the pressure in the sampling line for the micro gas chromatograph. The pressure transducers and associated displays are summarized in Table 2.3.

Table 2.3: Pressure Transducers and Pressure Displays

Equipment ID	Range	M &TE #
PT01	10, 000 Torr	HTS001669
PT02/PD02	10,000 Torr	HTS001562/HTS09144
PT03/PD03	10,000 Torr	HTS001667/HTS001562
PT04	10,000 Torr	HTS001640
PT05/PD05	10,343 Torr	HTS009145/HTS009152
PT06/PD06	10, 343 Torr	HTS009147/HTS006146
PT07/PD07	100 Torr	HTS001672/HTS001671
PT08/PD08	10,000 Torr	HTS001507/HTS001508
PT09/PT10	1 Torr	HTS001418/HTS001333

2.4 Performance Testing at SRNL

Additional performance testing of the Pd-Ag diffuser was completed at SRNL. The characterization of the diffuser was completed by varying the following parameters: feed gas composition, flow rate, pump type, and internal tube pressure. Three gas compositions of hydrogen-nitrogen mixtures were selected and fed through the diffuser at flow rates ranging from 100 sccm to 3000 sccm. The gas mixtures that were used during the characterization of the diffuser contained 96%, 50%, and 2% H_2, with the remainder balance of the gas mixture being nitrogen.

The diffuser's internal tube pressure was maintained at 700 Torr or 1140 Torr for testing. Some tritium processing systems operate at slightly sub-atmospheric conditions; therefore, it is necessary to determine the Pd-Ag diffuser performance at these conditions.

Two pump types, Normetex 15 and Edwards nXDS15iC, were selected as potential pumps to evacuate the permeate gas out of the diffuser shell space. Normetex pumps are currently used in various tritium facilities, due to their compatibility with tritium. It is recommended that tritium processing systems utilize Normetex scroll pumps for the pumping of tritated gas; however, these pumps are no longer manufactured. An alternative pump, Edwards nXDS15iC, was found to be a low cost replacement for the Normetex pump for tritium service with certain qualifications [5]. The Edwards nXDS15iC pump needs to be evaluated with the diffuser assembly to determine its performance characteristics. The performance of the Edwards pump will be evaluated and compared to the performance of the Normetex pump for diffuser performance characterization. Table 2.4 shows the experimental conditions that were evaluated during testing.

Table 2.4: Experimental Parameters for Pd-Ag Diffuser Characterization Testing

Gas Composition	Pump Type	Tube Pressure (Torr)
2% H_2 / 98% N_2	Normetex 15	700
50% H_2 / 50% N_2	Normetex 15	700
96% H_2 / 4% N_2	Normetex 15	700
2% H_2 / 98% N_2	Edwards nXDS15iR	700
50% H_2 / 50% N_2	Edwards nXDS15iR	700
96% H_2 / 4% N_2	Edwards nXDS15iR	700
2% H_2 / 98% N_2	Normetex 15	1140
50% H_2 / 50% N_2	Normetex 15	1140
96% H_2 / 4% N_2	Normetex 15	1140
2% H_2 / 98% N_2	Edwards nXDS15iR	1140
50% H_2 / 50% N_2	Edwards nXDS15iR	1140
96% H_2 / 4% N_2	Edwards nXDS15iR	1140

3.0 Results and Discussion

3.1 Results of the Performance Testing at the Vendor

Figure 3.1 shows the initial results of the performance testing of the Pd-Ag diffuser that was manufactured by Power and Energy. These tests were completed at the vendor facilities.

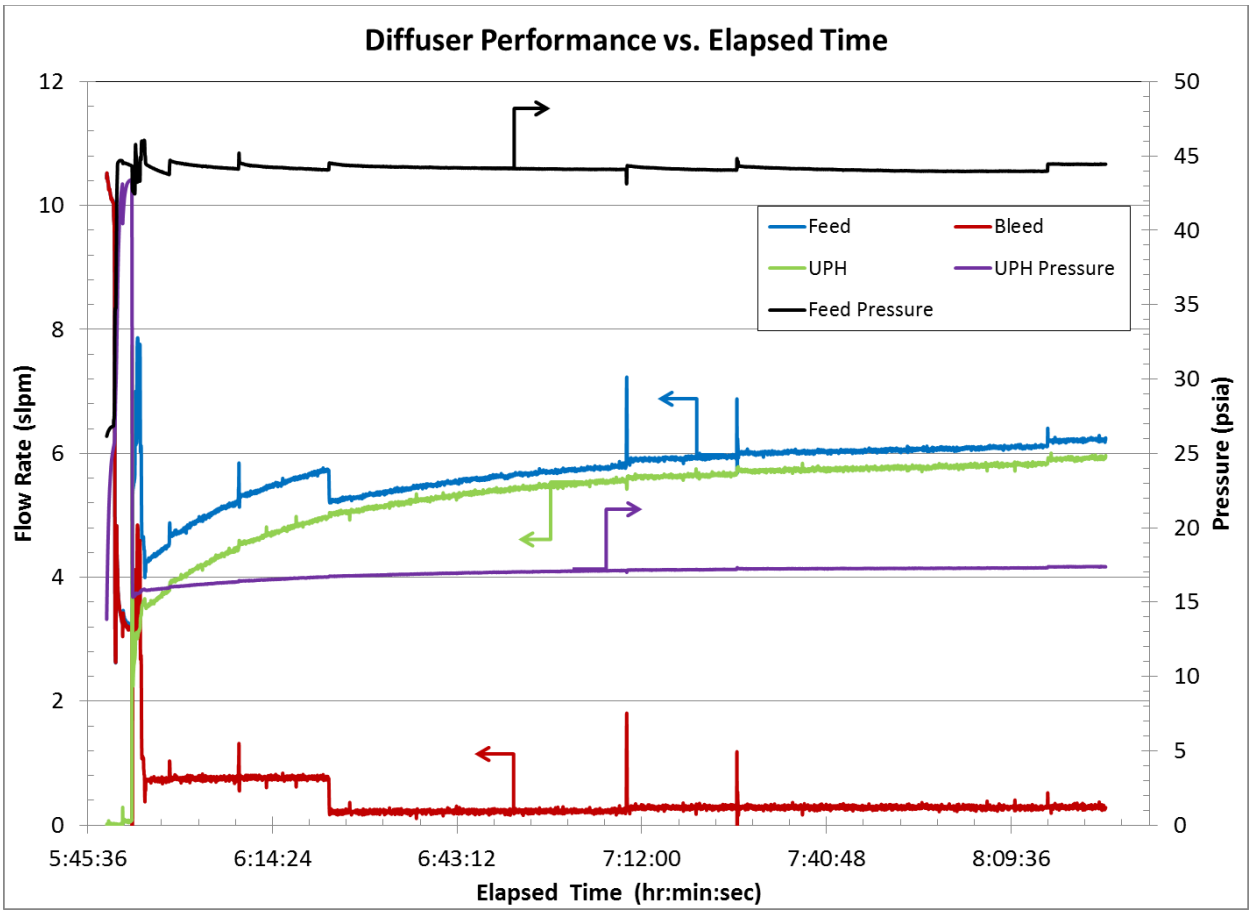

Figure 3.1: Initial Vendor Performance Testing of a Pd-Ag Diffuser Manufactured by Power and Energy

The feed pressure was controlled to about 2327 Torr for the duration of the test. The pressure measured at the permeate side was the permeate side was about 905 Torr.

The H_2 was fed to the Pd-Ag diffuser from a hydrogen cylinder at a flow rate of 6.2 standard liters per minute (SLPM) and the flow rate measured on the permeate side was 5.9 SLPM during the testing. The UPH flow rate was always less than the feed flow rate indicating that less than 100% of the available hydrogen was permeating through the Pd-Ag membrane. At various times during the flowing of hydrogen through the permeator, the UPH flow rate was typically 0.3 SLPM less than the feed flow rate. The flow rates that were used for this test may be above the capacity of the Pd-Ag diffuser, therefore not allowing all of the H_2 to permeate through the Pd-Ag membrane. It also may be possible that the pressure differential across the membrane may not have been large enough to facilitate complete diffusion of the hydrogen molecules.

The only criterionthat was evaluated at the vendor was the flow of pure hydrogen through the diffuser. There were no pre-determined criteria for acceptance at the vendor facility. Even though the Pd-Ag diffuser passed the preliminary performance testing at the vendor's facility, the actual application for the tritium fuel cycle is different than the set-up for the initial performance testing. For instance, the permeate (or UPH) side of the diffuser was discharged directly to the atmosphere during the performance testing of the diffuser. However, diffusers in tritium service

typically have vacuum pump(s) on the UPH outlet of the diffuser to ensure that the maximum amount of high purity hydrogen is removed from the feed stream. The particular application of this diffuser will utilize a Normetex 15 scroll pump (or an Edwards nXDS15iC) and a Metal Bellows pump to achieve a vacuum level of ~10^{-3} Torr on the permeate side of the diffuser. This will significantly affect the hydrogen permeation characteristics of the Pd-Ag diffuser.

3.2 Results of the Performance Testing at SRNL

Plots of bleed flow rate versus total (feed) flow rate are used to determine diffuser breakthrough curves. To determine diffuser performance, feed compositions and flow rates need to be known along with the flow rates of the bleed (raffinate) and pure (permeate) streams. The feed stream gas was produced by blending gases using mass flow controllers (MFCs) so feed flows and compositions can be calculated. The system is designed to evacuate the pure stream through a vacuum pump. The discharge of the vacuum pump is then directed to a user selectable range of mass flow meters (MFMs). The system is designed to check the measured flow of hydrogen through the MFMs by taking the hydrogen flow from the feed MFCs, bypassing the diffuser, passing this gas through the MFMs and comparing the MFC flow to the MFM flow. By knowing the feed flow rate and composition, measuring the pure/permeate flow (assuming no leaks so the permeate composition is 100% hydrogen), the flow rate and composition of the bleed/raffinate stream can be calculated.

A residual gas analyzer (RGA) will be employed to monitor the gas composition in the feed and/or bleed streams before and after the diffuser, respectively. Changes in the gas composition will be used to determine if the non-hydrogen species present in the feed stream are dissociating or otherwise reacting inside the diffuser. In addition, a micro gas chromatograph (μ-GC) will be used in an attempt to quantify the amount of hydrogen in the raffinate stream.

3.2.1 Results of the Performance Testing with Normetex Pump

A comparison of the bleed versus the total feed flow rate curves for the three gas compositions is shown in Figure 3.2. Experiments were conducted at 700 Torr and 1140 Torr with the permeate side of the diffuser pumped with the Normetex/Metal-Bellows pump combination. Each of the bleed vs. feed curves for the three compositions (96% H_2/4% N_2, 50%H_2/50% N_2, and 2%H_2/98% N_2) at 700 Torr overlap almost exactly with the curves at 1140 Torr. There is a slight difference between the 50%H_2/50% N_2 curves at flow rates above 1500 sccm.

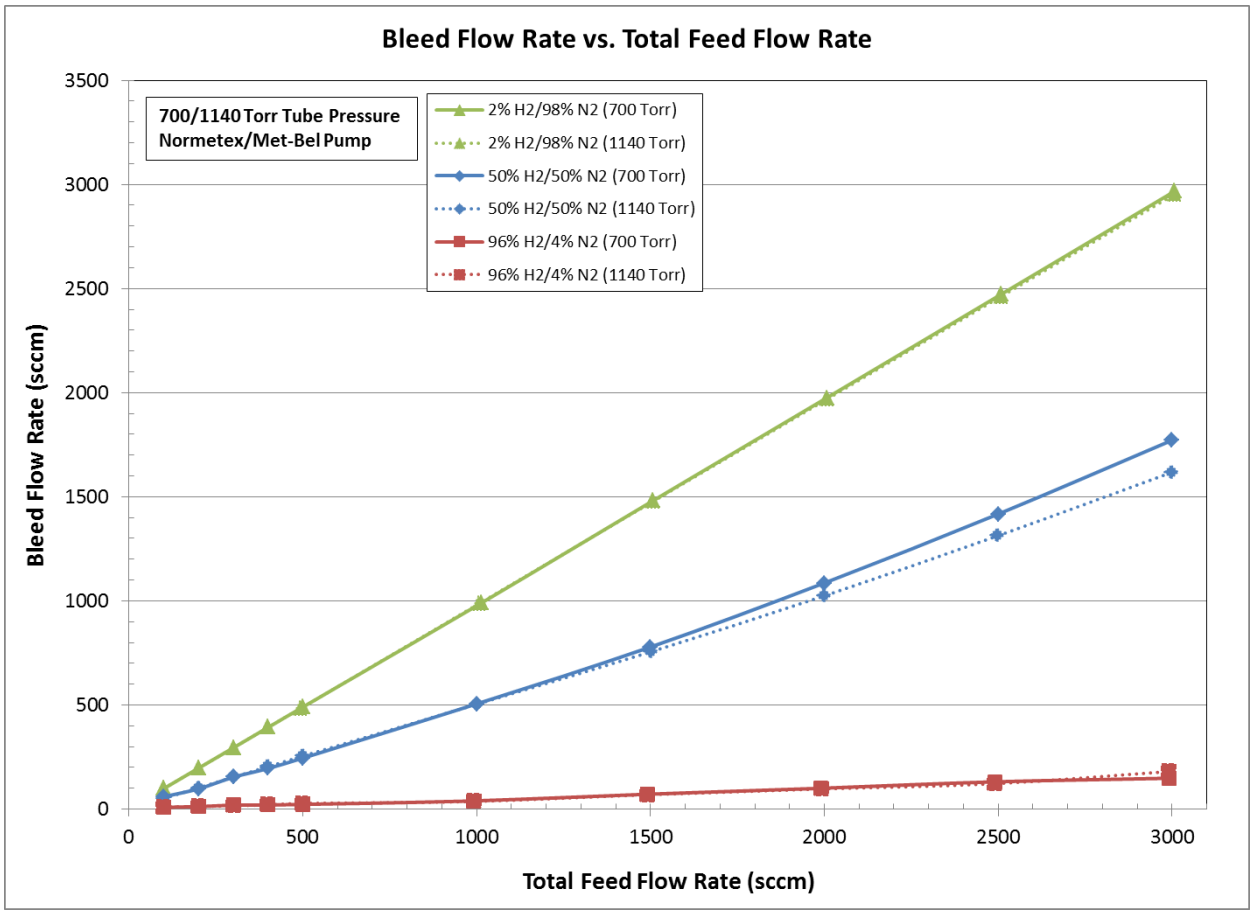

Figure 3.2: Plot of Bleed Flow Rate vs. Total Feed Flow Rate for Various Gas Compositions and Pressures with the Normetex Pump

Figure 3.3 shows the amount of hydrogen permeating through the Pd-Ag membrane versus the total feed flow rate. The amount of hydrogen permeating is calculated from the feed flow rate and the permeate flow rate. Both of the 96%H_2/4% N_2 curves indicate that above a total feed flow rate of 300 sccm nearly 100% of the available hydrogen is permeating through the membrane. For the 50%H_2/50% N_2 composition, there is only 100% permeation up to about 1000 sccm. Above 1000 sccm, the percentage of hydrogen that is permeating drops to ~82% and ~92% at internal tube pressures of 700 Torr and 1140 Torr, respectively. The two cases with 2% H_2/98% N_2 mixture never achieved 100% of the hydrogen permeating through the membrane. At flows above 500 sccm, the percentage of hydrogen that is permeating drops to ~66 % and ~90% at internal tube pressure of 700 Torr and 1140 Torr, respectively.

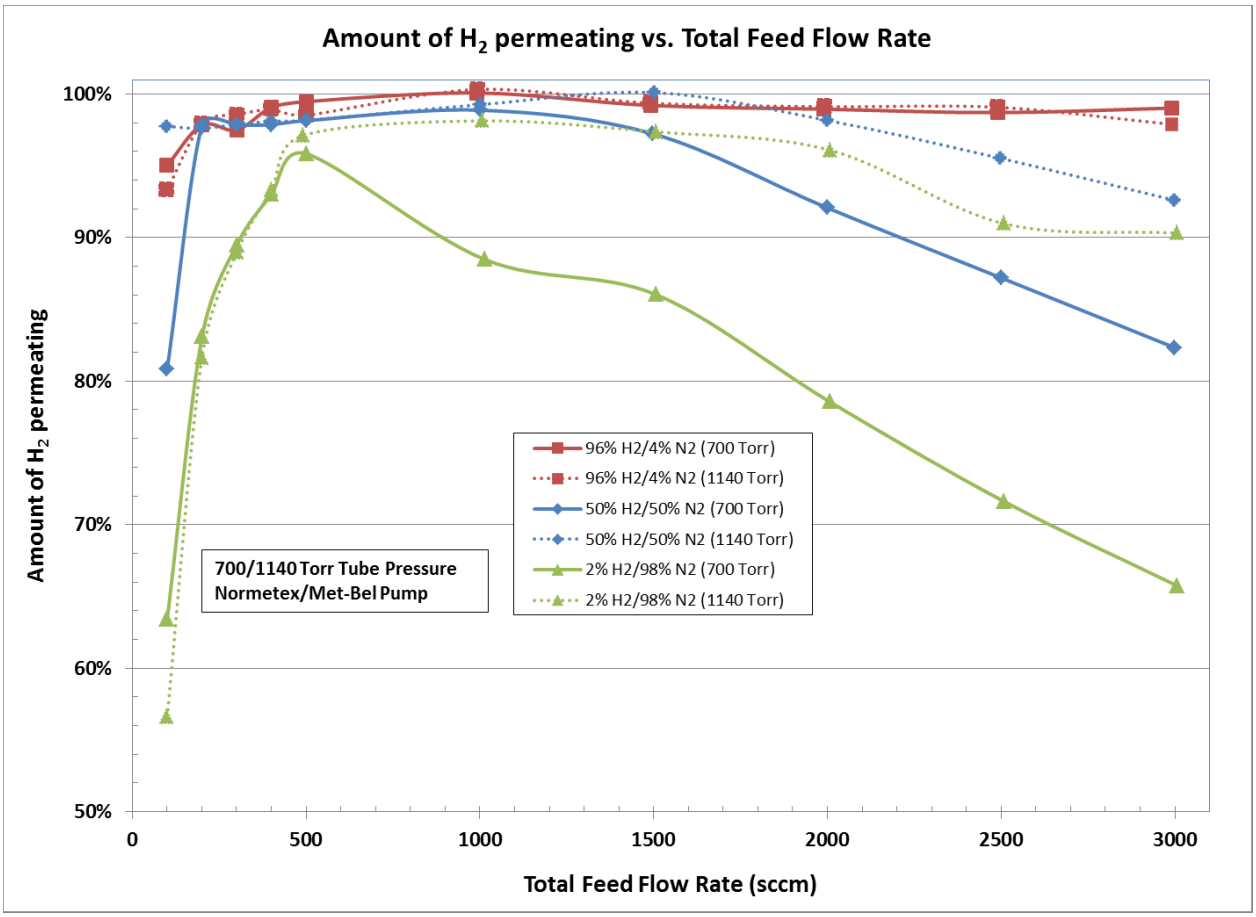

Figure 3.3: Plot of the Amount of H_2 Permeating vs. the Total Feed Flow Rate for Various Gas Compositions and Pressures with the Normetex Pump

Figure 3.4 shows the average shell pressure against the total feed flow rate using the Normetex/Met-Bel pump combination. The two cases with the feed composition of 96% H_2/4% N_2 had the highest shell pressures of the conditions tested. As the H_2 in the feed stream increases, more H_2 is available for the permeation through the Pd-Ag membranes, which results in an increased shell pressure. The increase in the shell pressure does not impede further diffusion of the hydrogen through the membrane for this particular diffuser configuration as there is still nearly 100% of the available hydrogen permeating at the 96% H_2/4% N_2 gas composition.

For the 96% H_2/4% N_2 and 2% H_2/98% N_2 cases, each of the bleed vs. feed curves for the two compositions at 700 Torr overlap almost exactly with the curve at 1140 Torr. The two curves for the 50% H_2/50% N_2 gas mixture are shown to slightly diverge from each other at feed flowrates above 1500 sccm. The shell pressure is not the limiting factor in the permeation of hydrogen through the Pd-Ag membrane.

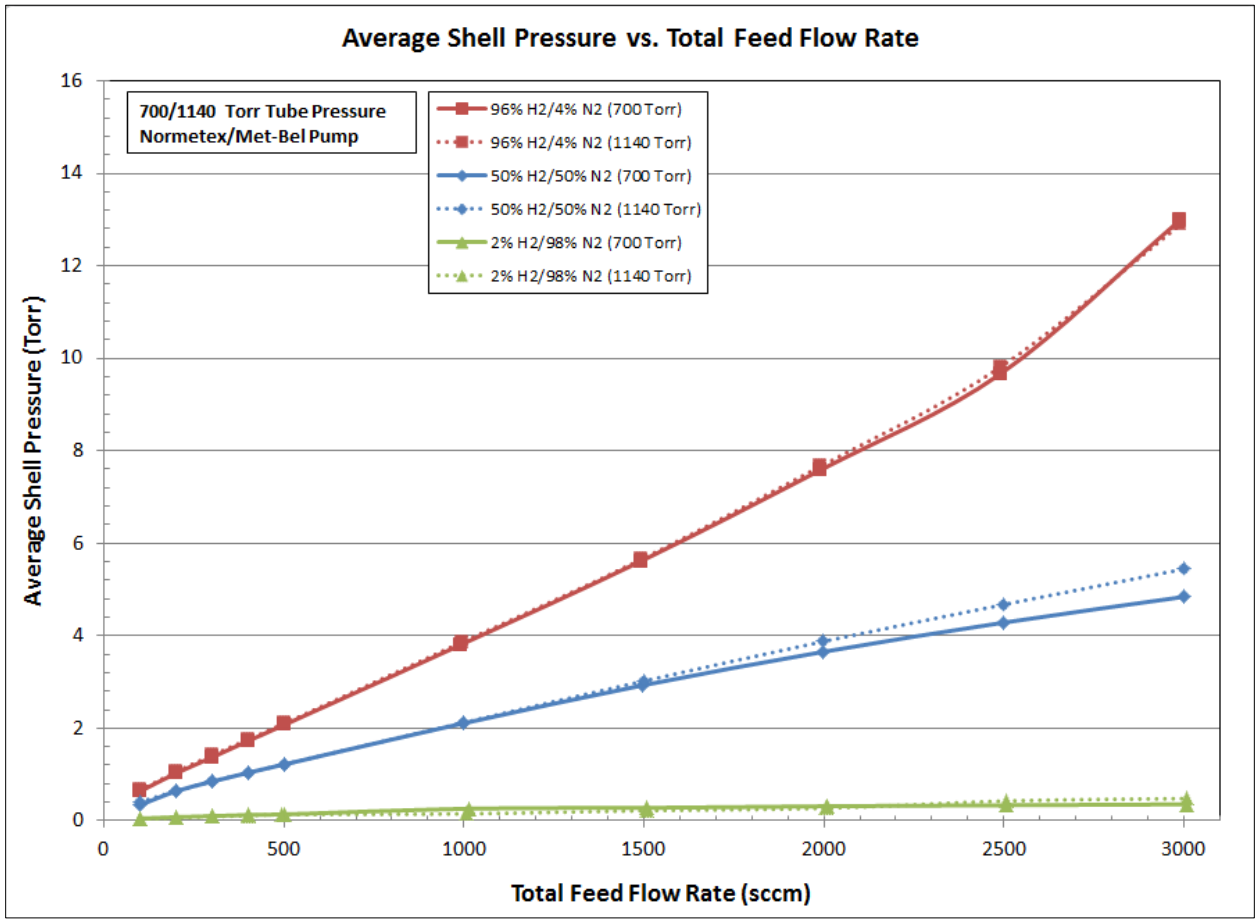

Figure 3.4: Plot of the Shell Pressure vs. the Total Feed Flow Rate for Various Gas Compositions and Pressures with the Normetex Pump

Figure 3.5 shows the pressure drop across the Pd-Ag diffuser versus the total feed flow rate. The pressure drop discussed here is the difference between the inlet feed stream pressure and the outlet bleed stream pressure . As the flow rate is increased for each of the gas compositions, there is a slight increase in the pressure drop across the diffuser. The Pd-Ag diffuser is originally designed for a maximum flow rate of 200 sccm. At a flow rate of 400 sccm for all of the gas compositions, the pressure drop across the diffuser assembly is less than 5 Torr.

The relatively small pressure drop across the diffuser assembly indicates that multiple diffuser stages could be placed in series to remove hydrogen isotopes from the bleed stream without the use of additional process equipment (i.e. pumps between the stages). The bleed stream from the first stage of the diffuser would become the feed stream of the second diffuser. Even though the pressure drop is negligible across the Pd-Ag diffuser, multiple diffusers connected in series will not dramatically increase the amount of hydrogen that can be removed from the feed stream.

The results presented in this paper indicate that at low-mid levels of H_2 in the feed stream that only a fraction of the available H_2 is able to permeate through the membrane. It is very unlikely that even with several Pd-Ag diffusers connected in series that the amount of H_2 remaining in the bleed stream can be reduced to levels acceptable for environmental discharge when the diffuser is used for tritium service. The residence time through the diffuser ranges from ~5.3 seconds at a flow rate of 100 sccm to ~0.17 seconds at a flow rate of 3000 sccm. In the presence of high concentrations of non-hydrogen species this residence time may not be long enough for the hydrogen to diffuser to the Pd-Ag membrane and dissociate.

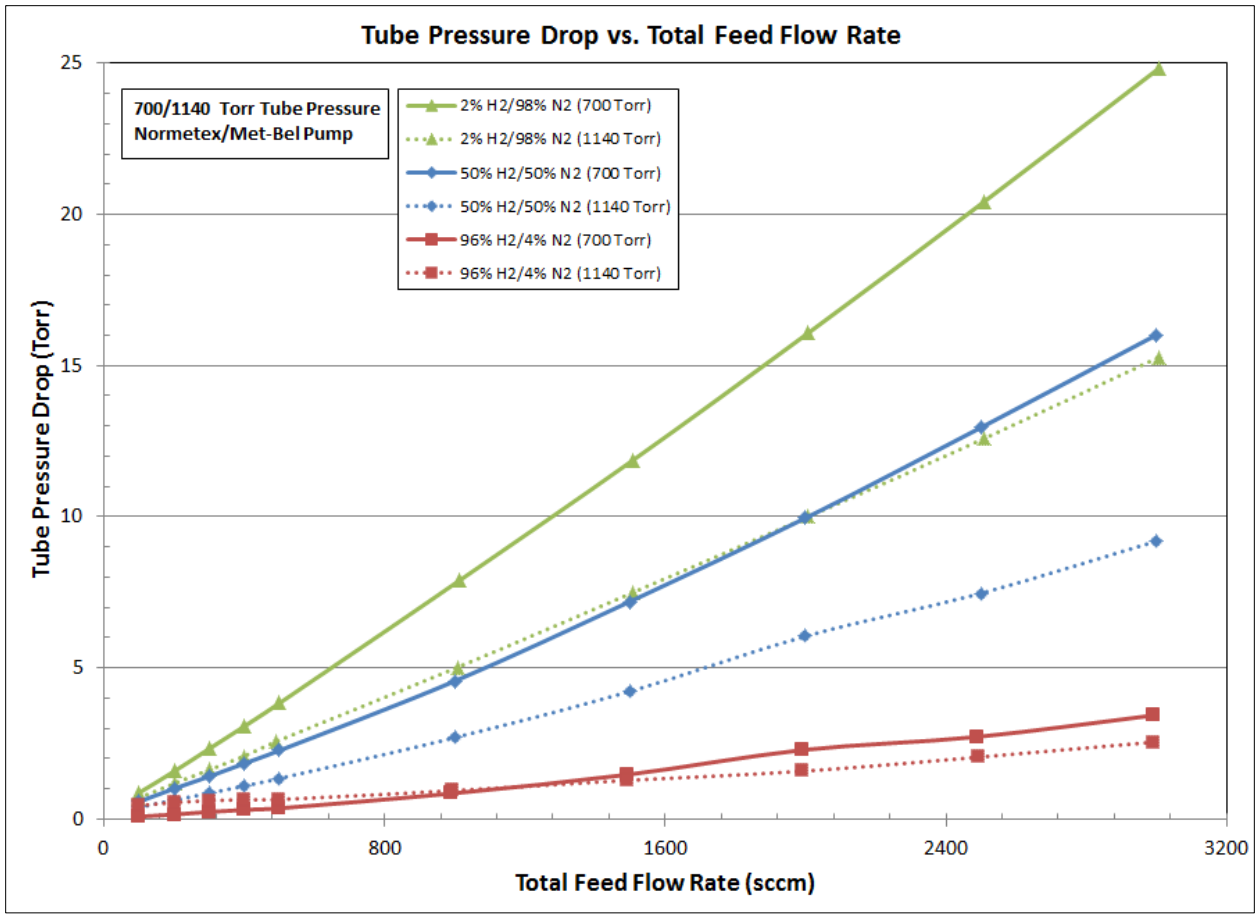

Figure 3.5: Plot of the Pressure Drop across the Diffuser vs. the Total Feed Flow Rate for Various Gas Compositions and Pressures with the Normetex Pump

3.2.2 Results of the Performance Testing with Edwards Pump

Figure 3.6 shows the bleed flow rate against the total feed flow rate curves for various gas compositions and internal tube pressures. Experiments were conducted with the permeate side of the diffuser pumped with the Edwards/Metal-Bellows pump combination. Each of the curves for the three gas compositions at an internal tube pressure of 700 Torr overlap almost exactly with the curves at 1140 Torr. The very slight differences in the curves can be attributed to the inconsistencies of the flow meters.

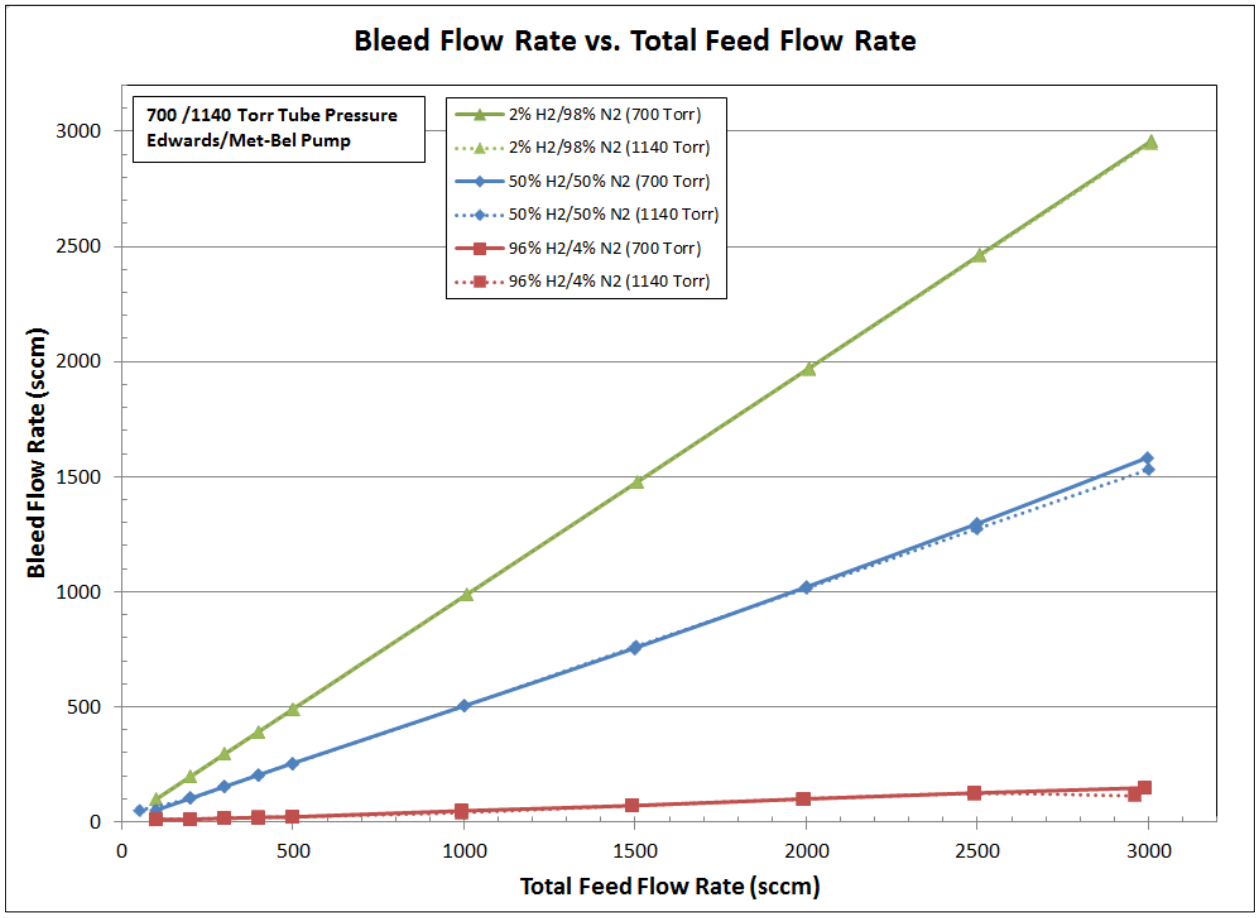

Figure 3.6: Plot of Bleed Flow Rate vs. Total Feed Flow Rate for Various Gas Compositions and Pressures with the Edwards Pump

Figure 3.7 shows the percentage of hydrogen permeating through the Pd-Ag membrane versus the total feed flow rate using an Edwards/Metal-Bellows pump combination. Both of the 96% H_2/4% N_2 and 50% H_2/50% N_2 curves indicate that above a total feed flow rate of 100 sccm, nearly 95-99% of the available hydrogen in the feed stream is permeating though the membrane. There were a few cases in which the percentage of hydrogen permeating was above 100%. This is due to a small amount of drift associated with the flow meters and controllers.

The 2% H_2/98% N_2 mixture was the only feed gas composition in which 100% H_2 permeation was not achieved. Even as the internal tube pressure was increased, the H_2 permeation through the Pd-Ag membranes only marginally increased. Above a total feed flow rate of 1000 sccm, the percentage of hydrogen permeating dropped to ~83% and ~95% at the internal tube pressures of 700 Torr and 1140 Torr, respectively. The percentage of available hydrogen permeating through the membrane is lower for the 2% H_2/98% N_2 cases compared to the 96% H_2/4% N_2 and 50% H_2/50% N_2. This reduced permeation is likely due to blanketing of the active permeation sites due to the increased concentration of nitrogen. The nitrogen molecules accumulate along the tube wall, impeding the hydrogen molecules from dissociating at the Pd-Ag surface and subsequently diffusing through the membrane.

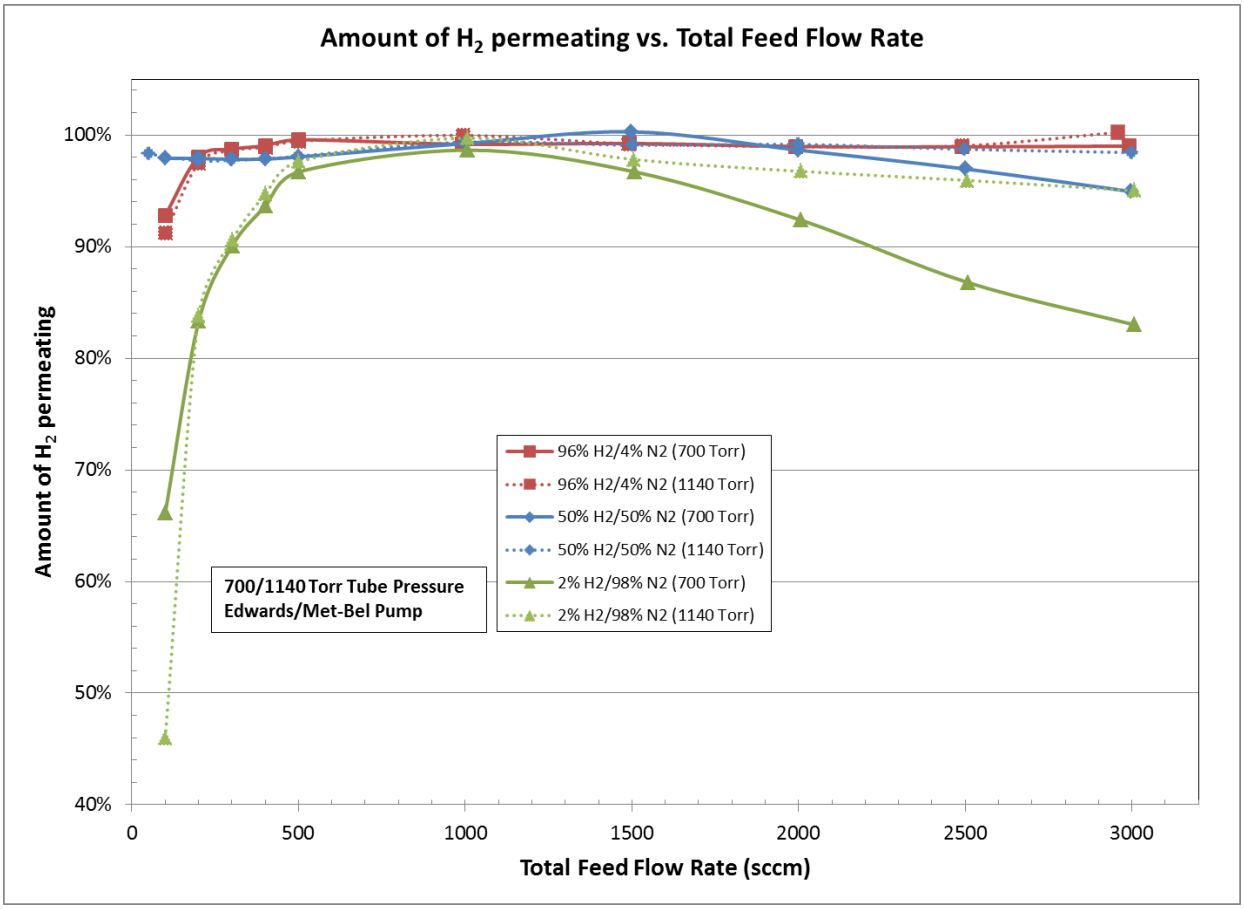

Figure 3.7: Plot of the Amount of H_2 Permeating vs. the Total Feed Flow Rate for Various Gas Compositions and Pressures with the Edwards Pump

Figure 3.8 shows a plot of the average shell pressure vs. total feed flow rate for the three feed gas compositions at two internal tube pressures. An Edwards/Metal-Bellows pump was used to pump the permeate side of the diffuser. Each of the shell pressure against feed rate curves for the three compositions at 700 Torr overlap almost exactly with the curves at 1140 Torr. There is a slight difference between the two 50%H_2/50% N_2 curves at flow rates above 1000 sccm. This similarity was noted for the same gas composition when using a Normetex/Metal-Bellows pump.

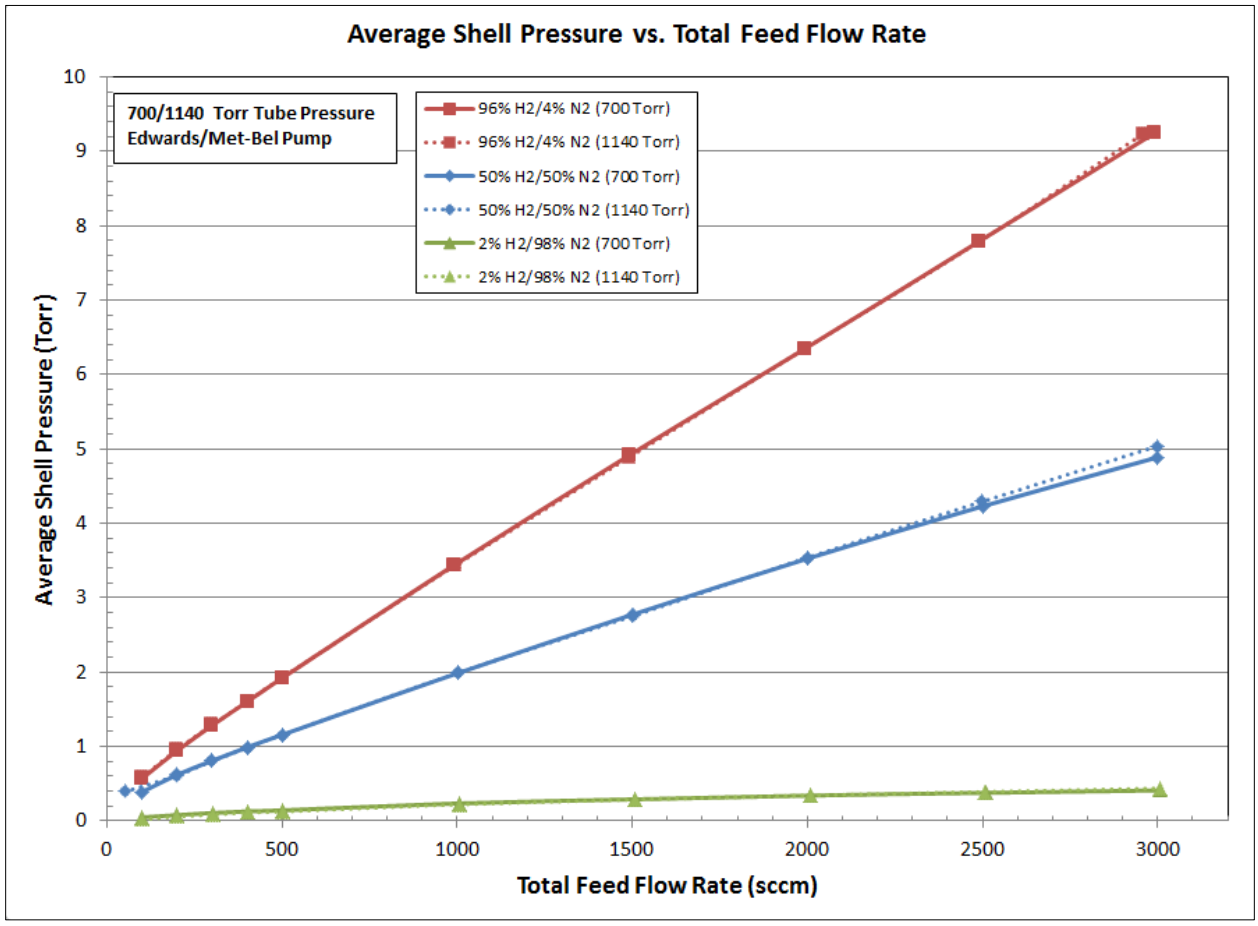

Figure 3.8: Plot of the Shell Pressure vs. the Total Feed Flow Rate for Various Gas Compositions and Pressures with the Edwards Pump

3.2.3 Comparison of the Normetex and Edwards Pumps

Figure 3.9 through Figure 3.14 show the comparison of the performance of the Normetex Model 15 pump and the Edwards nXDS15iC pump. The data in these figures is identical to the data plotted in Figure 3.2 through Figure 3.8. Figure 3.9 and Figure 3.10 show the amount of hydrogen permeating versus the total feed flow rate for 700 Torr and 1140 Torr, respectively.

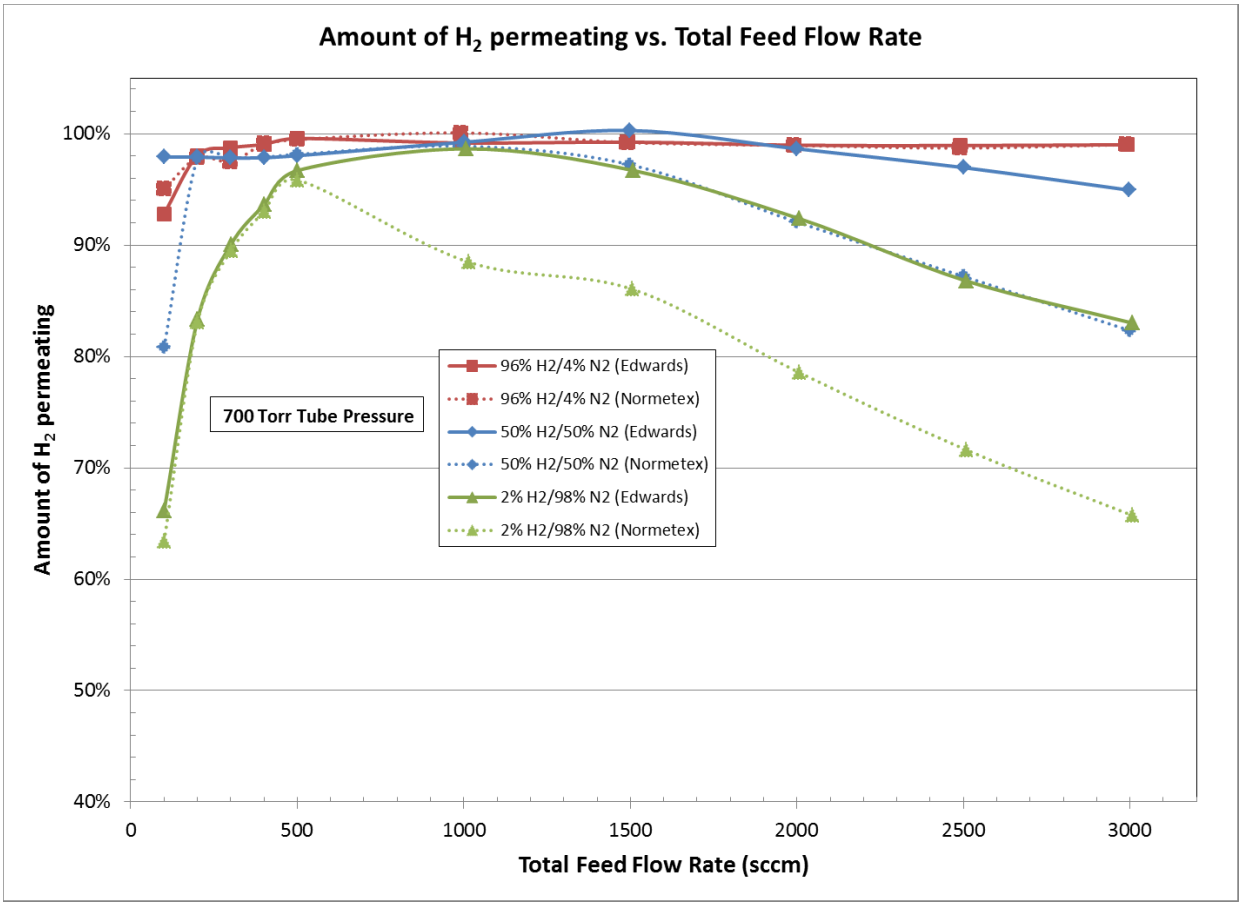

Figure 3.9: Plot of the Amount of H_2 Permeating vs. the Total Feed Flow Rate for Various Gas Compositions at 700 Torr

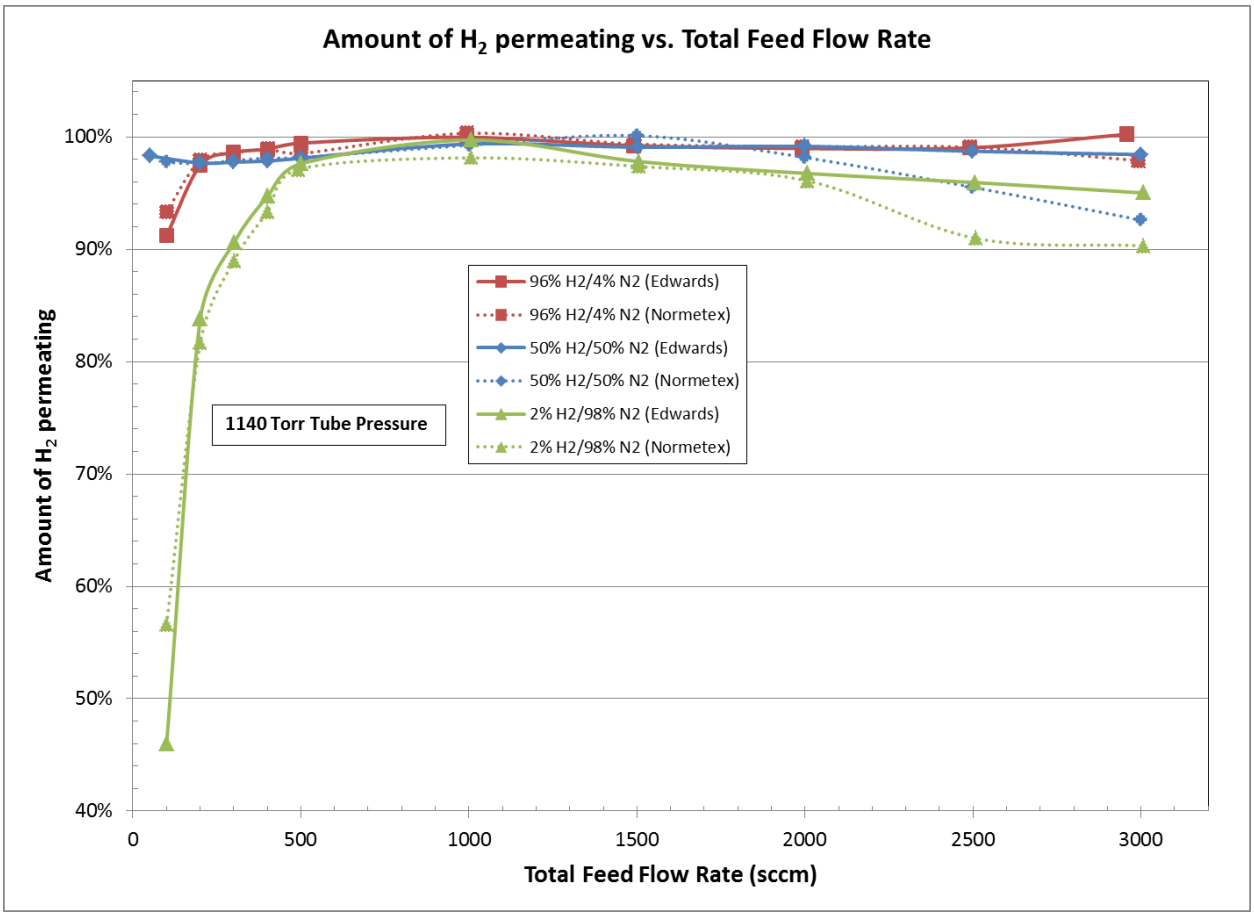

Figure 3.10: Plot of the Amount of H_2 Permeating vs. the Total Feed Flow Rate for Various Gas Compositions at 1140 Torr

Figure 3.11 and Figure 3.12 show the amount of hydrogen remaining in the bleed stream versus the total feed flow rate at 700 Torr and 1140 Torr, respectively. The amount of hydrogen in the bleed stream was calculated from the feed flow rates and permeate flow rates.

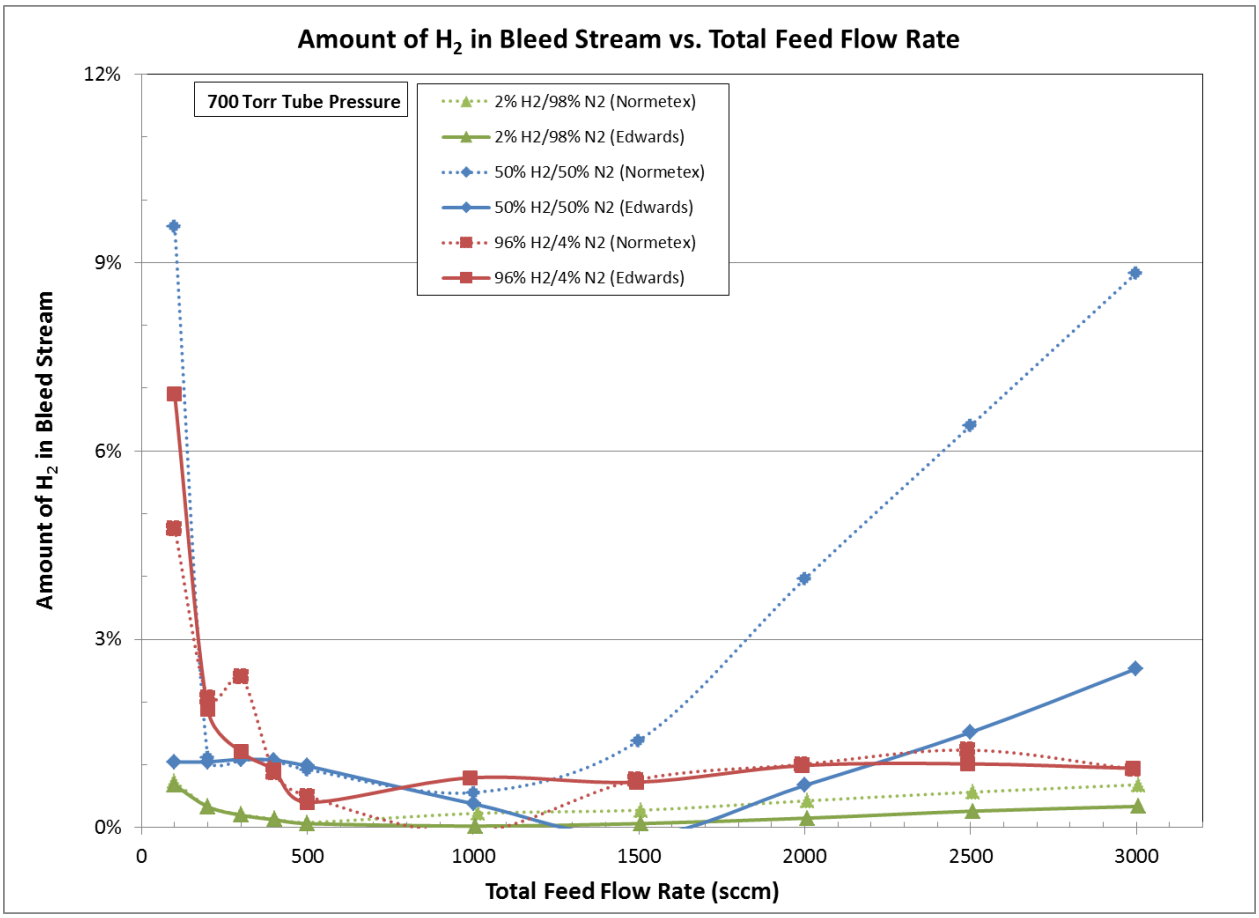

Figure 3.11: Plot of the Amount of H_2 in the Bleed Stream vs. the Total Feed Flow Rate for Various Gas Compositions at 700 Torr

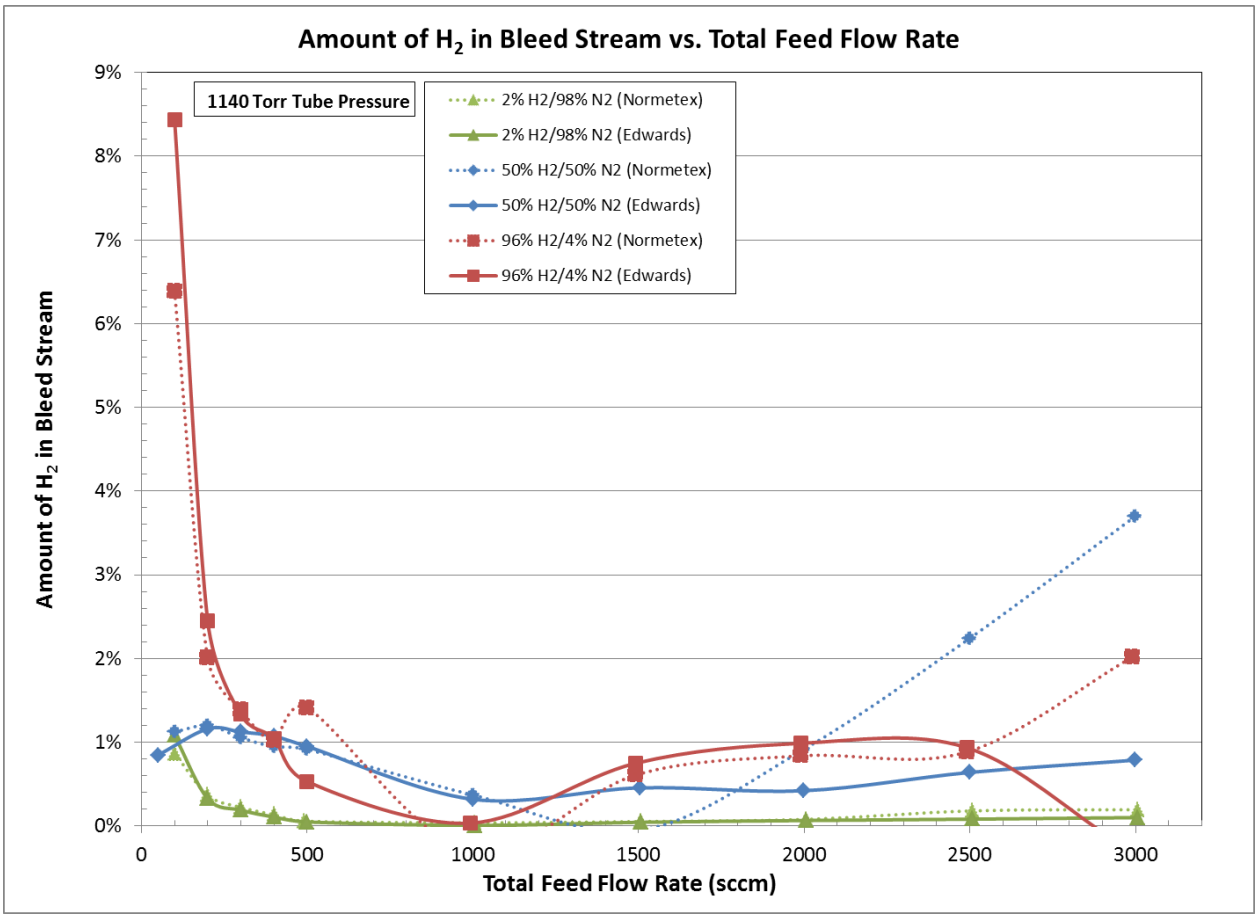

Figure 3.12: Plot of the Amount of H_2 in the Bleed Stream vs. the Total Feed Flow Rate for Various Gas Compositions at 1140 Torr

The average shell pressure versus the total feed flow rate for 700 Torr and 1140 Torr is shown in Figure 3.13 and Figure 3.14, respectively. For all three gas compositions at both 700 Torr and 1140 Torr, the shell pressure is lower when the permeate stream is pumped by the Edwards nXDS15iC compared to the Normetex Model 15 scroll pump.

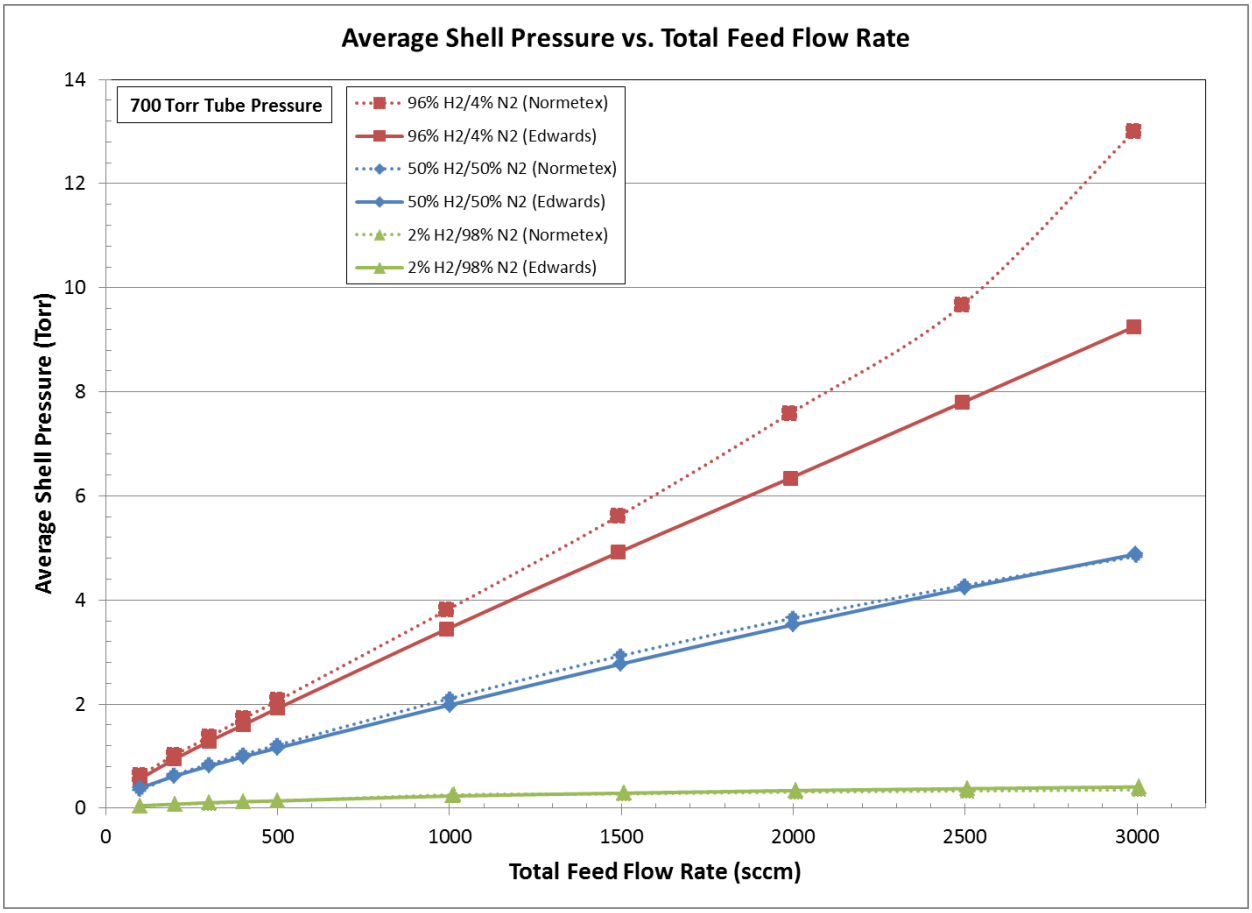

Figure 3.13: Plot of the Shell Pressure vs. the Total Feed Flow Rate for Various Gas Compositions at 700 Torr

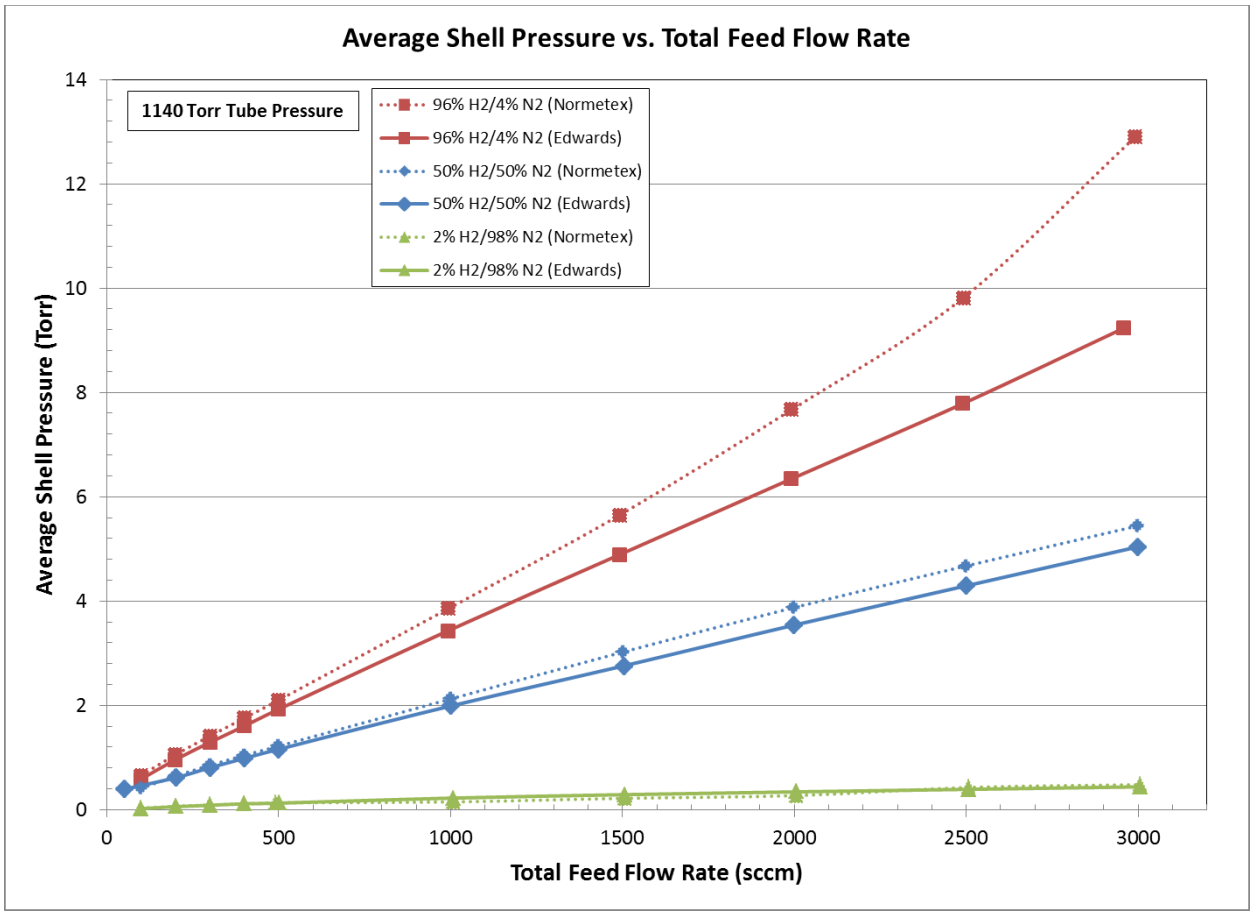

Figure 3.14: Plot of the Shell Pressure vs. the Total Feed Flow Rate for Various Gas Compositions at 1140 Torr

3.2.4 Residual Gas Analysis

The 2TS system is equipped with a Dycor Dymaxion DM200M Residual Gas Analyzer (RGA) for analysis of the gas compositions of the feed and bleeds streams. The hydrogen and nitrogen signal measured from the RGA for 96% H_2/4% N_2 mixture is shown in Figure 3.15 and Figure 3.16 for the Normetex and Edwards pump configurations, respectively. The sharp decrease and increase in the hydrogen and nitrogen signals, shown at the time interval of 40 to 80 minutes, corresponds to the time when the process gas is introduced into the diffuser. The periodic oscillations that can be seen in the nitrogen and hydrogen signals correspond to the μ-GC measurements and the re-stabilization of the pressure in the Pd-Ag tubes.

There are no oscillations in the hydrogen signal when the Edwards pump is used, as shown in Figure 3.16. The hydrogen signal is very low (~1 x 10^{-9}) for both of the cases indicating that the diffuser is removing most of the available hydrogen. There is only a slight increase in the hydrogen signal as the flow rate is increased. Most of the hydrogen in the feed stream is permeating through the Pd-Ag membrane into the shell space of the diffuser and exiting out of the permeate stream.

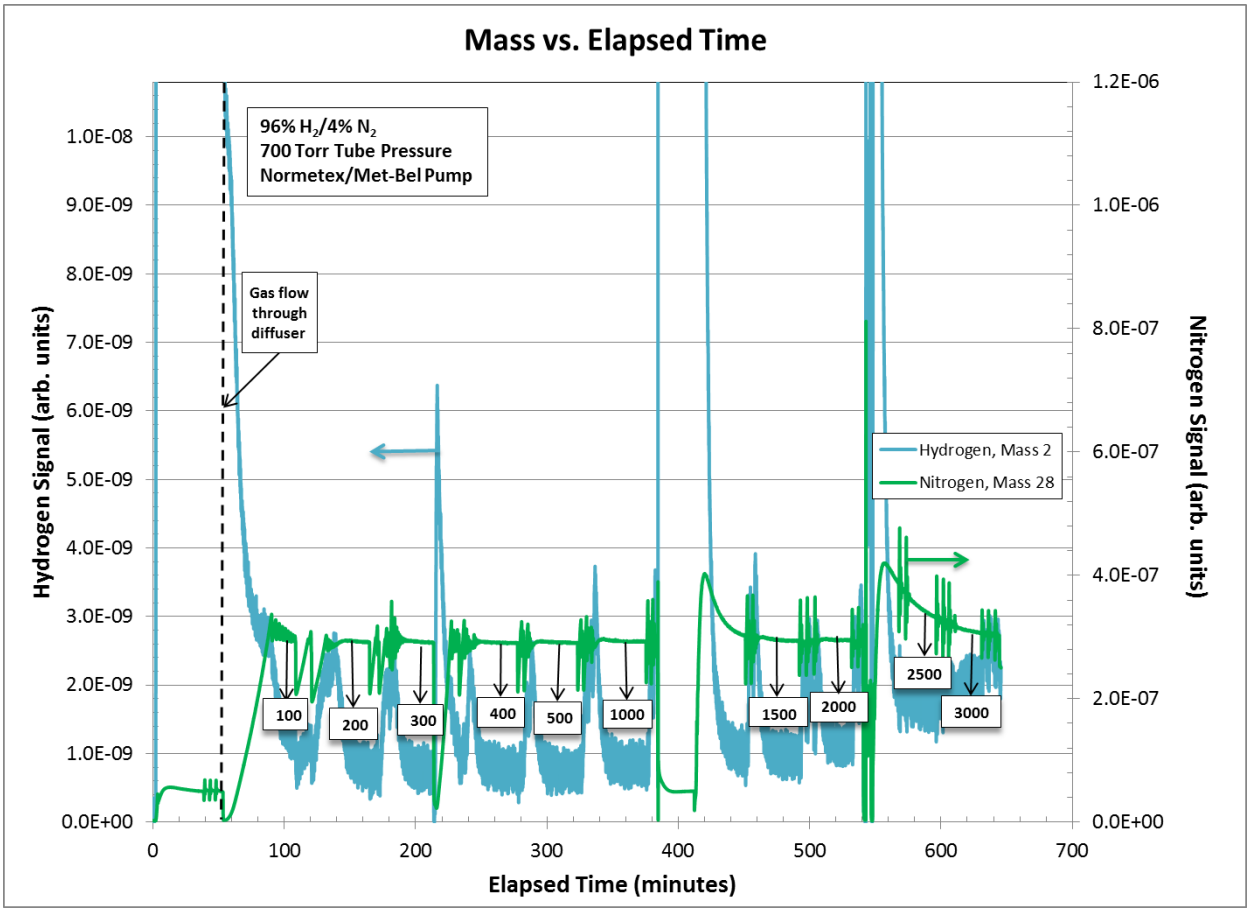

Figure 3.15: RGA Spectrum of 96% H_2/4% N_2 Gas Mixture at Various Flow Rates using a Normetex/Met-Bel pump combination

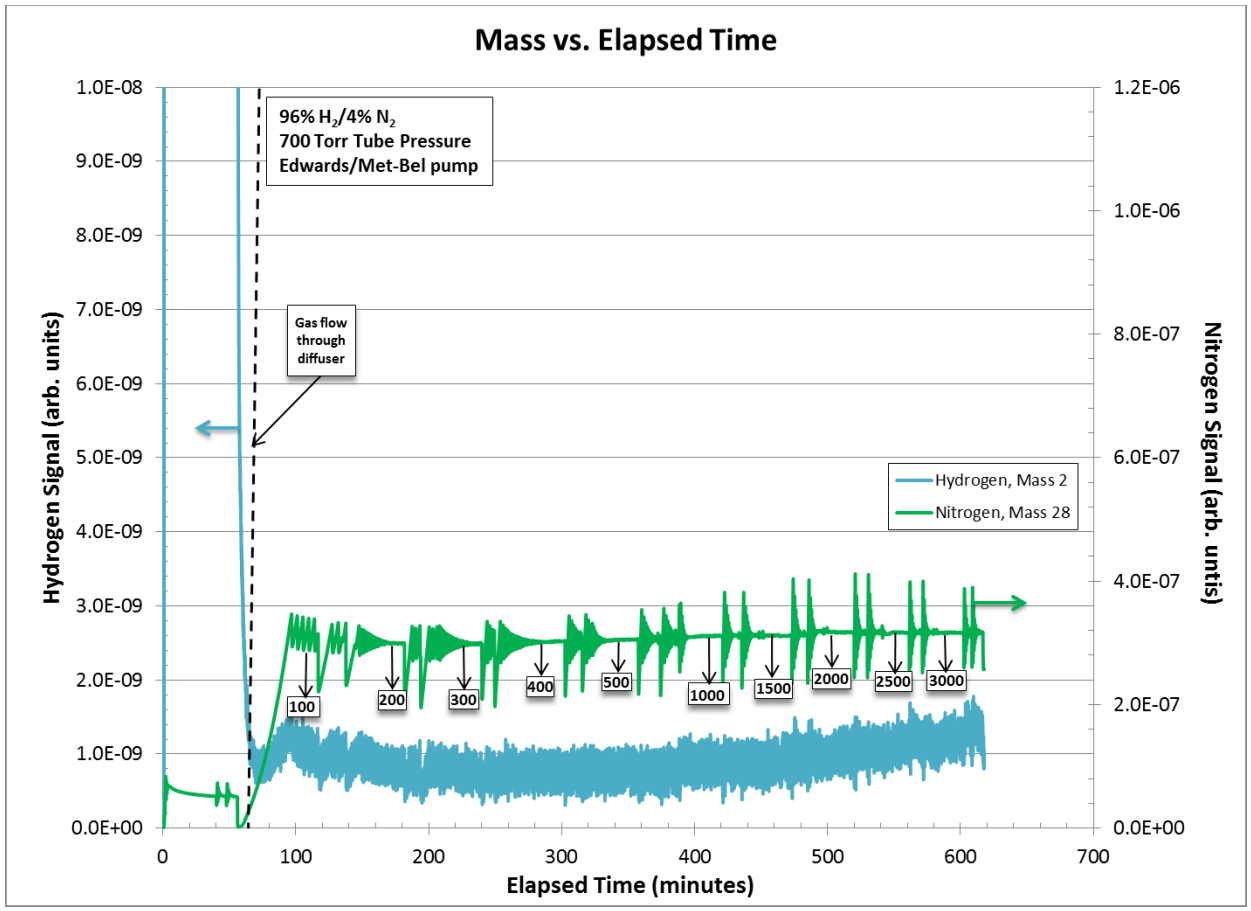

Figure 3.16: RGA Spectrum of 96% H_2/4% N_2 Gas Mixture at Various Flow Rates using an Edwards/Met-Bel pump combination

An RGA spectrum of a 50% H_2/50% N_2 mixture at various feed flow rates using the two pump configurations are shown in Figure 3.17 and Figure 3.18. The hydrogen signal is barely detectable on the RGA display at total feed flow rates up to 1500 sccm. At feed flow rates above 1500 sccm, a stepwise increase in the hydrogen signal is observed for both cases.

It is somewhat surprising that nearly all of the hydrogen is permeating through the Pd-Ag tubes for the 96% H_2/4% N_2 composition up to a flow rate of 3000 sccm, but with the 50% H_2/50%N_2 mixture nearly 100% permeation is only achieved up to 1500 sccm. This may be attributed to a blanketing effect caused by the nitrogen or to the small residence time through the diffuser. At higher flow rates, the residence time through the Pd-Ag tubes may not be long enough for all of the hydrogen molecules to permeate through the Pd-Ag membrane. In the presence of the nitrogen, some of the hydrogen is not able to diffuse through the nitrogen molecules and reach the Pd-Ag membrane. The region around 1000 sccm is determined to be the breakthrough region, where H_2 begins to be detected in the bleed stream.

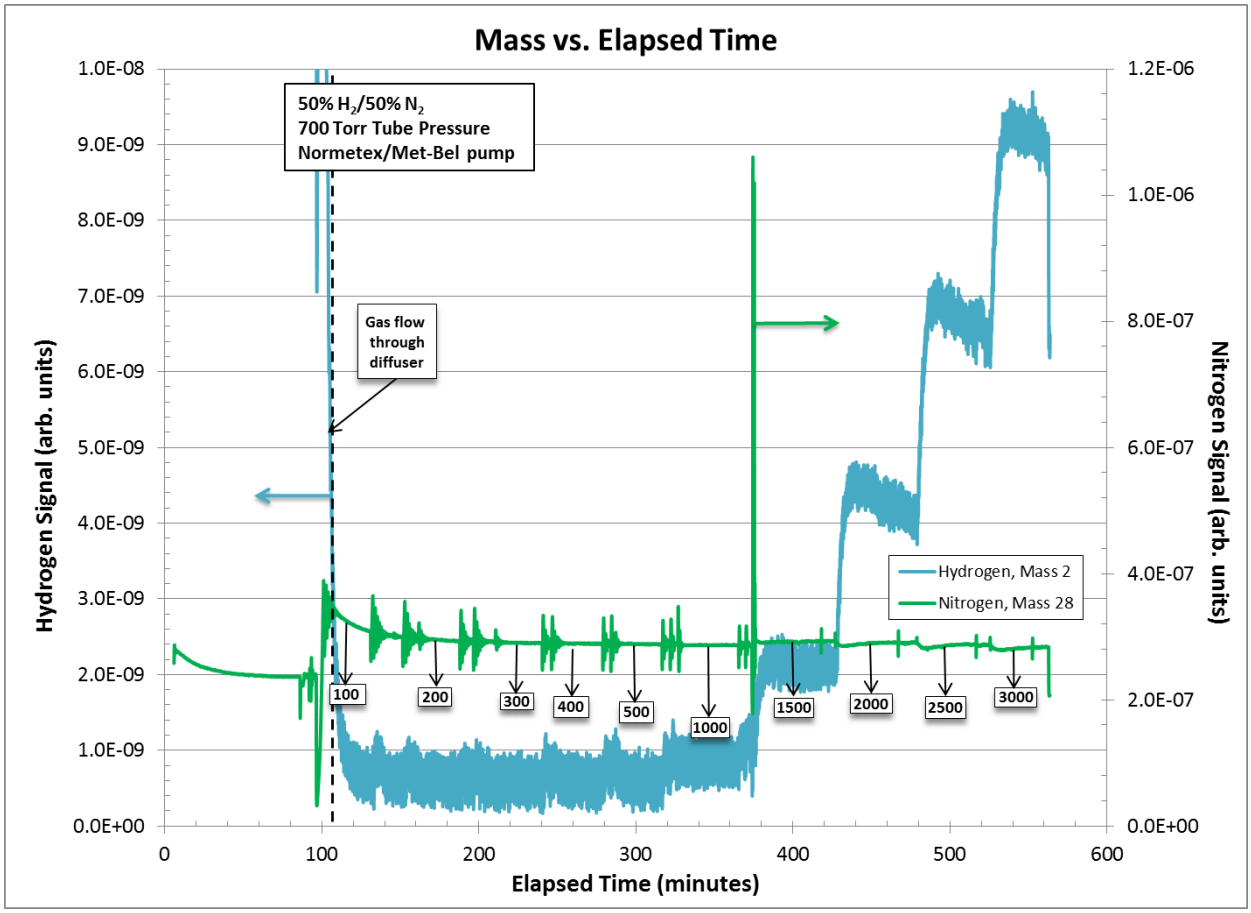

Figure 3.17: RGA Spectrum of 50% H_2/50% N_2 Gas Mixture at Various Flow Rates using an Normetex/Met-Bel pump combination

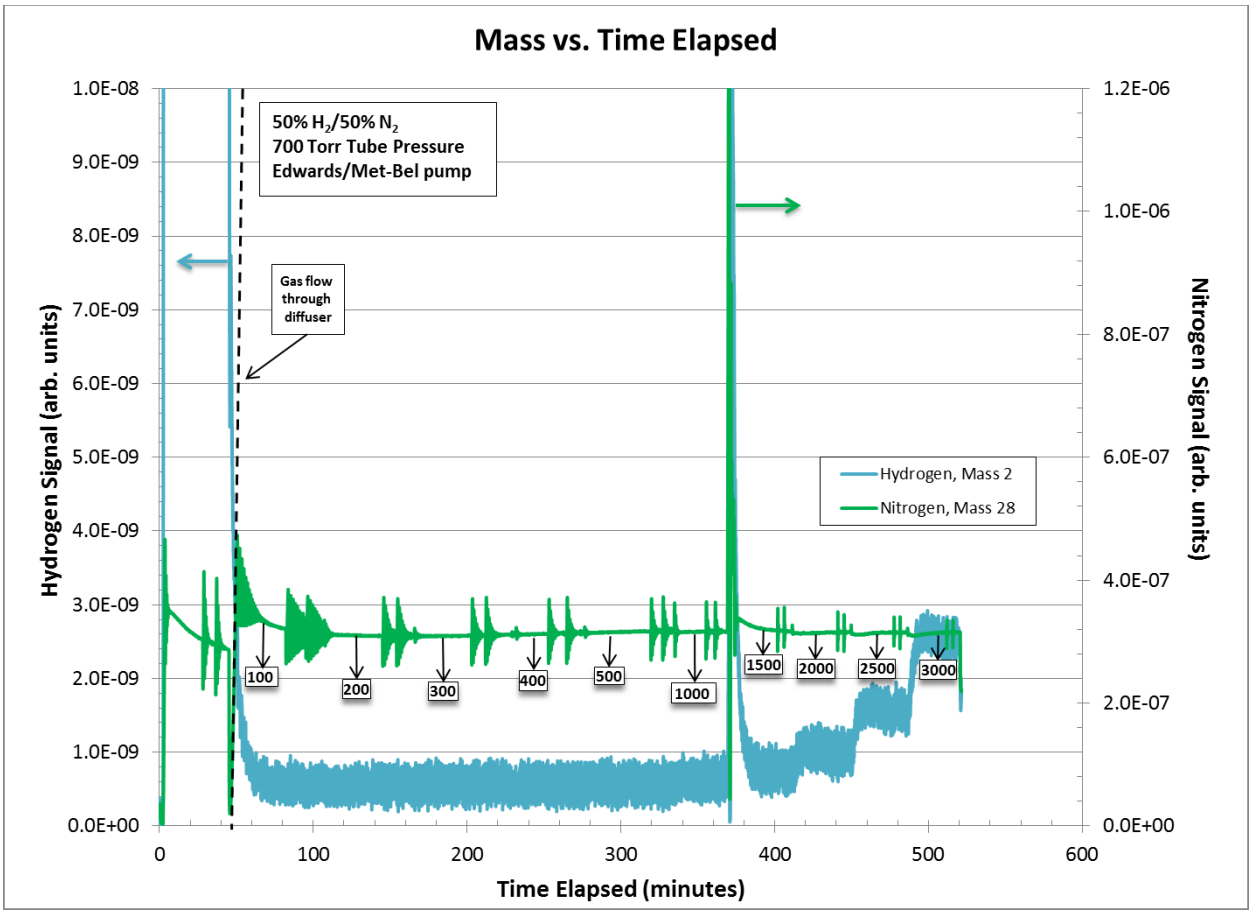

Figure 3.18: RGA Spectrum of 50% H_2/50% N_2 Gas Mixture at Various Flow Rates using an Edwards/Met-Bel pump combination

An RGA spectrum of a 2% H_2/98% N_2 gas mixture at various feed flow rates is shown in Figure 3.19 and Figure 3.20. The hydrogen and nitrogen signals remain relatively steady as the feed flow rates are increased. The hydrogen in the bleed stream is barely detectable in the RGA, indicating that very little hydrogen is present in the bleed stream. With a composition of only 2% H_2 in the feed stream, it would be expected that only a small amount of hydrogen would be evident in the bleed stream. The large spike at the time interval of 300 minutes correlates to the point at which pressure regulators were switched to the higher range. PV1A was used for bleed flow rates of up to 500 sccm, while PV2B was used for bleed flow rates above 500 sccm. As the flow rate is increased, there is a very gradual increase in the H_2 signal and a corresponding decrease in the N_2 signal.

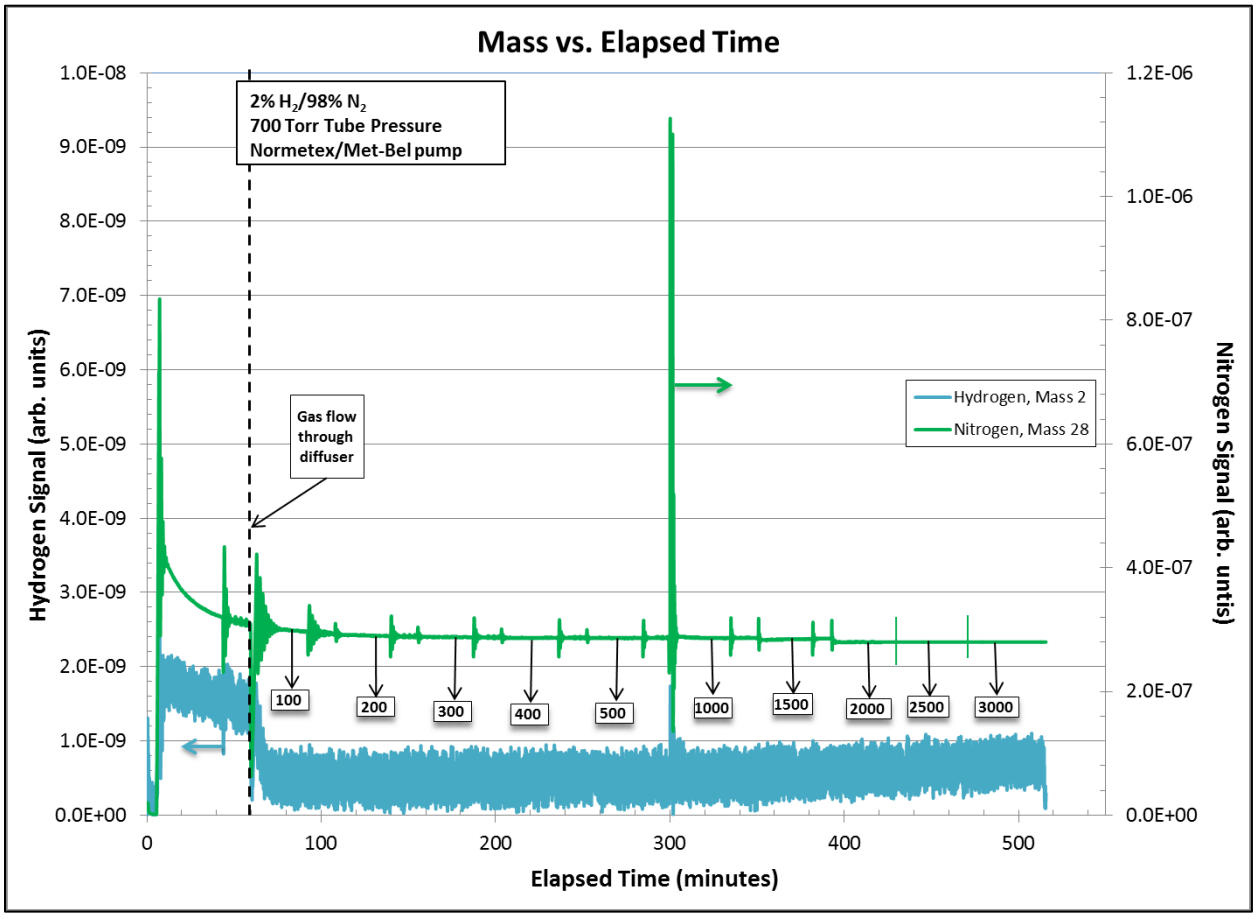

Figure 3.19: RGA spectrum of 2% H_2/98% N_2 Gas Mixture at Various Flow Rates using a Normetex/Met-Bel pump combination.

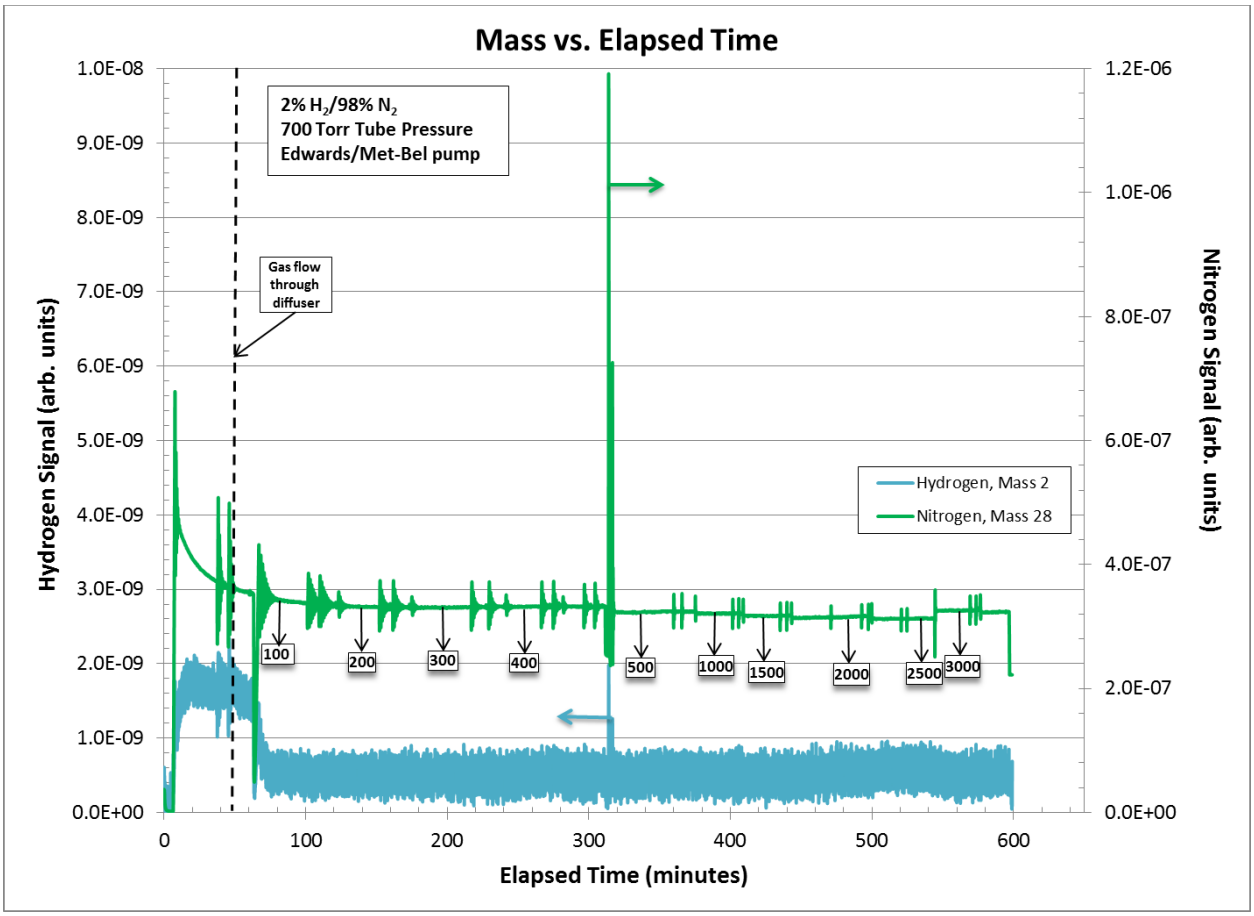

Figure 3.20: RGA Spectrum of 2% H_2/98% N_2 Gas Mixture at Various Flow Rates using an Edwards/Met-Bel pump combination.

3.2.5 Micro Gas Chromatograph Analysis

The composition of the bleed stream at each of the flow rates and compositions was analyzed using micro gas chromatography in an attempt to determine the amount of hydrogen that was present in the bleed stream. A calibration of the micro-GC was completed using gas compositions ranging from 0.5% H_2 to 25% H_2 with the remainder balance being nitrogen. The results of the μ–GC calibration are presented in Figure 3.21. The data was fit with a polynomial expression to determine the hydrogen concentration in the bleed stream. The polynomial expression was found to have the lowest least squares value.

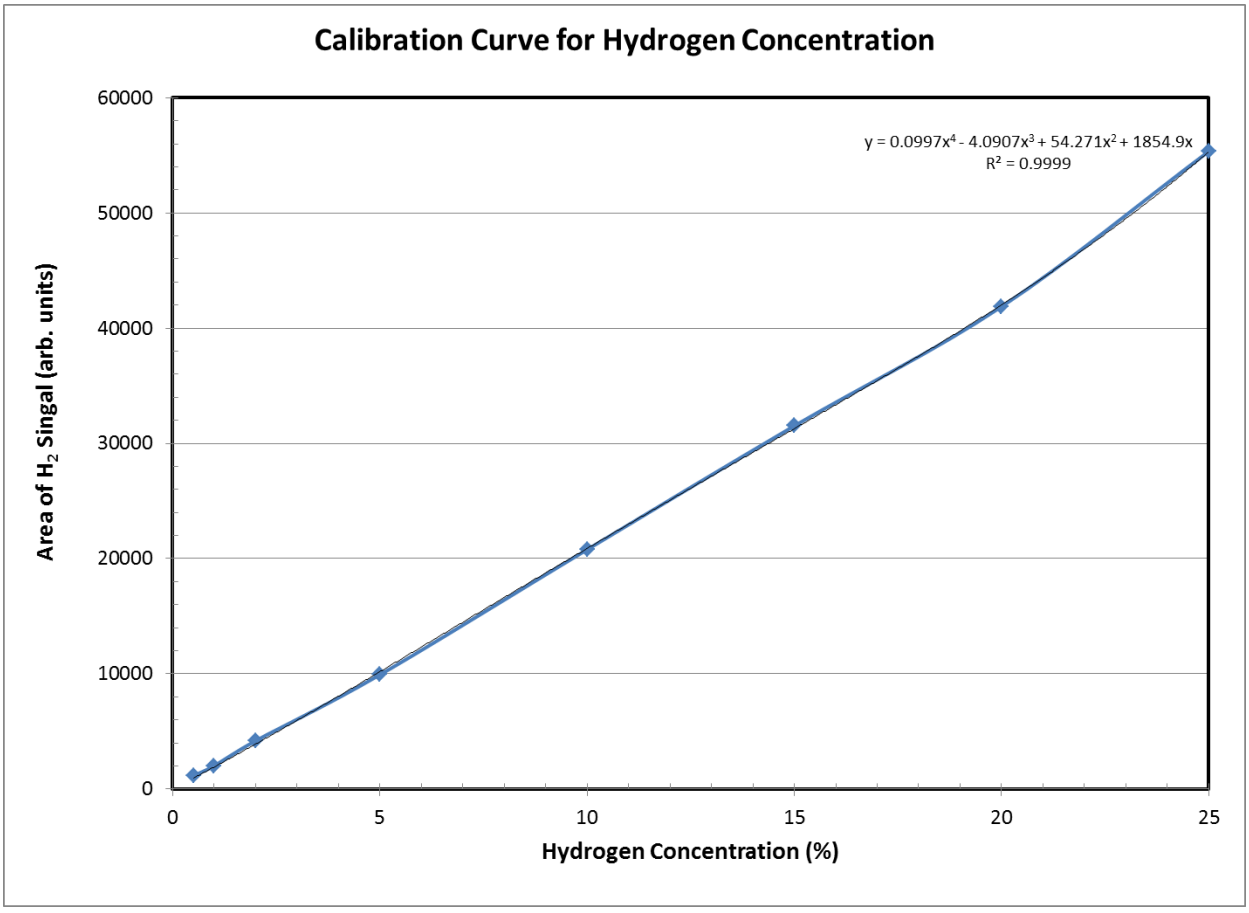

Figure 3.21: Calibration Curve for m-GC Analysis of Bleed Stream

The amount of hydrogen in the bleed stream versus the total feed flow rate is shown in Figure 3.22. Previous plots calculated from the flow controllers and flow meters have indicated that essentially all of the hydrogen is permeating for a gas composition of 96% H_2/4% N_2. The μ-gc analysis of the bleed stream ~~to determine the hydrogen concentration in the bleed stream~~ is more sensitive than the calculation from the flow controllers and flow meters.

The H_2 concentration in the bleed stream for the 96% H_2/4% N_2 ranges from 0.6% at 100 sccm to 1.6% at 3000 sccm. For the 50% H_2/50% N_2, the amount of hydrogen in the bleed stream ranges from 0.2% H_2 to 1.1% H_2 at total flow rates of 3000 sccm. The amount of H_2 in the bleed stream is less than 0.1 % up to a flow rate of 2000 sccm and then gradually increases to 0.3%. Based on the μ-GC measurements and the calculations of the amount of H_2 in the bleed stream from the flow controllers and flow meters, this prototype Pd-Ag diffuser does not meet the anticipated performance specifications.

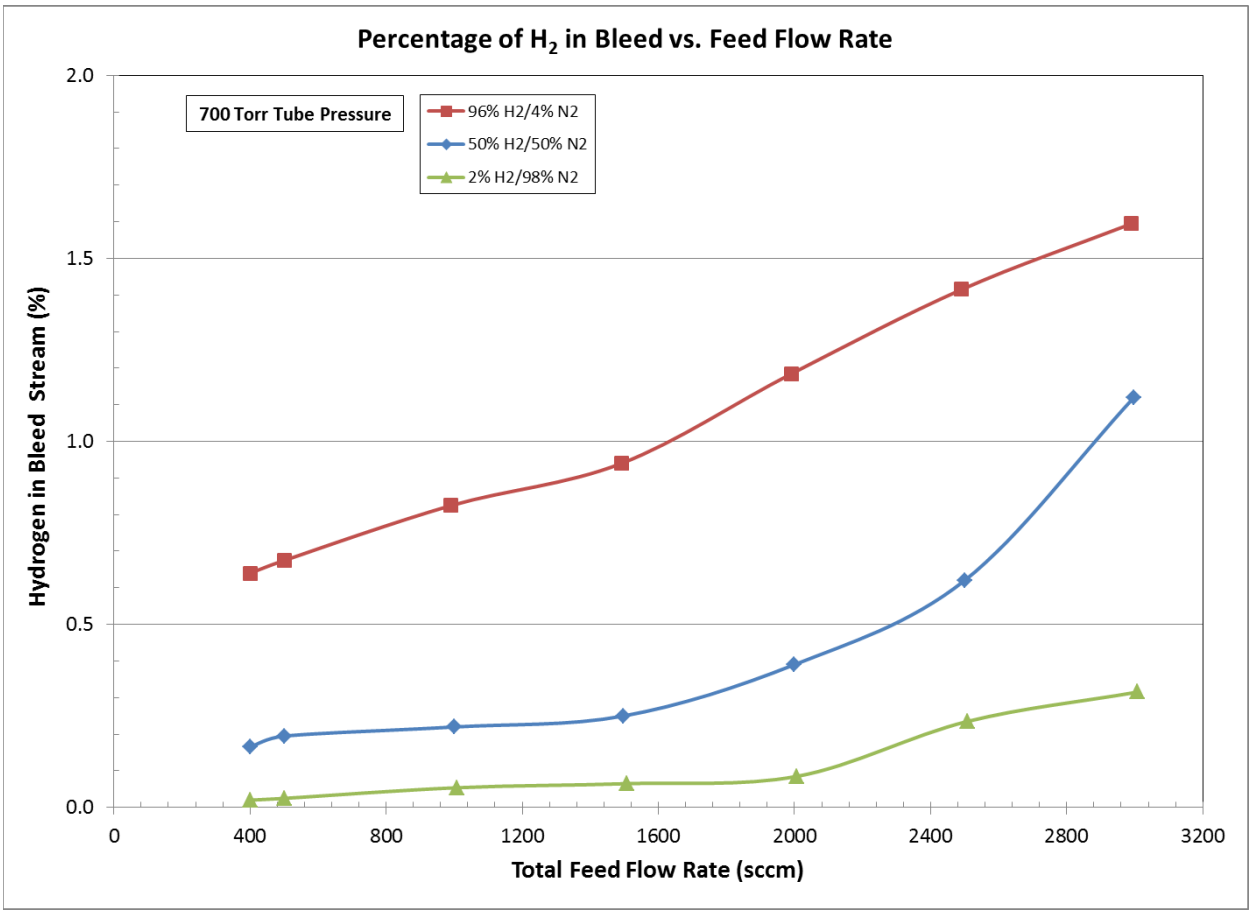

Figure 3.22: Amount of Hydrogen in the Bleed Stream at 700 Torr

4.0 Summary and Conclusions

Characterization tests on a new Pd-Ag diffuser design have been completed to gain an understanding into how the permeation of H_2 is affected by varying experimental test conditions. Testing of the diffuser involved varying the feed gas compositions, feed flow rates, internal tube pressuresand pump type configurations. The Pd-Ag diffuser was tested at 2% H_2/98% N_2, 50% H_2/50% N_2, and 96% H_2/4% N_2 at flow rates of 100 sccm to 3000 sccm and internal tube pressures of 700 and 1140 Torr.

The performance of the diffuser is characterized by the permeation of H_2 through the Pd-Ag membrane. The change in the permeation of H_2 through the membrane as the experimental parameters are varied is determined based on the location of the breakthrough point. For the 96% H_2/4% N_2 mixtures nearly all of the H_2 permeated through the Pd-Ag membrane. The breakthrough region for this particular case is likely well above a feed flow rate above 3000 sccm. However, the 50% H_2/50% N_2 gas composition results in nearly 100% permeation up to a flow rate of 1000 sccm and there is an increase in the amount of hydrogen remaining in the bleed stream at the higher flow rates. Blanketing of the permeation sites by nitrogen (or other non-hydrogen gas) is likely the cause for less than 100% H_2 permeation through the Pd-Ag membrane for the 2% H_2/98% N_2 and 50% H_2/50% N_2 mixtures. The very small residence time (<5 seconds at flow rates of 100 sccm) through the Pd-Ag diffuser may also have an impact on the hydrogen permeation

The results presented here indicate that this prototype Pd-Ag design is not acceptable for low to mid hydrogen gas compositions. Even at high hydrogen compositions, there is hydrogen remaining in the clled stream as indicated by the μ-GC measurements, RGA measurements, and the total feed and permeate flow measurements. Typical tritium purification systems can be expected to see a range of hydrogen isotope concentrations and this particular prototype diffuser is more suitable for process streams containing high concentrations of hydrogen isotopes. This prototype Pd-Ag diffuser design meets the design criteria for normal operations in a typical tritium purification system for fusion applications where there is high (> 90%) hydrogen isotopes and low non-hydrogen species in the process gas. However, during off-normal operations, where there may be low concentration of hydrogen isotopes and large amounts of non-hydrogen species, this diffuser design will not effectively remove the hydrogen isotopes from the process stream. If this particular diffuser design is used for tritium service, the hydrogen isotopes present in the bleed stream will need to be processed using additional techniques in order to maximize the tritium recovery and minimize the environmental discharges.

5.0 Recommendations, Path Forward or Future Work

Significant efforts should be undertaken to identify additional commercial vendors for Pd-Ag diffusers. It is of critical importance to identify, procure, and test different Pd-Ag designs that can perform well over a range of hydrogen isotope concentrations in the process gas.

6.0 Acknowledgements

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