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Evaluating All-Metal Valves for Use in a Tritium Environment

Levi R. Houk

Andrew N. Payton

September 2017

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EXECUTIVE SUMMARY

In the tritium gas processing system, it is desired to minimize polymer components due to their degradation from tritium exposure (beta decay). One source of polymers in the tritium process is valve components. A vendor has been identified that manufactures a valve that is marketed as being made from all-metal construction. This manufacturer, Ham-Let Group, manufactures a diaphragm valve (3LE series) that claims to be made entirely of metal.

SRNL procured twelve (12) Ham-Let diaphragm valves for characterization and evaluation. The characterization tests include identification of the maximum pressure of these valves by performing pressure and burst tests. Leak tests were performed to ensure the valves do not exceed the acceptable leak rate for tritium service. These valves were then cycled in a nitrogen gas and/or vacuum environment to ensure they would be durable in a process environment. They were subsequently leak tested per ASTM protocol to ensure that the valves maintained their leak tight integrity. A detailed material analysis was also conducted to determine hydrogen and tritium compatibility.

The Ham-Let diaphragm valves were initially characterized by pressure and burst tests. These valves exceeded the pressure specified by the manufacturer. The pressure tests supplied nitrogen to either the inlet or the outlet of the valve until the valve leaked. Approximately 3000 psig was required on the inlet side of the valve to lift the diaphragm and approximately 1000 psig was needed on the outlet side to lift the diaphragm. These pressures are much higher than the 150 psig rating stated by the manufacturer. The difference in maximum values for the inlet and the outlet pressure is due to the non-symmetric design of the valve. The burst test, applying pressure to both the inlet and the outlet, resulted in valve failure at over 16,600 psig. The only damage observed from this destructive test was a slightly dimpled diaphragm.

The leak rate specified by this experiment was 4.0×10^{-9} STD cc He/sec. A baseline leak test was performed on ten valves, including two valves that were initially pressure tested along with eight new valves. All but one of the valves passed the initial leak test. The seven new valves that passed the first leak test were then cycled in a nitrogen or nitrogen/vacuum gas environment. The valve cycle test protocol follows the ASTM "*Standard Test Method for Determination of Cycle Life of Automatic Valves for Gas Distribution System Components*." The valves have been cycled 200,000 times to date, with only one valve failing at 20,000 cycles. Initial failure analysis suggests the diaphragm was bent and would not seal. Whether this failure occurred from cycling or due to debris in the system is not yet determined. Two other valves failed at intermediate cycle test points, but recovered to meet leak rate requirements during further cycling when filters were added to remove any potential debris from the system.

One Ham-Let diaphragm valve was sectioned using Electrical Discharge Machining (EDM). While cutting the valve, an unknown material was observed right above the diaphragm. Further material testing by infrared spectroscopy showed that the unknown material was a polyimide material. Energy dispersive X-ray spectroscopy (EDS) showed that the diaphragms are made of two different alloys; one for the wetted diaphragm and another for the two non-wetted diaphragms. The materials were later confirmed by the manufacturer.

All wetted parts of Ham-Let diaphragm valves are metal; the process gas does not touch the polymer. The valve exceeded the pressure limits by over six times the manufacturer's stated pressure. Cycle testing has proved that these valves are durable; exceeding the manufacturer's stated life of 100,000 cycles. After 200,000 cycles, only one valve has failed. These valves should receive further testing to ensure compatibility with hydrogen and tritium.

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LIST OF ABBREVIATIONS

ATR-FTIR	Attenuated Total Reflection Fourier-Transform Infrared Spectroscopy
EDM	Electrical Discharge Machine
EDS	Energy-dispersive X-ray spectroscopy
Mo-99	Molybdenum-99
psi	pounds per square inch – a unit of pressure
PTFE	Polytetrafluoroethylene or Teflon
SEM	Scanning Electron Microscope
SRNL	Savannah River National Laboratory
STD cc He /sec	One cubic centimeter of helium gas flow per second at 14.7 psi and 77˚F
UHMW-PE	Ultra-High-Molecular-Weight Polyethylene

1.0 INTRODUCTION

Tritium gas reacts with and degrades polymer components in the tritium gas processing system. Tritium is a radioactive isotope of hydrogen that decays to helium-3 with a half-life of about 12.3 years. The conversion of the neutron to proton in the nucleus gives off beta radiation [1].

One potential source of polymers in a tritium gas processing system is valves. Some valves contain a polymer piece that is wetted, or is in contact with the process fluid. Polytetrafluoroethylene (PTFE or Teflon), UHMW-PE (Ultra High Molecular Weight Polyethylene) or Vespel (polyimide) are commonly used polymers. PTFE is usually avoided in tritium applications; Vespel is generally preferred over PTFE [2]. The fluoro- groups in PTFE can react to the tritium gas and create a vapor of either TF (tritium fluoride) or HF (hydrofluoric acid)**.** This vapor is very damaging in that it can corrode stainless steel.

Material properties are affected in the presence of tritium. This can be evidenced by the fact that the colors of some polymers change when in the presence of tritium, such as PTFE and UHMW-PE that are commonly used in valves. The polymer components turn from a whitish color to a noticeably darker color over several months [3-5]. This physical change may cause the polymer to become brittle, forming particulates that break apart into small pieces which may result in clogging equipment [6].

If polymers were eliminated from the valve's components, the valves could be in service for longer periods of time. More benefits, other than cost reduction from less frequent maintenance and replacing valves, include the decreased potential for byproduct gasses and particulate generation.

1.1 The Ham-Let All-Metal Valve

One manufacturer that produces an all-metal diaphragm valve that could be used in the tritium gas process has been identified. Ham-Let Group manufactures a diaphragm valve in which all wetted components are made of metal, meaning any fluid passing through the valve will only "wet" or touch metal. Figure 1-1 shows the selected diaphragm test valve, model number 3LES4C-FV-U [7]. This ultra clean valve boasts a small footprint, which is ideal for applications in gloveboxes. This valve measures less than 4" in width and height and its maximum operating pressure stated by the manufacturer is 150 psi with a working temperature range of -10 to 150˚C. The materials listed are a body of 316L stainless steel, cobalt-chromium-nickel alloy diaphragms, an actuator button set of 304 stainless steel and a button holder made from ASTM 630 H900 stainless steel. This valve has female ¼" VCR fittings. The actuation device itself is made from aluminum. From the diagram the only wetted components appear to be the body and the diaphragms. The "as-received valves", as seen in Figure 1-2, shows the ¼" VCR fittings of the valve. This test did not utilize limit switches on the valve, though the option to include limit switches is available.

3LE SERIES

COMPACT METAL SEAT MODEL

Metal Diaphragm Valves

The highest-ranking grade of compact models from the Ultra-Clean Valve Series are made according to UHP specifications. It is the ultimate in metallic diaphragm-operated valves with resins completely removed from their gas-contact areas. Their minimized valve internal volumes also best recommended them for use in liquid source supply applications.

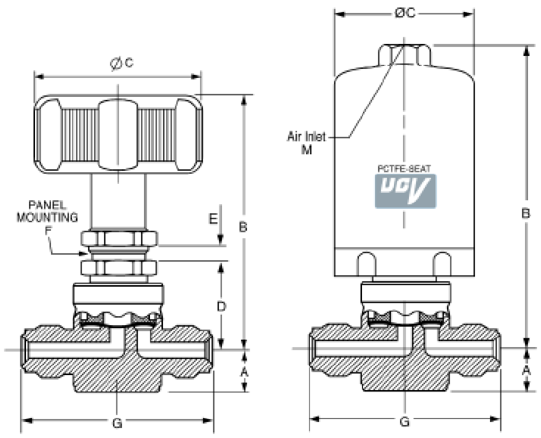
- Compact designs for minimum footprint.
- High-speed replacment of fluids in a gas or liquid state.
- Electropolished surfaces.

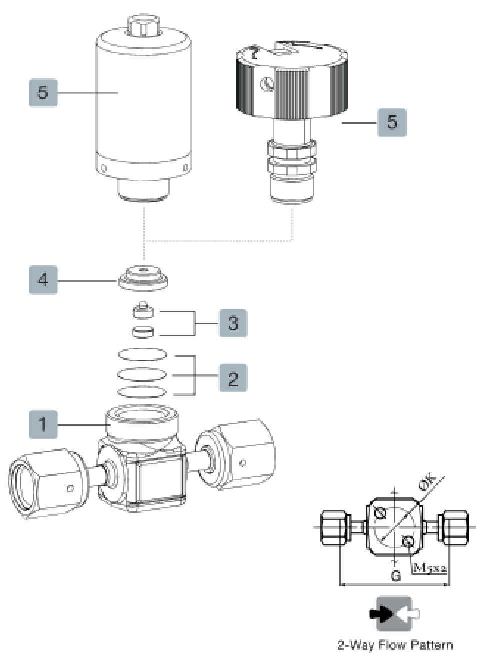
For details, please contact one of our field representatives.

ULTRA CLEAN VALVES

STANDARD CONFIGURATION DIMENSIONS

Part Number/ep	Size	End Connection	A	B	C	D	E	F	G	K	M
3LES2R-BV-U	1/8	Male HTC®	8	(51)	30	23	(4)	15	41	15	
3LES4R-W-U	1/4	Extended ButtWeld	11	(52)	30	24.5	(4)	17	47	17	
3LES4C-BW-U	1/4	Short ButtWeld	11	(86.2)	32	24.5	(4)	17	44.4	17	Rc1/8
3LES4C-FV-U	1/4	Swivel Female HTC®	11	(86.2)	32				66	17	Rc1/8





SPECIFICATIONS

Size	Pressure	Temp.	Cv	Leak Rates	
				Inboard	Across Seat
1/8	1MPa (150 psi)	-10 to 150°c	0.05	$3X10^{-12}$ pa•m³/sec Helium	$1X10^{-9}$ pa•m³/sec Helium
1/4			0.1		

STRUCTURE

	Parts	Material
1	Body	Stainlecc cteel, 316L Var or Vim/Var[1]
2	Diaphragm	Co-Cr-Ni Alloy
3	Act. Button Set	304 Stainless Steel
4	Act. Button Holder	Stainless Steel, ASTM 630 H900
5	Actuation Device	Aluminum

[1] Per SEMI F20-0305

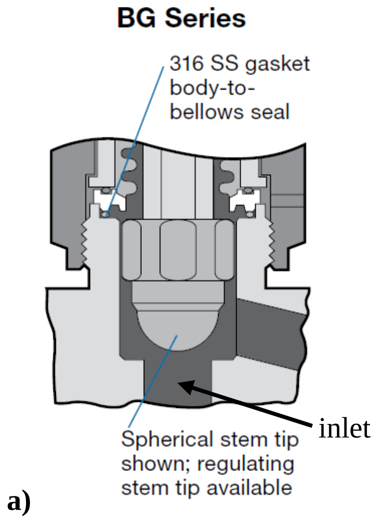
Figure 1-1. Product Description of All-Metal Valve from Ham-Let [7]

Figure 1-2. Pictures of As-Received Ham-let Valve

Comparing a typical metal bellows Swagelok valve [8], Figure 1-3a, to that of the Ham-Let diaphragm valve, Figure 1-3b, shows some noticeable differences. First, the Swagelok valve has a stem tip that sits in the inlet stream of the valve. The Swagelok BG series bellows valve, Figure 1-3a, is shown in the open, or up, position. When the BG valve is closed, the stem tip seals with the valve body which stops the gas flow. For the Ham-Let valve, the diaphragm, indicated as the dark wave in Figure 1-3b, is shown with the valve in the closed position, or with the diaphragm down, sealing off gas flow. When the valve is open, the diaphragm is lifted, allowing for the gas to pass on through to the outlet, with the diaphragm preventing the gas from passing through the body of the valve.

1.2 <u>Valve Testing Scope</u>

This work begins to evaluate whether Ham-Let valves are suitable for operation in a tritium environment. The manufacturer’s specifications are first verified to provide baseline data for the working conditions of the valve to ensure that the valve will safely operate in a gas process system.



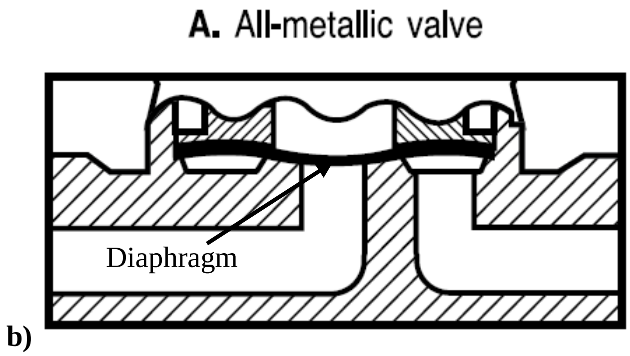


Figure 1-3. a) Typical Swagelok BG Series Bellows Valve, with a Stem Tip [8] b) Typical Design of a Ham-Let All-Metal Diaphragm Valve [7]

2.0 EXPERIMENTAL PROCEDURE

Twelve Ham-Let valves were purchased for testing and identified in Table 2-1. Testing was performed in four functional areas: Pressure and Burst Testing, Leak Testing, Cycle Testing, and Material Analysis. The test matrix used is shown in Table 2-1 below. Numbers designate the order in which testing was performed. All instrumentation and components used for testing measurements met SRNL quality assurance for M&TE.

A note about Table 2-1, a different nomenclature was used for the valves that were cycled and for the valves that were pressure and burst tested. Valves that were cycled are test valves (TV) and the valves that were pressure and burst tested are valves (V).

Table 2-1. Ham-Let Valve Test Matrix

Valve ID	Pressure/Burst Test	Leak Test	Cycle Test	Materials Analysis
TV00		1		
TV01		1	2	3
TV02		1	2	
TV03		1	2	
TV04		1	2	
TV05		1	2	
TV06		1	2	
TV07		1	2	
V01	1			2
V02	1	2		3
V03	1	2		
V04	1,2			

2.1 <u>Pressure and Burst Testing</u>

2.1.1 Pressure Test

Four of the as-received Ham-Let valves (Valves V01, V02, V03, and V04) were taken to the High Pressure Lab at SRNL for seat pressure testing. Two valves were pressurized from the inlet, two valves were pressurized from the outlet. Nitrogen was applied to one side of the valve with an evacuated line and a pressure transducer on the other side. Gas is slowly supplied to the valve, until the diaphragm of the valve is slightly lifted and the pressure gauge on the vacuum side registers a pressure increase.

2.1.2 Burst Pressure Test

After pressure testing, Valve V04 was burst tested by the High Pressure Lab at SRNL. Figure 2-1 shows the valve burst pressure testing set-up where both the inlet and outlet were pressurized with gas. Nitrogen was slowly added to both the valve inlet and the outlet, until the valve ultimately failed. Valve failure was indicated by a rapid pressure drop.

Figure 2-1. Set-up for Burst Test with Ham-Let Valve Attached

2.2 <u>Leak Testing</u>

The inlet, outlet, and body of each valve was leak tested using a calibrated helium leak detector. All of the leak tests were performed with 150 ± 5 psig of helium, which is the upper limit of the valve's pressure rating. This value was chosen to make sure the valves met maximum pressure limits.

2.2.1 Inlet Seat Leak Test

Inlet seat leak testing was performed by pressurizing the inlet side of the valve while the outlet was under vacuum. Leaks across the inlet seat were measured by the leak detector.

2.2.2 Outlet Seat Leak Test

Outlet seat leak testing was performed by pressurizing the outlet side of the valve while the inlet was under vacuum. Leaks across the outlet seat were measured by the leak detector.

2.2.3 Body Leak Test

The body leak test was performed by opening the valve, meaning the diaphragm is up and gas can pass through. Then gas was applied to one side and then the other side was then capped off. Any leakage from the body was measured.

2.3 <u>Cycle Testing</u>

The valve cycle test manifold (Figure 2-2) was designed to simultaneously test four scenarios in nitrogen or under vacuum. For all tests, nitrogen was supplied at 40-45 psig and vacuum was at 0.5 torr or less. The four different testing environments are:

1. Nitrogen/Nitrogen. The valves are cycled with nitrogen on both the inlet and outlet sides of the valves.

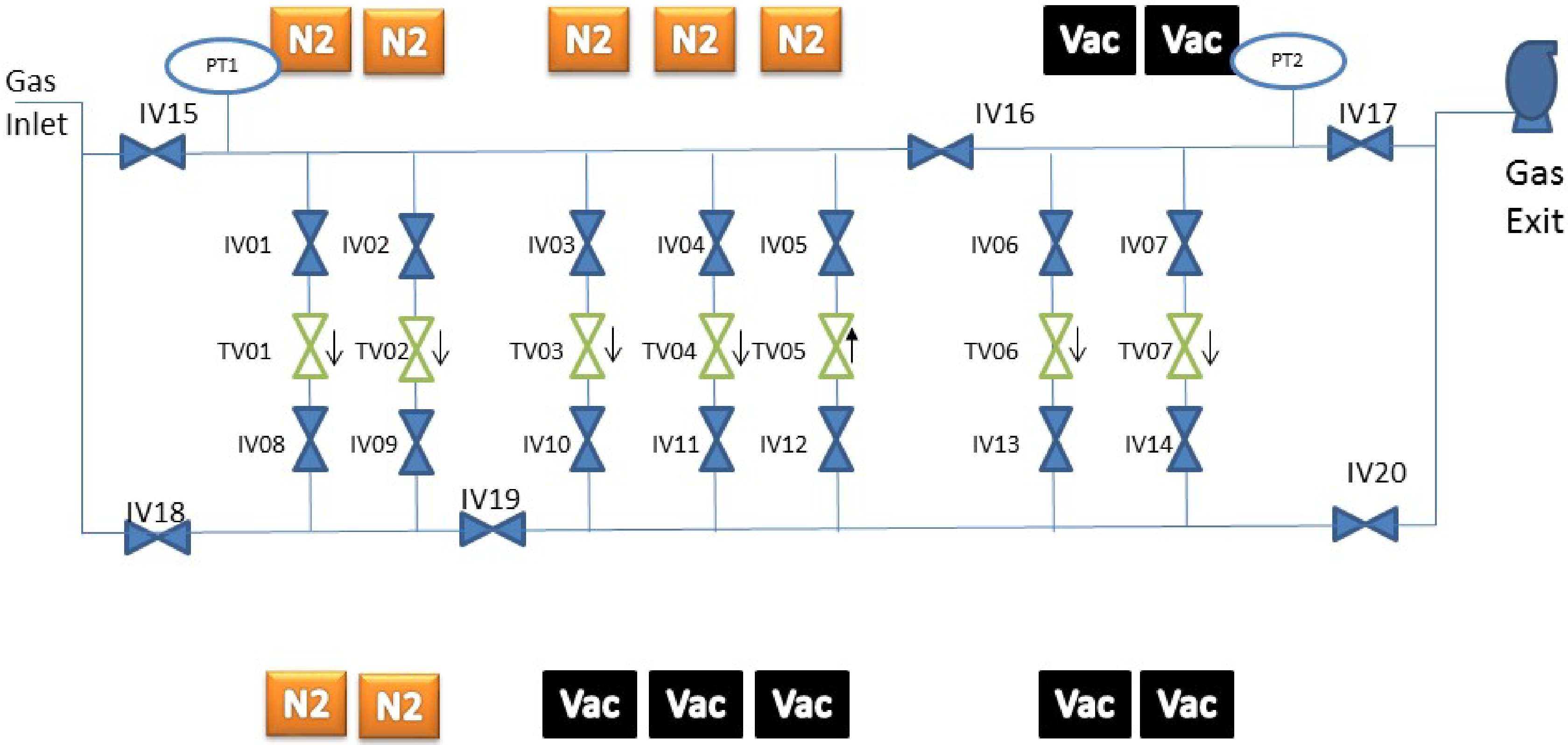


Figure 2-2. Diagram of Valve Cycling Set-Up

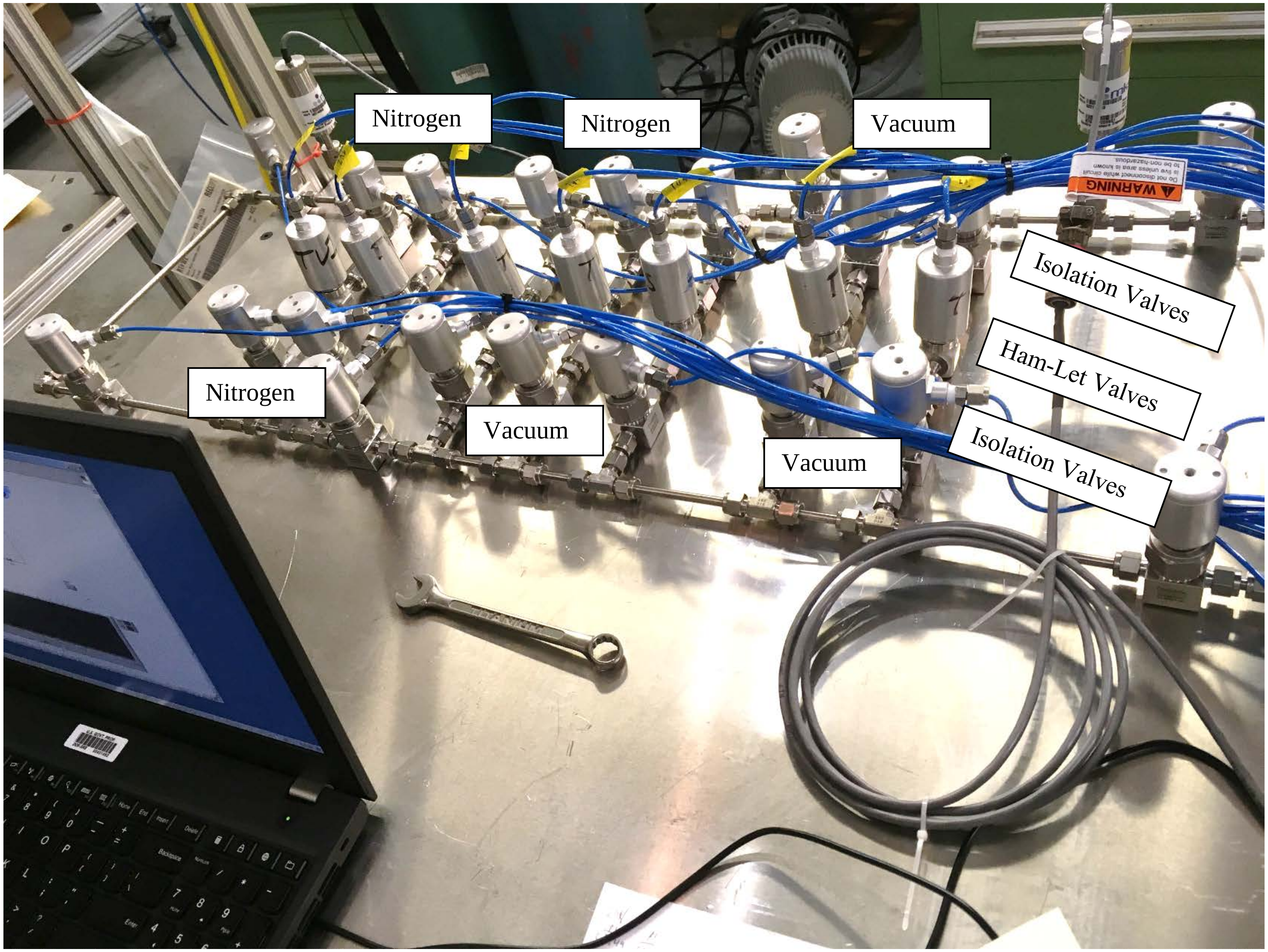


Figure 2-3. Picture of Valve Cycling Set-Up

2. Nitrogen/Vacuum. Nitrogen is supplied to the inlet of the valve, while the outlet of the valve is under vacuum. Each time the valve is cycled, the inlet must be re-pressurized with more nitrogen.
3. Vacuum/Nitrogen. Nitrogen is supplied to the outlet of the valve, while the inlet of the valve is under vacuum. Each time the valve is cycled, the outlet must be re-pressurized with more nitrogen.
4. Vacuum/Vacuum. The valves are cycled with a vacuum on both the inlet and outlet sides of the valves.

The ASTM standard, ASTM F1373 − 93 (2012) [9], only recommends testing condition 2, nitrogen/vacuum.

Simultaneous testing is achieved by using LabVIEW to control the valves. The isolation valves, labeled IVXX are used to control the test conditions.

Valve cycle testing follows the protocol of the ASTM standard, ASTM F1373 − 93 (2012), in which the valves are repeatedly cycled for 10% of their expected service life and then are leak tested. The manufacturer quotes these valves to have a cycle life of 100,000 cycles, so after every 10,000 cycles, the valves are leaked tested. For leak testing, the valves were removed from the manifold and transported to the High Pressure Lab. After the completion of the leak test, the valves are then connected back to the manifold for more testing until the valves were cycled 100,000 times. After the 100,000 cycles valve testing was accelerated to perform leak testing every 50,000 cycles.

Appendix A provides additional information on the LabVIEW program and for its control of the valve cycling. The pressure transducers are MKS model 870B, with a range of 0-100 psi. Nitrogen was supplied from liquid nitrogen tanks. The vacuum was supplied from an Edwards nXDS 15i Dry Scroll Pump. To control the automatic actuation of the valves, a FESTO pneumatic terminal block with Argon supplied at 100 psi was used.

The manifold, pictured in Figure 2-3, was assembled from ¼” 316L stainless steel tubing, connected with Swagelok compression fittings that were leak checked using a helium detector. The Ham-Let test valves were connected with VCR fittings. The vacuum exhaust was vented into a fume hood.

2.4 <u>Materials Analysis</u>

Valve structure was imaged optically, with X-rays (Figure 2-4), and with a Scanning Electron Microscope (SEM).

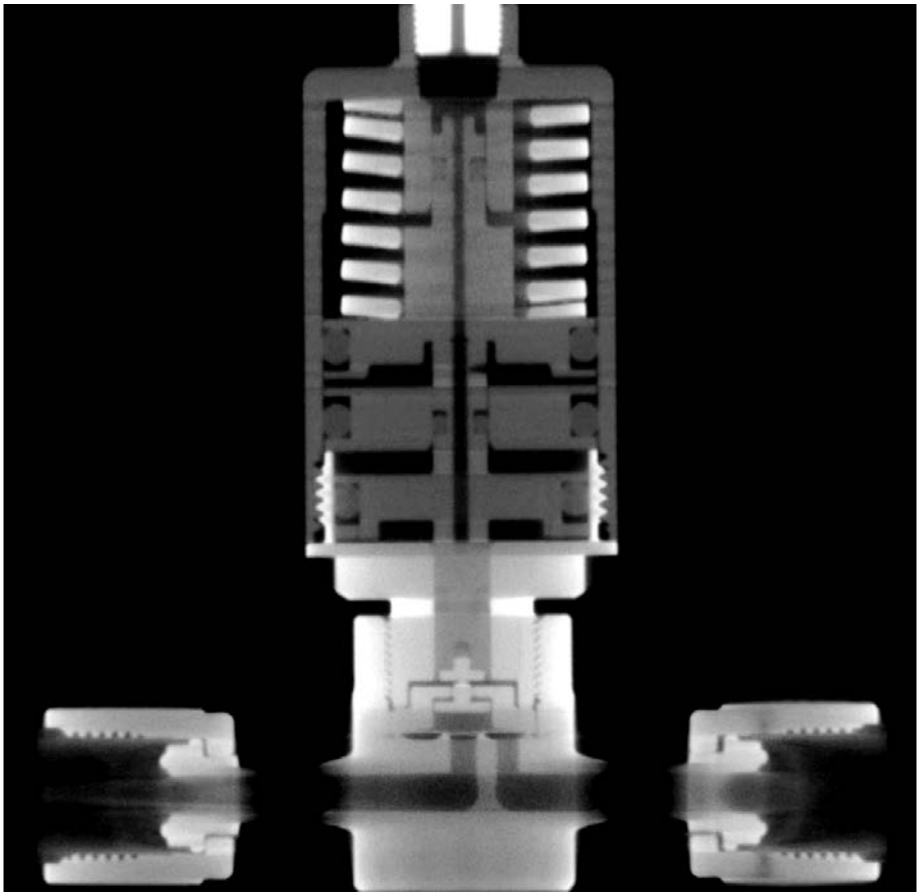


Figure 2-4. Digital Radiographed Valve

Valve materials were examined using Energy-dispersive X-ray spectroscopy (EDS) and Attenuated Total Reflection Fourier Transmission Infrared spectroscopy (ATR-FTIR). Samples were prepared using Electrical Discharge Machining (EDM) when necessary (Figure 2-5).

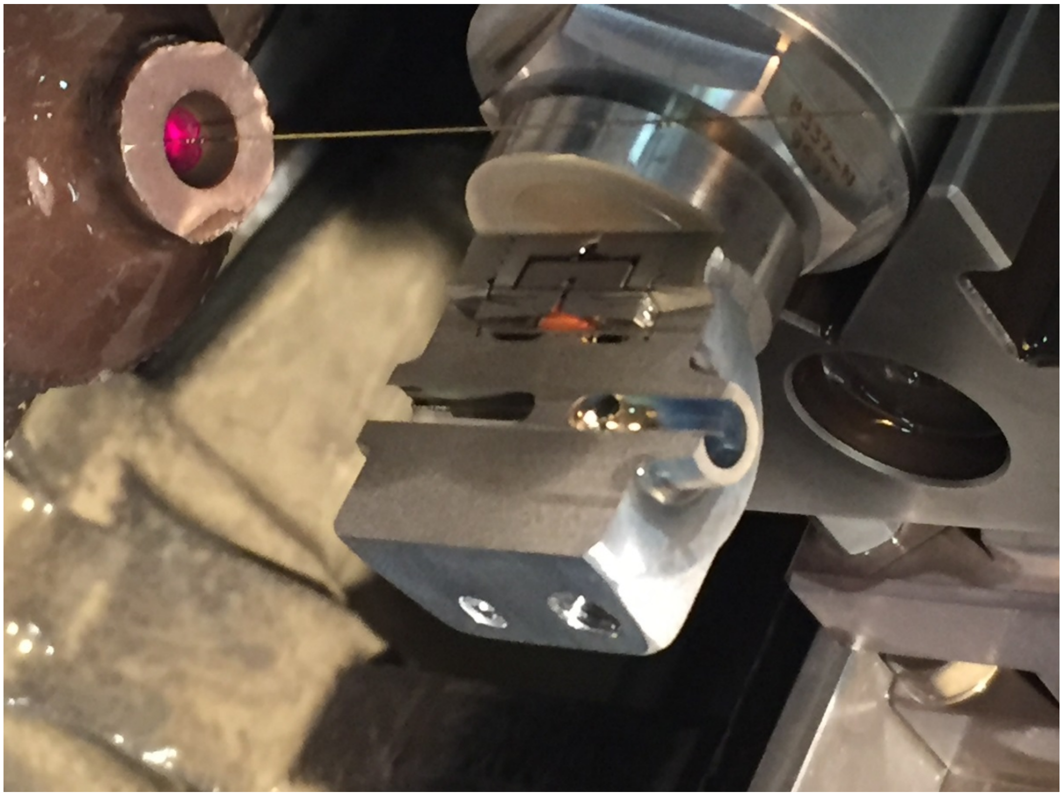


Figure 2-5. Ham-Let Valve in EDM Machine [10]

3.0 RESULTS AND DISCUSSION

3.1 Pressure and Burst Testing

3.1.1 Pressure Test

The first tests conducted on the Ham-Let valves were pressure and burst tests. These tests were conducted to verify that the valves met the manufacturer specifications and determined the safety envelope of the test manifold. The vendor-supplied maximum operating pressure of the valve is 150 psig Figure 3-1 depicts Ham-Let valve pressure testing. Test results are given in Table 3-1:

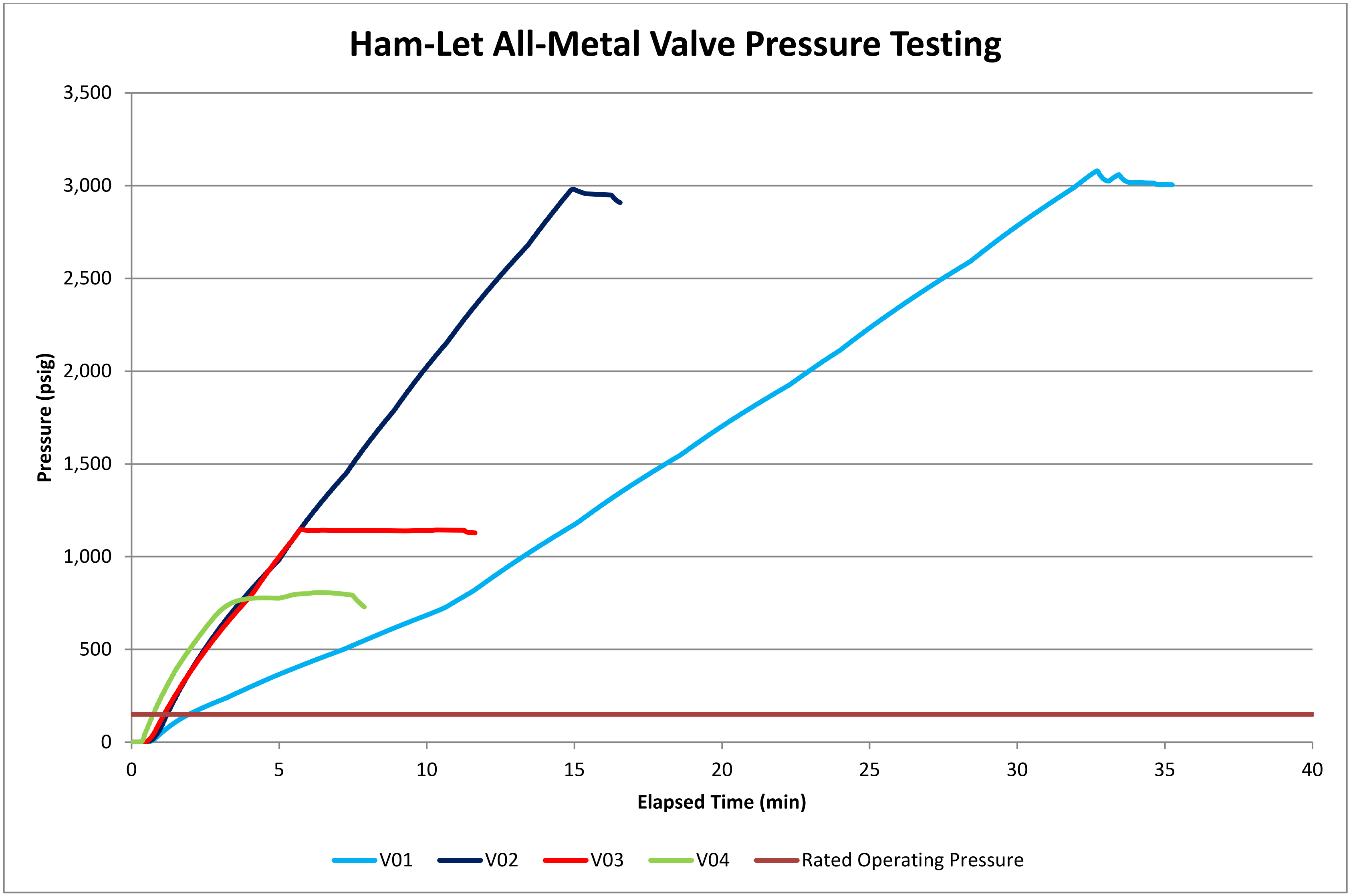


Figure 3-1. Graph of Pressure Testing of Ham-Let Valve

Table 3-1. Summary of Pressure and Burst Testing Pressures

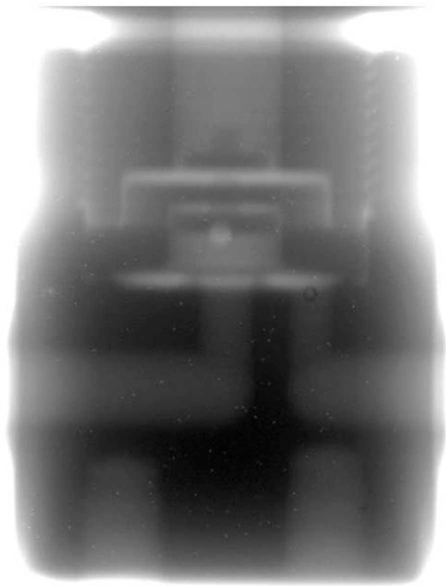
Valve ID	Pressure Test Inlet Pressure (psig)	Pressure Test Outlet Pressure (psig)	Burst Test (psig)
V01	3,059	vacuum	N/A
V02	2,981	vacuum	N/A
V03	vacuum	1,148	N/A
V04	vacuum	807	16,695

Examination of the valves after pressure testing, showed no obvious exterior damage Figure 3-2. One of the pressure tested valves was radiographed to determine whether any damage occurred internally. Radiographs (Figure 3-3) of the "as received" valve (a) and the pressure tested valve (b) show the asymmetry of the valve and do not reveal damage from pressure testing. This lack of symmetry is likely the source of the significant differences in the inlet or outlet pressures needed to cause the valve to leak.

a) 

b) 

Figure 3-2. Photos of Valves after Pressure Tests a) V01, Pressure on the Inlet b) V04, Pressure on the Outlet



a)

b)

Figure 3-3. Radiographs of a) "As-Received" Valve and b) Pressure Tested Valve

Figure 3-4, is another Ham-Let valve radiograph. The valve inlet is in the center of the of the valve body, the outlet is offset.

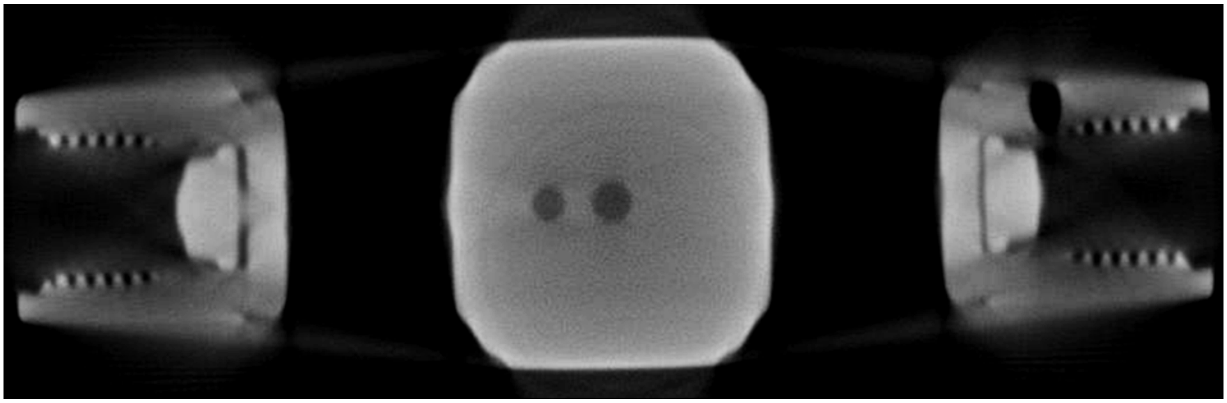


Figure 3-4. Digital Radiograph Top-Down Image Showing Asymmetric Valve

3.1.2 Burst Test

Valve V04 was selected for burst testing. Both the inlet and the outlet sides of the valve were pressurized with nitrogen until the valve failed near 16,695 psig. The test is depicted graphically in Figure 3-5.

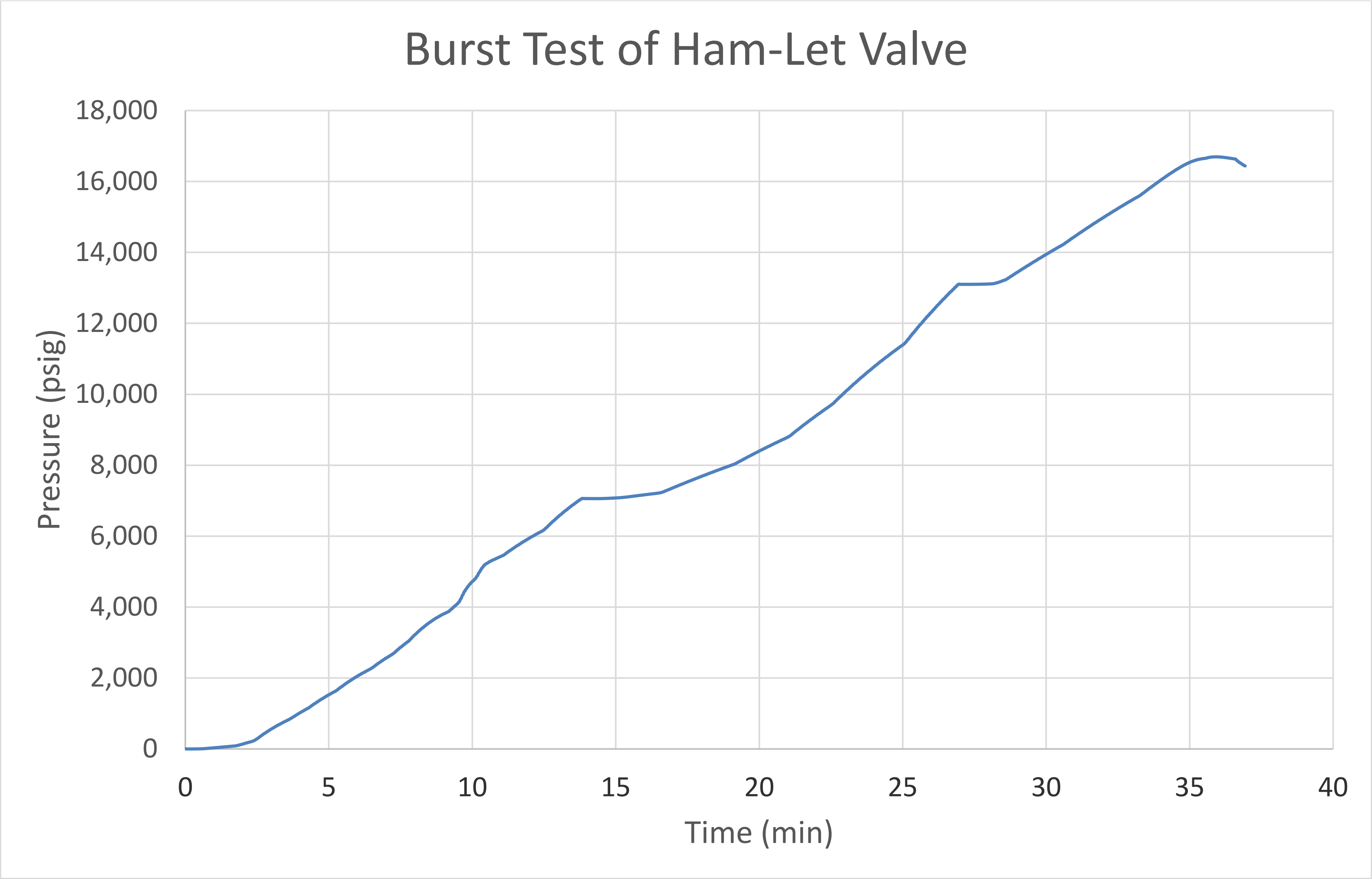


Figure 3-5. Graph of Burst Test of Ham-Let Valve

Photographs of the burst tested valve, Figure 3-6, revealed no exterior damage.

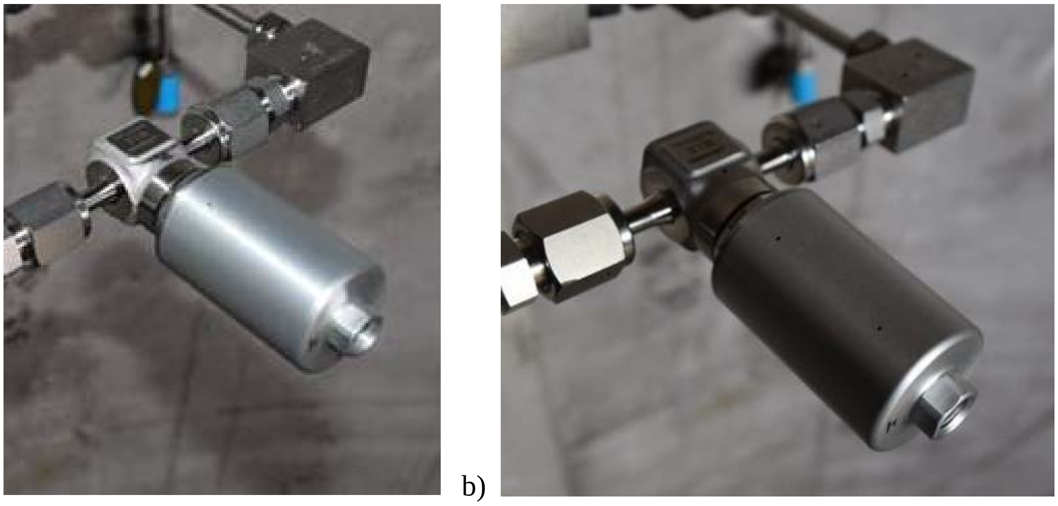
a)  b)

Figure 3-6. Burst Photographs a) Pre-Burst Test b) Post-Burst Test

The valve was then taken apart to inspect for damage to inner components. Photographs of the body and actuator, Figure 3-7, revels no exterior damage. Further investigation of the body components revealed damage to the diaphragms, Figure 3-8 and Figure 3-9.

Figure 3-7. Photographs of Burst Tested Valve Body and Actuator

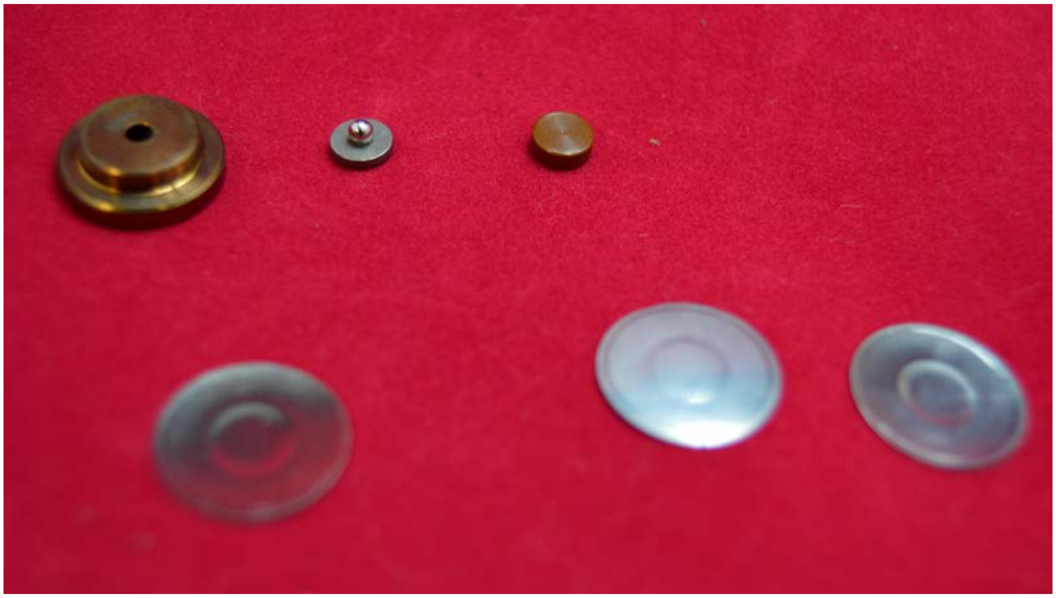


Figure 3-8. Photographs of Burst Tested Valve Inner Components

Figure 3-9. Photograph Comparing the Burst Tested Diaphragm (top) to the Pressure Tested Diaphragm (bottom)

Comparison of the burst test valve diaphragms (top) to diaphragms from a pressure tested valve (bottom) shows clear dimpling, Figure 3-9.

3.2 Leak Testing

Of the twelve valves initially ordered, ten were leaked tested (one was burst tested and one was cut apart for material testing). The leak acceptance criterion was 4.00 x 10^{-9} STD cc He/sec. Of the ten valves that were leak tested, one valve was shown to leak between the body and the actuator. Initial Leak test results (Table 3-2) were used as the baseline for the cycling test.

Table 3-2. Initial Leak Rates of Ham-Let Valves

Valve ID	Inlet Seat Leak Rate (STD cc He/sec)	Outlet Seat Leak Rate (STD cc He/sec)	Body Leak Test (STD cc He/sec)
TV00	$<7.9x10^{-10}$	$8.5x10^{-6}$	$8.6x10^{-6}$
TV01	$<8.4x10^{-10}$	$<8.4x10^{-10}$	*No body test performed
TV02	$<8.3x10^{-10}$	$<8.3x10^{-10}$	$<8.3x10^{-10}$
TV03	$<8.8x10^{-10}$	$<8.8x10^{-10}$	$<8.8x10^{-10}$
TV04	$<8.8x10^{-10}$	$<8.7x10^{-10}$	$<8.8x10^{-10}$
TV05	$<8.7x10^{-10}$	$<8.7x10^{-10}$	$<8.7x10^{-10}$
TV06	$<8.8x10^{-10}$	$<1.0x10^{-9}$	$<8.8x10^{-10}$
TV07	$<9.0x10^{-10}$	$<9.0x10^{-10}$	$<9.0x10^{-10}$
V02	$<8.4x10^{-10}$	$<8.2x10^{-10}$	*No body test performed
V03	$<8.3x10^{-10}$	$<8.3x10^{-10}$	*No body test performed

3.3 Cycle Testing

As described previously, cycle and leak testing was performed under four different conditions. Valves TV01 and TV02 were cycled with nitrogen on the inlet and outlet. Valves TV03 and TV04 were cycled with nitrogen on the inlet and vacuum on the outlet. TV05 valve was cycled with vacuum on the inlet and nitrogen on the outlet. Valves TV06 and TV07 were cycled with vacuum on the inlet and the outlet.

Leak rate test results are given in Table 3-3, valves that did not meet the required leak rate are highlighted in red. During testing, the LabVIEW program malfunctioned and data for 10,000 cycles and 30,000 cycles were not retrievable. TV01 passed the initial inlet seat leak test, 0 cycles, but failed at 20,000 cycles, Figure 3-10. After 50,000 cycles, two additional valves, TV02 and TV05, failed the inlet seat leak test. All of the failed valves were tested with nitrogen pressure on the outlet side. It is unknown whether this contributed to the valve failure. After TV02 and TV05 failed, Swagelok 0.003 µm filters, part number SS-SCF3-VR4-P-30, where installed in the nitrogen inlet line and at the outlet of isolation valves IV15 and IV18 and testing continued. TV01 testing continued without improvement. Both TV02 and TV05 passed leak testing after 60,000 cycles, suggesting the earlier leaks were a result of particulate contamination. It is unknown whether failure of TV01 was due to particulate contamination or valve failure.

One valve that was planned on being cycled failed the initial body test, TV00, this valve was never cycled as it was determined to have a leak in the actuator. Only one other valve had a leak from the body leak test, TV04 at 20,000 cycles, this was because this normally closed valve would not stay open for the duration on the test. The valve was still functioning and could be cycled, but it could not be body leak tested. Because of this, the High Pressure Lab determined that it would be best to only conduct outlet seat leak tests. For further cycling, all of the test valves have passed the outlet seat leak tests up to 200,000 cycles.

Table 3-3. Leak Rates of Cycle Tests

Test Valve	TV01 N_2/N_2		TV02 N_2/N_2		TV03 N_2/Vac		TV04 N_2/Vac		TV05 Vac/N_2		TV06 Vac/Vac		TV07 Vac/Vac	
		†Body Leak[1] / §Seat Outlet Leak [1]		†Body Leak[1] / §Seat Outlet Leak [1]		†Body Leak[1] / §Seat Outlet Leak [1]		†Body Leak[1] / §Seat Outlet Leak [1]		†Body Leak[1] / §Seat Outlet Leak [1]		†Body Leak[1] / §Seat Outlet Leak [1]		†Body Leak[1] / §Seat Outlet Leak [1]
0	< 8.4 x 10^{-10}	§< 8.4 x 10^{-10}	< 8.3 x 10^{-10}	†< 8.3 x 10^{-10}	< 8.8 x 10^{-10}	†< 8.8 x 10^{-10}	< 8.8 x 10^{-10}	†< 8.8 x 10^{-10}	< 8.7 x 10^{-10}	†< 8.7 x 10^{-10}	< 8.8 x 10^{-10}	†< 8.8 x 10^{-10}	< 9.0 x 10^{-10}	†< 9.0 x 10^{-10}
20,000	1.2 x 10^{-4}	†< 1.1 x 10^{-9}	< 1.2 x 10^{-9}	†< 1.2 x 10^{-9}	< 1.2 x 10^{-9}	†< 1.2 x 10^{-9}	1.7 x 10^{-9}	†1.4 x 10^{-8}	6.3 x 10^{-9}	†< 9.5 x 10^{-10}	< 9.5 x 10^{-10}	†< 9.5 x 10^{-10}	1.4 x 10^{-9}	†< 9.4 x 10^{-10}
40,000	9.2 x 10^{-4}	†< 8.4 x 10^{-10}	< 9.0 x 10^{-10}	†< 9.0 x 10^{-10}	< 8.6 x 10^{-10}	†< 8.6 x 10^{-10}	< 1.2 x 10^{-9}	†< 1.2 x 10^{-9}	< 1.0 x 10^{-09}	†< 1.0 x 10^{-09}	< 9.4 x 10^{-10}	†< 9.4 x 10^{-10}	< 1.0 x 10^{-9}	†< 1.0 x 10^{-9}
50,000	X	§X	8.6 x 10^{-4}	§1.6 x 10^{-9}	< 9.8 x 10^{-10}	§1.3 x 10^{-9}	1.7 x 10^{-9}	§2.6 x 10^{-9}	6.1 x 10^{-5}	§1.6 x 10^{-9}	1.5 x 10^{-9}	§2.3 x 10^{-9}	1.1 x 10^{-9}	§1.6 x 10^{-9}
60,000	NT	NT	< 1.0 x 10^{-9}	§< 1.0 x 10^{-9}	< 1.0 x 10^{-9}	§< 1.0 x 10^{-9}	< 1.0 x 10^{-9}	§< 1.0 x 10^{-9}	< 1.2 x 10^{-9}	§ 1.6 x 10^{-9}	< 1.2 x 10^{-9}	§1.6 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}
70,000	X	§X	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}	< 1.3 x 10^{-9}	§< 1.3 x 10^{-9}	< 1.3 x 10^{-9}	§< 1.3 x 10^{-9}
80,000	NT	NT	< 1.3 x 10^{-9}	§< 1.3 x 10^{-9}	< 1.3 x 10^{-9}	§< 1.3 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}
90,000	NT	NT	< 1.0 x 10^{-9}	§< 1.0 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}
100,000	NT	NT	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}
150,000	NT	NT	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}
200,000	NT	NT	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.3 x 10^{-9}	§< 1.3 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}	< 1.1 x 10^{-9}	§< 1.1 x 10^{-9}	< 1.2 x 10^{-9}	§< 1.2 x 10^{-9}

Notes:
[1] Leak Rates in STD cc He/sec
X Denotes the Leak Rate is Below Mass Spectrometer Detection
† Denotes Body Leak test was used
§ Denotes Seat Outlet Leak was used
NT Denotes Valve Not Tested

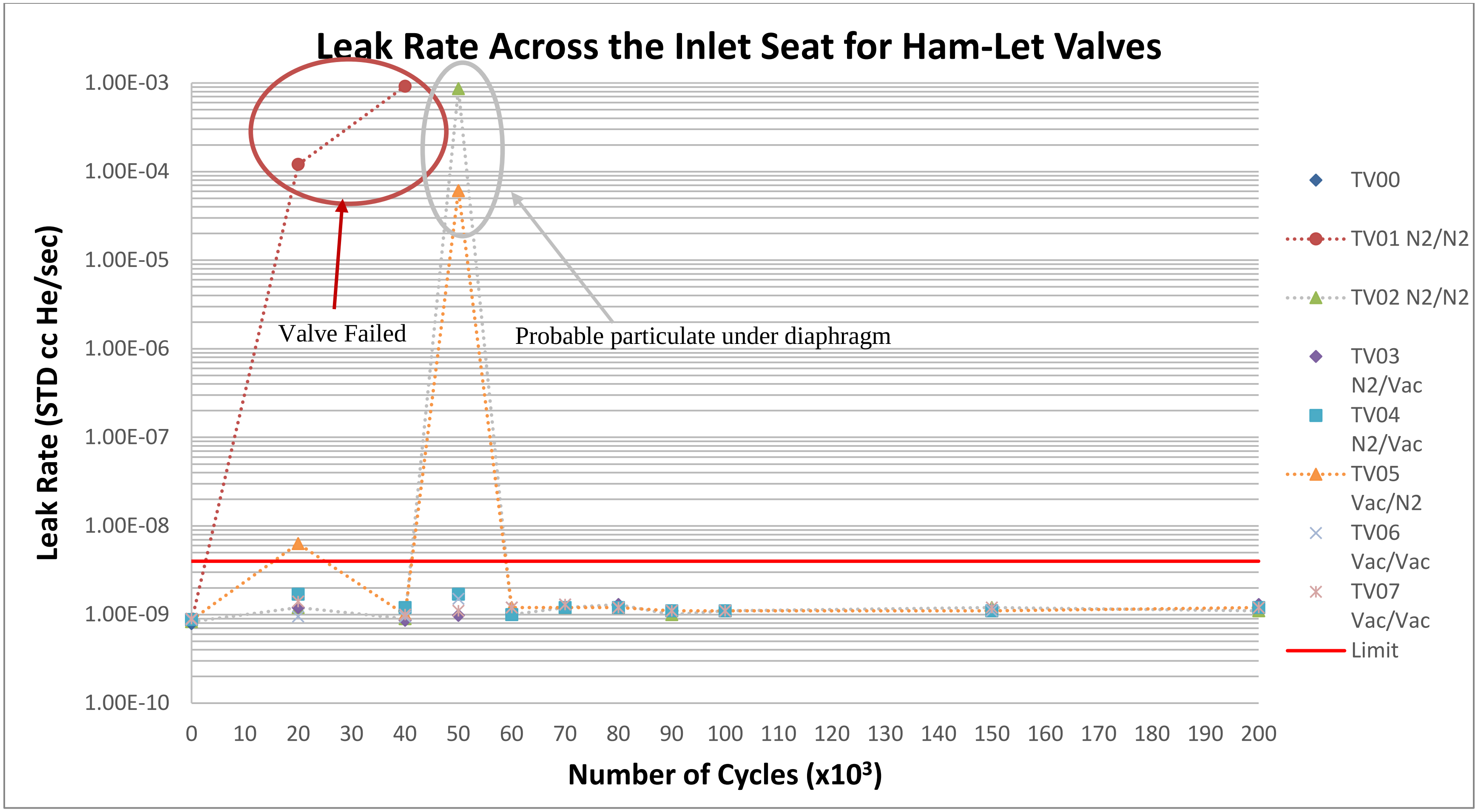


Figure 3-10. Graph of Leak Rates across the Inlet Seat of Ham-Let Valves

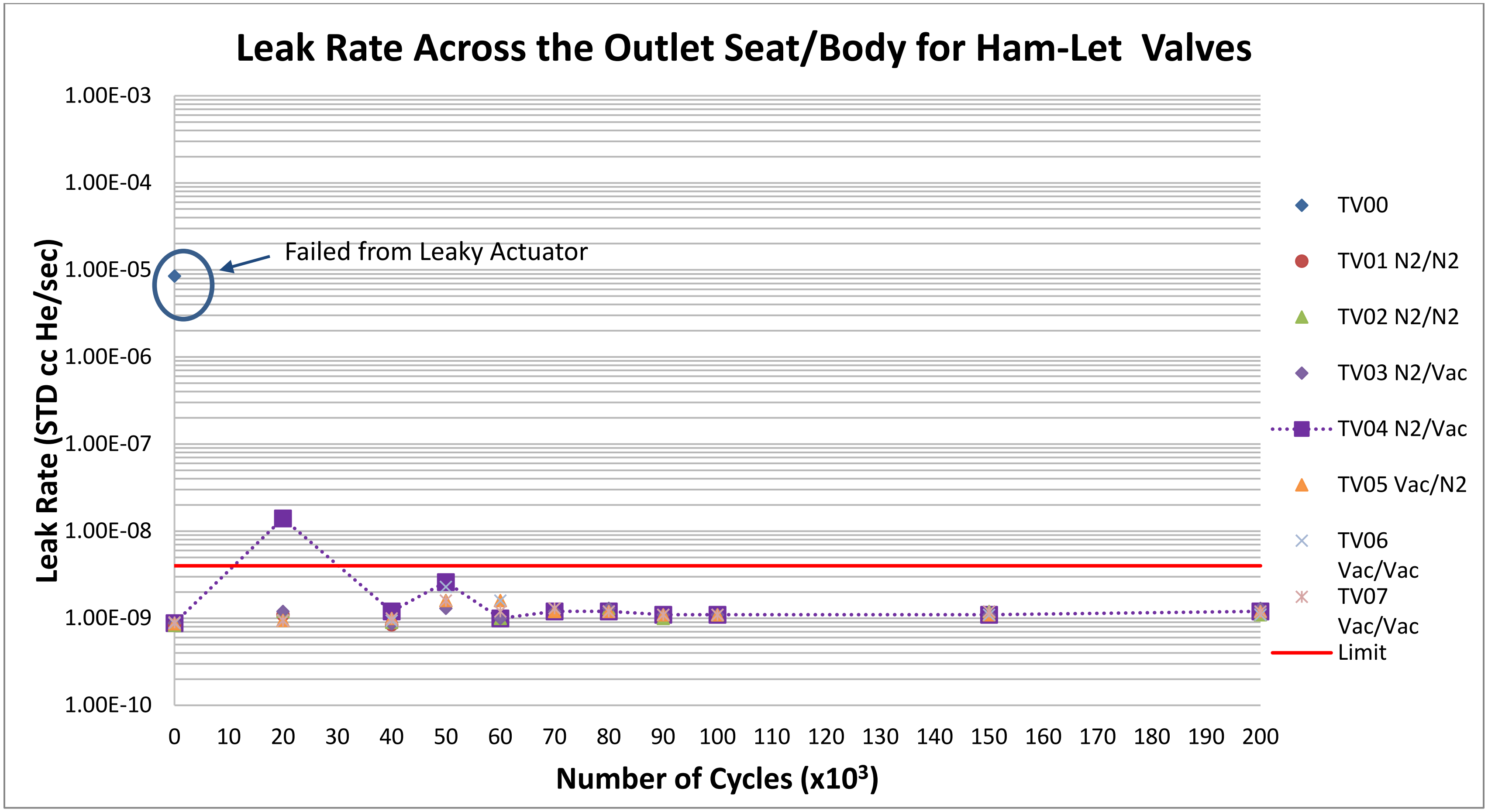


Figure 3-11. Graph of Leak Rates across the Outlet Seat or Body of Ham-Let Valve

3.4 Materials Analysis

Difficulty during Electrical Discharge Machining (EDM) of the valve indicated the possible presence of a non-conductive component [10]. Examination of the cut valve, Figure 3-12, revealed a small dark orange/brown disk.

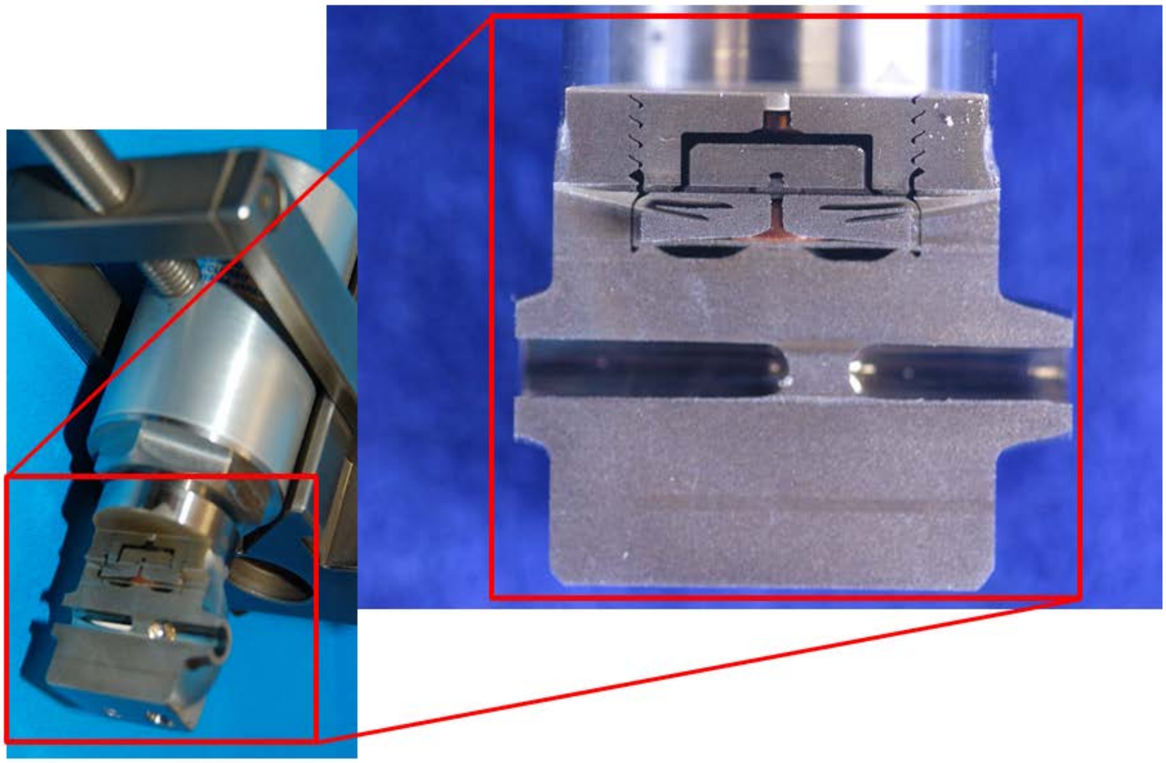


Figure 3-12. EDM Valve along with a detailed image of the cross section of the body

A closer look at the cross section of the body is given in Figure 3-13. The dark orange disk is easily seen. The Actuator Button Set rests between the Diaphragm, part 2 and the Actuator Button Holder, part 4. A different perspective, Figure 3-14, is of the same region as Figure 3-13, but was taken with an optical microscope.

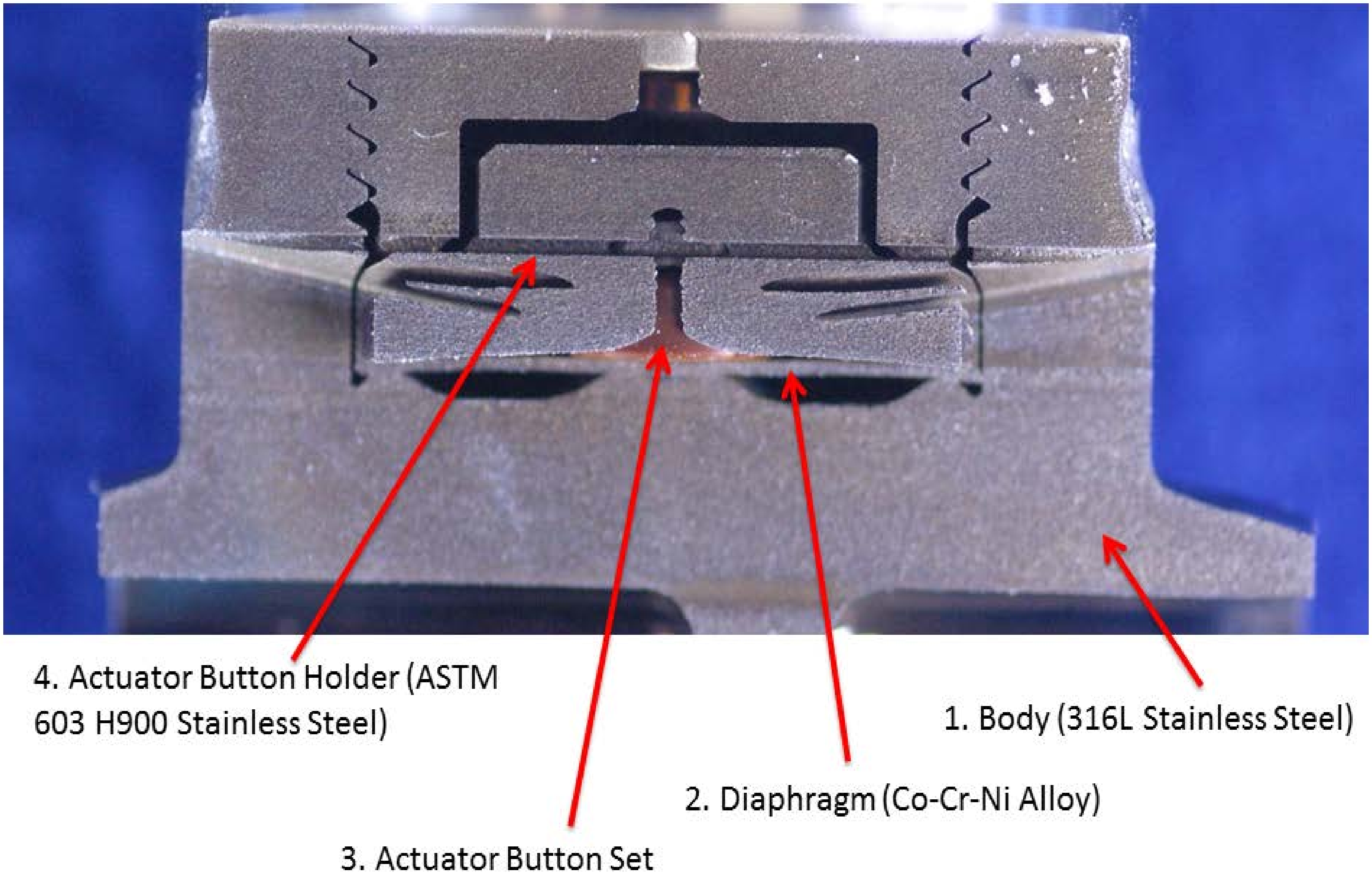


Figure 3-13. Detailed cross sectional image of valve body with main parts labeled and materials identified.

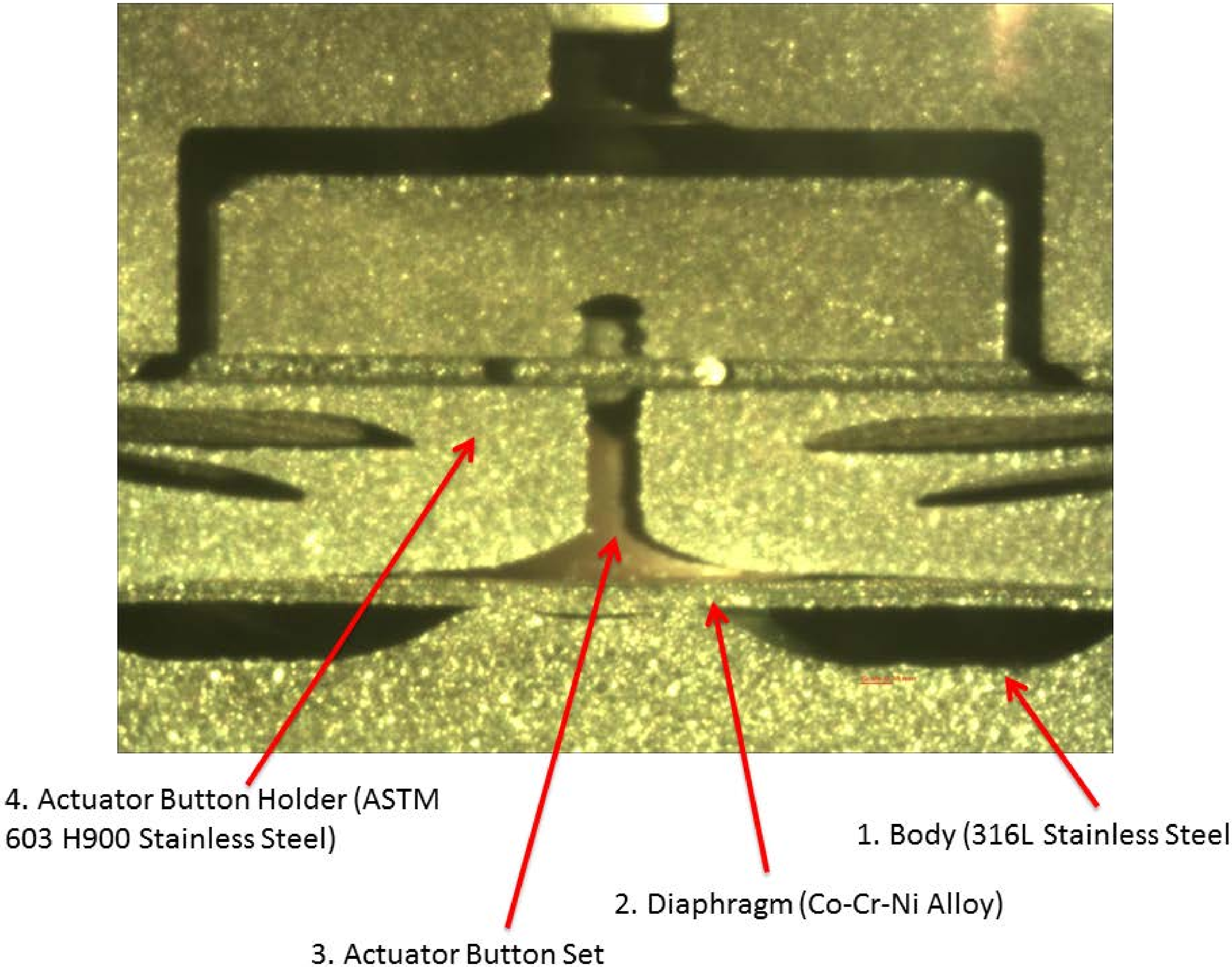


Figure 3-14. Optical Microscope view of cross section of the valve body, labeled for components and materials

3.4.1 Actuator Button Set Analysis

The images from the camera and optical microscope make it clear that both materials of Actuator Button Set are not the same material. To investigate further, a different valve was disassembled, Figure 3-15.

Figure 3-15. The two components of the Actuator Button Set, on the left is the top part of the set (closer to the actuator) and the right is the bottom part of the set (closer to the diaphragm)

The two parts of the Actuator Button Set were placed inside the Scanning Electron Microscope (SEM) together to perform a direct comparison of the two parts. However, a stable image could not be obtained due to off gassing.

Each sample was then analyzed separately in the SEM. The Top Actuator Button Set (on the left in Figure 3-15) was easy to image and it did not have any evidence of off-gassing. The surface of the Top Actuator Set appears metallic from imaging, Figure 3-16. The surface is easy to image and features are distinguishable. By using Energy-dispersive X-ray spectroscopy (EDS), a chemical fingerprint was obtained, Figure 3-17. This chemical footprint shows large amount iron and chromium.

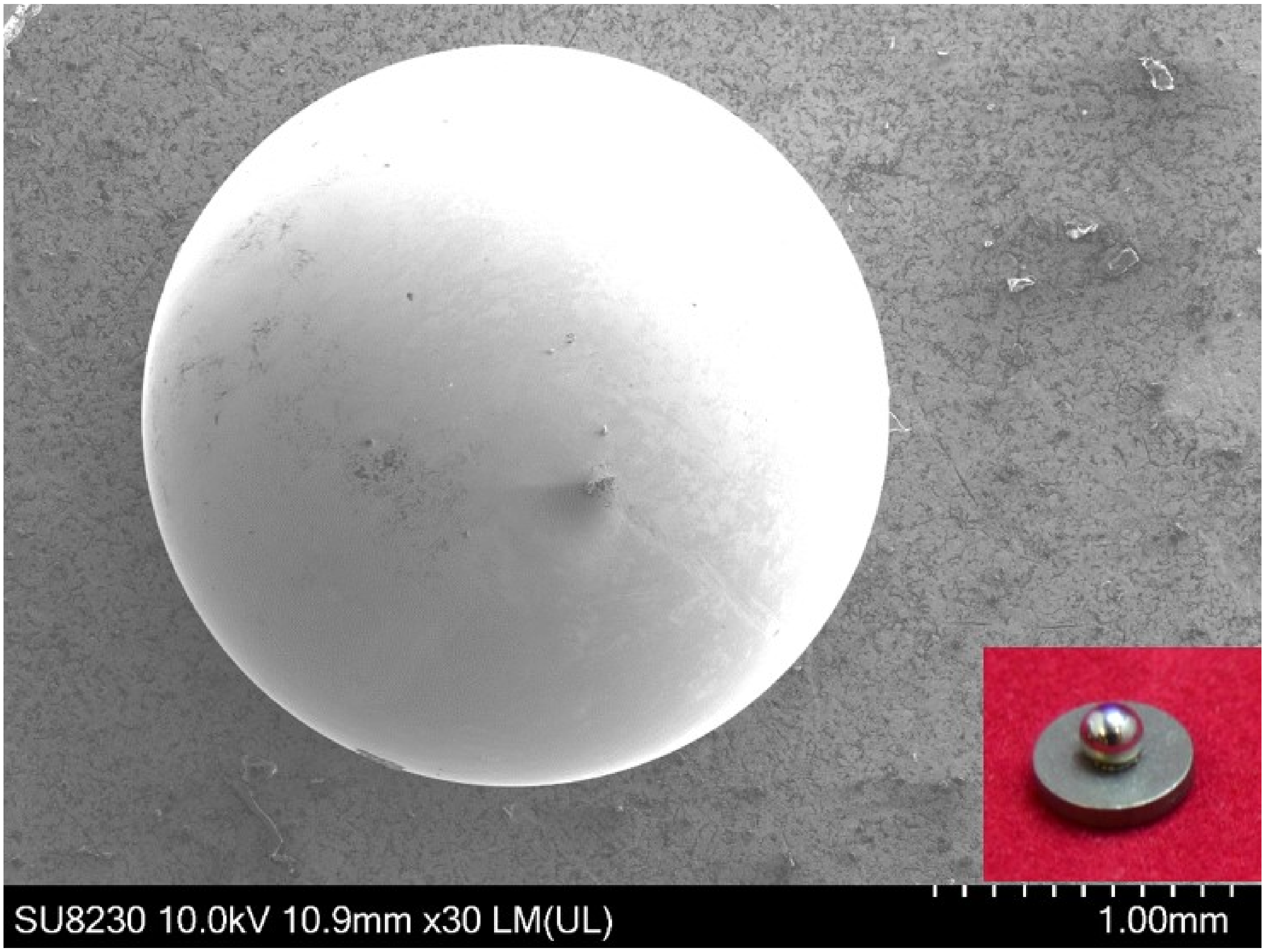
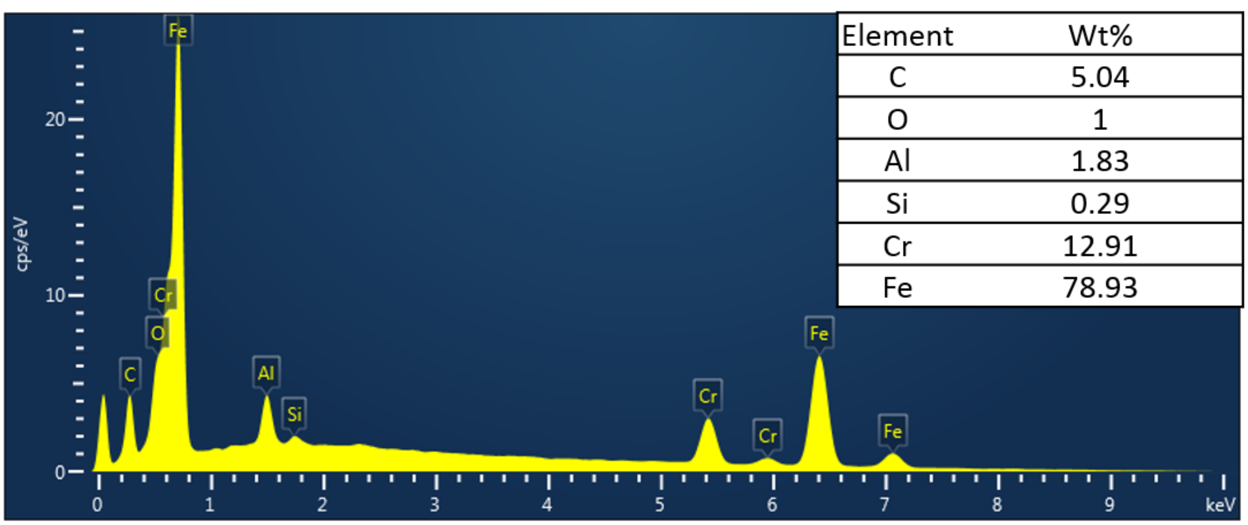


Figure 3-16. SEM image of Top Actuator Button Set with photograph inset



Element	Wt%
C	5.04
O	1
Al	1.83
Si	0.29
Cr	12.91
Fe	78.93

Figure 3-17. EDS Spectra of Top Actuator Button Set with Table of elemental compositions

The Bottom Actuator Button Set was then analyzed in the SEM, Figure 3-18. Elemental analysis performed with EDS, Figure 3-19, detected only carbon and oxygen.

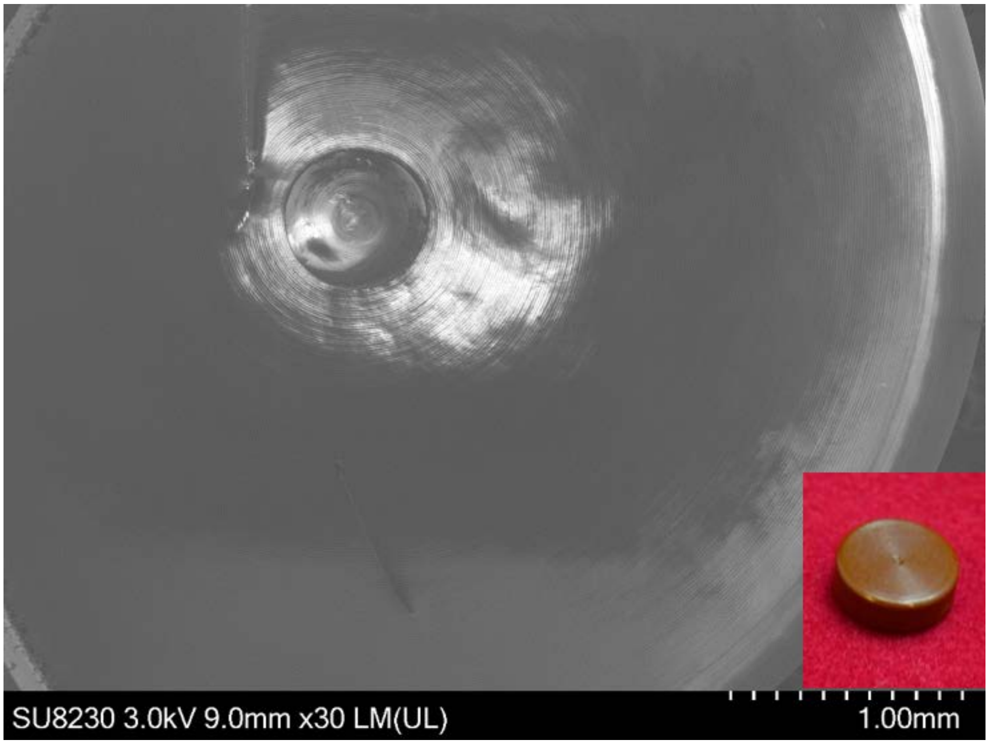
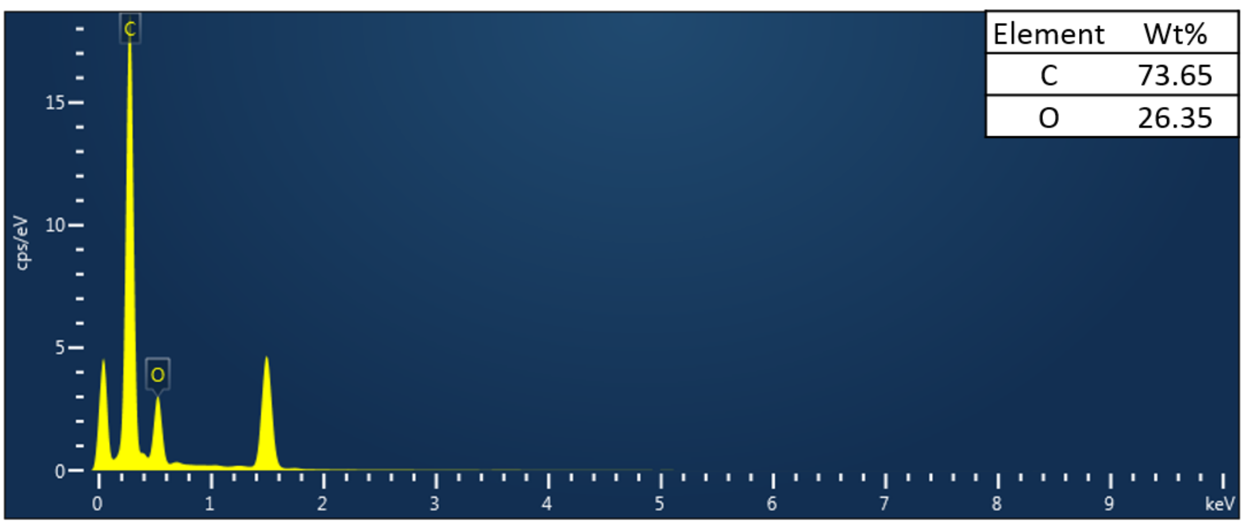


Figure 3-18. SEM image of Bottom Actuator Button Set with photograph inset



Element	Wt%
C	73.65
O	26.35

Figure 3-19. EDS Spectra of Bottom Actuator Button Set with Table of elemental compositions

The bottom actuator button set was then analyzed with an ATR-FTIR. The resulting spectra (Figure 3-20) was matched with that of a known polymer in a chemical database.

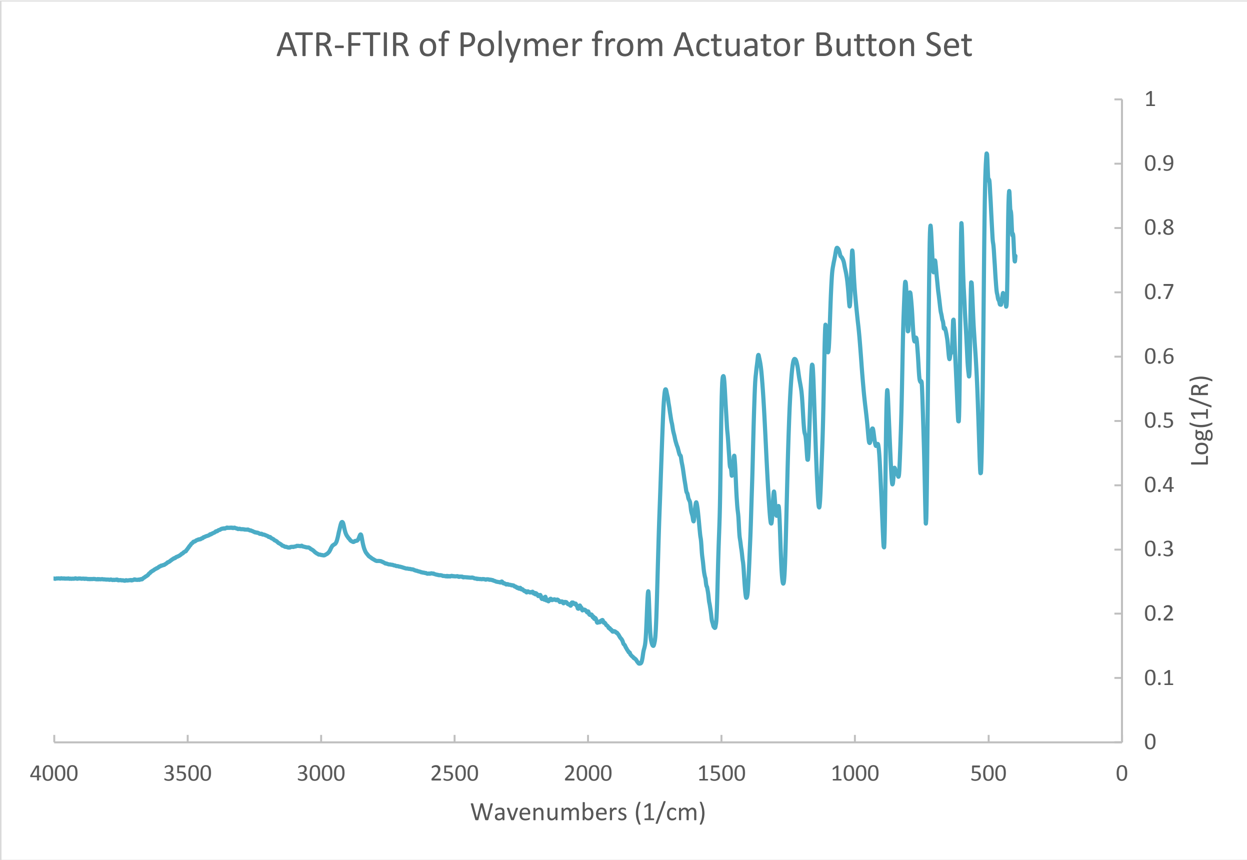


Figure 3-20. ATR-FTIR spectra of Bottom Actuator Button Set

Comparisons of the spectra obtained from the Bottom Actuator Button Set to Polyimide IV is given in Table 3-4 and Figure 3-21.

Table 3-4. Percent match to Button Actuator Button Set sample

Index	Match	Compound name
1070	64.73	Polyimide IV
274	43.19	Poly(trimellitamide imide), pyrol.
1080	37.93	Polyimide
199	36.96	DEXBROMPHENIRAMINE MALEATE IN KBR
34	36.68	POLY(TRIMELLITAMIDE IMIDE)
53	35.10	POLY(TRIMELLITIC AMIDE IMIDE)
342	32.64	Poly(trimellitamide imide)
1376	32.18	SELENITE(IV); BARIUM
924	31.80	Polyamide+imide, aromatic
1835	30.89	Bromodichloromethane

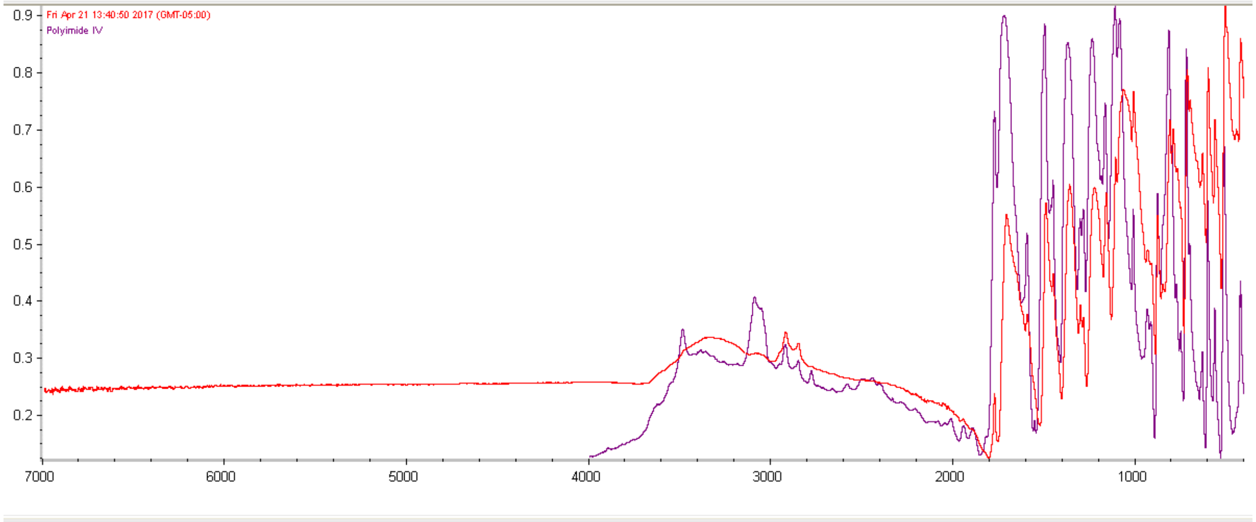


Figure 3-21. Comparison of Bottom Actuator Button Set (red) to Polyimide IV (purple)

From the SEM with EDS and ATR-FTIR, the Actuator Button Set is comprised of two materials, contradicting the specifications in the Ham-Let catalog. The top button is a metal, suspected to be 304L Stainless Steel, and the bottom button is a polymer, belonging to the polyimide family.

3.4.2 Diaphragm Analysis

The diaphragm of the valve was also characterized to ensure material compatibility with hydrogen gas. The Ham-Let catalog (Figure 1-1) lists the diaphragm as a cobalt-chromium-nickel alloy. The entire diaphragm assembly is composed of three different diaphragms, Figure 3-22, with the diaphragm that is wetted labeled Diaphragm 1.

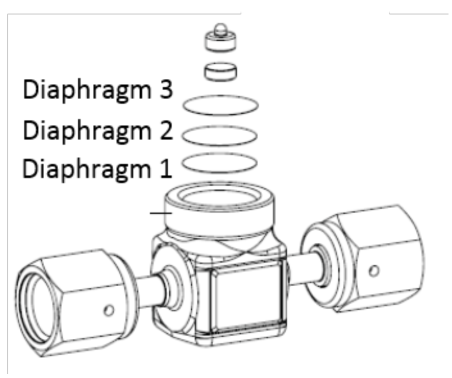


Figure 3-22. Schematic of Ham-Let Valve [7] with diaphragms labeled

The diaphragms were placed into the SEM and EDS was performed on each diaphragm individually. The results from the EDS are in Tables 3-5 and 3-6, along with literature compositions from matching cobalt-chromium-nickel alloys. The 95% confidence interval is also included in both tables, calculated from an analysis of several (10 to 15) random spots.

Table 3-5. EDS of wetted Diaphragm 1 with SPRON510 composition

Element	Diaphragm 01		SPRON510[11]
	Average wt. %	+/- 95% CI	Average wt.%
Co	33.03	0.11	35.40
Ni	32.43	0.13	32.50
Cr	20.21	0.16	20.00
Mo	10.89	0.15	10.00
Fe	1.90	0.04	1.60
Nb	0.97	0.16	--
Ti	0.56	0.04	0.5

Table 3-6. EDS of non-wetted Diaphragms 2 and 3 with Elgiloy composition

Element	Diaphragm 02		Diaphragm 03		Elgiloy[12]
	Average wt. %	+/- 95% CI	Average wt. %	+/- 95% CI	Average wt. %
Co	37.16	0.23	37.50	0.25	39.70
Cr	18.81	0.16	18.79	0.15	19.30
Fe	15.36	0.10	15.40	0.12	15.60
Ni	14.80	0.15	14.90	0.11	15.50
Mo	7.66	0.11	7.72	0.14	7.30
C	2.63	0.62	2.41	0.62	0.04
Mn	1.86	0.06	1.85	0.04	1.90
Al	1.21	0.09	0.89	0.05	0.01
Si	0.51	0.02	0.53	0.02	0.40

The EDS spectra reveal that two different cobalt-chromium-nickel alloys were used. The wetted diaphragm was SPRON510 and the non-wetted diaphragms were Elgiloy.

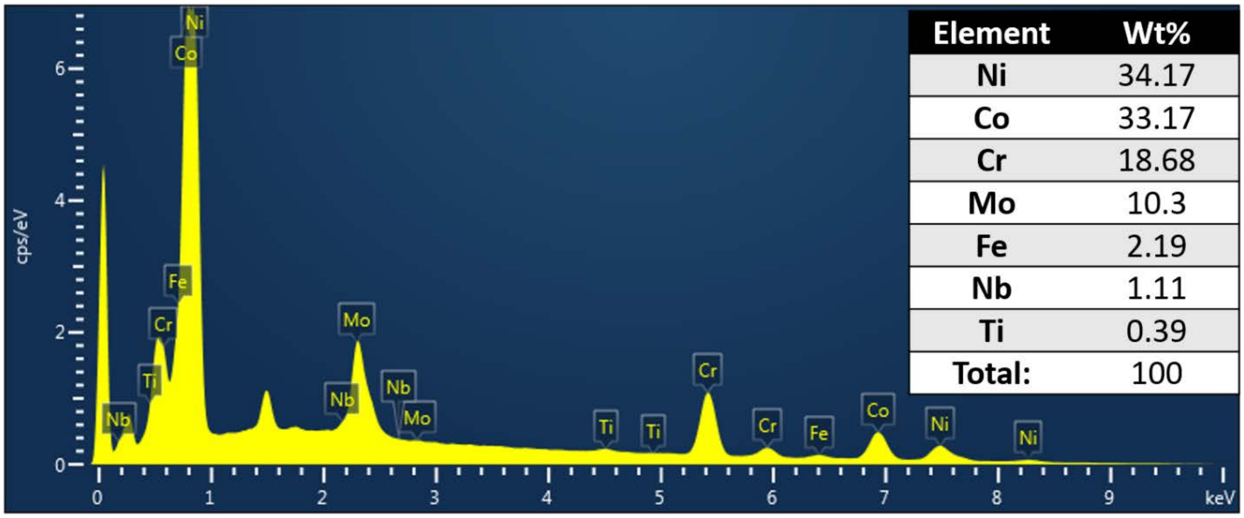


Figure 3-23. EDS Spectra of Diaphragm 1 with inset of atomic weight percent for this location

SEM images were also obtained to analyze the surface of the diaphragm. As shown in Figure 3-24, the surface appears somewhat rough with cracks all over. A higher resolution image of the diaphragm is given in Figure 3-25. A cross sectional image of the diaphragm that was cut by EDM was also viewed in the SEM, Figure 3-26.

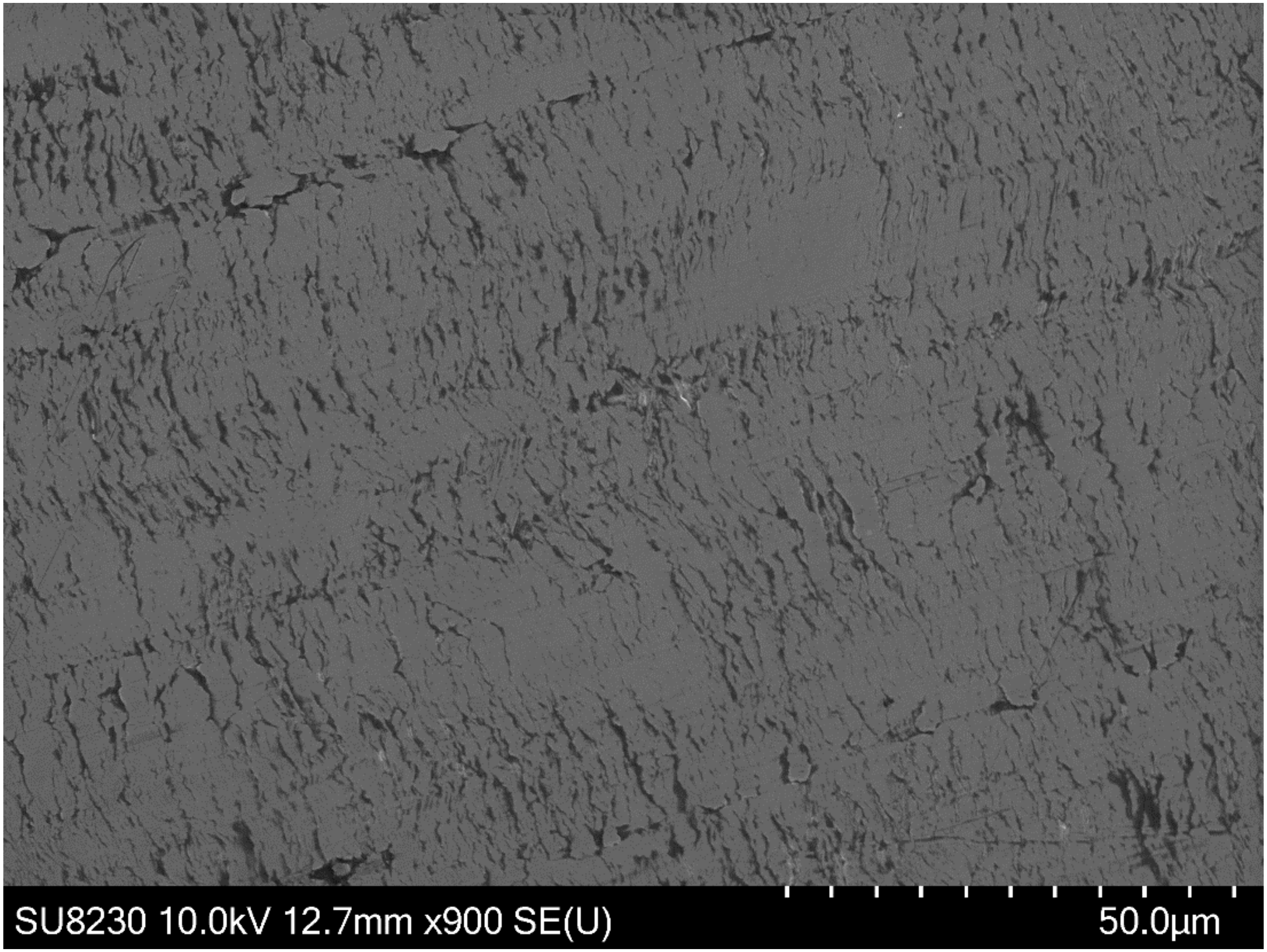


Figure 3-24. Low Resolution SEM Image of Diaphragm

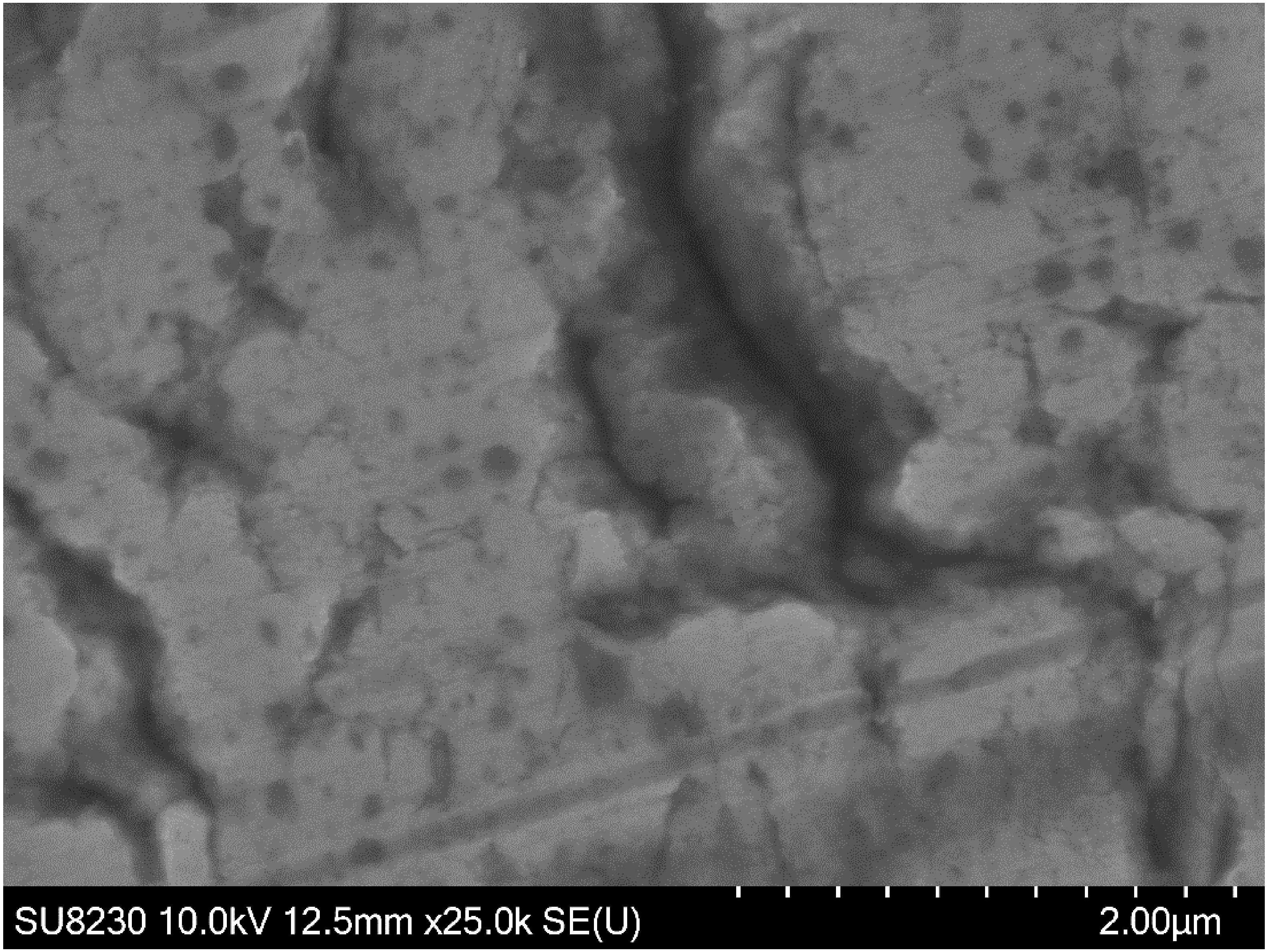


Figure 3-25. High Resolution SEM Image of Diaphragm

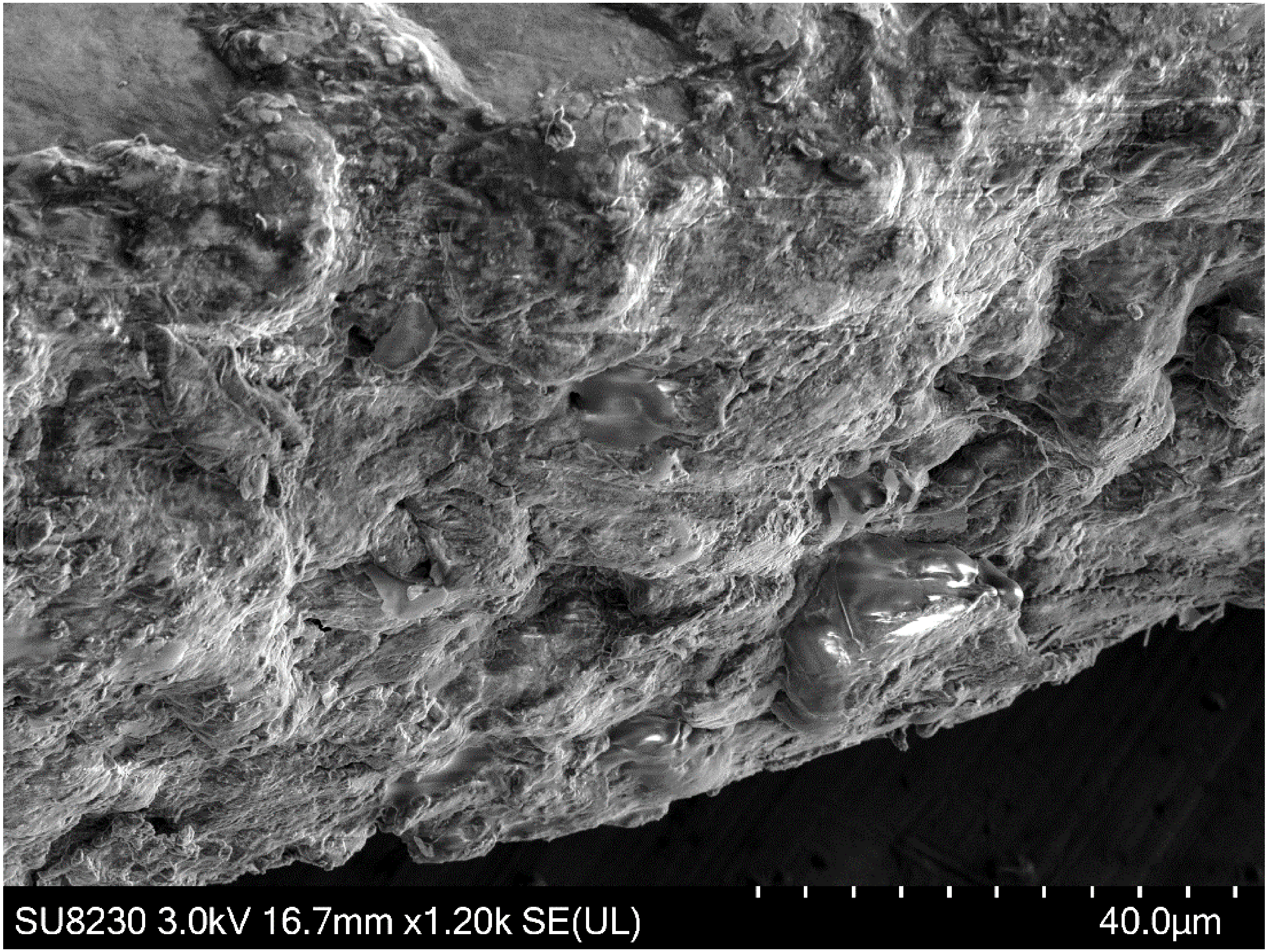


Figure 3-26. Cross Sectional SEM Image of Diaphragm that was EDM Cut

3.5 Failure Analysis

Although TV01 failed leak testing at 20,000 cycles, testing continued until it had been cycled for 150,000 times. At that point, the valve was radiographed to see if any failure mechanisms could be observed. The additional cycling was conducted to help accentuate any faults that may have caused the valve to fail. While inconclusive, it appears that there could be a small gap at the entrance of the inlet at the diaphragm, in Figure 3-27.

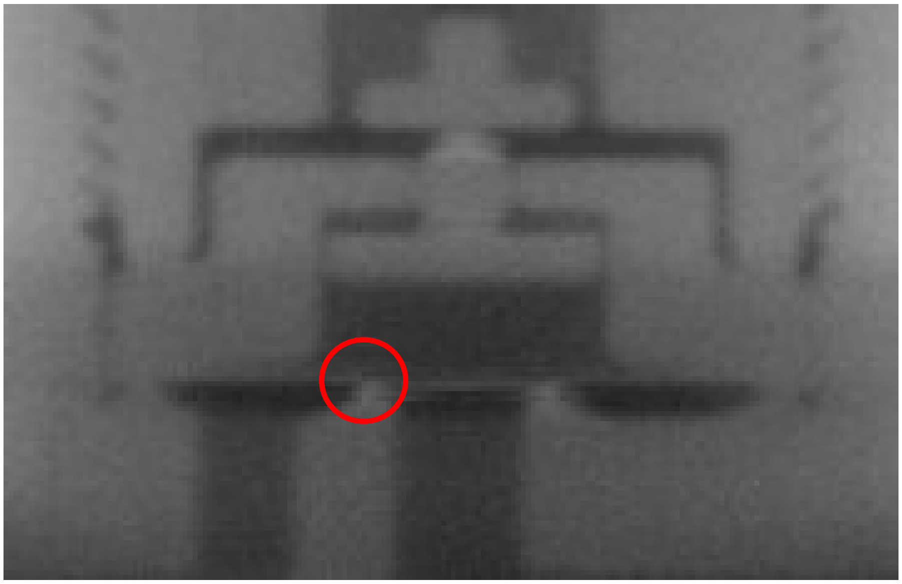


Figure 3-27. Possible Damage to Diaphragm in Radiographed Image

Viewing the diaphragm from the top-down, Figure 3-28, also shows an irregularity at the center of the diaphragm. The diaphragm may be slightly deformed due to particulate contamination or it could be an artifact from imagining. The polyimide Actuator Button Set, directly above the diaphragm, is not visible with X-Ray imaging.

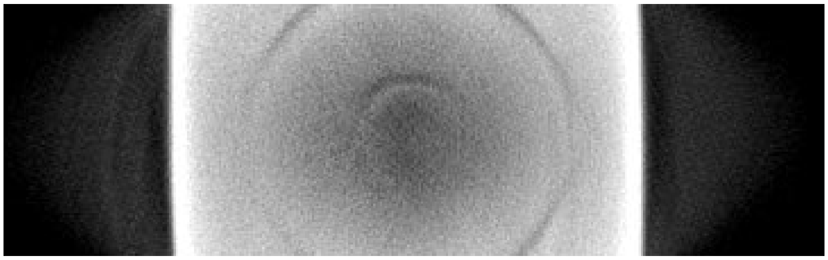


Figure 3-28. Top-Down View of Radiographed Image of Damaged Valve

After TV01 was radiographed, the valve was disassembled. Figure 3-29 shows the actuator facing side of all three diaphragms (from left: wetted, middle, actuator). Spots and/or divots were observed on one side of all of the diaphragms as was a slight irregularity in the wetted diaphragm, Diaphragm 1. Only the middle diaphragm, Diaphragm 2 had the markings on both sides, Figure 3-30 shows the diaphragms that face towards the fluid. Figure 3-31 is a closer view of Diaphragm 2. The source of these spots or divots is unknown; they were not detected in pressure and burst tested valves that were not cycled. Disassembly of other cycled valves may provide additional insight.

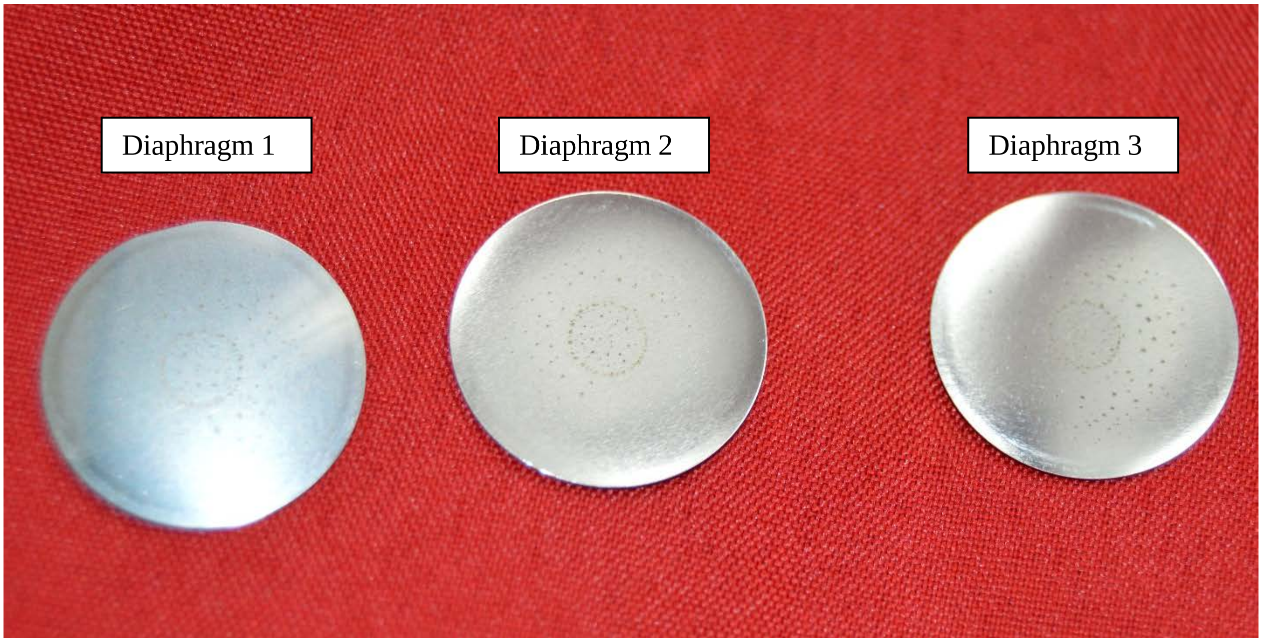


Figure 3-29. Spots on Actuator Facing Side of Diaphragms from TV01

Figure 3-30. Fluid Facing Side of Diaphragms from TV01

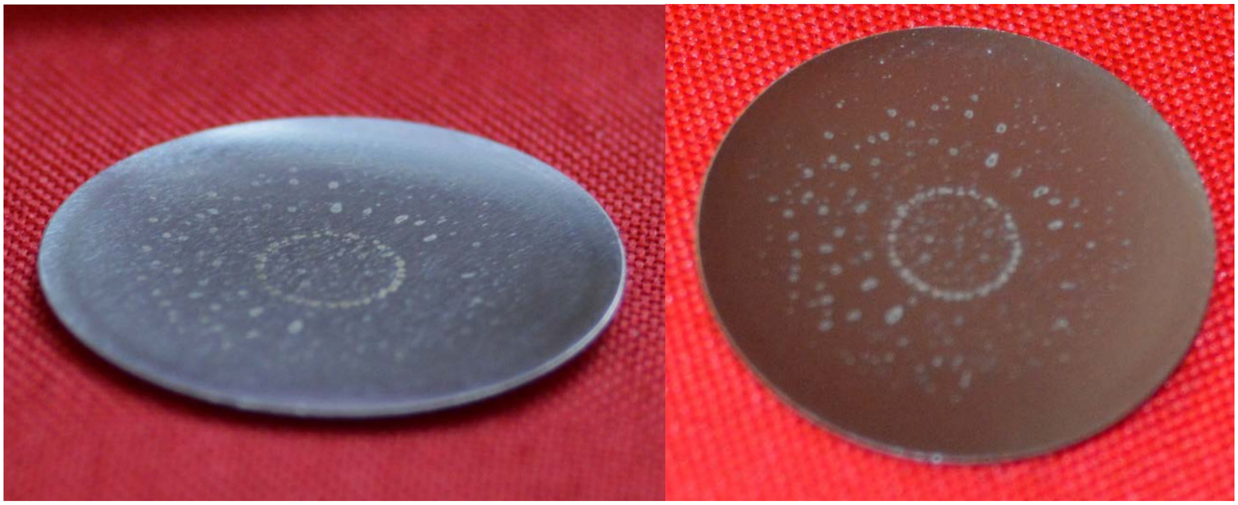


Figure 3-31. Closer Inspection of the Diaphragm 2 from TV01

4.0 CONCLUSIONS

SRNL procured twelve (12) all-metal Ham-Let diaphragm valves for characterization and evaluation. The characterization tests include identification of the maximum pressure of these valves by performing pressure and burst tests. Approximately 3000 psig was required on the inlet side of the valve to lift the diaphragm and approximately 1000 psig was needed on the outlet side to lift the diaphragm. These pressures are much higher than the 150 psig rating stated by the manufacturer. The difference in maximum values for the inlet and the outlet pressure is due to the non-symmetric design of the valve. The burst test, performed by applying pressure to both the inlet and the outlet, resulted in valve failure at over 16,600 psig. The only damage observed from this destructive test was a dimpled diaphragm.

Leak tests were performed to ensure the valves do not exceed the gas leak rate as defined by this study. Nine of ten valves passed the initial leak test. The valves that passed the first leak test were then cycled in a nitrogen or nitrogen/vacuum gas environment. The valve cycle test protocol follows the ASTM "*Standard Test Method for Determination of Cycle Life of Automatic Valves for Gas Distribution System Components*." The valves have been cycled 200,000 times to date, with only one valve failing at 20,000 cycles. Initial failure analysis suggests the diaphragm was bent and it would not seal. Whether this failure occurred due to cycling or due to debris is not yet determined. Two other valves failed at intermediate cycle test points, but recovered to meet leak rate requirements during further cycling, after filters were added to remove any potential debris from the system. These valves are cycling well beyond Ham-Let's expected life of 100,000 cycles; these valves will continue to be cycled.

A detailed material analysis was also conducted to identify the valve's materials of construction, and determine whether they might interact with hydrogen and tritium. The Ham-Let diaphragm valve contains only metal wetted parts; the process gas does not touch the polymer. The valve has held up well with the pressure tests, exceeding the pressure limits by over six times the manufacturer's stated pressure. Cycle testing has proved that these valves are durable; exceeding the manufacturer's stated life of 100,000 cycles. After 200,000 cycles, only one valve has failed. These valves should receive further testing to ensure compatibility with hydrogen and tritium.

5.0 RECOMMENDATIONS AND FUTURE WORK

It is recommended that the Ham-Let valves continue with further testing to ensure material compatibility with hydrogen and tritium. The use of the diaphragm valve in tritium service must consider safety implications due to diaphragm failures, in which tritium may leak through the valve body. Further evaluation is recommended for the materials and valve performance.

6.0 ACKNOWLEDGEMENTS

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Appendix A. Valve Cycle Test Description

Default/Start) The system is being evacuated, all of the isolation valves (IV) are open except for the valves that are for the gas inlet (IV15 and IV18), the test valves (TVs) are normally closed.

Step 1) Isolate the vacuum. To do this, close IV16 and IV19. All of the valves that are required to be under vacuum are under vacuum and the valves that will be pressurized will be ready to have pressure applied to them.

Step 2) Pressurize the line, open IV15 and IV18, now all of the test valves that will be pressurized under nitrogen will have 50 psi of Nitrogen on them.

Step 3) Isolate the TVs that will be tested under static conditions, two valves will be tested under vacuum (TV06 and TV07) and two will be tested in an atmosphere of nitrogen (TV01 and TV02). In order to do this close IV01, IV02, IV06, IV07, IV08, IV09, IV13, IV14, IV15, IV18. They will be closed for 10,000 cycles.

Step 4) Cycle all of the test valves, TV01-TV07. TV03-TV05 will have nitrogen on one side and vacuum on the other. The isolation valves around these test valves will be open, they will be static to the pressurized gas, but IV20 will remain open to allow the gas to that crosses the test valves to exit the system.

Step 5) Reapply pressure to test valves TV03-TV05 by opening IV15.

Step 6) Once the pressure has reached 50 psi, close IV15 to stop the pressurization of the line.

Step 7) Cycle all of the test valves, TV01-TV07. Once this has completed, loop back to 5 to keep reapplying pressure that has exited the system during the test cycle. Keep looping until number of test cycles is complete (for this case, we want 10,000 cycles).

Step 8) After loop is completed with 10,000 test cycles, get the system to a state where we can disassemble it and take the TVs to the High Pressure Lab. We will get the system under vacuum, by opening IV01, IV02, IV06, IV07, IV08, IV09, IV13, IV14, IV16, and IV19. Then shut the pump off.

The Following Page has a Table describing what is above

Table A-1. Steps of Valve Cycling

Step	Open IV	Close IV	Cycle TV	Time (seconds)
Start/Default	IV01, IV02,IV03, IV04, IV05, IV06 IV07, IV08, IV09, IV10, IV11, IV12, IV13, IV14, IV16 IV17, IV19, IV 20	IV15, IV18		5
1		IV16, 19		1
2	IV15, IV18			3
3		IV01, IV02, IV06, IV07, IV08, IV09, IV13, IV14, IV15, IV18		1
4			TV01-TV07	1 open 1 closed
5	IV15			2
6		IV15		1
7			TV01-TV07	1 open 1 closed
5*	IV15			2
6*		IV15		1
7*			TV01-TV07	1 open 1 closed
Stop/Default	IV01, IV02, IV06, IV07, IV08, IV09, IV13, IV14, IV16, IV19			1

*Indicates Steps in the Loop