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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois

HELIUM-PURIFICATION UNIT FOR HIGH-PURITY INERT-ATMOSPHERE BOXES

by

M. S. Foster, C. E. Johnson, and C. E. Crouthamel

Chemical Engineering Division

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DISCLAIMER

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SUMMARY

A device for purifying and recycling the helium atmosphere of two dryboxes having a combined volume of 60 ft^3 is described. Trace impurities are removed by passing the helium through a palladium-catalyst container and a molecular sieve bed, and then cooling to -195 C before passing through an activated charcoal bed. Flow rates of 10 scfm and purity levels are estimated. Regeneration procedures are given.

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I. INTRODUCTION

In the regenerative-cell program, nearly all experiments involve molten metals and salts which are extremely sensitive to the chemically active gases. Lithium, for example, may be expected to combine with oxygen, nitrogen, and water vapor. The commonly used lithium halides form hydrates at room temperature; at high temperatures, volatile halogen acids may be lost.

The problem, then, concerns not the absolute purity of the original charge of blanket gas, but maintaining the purity of the box atmosphere in the face of continuing contamination. Water vapor, for example, is continually diffusing into the box through rubber gloves. There is continual contamination of the inert gas atmosphere by chemically active gases released when new equipment is moved into the box and during heating operations.

There are at least two possible methods of maintaining a high-purity atmosphere in the dry box. One is to continually flush pure gas through the box. This method is wasteful of the inert gas, as a high flow rate must be maintained to provide for a complete change of box atmosphere every 3 to 5 min. The second method of maintaining a purified box atmosphere, in our opinion the only practical approach, is to cycle the inert gas through a purification unit and then return it to the box.

At least two general methods of gas purification under recycle are possible. One is the use of a hot active metal (such as titanium sponge at 800 C), which acts as a getter for all contaminants. The second, which was the method of choice, is the use of activated carbon or artificial zeolites at low temperatures. Both methods are capable of removing trace concentrations of the active gases. The latter procedure is similar to that used by the U. S. Bureau of Mines in the preparation of its "Grade A" helium. In their process, one of the last steps in purification is to pass the helium (at 2700 psi) through activated coconut charcoal at -320 F (-195.5 C). The resulting helium is 99.995 percent pure and contains <50 ppm by volume total impurities. Neon may be present to the extent of 1 to 20 ppm and all other constituents to the extent of <10 ppm. This procedure is suitable for purifying only helium among the inert gases.

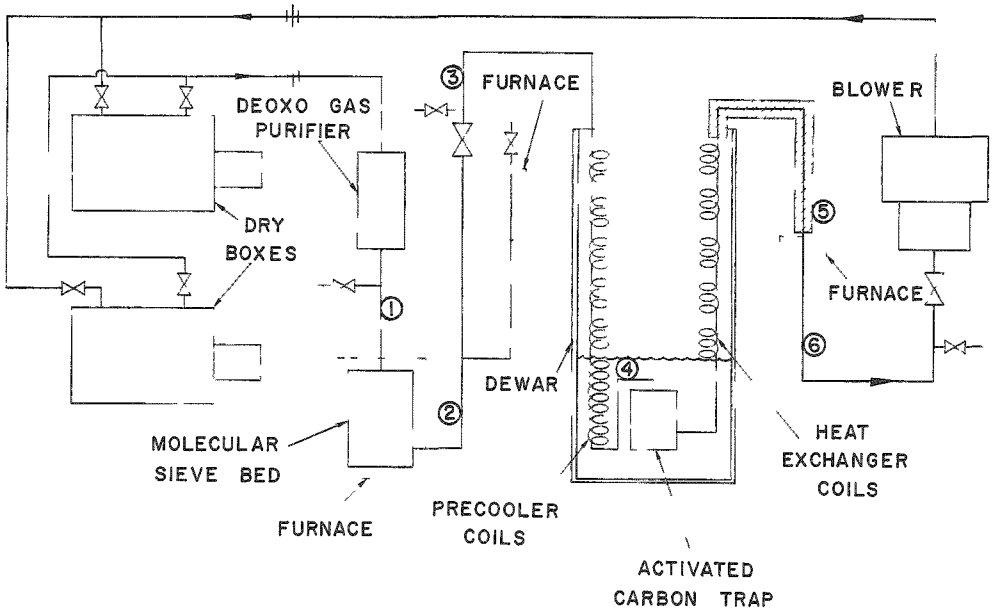
II. SYSTEM CONSTRUCTION AND OPERATION

A. Normal Operation

A schematic diagram of the helium-purification unit is shown in Figure 1. The unit was designed to operate at a helium flow rate of 6 to 10 scfm. This rate was considered adequate for purifying the atmosphere in two dry boxes, of conventional design, each having a volume of approximately 29.5 ft^3 and three glove ports. The piping arrangement was as shown (parallel hookup) in order that either or both boxes might be connected to the purification system at any time.

Figure 1

HELIUM-PURIFICATION SYSTEM



The sequence of steps in the purification system shown in Figure 1 was as follows. The contaminated gas from the dry boxes passed first through a palladium catalytic unit (Deoxo Model D-300-200, Engelhard Industries).* In this unit any hydrogen present in the gas stream was burned to water. It was assumed that there generally existed an excess of oxygen over hydrogen in the incoming atmosphere, thereby facilitating the combustion of hydrogen to water. The helium gas then passed through a dryer unit consisting of a bed, 8 in. in diameter and 12 in. deep, of $\frac{1}{8}$-in. Linde Type 5A Molecular Sieve pellets (dwg. CE-D5000**).

The gas was then allowed to cool by being passed through five sections of coiled copper tubing. Each coil consisted of three turns and was

*The addresses of manufacturers are given in the Appendix.

**All drawings are at end of report.

separated from each succeeding coil by a 2-in. section of stainless steel tubing inserted to reduce rapid longitudinal heat flow. These coils were wound into an 11-in.-ID helix and were placed in a large Dewar (dwg. CE-B4953), as shown in Figure 1. This Dewar was sufficiently deep so that none of the five sections of tubing were immersed in the liquid nitrogen, but rather were suspended in the nitrogen vapor escaping from the boiling liquid nitrogen. This allowed the incoming gas to be precooled by the nitrogen vapor before entering an eight-turn 11-in.-ID helix of copper tubing located below the liquid surface. After the gas passed through the eight-turn helix, the temperature was measured (at point 4 of Figure 1) as -195 C. Next, the gas passed through an activated charcoal bed immersed in liquid nitrogen. The bed was 8 in. in diameter and 12 in. high. Coconut charcoal for the bed came from Barnebey-Cheney Company and was size 8 x 14, activity 75 min as measured by an accelerated chlorpicrin test. The all-copper vessel containing this charcoal is described in drawing CE-D4796, sheet 2 of 3.

In an effort to utilize some of the cooling capability of the now purified helium gas, the gas was then allowed to pass out of the Dewar through five sections of coiled copper tubing, with coils again separated by 2-in. lengths of stainless steel tubing. This helix also was located in the nitrogen vapors above the liquid nitrogen surface and inside the Dewar.* The temperature of the gas was measured as -80 C (at point 5 of Figure 1) after it had passed through these "heat exchanger" coils and an additional 4 ft of insulated stainless steel tubing.

The helium stream was then heated to approximately 0 C by allowing it to pass through a length of copper tubing surrounded by furnace heating elements (two Hevi-Duty Type 77KSP, 480-w units). The control thermocouple is at point 6 of Figure 1.

The final step in the recycle process was to pass the helium into an "O"-ring-sealed chamber (dwg. CE-4922) containing a Rotron blower (Model MLPM, type A8-701). The flow rate maintained through the system by the Rotron blower was estmated as 10 scfm.

B. Regeneration

During regeneration of the two beds, the dry boxes and the blower chamber were isolated from the system. The valve in the main stream between the dryer and precooling coils was also closed. The carbon bed

*It was found that without a helium exchanger, cooling a 10-cfm helium stream to -195 C with a long copper coil immersed in liquid nitrogen required 800 liters of nitrogen per 24 hr of operation. The liquid nitrogen consumption was reduced to 100 liters per 24 hr by use of the heat exchanger. Since the cost of liquid nitrogen to the Laboratory is 7¢/liter, the 100 liters per 24 hr amounts to a cost of $7.00. Since it is not necessary to run the purification unit on a continuous basis, it is felt that this expense is not too great.

was regenerated by heating to approximately 100 C with hot argon entering the system through a small side valve located near the blower. This stream was heated in the furnace and passed through the charcoal bed in a reverse direction to normal helium flow. Additional heat was supplied by a 250-w infrared bulb suspended in the Dewar over the activated carbon trap to facilitate the evaporation of liquid nitrogen from the Dewar.

The Molecular Sieve pellets in the dryer were regenerated in a similar manner. Hot argon (at 400 C) was passed through the bed in a reverse direction while the outside of the bed was heated. The temperature of the entering argon was read at point 2 of Figure 1, and that of the existing argon at point 1.

After these units were both heated to the desired temperatures, they were evacuated to a pressure of perhaps 10 μ or less and maintained at this pressure for approximately 48 hr. Weekends were reserved for the long pumpdown periods necessary for good regeneration.

It was convenient to operate the system continuously during the week from Monday morning until Friday afternoon. The liquid nitrogen consumption during normal operation was approximately 5 liters/hr.

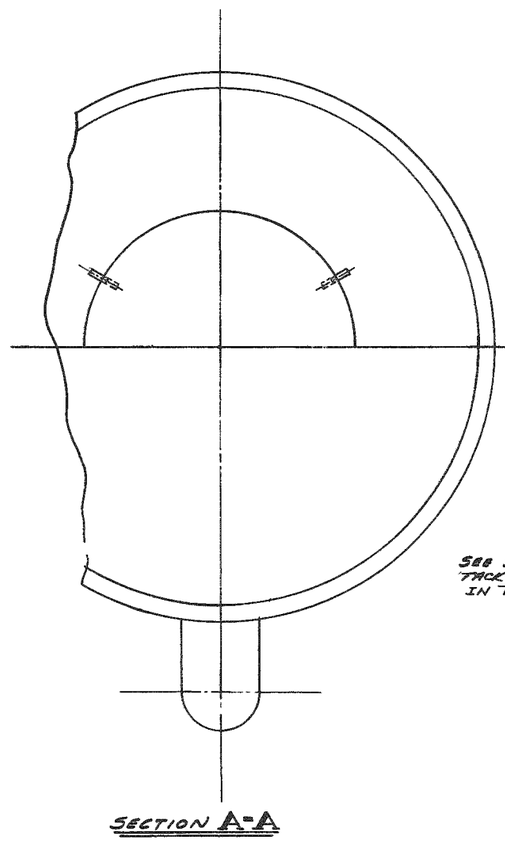
III. PURITY RESULTS

The amount of moisture in each box atmosphere was continuously monitored by Consolidated Electrodynamics Model 26-303MB Moisture Monitors (0-100 ppm by volume). During normal operation, readings were in the range of 0.5-5.0 ppm after the moisture from the box surfaces had been removed. Gas chromatographic analysis of the helium showed that oxygen and nitrogen concentrations were each below 10 ppm. Samples were taken from the exit line of the purification system and from the dry box atmosphere.

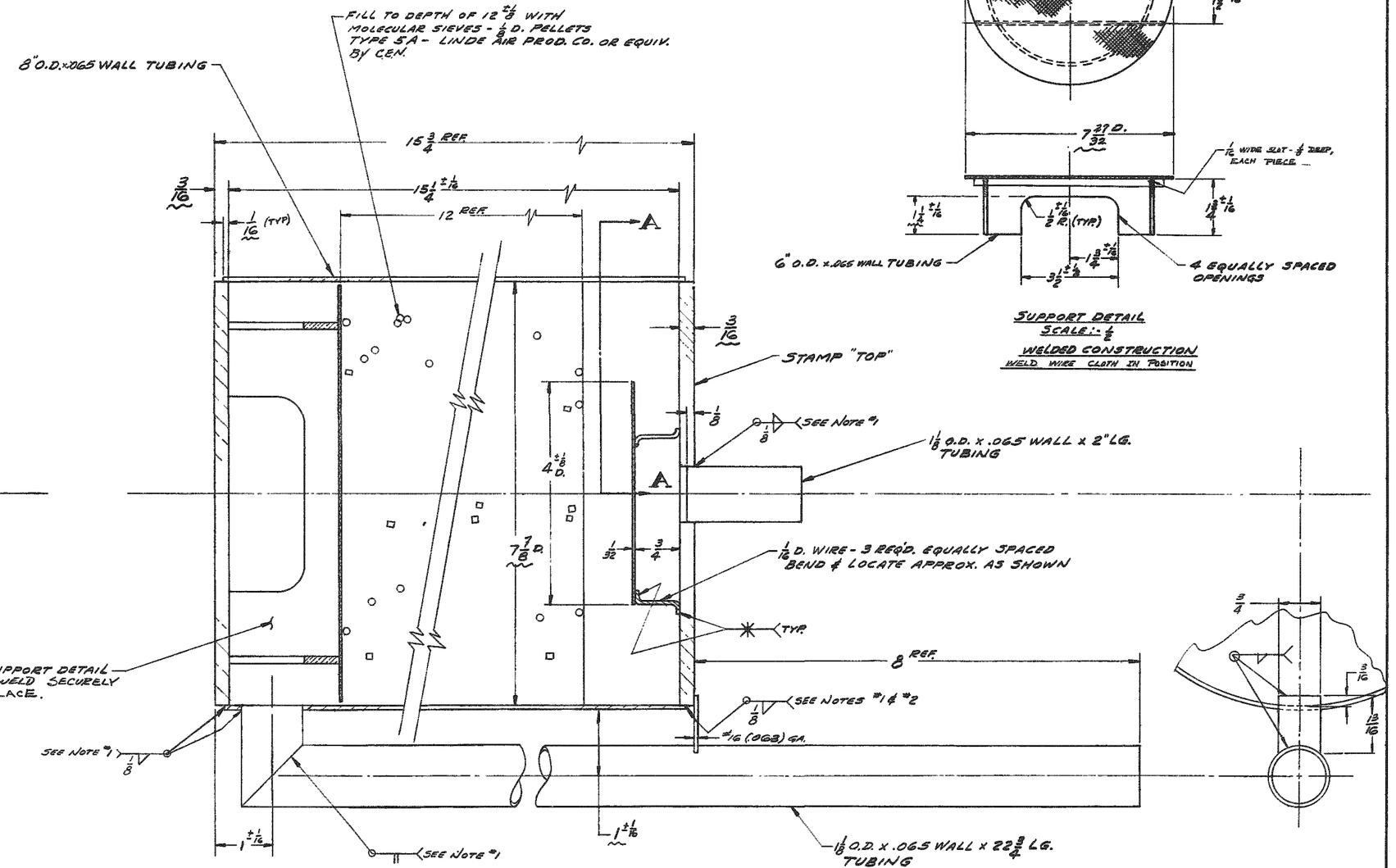
APPENDIX

Suppliers of Components

Deoxo Gas Purifier, Model D-300-200	Engelhard Industries, Inc. Gas Equipment Section 113 Astor Street Newark 2, New Jersey
$\frac{1}{8}$-in. Molecular Sieve pellets, Type 5A	Linde Company 230 North Michigan Avenue Chicago 1, Illinois
Activated Coconut Charcoal (size, 8 x 14; activity, 75 min, measured by accelerated chlorpicrin test)	Barnebey-Cheney Company Columbus 19, Ohio
Blower, Model MLPM, type A8-701	Rotron Manufacturing Company Woodstock, New York
Moisture Monitor Model 26-303MB	Consolidated Electrodynamics Corp. 2536 West Peterson Avenue Chicago 45, Illinois



SECTION A-A

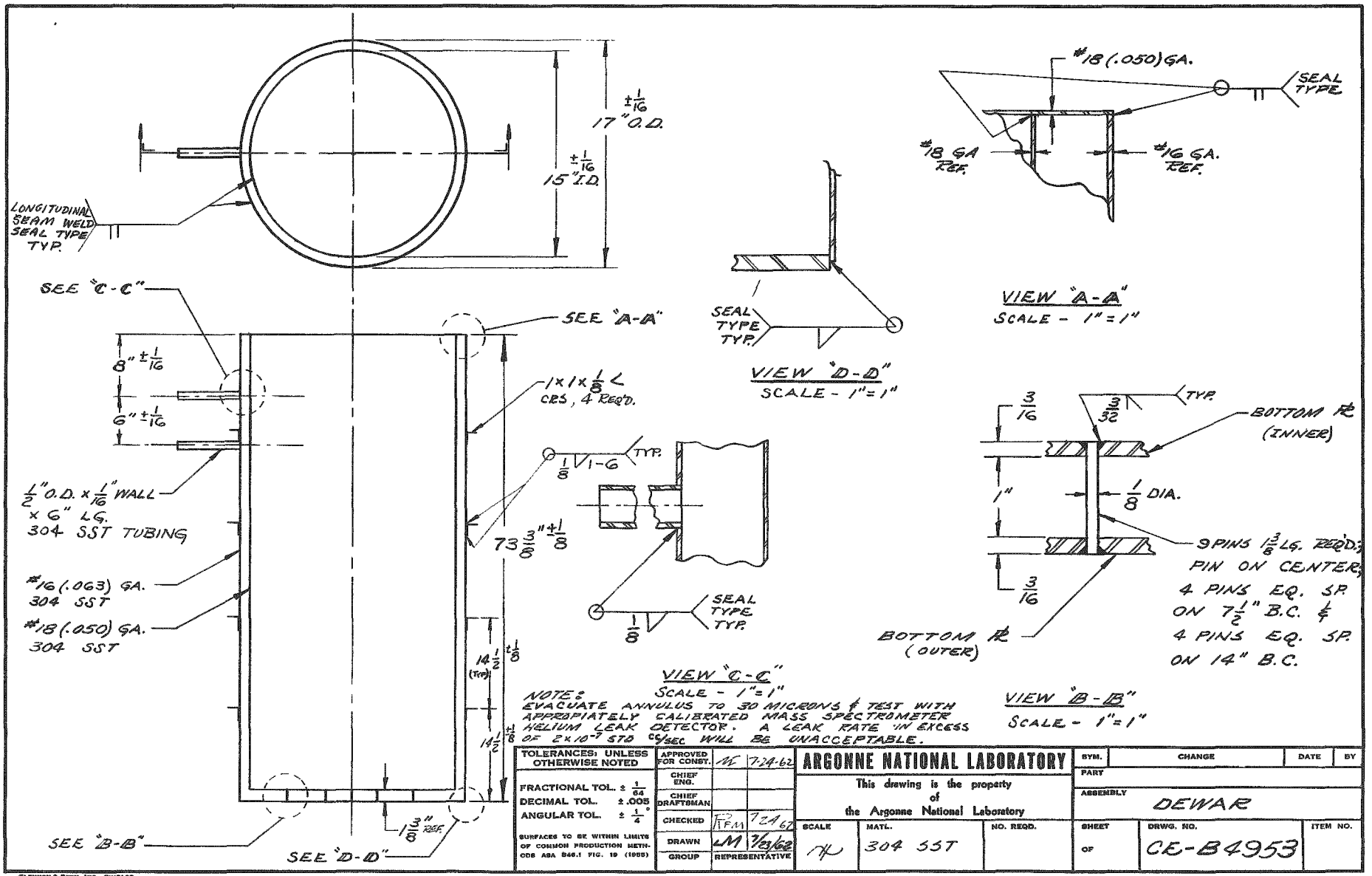


SUPPORT DETAIL
SCALE: 1/2
WELDED CONSTRUCTION
WELD WIRE CLOTH IN POSITION

NOTES:
1. SEAL TYPE WELD - TEST WITH MASS SPECTROMETER
2. WELD BEFORE FILLING WITH MOLECULAR SIEVE PELLETS
3. CLEAN & DEGREASE
4. OPERATING PRESSURE - VACUUM @ 800°F.

±1/32

TOLERANCES. FRACTIONAL TOL. ± 1/32 DECIMAL TOL. ± .005 ANGLE ± 1/2° ALL SURFACES TO HAVE AVERAGE FINE MICRO FINISH OR AS RECEIVED FINISH. UNLESS OTHERWISE NOTED	APPROVED FOR CONST.		11/8/61	ARGONNE NATIONAL LABORATORY This drawing is the property of the Argonne National Laboratory	A — REDESIGNED FROM DWG. CE-D 4796 — 2/9/62 — JK
	CHIEF ENG.				SYM — CHANGE — DATE — BY
	CHIEF DRAFTSMAN				PART: DRYER
	CHECKED				ASSEMBLY: HELIUM GAS PURIFICATION SYSTEM
	DRAWN	JK	11-8-61	SCALE: FULL & NOTED — MATL.: 304 SST & NOTED — NO REQD.	SHEET OF — DRWG. NO.: CE-D5000 — ITEM NO.
	GROUP		REPRESENTATIVE		



NOTE: EVACUATE ANNULUS TO 30 MICRONS & TEST WITH APPROPIATELY CALIBRATED MASS SPECTROMETER HELIUM LEAK DETECTOR. A LEAK RATE IN EXCESS OF 2×10^{-7} STD CC/SEC WILL BE UNACCEPTABLE.

TOLERANCES: UNLESS OTHERWISE NOTED	APPROVED FOR CONST.	ME	7-24-62
FRACTIONAL TOL. ± 1/64	CHIEF ENG.		
DECIMAL TOL. ± .005	CHIEF DRAFTSMAN		
ANGULAR TOL. ± 1/4°	CHECKED	TFM	7/24/62
SURFACES TO BE WITHIN LIMITS OF COMMON PRODUCTION METHODS ASA B46.1 FIG. 19 (1955)	DRAWN	LM	7/23/62
	GROUP	REPRESENTATIVE	

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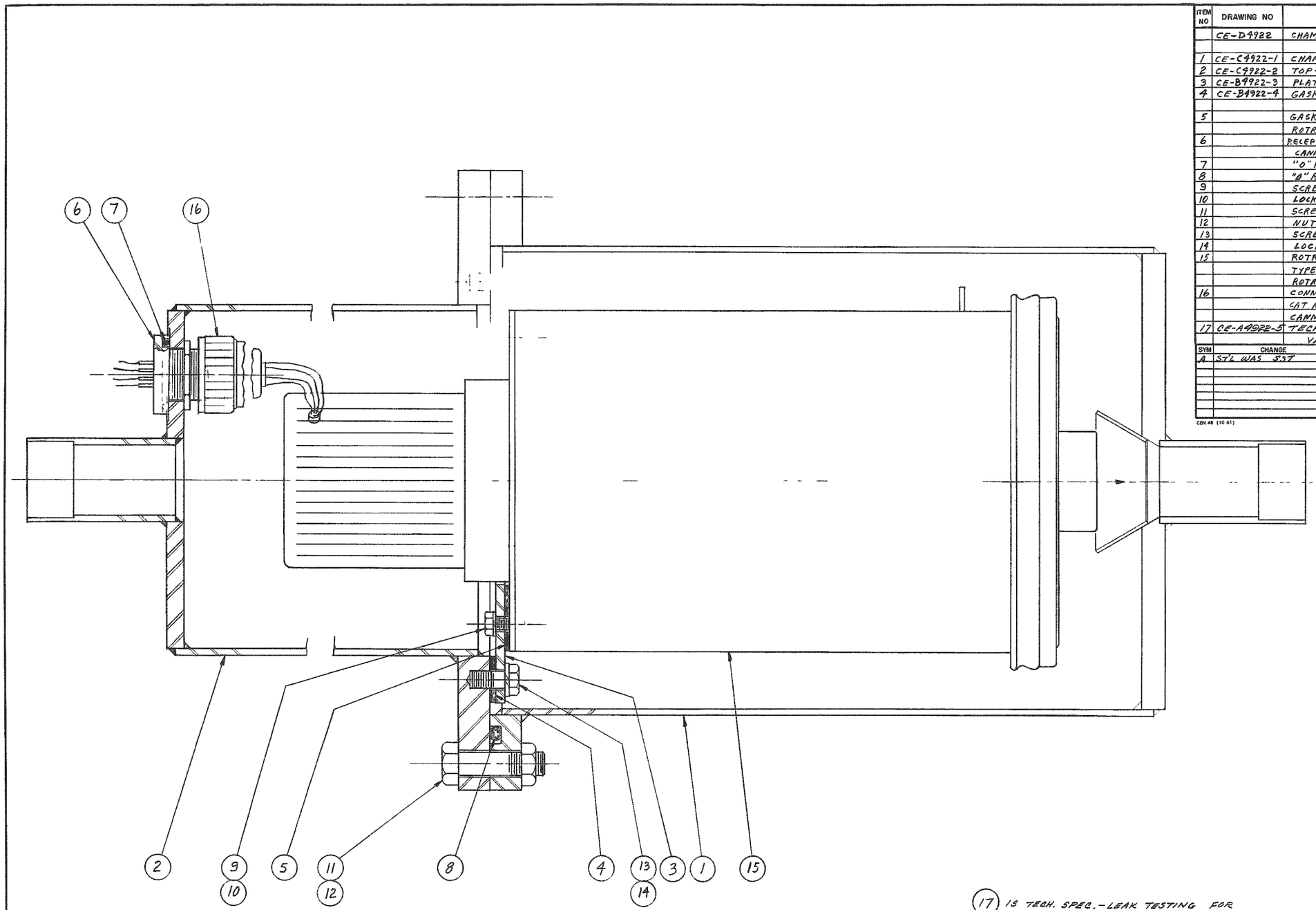
SYM.	CHANGE	DATE	BY

PART

ASSEMBLY: DEWAR

SCALE	MATL.	NO. REQD.	SHEET	DRWG. NO.	ITEM NO.
1/4	304 SST		OF	CE-B4953	

GLENNON & KEHN, INC., CHICAGO



ITEM NO	DRAWING NO	PART NAME OR DESCRIPTION	REMARKS	NO. REQ.
	CE-D4922	CHAMBER-ROTRON BLOWER		
1	CE-C4922-1	CHAMBER		1
2	CE-C4922-2	TOP-CHAMBER		1
3	CE-B4922-3	PLATE - SUPPORT		1
4	CE-B4922-4	GASKET		1
5		GASKET - ROTRON PART NO. 8936		
		ROTRON MANUFACTURING CO. INC.	BY ANL	1
6		RECEPTACLE CAT.# DPH-18-10S-002		
		CANNON ELECTRIC COMPANY		1
7		"O" RING - 1 1/4 I.D. X 1/8 W.	BUNA N	1
8		"O" RING - 10 1/2 I.D. X 1/4 W.	BUNA N	1
9		SCREW-HEX. HD. 1/4-20 UNC X 11/16 LG.	Δ ST. REC. CAD. PLT'D	8
10		LOCKWASHER 1/4 STD.	" .	8
11		SCREW-HEX. HD. 1/2-13 UNC X 1 1/4 LG.	" .	8
12		NUT-HEX. HD. 1/2-13 UNC	" .	8
13		SCREW-HEX. HD. 3/8-16 UNC X 3/4 LG.	" .	8
14		LOCKWASHER 3/8 STD.	" .	8
15		ROTRON BLOWER MODEL MLPM		
		TYPE AB-701 MOTOR SERIES 266JS		
		ROTRON MANUFACTURING CO.	BY ANL	1
16		CONNECTOR, STRAIGHT PLUG		
		CAT NO. MS3106A-18-10P (F16)		
		CANNON ELECTRIC CO.		1
17	CE-A4922-5	TECH. SPEC. - LEAK TESTING FOR VACUUM SERVICE		

SYM	CHANGE	BY	CHK	DATE
A	ST'L WAS S.S.T	MP		11-1-63

THIS PARTS LIST IS THE PROPERTY OF ARGONNE NATIONAL LABORATORY

ASSEMBLY: CHAMBER-ROTRON BLOWER

APPROVED: AE 6/3/63 | SHEET 1 OF 1

CHECKED: | DRWG NO

DRAWN: AB 6/8/63 | CE-D4922

CEN 48 (10 61)

(17) IS TECH. SPEC. - LEAK TESTING FOR VACUUM SERVICE

TOLERANCES: UNLESS OTHERWISE NOTED	APPROVED FOR CONST		6 3 63
FRACTIONAL TOL. ± 1/64	CHIEF ENG		
DECIMAL TOL. ± .005	CHIEF DRAFTSMAN		
ANGULAR TOL. ± 1/4°	CHECKED		
SURFACES TO BE WITHIN LIMITS OF COMMON PRODUCTION METH COS ABA 1346.1 FIG 19 (1000)	DRAWN	AB	6/8/63
	GROUP	REPRESENTATIVE	

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SCALE	MATL	ND REQD
1"=1"		

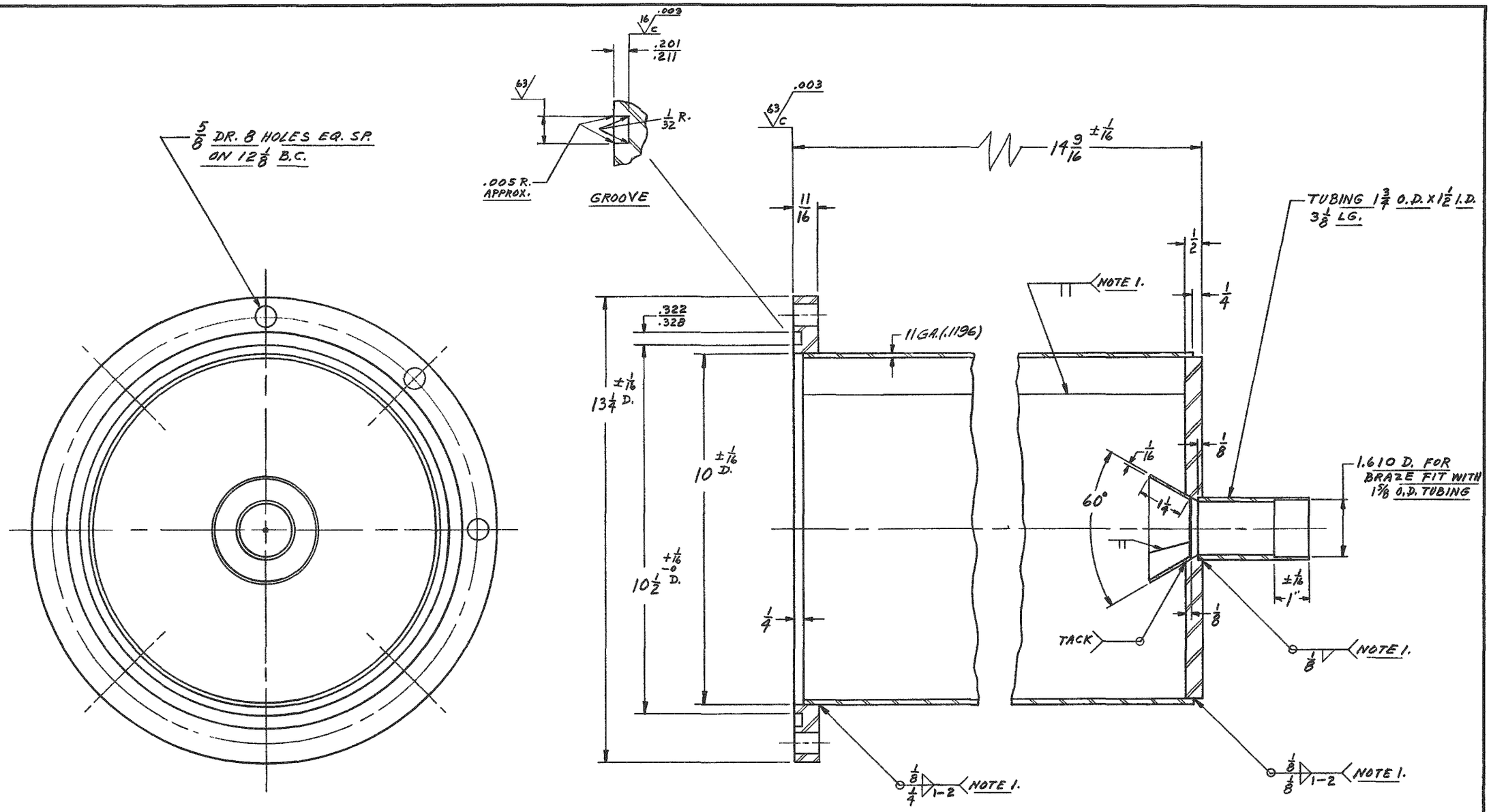
SYM	CHANGE	DATE	BY

PART

ASSEMBLY: CHAMBER-ROTRON BLOWER

SHEET	DRWG NO	ITEM NO
OF	CE-D4922	

GLENDON B. KEAN INC CHICAGO

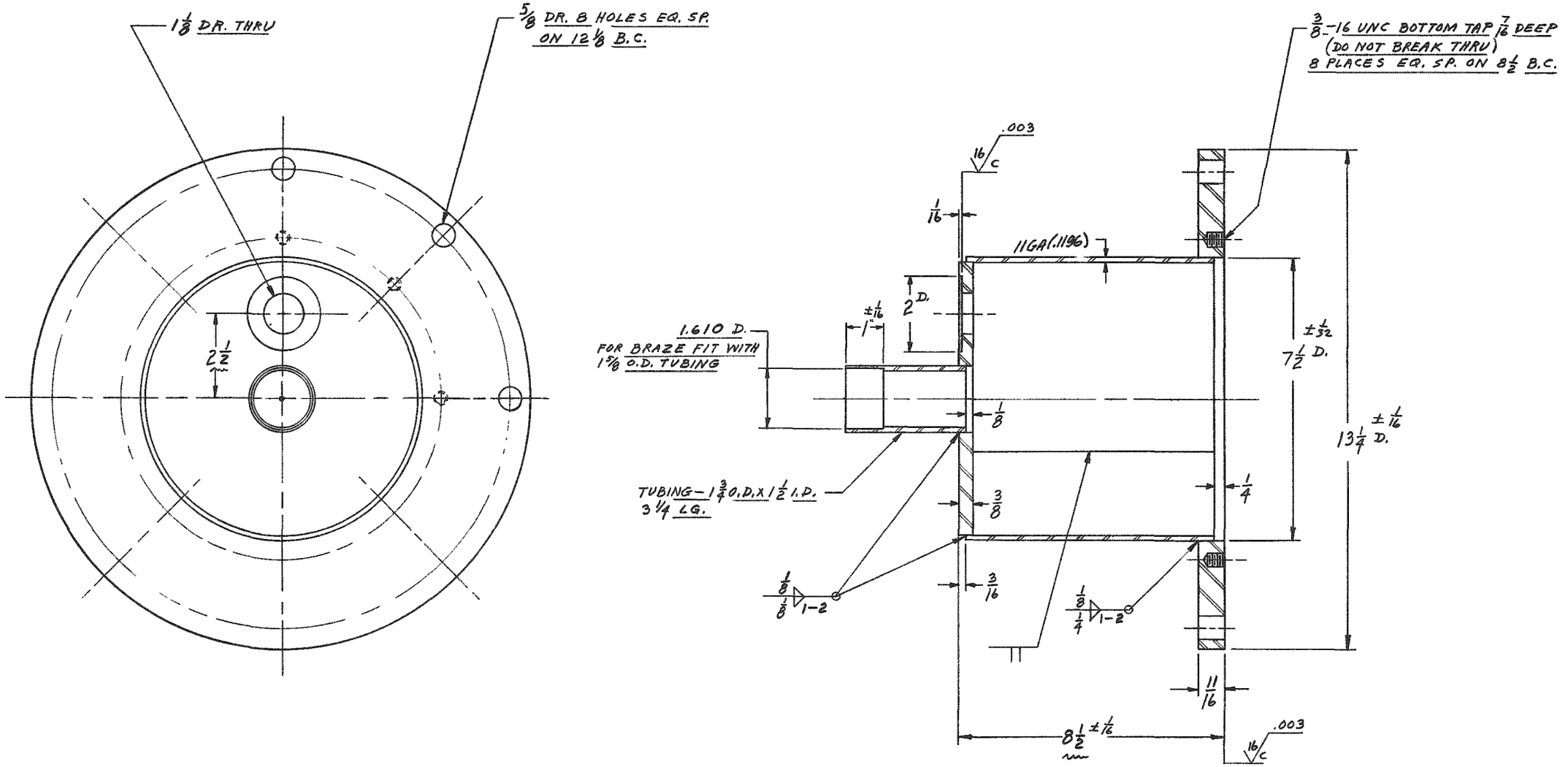


NOTE:

1. VACUUM TIGHT WELDS – HELIUM LEAK TEST WITH MASS SPECTROMETER AS PER SPEC.

TOLERANCES: UNLESS OTHERWISE NOTED	APPROVED FOR CONST.	JE	6-13-62	ARGONNE NATIONAL LABORATORY	A	STL WAS 304 SST	11-17-82 HWF
FRACTIONAL TOL. ± 1/64	CHIEF ENG.			This drawing is the property of the Argonne National Laboratory	SYM.	CHANGE	DATE BY
DECIMAL TOL. ± .005	CHIEF DRAFTSMAN				PART	CHAMBER	
ANGULAR TOL. ± 1/4°	CHECKED				ASSEMBLY	CHAMBER-ROTRON BLOWER	
SURFACES TO BE WITHIN LIMITS OF COMMON PRODUCTION METHODS ASA B46.1 FIG. 10 (1955)	DRAWN	RB	6-11-62	SCALE 1/2"=1" MATL. H.R. MILD STL (A) NO. REQD.	SHEET OF	DRWG. NO. CE-C4922-1	ITEM NO.
	GROUP	REPRESENTATIVE					

GLENNON & KERR, INC., CHICAGO



NOTE:

VACUUM TIGHT WELDS - HELIUM LEAK TEST WITH MASS SPECTROMETER

TOLERANCES: UNLESS OTHERWISE NOTED	APPROVED FOR CONST.		
FRACTIONAL TOL. ± 1/64	CHIEF ENG.		
DECIMAL TOL. ± .005	CHIEF DRAFTSMAN		
ANGULAR TOL. ± 1/4°	CHECKED		
SURFACES TO BE WITHIN LIMITS OF COMMON PRODUCTION METHODS ASA B46.1 FIG. 19 (1955)	DRAWN	NB	6/8/62
	GROUP	REPRESENTATIVE	

ARGONNE NATIONAL LABORATORY

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SCALE	MATL.	NO. REQD.
½" = 1"	HR. MILD STL (A)	

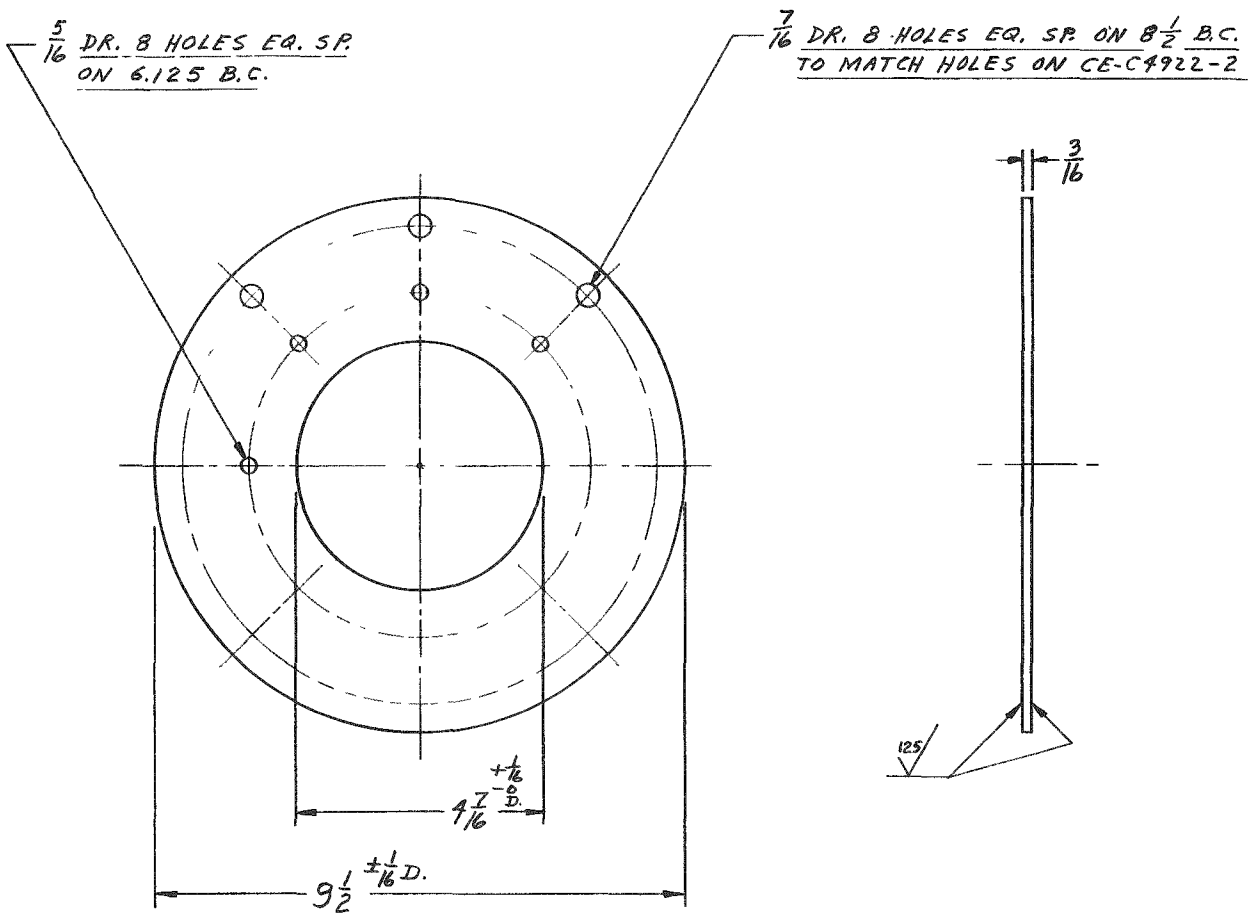
SHEET	DRWG. NO.	ITEM NO.
OF	CE-C4922-2	

SYM.	CHANGE	DATE	BY
A	STL WAS 304 SST	11-19-62	NB

PART TOP - CHAMBER

ASSEMBLY CHAMBER - ROTRON BLOWER

GLENNON & KERN, INC., CHICAGO

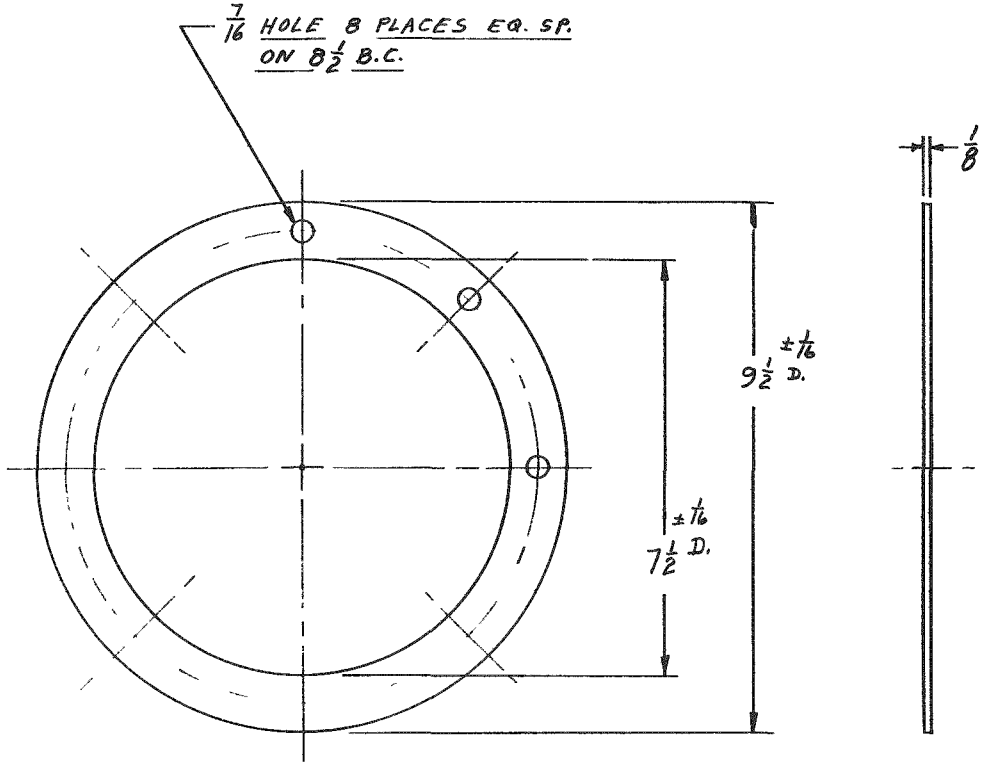


A	STL WAS 304 SST	11-19-62	HG				
SYM.	CHANGE	DATE	BY				

TOLERANCES: UNLESS OTHERWISE NOTED	APPROVED FOR CONST.	MF	6-13-62	ARGONNE NATIONAL LABORATORY			
FRACTIONAL TOL. ± 1/64	CHIEF ENG.			This drawing is the property of the Argonne National Laboratory			
DECIMAL TOL. ± .005	CHIEF DRAFTSMAN						
ANGULAR TOL. ± 1/4°	CHECKED			SCALE	MATL.	NO. REQD.	
SURFACES TO BE WITHIN LIMITS OF COMMON PRODUCTION METHODS ASA B46.1 FIG 19 (1955)	DRAWN	WB	6/11/62	1/2" = 1"	H.R. MILD STL (A)		
	GROUP	REPRESENTATIVE					

PART	PLATE - SUPPORT		
ASSEMBLY	CHAMBER - ROTRON BLOWER		
SHEET OF	DRWG. NO. CE-B4922-3	ITEM NO.	

GLENNON & KERN, INC., CHICAGO



TOLERANCES: UNLESS OTHERWISE NOTED	APPROVED FOR CONST.		6-13-62	ARGONNE NATIONAL LABORATORY	SYM.	CHANGE	DATE	BY
FRACTIONAL TOL. ± 1/64	CHIEF ENG.			This drawing is the property of the Argonne National Laboratory	PART GASKET			
DECIMAL TOL. ± .005	CHIEF DRAFTSMAN				ASSEMBLY CHAMBER-ROTRON BLOWER			
ANGULAR TOL. ± 1/4°	CHECKED				SCALE 1/2" = 1"	MATL. NEOPRENE	NO. REQD.	
SURFACES TO BE WITHIN LIMITS OF COMMON PRODUCTION METHODS ASA B46.1 FIG. 19 (1955)	DRAWN	RB	6/11/62		SHEET OF	DRWG. NO. CE-B4922-4	ITEM NO.	
	GROUP	REPRESENTATIVE						

GLENNON & KERN, INC., CHICAGO

1.0 LEAK TESTING FOR VACUUM SERVICE

Testing is to be done on items and assemblies as indicated and after all treatments where so indicated on the drawings (cleaning, heat treating, pickling, etc.) have been completed. All leaks found must be repaired. Prior to testing the repaired areas all treatments must be repeated unless specific permission to omit one or more of the steps is given in writing by the requestor.

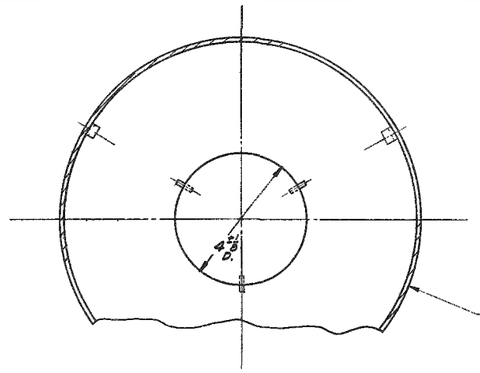
1.1 Leak Testing Method

Testing shall be performed using a mass spectrometer type helium leak detector which has been appropriately calibrated.

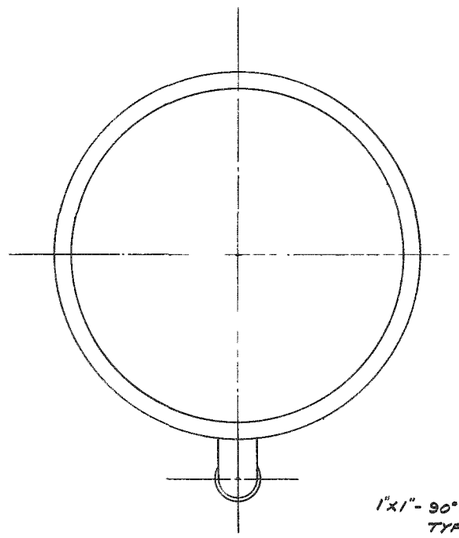
1.1.1 With the assembly evacuated to 50 microns, the entire exterior surface including connections is to be probed with a jet of helium. A leak rate in excess of 2×10^{-7} standard cubic centimeters per second of helium will be unacceptable.

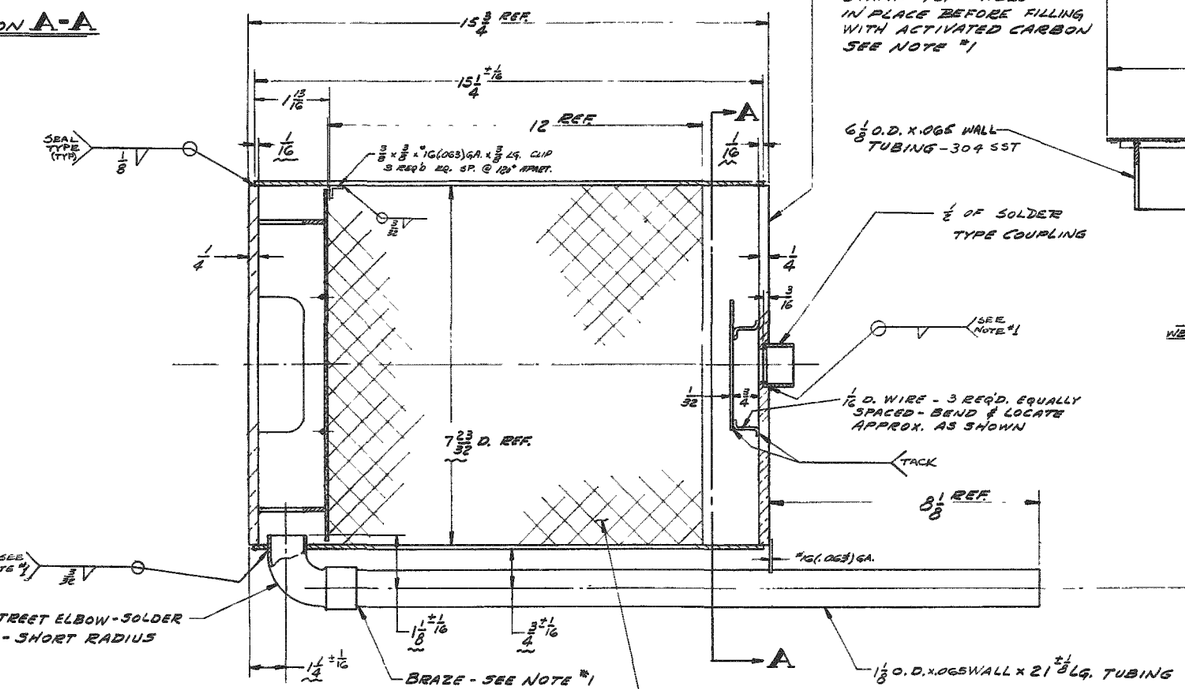
1.1.2 All welds designated as "seal type" on any detail or assembly drawing will be tested as specified above.

LEAK TESTING FOR VACUUM SERVICE	
Assembly: CHAMBER - ROTRON BLOWER	
Approved	Sheet 1 of 1
Drawing No.: CE-A-4922-5	



SECTION A-A





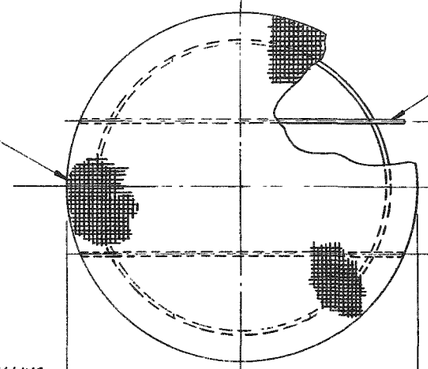
STAMP "TOP" - WELD IN PLACE BEFORE FILLING WITH ACTIVATED CARBON SEE NOTE #1

1/2 OF SOLDER TYPE COUPLING

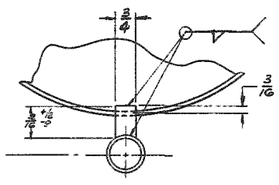
6 1/8 O.D. x .065 WALL TUBING - 304 SST

FILL TO DEPTH OF 12 ±1/8 WITH ACTIVATED CARBON - GRADE 5XC 8-14 MESH - NATIONAL CARBON Co. OR EQUIV. (FURNISHED & LOADED BY CEN)

304 SST WIRE CLOTH - DOUBLE CRIMP
CONST. - 14 MESH - .0286 D.
WIRE - .0428 SQ. OPENINGS
36% OPEN AREA



SUPPORT DETAIL
WELDED CONSTRUCTION
WELD WIRE CLOTH IN POSITION
SCALE - NONE



NOTES:
1. SEAL TYPE JOINT - TEST WITH MASS SPECTROMETER
2. CLEAN & DEGREASE
3. OPERATING PRESSURE - VACUUM @ 200°F.

TOLERANCES: FRACTIONAL TOL. ±1/32; DECIMAL TOL. ±.005; ANGLE ±1°; ALL SURFACES TO HAVE AVERAGE FINISH MICRO INCH OR AS RECEIVED FINISH UNLESS OTHERWISE NOTED	APPROVED FOR CONST.		ARGONNE NATIONAL LABORATORY	A	GENERAL REDESIGN WAS SHEET 2 OF 3	10/19/62	
	CHIEF ENG		This drawing is the property of the Argonne National Laboratory	SYM	CHANGE	DATE	BY
	CHIEF DRAFTSMAN			PART	ACTIVATED CARBON BED		
	CHECKED			ASSEMBLY	HELIUM GAS PURIFICATION SYSTEM		
	DRAWN	JK 11-3-61	SCALE 1/2	MATL. COPPER & NOTED	NO REQD.	SHEET OF	DRWG. NO. CE-D4796
	GROUP	REPRESENTATIVE					ITEM NO.