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DIELECTRIC TO METAL SEAL TECHNOLOGY STUDY

A.C. GRIMM AND P.D. STRUBHAR

RADIO CORPORATION OF AMERICA
ELECTRON TUBE DIVISION
INDUSTRIAL TUBE PRODUCTS
POWER TUBE OPERATIONS
LANCASTER, PENNSYLVANIA

THIRD QUARTERLY REPORT

CONTRACT AF 30(602) 2682

PREPARED FOR
ROME AIR DEVELOPMENT CENTER
AIR FORCE SYSTEMS COMMAND
UNITED STATES AIR FORCE
GRIFFISS AIR FORCE BASE
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CONTRACT AF 30(602) 2682
PROJECT 5573
TASK 557303

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ABSTRACT

This report covers the third quarter of work under a 19-month program to conduct a theoretical and experimental investigation leading to the development of improved dielectric-to-metal seals and sealing techniques for use with output windows for high-power microwave tubes.

A number of seal strength tests were performed to determine the effects of plating-metalizing combinations on seal strength. Various proportions of molybdenum and RCA S-641A metalizing mixtures as well as tungsten and RCA S-641A metalizing mixtures were tried on sapphire in order to find the optimum mixture. Available data indicate that the best combination is a 90% molybdenum-10% S-641A mixture fired at a metalizing temperature of 1750° Centigrade or higher.

A series of migration studies was made, using electron beam techniques, in an effort to better understand the migration of various elements in the seals and the effect of this migration on seal strength.

Efforts to metalize beryllium oxide with tungsten or molybdenum ink combinations will require more development to achieve success.

Compression-band window assemblies having dielectric discs made from beryllium oxide, boron nitride, magnesium fluoride, Pyroceram 9606, Pyroceram XM-1, and sapphire with a 60° crystal orientation were made and evaluated. Vacuum-tight window assemblies were made with all dielectrics except Pyroceram XM-1 and sapphire.

Tests have verified that compression bands fabricated from René 41, Waspalloy, or molybdenum will meet window design objectives. The choice of material for a specific window dielectric can, therefore, be based on thermal expansion considerations.

Experimental "butt-type" fused silica-to-metal seals have been successfully fabricated.

Title of Report RADC-TDR-63-249

PUBLICATION REVIEW

This report has been reviewed and is approved.

Approved: Paul M. Baldwin Lt Col USAF

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Acting Director
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Surveillance & Control

DIELECTRIC-TO-METAL SEAL
TECHNOLOGY STUDY

THIRD QUARTERLY INTERIM TECHNICAL REPORT
CONTRACT AF30(602)-2682

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INTRODUCTION

The objectives of the subject program are to conduct a theoretical and experimental investigation leading to the development of improved dielectric-to-metal seals and sealing techniques for use with output windows for high-power microwave tubes.

The investigation is being conducted in accordance with the requirements defined in Exhibit "A" entitled "Purchase Request Continuation Sheet for Dielectric-to-Metal Seal Technology Study", dated 12 October 1961, and as amended by Engineering Change A, dated 12 September 1962. The investigations to be conducted during the course of this contract are the development of a refractory metal sealing process for synthetic sapphire and beryllium oxide; an active metal sealing process for fused silica and Pyroceram; a compression-band technique for making window assemblies from beryllium oxide, boron nitride, magnesium fluoride, Pyroceram, and sapphire; and a "balanced-type" tapered fused silica-to-metal seal. In addition, evaluation tests will be conducted on completed window assemblies to determine vacuum tightness, mechanical strength, maximum possible bake temperature, electrical losses as required, and seal reliability when cycle life tested.

CONCLUSIONS

Test results for dielectric materials tested to date indicate that vacuum-tight compression-band window assemblies can be fabricated from beryllium oxide, magnesium fluoride, Pyroceram 9606, and boron nitride. Double vacuum bake test results have shown that beryllium oxide and Pyroceram 9606 can be successfully baked at 700° Centigrade. Optimum sealing parameters have been determined for fused silica-to-copper "butt-type" seals and two vacuum-tight seals have been fabricated using this configuration.

RECOMMENDATIONS

Materials research and the fabrication of experimental seals have crystallized the approaches to be used in successfully meeting objectives of the study. No recommendations regarding the scope or detailed aspects of the study appear necessary at this time.

DISCUSSION

The work performed during this report period was devoted to the development of metalized dielectric-to-metal seals and the measurement of their strength, evaluation of component parts for compression-band type seals, and fabrication of fused silica-to-metal seals.

I. Sapphire-Refractory Metal Seals

A. Molybdenum Metalizing

Work performed during the third quarter on seals using molybdenum-base metalizing consisted of the evaluation and verification of the metallographic information collected during the second quarter and the performance of additional tests to determine the effects of plating material selections on seal strengths.

Metallographic work performed prior to this period produced unsatisfactory results when 100% molybdenum metalizing ink (a combination of metals, additives, binders, and vehicles processed into a homogeneous mixture) was used. During this report period the molybdenum ink was combined with various percentages of RCA S-641A powder.

A strong reaction zone at the sapphire-metalizing interface and an open structure in the metalized area were obtained when 20% (by weight) of RCA S-641A powder was added to the molybdenum ink. The best structure metallographically was obtained at a 1500° Centigrade metalizing temperature.

A discernable reaction zone having a good appearing structure in the metalized area was obtained in tests with molybdenum ink containing 10% RCA S-641A powder (by weight).

Molybdenum ink containing 5% RCA S-641A powder (by weight) failed to form a sintered metal layer at 1500° Centigrade. Subsequent tests showed that for this mixture sintering of the metalizing mixture particles into a solid metallic layer became adequate at temperatures in excess of 1600° Centigrade (on either a sapphire or molybdenum metal substrate) and produced an excellent structure in the metalized area. No evidence of reaction between this layer and the sapphire was noted until a temperature of 1800° Centigrade was attained (see Table I).

Actual seal strength evaluations were initiated at the conclusion of the metalizing adherence tests.

The metalizing strength test procedure was standardized by using 1-inch long test bars with a cross section approximately 0.070 inch x 0.070 inch. These bars were cut and ground from 1/4-inch sapphire rods after brazing. This procedure was used to eliminate metalizing, plating, or brazing variations encountered in material near the perimeter of the processed specimen. The specimens were broken in a modulus of rupture jig mounted on a 0 to 30-pound load cell in a conventional testing machine. Both the width and length of the test specimens were measured to 0.0001 inch and the areas calculated to the nearest 0.0005 inch.

Observation of the metallographic specimens indicated that (1) decreases in the amount of RCA S-641A powder added to molybdenum ink increased the strength of the sintered metalized layer, and (2) increases in the amount of RCA S-641A added to molybdenum ink probably increased the strength at the sapphire-metalizing layer interface. This would indicate that there is an optimum addition of powder for highest overall strength. To verify these observations, a series of tests was performed to determine which molybdenum-

TABLE I. METALIZING ADHERENCE TESTS

SAPPHIRE-MOLYBDENUM-S-641A MIXTURES

SINTERED IN WET FORMING GAS - 90% N_2 - 10% H_2 - 18° ± 2° Dew Point

CuBraze

Metalizing Mixture	Metalizing Temperature	Scrape Test	Metallographic Examination	Remarks
80% Moly 20% S-641A	1500° C	Excellent	Good to Excellent	Good reaction probable; some braze penetration
80% Moly 20% S-641A	1600° C	Good	Fair	
80% Moly 20% S-641A	1700° C	Poor	-	Failed at plating
80% Moly 20% S-641A	1800° C	Good	Poor	Bad braze penetration
90% Moly 10% S-641A	1600° C	Good	Good	
90% Moly 10% S-641A	1700° C	Good	Excellent	
90% Moly 10% S-641A	1800° C	Good	Fair	Appears over-reacted
95% Moly 5% S-641A	1600° C	Fair	Fair	Not perfectly sintered
95% Moly 5% S-641A	1700° C	Good	Good	
95% Moly 5% S-641A	1800° C	Good	Excellent	Very dense layer

S-641A combination gave the greatest seal strength. A summary of the test results is given in items 1, 2, and 3 of Table II.

It became apparent during these tests that the location and appearance of the ruptured area as well as the calculated breaking strength of the specimen were of great importance. For example, the specimens made with 80% molybdenum - 20% RCA S-641A metalizing ink invariably broke in the metalizing layer, although the pressure necessary to break them varied between 26,500 psi to 54,000 psi. Whenever random breakage occurred, the break was caused by a high modulus value or a recognizable defect in the specimen. Data from defective specimens and breaks in the sapphire under 35,000 psi were discarded. Low pressure breaks in the sapphire were attributed to imperfections on the edges of the specimens or to internal strains set up in firing.

Specimens made with 90% molybdenum - 10% RCA S-641A metalizing ink showed a random breakage in the sapphire and at the sapphire-metalizing interface. However, no breakage occurred in the metalizing layer. It was concluded that in all cases the metalizing layer's strength was at least comparable to the strength of the sapphire, but the strength of the metalizing bond was not.

A reduction in the amount of RCA S-641A powder added to the metalizing ink to obtain a mixture of 95% molybdenum - 5% S-641A and an increase in the metalizing temperature produced results similar to those obtained with the 90% molybdenum - 10% metalizing ink combination. However, breaks occurred more frequently at the sapphire-metalizing interface.

Since it had been established that the 80% molybdenum - 20% S-641A metalizing ink did not produce promising results regardless of temperature, work with this combination was discontinued. To determine

TABLE II. MOLYBDENUM-S-641A METALIZING STRENGTH

Item	Metalizing Mixture	Metalizing Temperature	Plating	Braze Material	Modulus of Rupture (psi) Avg	Hi	Lo	Failure Location
1	80% Moly 20% S-641A	1500° C	Nickel	37.5 Au 62.5 Cu	36,300	54,000	26,500	Metalizing layer
2	90% Moly 10% S-641A	1700° C	Nickel	37.5 Au 62.5 Cu	43,700	48,000	39,000	Random 1. Sapphire 2. Sapphire-metalizing interface
3	95% Moly 5% S-641A	1800° C	Nickel	37.5 Au 62.5 Cu	41,900	47,500	36,300	Random 1. Sapphire-metalizing interface 2. Sapphire
4	90% Moly 10% S-641A	1600° C	Nickel	37.5 Au 62.5 Cu	40,400	45,600	39,000	Random 1. Sapphire-metalizing interface 2. Sapphire
5	90% Moly 10% S-641A	1750° C	Nickel	37.5 Au 62.5 Cu	47,000	51,400	37,900	Sapphire
6	92.5% Moly 7.5% S-641A	1750° C	Nickel	37.5 Au 62.5 Cu	34,500	49,300	12,800	Random 1. Metalizing and metalizing-sapphire interface
7	92.5% Moly 7.5% S-641A	1800° C	Nickel	37.5 Au 62.5 Cu	42,300	46,300	39,600	Sapphire

whether the optimum mixture combination for the best strength might lie between 90%-10% and 95%-5%, a 92-1/2% molybdenum - 7-1/2% S-641A metalizing ink was tested.

Items 3, 4, 5, 6, and 7 of Table II list the results obtained in tests of 90%-10% to 95%-5% combinations fired at temperatures from 1600° Centigrade to 1800° Centigrade. The following observations were made during these tests.

1. The number of sapphire-metalizing interface breaks increased when the seals were fired at 1600° Centigrade, and the average strength decreased.
2. The number of sapphire metalizing interface breaks decreased when the seals were fired at 1750° Centigrade, and the average strength increased.
3. The 92-1/2% molybdenum - 7-1/2% S-641A ink combination produced unsatisfactory results when fired at 1750° Centigrade. Although some specimens broke in the sapphire, most of the specimens broke at the sapphire-metalizing interface and within the metalized layer. It was concluded that the interface and the metalized layer were weak. Breaks in both areas were eliminated when the firing temperature was increased to 1800° Centigrade.

The work performed to date on molybdenum ink with an S-641A powder additive indicates that the best combination is between 90% molybdenum - 10% S-641A and 95% molybdenum - 5% S-641A fired at a temperature of at least 1750° Centigrade. Of the three combinations tried, the 90% molybdenum - 10% S-641A metalizing ink seems most promising. Tests to establish its acceptability will continue.

Another variable remaining to be investigated is the duration of the metalizing firing cycle. To date, a firing cycle of one-half hour has been used. Information presented in this report indicates that longer firing cycles may produce better results.

B. Tungsten Metalizing

Attempts to metalize sapphire with 100% tungsten ink were unsuccessful. Metallographic sections could not be obtained for study.

A decision to add S-641A powder to tungsten ink was made as a result of the success achieved with the addition of S-641A powder to molybdenum inks. The results were initially unfavorable; however, metallographic samples were made and evaluated. The structure of the metalized coatings was inferior to that obtained with molybdenum ink under the optimum conditions.

Table III summarizes the results of the metallographic examination of seals made with combinations of tungsten and S-641A powders. During the metallographic examination it was noted that the structures closely resembled those obtained with molybdenum ink; however, evidences of a reaction appeared only at a firing temperature 100° Centigrade higher than used on molybdenum ink. Very little evidence of reaction was noted with the 95% tungsten - 5% S-641A metalizing ink, although the samples appeared excellent in scrape tests.

Modulus of rupture test bars were prepared using the same procedure as that used for molybdenum ink. Firing temperatures were selected on the basis of the metallographic specimens. The modulus of rupture test results are summarized in items 1, 2, and 3 of Table IV.

It is apparent, after comparing items 1 and 2 of Table II and items 1 and 2 of Table IV, that the 80% tungsten - 20% S-641A and 90%-10%

TABLE III. METALIZING ADHERENCE TESTS, SAPPHIRE-TUNGSTEN-S-641A POWDER

Fired in 90% N_2-10% H_2 (Wet Forming Gas)

Average Dew Point 18° C

Nickel Plated, 37.5 Au-62.5 Cu Braze

Metalizing Mixture	Metalizing Temperature	Scrape Test	Metallographic Examination	Remarks
80% Tungsten 20% S-641A	1500° C	Good	Poor	Poor structure
90% Tungsten 10% S-641A	1500° C	Good	Poor	Poor structure
95% Tungsten 5% S-641A	1500° C	Failed	-	No sintering of tungsten. Evidence of attack on sapphire
80% Tungsten 20% S-641A	1700° C	Excellent	Very Poor	Over-reacted
80% Tungsten 20% S-641A	1800° C	Excellent	Very Poor	Over-reacted
90% Tungsten 10% S-641A	1700° C	Excellent	Poor	OK except badly blistered braze
90% Tungsten 10% S-641A	1800° C	Excellent	Poor	Badly blistered braze area.
95% Tungsten 5% S-641A	1700° C	Excellent	Poor	Poor structure, blistered
95% Tungsten 5% S-641A	1800° C	Excellent	Poor	Poor structure

TABLE IV. PLATING - BRAZING TESTS, SAPPHIRE-TUNGSTEN-S-641A POWDER

Fired in 90% N_2-10% H_2 (Wet Forming Gas)

Average Dew Point 18° C

Item	Metalizing Mixture	Metalizing Temperature	Plating	Braze Material	Modulus of Rupture (psi) Avg.	Hi	Lo	Failure Location
1	80% Tungsten 20% S-641A	1500° C	Nickel	37.5 Au 62.5 Cu	25,900	33,900	17,800	Metalizing layer
2	90% Tungsten 10% S-641A	1700° C	Nickel	37.5 Au 62.5 Cu	34,300	44,500	20,300	Metalizing layer
3	95% Tungsten 5% S-641A	1800° C	Nickel	37.5 Au 62.5 Cu	19,750	34,600	5,300	Metalizing-plating interferen
4	90% Tungsten 10% S-641A	1750° C	Chro-mium	Nioro	34,500	39,700	30,000	Random 1. Sapphire 2. Metalizing
5	90% Tungsten 10% S-641A	1800° C	Chro-mium	Nioro	40,200	49,700	25,100	Random 1. Sapphire 2. Sapphire-metalizing interface
6	90% Tungsten 10% S-641A	1800° C	None	37.5 Ag 62.5 Cu	35,500	41,000	29,200	Random 1. Sapphire-metalizing interface 2. Sapphire

tungsten ink mixtures failed at pressures approximately 10,000 psi lower than the corresponding molybdenum metalized samples. The 95% tungsten - 5% S-641A metalizing ink failed at a much lower pressure and nearly always at the interface of the metalized layer and braze. Chronic blistering was also encountered with tungsten inks.

During these tests a braze bead was often found adjacent to the larger blisters. Whether the blisters were caused by the braze bead or vice versa is yet to be determined. Brazing experiments were conducted to determine whether the braze disc was a factor. Since, in the fabrication of the test bars for the modulus of rupture determinations the perimeter of the 1/4-inch test rod was ground down 0.060 inch, three brazing disc diameters could be used: undersize discs of 0.230-inch diameter, normal size discs of 0.250-inch diameter, and oversize discs of 0.300-inch diameter. Samples fired with the oversize discs blistered badly at the bead. Samples fired with the undersize discs did not blister. Samples fired with the normal size discs blistered only when there was a streak of metalizing on the perimeter as a result of contact between the sample and firing boat during the previous metalizing firing. Because of the small number of samples processed, it was impossible to prove conclusively that the disc size caused the blistering; however, since the adoption of undersize discs, blistering has not recurred.

Iron plating, chromium plating, and the omission of plating with Nioro braze alloy were investigated to check the effect of plating on tungsten ink. Tests are still in progress; however, preliminary results indicate that chromium plating with a Nioro braze offers the best possibility for success. Complete test results are given in Table IV.

A definite pattern is yet to be established for the use of RCA S-641A powder with tungsten for metalizing sapphire. All tests with tungsten indicate that it is more difficult to acquire a strong bond at the sapphire-metalizing interface and a strong metalized layer than with molybdenum. It also appears that tungsten inks require substantially higher firing temperatures than molybdenum inks. The tungsten particle size is suspected of being an important factor in the strength developed.

C. Migration in Seals

In the fabrication of ceramic-metal seals, many variations have been observed when using similar techniques for metalizing different ceramics. Small amounts of elements other than the main constituent of the ceramic and the main constituent of the metalized layer seem to be necessary or helpful. In the case of alumina ceramics, for example, the difficulty of metalizing tends to increase with the purity of the ceramic when a single constituent metalizing layer is used. Results were poor until additives were introduced into the molybdenum and tungsten metalizing layers. It is assumed that the addition of magnesium and silicon in the form of a "steatite", which occurs with the use of RCA S-641A powder, must do two things: (1) it must aid in strengthening the metalizing layer, and (2) it must strengthen or promote the bond between the metalizing layer and the sapphire.

Observations made during this study contract have shown that a reaction zone with the sapphire is probably caused by the presence of the S-641A powder. At a given temperature, the reaction zone seems to increase when the percentage of S-641A powder in the metalizing ink is increased. It was also observed that there is an optimum range of S-641A powder percentages that aids in strengthening the metalizing layer at various metalizing temperatures.

To understand the effects observed, it was decided to make migration studies using electron probe techniques.[1] Because no data on seals using this technique were available to RCA, it was decided to scan sapphire seals and other ceramic seals.

The electron probe microanalyzer used to perform scans had a probing depth of 1 to 2 microns and a beam width of 1 to 3 microns. Figures 1 through 15 are charts of the data recorded on the microanalyzer. There are several precautions to be taken in interpreting the charts, namely:

1. The boundaries of the ceramic-metalizing layer and braze were determined by optical inspection of the location of the electron beam. Therefore, the charts are marked at the visual transition points. These may or may not coincide with the true chemical interface.

2. Where very small quantities of an element are indicated, the existence of the element is in doubt unless it exceeds twice the square root of 1 percent of the full-scale frequency reading of the chart. (Example: $\sqrt{.01 \times 1000} = 3.3$; any reading over 6.6 would be considered a trace of the element.)

3. Intensities of an element cannot be compared quantitatively in different materials since the intensity reading is influenced by the surrounding material.

4. The sapphire seal (Figures 9, 10, and 11) was cracked while being mounted, and as a result, the scan included

[1] Electron Beam Probe manufactured by Advanced Metals Research Company, Somerville, Massachusetts.

an intrusion of glass-fiber (silicon) filled diallyl phthalate used in mounting the specimen. It is for this reason that the silicon trace trails off the chart.

It is of interest that none of the seals scanned showed evidence of molybdenum migration into the braze or ceramic (Figures 4, 8, 11, and 15). This probably indicates that the reaction of the migrating materials with molybdenum is slight and occurs at the surface. Figures 1, 2, and 3 show scans of a molybdenum metalized seal of Frenchtown No. 4462 ceramic. The metalizing firing temperature on this seal was approximately 1520° Centigrade. Although the specific lot of ceramic material used in this particular seal was not tested, this ceramic-metalizing combination has consistently shown seal strengths of at least 60,000 psi modulus of rupture. Figure 1 shows a probable migration of aluminum from the ceramic into the metalizing layer. Figures 2 and 3 show very definite migration of silicon and manganese from the ceramic into this layer. The results of the electron probe microanalysis shows that diffusion of Si, Mn, and Al has occurred into the molybdenum layer. In fact, Si peaks in the layer and Mn tends to do so also.

Two different types of metalizing were used in the seal scanned in Figures 5, 6, 7, and 8. The Coors AD-94 ceramic which was used contains both magnesium and silicon. Coors AD-94 ceramics usually have seal strengths of 45,000 psi modulus of rupture when metalized with molybdenum at 1500° Centigrade.

To determine the effect of adding S-641A powder on a body containing magnesium and silicon, one side of the seal was metalized with molybdenum ink containing 10% S-641A. The aluminum trace (Figure 5) shows that there was more aluminum migration into the 90% molybdenum - 10% S-641A metalizing ink layer than into the 100% molybdenum metalizing ink layer. The slight rise of the aluminum trace

in the 100% molybdenum metalized layer is probably due solely to background "noise".

The silicon and magnesium traces, Figures 6 and 7, show no significant difference between metalized layers of different compositions. This might indicate that there is an optimum amount of silicon and magnesium that can diffuse into a molybdenum metalized layer and that the addition of 10% S-641A powder supplied this optimum quantity. There is some indication that the 90% molybdenum - 10% S-641A metalized layer is richer in magnesium than the 100% molybdenum layer.

Figures 9 through 15 are scans of two seals attempted with sapphire on one side and Wesgo Al-995 on the other. One seal used a 100% molybdenum metalized layer; the other used a 90% molybdenum - 10% S-641A powder metalized layer. The metalizing temperature for these seals was 1700° Centigrade.

Wesgo Al-995 ceramic seals metalized with 100% molybdenum usually have a 20,000 psi modulus of rupture. Wesgo Al-995 ceramic seals when metalized with the 90% molybdenum - 10% S-641A mixture usually break (in the ceramic) at pressures above 40,000 psi.

Comparison of the aluminum distribution curves for Wesgo Al-995 ceramic metalized with 100% molybdenum (Figure 9) and the same ceramic metalized with the 90% molybdenum - 10% S-641A mixture (Figure 12) indicates that there may be more aluminum crossing into the metalized layer in the 90%-10% specimen.

The silicon distribution (Figure 10) in the 100% molybdenum layer is puzzling. There is a heavy concentration of silicon at the metalizing braze layer, a concentration through the braze, and a concentration

in the metalizing on the sapphire side. No silicon should be present in any of these areas (note that the "black layer" on the sapphire side is resin). This is possibly caused by contamination of the braze or improper operation of the scanner. Although these conditions cannot be ruled out, the scanner showed approximately the trace expected at "dark phase", which is a high silica grain boundary. The braze disc used was cut from the same sheet as two other brazes used in this series and processed in the same manner. Thus, this unusual silicon migration may actually take place.

Another problem is presented by Figure 13. The screening of metalizing ink on the Wesgo Al-995 and sapphire was done at exactly the same time but the silicon traces indicate a definite difference in silicon concentration. Furthermore, a trace of silicon is indicated in the sapphire. This may mean that silicon can diffuse out of the 90% molybdenum - 10% S-641A metalizing layer and that it diffuses more easily in a body that contains silicon.

Figure 12 shows slight aluminum diffusion of the 90% molybdenum - 10% S-641A metalizing; Figure 9 shows no diffusion of the 100% molybdenum metalizing. Figure 14 (magnesium distribution) shows possible diffusion in the sapphire and probably no diffusion in the Wesgo Al-995 ceramic.

II. Beryllium Oxide - Refractory Metal Seals

A number of seals have been made using both molybdenum and tungsten inks of various compositions in an effort to metalize beryllium oxide. Further work is required to secure significant results.

III. Reactive Metal Seals

The equipment necessary to perform studies on reactive metal seals has been set up and actual seal studies will start in the next quarter.

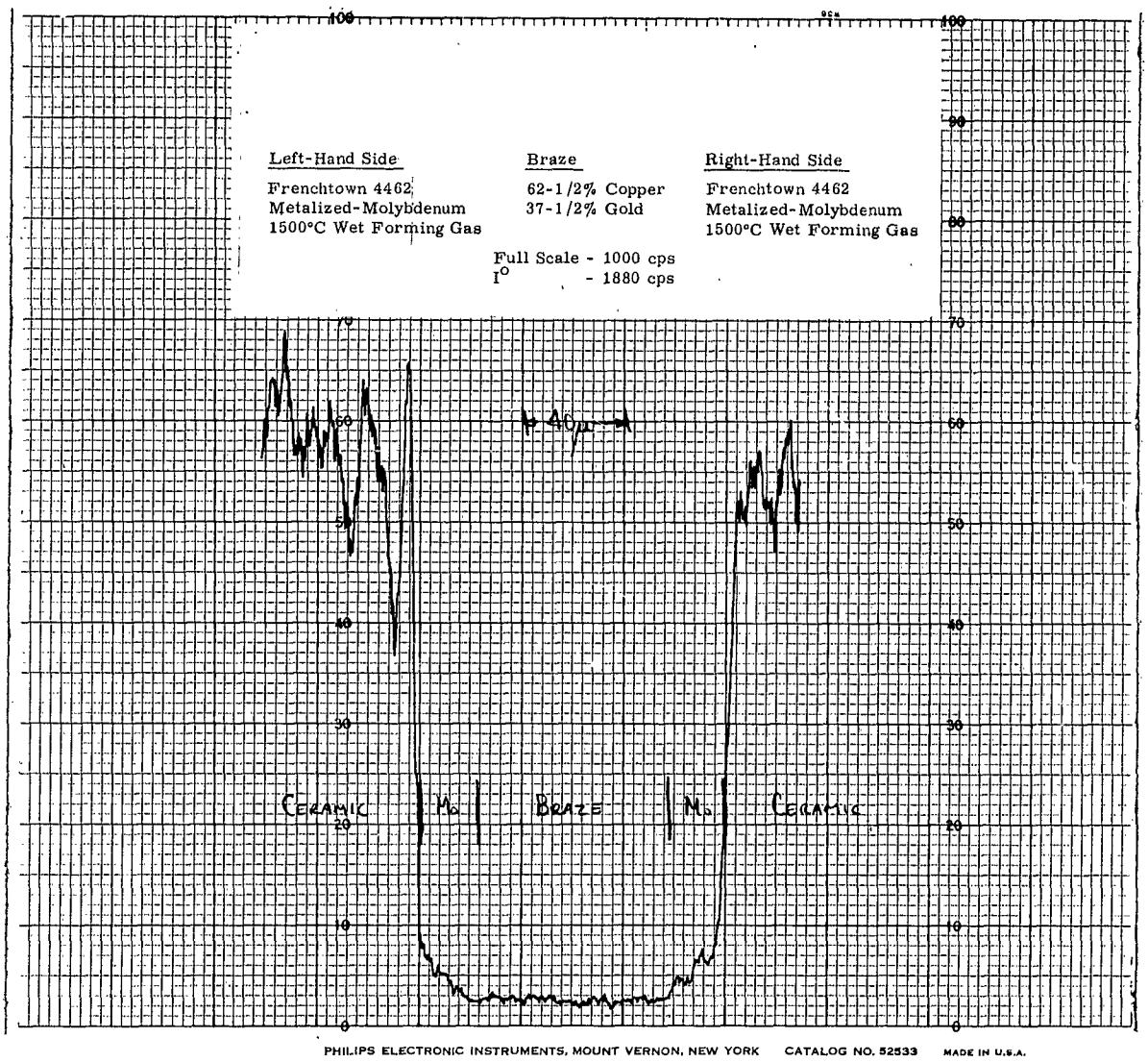


FIGURE 1 - ALUMINUM DISTRIBUTION IN
MOLYBDENUM METALIZED FRENCHTOWN 4462 CERAMIC SEAL

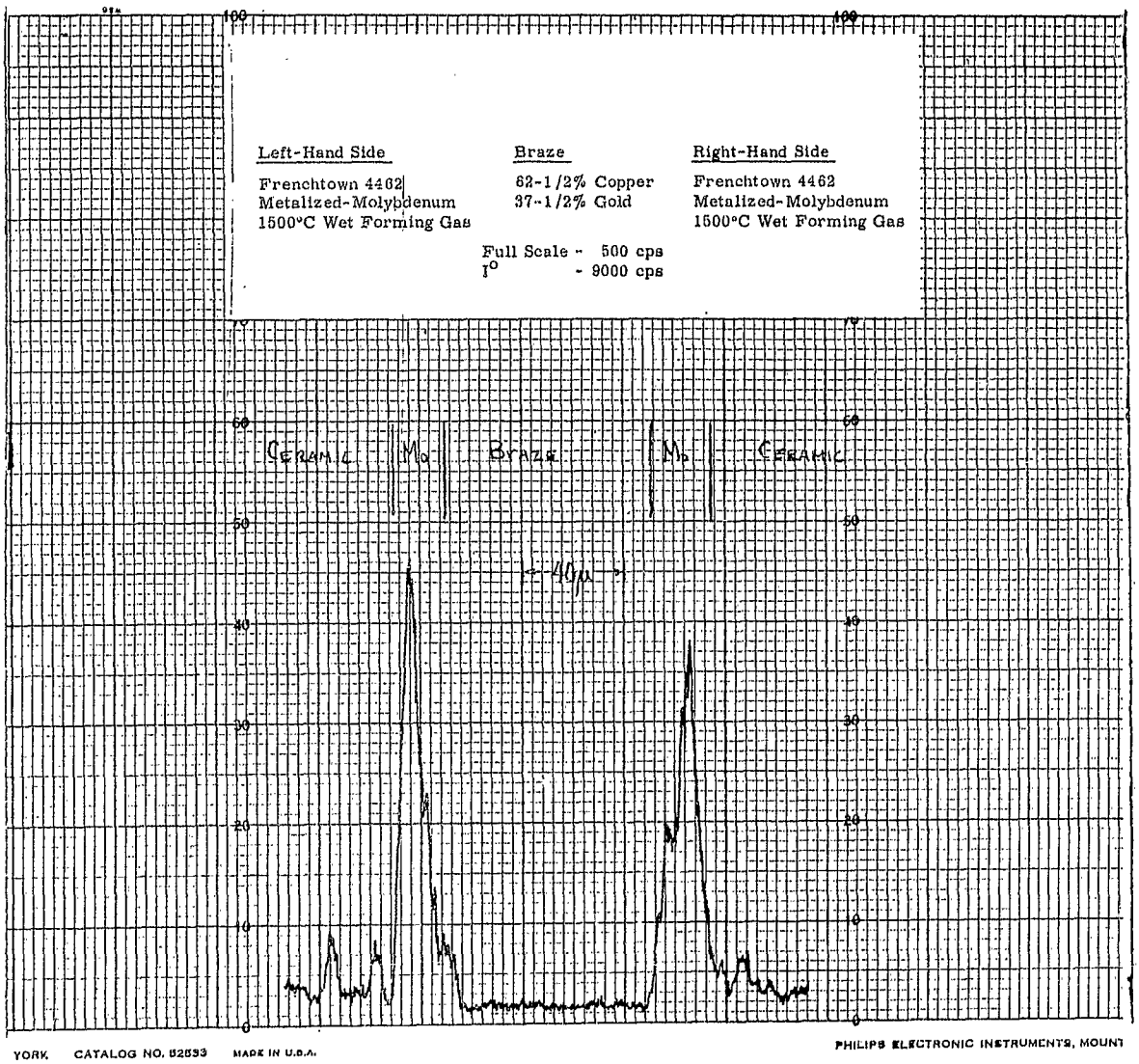


FIGURE 2 - SILICON DISTRIBUTION IN
MOLYBDENUM METALIZED FRENCHTOWN 4462 CERAMIC SEAL

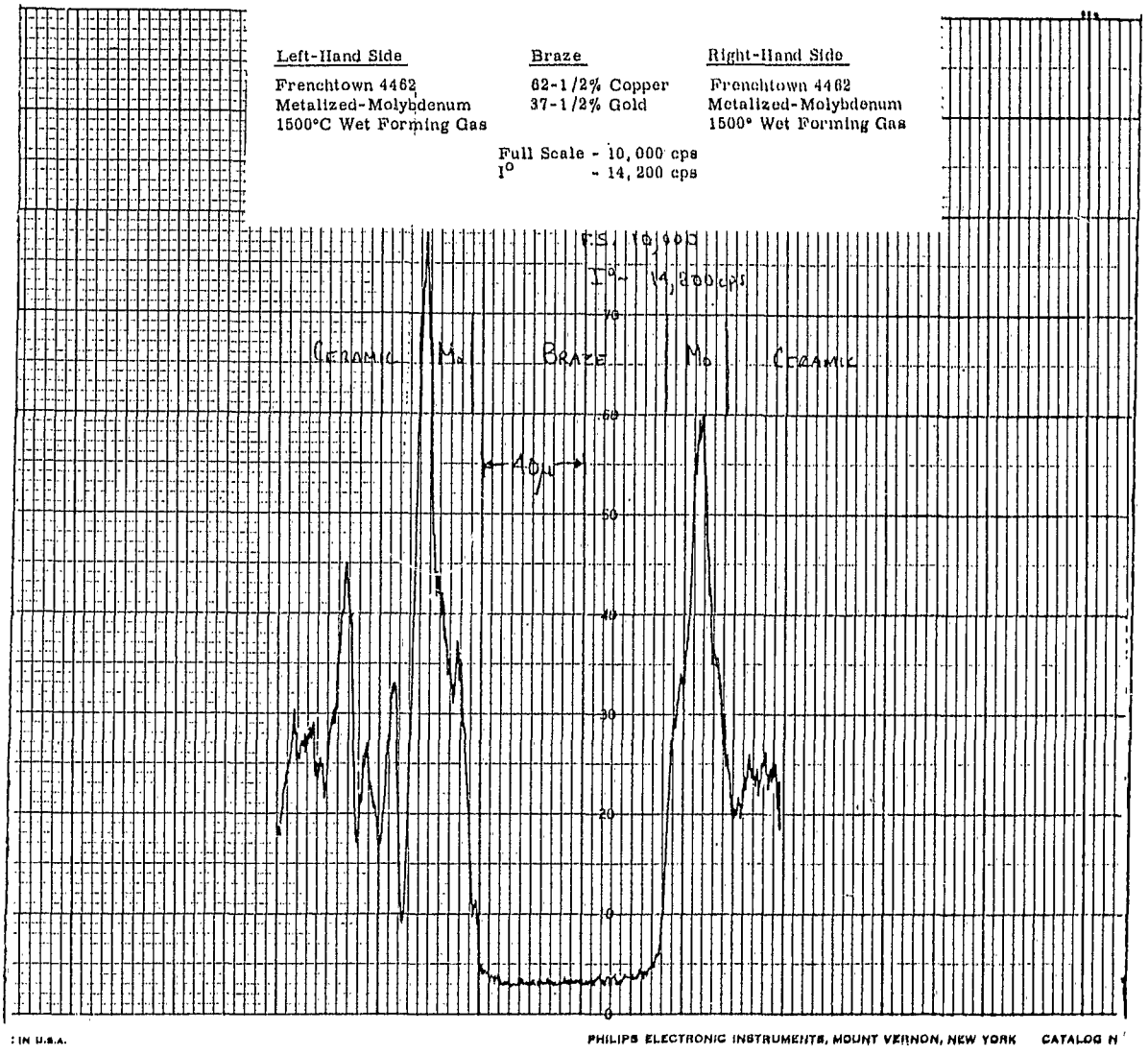


FIGURE 3 - MANGANESE DISTRIBUTION IN
MOLYBDENUM METALIZED FRENCHTOWN 4462 CERAMIC SEAL

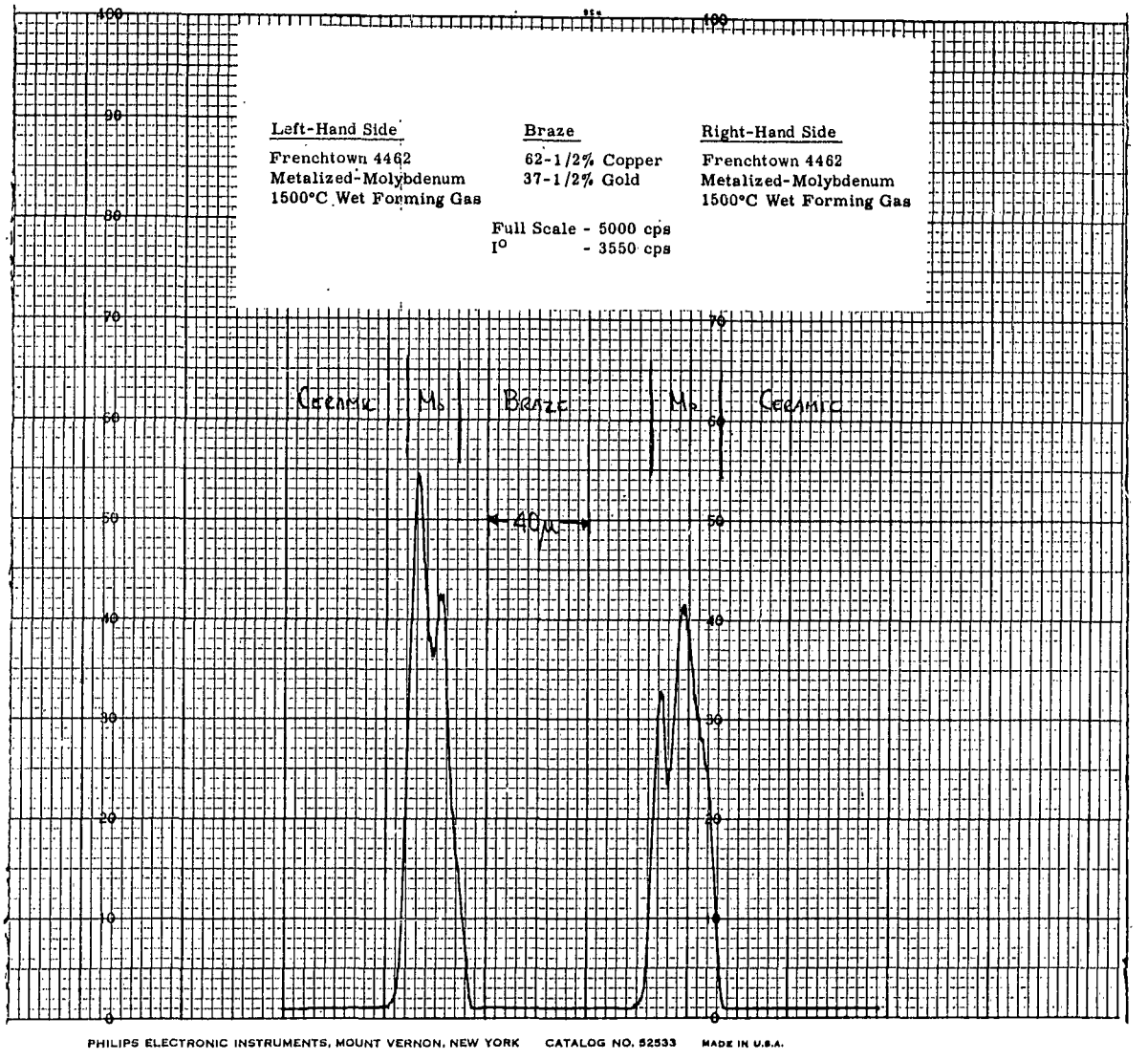


FIGURE 4 - MOLYBDENUM DISTRIBUTION IN
MOLYBDENUM METALIZED FRENCHTOWN 4462 CERAMIC SEAL

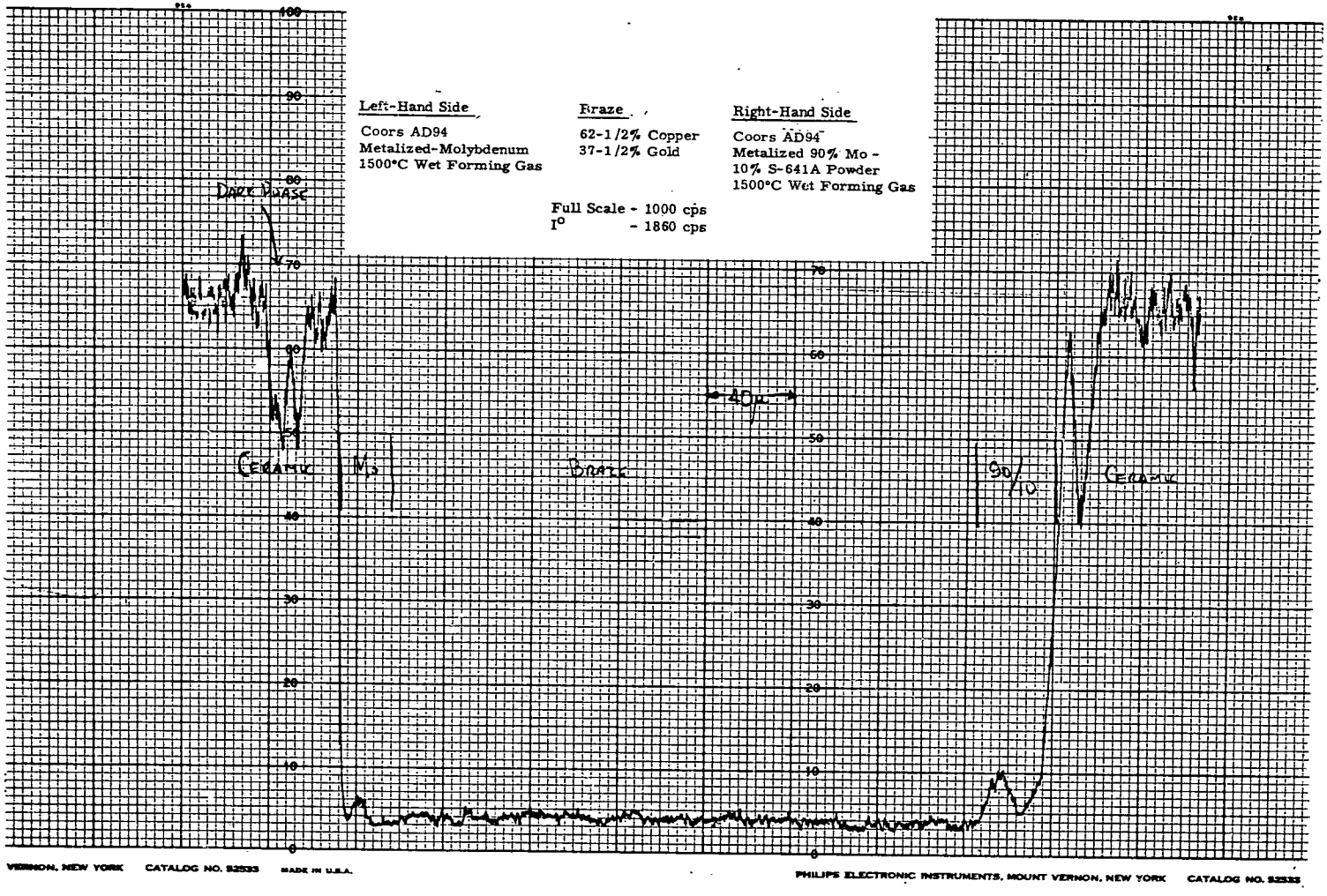


FIGURE 5 - ALUMINUM DISTRIBUTION IN
MOLYBDENUM METALIZED COORS AD94 CERAMIC SEAL

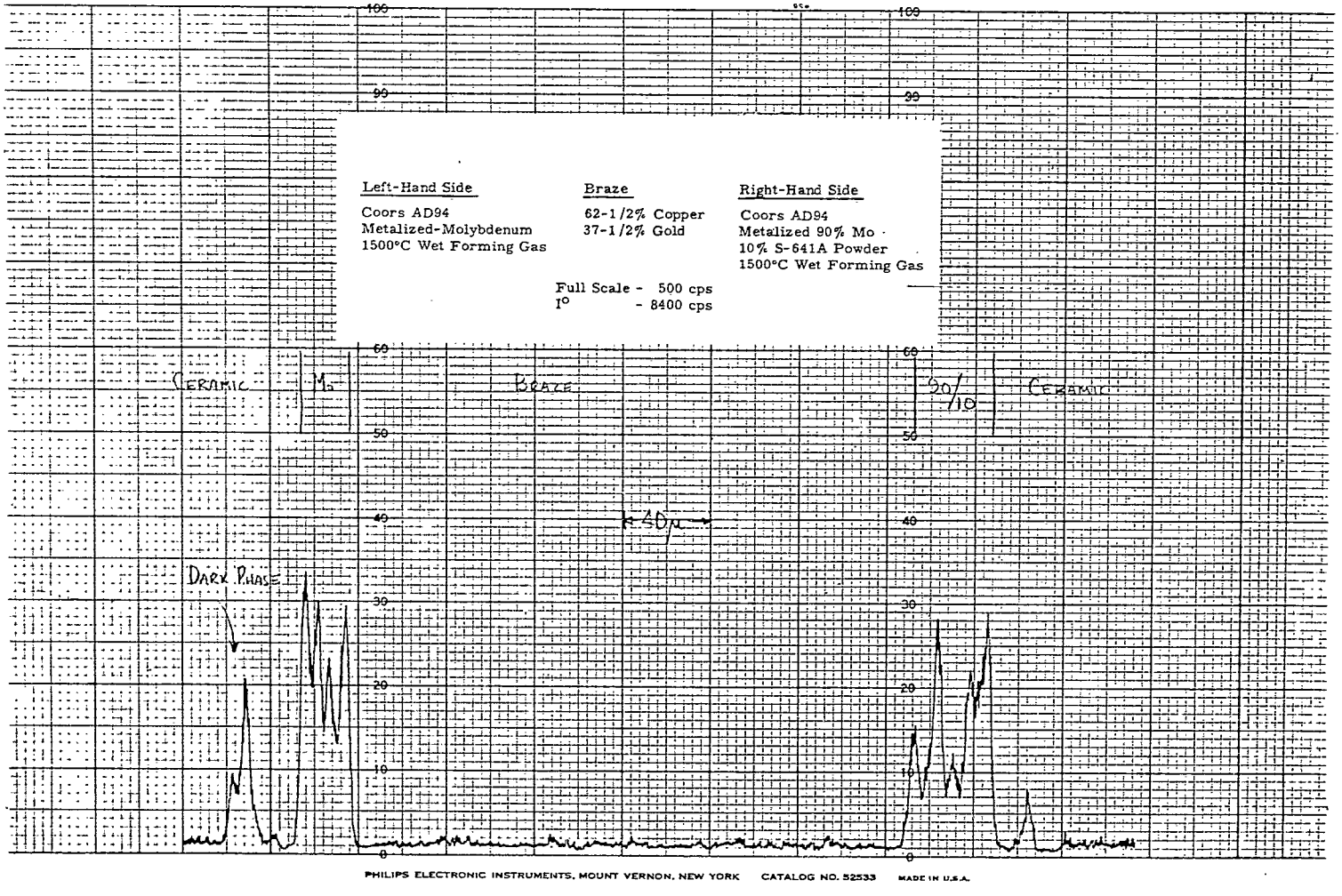


FIGURE 6 - SILICON DISTRIBUTION IN
MOLYBDENUM METALIZED COORS AD94 CERAMIC SEAL

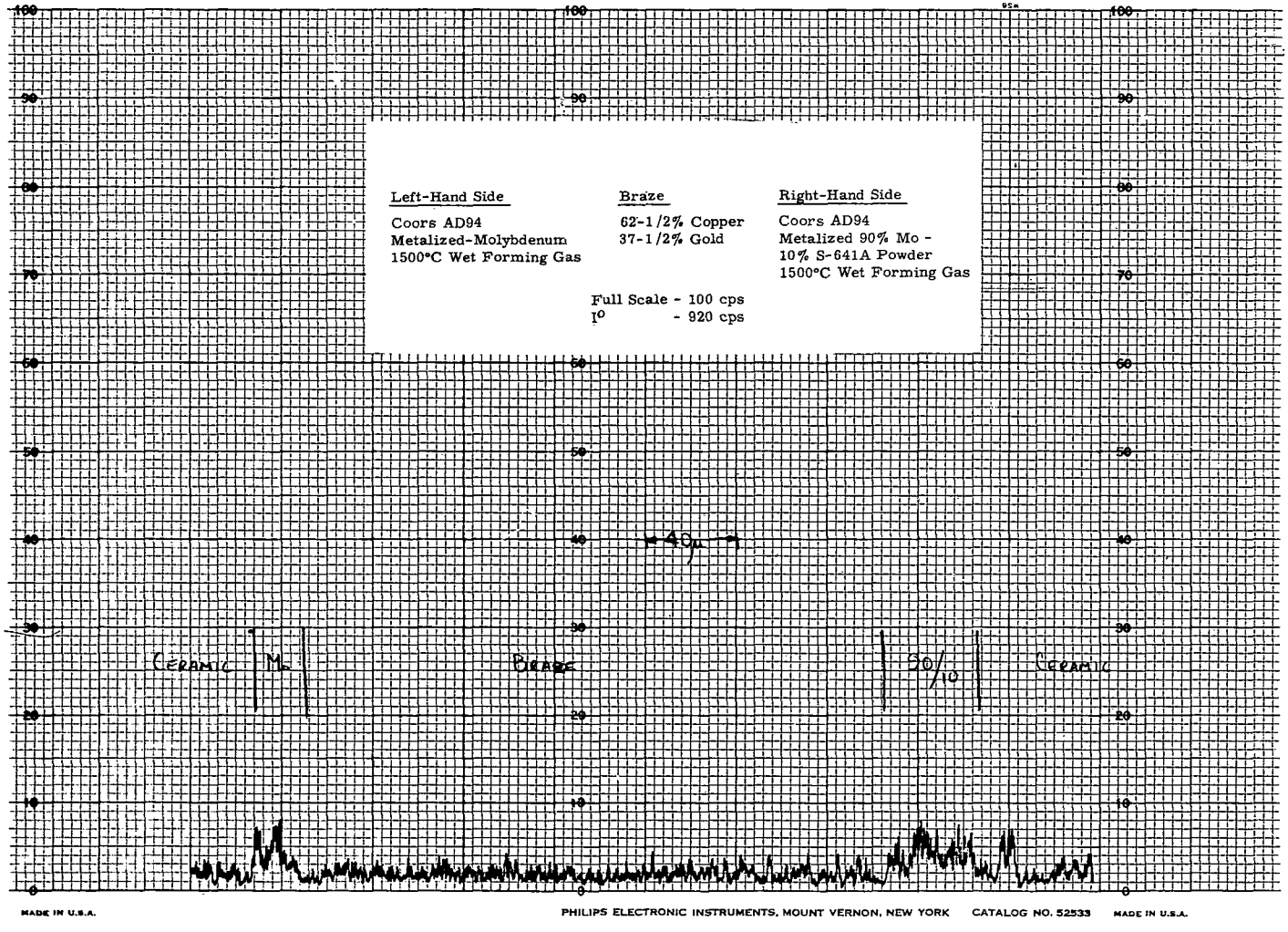


FIGURE 7 - MAGNESIUM DISTRIBUTION IN
MOLYBDENUM METALIZED COORS AD94 CERAMIC SEAL

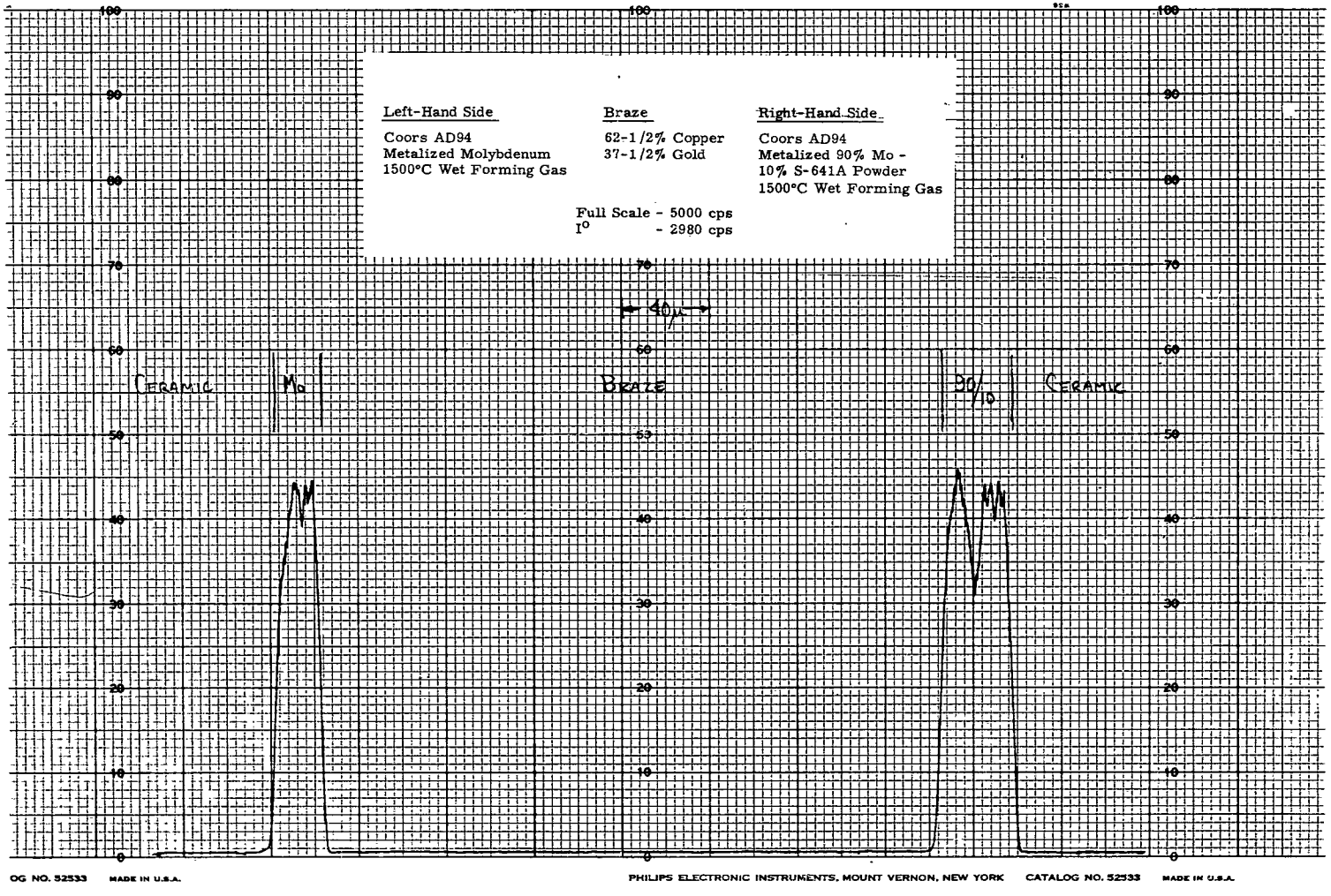


FIGURE 8 - MOLYBDENUM DISTRIBUTION IN
MOLYBDENUM METALIZED COORS AD94 CERAMIC SEAL

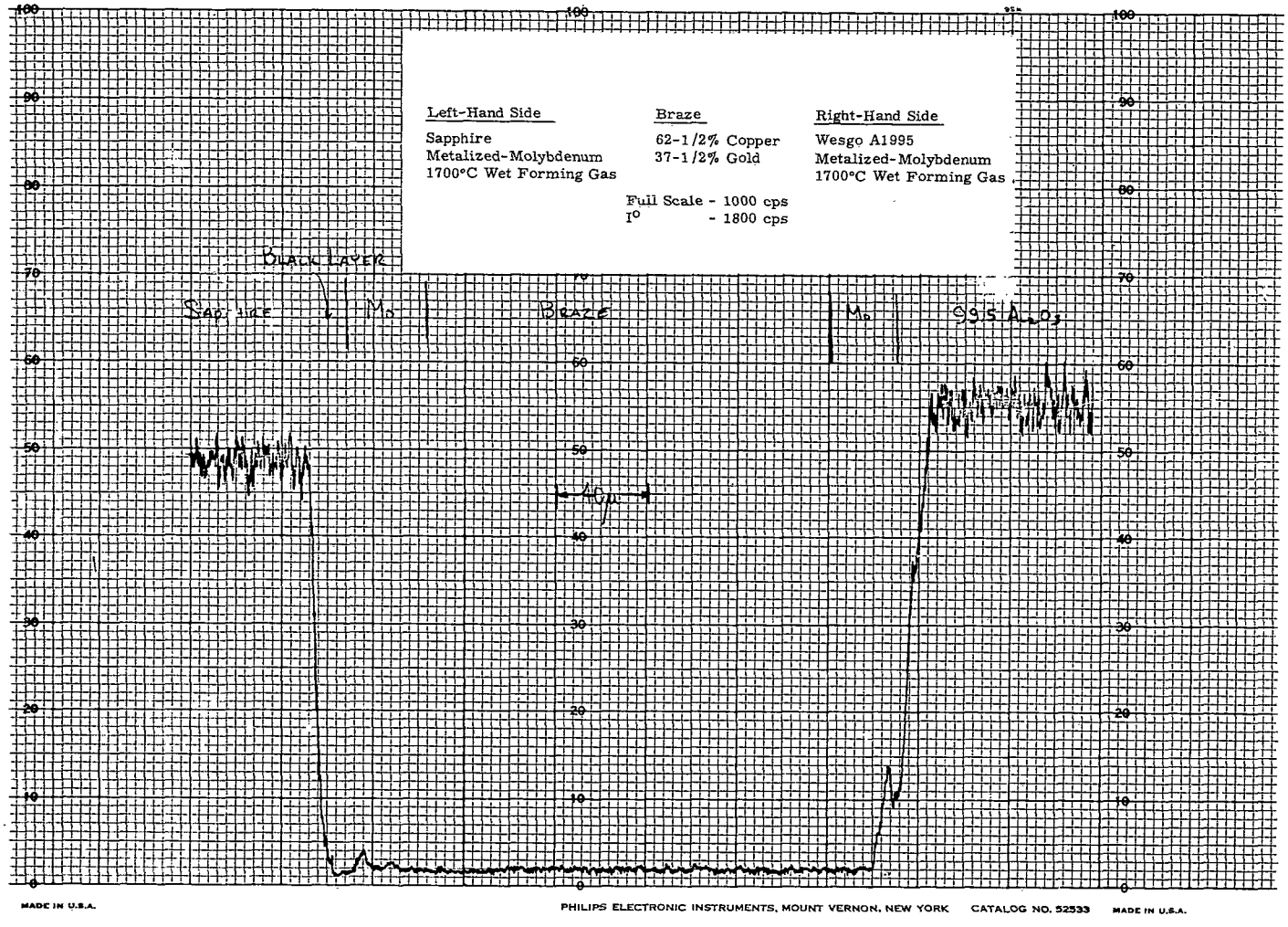


FIGURE 9 - ALUMINUM DISTRIBUTION IN
MOLYBDENUM METALIZED SAPPHIRE-WESGO A1995 SEAL

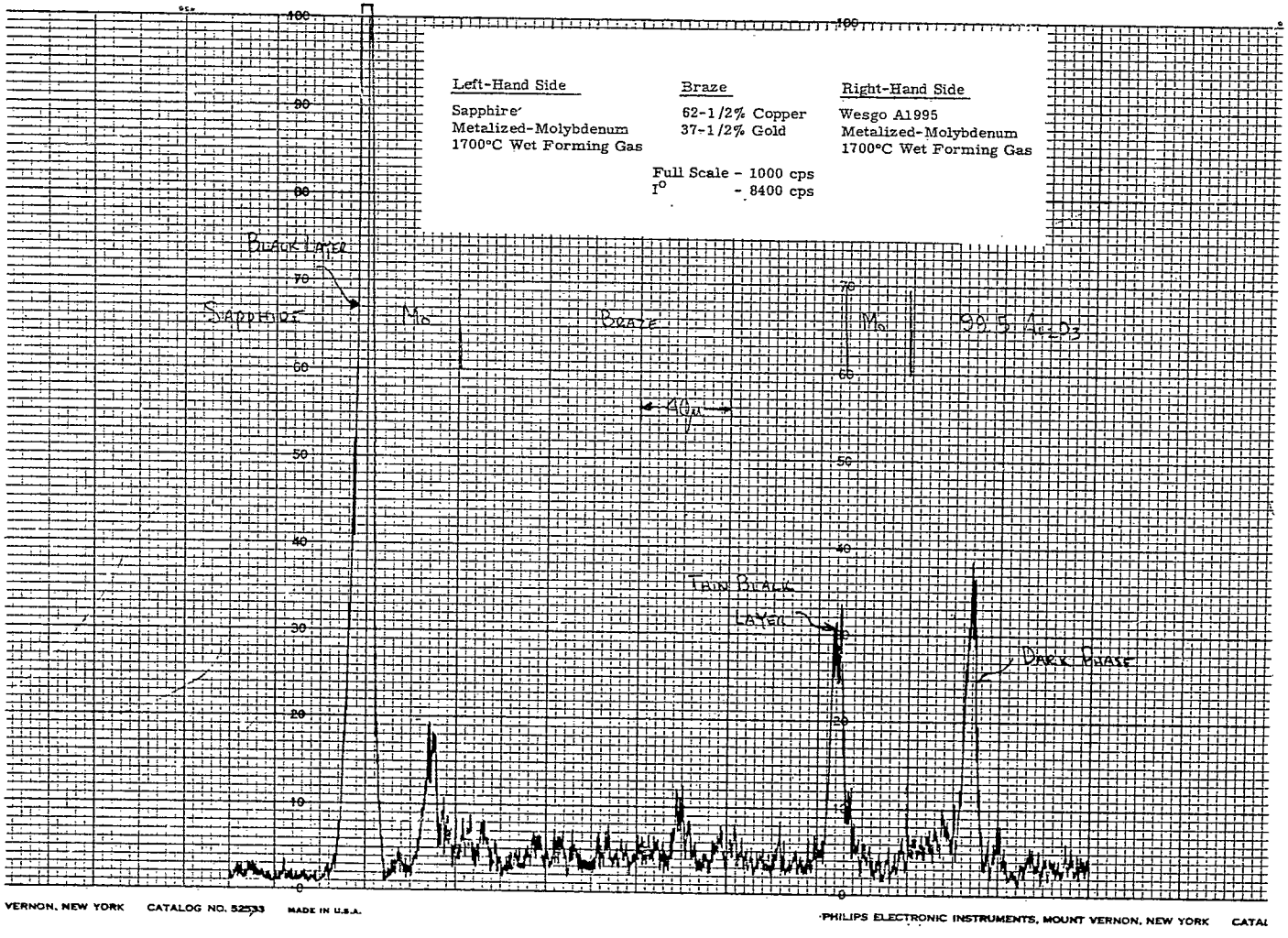


FIGURE 10 - SILICON DISTRIBUTION IN
MOLYBDENUM METALIZED SAPPHIRE-WESGO A1995 SEAL

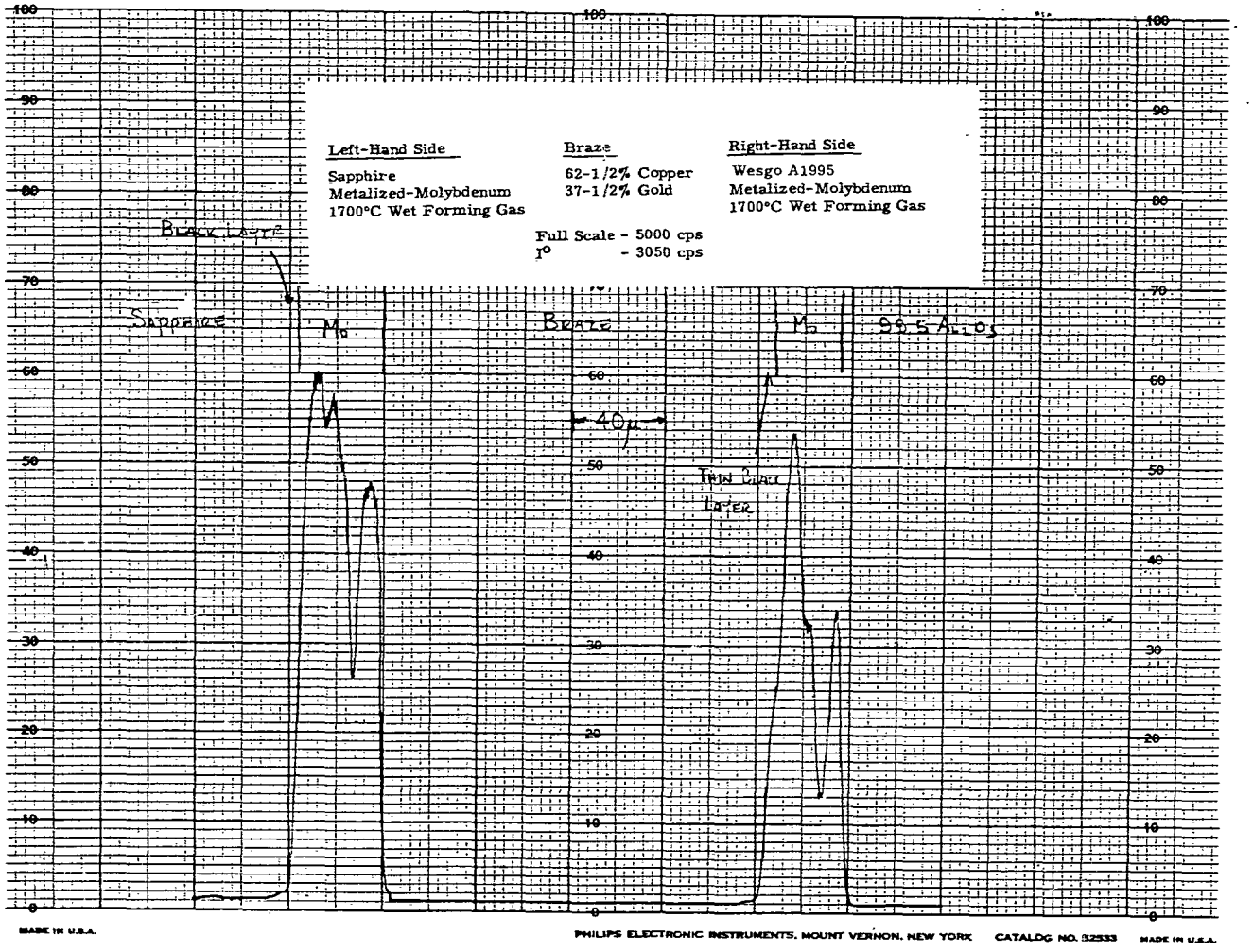


FIGURE 11 - MOLYBDENUM DISTRIBUTION IN
MOLYBDENUM METALIZED SAPPHIRE-WESGO A1995 SEAL

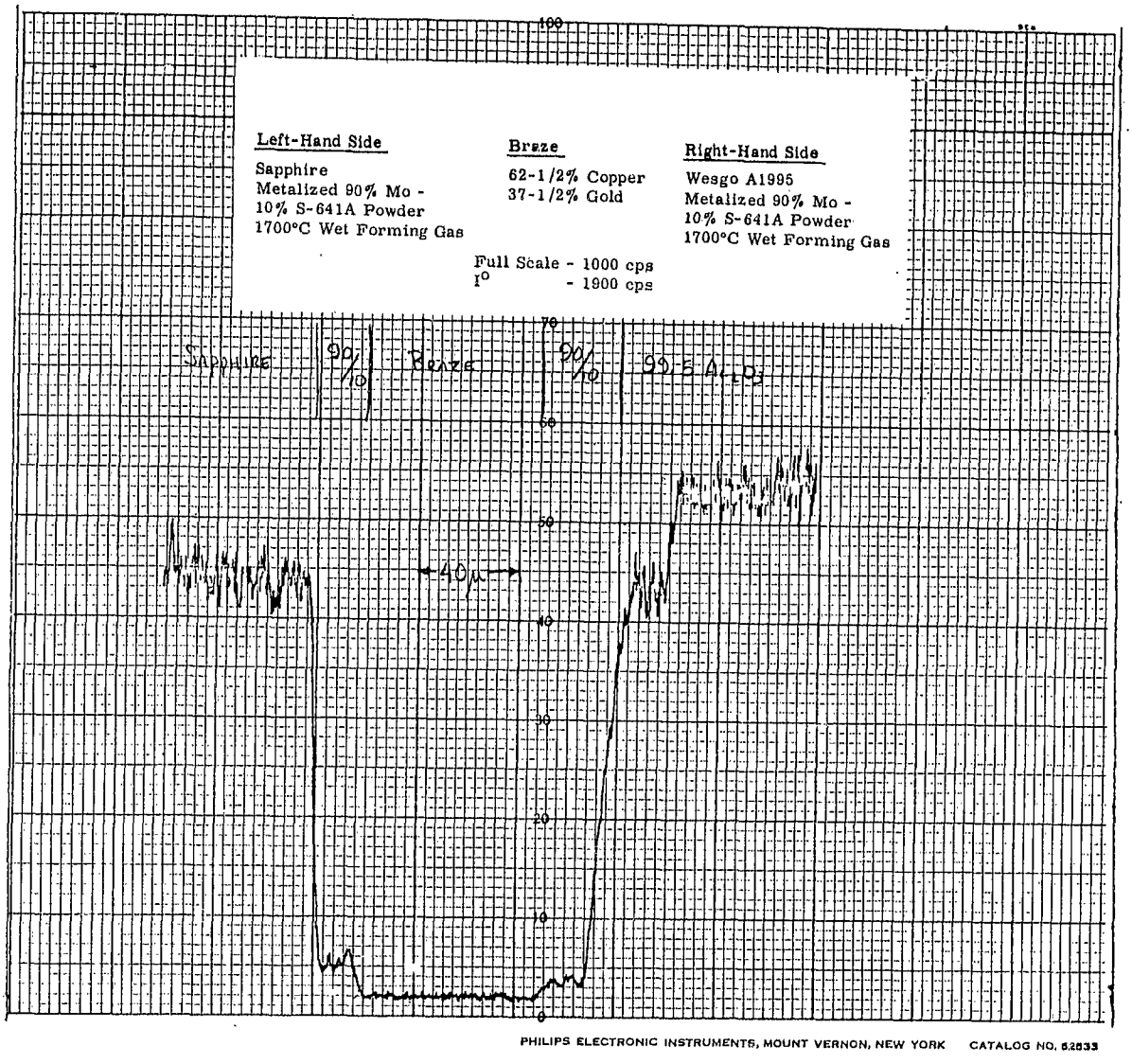


FIGURE 12 - ALUMINUM DISTRIBUTION IN
90% Mo-10% S-641A METALIZED SAPPHIRE-WESGO A1995 SEAL

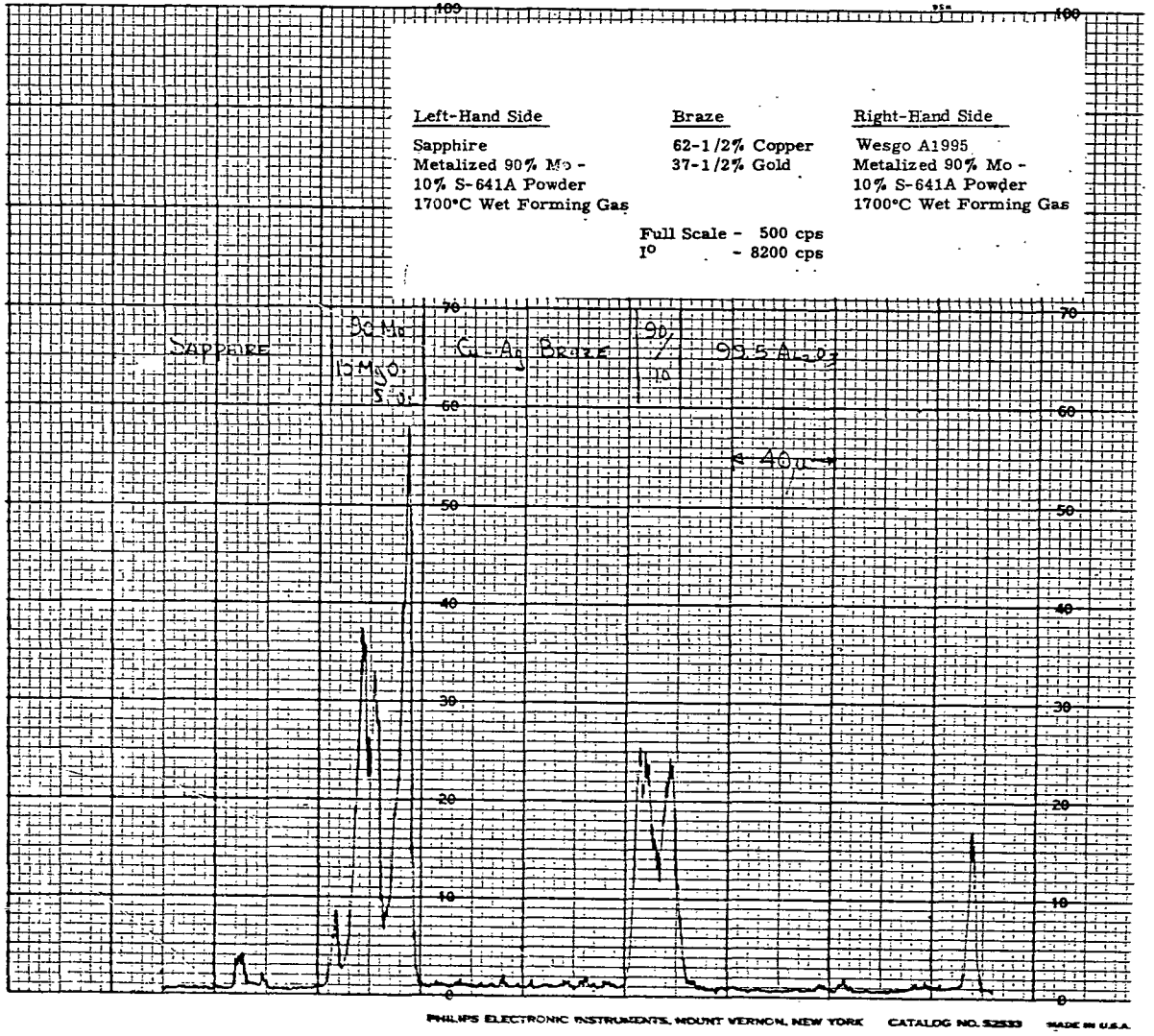


FIGURE 13 - SILICON DISTRIBUTION IN
90% Mo-10% S-641A METALIZED SAPPHIRE-WESGO A1995 SEAL

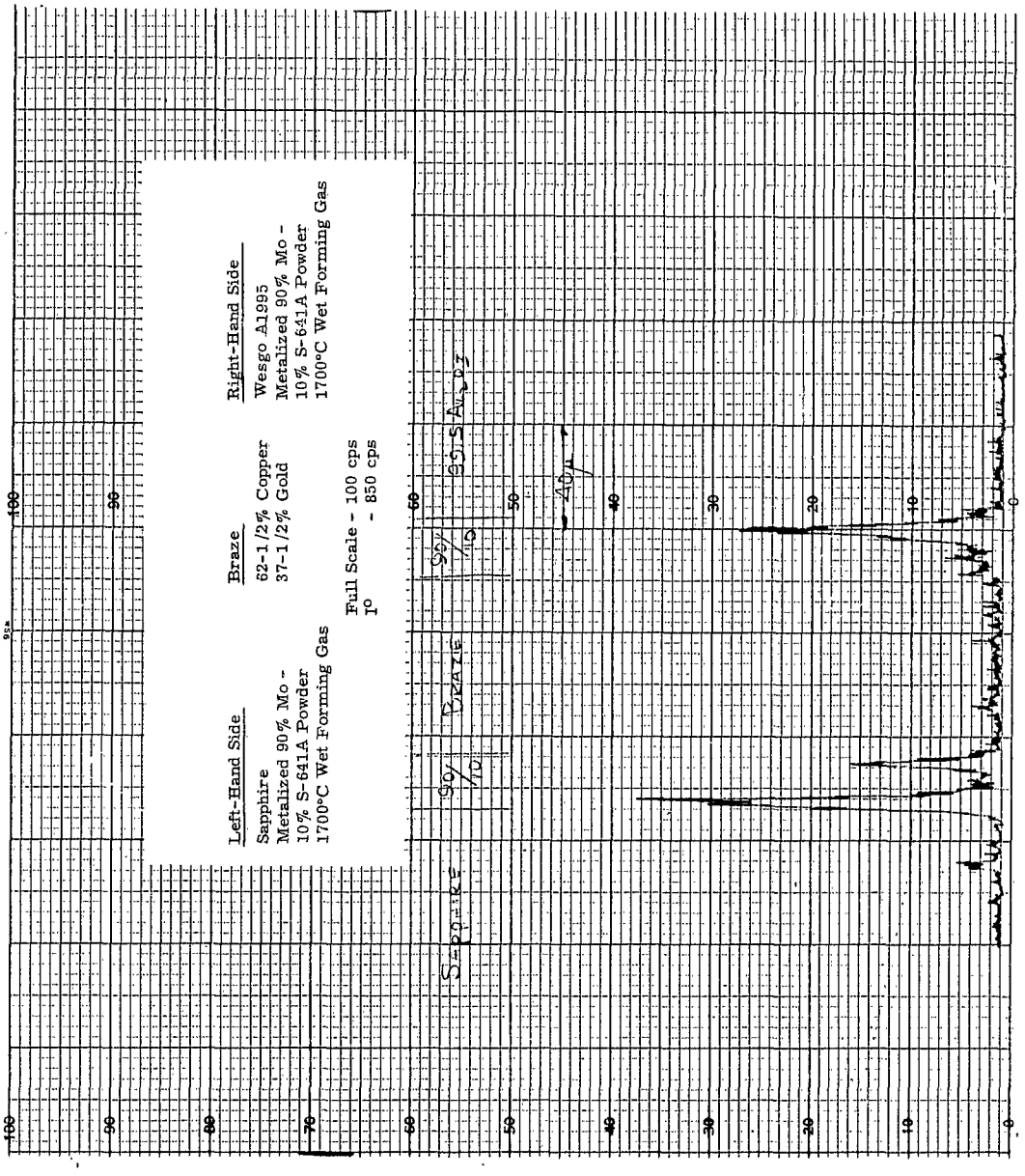


FIGURE 14 - MAGNESIUM DISTRIBUTION IN
90% Mo-10% S-641A METALIZED SAPPHIRE-WESGO A1995 SEAL

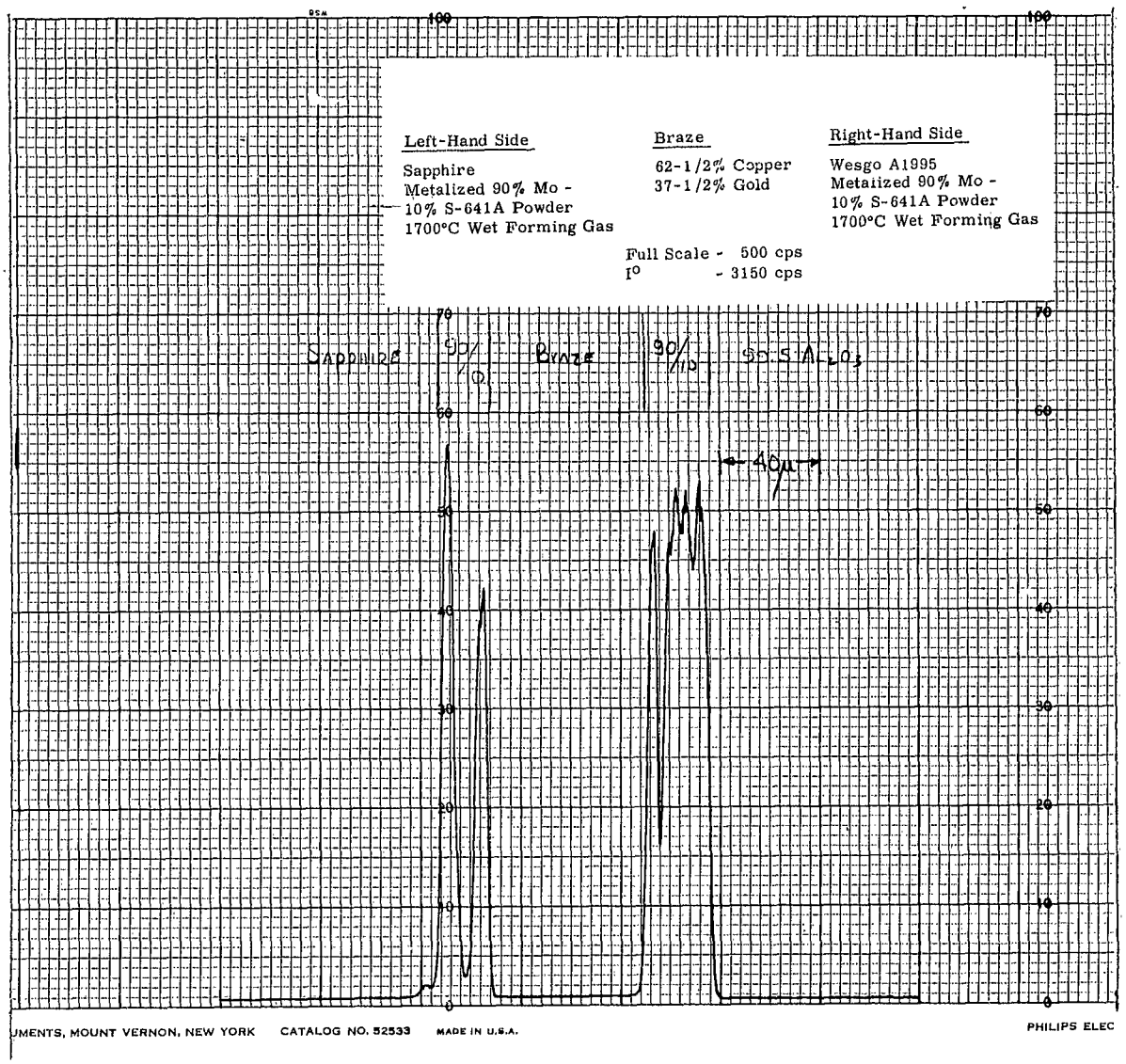


FIGURE 15 - MOLYBDENUM DISTRIBUTION IN
90% Mo-10% S-641A METALIZED SAPPHIRE-WESGO A1995 SEAL

IV. Metalized Window Assemblies

Jigs and fixtures have been made for the assembly of metalized sapphire windows. It is expected that several molybdenum metalized sapphire window assemblies will be ready for test in the near future.

V. Compression-Band Seals

The compression-band technique of making a vacuum-tight window assembly is similar to the technique of compressing a soft metal gasket between hard metal flanges which is used in making vacuum-tight seals. Metalizing and brazing are not required. In the compression-band window assemblies being developed under this contract, a soft copper sleeve is compressed against the edge of the dielectric disc by a high-strength compression band which exerts enough radial pressure to make a vacuum-tight seal.

A. Materials Investigation

During the report period, 41 compression-band seals were assembled to evaluate various dielectric and compression-band materials. Eighteen of the seals were vacuum tight. The work further defined the characteristics of the materials, provided valuable information on seal design parameters, and isolated the more common causes of seal failure.

1. Dielectric Materials for Windows

Developmental dielectric window assemblies were made using beryllium oxide, boron nitride, magnesium fluoride, Pyroceram XM-1, Pyroceram 9606, and sapphire having a 60° crystal orientation. For many of these assemblies, the Hot Form No. 2 compression band was used because of greater experience with these bands in previous tests. Their use also made it possible to accumulate comparative test data for the other compression-

band materials being evaluated. All seals were designed conservatively; generally the estimated seal pressure ranged from 50 percent to 100 percent below the published compressive strength of the dielectric material.

The beryllium oxide (Brush Beryllium Lot B-6-11) to be used in future tests will have a minimum modulus of rupture of 35,000 psi, which is approximately equivalent to a compressive strength of 280,000 psi. Also, for a slight additional cost the manufacturer has guaranteed that the discs will be selected for uniformity of compressive strength. It is expected that this beryllium will eliminate the difficulties experienced with Brush Beryllium Lot 1-B-7 which reportedly has a compressive strength of 240,000 psi but has fractured under much lower pressures.

The sapphire currently being used will be replaced by a sapphire with a 90° crystal orientation which places the weakest plane of the disc parallel to the direction of force and thus gives a greater working stress. This greater working stress should minimize the fracturing problem experienced with the 60° crystal orientation used in tests. The new discs will also be 0.012 inch thicker than those tested this period, providing a wider seal area.

2. Compression-Band Materials

Compression bands made from molybdenum, René 41, and Waspalloy metals have been fabricated and evaluated with Frenchtown No. 4462 ceramic discs. The strength and high-temperature characteristics of these band materials are adequate for the compression-band design objectives. The choice of a particular type of compression-band material will depend primarily on matching the expansion of the metal and the expansion of the dielectric

window material being used. The following are expected to be workable combinations of metals and dielectrics: (1) molybdenum-Pyroceram 9606, (2) René 41-sapphire 90° or beryllium oxide, and (3) Waspalloy-boron nitride, magnesium fluoride, or Pyroceram XM-1.

Compression bands made from molybdenum bar stock have been used successfully. The investigation of the molybdenum compression bands has shown that molybdenum bar stock is weaker than molybdenum flat stock having an equivalent cross-sectional area.

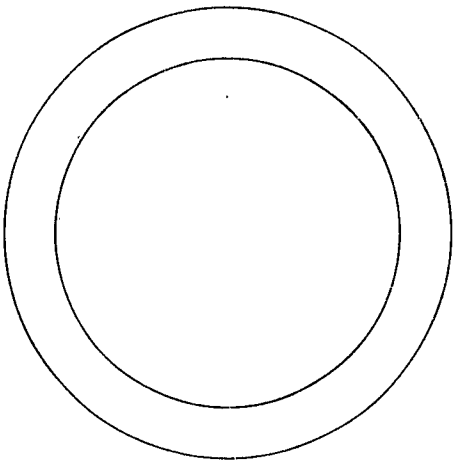
B. Seal Design Study

1. Pressure Calculations

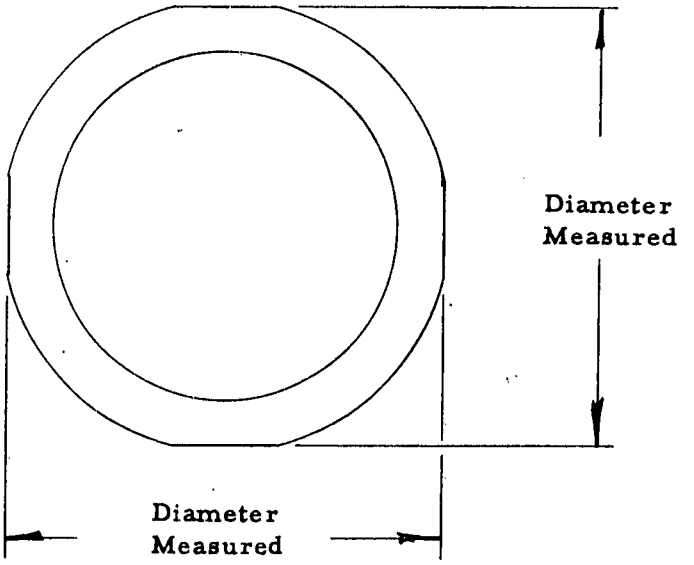
Initially, the only method for determining seal pressure, P_s, was to calculate the value by using an estimated value for the amount of copper deformation, d, in equation (1). This method is still used to estimate seal pressures.

$$P_s = E_b \frac{t_b}{t_c} \left(\frac{r_0^2 - r_1^2}{r_0^2 + r_1^2}\right) \left(\frac{r_c + t_s - d - r_1}{r_1}\right) \qquad (1)$$

By devising a method for determining the precise strain in assembled windows, it is now possible to calculate the actual seal pressure. Four small flat surfaces 90° apart (see Figure 16) were machined on the outside diameter of the compression band to provide accurate points for measuring the compression-band diameter before and after assembly. The difference in these diameters is the total strain ($r_c + t_s - d - r_1$) which can be substituted in equation (1) to give the actual seal pressure. Experience has shown that the actual seal pressure immediately after



(a) Former Configuration



(b) Modified Configuration

FIGURE 16 - COMPRESSION BAND MODIFIED TO MEASURE STRAIN ACCURATELY

sealing is as much as 50 percent below the estimated seal pressure. Experience has also shown that the difference between the actual and estimated pressures is larger at the higher seal pressures, reflecting the tendency of the copper sleeve to yield more at pressures above 30,000 psi.

C. Dielectric Window Test Summary

1. Beryllium Oxide Assemblies

Five beryllium oxide window assemblies were sealed using Hot Form No. 2 compression bands (see Table V). Three of the windows were vacuum tight. Test window 16 had a very small leak after the standard vacuum baking at 400° Centigrade and 450° Centigrade but became vacuum tight after 60 days. Test window 9 had a small leak initially but was vacuum tight when tested 30 days later. A number of surface cracks appeared in test window 11 when it was sealed, due partially to the high sealing pressure considered necessary during the earlier phases of this study and partially to the fact that the disc was not aligned properly in the compression band (refer to paragraph D for a discussion of window centering). Test window 11 became vacuum tight after vacuum baking.

2. Boron Nitride Window Assemblies

Three window assemblies (see Table VI) were made using boron nitride. One was vacuum tight. Difficulties in making compression-band seals with boron nitride were anticipated because of the material's low compressive strength (45,000 psi). Test window 48, with an actual seal pressure of 19,200 psi, was vacuum tight after vacuum baking at 400° Centigrade. Test window 47, which had an estimated and actual seal pressure of 9,600 psi, was made to determine if a window could be sealed vacuum

TABLE V. TEST RESULTS, BERYLLIUM OXIDE COMPRESSION-BAND WINDOW ASSEMBLIES

Window Test Number	Compression-Band Material	$P_{s(psi)}$ Estimated	$P_{s(psi)}$ Actual	Vacuum Tight	Comments
9	HF No. 2	85,000	-	Yes	-
10	HF No. 2	106,000	-	No	Initially good; fractured overnight
11	HF No. 2	122,000	-	Yes	Window not on center; surface cracks after sealing
16	HF No. 2	59,000	43,500	Yes	Window sealed after 2-month shelf life test
27	HF No. 2	61,800	-	-	Window fractured during assembly (off center)

TABLE VI. TEST RESULTS, BORON NITRIDE COMPRESSION-BAND WINDOW ASSEMBLIES

Window Test Number	Compression-Band Material	$P_{s(psi)}$ Estimated	$P_{s(psi)}$ Actual	Vacuum Tight	Comments
47	HF No. 2	9,600	9,600	No	-
48	HF No. 2	29,000	19,200	Yes	-
49	HF No. 2	31,800	18,700	No	Very small leak after baking

tight at this low pressure. The window was not vacuum tight. Test window 49 had a very small leak after vacuum baking, probably the result of slight imperfections in the seal surface of the dielectric material. The leak may also have been caused by out-of-round compression bands or variations in thickness of the copper sleeve wall. Generally, these three defects can be suspected as the causes of leaks in window assemblies fabricated from other dielectric materials and compression-band materials.

3. Frenchtown No. 4462 Ceramic Window Assemblies
To date, Frenchtown No. 4462 ceramic discs have been used for: (1) correlation of seal pressure design calculations, (2) evaluation of high-temperature bake test equipment, and (3) as a standard window material for evaluation of various compression-band materials. During this quarter, 14 ceramic discs were assembled using three compression-band materials: molybdenum, René 41, and Waspalloy. The test results are given in Table VII. No additional tests of these compression-band materials are planned because of the satisfactory results obtained. All three band materials have been found to be satisfactory.

4. Magnesium Fluoride Window Assemblies
Six magnesium fluoride window assemblies were sealed; one was vacuum tight, four fractured completely, and one partially fractured (see Figure 17). The estimated seal pressures for all of these assemblies were within the published compressive strength of 156,000 psi for the magnesium fluoride discs used in window assemblies 12 through 15 (see Table VIII) and well within the value for window assemblies 21 and 35. The exact

TABLE VII. TEST RESULTS, FRENCHTOWN NO. 4462
COMPRESSION-BAND WINDOW ASSEMBLIES

Window Test Number	Compression-Band Material	P_s(psi) Estimated	P_s(psi) Actual	Vacuum Tight	Comments
22	Molybdenum	52,000	-	-	Compression band cracked during sealing
23	Molybdenum	57,000	-	-	Compression band cracked during sealing
24	Molybdenum	48,000	48,000	Yes*	
25	Molybdenum	52,000	46,000	Yes*	
26	Molybdenum	-	-	Yes*	
28	Molybdenum	69,000	43,500	Yes	Leaked when silver solder brazed at 800° C
29	Molybdenum	78,000	59,400	Yes*	
38	Waspalloy	149,000	81,000	Yes**	
39	Waspalloy	146,500	76,000	Yes**	
40	Waspalloy	131,300	91,000	Yes**	
41	René 41	145,000	83,000	Yes**	
42	René 41	86,000	58,400	No	Small Leak
43	René 41	114,000	64,600	Yes**	
44	René 41	100,000	66,800	Yes*	

*Leaked initially but became vacuum tight without baking.
**Vacuum tight after sealing.

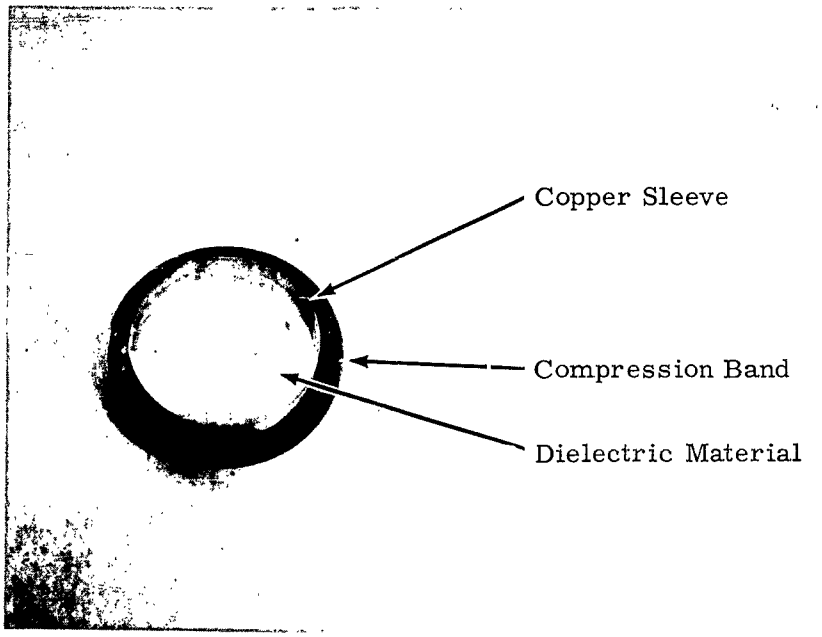


FIGURE 17 - MAGNESIUM FLUORIDE WINDOW ASSEMBLY

TABLE VIII. TEST RESULTS, MAGNESIUM FLUORIDE COMPRESSION-BAND WINDOW ASSEMBLIES

Window Test Number	Compression-Band Material	$P_{s(psi)}$ Estimated	$P_{s(psi)}$ Actual	Vacuum Tight	Comments
12	HF No. 2	63,000	-	-	Fractured completely
13	HF No. 2	63,000	-	-	Off center; fractured completely
14	HF No. 2	53,000	-	-	Fractured completely
15	HF No. 2	58,000	-	-	Fractured completely
21	HF No. 2	30,000	-	No	Surface cracks after baking
35	HF No. 2	21,200	15,700	Yes*	

*Leaked initially but became vacuum tight without baking.

cause of the failure of windows 12, 14, and 15 is yet to be determined. It is possible that the discs in these windows were not aligned and consequently fractured under the non-symmetrical compressive stress.

Additional magnesium fluoride windows have been ordered and tests will be conducted using Waspalloy compression bands and lower sealing pressures.

5. Pyroceram 9606 Window Assemblies

Seven Pyroceram 9606 window assemblies were sealed using both Hot Form No. 2 and molybdenum compression bands (see Table IX). Pyroceram 9606 is the first material to be evaluated with a matched expansion compression-band material

TABLE IX. TEST RESULTS, PYROCERAM 9606
COMPRESSION-BAND WINDOW ASSEMBLIES

Window Test Number	Compression-Band Material	$P_{s(psi)}$ Estimated	$P_{s(psi)}$ Actual	Vacuum Tight	Comments
30	HF No. 2	38,000	33,800	No	Parts reused
31	HF No. 2	40,000	31,400	No	Parts reused
36	Molybdenum	50,000	37,600	No	Off center
37	Molybdenum	48,000	20,000	No	
45	Molybdenum	70,500	-	No	Parts reused
46	Molybdenum	70,500	23,000	Yes	
50	HF No. 2	66,000	33,000	Yes	

(molybdenum). (Window discs and compression bands are considered to have matched expansions when the rate of expansion of the dielectric material is equal to or less than that of the compression-band material.) It is probable that test windows 30, 31, 36, and 45 leaked because the surface finish on the dielectric discs was only marginal. The Pyroceram 9606 discs used on test windows 46 and 50 were previously used discs repolished to a 16 micro-inch finish. Both of these test windows sealed at relatively low actual seal pressures.

6. Pyroceram XM-1 Window Assemblies

Two attempts were made to seal Pyroceram XM-1 window assemblies (see Table X). In both cases the dielectric material fractured. It is believed that test window 19 failed because the seal pressure at bake temperature (400° Centigrade) exceeded the compressive strength of the Pyroceram. The expansion of

TABLE X. TEST RESULTS, PYROCERAM XM-1
COMPRESSION-BAND WINDOW ASSEMBLIES

Window Test Number	Compression-Band Material	$P_{s(psi)}$ Estimated	$P_{s(psi)}$ Actual	Vacuum Tight	Comments
19	HF No. 2	30,000	-	No	Partial fracture at baking
20	HF No. 2	45,000	-	-	Complete fracture during assembly

Pyroceram XM-1 at 400° Centigrade is slightly greater than Hot Form No. 2, thus causing an increase in seal pressure with increased temperature. Further evaluation tests are planned using Waspalloy compression bands, which have a higher coefficient of expansion than Hot Form No. 2 and match more closely the expansion of Pyroceram XM-1. This will prevent an excessive pressure buildup as the bake temperature is increased to 400° Centigrade.

7. Sapphire Window Assemblies

Four sapphire (60°) window assemblies were sealed using Hot Form No. 2 compression bands (see Table XI). Window assembly 17 buckled and fractured completely. Window assemblies 18, 33, and 34 had only partial fractures as shown in Figures 18, 19, and 20. Window assemblies 33 and 34 cracked along the 60° crystal plane. The cause of these failures, i. e., the relatively low compressive limit of the 60° oriented windows, is expected to be eliminated by the use of sapphire windows with a 90° orientation. These have been ordered. Two other causes of failure, the lack of fine polishing of the seal edge of the sapphire

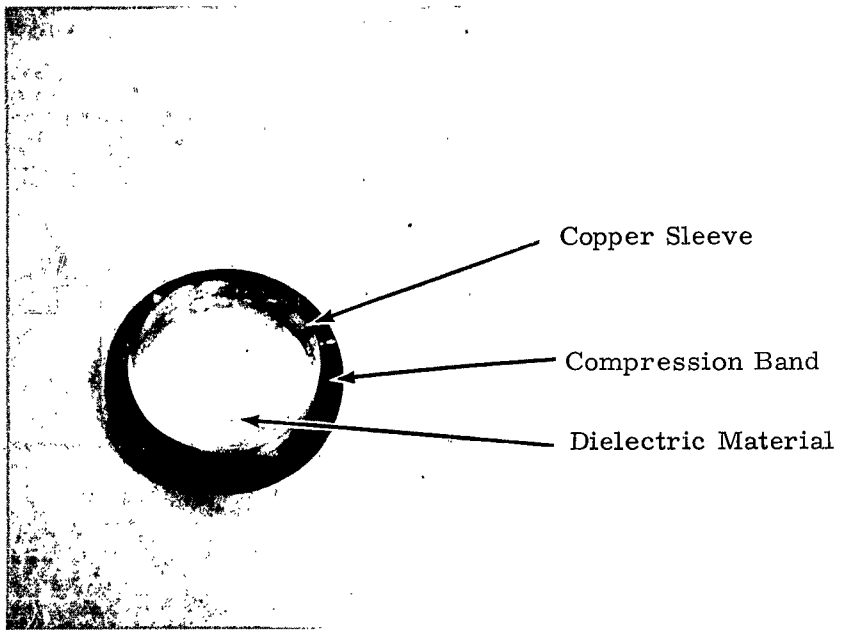


FIGURE 18 - SAPPHIRE WINDOW ASSEMBLY 18

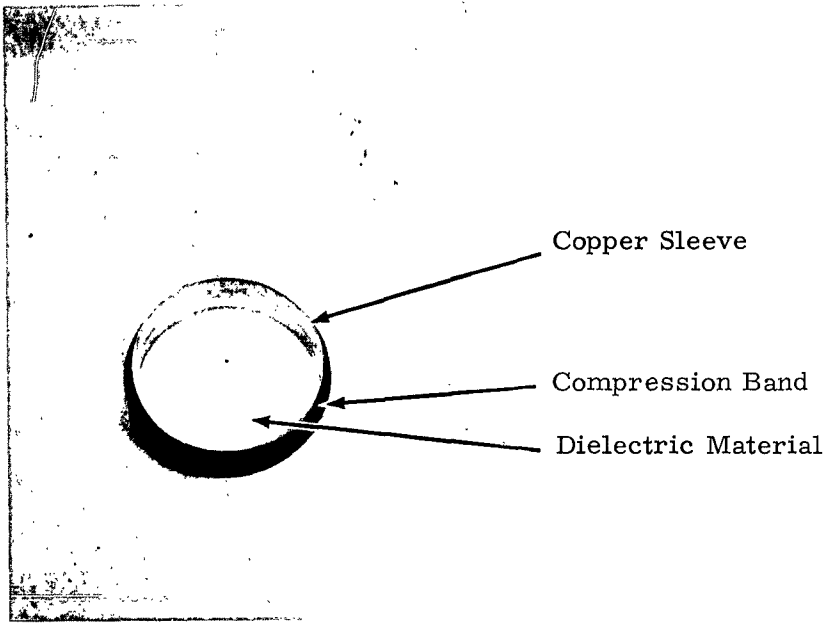


FIGURE 19 - SAPPHIRE WINDOW ASSEMBLY 33

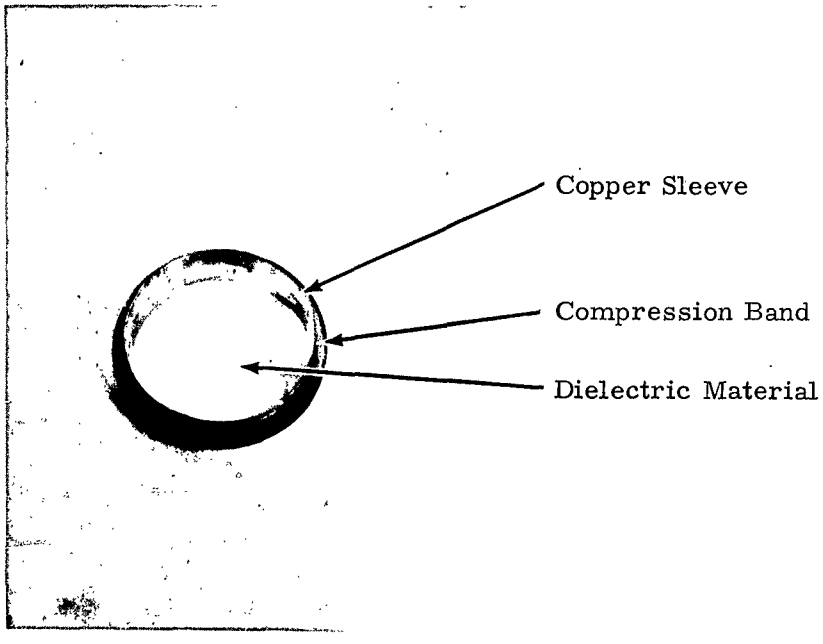


FIGURE 20 - SAPPHIRE WINDOW ASSEMBLY 34

TABLE XI. TEST RESULTS, SAPPHIRE COMPRESSION-BAND WINDOW ASSEMBLIES

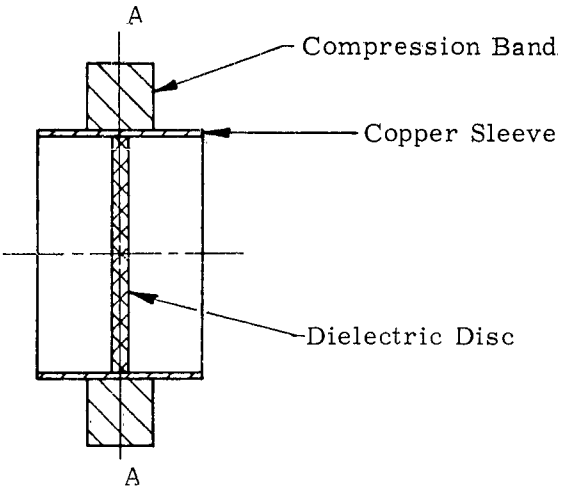
Window Test Number	Compression-Band Material	$P_{s(psi)}$ Estimated	$P_{s(psi)}$ Actual	Vacuum Tight	Comments
17	HF No. 2	76,000	-	-	Complete fracture
18	HF No. 2	62,000	-	No	Shear crack during sealing
33	HF No. 2	97,000	-	No	Random crack; window not centered
34	HF No. 2	81,500	-	No	Random crack; window not centered

disc, and off-center windows can be avoided by extreme care in fabrication and assembly.

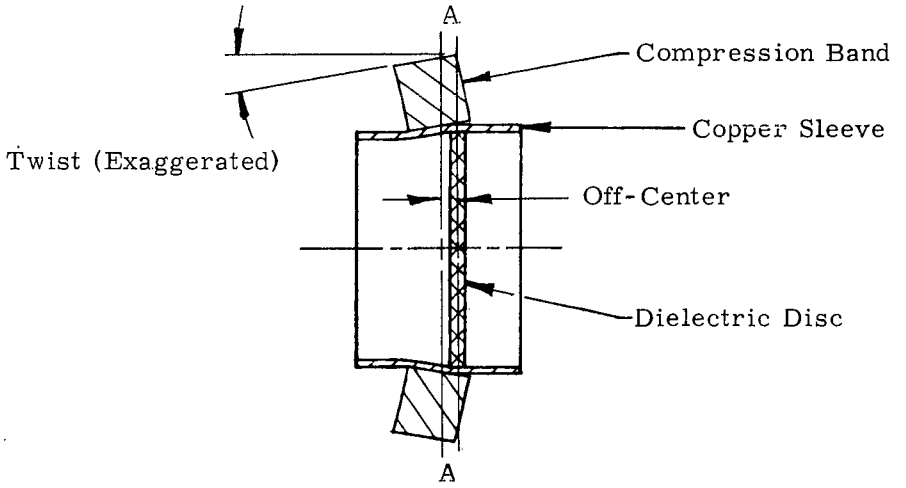
D. Window Fabrication

1. Assembly

A major problem encountered in the assembly operation has been the centering of the dielectric discs on the axial plane A'A of the compression bands (see Figure 21). Unless the compression band is expanded sufficiently when heated to provide the clearance necessary for assembly, the band seizes the disc before it is axially centered and the sealing pressure of the band is not uniform and buckles the window excessively and in the extreme case fractures the window. In properly assembled windows the maximum dielectric disc deviation from the center plane of the compression band is approximately 0.004 inch.



On-Center Window Assembly



Off-Center Window Assembly

FIGURE 21 - COMPRESSION-BAND WINDOW ASSEMBLY

When assembling windows using Hot Form No. 2 compression bands, it was necessary to limit the maximum assembly temperature to prevent rapid annealing and reduced tensile strength. The increased clearance necessary to prevent seizing during the assembly operation can readily be obtained by using a compression-band material with a higher maximum operating temperature or a higher rate of expansion than Hot Form No. 2. Compression-band materials such as Rene' 41 and Waspalloy have higher operating temperatures (the maximum temperature at which a metal retains for extended periods much of the strength it has at room temperature) and higher rates of expansion than Hot Form No. 2. Molybdenum has a comparatively low rate of expansion but can be expanded sufficiently because of its high (1100° Centigrade) operating temperature.

Off-centering of windows also introduces a second problem. Frequently the compression band twists, making it difficult to arrive at accurate strain measurements which are based on changes in the outside diameter of the band.

2. Brazing

Test window 8 made from Frenchtown No. 4462 ceramic and Hot Form No. 2 was successfully brazed with Incusil 15 (indium, copper, and silver composition) at 700° Centigrade.

Test window 28 made from Frenchtown No. 4462 ceramic and molybdenum developed a large leak when an attempt was made to BT braze (silver-copper eutectic, melting point 779° Centigrade) it to a vacuum bake test fixture. Because the expansion of ceramic is slightly higher than molybdenum, the increased seal pressure at a braze temperature of 800° Centigrade deformed the copper sleeve and caused the leak.

E. Window Evaluation Tests

1. High-Temperature Vacuum Bake Tests

During this report period, a compact, double vacuum bake testing system (see Figure 22) was constructed with RCA funds to provide readily available facilities for testing windows. It is assigned full time to this project. This double vacuum bake facility, which is similar to the systems used to bake electron tube devices at elevated temperatures, prevents oxidation by developing a vacuum inside and outside the device being bake tested. The system develops a maximum temperature of 780° Centigrade. A mass spectrometer can be attached to the window under test to detect leaks at test temperatures.

Test window 8 (ceramic and Hot Form No. 2) was subjected to three short high-temperature bake tests to determine if the window would remain vacuum tight after repeated bake testing. This window assembly was also leak checked at bake-out temperatures in the double vacuum system at a helium pressure of approximately 250 microns. Leaks were noted as the bake temperature was cycled to 600°, 550°, and 590° Centigrade from 300° Centigrade. This window was then life tested for 50 hours at a minimum temperature of 500° Centigrade. To check the results obtained in the short high-temperature bake tests, the temperature was raised repeatedly during the life test to approximately 600° Centigrade. Leaks were noted at an average temperature of 570° Centigrade. The window resealed at 500° Centigrade. At the conclusion of these vacuum bake tests, test window 8 was still vacuum tight at room temperature.

Test window 36 (Pyroceram 9606 and molybdenum) had a minute leak after assembly. When the window was subjected to a

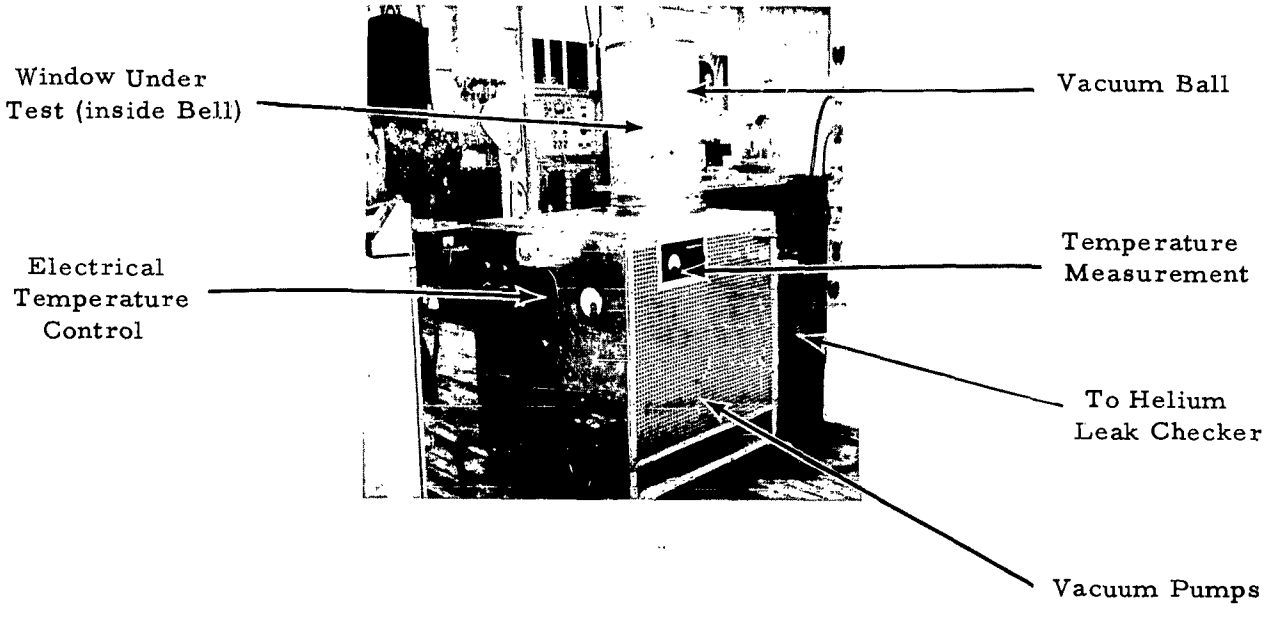


FIGURE 22 - DOUBLE VACUUM BAKE TEST EQUIPMENT

temperature of 700° Centigrade for two minutes, the leak diminished considerably. This would indicate that the Pyroceram 9606 and molybdenum combination has a good high-temperature vacuum bake-out potential.

2. Shelf Life Tests

All windows have remained vacuum tight in shelf life tests at room temperature (see Table XII). Two of the windows have also been tested for extended periods at elevated temperatures. Window 6 was life tested for 100 hours at 450° Centigrade; window 8 was life tested for 50 hours at 500° Centigrade. Both of these windows remained vacuum tight.

3. Cycle Life Test

The controls for the cycle life test equipment have been ordered and are expected during the fourth quarter. Present plans are to cycle life test several window assemblies simultaneously, using a common vacuum system with individual heating chambers from room temperature to 125° Centigrade.

VI. Fused Silica-to-Metal Seals

A. General

The objective during this quarter was to continue evaluating seal parameters and to determine the processing requirements of the copper and fused silica experimental parts.

Twenty-five experimental "butt-type" fused silica-to-metal copper seals (described in Second Quarterly Report) were fabricated. Two of these seals were vacuum tight. Experimental test seal 17, (see Table XIII) which is 1-inch in diameter, remained vacuum tight after being cycled four times to 200° Centigrade and 12 times

TABLE XII. RESULTS OF COMPRESSION-BAND WINDOW SHELF LIFE TESTS

Window Test Number	Window Type	Shelf Life Days at Room Temperature	Vacuum Tight
1	HF No. 2 & Ceramic	168	Yes
4	HF No. 2 & Ceramic	167	Yes
5	HF No. 2 & Ceramic	157	Yes
6	HF No. 2 & Ceramic	122	Yes
8	HF No. 2 & Ceramic	92	Yes
9	Beryllium Oxide and HF No. 2	90	Yes
35	Magnesium Fluoride and HF No. 2	42	Yes

TABLE XIII. SUMMARY OF FUSED SILICA-TO-METAL EXPERIMENTAL SEALS

Number	Fused Silica Dia. (in.)	Copper Thick. (in.)	Oxidation Time (sec)	Seal Pressure (psi)	Furnace Time (min)	Furnace Temp. (°C)	Comments
1	0.350	0.002	20	470	20	910	Partial reaction
2	Assembly not properly lined up						
3	0.350	0.002	20	470	20	900	Good adherence in some areas.
4	0.350	0.002	15	575	20	920	Good adherence in some areas.
5	0.350	0.002	15	470	15	910	One side cracked, good adherence.
6	0.350	0.002	15	575	15	940	Seal edge on fused silica polished, very good adherence, small leak.
7	0.350	0.002	10	715	15	970	Seal pressure too high, assembly collapsed.
8	0.350	0.002	15	620	15	950	Seal edge fine polished, vacuum tight.
9	-	-	-	-	-	-	Collapsed during sealing.
10	0.350	0.002	10	620	15	995	Copper deformed during sealing.
11	0.350	0.002	60	620	15	955	Spotty reaction, poor adherence.
12	0.350	0.010	20	620	15	955	Very good adherence, small leak.
13	0.350	0.010	20	620	30	955	Very good adherence, small leak.
14	0.350	0.002	15	620	30	955	Very good adherence, small leak.
15	0.350	0.010	Not Oxidized	620	15	955	0.001 inch gold plated, good adherence but not uniform.
16	0.350	0.007	Dark Oxide	620	15	970	Copper deformed during sealing.
17	1.0	0.006	3	155	15	970	Vacuum tight, cycle tested.
18	1.0	0.006	Light Oxide	155	15	960	Very good adherence, small leak.
19	1.0	0.007	10	155	15	980	Very good adherence, small leak.

TABLE XIII. SUMMARY OF FUSED SILICA-TO-METAL EXPERIMENTAL SEALS
(Continued)

Number	Fused Silica Dia. (in.)	Copper Thick. (in.)	Oxidation Time (sec)	Seal Pressure (psi)	Furnace Time (min)	Furnace Temp. (°C)	Comments
20	0.350	0.006	Light Oxide	17	15	1000	No adherence
21	0.350	0.006	Light Oxide	230	15	1000	Poor adherence
22	0.350	0.006	Light Oxide	230	15	1050	Small leak, copper disc melted.
23	0.350	0.007	3	230	15	1000	Very good reaction
24	0.350	0.007	3	230	15	1000	Poor adherence
25	0.350	0.007	3	230	15	1000	400-grit polish, poor adherence.

to 150° Centigrade. Experimental test seal 8, which has a diameter of 0.350 inch, was vacuum tight after being cycled eight times to 150° Centigrade.

To obtain preliminary structural data on the effect of temperature cycling on the tapered, balanced-type seal, a previously sealed window assembly (see First Quarterly Report), which had a small leak, was raised to above 800° Centigrade three times in a dry nitrogen atmosphere. There was no noticeable increase in the leak rate. A microscopic examination of this window assembly showed that some chipping of the fused silica disc had occurred in the vicinity of the seal edge. Tests with the butt-type experimental seals have proved that fire polishing of the sharp corners of the sealing edge of the fused silica will minimize chipping and thereby aid in eliminating seal leaks.

B. Summary of Seal Tests

Experimental seals 1 through 5 (see Table XIII) were made using the optimum seal parameters established in studies of seals made during the second quarter. The seal areas had a good uniform appearance but some fracturing of the fused silica on the edge of the seal area was noted. To correct this, the fused silica tubing was fire polished and then hand ground to produce a flat, uniform surface at the seal area.

Experimental test seals 6, 7, and 8 were fire polished before sealing, thereby eliminating the fracturing and chipping problem. Experimental seal 6 had a very small leak; seal 8 was vacuum tight.

Methods of cleaning the copper before the seals are oxidized were also investigated. In a few of the experimental seals, small particles were noted in the seal area. To keep the seal area particle

free, the final surface finish on the copper was obtained by using 400-grit fused silica powder. Although this procedure has eliminated the foreign particles, it has not materially aided in eliminating leaks.

Experimental seals 11 through 14 were sealed at a temperature of 955° Centigrade. The copper thickness, oxidation time, and sealing pressure on these windows were varied, as shown in Table XIII. Copper oxidation of experimental seal 11 for 60 seconds did not produce good seal adherence.

In fabricating the initial experimental butt-type seals, the thinnest practical copper discs were used so that the effects of variations in seal parameters (seal pressure, firing temperature, and firing time) could be observed more readily. It was recognized, of course, that a heavier disc would have to be used in any window designed for a vacuum envelope. After determining the optimum seal parameters with the thin discs, seals were fabricated with 0.010-inch copper discs (seals 12 and 13). These seals were judged equal to those obtained with the thinner discs. Seals are evaluated by the following routine procedure. After the seals are checked for leaks, the physical structure of the seal is microscopically studied for the presence of chips or cracks. This is followed by a microscopic examination of the seal area. This area is evaluated on the basis of color, uniformity of adherence, and the absence of foreign material. The color analysis is basically a comparison of the seal color with color common on the "Housekeeper"[1] (copper-to-glass seal). The seal evaluation is concluded by a break test to determine adherence of the fused silica to the copper oxide and adherence of the copper oxide to the copper.

[1] W. H. Kohl, Materials and Techniques for Electron Tubes, Pages 412-413, January 1960.

Experimental seals 13 and 14 were held at seal temperature for 30 minutes instead of 15 minutes; the seals were not vacuum tight and showed no improvement in the appearance of the seal area.

The 0.010-inch thick copper disc for experimental seal 15 was plated with 0.001 inch of gold. The gold diffused evenly into the quartz and the resulting seal was very strong. Because it eliminates oxidation of the copper and developes a high seal strength, this technique is very promising and more seals using gold-plated copper will be fabricated and evaluated.

The remaining experimental seals, 17 through 25, contained copper discs 0.006 and 0.007-inch thick and were sealed at temperatures between 960° and 1050° Centigrade. These seals were made with very lightly oxidized copper, sealing pressures of 17 psi to 230 psi, and a sealing time of 15 minutes.

Experimental seal 20 was made at a temperature of 1000° Centigrade and at a sealing pressure of 17 psi. There was no reaction. This duplicated results of a similar test conducted previously at a sealing temperature of approximately 910° Centigrade and sealing pressure of 17 psi. These test results indicate that below a certain seal pressure a seal reaction will not take place even though the sealing temperature approaches the melting point of the copper.

The amount of oxidation on the copper used in the tests was arbitrarily judged by visual comparison with the oxidation samples used for "Housekeeper" type seals. The two vacuum-tight seals that were made (8 and 17) had what would be considered lightly oxidized copper. Oxidized copper with this same appearance will be used for future experimental seals. One piece of copper oxidized for 30 seconds was cross-sectioned in order to determine the

thickness of the copper oxide. The copper oxide was approximately four microns thick (0.00016 inch). Additional samples will be made to check this initial measurement.

A few experimental seals were mechanically fractured in order to determine the strength and general appearance (see Figure 23). The seals fractured in a manner characteristic of standard glass-to-metal seal fractures. (See Figure 24 for copper-to-soft glass butt-type seal fracture.) A layer of the fused silica adhered to the copper disc, indicating that the fused silica-to-metal bond is the strongest part of the experimental seal.

Cross sections were made of the experimental seals, butt-type seals, and the "Housekeeper" seals. An examination of these under polarized light in high power and metallographic microscopes revealed that the reaction areas of these seals were similar in appearance.

The proposed tapered, balanced-type seal will be attempted during the next quarter when a larger induction heating unit and the necessary inconel enclosures will be available.

VII. Program for Next Quarter

1. Improve the reliability of molybdenum metalized-sapphire seal strength.
2. Increase the seal strength of tungsten metalized-sapphire systems.
3. Continue efforts on refractory metalizing of beryllium oxide including efforts to obtain more experimental latitude.
4. Produce metalized sapphire window assemblies for test.
5. Develop reactive metal seal systems.

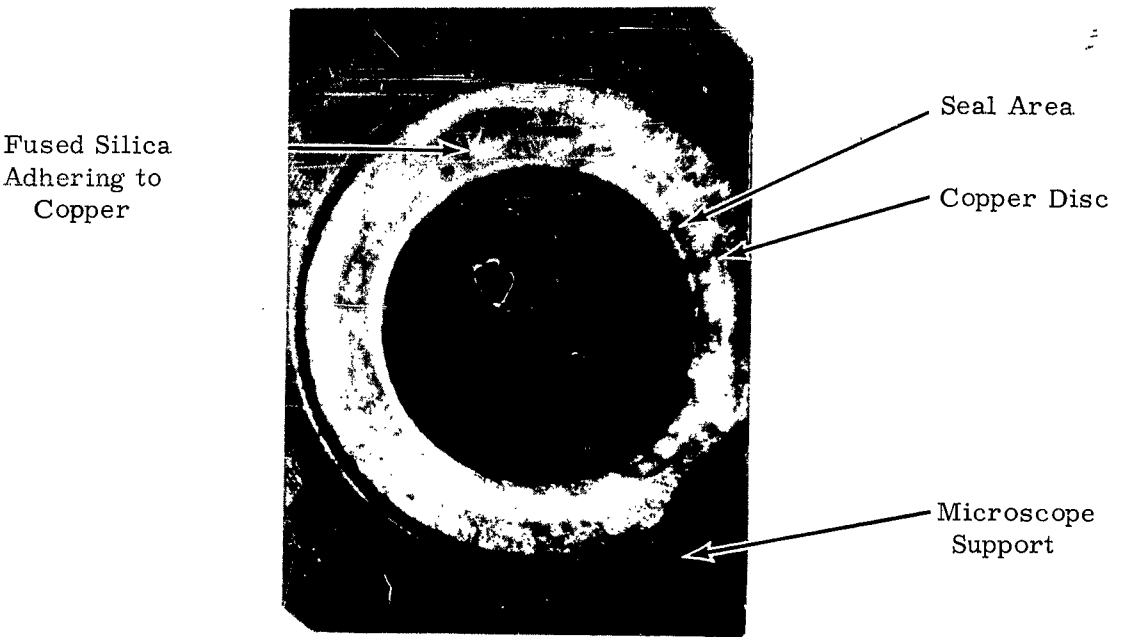


FIGURE 23 - FUSED SILICA-TO-METAL
EXPERIMENTAL SEAL NO. 23

6.9 Magnification

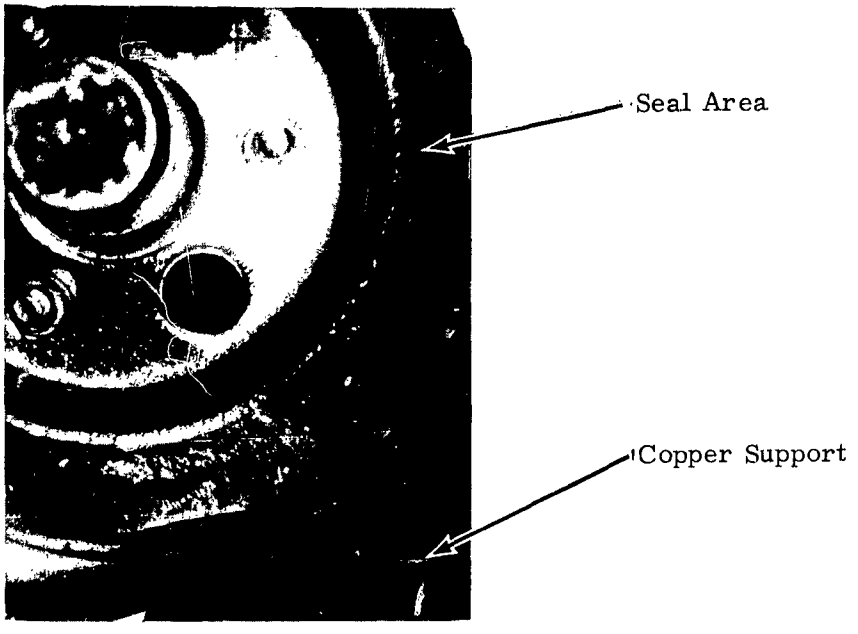


FIGURE 24 - PHOTOGRAPH SHOWING ADHERENCE OF SOFT GLASS TO COPPER IN BUTT-TYPE SEAL

6.9 Magnification

6. Construct and evaluate compression-band seal assemblies using metals and dielectrics with matched coefficients of expansion.

7. Evaluate copper sleeve thickness effects on compression-band window assemblies using Hot Form No. 2 and Frenchtown No. 4462 ceramic.

8. High-temperature bake test compression-band window assemblies fabricated from materials having matched coefficients of expansion.

9. Start cycle life tests on compression-band window assemblies.

10. Continue to evaluate optimum seal parameters for fused silica-to-copper seals using "butt-type" experimental test window assemblies.

11. Make tapered, balanced-type, fused silica-to-copper window seal assemblies.