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DESIGN AND OPERATION OF 1800°F PUMPED BOILING RUBIDIUM LOOP SYSTEM AND DETERMINATION OF DENSITY AND VAPOR PRESSURE RUBIDIUM BETWEEN 174° AND 1800°F

Final Report for Period February 1, 1960 Through May 31, 1961

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METALS, CERAMICS, AND MATERIALS

Final Report (Report No. 16)
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DESIGN AND OPERATION OF 1800°F PUMPED BOILING RUBIDIUM LOOP SYSTEM AND DETERMINATION OF DENSITY AND VAPOR PRESSURE RUBIDIUM BETWEEN 174° AND 1800°F

RUBIDIUM CORROSION AND PHYSICAL PROPERTIES
EVALUATION PROGRAM

Space Power Systems Technology Studies

USAEC Contract AT(04-3)-251
Project Agreement No. 5

Originated by: Peter F. Young
Peter F. Young, Project Engineer

Approved by: D E Deutsch
David E. Deutsch, Project Manager

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R. Arabian, M. Perry, R. Carey, J. Jouthas and G. Lundeen of the Metallurgy Department.

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E.Wolf and D. Bush of the Contracts Department.

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ABSTRACT

A pumped boiling rubidium loop was designed, operated, and analyzed by Aerojet-General Nucleonics. The loop simulated some of the characteristics of a boiling rubidium space power conversion system. Such a system would involve either a reactor or heat exchanger as the power source, and a turbine and radiator as the power conversion and vapor condensing equipment. This loop was used to study instrumentation and control problems and effects of rubidium corrosion on containment materials, and to gain some rough physical property and heat transfer data on rubidium. The first loop operated with 316 stainless steels and was run for 172 hours at various temperatures, the maximum being 1550°F boiling. The 316 stainless steel loop was operated only a short period since it was intended as a system shakedown measure.

The second loop was a Cb-1 Zr loop alloy operated for 455 hours total at 1840°F and a vapor quality of 9%. In the latter loop, the last 335 hours of operation were continuous. Stable boiling at this vapor quality level was achieved without the use of special liquid vapor phase separation devices. Operation of the loop was stable in all phases; however, overheating of the environmental chamber was observed due to insufficient insulation on the loop end resultant high heat losses from the loop heater and boiler. The columbium loop run was

terminated by a cracked weld at the entrance to the condenser section in the hot portion of the loop. Negligible corrosion by the rubidium on both loops was observed. Some carbide penetration on the outside of the columbium loop was observed which apparently was caused by carbonaceous impurities in the argon gas. The failure of the weld at the entrance to the condenser was believed to be caused by carbide penetration effects. The protective coating of zirconium foil wrapped around the columbium loop was observed to fulfill its function and to be entirely necessary to insure the long life operation of the columbium loop. However, some alloying of this foil with the columbium occurred at the hottest portions of the loop. Improved and refined existing metallographic techniques were developed for the evaluation of refractory metal corrosion. Results showed corrosion of refractory containment materials by rubidium is not critical in this temperature range. Rather, the critical corrosion occurs from the contaminants in protective and environmental atmospheres.

Preliminary boiling refluxing tensile tests utilizing 316 stainless steel and Cb-1 Zr alloy were operated for short times up to 1800°F. Negligible corrosion was observed. Longer time and more closely controlled tests are necessary to fully determine what the corrosion effect of rubidium on 316 stainless steel and Cb-1 Zr alloy is for this temperature range.

The density of rubidium was measured between 174° and 1307°F by an expansion method. The height of the rubidium in an enclosed tube was measured by electrodes which shorted out as the column of rubidium rose. The data conforms within ± 0.01 grams/cm to the equation:

$$B = 1.5005 - 2.62 \times 10^{-4} t + 3.94 \times 10^{-8} t^2$$

The total vapor pressure of rubidium was measured between 1000 - 1800°F by measuring the pressure of a sealed capsule. The data conforms to the following equation within ± 2% or ± 0.2 psi, whichever is higher over the range 1000 - 1800°F. The equation is as follows:

$$\log_{10} P = \frac{-4688}{T} - 1.758 \log_{10} T + 10.185$$

(Pressure in psia and temperatures in °K.)

This equation gives the boiling point of 1244°F. In the same apparatus used to measure vapor pressure an attempt was made to measure the latent heat of vaporization by a direct method at different temperatures. The design of the apparatus was basically correct, but an error in instrumentation installation prevented measurement of the actual boiling and condensing rates inside the apparatus. Modifications of the apparatus to enable the direct measurement of latent heat of vaporization are being made.

PROGRAM SUMMARY

The purpose of this program was to provide selected materials compatibility information and physical and thermodynamic properties data on rubidium metal at elevated temperatures in order to evaluate this material as a potential space power system turbine working fluid. The materials compatibility information was gained from brief static capsule tests and from a high temperature pumped boiling rubidium loop system. The physical and thermodynamic properties data were obtained from specially designed density, vapor pressure versus temperature, and latent heat of vaporization experimental apparatus. The milestones achieved on this program are as follows:

1. Design and operation of stainless steel and a columbium boiling refluxing corrosion capsule experiment.

2. Design of instrumentation and control system enabling stable boiling and unattended operation of a pumped-boiling-high-temperature liquid metal loop system.

3. Design, fabrication and operation of a stainless steel-contained pumped boiling rubidium corrosion loop system which operated for 172 hours. The last hour of operation was at 1550°F boiling.

4. Fabrication and operation of a columbium alloy-contained pumped boiling rubidium corrosion and thermodynamic test loop at a maximum temperature of 1840°F for a period of 330 hours. Total operating time of the loop was 455 hours. The 330 hours of continuous unattended operation of the loop in a stable boiling condition indicates the possibility of achieving reliable nuclear space power systems with this technology.

5. Measurement of the density of rubidium liquid from 100° to 1350°F with an accuracy of $\pm$ 0.005 gm/cm^3.

6. Design and construction of an apparatus to directly measure latent heat of vaporization of rubidium metal from its boiling point (1270°F) to 1800°F.

7. Design, construction, and operation of an apparatus for measurement of vapor pressure versus temperature of rubidium from 100° to 1800°F with an accuracy of $\pm$ 0.2 psi.

8. Design, building and operation of a pilot model filtering-gettering and distillation liquid metal purification train which produces high purity rubidium.

9. Improvement and refinement of existing metallographic techniques for evaluation of refractory metal corrosion.

The following pages give a brief summary of the work completed during this program. A detailed account of program accomplishment is contained in the Appendices.

I. CAPSULE CORROSION STUDIES

Two static corrosion capsule studies were made to determine the compatibility of 316 stainless steel and Cb-1% Zr alloy with rubidium at temperatures of approximately 1800°F. The first test evaluated 316 SS as a containment material for short time use and the second test merely confirmed the Cb-1% Zr alloy to be used as the reference material for fabrication of the pumped loop system. The results of the 316 stainless steel capsule test indicated that this material was adequate for containment of rubidium liquid metal at a temperature of 1800°F for periods up to 50 hours. It was thus shown that the test apparatus to be used during the thermodynamic and physical property measurements could be fabricated of 316 stainless steel or a like non-refractory metal alloy for short time operation with the assurance that this material would be adequate for this application.

A suitable heating element for the columbium test run was commercially unavailable and this forced termination of the capsule test short of the desired 1000 hour operation. This test was not critical, however, since prior work with the alloy has shown it to be satisfactory for containing more corrosive alkali metals at the same temperature.

This data is scheduled to be redetermined under more closely controlled environmental conditions in the rubidium corrosion capsule program, USAEC Contract AT(104-3)-368, Project Agreement No. 1. The results of this program will show related corrosion data and containment material solubility data in rubidium at temperatures from 1000° to 1800°F. Materials data will become available starting FY 62.

II. LOOP PROGRAM

The loop studies provided information on the design, control and instrumentation of boiling flowing liquid metals systems, the corrosive effects of boiling flowing rubidium, and the specific heat of rubidium. The temperature range of interest was 1000° to $1800^{\circ}F$.

A natural or thermal convection loop would have satisfied most of the requirements of this program. However, it was felt that a more significant state-of-the-art advancement was desired; a forced circulation loop was thus designed as the basis for this program. Many more problems of design, control and cost were inherent in such a forced convection loop system, but the tests performed and resulting data obtained are more applicable to actual full scale power system design.

A. LOOP SYSTEM DESIGN

The loop system consisted of three distinct parts or subassemblies: the rubidium or primary loop, the argon gas loop, and the water circulation loop. (A schematic of the loop system is shown in Figure B-1 following page 33.)

The rubidium or primary loop circulated the rubidium, heated it at one end, cooled it at the other, and maintained a pressure on it to control the boiling temperature. Heat was added to the system by electrical energy through direct resistance heating. The rubidium was circulated

by an electromagnetic pump. Pressure was maintained on the loop by balancing a pressure of argon gas over the rubidium in the surge tank against the vapor pressure of the boiling rubidium at the temperature of operation. Heat was extracted from the rubidium loop by a condenser-cooler situated on the opposite end of the loop from the heater. The condenser-cooler provided a temperature differential in the circulating rubidium, and simulated a space power system heat rejection radiator. In this simulated system, engineering considerations dictate that the heat be removed from the rubidium cooler by a flow of argon gas rather than by radiation heat transfer.

The argon gas loop served the double purpose of removing heat from the rubidium loop and of providing a protective atmosphere for the columbium loop in the environmental chamber. A large axial fan blew the argon through the system. A diversion valve and bypass line controlled the argon flow to the cooler and thus regulated the heat extraction from the rubidium condenser-cooler. After leaving the rubidium condenser-cooler, the argon flowed through a gas to water heat exchanger to remove heat picked up from the rubidium. About 1 to 2% of the argon flow was then diverted through a purification train to remove contaminants including O_2, N_2,H_2, and H_2O. This flow then rejoined the main stream, where it flowed into the environmental chamber and back to the blower. The argon was sampled at various points in the system to determine purity, problem areas of out-gassing, and effectiveness of the purification train.

The water circulation loop served the purpose of extracting the heat the argon picked up when it circulated over the rubidium to argon heat exchanger. A small pump circulated the water through a control valve to the heat exchanger and out to a cooling tower which recooled the water.

The three loop systems were interlocked by the necessary instrumentation and controls to provide stable, reliable operation. The instrumentation and control consisted of the following:

1. Temperature readout of loop, container system, and cooling system by chromel-alumel and platinum-platinum rhodium thermocouples.

2. Pressure readout and automatic control of loop system.

3. Rubidium flow rate readout and manual control by liquid metal pump and flowmeter system.

4. Argon and water cooling systems flow rates by automatic control of temperature and flow rate.

5. Manual loop heater control.

B. STAINLESS STEEL LOOP OPERATION

The initial program schedule called for a fabrication of only one loop system. This was to be made from the reference Cb-1% Zr alloy. After a thorough study was made of the problems involved in fabricating and operating refractory metal loop systems, it was decided to fabricate two loop systems -- the first, of 316 stainless steel, would be run at 1400° to 1600°F maximum for a limited time, and the second, of the Cb-1% Zr reference alloy, would run for full duration at 1800°F. The first, or stainless steel "shakedown" loop, was considered for the following reasons:

1. It would permit the shakedown of this system and auxiliary equipment at temperatures between 1400° and 1600°F with a relatively inexpensive and easy to fabricate stainless steel loop system without risking expensive, difficult to fabricate columbium material with an untried control system.

2. It would permit design modifications to be made to the loop system before operating the columbium alloy loop.

3. It would permit fabrication and assembly experience to be gained prior to the columbium alloy loop run.

The fabrication of the stainless steel loop proceeded without significant difficulties according to the blueprint and fabrication description as found in Appendix B, Section 11. Early tests showed that a sleeve-type joint with a saddle weld was most satisfactory considering joint stresses and weld penetration desirable for this application. Following fabrication of the stainless steel loop, it was pressure tested and helium leak checked. It was found to be leak tight and capable of withstanding anticipated pressures during the loop run. The stainless steel loop was then assembled on the loop support rack. The auxiliary equipment, such as pressurizing surge tank, liquid metal pump and flowmeter, insulation, thermocouples, etc., were mounted, the rack inserted into the environmental chamber, and the necessary connections made. The loop run was then started.

Loop startup procedures were initiated on 11 January 1961. Erratic flow in the liquid metal pump was attributed to gas bubbles in the loop caused by improper loading procedures. A small design change was indicated for the columbium loop run to eliminate this problem. The entrapped gas bubbles were finally removed from the loop system and the test proceeded with application of heat to the preheater and boiler.

Over a period of nine days from 11 January until 20 January various instrumentation and control and system shakedown tests were conducted. Modifications were made to the pressure control system to provide improved control. Additional sealing insulation was added to the argon to rubidium heat exchanger in an effort to keep the environmental chamber temperature below the desired maximum of 150°F. During this time specific heat determinations were conducted. The operation of the liquid metal flowmeter and pump was evaluated. On January 20 a successful attempt was made to boil the rubidium in the loop system. The boiling test was limited to the short run to determine whether stable boiling could be achieved in the loop to about 15% vapor quality. The length of this test run was limited because the environmental chamber temperature was increasing above 200°F due to the additional power input to the loop system. This increase in temperature was considered

dangerous to certain electrical equipment inside the chamber and it was deemed advisable not to risk failure of these electrical components by exposure to higher temperature.

During this boiling test checks were made to determine that boiling was actually taking place. When pressure was held constant and power was increased, no increase in temperature occurred, the extra power merely increasing the vapor quality. If power was held constant and pressure was increased, the temperature rose -- indicating the rubidium was then boiling at a higher temperature. The pressure and power were gradually increased in increments until the last test of the day was achieved. At this point the loop was operating at a maximum measured temperature of 1450°F (1550°F actual calculated from vapor pressure data) with a flow rate of 10 fps, a pressure of 60 lbs psia, a total power input of approximately 17.02 kilowatts (15.2 kilowatts net to the rubidium), and a vapor quality of 15%. Boiling was stable throughout the entire boiling test. No attempts were made to increase the temperature beyond the 1450°F measured temperature; the stainless steel material was in the region of maximum capabilities at this temperature and the environmental chamber temperature was higher than desirable. The test was then terminated, the loop shut down and the rubidium dumped back into the fill tank.

The stainless steel loop was then removed from the environmental chamber, cut up, and inspected to determine the corrosion effects on the tube material and weld joints.

Mass transfer effects were not present. This would be expected since the loop system did not operate at an elevated temperature for a sufficient length of time to produce these effects. Corrosion of the stainless steel was small. Results showed only minor penetration and solution of the tube wall. Thus, 316 SS seems to be adequate for short time operation at 1000° to 1550°F in lightly stressed applications.

No definite conclusions can be drawn as to the corrosive effects of rubidium on stainless steel in this system since generally low temperatures were observed throughout most of the duration of the test run and since the total loop running time was relatively short at temperatures where significant corrosion could be expected to result. Even the short time data, however, shows that for temperatures in the medium ranges, a non-refractory alloy could be useful as a containment material for liquid and vapor rubidium. Possibly a full scale loop test should be done in this time, temperature, material regime with one of the high strength non-refractory alloys to evaluate their use as economical solutions to the containment material problem.

C. COLUMBIUM LOOP OPERATION

Preparations were started for the columbium loop run on completion of the stainless steel loop test. Considerable care was taken in rendering the entire loop complex vacuum tight. Thermocouple packing glands were replaced, "O" ring seals of high-temperature resistant materials were respecified, leaking weld seams in the environmental chamber were repaired, and a general over-all vacuum system shakedown was conducted. These changes resulted in an empty chamber leak rate of less than 1500 micron liters per second at a chamber pressure of less than 15 microns after a pump down of approximately 24 hours from atmosphere. Further leak correction and design modification achieved a leak rate of 1500 micron liters per second at approximately 7 microns pressure after a chamber pump down of approximately 2 days with all equipment including the loop installed in the environmental chamber.

Methods of welding and forming the Cb-1% Zr alloy were studied. The columbium material received from Wah Chang Corporation was not stress relieved after the final drawing operation, and some difficulty was experienced in bending tubing and machining the plate material. Some vacuum stress relieving was done at approximately 1850°F to improve machinability. Welding tests were conducted in two different controlled environments. The first was a small evacuatable welding chamber. Weld tests were performed under argon cover gas to determine welding currents and minimum techniques necessary to obtain adequate

weld ductility. The second environment investigated was a small flush-box type apparatus shown in Figure B-8 following page 68. This enabled field welds to be made so that final assembly welds could be made with the loop assembled on the supporting rack. This was necessary since no welding chamber was available large enough to accept the loop for the last weld joints in the final assembly operation. Results of the chamber and the flush-box welding indicated that generally clean welds could be made with the Cb-1% Zr alloy. These welds, although not completely ductile, were sufficiently strong to satisfy the initial requirements of the program.

The fabrication of the actual loop system then proceeded to completion. The liquid metal pump cell was fabricated by an outside vendor. It consisted of a specially shaped columbium tube to which nickel bus bars were brazed. The resulting brazed joint was not entirely satisfactory; however, it was adequate for the purposes of this loop run. After the final weld assemblies were made, the loop was x-rayed and pressure and helium leak checked to determine if any cracks or voids existed in the welds. The rubidium to be used in this run was purified by filtration, gettering and distillation. This lowered the O_2 content from 500 ppm to 60 ppm and the N_2 content to below the limits of detection. The columbium loop was then insulated, thermocouple instrumentation attached, and the loop system inserted in the environmental chamber. All controls and instrumentation were checked out and the chamber and loop were pumped down to less than 10 microns and back flushed with argon gas.

The rubidium was loaded into the loop and final check out of the system was made. Difficulty was experienced in starting the rubidium flow in the loop system; apparently columbium is much more difficult to wet than stainless steel, and much higher temperatures are required for wetting. A heater had been inserted previously into the liquid metal pump; however, no heater was in the flowmeter and therefore flowmeter readings were initially erratic due to this wetting problem. Eventually the liquid metal pump cell was heated sufficiently to allow a satisfactory rubidium flow in the loop. Various adjustments were

made in the system and the actual loop run started. A number of temperature cyclings occurred due to minor adjustments in the instrumentation and control and auxiliary systems. Stable boiling operation in the loop was achieved at 1850°F at a vapor quality of approximately 9% and a flow rate of 5 fps. This loop run continued relatively unattended under stable boiling conditions for about 330 hours until voluntary shutdown procedures were initiated. A small leak appeared at the boiler outlet section and the loop was shut down to determine the cause of the exhaust of rubidium vapor from the loop system. It was not feasible to continue the operation or to attempt repair of the loop. The system was shut down and the loop extracted from the chamber, disassembled and analysis procedures started.

Traces of carbon or CO or CO_2, O_2 and N_2 in the argon environmental gas severely attacked the zirconium foil wrapped around the loop tubing and the columbium itself where it was exposed. This was especially true in the high temperature portions of the loop. In some cases the columbium was severely penetrated by carbide stringers. The corrosion was more severe on the outside of the tubing than on the inside next to the rubidium. No significant mass transfer effects were found, indicating the solubility of the columbium alloy in the rubidium was small. The loop cold trap was clean, indicating the loop system had generally low impurity content.

Corrosion of the columbium tube by the rubidium was negligible. The impurity level of the rubidium was low and relatively unchanged at the end of the test.

III. THERMODYNAMIC AND PHYSICAL PROPERTY MEASUREMENTS

Test equipment was designed and fabricated to measure the density, vapor pressure and latent heat of vaporization for rubidium over the temperature range 1000° to 1800°F. Measurement of the specific heat of liquid rubidium as a function of temperature was planned for the corrosion test loop by a heat balance technique.

The density of liquid rubidium was measured by an expansion-level wire method. Data points were obtained from the melting point (102°F) to 1307°F. At temperatures higher than 1307°F it was found that the increased amount of rubidium vapor interfered with the measurement apparatus. The following formula for liquid rubidium density as a function of temperature was developed from the measured data points:

$$\text{Density, g/cm}^3 = 1.5005 - 2.624 \times 10^{-4} t + 3.94 \times 10^{-8} t^2$$

Where t = temperature °F

It is estimated that this relationship will give the correct density to $\pm$ 0.005 g/cm^3 over the temperature range 102 to 1300°F. It is further estimated that its extrapolation into the range 1300 to 1800°F is within $\pm$ 0.01 g/cm^3.

The saturated vapor pressure of rubidium was measured over the temperature range 1000 to 1800°F in the boiling apparatus. Over this range a total of 60 data points was obtained. The following formula

for the saturated rubidium vapor pressure was developed from the data points:

$$\log_{10} P = \frac{-4688}{T} - 1.758 \log_{10} T + 10.185$$

where P = Vapor Pressure in Atmospheres
and T = Temperature ^{o}K

This equation gives a normal atmospheric pressure boiling point of 1244°F. It is estimated that this relationship will give the correct vapor pressure over the range 1000 to 1800°F to $\pm$ 0.2 psi or $\pm$ 2%, whichever is larger.

Accurate measurement of specific heat of liquid rubidium using the corrosion loop could not be obtained. Analysis of the test data led to the conclusion that the loop flow measurement apparatus was defective to a point where it was not sufficiently accurate to allow the desired specific heat measurements. Work will be undertaken on later projects to obtain accurate calibrations from these electromagnetic type flowmeters. Based on existing low temperature specific heat measurements, and assuming that the change with temperature is similar to that of sodium and potassium, the following relationship is recommended for use between the melting point and 1800°F:

$$\begin{bmatrix} \text{Specific Heat} \\ \text{of Liquid Rubidium} \\ \text{Btu/lb-}^{o}\text{F} \end{bmatrix} = C_{p_f} = 6.3 \times 10^{-9} t^2 - 1.26 \times 10^{-5} t + 0.0923$$

where t = Temperature ^{o}F

Direct measurement of the latent heat of vaporization for rubidium in the boiling apparatus was not obtained. Analysis of the test operational data has led to the conclusion that although the apparatus functioned as anticipated, in the effort to obtain a high degree of accuracy the radiation sources in the level measurement system were over-collimated. This actually resulted in poor radiation detector response and precluded the ability to measure the rate at which the rubidium was being boiled. It is expected that the apparatus can be satisfactorily modified to allow direct measurement of latent heat of vaporization in the Cesium Evaluation Program.

Using the measured density and vapor pressure data and standard thermodynamic relationships, the properties of specific volume, enthalpy and entropy phases have been calculated for temperatures up to 1800°F. This data is given in Appendix C, Table VIII, p. 152.

IV. CONCLUSIONS

Some general conclusions can be made as a result of studies undertaken on this program.

The static capsule test is valuable for corrosion screening tests. Additional refinements are being carried out on other programs in order to develop the static capsule test as a basic analytical tool.

The tests during this program showed that 316 stainless steel is a satisfactory short-time containment material for rubidium at temperatures up to 1800°F; additional tests should be undertaken to determine the time-temperature compatibility of rubidium with non-refractory alloys such as 316 stainless steel and nickel alloys. This data could be very important in considering the design of space power systems with selection of some materials for medium temperature applications.

The columbium corrosion capsule test did not operate long enough to gain meaningful corrosion data, and this work currently is being carried out on another program. Columbium should be a satisfactory containment material for high temperature rubidium under long time operation above 1800°F. The limits of time and temperature, however, are at present unknown.

The dynamic corrosion loop system designed for this program is highly satisfactory from an operational standpoint. System operation is stable and controllable with respect to flow, input, temperature, heat extraction, and readout of various system parameters. Small modifications should be made to the system to increase the versatility

of the system and to enable achievement of additional data. Some of these modifications are being made for the Cesium Evaluation Program, Contract AT(04-3)-368.

With respect to the loop as a tool for measuring corrosion, the design is basically satisfactory. Due to the relatively short test time, insufficient corrosion results were obtained to evaluate the loop as a basic corrosion tool. It is expected that operation with other materials at longer time and at higher temperature will provide more definite conclusions in the future on this subject.

The columbium loop failure primarily was due to the penetration of the top weld by carbides probably generated by the presence of hydrocarbon greases in the inert atmosphere. Additional work is needed on the elevated temperature protection of these alloys even in supposedly inert atmospheres containing less than 10 ppm total contaminants. Additional studies will have to be done on gas corrosion barriers, reducing the effects of diffusion of these barriers into the loop material and the formulation and use of advanced insulation techniques. These insulation materials should have low thermal conductivity, low outgassing rates and low reactivities with the loop materials. This problem is considered to be of prime importance for the successful ground testing of high temperature refractory metal systems.

Fabrication techniques were considered adequate. Considerable improvement is desired, however, to improve the capability of weld joints to withstand stress and high temperature during long time operation. The columbium loop failure may have been due partially to the fabrication techniques, or possibly unsatisfactory assembly and handling techniques before loop operation. These techniques are being improved for future loop operation.

Techniques for metallography and handling and purification of liquid metal are considered satisfactory for this type of operation. The chemical analysis procedures are considered only adequate at the present time. Additional work is being done in this area of endeavor. The lower limits of detection and the accurate detection of gas content

in the liquid metals and in the argon cover gas system should be improved. Efforts are being made to improve the analysis of oxygen, nitrogen, and other constituents in liquid metals, and additional instrumentation should be located so that continuous monitoring of nitrogen, water, hydrogen, CO, and CO_2 gases in the argon cover gas system can be facilitated.

The thermodynamic and physical properties apparatus was sufficient for this program. Density could be measured to the boiling point of the liquid metal. Vapor pressure versus temperature could be measured up to the limits of temperature design. The latent heat of vaporization apparatus was designed and functioned properly; however, the instrumentation and control should be improved so latent heat of vaporization can be determined directly instead of by calculation from vapor pressure versus temperature curves. Steps are being taken to correct the apparatus design for operation on another contract, the Cesium Evaluation Program.

Specific heat measurements undertaken by use of the corrosion loop system were basically well founded in concept; however, the problem of accurate temperature flow rate measurement rendered the data obtained from the loop run highly questionable. The temperature drop through the liquid metal to the thermocouple located on the outside tube could only be estimated and might be as much as 100°F. This error is not nearly as great as that introduced by the uncertain accuracy of the flowmeter in the loop system. Results of the test compared with relatively accurate calculated specific heat and heat transfer data indicate that the flowmeter used in the system may have been as much as 50% off in flow rate measurement. It is not known exactly what caused this; however, it may have been a combination of electrical feedback from the heating sections, improper calculations of the flowmeter calibration, insufficient knowledge of the temperature coefficient of the electrical resistivity of the columbium and rubidium metals, and insufficient knowledge of wetting phenomena between the rubidium and columbium metals. Unless more accurate methods of flow rate measurement can be employed, specific heat determinations should be done by standard calorimetry techniques. If heat transfer measurements are going to be determined from operation of a dynamic loop system, then more accurate flow rate measurements, by methods such as differential pressure meters coupled with an orifice, should be used to determine the flow rate to a more accurate degree.

APPENDIX A

CAPSULE TEST PROGRAM

1. Statement of Work Scope

The limited capsule test experiments were designed to verify that the materials selected do not undergo severe corrosion in rubidium at the desired temperatures of operation. Since only rough orders of magnitude of corrosion data were desired, the tests were designed to provide the necessary data with a minimum of effort. Originally only one test was to be performed: containment of 1800°F rubidium in a Cb-1% Zr alloy capsule for 500 hours to determine corrosion and possible mass transfer effects. A 316 stainless steel capsule, however, was also run to determine its resistance to corrosion by rubidium. This added test (possibly more important than the columbium alloy test) was performed to demonstrate the feasibility of using stainless steel containment vessels for thermodynamic studies later in the program. Considerable time and expense were saved by avoiding the use of columbium for this application.

2. Design

The boiling refluxing test capsule concept was employed at AGN to furnish a corrosion test method for analyzing effects of boiling liquid metals on containment materials. This method provides more data than the commonly used static capsule test procedures and bridges the gap between static capsule and dynamic loop corrosion tests. Figure A-1 shows the boiling refluxing capsule design. The capsule to be tested

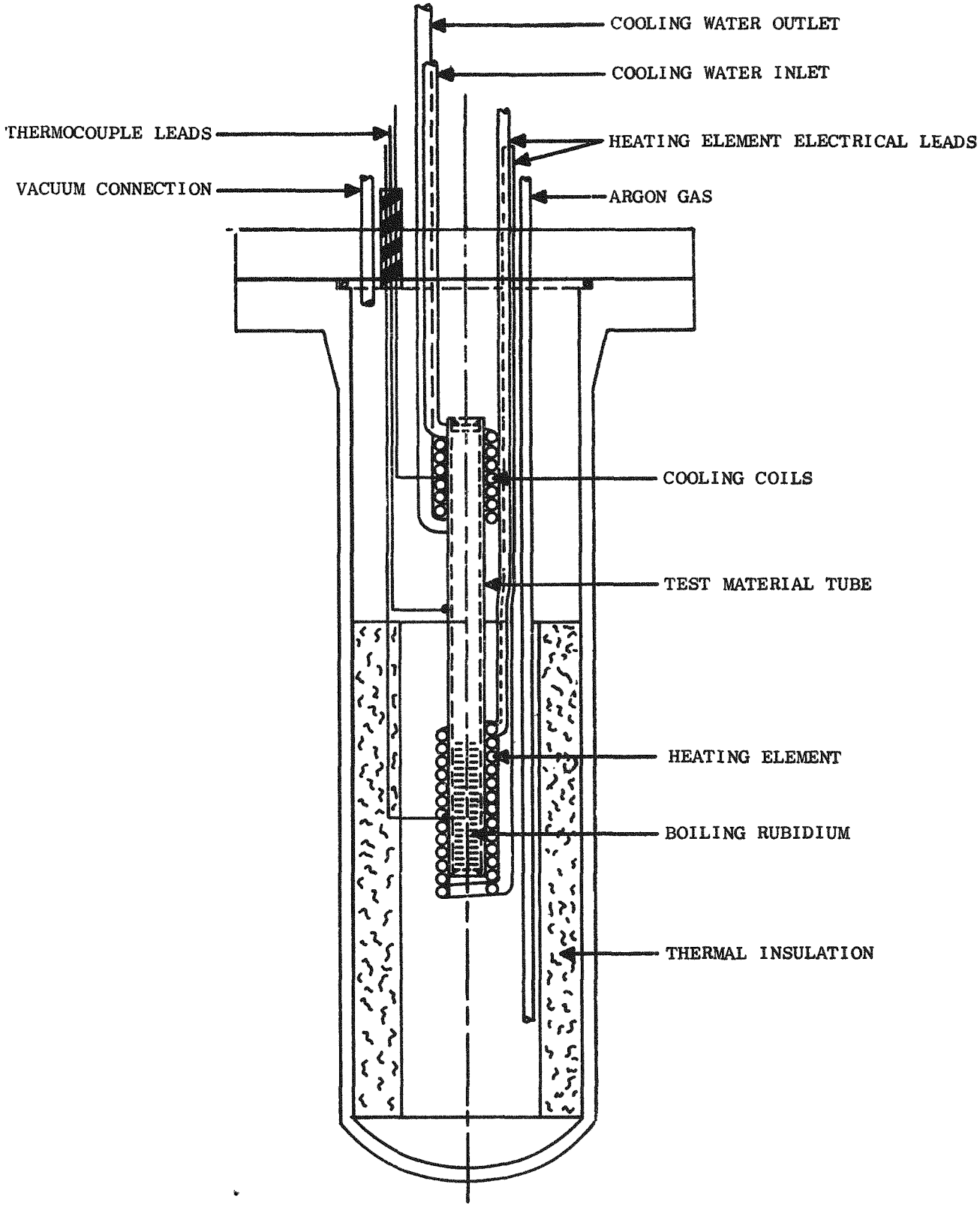


FIGURE A-1. BOILING RUBIDIUM TEST CAPSULE

was filled about half full of liquid metal. A heating coil was wrapped around the bottom and a cooling coil around the top (Figure A-2), and the assembly was inserted into a test bomb arrangement (Figure A-3).

When the heater was turned on, the rubidium boiled; the vapor rose and condensed on the cool upper wall of the capsule, ran down the wall (increasing in temperature as it went), and ran back into the pool of boiling rubidium. Since the condensed vapor was not saturated with dissolved foreign constituents when it was at a point just above the boiling pool, the liquid metal had the greatest capability for dissolving the wall material at this point. Because of the heat absorbed by the vaporizing liquid, the coolest part of the boiling pool was at the surface. The hottest part was at the bottom of the pool, but the actual gradient depended upon the degree of convective stirring. Since unsaturated, saturated, and super saturated conditions existed at different points in the system, varying degrees of corrosion and mass transfer effects were expected to occur along the length of the capsule. Thus, the results of this capsule test provided data on high temperature corrosive effects of liquid and vapor and gave some indications of mass transfer effects to be expected in dynamic loop systems.

The boiling refluxing capsule test system is shown in Figure A-4. The components of the system are (a) test capsule and containment vessel, (b) gas purification system, (c) vacuum purge system, (d) gas bubbler system, and (e) instrumentation and controls. A gas purification train consisting of a molecular sieve at room temperature and a zirconium chip furnace operating at 1500°F, was used to reduce the H_2O and O_2 content of the commercial argon before it passed into the containment vessel. The vacuum system was used to evacuate the containment system prior to backfilling with argon. The gas exhausted through a bubbler system which trapped any rubidium vapor escaping from the capsule and prevented back diffusion of any atmosphere into the container.

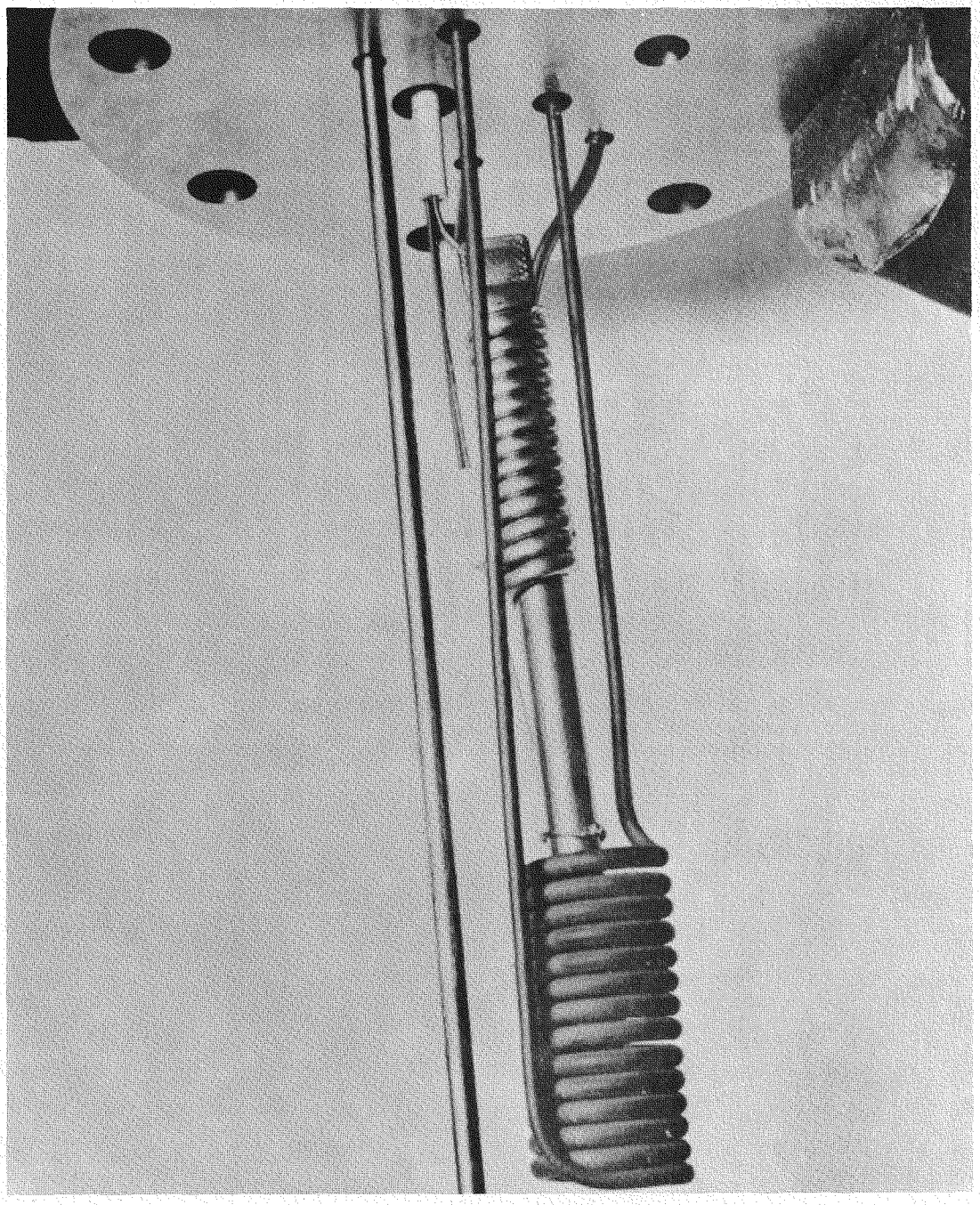


FIGURE A-2. CAPSULE HEATER - COOLER SUBASSEMBLY

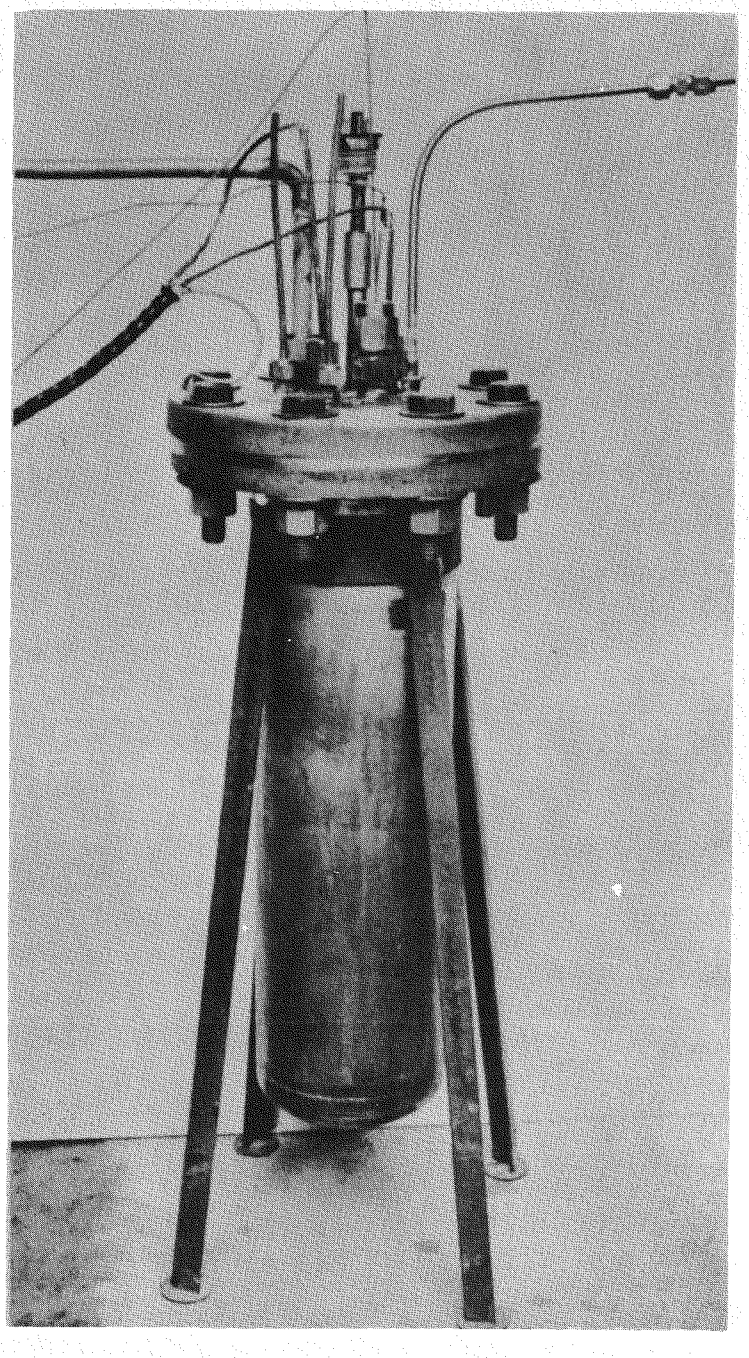


FIGURE A-3. CORROSION TEST CAPSULE FINAL ASSEMBLY

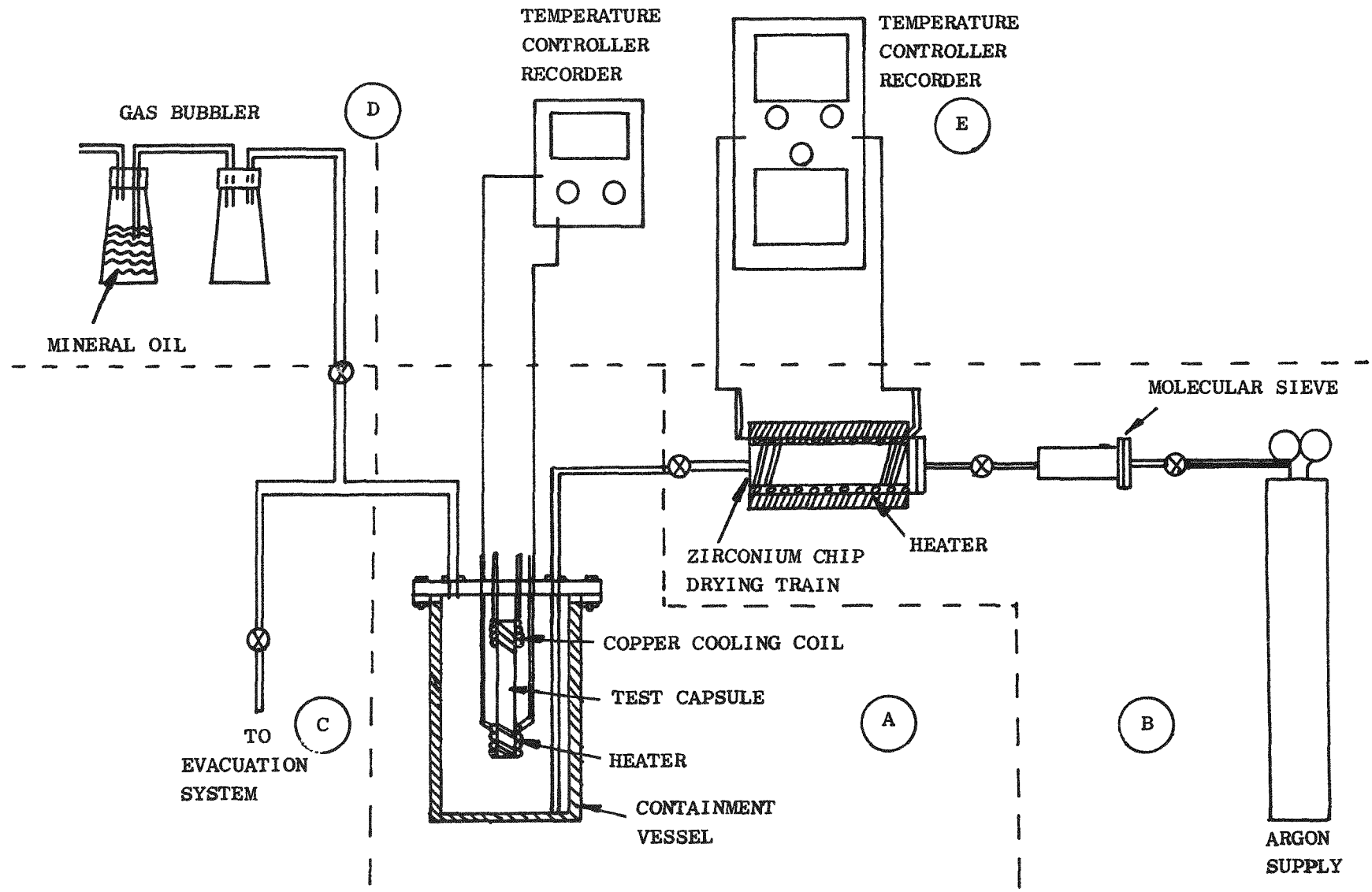


FIGURE A-4. BOILING REFLUXING CAPSULE TEST SYSTEM

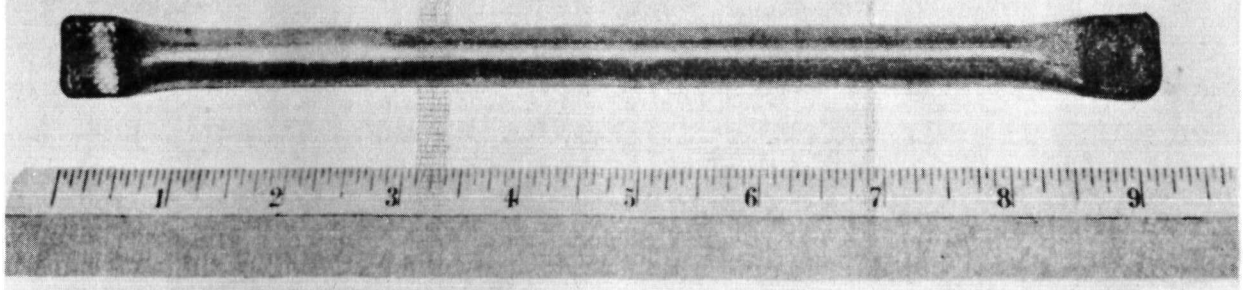
3. Stainless Steel Capsule Test

The first corrosion test capsule run was completed in May. A dry hydrogen annealed type 316 stainless steel capsule 1/2 in. O.D. with 0.049 in. wall containing 12.5 grams of rubidium was subjected to a boiling-refluxing type static capsule test for 50 hours at 1550°F and 50 hours at 1800°F. Estimated internal pressure was 192 psi at 1800°F, resulting in a stress of about 980 psi in the tube wall. The capsule filling and sealing operations were performed under argon gas in the liquid metal handling chamber. A second capsule was filled with rubidium and was used as a blank test for chemical analysis.

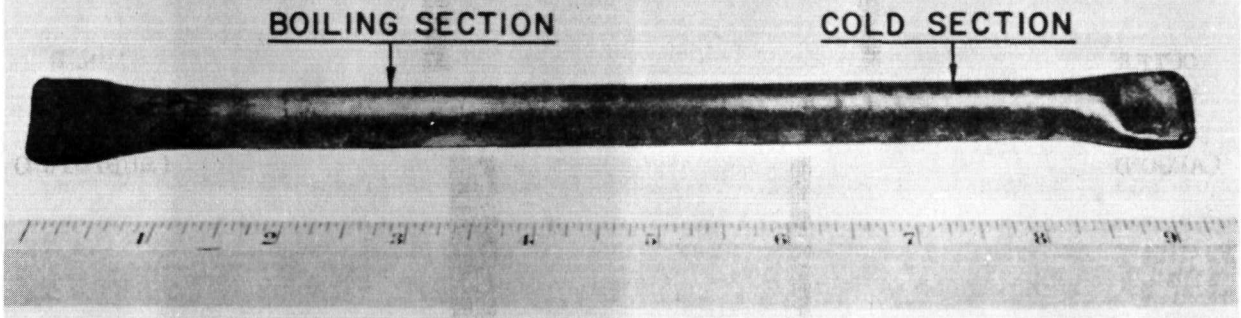
Figure A-5 shows the capsule before and after the test run and the location of the metallography samples. Figure A-6 shows the alloy microstructure. In Figure A-5 note the clear-etched surface in the area containing the boiling rubidium. The rubidium attacked the stainless steel to a small degree. The upper part of the tube containing the gas phase shows various amounts of deposits and discoloration. Figure A-6 shows the as-received stainless steel tubing, and the effects of the rubidium on the stainless steel in the gas, transition and liquid zones. Some surface solution has occurred along with a moderate amount of intergranular penetration. The microphotographs indicate the heaviest corrosion in the rubidium vapor region with relatively smaller effects in the liquid hot and cold zone. The results of this test indicate that 316 stainless steel is an adequate containment material for rubidium in short-time low stress level exposures at temperatures approaching 1800°F. A small deposit of crystals (Figure A-7) found in the cold zone is believed to result from mass transfer effects. The crystals, however, were not recovered in sufficient quantity to make positive identification. The mass transfer deposits occur just above the liquid-gas interface.

The chemical analysis procedures used for rubidium metal gas content were not sufficiently developed to render accurate results and are therefore not reported. Weight gain and loss data were not taken since corrosion effects were small.

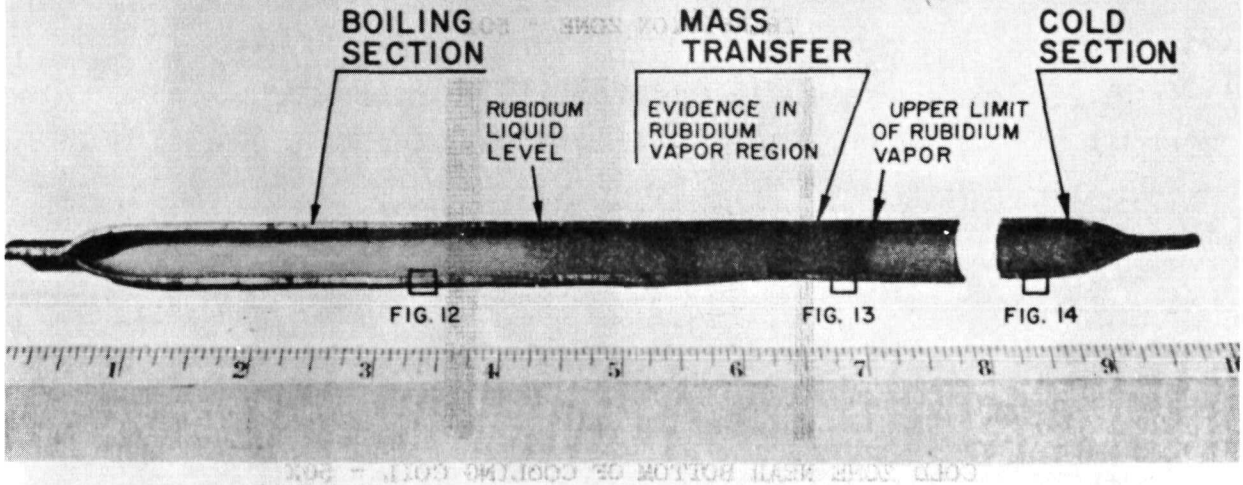
TYPE 316 STAINLESS STEEL BOILING–REFLUXING TEST CAPSULE.



BEFORE TEST

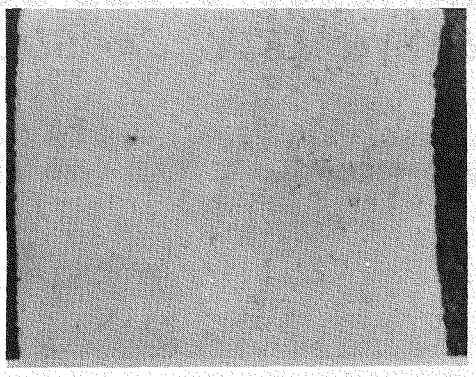


AFTER TEST

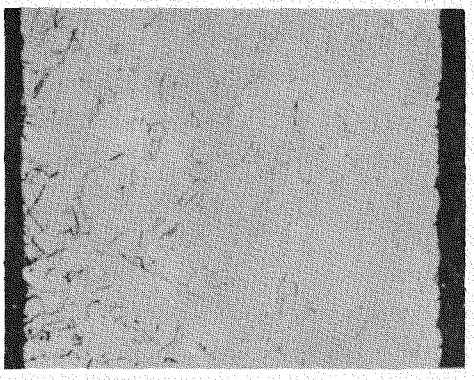


AS SECTIONED

FIGURE A-5



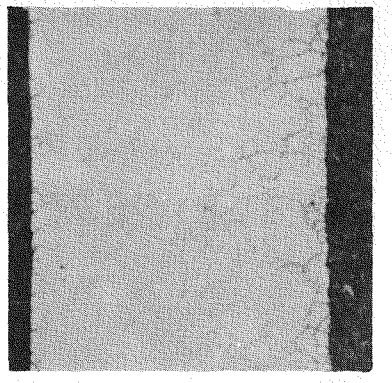
"AS RECEIVED" TYPE 316 STAINLESS STEEL TUBING - 75X



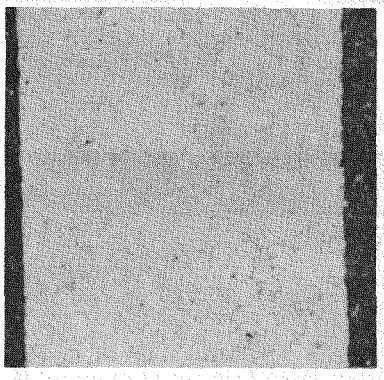
OUTER WALL (ARGON)

SECTION OF WALL FROM THE HOTZONE - 75X

INNER WALL (RUBIDIUM)



TRANSITION ZONE - 50X



COLD ZONE NEAR BOTTOM OF COOLING COIL - 50X

FIGURE A-6. METALLOGRAPHIC RESULTS OF 316 SS CORROSION CAPSULE TEST

ETCHED: 25 GLYCERINE 25 HCl 10 HNO_3

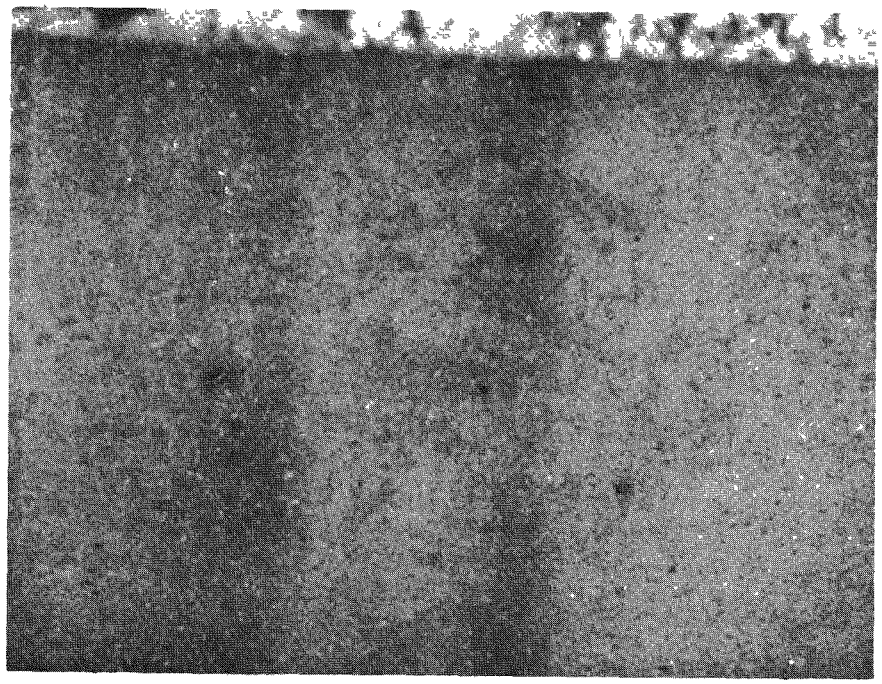


FIGURE A-7. MASS TRANSFER RESIDUE AREA SHOWN IN FIGURE A-5 - MAGNIFICATION 13X

4. Columbium Capsule Test

The Cb-1% Zr alloy test capsule was fabricated and assembled. The capsule was x-rayed to determine the exact height of the rubidium, and the test run started. The capsule was run at a maximum temperature of 1800°F and a minimum temperature of 1200°F. After 16 hours the heating element failed and the test was terminated. Attempts were made to procure a replacement heater; however, the current and heat loads necessary were higher than anticipated and major redesign of the equipment was necessary to continue the experiment. Since this is already in progress on Project Agreement No. 1, AT(04-3)-368, on a more refined and basic testing procedure, the use of this particular furnace design was terminated. This test will be duplicated during FY 1962 on the Rubidium Capsule Testing Program, Project Agreement No. 1, AT(04-3)-368. No results are reported here since the data from this short test run are of little significance.

APPENDIX B

CORROSION LOOP PROGRAM

1. Statement of Work Scope

The containment material found to have the most desirable characteristics was to be selected for the fabrication of a pumped loop to be used for dynamic testing. The pumped loop was to be designed and fabricated to simulate the characteristics of a boiling rubidium power conversion system. This dynamic testing was to be accomplished to investigate the nature and extent of corrosive and erosive attack by rubidium on containment materials in straight and curved pipes, valves, pumps, orifices, and associated equipment in a forced convection boiling heat transfer loop over a range of liquid velocities from 5 to 30 feet per second. The heater was to be capable of providing vaporization of the liquid rubidium up to 40 to 50% vapor quality, at least at the lower fluid velocities. Appropriate precautions were to be taken in the design and instrumentation of the system so that sufficient warning would be available of conditions detrimental to the continued operation of the system. Such conditions include excessive corrosion and attainment of heat fluxes such as might cause heater burnout.

The loop was to be operated with rubidium temperatures at the heater exit between 1800°F and 2000°F, and cooler outlet temperatures between 1000°F and 1200°F. Uninterrupted operation for 1000 hours was to be attempted. The effect of such operation on the system components and subassemblies were to be carefully studied. The following information was to be obtained and reported.

(a) Determination of qualitative corrosive and erosive effects of the rubidium liquid and vapor on the system materials. Special test sections were to be included to determine quantitative corrosion and erosion effects.

(b) Analysis of rubidium for foreign constituents prior to and after loop operation.

(c) Examination of corrosion products chemically and microscopically.

(d) Study of mass transfer effects in the system by sampling the tubing at various points. Loss or gain in weight and/or wall thickness changes were to be the major effects to be studied. Weight change in selected portions of the tubing were to be determined.

(e) A log of the loop operation was to be maintained.

Prior to fabrication and operation of the loop as set forth in the above paragraphs, a prototype loop was to be fabricated and operated utilizing all stainless steel components. The stainless steel loop was to be fabricated and operated (with rubidium at temperatures up to 1000-1500°F) so as to provide experience on system design, fabrication techniques, system control, and over-all system operation. This was to insure the satisfactory operation of the columbium loop later in the program. It would eliminate many of the problems inherent in the new, untried system without risking the expensive, difficult to fabricate columbium loop.

2. System Theory and Analysis

a. System Design Philosophy

The loop studies provided information on the design, control, and instrumentation of boiling flowing liquid metals systems, the corrosive effects of boiling flowing rubidium, and the specific heat of rubidium. The temperature range of interest was 1000 to 1800°F.

A natural or thermal convection loop would have satisfied most of the requirements of this program. However, a more significant state-of-the-art advancement was desired and so a forced circulation loop was designed as the basis for the program. Many more problems of design, control, and cost are inherent in a forced convection loop system such as this, but the tests performed and resulting data obtained are more applicable to actual full scale power system design. The loop system (Figure B-1) consisted of three distinct parts or subassemblies: the rubidium or primary loop, the argon gas loop, and the water circulation loop.

The rubidium or primary loop circulated the rubidium, heated it at one end, cooled it at the other, and maintained a pressure on it to control the boiling rate. Heat was added to the system by electrical energy through direct resistance heating. The rubidium was circulated by an electromagnetic pump. Pressure was maintained on the loop by balancing a pressure of argon gas over the rubidium in the surge tank against the vapor pressure of the boiling rubidium at the temperature of operation. Heat was extracted from the rubidium loop by a condenser-cooler situated on the opposite end of the loop from the heater. The condenser-cooler provided a temperature differential in the circulating

INSTRUMENTATION AND CONTROL SYSTEM FOR BOILING RUBIDIUM LOOP SYSTEM

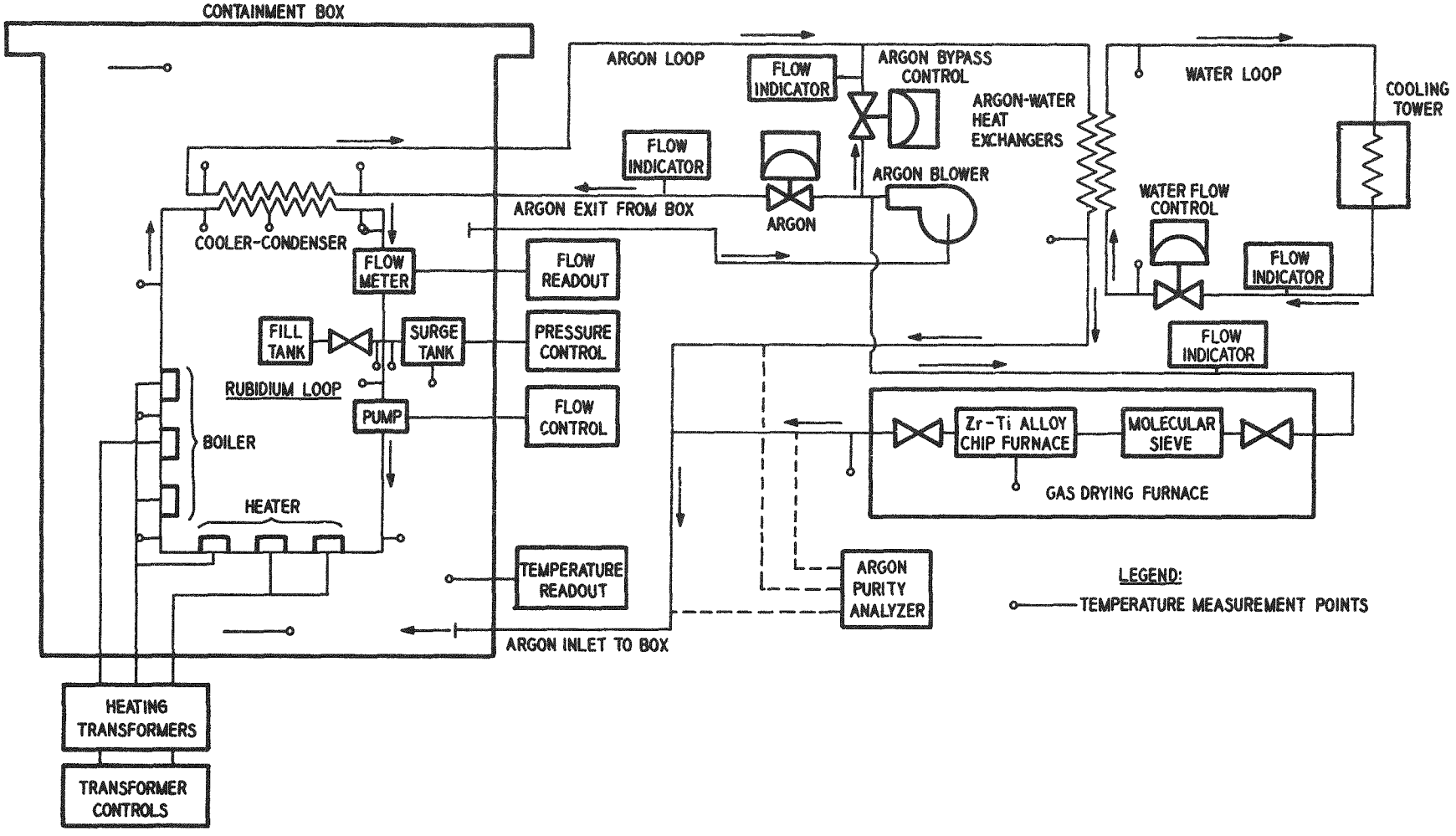


FIGURE B-1

rubidium, and simulated a space power system heat rejection radiator. In this system, engineering considerations dictated that the heat be removed from the rubidium cooler by a flow of argon gas rather than by radiation heat transfer.

The argon gas loop served the double purpose of removing heat from the rubidium loop and of providing a protective atmosphere for the columbium loop in the environmental chamber. A large axial fan blew the argon through the system. A diversion valve and bypass line controlled the argon flow to the cooler and thus regulated the heat extraction from the rubidium condenser-cooler. After leaving the rubidium condenser-cooler, the argon flowed through a gas-to-water heat exchanger to remove heat picked up from the rubidium. About 1 to 2% of the argon flow was then diverted through a purification train to remove contaminants including O_2, N_2, H_2, and H_2O. This flow then rejoined the main stream, where it flowed into the environmental chamber and back to the blower. The argon was sampled at various points in the system to determine purity, problem areas of out-gassing, and effectiveness of the purification train.

The water circulation loop extracted the heat the argon picked up when it circulated over the rubidium-to-argon heat exchanger. A small pump circulated the water through a control valve to the heat exchanger and out to a cooling tower which recooled the water.

The three loop systems were interlocked by the necessary instrumentation and controls to provide stable, reliable operation. The instrumentation and control consisted of the following:

(1) Temperature readout of loop, container system, and cooling system by chromel-alumel and platinum-platinum rhodium thermocouples,

(2) Pressure readout and automatic control of loop system,

(3) Rubidium flow rate readout and manual control by liquid metal pump and flowmeter system,

(4) Argon and water cooling systems flow rates by automatic control of temperature and flow rate,

(5) Manual loop heater control.

b. Corrosion Loop Design

The pressure drop, heating, cooling, and flow characteristics of the rubidium loop and heat exchanging systems were analyzed. This data provided basic information for good engineering design leading to stable loop operations. This analysis was necessary because a boiling two-phase loop is much more difficult to properly design than the single phase non-boiling loop systems generally in use at the present time. Basic heat transfer and fluid flow calculations determined the size of the heater and cooler section as follows:

(1) Heater

The heating system was divided into two sections. The first was to heat the rubidium to a temperature at or near the maximum of about 1900°F. The second, or boiler section, provided additional heat to boil the rubidium and raised the vapor quality to the desired amount at the boiler outlet. The dimensions, heat transfer and pressure drop characteristics of the heater and boiler were as follows:

<u>Loop Heater</u>

Length - 6 ft - 0 in.
Tubing Diameter - 1/4 in. O.D. x 0.150 in. I.D.
Maximum Liquid Velocity - 10 fps
Net Power Input - 7.85 kw
Maximum Heat Flux - 85,000 $\frac{Btu}{hr}$/sq ft
Maximum Temperature - 1900°F.
Maximum Pressure Drop - 0.94 psi
Maximum Vapor Quality - 0%
Mass Flow Rate @ 10 fps - 340 lb/hr

<u>Loop Boiler</u>

Length - 5 ft - 0 in.
Tubing Diameter - 1/2 in. O.D. x 0.400 in. I.D.
Maximum Vapor Velocity - 71 fps

Net Power Input - 15.6 kw

Maximum Heat Input - 117,000 $\frac{Btu}{hr}$/sq ft

Maximum Temperature - 1900°F

Maximum Pressure Drop - 2.3 psi

Maximum Design Vapor Quality - 50%

Operating Pressure @ 1900°F - 185 psia

The boiler was used for specific heat determinations later in the program. No vapor-liquid phase flow separator was used in the boiler or condenser since boiling and condensing were stable without it. These vortex generators consist of a 0.010 in. twisted tape inserted into the boiler section. This provided a rough separation of the liquid and vapor phases of the system and forced the liquid phase into more intimate contact with the tube wall, thereby reducing the possibility of burnout. (Burnout would have resulted in destruction of the loop system when operating at maximum heat fluxes.) This system will be used for heat transfer tests to be performed in a future program.

Direct resistance center tap heating was used in this application, and the heat input for the heater and boiler was separately controlled by two 8 volt 3750 amp transformers.

(2) Cooler

The cooler was designed to provide a maximum ΔT of 800°F when operating the loop at a maximum of 1900°F. The cooler consisted of a simple coil type design. Heat transfer was effected by forced draft of argon gas over the cooling coil. The gas flow was channeled by a cylinder and annulus arrangement. Cooler design performance data were as follows:

Length - 25.6 ft

Tubing Diameter - 0.500 O.D. x 0.049 in. wall

Maximum Temperature In - 1900°F

Minimum Rubidium Temperature Out - 1100°F

Maximum Rubidium Pressure Drop - 8.01 psi

Maximum Rubidium Liquid Velocity Out - 1.28 fps

Maximum Heat Extraction Potential - 96,900 $\frac{Btu}{hr}$

Over-all Heat Transfer Coefficient - 23.1 Btu/hr-ft^2-^oF

Cooling Method - closed cycle argon cooling system

(3) Pump and Flowmeter

The pump and flowmeter were the electromagnetic type. The pump was designed to impart a flow of about 1 gpm at a Δ P of 20 psi to the rubidium. The control system was manual since the electromagnetic pump is an inherently stable system.

(4) Surge Tank

A surge tank was included in the system between the pump and flowmeter. The surge tank acted as a reservoir for the rubidium metal and as an expansion chamber during boiling, and provided a means for prepressurizing the system before startup so that the rubidium would boil at a predetermined temperature and pressure. A rubidium fill line, pressure tap, and liquid level gages were included in the surge tank assembly.

(5) Evacuation Line

An evacuation line was attached to the loop system at the highest point between the end of the specific heat section and the cooler inlet. This line was used to evacuate the loop prior to filling.

(6) Drain-Fill Line

A drain-fill line was provided between the flowmeter and surge tank at the lowest point in the system. This line was used to fill and drain the loop system.

(7) Cold Trap

A cold trap was provided at the lowest coolest portion of the loop. This reduced plugging of the loop by corrosion products in the rubidium

(8) Specific Heat Test Section

The boiler section of the loop with modifications was reserved for liquid phase measurements in Phase II.

c. Argon Loop Design

A blower, mounted on the loop containment box, provided the flow of argon over the cooler to extract heat from the loop system.

This was a two stage axial fan rated at 960 cfm at a ΔP of 0.9 lb at 150°F. This fan was completely sealed into the system and utilized special lubricants and insulation to prevent outgassing.

The gas passed directly from the blower through a control valve and bypass line arrangement. After the gas left the loop heat exchanger, it passed through the argon-to-water heat exchanger, then into the loop environmental chamber where it provided a purging action, and then returned to the blower. Plumbing was 6 in. diameter carbon steel treated to suppress rusting and oxidation. All joints were welded or "O" ring sealed. Total pressure drop in the argon loop was about 0.5 psi.

d. Water Loop Design

The heat absorbed by the argon flow through the rubidium loop heat exchanger was removed by passing it through a gas-to-water heat exchanger. The design specifications were as follows:

Type - Closed cycle water cooling system
Maximum Inlet Temperature Argon - 300°F
Minimum Outlet Temperature Argon - 150°F
Maximum Outlet Temperature H_2O - 85°F
Maximum Inlet Temperature H_2O - 75°F
Maximum Flow Rate Argon - 51,500 lb/hr
Maximum Flow Rate H_2O - 10,000 lb/hr

The argon-to-water heat exchanger was a standard tube-in-shell design connected into the argon system by suitable "O" ring seals. The water cooling was obtained from a closed circuit pressurized plant water cooling system.

3. Instrumentation and Control System Design

In deciding on a rubidium corrosion loop control scheme, the first step was to define the requirements to be imposed on a system. These specifications led to the most direct method to achieve the desired results. In a complex system, there are usually several parameters which can be manipulated to provide control of a given parameter. From a control viewpoint, the choice of which parameter to manipulate goes to the one which has the largest influence on the system and is most easily controlled. Also important are the time lags encountered between manipulation of a variable and the resulting change in the system. The existence of two or more equal time lags (which give 180° or more phase lag at higher frequencies) may produce unstable servo mechanisms and rule out a particular control scheme.

A quantitative study of the influence of the various parameters upon one another was not completed for the rubidium corrosion loop control scheme because it was possible to select the most direct relationships without complex calculations. Table I shows the parameters, numbered 1 to 5, which could be regulated for this loop. The conceivable manipulated variables are lettered A through F. The letters in the boxes indicate the dependence of the parameter on the manipulated one.

O - - - - negligible
S - - - - small
L - - - - large
G - - - - great

TABLE I

LOOP DESIGN PARAMETERS

	Power to Boiler	Power to Heater	Flow Rate	Argon By-Pass Control	Pressure	Water Flow to Heat Exchanger
1. Boiling Point of Rubidium	O	O	O	O	G	O
2. Temperature in Box	S*	S*	O*	O*	O	G
3. Temperature into the Heater	S	S	L	G	O	S
4. Temperature out of Heater	S	G	L	L	O	S
5. Quality of Vapor	G	G	G	S	G	S
	A	B	C	D	E	F

* depending on loop insulation

The design specifications listing the operation required of the control system were:

(1) Continuous unattended operation, without severe drift or transients which would cause shutdown.

(2) Maintenance of ambient temperature in the environmental chamber box below 150°F.

(3) Prevention of boiling in the heater section.

(4) Regulation of boiling point of the rubudium, and maintenance of a given maximum loop temperature.

(5) Prevention of instabilities which might result in burnout of the heater or boiler.

The dependence of the boiling temperature on the pressure is so complete that the control methods suggested did not sense tempera-

ture and adjust pressure accordingly. Rather, pressure was adjusted, sensed, and regulated and the temperature at which boiling took place followed. The variation in the loop pressure drop amounted to a small perturbation but did not seriously influence the boiling point.

The ambient box temperature was most directly controlled by the gas-to-water heat exchanger (intersection 2F of Table I). Here, however, was the possibility of an unstable loop. The temperature of the gas leaving the box would lag behind the temperature entering the box by approximately 30 seconds. The argon flow rate was approximately 13 standard cu ft per second and the volume of the box was about 231 cu ft. The other time lag in the loop was between the movement of the water valve and the temperature of the gas entering the box. This constant was not known exactly but if it were close to 30 seconds, satisfactory operation of this loop would be difficult to achieve. One solution was to decrease the gain, and insert a slow reset correction, resulting in a slow responding control system and sensing the temperature of argon out of the rubidium-argon exchanger. As it turned out, the box temperature did not vary rapidly in relation to the water flow in the heat exchanger. This was mostly due to the large heat load in the chamber resulting from inadequate insulation on the loop heater and boiler.

Perhaps the most important requirement of the control system was to maintain the point at which boiling starts outside the heater but within the first part of the boiler tube. Two methods were suggested to accomplish this:

i) Regulation of the exchanger outlet temperature (intersection 4D of Table I), counting on a constant flow and constant heat power to bring the liquid to a boil at the desired location.

ii) Sensing of the temperature difference across the boiler, and forcing this temperature difference to be a certain small positive amount would actuate the argon bypass valve by balancing this signal against a setpoint signal. This second method adjusts for variations in flow rate and heater power.

Not enough was known about the characteristics of the loop to recommend which method to use. Method 1 would have been satisfactory if the variations in flow rate and heater power proved small during actual running conditions. Otherwise, Method 2, with its more complex sensing and larger control lag, would have been tried. It was found during the loop run that this problem was not critical for the low vapor qualities developed and essentially stable operation of the loop with regard to flow and power input was achieved.

It was assumed that the exact temperature of the rubidium on leaving the cooler would not be critical and would not require regulation, provided it were maintained below 1200°F. Also, the quality of vapor was assumed not critical enough to regulate as long as it remained below 50%; hence, perturbations might cause variations of this parameter. Note that in Table I three of the six possible manipulated variables have been used; control of the flow rate and power input were left available to both the heater and boiler if tighter requirements were imposed.

In comparing the two suggested methods of supplying power to the heater and boiler, the constant voltage source appeared more satisfactory than the constant current source method. The power dissipated in the load would vary inversely with the resistance in the former method (E^2/R) and directly with the resistance in the constant current method (I^2/R). Because the resistance of boiler would tend to rise as the liquid metal boiled, an unstable system could result if the power at any section of the pipe increased with resistance. Regeneration of this type would not be expected if constant voltage were applied.

The application of an analog or a digital computer for use in analysis of a rubidium loop control system was considered with respect to three applications. These reflect the complexity of the loops with the corresponding potential loss in the case of "miscontrol." The applications in order of increasing complexity were assumed to be (A) corrosion studies, (B) heat transfer studies, and (C) power plant operation.

For the corrosion loop it appeared that computer analysis was not needed. Hand calculations of closed loop control were adequate. These calculations showed that a three mode (proportional rate, reset) controller could be adjusted to provide pressure control corresponding to the accuracy of the pressure transducer used. This would give the temperature desired in the boiler portion of the rubidium loop. For the secondary and tertiary loops, two mode (proportional and reset) controllers could easily control the bypass of the argon loop, and the control valve in the water loop. The chance of loss of the system by inadequate control was small, as the controllers could be preset and then adjusted on-line starting with the water loop, followed by the argon loop, and then the rubidium loop. Low power tests could be run to check the response accuracy and sensitivity of each loop and the system before increasing the power.

For the second, or heat transfer, phase of the rubidium loop program, the same comments applied if the system were no more complicated than for Condition A; however, if more stringent requirements are imposed on the system, an analog computer analysis would be desirable.

Digital computer analysis for accurate steady state conditions, and analog computer analysis for dynamic study would have been imperative for Condition C, a rubidium loop type of power plant with a nuclear heat source, where large energy rates were involved. The dollar investment in a nuclear power plant loop, even in a scale model, and the danger of mishap where fission products are involved warrants detailed analysis prior to computer programming. The digital computer is excellent for checking steady state points for the analog computer and giving accurate energy balances around the loop for a multitude of given input conditions. The experience derived from analyzing the characteristics of the rubidium loop and its controls would make design and operation stages of subsequent systems much easier.

In conclusion, the recommended control scheme was based upon the assumed requirements:

(1) Regulation of the boiling point temperature,

(2) Regulation of the box ambient temperature, and

(3) Prevention of boiling in the heater.

The pressure of argon was automatically controlled, thereby setting the boiling temperature. The water valve to the argon-water heat exchanger was automatically controlled. The sensing point for this controller was the temperature of the argon entering the box. The argon bypass valve to the argon-rubidium heat exchanger was automatically controlled. The sensing point for this controller was the cooler outlet temperature.

A loop system control panel was set up as shown in Figure B-2. Nearly all the instrumentation and controls needed to operate the loop were mounted on this panel. The panel enabled the loop operators to monitor and control all the system parameters from one central point.

A system of fail-safe interlocks was used to prevent damage to the loop and the occurence of possible hazardous conditions. Any one of the following conditions would shut down the loop power and activate an alarm aystem:

(1) Low rubidium flow limiter indicating leakage or malfunction of pump or flowmeter.

(2) Loop low and high pressure limiter indicating leakage in the loop or failure of the pressurizing controls.

(3) Low or high rubidium level limiter in surge tank indicating leakage in loop or loss of pressurizing system.

(4) Chamber high temperature limiter indicating that chamber temperature was too high for electrical equipment to function properly.

(5) Argon gas low flow limiter indicating argon cooling loop was not functioning properly.

(6) Water low flow limiter indicating cooling water loop was failing to cool argon gas loop.

(7) Rubidium loop high temperature limiter indicating rubidium temperatures at exit from preheater was high, with resultant possibility of boiling in the preheater.

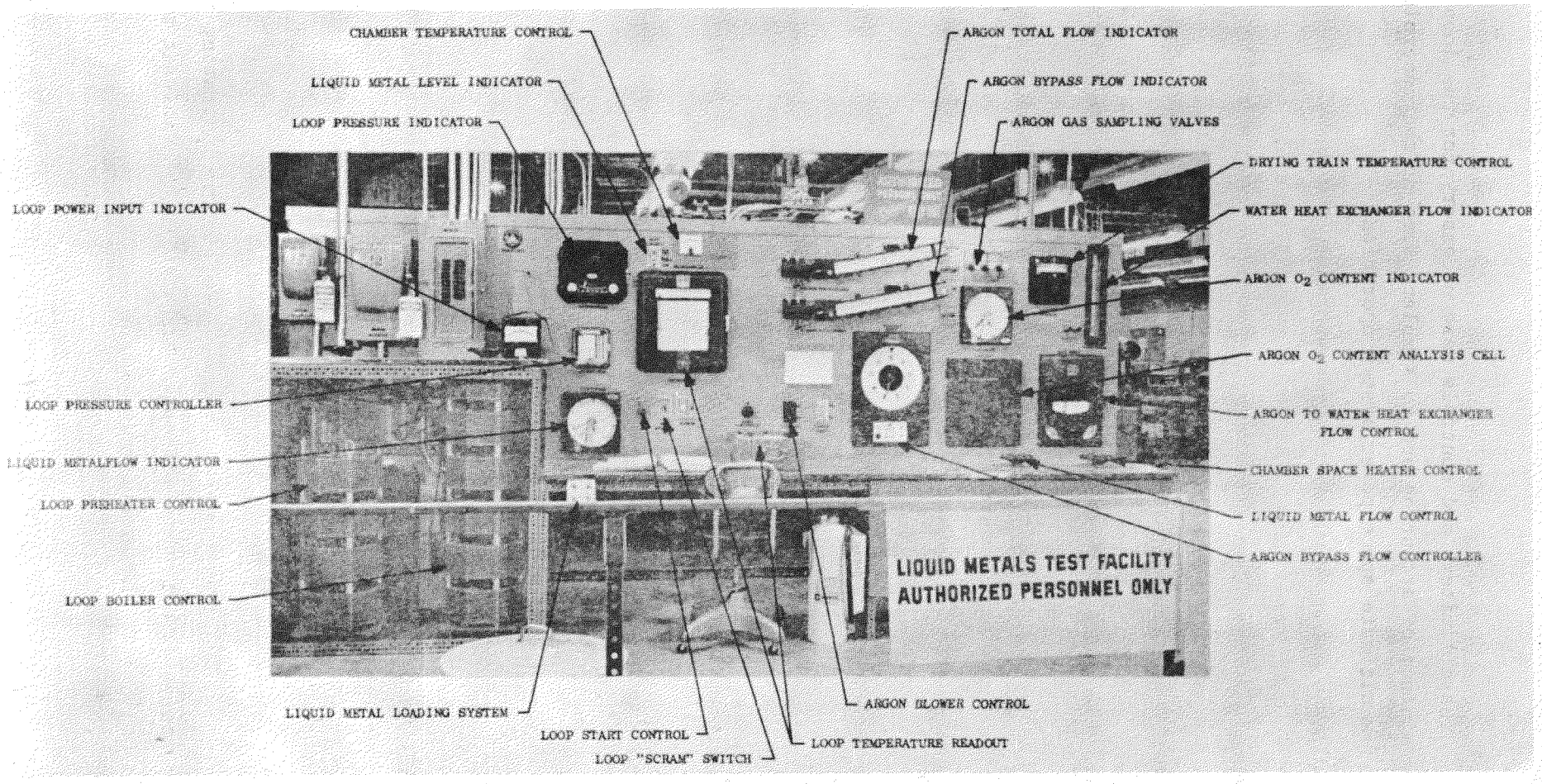


FIGURE B-2. BOILING RUBIDIUM LOOP CONTROL PANEL

These limiters and controls were necessary for the loop to run with a minimum of attention and to simulate some of the controls necessary for ultimate space power system operation.

4. Loop Environmental Facility

The loop environmental chamber shown in Figure B-3 is 7 ft long, 5.5 ft wide, and 6 ft high (inside dimensions). The chamber was designed for use either under an internal gas pressure or at least 1 atmosphere or under a vacuum of 10 microns. The chamber has the necessary lead throughs, view ports, and accessories to provide power, control, and instrumentation of the loop during operation.

Argon plumbing was 6 in. I.D. steel piping. The argon blower was a canned axial fan rated at 960 cfm @ a ΔP of 0.9 psi. The blower motor was glass insulated and had special lubrication to reduce outgassing. The argon-to-water heat exchanger was a steel welded tube type for vacuum service. All plumbing joints were flat gasket or "O" ring sealed for vacuum service. A positive argon pressure of 1 to 3 pounds above atmospheric was maintained in the chamber by a specially constructed valve.

After the loop system was inserted in the chamber and all power, instrumentation, and control leads connected, the chamber was pumped down and purged of air and then backflushed with argon. This process was repeated until the atmosphere was pure enough to prevent undesirable corrosion of the columbium loop in the chamber. No leak rate checks were taken on the empty chamber; however, a leak rate determination was made with the loop assembled and ready for operation. Under these conditions at 15μ to 30μ pressure, the leak rate was about 1200 μ-1/sec. A check was made to determine the O_2 content in the chamber after pumpdown to 10 μ and backflush with 6 to 8 ppm O_2 content argon. Backflush required about 20 minutes and O_2 content in chamber was 14 ppm after chamber was flushed to 1 atmosphere. This value agreed closely with what would be expected from calculated estimates.

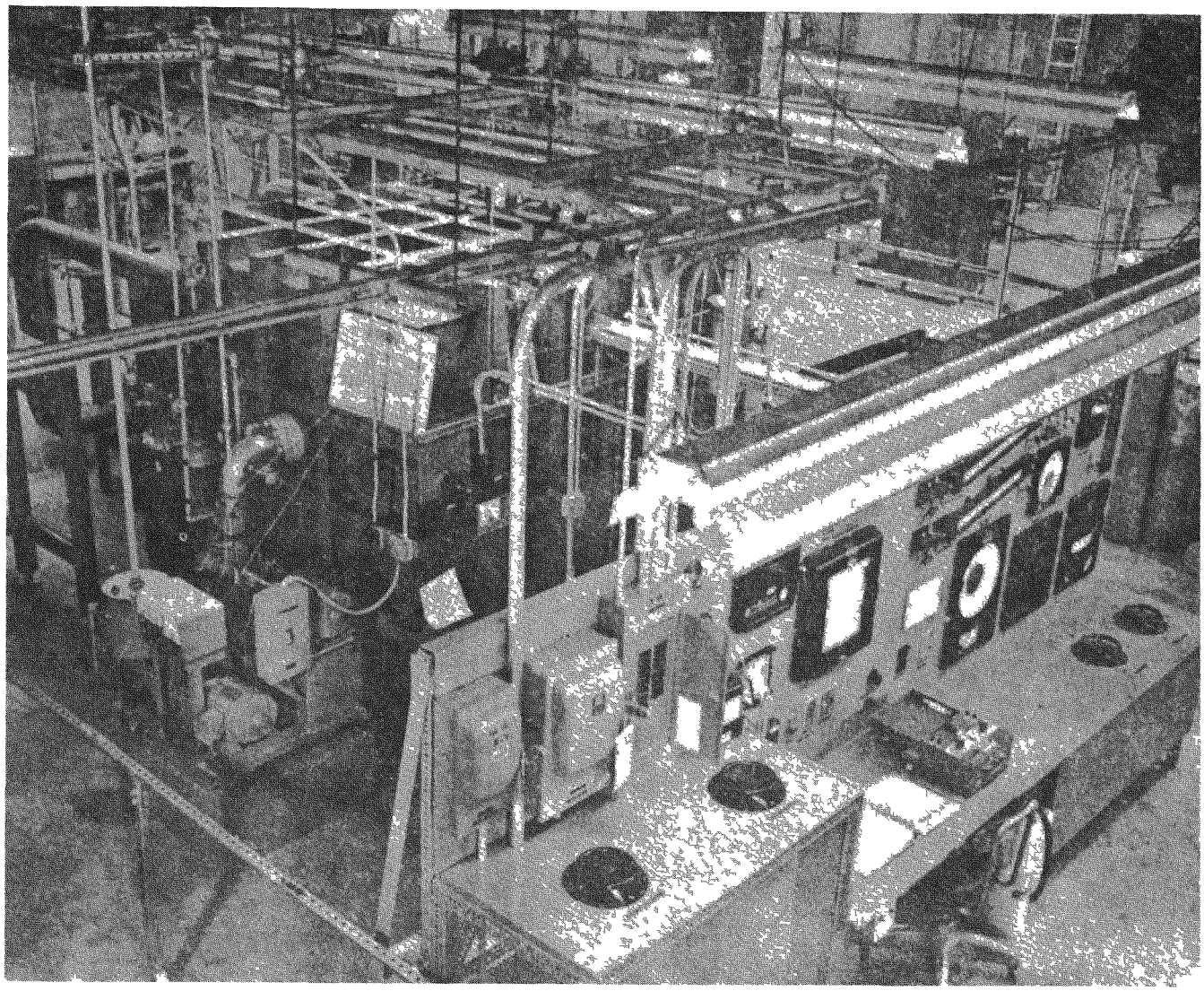


FIGURE B-3. LOOP TEST FACILITY

5. Corrosion Loop Design

a. Rubidium Loop System Breakdown

The rubidium loop (Figure B-4) was divided into seven subsections:

(1) Pump Section

This section was designed to pass through the electromagnetic pump which provides the desired flow and ΔP to rubidium. The size of this section (1/2 in. O.D. x 0.049 in. wall tubing) was dictated by pumping design factors. The electrodes on the electromagnetic pump were brazed into specially flattened sections of the pump cell by inert cover gas techniques using Nicrobraze type brazing alloys. The same procedure was used for both the columbium and stainless steel loops. Tests were conducted which indicated good joints could be achieved between stainless steel and columbium tube to the nickel bus bars. The pump cell was first brazed and then the bus bar tube assembly was welded into the loop system and the pump reassembled.

(2) Preheater Section

Immediately following the pump cell was the preheater section (0.250 in. O.D. x 0.049 in. wall tubing). The reduction in size was made necessary by the flow requirements which, if met in the larger tube size, would have resulted in a larger pumping system than was desired. The transition joint, a tapered section, was fabricated by taper machining of bar stock. Tabs were welded along the length of the preheater section at its beginning, middle, and end. Copper bus bars were clamped to these tabs and led to the main heater transformer outside the box.

(3) Boiler Section

The boiler section, connected to the preheater by a transition joint, was 1/2 in. O.D. x 0.049 in. wall tubing. The size increase was necessitated by heat transfer considerations. The

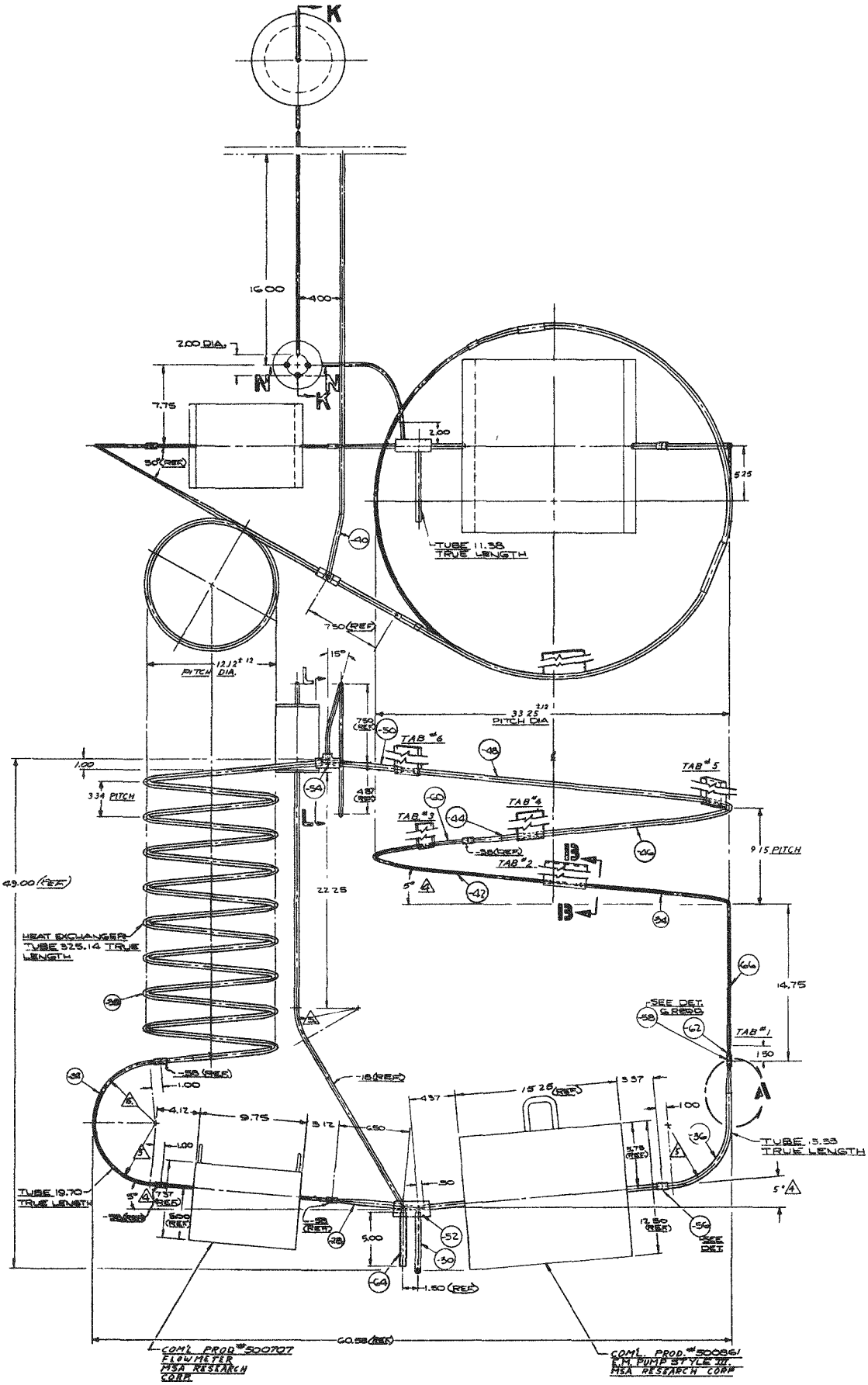


FIGURE B-4. DYNAMIC TEST LOOP FOR LIQUID RUBIDIUM

necessary heat for boiling the rubidium would have produced a higher than desired heat flux per unit area in the 1/4 in. O.D. tube used in the preheater. No flow swirl generators were used in the stainless steel or columbium loop boiler. The stainless steel loop achieved about 10 to 15% vapor quality in the boiler. If heat transfer limitations or unstable operation in the stainless steel loop had demonstrated the need for a flow swirl generator in the system, then it would have been added to the columbium loop. This was not found to be necessary. Other than the foregoing considerations, the boiler was essentially the same in design as the preheater.

(4) Condenser and Cooler

The outlet from the boiler passed directly into the condenser-cooler coil. A shroud and annulus was placed around the condenser-cooler and the heat was removed by a flow of argon gas over the coils.

(5) Flowmeter

The rubidium exited from the cooler and flowed through a transition section (from 1/2 in. O.D. to 1/4 in. O.D. tubing) and then through an electromagnetic flowmeter. The rubidium then passed into a 1/2 in. O.D. tube, flowed past the cold trap, pressurizing line, and fill line and then back into the electromagnetic pump to complete the circuit.

(6) Cold Trap

A 1/2 in. O.D. tube closed at one end was welded to the system at the lowest and coldest point. This cold trap was designed to catch and hold the insoluble impurities in the system, thus assisting in the prevention of loop plugging.

(7) Pressurizing System

A line, attached to the loop system at the lowest point, led up to the pressurized surge tank located alongside the point of the loop. This surge tank provided a reservoir of rubidium, acted as

an expansion chamber during boiling operation, and enabled pressure to be applied to the system to control boiling. The tank was cylindrical in shape with manually pre-set liquid level indicators of the spark plug type inserted from the top and hanging down into the surge tank. The rubidium surge tank, argon gas surge tank outside the chamber, and pressurizing lines were fabricated from 316 stainless steel in both the stainless steel and columbium loop systems, since these items operated at relatively low temperatures. A mechanical joint was provided between the stainless and columbium tubing at a point between the surge tank and the point where the pressurizing line entered the loop.

b. System Stress Calculations

Rough stress calculations were performed to determine the magnitude of stresses the system will be exposed to at maximum operating conditions.

(1) Tubing

Tubing wall and diameter dimensions were selected on the basis of heating method, expected corrosion effects, elevated temperature strength of the alloy, system pressures, heat transfer, and fabricability. Thus, a standard 0.049 in. wall was specified for the entire loop system. The nominal tubing diameter was 1/2 in. except for the heater section only, which was 1/4 in. O.D.

The most severe stresses in the 1/2 in. O.D. x 0.049 in. wall tubing at a T_{max} of 2000°F were roughly calculated as follows:

$$S = \frac{PD}{2t} = \frac{240 \text{ psi } (0.5 \text{ in.})}{2\ (0.049 \text{ in.})} = 1225 \text{ psi}$$

where S = tube wall loop stress in psi

P = system pressure

D = O.D. of tube

t = wall thickness

At 1600°F the stress would be

$$S = \frac{70 \text{ psi } (0.5 \text{ in.})}{2\ (0.049 \text{ in.})} = 357 \text{ psi}$$

Thus, the stress would be 1225 psi in the columbium loop under maximum operating conditions and 375 psi in the stainless steel loop under maximum conditions. These values were well within the operational capabilities of these materials.

(2) Surge Tank

The surge tank dimensions were selected on the basis of the rubidium inventory necessary to run the loop. The I.D. was about 4 in. and the height about 6 in. The columbium tank and stainless steel tank wall thickness were 0.100 in. The stress on the surge tank was as follows:

(a) Columbium loop maximum temperature - 2000°F with a surge tank maximum temperature of 1200°F.

$$S = \frac{PD}{2t} = \frac{240\ (4.2)}{2\ (0.1)} = 5040 \text{ psi}$$

(b) Stainless steel loop maximum temperature - 1600°F with a surge tank maximum temperature of 1000°F

$$S = \frac{PD}{2t} = \frac{70\ (4.2)}{2\ (0.1)} = 1470 \text{ psi}$$

These values were well within the operational capabilities of the columbium and stainless steel alloys at their respective operating temperatures.

6. Hazards Analysis

In any operation employing a basically reactive metal such as rubidium, certain hazards are present. Some hazards were eliminated entirely from the experimental area; in other cases the hazards were reduced to a point where it was considered that the operation was sufficiently safe to be carried out on an experimental basis.

The hazards fell into a number of categories:

1. Fire hazards resulting from exposure of liquid metal to air.
2. Hazard of exposure of liquid metal to operating personnel.
3. Hazard of exposure of liquid metal to water or materials having a high water content.
4. Explosion of a pressurized loop system which might endanger personnel and result in the release of quantities of liquid metal, causing a hazard under any of the first three categories.

These four hazard groups were carefully considered during the design, fabrication, and operation of these loop systems. The fire hazard problem was recognized as being of considerable importance if liquid metal were released into the air. Special liquid metal fire fighting equipment was available in the liquid metal laboratory for the suppression of liquid metal fires. These included Metal-X type extinguishers and other inert substances which smother a liquid metal fire by removing the air to allow the liquid metal to burn out. Neither the liquid metal used in this program (rubidium) nor its vapor is considered toxic; therefore, this problem was not important in the hazard analysis.

The exposure of personnel to liquid or vapor was considered to be of major importance; protective clothing, leather coveralls, and

approved liquid metal face shield and gloves were on hand in the liquid metal laboratory for emergency use. These were also used for routine operations where the slightest possibility of a hazardous condition existed. Mineral oil and eye wash solutions were provided. All personnel working with liquid metals were instructed in standard safety procedures for handling these materials.

The hazard of liquid metal contacting water or materials containing large amounts of water was eliminated insofar as possible. Rubidium, if it were allowed to run on the floor, might go down a normal floor drain and contact water in the drainage system. This problem was avoided by specifying that no floor drains be located in the Liquid Metals Laboratory. Rubidium might contact water if a normal building sprinkler system were to go into operation in case of a fire. It was specified that no sprinkler system be in the Liquid Metal Laboratory. There was a remote possibility that rubidium might contact water if the loop argon to water heat exchanger had developed a leak. The rubidium in the loop and the water in the heat exchanger would have had to leak at the same time and come together through the argon system. The chance of this occuring was so unlikely that no special precautions were taken. Even if this had occurred, the release of rubidium from the loop and/or the release of water from the heat exchanger into the argon stream would probably have been at such a low rate that any reaction would not have escaped from the environmental chamber system.

In a pressurized loop system the possibility always exists that a rupture could occur in the system, releasing rubidium vapor into the environmental chamber. No hazard was expected from contact of rubidium with air since an argon cover gas was used in the environmental system. Any leak which occurred would have been under the loop operating pressure; however, even if the loop had ruptured, the area inside the loop was small enough and the chamber large enough so that

the rupture would not have been of a violent nature since the argon pressurizing tank was relatively small. The amount of rubidium contained in the loop was small enough so that if it had ruptured one of the windows in the chamber, the resulting fire would not have been considered hazardous to personnel in the building. Only about 4 to 5 pounds of rubidium were used in each loop run. Passage of the rubidium throughout the environmental system was also not considered serious since it could only contact the materials which are inert or resistant to attack by rubidium at the temperatures employed.

7. Inert Atmosphere Purification

A duplex gas purification train was used to purify the argon gas circulating in the loop environmental chamber. The first stage of the drying system consisted of a tube 36 in. long and 8 in. in diameter filled with molecular sieve material at room temperature. This removed any water vapor from the argon gas. The second stage consisted of a high temperature oxygen and nitrogen gettering alloy of 50% titanium and 50% zirconium enclosed in an automatically controlled furnace (36 in. long and 8 in. in diameter) operating at about 1650°F. This combination of gettering materials and temperature provided one of the most efficient known methods of purifying the argon gas.

The full flow of argon through the main cooling system was not purified. Instead, a small bypass flow of 5 to 10 cfm was passed through the purification system. This purified gas was cycled back into the main cooling system. All gas in the loop system eventually passed through the purification train and was purified.

Efficiency studies of the purification train showed that it was able to reduce O_2 content of the 250 cu ft environmental chamber from 15 ppm to 0.5 ppm after a period of 120 minutes from startup with a flow of about 5 cfm through the drying train. The O_2 content of the chamber was held at between 0.5 and 5 ppm during the columbium loop run.

8. Liquid Metal Purification and Handling

No rubidium purification system (except a cold trap) was incorporated in the loop system since it was not thought necessary for these tests. This design philosophy required that the loop system be thoroughly cleaned before loading and that steps be taken to purify the rubidium metal before charging into the loop.

The as-received rubidium metal was approximately 99.9% pure. A three step purification process was settled upon. Step I involved passing the as-received rubidium through a filtering apparatus employing a micro-metallic filter to remove insoluble impurities from the rubidium. Hot reactive metal gettering of impurities at 1100°F from the rubidium was done as Step II. Step III involved vacuum distillation of the rubidium. Test results showed that O_2 contents of about 70 ppm and N_2 contents below limits of detection could be achieved. The purification apparatus is shown in Figure B-5.

The loop was loaded with rubidium inside the environmental chamber. Procedure called for evacuation of the chamber and back flush with argon. The loop meanwhile was being evacuated to about 10 μ and backflushed with argon. This step was repeated at least twice. The rubidium was then forced into the loop under argon gas pressure. These steps were controlled by remote solenoid operated vacuum, gas, and liquid metal valves. This system provides for emergency dumping of the rubidium back into the fill tank if desired.

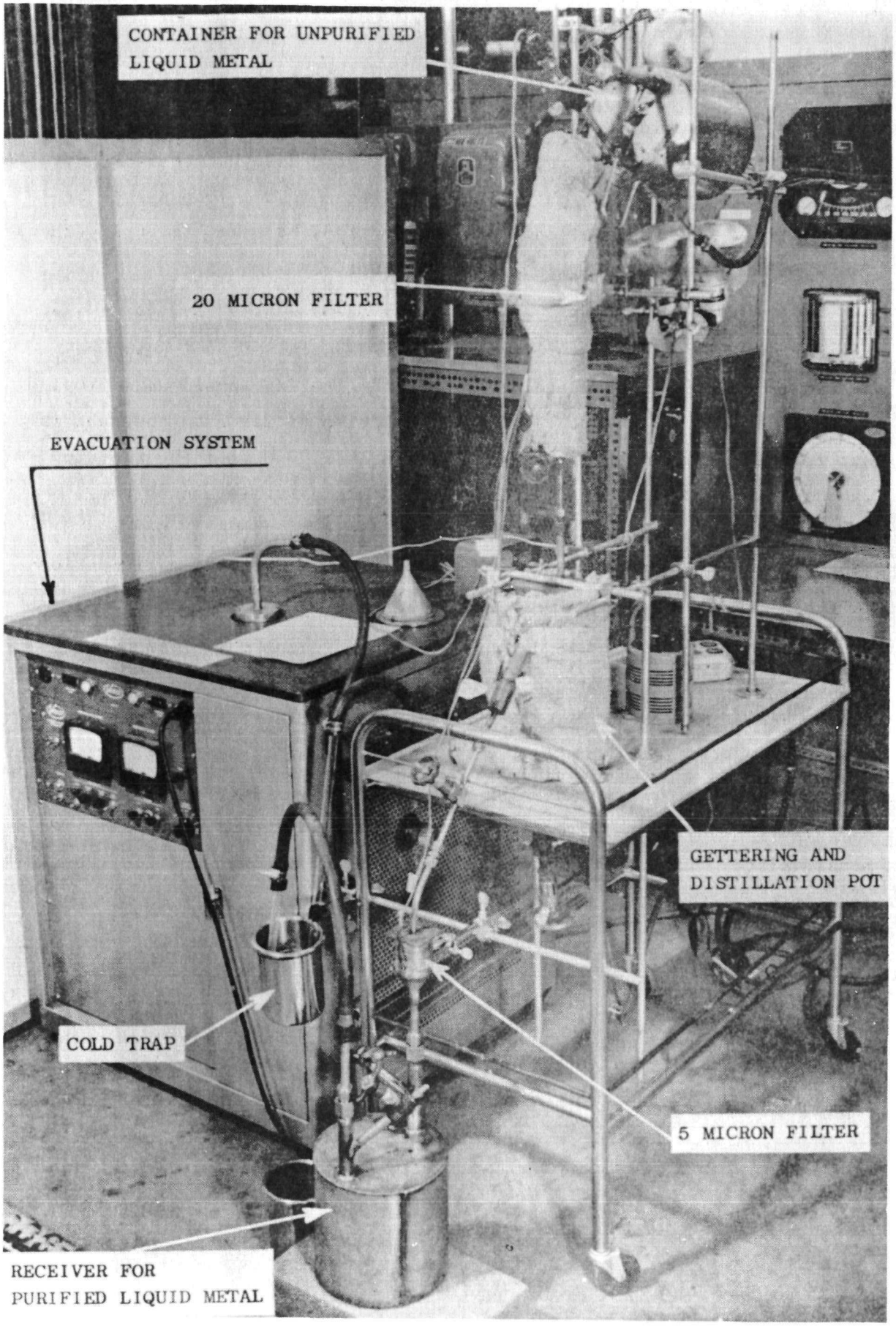


FIGURE B-5. LIQUID METAL PURIFICATION APPARATUS

9. Chemical Analysis of Materials

Chemical analyses studies were conducted and an analytical capability was set up at AGN. A purification still for repurifying the as-received rubidium was designed (see previous section). This enabled the project to obtain high purity rubidium for the loop and capsule operations and to repurify and reuse the spent rubidium after each test run. In evaluating the loop and capsule test results, the following chemical analyses were programmed.

a. Analysis of Cb-1% Zr Alloy

(1) Analysis for Base Metals and Rubidium

The analysis for base metals and rubidium was best accomplished by emission spectrography. A semi-quantitative analysis (± 50%) was specified for trace elements. In general, concentrations as low as 10 ppm were determined.

(2) Analysis for Interstitial Gases

Analysis for oxygen and hydrogen in columbium alloy were performed by a vacuum fusion technique. The analysis of nitrogen in columbium was done by the Kjeldahl method, and the final measurement of nitrogen for low concentrations was done by a colorimetric method using Nessler's reagent. The limits of detection were in the 1 to 10 ppm range. A sample of 1 to 5 grams was required.

b. Analysis of Rubidium Metal

A procedure for analyzing rubidium metal was established and included analyses for metallic impurities and gas content.

(1) Metallic Impurities Analysis

Since rubidium is highly reactive, the rubidium samples had to be carefully prepared under inert gas cover. Rubidium was reacted with one of the higher alcohols and then converted to a salt. The residue and the salt were then analyzed for trace elements by emission spectrography. A semiquantitative analysis was performed in a manner similar to that used for the columbium analysis above.

(2) Gas Content Analysis

The nitrogen analysis was also performed by the Kjeldahl method after the rubidium was first reacted with an alcohol and converted to a salt. An inert atmosphere was required for the initial sample preparations. The apparatus was the same as that used for the columbium work.

The oxygen content was determined by dissolving the rubidium in mercury. The oxygen which combines as rubidium oxide would float on the mercury, and rubidium was removed from the sample container with successive washings of mercury. The rubidium oxide was then dissolved in water and the base which formed was titrated with standard acid. The initial sample preparation was done in an inert atmosphere.

(3) Argon Purity Analysis

The analysis of the argon cover gas for total gas impurity content including oxygen, hydrogen, nitrogen, water and carbon dioxide appeared costly. With hot titanium and zirconium purification of the argon, analysis for oxygen only would probably be sufficient to indicate the general quality of the argon. A continuous type of analysis was the most desirable. The argon from the purification system was checked to indicate the quality of the purified argon. The chamber inlet and exit gas was also checked to indicate the degree of outgassing. When the columbium test loop was present, a check of only the exit gas was not sufficient since the columbium would also act as a purification system. A valving system to sample both the purified gas and the chamber inlet and exit gas was used in conjunction with one oxygen analyzer. The analyzer exit gas was not recycled since the oxygen analyzing system introduced large amounts of water into the argon. An oxygen analyzer, which included a built-in calibration system, was used to detect oxygen concentrations in the range 1 - 100 ppm.

10. Special Metallographic Procedures

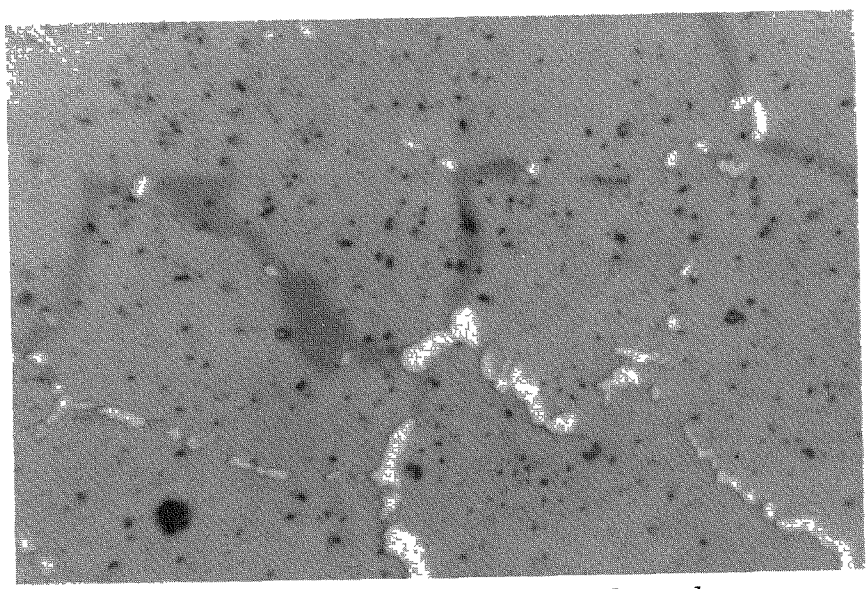
It was necessary to be able to analyze the corrosion products which occur in, and tend to degrade, the properties of containment materials in this system. These corrosion products are generally caused by a reaction of the system environment with the containment material and may be in the form of oxides, nitrides, carbides, hydrides, or other complex alloys of the environment specie with containment materials. The amount of corrosion products in the parent material is usually small.

Since the corrosion products are usually finely divided and mixed with the parent materials, analysis by conventional chemical or x-ray methods becomes extremely difficult, if not impossible.

Many research groups have experimented with heat tinting of steels and color etching of titanium alloys to determine the presence of various alloy phases in these materials. These experiments have been successful, but were not extended to include the identification of minor constituents in other materials. AGN has been experimenting with specialized metallographic preparation techniques which etch selectively and which allow visual identification of the various corrosion products in columbium metal. Results obtained indicate that these techniques, although still under development, are extremely useful and time-saving in analyzing materials corrosion data.

An example of this work is shown in Figure B-6. This figure is an early photograph of the results of a colorimetric etching technique performed on essentially pure columbium metal. The various corrosion products stand out clearly and are colored distinctively. This specific method is apparently accurate and reproducible. Identification is achieved by a color tinting process and involves specialized electrolytic techniques.

The specimen is mounted in conductive powder and polished by standard metallurgical techniques. After polishing, the specimen is



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1 1/2 Times in Reproduction

Figure B-6. Identification of Corrosion Products in Columbium Metal (Cb - Purple*, CbO - Blue†, and CbC - Yellow‡)

*Light gray background and splotches.
†Medium gray plus center of largest splotch.
‡White areas.

etched lightly to remove any smear material and to bring out the grain boundaries. The sample is then etched with a 10% oxalic acid solution by passing an electrical current of 20 VDC through the back of the sample mount for about 1/10 second. As shown in the figure, the columbium specimen turns purple. Metallographic examination of the specimen shows that if the following compounds are present, they will appear with the following colors:

CbC	Light Yellow
CbO	Light Blue
CbN	Greenish tint (?)
Cb	Purple

This technique was used in analyzing the columbium loop run.

11. Material Fabrication Development

The general loop configuration has been discussed in Section 5, Corrosion Loop Design. The specific materials and fabrication procedures are outlined here.

a. Stainless Steel Loop Procedures

The procedures, joint designs and techniques used on the stainless steel loop were essentially the same as used for the columbium loop, save for the specialized welding procedures outlined under "Columbium Loop Procedures" below. Welding was done by tungsten arc inert cover gas methods with a flow of cover gas being maintained outside and inside the joint during welding.

Joint designs considered were a sleeve joint with collar, without collar, and without collar but with center seal weld, and a butt joint. A simple butt type joint was rejected for this application since it is hard to set up properly in order to achieve maximum weld penetration. A sleeve joint with trial modifications was made to be the reference joint design. Of the three sleeve joint designs considered, the sleeve joint without a collar and without a center tube seal weld was chosen as being the least complicated and having the best penetration (75 to ±10%) and least weld porosity of the three designs. Tube bends were made by filling the tube with a low melting point alloy, bending the tube and then removing the bending alloy.

All welded joints were x-ray inspected for voids and penetration. The loop was pressure checked with helium and "sniffed" to find any joint weakness and to ascertain if any leaks were present.

b. Columbium Loop Procedures

The procedures for the columbium loop were essentially the same except for the welding techniques. Much greater care in cleaning and cover gas protection was used. The columbium material was welded either in the inert gas welding chamber (Figures B-7, 8) or by flush box bag techniques. The argon and helium cover gases used were

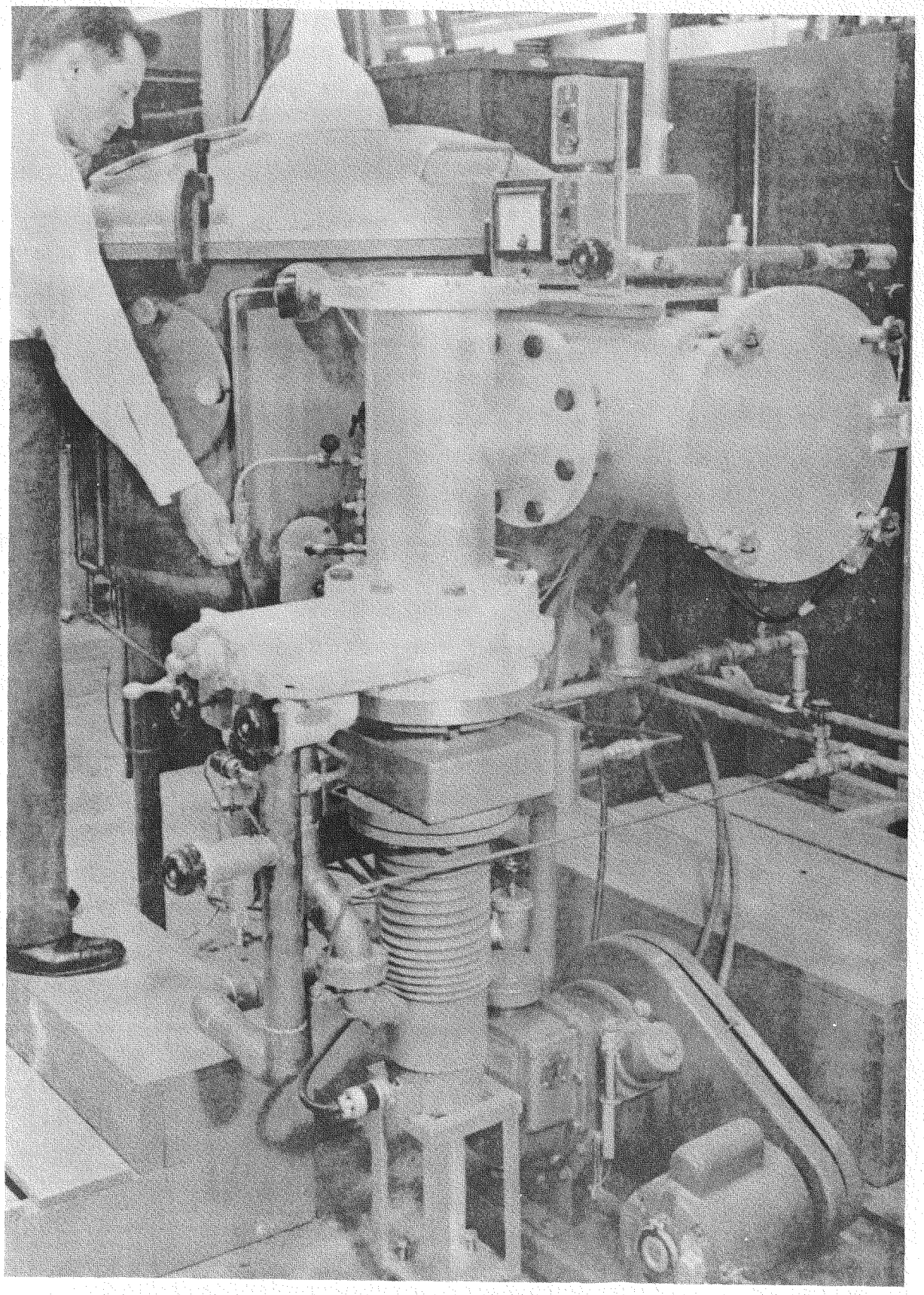


FIGURE B-7. CHAMBER FOR INERT GAS WELDING

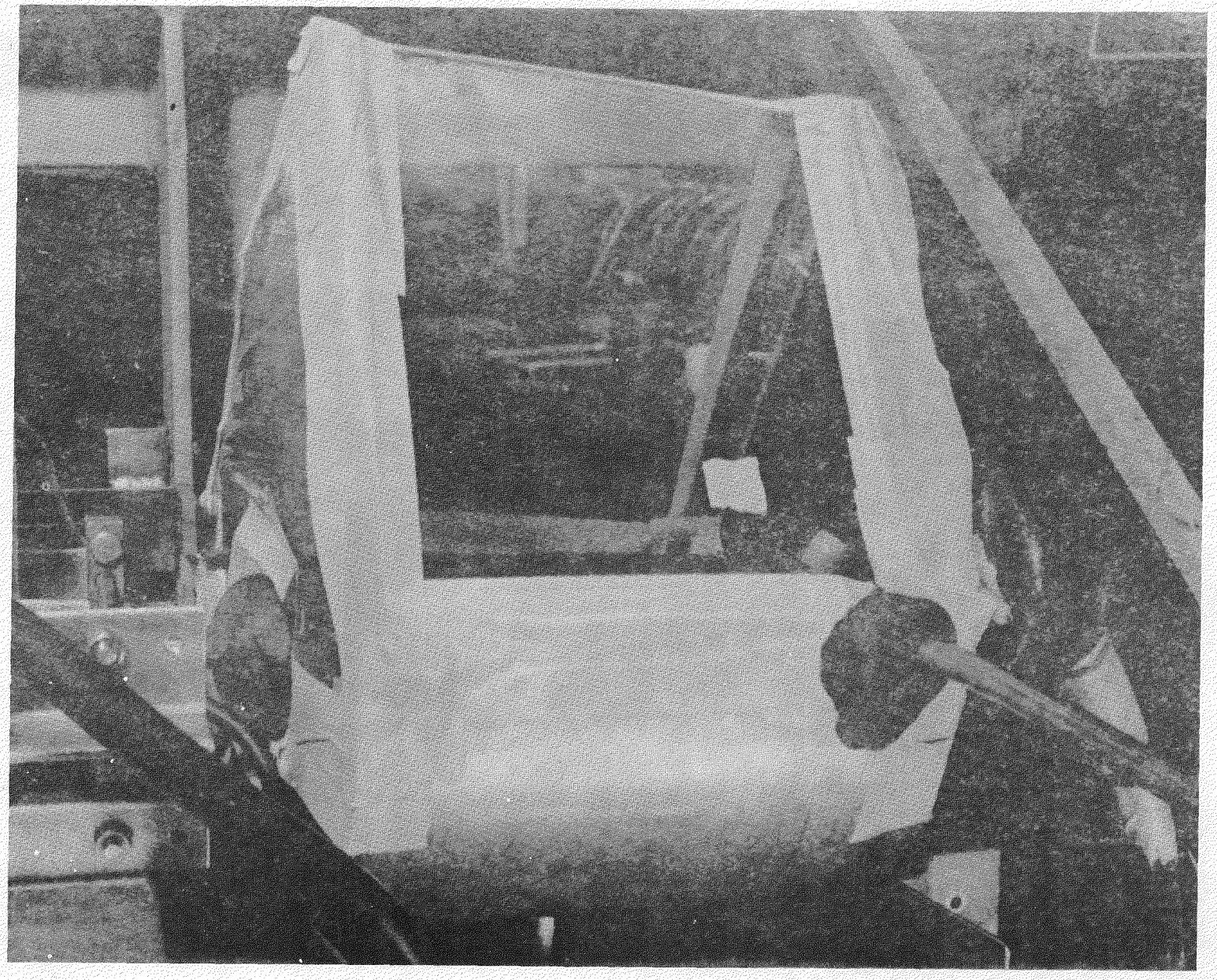


FIGURE B-8. FLUSH BOX FOR FINAL ASSEMBLY WELDING

the best commercially available (99.999% pure). No post weld annealing was done. The columbium tubing met AGC Specification 60064A, which was patterned after the use-tested PWAC Specifications SMS 1830A and 1831, especially with regard to maximum allowable impurity contents. Inspection of weld joints was the same as for the stainless steel loop.

After the columbium loop was fabricated and before thermocouples or insulation were attached, the loop was wrapped with a double layer of zirconium foil. This protective foil helped prevent air corrosion of the columbium should the environmental chamber atmosphere become contaminated.

c. Loop System Assembly

After assembly of the basic rubidium loop system, the loop was mounted on a portable support rack shown in Figures B-9 and B-10. The loop was suspended by a system of wires and counterweights so that it was subjected to a minimum of bending and twisting stresses. Thermocouples (chromel-alumel and platinum-platinum rhodium) were attached to the loop. These were applied over the protective foil wrap in the case of the columbium loop. Insulation was then applied over the loop. The two types considered were Foamsil and Thermoflex. The Foamsil has superior outgassing properties; however, it is not as easy to form into intricate shapes as the Thermoflex. Both outgassed Thermoflex and Foamsil were used on the stainless steel loop. Thermoflex was found to be unsatisfactory and was not used on the columbium loop run.

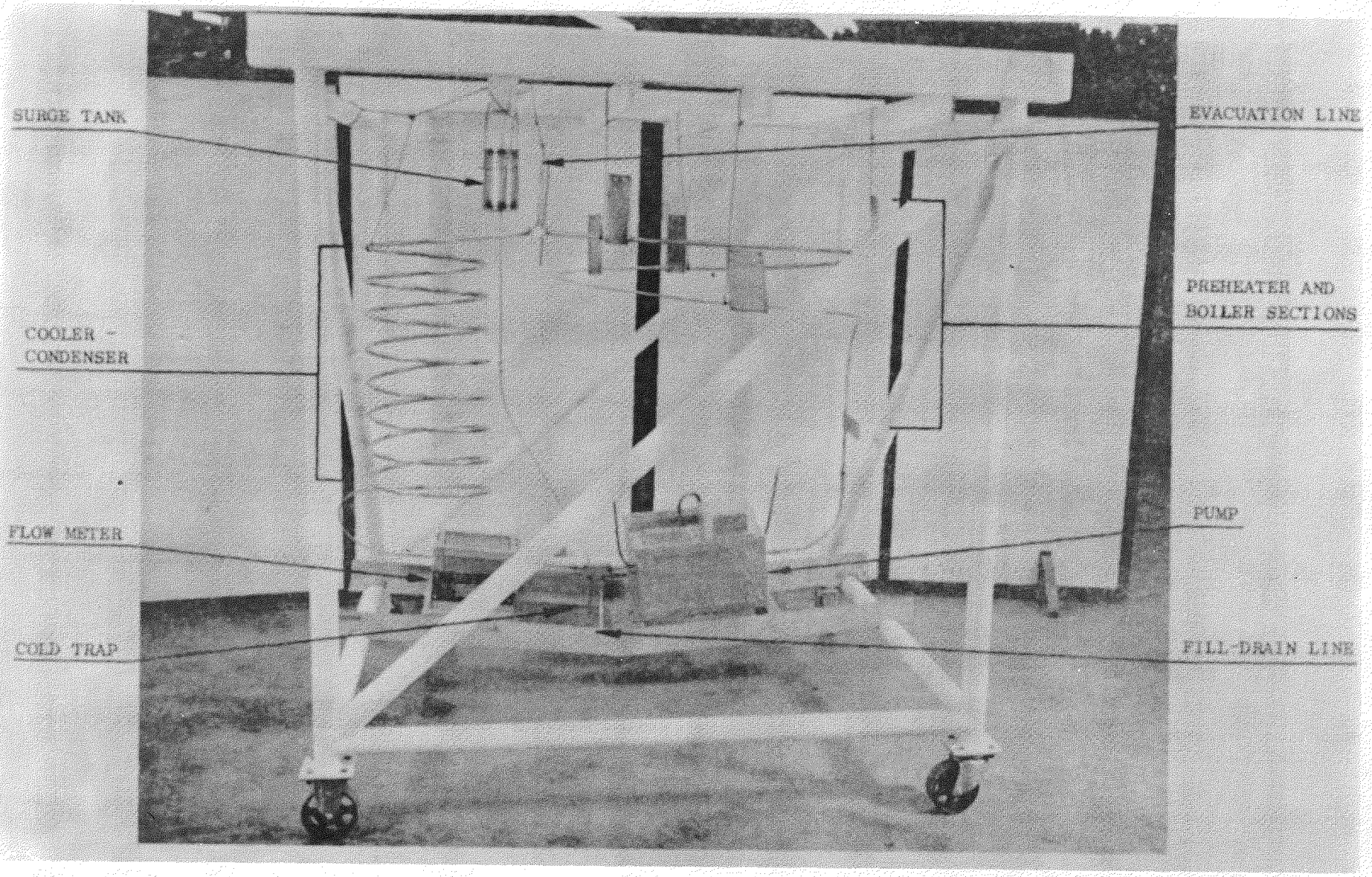


FIGURE B-9. RUBIDIUM - STAINLESS STEEL LOOP ASSEMBLED ON SUPPORT RACK

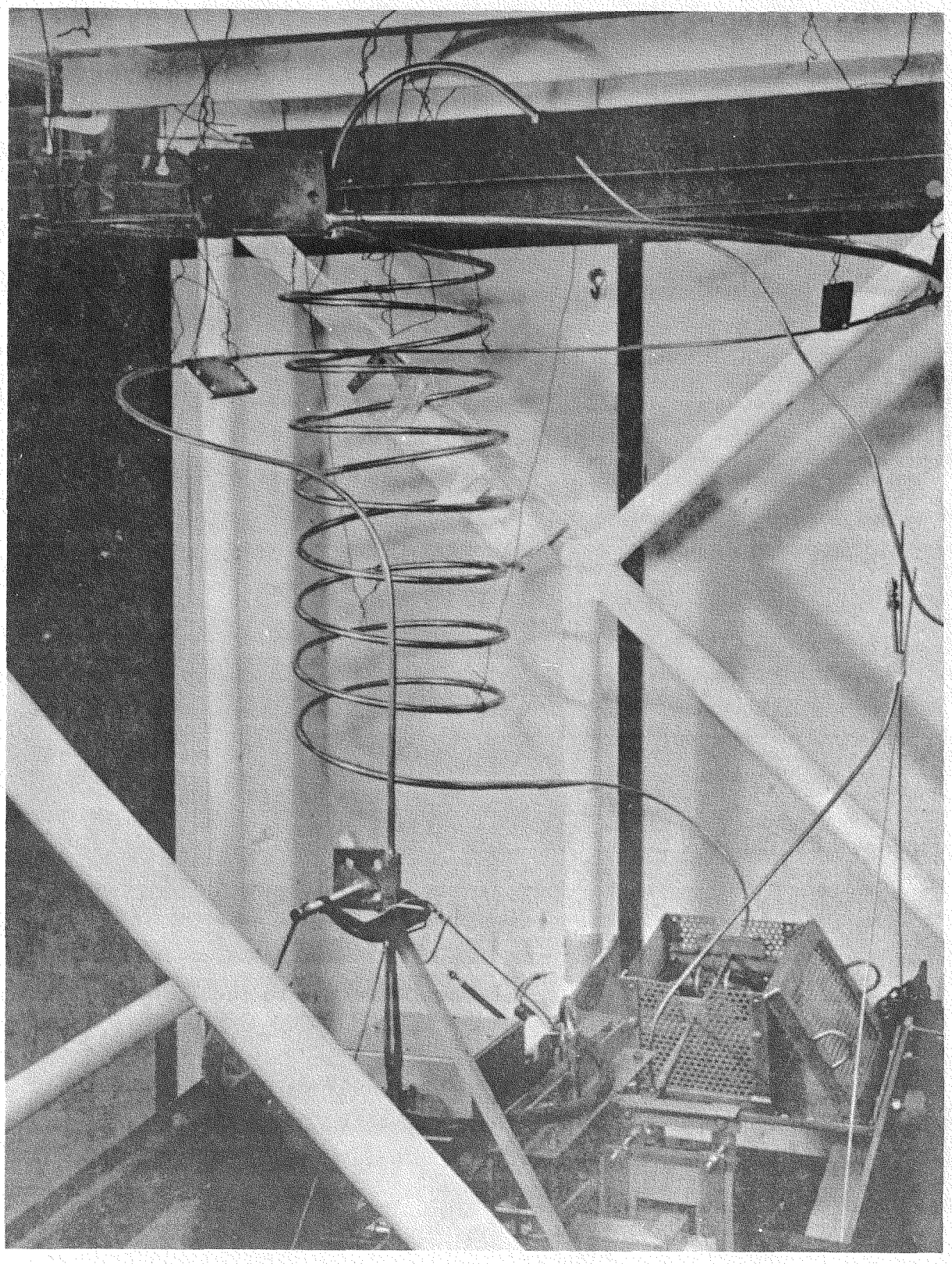


FIGURE B-10. RUBIDIUM-COLUMBIUM LOOP ASSEMBLED ON SUPPORT RACK

12. Loop Operations and Analysis

a. Stainless Steel Loop

(1) Loop Test Including Instrumentation and Controls, Facilities Preparation, and Loop Operation

The stainless steel loop was pressure and leak checked by helium mass spectrometer methods. Leakage rate was shown to be negligible. The associated loop pump and flowmeter, argon gas duct assembly, and flow controller were installed. The loop environmental chamber was found to have a larger number of air leaks than would be desirable for the columbium system test run. The system was run unchanged for the stainless steel loop, since these leaks would not affect this test. (After the stainless steel loop run was completed, the leaks in the environmental chamber were sealed to achieve the desired vacuum for the columbium loop run.)

The stainless steel loop was installed in the environmental chamber and the instrumentation and control systems hooked up and checked out. The rubidium loading system was checked out.

Loop start-up procedures were initiated on 11 January. The 316 stainless rubidium loop was evacuated for approximately 24 hours to purge it of atmospheric gases and to outgas any water present on the walls of the tubing. The vacuum chamber was pumped down and back flushed with argon gas. The loop itself was then back-filled with argon, evacuated again, back-filled a second time, and evacuated a third time. The chamber temperature was brought to 150°F and the valve opened from the rubidium fill tank leading to the loop. After the loop was filled with rubidium all valves to the loop were closed.

When the pressure on the surge tank was increased the liquid level fell in the surge tank, indicating there were gas bubbles trapped in the loop. Efforts were made to remove these trapped gas bubbles by alternately applying pressure and turning on the liquid metal pump and then releasing the pressure to move the gas

bubbles out of the loop and back into the surge tank. This procedure was continued until the level of rubidium in the surge tank did not fall appreciably upon application of pressure to the loop, thus indicating that most of the compressible gas contained in the loop system had been eliminated. The inclusion of gas in the rubidium during the loading process was due to a minor design error in the loop. This was remedied for the following loop run.

During the period when gas was being eliminated from the loop, the refilling procedure was repeated to add rubidium to the loop system. This made up for the volume of gas eliminated from the loop system. The rubidium flow through the preheater remained at less than 1 foot per second at the end of the day. At this point approximately 1 kw of heat had been applied to the preheater and the maximum loop temperature registered about 350°F. The low maximum flow rate obtainable from the loop at this point was due to the fact that the pump cell on the liquid metal pump had not wet, since the temperature in the pump cell was not above 150°F. The loop had achieved a state of continuous 24 hour operation and continued so until shut-down after the boiling test on January 20.

On January 12, certain aspects of the instrumentation and control system were adjusted and the maximum loop temperature attained was approximately 500°F at the exit from the preheater. On January 13, the last of the gas bubbles was eliminated from the system. The power in the loop was then increased to raise the maximum temperature in the pump cell, thus wetting the cell to obtain proper operation of the pump. To wet the pump cell, the argon flow through the rubidium cooler was cut off to bring the loop into essentially isothermal operation. The pump cell wet at approximately 350°F and flow increased to approximately 4 feet per second, with a maximum loop temperature of about 750° and a power input of 3.12 kw. The maximum temperature of the system then fell due to the increased circulation of the rubidium; a flow rate of approximately 4 ft per second at a maximum loop temperature of approximately 350°F was maintained overnight.

On January 14 the flow was increased to approximately 10 feet per second and power to 3.1 kw, resulting in maximum steady state operating temperature of about 550°F.

Steady state operating conditions continued over the 15th of January. Some difficulty was experienced with the pressure control system and an additional control unit was attached to the argon pressure control valve. The interior of the environmental chamber was overheating due to hot argon gas leaking out of the argon to water heat exchanger. More sealing and insulation was applied to the condenser can, but the resultant lowering of the chamber temperature was not as great as desired. The flow of rubidium in the loop system was increased to about 12 feet per second and the power to about 3.6 kw; this flow and power level was maintained overnight with a maximum temperature of approximately 500°F. The first specific heat determination was performed on January 17. The data resulting from this are reported in Appendix C. The maximum temperature obtained from the loop was about 850°F at a power input of 3 kw and a flow of about 13 feet per second. This is less power than was previously required to hold the loop at a lower maximum temperature. It is not known exactly what caused this effect; it may have had to do with wetting conditions in the loop.

On January 18 a final check showed that little or no gas remained in the loop system. Specific heat measurements were performed on January 19. Essentially the same loop operating conditions were maintained as those of January 17 with regard to maximum temperature and flow rate. On January 19 further specific heat measurements were made (reported in Appendix C of this report). The maximum loop temperature obtained was approximately 1150°F at a flow of about 6 feet per second in the preheater and a power input of about 9 kw. After these tests the maximum temperature of the loop was lowered to approximately 1000°F; the loop was run overnight at that value. Flow rate at this point was approximately 8 feet per second with a power input to the boiler of approximately 10 kw.

On January 20 a successful attempt was made to boil the rubidium in the loop system. A third specific heat measurement was taken prior to the test (data reported in Appendix C of this report). Due to the leaks in the protective can covering the argon to rubidium heat exchanger, and the subsequent increase in chamber temperature above 200°F, the boiling test was limited to a short run to determine whether stable boiling could be achieved in this loop to about 15% vapor quality. The flow rate was therefore set at approximately 10 feet per second and the loop pressure at 33 pounds per square inch absolute. At this point the total power to the loop was approximately 13 kw (11.3 kw net to the rubidium); the maximum temperature was approximately 1325°F uncorrected, and flow rate was approximately 10 feet per second when boiling was initiated. A quality of 7.2% was achieved. The power to the boiler was increased to approximately 15 kw at which point the vapor quality was about 13% (13.3 kw net to the rubidium). It was determined that boiling had been achieved since the increase of power input from 11 to 13 kw produced no increase in boiler outlet temperature with flow rate remaining constant. The additional power merely increased the vapor quality of the rubidium. A second check was made to determine if boiling was actually taking place; the pressure on the loop was increased in small increments and the resultant increase in boiler outlet temperature was noted. This check indicated that the pressure increase was causing the rubidium to boil at a higher temperature. This procedure was repeated several times to check the validity of the test. The pressure and power were gradually increased in increments until the last test of the day was achieved at a maximum measured temperature of 1450°F with a flow rate of 10 feet per second; a pressure of 65 pounds per square inch absolute; a total power input of approximately 17.0 kilowatts (15.2 kilowatts net to the rubidium), and a resulting vapor quality of 15%. Boiling was stable throughout the entire boiling test. This initial boiling test was satisfactory in most of the significant respects. No attempts were made to increase the temperature beyond the 1450° measured temperature since the stainless steel material is in the region of maximum capabilities at this temperature.

After termination of the test, the loop was shut down and the rubidium dumped back into the fill tank. The pump and heaters were turned off, and the loop was kept at a maximum temperature of 150°F until it was disconnected and removed from the environmental chamber at the end of the month.

(2) Heat Transfer and Specific Heat Analysis

The heat balance and vapor quality data obtained on this loop run are shown on Table II.

The specific heat data are discussed in the thermodynamic studies section.

Table II which lists three of the boiling conditions, shows that the measured temperature of the loop varies from 75° to 125° below the published boiling point of rubidium at the two pressures investigated. This discrepancy probably occurred because the thermocouples attached to the loop were not measuring the actual rubidium temperature, and were measuring instead the temperature at the surface of the tube to which they were attached. An analysis of the published data, system operating conditions, and thermocouple readout data, indicated that the actual temperature of the rubidium inside the loop was approximately 100°F higher than that actually measured. Therefore, the maximum temperature of the loop system was probably somewhere around 1550°F. In the next loop run steps were taken to obtain more accurate temperature measurement data in the system by use of more refined thermocouple readout techniques. It was estimated that the error in temperature measurement could be reduced to approximately 50°F by the procedures to be used.

(3) Corrosion Analysis

Only a cursory examination was made of the corrosive effects of rubidium in the stainless steel loop. Since the intended purpose was not as a corrosion run. The test was also of such short duration that no meaningful results could be obtained.

TABLE II

STAINLESS STEEL LOOP HEAT BALANCE CALCULATIONS

	System Parameters	Condition 1	Condition 2	Condition 3
1.	Rubidium flow rate, lb/hr	388	388	388
2.	Measured gross power input, kw	13.0	15.0	17.0
3.	Measured preheater power, kw	7.5	7.0	6.9
4.	Measured boiler power, kw	5.5	8.0	10.1
5.	Estimated losses from preheater/boiler, kw	0.9/0.8	0.9/0.8	0.9/0.9
6.	Net Power to Rb, kw	11.3	13.3	15.2
7.	Rubidium pressure, psia	33	33	65
8.	Published boiling point, ^{o}F (WADC TR 59-598	1410	1410	1550
9.	Measured boiling point (outside tubing), ^{o}F	1325	1325	1450
10.	Measured temperature at preheater inlet, ^{o}F	470	510	530
11.	Measured temperature at preheater outlet, or boiler inlet, ^{o}F	1090	1125	1124
12.	Rb quality	7.2±000.7	13.2±001.3	15.3±001.5

A sample of the original tubing and of the 1/2 in. O.D. tube at the boiler outlet in the hottest portion of the loop is shown in Figure B-11. Note that the corrosion of the 316 SS by the rubidium was very small. No definite evidence of mass transfer was found.

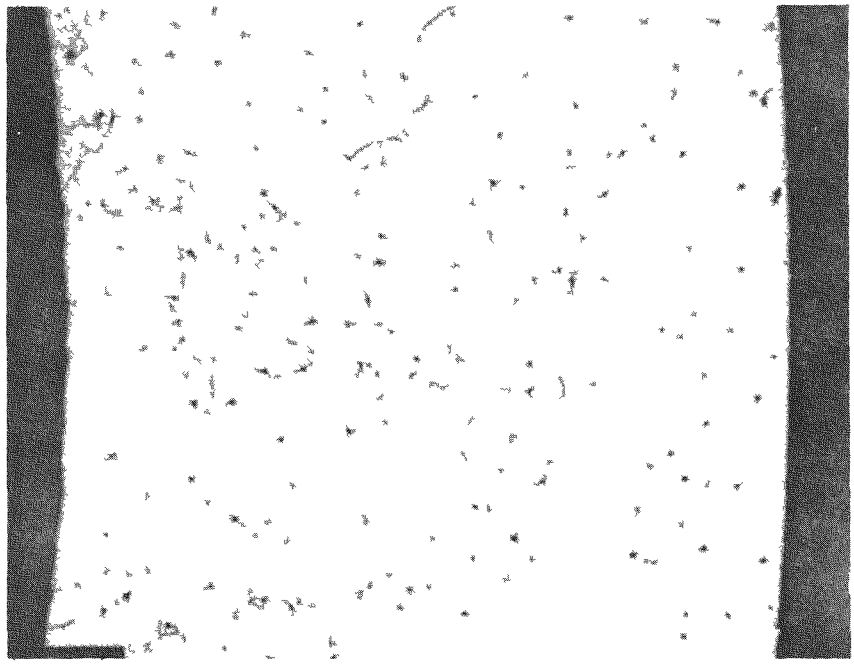
(4) Chemical Analysis

The rubidium used in the stainless steel loop was analyzed before-hand and found to have 114 ppm O_2 and N_2 below 10 ppm (below limits of detection). Complete analysis is as follows:

Cs	- 4700 ppm	Fe	- 70
K	- < 6000	Ni	- 30
Li	- < 300	Cr	- --
Na	- < 100	Cu	- 3
Mg	- 8	Si	- < 7
Al	- 40	Su	- 60
Ba	- 6	Pb	- 100
Ca	- 6	BE	- 30
N_2	- (not detected)	O_2	- 114 ppm

Sample taken after the loop run was lost in process of sampling.

As-Received 316 Stainless Steel Tubing, 1/2 in. O.D., Etched, Photo # 586, Mount G-8-9, Magnification 75 X



Post-Test Stainless Steel Tubing from First Turn of Condenser Coil, 1/2 in. O.D., Etched, Photo # 171, Mount Q-0-8, Magnification 75 X

FIGURE B-11

b. Columbium Loop

(1) Loop Test Including Instrumentation and Controls, Facilities, Preparation, and Loop Operation

The columbium loop was pressure and leak checked using mass spectrometer methods. Leaks were negligible and the final assembly of the loop was completed. The instrumentation and controls system was installed and checked out. The loop assembly was installed in the environmental chamber. Final connections were made and pump down operations started.

The environmental chamber was pumped down to 7 microns vacuum. At this vacuum the leak rate in the chamber was approximately 1600 micron liters per second. At the same time the second vacuum system was attached to the rubidium loop itself and the loop was pumped down. Pressure inside the loop was approximately 30 microns. Monday, 10 April the chamber was purged with argon and the oxygen content stabilized at approximately 16 ppm.

After the loop was loaded with rubidium, the next 17 days of operation included a number of adjustments, calibrations, and subordinate tests to assure a relatively uninterrupted corrosion test run. The drying train was checked to determine the efficiency of contamination removal. A baffle was inserted into the argon blower system to prevent overloading of the blower. The control thermocouple of the drying train was moved from the center of the gas stream to the outside wall of the furnace. Temperature drop from the outside wall to the center of the gas stream was as much as 1000°F with a maximum temperature of approximately 1500°F. Considerable difficulty was experienced in attempting to obtain a rubidium flow rate reading. Apparently the temperature of the liquid metal flowmeter was too low to allow the rubidium to wet the columbium flowmeter cell. An auxiliary heating element had been inserted into the liquid metal pump; however, none had been inserted into the flowmeter. A continuity check showed the flowmeter to be properly hooked up. At a temperature of about 400°, the flowmeter

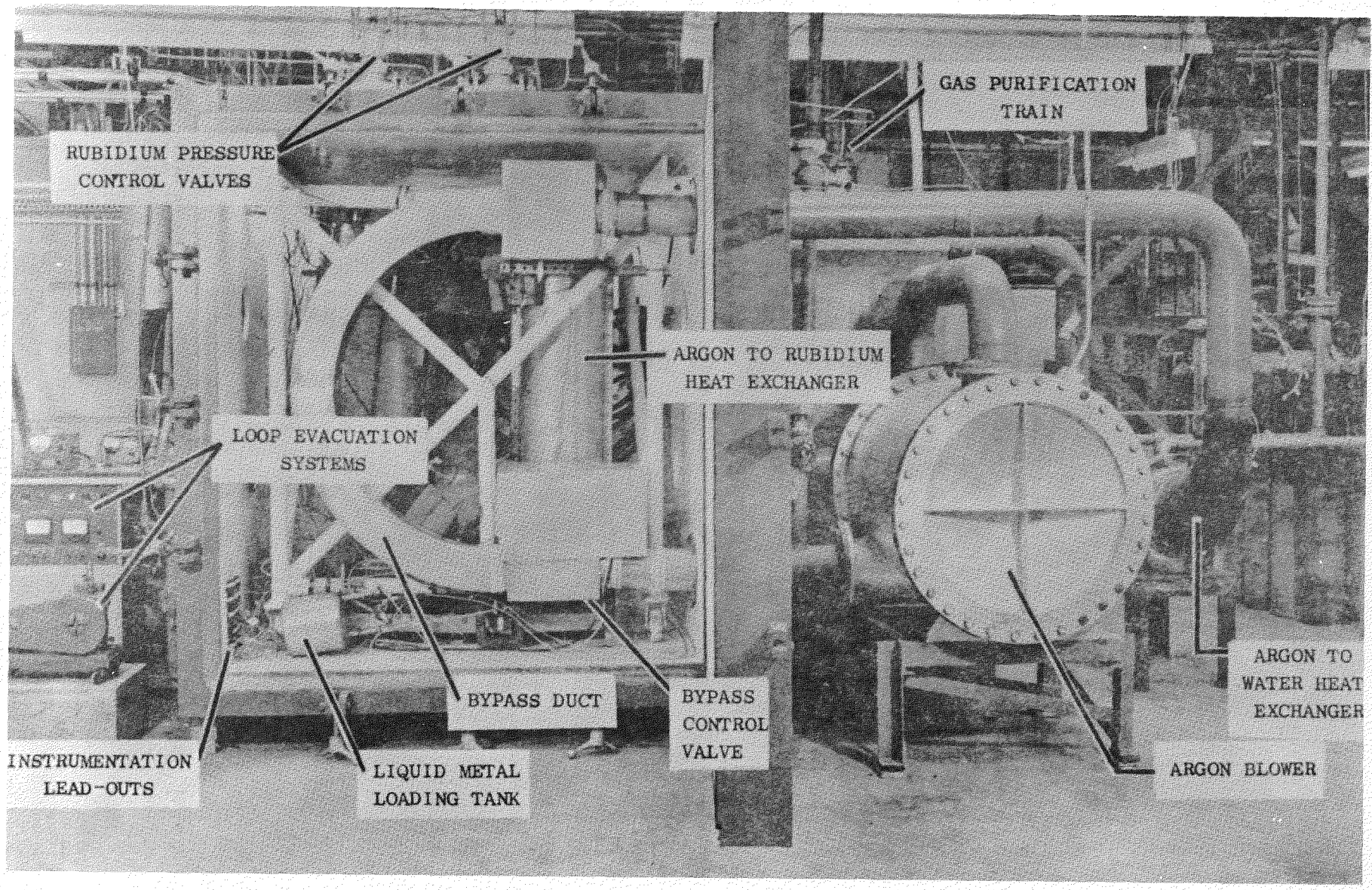


FIGURE B-12. RUBIDIUM COLUMBIUM LOOP INSTALLED IN ENVIRONMENTAL CHAMBER

began to register a flow in the loop.

It was noticed that the heating transformers were bucking each other at this point. The loop system was shut down and the transformers were rehooked in proper polarity and the loop was restarted. A specific heat determination was made and this data is reported on the section on thermodynamic and physical properties studies. A boiling test was then made at 1200°F (measured temperature). Stable boiling was achieved by setting the pressure at which corresponding temperature of boiling was desired. The preheater and boiler power were then increased gradually until surging in the flowmeter and raising of the level in the surge tank indicated the start of formation of vapor in the boiler. The power was increased further until the liquid vapor interface stabilized in the boiler and condenser and the desired vapor quality was obtained. Boiling was stable. Some difficulty was found in obtaining the correct level of rubidium in the surge tank and additional rubidium was loaded into the loop. A scheduled plant power outage forced shutdown of the loop at this time for approximately four hours. The loop was restarted, operating at a measured temperature of 1325°F boiling. Some problems were experienced with shutdown of loop power due to action of the low level limiter in the rubidium surge tank. It was discovered during this shutdown that additional rubidium was required for the loop. This leakage may have been caused by the pressure to hold the loading valve line closed being insufficient; some of the rubidium may have drained out of the loop back into the fill tank. This situation was corrected, the rubidium reloaded, and the loop restarted.

On April 24 the boiling corrosion run was started. Maximum measured loop temperature was 1735°F; however, pressure readings plus calculated temperature drop from the rubidium to thermocouples mounted on the outside tube wall indicated that temperature in the rubidium was of the order of 1840°F. The corrosion run continued until a momentary power outage shut the loop down, the high temperature

limiter malfunctioned, and the loop would not turn on again after regular power came back on. The malfunctioning high temperature limiter was removed and the corrosion run started again on the 28th of April. The corrosion loop run then continued normally for 355 hours until the 12th of May, when a vapor leak was discovered on the juncture of the boiler outlet and the condenser inlet. The loop was shut down and the leaking joint inspected. A decision was made to terminate loop operation and proceed with the metallurgical analysis, since it was not known at the time whether a successful repair weld could be made on material that had been exposed to rubidium.

(2) Heat Transfer and Specific Heat Analysis

The heat balance and heat transfer calculations on the columbium loop run are discussed below. The specific heat determination is discussed in the Thermodynamic Studies Section.

Liquid and vapor phase rubidium was circulated in the columbium tubing loop for 330 hours during April and May 1961. The objectives of the experiment were primarily of metallurgical nature; sufficient instrumentation was incorporated, however, to facilitate the establishment of rough heat balances and heat losses in the various parts of the loop. Calculations were carried out to find the over-all heat transfer coefficient in the rubidium condenser and to match the condenser design more closely to future loop requirements.

The loop operated approximately 330 hours at the following thermal equilibrium conditions:

Measured preheater inlet temperature	520°F
Measured preheater outlet temperature	1500°F
Measured boiler temperature	1790°F
Measured condenser inlet temperature	1770°F
Measured condenser outlet temperature	550°F
Measured rubidium flow rate	0.26 gpm at 520°F
Estimated rubidium flow rate	0.50 gpm

Estimated argon flow rate	1/2 capacity, i.e. 2550 lb/hr
Rubidium operating pressure	175 psia
Measured power to preheater	6.50 kw
Measured power to boiler	8.75 kw
Estimated losses from preheater	~2.00 kw
Estimated losses from boiler	4.00 kw
Approximate rubidium vapor quality	0.09 (9%)
Vapor in boiler by volume	92%

The above power and temperature data were for the heat balances carried out in various parts of the loop. The over-all heat transfer coefficient in the rubidium condenser was approximately 11.5 Btu/hr-ft^2-^{o}F, which is some 45% lower than previously estimated. This is probably due to insufficient knowledge of the temperature and vapor-liquid distribution in the condenser. The condenser heat balance showed furthermore that the rubidium flow rate should have read 0.5 gpm instead of 0.26 gpm on the flow meter to confirm the power and temperature measurements. Accuracy of the flow meter calibration is therefore doubtful since errors in all other measurements are of lower order.

The temperatures in the cold leg of the rubidium loop were consistently about 300° to 400°F lower than desired. Elimination of two condenser turns would reduce cooling and bring temperatures up to the desired levels.

(3) Corrosion Analysis

After loop decontamination, the insulation was removed. Figures B-13, 14 and 15 show various views of the failed joints. Note the blackening and discoloration of the foamsil, probably caused by impingement of rubidium vapor. The "T" section was cut out for analysis. X-ray techniques were insufficiently accurate to determine the location of the crack, and dye penetrant checking yielded no further information. The failed "T" was sectioned and inspected metallographically. The location of the cracks and metallurgical analysis is discussed in

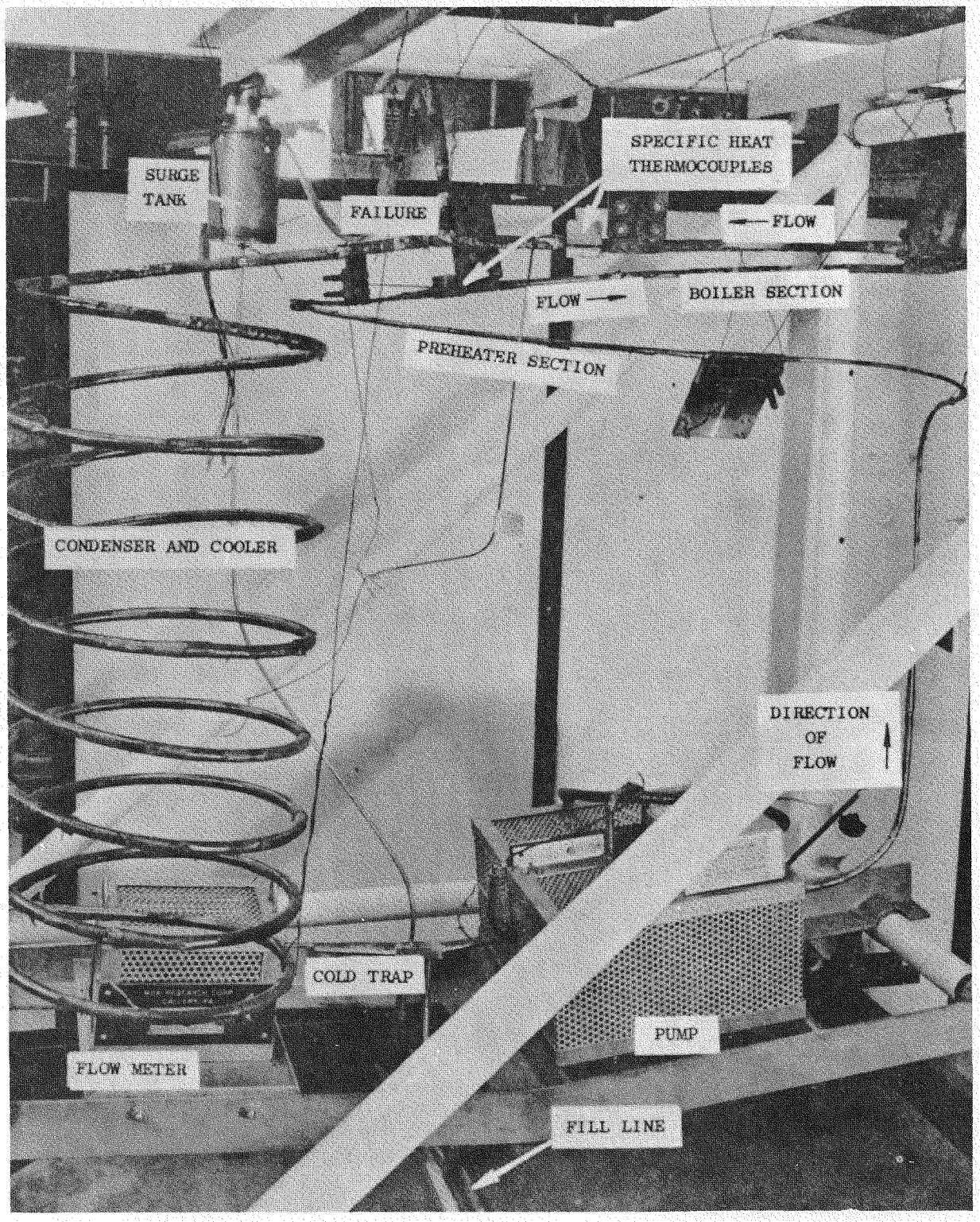


FIGURE B-13. COLUMBIUM LOOP POST-TEST AFTER REMOVAL OF INSULATION

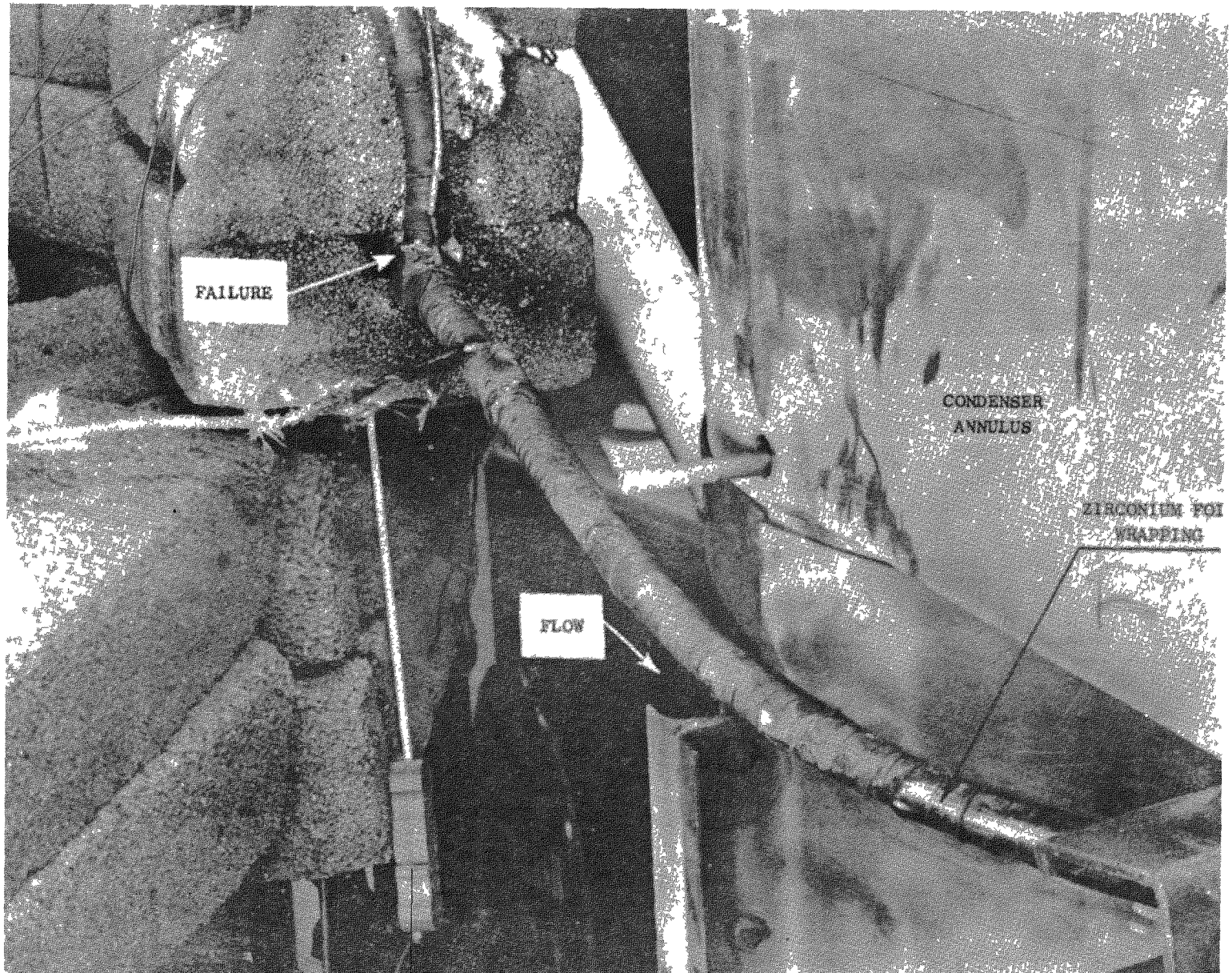


FIGURE B-14. CLOSEUP OF FAILURE AREA VIEWED FROM CONDENSER INLET SIDE

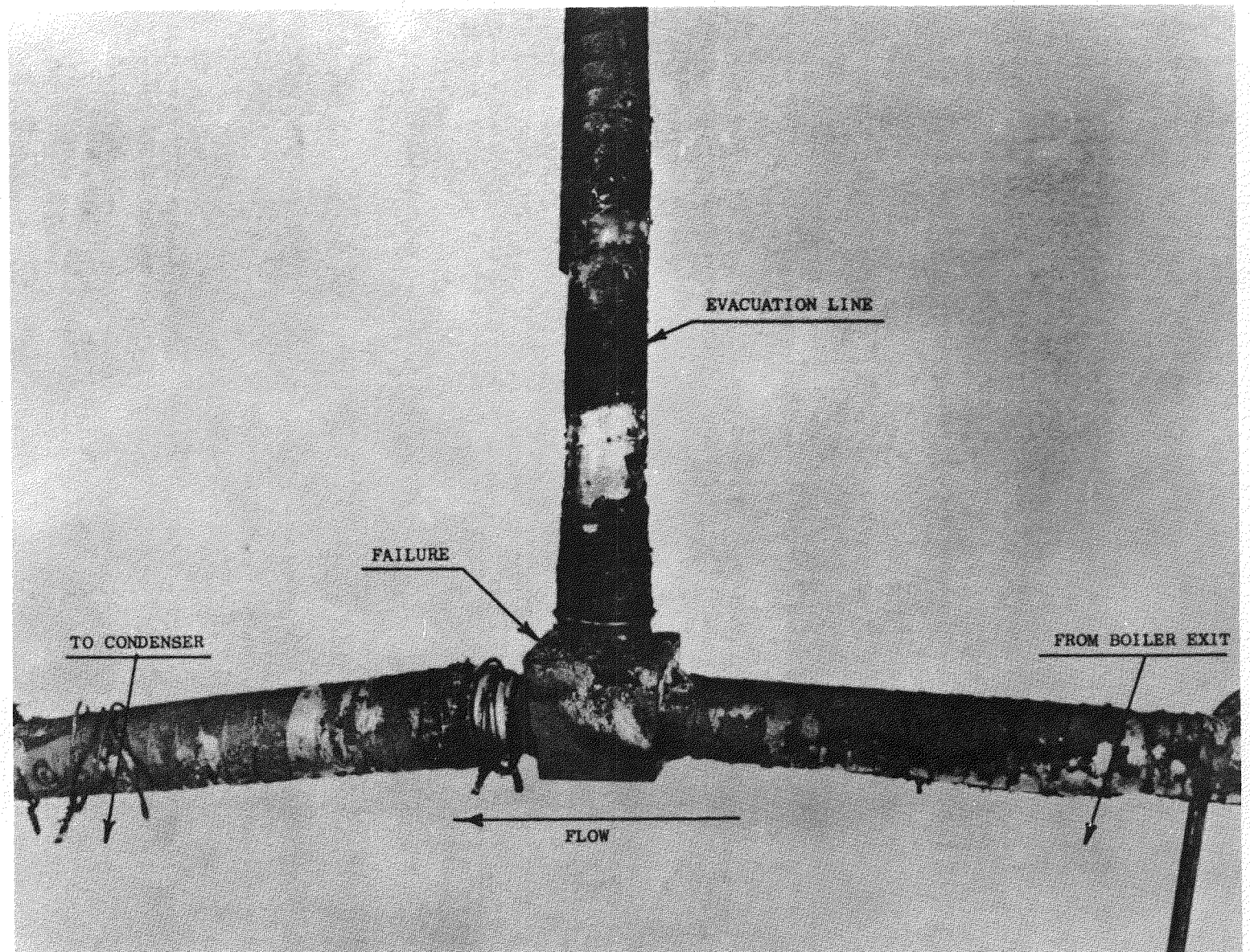


FIGURE B-15. CLOSEUP OF FAILURE AREA

a later portion of this section. Over-all examination of the loop revealed a number of interesting points.

In general, there was a large amount of crud encrusted on the zirconium foil wrap employed to protect the columbium tubing from attack from atmospheric gases. Results of the crud analysis are reported in the chemistry section. Source of the crud may have been due to:

i. Oil in the system coming from the lubricated bearings of the argon blower.
ii. Some contamination such as CO or CO_2 gas evolving from the Foamsil insulation.
iii. Contamination from organic materials used as insulation inside the vacuum chamber.

Analysis of this black crud did not reveal its origin. Steps will be taken in future loop runs to replace the heating cables, motor lubrication materials, and other possible sources of contamination by using non-outgassing materials.

The zirconium foil wrapped around the columbium tubing to protect it from atmospheric gas attack was severely embrittled in the first layer, which was exposed directly to the chamber atmosphere (Figure B-16). The underlined layer was in general relatively ductile and not attacked by the atmospheric gases. This foil was analyzed for the oxygen content of the various layers to determine what the impurity content gradient is from the surface of the foil to the surface of the columbium tubing. Note the blackened area, which is representative of spots around the loop where the zirconium foil failed and the columbium tubing was exposed to the chamber atmosphere. Note next to this blackened area the bright columbium tubing which was under the foil which had not failed. This indicates that the zirconium foil wrap is essential to prevent destructive attack by atmospheric gases on the columbium metal. Note also that the zirconium foil apparently diffused into the columbium alloy and adhered to it at certain points on the loop,

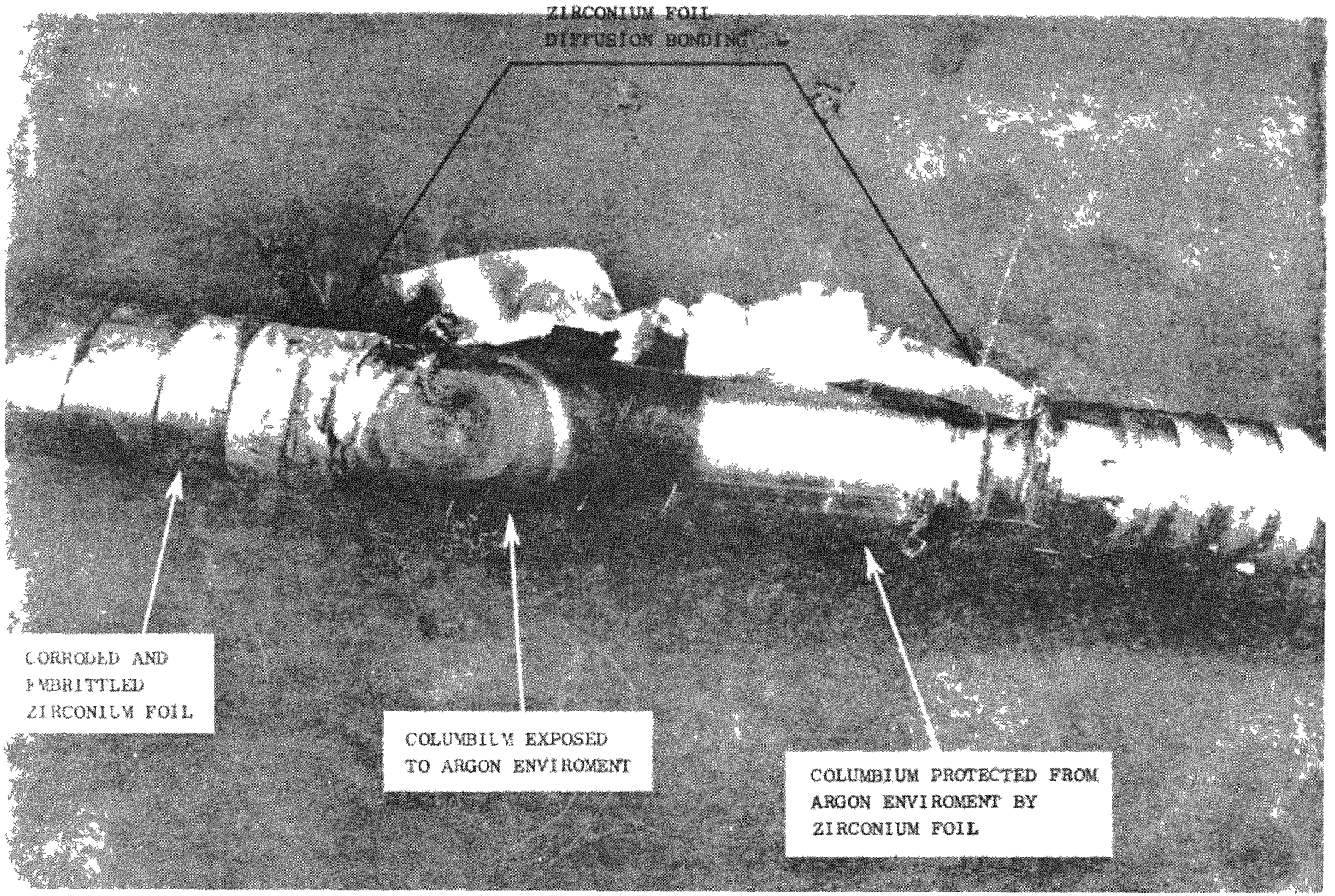


FIGURE B-16. CLOSEUP OF CONDENSER TUBE NEAR CONDENSER INLET SHOWING EFFECT OF ATMOSPHERE ON EXPOSED COLUMBIUM AND WELDING OF FOIL WRAP TO COLUMBIUM TUBE

especially in the hot sections. It is possible that for longer time and higher temperature operation, tantalum foil will have to be utilized as a substitute in an effort to avoid this problem.

Upon removal of the insulation from the loop several breaks occurred in the columbium tubing in the relatively cold section of the loop. One of these breaks can be seen in Figure B-17. One break occurred at the juncture of the surge tank line and the bottom portion of the loop and the second occurred at the start of the transition section at the exit from the flowmeter. When the half-inch O.D. tube leading into the liquid metal pump was sectioned with the tubing cutter, it was noted that tubing was very brittle as compared with the tubing in the hot portion of the loop in the condenser and the boiler. It was thought that possibly the brittle fractions which occurred in the tubing in the low portion of the loop could be due to a low temperature-aging effect in the columbium. It is known that columbium will age in the temperature range 1600° to 1800°F; however, it is suspected that the Cb-1% Zr alloy may also age at a lower temperature, possibly 500° to 800°F. Further investigations should be undertaken to determine what the temperature, time, composition relationship is in this aging effect. No other phenomenon of first-rank significance was noted on disassembly of the loop. The cold trap was removed and an examination revealed no corrosion products. No mass transfer products were found in the loop except for a small amount noted in Figure B-18 occurring at the condenser outlet. This evidence is not considered significant in this loop run. At higher temperatures, greater ΔT's and longer operational times, this mass transfer effect should be observed closely to determine if it increases in severity. This was expected since the solubility of columbium and rubidium at this temperature was known to be very low. The loop was sectioned at various points as shown in Figure B-19 and the metallurgical analysis proceeded. The columbium tubing as received from the vendor is shown in Figure B-20. No anneal was specified after the final drawing operation and this is shown very clearly in the cold worked structure of the as-received material.

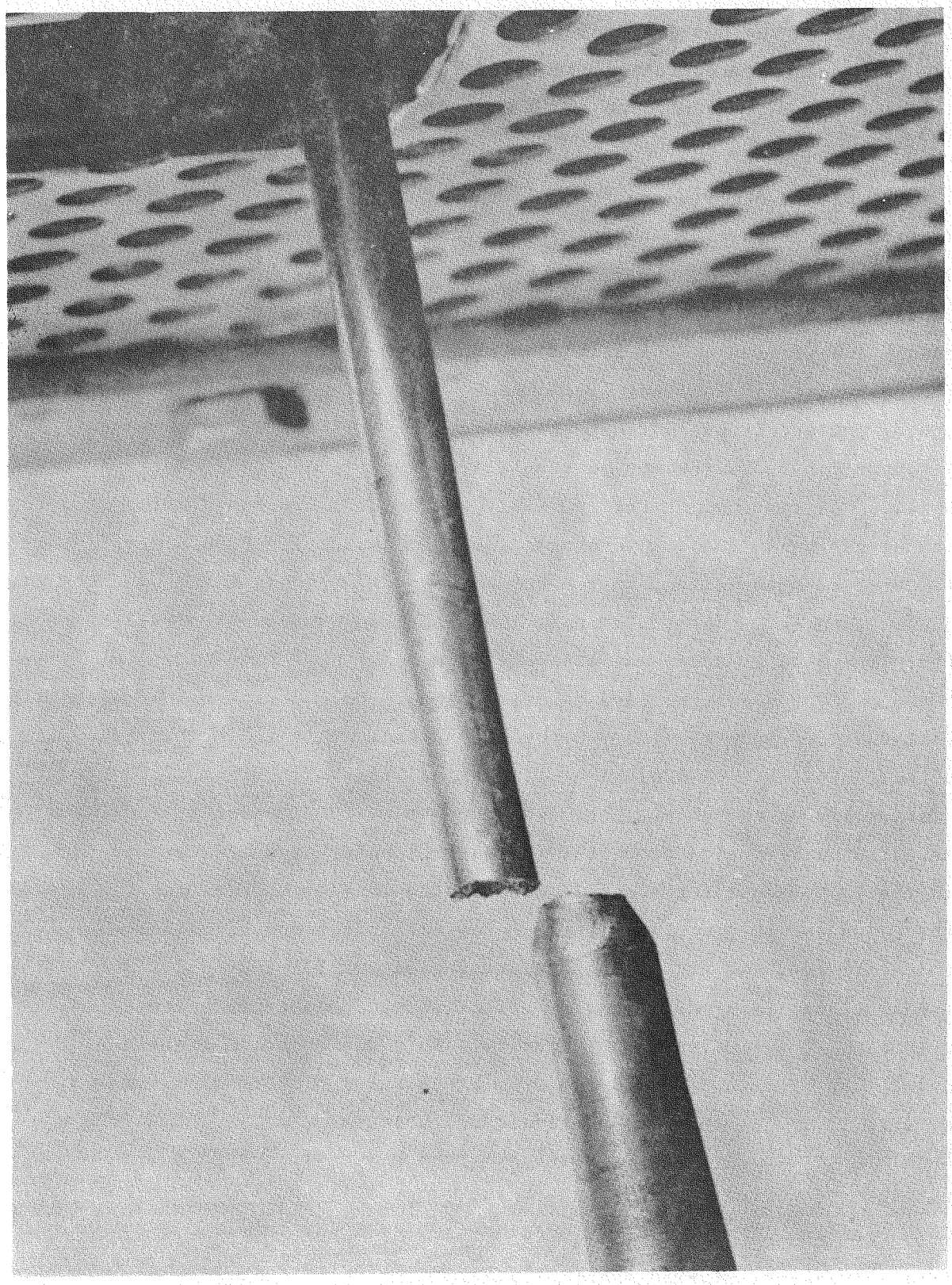
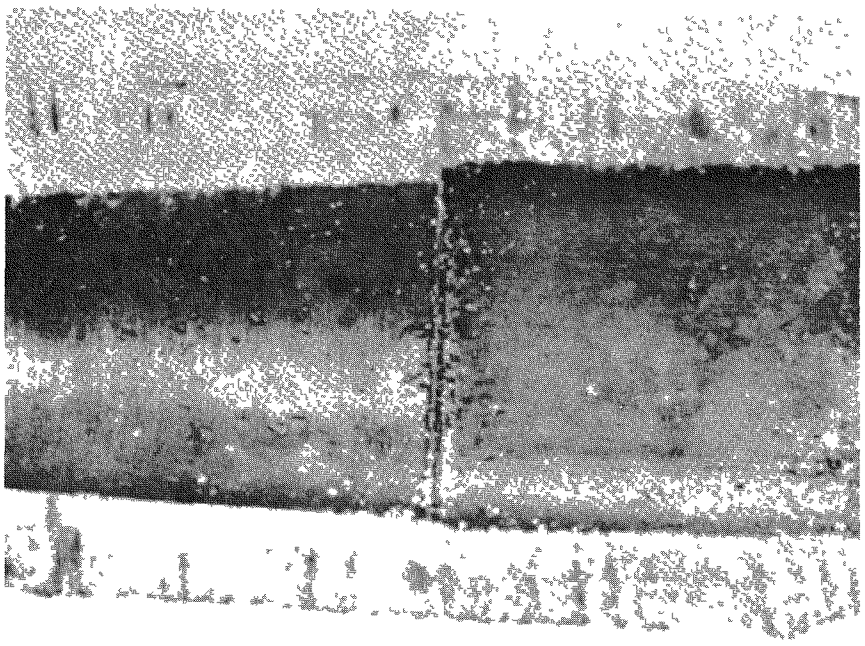


FIGURE B-17. CLOSEUP OF WELD BREAK AT FLOWMETER EXIT.



Evidence of Mass Transfer at Condenser Outlet
Photo # 589, Magnification 6 X

FIGURE B-18

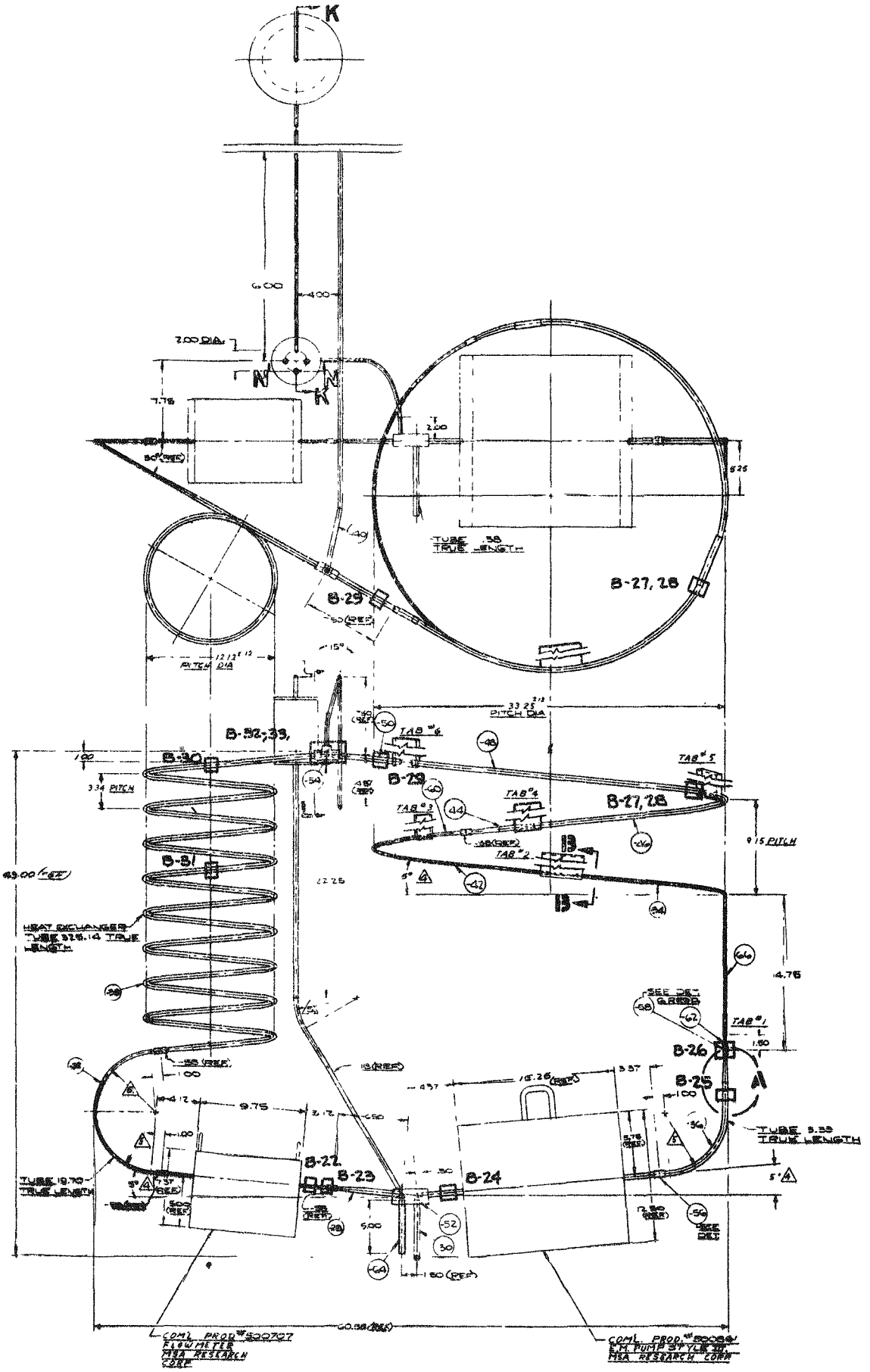
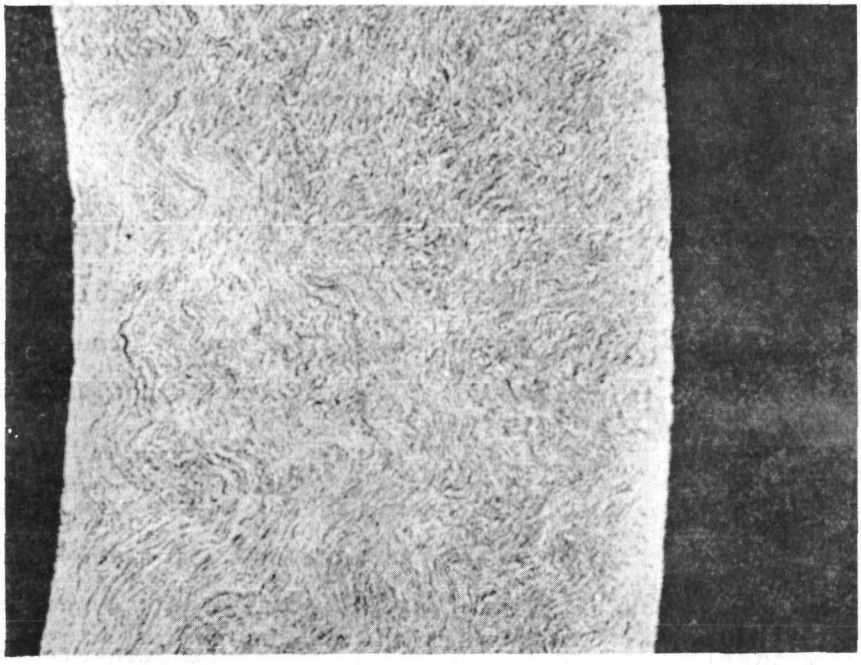
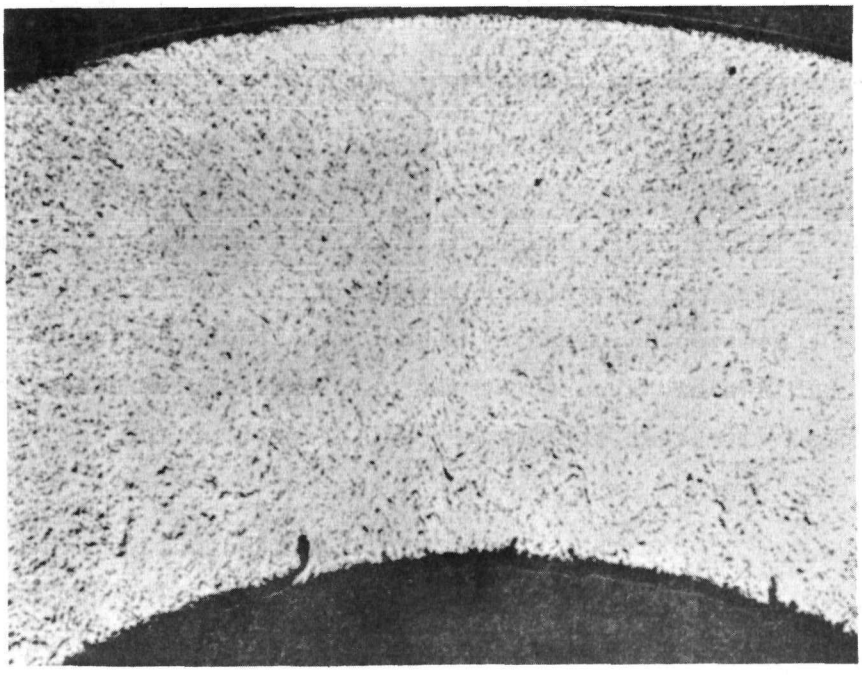


FIGURE B-19. LOCATION OF METALLOGRAPHY SECTIONS DISCUSSED IN TEXT (BY FIGURE NUMBER)

As-Received Columbium Tubing



1/2 in. O.D., Etched, Photo # 588, Mount S-6-3,
Magnification 50 X, R_B - 92.8 $\pm$ 0.3



1/4 in. O.D., Etched, Photo # 584, Mount S-5-9,
Magnification 50 X, R_B - 86.0 $\pm$ 1

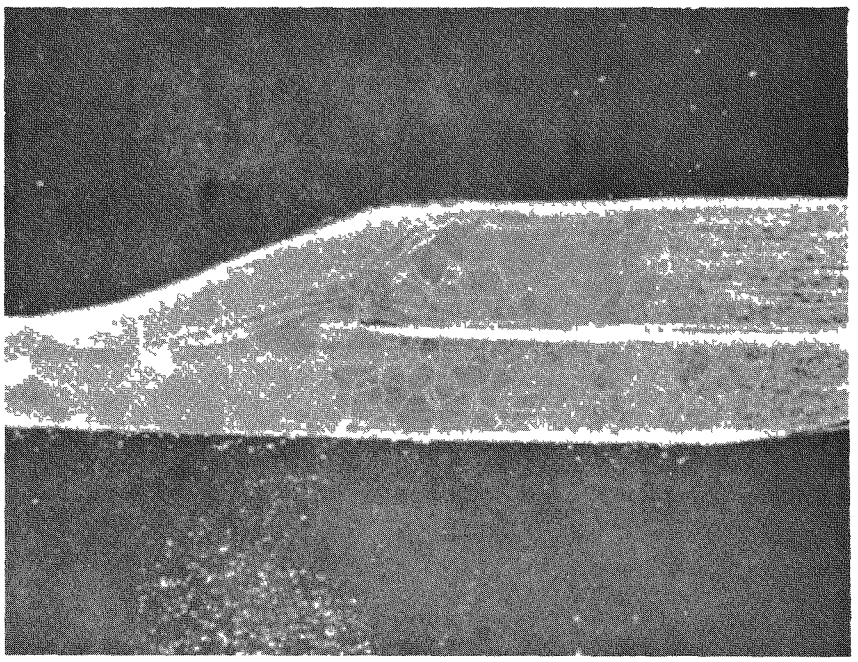
FIGURE B-20

Some "cracking" of the I.D. of the 1/4 in. O.D. tubing is evident. According to the vendor, these were not cracks but folds introduced during the drawing operation. The folds (or cracks) range from 0.5 to 4 mils deep with an average of 1 mil. The 1/4 in. O.D. tubing was R_B 86.0 $\pm$ 1 and the 1/2 in. O.D. tubing R_B 92.8 $\pm$ 0.3 in hardness. This difference was probably due to a difference in fabrication history since both sizes supposedly had the same composition. A typical weld test sample done prior to the loop fabrication is shown in Figure B-21. Note the grain growth in the weld and heat affected zone. Hardness was R_B 80.0 $\pm$ 10.0.

The 1/4 in. O.D. tubing at the flow meter exit is shown in Figure B-22. The microstructure is essentially unchanged from the as-received material; however, the hardness has increased from R_B 86 (cold worked) to R_B 91.5 (post-test). The temperature in this section was 500° to 700°F. The failed weld at the flowmeter exit is shown in Figure B-23. Pre-test hardness was not known so post-test hardness is not significant. The weld apparently was brittle and broke under the strain of loop disassembly. The surge tank line connection to the loop failed during disassembly for similar reasons.

The furnace brazed tube making up the pump cell is shown in Figure B-24. Note the thin nicrobraze coating on the O.D. of the tube. This tube fractured in a brittle manner during the loop sectioning operation. The high hardness (R_B 92.7 $\pm$ 1) would indicate that the tube was contaminated by a poor atmosphere during the brazing operation. This would seem to be the only explanation since, otherwise, it received no different treatment than the rest of the 1/2 in. O.D. tubing in the loop.

The 1/2 in. O.D. tube and weld at the entrance to the preheater are shown in Figures B-25 and 26. The hardness of the tube is R_B 95.2 $\pm$ 0.6, an increase from the as-received hardness of R_B 92.8. The weld has a post-test hardness of R_B 98.0 $\pm$ 0.2 but the change from pre-test, if any, is not known. The hardness of the 1/2 in. to 1/4 in. reducing section was R_B 69.0 $\pm$ 2. This material was fabri-



Typical Weld Test Sample
Etched, Photo # 601, Mount S-7-6, Magnification 8 X
R_B - 80.0 ± 10.0

FIGURE B-21

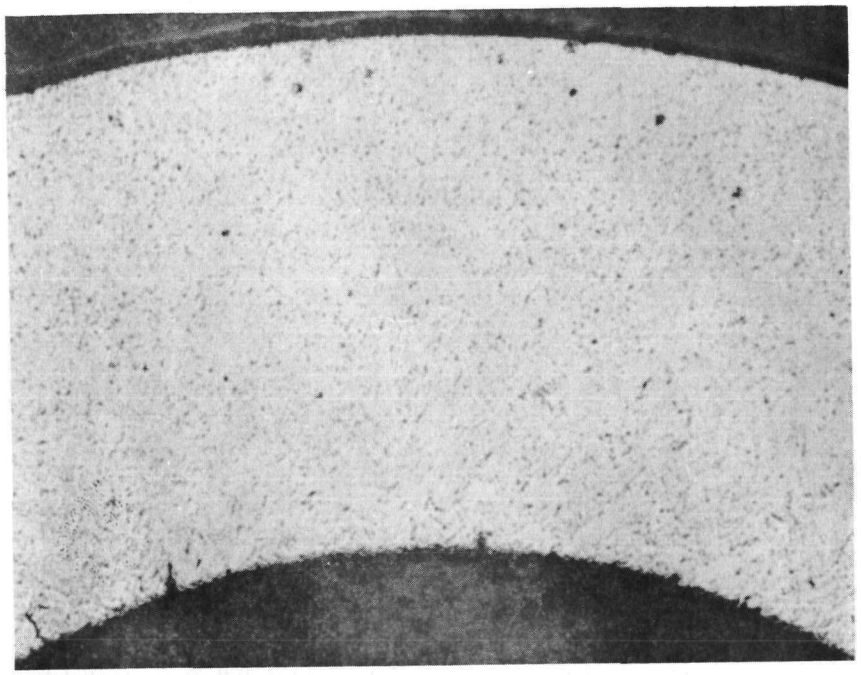


FIGURE B-22

1/4 in. O.D. Tubing at Flow Meter Exit. Etched, Mount S-4-0, Photo # 595, Magnification 60 X. R_B - 91.5 $\pm$ 1.5 (S-3-9)

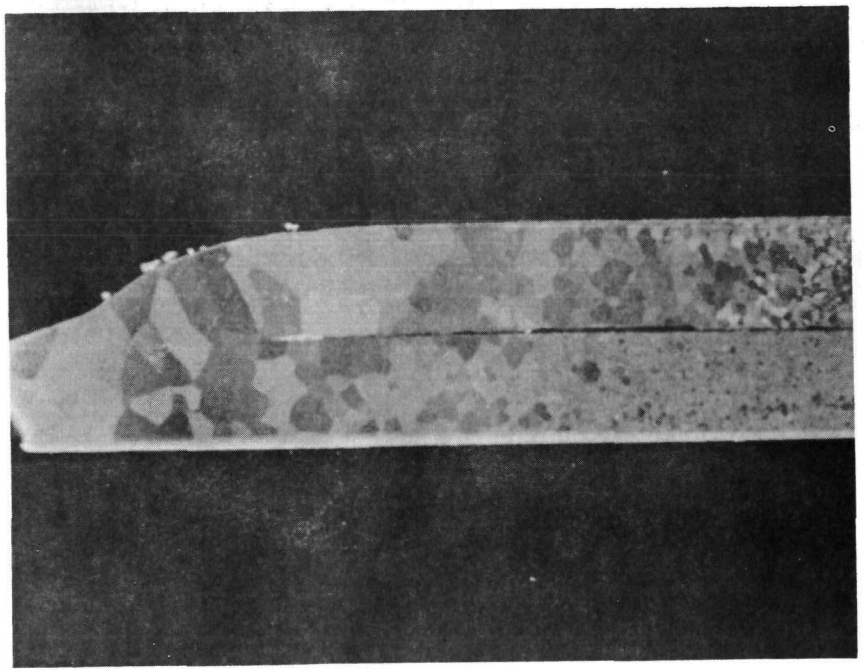
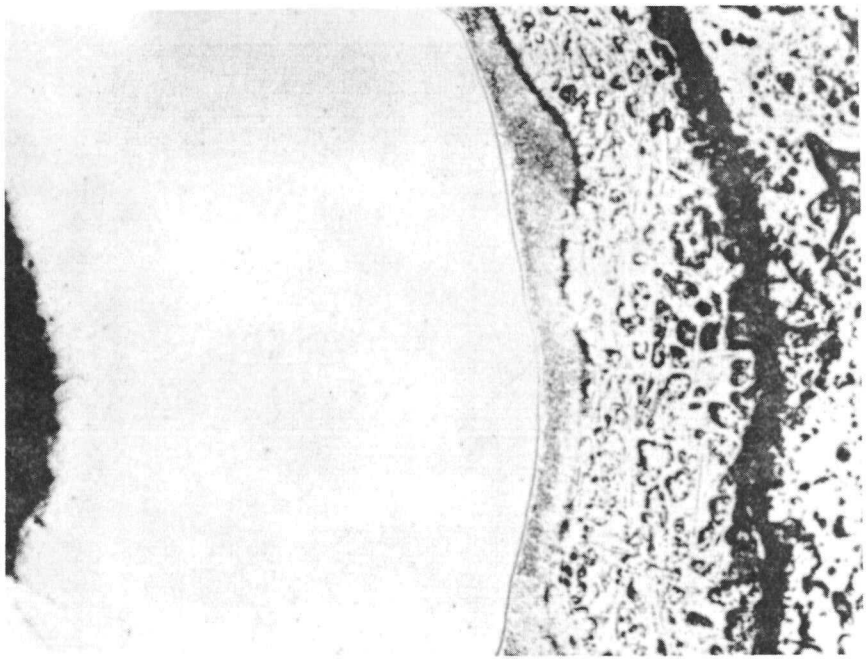


FIGURE B-23

Failed Weld at Flow Meter Outlet. Etched, Mount S-8-5, Photo # 596, Magnification 8 X. R_B - 81 $\pm$ 2 Tube, 73 $\pm$ 3 Plate, 95 $\pm$ 3 Weld



Columbium Tube Braze Nickel Buss

FIGURE B-24

Columbium Pump Cell Tube (Post-Test). Furnace Brazed Prior to Loop Run. Etched, Mount S-7-7, Photo # 594, Magnification 50 X

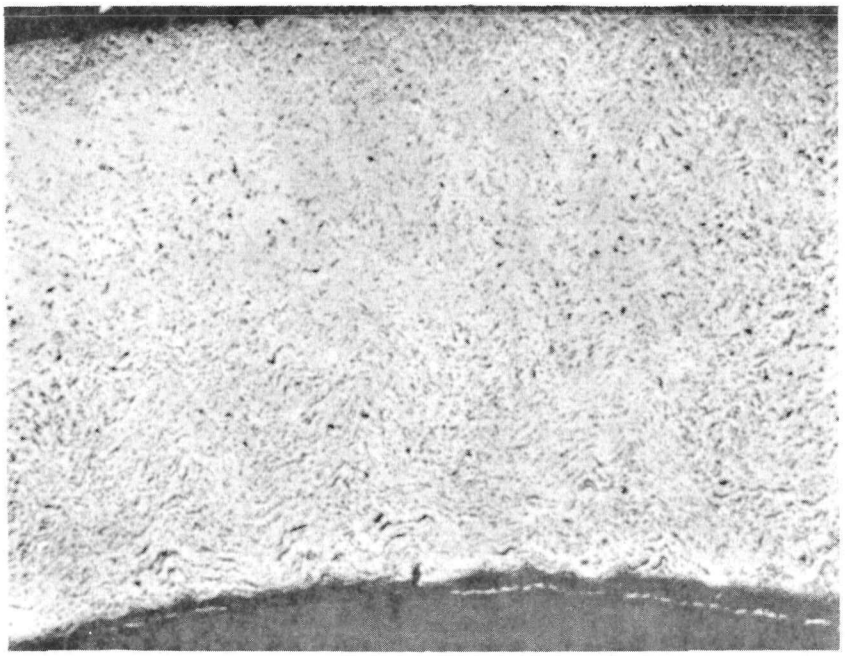


FIGURE B-25

1/2 in. O.D. Tubing (Post-Test) at Preheater Inlet. Etched, Mount S-3-4, Photo # 585, Magnification 50 X, R_B - 94.9 $\pm$ 0.6

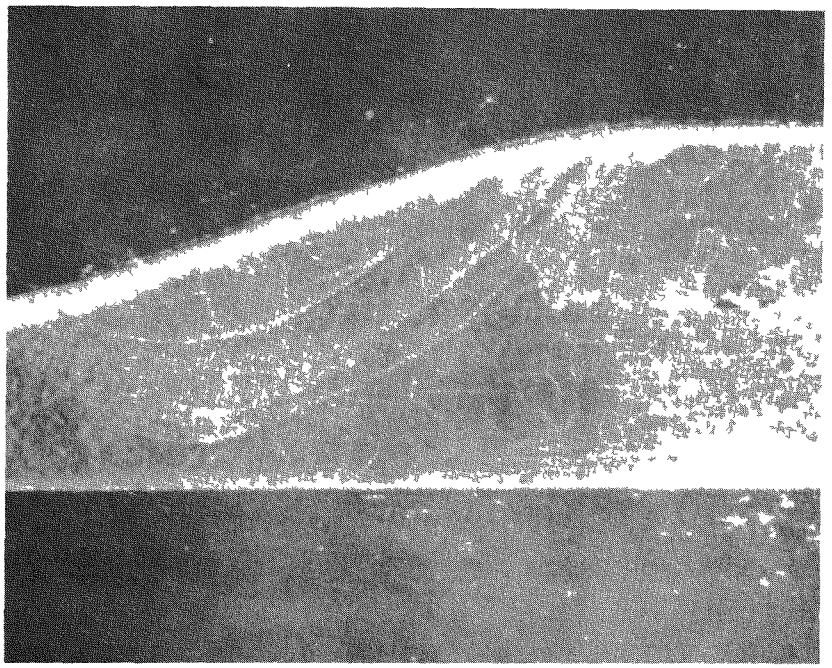


FIGURE B-26

Weld at Preheater Inlet (Post-Test). Etched, Mount S-3-3. Photo # 598, Magnification 15 X, R_B - 99.0 $\pm$ 0.7 (Weld) and R_B - 69.0 $\pm$ 2 in the Plate Reducing Section.

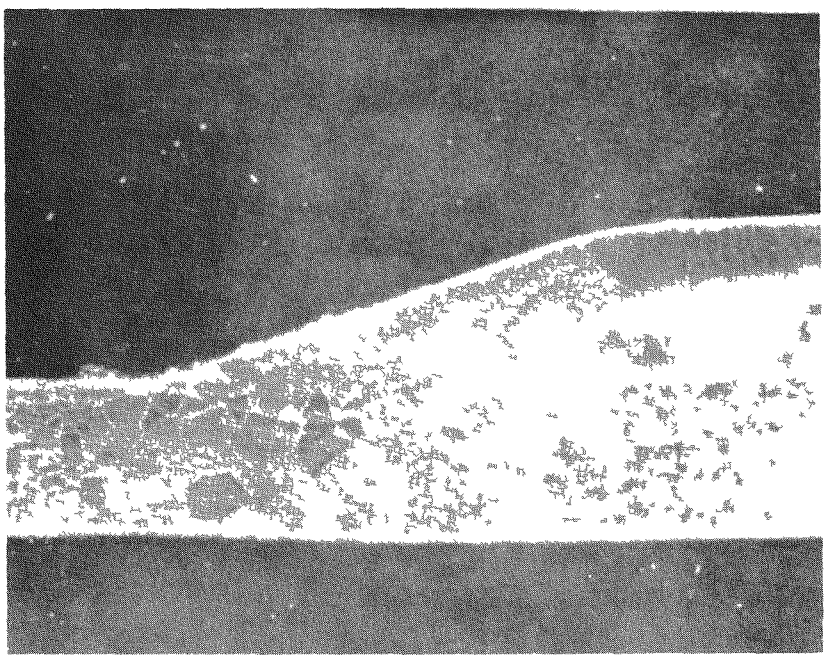


FIGURE B-27

Weld at Heater Tab # 5 (Post-Test). Etched, Mount S-7-9, Photo # 597, Magnification 15 X, R_B - 74.2 $\pm$ 0.2 Tube, 63.0 $\pm$ 10 Plate, 57.1 $\pm$ 0.2 Weld

cated from plate annealed at 1850°F before loop assembly, hence the low hardness. The cracks occurring in the weld are probably due to pre-test cracking resulting from stressing of the somewhat brittle weld during assembly of the loop. Note that neither the rubidium nor the chamber atmosphere attacked this weld.

The microstructure of the preheater and boiler tube and welds is much the same as the cold section of the loop. Again there was no perceptible corrosion by the rubidium and no corrosion by the atmosphere save where the zirconium foil became detached from the tube. Figures B-27 and B-28 show one of the welds at heater tab #5. Note the lack of any penetration of atmospheric constituents in the color photo. Compare with B-32, 33.

The problem of alloying of the zirconium foil with the columbium tubing can be seen in Figure B-29. The foil alloy has penetrated almost 10 mils into the tube wall (0.049 in.) in some places. A tantalum foil wrap may have to be substituted to avoid this effect.

Note that the first evidence of recrystallization and grain growth in the columbium occurs at the boiler exit (and probably along the latter part of the boiler). This section was running at over 1800°F. Again note that rubidium corrosion and atmospheric attack are nil.

The structures of the tube at the condenser inlet and three turns down are shown in Figures B-30 and 31, respectively. Recrystallization seems almost complete in the entrance (1800 + °F); however, three turns down the cold worked structure of the original pre-test tubing is again apparent.

The weld failure in the "T" joint which ultimately dictated the shutdown of the loop is shown in Figures B-32 and 33. Figure B-33 shows a cross section of the top weld on the "T" joint. The cracks shown are representative. An extension of a crack of this type into the I.D. of the tube caused the rubidium vapor leakage.

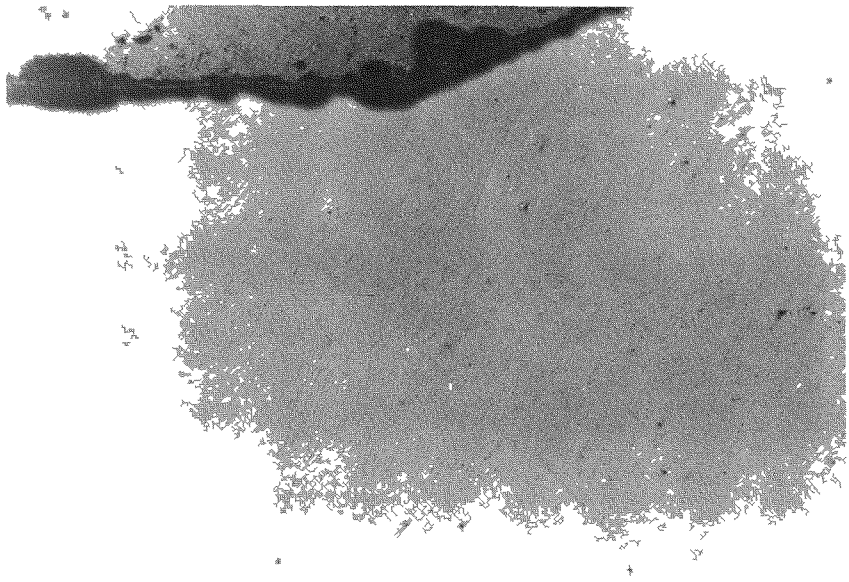


FIGURE B-28
Weld at Heater Tab # 5 (Post-Test). Etched, Mount S-7-9, Photo # Color-5, Magnification 75 X.

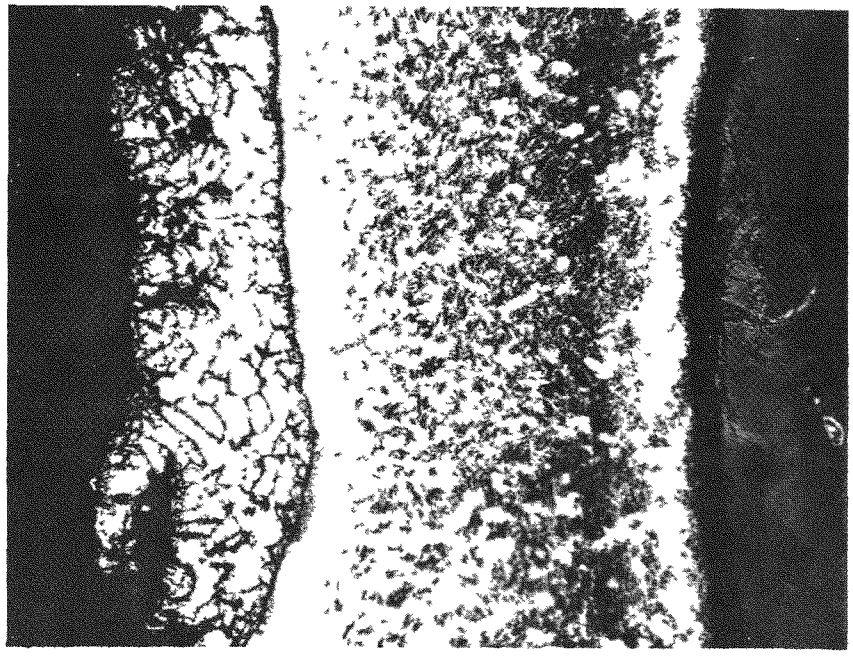


FIGURE B-29
Alloying of Zirconium Foil with Columbium Tube at Boiler Exit (Post-Test). Etched, Mount S-3-8, Photo # 488, Magnification 50 X

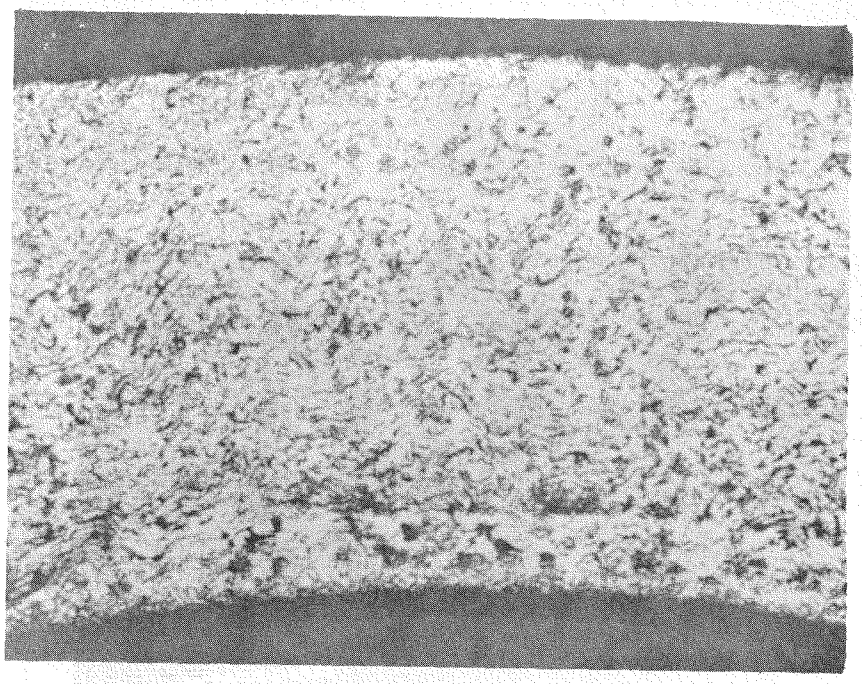


FIGURE B-30
Cross Section of Tube at Condenser Inlet (Post-Test).
Etched, Mount S-7-2, Photo # 587, Magnification 50 X,
R_B - 51 $\pm$ 1.0

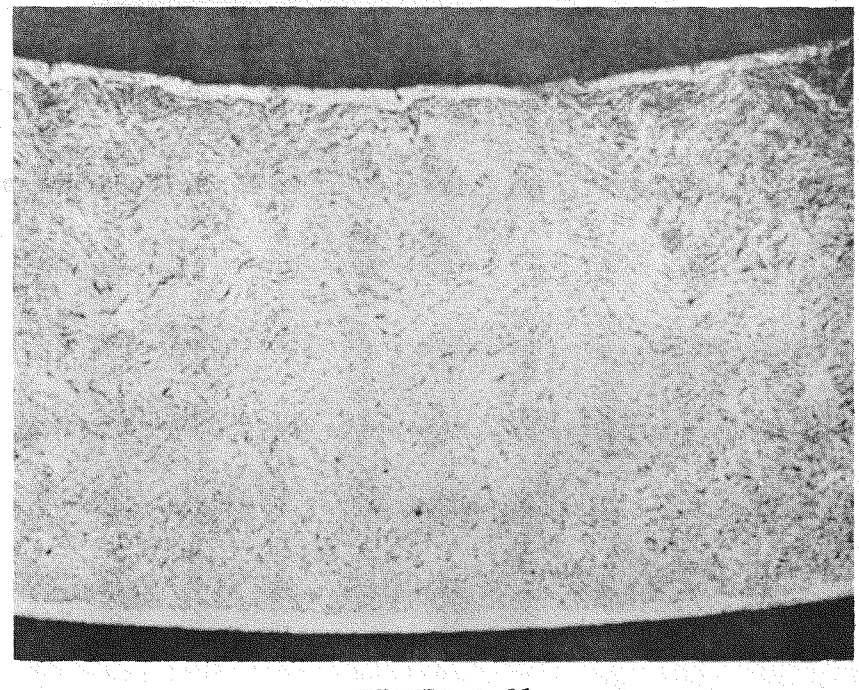
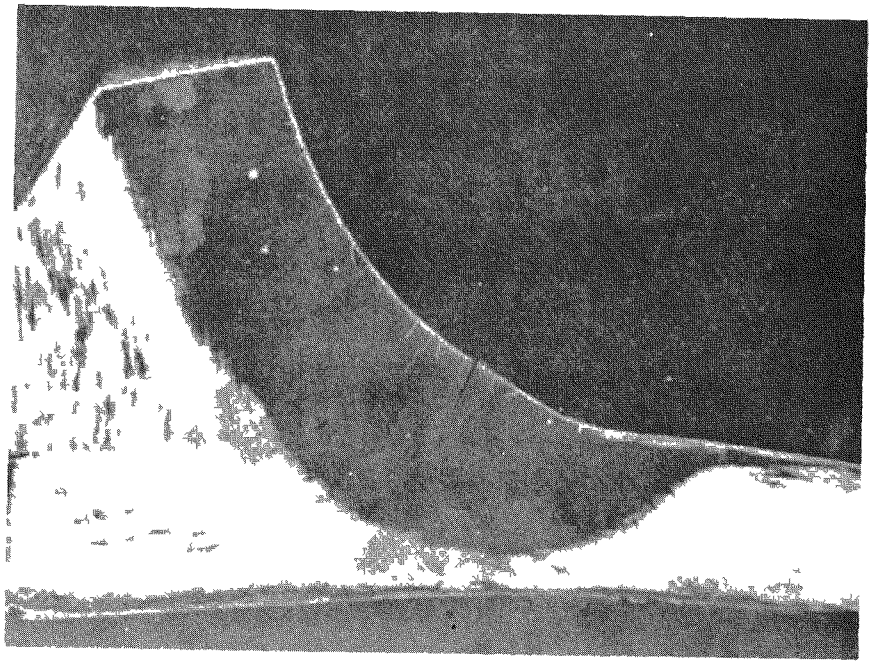


FIGURE B-31
Cross Section of Tube at Turn # 3 of Condenser (Post-Test).
Etched, Mount S-7-3, Photo # 561, Magnification 50 X

R_B - 85.2 $\pm$ 1.0



Cracks in Weld Near Evacuation Line Failure Area. Etched, Mount S-5-6, Photo # 591, Magnification 13 X, Weld Hardness R_B - 97.0 $\pm$ 2.0

FIGURE B-32

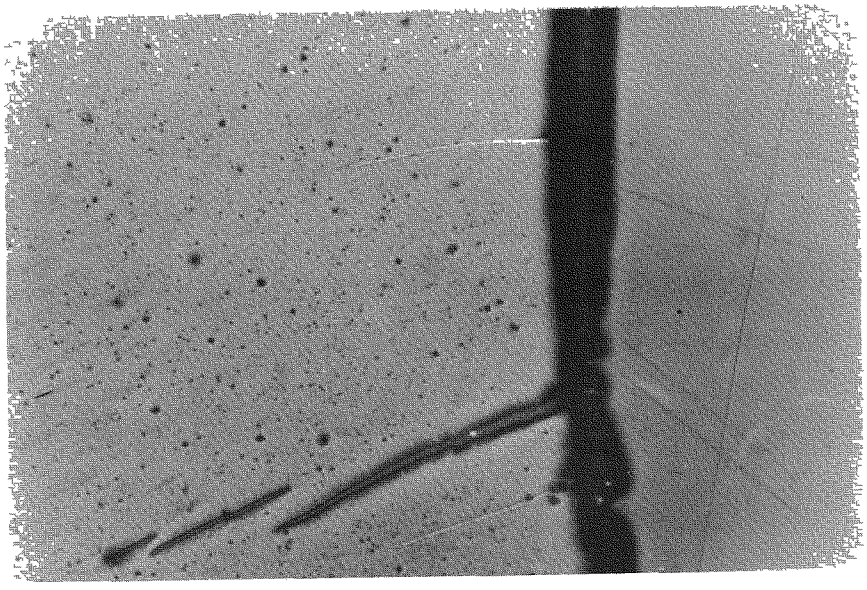


FIGURE B-33

Enlarged View of Major Crack and Associated Carbide Penetration Shown in Figure B-32. Etched, Mount S-5-6, Photo # Color-4, Magnification 100 X.

Note in Figure B-34 the carbide penetration parallel to the crack shown in Figure B-33. Apparently carbide penetration directly caused or aided the ultimate failure of this weld. The carbon could have come from a number of sources as explained on the first page of Section 12b(3). Probably the carbide penetration was not the single cause of failure. Rather, the failure most likely was caused by a combination of carbide penetration plus high weld stresses, low initial weld ductility, poor protection of the joint from atmospheric attack by the zirconium foil wrap, aging of the weld, and thermal cycling effects. Apparently the carbide preferentially attacks the weldments since this effect was not observed in other portions of the tubing which were exposed to the same temperature and environment. This may be due to the higher impurity content and lower ductility of the weld. These allow more strain to be built up in the joint. This results in greater internal energy, making the weld material more sensitive to corrosion than the unwelded tubing.

Some general observations on the test results can be made.

Little or no corrosion of the columbium by the rubidium was found even at the hottest portion of the loop.

Very minor mass transfer effects were found, indicating some solution of the columbium tubing into the rubidium occurred.

Little or no corrosion of the columbium by the chamber atmosphere occurred where the zirconium foil wrap remained "protective".

Where the zirconium foil wrap was not present originally, or where it fell away or was severely penetrated during the run, the columbium was attacked with varying severity. The welds seemed to be more sensitive to attack (especially carbide penetration than the tubing). Although the oxygen content of the argon gas was very low (in the neighborhood of 1 to

2 ppm with a light concentration of nitrogen), even this small amount was sufficient to severely attack any unprotected columbium. In most sections of the loop, the zirconium foil wrap remained continuous and acted as a protective barrier, preventing attack of the columbium.

The zirconium foil wrap tended to alloy with the columbium tubing by diffusion, especially in the hotter portions of the system.

Some age hardening of columbium apparently occurs at 1600° to $1800^{\circ}F$ and at 500° to $800^{\circ}F$.

The failure in the tee joint was due to carbide penetration of the unprotected weld, resulting in a brittle structure which broke under a combination of internal stress, bending, and thermal cycling after 455 hours of operation.

The failure in the flowmeter exit weld was due to strain applied to the brittle weld during loop disassembly operations, aggravated by poor initial weld ductility and possible age hardening of the columbium.

Ductility of the welds in the loop system was lower than that of the tubing. Hardness was higher. This is probably due to contamination in the welds.

Recrystallization temperature for this alloy is about $1850^{\circ}F$. However, the exact time-temperature-transformation relationship could not be determined from the limited data available.

(4) Chemical Analysis

The rubidium was analyzed for O_2, N_2 content before and after the loop run. The results are shown in Table III.

Metals content was not taken before or after the loop run. N_2 content was below limits of detection (< 10 ppm)

	Before	After
O_2	71 ppm	111 ppm
N_2	Not detectable	Not detectable

Analysis of the discolored foamsil insulation in the region around the leak showed zirconium and rubidium contamination. Black crud taken from the outside of the zirconium foil between heater tabs 3 and 4 showed zirconium nitride and carbon. This indicates that some carbide and nitride producing materials were present in the system. The nitrogen may have come from the argon gas. The source of the carbon is not known. It may have come from lubricating greases used in the argon blower system.

Gas analysis of the columbium tubing is shown in Table III. Note the large rise in O_2 content and small increase in N_2 content in the original versus post-test tubing. The reason for this is not apparent unless the zirconium foil wrap was less efficient as an oxygen getter than the columbium. Results of limited post-test analysis of the gas drying train showed a large increase in nitrogen content and only a small increase in oxygen content. This indicates either the absence of O_2 in the system or that the columbium and zirconium foil wrap gettered most of the oxygen. A large increase in O_2 content and negligible increase in N_2 content occurred in the zirconium foil wrap.

TABLE III
GAS ANALYSIS OF COLUMBIUM TUBING

	O_2 ppm	H_2 ppm	N_2 ppm
Original Columbium Tubing - 1/2" O.D.	160	3	80
Columbium Tubing After Test Exposed to Argon Atmosphere -1/2" O.D.	630	17	110
Columbium Tubing Weld Area After Test - 1/2" O.D.	1830	<1	110

APPENDIX C

THERMODYNAMIC AND PHYSICAL PROPERTIES PROGRAM

1. Statement of Work Scope

The thermodynamic property measurement part of the Rubidium Evaluation Program was termed Phase II. The soope of this part of the program was to design, build, and operate test equipment which would allow the direct measurement of the following properties for rubidium over the temperature range 1000 to 1800°F: (1) density, (2) vapor pressure, (3) latent heat of vaporization, and (4) specific heat. Using these measured properties, the additional properties of (1) enthalpy, (2) entropy, and (3) vapor specific volume were to be calculated.

2. Density Apparatus: Design and Operation

a. Design

After studying various possible methods of measuring the density of rubidium from its melting point to 1800°F, an expansion-level wire technique was chosen. This method consists of allowing a known weight of liquid rubidium to expand as the temperature is increased until it touches and electrically shorts a wire positioned above its surface. Then the density of the rubidium at the temperature at which the shorting occurred can be determined by dividing the weight of rubidium by the volume of the container to the tip of the level wire.

The density apparatus as designed and used is shown in Figures C-1 and C-2. The apparatus took the form of a small sealed capsule which would fit in a standard 4 in. x 4 in. laboratory type muffle furnace. It consisted of the following parts:

(1) Capsule Body

The capsule body involved a lower body, a hemispherical head, and head hold-down nut. These parts were made of type 316 stainless steel and were designed for 500 psi internal pressure at a temperature of 1800°F. A fitting was placed through the side of the capsule to allow pressure measurement and two thermocouple holes were drilled radially in the capsule base. One thermocouple extended halfway to the center and the other all the way to the center line.

RUBIDIUM DENSITY CAPSULE

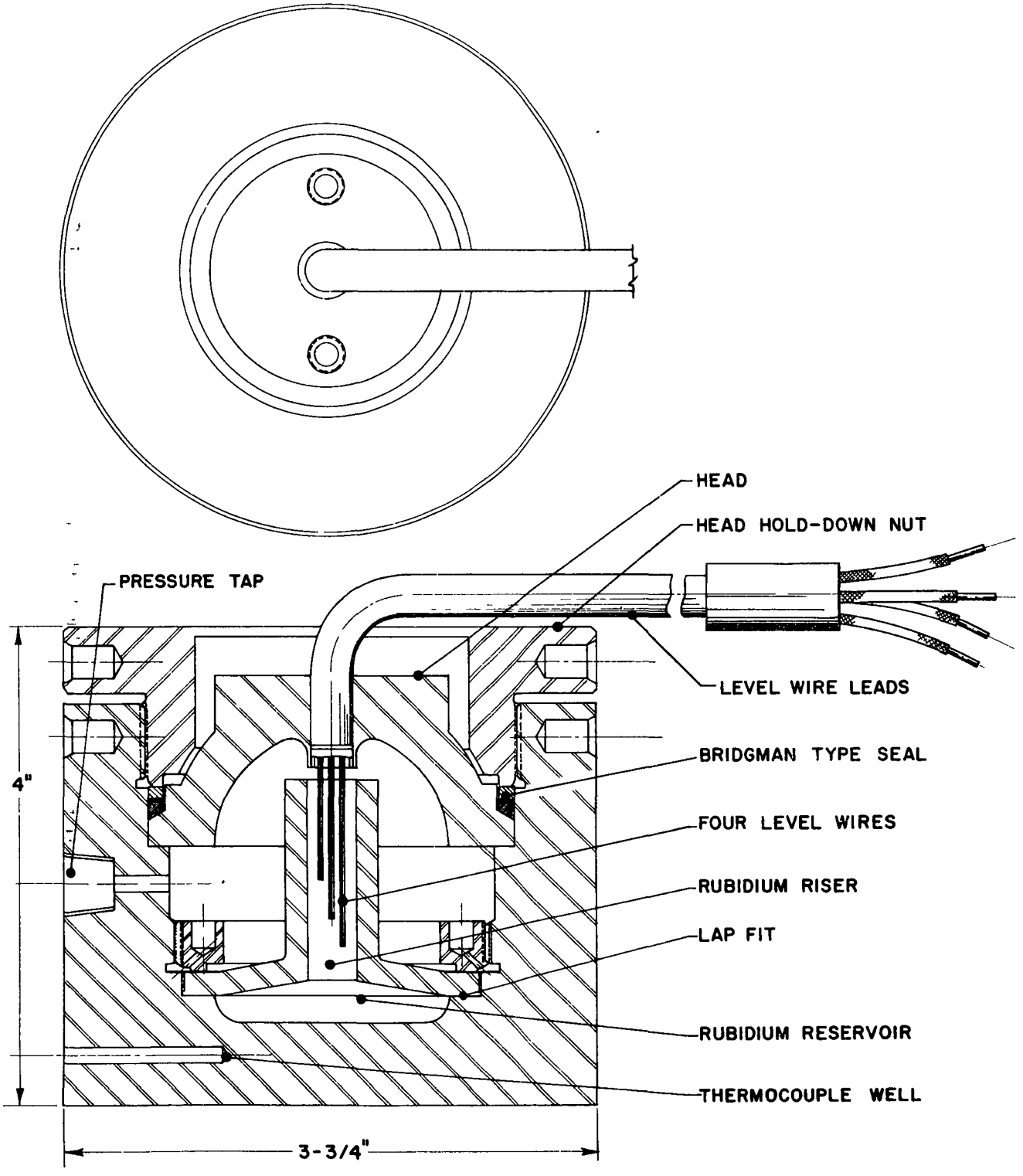


FIGURE C-1

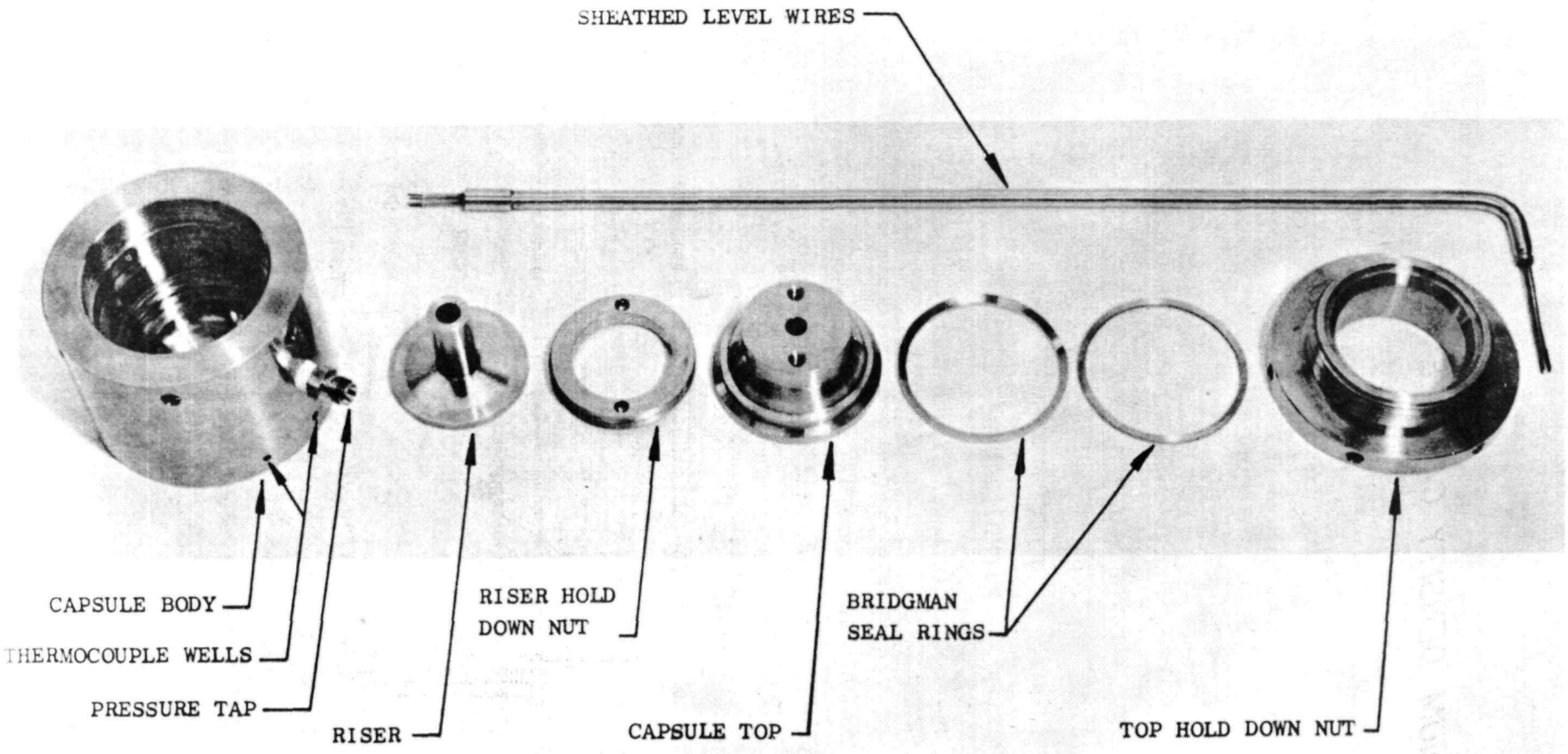


FIGURE C-2 RUBIDIUM DENSITY CAPSULE COMPONENT PARTS

(2) Capsule Seal

A "Bridgman Type" ring seal was used to seal the head in the capsule. The lower Bridgman seal ring was made of annealed A-nickel and the follower washer between the nickel ring and the head hold-down nut was of carbon steel.

(3) Capsule Internals

Inside the capsule body a small rubidium reservoir was created. The capsule body was the bottom of the reservoir and a riser section made up its top. This riser was lap-fit to the capsule body and was held in place by a threaded ring. Thus, the reservoir took a shape somewhat similar to a small volumetric flask.

(4) Capsule Level Wire Assembly

The level wire assembly consisted of four molyddenum wires swaged in MgO in a type 316 stainless steel sheath. These molybdenum wires extended past the end of the sheath so that they would reach down into the rubidium riser. The level wire sheath was seal welded into the capsule head. A 4-hole plug of high fired alumina was placed just inside the end of the sheath to prevent any MgO swage particles from falling into the rubidium pool.

The level wire electrical system consisted of a light and a bell connected to a 6-volt dry cell battery. One terminal of the battery was grounded to the capsule body and the other connected to the level wire of interest.

b. Operation

After fabrication of the apparatus, the four level wires were cut to different lengths. Thus four density points could be obtained in each run. The volume in the rubidium reservoir under each level wire was determined by introducing known amounts of mercury into the reservoir until each of the four level wires were electrically shorted by the mercury surface.

The capsule filling operation for each run was performed under an argon atmosphere in a vacuum glove box. A syringe and special pipette were used to transfer the rubidium from a glass ampoule into the reservoir in the capsule. Heat lamps were used to melt the rubidium and to maintain the transfer equipment and capsule above the melting point of rubidium. The weight of rubidium placed in the capsule was determined by weighing the rubidium ampoule and transfer pipette before and after the transfer.

After the rubidium was loaded into the capsule, the head was installed and the Bridgman seal ring assembly was torqued down. "Silver Goop" was used on the threads to prevent seizing. The seal was then tested by pressurizing the capsule to about 500 psi with argon and submerging it under water. Sufficient additional torque was applied to the head seal ring to stop any small leaks found.

A typical test run consisted of placing the capsule in a standard laboratory 4 in. x 4 in. muffle furnace. The two thermocouples (chromel-alumel) were then inserted in the holes in the base of the capsule. A special furnace door with holes drilled for the level wire sheath and thermocouples was then put in place and its edges were packed with thermoflex insulation. The temperature was then slowly raised. The temperature at which each level wire was shotted by the expanding rubidium was recorded.

At the completion of a run, the capsule was cooled down, opened, and cleaned out in preparation for the next run.

c. Test Runs

Three 4-point density runs were made during the program.

(1) Test Run #1 (November 14, 1960)

Based on the predicted density for rubidium[1], the level wire lengths were chosen to obtain four density points in the

range 500 to 1500°F. Erratic shorting of level wires #1 and #2 was observed in the range 200-600°F and then four distinct density points were obtained at 717, 934, 1229, and 1307°F.

Analysis after the run led to the conclusion that the erratic low temperature shorting was due to argon gas bubbles which were trapped during the loading operation. These bubbles then expanded and left the rubidium pool as the temperature was raised. Care was taken during subsequent capsule loadings to prevent gas entrapment by ensuring that the entire capsule was heated above the melting point of rubidium during the entire filling operation.

(2) Test Run #2 (January 24, 1961)

New level wires were prepared for the second run to cover the temperature range 200°F to 1000°F. No erratic shorting was observed and four distinct density points were obtained at 174, 332, 504, and707°F.

(3) Test Run #3 (February 2, 1961)

The level wires used in Test Run #2 were re-used for Test Run #3 and the quantity of rubidium loaded was reduced so that density points could be obtained in the range 1400 to 1800°F. At a temperature of about 1350°F weak shorting was observed on all four level wires. As the temperature was raised further, these weak signals gradually grew stronger. No distinct density points were obtained during this run.

Analysis of the test results after this test run led to the conclusion that at a temperature above about 1350°F there was enough rubidium vapor present around the wires to cause this effect. Possibly some condensation occurred at the top of the level wire assembly or the vapor itself was enough to complete the electrical circuit. It has been concluded from this run that the level wire method used in this test work is not applicable above a temperature of about 1350°F.

d. Rubidium Density Results

The eight density points obtained in Test Runs #1 and #2 are given in Table IV. The temperature given for each point is as measured by the chromel-alumel thermocouples mounted in the density capsule. These thermocouples were calibrated to a National Bureau of Standards standardized platinum-platinum +13% rhodium thermocouple. The range for error in temperature was estimated as the maximum gradiant existing between the thermocouple position and the average rubidium pool temperature.

The density values shown in Table IV were calculated from the loaded weight of rubidium and the volume of the reservoir to each level wire. This volume was corrected for thermal expansion of the capsule to the particular datum point temperature. The mean thermal coefficients of expansion for type 316 stainless steel and molybdenum listed in the Metals Handbook[2] were used. Also, the nominal volumes were corrected to a flat meniscus from the convex mercury meniscus which existed during the room temperature calibration of volume. The estimated possible error in each density point includes the accuracy of the weighing and volume measurements, the impurities (see Table VI) and possible meniscus error. Meniscus error accounts for the major part of the estimated error. If the rubidium meniscus was convex the estimated error should be added to the density and if it was concave the error should be subtracted.

TABLE IV

MEASURED DENSITY OF RUBIDIUM* VS TEMPERATURE

1. Melting Point Density for Rubidium[3] = 1.472 g/cc @ 102°F

2. Test Run #2

Date of Run: January 24, 1961

Rubidium Loading: 12.7271 grams

Level Wire #	Temperature °F	Density g/cc
1	174 ± 5	1.459 ± 0.005
2	332 ± 5	1.419 ± 0.005
3	504 ± 5	1.377 ± 0.005
4	707 ± 5	1.337 ± 0.005

3. Test Run #1

Date of Run: November 14, 1960

Rubidium Loading 11.7881 grams

Level Wire #	Temperature °F	Density g/cc
1	717 ± 5	1.329 ± 0.005
2	934 ± 5	1.288 ± 0.005
3	1229 ± 5	1.248 ± 0.005
4	1307 ± 5	1.217 ± 0.005

* See Table VI for chemical analysis of rubidium used.

3. Boiling Apparatus: Design and Operation

The rubidium boiling apparatus was designed to allow direct measurement of the vapor pressure and latent heat of vaporization of rubidium over the temperature range 1000 to 1800°F.

a. Operational Concept

The concept of operation was to heat, boil, and condense rubidium at a measured rate within a sealed container which was adiabatically shielded from its surroundings. A test run consisted of establishing temperature equilibria at a number of successive temperatures between 1000 and 1800°F. At each equilibrium datum point, the rubidium temperature and vapor pressure was recorded and the latent heat of vaporization measured.

The vapor pressure measurement device was a diaphragm type strain gage transducer. This transducer was fitted at the end of a tube which extended down from the boiling metal pool into a cool region (between 200 and 300°F). The vapor pressure of the rubidium was transmitted to the transducer by the column of liquid metal above it.

Latent heat of vaporization was determined at a datum temperature by measuring the following quantities:

The heat loss from the boiling vessel with no boiling occurring. (This heat loss was set to a minimum (or zero) by adjusting the adiabatic guard shielding.)

The total heat input to the rubidium boiling vessel.

The rate of boiling.

Subcooling of condensed rubidium.

Knowing these quantities, the latent heat of vaporization at the datum temperature was calculated by dividing the total heat input corrected for heat loss and condensate subcooling by the boiling rate.

Determination of the boiling rate was the difficult measurement. A detailed discussion of how this measurement was made is given in the next section. Briefly, the measurement consisted of collecting all of the rubidium condensate in a reservoir which was periodically emptied by a siphon. The time to fill the calibrated volume of this reservoir was determined, using a radiation-source level measurement technique. Then the mass flow rate of condensate (i.e., the boiling rate) was determined by multiplying the measured volume flow rate by the density.

A preliminary check-out of this experimental method was made in a glass-water mock-up. In operation, it showed good siphoning stability and suggested several design features which were incorporated into the actual apparatus design.

An error analysis was made of this measurement method which indicated that latent heat of vaporization could be measured to within $\pm$ 3% at 1000°F and to within about $\pm$ 5% at 1800°F.

b. Boiling Apparatus Equipment

The boiling apparatus consisted of two basic parts: a vessel containing the boiling rubidium and a thermal radiation shield assembly. Figure C-3 is a cutaway drawing of the apparatus.

(1) The Boiling Vessel

Details of the boiling vessel and its internal parts are shown in Figures C-4, C-5, C-6, and C-7. The cylindrical-shaped vessel had a flanged head. It was made of Inconel with an I.D. of about 6 in. and a height of about 12 in. The flanged head was first seal welded to the vessel body and then held in place by 16 Haynes-25 1/2 in. bolts. The vessel was designed to contain 500 psi at 1800°F.

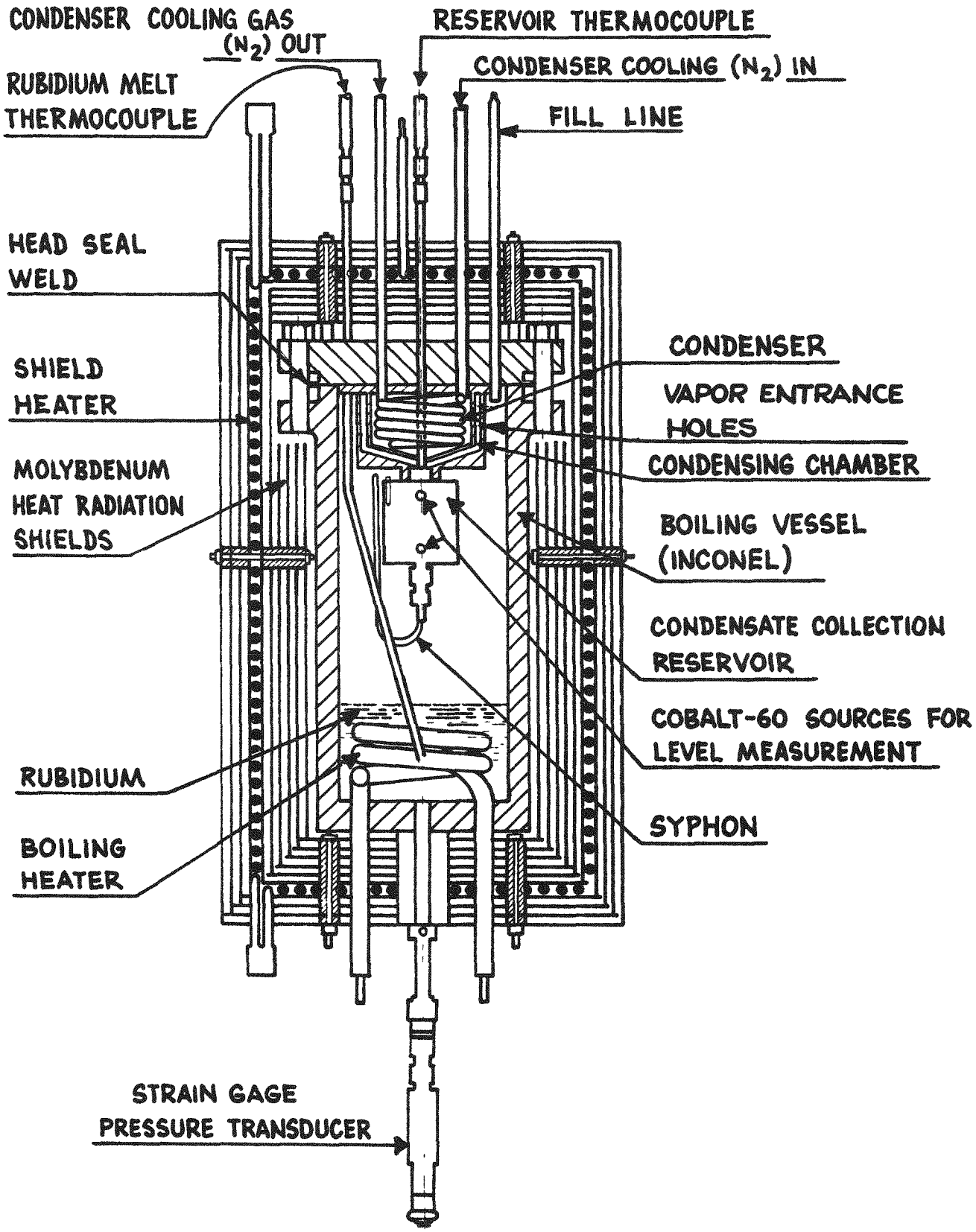


FIGURE C-3 RUBIDIUM BOILING APPARATUS

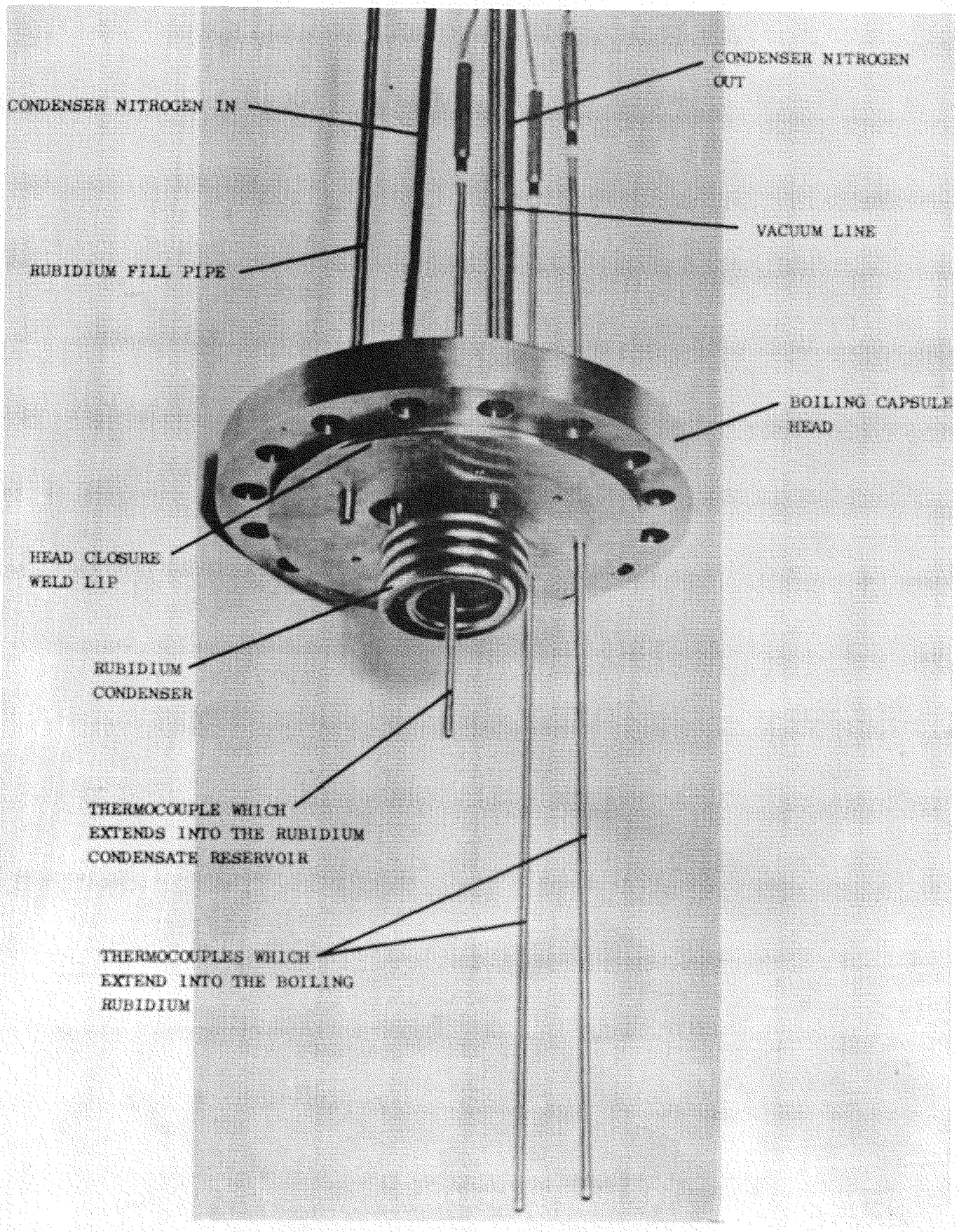


FIGURE C-4 HEAD OF BOILING RUBIDIUM CAPSULE SHOWING CONDENSER

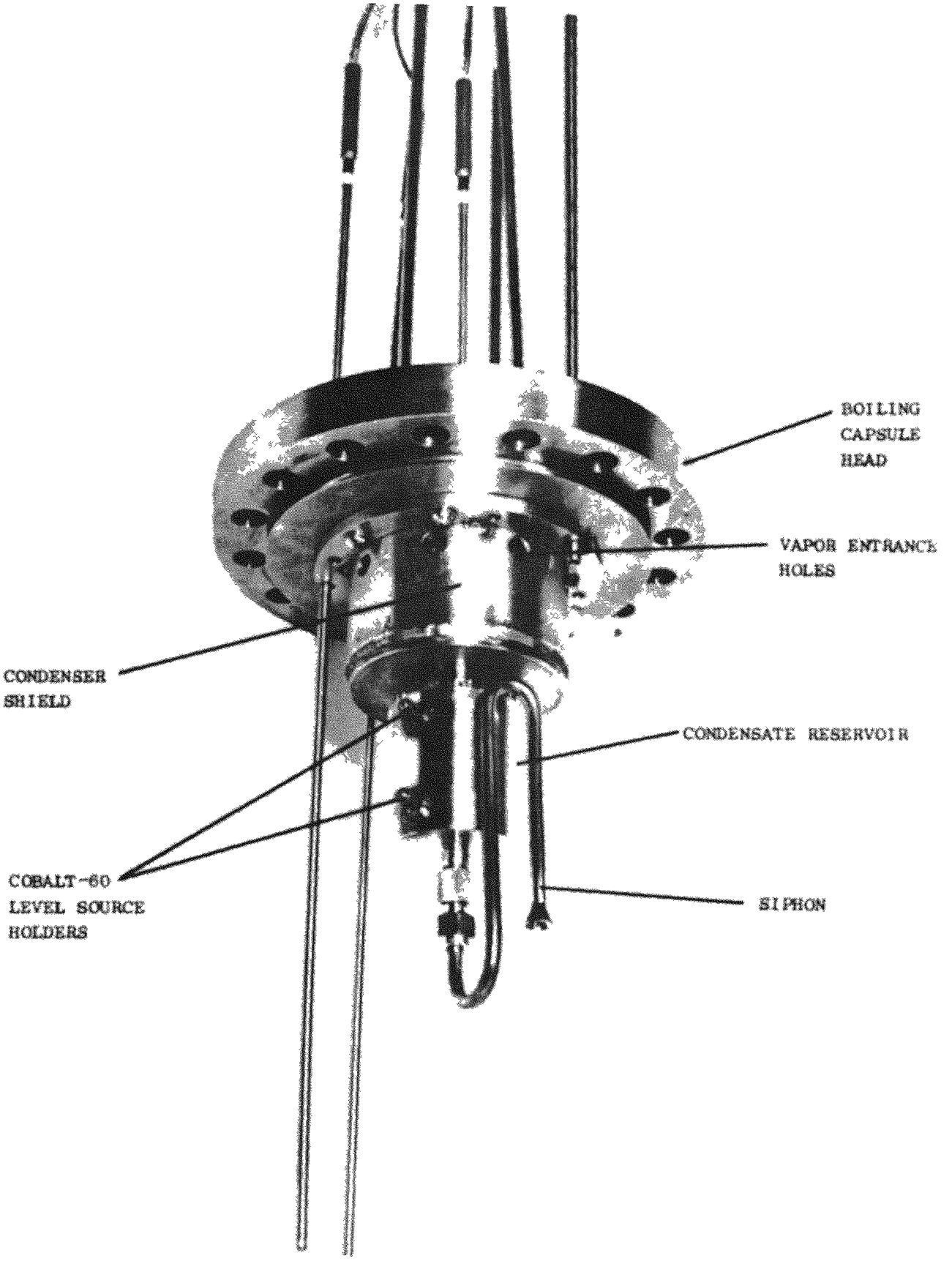


FIGURE C-5. HEAD OF BOILING RUBIDIUM CAPSULE WITH CONDENSER SHIELD, CONDENSATE RESERVOIR AND SIPHON ATTACHED.

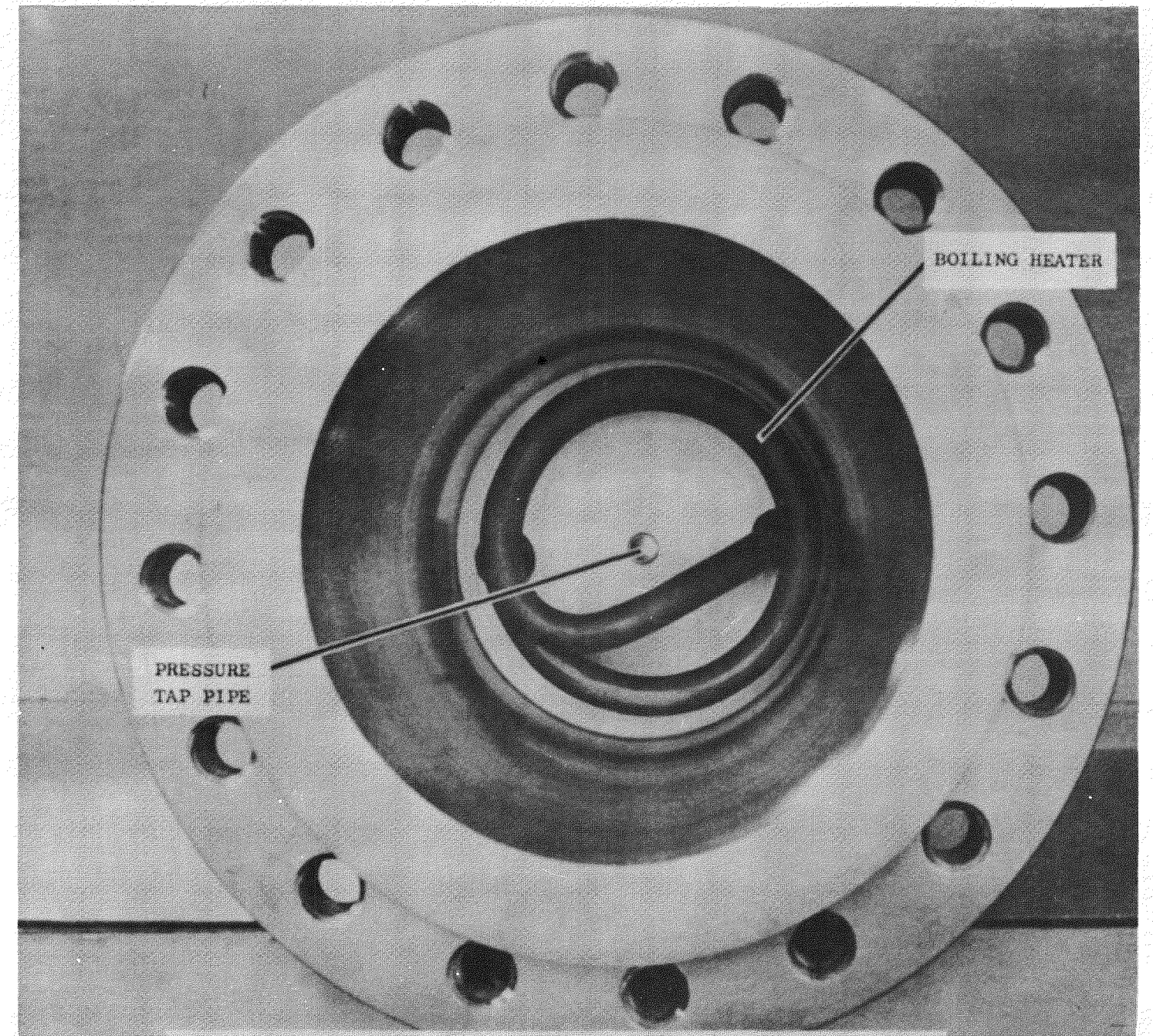


FIGURE C-6 TOP VIEW OF OPEN BOILING RUBIDIUM CAPSULE BODY

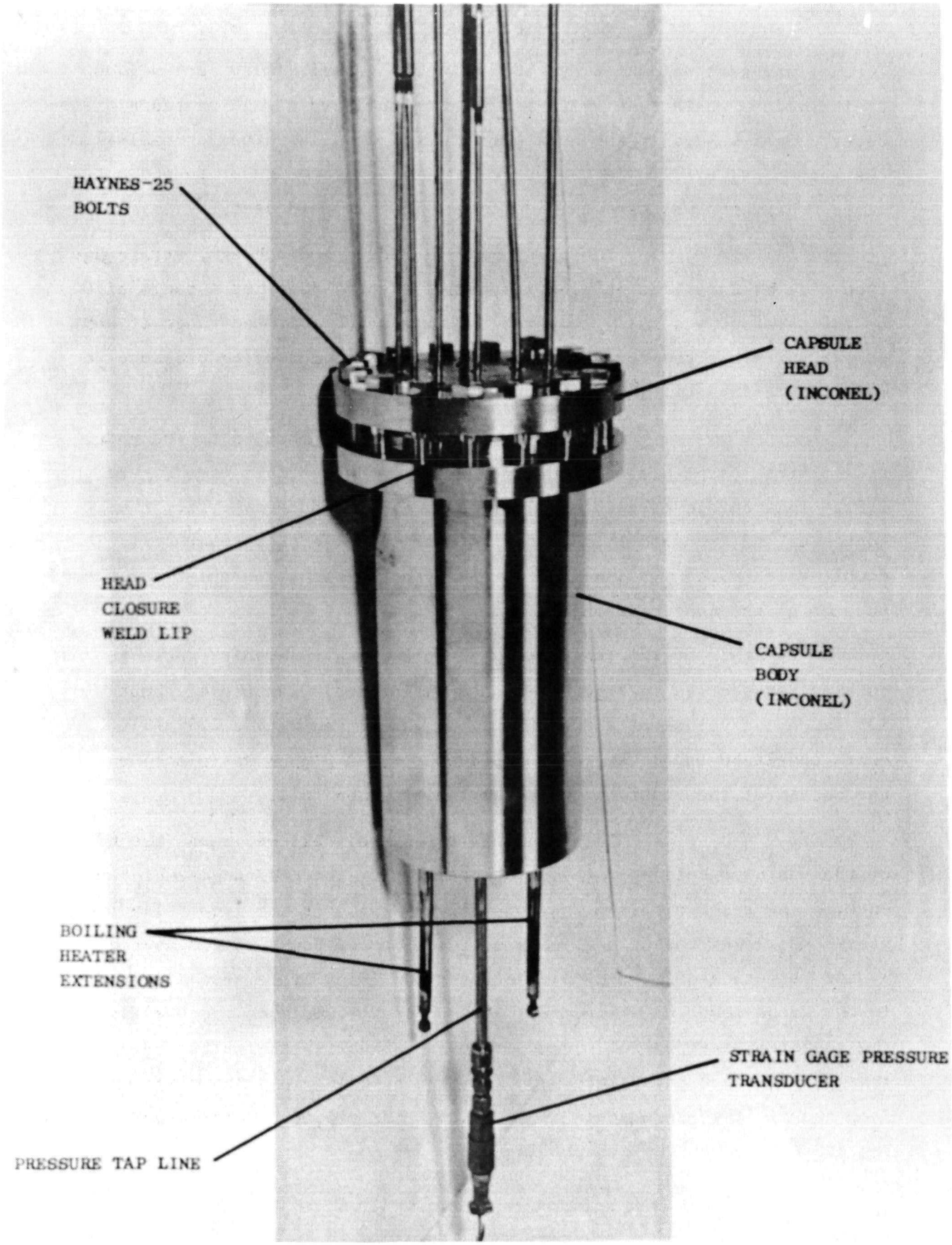


FIGURE C-7. ASSEMBLED BOILING RUBIDIUM CAPSULE

A Calrod type resistance heater sheathed with Inconel was seal welded in the bottom of the vessel. Also seal welded to its bottom was a tube which led down to the strain gage pressure transducer. Seal welded in the head were three Inconel sheathed chromel-alumel thermocouples, a fill line, a vacuum line and the condenser in and out lines. Nitrogen was used as the coolant in this condenser.

The pressure transducer, a diaphragm, strain gage type made by Consolidated Electrodynamics (Type 4-326) had a range of 0 to 250 psia and was fully temperature compensated over the range of -65 to +250°F. This type of transducer is capable of measuring pressure to $\pm$ 0.1 psi or about $\pm$ 1%, whichever is larger.

A double walled shield was mounted to the under side of the vessel head and surrounded the condenser coil (see Figure C-5). This shield collected all of the rubidium condensate from the condenser and funneled it into the condensate collection reservoir. The double wall reduced radiant heat transfer between the condenser and the outside of the condenser shield.

The condensate collection reservoir (shown in cutaway perspective in Figure C-8) had an internal diameter of about 1.5 in. and a volume of about 30 cc. It was fitted with a 0.5 millicurie cobalt-60 source at its top and bottom. A siphon was placed on the bottom of the reservoir which emptied it when full.

This condensate reservoir allowed measurement of the rubidium boiling rate during operation. The condensed rubidium entered the reservoir from the top (Figure C-8). As the liquid level passed the lower cobalt source, a decrease in the count rate measured by the radiation detector would be observed. Finally, a second decrease in the gross count rate measured by the detector would be observed as the rubidium level passed the upper source. Shortly thereafter, the level would reach the point where the siphon would empty the reservoir. Then the filling cycle would be repeated. The predicted radiation detector response curve is shown in Figure C-9.

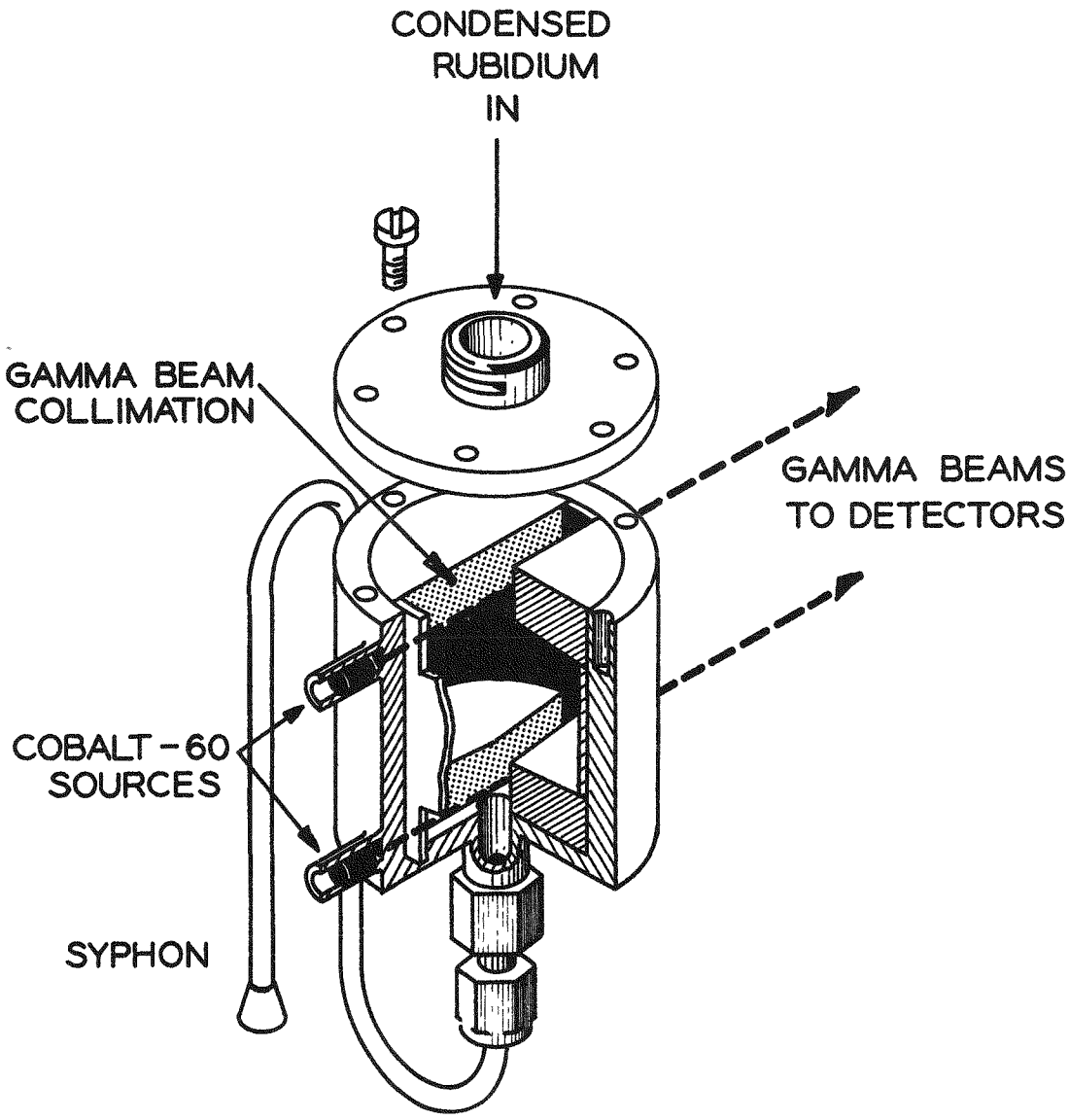
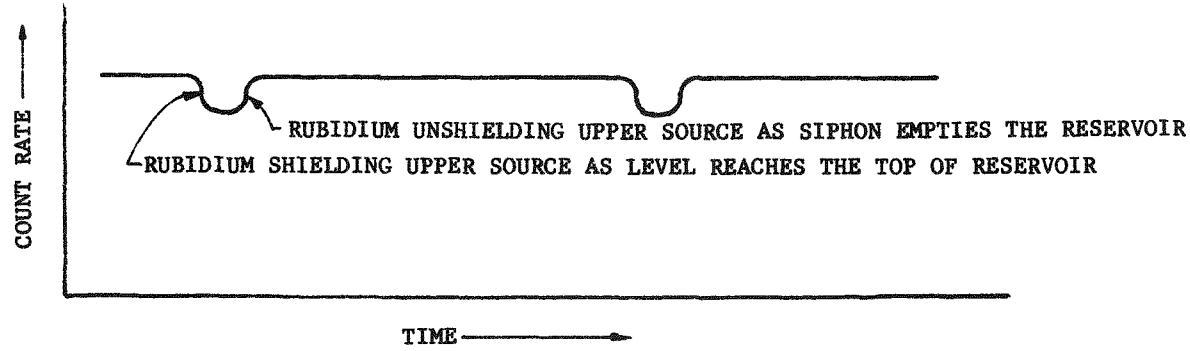
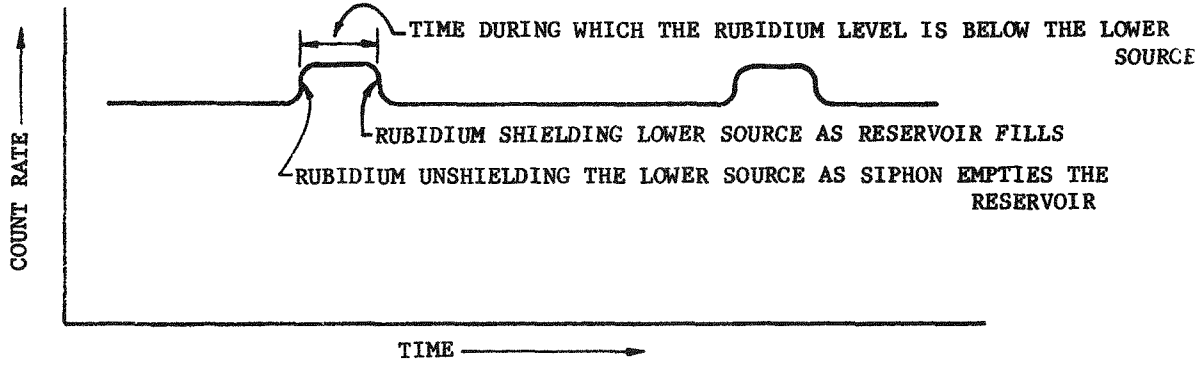


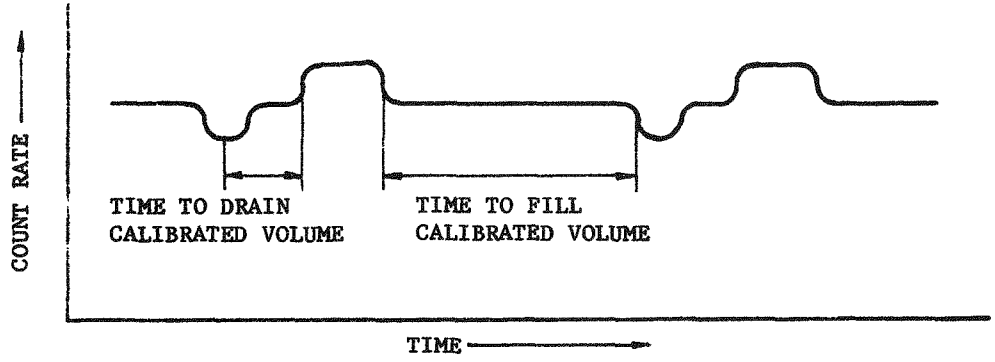
FIG. C-8 CUTAWAY OF CONDENSATE RESERVOIR



(a) COMPONENT OF DETECTOR RESPONSE DUE TO UPPER SOURCE



(b) COMPONENT OF DETECTOR RESPONSE DUE TO LOWER SOURCE



(c) TOTAL RADIATION DETECTOR RESPONSE CURVE

FIGURE C-9 PREDICTED RADIATION DETECTOR RESPONSE CURVE

The condensate flow rate (i.e., boiling rate) would then be determined by dividing the calibrated volume of the reservoir between the sources by the time to fill and multiplying by the liquid density at the datum temperature. As shown in Figure C-8, radiation source collimation was built into the condensate reservoir at its top and bottom so that an error in level detection would cause only a small error in volume determination. This collimation was 1/16 in. wide and the full 1.5 in. diameter long. It was expected that about a 10 to 12% decrease would be observed in total count rate as the liquid level passes each source.

All of these internal parts were fabricated from Type 316 stainless steel.

(2) The Thermal Radiation Shielding

An accurate measurement of the heat input to the boiling apparatus was required to measure latent heat of vaporization. Therefore, the boiling vessel was surrounded by an adiabatic barrier of heated thermal radiation shields (see Figure C-3). These radiation shields were made from 10 mil thick molybdenum sheet and shaped in the form of concentric cylindrical cans. There were ten such concentric radiation shields, each separated by ceramic spacers. A Calrod type heating element was placed in the radiation shielding between cans 6 and 7. The side section of this heater was in the form of a coil and the top and bottom sections were in the form of flat spirals.

During operation, the temperature of the shield heater was controlled by differential thermocouples placed between it and the boiling capsule. The shield heater controller was set to hold a zero differential thermocouple reading between the boiling capsule and the shield heater. In this way, the heat loss from the boiling apparatus was small as compared to the heat input required to boil the rubidium. The six molybdenum shields between the shield heater and the boiling capsule reduced the amount of heat lost (or gained), due to mis-

match of the shield heater temperature. The three molybdenum shields between the shield heater and the vacuum box reduced the heat loss from the shield assembly and thus minimized the shield heater power requirement.

Four sets of differential thermocouples were placed in the radiation shield assembly. One set was placed on the top, one on the bottom and two sets on the side (high and low). All of these couples could be read for information and either the upper side or lower side differential couple could be used as the input to the shield heater controller. A pre-amplifier was used to magnify the small differential thermocouple signal so that fine control could be achieved.

(3) The Over-All System

Figure C-10 is a photograph showing the boiling apparatus installed in the vacuum chamber. The entire inside surface of the vacuum chamber was covered with two layers of aluminum foil to reduce radiant heat loss from the apparatus and to prevent overheating of the vacuum chamber walls. The scintillation gamma detector with its cooling coil and radiation shield was mounted in the back of the chamber. The strain gage pressure transducer and its cooling coil extended down from the apparatus into a well in the vacuum chamber. The power, thermocouple and cooling lines were led out through connectors in a plate which fitted into one of the glove ports in the vacuum chamber door.

The gamma detector high voltage line (1500 volts) was led through a ceramic insulated welding connection in the side of the vacuum chamber.

The control panel used with the apparatus is shown in Figure C-11 and the control bench is shown in Figure C-12.

c. Test Operations

(1) Calibrations

The strain gage pressure transducer was calibrated over its range of 0-250 psia both before and after the test runs.

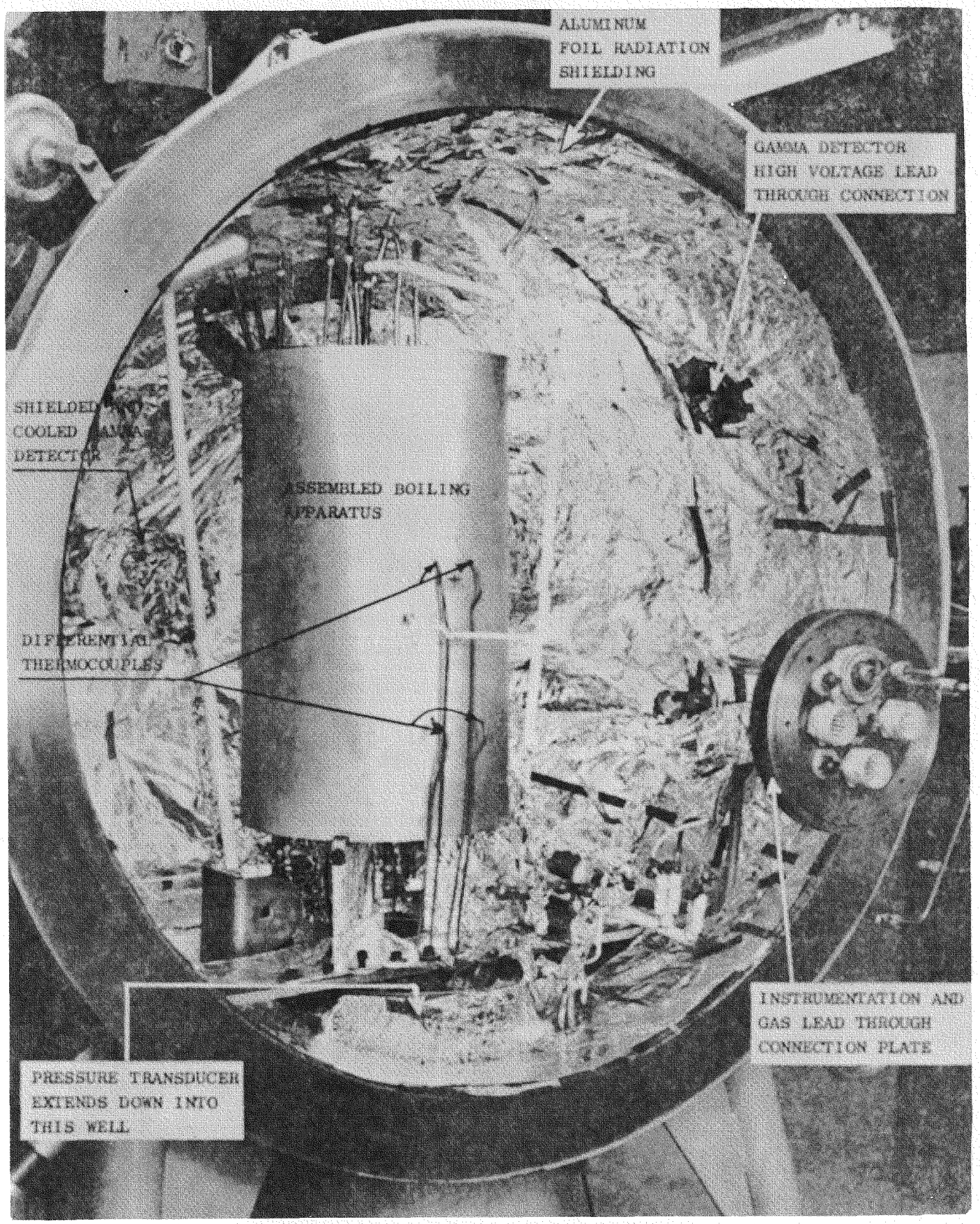


FIGURE C-10. ASSEMBLED BOILING APPARATUS INSTALLED IN THE VACUUM CHAMBER (DOOR OPEN)

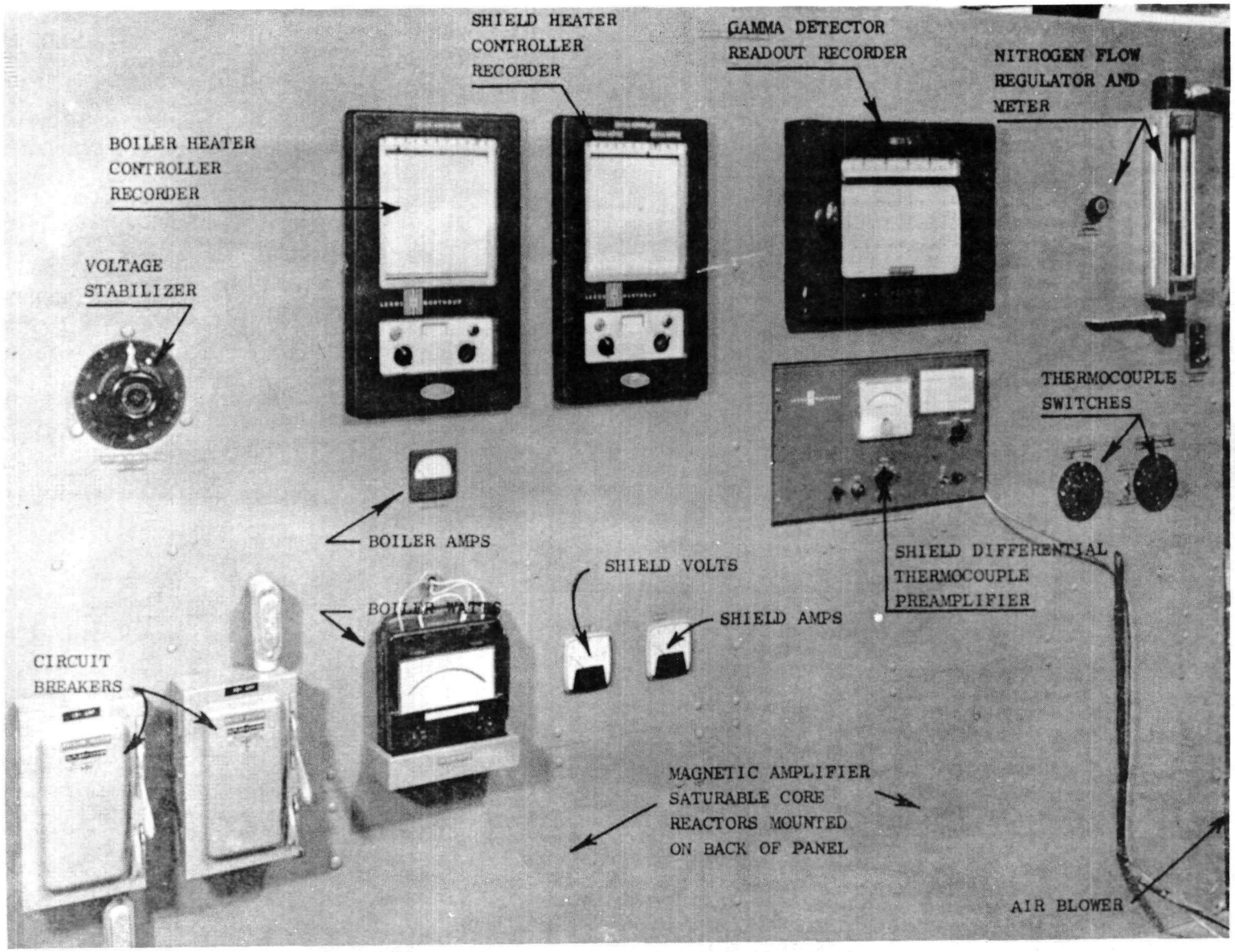


FIGURE C-11. BOILING APPARATUS CONTROL PANEL

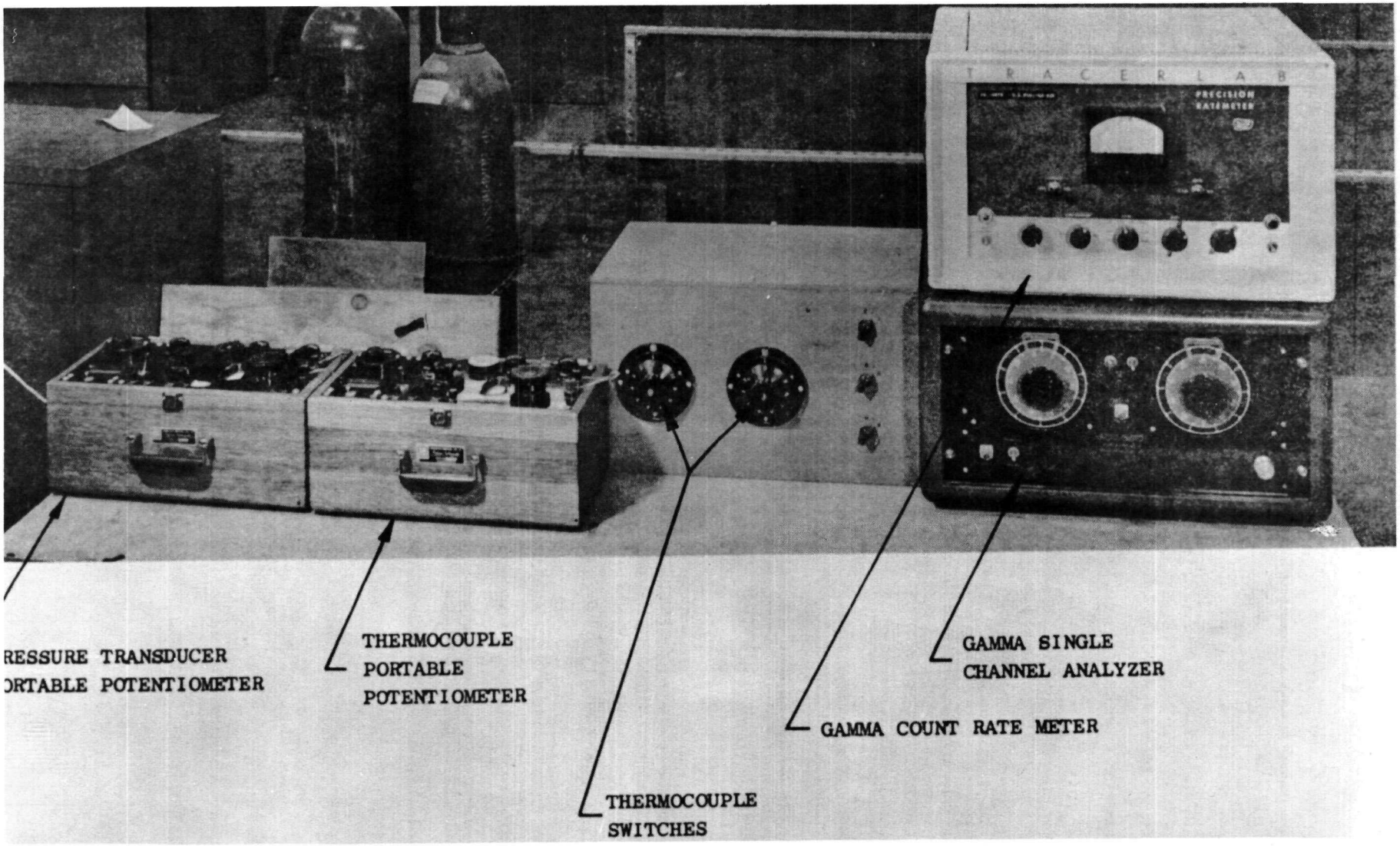


FIGURE C-12. BOILING APPARATUS TEST BENCH

These calibrations were made at room temperature and at 200°F to check the temperature compensation characteristics. A mercury manometer was used as the standard from 0 to 20 psia and a large ± 1/4% bourdon tube pressure gage was used between 20 and 250 psia. Corrections were made for barometric pressure. The 10 volt excitation current was supplied by a 12 volt auto battery, variable resistor, and a ± 1/4% voltmeter.

Prior to assembly of the boiling apparatus, the volume of the condensate collection reservoir between the radiation sources was carefully measured. The volume at room temperature was found to be 32.99 cc.

The chromel-alumel thermocouples in the boiling vessel used to measure the boiling and condensed rubidium temperatures were calibrated against a National Bureau of Standards standardized platinum-platinum +13% rhodium thermocouple.

The gamma energy spectrum seen by the gamma detector was determined so that during operation the single channel analyzer could be set to count only primary uncollided gammas. This increased the sensitivity of the condensate level measurement.

(2) Rubidium Loading

About 2.5 pounds of rubidium was placed in a special glass flask, which was then sealed under vacuum. A helium leak-tight, bellows type, liquid metal valve was fitted to the bottom of the flask by a Kovar glass to metal seal.

This loading flask was then connected to the fill line on the boiling vessel and a vacuum was drawn on the boiling vessel through the vacuum line. All welds and fittings were helium leak tested and a vacuum of about 1×10^{-5} microns was achieved. Following the leak testing, the valve to the rubidium loader was opened. With the vessel heated to about 140°F, the rubidium was melted and allowed to flow into the vessel.

The technique developed to seal off the fill and vacuum lines consisted of first flattening a section of the tubing with a special hydraulic press. Then, while the hydraulic press was held in place on the tubing, the tube was quickly cut and seal welded. Preliminary tests made in development of this method indicated that the seal was helium leak tight throughout the procedure.

(3) Test Runs

During each of three test runs it was found that the vacuum in the containment chamber could be held between 1×10^{-5} and 1×10^{-4} microns. It was also found that the net heat loss from the boiling vessel to the surroundings could be held below 5 watts by proper adjustment of the radiation shield guard heater.

(a) Test Run #1 (March 21, 1961)

In Test Run #1, an orderly increase in boiler temperature was made to 1000°F. It was found that the system temperature control was stable and easy to maintain. The boiler temperature was then raised to 1200°F. Before system equlibrium could be achieved at 1200°F, the gamma count rate signal was lost and the run was terminated. No test data was obtained in this run.

Upon inspection after cool-down, it was found that sections of the gamma detector high voltage and signal coaxial cables had melted, causing signal to shield shorts. In preparation for the next test run, these two coaxial cables were relocated to a cooler position in the vacuum chamber. The air cooling system to the pressure transducer was also modified to increase efficienty.

(b) Test Run #2 (March 29, 1961)

In Test Run #2, equilibrium boiler datum temperatures of 1000, 1200, 1300, 1400, and 1500°F were successively obtained. The vapor pressure was measured at each of these temperatures. Since no significant variations in the gamma read out could be detected during boiling, however, no measurement of the boiling rate could be obtained.

During the increase in temperature to 1600°F, the fuses in the boiler and shield heater power lines were lost and the run was terminated. The insulation on the power cables had melted, causing a short circuit. This was repaired by changing to ceramic insulators. During the shutdown, the cooling system for the gamma detector and the pressure transducer was changed to use water instead of air since difficulty was encountered in maintaining sufficient air cooling to these components at high boiler temperatures.

(c) Test Run #3 (April 5, 1961

In Test Run #3 successive equilibrium temperatures up to 1800°F were obtained. Vapor pressure was measured over the range 1000 to 1800°F during the heat-up and the subsequent cool-down. As in Test Run #2, however, no measurements of the boiling rate could be made. Therefore, no direct measurements of latent heat of vaporization were obtained.

Analysis of the test data after this run indicated that the siphon action on the condensate reservoir was operating. This could be concluded from the periodic temperature cycling seen by the condensate thermocouple. It was finally concluded that the difficulty was that the 1/16 in. wide collimation regions at the top and bottom of the condensate reservoir were too narrow. This resulted in over collimation of the cobalt-60 gamma beam and the change in count rate caused by the rising liquid rubidium level was not significant as compared with random count rate variations (less than 5% change).

During the modification of this apparatus in preparation for similar measurements on cesium, the gamma collimation regions in the condensate reservoir will be increased to a width of 1/4 in. It is expected that this change will allow the direct measurement of latent heat of vaporization.

4. Measurement of Rubidium Specific Heat

The measurement of specific heat of liquid rubidium was planned to be made using the rubidium corrosion test loop. The technique consisted of a heat balance on the boiling heater section of the loop (no boiling would be allowed during this measurement).

The rate of heat loss from the boiling heater section of the loop was first determined vs temperature. This was done by setting the inlet temperature to the boiler section to a desired value and then putting just enough power into the boiling section heater to make the outlet temperature equal to inlet temperature. Thus, the power required to maintain a zero temperature drop was equal to the heat loss at this temperature. In this manner the heat loss from the boiling section of the loop was determined over the temperature range of interest.

Following this heat loss calibration the specific heat was to be determined. This was done by putting enough power in the boiling section of the loop to create a positive ΔT across the section. The specific heat at the mean temperature in the boiling section could then be calculated by the following formula:

$$C_p = \frac{\left[\begin{array}{l}\text{Boiling heater power}\\ \text{required to produce } \Delta T\end{array}\right] - \left[\begin{array}{l}\text{Boiling section heat}\\ \text{loss integrated over } \Delta T\end{array}\right]}{\left[\Delta T\right]\left[\text{Flow rate}\right]} \frac{\text{Btu}}{\text{lb}^{\circ}\text{F}}$$

During initial operation of the stainless steel corrosion test loop specific heat measurements were made between 600 and 1000°F using this method. Data obtained were found to be erratic; it was believed that poor thermocouple arrangement was the major cause. In

addition, it was thought that heater power feedback through the grounded thermocouples and poor knowledge of flow rate also contributed.

The thermocouples were changed to an ungrounded type and mounted in a heat sink block to the columbium corrosion test loop. It was hoped that this modification would reduce the major cause of erratic results. However, erratic results were again obtained when the measurement was attempted on the columbium loop.

The specific heat values which were obtained are shown in Figure C-13. Also included in this figure are the specific heat values extrapolated by Weatherford[1] and Kelly[4] from room temperature data. As can be seen, the AGN measured specific heat values are as much as a factor of 1.5 to 2 higher than the values predicted by Weatherford and Kelley.

The spread of the measured data points at 1000°F is of particular interest. These points were run at a number of different ΔT values and flow rates. Examination of this data indicated that regardless of the ΔT which was used, the data spread was primarily a function of the flow rate. Figure C-14 was prepared to show the effect of flow rate on the measured C_p. In this figure, the measured C_p divided by the predicted C_p is plotted against the flow rate. Assuming the predicted C_p values are correct, this plot indicates that the flow meter reading was lower than the actual flow. It can also be concluded that the flow meter reading accuracy varies from as much as 50% low at a flow rate of 0.2 gpm to about 20% low at a flow rate of 0.7 apm.

Based on this evaluation, it has been concluded that poor knowledge of loop flow rate was the major factor in the inability to accurately measure C_p on the corrosion test loops. It has been further concluded that the liquid metal wetting characteristics in the flow meter and the electrical conductivity of the system are the basic knowns in the accurate interpretation of the EM flow meter output signal.

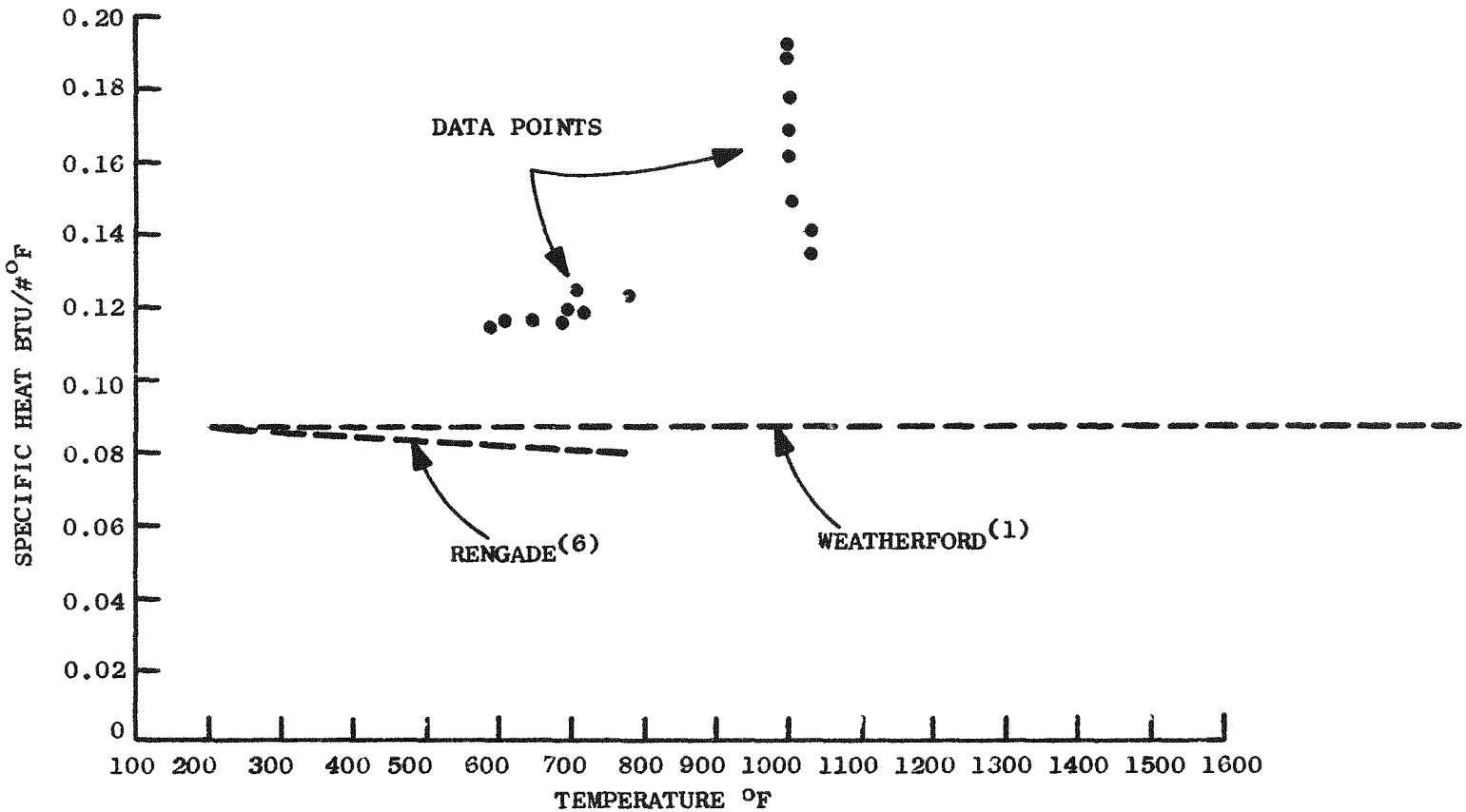


FIGURE C-13. SPECIFIC HEAT OF RUBIDIUM VERSUS TEMPERATURE

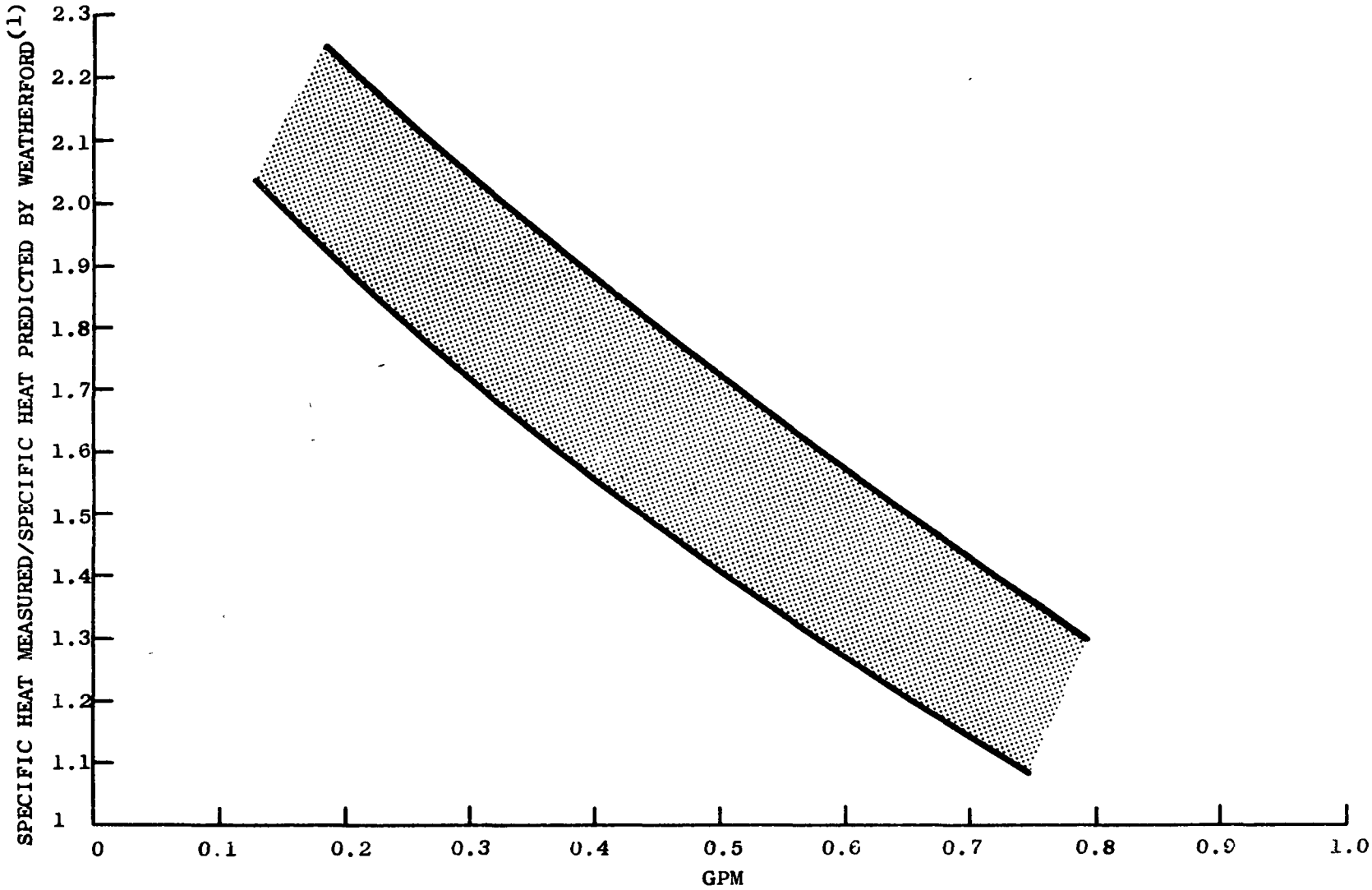


FIGURE C-14. EFFECT OF LOOP FLOW RATE ON SPECIFIC HEAT MEASUREMENT

5. Discussion and Presentation of Results

The thermodynamic properties for the saturated phases of rubidium are presented in Table VII. The density of the liquid and the vapor pressure were obtained by direct measurement while the remainder of the properties were calculated. The method used in the determination of each of these properties is discussed below. The nomenclature used in the following discussion is defined in Table V. The chemical analysis for the rubidium which was used in the determination of density and vapor pressure is given in Table VIII.

a. Density and Specific Gravity of the Liquid

The eight measured density points and the melting point have been plotted and are shown in Figure C-15. It was assumed that the density followed a quadratic equation of the form:

$$d = a + bt + ct^2$$

The following equation was then obtained using the least square analysis method:

$$d = 1.5005 - 2.624 \times 10^{-4} t + 3.94 \times 10^{-8} t^2 \qquad (1)$$

where

$$t = {}^{o}F$$

The accuracy was estimated to be $\pm$ 0.005 gm/cm^3 over the measured range of 102° to 1300°F. The accuracy of this equation between 1300° and 1800°F is estimated to be $\pm$ 0.01 gm/cm^3.

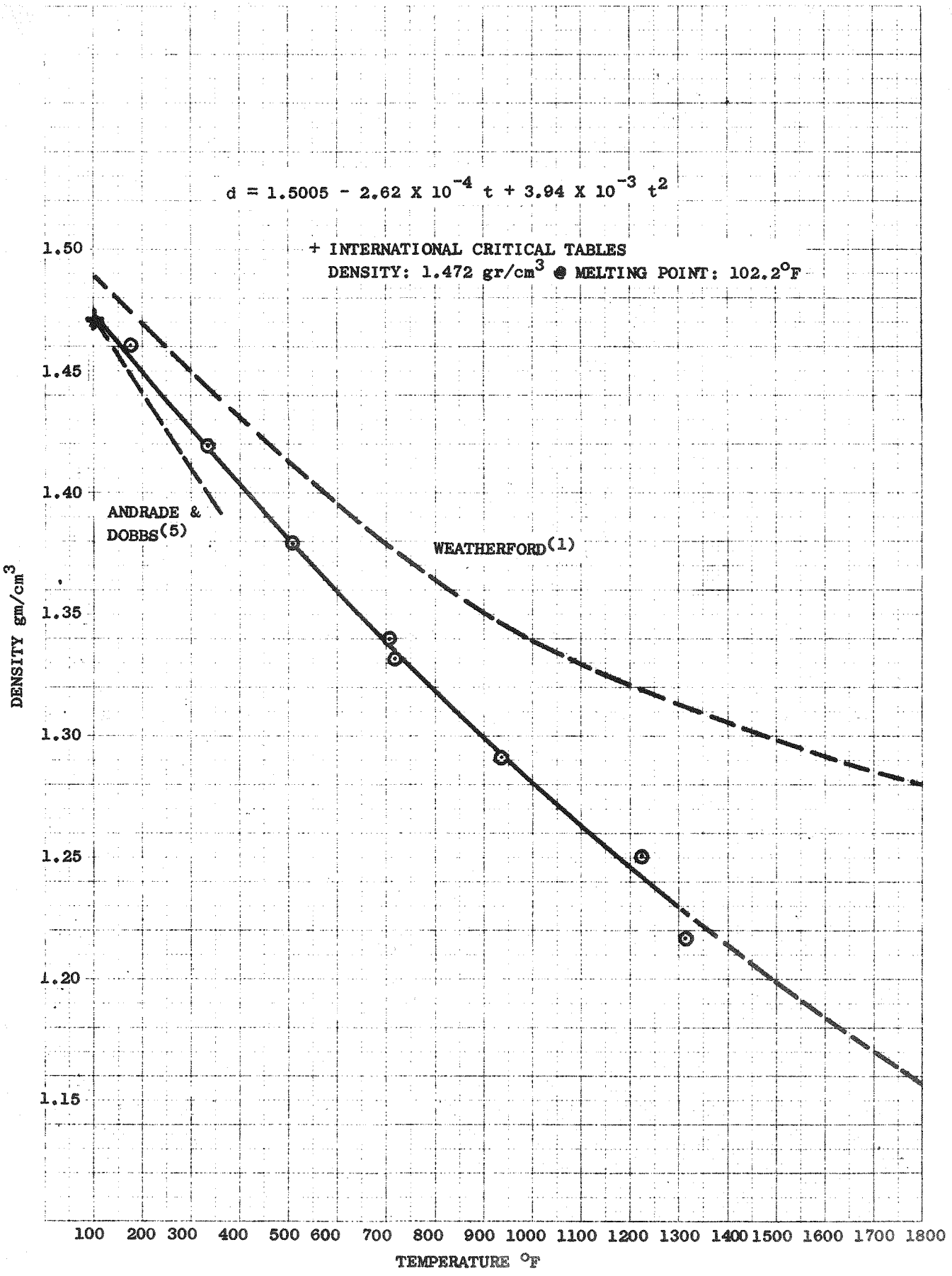


FIGURE C-15. DENSITY OF LIQUID RUBIDIUM

TABLE V

T	temperature, ^{o}R or ^{o}K as noted
t	temperature, ^{o}C or ^{o}F as noted
d	density, gm/cm^3
v_f	specific volume of the liquid, ft^3/lb
v_g	specific volume of the vapor, ft^3/lb
h_f	enthalpy of saturated liquid, Btu/lb
H_{fg}	enthalpy of vaporization, Btu/lb
h_g	enthalpy of saturated vapor, Btu/lb
R_o	universal gas constant, 1545 ft lb/mole^{o}R
M	molecular weight of the mixed vapor phase
p_1	partial pressure of monoatomic vapor, atmosphere
p_2	partial pressure of diatomic vapor, atmosphere
P	total pressure, atmosphere
K_p	equilibrium constant of dimerization
x_1	mole fraction of Rb_1 in the vapor phase
x_2	mole fraction of Rb_2 in the vapor phase
c_{p_f}	specific heat of the liquid, Btu/lb^{o}F
c_{p_s}	specific heat of the solid, Btu/lb^{o}F
ΔH_m	heat of fusion, Btu/lb
S_f	entropy of saturated liquid, Btu/lb-^{o}R
S_{fg}	entropy of vaporization, Btu/lb-^{o}R
S_g	entropy of saturated vapor, Btu/lb-^{o}R

The results were compared to the data reported in the literature. Andrade and Dobbs[5] in analyzing the data of Hackspill[7] and those of Rengade[6] gave values up to 428°F. These values, as shown in Figure C-16, are within 0.5% of the values which were measured by AGN. On the other hand, the density values predicted by Weatherford[1] are somewhat higher than those measured by AGN. It should be noted the AGN density curve passes through the melting point density given in the International Critical Tables.[3]

The specific volume of the liquid (v_f) was derived from the density equation and expressed in ft^3/lb as follows:

$$v_f = \frac{1}{d \times 62.43} \qquad (2)$$

b. Vapor Pressure Versus Temperature

The vapor pressure of rubidium was measured from 1000 to 1800°F. Sixty vapor pressure points over this temperature range were recorded and used to derive the "best" curve to fit the data. This curve is shown on Figure C-16. Of the many equations that can be used to describe such a curve, two forms have been widely used for the vapor pressure of the liquid metals.

(1) The first one of these equations is derived from the Clausius-Calpeyron equation and makes the assumption that the volume of liquid can be neglected in comparison to that of the vapor. It is also assumed that the vapor is an ideal gas and that the latent heat of vaporization is independent of temperature. This results in a linear relationship between log P and $\frac{1}{T}$.

$$\log P = \frac{A}{T} + B$$

(2) The second type of equation that was considered also neglects the volume of the liquid and assumes the perfect gas law, but is assuming that the latent heat of vaporization is related to the temperature by the equation:

$$h_{fg} = (h_{fg})_{\rho} + aT$$

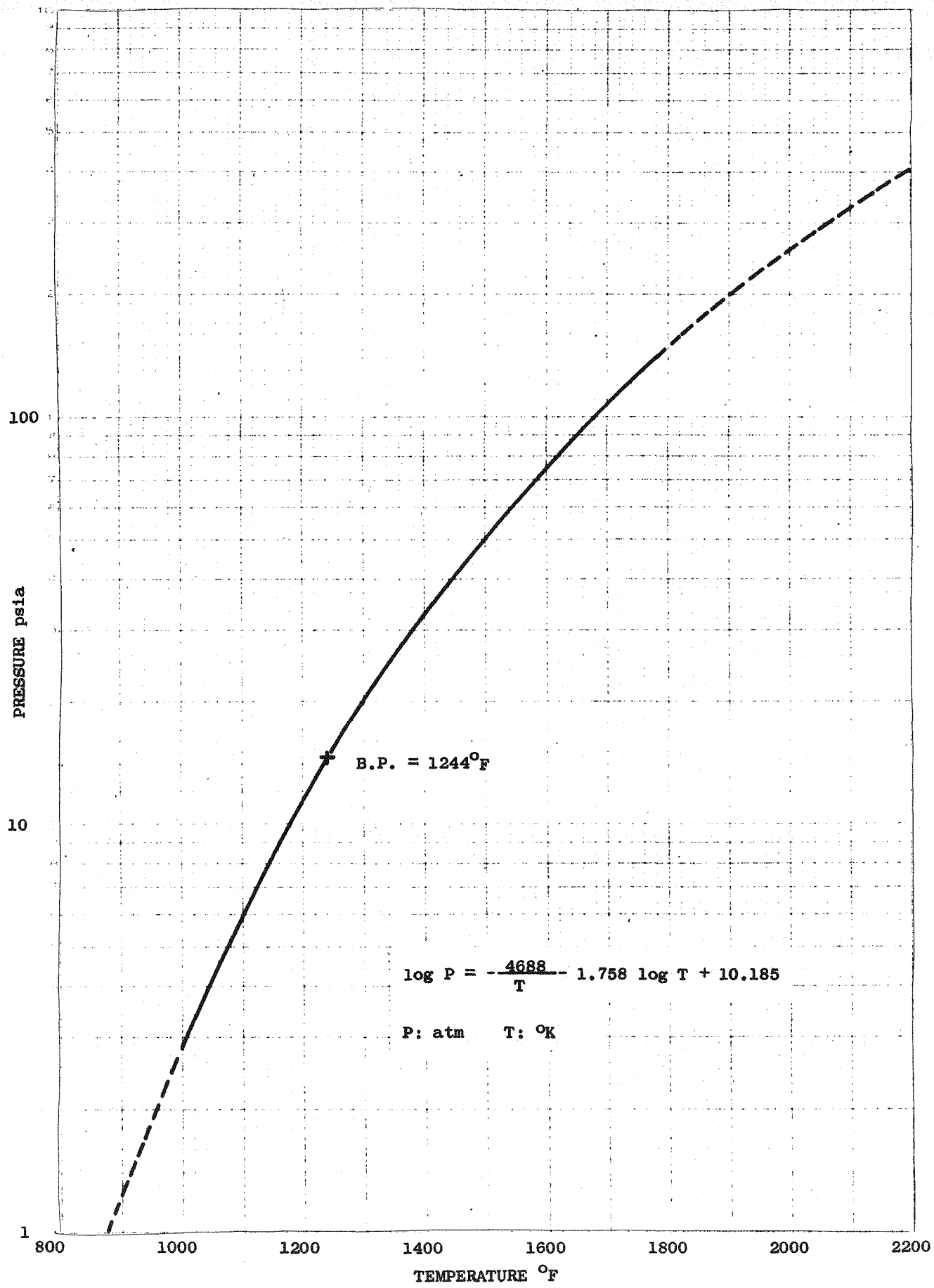


FIGURE C-16. SATURATED RUBIDIUM VAPOR PRESSURE

Integrating the Clausius-Clapeyron equation under these conditions gives the following empirical relationship, known as the Kirchhoff equation:

$$\log P = \frac{A}{T} + B \log T + C$$

Since it is well known that the latent heat of vaporization is highly dependent on temperature, it was decided to use the second form, and the following equation was obtained for the vapor pressure of saturated rubidium:

$$\log_{10} P = - \frac{4688}{T} - 1.758 \log_{10} T + 10.185 \qquad (3)$$

where

$$T = {}^{o}K.$$

This equation gives a normal boiling point of 1244°F at atmospheric pressure.

It was estimated that the accuracy of this curve is $\pm$ 0.2 psi or $\pm$ 2% whichever is higher. The accuracy of the pressure transducer and thermocouples and the effect of impurities (see Table VI) were considered in making this error analysis. The measured vapor pressure at a given temperature is slightly higher than that predicted by Weatherford.[1]

c. Vapor Dimerization and Molecular Weight

Rubidium vapor is essentially monoatomic at low temperature and dimerizes as the temperature increases:

$$Z\ Rb_1 \rightleftarrows Rb_2$$

The molecular weight of the vapor mixture was calculated using the following equations:

$$K_p = \frac{P_2}{(P_1)^2} \qquad (4)$$

TABLE VI

CHEMICAL ANALYSIS OF RUBIDIUM

A sample of the rubidium used for the density and vapor pressure measurements was analysed by the emission spectrographic method. The following metals content was found:

Rubidium	Greater than	98.86 %
Cesium		0.47
Potassium	less than	0.60
Lithium	" "	0.03
Sodium	" "	0.01
Lead		0.01
Iron		0.007
Tin		0.006
Aluminum		0.004
Beryllium		0.003
Nickel		0.003
Magnesium		0.0008
Silicon	less than	0.0007
Barium		0.0006
Calcium		0.0006
Copper		0.0003

where K_p is the equilibrium constant of dimerization whose value as a function of temperature is given by Stull and Sinke[8] and Evans.[9] The partial pressures of Rb_1 and Rb_2 vapors are represented by p, and p_2, respectively.

The Dalton Law of partial pressures was assumed to hold:

$$p_1 + p_2 = P = \text{total pressure} \tag{5}$$

by solving for p_1 in equations (4) and (5), the partial pressure of the monoatomic vapor Rb_1 was obtained:

$$p_1 = \frac{-1 + \sqrt{1 + 4 K_p P}}{2 K_p} \tag{6}$$

The mole fraction of the monoatomic vapor (x_1) was then calculated:

$$x_1 = \frac{p_1}{P} \tag{7}$$

and finally the net molecular weight of the Rb_1, Rb_2 vapor mixture was obtained as follows:

$$M = x_1 M_1 + 2 x_2 M_1$$

$$M = M_1 (2 - x_1) \tag{8}$$

where M_1 = molecular weight of Rb_1 = 85.48.

The calculated molecular weight of rubidium vapor vs temperature is given in Table VII.

d. Specific Heat of the Liquid

This property, needed for the calculation of the enthalpy and entropy, was correlated from various sources.

TABLE VII

CALCULATION OF RUBIDIUM VAPOR DIMERIZATION AND MOLECULAR WEIGHT

Temperature ^oF	Temperature ^oR	Vapor Molecular Weight M	Vapor Pressure atm. P	Equilibrium constant for K_p	Partial Pressure of Rb_1 P_1	Vapor Molecular Fraction of Rb_1 X_1
200	660	85.49	7.787×10^{-8}	1.995×10^{3}	7.786×10^{-8}	0.9998
400	860	85.67	4.599×10^{-5}	47.32	4.589×10^{-5}	0.9978
600	1060	86.39	2.263×10^{-3}	4.732	2.239×10^{-3}	0.9894
800	1260	87.79	3.063×10^{-2}	0.9772	2.976×10^{-2}	0.9716
900	1360	88.80	0.083	0.5129	0.07961	0.9592
950	1410	89.29	0.130	0.3890	0.1240	0.9538
1000	1460	89.80	0.195	0.2951	0.1848	0.9477
1050	1510	90.36	0.286	0.2239	0.2697	0.9430
1100	1560	90.92	0.408	0.1778	0.3818	0.9358
1150	1610	91.49	0.569	0.1413	0.5294	0.9304
1200	1660	92.00	0.766	0.1172	0.7073	0.9234
1250	1710	92.50	1.04	0.0944	0.9540	0.9173
1300	1760	93.07	1.35	0.0792	1.2302	0.9113
1350	1810	93.65	1.75	0.0667	1.5829	0.9045
1400	1860	94.21	2.23	0.0569	2.0018	0.8977
1450	1910	94.73	2.80	0.0487	2.4965	0.8916
1500	1960	95.22	3.46	0.0419	3.0666	0.8863
1550	2010	95.69	4.25	0.0362	3.7429	0.8807
1600	2060	96.15	5.14	0.0317	4.4985	0.8752
1650	2110	96.61	6.19	0.0281	5.3774	0.8687
1700	2160	97.08	7.33	0.0248	6.3348	0.8642
1750	2210	97.52	8.63	0.0221	7.4149	0.8592
1800	2260	97.92	10.08	0.0196	8.6227	0.8554

Rengade[6] gives a decreasing value in the temperature range from 39 to 126°C expressed by:

$$c_{p_f} = 0.0921 - 0.000026t \qquad (9)$$

where

$t = {}^{o}C$, and

$$c_{p_f} = \frac{Btu}{lb\,^{o}F}$$

On the other hand, such liquid metals as sodium and potassium exhibit a c_p curve of a quadratic form with a minimum around 1000°F. Stull and Sinke[8] give a constant value which is an average over the liquid phase equal to:

$$c_{p_f} = 7.50 \frac{cal}{mol - {}^{o}C} = 0.0877 \frac{Btu}{lb\,^{o}F}$$

Using the slope given by Rengade in the low range and the average value given by Stull and Sinke, the following equation was derived and is recommended for use in the temperature range from the melting point to 1800°F:

$$c_{p_f} = 6.3 \times 10^{-9}\, t^2 - 1.26 \times 10^{-5} t + 0.0923 \qquad (10)$$

where t is in degrees F

or

$$c_{p_f} = 6.3 \times 10^{-9} T^2 - 1.839 \times 10^{-5} T + 0.0994 \qquad (11)$$

where T is in degrees R.

e. Enthalpy

The reference level for all enthalpy calculations was taken as

$$298.15\,^{o}K \cong 537\,^{o}R$$

This reference level was chosen, rather than the melting point used by some authors, to be consistent with most reported thermodynamic data. This greatly facilitates the comparison of the properties of the various liquid metals.

(1) Enthalpy of the Saturated Liquid

The enthalpy of the saturated liquid was obtained from the following equation:

$$h_f = \int_{537}^{Tm} (c_{p_s})\, dT + (\Delta H_m) + \int_{Tm}^{T} (c_{p_f})\, dT \qquad (12)$$

The first term represents the enthalpy in the solid phase. The specific heat of the solid metal (c_{p_s}) from the reference level to the melting point ($Tm = 562^{o}R$) was averaged from Stull and Sinke data:

$$c_{p_s} = 0.088 \text{ BTU/lb}$$

The second term, or heat of fusion, was also obtained from Stull and Sinke and its value is given as:

$$\Delta H_m = 11.79 \frac{\text{BTU}}{\text{lb}} .$$

The third term, which represents the enthalpy of the saturated liquid, was calculated by replacing c_{p_f} by the equation (11) derived above:

(2) Latent Heat of Vaporization

The latent heat of vaporization (H_{fg}) was calculated using the Clausius-Clapeyron equation:

$$\frac{dP}{dT} = \frac{h_{fg}}{T(v_g - v_f)} \qquad (13)$$

$\frac{dP}{dT}$ is obtained from the measured vapor pressure relationship, Equation (3):

$$\frac{dP}{dT} = \frac{P}{T} \left(\frac{4688}{\log_{10} e \times T} - 1.758\right) \qquad (14)$$

The specific volume of the liquid (v_f) is neglected here, and for the specific volume of the vapor (v_g), the perfect gas law is substituted in the equation:

$$v_g = \frac{R_o T}{MP} \tag{15}$$

Substituting into equation (13) the latent heat of vaporization is given by:

$$h_{fg} = 1.987 \frac{T_{^oR}}{M} \left(\frac{19430}{T_{^oR}}\right) - 1.758 \frac{Btu}{lb} \tag{16}$$

(3) Enthalpy of the Saturated Vapor

The enthalpy of the saturated vapor was calculated from the equation:

$$h_g = h_f + h_{fg} \tag{17}$$

f. Entropy

As for enthalpy, the reference level chosen for all entropy values was taken at $298.15^oK = 537^oR$.

(1) Entropy of the Saturated Liquid

The entropy change of the saturated liquid was obtained by the following relationship:

$$S_f = \int_{537}^{Tm} (c_{p_s}) \frac{dT}{T} + \frac{\Delta H_m}{T_m} + \int_{Tm}^{T} (c_{p_f}) \frac{dT}{T} \tag{18}$$

The first term is the entropy change from 537^oR to the melting point ($Tm = 562^oR$) is evaluated by taking an average value for the specific heat of the solid:

$$c_{p_s} = 0.088 \frac{Btu}{lb} .$$

The second term represents the entropy of fusion and is calculated using the value of Stull and Sinke, $\Delta H_m = 11.79 \frac{Btu}{lb}$ for the latent heat of fusion.

The third term, or change of entropy in the liquid phase, was calculated using the equation (11) for the specific heat of the liquid.

(2) Entropy of Vaporization

The entropy of vaporization was obtained from the equation:

$$S_{fg} = \frac{h_{fg}}{T} \frac{Btu}{lb\,^{o}R} \tag{19}$$

(3) Entropy of the Saturated Vapor

The entropy of the saturated vapor was calculated by the equation:

$$S_g = S_f + S_{fg} \tag{20}$$

TABLE VIII

THERMODYNAMIC PROPERTIES OF RUBIDIUM – SATURATED PHASES

Temp.	Pressure		Specific Vol. ft^3/lb		Enthalpy Btu/lb			Entropy Btu/lb – °R		
°F	atm	psia	v_f	v_g	h_f	h_{fg}	h_g	s_f	s_{fg}	s_g
77					0			0		
200			0.01105		23.02			0.040		
400			0.01143		40.84			0.063		
600			0.01180		58.35			0.081		
800			0.01217		75.67			0.096		
900	0.083	1.22	0.01236	122.	84.28	381.3	465.6	0.103	0.280	0.383
950	0.130	1.90	0.01245	81.	88.58	377.2	465.8	0.106	0.268	0.374
1000	0.195	2.87	0.01253	58.	92.88	373.1	466.0	0.109	0.256	0.365
1050	0.286	4.21	0.01263	42.5	97.18	368.9	466.1	0.112	0.244	0.356
1100	0.408	6.00	0.01271	31.1	101.48	364.7	466.2	0.144	0.234	0.348
1150	0.569	8.36	0.01280	26.6	105.78	360.5	466.3	0.117	0.224	0.341
1200	0.766	11.26	0.01289	17.0	110.09	356.6	466.7	0.120	0.215	0.335
1250	1.04	15.22	0.01298	13.02	114.41	352.8	467.2	0.123	0.206	0.329
1300	1.35	19.84	0.01307	10.05	118.73	348.8	467.5	0.126	0.198	0.324
1350	1.75	25.72	0.01315	8.00	123.06	344.7	467.8	0.128	0.190	0.318
1400	2.23	32.77	0.01323	6.93	127.40	340.8	468.2	0.130	0.183	0.313
1450	2.80	41.15	0.01332	5.25	131.76	337.1	468.9	0.132	0.176	0.308
1500	3.46	50.85	0.01339	4.30	136.13	333.6	469.7	0.135	0.170	0.305
1550	4.25	62.46	0.01348	3.56	140.52	330.1	470.6	0.137	0.164	0.301
1600	5.14	75.54	0.01356	2.98	144.92	326.7	471.6	0.139	0.159	0.298
1650	6.19	90.97	0.01364	2.55	149.34	323.3	472.6	0.141	0.153	0.294
1700	7.33	107.7	0.01371	2.22	153.79	320.0	473.8	0.143	0.148	0.291
1750	8.63	126.8	0.01378	1.92	158.25	316.7	475.0	0.145	0.143	0.288
1800	10.08	148.2	0.01386	1.68	162.74	313.7	476.4	0.147	0.139	0.286

REFERENCES

1. W. D. Weatherford, Jr., et al., "Properties of Inorganic Working Fluids and Coolant for Space Applications", Southwest Research Institute (December 1959), WADC Technical Report 59-598

2. Metals Handbook, American Society for Metals, 8th ed. (1961)

3. International Critical Tables, Vols. I, III, and V, Mc-Graw Hill (1933)

4. K. K. Kelley, U. S. Bureau of Mines, Bulletin 476 (1949)

5. E. N. da C. Andrade and Dobbs, The Viscosity of Liquid Lithium, Rubidium and Cesium, Journal of the Royal Society, 211-12 (1952)

6. E. Rengade, Bulletin de la Societé Chimique de France, 15, 130 (1914)

7. L. Hackspill, Recherches sur les Métaux Alcalins, Annales de Chimie et de Physique (8), 28-30, 613, (1913)

8. D. R. Stull and G. C. Sinke, Thermodynamic Properties of the Elements, American Chemical Society, Washington, D. C. (1956)

9. William H. Evans, Rosemary Jacobson, Thomas R. Munson, and Donald D. Wagman, Thermodynamic Properties of the Alkali Metals, Journal of Research of the National Bureau of Standards, 55, 83 (1955)

10. Samuel Glasstone, Textbook of Physical Chemistry, 2nd ed., Van Nostrand, p. 443 to 452 and 828 to 833 (1946)

11. B. F. Dodge, Chemical Engineering Thermodynamics, 1st ed., Mc-Graw Hill (1944)

12. W. D. Kingery, Properties Measurements at High Temperatures, John Wiley and Sons