MW

1

NASA CONTRACTOR REPORT

NASA CR-1276

NASA CR-1276

[redacted]

[redacted]

N69-19884

SOLUBILITY AND DIFFUSION OF OXYGEN IN TANTALUM

by M. Parkman, R. Pape, R. McRae,
D. Brayton, and L. Reed

Mary! Same as CR.
We have it.

Prepared by
VARIAN ASSOCIATES
San Carlos, Calif.
for Lewis Research Center

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION • WASHINGTON, D. C. • FEBRUARY 1969

NASA CR-1276

SOLUBILITY AND DIFFUSION OF OXYGEN IN TANTALUM

By M. Parkman, R. Pape, R. McRae,
D. Brayton, and L. Reed

Prepared under Contract No. NAS 3-7626 by
EIMAC
Varian Associates
San Carlos, Calif.

for Lewis Research Center

NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

For sale by the Clearinghouse for Federal Scientific and Technical Information
Springfield, Virginia 22151 – CFSTI price $3.00

Abstract

The solubility of oxygen in tantalum between 600° and 900°C was determined to be

$$\log(\text{ppm O}) = 4.130 - \frac{1279}{T°K}$$

using a resistivity technique. Carefully reproduced pulses of oxygen were admitted to a high vacuum quartz tube furnace containing a tantalum wire. During each pulse, oxygen was maintained in contact with the specimen for a standard reproduced time after which the oxygen was pumped out and the resistance measured. A plot of change in resistance from the starting value versus number of oxygen charges resulted in a linear curve followed by one or more parabolic curves. The end of the linear section was interpreted to be the phase boundary. Specimens were analyzed by neutron activation to relate change in resistance to oxygen content.

Changes in slope of the resistance versus temperature curve for Ta containing between 0.007% and 0.12% oxygen were found at about 450°C and 750°C. An anomoly in the curve of resistance versus temperature for oxygen-doped Ta was found at about 900°C. Both of these effects were found in the 2-phase region and appear to be associated with the oxides.

Preliminary solubility data were obtained with columbium and Ta-10W alloy.

A trial run was made to test a proposed method of measuring diffusion of oxygen in refractory metals.

FOREWORD

The research described herein, which was conducted by EIMAC, Division of Varian Associates, was done under NASA Contract NAS 3-7626. The Project Manager was Mr. Russell A. Lindberg, Space Power Systems Division, NASA-Lewis Research Center. The report was originally issued as EIMAC Report No. APO 68-1101.

Page Intentionally Left Blank

TABLE OF CONTENTS

LIST OF TABLES

Page Intentionally Left Blank

LIST OF FIGURES

I. INTRODUCTION

Space power systems and components made from refractory alloys must be tested for long periods on earth before being used in space. High vacuum systems are used to protect the alloys from air. Although vacuums of the order of 10^{-8} torr and higher are used, some oxygen-containing species remain in the chamber, or leak in, and can react with the alloys, altering their properties. Also, start up, or accidents, can permit a larger amount of impurities to contact the alloys.

Engineers need to know the rate that such impurities arrive at the alloy surface, react with it, and diffuse inward. They also need to know the effects of the impurities thus dissolved. The rate of diffusion of impurities and the capacity of the alloy to react with impurities are a function of the solubility. If the solubility and diffusion coefficients were known, then engineers could better estimate the consequences of exposure of power system components to given environmental conditions.

This program is a part of the attack on the overall problem and is to measure oxygen diffusion and solubility in commercially pure refractory metals and advanced alloys of interest for space power system components. The limits of oxygen solubility were to be determined commercially pure tantalum and columbium, and in the alloys Cb-1Zr, Ta-10W, FS-85 (Cb-28Ta-10.5W-0.9Zr), and T222 (Ta-9.8W-2.4Hf-0.01C), at five temperatures in the temperature range of 1000°F to 2400°F. This report is an account of activity to determine the solubility of O_2 in Ta and Cb. In addition, preliminary solubility runs were made with alloys. A run to evaluate the method of measuring the diffusion coefficient was also made.

Existing information on the solubility and diffusion of oxygen in refractory metals was summarized in several papers presented at the AGARD Conference on Refractory Metals held in Norway in 1963. The proceedings were published as a book.[1] Poor agreement exists between investigators of the solubility of O_2 in Cb and Ta. Reasons for this include the complexity of the systems, uncertainty as to the type of oxide in equilibrium in some cases, and differences in purities of the materials and qualities of test apparatus. The uncertainty of the analysis of O_2 in Ta and Cb may also have contributed, but it is not considered responsible for a major part of the confusion. Measurements have been largely confined to temperatures above 800°C where the solubilities reported are largely above 1 atomic per cent. Discrepancies are much larger than the available analytical accuracies.

The oxidation characteristics of Ta and Cb have been extensively studied.[1] A proper discussion of solubility should consider this information, because 1) the methods used to form metal-oxygen alloys are similar to the oxidation studies, and 2) the treatment of raw data from which solubility values are derived (from some methods) involves interpretation of what is essentially an oxidation process.

Various methods have been used to derive solubility data, including 1) metallography, microhardness, x-ray diffraction, and chemical analysis of alloys of varying composition so as to detect the first presence of a second phase; 2) internal friction (at low temperatures); and 3) the change in electrical resistance of an alloy at the phase boundary.

A method was proposed for this program wherein an attempt was made to minimize some of the problems of previous workers while being able to obtain data reasonably rapidly. An ultra high vacuum, ion-pumped apparatus was used to minimize contamination. The phase boundary was determined by the difference in electrical resistivity characteristics in the single and 2-phase regions.

Diffusion of O_2 in Ta and Cb has been calculated from oxidation rates, and from concentration gradients as determined by microhardness.[1] It was proposed herein to deduce the diffusion coefficient by observing the change in resistance with time as oxygen diffused along a wire.

II. EQUIPMENT AND EXPERIMENTAL PROCEDURES

A. Method of Approach

1. SOLUBILITY

Two procedures for phase boundary determination from resistivity measurements were considered at the start of the program:

1) Isothermal Pulse Technique

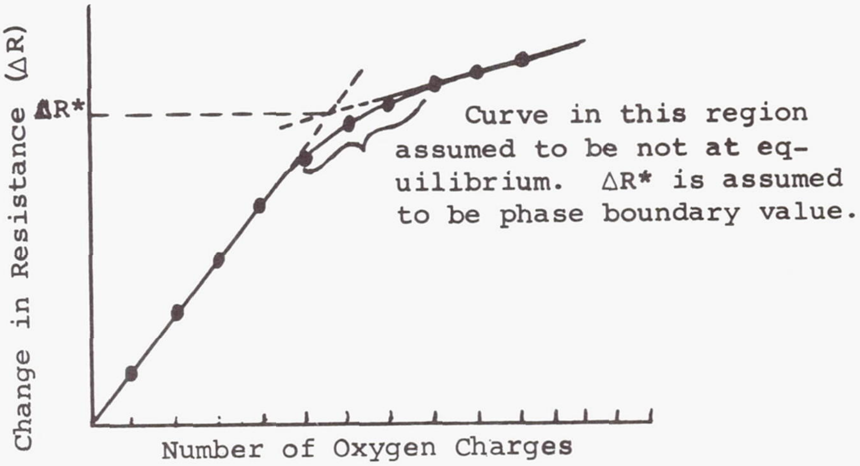
Measure room temperature resistance of wire specimen with the 4-wire Kelvin bridge method. Heat specimen to temperature in ultra high vacuum furnace and add carefully reproduced charges of O_2. Measure the resistance at the end of the charge after the specimen has reacted with the O_2. Add O_2 until the phase boundary is crossed. Detect the boundary from the difference in slopes of the resistance change versus number of oxygen charges data corresponding to the single phase and 2-phase regions (Figure 1a).

Conduct calibration runs with the same charge cycle to relate the oxygen content to the resistance change in the wire. This was to be done by charging the wire with oxygen to a given change in resistance (shown as α in Figure 1b after 5 charges), then analyzing the wire by neutron activation for oxygen content. A calibration curve was then to be constructed (Figure 1c) from analysis of calibration specimens charged with different numbers of oxygen charges. The calibration curve would relate change in resistance to oxygen content. It was found late in the program that the change in resistance had to be normalized by dividing by the room temperature starting resistance to eliminate the effect of geometric differences between samples. The oxygen content corresponding to the critical change in resistance ΔR^* at the intersection of the curve in Figure 1a was then to be obtained by interpolation from the calibration curve.

The method depended upon the applicability of Nordheim's rule and the constant absorption of oxygen during each pulse.

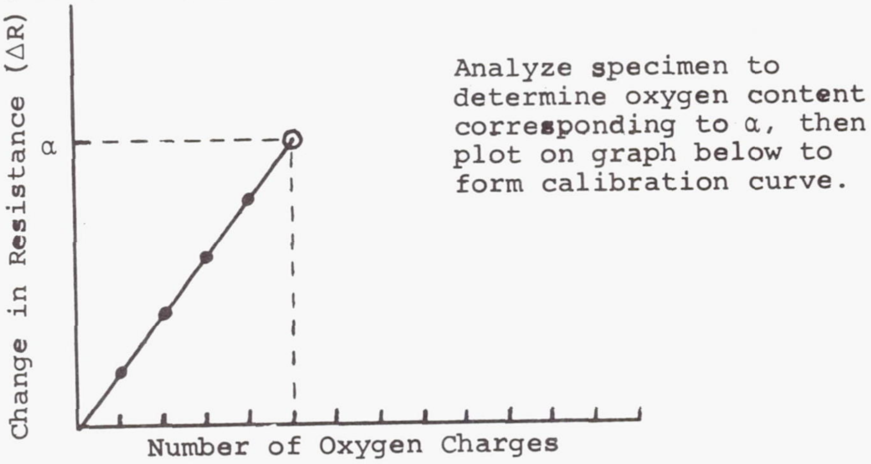
2) Isoconcentration Technique

Heat specimen to temperature in ultra high vacuum furnace. Expose specimen to oxygen until a given ΔR is reached. cool the specimen in steps, measuring resistance of the specimen at each step. The coefficient of resistance was thought to be slightly different above and below the solubility limit so that the phase boundary between the single phase region, and the 2-phase region could be



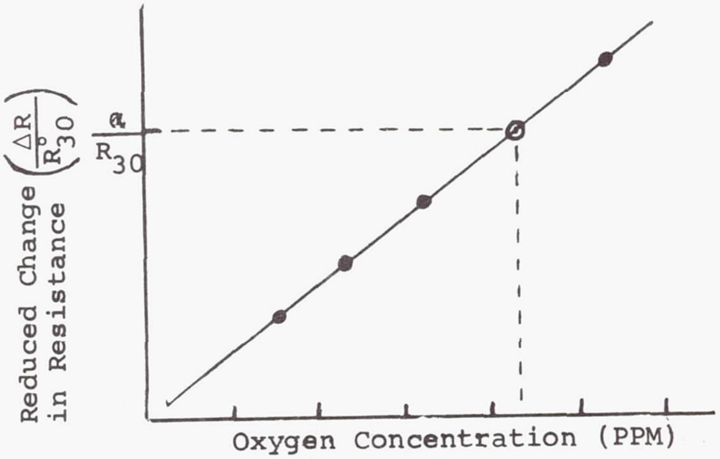
Curve in this region assumed to be not at equilibrium. ΔR* is assumed to be phase boundary value.

(a) Expected form of data from the Isothermal Pulse Technique



Analyze specimen to determine oxygen content corresponding to α, then plot on graph below to form calibration curve.

(b) Expected form of data from calibration run.



(c) Expected form of calibration curve.

FIGURE 1 - Expected Method of Obtaining Solubility Data with the Isothermal Pulse Technique.

determined by the intersection of the slopes. The specimens would be analyzed for O_2 after the run.

The method depended on a reasonably rapid precipitation process and different temperature coefficients of resistance in the two regions.

Experimental work was started using Technique 2) because this appeared to be the faster way to obtain data; however, no change in slope was detected during any trial run. Runs with commercially pure Ta and Cb were then made with Technique 1). A "reasonable looking" solubility curve was obtained for Ta. The data for Cb, however, could not be interpreted.* At this time, an extended shutdown of the project took place because of personnel changes. On restarting the program, it was decided to conduct a run with Ta using a third technique to verify the earlier data and to re-examine the isoconcentration technique over a wider temperature range. An outline of this third procedure follows:

3) Combined Isothermal Pulse, Isoconcentration Technique.

Measure resistance of specimen at room temperature and at 50°C intervals between 500°C and 950°C in the ultra high vacuum furnace. At 950°C, add one charge of oxygen. Measure resistance versus temperature between 950°C and 500°C. Repeat previous two steps, obtaining a series of R versus T curves, from which the phase boundary is determined from changes in slope as with Techniques 1) and 2). Changes in slope were found, but these apparently were associated with oxide transitions rather than the formation of a second phase. This method was therefore thought to be unsuitable for the main purpose of the program.

The interpretation of the Technique 3) data led to the re-examination of earlier runs with the result that some assumptions associated with the first two procedures were found to be erroneous. The 2 linear regions shown in Figure la were found actually to be a linear region followed by one or more parabolic regions as shown in Figure 2. This finding is similar to results from oxidation studies and suggests that a constant amount of oxygen was not absorbed in each charge as was required for procedure 1). In fact, the amount decreases in a manner controlled by the oxidation characteristics. This infers that the solubility was greatly exceeded in all runs. Independent verification of this was found in another run made with Technique 1). At this time the solubility limit was interpreted to occur at the start of the first parabolic region. Recalculation of

---

*They were interpreted at a later time, but found to be more inaccurate than desired.

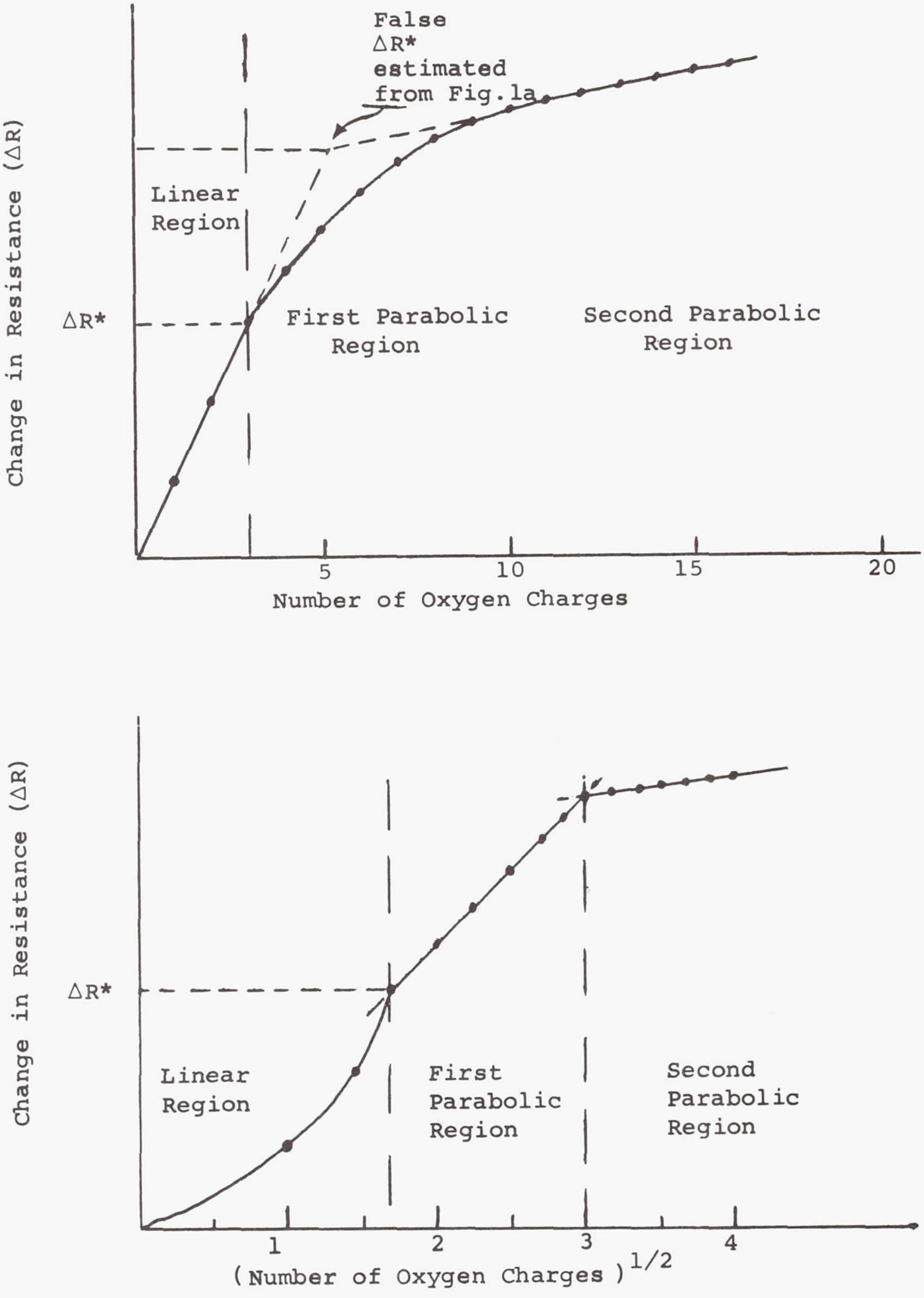


Figure 2 - Corrected Interpretation of Data from the Isothermal Pulse Technique

the data produced a "final" curve. Only the "final" curve, and calculations and interpretations thereof, are included in this report.

This final curve disagreed with the data of Gebhardt and Seghezzi[2], which we regarded as most reliable. A final run with Ta was made using the method of these investigators, as outlined below.

4) Isothermal, Isobaric Technique

Measure resistance of specimen at room temperature and at selected temperature in the ultra high vacuum furnace. While at selected temperature, adjust oxygen pressure to constant value and measure resistance versus time. A linear region followed by a parabolic region is obtained (Figure 3). The first departure from linearity is interpreted as the phase boundary. Results of our run agreed with an extrapolation of the data of Gebhardt and Seghezzi, but disagreed with the data from Technique 1).

Although four approaches to determining solubility from resistivity data were tried, ambiguous interpretation of these data left the best approach unproven. It was clear that independent verification of the phase or solid solution changes associated with resistivity changes required independent verification. Specimens were examined by metallography, and a preliminary diffusion experiment was conducted to obtain further evidence of the proper interpretation so that a best approach could be recommended. Results of these experiments leave our final interpretation in doubt as well as those of other workers. Further work is needed to support the proper interpretation of data and to select the best approach to obtaining the solubility.

2. DIFFUSION

Several procedures for measuring diffusion were considered. The one finally selected for trial was devised to make use of the method for measuring resistance. Leads were attached to a wire specimen as shown in Figure 4. The resistance of regions between D and G could be measured independently as in the solubility studies. The following procedure was used.

Measure starting resistance of all regions at room temperature and at elevated temperature in the ultra high vacuum furnace. With specimen at room temperature, and with O_2 in the furnace, pass current through Region A - B until the temperature of this region is above 1000°C. Hold until resistance of Region A - B has increased to value corresponding to the desired concentration

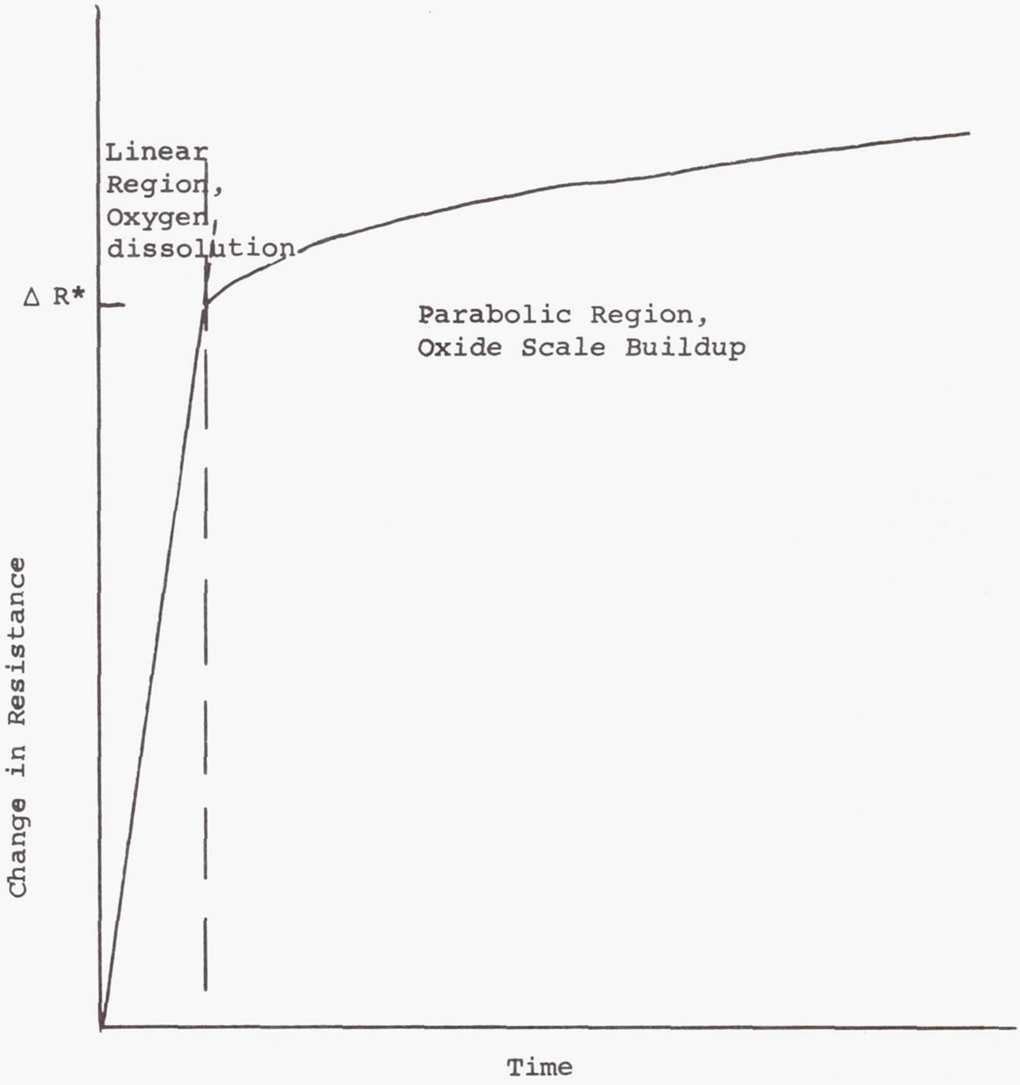


Figure 3 - Form of Data From the Isothermal, Isobaric Technique (after Gebhardt and Seghezzi[2])

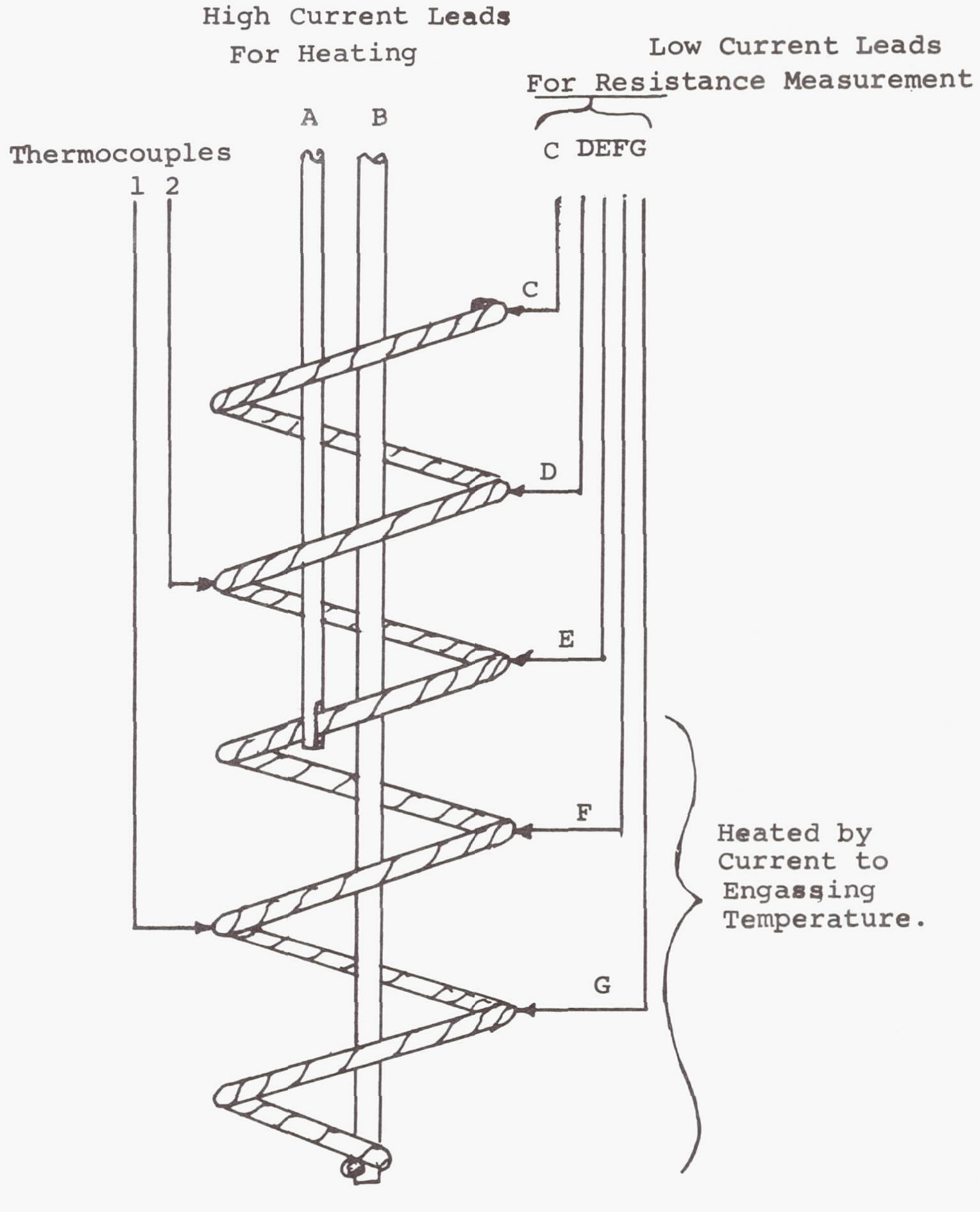


Figure 4 - Diffusion Specimen and Electrical Hookup.

couple between Regions A - B and A - C. Stop current and heat entire specimen to desired temperature with the furnace. Follow diffusion by changes in resistance of regions between D and G. Verify concentration gradients with a microhardness traverse along the wire after the run.

The one trial run showed the above procedure to be useful, but diffusion proved to be too slow to see any change in resistance. A microhardness traverse after the run showed the concentration gradient quite well, however.

B. Apparatus

The apparatus (Figures 5-8) consisted of a quartz tube furnace to contain the wire specimen and suitable auxilliary apparatus to measure resistance and control the atmosphere in the furnace. The auxilliary apparatus included an ultra-high vacuum, ion pumped vacuum system with a residual gas analyzer; the temperature measuring and control system; the resistance measuring system; and the oxygen feed system.

The investigation required measurements to be made to 1500°C. The existing quartz tube furnace was limited to about 950°C. It was used while a large, double pumped, alumina tube furnace capable of 1600°C was constructed (Figure 9). An oil diffusion pumped chamber outside the Triangle RR alumina tube (Figure 10) containing W hairpin resistance heaters (Figure 11) was capable of about 1 x 10^{-6} torr. This permitted much lower pressures inside the tube and protected the W heaters. The 1600°C furnace was checked out, but was not used. The 950°C system was used for all data reported herein and is described in the following sections.

1. VACUUM SYSTEM

A conventional ion-pumped system (Figure 5) was used having a base pressure in the 10^{-9} torr range. Rough pumping was accomplished with a liquid-nitrogen-cooled sorption pump. High vacuum was achieved with a Varian Vacion pump of 40 liters/sec capacity. A one liter/sec orifice was located above the ion pump to provide pumping speed measurements, if desired. A valved bypass was usually used during runs. Pressure was measured in the manifold above the limiting orifice with a Bayard-Alpert nude ionization gauge. The Diatron for the C.E.C. 21-612 residual gas analyzer was also located above the pump. A manifold was used to connect the pump, the furnace and a Millitorr ionization gauge. A Mosely chart recorder monitored the output of the Millitorr gauge. (At the time the photographs for Figures 6 and 7 were taken, the Millitorr gauge was mounted in the expansion volume of the O_2 feed system. Later, the gage was moved to the manifold, and thermocouple gages were installed on the O_2 feed system.)

The 12-in.-long quartz reaction tube was attached to the manifold bottom flange by means of a graded glass section three in. long and a Kovar tube. The specimen support assembly was attached to the top flange as described in Section II,C.

2. FURNACE AND TEMPERATURE CONTROL SYSTEM

The 1000°C resistance-heated furnace was mounted on tracks (Figure 8) below the manifold. The furnace could be raised to surround the quartz reaction tube. The furnace was 16 in. long, 6 in. diameter, and had a 1-1/4 in. bore. The temperature profile is described in Section II,D.

A Chromel-Alumel thermocouple at the middle of the hot zone outside the quartz reaction tube was used with a Leeds and Northrup Series 60 3-action C.A.T. control (Model C-1) to control the temperature and determine the nominal furnace temperature. The correction signal and a chart recording of temperature was supplied by a Leeds and Northrup Speedomax H A.Z.A.R. recorder. In practice, the control system was capable of holding the temperature within ±0.1°C.

3. OXYGEN FEED SYSTEM

The purpose of the oxygen feed system (Figure 5) was to provide a precisely reproducible quantity of oxygen for injection into the furnace volume at a controlled rate. This was done by filling the measured volume with O_2 to a known pressure and then admitting this to the furnace through a Granville-Phillips variable leak valve.

The O_2 supply was 99.999% research grade purchased from the Liquid Carbonic Division of General Dynamics. Analysis is shown in Table 1.

4. SPECIMEN MATERIALS AND GEOMETRY

All specimens were made from 0.020-in.-dia. wire obtained from commerical sources. Commercial purity was used as specified in Appendix B. Analysis is also reported in Appendix B. The following wire was obtained:

TABLE 1

ANALYSIS OF OXYGEN USED IN SOLUBILITY RUNS

IMPURITY	CONTENT, ppm
Nitrogen	<4
Hydrogen	<3
Water	1
CO	0.8
CO_2	0.2
Halogenated Hydrocarbon (probably trichlorethylene)	0.1

Liquid Carbonic Reagent Grade Oxygen.
Cylinder #3710. 99.999% O_2.

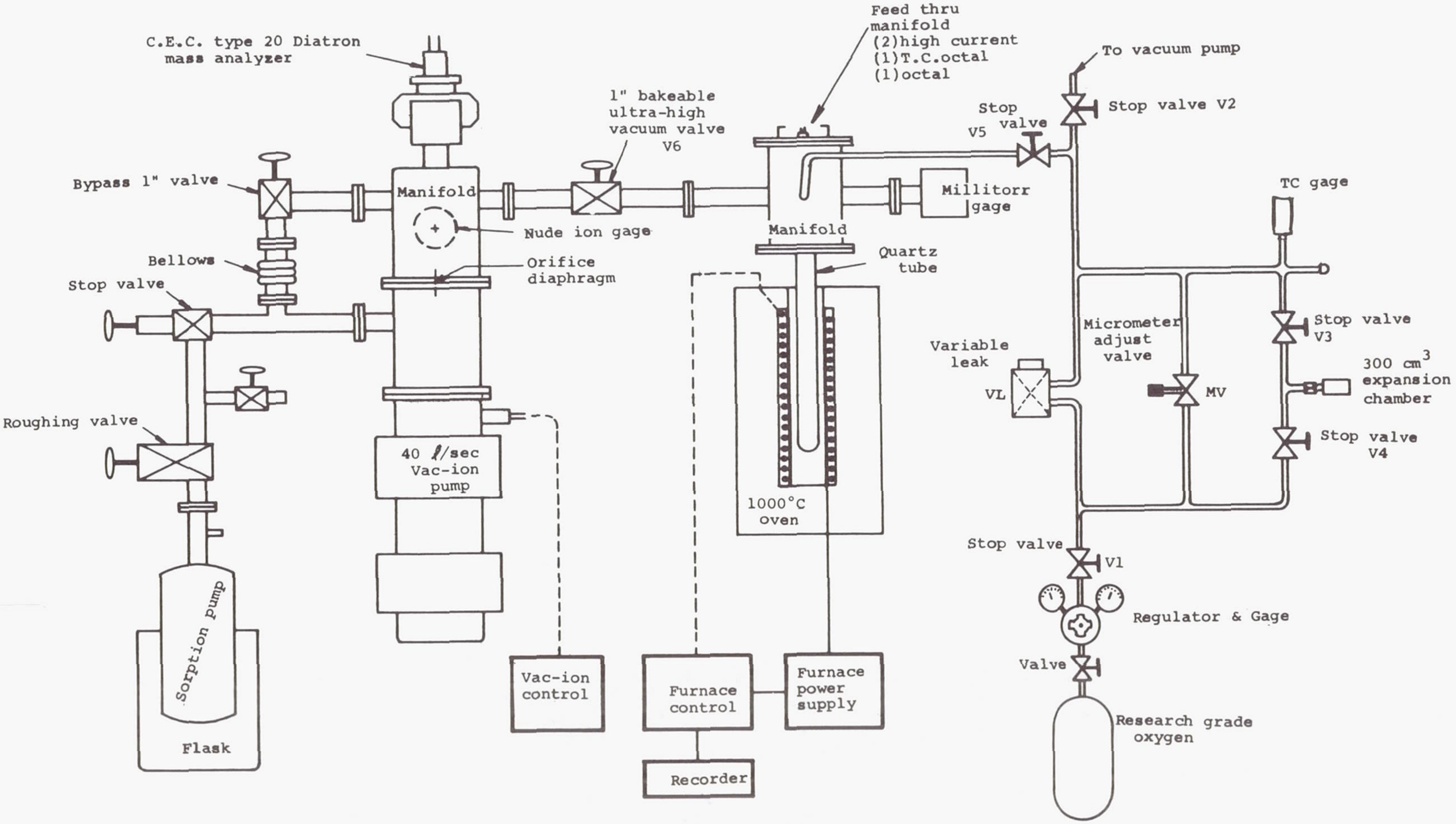


Figure 5 - Solubility Apparatus

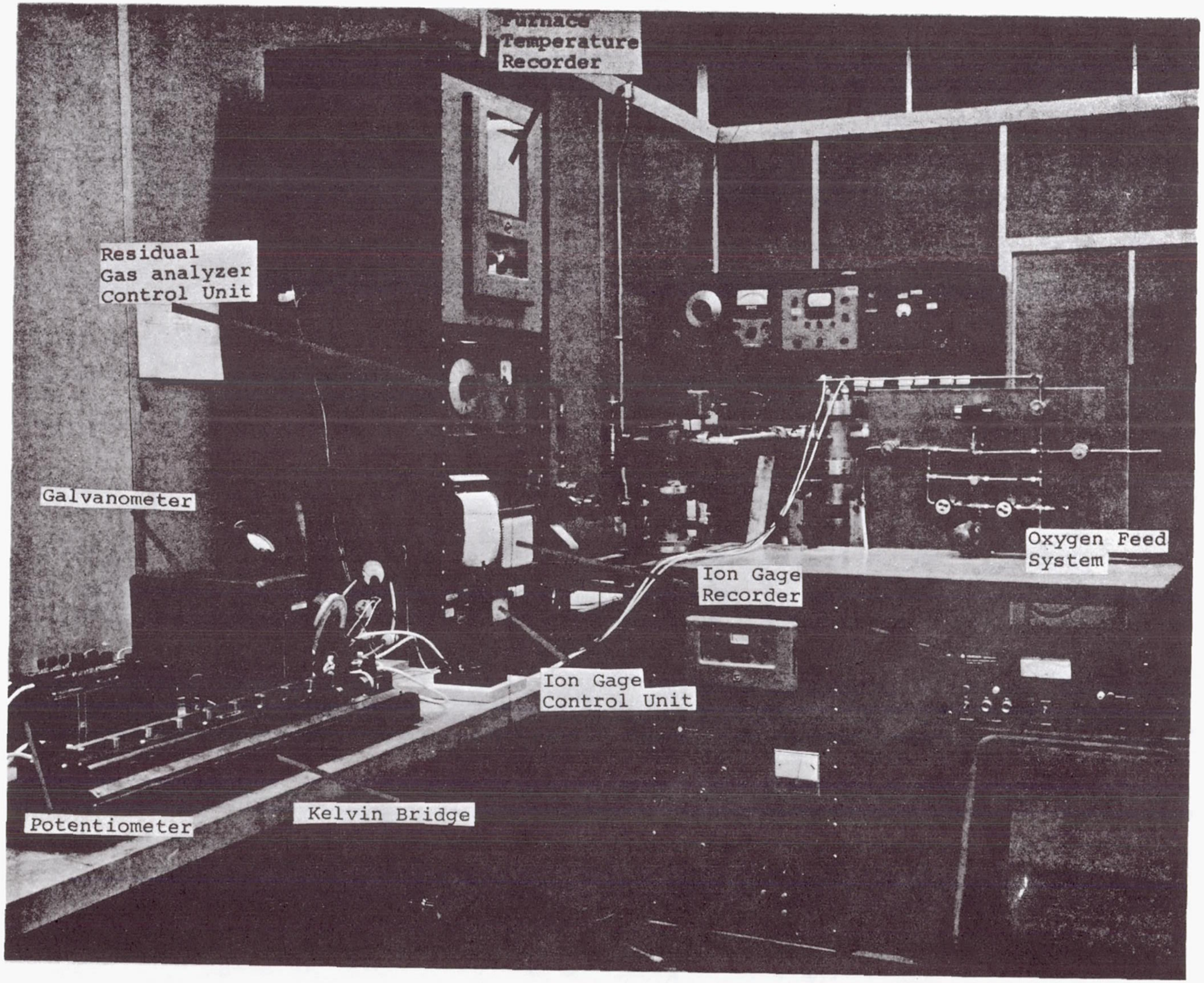


Figure 6 - Solubility Apparatus

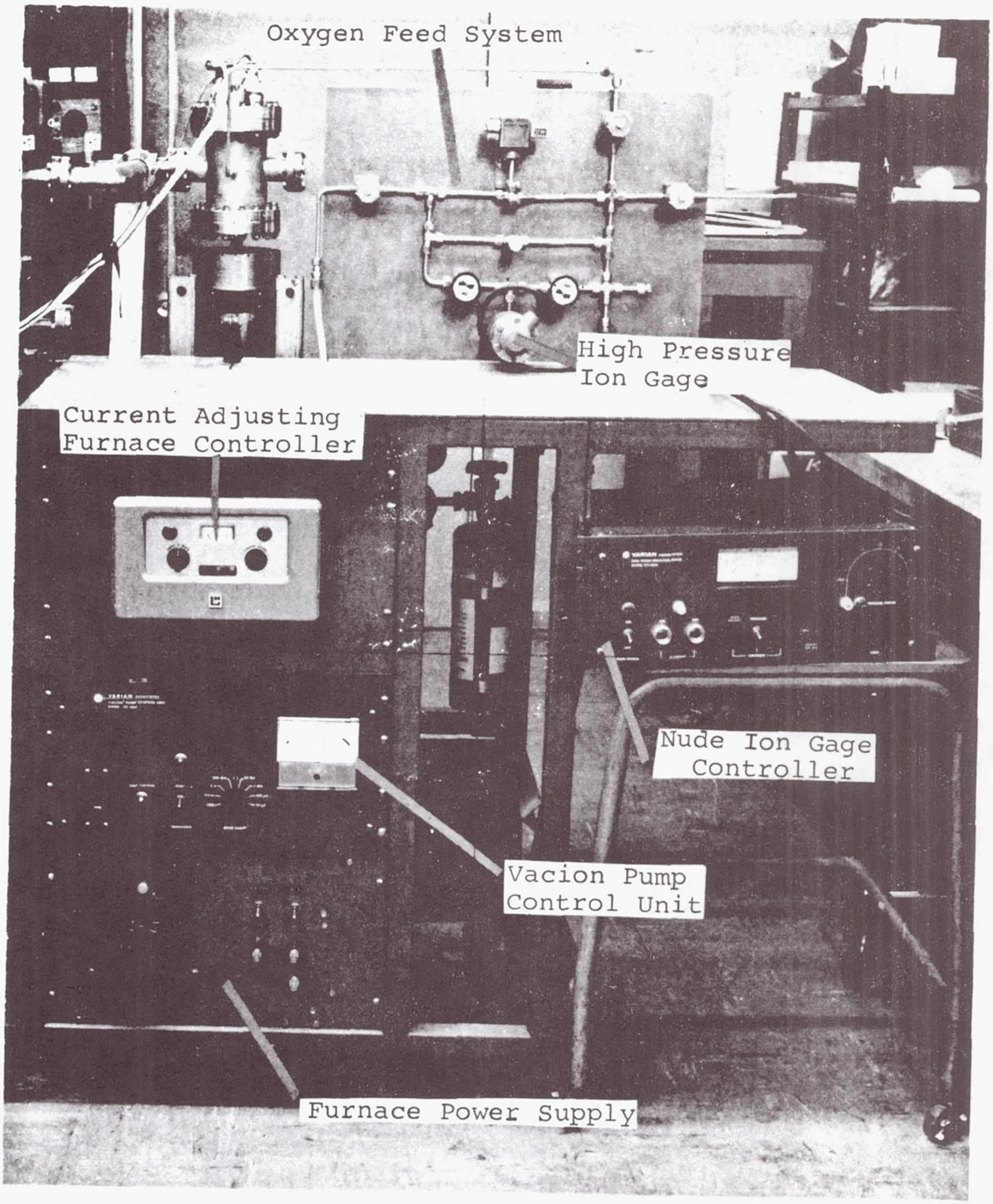

Oxygen Feed System
High Pressure Ion Gage
Current Adjusting Furnace Controller
Nude Ion Gage Controller
Vacion Pump Control Unit
Furnace Power Supply

Figure 7

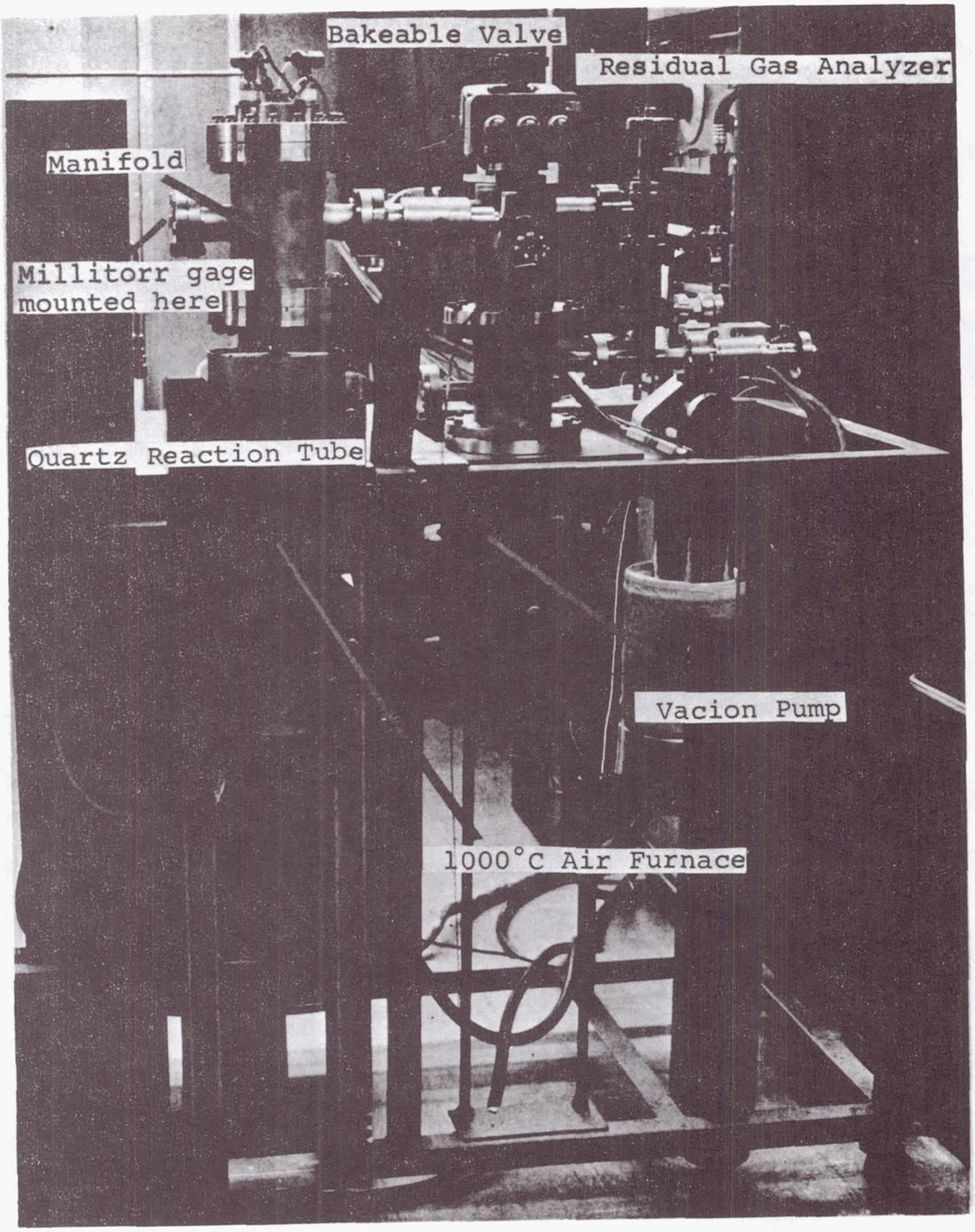

Bakeable Valve
Residual Gas Analyzer
Manifold
Millitorr gage mounted here
Quartz Reaction Tube
Vacion Pump
1000°C Air Furnace

Figure 8

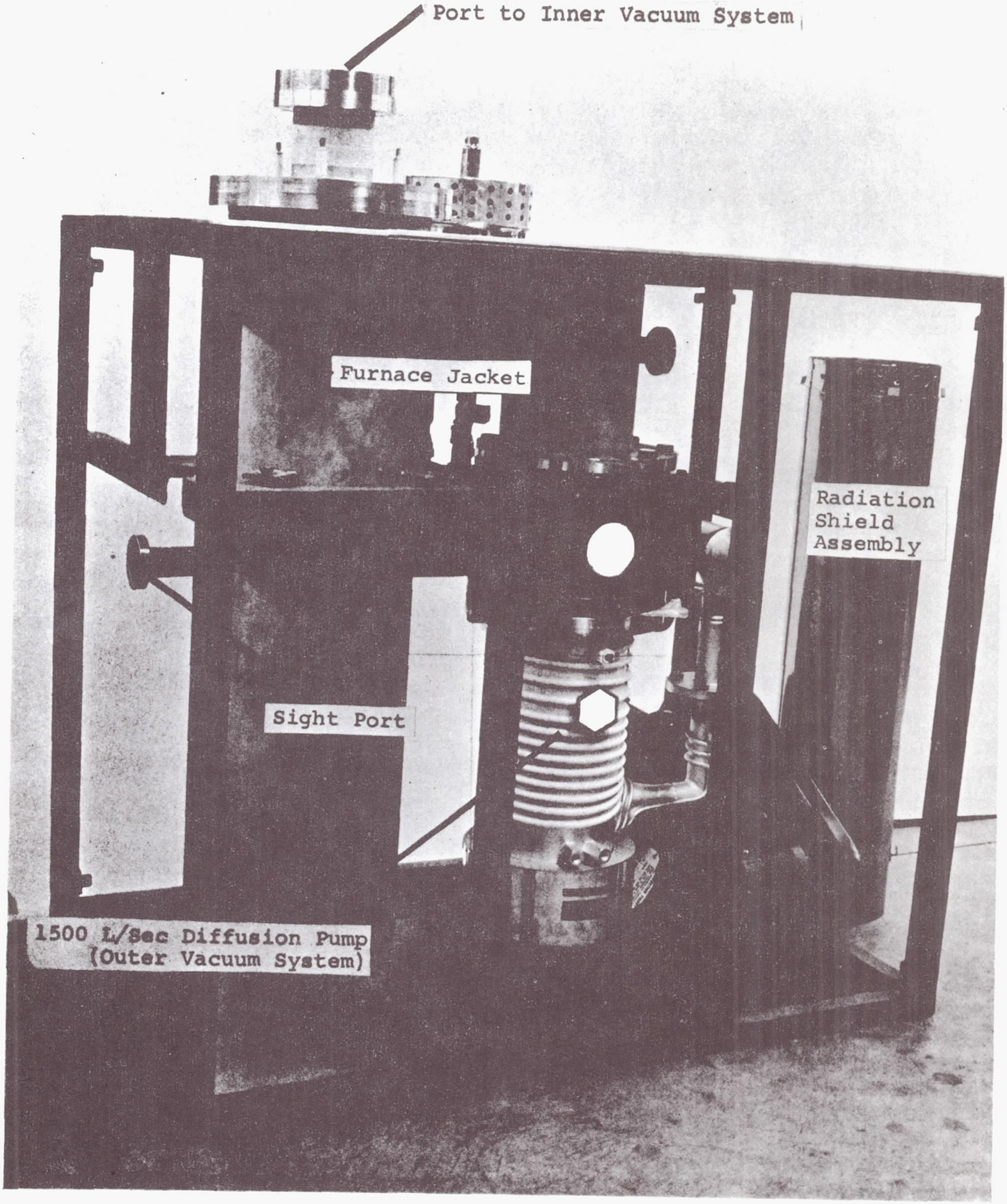


Figure 9 - 1600°C Double Vacuum Furnace

Figure 10 - Alumina Tube for 1600°F Double Vacuum Furnace

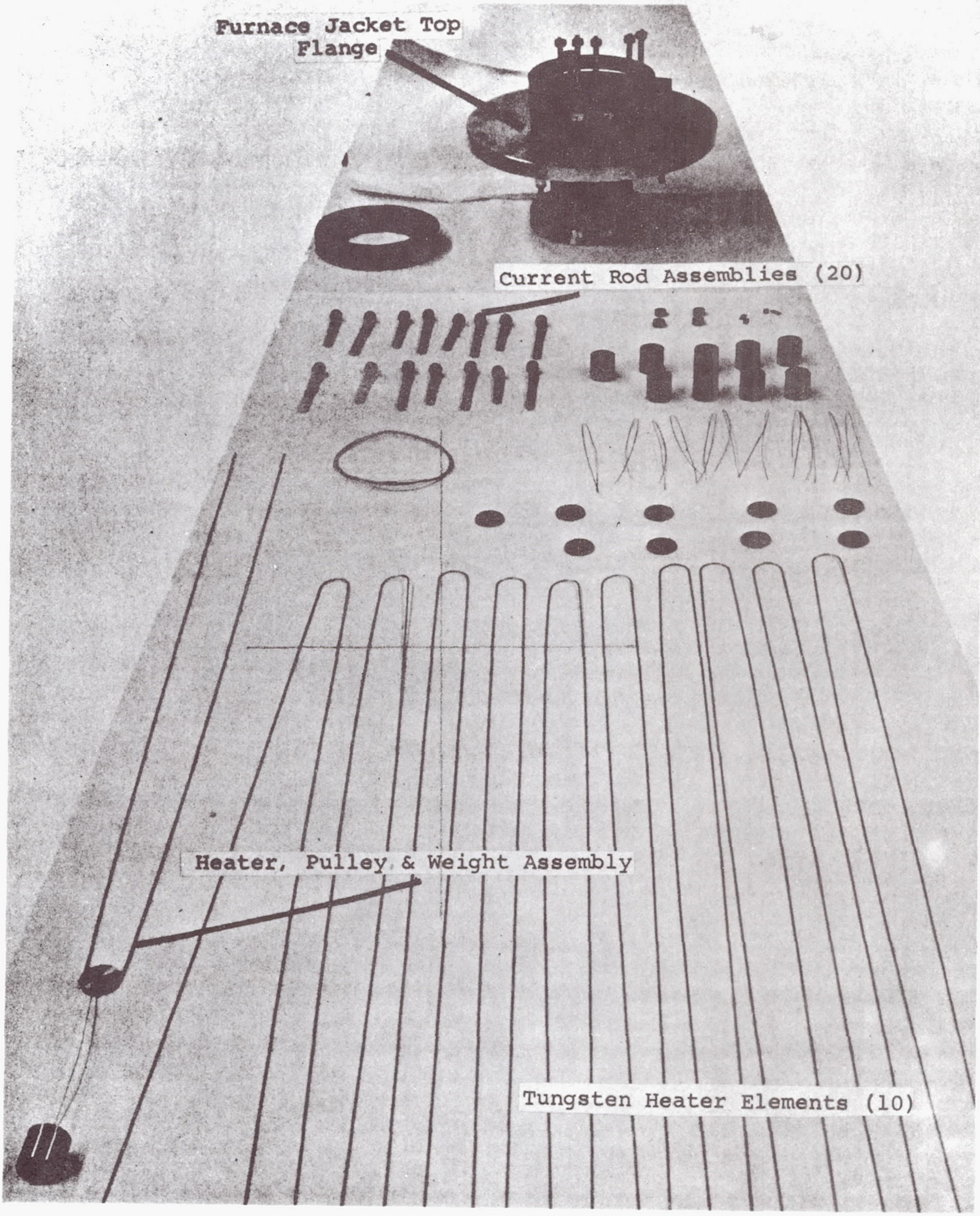


Figure 11 - Heater Elements for 1600°C Double Vacuum Furnace

Material	Quantity, ft.	Supplier
Tantalum	260	Fansteel
Tantalum - 10% Tungsten	225	"
T-222 Alloy	170	"
Columbium	200	"
Columbium - 1% Zirconium	215	"
FS-85 Alloy	200	"

a. Solubility Specimen

Three lengths of wire were braided together to form a specimen having about 0.2 ohm resistance at room temperature. The specimen was then coiled into a helix and spot welded under methanol to Mo rods as shown in Figure 12. Platinum sheet was placed between the specimen and rods to permit a ductile weld area. Two Ta leads for measuring voltage drop were spot welded to the top and bottom coils. The part of the specimen between these two leads was thus the length over which measurements were made. A Pt-Pt/10Rh thermocouple (5 mil wire) was attached to the center of the specimen by pinching the bead between a pair of the braided wires. This thermocouple was abandoned after Run Ta19 when it was found that the other furnace thermocouples had the same output.

The specimen, leads, and thermocouple were held in the top flange as shown in Figure 13. Ceramic-to-metal feedthroughs using the thermocouple wire were used to make connection to external leads.

Two additional thermocouples were located as shown in Figure 12, being freely suspended in the furnace volume. In general, readings from the three thermocouples were insignificantly different except above 900°C (see Section II,C,1 and tables of data, Appendix C).

b. Diffusion Specimen

A coil specimen (Figure 4) was made from two 0.020-in.-dia. Ta wires twisted together and coiled around a 0.7-in.-dia. mandrel. The center turn was flattened for spot welding to the longer Mo rod. The upper coil was attached to the shorter Mo rod, and the bottom turn attached to a single lead of 0.020-in.-dia. Ta wire which was insulated with quartz tubing.

The specimen was positioned as for the solubility specimen. The resistance leads were connected through an octal feedthrough to a rotary switch which allowed a selection of leads

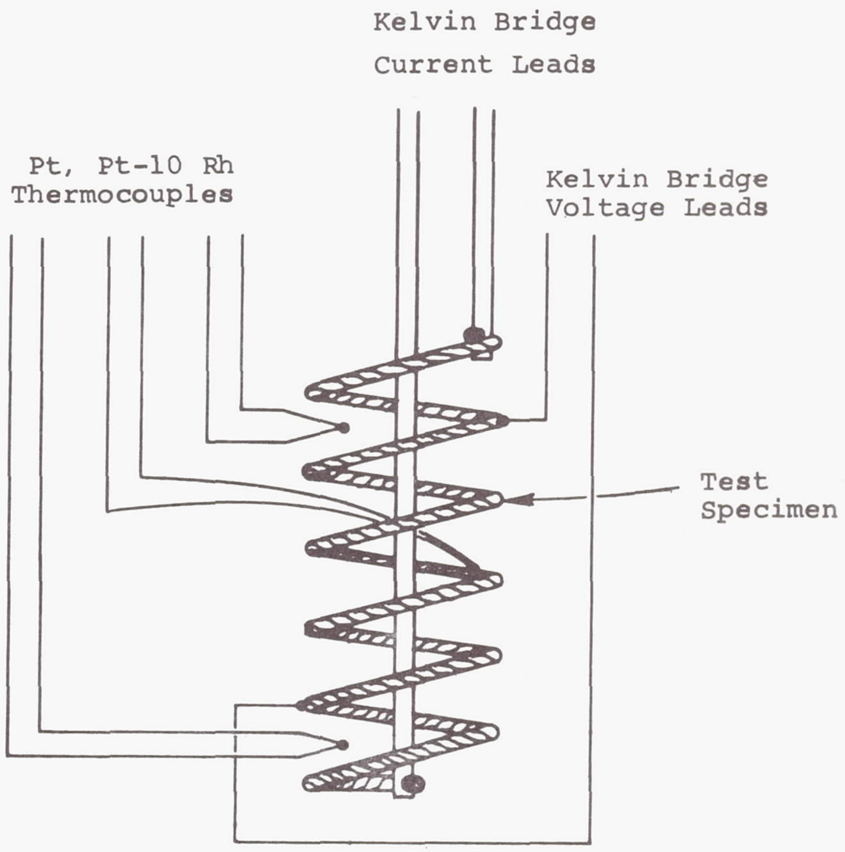


Figure 12 - Solubility Specimen

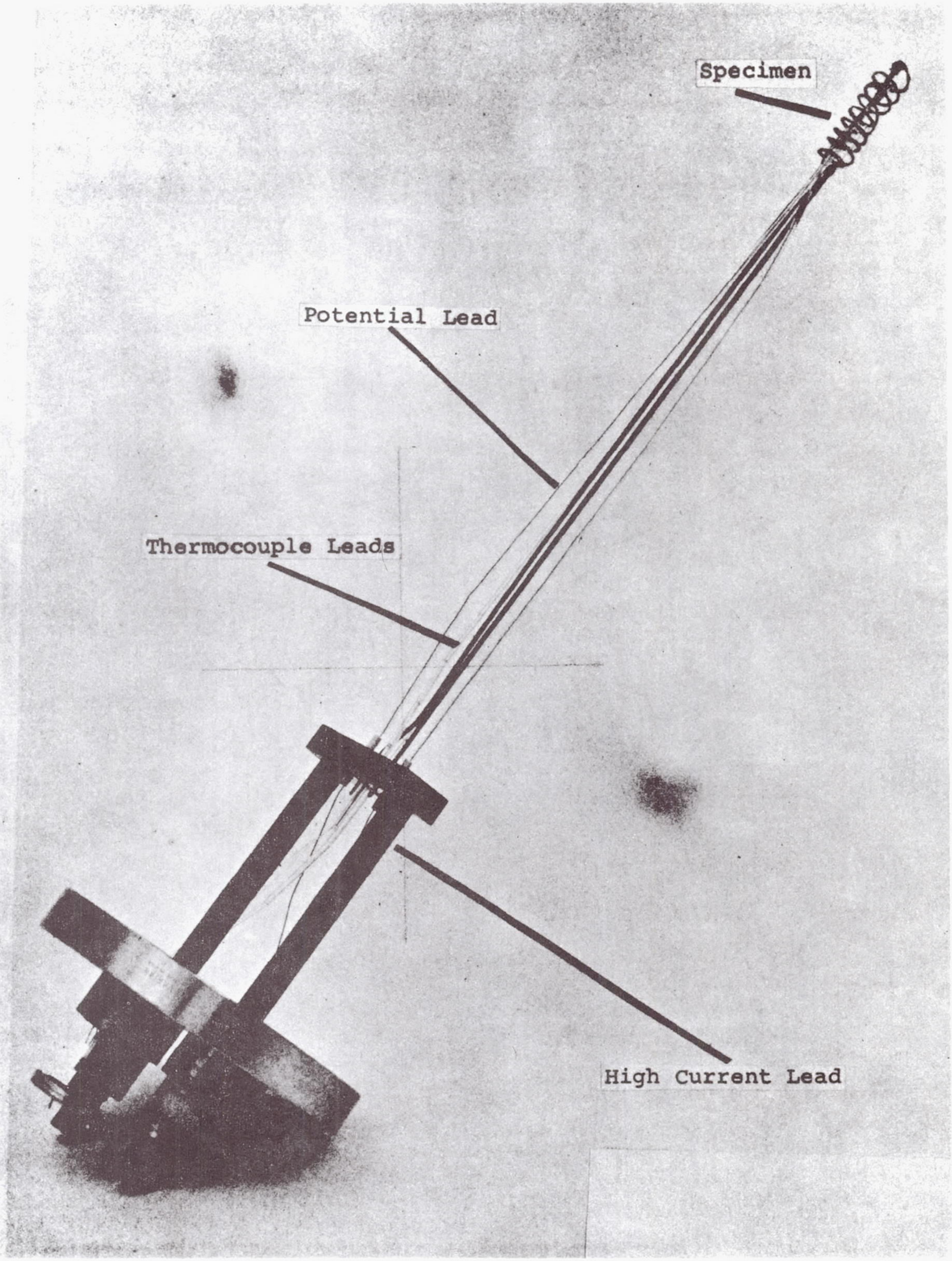


Figure 13 - Specimen Assembly

D-E, E-A, A-F, F-G, E-F, and D-G to be connected into the bridge circuit while the bridge current was passed the entire length of the specimen (leads C-B).

5. RESISTANCE AND TEMPERATURE MEASURING SYSTEM

Specimen resistance was determined by the 4 wire Kelvin bridge method. A Leeds and Northrup #4300 Adjustable Standard, #4320 ratio box and #2430-C galvanometer having a sensitivity of 5.02 microvolt/cm were the components of the bridge (Figure 14). The solubility specimen was included in the circuit by passing the bridge current through the heavy Cu posts and Mo rods attached to the ends of the specimen. Twenty gauge OFHC Cu wire was used to connect the Ta leads and the bridge. The bridge current was supplied by two 6-volt automotive type batteries in series. The bridge current was controlled by two 10-ohm variable resistors in series and read by a zero-to-1.0 amp D.C. ammeter. A reversing switch was situated between the power supply and the bridge. Precision of better than ±.01% was achieved in reading the resistance.

The temperature of the specimen was measured with Pt-Pt/10Rh thermocouples and a Leeds and Northrup Model K-3 Potentiometer. The thermocouple wire was joined to Cu wire in an insulated, distilled water ice bath. The same potentiometer and the bridge circuit galvanometer were used to measure the output of the thermocouples through a Leeds and Northrup Model 31-3 thermocouple switch. In general, temperatures could be read to ±0.1°C. Accuracy of temperature readings varied as discussed in the following section.

C. Apparatus Calibration

1. THERMOCOUPLES

Runs with Cb and runs with Ta01 through Ta19 were made with uncalibrated thermocouples. A standard couple, from the same coils of wire, was made against which the thermocouples used for the runs listed above were planned to be calibrated against the EIMAC NBS-certified standard. Both were destroyed by a melt-down in the calibration furnace when a controller failed.

Prior to the above accident, a new thermocouple, used in Runs Ta20 - Ta22, was calibrated against the EIMAC standard. The correction found is shown in Figure 15 as that marked TC 1. This new thermocouple was heated in the solubility furnace with the other two working couples. The hot junctions were placed together in a Nichrome cylinder covered with asbestos. All three couples read within 1/2° to 1-1/2°C of each other, with the wider variation at the highest temperatures.

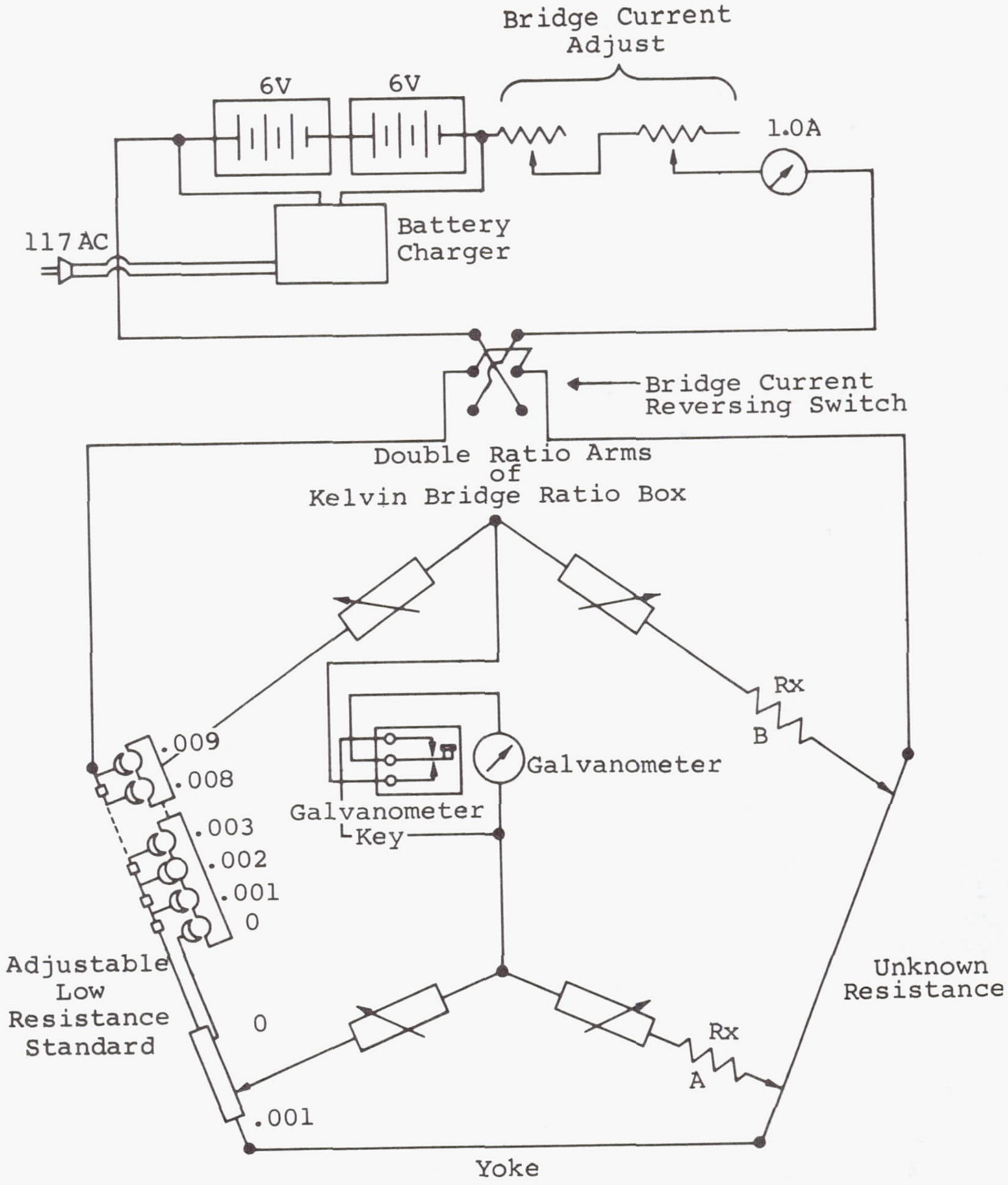


Figure 14 - Resistance Measuring Circuit

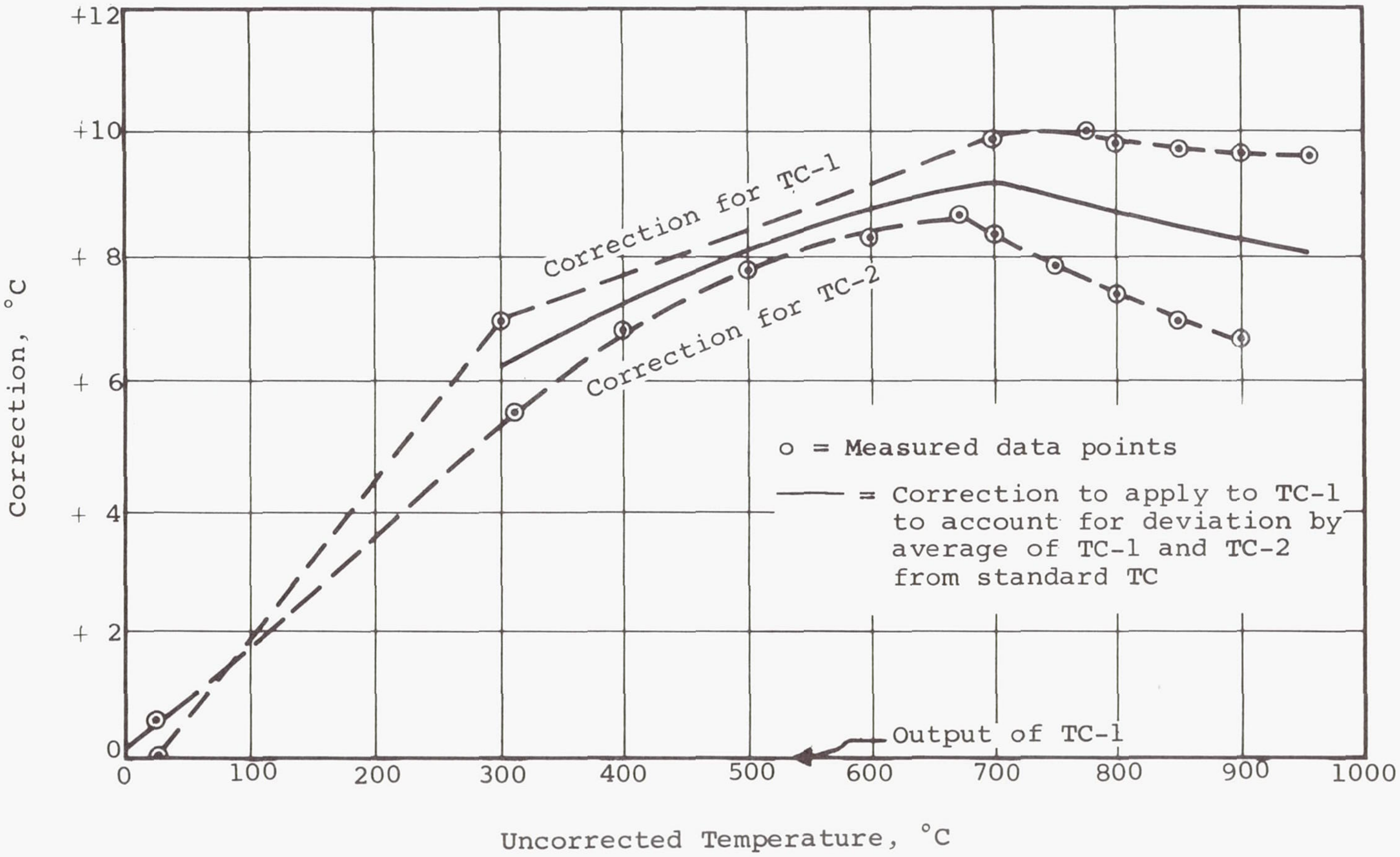


Figure 15 - Thermocouple Calibration Data

At the start of Run Ta20 it was decided to use the average of the outputs of TC 1 and TC 2, hanging free at the top and bottom of the specimen, respectively, as the specimen temperature. The final correction curve was then formed as shown in Figure 15 from data in Appendix B, Table C14, Event 3.

The calibrated TC 1 was in the calibration furnace for post-test calibration when it too was destroyed in the furnace melt down. Temperatures for the remaining test were measured with uncalibrated thermocouples.

It is noted that the correction for much of the temperature range is outside the normal variation for thermocouple wire. Since new, unworked wire is usually within calibration, the standard thermocouple was suspect. Other lots of wire were found to be equally far out of calibration. The standardization run that resulted in destruction of the relevant thermocouples was being conducted to obtain reference curves prior to having the standard recertified.

Considering the uncertainties, we regard the data accurate only within the usual variation of thermocouple wire (±3/4%). It is noted that this error is small compared to the oxygen analysis data so that the solubility data were negligibly affected.

2. FURNACE TEMPERATURE PROFILE

The temperature profile of the furnace was determined (Figure 16) at the start of the program by sliding a thermocouple up and down the furnace. Prior to Run TA20, the profile was determined by the following procedure:

The length of the furnace was 16 inches. The bottom of the 1-1/4-in.-dia. working volume was filled to 1-1/2 in. with Fibrefrax. The quartz tube was inserted so that its bottom was 2-1/2 in. below the center. The annulus between the quartz tube and the furnace was filled with Fibrefrax to a depth of 1 in. The calibrated thermocouple (TC-1) was inserted to the bottom of the quartz tube and connected to the ice bath. The top of the quartz tube was sealed to the thermocouple insulator with Fibrefrax. A scale was attached to the thermocouple holder.

After thermal equilibrium was reached at approximately 690°C, the thermocouple was withdrawn in steps of 1/4 in. and the temperature measured at each position. The results are plotted in Figure 17, along with a similar plot at 890°C. The uniform temperature zone shifted downward somewhat at the higher temperature.

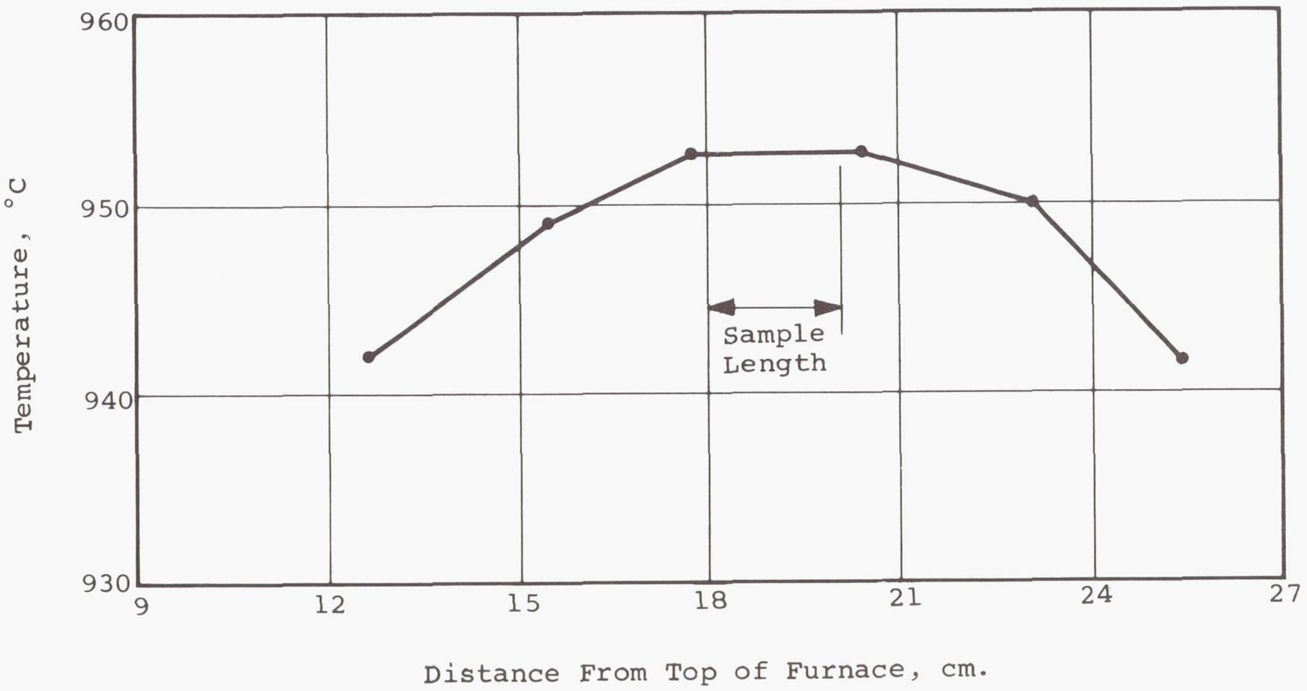


Figure 16 - Furnace Temperature Profile at Start of Program

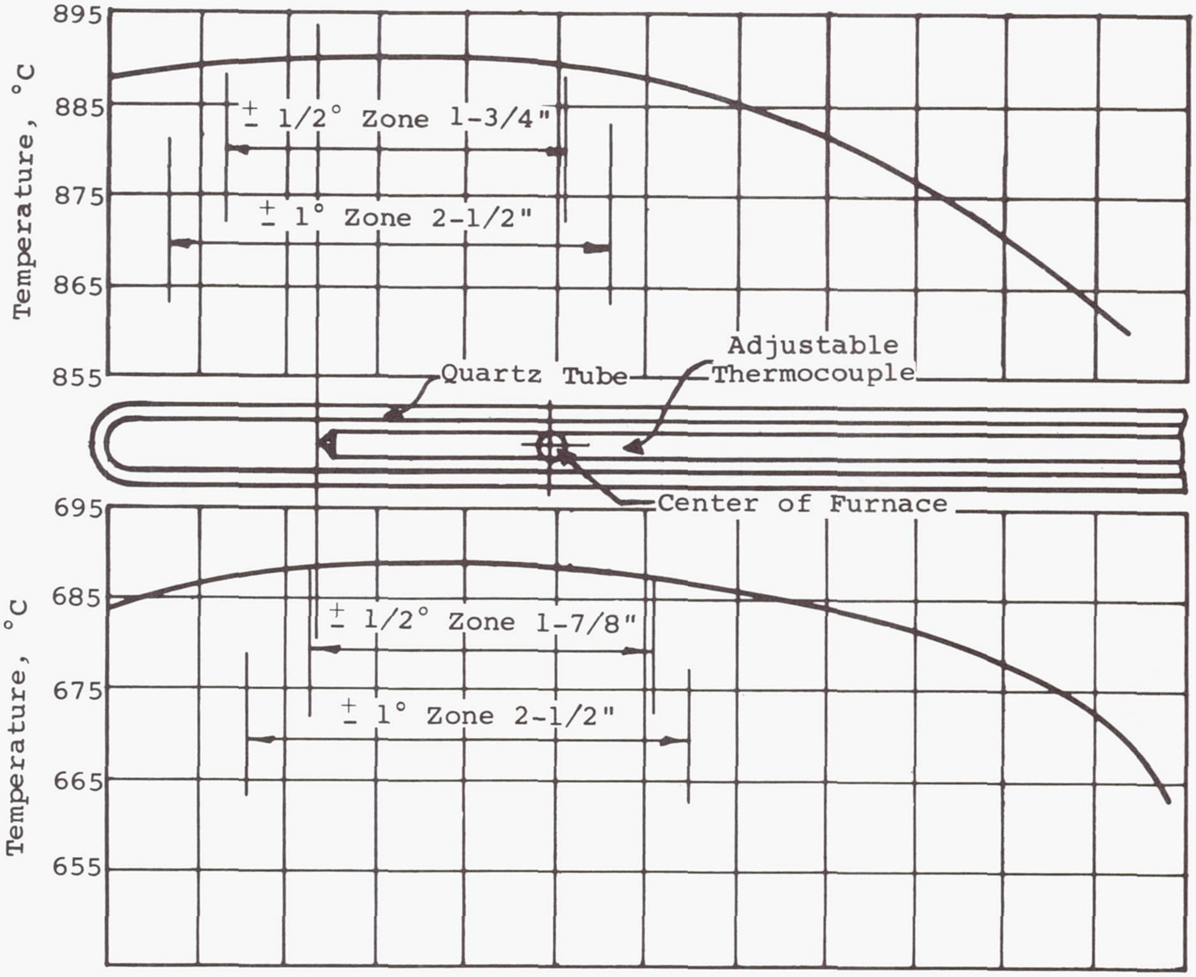


Figure 17 - Furnace Temperature Profile for Run Ta20

3. REPRODUCIBILITY OF RESISTANCE MEASUREMENT AND PRECISION BETWEEN RANGES

Tests were conducted with a dummy specimen at room and liquid nitrogen temperatures to determine the resistance measurement characteristics.

The effect of changing the resistance ratio is shown in Table 2 to be negligible. No change in resistance by changing the bridge current (Table 3) indicates negligible heating of the specimen in air. Later evidence indicated a slight effect which was associated with the specimen in vacuum at room temperature.

The effect of resistance asymmetry in the bridge arms is shown in Table 4 (see Section II.D.1.d).

Table 2. Effect of Bridge Ratio on Precision of Measuring Resistance.

Ratio:	10,000/100		10,000/300		10,000/400	
Temperature, °C	Resistance, ohms		RSTD	RSPEC	RSTD	RSPEC
	Standard	Specimen	ohms		ohms	
26	.002243	.2243	.006741	.2247	.008999	.2245
-195	.002091	.2091	.006278	.2093	.008370	.2093
26	.002246	.2246	.006733	.2244	.008975	.2244
-195	.002092	.2292	.007280	.2093	.008372	.2093
26	.002244	.2244	.006732	.2244	.008974	.2244

Table 3. Effect of Bridge Current on Specimen Resistance.

Bridge Current, amp	Resistance, ohms	
	Standard (1)	Specimen
0.64	.006732	.2244
0.80	.006732	.2244
1.00	.006732	.2244
1.3	.006732	.2244

(1) 10,000/300 ratio

Table 4. Effect of Resistance Asymmetry in Bridge Arms.

R_x, ohms	RESISTANCE OF DUMMY RESISTOR			% Error due to uneven Resistance	
	Equal Resistance in Arms	Add R_x to Arm A	Add R_x to Arm B		
10	.006732	.006723	.006734	-.13	+.03
47	.006732	.006690	.006741	-.62	+.13
100	.006732	.006639	.006740	-1.5	+.25
330	.006732	.006452	.006804	-4.2	+1.1

D. Operating Procedures

1. SOLUBILITY RUNS

a. Specimen Preparation

The specimen was formed by braiding three 15-in. lengths of 0.020-in.-dia. wire and coiling this into a helix on a 7/16-in.-dia. mandrel. Approximately 7 turns were made. The coil was chemically cleaned in a HNO_3 + 10% HF etchant and rinsed with de-ionized water. The initial weight was then recorded to 0.0001 g. The specimen was spot welded at the top near the end of a .075-in.-dia. Mo rod and at the bottom to a similar Mo rod which passed down the center of the helix. These rods supported the specimen and provided current leads for resistance heating of the specimen and current for the resistance measurements. The security of the spot welds was assisted by an intermediate layer of platinum foil. The two 0.020-in.-dia. Ta resistance measuring leads (potential leads) were attached, one turn from each end of the specimen, by separating the braid, inserting the lead, crimping, and spot welding. The thermocouples were attached in a similar manner by inserting the hot junction in the braid but securing it only by closing the braid behind the bead. The thermocouples were inserted one or two turns above and below the center of the specimen and below the center of the specimen and on opposite sides. Both the potential and thermocouple leads were quartz insulated and were brought down inside the helix.

The Conflat flange which contained the feed throughs for the temperature and resistance circuits and also the mounting posts to which the Mo rods were attached was bolted in place before the start of the run.

b. Pump out, Bake out, and Anneal

Roughing down was performed through the oxygen metering system to 100 torr or less. The roughing valve (V5) was closed and the Varian bakeable valve (V6) opened, Figure 5.

The system pumped rapidly to the 10^{-5} torr scale unless a general bake out of the system was necessary. If this was required, heating tape was used to raise the metal surfaces to 200° to 300°C for 1 to 5 hours as necessary. The heat was then removed. After a few hours the vacuum in the specimen chamber was on the 10^{-6} torr scale.

In all runs except Ta20, 21, and 22 the specimens were heated slowly with a current through the specimen until the annealing temperature (usually 1400°C) was reached and held for one hour. For the final three runs, the specimen was heated with the furnace in steps of 100°C. The pressure was monitored and the heating discontinued if a reading of greater than 5 x 10^{-5} torr was encountered. After thermal equilibrium was achieved at each 100°C, a resistance measurement was taken and a scan made with the residual gas analyzer. When 900°C was reached, the specimen was heated to 1400°C by means of a current through the specimen for the annealing interval of one hour. Some specimens were not heated as much because of insufficient power capacity.

The specimen was cooled to room temperature for a resistance measurement to be used for normalizing the subsequent resistance measurements. Base pressure at this time was about 10^{-8} torr with only H_2, CO and CH_3 showing on the residual gas analyzer scan. In some extended runs, base pressure dropped into the 10^{-9} torr range.

c. Engassing, Isothermal, Pulse Method

The method depended on precisely reproducable quantities of oxygen being added to the wire in pulses. A run consisted of measuring the resistance at room temperature, raising the specimen to temperature, repeating the resistance measurement, and then adding pulses of oxygen. Resistance was measured after each pulse or group of pulses, as seemed prudent.

The furnace temperature was monitored continuously on the controller chart. The sample temperature was read with the potentiometer at regular intervals. Once set, it usually remained essentially constant. The pressure in the manifold and furnace was measured with the Millitorr gauge, with the output continuously recorded on a logarithmic chart.

A pulse cycle was conducted as follows. Evacuate the entire oxygen fill system (Figure 5) through a liquid nitrogen trap with a mechanical pump. Fill the 300 cc volume to the desired pressure (0.5 or 2 torr) by cracking V1 and again evacuating the system to the exact pressure. Admit oxygen into the sample chamber through the variable leak, (V1).

Start cycle shortly before t = 0 and after the sample resistance is read, by closing V6 and V2. At t = 0, open V5 and V4 admitting oxygen to the system. Open calibrated leak to permit rapid equalization of pressure. At t = 3-3/4 min. and t = 7-1/2 min., fill the 300 cc volume with oxygen by cracking V1. Evacuate the oxygen fill system by opening V2. This completes the valving cycle which repeats every 7-1/2 minutes. The times were selected because each division on the pressure chart (from which time was measured) happened to correspond to 3-3/4 min.

The resistance readings were plotted and the cycling continued until the desired oxygen level was achieved.

d. Details of Resistance and Temperature Measurement

A reversing switch was installed in the bridge current circuit and was used for the following: 1) to test the Kelvin bridge circuit for unequal resistance in the potential leads, 2) to test for thermoelectric errors, 3) to test for interference between the bridge current and the thermocouple leads.

In the first case, since the Kelvin circuit is not a null circuit except in the galvanometer leg, a resistance asymmetry of an order comparable to the resistances in the ratio box will cause an erroneous reading and will be dependent on bridge current. For example, if the ratio is 10,000/400 and the resistance assymetry amounts to 1 ohm, the ratio for sample resistance will be roughly $(1/10^4)$ x 100% or 0.01% in error. Errors of this type may be reduced but not eliminated by averaging the two readings.

A thermoelectric error may arise in the potential leads by the temperature gradient and chemical or cold work gradients. Both of these are secondary effects and are apt to be small but can be eliminated by averaging the two readings.

Any difference in the temperature readings when the bridge current is reversed indicates that the leads near the junction are in contact with the wires at different points in the potential gradient along the braid. The error involved can be eliminated by averaging the two temperature readings.

e. Isoconcentration Run

The isoconcentration run was performed with an engassed specimen simply by starting at a high temperature (in the single phase region) and taking resistance and temperature readings there and at every 50°C as the temperature was lowered stepwise to a point in the two phase region.

f. Ta20 Procedure

The procedure for Run Ta20 was a series of closely spaced isoconcentration runs, one of which was performed after each oxygen pulse. This was continued for 19 pulses after which an isoconcentration run was performed after each four pulses to a total of 38 pulses.

g. Ta22 Procedure

The procedure of Gebhardt[2] was followed as closely as possible. The oxygen was admitted so as to maintain a constant pressure over the specimen. The temperature was held constant and the resistance measured as a function of time. The oxygen rate of absorption changed with time, therefore, the flow rate into the sample chamber had to be changed. Flow rate was adjusted both by adjusting the pressure behind valve V5 and by adjusting V5.

2. DIFFUSION RUN

After the specimen was baked out at 900°C, it was cooled to room temperature. The segment A-B between the Mo rods (Figure 4) was heated to 900°C for engassing. The amount of oxygen was calculated to reach approximately the solubility limit. Eight 0.5-torr, 300 cc pulses were admitted to the specimen chamber. Only the hot half of the specimen absorbed the oxygen, as determined from resistance measurement.

The specimen was cooled to room temperature and the lowered furnace brought to 600°C. At time zero, the furnace was raised to enclose the specimen area. The specimen thermocouple output was continuously recorded, and resistance readings taken at one minute intervals once thermal equilibrium was established. This was reduced to 5 minute intervals after 15 minutes.

The specimen segments over which resistance was measured were D-E and F-G. Resistance asymmetry between the Mo rod and tantalum wire leads made the E-A and A-F readings inaccurate as explained in section 1d. No change in resistance was noted after 3 hours so the temperature was raised to 700°C. After 3 hours the furnace was shut down overnight.

The next day the furnace was heated to 800°C in the lowered position. It was then raised and readings taken as before for 3 hours. There was no change, so the temperature was again increased to 900°C, where it remained for 6 hours. After 3 hours the readings were reduced to one every half hour. No resistance change was noted so the run was terminated.

The specimen was removed and subjected to microhardness tests.

III. RESULTS

Twenty-two runs were made to obtain data on the solubility of oxygen in Ta (Section A). Of these, five were to obtain calibration data and six were used to plot a solubility curve.

Ten runs were made to obtain data on the solubility of oxygen in Cb (Section B). These data were not accurate enough to be useful.

Single trial runs were made with Cb-1Zr and Ta-10W alloys (Section C). A single run was completed to measure diffusion of oxygen in Ta (Section C). Data were obtained useful to plan further runs.

Other data were obtained from a special run with Ta. Results are useful beyond the contracted scope of the program (Section A3).

A. Solubility of Oxygen in Tantalum

1. Calibration Data

Runs Ta10 to Ta14 were intended as calibration runs. Samples were charged with oxygen in pulses at 900°C as shown in Appendix B, Tables B5 to B9. The samples were then analyzed for oxygen by the neutron activation method by General Atomics with other samples as shown in Table 5. Results were plotted in the quarterly reports as a calibration curve of R/R_0 at 900° versus oxygen content. The solubility points given in the quarterly reports were calculated from this graph. It was later found that R/R_0 varies with temperature, resulting in an error if R/R_0 calibration data taken at 900°C are used at other temperatures without correction. The method for determining and using the corrected calibration curve is described below.

The resistance data from Runs Ta10, 11, and 12 (given in Tables B5, B6, and B7) are plotted in Figure 18. Other data in this figure will be discussed later. The sample from Run Ta13 was not charged with oxygen. The run was intended to show the blank change in resistance due to the outgassing and annealling step. Essentially no blank correction was needed. This conclusion appears to be at odds with the data in Table 5 which show about a 70 ppm difference between as received wire and the Ta13 sample. As shown later, this difference is not necessarily significant. More data are needed to resolve the discrepancy. Linear, coinciding plots of R vs. N_0 are shown in Fig. 18 for Runs Ta10, 11, and 12 suggesting that the solubility had not been exceeded and indicating excellent reproducibility between runs.

A plot of the oxygen concentration vs. ΔR reached at the end of the calibration runs for Runs Ta10-13 is shown in

Table 5. Calibration Data for Tantalum

	Prerun Data				
Sample No.	R, Ohms	Temp., °C	R°_{30}(1), Ohms	R°_{900}(2), Ohms	Weight, g
As rec'd	-	-	-	-	3.1523
Ta10	.0642	41.9	.0625	.2077	2.4341
Ta11	.0702	53.3	.0666	.2207	2.6425
Ta12	.0689	33.1	.0684	.2270	2.7401
Ta13	.0686	38.1	.0673	.2238	2.6639
Ta05	.0619	32.0	.0615	.1912	2.5182
Ta07	.0700	25.3	.0709	-	2.8456
Ta14	.0742	36.3	.0731	-	2.6231

	Post Run Data						Oxygen Concentration(4) ppm by weight		Std. Dev. ppm(5)
Sample No.	R, Ohms	Temp., °C	R^f_{30}, Ohms	R^f_{900}, Ohms	ΔR_{900},(3) Ohms	$\frac{\Delta R_{900}}{R^\circ_{30}}$	1	2	
As rec'd	-	-	-	-	-	-	159	133	20
Ta10	.0922	49.0	.0894	.2324	.0247	.396	1350	1300	50
Ta11	.0785	42.4	.0765	.2318	.0111	.167	817	764	50
Ta12	.1110	32.0	.1107	.2679	.0409	.598	1640	1710	60
Ta13	.0708	48.7	.0678	-	0	0	206	225	20
Ta05	-	-	-	-	.073(6)	1.19	4370	4370	40
Ta07	-	-	-	-	.034(6)	.48	2450	2430	10
Ta14	-	-	-	-	.0882(6)	1.21	2440	2490	50

(1) Prerun resistance at room temperature corrected to 30°C.

(2) Prerun resistance at engassing temperature corrected to 900°C.

(3) Difference between prerun and post run resistance at 900°C.

(4) Determined by activation analysis at General Atomic, San Diego, California. Each specimen was run twice.

(5) Determined from counting statistics by General Atomic.

(6) Determined by graphical extrapolation in Figure 18.

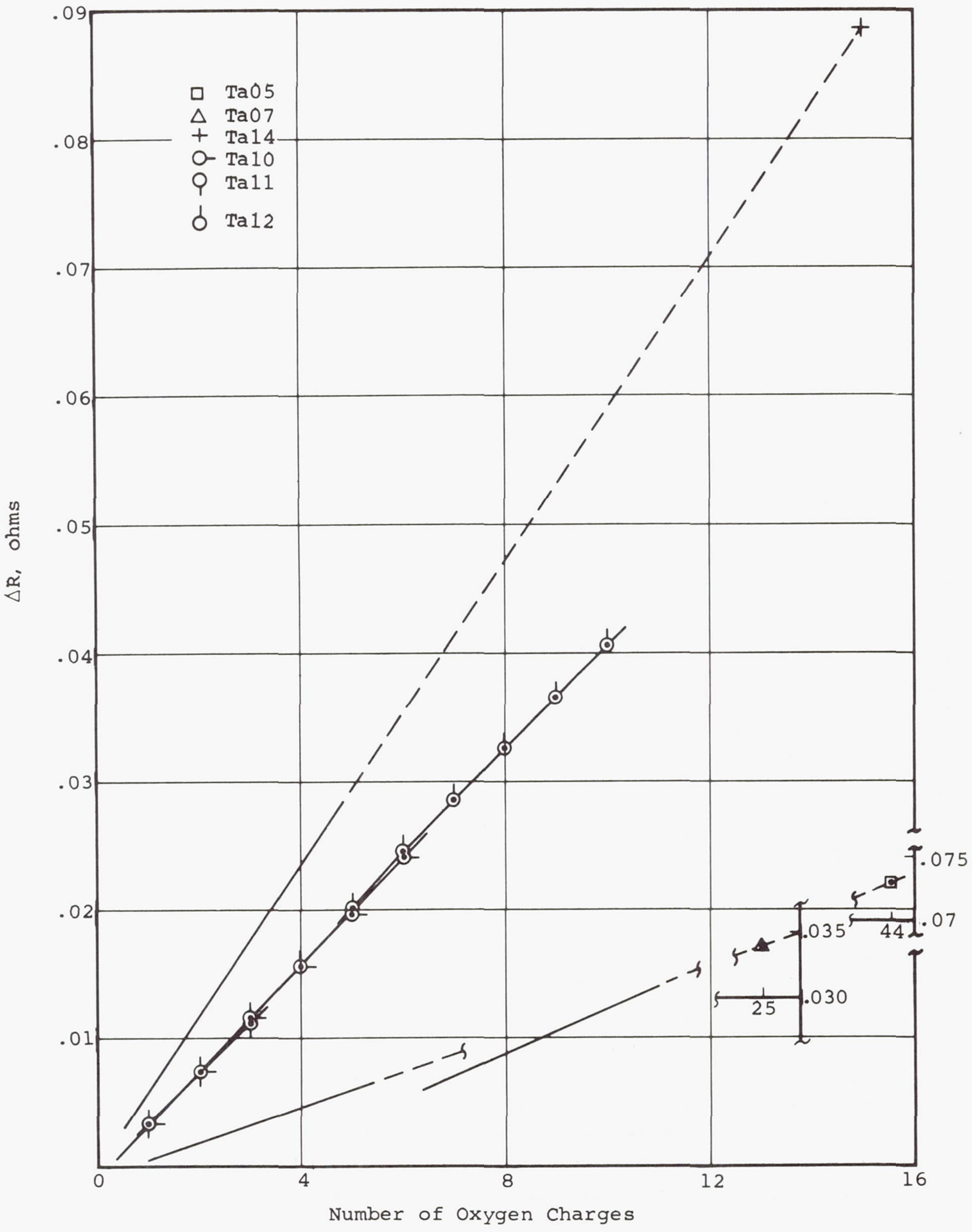


Figure 18 - Linear Plot of Resistance Change vs. Oxygen Charge Number for Tantalum Calibration Runs.

Fig. 19. Also shown are points from Runs Ta05, 07, and 14. Various interpretations of the data are possible. These are described below, with reasons for selecting a preferred interpretation for use in calculating solubility points. First, however, a discussion of the data from Runs Ta05, 07, and 14 is given.

1) Data from Runs Ta05, 07, and 14.

Of these 3 runs, Ta05 was a trial run to test the solubility technique, Ta07 was a solubility run, and Ta14 was a calibration run. There was no reason to invalidate the data from any of the 3 runs, so all must be interpreted. All are included in the calibration analysis because the final samples were all analyzed for oxygen after the run. Data from all 3 runs are also used to estimate the solubility curve, as described in a later section.

The linear sections of the Ta05, Ta07 and Ta14 curves from Fig. 20 are reproduced in Fig. 18. It is apparent from the differing slopes that different amounts of oxygen were taken up by the samples in each charge. The amount per charge was nearly identical for Runs Ta10, 11 and 12. Although charged under essentially identical conditions, a greater amount of oxygen per charge apparently was taken up in Ta14. The only difference in pre-engassing procedure was that the Ta14 sample was held at 900° for 3 days rather than overnight for Runs Ta10-13. The lower rate of oxygen uptake for Runs Ta05 and 07 can be accounted for by the fact that they were engassed at lower temperatures, and the charge pressure was one half that of the calibration runs.

Extrapolations of the linear sections of the Ta05, 07, and 14 curves to the total number of charges are shown because of the following reasoning. The interpretation of the calibration data involves the slope of the $\Delta R/R_{30}$ vs. oxygen concentration curve (Fig. 19). We knew of no *a priori* reason why the slope should be constant over the entire oxygen content range. In particular, the slope below the solubility might be different from that above. Gebhardt(1) shows the resistance vs. oxygen content curve at 10°C to be linear from 0 to 5 atomic %. However, to get the curve, he engassed at a high temperature and quenched to 10°C. All his data are thus equivalent to $\Delta R_{10}/R_{10}$, where the R is always measured on the 2-phase quenched structure. Our data are all $\Delta R_{900}/R_{30}$ where R is measured on the 1-phase structure at 900°C, up to the solubility limit, and on the 2-phase structure above the solubility limit. The nearest direct comparison is between our $\Delta R_{30}/R_{30}$ at the end of each run and his $\Delta R_{10}/R_{10}$ at an equivalent oxygen concentration. More will be said on this later.

Another factor leading to the possibility of 2 (or more) slopes is the fact that the oxygen in solution is in

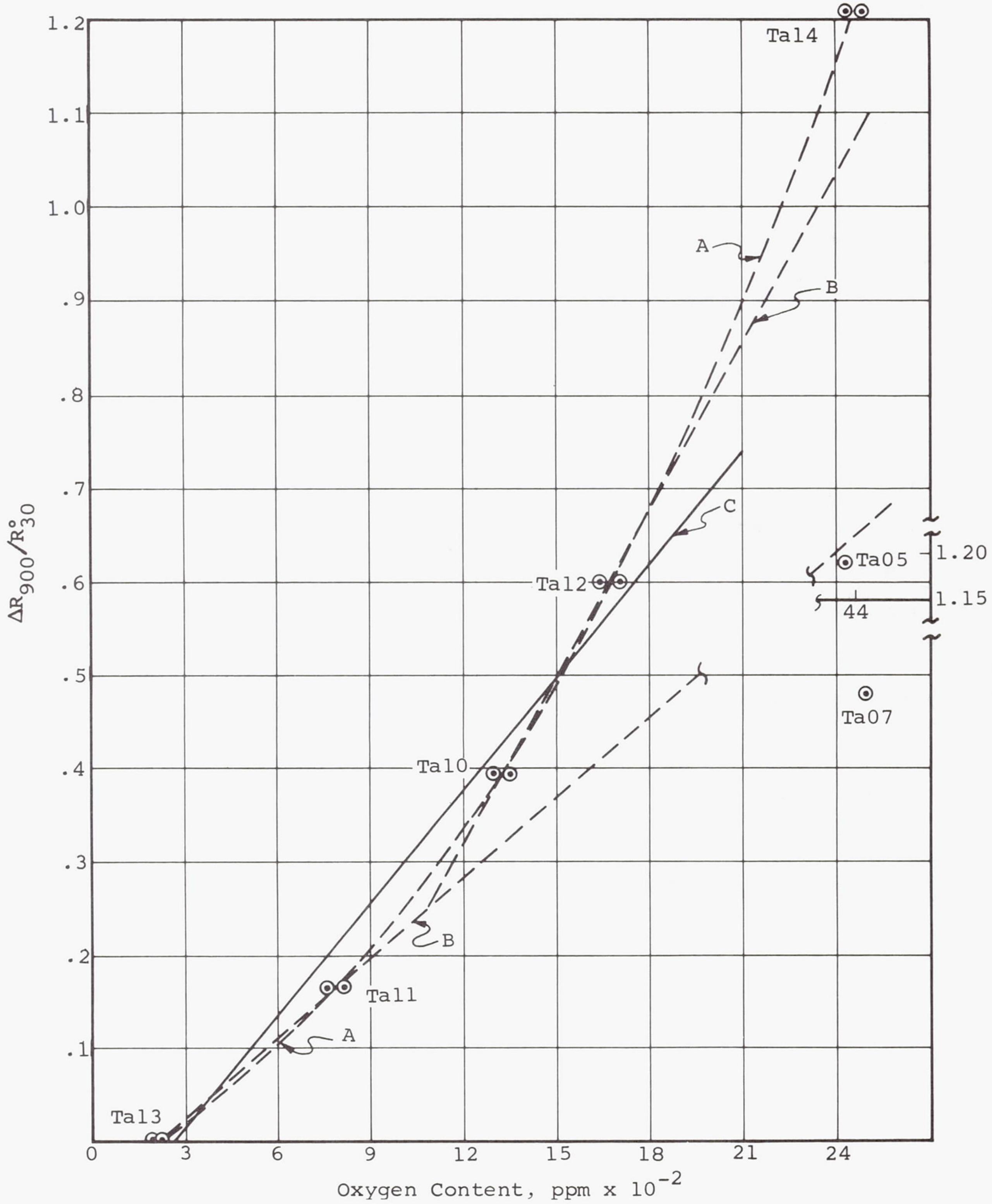


Figure 19 - Calibration Curve for Tantalum

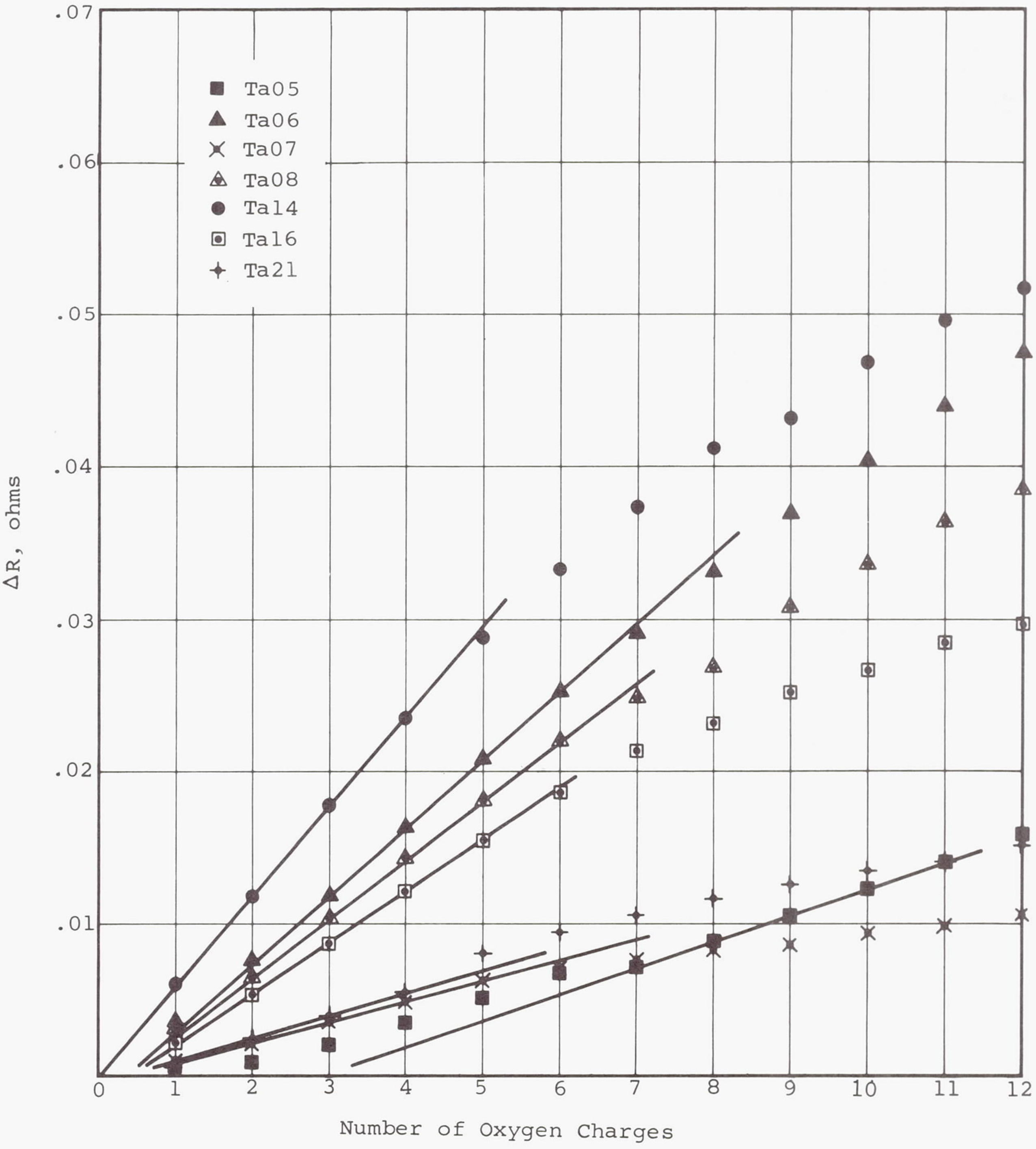


Figure 19 - Linear Plot of Resistance Change vs Oxygen Change Data for Tantalum.

equilibrium with different oxides at different temperatures. Thus, engassing above 750° where the stable oxide is Ta_2O_5 might result in a slightly different slope than if engassed below that temperature where other oxides appear to be the stable species. Gebhardt's work would not show this type of slope change. The two Runs Ta05 and Ta07 in our work, in comparison with runs engassed at higher temperature, might disclose a slope change.

The solubility was exceeded during the engassing on all 3 Runs, Ta05, 07, and 14, (again, explained in the next section) with parabolic portions of the curves resulting from scale formation above the solubility limit. If a constant oxygen pickup per charge is assumed (see Appendix F), then if the oxygen had been able to go into solution at the same rate for the total number of charges, the ΔR that would have resulted is shown as the extrapolated points in Figure 18. These ΔR values, converted to values of $\Delta R/R_{30}$, are also plotted in Fig. 19. The significance of the extrapolated points is covered in the following interpretation paragraphs.

2) Interpretation 1 - Curve A.

A smooth logarithmic curve can be drawn through the data from the first 4 calibration runs and the extrapolated Ta14 point. Although this gives the best fit to the data, the curve would appear to have no theoretical significance and is grossly deviant from Gebhardt's curve. This interpretation is rejected.

3) Interpretation 2 - Curve B.

The next best fit to the data is shown by curve B. The arguments in the section 1) above apply to this interpretation. The break in the curve at 1150 ppm corresponds approximately to the solubility at 900°C, the temperature of engassing of the calibration runs. The extrapolated Ta05 and Ta14 points also fall approximately on the two parts of the curve as postulated in section 1). The extrapolated Ta07 curve falls below the curve. We can postulate no likely reason for this. If some of the scale fell off the wire before it was analyzed it would have the effect of placing the point to the left of where it should be rather than to the right.

The slope of neither part of the curve is similar to Gebhardt's. Despite the interesting coincidences pointed out, the data are not numerous enough to support this interpretation or refute the data of Gebhardt. The assumptions required to support the validity of the extrapolated points are also tenuous. In particular, despite the evidence given in Appendix F, it is difficult to believe that the amount of O_2 taken up in each charge by the specimens remains constant after the solubility point is passed.

We reject this curve also, recognizing that a break in the curve could still exist.

4) Interpretation 3 - Curve C.

If data from Runs Ta05, 07, and 14 are rejected because the solubility was exceeded and the chemical analysis was conducted on an imperfectly characterized and understood 2-phase structure, one can draw a best straight line through the 4 remaining points. To do this, one must accept a between-samples analytical error substantially larger than the within-sample error.

If this curve is extrapolated to intersect with the ordinate, one could then renumber the ordinate to reflect change in resistance from zero oxygen concentration rather than the oxygen concentration of the starting material. If we also assume that Gebhardt's degassing process also reduced his starting O_2 content to near zero, then his % change in resistance data correspond to our $\Delta R/R_{30}$ data (after dividing by 100). If we do the above, we find that curve C is identical to Gebhardt's curve.

Because we have insufficient data and reasoning to support any other interpretation and to refute Gebhardt's data, we adopted this curve for calculating the solubility points discussed in the next section. The total uncertainty in $\Delta R/R_{30}$ value taken from curve C is seen to be of the order of ±100 ppm.

2. SOLUBILITY DATA

Runs Ta01 to Ta04 were exploratory runs and data were not used. Data from Runs Ta05 - Ta09, Ta14, Ta16, and Ta21 were used to calculate solubility points. The curve published in the quarterly reports was found to be in error when assumptions used in interpreting the data were found to be incorrect. This was discovered during Run Ta21, as outlined below.

Run Ta21 was conducted at 700°C with the procedure used for Runs Ta01 through Ta19. Data from this run resulted in important evidence concerning the metallurgical phenomena which determine the nature of the ΔR versus number-of-oxygen-pulse curve. An initial linear portion was found as expected from prior similar runs; also, after 6 oxygen pulses, the slope began to decrease as before. The run was continued to 48 pulses. The unexpected feature was that the slope never became constant as was thought to occur earlier.

The data from the curved portion of the curve were fitted to various functions and found to be parabolic. Furthermore, the curve contained 4 parabolic regions: first, from pulse 5 to pulse 14; second, pulse 14 to pulse 32; third, 32 to 42; fourth, 42 to 48. In the Cb-O system, a similar effect was found by Hurlen(3) by measuring the weight gain versus time at constant temperature and oxygen pressure. He found an initial linear segment followed by several parabolic regions. He interpreted the linear region to indicate a boundary-controlled rate, while a parabolic region indicated that diffusion through a growing layer controlled the rate.

The implication is that the solubility limit is reached at the end of the linear region, and not at the intersection of the extrapolated lines from the initial linear and a later linear region, as we previously thought.

As a result of the above implication, we re-examined the data from the prior Runs Ta05 - Ta08 and Ta10 - Ta16. Furthermore, since R/R_o is dependent on temperature, the basic parameter was changed to ΔR. For each run, ΔR was plotted against the number of oxygen pulses (Figure 20) and again versus the square root of the number of oxygen pulses (Figure 21) in order to identify the parabolic

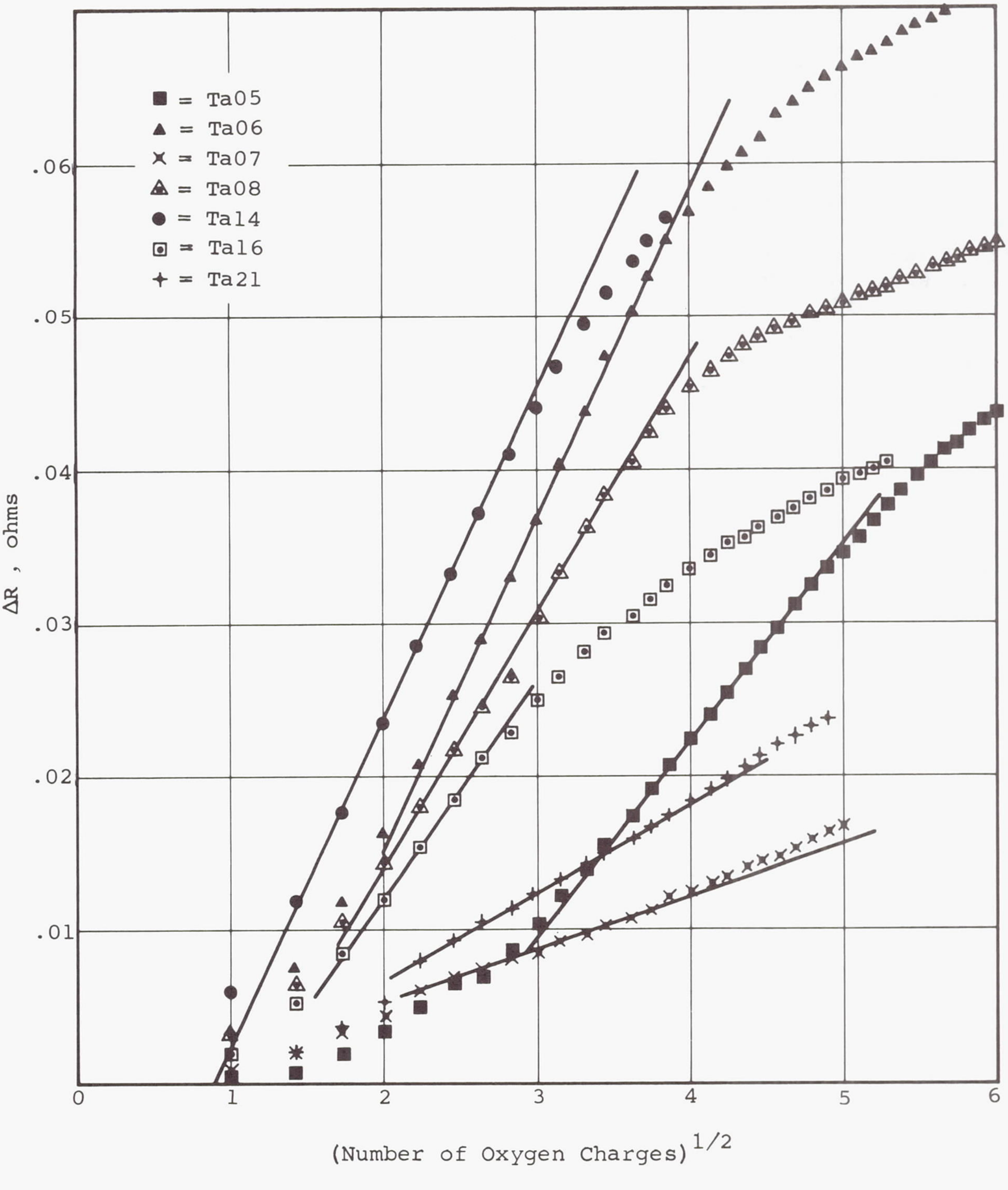


Figure 21 - Parabolic Plot of Resistance Change vs. Oxygen Charge Data for Tantalum.

regions. From these, the equation $R = a + b N_O$ was determined for the initial linear segment and the equation $N_O = a + b(\Delta R) + c(\Delta R)^2$ was determined for the first parabolic region. The simultaneous solution of these two equations resulted in the first departure from linearity, designated as ΔR^*. See Appendix C for details of the calculation.

Values of ΔR were divided by the room temperature resistance to normalize geometric variations. The resulting number was then correlated to ppm oxygen content with Figure 19. This was done for the prior Runs Ta05 through Ta08, Ta14, and Ta21, and the results replotted as log of solubilities versus reciprocal temperatures in Figure 22. Data are summarized in Table 6.

Ta14 was planned as the highest oxygen content calibration point, but was questionable as a calibration point because the engassing was found to have entered the first parabolic region. However, a solubility limit point could be derived from the data.

Before the recalculation of the solubility for Figure 22, we reported our curve in the quarterly reports to be much higher. We were unable to account for the large difference between our data and that of Gebhardt(2). Run Ta22 was conducted using the latter's method to investigate this difference.

Preparations and samples were the same as for Run Ta21. The O_2 pressure in the furnace was adjusted as closely as possible to 0.02 torr (same as Gebhardt), and the resistance change measured as a function of time (Table B15, Figure 23).

The data were interpreted in the same manner as for Ta21, by plotting ΔR versus both time and the square root of time. Since there is such a sharp decrease in slope, a ΔR^* was obtained by visual estimate and divided by the room temperature starting resistance to obtain the solubility point from Figure 19. The temperature of the run was 700°C. Gebhardt's(2) lowest temperature was 900°C. The Ta22 point is shown in Figure 22 to coincide with an extrapolation of Gebhardt's data.

After the steep linear section, two parabolic regions are shown in Figure 23. The first one is very short and may indicate a transition from the linear to a parabolic region rather than

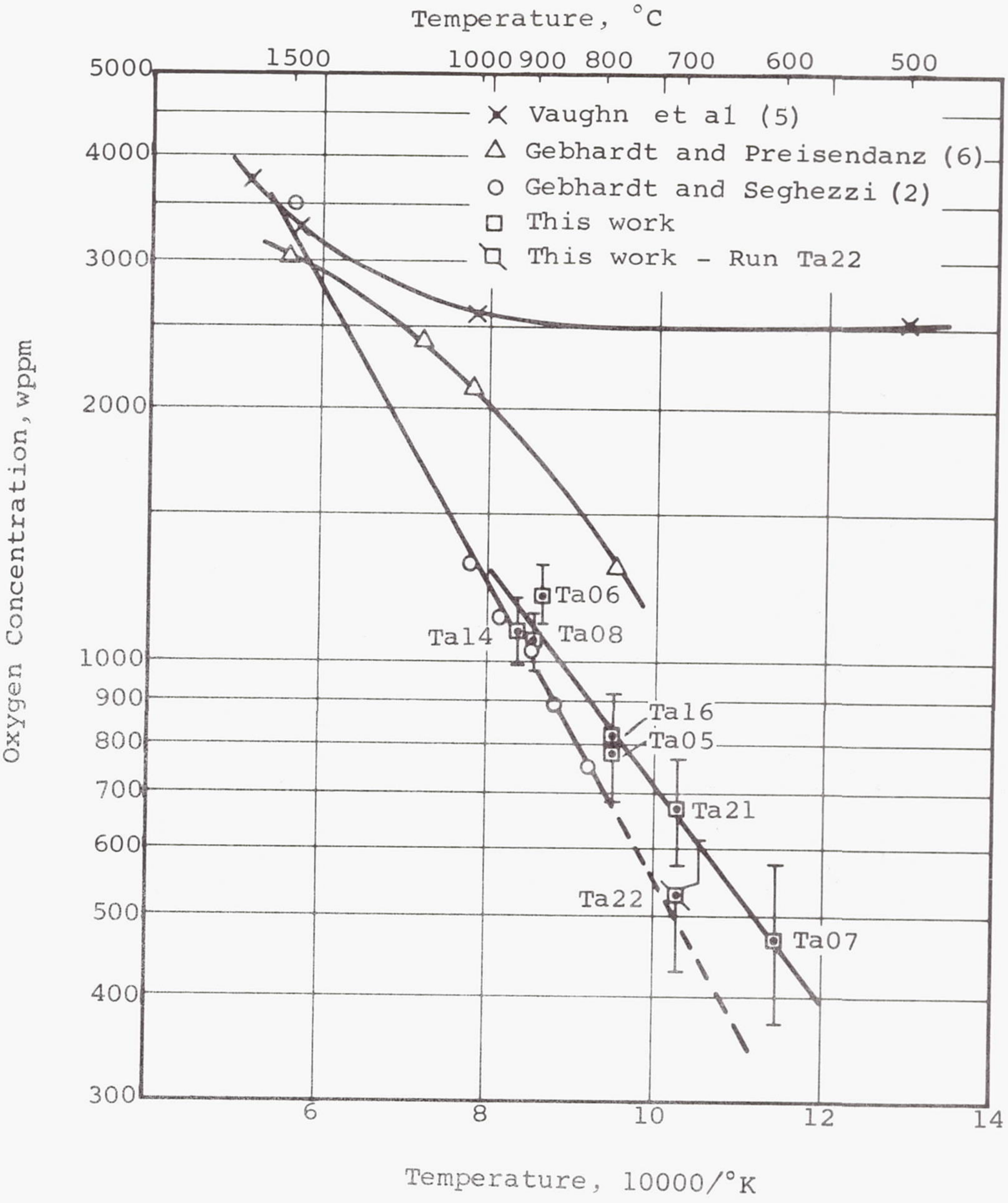


Figure 22 - Solubility of Oxygen in Tantalum

Table 6. Summary of Tantalum Solubility Data

Run No.	ΔR^*(1)	R°_{30}	$\frac{\Delta R^*}{R^\circ_{30}}$	T°C	Oxygen Concentration ppm	Method (1)
Ta05	.0130	.0615	.212	780	780	E
Ta06	.0265	.0738	.372	880	1190	G
Ta07	.0060	.0709	.085	600	470	E
Ta08	.0166	.0500	.332	920	1080	G
Ta14	.0233	.0731	.319	900	1050	G
Ta16	.0219	.0977	.224	780	820	G
Ta21	.0081	.0488	.166	700	670	E
Ta22	.0068	.0630	.108	700	530	E

(1) Intercept between linear and parabolic sections of $\Delta R^*/R_{30}$ vs. number-of-oxygen-charge curve determined by simultaneous solution of slope equations (E) or by visual estimate from a graph (G).

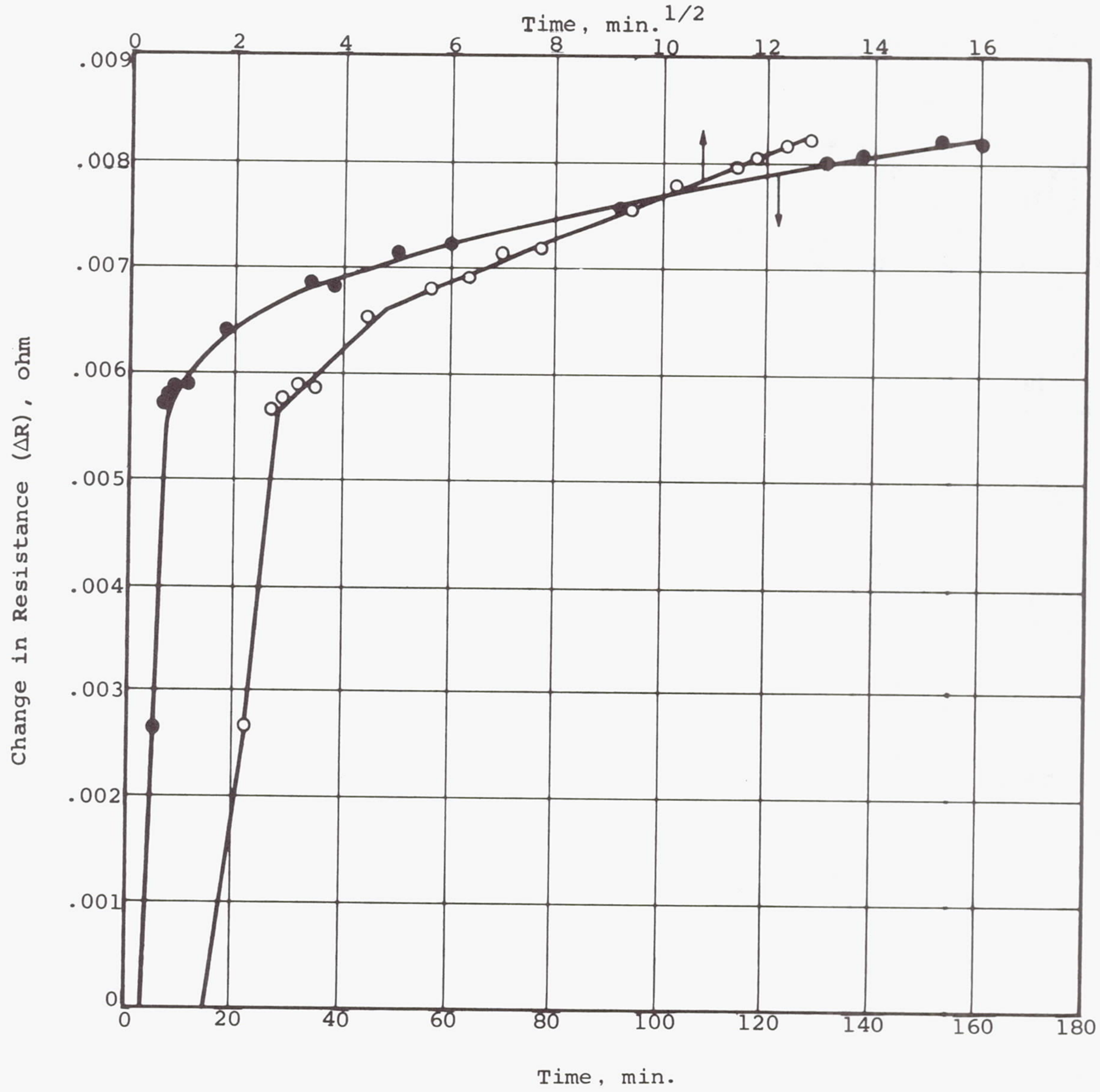


Figure 23 - Change in Resistance vs. Time for Tantalum at 700°C - Run Ta22.

be a separate parabolic region.

3. RUN Ta20

Under the procedure used in prior runs, the determination of five points along the solubility versus temperature curve for a material required at least ten separate runs: Five isothermal runs to determine solubility versus reduced resistance and five or so runs to calibrate reduced resistance versus oxygen content.

The extended program shutdown had occurred after Run Ta19. Because of the change in personnel, it was decided to conduct a trial run to check out the apparatus thoroughly and gain experience with the operating technique. It was also decided to conduct the run in a way, which, if successful, would greatly shorten the time to obtain a solubility curve for a material. With this new technique, resistance of the starting wire was to be measured at 500, 600, 700, 800, and 950°C. Increments of oxygen were to be added at 950°C as in the isothermal pulse method. However, after each increment, the resistance was to be measured at the four other temperatures. The experiment was to be shut down daily to obtain the room temperature resistance. A series of parallel R versus T curves was expected in which a change of slope was expected at a phase boundary.

Two changes in slope were found immediately in the R versus T curves (Figure 24), one at about 450°C and the other at about 750°C. Because of these breaks, resistance was measured at additional temperatures to clarify them. Twenty charges were completed after which the R versus T determination was reduced to once every 4 charges to a total of 38 charges. A plot of the raw data, resistance versus temperature, resulted in parallel lines for each charge, confirming Nordheim's Rule. This rule, simply stated, is that the change in resistance for a given change in oxygen content is independent of temperature. The major features of this set of data are outlined below.

1) A series of R versus T curves was obtained which were roughly parallel, as would be expected from Nordheim's rule (e.g., see Figure 24). The large amount of data taken are listed in Table B14 in the order taken. To extract the maximum value from the data, they must be plotted on a scale of at least 100°C = 4 in. versus .01 ohm = 4 in. The resulting figure

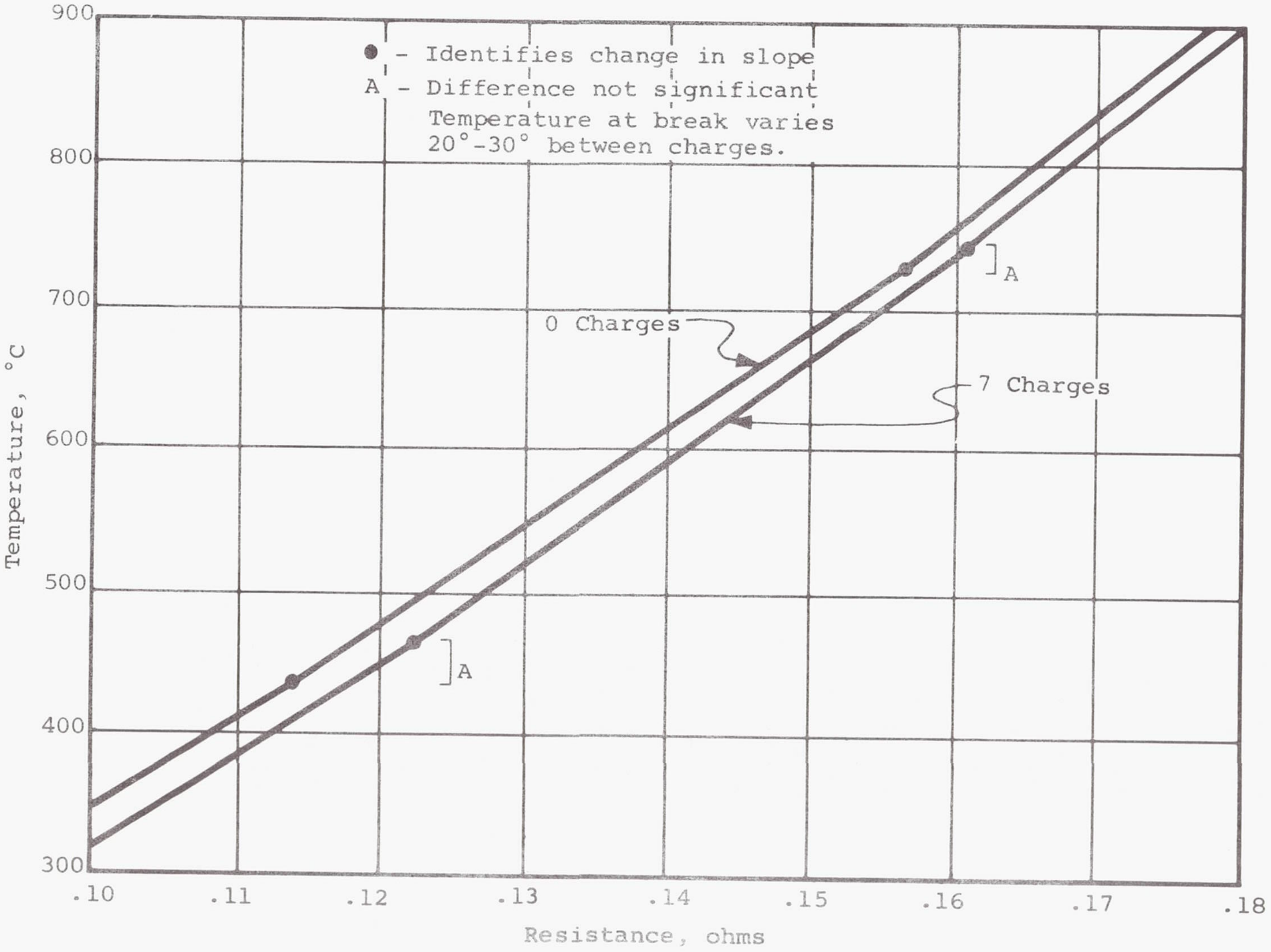


Figure 24 - Resistance vs. Temperature of Tantalum - Run Ta20

was much too complex to reproduce on a page smaller than 2 ft. x 3 ft. so only the principal features are illustrated by figures in this report. A detailed evaluation of the data was beyond the scope of the program.

2) The breaks at about 450°C and 750°C were found to persist as oxygen was added, with no significant change as a function of O_2 content. The breaks varied around average values in a more or less random fashion between oxygen charges. The lower values varied ±30°C and the upper values ±45°C. The presence of the break at 750°C became uncertain after 11 charges because of the obscuring effect of the following result.

3) A shift in resistance was found to occur after 7 charges. This varied in a systematic, but complex way (Figure 25). At the same time the change in resistance resulting from a charge of oxygen became a function of temperature when previously it was independent of temperature (Figure 26). The resistance shift at 900°C (Figure 25) could be made to disappear and reappear by a suitable heat treatment. This shift was found later to be a part of an offset in the R versus T curve similar to that for α, γ and δ Fe (Figure 27). However, the offset amounted to only about .3% change in resistance. These phenomena are described in more detail later.

4) Reproducibility of the resistance and temperature was about 0.0001 ohm (in a 0.2 ohm sample) and 0.2°C respectively.

5) Although the temperature range from 300°C to 950°C, and oxygen content range from about 250 ppm to about 1200 ppm O_2 was covered in this run, no series of breaks that could be interpreted as a solubility curve was found.

6) The breaks that were found are interpreted as transitions between Ta oxides, since the temperatures approximately correspond to transitions reported by others in oxidation studies. If this interpretation

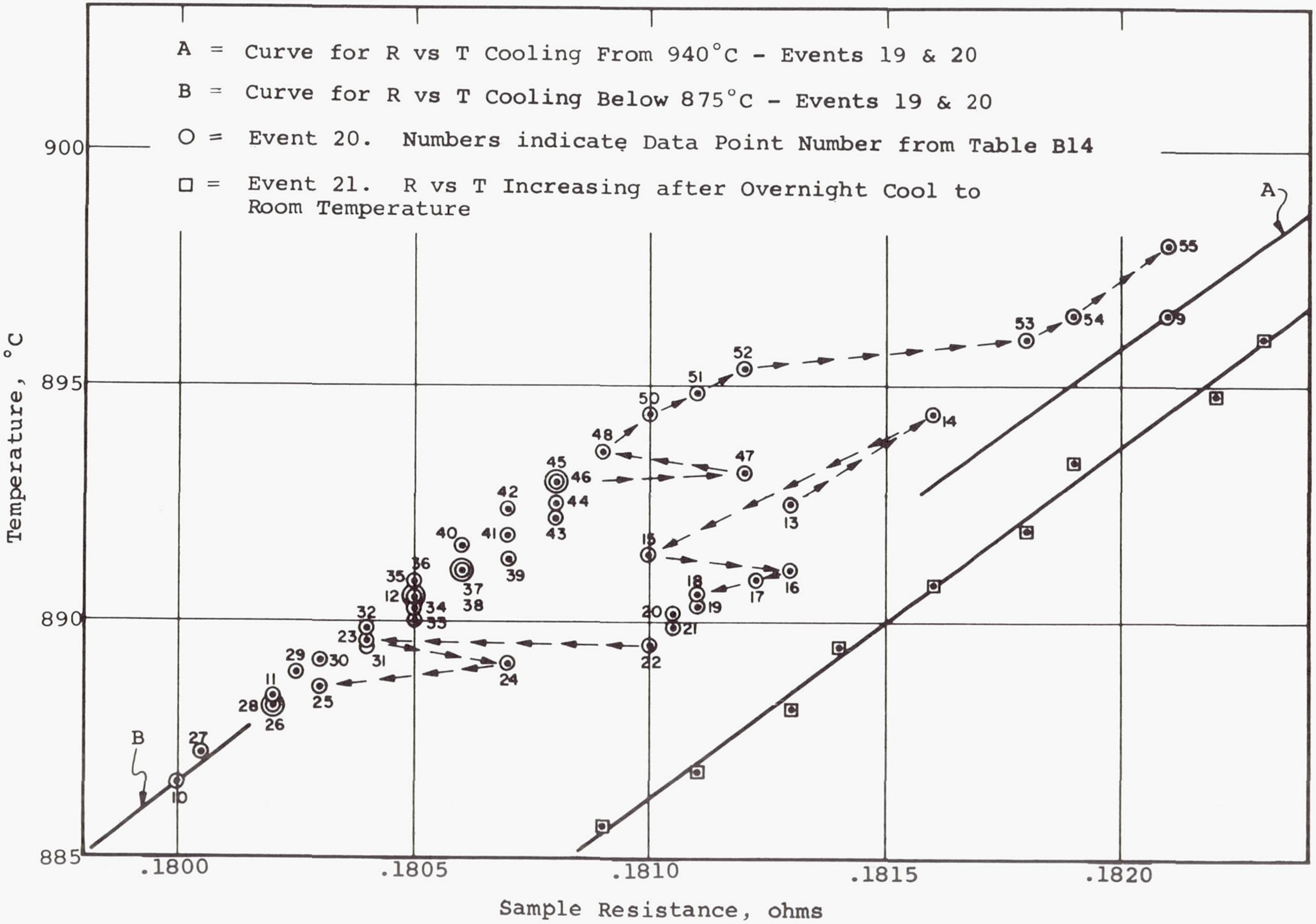


Figure 25 - Anomoly in Resistance vs Temperature Curve for Tantalum Containing Oxygen.

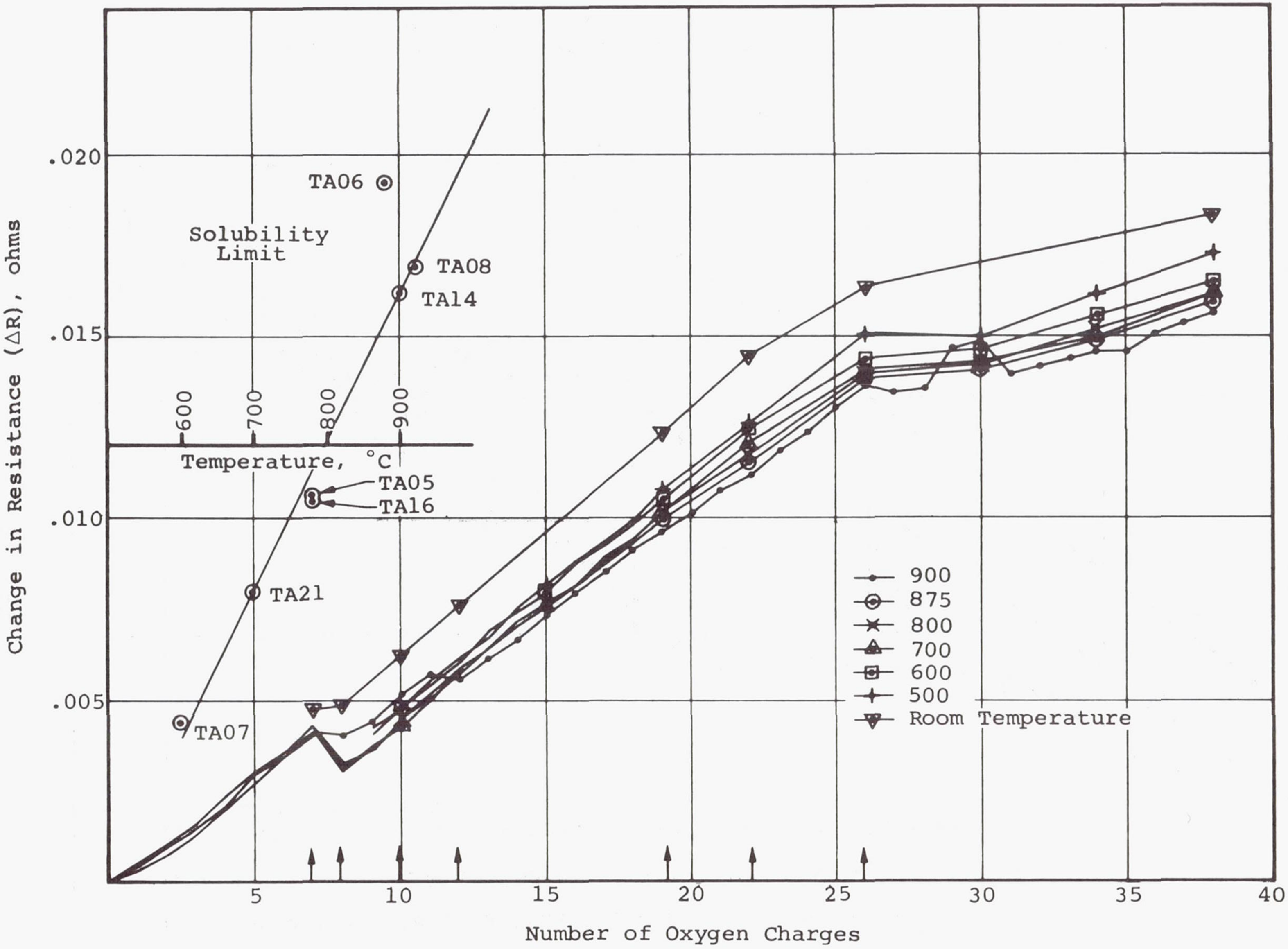
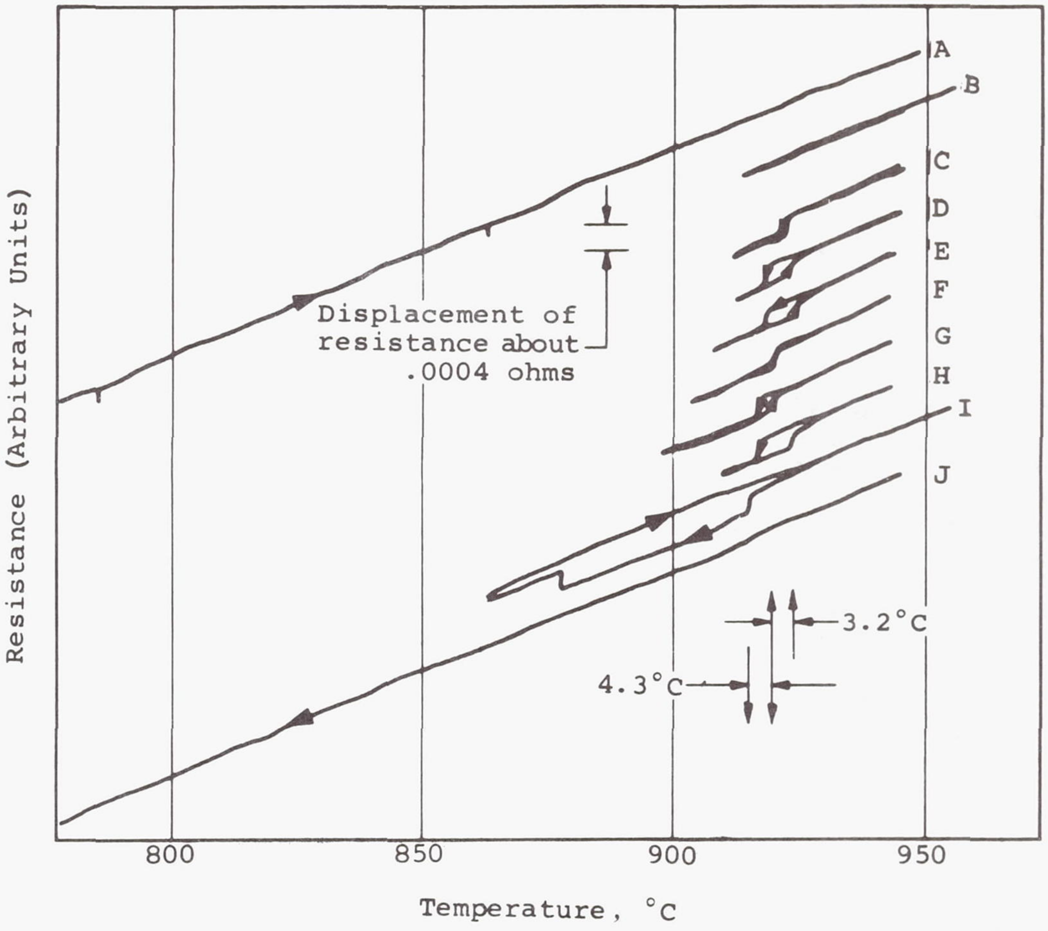


Figure 26 - Change in Resistance vs. Number of Oxygen Charges and Temperature for Tantalum - Run Ta20.



KEY

A	Heating from room temperature to 950°C.
B	Cooling from 950°C (3.5 ma*) and return to 950°C. Attempt to find jog. No jog found. Heat to 1400°C for 10 min.
C	Cooling from 950°C (3.0 ma Current) and return to 950°C. Jog found.
D	Reproduce C.
E	Cooling at furnace setting of 2.0 ma, followed by reheating at maximum rate.
F	Same as E, but with 1.0 ma.
G	Same as E, but power off.
H	Same as E, but with 3.5 ma.
I	Same as G.
J	Cool to room temperature with power off.

* Furnace set current, used to control rate of furnace heating or cooling.

Figure 27 - X-Y Recorder Plots of Resistance vs Temperature Offset for Oxygen-doped Tantalum - Run Ta20.

is correct, then the presence of oxides indicates that the solubility had been exceeded. This indicated to us that the solubility was lower than previously reported and led to our reinterpreting the previous data as outlined in the preceding section.

7) Although two significant changes were found in the slope of the curve of ΔR versus number of changes (Figure 25), these occurred after an overnight shutdown. Further work is required to learn whether this was coincidence.

The data from Run Ta20 were reduced to ΔR versus the number of charges of oxygen for each temperature (Figure 25). Overnight shutdowns are indicated by the vertical arrows. Nordheim's Rule is demonstrated by the equal slopes (ΔR vs O_2 pulse) for each temperature through the 7th pulse. Between the 7th and 8th pulses (an overnight room temperature shutdown), the resistance dropped at all temperatures except room temperature. The same slope, after this disturbance is carried to pulse 26 after which (again an overnight shutdown) the slope decreases for all temperatures; the data also show more scatter. We are uncertain whether the very slight divergence of slopes at different temperatures after the 8th pulse is significant.

Figure 25 includes the solubility curve from Figure 21 plotted at the left. We partially interpret the data as follows: The first 7 pulses show a consistent increase in ΔR according to Nordheim's Rule, even though the solubility at 500°C (the lowest temperature reached during the R versus T traverse after each pulse) appears to have been exceeded at about the 2nd or 3rd pulse. Even the shutdown to room temperature after the 3rd pulse had no effect on the data, although the uncertainty in the calibration curve is such that possibly we had not reached the solubility limit by the third pulse. We interpret the data as showing that oxygen remained in solution until the room temperature shutdown after the 7th pulse. Thereafter, some fraction of the precipitated oxide failed to redissolve upon heating.

It is reasonable to expect that precipitation reactions will be sluggish around 500°C. Gebhardt[1] has demonstrated that days are required to fully precipitate the metallic oxide in the Cb-0 system at 500°C. If the solubility is exceeded in this temperature range, there will be a set of cooling rates and excess concentration at which precipitation can be detected. Apparently, we did not reach these conditions until after the 7th pulse when the oxygen content corresponded to a solubility limit near 600°C. An unexplained aspect is that the precipitates which (presumably) formed overnight between the 7th and 8th pulses could not be redissolved on heating to 1200°C. Also unexplained is why the ΔR at 900°C did not drop to its "normal" position during the 8th through 11th pulses, as it did for pulse 12 and beyond. This may be related to the resistance shift at 900°C mentioned earlier because the amount of the discrepancy is about the same as the offset. After pulse 12, the curves lie in the order of temperature with the lowest temperature (ambient) showing the highest ΔR. This departure from Nordheim's Rule appears to be about inversely proportional to absolute temperature, so, possibly, represents a thermally activated precipitation process.

No further discontinuities appear although the data become more irregular after the slope decreases at pulse 26. The change in slope corresponds to a solubility limit at 850° to 900°C. Possibly a stable oxide coating is forming and is interfering with the dissolution of oxygen after this point.

There were two additional features which remain unexplained:

1) In the later pulses of both Ta20 and Ta21, the resistance changed more rapidly during the first one quarter or one third of the oxygen exposure part of the pulse, and, in fact, completed approximately 90% of the change during this time (see Appendix D).

2) It was observed in Ta20 on pulses 27, 31 and 35 (those after a temperature traverse to 500°C) that the resistance at 950° was higher than before the excursion to 500°C and would drop when oxygen was admitted rather than rise as usually occurred.

The resistance shift at 900°C was found after the 7th pulse when the 950°C resistance point was found to be displaced from a line extrapolated through the 800° and 875°C points. Assuming at first that a new slope had been established somewhere above 875°C, a series of readings were taken at 10°C intervals downward from 950°C to determine the break point.

A shift was found between the 906° and 896°C points. Resistance was measured at increasingly smaller increments with the results shown in Figure 25. It was first thought that a hystersis effect was being shown, but later the true nature of the shift was found. The resistance shift for a tiny change in temperature was as much as .0006 ohms. Moreover, the shift was very rapid, much faster than could be followed by the galvanometer.

At the end of the run, further investigation was made with an x-y recorder hooked to the specimen thermocouple and in place of the galvanometer in the bridge circuit. The specimen was heated and cooled so that a resistance versus temperature curve was plotted. Many traverses were made but the series shown in Figure 27 is most illustrative of the results.

Curve A traces the heating of the specimen from room temperature to 950°C. The series of traverses is outlined in the figure. After heating from room temperature, no shift was found. After annealling at 1400°C, however, a downward shift was noted at about 920°C. In 7 traverses, the downward shifts fell within a temperature range of 3.2°C.

An attempt was made to relate the temperature at which the downward jog occurred to cooling rate. The furnace control current was manually set lower than that required to maintain the sample temperature above 900°C. The control currents are shown in the figure. The lower selected currents produced, of course, the faster cooling rates. The actual cooling rates were not measured, but varied between about 0.5 and 5°C per second. It might be expected that the lower shift temperatures might be associated with the higher cooling rates. A poor correlation was found (Table 7).

Trace I was to be the shutdown run to room temperature. The cooling rate was the fastest obtainable with the furnace in place. The downward jog occurred as expected, but an upward jog occurred on further cooling at approximately 878°C. This was the first time this upward jog had been seen. No jog was seen on reheating. After cooling once more from 950°C (Trace J), only the slightest

Table 7. Correlation between Cooling Rate and Temperature of Resistance Offset in Curves of Figure 27.

Temperature of Offset (highest to lowest)	C	F	E	D	G	H	J
Cooling Rate (slowest to fastest)	H	C	D	E	F	G	J
Furnace Controller Current, amps.	3.5	3.0	3.0	2.0	1.0	0.0	0.0

suggestion of an offset is visible. Presumably the effect could be fully recovered by heating to 1400°C for ten minutes as had been done between Trace B and Trace C.

No reference to such a phenomenon was found in the literature investigation that followed. The characteristics of the offset are similar to effects observed in diffusionless transformations such as Fe to martensite and metastable β brass to martensitic brass. It is possible that at 1400°C some oxide is taken into solution. Upon cooling, a metastable metallic phase is formed at 940°C. Its disappearance at 878°C indicated by another precipitous change in resistance suggests another diffusionless transformation to the stable oxide.

The above hypothesis is only tentative and serves only as a guideline in planning further research.

B. The Solubility of Oxygen in Cb

Ten runs were made in an attempt to obtain solubility data (Tables B18 to B26, Appendix B). Data from four runs were selected for reduction because the runs started with unengassed Cb and showed an initial linear segment: runs Cb04,03, 09 and 08, which were performed at 909, 906, 800 and 700°C, respectively. The data were plotted as ΔR versus number of oxygen pulses, and ΔR versus square root of number of oxygen pulses (Figure 28). The first departure from linearity was determined as outlined for tantalum. These values are listed in Table 8 and plotted in reduced form in Figure 29. Results were too inaccurate to warrant obtaining a calibration curve.

C. Trial Runs

1. Ta-10W

One trial run was made with Ta-10W alloy to determine the engassing characteristics of this alloy. The data for this run are tabulated in Appendix B, Table B27. Engassing was done at 900°C using 2.00 torr pulses. The first 10 pulses and resistances are reported in Table 9. These data are plotted as ΔR versus number of oxygen pulse in Figure 30. The remaining pulses gave erratic readings, and ultimately an open circuit developed in the bridge at the specimen.

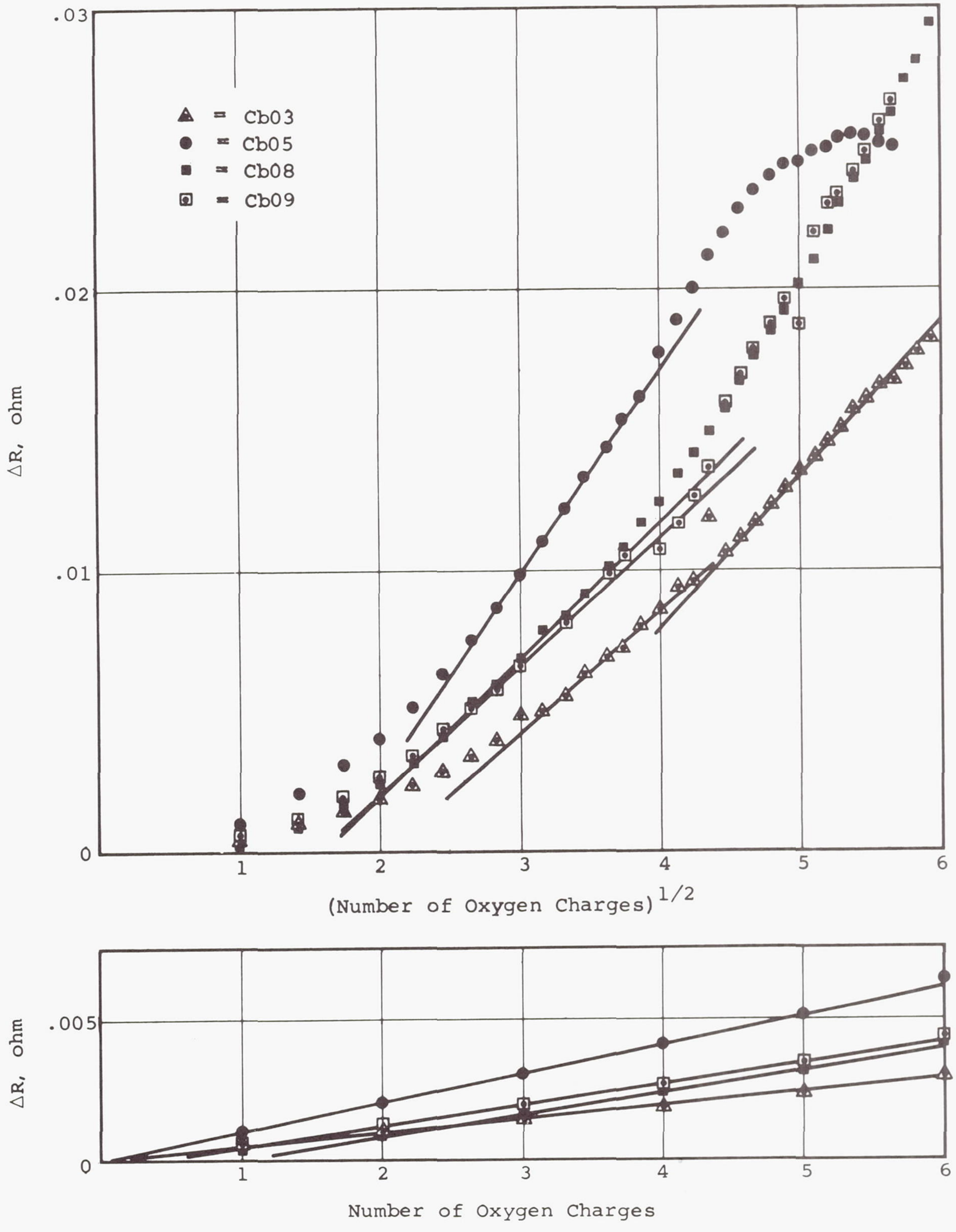


Figure 28 - Change in Resistance vs Number of Oxygen Charges for Columbium.

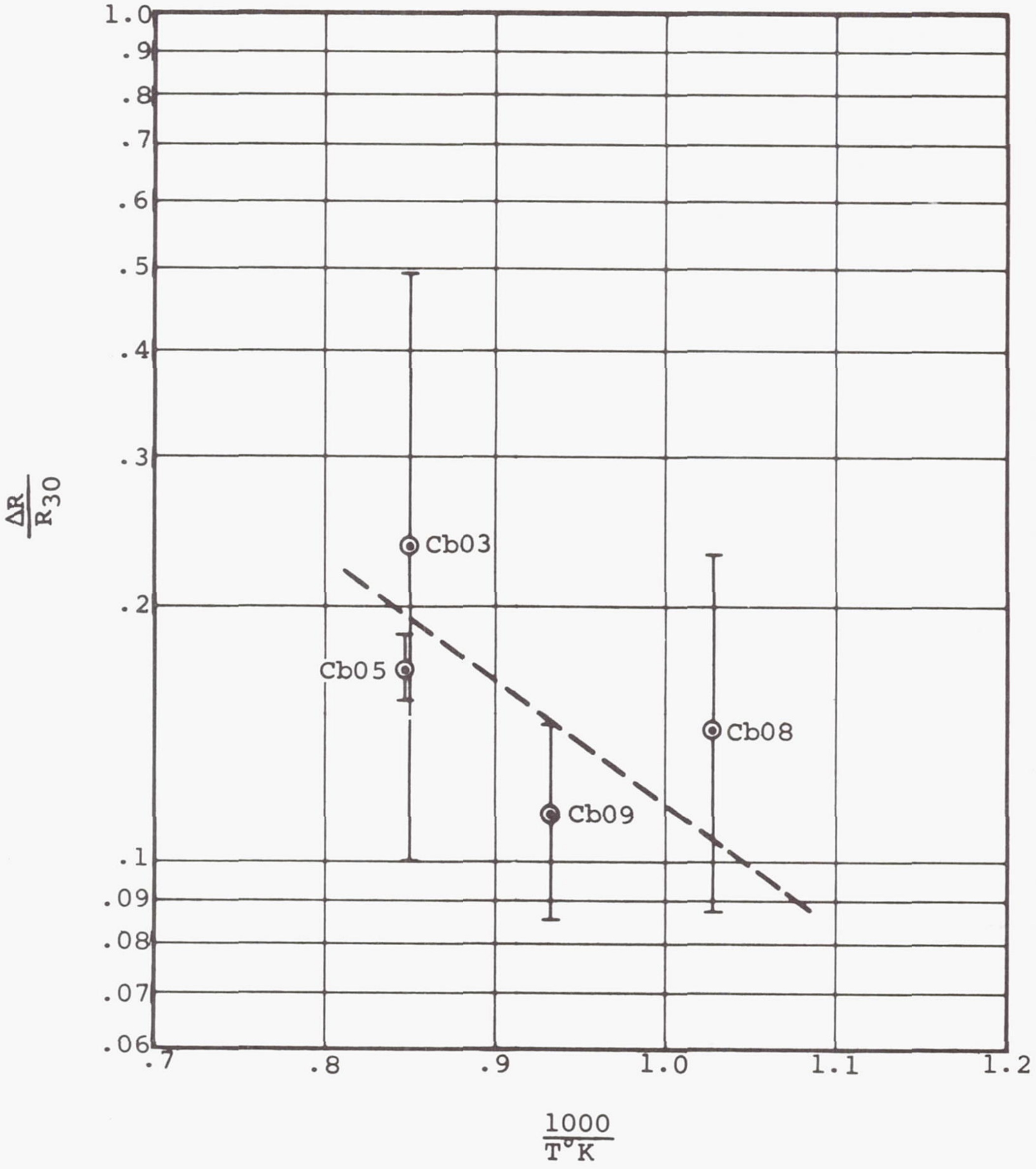


Figure 29 - Intercept of Inital Linear and First Parabolic Sections of Curves of Reduced Resistance Change vs. Number of Oxygen Charges as Function of Reciprocal Temperature.

Table 8. Intercept of Initial Linear and First Parabolic Sections of Curves of Resistance versus Number of Oxygen Pulses for Columbium.

Run No.	Temp. °C	ΔR^*	Estimated Error	R_{30}	$\Delta R^*/R_{30}$
Cb05	909	.0108	.0009	.0643	.168
Cb03	906	.0145	.0337	.0619	.234
Cb09	800	.0067	.0017	.0585	.114
Cb08	700	.0088	.0051	.0610	.142

Table 9. Resistance Data from Trial Run with Ta-10W.

Pulse Number	R, Ohms	ΔR, Ohms
0	.1232	0
1	.1253	.0021
2	.1273	.0041
3	.1310	.0078
4	.1334	.0102
5	.1372	.0140
6	.1423	.0191
7	.1477	.0245
8	.1521	.0489
9	.1622	.0590
10	.1739	.0707

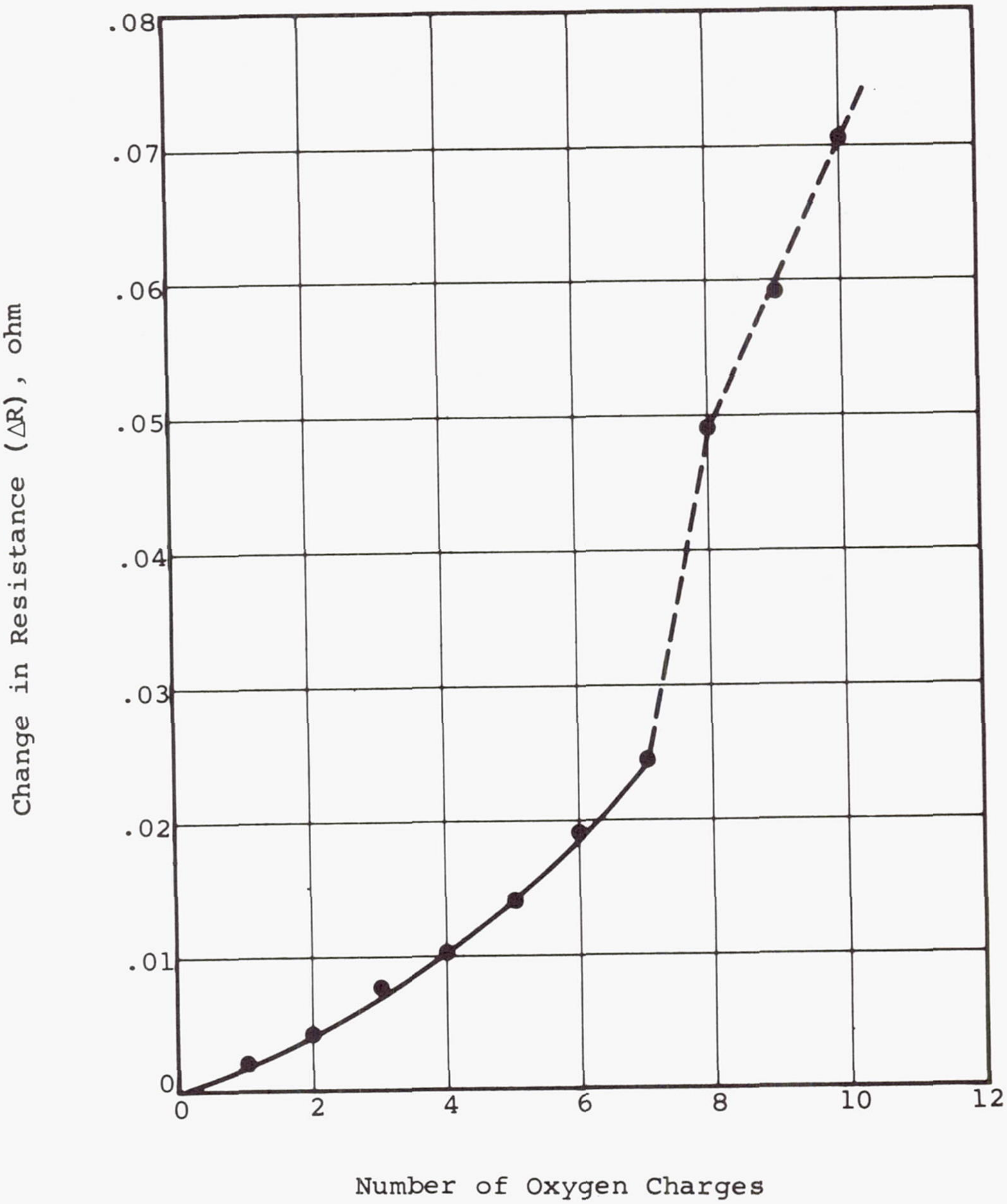


Figure 30 - Change in Resistance vs. Number of Oxygen Charges for Ta-10W Alloy.

The resistance pattern was different from that of Cb and Ta. More preliminary work is required with this alloy.

2. Cb-1Zr

An isothermal pulse run was performed with Cb-1Zr but the data provided no information due to its erratic nature.

3. DIFFUSION OF OXYGEN IN Ta

The prior work on the diffusion of oxygen in Ta by Gebhardt(4) using a technique similar to ours, showed a very small change in slope at the interface after 3 hours at 1010°C. The general oxygen level in the unengassed half had risen from approximately .03 to .10 atom percent during this time. The maximum time used by Gebhardt to obtain a measureable gradient between the two halves of the specimen was 278 hours. The longer time specimens contained progressively higher general oxygen levels in the unengassed end. The 278 hour specimen contained approximately 0.2 atom percent oxygen in this region. This suggested either a much more rapid diffusion at low oxygen concentration or gradual contamination from the furnace atmosphere.

On the assumption that the former was true, Ta22 was designed to detect and measure this rapid low oxygen concentration diffusion. The six hours at 900°C did not show any change in resistance in the specimen segments D-E and F-G. Neither could the microhardness gradient be unambiguously interpreted as representing oxygen diffusion.

The microhardness readings are tabulated in Table 10 and plotted in Figure 31. Although a definite diffusion gradient is shown, the data must be compared to those from a run which was shut down immediately after engassing to determine whether the gradient is different from the starting gradient.

Results demonstrated the feasibility of the approach, and useful data should be expected from the technique, particularly at higher temperatures.

Interference by the attached leads might be expected, but the use of Pt barriers between the sample and leads was expected to minimize this.

Table 10. Microhardness Data from Trial Diffusion Run.

Distance Along Wire[1] Microns	INDENTATION LENGTH, MICRONS[2]		
	Transverse	Axial	Average
0	67.0	58.6	62.8
200	67.1	58.2	62.6
400	65.9	60.7	63.2
600	86.3	62.3	64.3
800	66.0	61.9	63.9
1,000	66.3	59.1	62.7
1,200	67.0	63.2	65.1
1,400	64.5	59.6	62.0
2,200	69.0	56.0	62.5
2,375	62.5	59.8	61.2
2,575	54.1	63.3	58.7
2,955	60.2	55.8	58.0
3,805	65.0	64.8	64.9
6,485	61.4	57.3	59.3
9,642	60.1	53.0	56.5
9,787	60.2	47.3	58.7
11,780	58.0	51.5	54.7
12,885	54.7	46.5	51.6
13,885	54.3	46.3	50.3

(1) Measured from center line of Mo lead # F (see Figure 4).
(2) Indenter is square, but because of wire curvature, indentation lengths are unequal. Distances measured across diagnals.
(3) Scatter due in part to rough as-received surface.

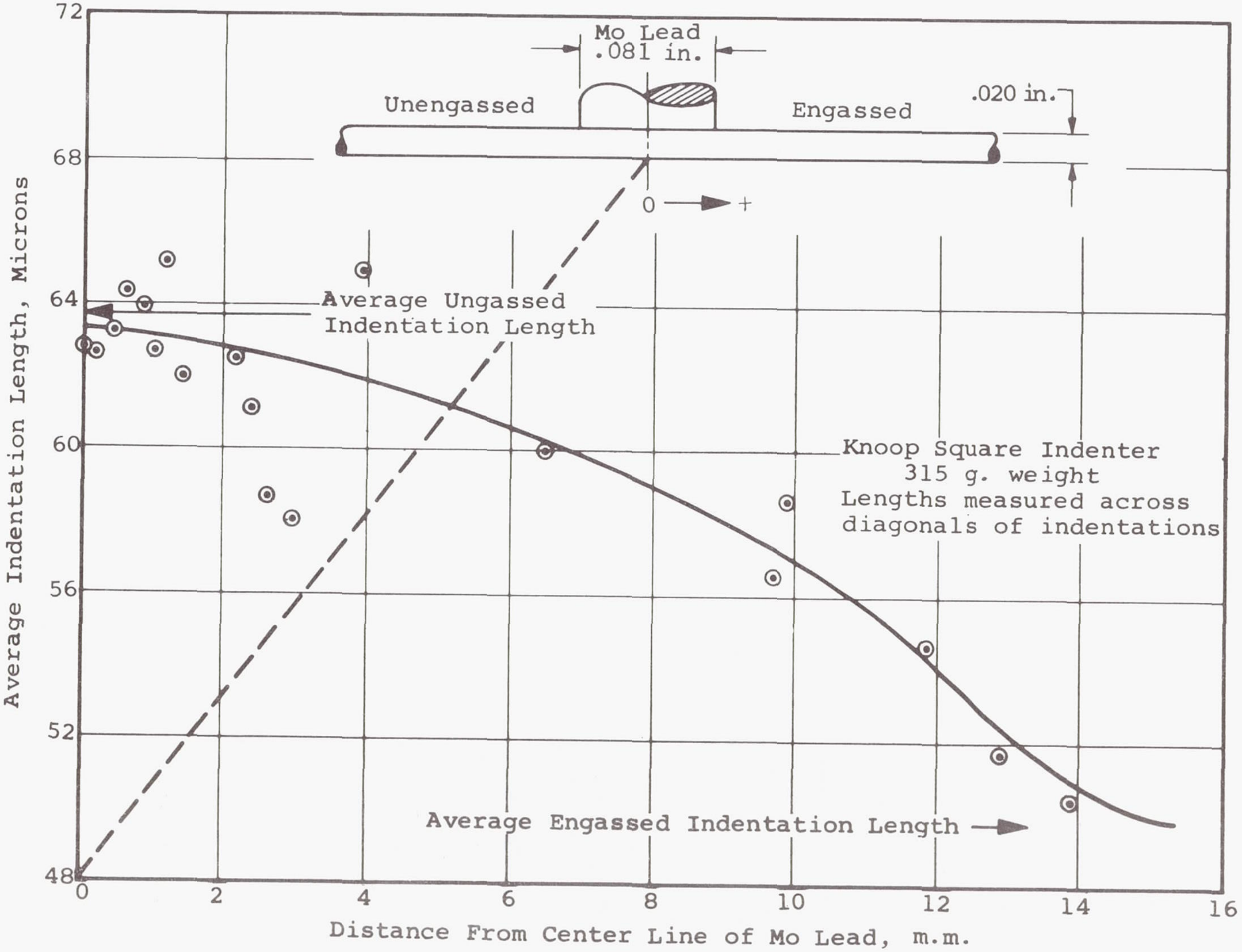


Figure 31 - Hardness Profile of Tantalum Diffusion Specimen

IV. CONCLUSIONS AND RECOMMENDATIONS

1. An apparatus was constructed in which resistance of wire specimens could be measured up to 950°C. Principal features follow:

a) Base pressure: $<1 \times 10^{-8}$ torr

b) Temperature stability: ±0.02°C

c) Precision of resistance measurement: 0.02% at 0.2 ohm level.

d) Temperature control precision: if the resistance of a specimen has been measured at a temperature and the furnace then cooled to room temperature, the resistance could be reproduced within ± 0.0001 ohm by dialing the previous temperature setting in the controller and waiting for the temperature to stabilize (provided no metallurgical change occurred in the sample).

e) Determination of partial pressure of gases contributing to the furnace pressure.

2. A method was developed for determining the location of phase boundaries in a metal-oxygen system. With this method, a metal wire sample is exposed to reproducible pulses of oxygen and the resistance of the wire measured after each pulse or group of pulses. Phase boundaries are located from changes in slope of the resulting curve of change in resistance vs number of pulses.

3. The ΔR vs number-of-pulse data for commercially pure Ta showed an initial linear relationship followed by up to 4 parabolic sections. By interpreting the intersection of the linear and first parabolic sections as the phase boundary, a curve was derived for the solubility of oxygen in Ta. The equation for this curve is

$$\log \text{concentration (ppm)} = 4.130 - \frac{1279}{T^{\circ}K}$$

4. Similar data were obtained for Cb but accuracy was not thought to be sufficient to warrant obtaining the calibration curve necessary to convert the data to solubility values.

5. An anomoly in the resistance vs temperature curve for oxygen-doped Ta was found at about 900°C.

6. Changes in slope of the resistance vs temperature curve for oxygen-doped Ta were found at about 450°C and 750°C. These slope changes persisted as oxygen was added, up to about 0.12 wt.%. They correspond roughly to transition of the metal suboxides reported by others.

7. Analysis of Ta specimens by neutron activation analysis resulted in data about 1 order of magnitude less precise than desired. This was largely caused by the coil shape of the specimen. A more compact form of the sample should result in a smaller error.

8. Because of erroneous interpretation of the data through most of the program, most of the data were taken in the 2 phase region. The limited single phase data often resulted in a higher than desired uncertainty in the intersection of the curve segments mentioned in conclusion 3.

9. Although the method appeared to produce rapid and very precise data, further work should include the following improvements:

   a) Independent verification of the presence of a second phase. A hot stage x-ray diffraction unit should be used because of the extremely rapid transitions observed. More liberal use of metallographic analysis is also recommended.

   b) Use of a method of oxygen analysis capable of rapid answers. A replicate sample suspended from a recording micro-balance is suggested.

   c) Inclusion of very pure specimens for reference purposes.

   d) Use of the 1500°C furnace.

10. Certain discrepancies and unexplained phenomena should be resolved.

   a) The difference in "solubility" points obtained by the isothermal pulse and isothermal,isobaric methods.

   b) The reason for oxygen entering the specimen only in the early part of the pulse. This phenomenon may be tied in with a).

c) The rapid entry of oxygen into the specimen in the linear region and the slow diffusion rate observed in the trial diffusion run.

d) The anomolies in the R and ΔR vs T data for the Ta-0 system.

V. REFERENCES

1. Promisel, N., ed., *Science and Technology of Selected Refractory Metals*, McMillan, New York, 1964.
2. Gebhardt, E., and Seghezzi, H.D., *Plansee Proc*, Ruette, Austria, 1958, p. 280, also see Ref. 1.
3. Hurlen, R., *J. Inst. Metals*, 87, 235 (1958).
4. Gebhardt, E., Seghezzi, H.D., and Stegherr, A., *Z. Metallkde 48*, 624 (1957).
5. Vaughn, D.A., Stewart, O.M., and Schwartz, C.M., *Trans AIME 221*: 937 (1961).
6. Gebhardt, E. and Preisendanz, H., *Z. Metallkde 46*: 560 (1955).

APPENDIX A

MATERIALS SPECIFICATIONS AND ANALYSIS

I. CHEMICAL SPECIFICATIONS FOR TEST MATERIALS

1) Columbium (Niobium)

1. FORM - Wire

2. COMPOSITION - The chemical composition of wire shall be as follows:

ELEMENT	WT. %		ELEMENT	WT. %	
Cb	99.8	Min.	Mo	.02	Max.
C	.005	Max.	Ni	.02	Max.
O_2	.03	Max.	W	.02	Max.
N_2	.015	Max.	Zr	.02	Max.
H_2	.0005	Max.	V	.01	Max.
Fe	.01	Max.			
Si	.01	Max.			
Ta	.05	Max.			
Ti	.02	Max.			

3. CONDITION - Fully recrystallized

2) Columbium - 1Zr

1. FORM - Fully recrystallized wire

2. COMPOSITION - The chemical composition of the wire shall be as follows:

ELEMENT	MAXIMUM	MINIMUM
Zirconium	1.2%	0.8%
Carbon	100 p.p.m.	-
Oxygen	300 "	-
Nitrogen	300 "	-
Hydrogen	10 "	-
Tantalum	1000 "	-
Tungsten	500 "	-
Iron	500 "	-
Silicon	300 "	-
Titanium	500 "	-
Molybdenum	1000 "	-

ELEMENT	MAXIMUM	MINIMUM
Boron	1	-
Cadmium	5	-
Cobalt	30	-
Hafnium	100	-
Lead	50	-
Manganese	100	-
Nickel	200	-
Vanadium	200	-
Columbium	Balance	-

3. CONDITION - All wire shall be in the fully recrystallized condition.

3) FS-85

1. FORM - Fully recrystallized wire

2. COMPOSITION - FS-85 (Cb-28Ta-10.5W-0.9Zr) Alloy

ELEMENT	MINIMUM CONTENT ppm	MAXIMUM CONTENT ppm
Carbon	-	100
Nitrogen	-	75
Oxygen	-	300
Hydrogen	-	10
Molybdenum	-	200
Nickel	-	50
Cobalt	-	50
Iron	-	50
Tantalum	26 w/o	29 w/o
Tungsten	10 w/o	12 w/o
Zirconium	0.6 w/o	1.1 w/o
Columbium	Remainder	-

3. CONDITION - All wire shall be in the full recrystallized condition.

Tantalum

1. FORM - Wire

2. COMPOSITION - The chemical composition of the material shall be the following:

ELEMENT	MAXIMUM WT %	MINIMUM WT %
Tantalum		99.90
Carbon	.010	
Oxygen	.010	
Nitrogen	.005	
Hydrogen	.002	

3. CONDITION - Material shall be in the recrystallized state.

5) Tantalum - 10 Tungsten

1. FORM - Fully annealed wire

2. COMPOSITION - The chemical composition of the material shall be as follows:

ELEMENT	
Carbon	50 PPM Max.
Oxygen	70 PPM Max.
Nitrogen	30 PPM Max.
Hydrogen	6 PPM Max.
Columbium	1000 PPM Max.
Iron	70 PPM Max.
Molybdenum	300 PPM Max.
Nickel	70 PPM Max.
Tungsten	8.5-11 wt. %
Tantalum	Balance

3. CONDITION - All material will be in the fully recrystallized condition.

6) T-222

1. FORM - Fully recrystallized wire

2. COMPOSITION - T222 (Ta-9.8W-2.4Hf-0.01C) Alloy

ELEMENT	MINIMUM CONTENT ppm	MAXIMUM CONTENT ppm
Carbon	80	175
Nitrogen	-	50
Oxygen	-	100
Hydrogen	-	10
Columbium	-	1000
Molybdenum	-	200
Nickel	-	50
Cobalt	-	50
Iron	-	50
Vanadium	-	20
Tungsten	9.6 w/o	11.2 w/o
Hafnium	2.2 w/o	2.8 w/o
Tantalum	Remainder	-

3. CONDITION - All wire shall be in the fully recrystallized condition.

II. MATERIALS LOT ANALYSIS

MATERIAL		Cb-1Zr	FS-85	Ta-10W	T-222	**Ta**
Mfg Lot No.:		80B2341 80B781	85D695	60B901	22D1002	MG 41
	C	20	20	10	139	10
	O	50	40	10	10	52
	N	60	20	10	10	10
	Zr	0.90%	0.97%	10	500	5
	Ta	600	28.0 %	bal.	bal.	bal.
	Ti	50	-	10	10	5
	Fe	70	50	10	10	50
	Ni	70	-	10	10	5
ANALYSIS PPM	W	260	10.25%	9.5%	8.9%	5
	Si	100	100	-	10	5
	Mo	200	-	50	10	30
	Cb	bal.	bal.	90	500	470
	V	-	-	10	10	5
	Co	-	-	10	10	5
	Mn	-	-	-	10	5
	Pb	-	-	-	10	-
	Cu	-	-	-	10	5
	Cr	-	-	-	10	5
	H	-	-	-	3.7	-
	Hf	-	-	-	2.21%	-
	Mg	-	-	-	10	5
	Ca	-	-	-	-	5
	Al	-	-	-	-	5
	Sn	-	-	-	-	5

APPENDIX B

RAW DATA FROM INDIVIDUAL RUNS

TABLE B1

Run Ta05 Data
Isothermal Pulse Run at 780°C

Sample Wt.: 3.689 g
Pre run resistance = 0.0619 ohm at 32.0°C
Post run resistance = 0.1056 ohm at 33.3°C
Oxygen charge size = 300 cc at 1 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
0	.1912	0
1	.1919	.0007
2	.1920	.0008
3	.1932	.0020
4	.1947	.0035
5	.1962	.0050
6	.1978	.0066
7	.1982	.0070
8	.2000	.0088
9	.2016	.0104
10	.2034	.0122
11	.2052	.0140
12	.2069	.0157
13	.2087	.0175
14	.2104	.0192
15	.2120	.0208
16	.2136	.0224
17	.2152	.0240
18	.2167	.0255
19	.2182	.0270
20	.2196	.0284
21	.2210	.0298
22	.2223	.0311
23	.2237	.0325
24	.2249	.0337
25	.2259	.0347
26	.2269	.0357
27	.2280	.0368
28	.2290	.0378
29	.2299	.0387
30	.2308	.0396
31	.2316	.0404
32	.2324	.0412
33	.2330	.0418
34	.2337	.0425
35	.2343	.0431
36	.2348	.0436
37	.2353	.0441
38	.2358	.0446
39	.2361	.0449
40	.2366	.0454
41	.2370	.0458
42	.2373	.0461
43	.2377	.0465
44	.2380	.0468

TABLE B2

Run Ta06 Data
Isothermal Pulse Run at 880°C

Sample Wt.: 4.231 g
Pre run resistance = .0738 ohm at 29.8°C
Post run resistance = .1139 ohm at 24.8°C
Oxygen charge size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
0	.2468	0
1	.2503	.0035
2	.2544	.0076
3	.2587	.0119
4	.2631	.0163
5	.2676	.0208
6	.2719	.0251
7	.2758	.0290
8	.2798	.0330
9	.2836	.0368
10	.2871	.0403
11	.2906	.0438
12	.2940	.0472
13	.2969	.0501
14	.2995	.0527
15	.3017	.0549
16	.3036	.0568
17	.3052	.0584
18	.3067	.0599
19	.3079	.0607
20	.3089	.0617
21	.3100	.0632
22	.3108	.0640
23	.3116	.0648
24	.3124	.0656
25	.3130	.0662
26	.3137	.0669
27	.3141	.0673
28	.3147	.0679
29	.3153	.0685
30	.3158	.0690
31	.3161	.0693
32	.3166	.0698
33	.3171	.0703

TABLE B3

Run Ta07 Data
Isothermal Pulse Run at 598°C

Sample Wt.: 4.0131 g
Pre run resistance = .0700 ohm at 25.3°C
Post run resistance = Not Recorded
Oxygen charge size: 300 cc at 1 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
0	.1838	.0008
1	.1847	.0009
2	.1859	.0021
3	.1872	.0034
4	.1885	.0047
5	.1899	.0061
6	.1906	.0068
7	.1912	.0074
8	.1920	.0082
9	.1924	.0086
10	.1931	.0093
11	.1937	.0099
12	.1942	.0104
13	.1947	.0109
14	.1952	.0114
15	.1960	.0122
16	.1965	.0127
17	.1969	.0131
18	.1974	.0136
19	.1979	.0141
20	.1983	.0145
21	.1987	.0149
22	.1992	.0154
23	.1997	.0159
24	.2002	.0164
25	.2006	.0168
25	.2012	(1)
25	.2049	(2)

(1) Recorded temperature 596.4°C
(2) Recorded temperature 601.1°C

TABLE B4

Run Ta08 Data
Isothermal Pulse Run at 921°C

Sample Wt: 3.418 g
Pre run resistance = Not recorded
Post run resistance = .0957 ohm at 39.2°C
Oxygen charge size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS	
0	.1734	--	(1)
0	.1706	0	(2)
1	.1738	.0032	
2	.1770	.0064	
3	.1810	.0104	
4	.1850	.0144	
5	.1886	.0180	
6	.1924	.0218	
7	.1953	.0247	
8	.1973	.0267	
9	.2012	.0306	
10	.2039	.0333	
11	.2067	.0361	
12	.2089	.0383	
13	.2110	.0404	
14	.2129	.0423	
15	.2145	.0439	
16	.2158	.0452	
17	.2169	.0463	
18	.2178	.0472	
19	.2186	.0480	
20	.2192	.0486	
21	.2197	.0491	
22	.2203	.0497	
23	.2207	.0501	
24	.2211	.0505	
25	.2215	.0509	
26	.2219	.0513	
27	.2222	.0516	
28	.2225	.0519	
29	.2229	.0523	
30	.2233	.0527	
31	.2237	.0531	
32	.2240	.0534	
33	.2244	.0538	
34	.2247	.0541	
35	.2250	.0544	
36	.2254	.0548	
36	.2236	.0530	
36	.2245	.0539	

(1) Temperature 943.7°C
(2) Temperature 921.4°C

TABLE B5

Run Ta10 Data
Isothermal Pulse Run for Calibration Curve
Run Temperature 900°C

Sample Wt.: 3.660 g
Pre run resistance = .0642 ohm at 41.9°C
Post run resistance = .0922 ohm at 49.0°C
Oxygen charge size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
0	.2080 (1)	0
1	.2113	.0033
2	.2154	.0074
3	.2195	.0115
4	.2236	.0156
5	.2277	.0197
6 (2)	.2321	.0241
6 (2)	.2322	.0242
6 (2)	.2321	.0241
6 (2)	.2323	.0243
6 (2)	.2324	.0244
6 (2)	.2324	.0244
6 (2)	.2325	.0245
6 (2)	.2325	.0245
6 (2)	.2326	.0246

(1) At 902.2°C.
(2) Resistance after 6 pulses monitored over 3 hour period

TABLE B6

Run Tall Data
Isothermal Pulse Run for Calibration Curve
Run Temperature 901°C

Sample Wt.: 3.919 g
Pre run resistance = .0702 ohm at 53.3
Post run resistance = .0785 ohm at 42.4
Oxygen charge size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
0	.2207	0
1	.2240	.0033
2	.2279	.0072
3	.2318	.0111 (1)
3	.2319	.0112 (1)
3	.2319	.0112 (1)

(1) Resistance after 3 pulses monitored over 2 hour period.

TABLE B 7

Run Ta12 Data
Isothermal Pulse Run for Calibration Curve
Run Temperature 900°C

Sample Wt.: 4.042 g
Pre run resistance = .0689 ohm at 33.1°C
Post run resistance = .1110 ohm at 32.0°C
Oxygen charge size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
0	.2271	0
1	.2305	.0034
2	.2344	.0073
3	.2386	.0115
4	.2429	.0158
5	.2473	.0202
6	.2517	.0246
7	.2558	.0287
8	.2598	.0327
9	.2637	.0366
10	.2678	.0407
10 (1)	.2680	.0409
10 (1)	.2681	.0410
10 (1)	.2672	.0401
10 (1)	.2671	.0400
10 (1)	.2678	.0407
10 (1)	.2679	.0408
10 (1)	.2679	.0408

(1) Resistance after 10 pulses monitored over 4 hour period

TABLE B8

Run Ta13 Data
Isothermal Pulse Run for Calibration Curve
Run Temperature 901°C

Sample Wt.: 4.026 g
Pre run resistance = .0686 ohm at 38.1°C
Post run resistance = .0708 ohm at 48.7°C
Oxygen charge size: Not Applicable

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
0	.2238	0

TABLE B9

Run Ta14 Data
Isothermal Pulse Run at 901°C

Sample Wt.: 4.037 g
Pre run resistance = .0742 at 36.3°C
Post run resistance = .1271 at 44.9°C
Oxygen pulse size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS		CHANGE IN RESISTANCE, OHMS
0	.2483		0
1	.2542		.0059
2	.2602		.0119
3	.2661		.0178
4	.2717		.0234
5	.2769		.0286
6	.2814		.0331
7	.2854		.0371
8	.2892		.0409
9	.2921		.0438
10	.2950		.0467
11	.2977		.0494
12	.2997		.0514
13	.3019		.0536
14	.3032		.0549
15	.3045		.0562
15	.2991	(1)	.0508
15	.1628	(2)	-
15	.3135	(3)	.0652

(1) After 12 hours at 900°C.
(2) After 1000°C 15 min. and 1200°C 1 hour at 63°C.
(3) After 1200°C 45 min. and 1350°C 30 min. and 900°C overnight.

TABLE B10

Run Ta15 Data
Isothermal Pulse Run at 781°C

Sample Wt.: 3.6891 g
Pre run resistance = .0619 (temperature no recorded)
Post run resistance = Not Recorded
Oxygen pulse size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
0	.1941 (1)	
0	.1932 (2)	
0	.1785 (3)	
0	.1756	0
1	.1774 (4)	.0018
1	.1770	.0014
1	.1773	.0017
2	.1811	.0055
2	.1811	.0055
2	.1814	.0058
3	.1843	.0087
3	.1847	.0091
3	.1847	.0091
4	.1883	.0127
4	.1883	.0127
4	.1882	.0126
5	.1912	.0157
5	.1918	.0163
5	.1918	.0163
6	.1959	.0203
6	.1962	.0206
6	.1963	.0207
7	.1988	.0232
7	.1984	.0228
7	.1989	.0233
8	.2019	.0263
8	.2022	.0266
8	.2021	.0265
9	.2046	.0290
9	.2045	.0289
9	.2045	.0289
9	.2066 (5)	.0310
9	.2067	.0311
9	.2067	.0311
9	.2078	.0322

(1) Temperature = 899°C
(2) Temperature = 903°C
(3) Temperature = 901°C
(4) 30 minute cycle with 3 resistance readings.
(5) Remaining readings unrecorded. Taken over unknown time. Terminated in sample short circuit.

TABLE B11

Run Ta16 Data
Isothermal Pulse Run at 781°C

Sample Wt.: 3.666 g (initial)
3.724 g (final)
Pre run resistance = .061 (temperature not recorded)
Post run resistance = .0903 at 34.0°C
.0896 at 28.0°C
Oxygen pulse size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
0	.061	
1	.1578	0
1 (1)	.1598	.0020
1	.1599	.0021
1	.1600	.0022
2	.1630	.0052
2	.1631	.0053
2	.1632	.0054
3	.1665	.0087
3	.1667	.0089
3	.1667	.0089
4	.1698	.0120
4	.1699	.0121
4	.1702	.0124
5	.1731	.0153
5	.1732	.0154
5	.1732	.0154
6	.1764	.0186
6	.1764	.0186
6	.1765	.0187
7	.1790	.0212
7	.1790	.0212
7	.1790	.0212
8	.1807	.0232
8	.1809	.0234
8	.1810	.0235

TABLE B 11

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
9	.1828	.0250
9	.1828	.0250
9	.1828	.0250
10	.1844	.0266
10	.1844	.0266
10	.1845	.0267
11	.1860	.0282
11	.1860	.0282
11	.1860	.0282
12	.1873	.0295
12	.1873	.0295
13	.1881	.0303
13	.1883	.0305
13	.1883	.0305
14	.1895	.0317
14	.1895	.0317
14	.1895	.0317
15	.1904	.0326
15	.1904	.0326
15	.1905	.0327
15	.1914	.0336
15	.1921	.0343
15	.1922	.0344
15	.1922	.0344
15	.1929	.0351
15	.1930	.0352
15	.1932	.0354
15	.1932	.0354
15	.1939	.0361
15	.1940	.0362
15	.1946	.0368
15	.1947	.0369
15	.1954	.0376
15	.1954	.0376

TABLE B11

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
15	.1959	.0381
15	.1964	.0386
15	.1964	.0386
15	.1970	.0392
15	.1971	.0393
15	.1971	.0393
15	.1975	.0397
15	.1977	.0399
15	.1981	.0403
15	.1982	.0404
15 (2)	.1987	.0409

(1) 30 minute cycle with 3 resistance readings
(2) Taken after 2 days at 780°C

TABLE B12

Run Ta17 Data
Isothermal Pulse Engassing at 900°C and Isoconcentration Runs

Sample Wt.: 3.655 g
Pre run resistance = .0617 at 28°C

Oxygen charge size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
0	.1995	0
1	--	--
2	.2077	.0082
3	.2093	.0098
4	.2151	.0156
5	.2213	.0218
6	.2246	.0251
7	.2283	.0288
8	.2314	.0319
9	.2352	.0357
10	.2394	.0399
11	.2429	.0434
12	.2455	.0460
13	.2487	.0492

TEMPERATURE °C		
924.7	.2487 (1)	.0492
"	.2486	.0491
908.1	.2460	.0465
"	.2459	.0464
891.7	.2434	.0439
"	.2434	.0439
875.0	.2409	.0414
"	.2412	.0417
858.4	.2392	.0397
"	.2385	.0390

TABLE B12

TEMPERATURE °C	RESISTANCE OF SAMPLE, OHMS		CHANGE IN RESISTANCE, OHMS
841.7	.2362		.0367
"	.2362		.0367
825.0	.2339		.0344
"	.2339		.0344
801.9	.2328	(2)	.0333
809.0	.2309		.0314
792.0	.2278		.0283
792.9	.2297		.0302
"	.2257		.0262
771.0	.2276		.0281
771.0	.2260		.0265
756.5	.2251		.0256
733.0	.2225		.0230
742	.2238		.0243
726	.2214		.0219
725.6	.2213		.0218
725.8	.2213	(3)	.0218
821.5	.2364		.0369
822.0	.2362		.0367
808.0	.2346		.0351
808.2	.2345		.0350
791.4	.2321		.0326
791.5	.2321		.0326
775.7	.2295		.0300
775.8	.2297		.0302
741.0	.2242		.0247
751.6	.2243		.0248
726.0	.2218		.0223
"	.2218		.0223
699.5	.2173		.0178
700.0	.2176		.0181
700.0	.2169	(4)	.0174
927	.2488		.0493
950	.2500		.0505
900	.2455		.0460
893	.2436		.0441

TABLE B12

TEMPERATURE °C	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
873	.2427	.0432
850	.2390	.0395
842	.2371	.0376
823	.2354	.0359
791	.2306	.0311
774	.2275	.0280
755	.2253	.0258
725	.2207	.0212
740	.2226	.0231
720	.2198	.0203
704	.2171	.0176
950.9	.2533 (5)	.0538
"	.2532	.0537
927.3	.2496	.0501
"	.2496	.0501
910.0	.2469	.0474
"	.2469	.0474
889.5	.2443	.0448
889.7	.2443	.0448
874.7	.2426	.0431
"	.2426	.0431
858.0	.2400	.0405
"	.2400	.0405
841.3	.2372	.0377
"	.2373	.0378
825.3	.2351	.0356
"	.2352	.0357
808.0	.2325	.0330
"	.2326	.0331
791.3	.2303	.0308
"	.2303	.0308
774.7	.2279	.0284
"	.2279	.0284
758.4	.2253	.0258
"	.2253	.0258

TABLE B12

TEMPERATURE °C	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
741.7	.2228	.0233
742.1	.2229	.0234
726.0	.2204	.0209
726.2	.2204	.0209
710.0	.2178	.0183
669.0	.2159	.0164
950.3	.2550 (6)	.0555
924.7	.2510	.0515
924.9	.2510	.0515
901.5	.2480	.0485
900.6	.2480	.0485
874.7	.2442	.0447
"	.2442	.0447
850.6	.2406	.0411
"	.2406	.0411
825.4	.2366	.0371
"	.2366	.0371
800.6	.2329	.0334
801.5	.2329	.0334
773.9	.2287	.0292
774.1	.2287	.0292
751.6	.2255	.0260
752.4	.2255	.0260
724.1	.2210	.0215
	.2210	.0215
702.9	.2174	.0179
	.2174	.0179
950.4	.2536 (7)	.0541
925.2	.2507	.0512
900.8	.2476	.0481
874.1	.2437	.0442
850.8	.2401	.0406
823.0	.2364	.0369
802.9	.2333	.0338
776.4	.2295	.0300

TABLE B12

TEMPERATURE °C	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
749.3	.2252	.0257
725.3	.2217	.0222
703.8	.2181	.0186

(1) Isoconcentration run: readings taken 15 minutes and 25 minutes after resetting temperature.
(2) Remaining data appeared erratic in both resistance and temperature. T.C. interference with I_B.
(3) Resistance heat to 1400°C for 1 hour
(4) After 700°C overnight
(5) New isoconcentration run
(6) 1450°C for 1 hour
(7) 1450°C for 1 hour

TABLE B13

Run Ta 19 Data
Isothermal Pulse Run at 800°C

Sample Wt.: 3.666 g
Pre run resistance = .0635 at 29.2°C
Post run resistance = Not Recorded
Oxygen pulse size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
0	.1933	--
0	.1933 (1)	--
0	.1932 (2)	--
0	.1923	0
1	.1934	.0011
2	.2148	.0125
3	.2174	.0151
4	.2189	.0166
5	.2200	.0177
6	.2203	.0180
7	.2208 (3)	.0185
8	.2237	.0214
9	.2254	.0231
10	.2268	.0245
11	.2285	.0262
12	.2296	.0273
13	.2314	.0291
14	.2319	.0296
15	.2329	.0306
16	.2344	.0321
17	.2356	.0333
18	.2361	.0338
19	.2375	.0352
20	.2385	.0362
21	.2392	.0369
22	.2404	.0381
23	.2412	.0389
24	.2420	.0397
25	.2428	.0405

TABLE B13

NUMBER OF OXYGEN PULSE	RESISTANCE OF SAMPLE, OHMS	CHANGE IN RESISTANCE, OHMS
26	.2436	.0413
27	.2451	.0428
28	.2460	.0437
29	.2466	.0443
30	.2471	.0448
31	.2474	.0451
32	.2480	.0457
33	.2487	.0464
34	.2495	.0472
35	.2509	.0486
36	.2515	.0492
37	.2516	.0493

(1) After 1 hour at 1400°C resistance heating
(2) After 1 hour at 1400°C resistance heating
(3) Operational error pulses 1 through 7, air in pulse not oxygen

TABLE B14

Run Ta20 Data
Combination Isothermal Pulse, Isoconcentration Run

TEMPERATURE, °C (1)					SAMPLE RESIST-ANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
EVENT	1 - Preanneal Data, 1/20/67								
36.3	-	37.8	-	37.0	.0517	1.1	-	-	1.01
407.5	8.0	406.4	8.0	416.2	.1102	-	-	-	1.02
604.0	9.3	604.2	9.3	613.4	.1375	1.4	600	.1356	1.03
701.9	9.9	-	-	711.8	.1510	1.4	700	.1493	1.04
EVENT	2 - Measure Slope of R vs T Near 500°C, 1/23/67								
503.8					.1241				2.01
505.3					.1243				2.02
501.3					.1238				2.03
503.8					.1241				2.04
501.5					.1238				2.05
500.9					.1237				2.06
						1.3	500	.1236	
EVENT	3 - Measure R vs T At Increasing Temperatures After Anneal At 1350°C, 1/24/67								
496.	8.6	495.4	8.6	504.3	.1242	1.40	500	.1236	3.01
539.5	8.9	538.8	8.9	548.0	.1304	1.40	550	.1307	3.02
584.2	9.3	588.2	9.3	598.0	.1373	1.40	600	.1376	3.03
639.0	9.6	637.8	9.6	648.0	.1442	1.40	650	.1445	3.04
689.5	9.2	688.4		698.7	.1510	1.40	700	.1512	3.05(3)
790.7	8.7	788.4		799.4	.1643	1.32	800	.1644	3.06(3)
841.8	8.5	839.3		850.3	.1712	1.32	850	.1712	3.07
892.9	8.3	890.8		901.2	.1778	1.32	900	.1776	3.08(3)
942.6	8.1	940.1		950.7	.1842	1.32	950	.1841	3.09(3)
738.0	9.0	737.5		747.0	.1575	1.32	750	.1579	3.10
EVENT	4 - Measure R vs T at Decreasing Temperatures After Above, 1/24/67								
942.3	8.1			950.4	.1842	1.32	950	.1841	4.01(3)
893.3	8.3			901.6	.1779	1.32	900	.1777	4.02(3)

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
790.9	8.7			799.6	.1644	1.32	800	.1645	4.03(3)
690.9	9.2			699.5	.1509	1.40	700	.1510	4.04(3)
589.4	9.3			598.7	.1370	1.40	600	.1368	4.05(3)
494.7	8.6			503.3	.1235	1.40	500	.1231	4.06(3)
393.8	7.2			401.0	.1087	1.40	400	.1086	4.07(3)
29.7	-.2			29.5	.0507	1.4	30	.0508	4.08(4)
EVENT 5 - Check R vs T Slope Near RT, 1/30/67									
29.6					.0506				5.01
26.2					.0500	1.1			5.02
28.4					.0507	±.1			5.03
31.3					.0509				5.04
EVENT 6 - Measure R vs T Upward After Furnace Off For 5 Days and Specimen Under High Vacuum $(<2 \times 10^{-8})$, 1/30/67									
538.0	8.9			546.9	.1298	1.40			6.01
588.0	9.3			597.3	.1368	1.40	600	.1372	6.02(3)
637.8	9.6			647.4	.1437	1.40			6.03
687.9	9.2			696.1	.1505	1.40	700	.1510	6.04(3)
736.5	9.0			745.5	.1572	1.40			6.05
787.7	8.7			797.4	.1639	1.32	800	.1643	6.06(3)
837.6	8.5			846.1	.1705	1.32			6.07
888.1	8.3			896.4	.1771	1.32	900	.1776	6.08(3)
916.1	8.2			924.3	.1808	1.32			6.09
946.4	8.1			954.5	.1847	1.32	950	.1841	6.10(3)
493.6	8.6			502.2	.1234+	1.40	500	.1231	6.11(3)
EVENT 7 - R vs T After Add 1 Charge of O_2, 1/30/67									
946.1	8.1			954.2	.1851	1.32	950	.1845	7.01
791.2	8.7			799.9	.1649	1.32	800	.1649	7.02
691.1	9.2			700.3	.1515	1.40	700	.1515	7.03
591.2	9.3			600.5	.1376	1.40	600	.1375	7.04
496.9	8.6			505.5	.1243	1.40	500	.1235	7.05
945.8	8.1			953.9	.1850	1.32	950	.1845	7.06

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESIST-ANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
EVENT 8 - R vs T After Add 2 Charges, 1/30/67									
945.7	8.1			953.8	.1855	1.32	950	.1850	8.01
866.8	8.4			875.2	.1753	1.32	875	.1753	8.02
790.6	8.7			799.3	.1653	1.32	800	.1654	8.03
690.4	9.2			799.6	.1518	1.40	400	.1519	8.04
590.1	9.3			599.4	.1380	1.40	600	.1381	8.05
496.1	8.1			504.2	.1247	1.40	500	.1241	8.06
EVENT 9 - R vs T After Add 3 Charges With Overnight Shutdown Between 2nd & 3rd Charge, 1/31/67									
948.4	8.1			956.5	.1858	1.32	950	.1849	9.01(5)
948.8	8.1			956.9	.1865	1.32	950	.1856	9.02(6)
896.6	8.4			878.0	.1763	1.32	875	.1759	9.03
793.6	8.7			802.3	.1663	1.32	800	.1660	9.04
693.5	9.2			702.7	.1529	1.40	700	.1525	9.05
593.1	9.3			602.4	.1390	1.40	600	.1387	9.06
499.4	8.1			507.5	.1257	1.40	500	.1246	9.07
949.7	8.1			957.8	.1866	1.32	950	.1856	9.08
EVENT 10 - R vs T After Add 4 Charges, 1/31/67									
949.3	8.1			957.4	.1872	1.32	950	.1862	10.01
869.9	8.4			878.3	.1770	1.32	875	.1766	10.02
793.8	8.7			802.5	.1670	1.32	800	.1667	10.03
693.9	9.2			703.1	.1536	1.40	700	.1532	10.04
593.6	9.3			602.9	.1400	1.40	600	.1396	10.05
499.6	8.1			507.7	.1265	1.40	500	.1254	10.06
949.2	8.1			957.3	.1872	1.32	950	.1862	10.07
EVENT 11 - R vs T Decreasing, After 5 Charges, 1/31/67									
949.0	8.1			957.1	.1879	1.32	950	.1870	11.01
869.8	8.4			878.2	.1777	1.32	875	.1773	11.02
793.8	8.7			802.5	.1677	1.32	800	.1674	11.03
694.0	9.2			703.2	.1543	1.40	700	.1539	11.04
593.5	9.3			602.8	.1404	1.40	600	.1400	11.05

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESIST- ANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
499.8	8.1			507.9	.1272	1.40	500	.1261	11.06
949.5	8.1			957.6	.1879	1.32	950	.1869	11.07
EVENT 12 - R vs T Decreasing After 6 Charges, 1/31/67									
949.6	8.1			957.7	.1885	1.32	950	.1875	12.01
870.2	8.4			878.6	.1783	1.32	875	.1778	12.02
793.7	8.7			802.4	.1684	1.32	800	.1681	12.03
694.1	9.2			703.3	.1550	1.40	700	.1546	12.04
593.7	9.3			603.0	.1412	1.40	600	.1408	12.05
499.8	8.1			507.9	.1278	1.40	500	.1267	12.06
949.8	8.1			957.9	.1886	1.32	950	.1875	12.07
EVENT 13 - R vs T Decreasing After 7 Charges, 1/31/67									
949.5	8.1			957.6	.1892	1.32	950	.1882	13.01
870.2	8.4			878.6	.1789	1.32	875	.1784	13.02
793.8	8.7			802.5	.1689	1.32	800	.1686	13.03
693.4	9.2			702.6	.1555	1.40	700	.1552	13.04
593.1	9.3			602.4	.1417	1.40	600	.1414	13.05
499.1	8.1			507.2	.1283	1.40	500	.1274	13.06
379.9	7.3			405.2	.1135	1.44			13.07
297.6	6.1			303.7	.0981	1.44			13.08
236.9	4.7			238.6	.0887	1.44			13.09
281.4	6.0			287.4	.0995	1.44			13.10 (7)
204.1	4.0			208.1	.0833	1.44			13.11 (7)
EVENT 14 - Planned to be R vs T After 8th Charge. R at 950 was substantially different from previous event so changed objective to measuring R vs T Downward to compare curves.									
28.1					.0550	1.44			14.01 (4)
948.9	8.1	957.0			.1874	1.32	950	.1864	14.02
871.1					.1775	1.32	875		14.03
793.0					.1674	1.32	800		14.04
692.4					.1541	1.40	700		14.05
592.6					.1404	1.40	600		14.06

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
498.6					.1273	1.40			14.07
398.3					.1127	1.44			14.08
377.1					.1096	1.44			14.09
340.6					.1040	1.44			14.10
EVENT 15 - R vs T Increasing to More Closely Define New R vs T Curve, 2/1/67									
359.7					.1070				15.01
379.0					.1101				15.02
379.9					.1130				15.03
417.7					.1160				15.04
437.6					.1190				15.05
457.1					.1220				15.06
478.4					.1249				15.07
499.4					.1279				15.08
519.6					.1308				15.09
560.7					.1366				15.10
581.4					.1395				15.11
693.6					.1550				15.12
794.5					.1684				15.13
870.3					.1783				15.14
949.0					.1885				15.15
EVENT 16 - R After High Temperature Anneals									
949.0					.1882	1.32			16.01
949.0					.1886	1.32			16.02
949.1	8.1			957.2	.1890	1.32	950	.1881	16.03(8)
949.1	8.1			957.1	.1888	1.32	950	.1879	16.04(9)
949.0	8.1			957.1	.1889	1.32	950	.1880	16.04(9)
EVENT 17 - R vs T Decreasing. Determine Curve After Anneal, 2/1/67									
869.8	8.4			878.2	.1778	1.32	875	.1774	17.01
793.1	8.7			801.8	.1678	1.32	800	.1676	17.02
691.7	8.2			700.9	.1543	1.40	700	.1542	17.03

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
591.8	8.7			600.5	.1406	1.40	600	.1405	17.04
559.5					.1362				17.05
519.1					.1305				17.06
478.3	8.2			486.5	.1247	1.40	500	.1266	17.07
418.6					.1160				17.08
379.5					.1102				17.09
340.4					.1043				17.10
330.9					.1008				17.11
EVENT 18 - R vs T Increasing. Warmup for 8th Engass, 2/2/67									
28.7					.0553				18.01
420.4					.1168				18.02
501.5					.1284				18.03
595.5					.1414				18.04
695.9					.1555				18.05
795.0					.1687				18.06
871.4					.1784				18.07
950.8					.1889				18.08
EVENT 19 - R vs T Decreasing After Add 8 Charges, 2/2/67									
951.1	8.1			959.2	.1893	1.32	950	.1881	19.01
866.3	8.4			874.7	.1773	1.32	875	.1773	19.02
788.7	8.8			797.5	.1673	1.32	800	.1676	19.03
687.1	9.2			696.3	.1538	1.40	700	.1543	19.04
EVENT 20 - R vs T To Search For Resistance Offset Found in Preceding Event, 2/2/67									
938.3					.1876				20.01
928.0					.1863				20.02
917.8					.1850				20.03
907.5					.1837				20.04
863.4					.1770				20.05
907.2					.1833				20.06
917.3					.1848				20.07

TABLE B14

TEMPERATURE, °C (1)	SAMPLE RESISTANCE, OHMS (1)	DATA POINT NUMBER	TEMPERATURE, °C (1)	SAMPLE RESISTANCE, OHMS (1)	DATA POINT NUMBER
906.6	.1834	20.08	890.8	.1805	20.36
896.5	.1821	20.09	891.1	.1806	20.37
886.6	.1800	20.10	891.2	.1806	20.38
888.4	.1802	20.11	891.3	.1807	20.39
890.5	.1805	20.12	891.6	.1806	20.40
892.5	.1813	20.13	891.8	.1807	20.41
894.4	.1816	20.14	892.4	.1807	20.42
891.4	.1810	20.15	892.2	.1808	20.43
891.1	.1813	20.16	892.5	.1808	20.44
890.9	.1812	20.17	892.9	.1808	20.45
890.6	.1811	20.18	893.0	.1808	20.46
890.3	.1811	20.19	893.2	.1812	20.47
890.2	.18105	20.20	893.6	.1809	20.48
889.8	.18105	20.21	893.8	.1809	20.49
889.5	.1810	20.22	894.4	.1810	20.50
889.3	.1804	20.23	894.8	.1811	20.51
889.1	.1807	20.24	895.4	.1812	20.52
888.6	.1803	20.25	896.0	.1818	20.53
888.2	.1802	20.26	896.5	.1819	20.54
887.2	.18005	20.27	898.0	.1821	20.55
888.2	.1802	20.28	862.9	.1770	20.56
888.9	.18025	20.29	786.4	.1670	20.57
889.2	.1803	20.30	684.8	.1535	20.58
889.5	.1804	20.31	584.9	.1398	20.59
889.8	.1804	20.32	488.8	.1262	20.60
890.0	.1805	20.33	416.7	.1158	20.61
890.3	.1805	20.34	338.9	.1042	20.62
890.5	.1805	20.35			
EVENT 21 - R vs T Increasing. Search For Offset, 2/3/67					
28.9	.0553	21.01(4)	886.8	.1811	21.08
860.6	.1777	21.02	888.2	.1813	21.09
868.5	.1787	21.03	889.5	.1814	21.10
873.8	.1793	21.04	890.8	.1816	21.11
880.0	.1802	21.05	892.0	.1818	21.12
884.0	.1807	21.06	893.4	.1819	21.13
885.7	.1809	21.07	894.8	.1822	21.14

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
896.1					.1823				21.15
897.5					.1825				21.16
898.9					.1827				21.17
900.3					.1829				21.18
EVENT 22 - R vs T Decreasing, 2/3/67									
938.3					.1878				22.01
864.5					.1782				22.02
788.1					.1673				22.03
EVENT 23 - R vs T Increasing to 950°C For Engass, 2/3/67									
798.9					.1684				23.01
808.9					.1699				23.02
819.2					.1717				23.03
823.5					.1724				23.04
828.3					.1731				23.05
833.6					.1739				23.06
838.2					.1745				23.07
839.0					.1746				23.08
866.1					.1782				23.09
EVENT 24 - R vs T After 9 Charges									
939.9	8.1			948.0	.1880				24.01 (5)
940.6	8.1			948.7	.1883	1.32	950	.1885	24.02 (6)
866.1	8.4			874.5	.1779	1.32	875	.1780	24.03
789.7	8.8			798.5	.1678	1.32	800	.1680	24.04
688.1	9.2			697.3	.1545	1.40	700	.1549	24.05
588.1	8.7			596.8	.1408	1.40	600	.1412	24.06
492.1	8.2			500.3	.1273	1.40	500	.1273	24.07
939.5					.1884		950		24.08
EVENT 25 - R vs T After 10 Charges, 2/3/67									
938.1	8.1			946.2	.1887	1.32	950	.1892	25.01
863.6	8.4			872.0	.1783	1.32	875	.1787	25.02
787.1	8.8			795.9	.1683	1.32	800	.1688	25.03

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
685.3	9.2			694.5	.1548	1.40	700	.1555	25.04
586.2	8.7			594.9	.1412	1.40	600	.1419	25.05
491.7	8.7			499.4	.1277	1.40	500	.1278	25.06
398.1	8.2				.1145	1.44			25.07
EVENT 26 - R vs T Increasing After Overnight Shutdown, 2/6/67									
30.0					.0569		30		26.01 (4)
391.9					.1134		400		26.02
485.2					.1275		500		26.03
584.0					.1411		600		26.04
684.2					.1555		700		26.05
785.4					.1691		800		26.06
861.9					.1791		875		26.07
936.6					.1888		950		26.08
EVENT 27 - R vs T After 11 Charges, 2/6/67									
937.0	8.1			945.1	.1892	1.32	950	.1898	27.01
862.9	8.4			871.3	.1788	1.32	875	.1793	27.02
786.6	8.8			795.4	.1689	1.32	800	.1695	27.03
685.3	9.2			694.5	.1554	1.40	700	.1562	27.04
585.6	8.7			594.3	.1418	1.40	600	.1426	27.05
489.7	8.2			497.9	.1283	1.40	500	.1286	27.06
905.0					.1853				27.07
922.6					.1875				27.08
938.0					.1895				27.09
EVENT 28 - R vs T After 12 Charges, 2/6/67									
938.6	8.1			946.7	.1892	1.32	950	.1896	28.01
864.3	8.4			972.7	.1797	1.32	875	.1800	28.02
787.9	8.8			796.7	.1698	1.32	800	.1702	28.03
686.3	9.2			695.5	.1563	1.40	700	.1569	28.04
586.4	8.7			595.1	.1426	1.40	600	.1432	28.05
489.7	8.2			497.9	.1289	1.40	500	.1292	28.06
542.8					.1365				28.07
938.5					.1900				28.08

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
489.7					.1355				
587.8					.1492				41.04
687.2					.1627				41.05
788.1					.1627				41.06
861.8					.1855				41.07
937.0					.1949				41.08
936.2					.1962				41.09
944.7					.1971				41.10
EVENT 42 - R vs T After 26 Charges, 2/13/67									
944.7	8.1			952.8	.1963	1.32	950	.1959	42.01 (25)
944.7	8.1			952.8	.1968	1.32	950	.1964	42.02
944.5	8.1			952.6	.1974	1.32	950	.1971	42.03 (27)
944.5	8.1			952.6	.1980	1.32	950	.1977	42.04 (28)
869.2	8.4			877.6	.1884	1.32	875	.1881	42.05
790.7	8.8			799.5	.1782	1.32	800	.1783	42.06
689.8	9.2			698.0	.1649	1.40	700	.1652	42.07
582.5	8.7			591.2	.1503	1.40	600	.1515	42.08
491.8	8.2			500.0	.1381	1.40	500	.1381	42.09
542.4					.1453				42.10
EVENT 43 - Search for Resistance Anomaly, 2/13/67									
590.6					.1519				43.01
588.2					.1516				43.02
588.8					.1512				43.03
583.5					.1508				42.04
581.0					.1504				43.05
578.7					.1499				43.06
576.2					.1495				43.07
573.9					.1492				43.08
573.6					.1491				43.09
569.3					.1485				43.10
567.1					.1482				43.11
562.2					.1475				43.12
557.6					.1468				43.13

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
938.5					.1899				28.09(10)
938.2					.1891				28.10(11)
863.2					.1795				28.11
EVENT 29 - R vs T Increasing After Overnight Shutdown, 2/7/67									
31.6					.0585				29.01 (4)
395.0					.1154				29.02
491.6					.1298				29.03
									29.04
689.3					.1568				29.05
789.3					.1706				29.06
865.2					.1797				29.07
938.2					.1898				29.081
938.2					.1893				29.082(12)
938.2					.1890				29.083(13)
938.2					.1890				29.084(14)
EVENT 30 - R vs T After Anneal at 1200°C for 15 Minutes, 2/7/67									
938.4					.1896				30.01
938.2					.1895				30.02(15)
938.2					.1894				30.03(16)
938.2					.18935				30.04(17)
938.2					.1893				30.05(18)
EVENT 31 - R vs T After 13 Charges, 2/7/67									
938.2	8.1				.1898				31.01(15) (19)
938.2	8.1				.1899				31.02(20)
938.2	8.1				.1899				31.03(21)
937.5	8.1			945.6	.1896	1.32	950	.1902	31.04
862.9	8.4			871.3	.1802	1.32	875	.1807	31.05
786.8	8.8			795.6	.1702	1.32	800	.1708	31.06
685.0	9.2			694.2	.1568	1.40	700	.1576	31.07
585.2	8.7			593.9	.1432	1.40	600	.1441	31.08

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESIST-	SLOPE,	INTERPOLATED RESISTANCE		DATA
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.	ANCE, OHMS (1)	OHMS/°C x 10^4	TEMP. CORR. TO, °C	OHMS	PT. NO.
489.2	8.2			497.4	.1294	1.40	500	.1298	31.09
937.9					.1897				31.10
EVENT 32 - R vs T After 14 Charges, 2/7/67									
937.9	8.1			946.0	.1902	1.32	950	.1907	32.01
863.8	8.4			872.2	.1809	1.32	875	.1813	32.02
787.6	8.8			796.4	.1710	1.32	800	.1715	32.03
686.3	9.2			695.5	.1576	1.40	700	.1582	32.04
586.2	8.7			594.9	.1439	1.40	600	.1446	32.05
490.1	8.2			498.2	.1304	1.40	500	.1306	32.06
939.2					.1905				32.07
939.0					.1904				32.08
EVENT 33 - R vs T After 15 Charges, 2/7/67									
939.0					.1911				33.01
939.0	8.1			947.1	.1910	1.32	950	.1914	33.02 (20)
863.8	8.4			872.2	.1814	1.32	875	.1818	33.03
787.4	8.8			796.2	.1715	1.32	800	.1720	33.04
686.5	9.2			695.7	.1581	1.40	700	.1587	33.05
586.8	8.7			595.5	.1445	1.40	600	.1451	33.06
490.3	8.2			498.5	.1310	1.40	500	.1312	33.07
398.2					.1176				33.08
EVENT 34 - R vs T Increasing After Overnight Shutdown, 2/8/67									
30.0					.0603	1.44	30	.0603	34.01 (4)
397.3					.1176				34.02
493.5					.1317				34.03
590.6					.1453				34.04
690.4					.1588				34.05
791.2					.1722				34.06
866.7					.1818				34.07
941.0					.1912				34.08

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
EVENT 35 - R vs T After 16 Charges, 2/8/67									
941.0	8.1			949.1	.1919	1.32	950	.1920	35.01
866.3	8.4			874.7	.1824	1.32	875	.1824	35.02
789.5	8.8			799.3	.1724	1.32	800	.1725	35.03
687.8	9.2			697.0	.1589	1.40	700	.1593	35.04
588.2	8.7			596.9	.1454	1.40	600	.1458	35.05
491.7	8.2			499.9	.1319	1.40	500	.1319	35.06
545.4					.1395				35.07
941.0					.1919				35.08
EVENT 36 - R vs T After 17 Charges, 2/8/67									
941.1	8.1			949.2	.1925	1.32	950	.1926	36.01
866.4	8.4			874.8	.1830	1.32	875	.1830	36.02
789.9	8.8			798.7	.1731	1.32	800	.1733	36.03
688.4	9.2			697.6	.1597	1.40	700	.1600	36.04
588.2	8.7			596.9	.1460	1.40	600	.1464	36.05
544.8					.1400	1.40			36.06
492.2	8.2			500.4	.1325	1.40	500	.1324	36.07
680.4					.1586	1.40			36.08
720.4					.1639				36.09
941.1					.1925				36.10
EVENT 37 - R vs T After 18 Charges, 2/8/67									
941.1	8.1			949.2	.1931	1.32	950	.1932	37.01
866.0	8.4			874.4	.1835	1.32	875	.1836	37.02
790.3	8.8			789.2	.1737	1.32	800	.1738	37.03
688.7	9.2			697.9	.1603	1.4	700	.1606	37.04
588.8	8.7			597.5	.1407	1.4	600	.1470	37.05
545.2					.1407	1.4			37.06
492.9	8.2			501.1	.1332	1.4	500	.1330	37.07
645.0					.1545				37.08
719.3					.1644				37.09
939.4					.1929				37.10

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
EVENT 38 - R vs T After 19 Charges, 2/8/67									
939.3	8.1			947.4	.1934	1.32	950	.1937	38.01
864.2	8.4			872.6	.1829	1.32	875	.1842	38.02
787.7	8.8			796.5	.1740	1.32	800	.1745	38.03
685.9	9.2			695.1	.1606	1.40	700	.1613	38.04
586.4	8.7			595.1	.1470	1.40	600	.1477	38.05
490.0	8.2			498.2	.1335	1.40	500	.1338	38.06
398.0					.1202				38.07
30.3					.0630				38.08
EVENT 39 - R vs T Increasing After Overnight Shutdown, 2/10/67									
30.5					.0627		30	.0626	39.01(4)
398.7					.1204				39.02
491.1					.1339				39.03
588.3					.1475				39.04
688.1					.1610				39.05
790.5					.1743				39.06
864.5					.1838				39.07
938.4					.1933				39.08
946.0					.1942				39.09
EVENT 40 - R vs T After 22 Charges, 2/10/67									
946.1				954.2	.1948	1.32	950	.1942	40.01(22)
945.9				954.0	.1954	1.32	950	.1948	40.02(23)
946.0	8.1			954.1	.1958	1.32			40.03(24)
869.7	8.4			878.1	.1862	1.32	875	.1858	40.04
792.2	8.8			801.0	.1762	1.32	800	.1761	40.05
689.5	9.2			698.7	.1630	1.40	700	.1632	40.06
583.5	8.7			592.2	.1485	1.40	600	.1496	40.07
487.4	8.2			495.6	.1350	1.40	500	.1356	40.08
540.8					.1426				40.09
EVENT 41 - R vs T Increasing After Overnight Shutdown, 2/13/67									
26.7				26.7	.0646	1.73	30	.0651	41.01
384.1					.1216				41.02

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
552.8					.1462				43.14
548.0					.1455				43.15
545.6					.1452				43.16
543.4					.1449				43.17
541.0					.1443				43.18
538.6					.1442				43.19
536.2					.1439				43.20
534.0					.1435				43.21
529.4					.1429				43.22
508.3					.1400				43.23
491.9					.1377				43.24
436.4					.1298				43.25
EVENT 44 - R vs T Increasing After Overnight Shutdown, 2/14/67									
30.6				30.6	.0671	1.73	30	.0670	44.01(4)
398.6					.1247				44.02
494.0	8.2			502.2	.1387	1.40	500	.1385	44.03
592.9	8.7			801.6	.1524	1.40	600	.1522	44.04
693.5	9.2			702.7	.1654	1.40	700	.1650	44.05
793.7	8.8			802.5	.1786	1.32	800	.1783	44.06
871.0	8.4			879.4	.1892	1.32	875	.1893	44.07
945.4	8.1			953.5	.1991	1.32	950	.1986	44.08
EVENT 45 - R vs T After 30 Charges, 2/14/67									
945.3	8.1			953.4	.1979	1.32	950	.1975	45.01(29)
945.3	8.1			953.4	.1980	1.32	950	.1976	45.02(30)
945.0	8.1			953.1	.1991	1.32	950	.1987	45.03(31)
945.0	8.1			953.1	.1993	1.32	950	.1989	45.04(32)
868.9	8.4			877.3	.1886	1.32	875	.1883	45.05
790.6	8.8			799.4	.1784	1.32	800	.1785	45.06
690.3	9.2			699.5	.1653	1.40	700	.1654	45.07
589.8	8.7			598.5	.1516	1.40	600	.1518	45.08
492.4	8.2			500.6	.1381	1.40	500	.1380	45.09
492.4					.1380				45.10(15)
492.5					.1384				45.11(33)
942.7	8.1			950.8	.1992	1.32	950	.1991	34.12

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
EVENT 46 - R vs T After 34 Charges, 2/14/67									
942.4	8.1			950.5	.1980	1.32	950	.1980	46.01(34)
942.2	8.1			950.3	.1982	1.32	950	.1982	46.02(35)
942.2	8.1			950.3	.1984	1.32	950	.1984	46.03(36)
942.1	8.1			950.2	.1986	1.32	950	.1986	46.04(37)
870.2	8.4			878.6	.1896	1.32	875	.1891	46.05
792.2	8.8			801.0	.1796	1.32	800	.1795	46.06
692.9	9.2			702.1	.1664	1.40	700	.1661	46.07
592.7	8.7			601.4	.1529	1.40	600	.1527	46.08
497.0	8.2			505.2	.1399	1.40	500	.1392	46.09
943.7	8.1			951.8	.2001	1.32	950	.2000	46.10
943.3					.2000				46.11
EVENT 47 - R vs T After 38 Charges, 2/14/67									
942.9	8.1			951.0	.1987	1.32	950	.1986	47.01(38)
942.7	8.1			950.8	.1992	1.32	950	.1991	47.02(39)
942.8	8.1			950.9	.1995	1.32	950	.1994	47.03(40)
942.4	8.1			950.5	.1998	1.32	950	.1997	47.04(41)
869.0	8.4			877.4	.1905	1.32	875	.1902	47.05
790.5	8.8			799.3	.1804	1.32	800	.1805	47.06
690.3	9.2			699.5	.1671	1.40	700	.1672	47.07
590.0	8.7			598.7	.1535	1.40	600	.1536	47.08
284.7	8.				.1096				47.09
493.2	8.2			501.4	.1404	1.40	500	.1403	47.10
30.1					.0690				47.11
EVENT 48 - R vs T Increasing After 2 Day Shutdown, 2/16/67									
33.3					.0696				48.01
398.6					.1266				48.02
499.5	8.2			507.7	.1412	1.4	500	.1401	48.03
									48.04
593.2	8.7			601.9	.1541	1.4	600	.1538	48.05
692.3	9.2			701.5	.1674	1.4	700	.1672	48.06
792.7	8.8			801.5	.1812	1.32	800	.1810	48.07
870.4	8.4			878.8	.1917	1.32	875	.1912	48.08
943.0	8.1			951.1	.2010	1.32	950	.2008	48.09

TABLE B14

TEMPERATURE, °C (1)					SAMPLE RESISTANCE, OHMS (1)	SLOPE, OHMS/°C x 10^4	INTERPOLATED RESISTANCE		DATA PT. NO.
TC 1	CORR. (2)	TC 2	CORR. (2)	AVE. CORR.			TEMP. CORR. TO, °C	OHMS	
EVENT 49 - Effect of Annealing at 950°C on R, 2/16/67									
942.8					.2013				49.01(42)
943.0					.2004				49.02(43)
942.0					.2000				49.03(44)
942.6					.2005				49.04(45)
943.2					.2015				49.05(46)
943.2					.2017				49.06(46)
943.7					.2020				49.07(46)
942.8					.2015				49.08(46)
942.9					.2015				49.09(46)
EVENT 50 - R vs T Increasing After Overnight Shutdown, 2/17/67									
31.7					.0699				50.01
400.5					.1277				50.02
597.5				596.5	.1559				50.03
696.9				696.5	.1693				50.04
795.9				794.8	.1822				50.05
876.7				866.3	.1977				50.06
944.8				943.0	.2016				50.07
944.1					.2014				50.08(46)
EVENT 51 - Effect of Annealing and Hold on R at 950°C, 2/17/67									
944.5					.2019				51.01(45)
944.5					.2019				51.02(46)
944.4					.2018				51.03(46)
944.4					.2018				51.04(46)
944.4					.2018				51.05(46)
944.2					.2017				51.06(46)
944.6					.2016				51.07(46)

TABLE B14

Footnotes

(1) Most readings taken 10 minutes apart. Resistance and temperature measured near end of 10 minute cycle. Temperature change and equilibrium usually complete in first 3-5 minutes of cycle.
(2) Correction factor taken from Figure 25.
(3) Interpolated value of resistance used to calculate R_o. Table B15.
(4) Measured after overnight cool to room temperature.
(5) Precharge
(6) Postcharge
(7) Resistance measured while sample cooling.
(8) After heating specimen to 1050°C for 5 minutes with current through specimen.
(9) After heating specimen to 1200°C for 5 minutes with current through specimen.
(10) After 10 minutes.
(11) After 35 minutes.
(12) After 5 minutes.
(13) After 25 minutes.
(14) After 50 minutes.
(15) After 3 minutes.
(16) After 7 minutes.
(17) After 11 minutes.
(18) After 24 minutes.
(19) Taken during the engassing.
(20) After 4 minutes.
(21) After 8 minutes.
(22) After charge 20.
(23) After charge 21.
(24) After charge 22.
(25) After charge 23.
(26) After charge 24.
(27) After charge 25.
(28) After charge 26.
(29) After charge 27.
(30) After charge 28.
(31) After charge 29.
(32) After charge 30.
(33) After 30 minutes.
(34) After charge 31.

TABLE B14

(35) After charge 32.
(36) After charge 33.
(37) After charge 34.
(38) After charge 35.
(39) After charge 36.
(40) After charge 37.
(41) After charge 38.
(42) After heating specimen to 1050°C for 10 minutes with current through the specimen.
(43) After heating specimen to 1100°C for 10 minutes.
(44) After 1.25 hour.
(45) After heating specimen to 1200°C for 10 minutes.
(46) After periods of scanning R vs T with x-y recorder.

TABLE B15

Average Values of R_o For Run Ta20

TEMPERATURE, °C	RESISTANCE, OHM			
	EVENT 3	EVENT 4	EVENT 6	AVERAGE
500	--	.1231	.1231	.1231
600	--	.1368	.1372	.1370
700	.1512	.1510	.1510	.1511
800	.1644	.1645	.1643	.1644
950	.1841	.1841	.1841	.1841

TABLE B16

Run Ta21 Data
Isothermal Pulse Run at 700°C

Sample weight: 3.1019 g
Pre run resistance = .0488 ohm at 29.0°C
Post run resistance = .0792 ohm at 24.7°C

TEMPERATURE, °C		SAMPLE RESISTANCE, OHMS	NOMINAL TEMPERATURE °C	DATA POINT NUMBER	BRIDGE CURRENT
TC 1	TC 2				
EVENT 1 - Bake Out, 2/23/67					
29.0	23.8	.1488	R.T.	1.01	
30.1		.0488	R.T.	1.02	
102.0		.0601	100	1.03	
104.5		.0606	100	1.04	
	102.4	.0606	100	1.05	
	189.8	.0742	200	1.06	
195.6		.0749	200	1.07	
	193.0	.0749	200	1.08	
293.7		.0900	300	1.09	
	290.2	.0900	300	1.10	
399.1		.1057	400	1.11	
	395.8	.1057	400	1.12	
494.1		.1193	500	1.13	
	490.4	.1193	500	1.14	
595.3		.1330	600	1.15	
	589.5	.1330	600	1.16	
498.4		.1197	500	1.17	
	492.5	.1197	500	1.18	
494.9		.1197	500	1.19	
	493.5	.1197	500	1.20	
119.7		.0757	200	1.21	
	196.0	.0757		1.22	
EVENT 2 - Continue Bake Out After Overnight at 200°C, 2/27/67					
197.3		.0756	200	2.01	+.45
	195.6	.0756	200	2.02	+.45
292.2		.0899	300	2.03	+.45
	289.8	.0899	300	2.04	+.45
292.4		.0899	300	2.05	-.45

TABLE B16

TEMPERATURE, °C		SAMPLE RESISTANCE, OHMS	NOMINAL TEMPERATURE °C	DATA POINT NUMBER	BRIDGE CURRENT
TC 1	TC 2				
398.7		.1057	400	2.06	+.45
	396.3	.1057	400	2.07	+.45
494.2		.1193	500	2.08	+.45
	491.2	.1193	500	2.09	+.45
491.9		.1193	500	2.10	-.45
	491.0	.1193	500	2.11	-.45
494.2		.1193	500	2.12	+.45
594.0		.1329	600	2.13	+.45
	589.6	.1329	600	2.14	+.45
695.5		.1464	700	2.15	+.45
	689.9	.1464	700	2.16	+.45
688.4		.1464	700	2.17	-.45
796.1		.1596	800	2.18	+.45
	789.7	.1596	800	2.19	+.45
788.2		.1596	800	2.20	-.45
	789.7	.1596	800	2.21	-.45
895.2		.1723	900	2.22	+.45
	889.6	.1723	900	2.23	+.45
895.8		.1724	900	2.24	+.45
	890.2	.1724	900	2.25	+.45
889.6		.1723	900	2.26	-.45
	890.2	.1723	900	2.27	-.45
948.0		.1789	950	2.28	+.45
	941.9	.1789	950	2.29	+.45
941.2		.1788	950	2.30	-.45
	941.9	.1788	950	2.31	-.45
951.6		.1790	950	2.32	+.90
	942.4	.1790	950	2.34	-.90
393.2		.1790	950	2.35	-.90
EVENT 3 - Establish Base Line (R vs T), 2/27/67					
947.9		.1789	950	3.01	+.45
	933.6	.1778	940	3.02	+.45
	943.2	.1791	950	3.03	+.88
946.5		.1791	950	3.04	+.88
947.3		.1791	950	3.05	+.88
	943.1	.1791	950	3.06	+.88

TABLE B16

TEMPERATURE, °C		SAMPLE RESISTANCE, OHMS	NOMINAL TEMPERATURE °C	DATA POINT NUMBER	BRIDGE CURRENT
TC 1	TC 2				
	943.1	.1791	950	3.07	-.88
943.3		.1791	950	3.08	-.88
874.2		.1700	875	3.09	+.88
	869.8	.1700	875	3.10	+.88
	869.8	.1700	875	3.11	-.88
879.6		.1700	875	3.12	-.88
795.7		.1600	800	3.13	+.88
	791.2	.1600	800	3.14	+.88
	791.2	.1600	800	3.15	-.88
721.2		.1600	800	3.16	-.88
695.2		.1468	700	3.17	+.88
	690.3	.1468	700	3.18	+.88
	690.3	.1468	700	3.19	-.88
691.8		.1468	700	3.20	-.88
594.0		.1333	600	3.21	+.88
	588.6	.1333	600	3.22	+.88
	588.6	.1333	600	3.23	-.88
591.0		.1333	600	3.24	-.88
496.2		.1198	500	3.25	+.88
	490.5	.1198	500	3.26	+.88
	490.5	.1198	500	3.27	-.88
493.6		.1198	500	3.28	-.88
EVENT 4	- Warm up After Overnight at R.T., 2/28/67				
	27.0	.0510	30	4.01	+.45
	24.0	.0488	30	4.02	+.45
30.1		.0488	30	4.03	+.45
	25.0	.0488	30	4.04	+.45
	25.0	.0488	30	4.05	+.45
	25.0	.0488	30	4.06	+.45
500.9		.1205	500	4.07	+.90
	497.1	.1205	500	4.08	+.90
	497.4	.1206	500	4.09	-.90
500.1		.1206	500	4.10	-.90
601.8		.1342		4.11	+.90
598.8		.1342		4.12	-.90
	597.2	.1342		4.13	+.90

TABLE B16

TEMPERATURE, °C		SAMPLE RESISTANCE, OHMS	NOMINAL TEMPERATURE °C	DATA POINT NUMBER	BRIDGE CURRENT
TC 1	TC 2				
	597.2	.1342		4.14	-.90
701.8		.1475		4.15	+.90
	697.4	.1475		4.16	+.90
	697.4	.1475		4.17	-.90
698.4		.1475		4.18	-.90
799.8		.1603	800	4.19	+.90
	795.3	.1603	800	4.20	+.90
	795.3	.1603	800	4.21	-.90
796.2		.1603	800	4.22	-.90
693.8		.1467	700	4.23	+.45
	690.8	.1467	700	4.24	+.45
	690.8	.1467	700	4.25	-.45
692.1		.1467	700	4.26	-.45
692.1		.1467	700	4.27	+.45
	690.0		700	4.28	+.45
	690.0		700	4.29	-.45
691.8			700	4.30	-.45

TABLE B16

NUMBER OF OXYGEN PULSE (1)	TEMPERATURE, °C TC 2	SAMPLE RESISTANCE, OHMS	NOMINAL TEMPERATURE °C	DATA POINT NUMBER
EVENT 5 - Isothermal Run at 700°C, 3/1/67				
0	688.4	.1466		5.00
1(2)		.1472		5.01
2	688.3	.1488		5.02
3	688.3	.1504		5.03
4	688.3	.1520		5.04
5(3)	688.3	.1546		5.05
6	688.3	.1560		5.06
7	688.3	.1570		5.07
8	688.3	.1581		5.08
9	688.3	.1591		5.09
10	688.3	.1599		5.10
11	688.3	.1609		5.11
12	688.3	.1617		5.12
13	688.2	.1626		5.13
14	688.2	.1634		5.14
15	688.2	.1642		5.15
16	688.3	.1650		5.16
17	688.6	.1658		5.17
18	688.6	.1665		5.18
19	688.5	.1672		5.19
20	688.7	.1679		5.20
21	688.7	.1686		5.21
22	688.7	.1692		5.22
23	688.7	.1698		5.23
24	688.7	.1704		5.24
25	688.7	.1709		5.25
26	688.7	.1715		5.26
27	688.7	.1720		5.27
28	688.7	.1725		5.28
29	689.0	.1728		5.29
30	689.1	.1735		5.30
31	688.9	.1740		5.31
32	689.0	.1744		5.32
33	689.0	.1749		5.33
34	689.1	.1753		5.34
35	688.8	.1758		5.35

TABLE B16

NUMBER OF OXYGEN PULSE (1)	TEMPERATURE, °C TC 2	SAMPLE RESISTANCE, OHMS	NOMINAL TEMPERATURE °C	DATA POINT NUMBER
36	688.6	.1762		5.36
37	688.6	.1766		5.37
38	688.6	.1770		5.38
39	688.6	.1774		5.39
40	688.6	.1777		5.40
41	688.6	.1781		5.41
42	688.6	.1784		5.42
43	688.6	.1789		5.43
44	688.6	.1792		5.44
45	687.8	.1795		5.45
46	687.8	.1799		5.46
47	687.8	.1802		5.47
48	687.8	.1806		5.48
EVENT 6 - Shutdown After Isothermal Run, 3/1/67				
	638.4	.1735	650	6.01
	587.0	.1661	600	6.02
	538.0	.1590	550	6.03
	490.3	.1520	500	6.04
	444.1	.1435	450	6.05
	398.3	.1385	400	6.06
	342.8	.1300	350	6.07
	295.9	.1228	300	6.08
	24.9	.0792	R.T.	6.09

(1) Oxygen charge size 1000 μ at 300 cc.
(2) 500 μ charge, experimental error.
(3) Oxygen Burst, unknown quantity.

TABLE B17

Run Ta22 Data
Isothermal-Isobaric Run (after Gebhardt)

Sample Weight: not recorded
Pre run resistance = .0612 ohm at 51.2°C
Post run resistance = not recorded

TEMPERATURE, °C		SAMPLE RESISTANCE, OHMS	NOMINAL TEMPERATURE °C	DATA POINT NUMBER	BRIDGE CURRENT
TC 1	TC 2				
EVENT 1 - (Data taken before specimen was rewelded), 3/6/67					
EVENT 2 - Bake Out After Repair of Short Circuit, 3/7/67					
51.2		.0612	R.T.	2.01	+.45
	51.4	.0611		2.02	+.45
	51.4	.0611		2.03	-.45
50.7		.0611		2.04	-.45
188.4		.0853	200	2.05	+.45
	188.4	.0853		2.06	+.45
	188.4	.0854		2.07	-.45
188.4		.0853		2.08	-.45
286.2		.1034	300	2.09	+.45
	188.4	.1034		2.10	+.45
	188.4	.1034		2.11	-.45
286.2	.1034			2.12	.-45
392.0		.1213	400	2.13	+.45
	392.2	.1213		2.14	+.45
486.6		.1372	500	2.15	+.45
	487.2	.1372		2.16	+.45
	586.1	.1530	600	2.17	+.45
586.6		.1530		2.18	+.45
685.7		.1686	700	2.19	+.45
	686.3	.1686	700	2.20	+.45
685.6		.1686		2.21	+.45
	685.4	.1686		2.22	+.45
	685.3	.1686		2.23	-.45
685.6		.1686		2.24	-.45
685.7		.1686		2.25	+.45

TABLE B17

TEMPERATURE, °C		SAMPLE RESISTANCE, OHMS	NOMINAL TEMPERATURE °C	DATA POINT NUMBER	BRIDGE CURRENT
TC 1	TC 2				
EVENT 3	- Establish Base Line R vs T after Anneal 1300°, 15 Minutes, 3/9/67				
935.9		.2060	950	3.01	+.90
	935.9			3.02	+.90
935.6		.2061		3.03	+.90
862.6		.1956	875	3.04	+.90
784.4		.1842	800	3.05	+.90
735.3		.1769	750	3.06	+.90
685.5		.1695	700	3.07	+.90
636.3		.1619	650	3.08	+.90
585.2		.1540	600	3.09	+.90
536.6		.1464	550	3.10	+.90
486.0		.1383	500	3.11	+.90
439.2		.1309	450	3.12	+.90
391.8		.1233	400	3.13	+.90
338.5		.1144	350	3.14	+.90
288.4		.1059	300	3.15	+.90
245.6		.0989	250	3.16	+.90
41.6		.0681	R.T.	3.17	+.90
26.2		.0624	R.T.	3.18	+.90
EVENT 4	- Warm up For Run After Overnight R.T., 3/10/67				
26.2		.0630	R.T.	4.01	
	25.4	.0630	R.T.	4.02	
692.8		.1700	700	4.03	
	693.1	.1700	700	4.04	
690.5		.1699	700	4.05	
	690.9	.1699	700	4.06	

TABLE B17

TIME (MINUTES)	$(\text{TIME})^{1/2}$	SAMPLE RESISTANCE, OHMS	CHANGE IN RESISTANCE, OHMS
EVENT 5 - Isothermal-Isobaric Run at 700°C			
0	0	.1699	.0000
2.25	1.50	.1700	.0001
4.67	2.16	.1725	.0026
7.50	2.74	.1755	.0056
8.00	2.83	.1756	.0057
9.00	3.00	.1757	.0058
11.00	3.32	.1757	.0058
18.00	4.24	.1763	.0064
33.00	5.74	.1767	.0068
39.00	6.24	.1768	.0069
49.00	7.00	.1770	.0071
60.00	7.75	.1771	.0072
93.00	9.65	.1775	.0076
103.00	10.15	.1777	.0078
128.00	11.32	.1779	.0080
138.00	11.75	.1780	.0081
152.00	12.33	.1781	.0082
160.00	12.65	.1781	.0082

TABLE B18

Run Cb01 Data
Isothermal Pulse Run at 906°C

Initial Weight: 1.6453 g
Pre run resistance = .0570 ohm at 36.2°C
Post run resistance = not recorded
Oxygen charge size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	TEMPERATURE °C	SAMPLE RESISTANCE, OHMS
0	36.2	.0570
0	905.5	.1752
1	905.7	.1753
2	905.7	.1762
3	905.8	.1794
4	905.8	.1811
5	905.8	.1827
6	905.8	.1844
7	906.0	.1948
8	906.0	.1972
9	906.0	.2016
10	906.0	.2025
11	906.0	.2025
12	906.0	.2022
13	906.0	.2018
14	906.0	.2014
15	906.0	.2010
16	906.0	.2005
16	906.0	.1993
16	906.0	.1993(1)
16	906.0	.1983(1)
16	906.0	.1979(1)
16	906.0	.1960(1)
16	906.0	.1975(1)
16	906.0	.1965(1)
16	906.0	.1973(1)
16	906.0	.1972(1)
16	906.0	.1972(1)
16	906.0	.1971(1)
16	906.0	.1969(1)

(1) Resistance readings taken one every 7-1/2 minutes.

TABLE B19

Run Cb02 Data
Isothermal Pulse Run at 906°C

Cb02 Speciman From Run Cb01
Oxygen charge size: 300 cc at 0.5 torr
Final Weight: 1.698 g

NUMBER OF OXYGEN PULSE	TEMPERATURE C°	SAMPLE RESISTANCE, OHMS
0	906.0	.1950
1	905.4	.1955
2	905.3	.1960
3	905.0	.1969
4	905.0	.1974
5	905.0	.1978
6	905.2	.1983
7	905.6	.1987
8	905.7	.1989
9	905.7	.1991
10	905.7	.1991
11	905.8	.1991
12	905.7	.1990
13	905.7	.1989
14	905.7	.1989
15	905.7	.1988
16	905.7	.1987
17	905.7	.1988
18	905.7	.1989
18	905.7	.1990 (1)
18	905.7	.1979 (1)
18	905.7	.1975 (1)
18	905.7	.1972 (1)
18	905.7	.1969 (1)
18	905.7	.1965 (1)
18	905.7	.1965 (1)
18	905.7	.1965 (1)
18	905.7	.1965 (2)
18	905.7	.1913 (3)
18	905.7	.1956 (4)
18	905.7	.1956 (5)
18	905.7	.1956 (5)
18	905.7	.1957 (5)
18	905.7	.1916 (5)

(1) Readings every one-half hour.
(2) Reading after 1-1/2 hour.
(3) Reading after overnight. Bridge contacts dirty.
(4) Reading after one hour. Bridge contacts cleaned.
(5) Readings every hour.

TABLE B20

Run Cb03 Data
Isothermal Pulse Run at 902°C

Initial Weight: 1.654 g
Pre run resistance = .0619 ohm at 30.9°C
Post run resistance = not recorded
Oxygen charge size: 300 cc at 0.25 torr

NUMBER OF OXYGEN PULSE	TEMPERATURE °C	SAMPLE RESISTANCE, OHMS
0	30.9	.0619
1	906	.1900
2	905.6	.1904
3	905.6	.1910
4	903.9	.1914
5	903.9	.1918
6	903.7	.1923
7	902.9	.1928
8	904.3	.1934
9	902.5	.1940
10	903.1	.1948
11	903.7	.1950
12	903.1	.1955
13	901.6	.1963
14	903.1	.1969
15	903.1	.1972
16	903.0	.1980
17	903.1	.1986
18	902.5	.1994
19	902.5	.1996
20	902.5	.2002
21	900.0	.2006
22	900.1	.2012
23	900.1	.2017
24	901.6	.2023
25	899.8	.2028
26	901.7	.2035
27	901.5	.2040
28	901.5	.2045
29	904.7	.2050
30	904.2	.2057
31	905.5	.2060
32	908.0	.2065
33	907.4	.2067
34	903.8	.2072
35	905.0	.2077
36	904.7	.2082

TABLE B21

Run Cb04 Data
Isothermal Pulse Run at 902°C

Cb04 Specimen From Run Cb03
Oxygen charge size: 300 cc at 0.5 torr
Final Weight: 1.633 g

NUMBER OF OXYGEN PULSE	TEMPERATURE °C	SAMPLE RESISTANCE, OHMS
0	904.9	.2064
1	902.7	.2076
2	902.7	.2085
3	902.7	.2097
4	902.7	.2105
5	903.7	.2117
6	902.0	.2124
7	902.0	.2133
8	902.0	.2142
9	902.0	.2148
10	902.0	.2155
11	903.7	.2161
12	903.7	.2164
13	902.5	.2172
14	901.6	.2176
15	901.6	.2176
16	901.6	.2177
17	901.5	.2175
18	901.5	.2176
19	901.9	.2176
20	901.9	.2175
21	901.9	.2167
21	901.9	.2160(1)
21	901.1	.2146(1)
21	901.9	.2135(1)
21	901.9	.2137(1)
21	901.9	.2121(1)
21	901.9	.2110(1)
21	901.9	.2095(2)
21	901.9	.2105(3)
21	903.4	.2108(4)
21	904.4	.2108(5)
21	903.4	.2100(6)
21	903.4	.2097(6)

(1) Readings approximately every 20 minutes.
(2) After 1450°C for 1 hours.
(3) After 20 hours
(4) After 1 hour.
(5) After 5 hours.
(6) After 24 hours.

TABLE B22

Run Cb05 Data
Isothermal Pulse Run

Initial Weight: 1.661 g
Pre run resistance = .0643 at 30.5°C
Post run resistance = not recorded
Oxygen charge size: 300 cc at 0.5 torr

NUMBER OF OXYGEN PULSE	TEMPERATURE C°	SAMPLE RESISTANCE, OHMS
0	30.5	.0643
1	908.1	.1853
2	908.6	.1863
3	908.6	.1874
4	908.7	.1884
5	908.7	.1894
6	908.8	.1905
7	908.8	.1917
8	908.7	.1928
9	908.7	.1940
10	909.0	.1951
11	908.7	.1963
12	908.7	.1975
13	908.7	.1986
14	908.7	.1996
15	908.7	.2006
16	908.7	.2018
17	908.7	.2030
18	908.7	.2042
19	908.7	.2053
20	908.8	.2065
21	908.8	.2073
22	908.8	.2081
23	909.0	.2088
24	909.0	.2093
25	909.1	.2097
26	909.2	.2098
27	909.3	.2101
28	909.3	.2103
29	909.3	.2106
30	909.2	.2107
31	909.2	.2106
32	909.3	.2104
33	909.3	.2103
33	909.2	.2077
33	909.2	.2070

TABLE B23

Run Cb06 Data
Isoconcentration Run

Cb06 Specimen From Cb05, Engassed.
Final Weight: 1.699 g

NUMBER OF OXYGEN PULSE	TEMPERATURE °C	SAMPLE RESISTANCE, OHMS
33	906.0	.2049
33	963.4	.2178
33	953.0	.2164
33	943.2	.2150
33	933.9	.2135
33	924.3	.2117
33	914.3	.2103
33	904.2	.2122
33	894.4	.2098
33	885.0	.2089
33	875.0	.2077
33	864.3	.2055
33	855.2	.2054
33	845.2	.2044
33	835.5	.2021
33	826.1	.2003
33	814.4	.1992
33	804.7	.1977
33	795.2	.1960
33	784.9	.1947
33	774.8	.1936
33	905.1	.2081
33	951.7	.2153
33	942.0	.2139
33	931.2	.2125
33	921.6	.2109
33	911.8	.2096
33	902.0	.2071
33	890.7	.2067
33	881.0	.2048
33	870.6	.2034
33	860.1	.2019
33	850.0	.2003
33	839.8	.1990
33	829.8	.1976
33	820.0	.1961
33	809.7	.1947
33	801.0	.1935
33	790.4	.1921

TABLE B24

Run Cb08 Data
Isothermal Pulse Run

Initial Weight: 1.6487 g
Pre run resistance = .0607 at 27.5°C
Post run resistance = not recorded
Final Weight: 1.7138 g
Oxygen charge size: 300 cc at 0.5 torr

NUMBER OF OXYGEN PULSE	TEMPERATURE °C	SAMPLE RESISTANCE, OHMS
0	27.5	.0607
0	92.1	.0741
0	699.6	.1577 (1)
0	696.0	.1570
0	93.4	.0921
0	696.8	.1575
1	696.4	.1578
2	695.3	.1583
3	698.0	.1592
4	698.0	.1599
5	698.0	.1607
6	701.7	.1616
7	703.8	.1628
8	704.0	.1635
9	704.1	.1644
10	704.3	.1652
11	704.7	.1659
12	704.7	.1667
13	704.6	.1676
14	704.6	.1683
15	704.6	.1692
16	705.0	.1699
17	703.5	.1709
18	702.6	.1717
19	702.7	.1724
20	703.8	.1732
21	704.4	.1742
22	705.2	.1750

TABLE B24

NUMBER OF OXYGEN PULSE	TEMPERATURE °C	SAMPLE RESISTANCE, OHMS
23	705.1	.1759
24	705.1	.1767
25	702.5	.1777
26	705.3	.1785
27	705.2	.1794
28	706.0	.1805
29	705.0	.1813
30	705.7	.1820
31	705.7	.1830
32	703.8	.1837
33	703.8	.1848
34	704.8	.1860
35	704.8	.1869
36	704.8	.1878
37	704.8	.1887
38	704.8	.1894
39	703.8	.1904
39	701.0	.1765 (2)
40	700.0	.1772
41	700.5	.1777
42	700.7	.1783
43	701.0	.1788
44	700.6	.1792
45	700.7	.1797
46	700.7	.1799
47	700.8	.1804
48	700.5	.1808
49	700.5	.1810
50	701.0	.1815
51	701.3	.1818
52	701.9	.1820
53	701.4	.1822
54	701.3	.1822
55	701.4	.1822
56	701.5	.1822
57	701.9	.1823

(1) After 1400°C for one hour. (2) After 700°C overnight.

TABLE B25

Run Cb09 Data
Isothermal Pulse Run

Initial Weight: 1.6477 g
Pre run resistance = .0583
Post run resistance = not recorded
Final Weight: 1.702 g

NUMBER OF OXYGEN PULSE	TEMPERATURE °C	SAMPLE RESISTANCE, OHMS
0	27.8	.0583
0	100.9	.0704
0	798.9	.1639(1)
0	791.8	.1638
0	799.7	.1668
1	798.8	.1674
2	798.8	.1679
3	796.4	.1687
4	798.0	.1695
5	798.5	.1702
6	798.5	.1711
7	797.0	.1719
8	796.4	.1726
9	798.0	.1734
10	797.5	.1725
11	798.7	.1750 .1735(3)
12	798.7	.1738 .1741(3)
13	799.4	.1767 .1751(3)
14	800.0	.1773 .1759(3)
15	800.0	.1767(4)
16	800.0	.1775
17	801.2	.1785
18	801.2	.1794
19	801.5	.1804
20	801.4	.1826
21	801.5	.1837
22	801.6	.1845
23	799.2	.1855
24	798.2	.1864

TABLE B25

NUMBER OF OXYGEN PULSE	TEMPERATURE °C	SAMPLE RESISTANCE, OHMS
25	798.2	.1855
26	801.0	.1882
27	801.0	.1892
28	801.0	.1901
29	801.0	.1909
30	801.0	.1916
31	801.0	.1926
32	801.0	.1934
33	801.0	.1942
34	799.2	.1950
35	797.5	.1956
36	800.1	.1940
37	800.1	.1942
38	799.2	.1964
39	799.7	.1940
40	799.7	.1941
41	799.2	.1942
42	799.2	.1942
42	797.3	.1815(5)

(1) After 1425°C for one hour and overnight at temperature.
(2) After 24 hours.
(3) Second reading after 4 minutes.
(4) Second readings taken for each pulse showed no change.
(5) After overnight.

TABLE B26

Run Ta10W01 Data
Isothermal Pulse Run

Initial Weight: 2.8962 g
Pre run resistance = .0458 ohm at 44.6°C
Post run resistance = not recorded
Final Weight: not recorded
Oxygen charge size: 300 cc at 2 torr

NUMBER OF OXYGEN PULSE	TEMPERATURE C°	SAMPLE RESISTANCE, OHMS
0	44.6	.0458
0	902.9	.1236
0	1375	.1737(1)
0	1200	.1537(1)
0	915	.1282(1)
0	1200	.1560(1)
0	920	.1304(1)
0	707	.1072
0	506.4	.0889
0	900.9	.1247
0	1425	.1736(1)
0	1195	.1530(1)
0	900.9	.1245
0	706.5	.1077
0	899.1	.1228
0	900	.1232(2)
1	900	.1253
2	900	.1273
3	900	.1310
4	900	.1334
5	900	.1372
6	900	.1423
7	900	.1477
8	900	.1521
9	900	.1622
10	900	.1739
10	900	.1745
10	900	.1768
10	900	.1806
10	900	.1821
10	900	.1852
10	900	.1888
10	900	.1934
10	900	.1970
10	900	.2050(3)

(1) Self-heated
(2) Approximately ±1.5°C
(3) Resistance leads deteriorated.

APPENDIX C

CALCULATIONS

METHOD OF SOLVING GRAPHICALLY FOR SOLUBILITY POINT

1) Determine equation for linear segment

$$\Delta R = a_1 + b_1 N_O \tag{1.1}$$

from graph of data plotted ΔR vs N_O where N_O = no. of pulses. Choose 2 points on initial straight line $(\Delta R_1, 0_1)$ and $(\Delta R_2, 0_2)$ $(\Delta R_1 = 0$ and $\Delta R_2 =$ large)

$$\text{then} \qquad b_1 = \frac{\Delta R_1 - \Delta R_2}{0_1 - 0_2}$$

$$\text{and} \qquad a_1 = \frac{0_1 \Delta R_2 - 0_2 \Delta R_1}{0_1 - 0_2}$$

2) Determine equation for parabolic segment

$$N_O = a_2 + b_2 (\Delta R) + c_2 (\Delta R)^2 \tag{2.1}$$

from graph of data plotted ΔR vs $N_O^{1/2}$. Choose 3 points $(\Delta R_1, 0_1)$ $(\Delta R_2, 0_2)$ $(\Delta R_3, 0_3)$. Since the above parabolic equation can be written

$$\frac{N_O - 0_1}{\Delta R - \Delta R_1} = a_2' + b_2' (\Delta R), \tag{2.2}$$

find a_2' and b_2', where

$$b_2' = \frac{X_2' - X_3'}{\Delta R_2 - \Delta R_3} \quad \text{and} \quad a_2' = \frac{\Delta R_2 X_3' - \Delta R_3 X_2'}{\Delta R_2 - \Delta R_3}, \text{ and}$$

where

$$X'_2 = \frac{O_2 - O_1}{\Delta R_2 - \Delta R_1} \text{ and } X'_3 = \frac{O_3 - O_1}{\Delta R_3 - \Delta R_1} .$$

Having equation (2.2), cross multiply to get equation (2.1).

3) Solve linear and parabolic equations by substituting equation (2.1) into equation (1.1) such that

$$\Delta R = a_1 + b_1 \left[a_2 + b_2(\Delta R) + c_2(\Delta R)^2\right]. \qquad (3.1)$$

Rearrange to the form

$$\text{zero} = a_3 + b_3(\Delta R) + c_3(\Delta R)^2. \qquad (3.2)$$

Find the roots from the quadratic formula:

$$\Delta R^* = \frac{-b_3}{2c_3} \pm \frac{b_3^2 - 4a_3c_3}{2c_2} \qquad (3.3)$$

4) Divide ΔR^* by the room temperature initial resistance and find the equivalent oxygen in ppm by means of the calibration curve.

Note on part 3)

If the two segments meet but do not cross there will be only one root. That is, the radical will be zero. If they cross, the two roots will be real; if not, they will be imaginary. The difference $b_3^2 - 4a_3c_3$ is a good indication of the precision of the data. The intersection (and, therefore, the solubility point) is taken as the average of the 2 roots. For example, this was done in the two runs below 750°, Ta07 and Ta21.

This method is not intended to be rigorous, but was used later after finding that the visual estimates for the position of the break depended on whether the data were plotted linearly or parabolically. The accuracy is still very sensitive to the scatter in the data.

APPENDIX D

DISCUSSION OF THE SHAPE OF THE OXYGEN PRESSURE CURVE DURING A CYCLE IN THE ISOTHERMAL PULSE METHOD (RUN Ta21)

The timing of the oxygen charges in the isothermal pulse method was controlled from the pressure recording of the O_2 in the manifold as measured by the Millitorr gage. The recording was made with a Moseley Model 680 recorder with a chart speed of 8 in./hr. A cycle length of 7-1/2 min. was selected because this coincided with the major lines on the chart from which the opening and closing of valves could be easily judged. Oxygen exposure occurred during 3-3/4 min. followed by 3-3/4 min. equilibration hold in vacuum. The output of the Millitorr gage was converted within the control unit so that a pressure range of about 6 orders of magnitude (log pressure) was recorded linearly on the 4-in.-wide chart.

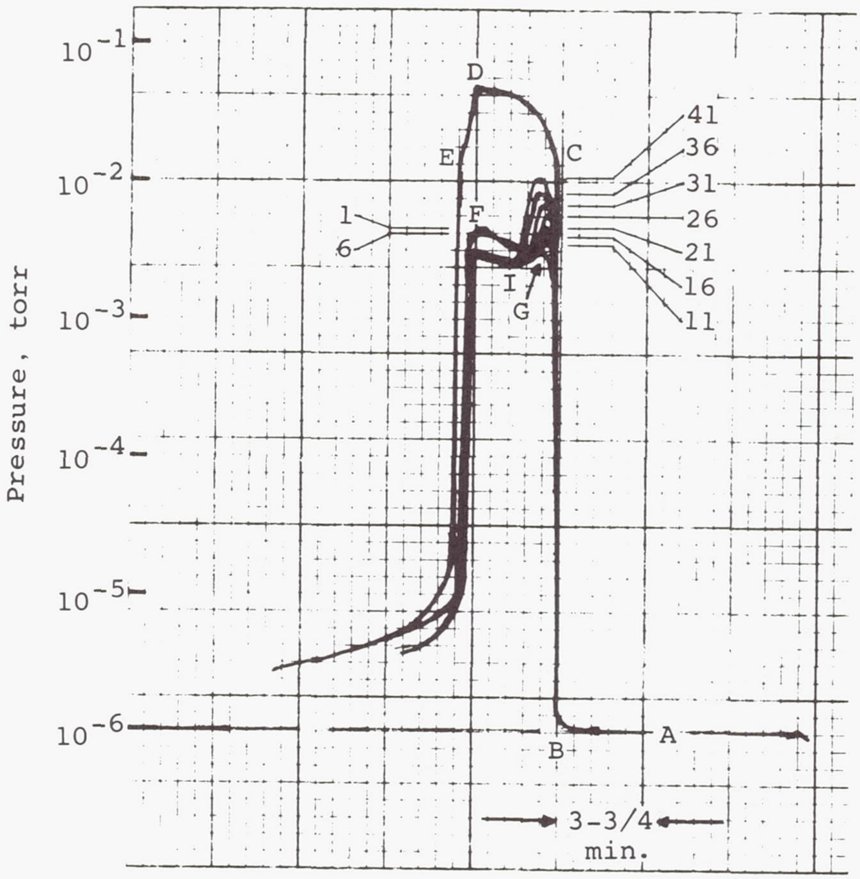
During Run Ta21 a gradually changing shape of each cycle of the pressure recording was noticed. Careful study revealed a systematic change in the shape that correlated with other observations. The curve for every fifth charge is reproduced in Figure 32, superimposed so that the start of each cycle exactly coincides. Before discussing and interpreting the curve, a review of the cycle will be given.

Referring to Figure 5 and the procedure in section II,D,l,c, the cycle consisted of expanding a 1 torr charge of O_2 contained in the 300 cc volume into the furnace and manifold (1700 cc), holding for 3-3/4 min., then evacuating for 3-3/4 min. while the 300 cc volume was being refilled.

The highest curve in Figure 32 is a trace of the pressure with the furnace and specimen at room temperature. This is thus a no-O_2-consumption reference. The correlation of this curve with the cycle steps follows:

1) The base pressure in the furnace at the start of the cycle (A) was less than 1×10^{-6} torr.

2) Valve 2 was opened at the start of the cycle (B), coinciding with the line on the chart.



← time

Figure 32 - Manifold Pressure During Oxgyen Pulse Cycles From Run Ta21.

3) The pressure in the manifold and furnace tube rose very rapidly to .01 torr at C and then rose more slowly to about .045 torr, nearly reaching equilibrium at the end of 3-3/4 min. at (D). This slow rise during most of the cycle was the result of the low pressure gradient between the 300 cc volume and the manifold, and of the small tubing (1/4 in. O.D.) between them. Slow response of the gage could also have contributed, but, as indicated later, we believe this contribution was small.

4) As Valve 2 was closed at (D) and the bakeable valve opened, the pressure dropped rapidly as the O_2 was pumped out. The slow change between (D) and (E) was caused by a slow opening of the bakeable valve to keep from swamping the ion pump. This usually was not necessary when the furnace was hot, as shown by the remaining curves.

The equilibrium pressure of 1 torr of gas in 300 cc being expanded into a connecting 1700 cc volume is about .15 torr. Some of the difference between .15 torr and the observed .045 torr can be accounted for by adsorption of O_2 on the walls. However, it is more likely that the gages were not in calibration. (The method did not require that they be calibrated; it required only good reproducibility, which is demonstrated in Figure 32.)

When the sample was at temperature, the cycle and the recorded pressure were similar except for differences caused by absorption of some of the oxygen by the sample. Of the 48 cycles in Run Ta21, 9 are plotted in Figure 32. The features of this set of curves are discussed below:

1) The maximum equilibrium pressure reached after 3-3/4 min. of O_2 exposed to the specimen was about .0030 to .0042 torr. Comparing this with the .045 value in the no-consumption run, we find that more than 90% of the oxygen was consumed during the cycle.

2) The shape of the curves indicates a changing competition in the rate at which O_2 entered and the rate at which it was consumed. Moreover, this competition changed significantly between the 6th and 7th charges. This change coincided with the passage of the solubility limit as determined from the resistance data.

a) For charges 1 to 6, the pressure rose rapidly to about .003 torr, then increased slowly to about .0042 torr. At this point (F), the remaining oxygen was pumped out. The slight differences in the curves for charge 1 and 6 were caused by slight differences in the time and speed that the bakeable valve was opened. It was apparent that nearly identical amounts of O_2 were consumed during charges 1 to 6. Moreover, the O_2 was consumed very rapidly at the start of the pulse, and little or no O_2 was consumed after the first 30 sec. (G). The pressure rise from (G) to (F) appears to be one of pressure equalization between the volumes, as between (C) and (D). (Since the pressure gradient is lower for charges 1 to 6, the rate of equalization would be lower.) There were confirming indications in the resistance measurements that O_2 was consumed only early in the cycle. Because of the slow moving galvanometer, this could not be traced accurately, but the change was essentially complete within the first minute.

The questions arise as to why (1) there was no reaction between the specimen and the O_2 at a pressure of .0025 torr and below, and (2) once the oxygen was pumped out, the specimen freely reacted with the new charge of gas in the same manner as before.

b) For charges 7 and afterward, the rate of O_2 input to the manifold was increasingly greater than the rate of consumption. In each case the pressure reached a peak (H) until the consumption rate exceeded the input rate. At this time the pressure fell until at (I) the inlet rate was once again greater than the consumption rate. The same final pressure was reached after each charge indicating that a constant amount of O_2 was consumed during each charge. Moreover, the amount consumed was slightly greater than during charges 1 to 6. The same questions arise as listed earlier.

The conclusion reached from the resistance data was that the parabolic sections of the ΔR versus number-of-charge curve resulted from a decreasing amount of oxygen entering the specimen. This conclusion is incompatible with the above observation. Further work is needed to resolve this discrepancy. The following observations must also be taken into account.

1) Some of the O_2 reacted with the Mo rods forming MoO_3. The yellow oxide was found in the bottom of the furnace tube after the program. The amount was small and accumulated over Runs Ta19 to 22. No more than a minor effect can be attributed to this cause.

2) Even though O_2 entered the specimen rapidly while in the single phase region, we have no evidence to indicate that the O_2 is evenly distributed in the lattice. Micrographs of a few specimens disclosed the typical platelet oxide formation along favored crystal directions in the grains. Although substantial amounts of the oxide were observed, the precipitation was uniform across the wire and there was no build up of a scale on the outside of the wire.

APPENDIX E

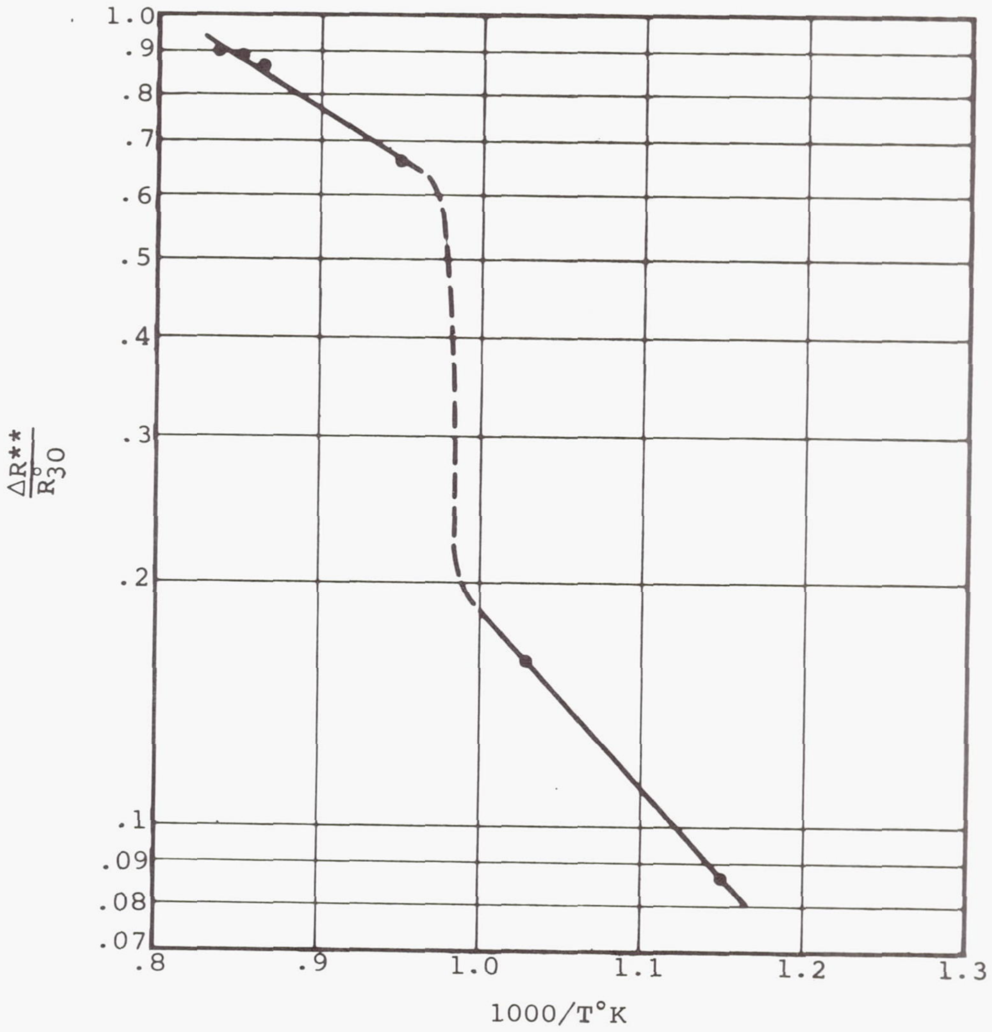
Other Data

In figure 21 it can be seen that values of $\Delta R/R^{\circ}_{30}$ corresponding to the transition between the first and second parabolic regions can be obtained. These were calculated and are given in Figure 33.

The project ended before proper correlation of these data with the Ta oxidation literature could be made. However, the following features are noted:

1) The lower part of the curve is identical to the solubility curve (Figure 22) if oxygen concentration values corresponding to values of $\Delta R/R^{\circ}_{30}$ are taken from Figure 19. This results because the first parabolic section for runs Ta07 and 21 (Figure 21) is very short (or possibly non-existent. We note that the slope increases for the first parabolic region for runs above 750°C, and decreases for the second parabolic region. The slope decreases after the linear section for runs under 750°C; however, a suggestion of a very short section of increasing slope is shown. For figure 33, we interpreted this very short section as being real and interpreted it to be the first parabolic region so that all curves are consistent).

2) The transition between the upper and lower curves is consistent with the anomolies in the Arrhenius plots of linear oxidation rates as compiled by Ong[1].



Run No.	Temperature °C	ΔR^{**}	$\frac{\Delta R^{**}}{R^{\circ}_{30}}$	Equivalent Oxygen Concentration, ppm
Ta14	900°	.065	.89	2700
Ta08	920°	.045	.90	2740
Ta06	880°	.062	.84	2570
Ta05	780°	.041	.66	2070
Ta21	700°	.008	.16	680
Ta07	600°	.006	.086	480

Figure 33 - Intercept of First and Second Parabolic Sections of Curves of Reduced Resistance Change vs. Number of Oxygen Charges as Function of Reciprocal Temperature.

NASA-Langley, 1969 —— 17