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CONVERTER PERFORMANCE TEST OF A RHENIUM EMITTER IN CONTACT WITH A CARBIDE FUEL

by

R. C. Howard

Prepared for

National Aeronautics and Space Administration

Contract No. NAS 3-4164

THERMO ELECTRON ENGINEERING CORPORATION

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FINAL REPORT

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I. INTRODUCTION

In the spring of 1963, Thermo Electron operated a thermionic converter which had a rhenium emitter in contact with a uranium carbide disk. This device, which was partially constructed under Air Force support, operated for about 200 hours at a power output between 5 and 8 watts/cm^2 without degradation. These results were encouraging evidence of the suitability of the rhenium-uranium carbide based fuel-emitter concept. However, since the carbide fuel was bonded to the rhenium emitter with a vanadium braze, some doubt existed regarding the applicability of the results to carbide-rhenium fueled-emitters bonded in other ways. Further, the duration of the test was far short of the life desired for nuclear thermionic systems. It was quite obvious that much more extensive tests were required before the feasibility of the carbide-rhenium concept could be established.

As an initial step toward this required testing, Thermo Electron contracted with the Lewis Research Center of NASA to perform another fueled-diode test similar to the one just described. The purpose of the test was to demonstrate the feasibility and desirability of rhenium emitters in nuclear heated converters. The goals were to extend the test duration beyond that already demonstrated and to operate with a mechanical bond between rhenium and uranium carbide without the use of a vanadium braze.

The fuel disk was to be composed of ceramic uranium monocarbide without a metal or other carbide diluent. This imposed a severe restriction on the thermal-mechanical operation of the fuel-emitter structure because of the large mismatch in expansion coefficient between rhenium and ceramic uranium carbide. This mismatch can be avoided if a carbide cermet is used instead of a ceramic. However, a ceramic was used in order to conduct the test under the most pessimistic conditions possible relative to rhenium-uranium carbide compatibility.

Two diode test devices were built and operated under this program. The following sections describe the construction of these devices and summarize the test data obtained from their operation.

II. TEST DEVICE DC-I

A. Design and Fabrication

In Figure 1, a simplified drawing of DC-I is shown. The basic structure was a slightly modified version of a standard planar diode used by Thermo Electron in several other programs. The collector was fabricated from a molybdenum rod with a larger diameter base which could be bolted-on to a heat rejection stand. The collector was joined to an alumina insulator seal through concentric kovar cups. This seal was joined to the emitter thin-walled sleeve by another kovar cup. The kovar-to-kovar joints and the kovar to tantalum joint were made by electron beam welding. A thin tantalum tube rested on an insulator-shoulder on the sides of the collector and served as a shield to better define the emission area.

The fuel-emitter structure was a complex assembly of six components. A molybdenum adapter ring was electron beam welded to the tantalum emitter sleeve and to the rhenium emitter disk. This ring had a 2.5 to 1 hole for pyrometric temperature measurements. The uranium carbide disk was held in place by a molybdenum cup which was electron beam welded to the exterior of the rhenium emitter. A 0.010 inch thick tungsten diffusion barrier disk was brazed with vanadium to the inside of the molybdenum cup to minimize reaction between the cup and the uranium carbide fuel. The outside diameter of the fuel disk was 0.020 inch smaller than the inside diameter of the cup for the same reason. No braze was used on either surface of the uranium carbide. All three of the molybdenum welds consisted of melting the thin lips which extended from the adapter ring and the fuel cap. No rhenium or tantalum was intended to be melted in making these welds. Temperature measurement holes of 2.5-1 were made in both the fuel cap and in the fuel disk. A hole in the fuel cap adjacent to the hole

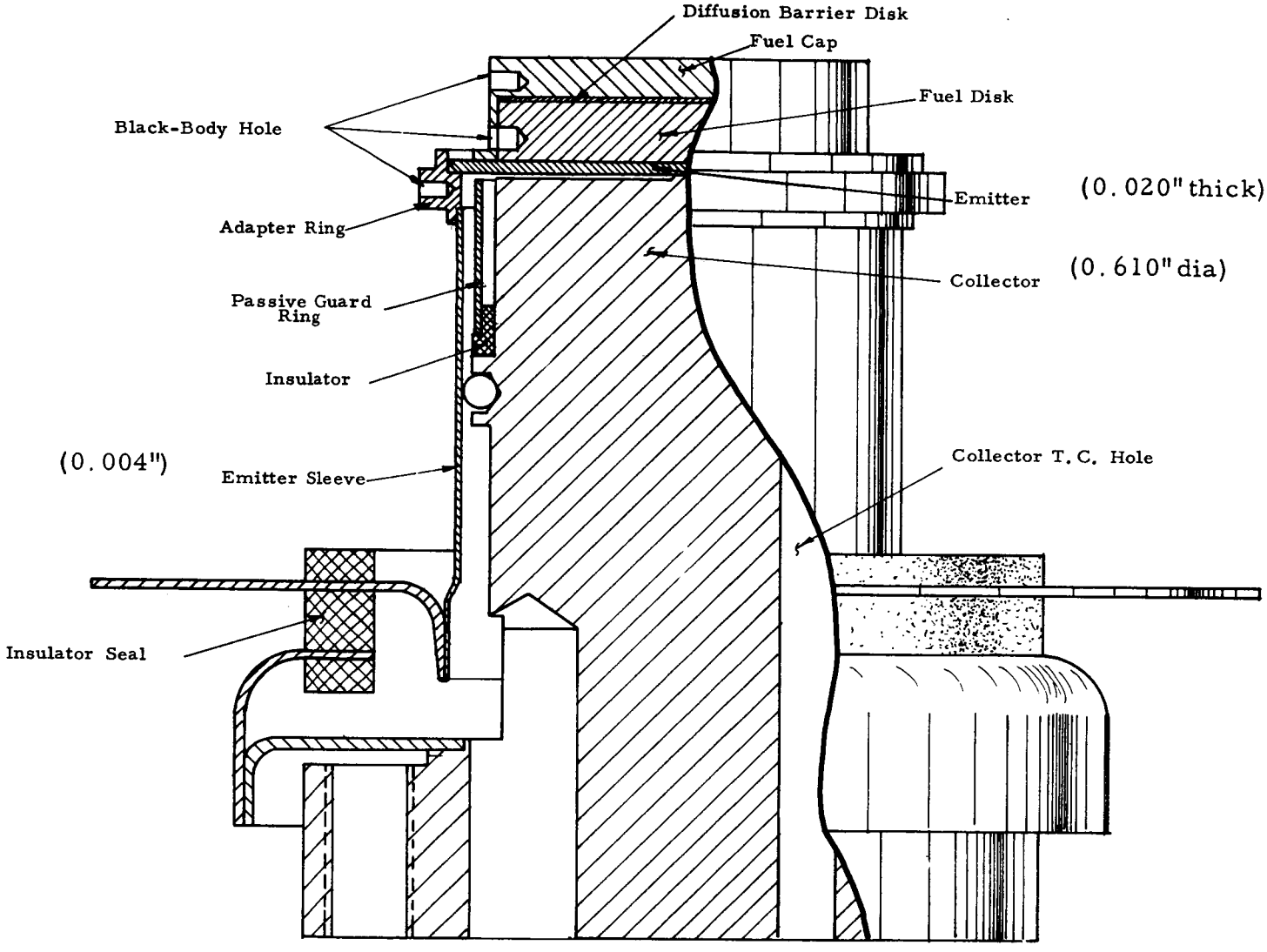


Figure 1. Design of Converter DC-1

in the fuel body was provided to permit observation of the fuel temperature. The assembly procedure for this emitter structure was as follows:

1. The adapter ring was welded to the emitter sleeve.

2. The fuel disk was positioned on the emitter and the fuel cap was welded into place with a force applied to compress the fuel cap on the fuel disk. This weld was performed with a 0.003 inch clearance between the lip of the fuel cap and the emitter to insure a tight fit between fuel disk and the emitter.

3. The emitter was welded to the adapter ring.

During the weld of the fuel cap to the emitter, the temperature of the fuel disk was kept well below the temperature of the sides of the fuel cap. This procedure plus the fact that the thermal expansion coefficient of uranium carbide is much greater than molybdenum insured that very high contact pressure existed at the molybdenum-uranium carbide and the uranium carbide-rhenium interfaces at operating temperatures.

B. Temperature Calibrations

Prior to the welding of the emitter sleeve to the insulator flange, a series of fuel-emitter temperature calibration tests were performed. The purpose of these tests was to relate the emitter surface temperature to reference temperatures which could be measured during diode operation. The test set-up for performing these measurements is shown in Figure 2.

The fueled emitter and sleeve assembly were held in a massive copper heat sink which was maintained at about 300°C. An electron bombardment filament was placed above the fuel cap to heat the fueled-emitter assembly. A prism was positioned beneath the sleeve, providing a view of the entire emitter surface.

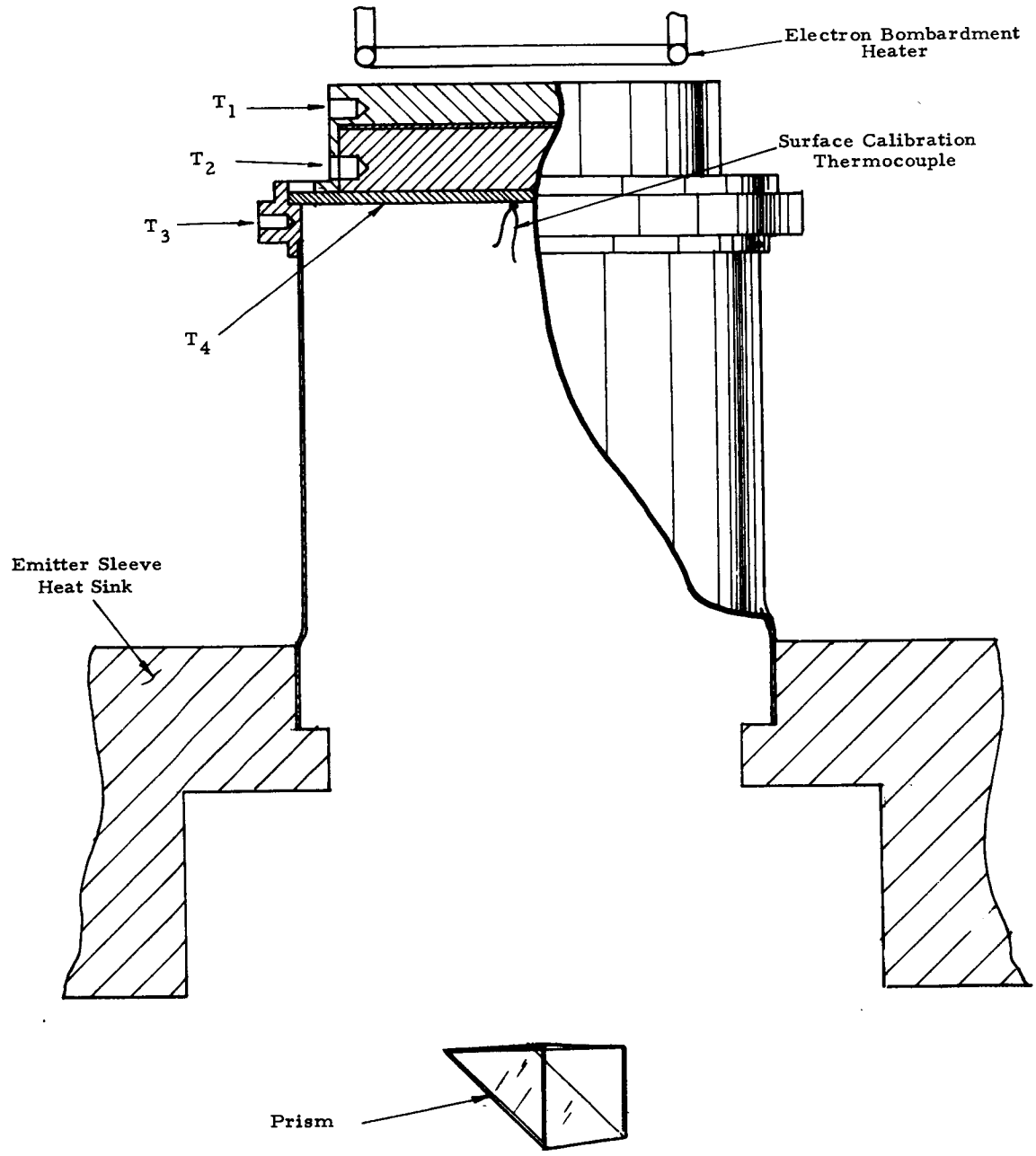


Figure 2. Fuel-Emitter Temperature Calibration Set-up

A tungsten versus tungsten-rhenium thermocouple was spot welded near the center of the emitter surface. The entire assembly was placed inside a vacuum bell jar for operation. When the fuel cap was heated by the filament, pyrometric temperature measurements were taken at T_1, T_2, and T_3. These temperature readings were corrected for the difference between a 2.5-1 hole and a true blackbody hole and for the glass bell jar. Temperature measurement scans of the emitter surface were taken pyrometrically through the prism at the base of the sleeve. By comparing the pyrometer reading to the thermocouple reading at the thermocouple location, it was possible to obtain a relationship between the "true" temperature and the pyrometrically observed temperature at any point on the surface. This relationship which is plotted in Figure 3, was used to correct all the temperatures read on the emitter face. The temperature, T_4 was measured at the center of the emitter which had a variation of approximately 30°C over its surface.

The accuracy of the fueled emitter temperature calibration procedure was affected by several sources of error. The presence of the thermocouple on the emitter surface locally perturbed the measured temperature. Although very thin wires were used, some conduction down the wires was unavoidable. In addition, the variation in the thermocouple properties of tungsten versus tungsten-rhenium wires introduces some error. Finally, there may have been emissivity variations and light condensation on the bell jar and prism. The latter effect was minimized by frequent cleaning and by remotely operated shields. It is estimated that the errors in absolute temperature were about ±20°C.

The results of the temperature calibrations are shown in Figure 4 with the fuel cap temperature (T_1) used as a reference. The points marked "initial" describe the temperature distribution during the first heating. The curves labeled "A", "B", and "C" represent the temperature distributions after 25 hours of operation at a power input corresponding to the first heating. After one cycle

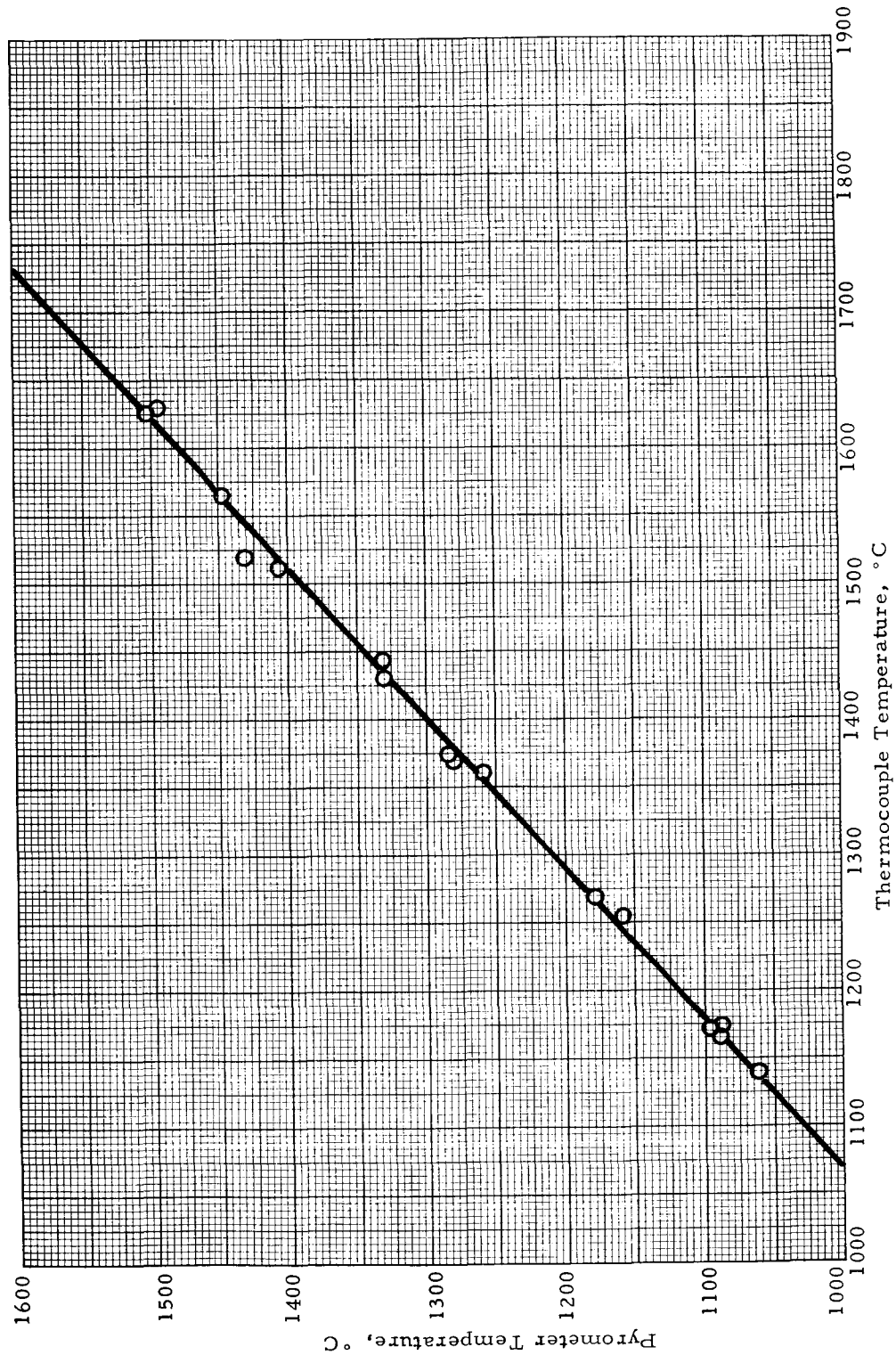


Figure 3. Brightness Temperature Correction for Rhenium Emitter

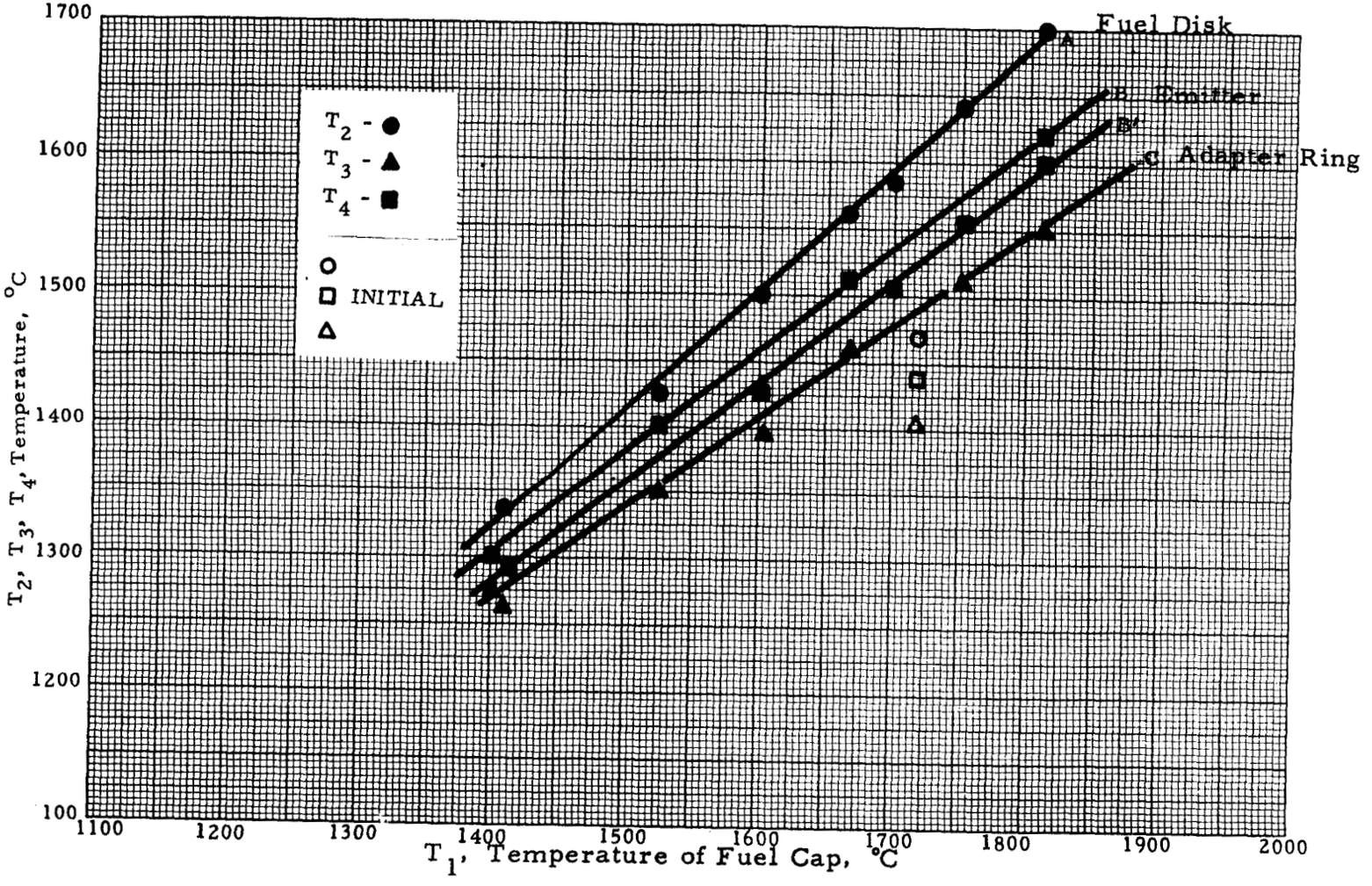


Figure 4. Results of DC-1 Fuel-Emitter Temperature Calibration

down to room temperature, curve "B'" was obtained for the emitter surface temperature with curves "A" and "C" unchanged. An additional 15 hours of operation at the same conditions caused no further change in any of the curves. The major effect of the 25-hour "soaking" was to greatly improve the load between the fuel cap and the fuel. The shift between "B" and "B'" was apparently due to a change in the bond between the rhenium emitter and the UC disk. Further soaking seemed to have no effect in restoring this bond to its former quality.

It is difficult to determine whether the final temperature distribution shown in Figure 4 persisted during subsequent cycling and operation in the converter. Previous tests on similar samples have indicated that after the first 25 to 50 hours and the first few cycles, no large changes occur as long as severe thermal cycling is avoided. Any changes that do occur are most likely to be in the direction of a decreasing emitter temperature in relation to fuel cap and fuel temperatures.

During diode operation, it was not possible to directly measure emitter surface temperature. However, by monitoring T_1, T_2, and T_3, a good estimate of emitter temperature could be obtained. As long as no drastic changes occurred in the relative levels of these temperatures, the use of Figure 4 to estimate emitter temperature should be valid. Of course, this procedure can only be followed for diode operation at open circuit conditions. When current is being drawn, the increased heat transfer rate through the emitter changes the temperature distribution shown in Figure 4. Consequently, all temperature measurements taken during diode operation were at open circuit conditions. These measurements are shown in Figure 5 for the entire test duration. The lines labeled "final calibration level" correspond to curves "A", "B'", and "C" of Figure 4. Each of the other points were taken at some stage of diode operation. The lines connecting them are drawn only to make it easier to read the points. No temperature

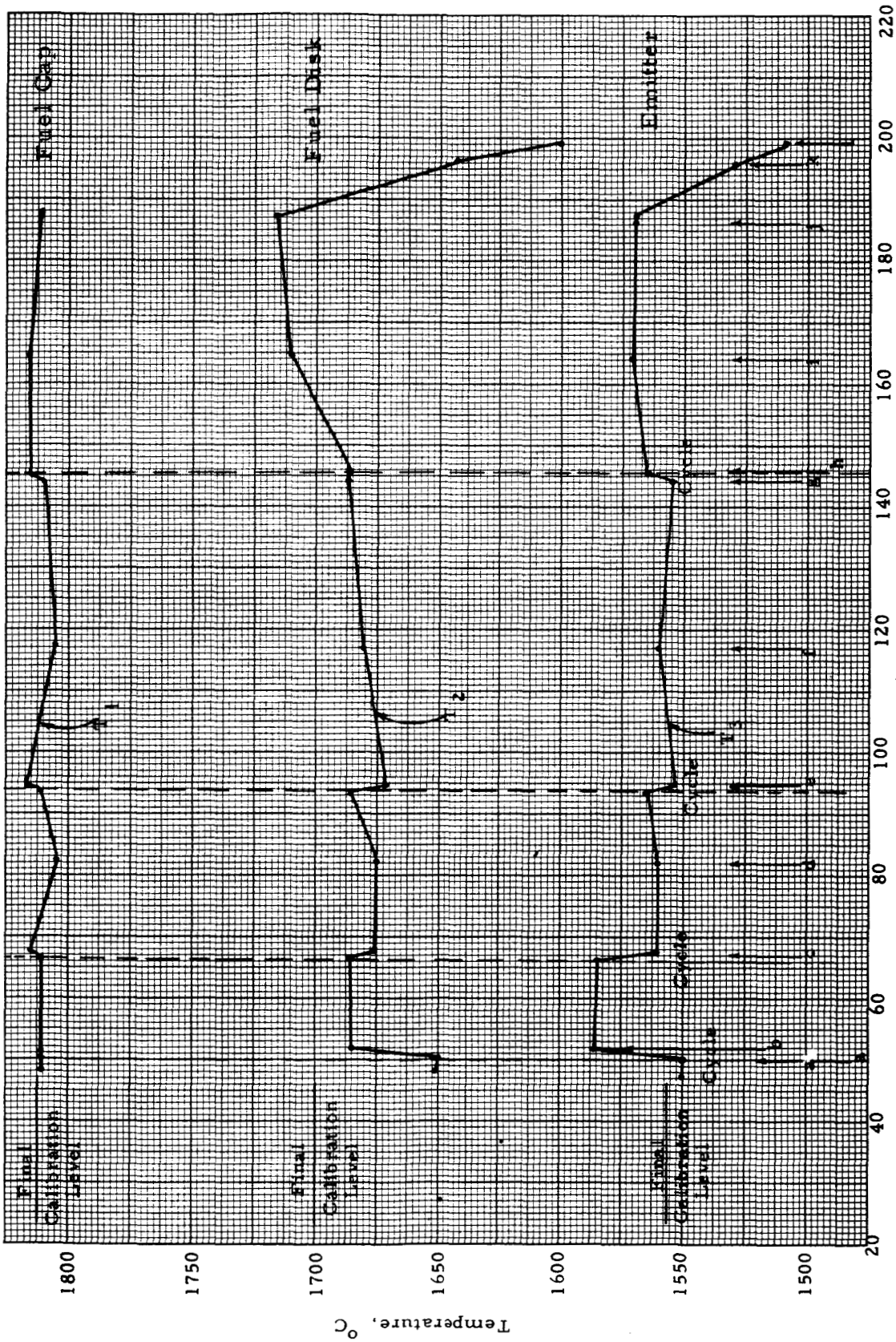


Figure 5. Variation of Fuel-Emitter Temperature versus Time for DC-1

measurements were taken between points. Also indicated in Figure 5 are the temperature cycles resulting from diode shutdowns to room temperature. Other cycles, between different high temperature levels, are not shown.

C. Converter Testing

After the temperature calibrations were completed, the fueled emitter was installed in the converter and diode testing was initiated. During the claibration tests and the diode outgassing, a total of about 50 hours of emitter soaking above 1600°C was accumulated. The diode testing was started by raising the heat input until the fuel cap reached approximately 1800°C which corresponded to a central emitter temperature of about 1600°C with an approximate variation of 30°C. In order that the temperature calibrations of Figure 4 could be used, the diode was operated at open circuit conditions. When current-voltage curves were taken, the output leads were momentarily placed across a sweep circuit load and the results were recorded by an oscilliscope camera.

Figure 6 shows the first output characteristic taken at the beginning of diode operation (after about 50 hours of emitter soaking). The thickness of the curve is due to the temperature drop which occurred while the picture was being taken. The appropriate I-V characteristic is obtained by observing the upper side of the current curve at all voltages. The performance at 0.5 volt output was about 7.5 watts/cm^2. Within the uncertainties in temperature and emitter area, this output is close to normal rhenium performance. The temperature distribution for this data point is shown in Figure 5 at the 50-hour time as point (a).

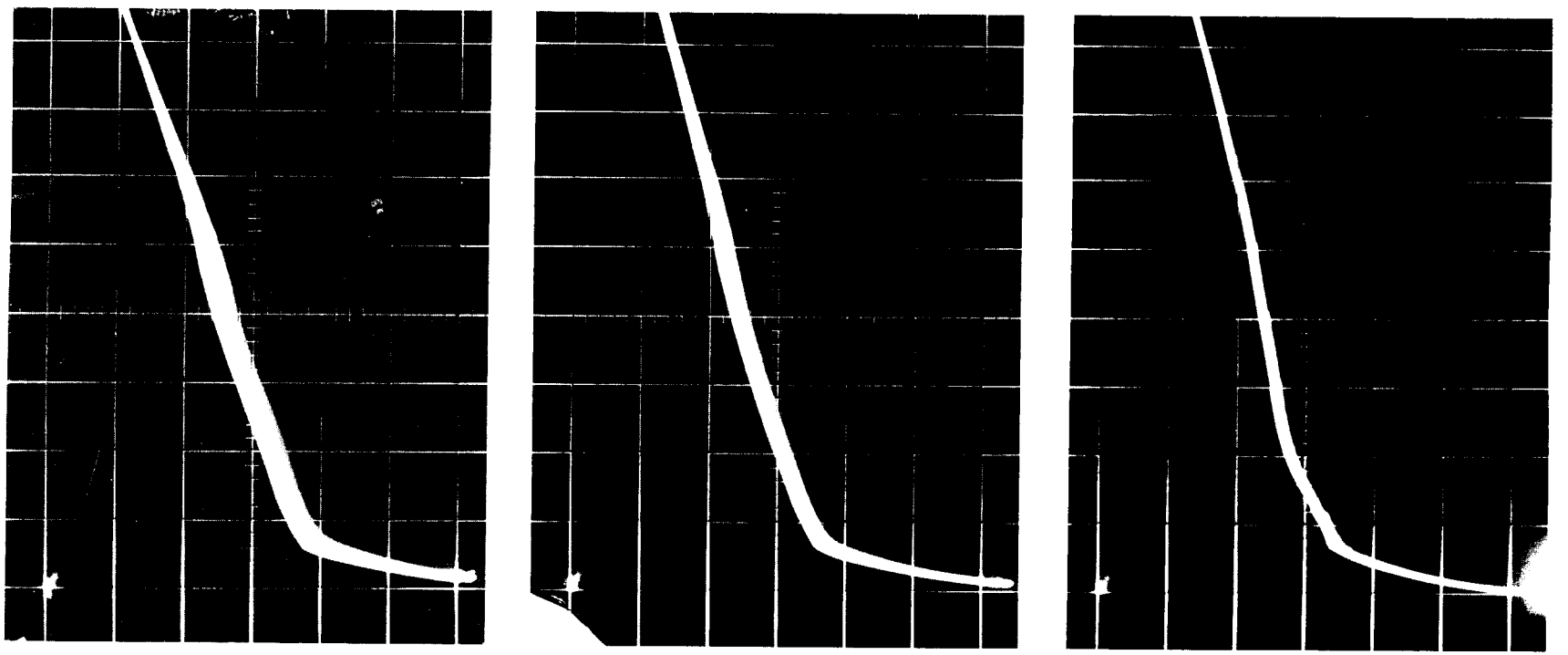
After this initial performance was observed, the diode was shut down and installed into an ion-getter vacuum life test station. After it was brought

back to operating temperature, the output characteristic was as shown in Figure 7. The temperature distribution for this point is shown as point (b) in Figure 5. An increase of about 35°C in T_2 and T_3 occurred as a result of the temperature cycle due to the shutdown. This effect could have been caused by a partial bond separation between the fuel and the emitter which forced more of the heat to flow through the sides of the fuel cap. This could have resulted in a slightly lower emitter central temperature but a slightly higher emitter edge temperature. Although some differences exist in the I-V curves of Figures 6 and 7, no major change in characteristics is evident.

At the end of about 66 hours (total emitter soak time), the diode was shut down to repair a filament lead. After returning to operating temperature for several hours, the output characteristic was as shown in Figure 8 and the temperature distribution was as shown as point (c) in Figure 5. As before, no major change in output is evident. It is interesting to note that through the first two cycles, the operating temperature distributions stayed within 30°C of the final calibration levels.

Figure 9 shows the output and point (d) on Figure 5 shows the temperature distribution after 82 hours. After 93 hours, a high voltage breakdown caused the electron gun to shut off during a night period of unattended operation. When the diode was brought back to operating temperature, the output was as shown in Figure 10 and the temperature distribution was as shown as point (e) in Figure 5. Figure 11 and point (f) in Figure 5 were taken after 117 hours. Figure 12 and point (g) in Figure 5 were taken after 144 hours.

Throughout the first 144 hours, no major change in output characteristics or temperature distribution occurred. The slight variation in output was well within the fluxuations in cesium reservoir, collector, and emitter temperatures.

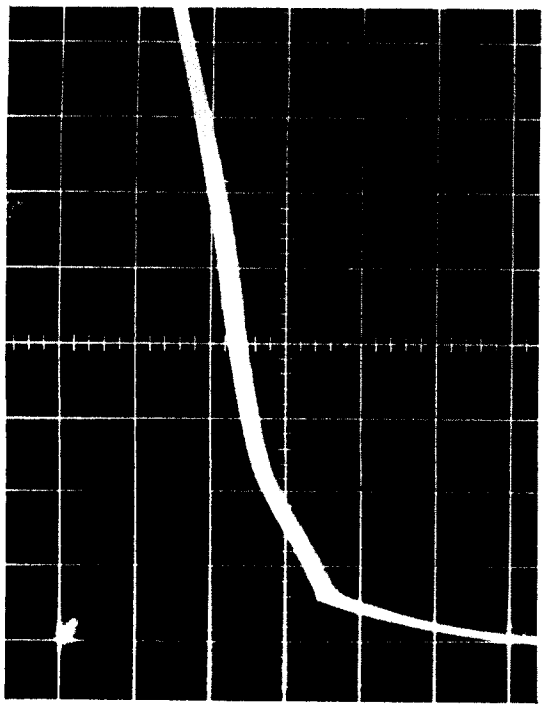


Collector Temperature - 492°C
Reservoir Temperature - 331°C

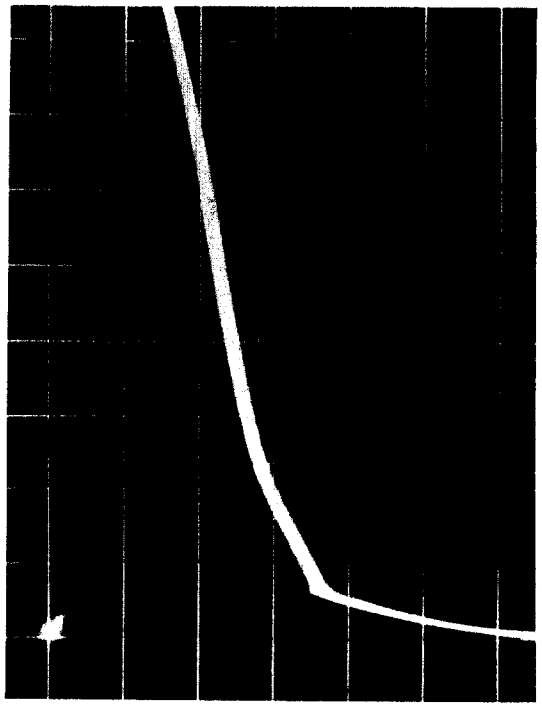
Collector Temperature - 407°C
Reservoir Temperature - 331°C

Collector Temperature - 483°C
Reservoir Temperature - 324°C

Figures 6, 7, 8. Converter Performance Curves



Collector Temperature - 471°C
Reservoir Temperature - 321°C



Collector Temperature - 447°C
Reservoir Temperature - 324°C

Collector Temperature - 452°C
Reservoir Temperature - 324°C

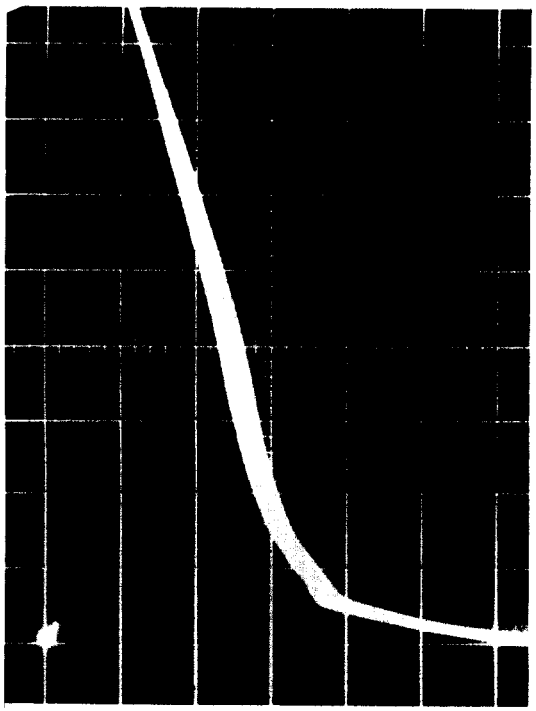
Figures 9, 10, 11. Converter Performance Curves

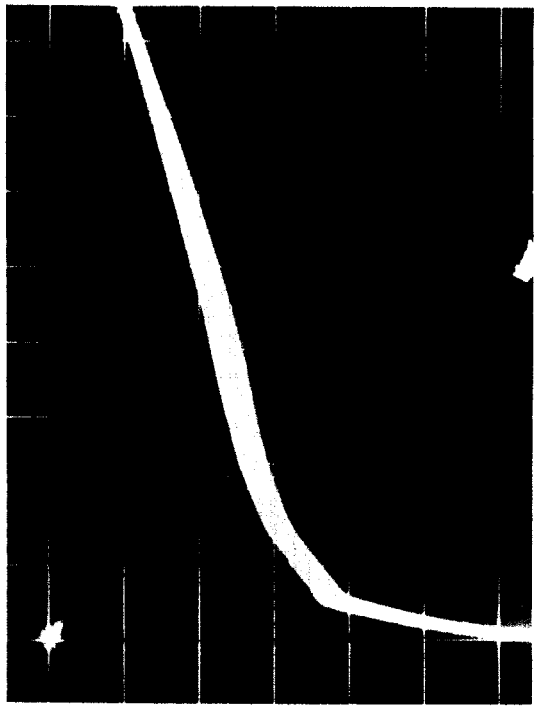
No significant trends were evident. At the 144 hour mark, the diode was again shut down to clean off bell jar deposits and inspect the fueled-emitter. It was noticed at this time, that some apparent reaction between the UC fuel and the sides of the molybdenum fuel cap near its base had occurred. It appeared that the reaction was penetrating the sides of the cap and would eventually cause the separation of the cap from the emitter.

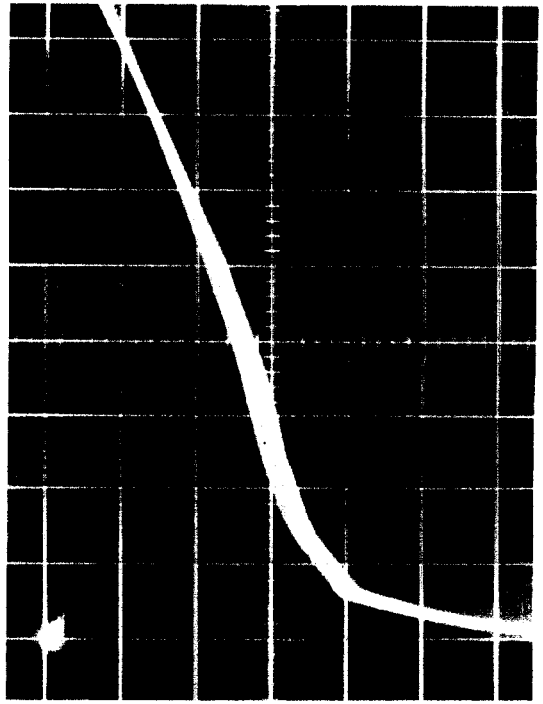
After the diode was brought back to operating temperature, the output was as shown in Figure 13 and the temperature distribution was as shown as point (h) in Figure 5. Still no significant changes had occurred. At 164 hours, Figure 14 shows that still no significant change in output had occurred. However, point (i) in Figure 5 shows that the fuel temperature (T_2) had risen by 25°C. At 187 hours, the first definite evidence of output change was observed. Figure 15 and point (j) of Figure 5 show the conditions at this time. At 196 hours, it was noticed that the molybdenum cap had completely separated from the emitter, apparently due to the Mo-UC reaction observed previously. The black-body hole in the fuel cap was no longer discernible because of this reaction. The output at 196 hours is shown in Figure 16, down by about 20% from earlier operation. Point (k) of Figure 5 shows the temperatures at this time to be down markedly. Although an additional 35 hours of testing was conducted, the useful life of the experiment terminated at about 200 hours.

D. Analysis of Results

When the converter was removed from its test stand, the molybdenum cap containing the UC fuel was completely severed from the back side of the emitter. Figure 17 shows the fuel cap, the UC fuel, the back side of the emitter, and the inside of the emitter. There was evidence of some reaction at the outer







Collector Temperature - 452°C
Reservoir Temperature - 329°C

Collector Temperature - 436°C
Reservoir Temperature - 326°C

Collector Temperature - 464°C
Reservoir Temperature - 329°C

Figures 12, 13, 14. Converter Performance Curves

Collector Temperature - 476°C
Reservoir Temperature - 326°C

Collector Temperature - 468°C
Reservoir Temperature - 329°C

Figures 15, 16. Converter Performance Curves

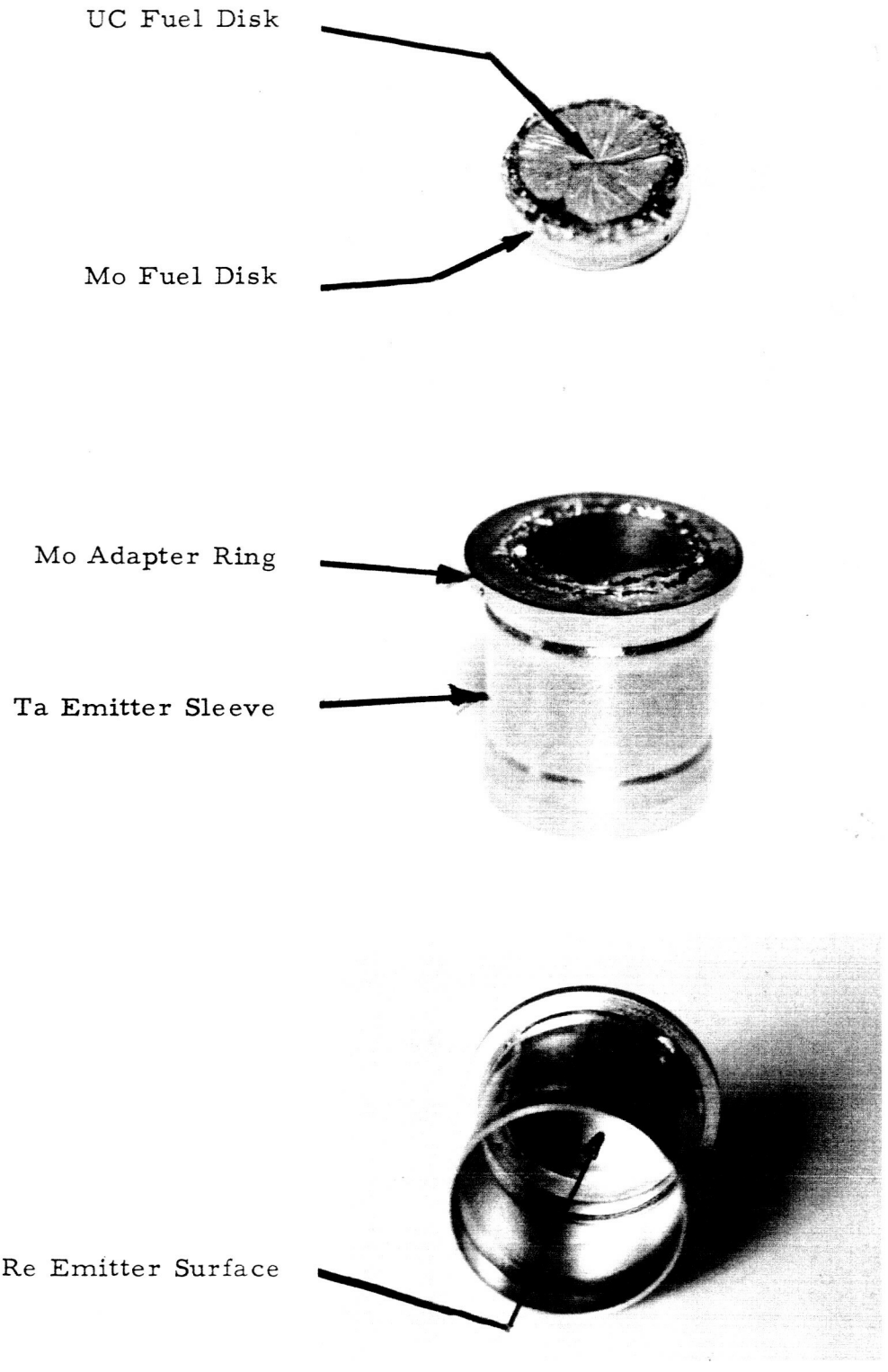


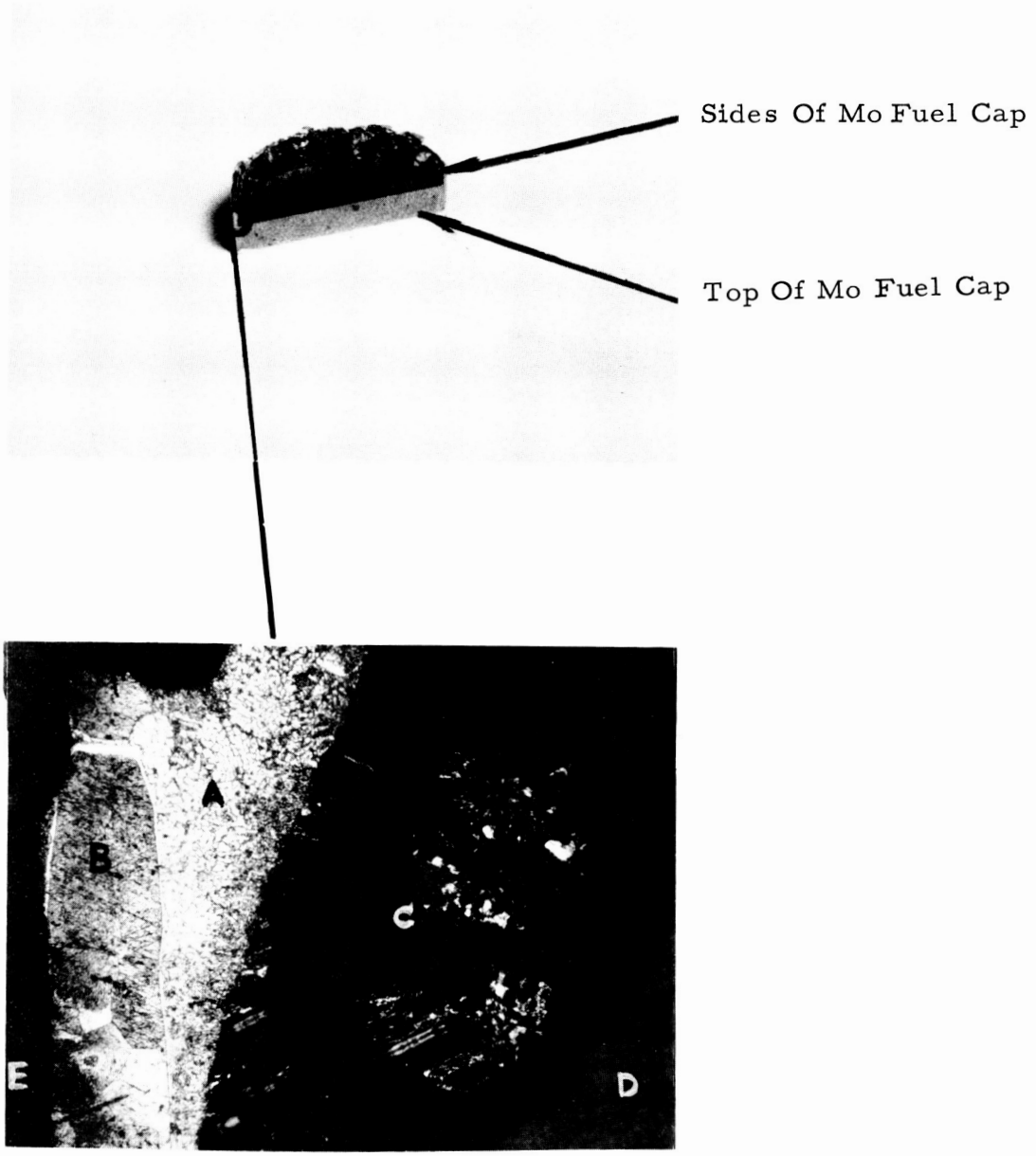
Figure 17. Components of DC-1 Fuel-Emitter After Disassembly

edge of the fuel with the fuel cap. It appeared that during the weld of the fuel cap lip to the emitter, a reaction occurred between the fuel and the sides of the fuel cap and between the emitter and the sides of the fuel cap. The emitter at the outer edge was very brittle, typical of Mo to Re welds in which excessive times and/or temperatures are involved. the UC adjacent to the weld area apparently melted during the weld and caused embrittlement of the sides of the molybdenum fuel cap. Figure 18 shows the remains of the UC fuel disk which had reacted and bonded to the sides of the molybdenum cap. The photomicrograph in Figure 18 shows the reaction zone between the UC and Mo at a magnification of 150X. It is obvious that the failure in the sides of the fuel cap resulted from the embrittlement caused by the UC-Mo reaction which occurred during welding. What had been observed during the testing as a UC-Mo reaction was apparently thermal stress cracking of the fuel cap sides which had already been embrittled during welding.

Aside from the effects of this weld, the over-all condition of the fueled emitter was quite encouraging. Althouth no chemical analysis of any component was attempted, the emitting surface of the rhenium looked bright and shiny with no visible evidence of contamination. The surface of the rhenium in contact with the fuel had only a slight tarnish over part of its area. Except at its edges, there was no visible evidence of reaction between the Re and the UC which was hyper-stoichiometric prior to test. The photomicrographs presented in Figures 19 and 20 also show no evidence of a reaction. Figure 19 gives a view of the entire cross section of the emitter from DC-1 and compares it to an "as received" rhenium foil. Except for slight differences in polish and film exposure, the emitter and the control specimen are quite similar. Figure 20 shows a similar comparison at higher magnification of the side of the DC-1 emitter which was next to the UC fuel. Again, the appearance of the two specimens is nearly the same. There was no noticeable contamination evident on the surface of the collector or on the emitter support tube.

Based on the inspection and the metallographs, it is concluded that no significant reaction occurred between the UC and the rhenium emitter. It is

apparent that much longer operation of the converter could have been performed without degradation had the mechanical failure in the structure not occurred.



(A) Reaction Zone of Mo and UC
(B) Side of Mo Fuel Cap
(C) Piece of UC Fuel
(D) Plastic Mounting
(E) Plastic Mounting

Magnification X150 , White Light,
Electropolished, Electroetched

Figure 18. Reaction of Molybdenum Fuel Cap with Uranium Carbide Fuel Disk

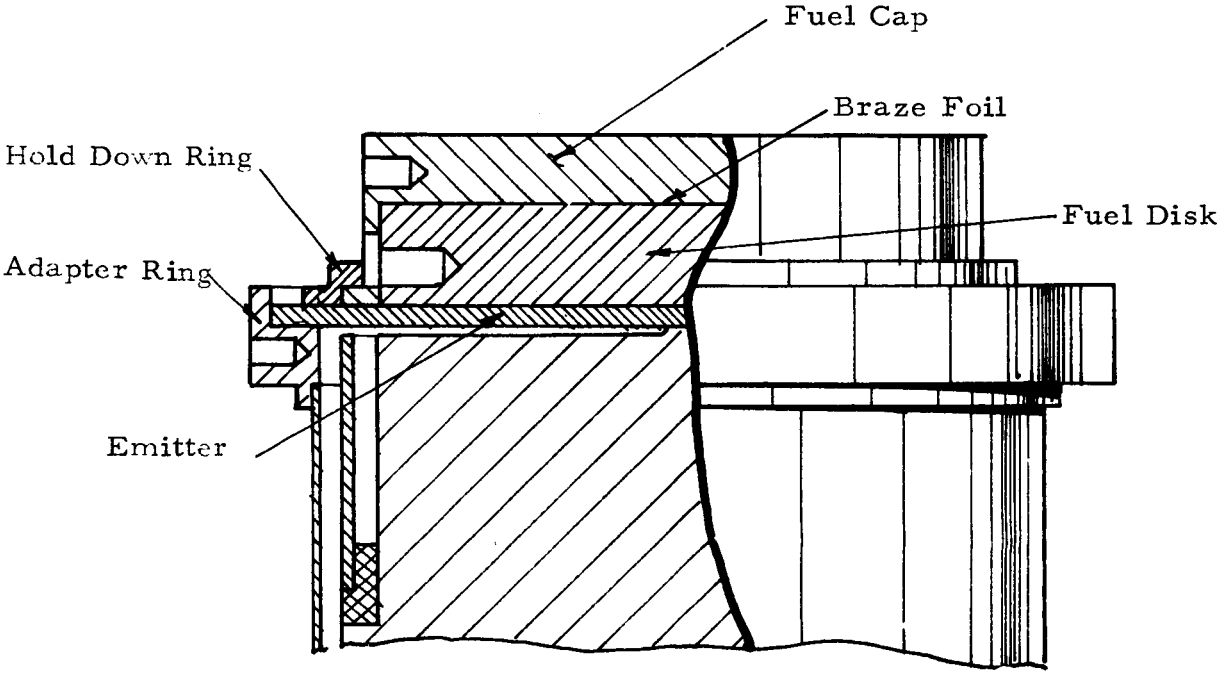
III. TEST DEVICE DC-II

A. Design and Fabrication

After the embrittlement of the Mo fuel cap curtailed the operation of DC-I, a second test device was designed with modifications in the fuel-emitter structure. As shown in Figure 21, a tungsten fuel cap was employed instead of molybdenum to avoid the Mo-UC problem. Since it was not possible to weld this tungsten cap to the rhenium emitter without melting the rhenium, a molybdenum "hold-down" ring was used to keep the fuel tightly against the emitter. This ring was separated from the UC fuel by the tungsten fuel cap and was electron beam welded to the rhenium emitter by melting its outer lip on the emitter. During the weld, a force was applied on the inner lip of the hold-down ring to achieve a tight fit of the fuel cap to the fuel and of the fuel to the emitter. To insure these fits, a clearance previous to welding of about 0.003 inch was left between the lip of the tungsten cap and the emitter and of about 0.001 inch between the hold-down ring and the emitter. To reduce the temperature differences in the fuel-emitter structure, the UC disk was brazed to the tungsten fuel cap with a 0.001 inch foil of platinum. Since this braze was on the face of the UC disk opposite to the face in contact with the emitter, the possibility of interaction between the platinum and the emitter was minimized. The rest of the fuel-emitter assembly and the diode was identical to that used in DC-I.

B. Temperature Calibrations

The calibration set-up used for DC-II was identical to that used for DC-I. The relationship between the temperatures of the fuel cap, the fuel disk, the adapter ring, and the emitter face was measured for a range of power inputs.


Fuel Cap
Braze Foil
Hold Down Ring
Fuel Disk
Adapter Ring
Emitter

The data obtained initially and after a 45-hour soak are shown in Figure 22. The curves labeled "A", "B", and "C" represent the temperature distributions during the first heating. After soaking for 45 hours with T_4 maintained at 1650°C, curves "B" and "C" shifted to curves "B'" and "C'". No change occurred in curve "A". The variation across the emitter surface was comparable to that observed in DC-1.

In comparing Figure 22 to Figure 4, two differences are evident. For a given value of the fuel cap temperature (T_1), both the fuel temperature (T_2) and the emitter temperature (T_4) are substantially higher (>90°C) in the DC-II calibration. Since the fuel was brazed to the fuel cap in DC-II, this result was not unexpected. The second difference is the shift which occurred in the adapter ring temperature (T_3) of DC-II after soaking. Whereas no shift in T_3 occurred in DC-I, an upward shift of about 60°C in T_3 occurred in DC-II. This increase was probably due to diffusion bonding between the molybdenum hold-down ring and the tungsten fuel cap. As this bonding improved, more heat transfer occurred down the walls of the fuel cap to the outer periphery of the emitter. Consequently, the temperature of the adapter ring increased relative to the temperature of the other parts of the emitter structure. It is interesting to note that the final relationship of T_3 to T_1 in Figure 22 (curve "C'") is virtually identical to that in Figure 4 (curve "C"). Thus, either errors in temperature measurements were offset by differences in thermal bonds between DC-I and DC-II, or the temperature measurements were accurate and the thermal bonds in DC-I and DC-II were about equal.

C. Converter Testing

In testing DC-II, the procedures used in DC-I were repeated. All temperature measurements were taken at open circuit conditions and all output

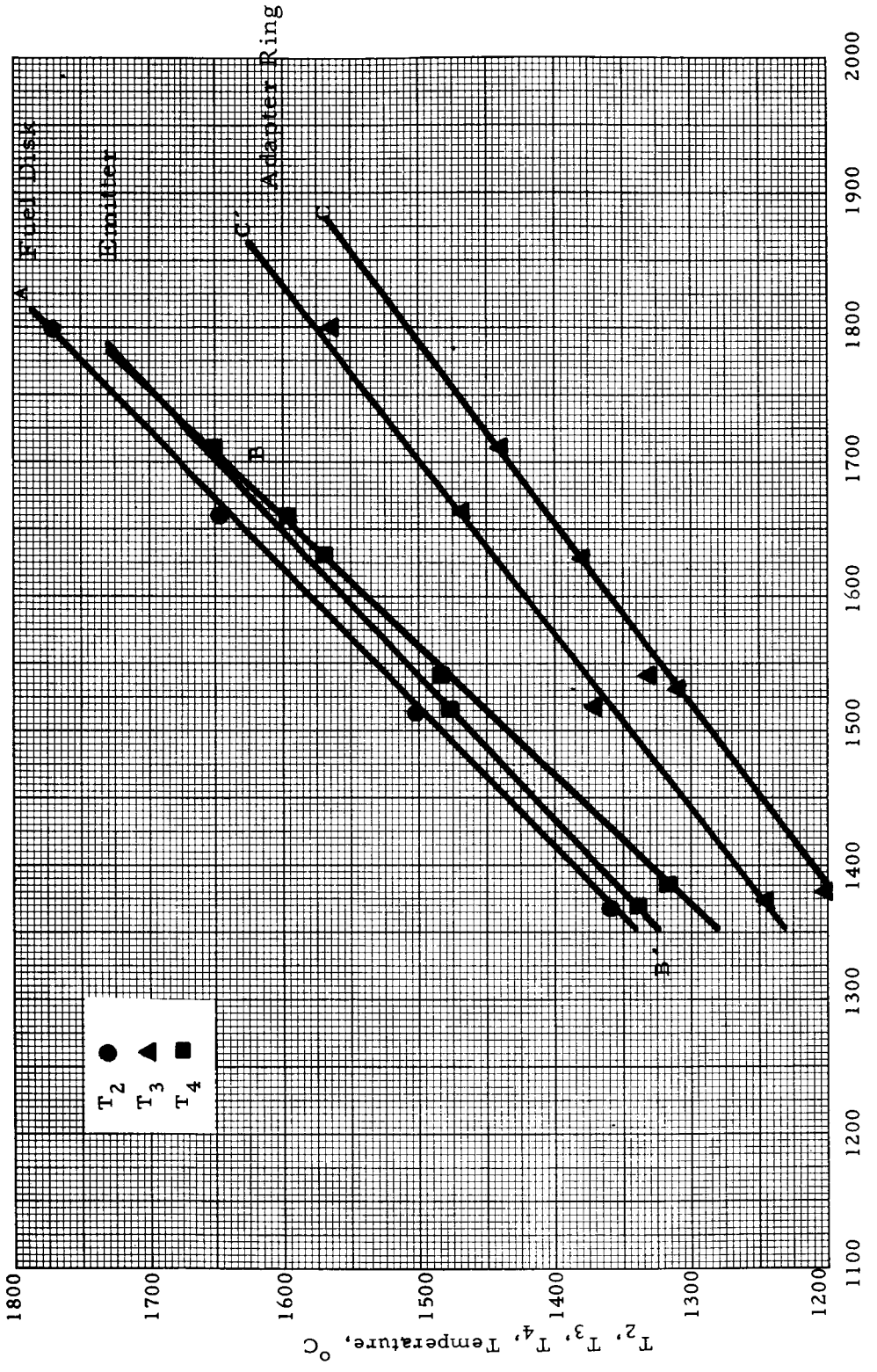


Figure 22. Results of DC-2 Fuel Emitter Temperature Calibration

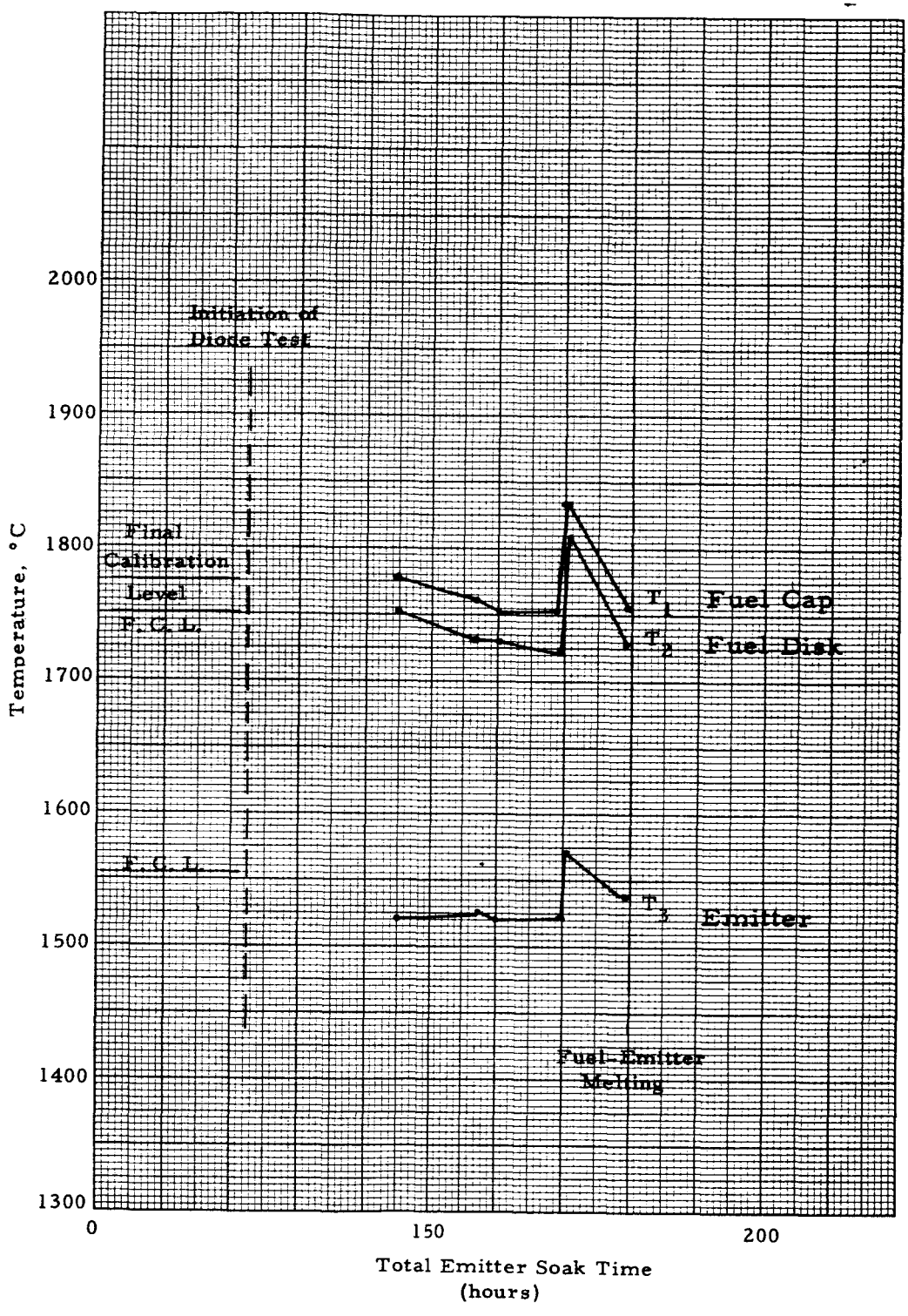


Figure 23. Variation of Fuel-Emitter Temperatures versus Time for DC-2

measurements were taken by momentary sampling techniques. This permitted the use of the temperature measurement data of Figure 22.

It was evident from the initial operation of the diode that the performance was well below the level normally observed for rhenium emitters. Figure 24 shows an I-V characteristic after 75 hours of diode operation (120 hours of total soak time). The estimated emitter temperature was 1690°C and the cesium reservoir temperature was 364°C. The output was about one-fourth of what rhenium should produce at this emitter temperature. The variation in output with cesium pressure was also quite different from typical rhenium behavior. In Figure 25, the output is shown at the same time with the emitter temperature increased to 1770°C. Although the output was substantially higher than in Figure 24, it is still far below the capability of rhenium.

Operation was continued at about this temperature level for another 44 hours without any significant change in output. However, at the 119 hour point in diode operation, it was observed that the entire fuel-emitter structure had melted into a single mass. Since this structure was barely visible from the outside of the vacuum bell jar during the test, it was not known how long this melting process had been going on. It is quite possible that it was a gradual process which occurred throughout the duration of the high temperature operation of the fuel-emitter structure. At a very early stage of testing, there was some evidence of diffusion of the fuel disk material out through the black-body hole in the side of the fuel cap. It also appeared at this same time that the black-body hole in the fuel disk had deformed due to migration of the fuel material.

Some further testing was conducted after the melting of the fuel-emitter structure was observed. However, since the temperatures were unknown and the emitter was obviously poisoned, no attempt was made to analyze the data obtained.

IV. SUMMARY

Although structural problems with the fuel-emitter assemblies prevented complete attainment of the program objectives, meaningful information was produced. The 200-hour results previously obtained on the vanadium-brazed fue-emitter interface were repeated on DC-1 with a braze free interface. These results prove that uranium carbide does not affect the emission from rhenium at temperatures consistent with high power density for at least 200 hours. The doubts about the possible effects of vanadium on the previously obtained results were convincingly removed.

Furthermore, the consistency of the temperature measurements obtained for DC-1 and DC-2 demonstrated the reliability and reproducibility of the temperature calibration technique employed. This same technique can now be applied in future diffusion-emission tests with increased confidence in its meaningfulness.

The trouble free performance of the basic diode structure demonstrated its suitability for this type of testing. Except for the fuel-emitter structural problems, this diode has proven to be a remarkably rugged, reproducible, flexible, precise, and yet inexpensive vehicle for diffusion-emission tests. For tests of cermet fuels in which metallurgical bonds between the fuel and emitter can be maintained despite thermal cycling, this diode structure should be ideal.

The temperature calibration results also produced a quantitative measure of the temperature drop penalty at the fuel-emitter interface inherent in ceramic fuels. Although very high contact pressures were obtained between the fuel and the refractory metal surfaces in contact with it, interface temperature differences of 30°C to 60°C were observed even after prolonged soaking. After cycling, these temperature drops became as high as 100°C. When these measurements were

taken, the fuel-emitter interface was at the equivalent of an "open-circuit" condition. It is obvious that the increased heat fluxes required for power producing operation greatly increased these temperature differences.

Since the lifetime of a fuel in a reactor environment is strongly dependent upon temperature, these temperature differences represent a severe penalty for carbide ceramic fuels. Because cermet fuels can be metallurgically bonded to emitter materials, such severe temperature drops can be avoided by their use. The temperature measurements taken in this study provide an indication of a major advantage of cermet fuels for thermionic applications.

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