FABRICATION AND LIFE TESTING OF THERMIONIC CONVERTERS

FINAL REPORT

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An unfueled converter containing a chloride-fluoride duplex tungsten emitter of 4.78 eV vacuum work function was tested for 46,647 hours at an emitter temperature of 1973°K and an electrode power output of about 8 watts/cm². The test demonstrated the superior and stable performance of the (110) oriented tungsten emitter at high temperatures. Three UC-10 ZrC (C/U = 1.04, tungsten additive = 4 wt %) fueled converters were fabricated and tested at an emitter temperature of 1873°K. Converter containing chloride-arc-cast duplex tungsten cladding showed higher initial thermionic performance and slower rate of performance drop than converter containing chloride-fluoride duplex tungsten cladding. This is believed to be due to the superior fuel component diffusion resistance of the arc-cast tungsten substrate used in the fuel cladding. It was shown that a converter containing a carbide fueled chloride-arc-cast duplex tungsten emitter with an initial electrode power output of 6.80 watts/cm² could still deliver an electrode power output of 6.16 watts/cm² after 18,632 hours of operation at an emitter temperature of 1873°K.
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THERMIONIC CONVERTERS

FINAL REPORT

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INTRODUCTION

This report describes the work carried out from July 3, 1969 through January 18, 1973 under Contract NAS3-12980 on the fabrication and out-of-pile testing of thermionic converters. Due to the cancellation of the U.S. thermionic program in January, 1973, the testing originally planned for this contract had to be reduced. The content of this report is therefore limited to the following subjects:

1. Continuation of the life-testing of the unfueled converter LC-9, which was carried out previously under Contract NAS3-8504.

2. Fabrication, life-testing and post-operational examinations of the 90UC-10ZrC* fueled converter LC-10.

3. Fabrication and life-testing of the 90UC-10ZrC fueled converter LC-11.

4. Fabrication and short term testing of the 90UC-10ZrC fueled converter LC-12.

A list of reports published previously on thermionic material work sponsored by NASA at Gulf General Atomic is shown on the page next to the title page.

*Concentration expressed in mole percent.
SUMMARY

This report concerns the fabrication, testing and post-operational examinations of out-of-pile converters.

Life testing of an unfueled converter designated as LC-9 was continued at an emitter temperature of 1973°K. LC-9 contains an unfueled chloride-fluoride duplex tungsten emitter of 4.78 eV vacuum work function, which was tested previously under Contract NAS 3-8504. A total of 46,647 operating hours was accumulated, at an electrode power output of about 8 W/cm². The test demonstrated that an unfueled converter containing a (110) oriented emitter can maintain superior and stable performance for a long period of time.

Three carbide fueled converters, designated as LC-10, LC-11, and LC-12, were fabricated. LC-10 contained a 90UC-1OZrC (C/U = 1.04, tungsten additive = 4 wt-%) fueled chloride-fluoride duplex tungsten emitter and a niobium collector. The emitter had an as-fabricated vacuum work function of 4.9 eV which decreased to 4.74 eV after it was incorporated into the converter, presumably because of contamination of the emitter surface during the assembly of the converter. LC-10 was tested for a period of 8244 hours at an emitter temperature of 1873°K. Performance mappings, electrode work function measurements and electrode relative emittance determinations were made initially and at 1338 and 5289 hours of operation. The decrease of the electrode power output during the first 1338 hours from 5.94 W/cm² to 5.46 W/cm² is believed to be due to an increase in cesiated collector work function by 0.2 to 0.3 eV from the initial value due to fuel components diffusing through the cladding and depositing on the collector surface. This was accompanied by an increase in the electrode emittance. Performance decreases after 1338 hours were not associated with electrode work function or electrode emittance change. It appears that such performance decreases were
caused mainly by a shift of the electron gun filament in the emitter cavity causing a change in the temperature profile of the emitter. The test was terminated at 8244 hours because of cesium leakage through the converter envelope. Post-operational examinations showed that the leakage occurred at the welded joint between the niobium cesium lead tube and the copper cesium reservoir. There was no significant change in the emitter diameter, and the bond between the chloride tungsten and the fluoride tungsten remained in excellent condition. A thin layer (~0.3 mil) of UWC₂ interaction layer was present at the fuel-cladding interface but no uranium (within a few tenths of a percent) was detected in the grain boundaries of the tungsten cladding. The collector surface had a dark appearance and a reaction layer of a few microns thickness. Analytical results obtained on samples taken from the collector surface indicated an average carbon flux of $1.2 \times 10^{-10}$ gm per cm² of fuel-cladding interface area per hour over the 8244 hours of testing period. The corresponding uranium and zirconium fluxes were $4 \times 10^{-11}$ and $2.2 \times 10^{-12}$ gm per cm² per hour, respectively.

LC-11 contained a 90UC-10ZrC (C/U = 1.04, tungsten additive = 4 wt-%) fueled chloride-arc-cast duplex tungsten emitter and a niobium collector. The emitter had a vacuum work function of 4.9 eV. The converter was tested at an emitter temperature of 1873°K for a period of 18,632 hours. The initial electrode power output was 6.80 W/cm² which was much higher than that of LC-10 (5.94 W/cm²). During the first 5000 hours, the electrode power density decreased to 6.53 W/cm²; however, the 6.8 W/cm² value was regained after thermocouple and electron gun filament replacement. At the end of 18,632 hours of operation, the electrode power output was 6.16 W/cm². Compared with the LC-10 test data, LC-11 had a higher initial thermionic performance and a slower rate of performance drop. The slower rate of performance drop is believed to be due to the superior fuel component diffusion resistance of the arc-cast tungsten substrate used in the fuel cladding.
LC-12 contained a 90UC-10ZrC (C/U = 1.015, tungsten = 4 wt-%) fueled chloride-fluoride duplex tungsten emitter and a niobium collector. While the cladding material was the same as that of LC-10, the converter design was modified in order to minimize emitter contamination during assembly and to improve converter life expectancy. The emitter had a vacuum work function of 4.9 eV determined from measurements made on the assembled converter. Due to the termination of the U.S. thermionic program, the converter was tested for only 313 hours. The initial performance mapping results showed that the performance of LC-12 was better than that of LC-10, but poorer than that of LC-11. The initial electrode power output 6.2 W/cm² decreased to 5.81 W/cm² at an operating time of 313 hours. Since the latter is lower than that of LC-11 (6.16 W/cm²) even after LC-11 was operated for 18,632 hours, it is believed that the chloride-arc-cast duplex tungsten represents a better cladding material for thermionic fuel element application from the point of view of thermionic performance. An in-pile test of a carbide fueled converter containing such cladding material should be carried out in order to establish its life and performance stability.
1. LIFE TESTING OF UNFUELED CONVERTER LC-9

1.1 TESTING OBJECTIVE

LC-9 was fabricated in 1966 under Contract NAS3-6471. The design characteristics, components and fabrication techniques for LC-9 have been described in previous summary reports for Contracts NAS3-6471 and NAS3-8504. The unique feature of this converter lies in its partially (110) oriented chloride-fluoride duplex tungsten emitter* of 4.78 eV vacuum work function, as versus 4.5 eV for the (100) oriented fluoride tungsten emitters** used in converters fabricated on this program prior to LC-9. The specific objective of the test is to evaluate the performance improvement and the performance stability of a converter containing an emitter of high work function.

1.2 PREVIOUS TESTING RESULTS

Life-testing of LC-9 was carried out at an average emitter temperature of 1973°K. The test began on December 27, 1966 under Contract NAS3-8504, and was continued under the same contract until December 7, 1970 for a total testing time of 29,600 hours. The output remained constant (within 3%) after the replacement of the electron gun filament at 2453 hours for better alignment in the emitter cavity. Compared with another unfueled converter, LC-7, containing a fluoride tungsten emitter of 4.5 eV vacuum work function under the same operating conditions, the output of LC-9 was about 25% higher. Although the emitter thermocouple readings showed significant changes with time, the power output remained the same for the same power input and the same operating conditions. It is believed that the observed changes in emitter thermocouple readings were caused by thermocouple degradation at the operating temperature (1973°K average) of the emitter, and that the emitter temperature stayed essentially constant during the test.

*12.5 mil chloride tungsten emitting layer prepared by the hydrogen reduction of tungsten chloride over a fluoride tungsten substrate prepared by the hydrogen reduction of WF₆.

**Prepared by the hydrogen reduction of WF₆.
1.3 TESTING RESULTS FOR THE PRESENT REPORTING PERIOD

The life-testing of LC-9 was continued under Contract NAS3-12980 from December 8, 1970. The operating history is shown in Fig. 1. The total testing time accumulated was 46,647 hours. The test results clearly demonstrate that an unfueled converter containing chloride-fluoride duplex tungsten emitter of (110) preferred crystal orientation can maintain high and stable thermionic performance for a long period of time.
Fig. 1. Power output and operating parameters of LC-9 (Sheet 1 of 2)
Fig. 1. Power output and operating parameters of LC-9 (Sheet 2 of 2)
2. **FABRICATION, LIFE-TESTING AND POST-OPERATIONAL EXAMINATION OF CARBIDE FUELED CONVERTER LC-10**

2.1 **TESTING OBJECTIVE**

Previously under Contract NAS3-6471\(^1\), a thermionic converter containing a 90UC-10ZrC fueled fluoride tungsten emitter was fabricated. This converter, designated as LC-8, was life-tested in the temperature range 1673-2073°K to determine if the addition of 4 wt-% of tungsten to the carbide fuel material and the close control of fuel stoichiometry to insure a slight excess in carbon (C/U = 1.04) would improve the thermionic performance stability of the converter. The test results\(^3\)\(^4\) showed that while the converter performance remained relatively stable at 1673°K, the power output decreased with time at 1873° and 2073°K. Diagnostic studies indicated that the observed decreases in converter output were largely due to the increase of the cesiated collector work function. Post-operational examinations of the components of LC-8 showed that the work function changes were most probably caused by the accumulation on the collector of fuel components diffusing through the fluoride tungsten cladding.

To improve the thermionic performance stability of carbide fueled converters, it was believed necessary to reduce the transport of fuel components through the tungsten cladding. Out-of-pile measurements\(^5\) made on the transport rates of fuel components from 90UC-10ZrC (4 wt % tungsten, C/U = 1.04) through various types of tungsten claddings showed that the transport rates were strongly dependent upon the microstructures of the tungsten cladding. The rates could be lowered significantly if the fluoride tungsten cladding of columnar grain structures was replaced by chloride tungsten or arc-cast tungsten claddings of equiaxial grain structures.
Based on this data, it was decided to evaluate a carbide fueled-duplex (chloride on fluoride) tungsten clad emitter in a cesiated converter. The chloride tungsten provided an emitting surface of (110) preferred orientation and high vacuum work function together with a layer of equiaxial grain structure of low fuel transport rate; the fluoride tungsten contributed a substrate of stable grain structure and high creep strength. Although it was anticipated that the use of a chloride-fluoride duplex tungsten cladding would improve the electrical power output and thermionic performance stability of carbide fueled converters, no experimental data were available to substantiate such anticipation. The test of LC-10 was aimed at providing long-term, out-of-pile information to determine whether its duplex clad emitter would provide significantly better performance than the fluoride clad emitter in LC-8.

The essential components and characteristics of LC-10 are summarized in Table 1. Compared with LC-8, the major difference was that LC-8 had a fluoride tungsten cladding of 1 mm thickness, while LC-10 had a chloride-fluoride duplex tungsten cladding consisting of a chloride tungsten emitting layer of 0.5 mm thickness over a fluoride tungsten substrate of 0.75 mm thickness.

**TABLE 1**

**COMPONENTS AND CHARACTERISTICS OF LC-10**

<table>
<thead>
<tr>
<th>Fuel</th>
<th>90UC-10ZrC, 4 wt-% tungsten, C/U = 1.04</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cladding</td>
<td>Chloride tungsten emitting layer, 0.5 mm thick Fluoride tungsten substrate, 0.75 mm thick Emitting area 16.43 cm²</td>
</tr>
<tr>
<td>Collector</td>
<td>Niobium</td>
</tr>
<tr>
<td>Insulator Seal</td>
<td>Niobium sleeves</td>
</tr>
<tr>
<td></td>
<td>Lucalox insulation</td>
</tr>
<tr>
<td></td>
<td>Litton metallising layer</td>
</tr>
<tr>
<td></td>
<td>Cu-10 wt-% Ni braze</td>
</tr>
<tr>
<td>Interelectrode spacing</td>
<td>Hot 0.25 mm</td>
</tr>
<tr>
<td></td>
<td>Cold 0.23 mm</td>
</tr>
</tbody>
</table>
2.2 DESIGN AND FABRICATION PROCEDURES

The design and fabrication procedures for LC-10 were essentially the same as that for LC-8, as described in a previous summary report. The major differences involve the fueled emitter design and fabrication in the following areas. First, to increase the sensitivity for determining the amount of carbon deposited on the collector surface, $^{14}C$ was incorporated into the carbide fuel material to allow the detection of at least $10^{-7}$ gm of carbon by beta-counting, as compared to a sensitivity of $2 \times 10^{-6}$ gm by the combustion and gas chromatographic techniques used previously. Secondly, the carbide fuel was clad with chloride-fluoride duplex tungsten instead of fluoride tungsten. The chloride tungsten layer was thoroughly evaluated with respect to its degree of (110) preferred orientation and its vacuum work function was determined both before and after the emitter was incorporated into the converter. Thirdly, the sealing of the end of the emitter after the fuel slabs were loaded into the emitter was made with chloride tungsten instead of fluoride tungsten and the deposition of the chloride tungsten emitting layer over the fluoride tungsten substrate was accomplished at the same time. The deposition of the chloride tungsten was carried out under the following conditions to insure the presence of a high degree of (110) preferred orientation in the deposit: Substrate temperature, 1373 K; hydrogen flow rate, 240 c.c./min; tungsten chloride flow rate, 230 c.c./min; tungsten chip temperature, 1123 K; residual gas pressure in deposition chamber, ~3 torr.

2.2.1. Emitter Configuration

The configuration and dimensions of LC-10 emitter are shown in Fig. 2 which is self-explanatory.

2.2.2. Emitter Characterization

After the completion of the chloride tungsten deposition, the emitter was ground to the dimensions required, electropolished in 1 wt-% NaOH, and outgassed in vacuum at 1873 K for 100 hours. The distributions of $\langle 110 \rangle$ crystal axes in the deposit was then determined
Note: Emitting area is defined by emitter length (3.195 cm) and emitter diameter (1.638 cm) at 1600°C and equals 16.43 cm².

Fig. 2. Configuration and dimensions (cold) of LC-10 emitter

1. Carbide fuel slab
2. Chloride tungsten cladding, 0.5 mm thick on side wall; 0.625 mm thick on bottom
3. Fluoride tungsten cladding and stem, 0.75 mm thick
4. Fluoride tungsten cup, 2.75 mm high, 0.25 mm side wall thickness, 20 mil bottom thickness
5. Fluoride tungsten spacer, 1 mm thick, gap between spacer and cup 0.25 mm
6. Collector
7. Fluoride tungsten blank
by X-rays\textsuperscript{5}. Figure 3 shows the distributions of the \textlangle 110 \rangle crystal axes at 0.32, 1.27, and 2.54 cms from the closed end of the outgassed emitter. The corresponding vacuum work functions determined in a vacuum emission cell for cylindrical emitters at 1873°K were 4.90, 4.90 and 4.91 eV, respectively.

The emitter was then brazed to the tantalum transition with vanadium, and the tantalum transition was machined to fit the upper sleeve of the insulator seal. After degreasing with a mixture of xylene-acetone, the temperature profile and the vacuum work function of the emitter were determined in an apparatus described in a previous report\textsuperscript{1}.

Figure 4 shows the temperature profile of the emitter at an average surface temperature of 1866°K. The average surface temperature deduced from optical pyrometer readings was 49° and 57° lower than the readings of the two emitter thermocouples, T\textsubscript{E1} and T\textsubscript{E2}, respectively.

The vacuum work function of the overall emitter surface was measured as a function of time for average emitter temDeratures of 1836-1859°K. The results are shown on the left hand side of Fig. 5. The vacuum work function started at about 4.0 eV and increased gradually to about 4.68 eV in 52 hours, which was much lower than the value 4.90 eV obtained before the emitter was brazed to the tantalum transition.

At this point, it was decided to stop the testing in the apparatus for emitter temperature profile determination, and to carry out the rest of the converter assembly steps before final outgassing in the assembled converter, since the emitter would have to be handled in the electron beam welder and in the shop. The emitter structure was then welded to the insulator in the electron beam welder in a vacuum of 10\textsuperscript{-5} torr range maintained with a baffled oil diffusion pump. The welded emitter structure was then sent to the
Fig. 3. Distribution of the <110> axes in LC-10 emitter after electropolishing
Fig. 4. LC-10 emitter temperature profile after brazing to tantalum transition but prior to welding to insulator
In apparatus for temperature profile measurement after brazing to tantalum transition during bake-out after assembly into converter 1857°-1873°K

Fig. 5. Effective vacuum work function of LC-10 emitter at different stages of converter fabrication
shop for machining the lower insulator skirt to fit the collector. Final assembly of the emitter and the collector was carried out in the remote assembly apparatus in $10^{-7}$ torr range vacuum maintained with an ion pump. Prior to the assembly, the emitter and the collector were outgassed at 1873°K and 1073°K, respectively, for 3 hours. They were then brought together through the remote assembly arrangement and the Cu-Ti final closure braze was melted by induction heating to complete the converter assembly.

The assembled converter was then installed in the bakeout station, the emitter was brought to about 1873°K and the vacuum work function was followed as a function of time. The results are shown in Fig. 5 (52 hr-125 hr.). It can be seen that the vacuum work function remained at about 4.66-4.69 eV. At the end of 125 hours of accumulated testing time, the emitter temperature was raised to 1973°K for 8-1/2 hours. The vacuum work function results obtained are also included in Fig. 5 (region A-A) and shown in an expanded form in Fig. 6. The vacuum work function stayed at about 4.73 eV which dropped to 4.70 eV after the emitter temperature was brought back to 1873°K. These vacuum work functions were still much lower than that obtained in the cylindrical vacuum work function apparatus (~ 4.90 eV) before the emitter was brazed to the tantalum transition. It was suspected that the emitter surface was contaminated. A decision was made to remove the emitter from the converter and clean the emitting surface.

The emitter subassembly, including the emitter, the tantalum transition and the insulator seal, was separated from the collector with a jeweler's saw at a point just below the bottom of the lower skirt of the insulator seal. Both the emitter surface and the collector surface were found to be shiny, showing no evidence of macroscopic contamination. An attempt was made to clean the emitter surface by heating the subassembly in vacuum in the apparatus used for determining the emitter temperature profile, which had a much higher conductance for pumping than a converter under final bakeout. The effective vacuum work function at 1873°K was 4.82 eV at the
Fig. 6. Effective vacuum work function of LC-10 emitter as a function of bakeout time at 1973°K in the assembled converter. (Expanded version of the region marked A–A in Fig. 5)
beginning, 4.81 eV after 2 hours, and stayed at 4.76 eV between 19 hours and 49 hours of heating time. This is slightly higher than the 4.70 eV observed during the bakeout of the assembled converter but fell short of the 4.90 eV determined in the cylindrical vacuum emission apparatus.

For a more effective cleaning of the emitter surface, the emitter was electropolished to remove .0013 cm from its radius. After the electropolishing, the emitter was thoroughly washed in distilled water, and successively cleaned ultrasonically in the following media, a mixture of acetone and xylene, distilled water, xylene, distilled water and acetone. The vacuum work function of the electropolished emitter was then determined at 1874-1896°K. The results are shown in Fig. 7. It can be seen that the effective vacuum work function stayed at 4.86 eV after 20 hours. The work function results in Fig. 7 (also all the other work function results reported previously) were calculated from the observed emission current by applying the average surface temperature to the Richardson equation. However, if consideration is given to the exponential variation of the emission current with temperature over the emitter surface according to the temperature profile curve, then the work function obtained was about 0.02 eV higher. On this basis, the vacuum work function was 4.88 eV, which is within the experimental error range of the 4.90 eV value originally obtained. Figure 8 shows the temperature profile of the emitter at an average surface temperature of 1874°K. The average emitter surface temperature deduced from the pyrometer readings was 14° and 28° lower than the readings of the emitter thermocouples T_E1 and T_E2 respectively. This difference between T_E2 and the average surface temperature of the emitter was used during the early stage of life-testing to establish the emitter temperature T_E.
Fig. 7. Effective vacuum work function of LC-10 emitter after removal from converter and cleaning the surface by electropolishing 0.5 mil off its radius. The numbers beside each point are the effective vacuum work function in eV and the average surface temperature in °K.
Fig. 8. LC-10 emitter temperature profile after removing from converter and electropolishing. The emitter was attached to the tantalum transition and the insulator seal and a small part of the collector top was left on the lower skirt of the insulator seal during the removal of the emitter from the converter with a jeweler's saw.
With the cleaning of the emitter surface accomplished, a new collector was made and the converter was reassembled and again installed in the bakeout station. The vacuum work function of the emitter was determined after the emitter was brought to an average temperature of 1873°K. A value of 4.74 eV was obtained, which stayed constant for a period of 115 hours. This was again lower than the desired value of 4.90 eV. However, it was decided after receiving the approval of the NASA Project Manager, to complete the processing steps and to initiate converter life-testing.

2.3 TESTING RESULTS

The emitter of LC-10 was brought to an average temperature of 1873°K on August 21, 1970. Performance mapping and diagnostic studies were then carried out to establish the initial electrode work functions and emittance, and the optimum operating conditions for the life-testing operation. Figure 9 shows the initial optimum electrode power output as a function of current density for average emitter temperatures of 1673°, 1773° and 1873°K. Figure 10 contains the measured effective work functions of LC-10 emitter superposed on a Rasor-Warner plot for an operating time of 128 hours. The results indicate that the bare work function of the emitter was slightly higher than 4.5 eV. The discrepancy between the vacuum work function obtained in the bakeout station (4.74 eV) and the bare work function deduced here from the Rasor-Warner plot is considered to be unimportant. For the fueled emitter, this method is probably useful only for following the change in work function rather than for determining the absolute value of the work function. Figure 11 shows the effective work function of the collector as a function of $T_c/T_R$ ($T_c$ = collector temperature, $T_R$ = cesium reservoir temperature, both in °K) at an operating time of 135 hours. These data points agree within 0.1 eV with the corresponding results for the LC-9 emitter². The power input for maintaining the emitter at 1873°K, with the collector at 1006°K and cesium reservoir at 631°K under open circuit condition was 397 watts. These are the baseline
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<th>$T_R(°K)$</th>
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Fig. 9. Initial optimum electrode density of LC-10 as a function of current density at various emitter temperatures.
Fig. 10. Effective emitter work function of LC-10 for various $\frac{T_E}{T_R}$ ratios at 128 hours of operating time. $T_E =$ Emitter temperature, $T_R =$ cesium reservoir temperature.
Fig. 11. Effective collector work function of LC-1U for various $T_c/T_R$ ratios at 135 hours of operating time. $T_c$ = collector temperature, $T_R$ = cesium reservoir temperature.
conditions used for the evaluation of any change in electrode emittance during the life-testing.

Life-testing of LC-10 began on August 27, 1970 at an average emitter temperature of 1873°C, a collector temperature of 1073°C, a cesium reservoir temperature of 625°C, and at the same current density (10.60 amp/cm²) as that used for the life testing of LC-8. These collector and cesium reservoir temperatures represent the optimum values for the emitter temperature and current density chosen for the test, as determined by the initial thermionic performance mapping. The total operating time for LC-10 at the initiation of life-testing was 151 hours. The electrode power output at that time was 5.94 W/cm².

The power output and operating parameters for LC-10 are shown in Fig. 12 as a function of time. The electrode power output dropped gradually from the initial value of 5.94 W/cm² to 5.46 W/cm² at an operating time of 1338 hours. Performance mapping and diagnostic studies were initiated in order to throw light on the causes of the observed thermionic performance degradation. It was found that there was no significant change in the optimum cesium reservoir temperature, the optimum collector temperature, and the emitter work function. However, the effective collector work function was raised by 0.2 to 0.3 volt (see Fig. 13). At the same time the power input needed for maintaining an average emitter temperature of 1873°C, with the collector at 1006°C and cesium reservoir at 631°C under open circuit condition was 453 watts, an increase of 56 watts from the baseline value. These results imply that the observed degradation in converter performance was probably caused by the contamination of the collector surface by fuel components diffusing through the cladding, which raised both the effective collector work function and the electrode emittance and thus lowered the thermionic and the thermal performance of the converter. Figure 14 compares the optimum electrode power output as a function of current density for average
Fig. 12. Power output and operating parameters of LC-10 (Sheet 1 of 2)
Fig. 12. Power output and operating parameters of LC-10 'Sheet 2 of 2')
Fig. 13. Effective collector work function of LC-10 as a function of $T_C/T_R$ ($T_C =$ collector temperature, $T_R =$ cesium reservoir temperature) at operating times of 135 and 1648 hours.
Fig. 14. Optimum electrode power density of LC-10 as a function of current density for various emitter temperatures at the operating hours indicated.
emitter temperatures of 1673°, 1773° and 1873°K with the initial optimum electrode power output. The loss of electrode power output amounted to about 0.5 W/cm² at the current density (10.6 amp/cm²) used for the life-testing for all the three emitter temperatures studied.

Test of LC-10 was continued after completing the performance mapping and diagnostic studies carried out between 1338 and 1648 hours of operating time. The converter output continued to decrease until a value of 5.10 W/cm² was reached at an operating time of 2920 hours. Examination of the electron bombardment filament showed a shift in its position in the central cavity of the emitter. This shift in position resulted in a change in emitter temperature profile so that a slight decrease in input power (by 5 watts in a total of 865 watts) was needed in order to keep the reference emitter thermocouple No. 2 at the normal operating temperature, while emitter thermocouple No. 1 was cooler by 10°C than the normal operating temperature. Thus, the decrease in power output could be due to a change in emitter temperature profile because of the shift in the position of the electron bombardment filament in the emitter cavity. The converter was shutdown on January 7, 1971 at an operating time of 3080 hours for the replacement of the electron gun filament and the two emitter thermocouples. Emittance measurements and cesium reservoir temperature optimization study carried out during converter startup showed no significant changes from the results obtained during the diagnostic studies at 1338 to 1648 hours of operating time. The change of the filament and emitter thermocouples caused a shift in the emitter thermocouple behaviors. Prior to the filament and emitter thermocouple changes, emitter thermocouple No. 2 was reading higher than emitter thermocouple No. 1. The order was reversed after the filament and emitter thermocouple changes, with emitter thermocouple No. 1 reading higher than emitter thermocouple No. 2. Using emitter thermocouple No. 1 as the reference, the electrode power output was 5.36 W/cm² at an average emitter temperature of 1873°K, which was the same as the electrode power output after the last diagnostic studies. Thus, the decrease in electrode power output observed after the last diagnostic
studied (i.e., between about 1500 to 3000 hour operating time) was probably due to the change in emitter temperature profile when the electron gun filament shifted its position gradually in the emitter cavity.

The test was continued and the power output dropped from 5.36 W/cm² at 3080 hours to 5.00 W/cm² at 5289 hours. Since the decrease was more than 5%, performance mapping and diagnostic studies were therefore carried out according to the contractual requirements. Figure 15 shows the optimum electrode power output as a function of current density and emitter temperature. The data obtained in two previous diagnostic studies are included for comparison. The results clearly indicate the decrease of converter output with operating time. However, no significant change in emitter work function was observed when measurements made at 128, 1455, and 5415 hours of operating time were compared (see Fig. 16). Change in collector work function was noted between 135 and 1648 hours of operating time but not between 1648 and 5416 hours of operating time (see Fig. 17). The input power needed for maintaining an average emitter temperature of 1873°K at reference collector and cesium reservoir temperatures (1006°K at reference collector and cesium reservoir temperatures (1006°K and 631°K, respectively) under open circuit conditions (455 watts) did not differ significantly from the value of 453 watts determined during the last diagnostic studies. It appears that although the power output of LC-10 decreased with time, significant changes in electrode emittance before an operating time of 1648 hours. The performance change between 1648 hours and 5289 hours, was not accompanied by electrode work function or electrode emittance change.
Fig. 15. Optimum electrode power density of LC-10 as a function of current density for various emitter temperatures at the operating hours indicated.
Fig. 16. Effective emitter work function of LC-10 as a function of $T_E/T_R$ ($T_E$ = emitter temperature, $T_R$ = cesium reservoir temperature) at different operating times.
Fig. 17. Effective collector work function of LC-10 as a function of $T_C/T_R$ ($T_C =$ collector temperature, $T_R =$ cesium reservoir temperature) at operating times of 135, 1648, and 5416 hours.
The electrode power output increased to 5.3 W/cm² when the converter was brought back to normal operating conditions after completion of the diagnostic studies. The output again dropped to 5.0 W/cm² at 6350 hours of operating time. The power input for maintaining an average emitter temperature of 1873°K at reference collector and cesium reservoir temperatures under open circuit conditions was found to be 463 watts, which was not significantly different from the 453 watts determined at an operating time of 1358 hours. The cesiated collector work function was measured at 6355 hours and a $\frac{T_c}{T_R}$ value of 1.49 ($T_c = 634°K$, $T_R = 424°K$, $T_E = 1216°K$). The measurement was completed in two hours from the time the normal operating temperatures of emitter, collector, and cesium reservoir were changed to the values indicated. It was hoped that in doing so the collector surface conditions might not be changed significantly from that in the operating converter and the cesiated collector work function obtained would be closer to the true operating value. The experimental point (shown in Fig. 13) was found to agree reasonably well with those relating the cesiated collector work function to $T_c/T_K$ at an operating time of 1648 hours (see Fig. 13). Here again, the observed performance change cannot be attributed to any change in collector work function or electrode emittance.

The converter was shutdown at an operating time of 6525 hours because of an electric power failure. Upon startup, the electrode power output was 5.22 W/cm², but returned to 5.0 W/cm² in about 40 hours. On August 16, 1971 at 7598 hours of operation, the converter was again shutdown for the replacement of the electron gun filament, the two emitter thermocouples and the three collector thermocouples. The electrode power output prior to the shutdown was 4.62 W/cm². The emittance data taken prior to the shutdown agreed with that obtained at 6351 hours of operation, indicating no change in electrode surface conditions. Upon restart, it was found that the optimum cesium reservoir temperature had increased from 625°K to 643°K. The electrode output was 5.18 W/cm². If the cesium reservoir
temperature was kept at the original value of 625°C, the output voltage of the converter was too low to maintain the operating current of 10.6 amp/cm$^2$ with the load maintained at the minimum resistance position. At a converter current of 10.5 amp/cm$^2$ electrode output was only 3.76 W/cm$^2$. Since the converter current output and therefore the emitter temperature was too sensitive to small change in cesium temperature, it was considered risky to leave the converter at such an off-optimum condition. The test was therefore continued at a cesium reservoir temperature of 643°C.

On September 8, 1971, at a total operating time of 7788 hours, converter performance mapping and diagnostic studies were initiated. Figure 18 shows the optimum electrode output as a function of current density and emitter temperature. When Fig. 18 is superposed on Fig. 15, it can be seen that no significant changes have occurred in the shapes of these curves from that obtained at 5295 hours except in the region of 1873°C emitter temperature and current densities higher than 10 amperes/cm$^2$. There were also no changes observed in the emitter and collector work functions and electrode emittance from the last previous set of diagnostic data (compare Fig. 19 with Fig. 16 and Fig. 20 with Fig. 17).

On September 20, 1971, at 8080 hours, a cesium leak was detected by partial-pressure-gage scan of the residual gas in the bell jar. The converter became so starved of cesium vapor it was not possible to maintain the 10.6 amp/cm$^2$ with the minimum resistance. Test of the converter was therefore terminated on October 4, 1971, after a total operating time of 8244 hours.

2.4 POST-OPERATIONAL EXAMINATIONS

2.4.1. Leak Location

Upon the termination of the test, the converter envelope was checked with a helium mass spectrometer for the source of the cesium leak. It was found that the leak occurred at the weld between
Fig. 18. Optimum converter output of LC-10 as a function of emitter temperature and current density at 7788 to 7816 hours.
Fig. 19. LC-10 emitter work function measurements at 7956 hour operating time
Fig. 20. LC-10 collector work function measurements at 7981 hour operating time
The stainless steel cooling line attached to the cesium reservoir was apparently too rigid. When the converter was brought to temperature, the stress generated caused the cesium lead tube to bend at the weld joint. Since there is no appreciable diffusion between copper and niobium, the joint opened up where the bending stress was the highest. Figure 21 shows the weld joint in both the unetched and the etched conditions. It can be seen that the leak occurred through intergranular cracks of the copper. It is possible that the grain boundaries may contain traces of oxygen impurities and cesium vapor corrodes these areas under stress to cause stress corrosion cracking. The life of the converter envelope could conceivably be prolonged if this joint were eliminated.

2.4.2. Work Function and Temperature Profile Measurements

After leak checking, the emitter was removed from the converter by cutting the joint between the tantalum transition piece of the emitter and the top niobium sleeve of the insulator with a jeweler's saw. The surface of the emitter was found to be as shiny as in the pre-test condition. The vacuum work function and the axial temperature profile of the emitter were then determined in the same apparatus as that used for the emitter in its pre-test state. The measurements were made both with a fuel slab facing the sight holes in the collector and with a tungsten web facing these sight holes.

The work function results are shown in Fig. 22, which fall in the range of 4.86 to 4.88 eV for average emitter temperatures 1859 to 1889°K. These results agree with that reported for the emitter prior to its incorporation into the converter (see Fig. 7) but are higher than that observed (4.74 eV) after the emitter was assembled into the collector cavity.
Fig. 21. Microstructures of the joint between the niobium cesium lead tube and the copper cesium reservoir of LC-10 after life-testing.
Fig. 22. LC-10 emitter vacuum work functions after life test. The work function and the average temperature of the emitter are indicated for each data point.
The results on axial temperature profiles are shown in Fig. 23 through Fig. 27. It can be seen from Fig. 23 that in the post-operational state, 29 more watts were needed to attain an average emitter temperature of 1859°K than that needed for maintaining an average emitter temperature of 1866°K in the pre-operational state. The axial temperature profiles determined before and after the life-testing, however, did not differ significantly. For the same power input (Fig. 24), the average emitter temperature is 50° lower in the post-operational state. There is no significant difference in the axial temperature profiles determined at various circumferential directions, both over the fuel slabs and over the tungsten webs (Figs. 25, 26 and 27). The higher power input needed (~10%) to maintain the required emitter temperature in the post-operational state may be associated with changes in the emittance of the emitter surface, or with the electron gun filament geometry.

2.4.3. Examination of Emitter Assembly

The emitter diameter was measured as a function of axial position at three circumferential locations. The change from the pre-test dimension was less than 0.0013 cm. The emitter was then sectioned across the middle of the fuel slabs in a direction perpendicular to its cylindrical axis for the examinations of the microstructures of the fuel, the cladding and the fuel-cladding interface. Figure 28(a) shows the microstructures of the duplex tungsten cladding over the carbide fuel slabs. No porosity was found in the fluoride tungsten and the bond between the fluoride tungsten and the chloride tungsten was excellent. Figure 29(b) shows the carbide fuel in unetched conditions and Fig. 28(c) shows the carbide fuel in etched conditions. Figure 28(d) shows the presence of a thin interaction layer (~0.0008 cm) at the fuel-cladding interface. Electron microprobe studies identified the
Fig. 23. Comparison of pre-operational (Fig. 4) and post-operational axial temperature profile of LC-10 emitter. Input power was adjusted during post-operational measurements to yield an average emitter temperature which was about the same as that in the pre-operational measurements. Sight holes facing edge of one of the fuel slabs.
Comparison of pre-operational (Fig. 4) and post-operational axial temperature profile of LC-10 emitter at equal input power. Sight holes facing edge of one of the fuel slabs.
Fig. 25. Post-operational axial temperature distribution of LC-10 emitter at three azimuthal positions, 120° from one another. Sight holes face fuel slab.
Fig. 26. Post-operational axial temperature distribution of LC-10 emitter at three azimuthal positions, 120° from one another. Sight holes face tungsten web.
Fig. 27. Comparison of axial temperature profile of LC-10 emitter over the fuel slabs with that over the tungsten webs at same power input (317 watts)
Fig. 28. Microstructures of fuel, cladding, and fuel-cladding interface of LC-10 emitter after life-testing
dispersions in the carbide fuel (Fig. 28c) and the fuel-cladding interaction layer (Fig. 28d) as the UWC$_2$ phase. No uranium was detected in the grain boundaries of the cladding (within a few tenths of one percent). Chemical analysis of the fuel material recovered from the fuel cavities yielded a C/U ratio of 1.03 and a tungsten concentration of 4.1 wt-%, which do not differ significantly from the pre-test composition (C/U = 1.04, tungsten = 4.2 wt-%).

2.4.4. Examination of Collector

Attempts to remove the collector structure from the heat sink were unsuccessful. The collector surface was then wiped with cotton Q-tips wetted with xylene. There was no loose deposit on the collector surface. The Q-tips were counted for C-14 with a liquid scintillator. The total amount of carbon deduced from the counting results was about $2 \times 10^{-9}$ grams. Analysis of the wipe for uranium yielded negative results; the sensitivity of the colorimetric method used was 0.05 micrograms. The collector was sawed into two halves under xylene with a jeweler's saw, which were then pushed out of the heat sink. Figure 29 shows the surfaces of these pieces, which were facing the emitter. It can be seen that these surfaces are smooth and free from any deposit and large grains of niobium are clearly visible. There is a general dark appearance and some of the grains appear darker than the others.

One of the collector pieces was examined for its microstructures. Figure 30(a) shows a cross section of this piece. The niobium exhibits large grains and clean grain boundaries. There appears to be a thin reaction layer on the surface facing the emitter. Figure 30(b) represents a magnified view of Fig. 30(a). The undulating nature of the surface is probably associated with the machining operation for the collector surface. The reaction layer is about a few micron thick. Radioactive counting indicated the presence of carbon and uranium on the surface. To gain a
Fig. 29. Appearance of collector surface of LC-10 after life-testing
(a) Thin reaction layer on surface facing the emitter

(b) Magnified view of (a)

Fig. 30. LC-10 collector after life-testing
quantitative idea of the amounts of fuel components near the surface of LC-10 collector, about 1 mil of material was milled off under xylene from the surface of the other half of the collector. The amounts of carbon, uranium and zirconium in the material recovered were determined by the combustion, radioactive counting and colorimetric techniques described in a previous report. The results yielded an average carbon flux of $1.2 \times 10^{-10}$ gram per cm$^2$ of fuel-cladding interfacial area (based on fuel O. D. surface area in contact with the cladding) per hour for the 8244 hours of operation. The corresponding uranium and zirconium fluxes were $4 \times 10^{-11}$ gram/cm$^2$/hr and $2.2 \times 10^{-12}$ gram/cm$^2$/hr, respectively. If it were assumed that a monolayer adsorbed on the collector surface consists of $5 \times 10^{14}$ atoms, then for the fuel component fluxes described facing the fuel slab with a monolayer of carbon, while about 5000 hours and 40,000 hours are needed to attain monolayers of uranium and zirconium, respectively. These estimates are made to show the relative magnitude of collector contamination by various fuel component transports through the cladding. Carbon seems to be the major source of contaminants.

2.4.5. Examination of Other Converter Components

Figure 31 shows the vanadium braze region between the tantalum transition and the tungsten emitter stem. Some defects were observed near the outer edge but the joint remained in good condition. No vanadium was detected in tantalum or tungsten but about 50% of tantalum and a few percent of tungsten were found in the vanadium braze.

Figure 32 shows a cross section of the interface between the Lucalox insulator and the niobium sleeve of the seal. The bond appears to be in excellent condition.
Fig. 31. Post operational appearance of the vanadium braze region between tantalum transition and tungsten emitter stem
Fig. 32. Post operational appearance of the interface between the Lucalox insulator and the niobium sleeve of LC-10 insulator seal
2.4.6. **Emitter Thermocouples**

A total of four emitter thermocouples were replaced during the LC-10 life test. These thermocouples were designated as C-148, C-150, C-152, and C-153. C-148 and C-150 were the initial emitter thermocouples and were replaced at an operating time of 3080 hours. Post-test calibration indicated that no appreciable degradation (6° at 1923°K) occurred in C-150 which was the reference thermocouple used for deducing the average emitter temperature. The post-test readings from C-148, however, showed significant deviation from its pre-test value. At a temperature of 1873°K, the post-test thermal emf of C-148 was about 1 mv lower than the pre-test value, which is equivalent to a temperature difference of 75°. However, since C-148 was not used as the reference thermocouple, its degradation should not affect the LC-10 life testing results.

C-152 and C-153 were replaced at a converter testing time of 7598 hours after 4518 hours of operation. Post-test calibration showed that at 1873°K, the temperature deduced from the thermal emf of each of these thermocouples was 30° lower than the true temperature. Part of the observed degradation could be due to experimental error involved in the calibration process and part could be caused by the handling of these thermocouples when they were removed from the thermocouple wells in the emitter cavity. The true degradation may be much less than the observed change.

2.5 **DISCUSSION**

At an average emitter temperature of 1873°K, LC-10 had an initial electrode power output of 5.94 W/cm² at a current density of 10.6 amp/cm², which decreased to 5.46 W/cm² in 1338 hours. For the same average emitter temperature and current density, LC-8 had an initial electrode power output of 5.5 W/cm² which increased to 5.8 W/cm² in about 100 hours and then decreased to 5 W/cm² in about 1000 hours. The maximum difference in electrode power output between LC-10 and LC-8 is thus less than ten percent. The failure to achieve
a larger gain in power output when the cladding was changed from fluoride tungsten of 4.5 eV vacuum work function to chloride-fluoride duplex tungsten cladding is probably caused by the degradation of the vacuum work function of the duplex tungsten cladding from 4.9 eV to 4.7 eV after the emitter was assembled into the converter. The decrease of the power output, however, appears to occur at a slower rate for LC-10 than for LC-8, presumably because the fuel component transport rates are slower through the chloride-fluoride duplex tungsten cladding than the fluoride tungsten cladding.

To assess the major cause for the performance changes observed during LC-10 life testing, pertinent test data at various operating times and major events occurred during the test are summarized in Table 2. Prior to 1338 hours, the decrease of power output is believed to be mainly due to the contamination of the collector surface by fuel components diffusing through the tungsten cladding, as shown by the observed increase of collector work function and electrode emittance. It appears that although the fuel component transport rates are reduced by the use of chloride–fluoride duplex tungsten cladding, the transport rates are still high enough to cause significant contamination of the collector surface in about 1000 hours. The decrease of converter power output after 1338 hours, however, is not associated with any electrode work function or emittance change; therefore, the decrease must be caused by other reasons. One of the most likely contributors to the observed decrease in converter power output is the change in electron gun filament position in the emitter cavity, which in turn causes a change in emitter temperature profile. Thus, the replacement of the electron gun filament at 3080 hours improved the converter output from 5.10 W/cm² to 5.36 W/cm² (see Table 2). As the replaced filament became aged, its position in the emitter cavity again shifted and the converter power output decreased again.
### TABLE 2
PERTINENT TEST DATA AT VARIOUS OPERATING TIMES AND MAJOR EVENTS DURING LC-10 LIFE TEST

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<td>5.31</td>
<td>5.10</td>
<td>5.36</td>
<td>5.28</td>
<td>5.20</td>
<td>5.00</td>
<td>5.10</td>
<td>5.22</td>
<td>4.80</td>
<td>4.62</td>
<td>5.27</td>
<td>5.31</td>
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<tr>
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<td>(1)</td>
<td>(2)</td>
<td>(3)</td>
<td>(4)</td>
<td>(5)</td>
<td>(6)</td>
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<td></td>
<td></td>
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</table>

(1) Life testing initiated. Initial performance mapping and diagnostic studies completed. \( P_R \) is power needed to maintain converter at reference temperatures (\( T_E \) average = 1873°K, \( T_C \) = 1006°K, \( T_R \) = 631°K) under open circuit conditions (\( \approx 397 \) watts initially). \( T_{E2} \) was the control thermocouple.

(2) Performance mapping and diagnostic studies at 1338 - 1446 hours. No change in \( T_E \), but \( T_C \) increased by 0.2 - 0.3 eV. \( P_R = 453 \) watts, an increase of 56 watts.

(3) Shift in electron gun filament position in emitter cavity noted. Converter shutdown at 3080 hours for replacement of electron gun filament and emitter thermocouples. Upon restart, power output returned to 1550 hour value. No change in electrode emittance. \( T_{E1} \) was the control thermocouple.

(4) Performance mapping and diagnostic studies at 5289 hours. No significant change in \( T_E \), \( T_C \) or electrode emittance was observed. \( P_R = 455 \) watts.

(5) Spot check at 6351 hours showed no significant change in \( T_C \) or electrode emittance. \( P_R = 463 \) watts.

(6) Converter shutdown due to power failure. Power output went up to 5.22 W/cm² upon restart.

(7) Converter shutdown for replacement of electron gun filament, emitter and collector thermocouples at 7598 hours. When converter was restarted, optimum cesium reservoir temperature increased from 625°K to 643°K. Performance mapping and diagnostic studies at 7788 hours. No significant change in \( T_E \), \( T_C \) or electrode emittance was observed.

(8) Test terminated because of cesium leakage through converter envelope.
The replacement of the electron gun filament at 7598 hours also brought an increase in converter power output. However, the optimum cesium reservoir temperature also increased by 18°. This increase in optimum cesium reservoir temperature could not be caused by electrode work function changes, since none was observed. The exact reason for this increase remains unknown. It is interesting to note that although the amounts of fuel components accumulated on the collector surface must have increased with time, there was no appreciable change in collector work function or electrode emittance after about 1000 hours. It appears that the effect of collector contamination on collector work function and emittance became saturated at early stage of the test, and subsequent arrival of more fuel components at the collector surface did not cause more changes in these surface properties.

The test data on LC-10 show that the use of a chloride-fluoride duplex tungsten cladding for carbide fuel did not lead to significant improvement in converter power output and thermionic performance stability. However, it must be pointed out that the emitter of LC-10 was probably contaminated by trace impurity so that its vacuum work function (4.7 eV) did not equal the 4.9 eV value measured in the early stages of emitter processing. The lack of significant improvement in initial power output is therefore understandable. The uncertainty in electron gun position in emitter cavity and thus the emitter temperature profile complicates the interpretation of the test data on thermionic performance stability. However, the analytical results on fuel component transport and the diagnostic data at 1338 hours point out that the chloride-fluoride duplex tungsten cladding did not completely rid the carbide fueled converter of fuel effect on converter performance. It would be of interest to check the
LC-10 test results using a similar converter containing an uncontaminated emitter. It would be of interest to find out whether substituting equiaxed tungsten for the fluoride tungsten substrate over the fuel would improve the thermionic performance stability of the carbide fueled converter. The next two parts of this report will deal with these cases.
3. FABRICATION AND LIFE-TESTING OF CARBIDE FUELED CONVERTER LC-11

3.1 TESTING OBJECTIVE

The testing results obtained on LC-10 described in the last section indicate that the transport rates of carbide fuel components through a chloride-fluoride duplex tungsten cladding were high enough to cause a decrease in converter output during long term operation. Since the transport rates of carbide fuel components through arc-cast tungsten were orders of magnitude lower than that through fluoride tungsten, the substitution of an arc-cast tungsten substrate for the fluoride tungsten substrate in the duplex tungsten cladding should help to improve the performance stability of a carbide fueled converter. The 90 UC-10 ZrC fueled converter LC-11 was therefore fabricated and life-tested in order to evaluate the effect of chloride-arc-cast duplex tungsten cladding on the power output and thermionic performance stability of carbide fueled converter.

The components and characteristics of LC-11 are essentially the same as for LC-10 except that the cladding consisted of 0.5 mm of chloride tungsten over a 0.75 mm thick substrate of arc-cast tungsten.

3.2 DESIGN AND FABRICATION PROCEDURES

The design and fabrication procedures for LC-10 and LC-11 were similar. The major differences were in the fueled emitter design and fabrication, which are described as follows.
3.2.1. **Emitter Configuration**

Figure 33 shows the configuration and dimensions for the LC-11 emitter. The emitting area is defined by the collector length rather than by the emitter length as in the case of LC-10. The emitting area, 17.98 cm$^2$, is slightly larger than that of LC-10 (16.43 cm$^2$). Although an arc-cast tungsten emitter body was used, the emitter stem still consisted of fluoride tungsten. This was because the arc-cast tungsten is more susceptible to grain growth which might cause embrittlement and cracking of the emitter stem upon thermal cycling, while the grain structure stability of fluoride tungsten at high temperatures is well established.

3.2.2. **Emitter Fabrication**

An arc-cast tungsten billet (19.05 cm diameter, 15.24 cm length) for the emitter blank was procured from Therm0 Electron Corporation (TECO). The major impurity contents were (in ppm): Cu, 10; Mo, 360; C, 10; O, 0.7; N, 1. To insure that the high molybdenum content would not cause an interaction with the carbide fuel, a compatibility test between the tungsten and the 90 UC 10 ZrC fuel material used in LC-11 was carried out at 1973 K for 100 hours. Metallographic and electron microprobe examinations failed to indicate any interaction. The TECO billet was therefore selected as the emitter blank material.

The fluoride tungsten stem was deposited onto the arc-cast tungsten blank by the hydrogen reduction of WF$_6$. After machining to the dimensions shown in Figure 33, the assembly was thermal cycled.
Note: Emitting area defined by collector length (3.505 cm) and emitter diameter (1.635 cm) at 1600°C and equals 17.98 cms.

1. Carbide fuel
2. Chloride tungsten cladding, 0.488 mm thick on side wall, 0.625 mm thick on bottom.
3. Cast tungsten billet; 0.75 mm thick cladding on fuel O.D.
4. Fluoride tungsten cup, 2.75 mm high, 0.25 mm side wall thickness, 0.5 mm bottom thickness.
5. Fluoride tungsten spacer, 1 mm thick, gap between spacer and cup 0.25 mm
6. Fluoride tungsten stem, 0.75 mm thick
7. Collector

Fig. 33. Configuration and dimension of LC-11 emitter
between 1873 K and room temperature for 20 times with no leak detected. Six fuel cavities in the emitter structure were formed by electrical-discharge-machining; their dimensions were the same as those in the LC-10 emitter. Major part of the material was removed at a rate of about 0.2 in$^3$/hr. The last 5 mils were removed at a much slower rate ($\sim$0.0001 in$^3$/hr) in order to minimize the formation of surface microcracks. Any shallow surface microcracks inside the fuel cavities were removed by rubbing with a tungsten slab smeared with 5 micron size diamond paste. The diamond powder left in the fuel cavities was rinsed off ultrasonically in acetone. Encapsulation of 90 UC-10 ZrC (C/U = 1.04, tungsten = 4 wt %) fuel slabs into the fuel cavities and deposition of the chloride tungsten emitting layer were carried out simultaneously by the hydrogen reduction of tungsten chloride under the same conditions as that used for LC-10 emitter.

Figure 34 shows the distribution of the $\langle 110 \rangle$ crystal axes in the chloride tungsten after the emitter was machined to dimension and electropolished. Vacuum work function measurements made in the cylindrical emitter apparatus yielded 4.92, 4.93 and 4.95 eV at 1/8 inch, 1/2 inch and 1 inch from the closed end of the emitter. Figure 35 shows the axial temperature profile of the emitter after brazing to the tantalum transition. Figure 36 represents the axial temperature profile of the emitter after brazing to tantalum transition and welding to the insulator seal. Emitter thermocouple $T_{E1}$ which was 59° higher then the average surface temperature of the emitter, was selected as the reference for deducing the average temperature of the surface of the emitter during converter operation. Figures 37, 38 and 39 show the average vacuum work function of the emitter surface at different stages of the emitter assembly and converter fabrication. The value obtained after the final bakeout was 4.85 eV which agreed within the limit of experimental error with the desired value of 4.9 eV.
Fig. 34. Distribution of the <110> axes in LC-11 emitter after electropolishing
**Fig. 35.** LC-11 emitter temperature profile after brazing to tantalum transition but prior to welding to insulator
\[ V = T = 1930^\circ K \]
\[ B = T = 1933^\circ K \]

Average Surface Temperature
\[ 1874^\circ K \]

Fig. 36. LC-11 emitter temperature profile after brazing to tantalum transition and welding to insulator but prior to converter assembly
Effective vacuum work function of LC-11 emitter determined in the apparatus for temperature profile measurement after brazing to tantalum transition. The numbers beside each point are the effective vacuum work function in eV and the average surface temperature in °K.
Fig. 38. Effective vacuum work function of LC-11 emitter determined in the apparatus for temperature profile measurement after brazing to tantalum transition and welding to insulator. The numbers beside each point are the effective vacuum work function in eV and the average surface temperature in °K.
Fig. 39. Vacuum work function (eV) of LC-11 emitter as a function of time during final bakeout at 1873°C.
3.3 TESTING RESULTS

Testing of LC-11 was initiated on July 16, 1970. During the period July 16 to July 30, 1970, efforts were devoted to the mapping of the converter performance, the measurements of the emitter and collector work functions and the establishment of the baseline conditions for the evaluation of any change of electrode emittance during the life testing of the converter. Figure 40 shows the initial optimum electrode power output as a function of current density and emitter temperature. Figure 41 contains the measured effective emitter work function superposed on a Rrasor-Warner plot. It can be seen that the base work function of the emitter lies between 4.5 to 5.0 eV. Figure 42 compares the effective work functions of the collector of LC-11 with that of the unfueled converter LC-9 which also contains a niobium collector. It can be seen that the agreement is good. The power input needed to maintain an average emitter temperature of 1873K, with collector at 938K and cesium reservoir at 631K under open circuit condition was 448 watts. These were the baseline parameters for the evaluation of change in electrode emittance during the life-testing of LC-11.
Fig. 40. Initial optimum power output of LC-11 as a function of current density at various emitter temperatures.
Fig. 41. Effective emitter work function of LC-11 for various $T_E/T_R$ ratios at 113 hours of operating time.
Fig. 42 Effective collector work functions of LC-11 for various $T_c/T_R$ ratios at 95 hour of operating time
Life testing of LC-11 began on July 30, 1973 at an average emitter temperature of 1873°K, a collector temperature of 1073°K, a cesium reservoir temperature of 625°K and a current density of 10.57 amperes/cm². The power output and the operating parameters as a function of testing time are shown in Figure 43. The important events concerning the performance of LC-11 throughout the test are described as follows.

The converter performance remained relatively stable during the first 5000 hours of operation. Starting from an electrode power output of 6.8 W/cm² at 500 hours, the electrode power output decreased to 6.53 W/cm² at 4903 hours, and the input power increased by 3 watts from 943 to 946 watts. The two emitter thermocouple readings which were only a few degrees apart at the initiation of the test, differed by 18° at 4903 hours. This indicates either a change in electron gun filament position in the emitter cavity or the degradation of the emitter thermocouples, or both.

The electron gun filament and the emitter thermocouples were replaced at an operating time of 5051 hours. Upon restart, the power input needed to maintain an average emitter temperature of 1873°K increased from 946 watts before the shutdown to 975 watts. There was no significant change in the optimum cesium reservoir temperature and the electrode power output returned to the initial value of 6.8 W/cm². This implies that there were no significant electrode work function changes during the first 5000 hours of operation. The power input needed to maintain the initial temperatures of the emitter, the collector and the cesium
Fig. 43. Power output and operating parameters of LC-11 (Sheet 1 of 2)
Fig. 43. Power output and operating parameters of LC-11 (Sheet 2 of 2)
reservoir under open circuit condition increased from 448 watts to 486 watts at the restart of the converter at 5051 hours. This indicates an increase in electrode emittance, presumably due to contamination of the collector surface by carbide fuel components diffusing through the emitter cladding.

The test was continued from 5051 hours until the electrode power output decreased from 6.8 W/cm\(^2\) to 6.50 W/cm\(^2\) at 10,143 hours, with the power input staying at 970 ± 5 watts and the two emitter thermocouple readings differing by 2° to 13° during this entire testing period. On November 1, 1971, at an operating time of 10599 hours, however, it was found that the input power had to be reduced from 970 watts to 955 watts in order to maintain the emitter at an average temperature of 1873°K. This decrease in power input was accompanied by a drop in electrode power output from 6.50 W/cm\(^2\) to 6.42 W/cm\(^2\). It appeared that the electron gun filament had developed a hot spot. The converter was shut down on November 18, 1971 at 10599 hours for the replacement of the electron gun filament, both emitter thermocouples and two collector thermocouples which had opened up. Electrode emittance measurement made after the converter was restarted on November 30, 1971 showed that the change was small during the 5051 to 10,662 hours testing period; the power input needed to maintain the converter at reference temperatures under open circuit condition increased only from 486 watts to 490 watts. There was no significant change in the optimum cesium reservoir temperature. Thus the observed decrease of electrode power output from 6.8 W/cm\(^2\) at 5051 hours to 6.50 W/cm\(^2\) at 10,143 hours was probably due to an increase in the collector work function when more fuel components were condensed on the collector surface.

Performance data taken on December 2, 1971 at 10,662 hours indicated that the input power needed for maintaining an average emitter temperature of 1873°K was 947 watts, and the electrode power output was 6.40 W/cm\(^2\). These values were about the same as that before the
replacement of the electron gun filament and the emitter thermocouples. The power output, however, dropped to $6.22 \text{ W/cm}^2$ at 12,603 hours, and the input power needed for maintaining an average emitter temperature at $1873^\circ\text{K}$ increased to 959 watts. The difference in the readings of the two thermocouples remained at about $28^\circ-29^\circ$ during the entire period from 11,307 hours to 12,603 hours, as compared to a normal value of less than a few degrees. It appeared that either the replacement of the electron gun filament at 10,622 hour did not improve the situation, or the replaced emitter thermocouples were defective. A decision was therefore reached at 13084 hours to shut down the converter for inspecting the electron gun filament and the emitter thermocouples. It was found that there was no visible distortion of the electron gun filament. The filament was therefore not replaced but the two emitter thermocouples were replaced. Upon restart, the two emitter thermocouple readings were within 1$^\circ$ of each other and the electrode power output was $6.22 \text{ W/cm}^2$, which was the same as that before the replacement of the two emitter thermocouples. The power input needed for maintaining an average emitter temperature of $1873^\circ\text{K}$ was 973 watts which was about the same as that at 10,143 hours. Since the electrode power output, $6.22 \text{ W/cm}^2$, was lower than that at 10,143 hours ($6.5 \text{ W/cm}^2$), it was concluded that there was further increase in the cesiated collector work function since 10,143 hours. Electrode emittance measurement also indicated an increase of power input from 490 watts at 10,622 hours to 498 watts at 13,089 hours for maintaining the converter at reference temperatures under open circuit condition. Both the reduced electrode power output and the increased electrode emittance were probably caused by further condensation of fuel components on the collector surface.
The electrode power output subsequently decreased to 6.16 W/cm² at 14,592 hours while the power input required for maintaining the average emitter temperature at 1873°K increased from 973 watts to 989 watts, with the readings of the two emitter thermocouples within a few degrees from each other. Measurements taken after an involuntary shutdown at 15,116 hours showed significant changes in the input power (from 989 watts at 14,592 hours to 971 watts at 15,112 hours) and output power (from 6.16 W/cm² at 14,592 hours to 5.95 W/cm² at 15,112 hours). In addition, the deviation of the two emitter thermocouple readings increased from 2° at 14,592 hours to 16° at 15,112 hours. The shutdown probably changed the configuration of the electron gun filament in the emitter cavity and thus increased the non-uniformity in emitter surface temperature and lowered the power output. With the approval of the NASA Project Manager, the converter was shutdown at 15,112 hours for the replacement of the electron gun filament and both emitter thermocouples.

Following start-up, at 15,114 hours, the electrode power output was 6.41 W/cm² versus 5.95 W/cm² before the shutdown and the power input required to maintain the average emitter temperature at 1873°K was 1001 watts. During subsequent testing to 15,543 hours, the input power had to be increased gradually from 1001 watts to 1013 watts in order to maintain the average emitter temperature at 1873°K. It was suspected that the thermocouples were defective. The input power was maintained at 1013 watts from 15,543 hours to 15,784 hours. The average emitter temperature derived from the thermocouple readings, decreased gradually from 1873°K to
1856°K, with the output power remaining constant at 6.37 W/cm². Since both the input and the output power remained constant, the observed change in emitter temperature was probably due to emitter thermocouple degradation. At a derived average emitter temperature of 1873°K, 1026 watts of input power was needed at 15,784 hours and the electrode power density was 6.73 W/cm². To prevent the emitter from getting excessively hot because of defective thermocouples, the input power was kept at 1000 watts which was the power input at the startup of the converter at 15,114 hours. The derived average emitter temperature dropped to 1854°K and the electrode power output decreased to 6.36 W/cm². Since there was uncertainty in emitter temperature, the converter was shutdown at 16,145 hours for the replacement of the electron gun filament and the emitter thermocouples.

After the converter was restarted, electrode emittance data indicated that 511 watts of power input was required to maintain the converter at reference temperatures under open circuit conditions, which was 13 watts more than that at 13,089 hours after the replacement of emitter thermocouples. The electrode power output at 16,301 hours was 6.11 W/cm² and the input power needed for maintaining an average emitter temperature of 1873°K was 986 watts. Both the input power and the output power remained relatively constant until 17,772 hours when the input power was 991 watts and the electrode power output was 6.21 W/cm². The difference between the two emitter thermocouple readings was about 22° during this testing period. Degradation of emitter thermocouples was evident at 18,468 hours, since with about the same power input (991 watts), the two emitter thermocouple readings were lower by 18° and 13° respectively than that at 16,301 hours but the power output (6.13 W/cm²) was about the same. Such emitter thermocouple degradation continued until at 18,632 hours, when the thermionic program was terminated, the two emitter thermocouple readings were
23° and 19° lower than that at 16,301 hours. No significant
difference was observed in power input (986 watts at 16,301 hours and
993 watts at 18,632 hours) and electrode power output (6.11 W/cm²
at 16,301 hours and 6.16 W/cm² at 18,632 hours). It appeared that
no significant change in converter performance occurred between
16,301 hours and 18,632 hours.

Post-test calibration was carried out on the emitter thermocouples
replaced at 5051 hours (C-149 and C-151), 10,622 hours (C-154, C-155),
13084 hours (C-161, C-162), 15,112 hours (C-163, C-164) and 16,145 hours
(C-167, C-168). In the operating temperature range for the emitter
all of these thermocouples except C-167 and C-168 were found to read
between 15° to 30° lower than the true temperature. Part of these
discrepancies could be due to experimental error involved in the
calibration operation and part could be caused by handling when
these thermocouples were removed from the thermocouple wells in the
emitter structure. C-167 and C-168, however, read 75° and 80° lower
than the true temperature. This is consistent with the observation that
much higher input power was needed to maintain the desired emitter
temperature after these thermocouples were installed in the emitter
structure after the shutdown at 15,112 hours.

3.4 DISCUSSION

Even though the interpretation of the test data of LC-11 is
complicated by the change in electron gun filament configuration in
the emitter cavity and the degradation of emitter thermocouples,
some insights on the change of converter performance with time can be
gained by examining test data obtained immediately after the electron gun
filament and (or) the emitter thermocouples were replaced, provided
there were no obvious irregularities in the test data after these
replacements were made. Test data selected on the basis of these
criteria are listed in Table 3.
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(1) Initiation of life test at 237 hours.
(2) Replacement of electron gun filament and emitter thermocouples.
(3) Replacement of electron gun filament, emitter thermocouples and collector thermocouples.
(4) Replacement of emitter thermocouples.
(5) Replacement of electron gun filament and emitter thermocouples.
(6) Termination of test.
The data shown in Table 3 indicate that the electrode power output of LC-11 decreased from 6.8 W/cm\(^2\) to about 6.1 W/cm\(^2\) in 18,632 hours, representing a loss of thermionic performance of about 10%. At the same time, the input power needed to maintain an average emitter temperature of 1873\(^o\)K increased about 50 watts which is equivalent to about 5% of the power input. Both the thermal performance and the thermionic performance degradations are believed to be due to the contamination of the collector surface by fuel components diffusing through the emitter cladding. This is borne out by the electrode emittance data shown in Table 3. Compared with the test data of LC-10 (see Table 2), LC-11 has a higher initial thermionic performance (6.80 W/cm\(^2\) versus 5.92 W/cm\(^2\)) and a slower rate of thermionic performance degradation (10% in 18,632 hours versus 10% in 3233 hours). The higher initial thermionic performance results from the higher vacuum work function of the LC-11 emitter (4.85 eV versus 4.75 eV). The slower rate of thermionic performance degradation is probably due to the lower fuel transport rates through chloride-arc-cast duplex tungsten cladding than chloride-fluoride tungsten cladding. Although the advantage of chloride-arc-cast duplex tungsten over chloride-fluoride duplex tungsten as cladding material in carbide-fueled converters was demonstrated, some thermionic performance degradation still exists. Nevertheless, the 10% drop in electrode power output in 18,632 hours does not appear to be intolerable and should be taken into account in the design of thermionic fuel element for long term application.
4. FABRICATION AND PERFORMANCE MAPPING
OF CARBIDE FUELED CONVERTER LC-12

4.1 TESTING OBJECTIVE

In Section 2 of this report, the carbide fueled converter LC-10 was fabricated and tested in order to determine the electrical power output and thermionic performance stability of a converter containing a conventional duplex tungsten emitter. Since the insitu vacuum work function of the emitter of LC-10 was lower than desired (4.7 eV versus 4.9 eV), it was decided to repeat the study with another carbide fueled converter, designated as LC-12, the emitter work function of which has the reference design value of 4.9 eV. The termination of the U. S. thermionic program interrupted the life-test. Special design and fabrication features, initial performance mapping results and test data for its 313 hours of operation are described under this section.

4.2 DESIGN AND FABRICATION PROCEDURES

LC-12 had the same converter design and chloride-fluoride duplex emitter configuration as LC-10 except the following:

1. The 90UC-10ZrC had a C/U of 1.015 instead of 1.04. It was believed that a lower C/U should lower the carbon transport rate through the cladding and thus should improve the thermionic performance stability of the converter.

2. A W-Y2O3 metallized, Nb-V brazed high temperature insulator seal was used instead of the Litton insulator seal.
3. The joint between the top of the collector and the bottom of the lower insulator sleeve (the final assembly of the emitter and the collector) was made by electron beam welding rather than Cu-Ti brazing. The Cu-Ti brazed joint is easily contaminated and becomes embrittled. In addition, the spraying of the braze components during the brazing operation may contribute to the contamination of the emitter and collector surfaces.

4. The Nb-Cu weld joint in the cesium reservoir lead tube was eliminated by using a niobium reservoir instead of a copper reservoir, since the Nb-Cu joint failure led to the termination of LC-10.

The fabrication procedures for LC-12 were similar to that for LC-10 except for items (3) and (4) listed above. Characterization of the emitter with respect to the degree of (110) preferred orientation, temperature profile and vacuum work function was carried out in the same way as that used for the LC-10 and LC-11 emitters.

Figure 44 shows the distribution of the <110> crystal axis in the chloride tungsten layer of LC-12 emitter. Vacuum work function measurements made in the cylindrical emitter apparatus yielded 4.90, 4.92 and 4.93 eV at 1/8, 1/2, and 1 inch from the closed end of the emitter. Figure 45 represents the temperature profile of the emitter after it was vanadium brazed to the tantalum transition and the tantalum transition was electron beam welded to the insulator seal. The emitter surface temperature readings read with a calibrated optical pyrometer at three circumferential positions, were in good agreement. The two emitter thermocouple readings also agreed within a few degrees.
Fig. 44. Distribution of the <110> axes in LC-12 emitter after electropolishing.
Fig. 45. LC-12 emitter temperature profile after brazing to tantalum transition and welding to insulator seal
The average emitter surface temperature read with the pyrometer was 9° lower than the temperature given by emitter thermocouple $T_E$. In the apparatus used for the determination of the emitter temperature profile, the emitter yielded an average vacuum work function of 4.88 eV over the entire emitting surface, which was stable for the 100 hour measurement period (Fig. 46). After the assembled converter was installed in the bakeout station with the emitter at an average temperature of 1873°K, the vacuum work function observed was 4.87 eV which remained at the same value for 61 hours. This agreed within experimental error with the desired value of 4.90 eV.

4.3 TESTING RESULTS

Test of LC-12 was initiated on January 9, 1973. Electrode emittance data were taken after the emitter was brought to an average temperature of 1873°K. It was found that under open circuit condition a power input of 390 watts was needed to maintain the emitter at 1873°K, the collector at 938°K and the cesium reservoir at 631°K. These were taken as the reference conditions for the future electrode emittance measurements. At the same current density (10.6 amperes/cm$^2$) as that used for LC-10 and an emitter temperature of 1873°K, the optimum cesium reservoir temperature was 623°K and the optimum collector temperature was 1073°K. These were very close to the operating conditions for LC-10. At an operating time of 7 hours under these conditions, the electrode power output was 6.70 W/cm$^2$ and the input power refined was 817 watts. The electrode power output, however, dropped to 6.06 W/cm$^2$ at an operating time of 22 hours under the same operating conditions. Performance mapping and electrode work function measurements were initiated on January 10, 1973 at an operating time of 29 hours. The performance mapping results are shown in Fig. 47 together with similar data for LC-10 and LC-11. It can be seen that for current densities below 6 amperes/cm$^2$, the differences in the performances of these converters are insignificant. At higher current densities, however, both LC-11 and LC-12 perform better than LC-10. This is consistent with the higher vacuum functions of LC-11 and LC-12 emitters.
Fig. 46. Effective vacuum work function of LC-12 emitter. The work function and the temperature are indicated beside each data point.
Fig. 47. Initial optimum power output of LC-12 as a function of current density at various emitter temperatures. Similar data for LC-10 and LC-11 are included for comparison.
Figure 48 shows the effective emitter work functions at various $\frac{T_E}{T_R}$ values superposed on Rasor-Warner plots. The results obtained are not significantly different from that obtained for LC-10 (Fig. 10) and LC-11 (Fig. 41). This further indicates that such measurements are probably useful only for indicating any change of work function rather than the absolute value. Figure 49 shows the effective collector work functions at various $\frac{T_C}{T_R}$ values. Compared with that for LC-11 (Fig. 42), the data points for LC-12 lie slightly higher on the plot. This may be the reason why the thermionic performance of LC-12 is slightly lower than that for LC-11 (see Fig. 47). The exact reason for this difference in collector work function is unknown. One possible explanation may be that this was caused by traces of fuel components diffusing through the chloride-fluoride duplex tungsten cladding which has higher transport rates for the fuel components than the chloride-arc-cast duplex tungsten cladding.

After the completion of the initial performance mapping and electrode work function measurements, test of LC-12 was continued at an emitter temperature of 1873°K, a collector temperature of 1073°K, a cesium reservoir temperature of 623°K and a current density of 10.6 amperes/cm² from 215 hours to 313 hours. The electrode power output stayed between 5.79 and 5.88 W/cm² and the power input needed for maintaining the above described operating conditions was between 811 and 820 watts. The power output appeared to have decreased appreciably from the initial value of 6.20 W/cm², but the power input needed stayed essentially the same. The test was terminated at an operating time of 313 hours. The test results and operating parameters are summarized in Fig. 50.

4.4 DISCUSSION

The test of LC-12 was not carried out for a long enough period of time to demonstrate its performance stability. The test, however, did indicate that the thermionic performance of a converter containing a high work function chloride tungsten
Fig. 48. Effective emitter work functions by LC-12 for various $T_E/T_R$ ratios at 160 hours of operating time.
Fig. 49. Effective collector work functions of LC-12 for various $T_C/T_R$ ratios at 190 hours of operating time.
Fig. 50. Power output and operating parameters of LC-12
Fig. 50. Power output and operating parameters of LC-12
emitting layer on a fluoride tungsten substrate was inferior to a converter with a similar emitting surface but an arc-cast tungsten substrate. For instance at an operating time of 313 hours, LC-12 delivered an optimum electrode output of 5.81 W/cm$^2$ at an emitter temperature of 1873°K, a collector temperature of 1073°K and a current density of 10.6 ampere/cm$^2$. Under the same operating conditions, LC-11 delivered optimum electrode outputs of 6.8 W/cm$^2$ at an operating time of 500 hours and 6.16 W/cm$^2$ at an operating time of 18,632 hours. Since the thermionic performance of LC-12 is likely to be lower at 18,632 hours of operation than the initial value, the previously demonstrated superiority of the chloride-arc-cast duplex tungsten over the chloride-fluoride duplex tungsten as the cladding material for carbide fueled converter is reinforced.

It must be pointed out, however, that the above discussions were based on the thermionic performance of the fueled emitter. For the operation of a thermionic fuel element under neutron environment, other properties of the cladding material, such as structural integrity and creep resistance to fuel swelling under irradiation, may also be of importance. In-pile evaluation of a converter containing a carbide fueled chloride-arc-cast duplex tungsten emitter should be of great interest in the development of thermionic fuel element of improved life and performance.
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