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GENERAL ATOMIC

DIVISION OF GENERAL DYNAMICS

GA-4511

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HIGH-TEMPERATURE VAPOR-FILLED THERMIONIC CONVERTER

QUARTERLY TECHNICAL PROGRESS REPORT
FOR THE PERIOD ENDING JULY 31, 1963

Contract AF 33(657)-8563
Project No. 8173, Task No. 817305-5

Aeronautical Systems Division
Air Force Systems Command
U.S. Air Force
Wright-Patterson Air Force Base, Ohio

August 26, 1963

CATALOGED BY DDC

GENERAL ATOMIC
DIVISION OF
GENERAL DYNAMICS

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Wright-Patterson Air Force Base, Ohio

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The work covered by this report was accomplished under Air Force Contract AF 33(657)-8563, but this report is being published and distributed prior to Air Force review. The publication of this report, therefore, does not constitute approval by the Air Force of the findings or conclusions contained herein. It is published for the exchange and stimulation of ideas.

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FOREWARD

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INTRODUCTION AND SUMMARY

This report covers the quarterly period ending July 31, 1963. The program was initiated to evaluate materials, fabrication and testing problems associated with the operation of high-temperature vapor-filled thermionic converters employing uranium-zirconium carbide emitters of cylindrical geometry. To date five of these converters have operated a total of 5300 hr. The last converter to be tested in this program is Cell K, which will soon be operable.

During the previous reporting period, Cells G and J were operated until loss of cesium caused discontinuation of the test (Ref. 1). In this reporting period, Cell H, which had two cesium vials, was placed in operation, while final assembly of Cell K was completed. Cell H output was 56 watts at an emitter temperature of 2380°K . The cesium temperature optimum for maximum power was 637°K . A collector temperature optimum was not obtained due to cooling limitations.

After 530 hr of Cell H operation at an average power of 20 watts, and at an emitter temperature of about 2220°K , a 50% power degradation was observed. At 608 hr the bombardment filament was replaced and the second cesium vial was opened. Subsequent operation yielded a power output of 48 watts at 2270°K . This performance was basically the same as that observed during the initial operation. It is concluded that the degradation of this cell at 608 hr was primarily due to loss of cesium through the leaks in the cell envelope. At 700 operating hours the test was concluded due to the development of an interelectrode short.

Vacuum emission data obtained with a test collector gave current values a factor of 8 greater than the initial data at a corresponding emitter

temperature. The Cell H emitter appeared to be essentially unaltered by the test except for slight dimensional changes. In the emitter center portion, the diameter had decreased from 0.674 to 0.654. A deposit was found in the opposing area of the collector cavity, which reduced its diameter from 0.698 to 0.680. Chemical analysis of the emitter showed that the uranium content had decreased from 20% to 15%. A recalibration of the emitter thermocouples against emitter surface pyrometer readings indicated that they had maintained their calibration. Metallographical examination of the emitter thermocouples showed a heavy oxide formation on the tantalum sheath and a considerable loss of the beryllium oxide insulating material.

Post-test analysis on the collector deposits of Cells G and J was obtained during this report period. The analysis showed a 1% uranium and 90% zirconium content. The deposit in the Cell H collector, however, had 80% uranium and only 17% zirconium. An intermetallic reaction had taken place in these collectors between the deposit and the nickel substrate which extended deep into the nickel.

Preliminary tests on Cell K are completed and cell operation is anticipated in the very near future. Vacuum emission data is in good agreement with previous cells. An emitter temperature profile study gave 100°K variations over the emitter surface.

GENERAL

OPERATION AND POST-TEST ANALYSIS OF CELL H

Cell H was completely assembled during the previous quarter and held pending completion of tests of Cells G and J. To assure that the cell had remained leak-tight and that the emitter had not become contaminated, another vacuum emission check was conducted on Cell H. The data were in good agreement with those obtained a few months earlier (Fig. 1).

The physical dimension and construction of Cell H is the same as Cells F, G, and J in every respect except that Cell H has two cesium vials. Final power degradation of all previous cells had been traced to the loss of cesium through cracks in the cell envelope. It remains to be shown that the introduction of an additional amount of cesium will restore the power output of the cell.

One of the two cesium vials was broken and the cell was placed in operation. The effect of cesium temperature on cell output was investigated at the emitter temperatures of 2000°K and 2200°K (Fig. 2). As in previous cells, a peak was observed, although it was shifted in this case from 620°K to $637 \pm 5^{\circ}\text{K}$. The peak occurred at essentially the same cesium temperature for the two emitter temperatures.

A large coil cooled with Dowherm oil was attached to the collector of Cell H because the post-test analysis of Cells G and F indicated that the collectors had been operated at excessively high temperatures. Fig. 3 shows cell power output as a function of emitter temperature with and without collector cooling. At an emitter temperature of 2200°K , there is an increase in power output of about 20% when the collector temperature is decreased by 120°K . The improved collector cooling coil, however, was still inadequate to obtain substantially lower collector temperatures, since the collector temperature could only be investigated over the limited range of about 120°K .

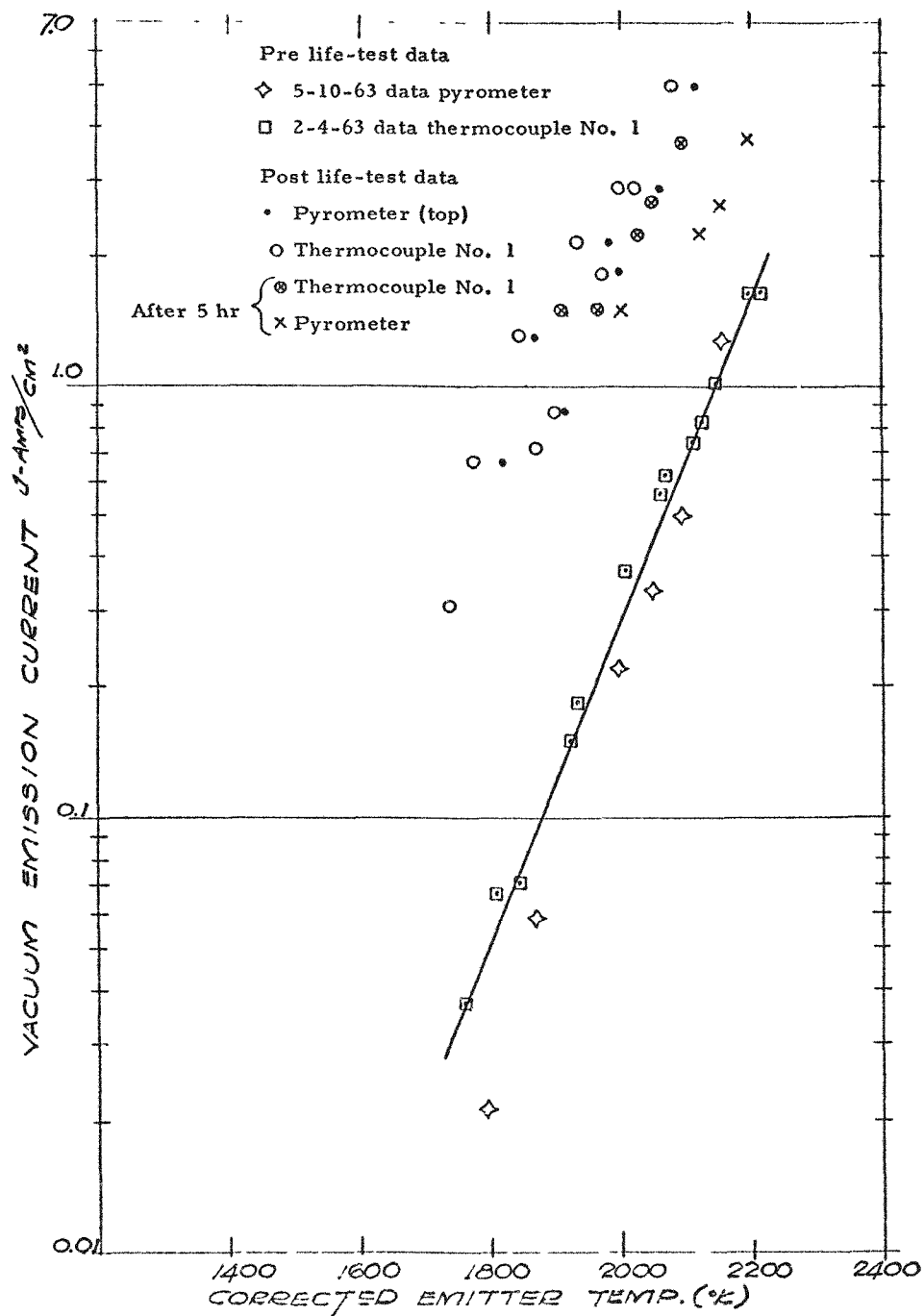


Fig. 1--Vacuum emission of Cell H,
emitter No. 40

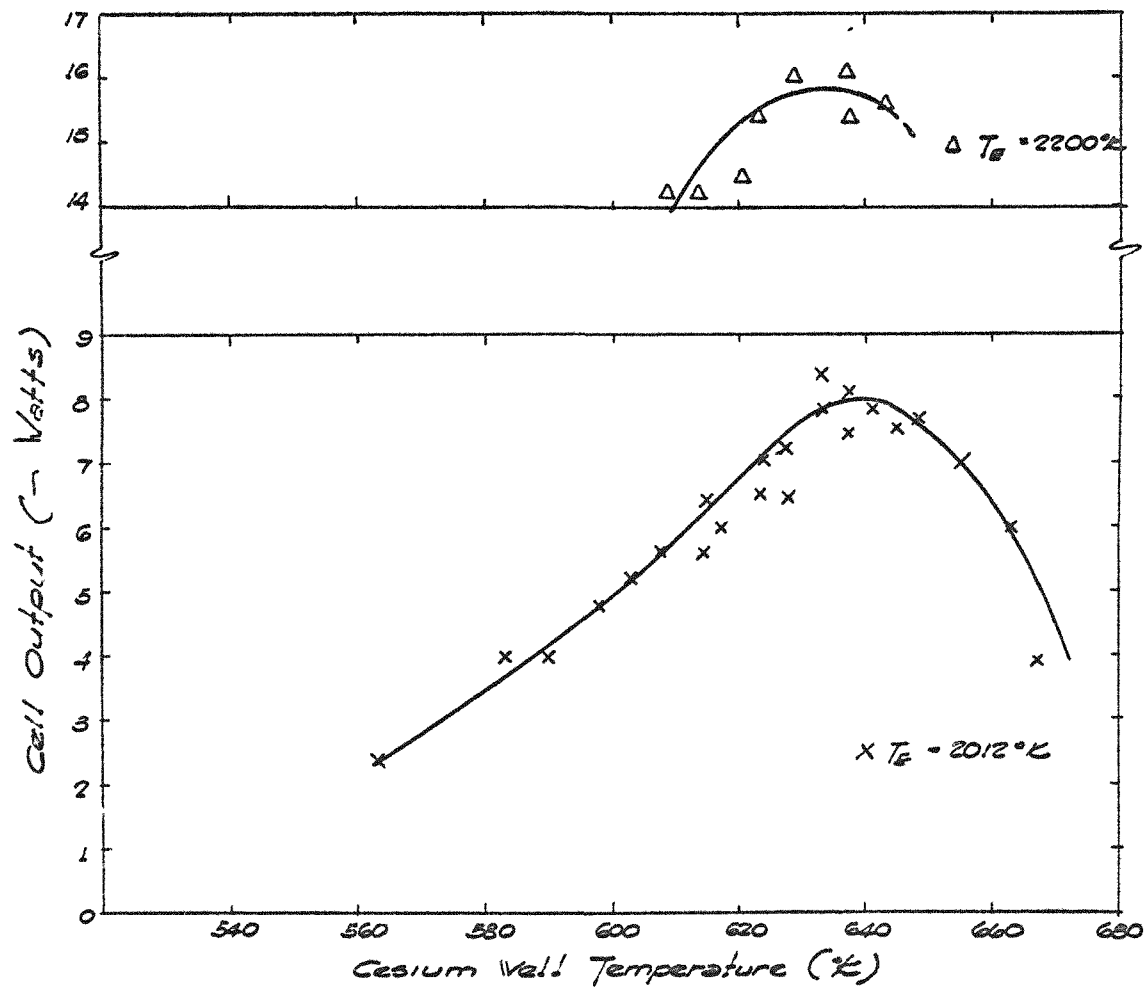


Fig. 2--Effect of cesium temperature on output of Cell H

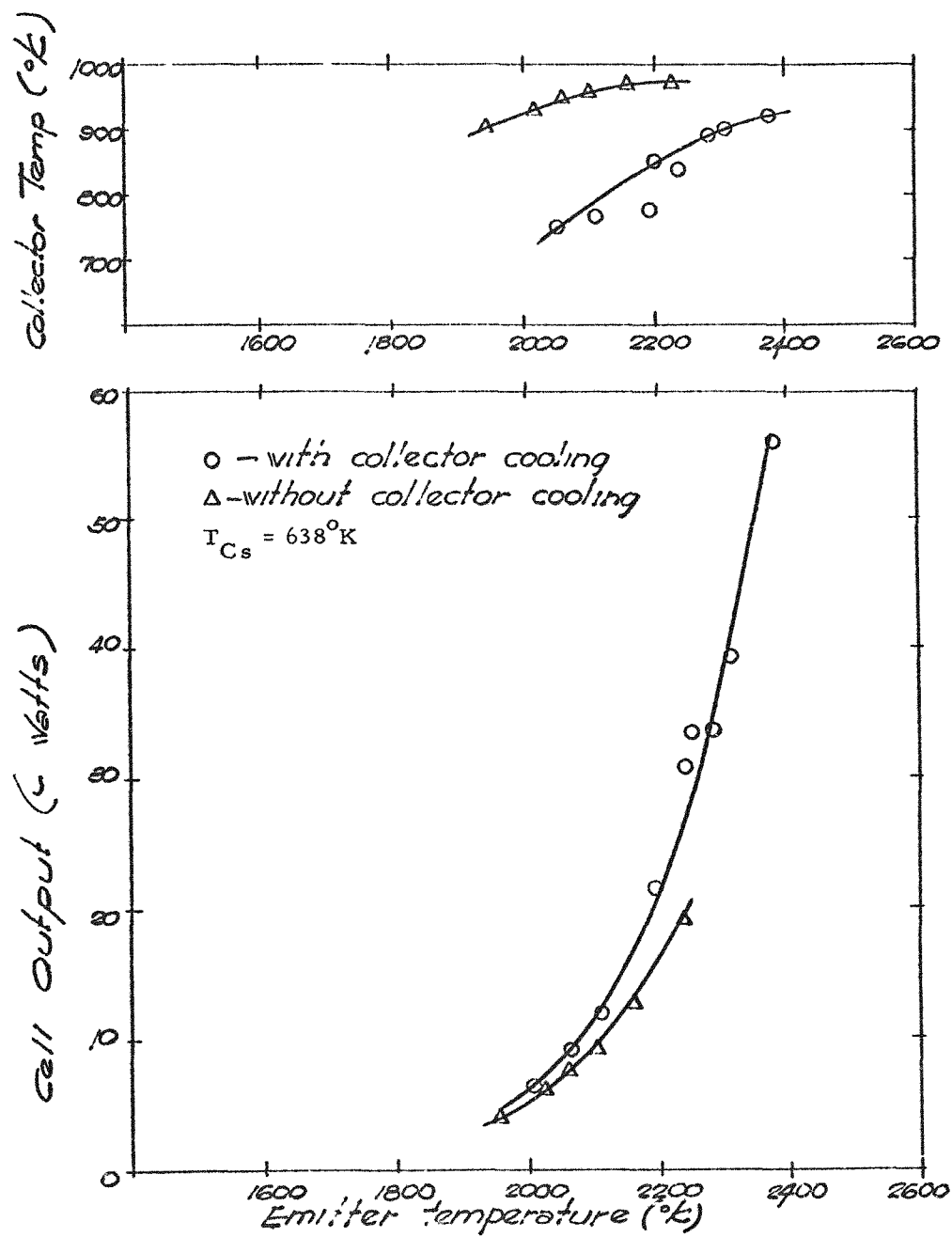


Fig. 3--Effect of collector cooling on power output and collector temperature of Cell H at various emitter temperatures

The output from Cell H was lower by a factor of 3 as compared to Cell G at equivalent conditions. It is noteworthy that the vacuum emission current of Cell H was also lower by a factor of 3 than that of Cell G. The maximum output recorded on Cell H was 56 watts; it was obtained at an emitter temperature of 2380°K , a collector temperature of 1023°K , and a cesium well temperature of 638°K (Fig. 3). In Fig. 4 the short-circuit current density is plotted as a function of emitter temperature at a cesium temperature of 638°K . The maximum current density of 16.5 amp/cm^2 was recorded at an emitter temperature of 2380°K . While these currents were about a factor of 8 higher than the vacuum emission currents, the slope of the two curves (Figs. 1 and 4) are the same.

After the characteristics of the cell had been established, operation at a continuous nominal cell output of 20 watts was begun. The emitter temperature varied between 2100°K and 2270°K . The cesium temperature was maintained between 628°K and 638°K . During the first 530 hr of operation, only a slight power degradation was observed. In the following 78 hr this degradation became considerable.

Increasing the cesium well temperature at this time to 700°K did not change the cell output, as normally observed. This indicated, as it did in previous cells, that the liquid phase of cesium had disappeared and total depletion of cesium might be anticipated soon. Condensation of a cesium compound was noted on the bell jar inner surface also indicating a cell leak.

The I-V sweep pictures taken over the period of the next few days showed shapes observed in other cells when power degradation had occurred. Also, in this case, the ion current became negligible and the short-circuit current decreased by a factor of 2. The cell output dropped to 12 watts at a 2230°K emitter temperature (Fig. 5). After a total of 608 hr, the filament burned out and was replaced. After replacing the filament, the second cesium vial was broken.

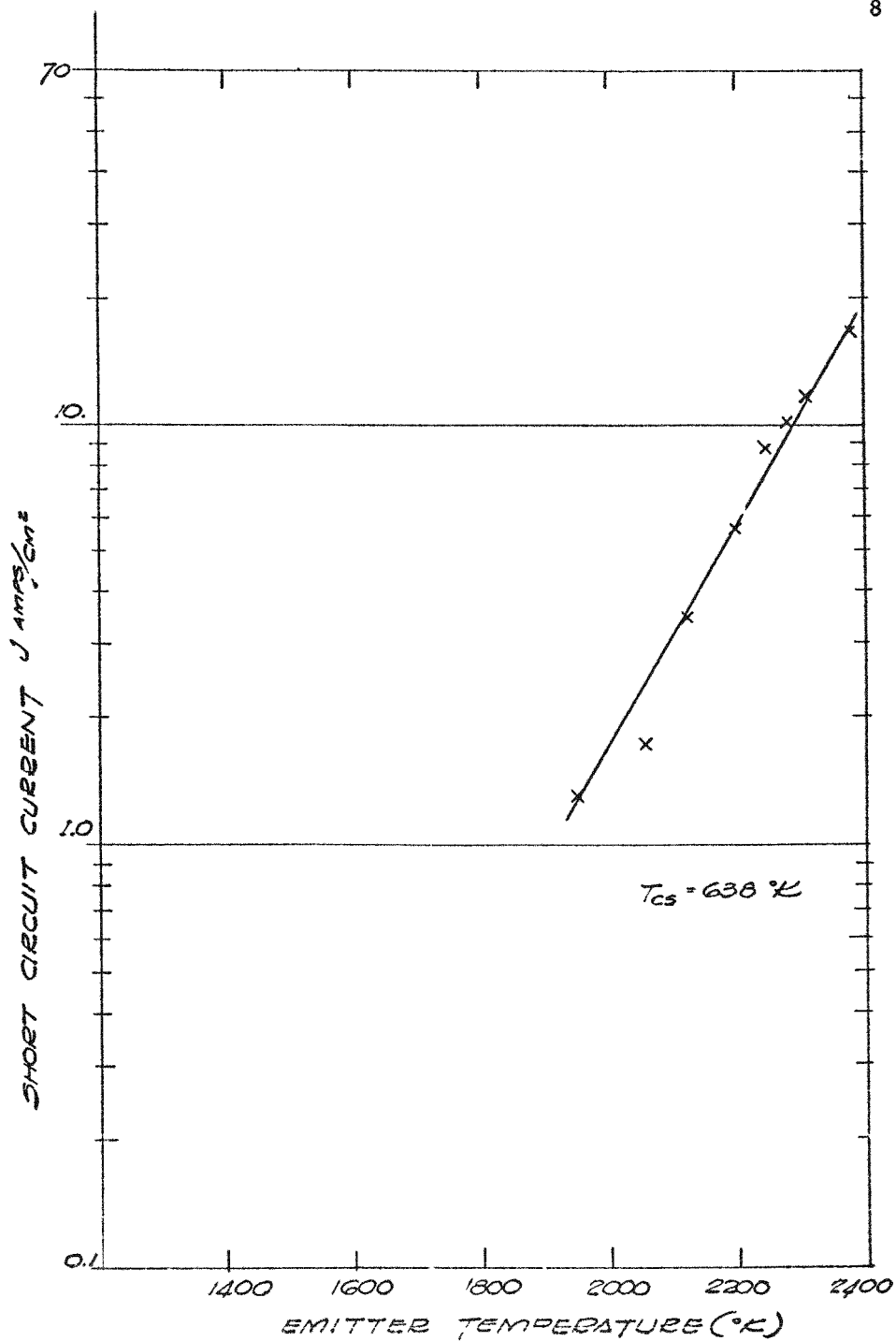


Fig. 4--Short-circuit current of Cell H

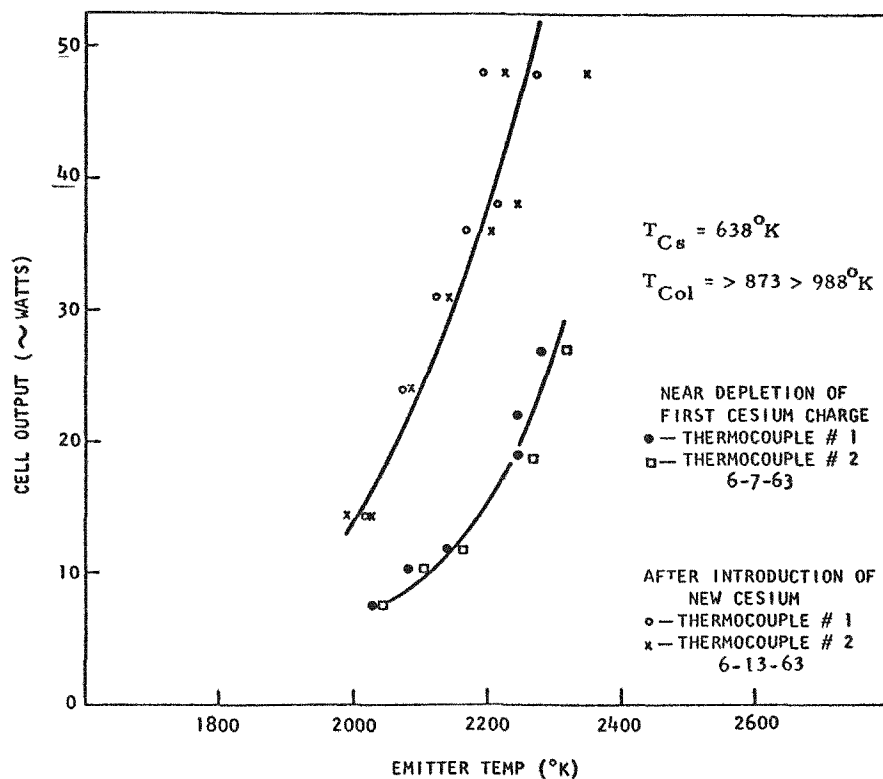


Fig. 5--Output of Cell H versus temperature for operation with first and second charge of cesium

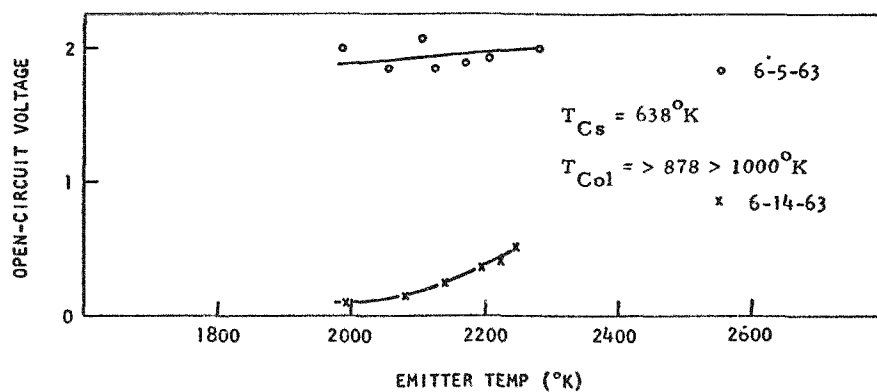


Fig. 6--Open-circuit voltage of Cell H versus emitter temperature with normal and degraded operating conditions

An initial power output of 48 watts was observed upon restart up at an emitter temperature of 2270°K , a collector temperature of 988°K , and a cesium well temperature of 638°K . A study of cell output versus emitter temperature showed that the performance had improved slightly over the previous data shown in Fig. 5. Thus, it was shown that the degradation observed during the last few days of operation just prior to filament failure was due to the depletion of cesium and not a deterioration of the carbide emitter.

Within 24 hr after breaking the second cesium vial, another sharp degradation was observed. The cell voltage was abnormally low. An open-circuit voltage study showed that the cell voltage was down by a factor of 4 for corresponding emitter temperatures (Fig. 6). A partial internal short existed between the emitter and collector. Because this short would not open, after several days, cell operation was discontinued. A total of 700 operating hours were accumulated on this cell.

An inspection revealed a large leak in the lower weld between the copper base and the Monel ring of the insulator. The insulator itself did not leak.

The emitter appeared to be in good condition. The bottom edge of the emitter flared out to a diameter of 0.685 and 0.690 in. In other areas the emitter diameter varied from a low of 0.654 in. near the center portion to 0.670 in. at the top. The original dimension was 0.674 ± 0.001 in. There was little noticeable deterioration and the emitter top appeared to be in excellent condition. An alignment check showed the emitter to be canted, resulting in one side being off by 0.012 in.

The collector cavity had an uneven deposit on all surfaces. The hole diameter varied between 0.680 and 0.693 in. The original dimension was 0.698 ± 0.001 in. The contact between collector and emitter probably occurred in the area where the emitter had flared out at the bottom edge,

or perhaps at the top due to the emitter inclination with respect to the collector cavity.

A vacuum emission study was conducted on the emitter after the life test with a test collector. The current values were higher by a factor of 8 than the initial vacuum emission data taken prior to the introduction of cesium (Fig. 1). These high values were in good agreement with the short-circuit current (Fig. 4). The emitter was allowed to cool and then it was heated and maintained at 2100°K for 5 hr to see if operation in the bell jar vacuum of 1×10^{-5} mm Hg would have an effect on the emission. The data showed a slight decrease in emission from the previous values.

During the vacuum emission measurements the emitter temperature was recorded on both thermocouples and sighted through the two holes in the test collector. In most cases the two pyrometer readings agreed within 10° . Both thermocouples were in agreement with the corrected pyrometer temperatures. This test has shown that the emitter temperature readings obtained during the life test were reliable.

The tip of the emitter thermocouple broke off when the emitter was cut off for analysis. It was found that the BeO insulation used in the thermocouple had disappeared in the upper 3/8 in. This same depletion of the BeO insulation had been observed in Cells G and J. Metallographical examination showed that the tantalum sheath had severely oxidized and failure was imminent. It is theorized that the tantalum reduces the BeO at emitter operating temperatures. Therefore, long-term operation of tantalum sheath thermocouples with BeO insulation at temperatures in excess of 2100°K may not be successful.

POST-TEST ANALYSIS OF CELLS G AND J

Cell G

Operation and the cause for termination of operation of Cells G and J is described in Ref. 1. To conduct a metallurgical and chemical analysis

on the emitter and collectors of both cells, the collector had to be removed. During this procedure, the emitter of Cell G was removed with the collector because dimensional changes of the two had caused them to fuse together. A dimensional check of either the emitter or the collector was not possible.

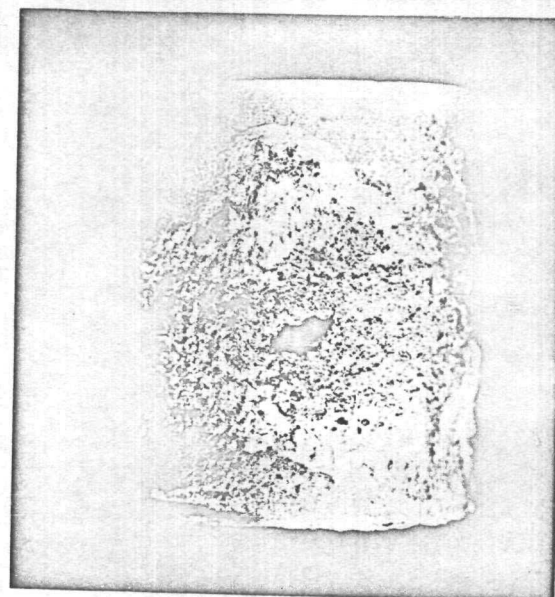
The emitter of Cell G had a number of large areas which showed no apparent sign of operating for 1700 hr in excess of 2000°K , while other areas exhibited deep erosions, cavities, and low carbide density (Fig. 7). These low-density areas protruded beyond the original dimensions of the emitter.

A longitudinal section of the emitter revealed that the serrations of the tantalum slug had melted away on the top and partially on the left-hand side while they had maintained their shape on the right-hand lower edge (Fig. 8). Since the outside edge of the emitter as well as the filament cavity were sharp straight edges, it is reasoned that the serration had not melted during cell operation but during hot pressing, which preceded the final machining on the inside and outside surfaces.

The photomicrograph of one of the serrations shows the extensive metallurgical interaction which has taken place during the 1700 hr of cell operation (Fig. 9). Large voids have opened up between the tantalum substrate and the carbide overlay as well as in the carbide itself. A considerable amount of tantalum has diffused into the carbide, while the latter has diffused throughout the tantalum substrate.

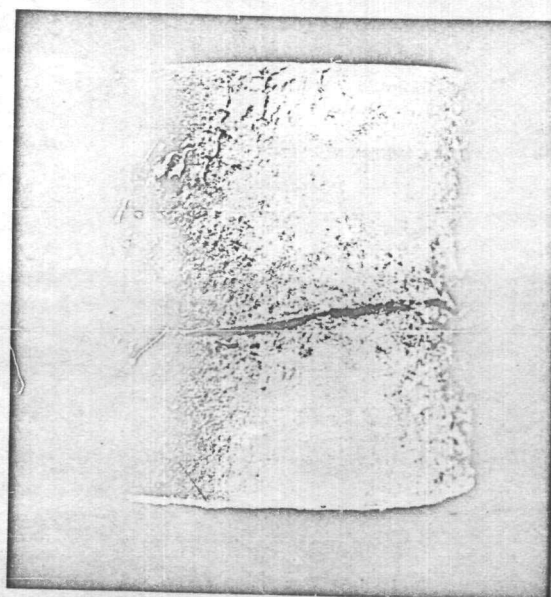
Cell J

In this cell the collector and emitter were not making contact at the end of the life test; however, a deposit in the collector prevented the complete withdrawal of the collector without removing the emitter as well. By heating the collector and applying some force, the emitter could be removed from the collector cavity.



← Note sharp edge on top of emitter

← Note heavily eroded area and low-density material



← Note clear areas showing no effects of operation at elevated temperature

Fig. 7 --Cell G emitter after 1753 hr of operation

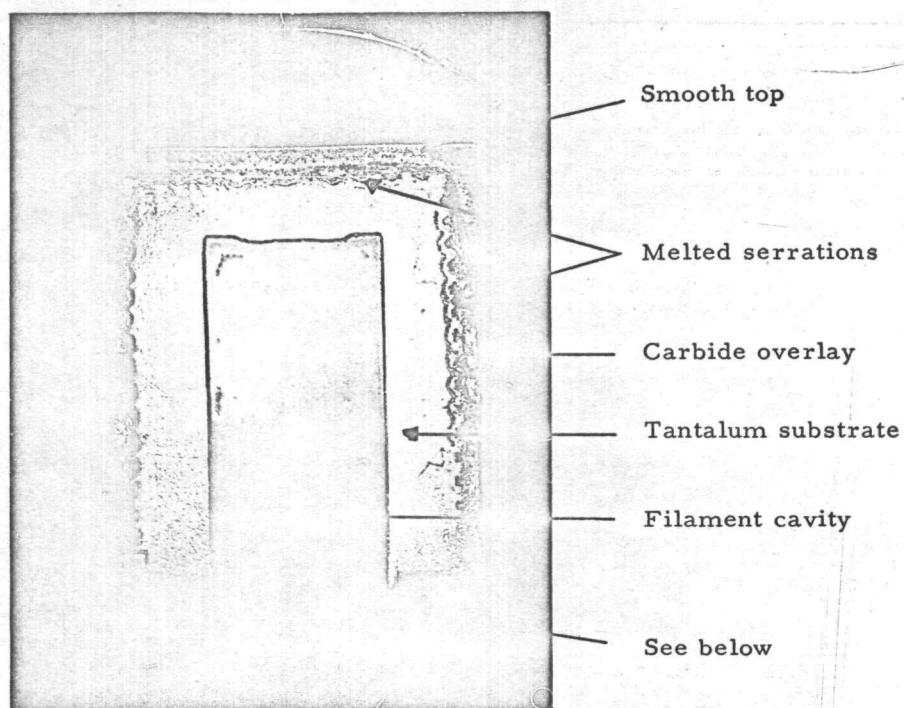


Fig. 8--Section of Cell G emitter

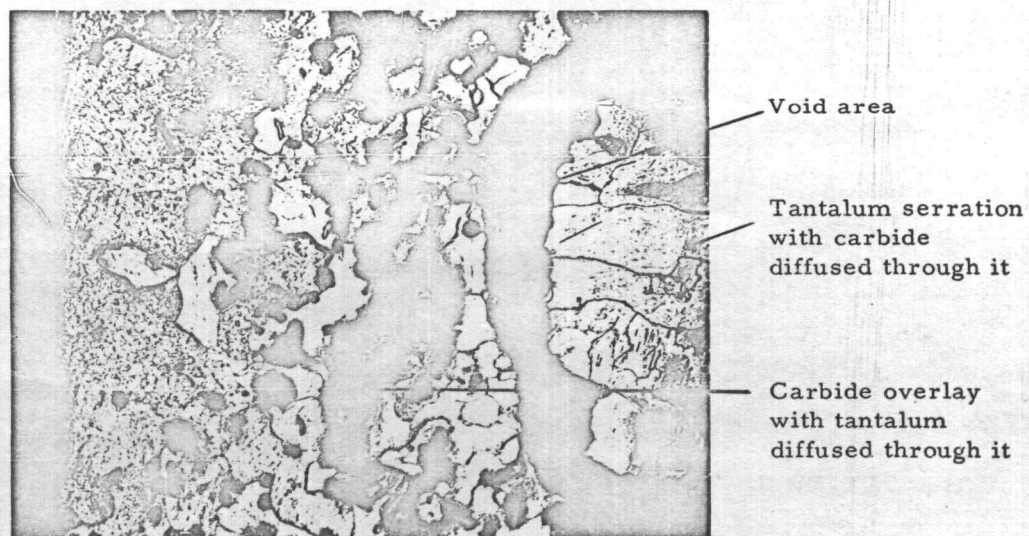


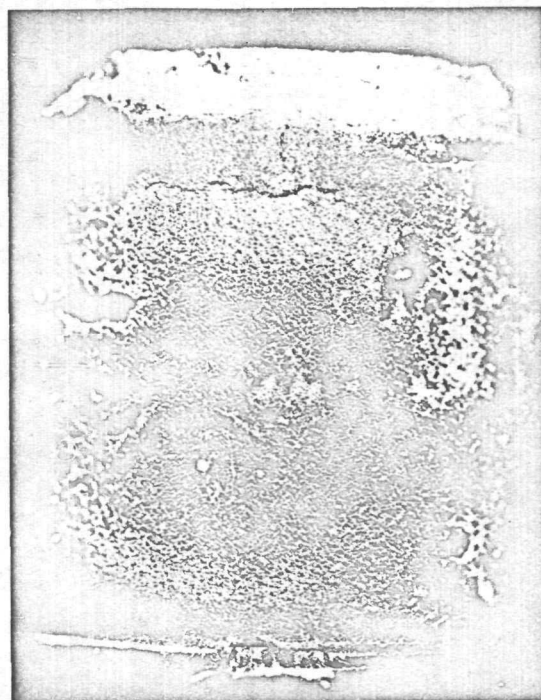
Fig. 9--Photomicrograph of Cell G emitter

A dimensional check of the emitter and collector diameters indicated why the emitter could not be withdrawn from the collector cavity. The original cold dimensions of the emitter and collector were 0.675 in. diameter and 0.700 in. diameter, respectively. After the long-term operation, the diameter at the middle section of the collector cavity had decreased to 0.674 in. At the same location the emitter diameter had decreased to 0.647 in. However, the very top of the emitter had retained its original dimension of 0.675 in., which produced an interference fit of 0.001 in. when these surfaces approached each other during removal of the collector. It is apparent that since the highest emitter temperature occurred at the middle section of the emitter, a higher rate of evaporation of UC-ZrC occurred there. The evaporated material was deposited at the same elevation in the collector, so that the original 10- to 20-mil spacing appears to have been maintained throughout the life of the experiment.

The emitter of Cell J was very porous over the entire cylindrical surface (Fig. 10), while the top (which had been cooler during operation) was much smoother and far less porous. Since the emitter operated at $2500^{\circ}\text{K} \pm 50^{\circ}$ for 90 hr (and in excess of 2000°K for 1427 hr), it is readily seen why the emitter showed signs of deterioration.

Chemical Analysis of Collectors of Cells G, J, and H

The deposit in the collector cavities of Cells G, J, and H was found to be very hard, and only very little could be scraped off. Additional material was machined off with a carbide tool. The metallurgical reaction of the deposit with the nickel substrate is attributed to the high collector temperature. Operating temperatures up to 1250°K were recorded. The chemical analysis of the material removed is tabulated below. Visual inspection of the collector cavities of Cells G and J after the removal of the deposit revealed that the penetration of the deposit is still deeper. This may account for the much lower uranium concentration in these two samples. A metallographical examination on a section of Cell G collector



← Top edge broke off
when emitter was
forcibly removed
from collector
cavity

Fig. 10--Cell J emitter (after 1427 hr of operation)

showed that the depth of penetration was 0.020 in. after removal of the deposit used for the chemical analysis below (Fig. 11).

Table I

	Collector Cell G	Collector Cell J	Collector Cell H
Total Deposit Removed (gm)	3.1	1.7	16.96
Nickel	93.0%	93.0%	97.0%
Analysis of Remainder of Deposit	in %		
Ta	3.6	3.1	<2.5
U	1.4	0.9	80.5
Zr	90.9	91.9	17.0
C	<1.2	4.0	Not Checked

The chemical analysis of the uranium-zirconium carbide overlay of the emitter from Cell H is tabulated below. The uranium content has depleted from 20% to 15%. The zirconium content has increased slightly.

Table II

ANALYSIS OF UC-ZrC OF CELL H EMITTER IN %

Composition	Carbon	Uranium	Zirconium	Tungsten	Tantalum
Original	10.32-10.50	20.37-20.40	68.50-68.81	1	*
After 700 Hr	11.60-12.00	15	70	*	2

*Not Detected

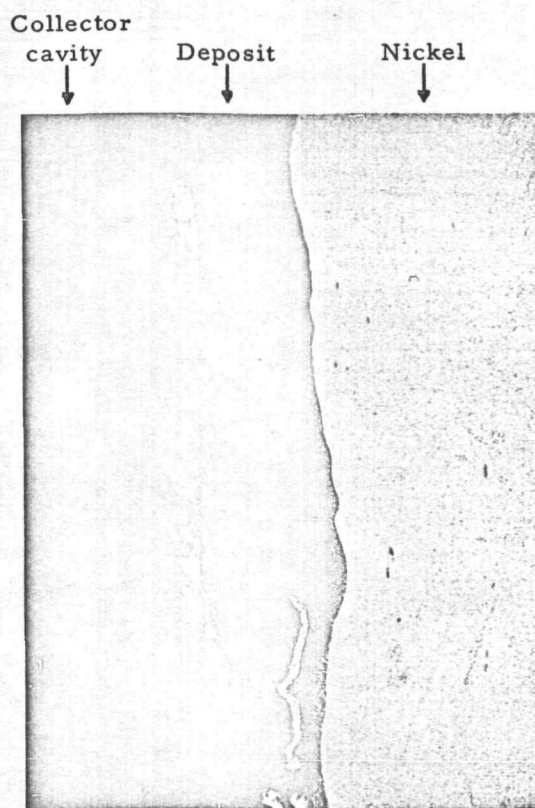


Fig. 11--Cell G collector with deposit
(Magnification - 50)

CELL K STATUS

Two additional emitters with the same 10 UC - 90 ZrC composition were fabricated for use in Cell K. Dimensionally, this cell is the same as previous cells; however, the electron-beam weld between the copper base and the Monel ring of the insulator which had leaked in all cells was eliminated by brazing the entire assembly in a hydrogen furnace. Besides eliminating the unreliable weld, the resulting assembly was free of contamination.

Vacuum emission studies were performed and are shown in Fig. 12. The values are comparable with those of Cell G, which were above average. They are about a factor of 3 higher than the data obtained from Cell H. Since in the past high vacuum emission and good cell performance were closely related, it is expected that Cell K will give high power output.

A temperature profile study on the emitter was conducted. The results are listed below (Table III). Points A, B, and C are three angular positions approximately 120°C apart. In the same radial plane the

Table III

	Run 1			Run 2		
Axial Position	Angular Position			Angular Position		
		Temperature in °K				
	A	B	C	A	B	C
Top	1770	1735	1745	1875	1891	1882
Upper	1800	1800	1785	1915	1950	1930
Middle	1811	1811	1805	1949	1970	1972
Lower	1735	1778	1751	1909	1908	1899
Bottom	1708	1708	1710	1852	1868	1842

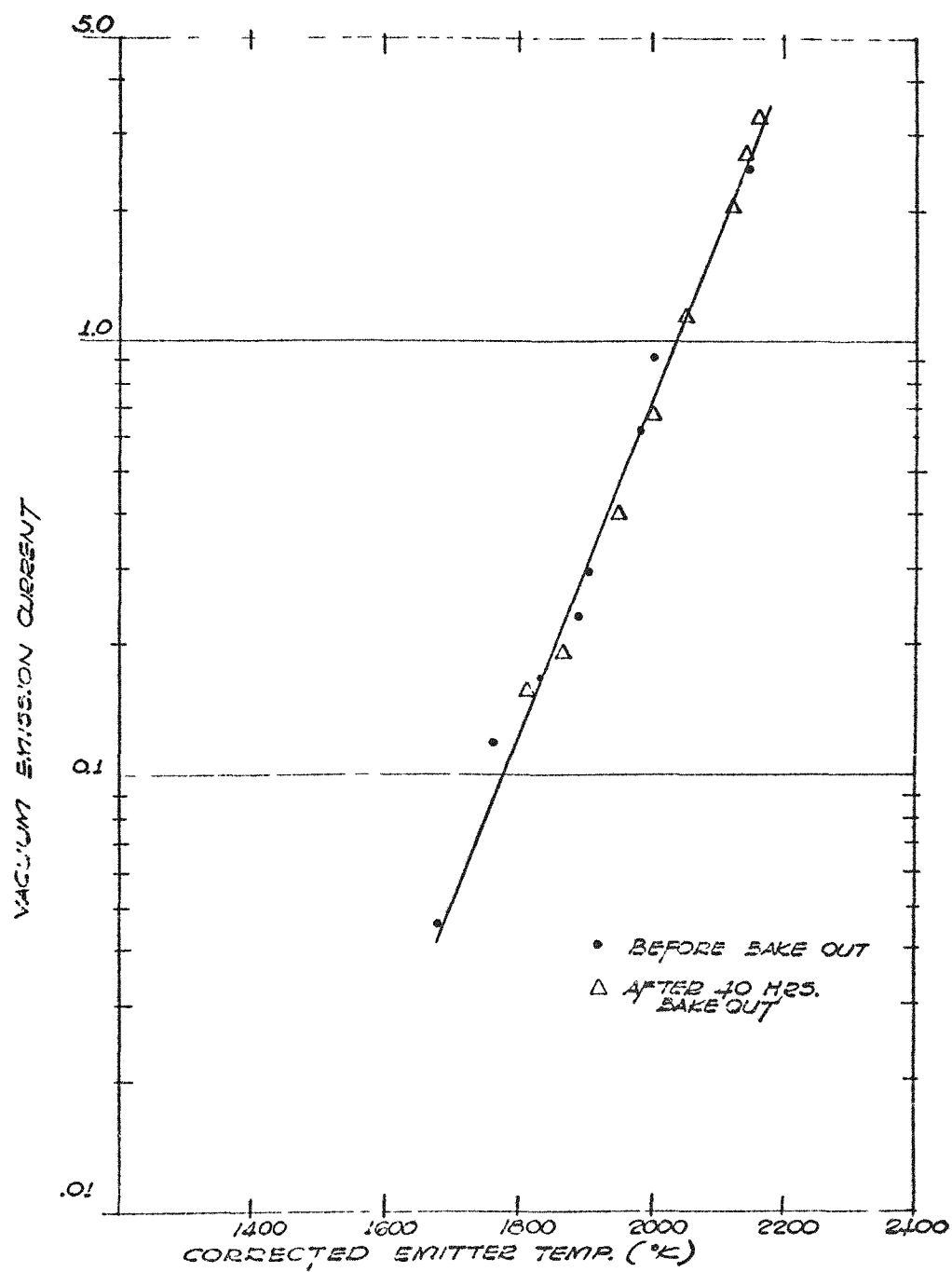


Fig. 12--Vacuum emission of Cell K,
emitter No. 43

maximum deviation was 35°K , while in the axial plane differences up to 130°K were observed.

Final bakeout of Cell K is now in progress. With the cell body at 670 to 770°K and the emitter at 2140°K , the pressure was 3×10^{-6} torr. Cell operation is anticipated in the next few days.

REFERENCE

1. Godsin, W., and R. Skoff, High-Temperature Vapor-Filled Thermionic Converter, Quarterly Technical Progress Report for the Period Ending April 30, 1963, General Atomic Report GA-4358, May 17, 1963.

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