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GENERAL ATOMIC
DIVISION OF GENERAL DYNAMICS

GA-3330

HIGH-TEMPERATURE VAPOR-FILLED THERMIONIC CONVERTER

QUARTERLY TECHNICAL PROGRESS REPORT

FOR THE PERIOD

MARCH 15 THROUGH JUNE 30, 1962

Contract AF 33(657)-8563
Project No. 3145, Task No. 60962-5

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

July 13, 1962

GENERAL ATOMIC

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GENERAL DYNAMICS

JOHN JAY HOPKINS LABORATORY FOR PURE AND APPLIED SCIENCE

P O BOX 608 SAN DIEGO 12 CALIFORNIA

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July 13, 1962

FOREWORD

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.

INTRODUCTION

The existing program of research on a high-temperature, vapor-filled thermionic converter is a continuation of the efforts initiated under Contract AF 33(616)-7422, under which a cell was operated for 400 hr and produced up to 1.7 watts/cm^2 of power.*

This quarterly report covers the work done during the period from March 15 to June 30, 1962, on a bench test of an unclad thermionic converter. The objective of this work is to produce an efficient, high-power-density, long-life thermionic power converter using a uranium-zirconium-carbide emitter and a high-temperature nickel collector.

At the start of this contract a cell (referred to as Mark V, Cell E) complete with an emitter was available. The collector, made of "A" nickel, was installed, and the cell operated for 1034 hr and produced up to 14 watts of power. The emitter used in Cell E was made of 10% UC and 90% ZrC powder hot-pressed onto a hollow tantalum stem. Although a number of minor failures occurred, the cell was still in good condition at the end of the run, though a decrease in power output had been observed. Cell operation was discontinued in order to study the effect of temperature and cesium on all critical components. Further bench testing of 10% UC – 90% ZrC has been discontinued, since higher uranium concentrations offer greater promise for use in thermionic reactor fuel elements.

OPERATION OF CELL E

The partially assembled cell was placed in a bell jar (see Fig. 1) and the emitter was heated to 2200°K in vacuum to drive off all gases. The collector was outgassed separately, then the two parts were joined and sealed by electron-beam welding. A small leak in the emitter thermocouple was detected, which was sealed by potting the external end of the thermocouple sheath with a low-vapor-pressure epoxy resin. After final bakeout, the high-temperature, high-vacuum valve was closed, the cesium ampule was broken, and the cell temperature was raised to 500°K.

The cell was operated continuously except for a few instances which will be discussed later. During periods of unattended operation, bihourly

*A. E. Campbell, F. D. Carpenter, J. B. Dunlay, and R. W. Pidd, High-temperature, Vapor-filled Thermionic Converter, General Atomic, Report GA-2911, April 4, 1962.

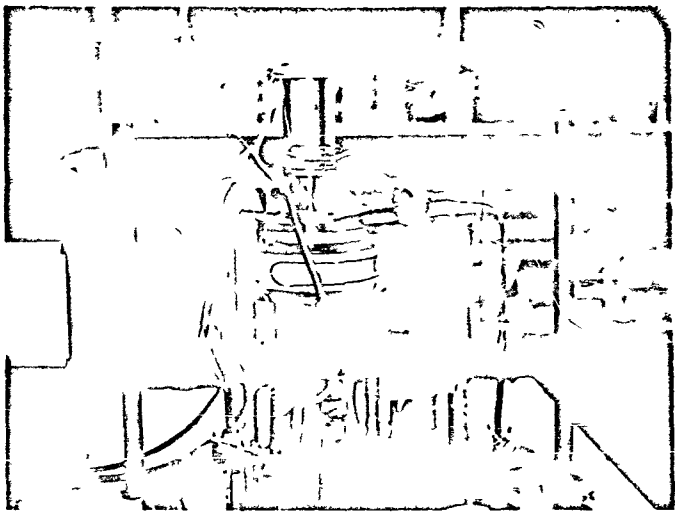


Fig. 1--Cell E in bell jar, with cooling coils, heaters, and thermocouples installed

checks were made on the pressure gauges, flow meters, ammeters, and volt meters. Safety circuits prevented damage to the cell and the experiment. During normal working hours, the emitter temperature and the cesium pressure were treated as variable test parameters. A fairly constant collector temperature of 750° to 800°K was maintained during the first 8 days of operation by the use of a water cooling coil. When this developed a leak, it was replaced and the collector floated up to higher temperatures. The emitter thermocouple began to act erratically and showed very low temperatures. It was believed that cesium had penetrated through the crack in the sheath and was condensing at a cooler point, shorting out the thermocouple leads. From a previously determined power-temperature correlation, however, the emitter temperature could be estimated with reasonable accuracy. All subsequent data were based on this emitter temperature estimation.

A loss of Dowtherm coolant flow due to low reservoir level occurred twice during the 1034 hr of operation and the cell was operated at a reduced input power while Dowtherm was added to the system (approximately 4 hr of refilling time). After 600 hr of operation, the bombardment filament burned out and was replaced. The ambient pressure surrounding the cell had to be brought to atmospheric pressure during this operation by introducing argon into the bell jar. Subsequent cell output was much lower than that previously obtained, but after a few days of operation it increased to a level of about one-half the original output. It is feared that the cell might

have been leaking, and thus a small amount of oxygen could have entered the cell while the filament was being replaced. The cell continued to operate at the reduced level until 1008 hr had been accumulated. Then the emitter temperature was raised to 2500°K to study the output at that temperature.

Low-cesium-pressure studies and back-emission studies were conducted just prior to dismantling of the cell.

POSTOPERATIVE ANALYSIS

After the conclusion of the run, an extensive postoperative analysis was initiated. The most important parts of this analysis were studies of the integrity of the cell and its components; the determination of the presence of gases, if any in the cell; a recalibration of the emitter surface temperature versus power input; and, finally, metallurgical and chemical examination of all major components.

The failure of Cell D (see Report GA-2911) had, in part, been attributed to contaminating gases within the cell. To obtain some information on these gases in Cell E, silica gel and a gas bulb were inserted into the external cell system upstream from the diffusion pump by the addition of a 3-way valve (see Fig. 2). During this process the external tubing was in contact with air and it was noted that a metallic deposit, which had formed in this external tubing during the 1000-hr run, had turned powdery white, a clear indication that cesium had leaked across the seat of the high-temperature, high-vacuum valve. Later it was found that the cell itself had also developed two leaks, one in the tantalum thermocouple sheath and the other in the copper-to-Kovar weld at the insulator. Gas analysis indicated the presence of nitrogen, oxygen, and argon, which was introduced when the cell was brought to atmospheric pressure. Through two holes drilled into the collector, a temperature calibration was made; the collector was then cut off and the calibration was repeated. The thermocouple began to function properly again once the cesium was pumped out of the cell. Detailed metallurgical examination and chemical analyses are now in progress on the emitter, the collector, and the insulator. Included in this examination is a quantitative analysis for uranium in deposits on the collector and for cesium penetration of the insulator, and a metallographic study of the UC – ZrC bond area. Preliminary analyses indicate that the condensate on the collector is primarily uranium.

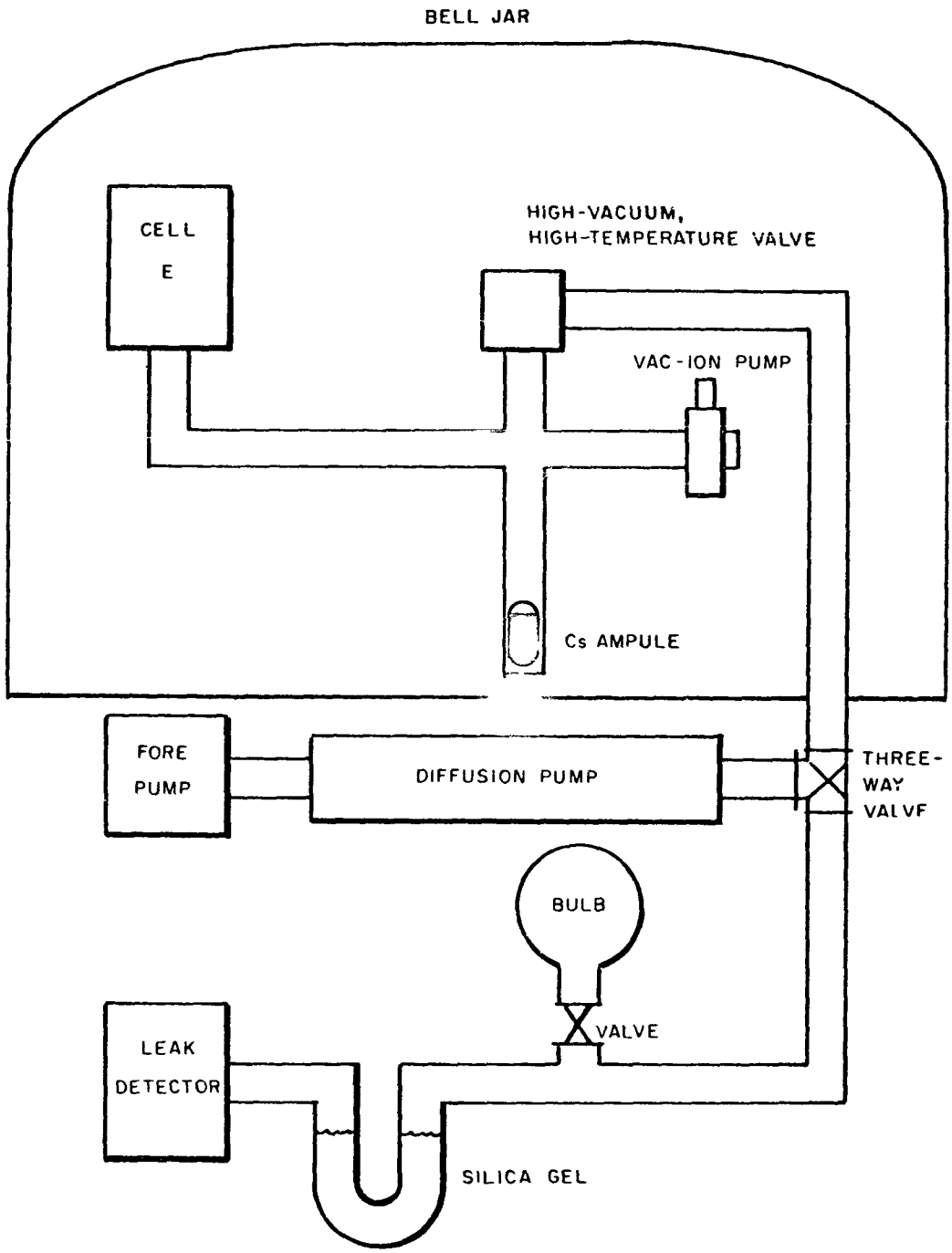


Fig. 2--Schematic of cell set up in bell jar

CELL E DATA

While the object of this project is primarily to demonstrate the long-life performance and high power density of an unclad thermionic converter, considerable data to broaden the understanding of such a cell was obtained from Cell E.

The maximum power output was 14 watts; this was obtained at an emitter temperature of 2414°K, a collector temperature of 1023°K, and a cesium pressure of 9 mm Hg. This output resulted in an efficiency of 2%. The cell output is plotted for a cesium pressure of 4 mm Hg and an emitter temperature of 2323°K over the total length of the run in Fig. 3. In many instances, the output for the exact emitter temperature was interpolated from curves of cell power output versus emitter temperature (see Fig. 4).

During the days for which no data point is shown in Fig. 3, the cell was operated either at higher or lower cesium pressures. A maximum occurs for the period between May 10 and May 18. When analyzing the history of the cell output data, it was found that higher cell output always followed periods of cesium pressure higher than 4 mm Hg. Although after operating conditions were changed, several hours elapsed before another set of data was taken, it is possible that this is insufficient time to allow for complete pressure equalization. It might be pointed out here that the cell has relatively large volume and that the connecting tubing to the cesium reservoir is long and of small diameter. This might also account for the lower points at the beginning of the run.

Following the failure of the filament (May 22), there was a very large drop in output power, which increased after several days of operation by a factor of two. Since the cell was at very low temperatures during the one day used for filament replacement and the next day, this may in part account for the low values. Aside from the fluctuations, there was a decrease in power output over the total operating period.

The relation of cesium pressure to cell power output is plotted for two different periods in Fig. 5. The five points obtained on April 26 were taken within a period of eight hours by starting at 0.5 mm Hg, then decreasing the pressure, and finally increasing it. The curve is relatively flat and shows a slight peak at 0.5 mm Hg. The next set of data was taken over a period of ten days. Cesium pressures were varied over the range from 1 mm to 9 mm Hg, but specific pressures were maintained for longer periods (up to 24 hr). These data show a definite increase in cell power output with increasing cesium pressure. A maximum output of 5.5 watts was obtained at a cesium pressure of 6 mm Hg, which had been maintained

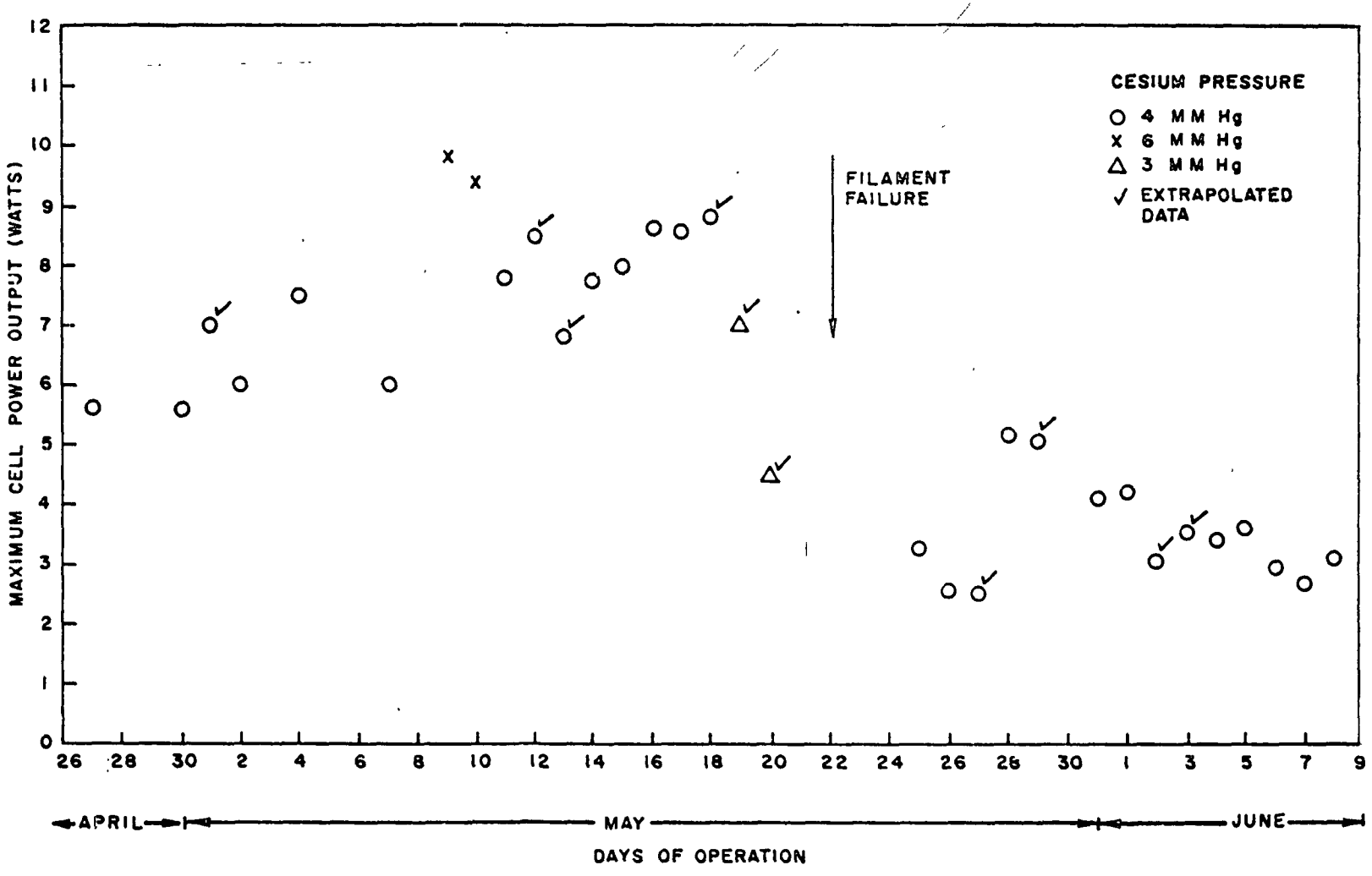


Fig. 3--Maximum cell power output versus day of operation for an emitter temperature of 2323°K

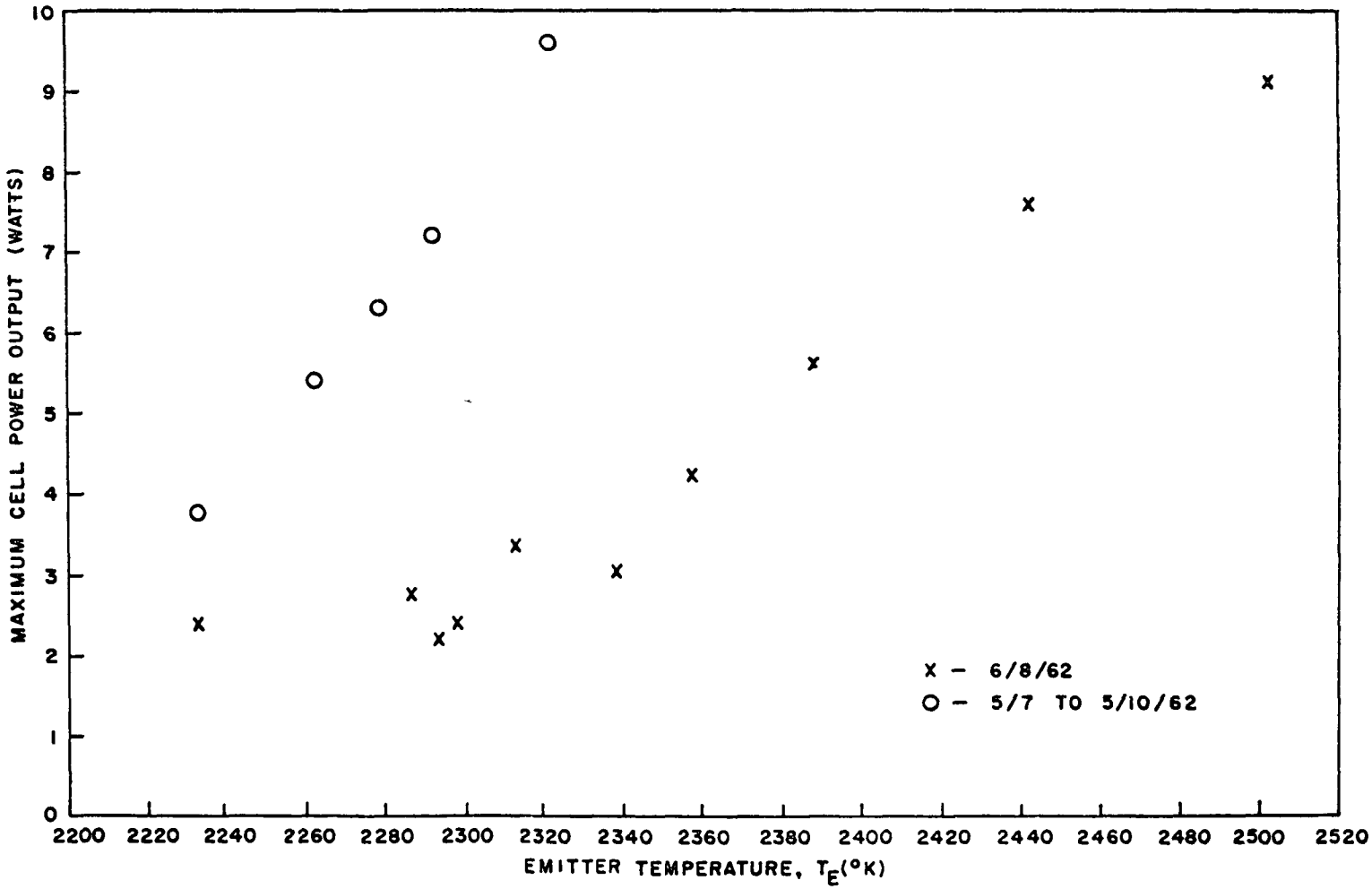


Fig. 4--Maximum cell power output versus emitter temperature for a cesium pressure of 4 mm Hg

for 24 hr. However, there is no indication from these data where a maximum occurs. For the bulk of the 1000-hr period the cell was operated at a cesium pressure of 4 mm Hg.

The emitter temperature determination was somewhat inaccurate during this test because of the failure of the emitter thermocouple. A careful comparison of the power input data versus surface temperature before and after the 1034 hr showed very close correlation for a bare emitter, i.e., with no collector (see Fig. 6). The good agreement of the thermocouple calibrations before and after the run substantiates the power input versus surface temperature correlation obtained during the early days of operation, from which all subsequent emitter temperature data were computed. A large difference existed between the power input requirement for a given emitter temperature before and after the collector was installed; this, however, is in agreement with the change in effective emissivity.

Since

$$p = \sigma E_b A T^4 ,$$

where p = power input = 600 watts,*
T = bare emitter temperature = 1800°K,
A = total emitter surface area = 14.5 cm^2,
σ = constant = 5.67×10^{-12} watts $cm^{-2}\,{}^{o}K^{-4}$,

then the emissivity for the bare emitter, E_b, is

$$E_b = \frac{p}{\sigma A T^4}$$

$$= \frac{600}{5.67 \times 14.5 \times 1.8^4}$$

$$= 0.7 .$$

The effective emissivity with the collector installed can be computed from the data obtained on April 26:

*It is assumed that (1) the heat lost by conduction is small in this design; (2) the power is consumed in the filament only--all other losses are negligible; (3) the emitter temperature is uniform; and (4) there is no radiation from the emitter stem.

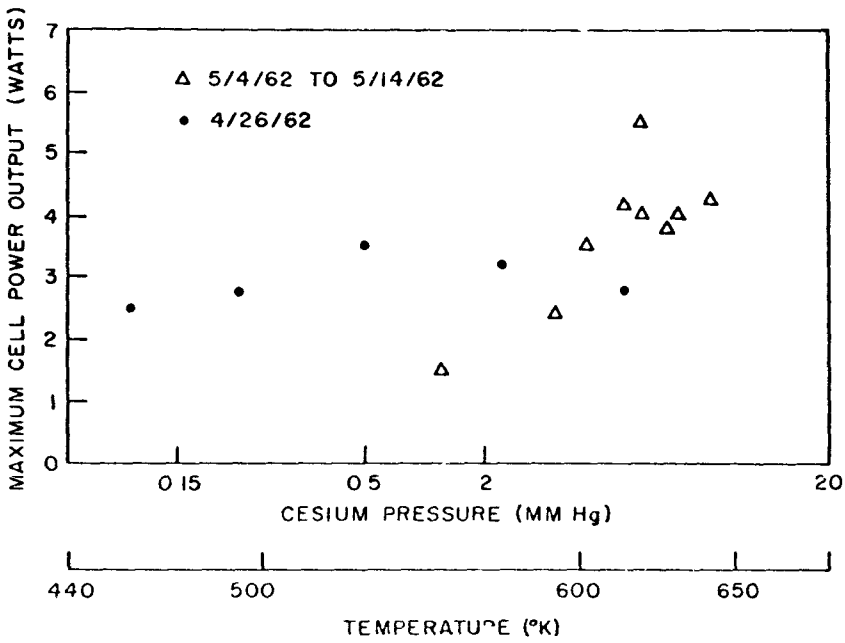


Fig. 5--Maximum cell power output versus cesium pressure for an emitter temperature of 2233°K

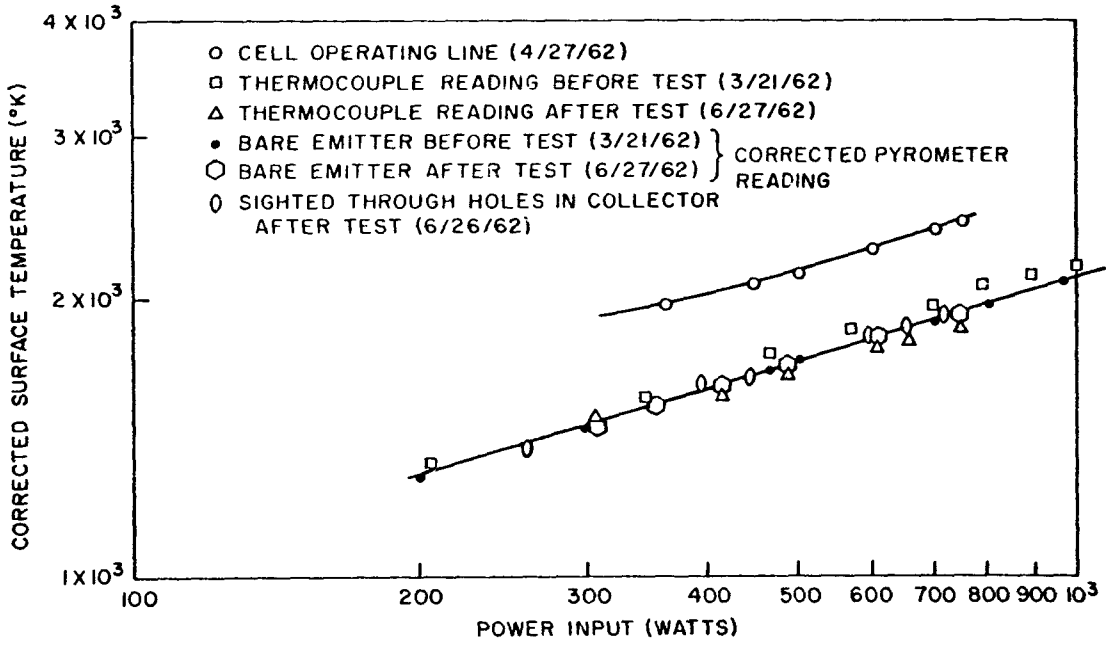


Fig. 6--Emitter temperature versus power input

$$E_{eff} = \frac{P}{\sigma A T^4}$$

$$= \frac{390}{5.67 \times 14.5 \times 2^4}$$

$$= 0.30,$$

where $T = 2000^{\circ}K$,
$P = 390$ watts.

The effective emissivity is also given as

$$E_{eff} = \frac{E_1 E_2}{E_2 + (1 - E_2)E_1},$$

where, for long coaxial cylinders,
E_1 = emissivity of emitter = 0.7,
E_2 = emissivity of collector.

Then,

$$E_2 = \frac{1}{\frac{1}{E_{eff}} + 1 - \frac{1}{E_1}}$$

$$= \frac{1}{\frac{1}{0.3} + 1 - \frac{1}{0.7}}$$

$$= 0.35 .$$

This high value of the emissivity of the collector indicates that it has become tarnished.

By increasing the collector temperature to $1040^{\circ}K$, it was noted that an increase in maximum cell output occurred (see Fig. 7). Reducing the collector temperature again did not reduce the power output to its original value.

It can be shown that a 10% decrease in effective emissivity can account for the increase in cell output observed upon returning to the original conditions. General Atomic sponsored research on deposition of carbides on nickel indicates a considerable decrease in deposition rate as the nickel temperature is increased from 400° to $1000^{\circ}K$. The evaporation

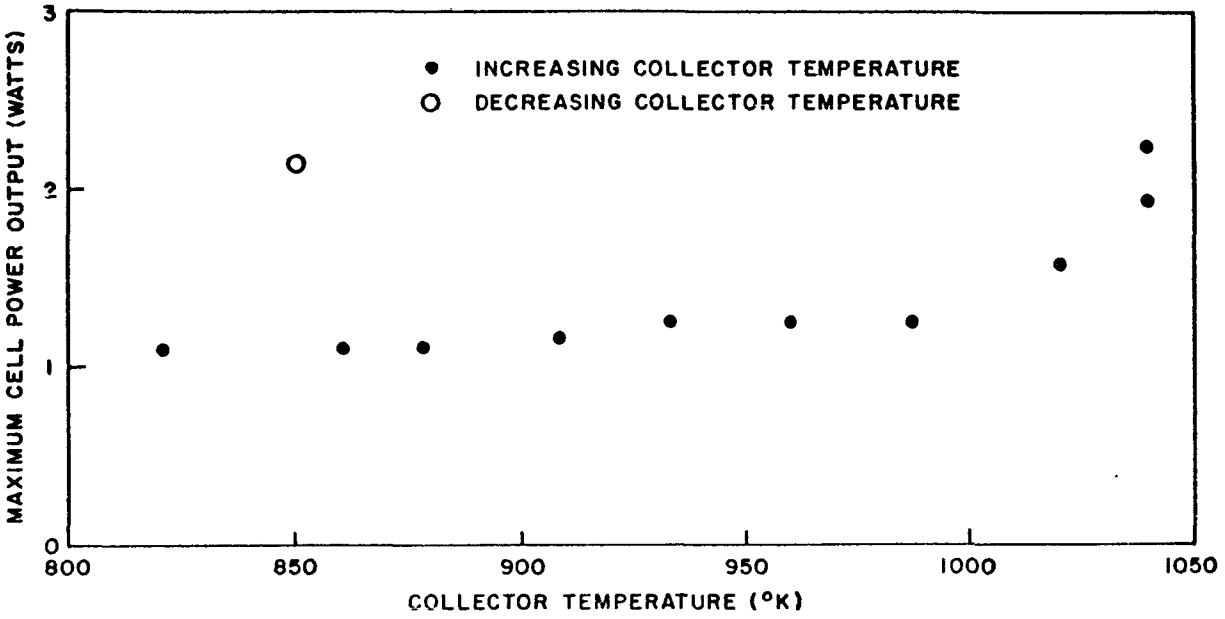


Fig. 7--Maximum cell power output versus collector temperature for an emitter temperature of 2113°K and a cesium pressure of 4 mm hg

rate of carbides from a nickel surface at 1000°K has not been studied as such; however, the performance of Cell E could be explained on the basis of a decrease in effective emissivity due to evaporation from the nickel surface.

The last tests on Cell E were back-emission studies at three different cesium pressures and an emitter temperature of 2070°K. The results are presented in Fig. 8, and some of the raw data are given in Fig. 9. There are two knees in the lower photograph of Fig. 9. The second one occurs when the voltage in the cell is equal to the ionization potential of cesium and could be attributed to a gas breakdown in the cell.

It may be stated that increasing collector temperature results in an increase in effective work function as well as a decrease in contact potential, which may be attributed to a decrease in wetting of cesium on the collector. The very sharp rise in current noted at a collector temperature of 940°K may be the formation of a significant ion current. It must be kept in mind that this cell was not designed to obtain precise physics data, and the resulting current is a complex sum of ion current and collector electron emission. Furthermore, the pressure measurement is somewhat inaccurate and equilibrium between the cell proper and its cesium reservoir

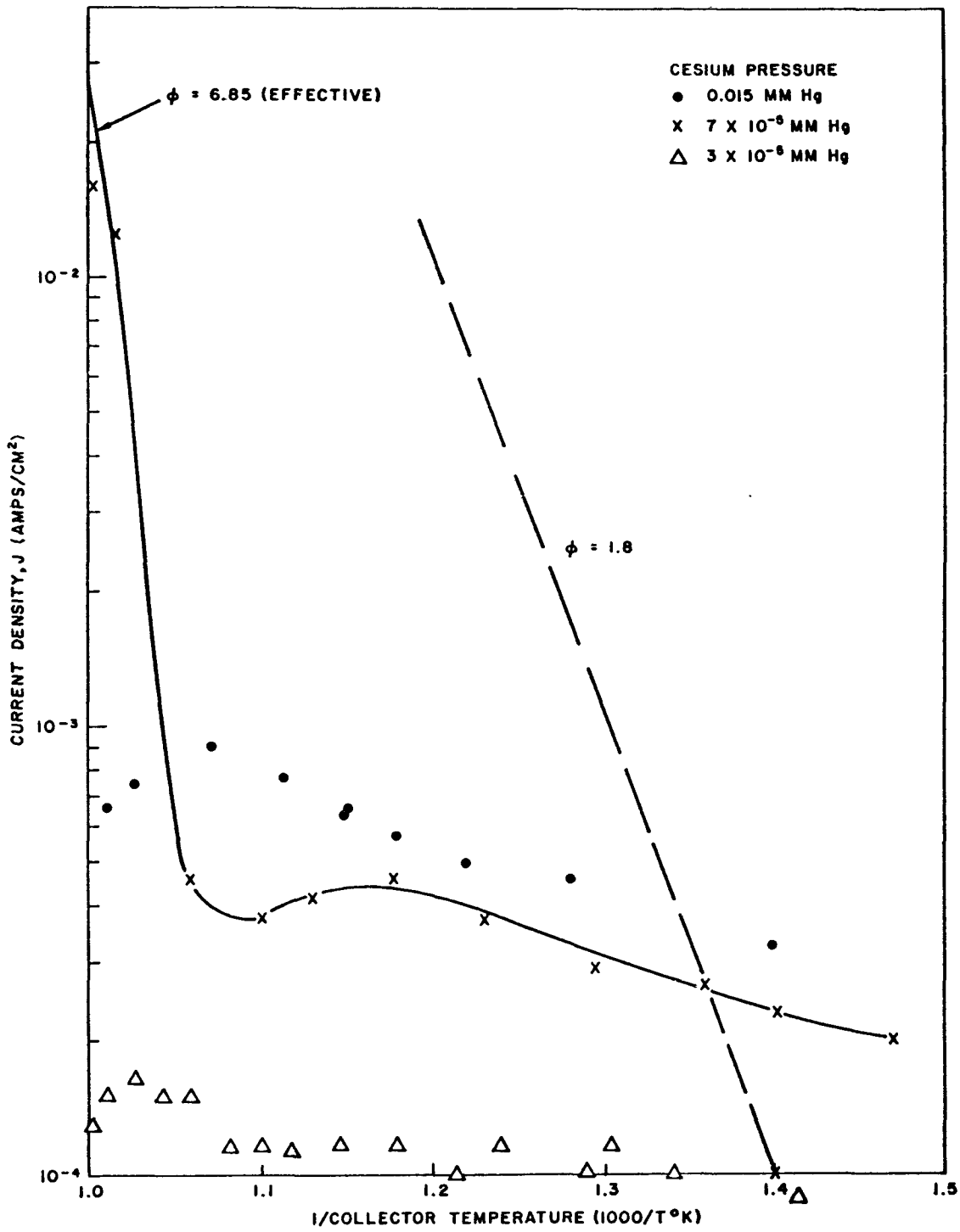
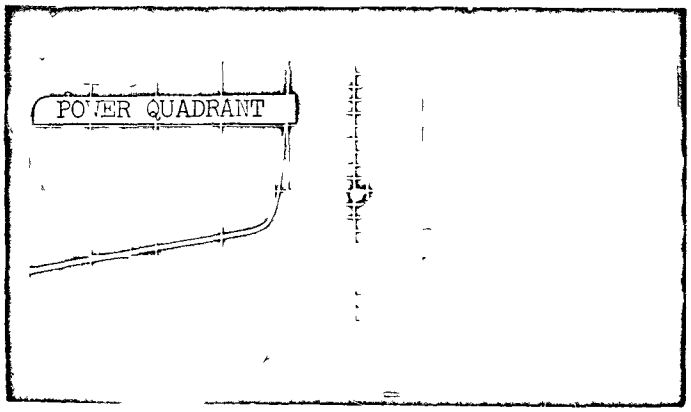
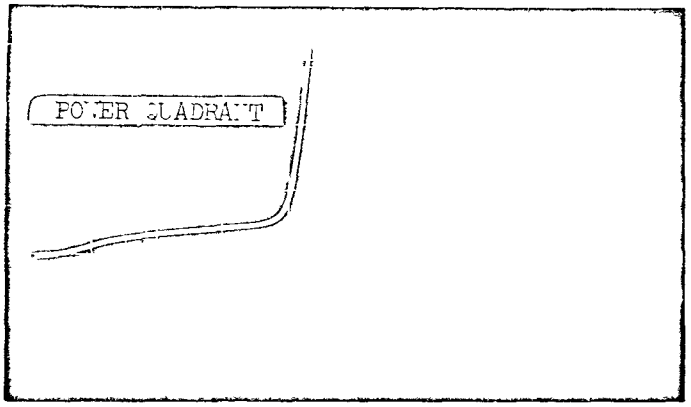


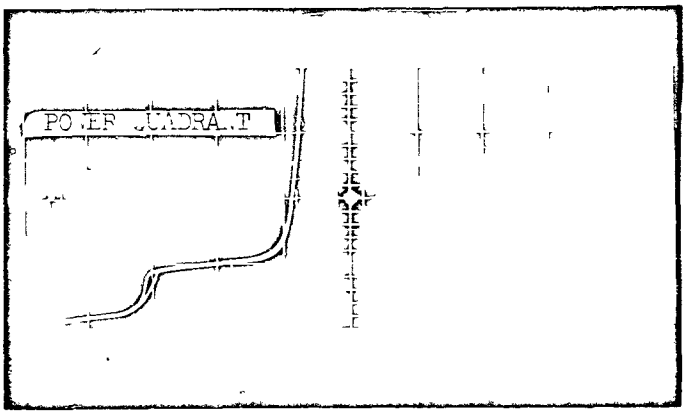
Fig. 8--Back emission versus collector temperature



Date, 6/12/62
Abscissa, 2 v/cm
Ordinate, 5 ma/cm
Emitter temperature, 2073°K
Collector temperature, 713°K
Cesium pressure, 0.015 mm Hg



Date, 6/12/62
Abscissa, 2 v/cm
Ordinate, 10 ma/cm
Emitter temperature, 2078°K
Collector temperature, 848°K
Cesium pressure, 0.015 mm Hg



Date, 6/12/62
Abscissa, 2 v/cm
Ordinate, 10 ma/cm
Emitter temperature, 2070°K
Collector temperature, 933°K
Cesium pressure, 0.016 mm Hg

Fig. 9--Back-emission studies at low cesium pressures

may not have been reached in the short time of the run at these low pressures. There is also good reason to question the effective area used in the computation of the data.

It is significant to note however, that there was cesium in the cell and on the collector up to the completion of the 1000-hr run and, furthermore, that no significant decrease in contact potential or increase in back emission occurred up to collector temperatures of 1000°K for cesium pressures up to 0.01 mm Hg.

CONCLUSIONS

Operation of Cell E has shown the feasibility of continuous, unattended operation of a thermionic converter. The high operating temperature of the collector makes it especially attractive for space-flight application. The small decrease in power output over the total 1034-hr run is not detrimental. Higher cesium pressures might result in higher output power. The maximum power output and the efficiency were lower than anticipated.

The successful operation of Cell E has shown that basic design of the hardware for Cell E was sound and reliable. New cells of the same basic design have been fabricated with the only major change being the addition of another emitter thermocouple and the increase in size of the thermocouples from 0.040 to 0.062 in. in diameter. Future cells will be sealed by pinching off the evacuating tube rather than by using a high-vacuum valve, which leaked across the seat during routine operation.

The major change anticipated in the next cells is higher uranium carbide concentration in the emitter. The bonding of a high-uranium-content carbide to the all-metal stem is the most severe problem at hand. It is presently believed that only a tungsten stem would be satisfactory because of the reaction of high mole per cent uranium carbide with tantalum. The major difficulty, however, is the large difference between the coefficient of thermal expansion of the tungsten and the uranium-zirconium carbide.

A number of methods to produce a vacuum-tight hollow support for the emitter have been designed, but only a few have been tried. An unsuccessful attempt was made to bond the uranium-zirconium-carbide powder to a tungsten-molybdenum alloy (85% W – 15% Mo) using a tantalum interfacial layer. More success was achieved with a low uranium concentration adjacent to the tungsten and a high concentration in the external layers. A continuing development program is under way to produce a high-UC emitter structure for life tests in the Mark V test stands.